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(73) 特許権者 000001926  
 塩野義製薬株式会社  
 大阪府大阪市中央区道修町3丁目1番8号  
 (74) 代理人 100113789  
 弁理士 杉田 健一  
 (74) 代理人 100103230  
 弁理士 高山 裕貢  
 (72) 発明者 糸 昌治  
 大阪府大阪市福島区鷺洲5丁目12番4号  
 塩野義製薬株式会社内  
 (72) 発明者 松尾 健二  
 大阪府大阪市福島区鷺洲5丁目12番4号  
 塩野義製薬株式会社内

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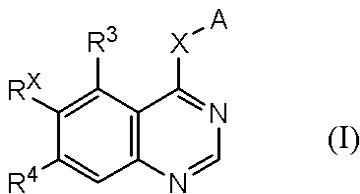
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(57) 【特許請求の範囲】

【請求項1】

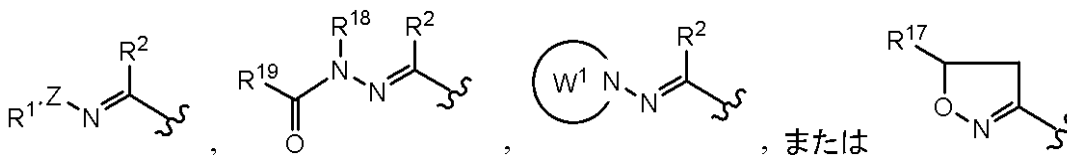
一般式(I) :

【化1】



[式中、R<sup>x</sup>は、式：

【化2】



(式中、R<sup>1</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアリール、または置換されていてもよい非芳香族含窒素複素環基；

Zは、 $-O-$ 、 $-N(R^{10})-$ 、または $-O-$ もしくは $-N(R^{11})-$ を介在してもよいアルキレン； $R^{10}$ および $R^{11}$ は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル；

$R^2$ は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいシクロアルキル、置換されていてもよいアリール、または置換されていてもよいヘテロアリール；

$R^{19}$ は、置換されていてもよいアルキル、置換されていてもよいアリール、置換されていてもよいヘテロアリール、または置換されていてもよい非芳香族複素環基；

$R^{18}$ は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよいアルキニル；

$W^1$ は、置換されていてもよい非芳香族含窒素複素環基または置換されていてもよいヘテロアリール；

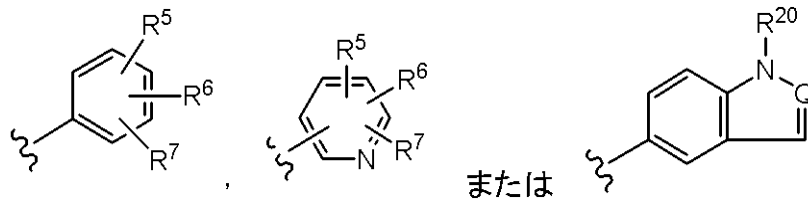
$R^{17}$ は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよい非芳香族含窒素複素環基)で表される基；

$R^3$ および $R^4$ は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、ハロゲン、ヒドロキシ、メルカプト、または置換されていてもよいアミノ；

Xは、 $-O-$ 、 $-S-$ 、 $-N(R^{12})-$ 、または $-O-$ 、 $-S-$ 、もしくは $-N(R^{13})-$ を介在してもよいアルキレン； $R^{12}$ および $R^{13}$ は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル；および

Aは、式：

【化3】



(式中、 $R^5$ は、水素原子、ハロゲン、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、または式： $-Y-R^8$  (式中、Yは $-O-$ 、 $-S-$ 、 $-SO_2-$ 、または $-O-$ 、 $-S-$ 、もしくは $-N(R^9)-$ を介在してもよいアルキレン； $R^8$ は置換されていてもよいアリールまたは置換されていてもよいヘテロアリール； $R^9$ は水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル)で表される基；

$R^6$ および $R^7$ は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、ハロゲン、ヒドロキシ、メルカプト、シアノ、または置換されていてもよいアミノ；

$R^{20}$ は、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、または式：

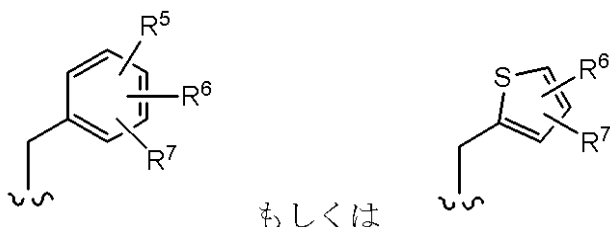
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## 【化4】



(式中、 $R^5$ 、 $R^6$ 、および $R^7$ は、前記と同意義)で表される基；

QはNまたは $CH_2$ )で表される基]

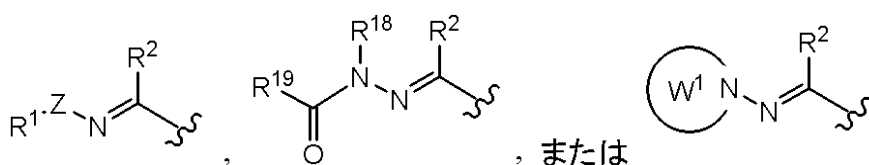
で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項2】

$R^x$ が、式：

## 【化5】



(式中、 $R^1$ 、 $R^2$ 、 $R^{18}$ 、 $R^{19}$ 、Z、および $W^1$ は、請求項1と同意義)で表される基である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項3】

$R^1$ が、置換基群A(置換基群A:ヒドロキシ、ヘテロアリール、カルボキシ、置換されていてもよいアミノ、置換されていてもよい非芳香族含窒素複素環基、置換されていてもよいアミノカルボニル、または置換されていてもよい非芳香族含窒素複素環カルボニル)から選択される置換基で置換されていてもよいアルキル、または置換基群B(置換基群B:アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、オキソ、アミノ、カルボキシ、シアノ、シアノアルキル、ヒドロキシアルキル、アルキルカルボニルアミノ、アルキルスルホニルアミノ、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項4】

$R^1$ が、置換基群C(置換基群C:アルキル、アルケニル、アルキニル、置換されていてもよいアリール、アラルキル、アルキルオキシ、ヒドロキシアルキル、ヒドロキシアルキルオキシアルキル、ハロアルキル、1または2のアルキルで置換されていてもよいアミノアルキル、アルキルスルホニル、アルキルスルホニルアルキル、ハロゲンもしくはアルキルオキシで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、置換されていてもよいシクロアルキル、カルボキシアルキル、置換されていてもよいアミノカルボニルアルキル、置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、アルキルチオアルキル、アルキルカルボニルアミノアルキル、アルキルスルホニルアミノアルキル、アルキルスルホニル(アルキル)アミノアルキル、アルキルオキシカルボニルアルキル、アルキルチオ(ヒドロキシ)アルキル、シクロアルキルアルキル、シアノアルキル、置換されていてもよいアミノアルキルカルボニル、置換されていてもよいヘテロアリール、ヘテロアリールアルキル、ヒドロキシアルキルオキシアルキル、置換されていてもよい非芳香族複素環基、置換されていてもよい非芳香族複素環アルキル)から選択される置換基で置換されていてもよいアミノで置換されたアルキル、置換基群D(置換基群D:ハロゲン、アリール、ヒドロキシ、オキソ、置換されていてもよいアミノカルボニル、アルキルオキシカルボニル、置換基群H(置換基群H:置換されていてもよいアミノカルボニル、シアノ、アルキルオキシ、アルキルスルホニルアミノ、アミノ、

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カルボキシ、アルキルオキシカルボニル、およびヒドロキシ)から選択される置換基で置換されていてもよいアルキル、アルキルアミノカルボニル、カルボキシ、シアノ、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル、アルキルオキシアルキルカルボニル、アルキルカルボニルアミノ、アミノカルボニルオキシ、非芳香族含窒素複素環基、および非芳香族複素環カルボニル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキル、または置換基群B(置換基群B:アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、オキソ、アミノ、カルボキシ、シアノ、シアノアルキル、ヒドロキシアルキル、アルキルカルボニルアミノ、アルキルスルホニルアミノ、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項5】

R<sup>19</sup>が、置換基群A(置換基群A:ヒドロキシ、ヘテロアリール、カルボキシ、置換されていてもよいアミノ、置換されていてもよい非芳香族含窒素複素環基、置換されていてもよいアミノカルボニル、または置換されていてもよい非芳香族含窒素複素環カルボニル)から選択される置換基で置換されていてもよいアルキル、置換基群I(置換基群I:アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、シアノアルキル、ヒドロキシアルキル、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよいアリール、または置換基群Iから選択される置換基で置換されていてもよいヘテロアリールである、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項6】

R<sup>19</sup>が、置換基群C(置換基群C:アルキル、アルケニル、アルキニル、置換されていてもよいアリール、アラルキル、アルキルオキシ、ヒドロキシアルキル、ヒドロキシアルキルオキシアルキル、ハロアルキル、1または2のアルキルで置換されていてもよいアミノアルキル、アルキルスルホニル、アルキルスルホニルアルキル、ハロゲンもしくはアルキルオキシで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、置換されていてもよいシクロアルキル、カルボキシアルキル、置換されていてもよいアミノカルボニルアルキル、置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、アルキルチオアルキル、アルキルカルボニルアミノアルキル、アルキルスルホニルアミノアルキル、アルキルスルホニル(アルキル)アミノアルキル、アルキルオキシカルボニルアルキル、アルキルチオ(ヒドロキシ)アルキル、シクロアルキルアルキル、シアノアルキル、置換されていてもよいアミノアルキルカルボニル、置換されていてもよいヘテロアリール、ヘテロアリールアルキル、ヒドロキシアルキルオキシアルキル、置換されていてもよい非芳香族複素環基、置換されていてもよい非芳香族複素環アルキル)から選択される置換基で置換されていてもよいアミノで置換されたアルキル、または置換基群D(置換基群D:ハロゲン、アリール、ヒドロキシ、オキソ、置換されていてもよいアミノカルボニル、アルキルオキシカルボニル、置換基群H(置換基群H:置換されていてもよいアミノカルボニル、シアノ、アルキルオキシ、アルキルスルホニルアミノ、アミノ、カルボキシ、アルキルオキシカルボニル、およびヒドロキシ)から選択される置換基で置換されていてもよいアルキル、アルキルアミノカルボニル、カルボキシ、シアノ、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル、アルキルオキシアルキルカルボニル、アルキルカルボニルアミノ、アミノカルボニルオキシ、非芳香族含窒素複素環基、および非芳香族複素環カルボニル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキルである、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項7】

W<sup>1</sup>が、置換基群J(置換基群J:アルキル、シアノアルキル、ヒドロキシアルキル、アミノカルボニルアルキル、アルキルオキシ、アルキルオキシカルボニル、置換されていて

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もよいアミノカルボニル、およびシアノ)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で表される基または置換基群Jから選択される置換基で置換されていてもよいヘテロアリールで表される基である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【請求項8】

R<sup>2</sup>が、水素原子、ハロゲンで置換されていてもよいC1 - C6アルキル、C2 - C6アルケニル、C2 - C6アルキニル、ハロゲン、またはフェニルである、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【請求項9】

R<sup>3</sup>およびR<sup>4</sup>の一方が水素原子、他方が水素原子、置換されていてもよいアルキルオキシまたは置換されていてもよいアルケニルオキシである、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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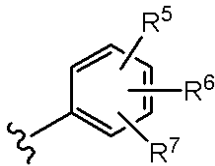
【請求項10】

Xが、-NH-である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【請求項11】

Aが、式：

【化6】



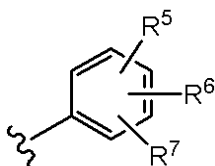
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(式中、R<sup>5</sup>は、水素原子またはハロゲン；R<sup>6</sup>は、ハロゲンまたはアルキニル；およびR<sup>7</sup>は水素原子)で表される基である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【請求項12】

Aが、式：

【化7】



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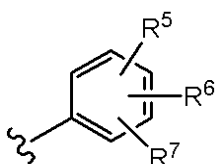
[式中、R<sup>5</sup>は、置換基群E(置換基群E：カルボキシ、アルキルオキシカルボニル、シクロアルキル、および置換されていてもよいアミノカルボニル)から選択される置換基で置換されていてもよいアルキルオキシ、置換基群Eから選択される置換基で置換されていてもよいアルケニルオキシ、または置換基群Eから選択される置換基で置換されていてもよいアルキニルオキシ；R<sup>6</sup>は、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；およびR<sup>7</sup>は水素原子]で表される基である、請求項1記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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【請求項13】

Aが、式：

【化8】



[式中、R<sup>5</sup>は、式：-Y-R<sup>8</sup>(式中、Yは、-O-を介在してもよいアルキレン；R

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<sup>8</sup> は、置換基群 F (置換基群 F : ハロゲン、カルボキシ、アルキル、ハロアルキル、ヒドロキシアルキル、アルキルオキシ、アルキルオキシカルボニル、および置換されていてもよいアミノ) から選択される置換基で置換されていてもよいフェニル、置換基群 F から選択される置換基で置換されていてもよいピリジル、置換基群 F から選択される置換基で置換されていてもよいフリル、置換基群 F から選択される置換基で置換されていてもよいチエニル、置換基群 F から選択される置換基で置換されていてもよいチアゾリル、または置換基群 F から選択される置換基で置換されていてもよいオキサゾリル) で表される基 ; R<sup>6</sup> が、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン ; および R<sup>7</sup> が水素原子] で表される基である、請求項 1 記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【請求項 14】

請求項 1 記載の化合物を有効成分として含有する医薬組成物。

## 【請求項 15】

請求項 1 記載の化合物を有効成分として含有する抗癌剤。

## 【発明の詳細な説明】

## 【技術分野】

## 【0001】

本発明は、EGF 受容体チロシンキナーゼおよび HER2 チロシンキナーゼの両方を阻害する化合物およびそれらを有効成分として含有する医薬組成物に関する。

## 【背景技術】

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## 【0002】

チロシンキナーゼは蛋白質中のチロシン残基をリン酸化する酵素であり、細胞の分化・増殖や細胞内情報伝達系において重要な役割を果たすことが知られている。特に、HER2 (ErbB2 または Neu と呼ばれる) および EGF 受容体等の増殖因子受容体チロシンキナーゼ (以下、受容体チロシンキナーゼ) が癌の形成に深く関与していること、ヒト癌において受容体チロシンキナーゼ活性が亢進していることが知られている (非特許文献 1、非特許文献 2、および非特許文献 3)。さらに、これら受容体チロシンキナーゼは脳、肺、胃、腸、膵臓、頭頸部、食道、膀胱、腎臓、前立腺、卵巣、乳、子宮、甲状腺等における腫瘍で過剰に発現していることが示されている (非特許文献 4 および特許文献 1)。したがって、チロシンキナーゼ阻害剤が、多くの癌種に適応しうる副作用の少ない抗癌剤として有用であると考えられる。チロシンキナーゼ阻害剤としては、特許文献 2、特許文献 3、特許文献 4、特許文献 5、特許文献 6、特許文献 7 等に記載の化合物が知られている。

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また、EGF 受容体と HER2 の共発現により EGF 受容体単独による癌化がさらに加速されることが知られている (非特許文献 5)。さらに、乳癌、口腔癌、肺癌等において EGF 受容体と HER2 の共発現があると予後不良になるとの報告がある (非特許文献 6)。EGF 受容体チロシンキナーゼと HER2 チロシンキナーゼの両方を阻害する薬剤としては、特許文献 8 等に記載の化合物が知られている。

キナゾリン誘導体の 4 位にオキシム型置換基を有する抗癌剤が特許文献 9 および特許文献 10 に記載されている。

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【特許文献 1】特開平 5 - 208911 号公報

【特許文献 2】国際公開第 92 / 20642 号パンフレット

【特許文献 3】欧州特許出願公開第 92305703 . 8 号明細書

【特許文献 4】欧州特許出願公開第 0566266 号明細書

【特許文献 5】欧州特許出願公開第 0602851 号明細書

【特許文献 6】欧州特許出願公開第 0520722 号明細書

【特許文献 7】国際公開第 98 / 02434 号パンフレット

【特許文献 8】国際公開第 02 / 066445 号パンフレット

【特許文献 9】国際公開第 2004 / 069145 号パンフレット

【特許文献 10】国際公開第 2004 / 105765 号パンフレット

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【非特許文献1】 キャンサー リサーチ (Cancer Res.) 1991年、第51巻、p.4430-4435

【非特許文献2】 キャンサー リサーチ (Cancer Res.) 1992年、第52巻、p.3636-3641

【非特許文献3】 キャンサー ケモセラピー アンド ファーマコロジー (Cancer Chemother. Pharmacol.)、1993年、第32巻、p.1-19

【非特許文献4】 エクスパート オピニオン オン インベスティゲーショナル ドラッグス (Expert. Opin. Invest. Drugs) 1994年、第3巻、第6号、p.577-595

【非特許文献5】 セル (Cell) 1987年、第58巻、p.287-292

【非特許文献6】 クリニカル キャンサー リサーチ (Clin. Cancer Res.) 1999年、第5巻、p.4164-4174

【発明の開示】

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【発明が解決しようとする課題】

【0003】

E G F 受容体と H E R 2 のデュアル阻害剤は単独のキナーゼにのみ作用する化合物と比較して、適応疾患が広く、デュアル阻害の相乗作用によってより強い治療効果が得られる点で優れている。したがって、E G F 受容体チロシンキナーゼおよび H E R 2 チロシンキナーゼの両方を阻害する化合物が求められていた。

【課題を解決するための手段】

【0004】

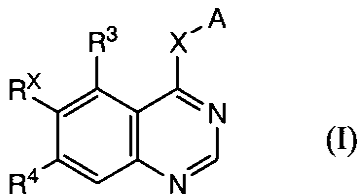
そこで、本発明者らは鋭意研究の結果、6位にある種の置換基を有するキナゾリン誘導体が、優れた E G F 受容体および H E R 2 のデュアル阻害作用を有することを見出した。

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すなわち、本発明は、

(1) 一般式 (I) :

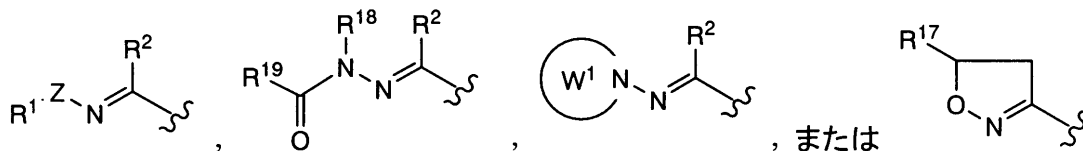
【化1】



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[式中、R<sup>X</sup>は、式：

【化2】



(式中、R<sup>1</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアリール、または置換されていてもよい非芳香族含窒素複素環基；

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Zは、-O-、-N(R<sup>10</sup>)-、または-O-もしくは-N(R<sup>11</sup>)-を介在してもよいアルキレン；R<sup>10</sup>およびR<sup>11</sup>は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラールキルオキシカルボニル；

R<sup>2</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいシクロアルキル、置換されていてもよいアリール、または置換されていてもよいヘテロアリール、；

R<sup>19</sup>は、置換されていてもよいアルキル、置換されていてもよいアリール、置換されていてもよいヘテロアリール、または置換されていてもよい非芳香族複素環基；

R<sup>18</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル

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、または置換されていてもよいアルキニル；

W<sup>1</sup> は、置換されていてもよい非芳香族含窒素複素環基または置換されていてもよいヘテロアリール；

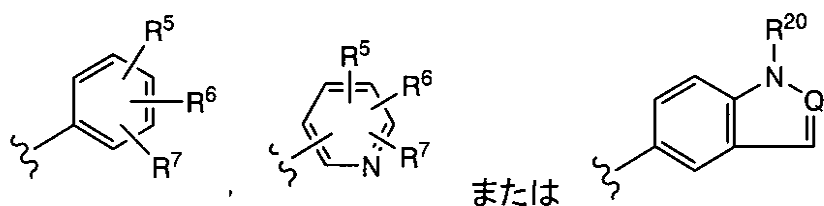
R<sup>1-7</sup> は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよい非芳香族含窒素複素環基) で表される基；

R<sup>3</sup> および R<sup>4</sup> は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、ハロゲン、ヒドロキシ、メルカプト、または置換されていてもよいアミノ；

X は、- O -、- S -、- N ( R<sup>1-2</sup> ) -、または - O -、- S -、もしくは - N ( R<sup>1-3</sup> ) - を介在してもよいアルキレン； R<sup>1-2</sup> および R<sup>1-3</sup> は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル；および

A は、式：

【化 3】

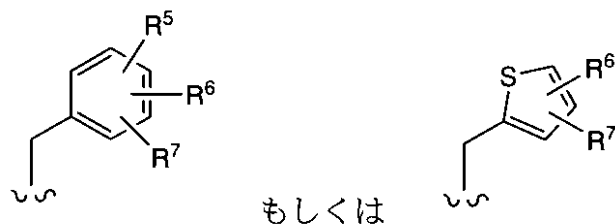


(式中、R<sup>5</sup> は、水素原子、ハロゲン、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、または式： - Y - R<sup>8</sup> (式中、Y は - O -、- S -、- S O<sub>2</sub> -、または - O -、- S -、もしくは - N ( R<sup>9</sup> ) - を介在してもよいアルキレン； R<sup>8</sup> は置換されていてもよいアリールまたは置換されていてもよいヘテロアリール； R<sup>9</sup> は水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル) で表される基；

R<sup>6</sup> および R<sup>7</sup> は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、ハロゲン、ヒドロキシ、メルカプト、シアノ、または置換されていてもよいアミノ；

R<sup>20</sup> は、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、または式：

【化 4】



(式中、R<sup>5</sup>、R<sup>6</sup>、および R<sup>7</sup> は、前記と同意義) で表される基；

Q は N または C H<sub>2</sub>) で表される基]

で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物、に関する。

【0005】

さらに詳しくは、以下の(2)~(20)に関する。

(2) R<sup>x</sup> が、式：

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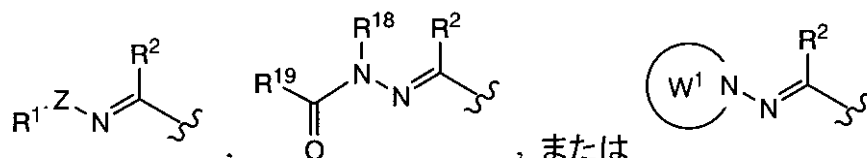
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## 【化5】



(式中、 $R^1$ 、 $R^2$ 、 $R^{18}$ 、 $R^{19}$ 、 $Z$ 、および $W^1$ は、(1)と同意義)で表される基である、(1)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(3)  $R^1$ が、置換基群A(置換基群A:ヒドロキシ、ヘテロアリール、カルボキシ、置換されていてもよいアミノ、置換されていてもよい非芳香族含窒素複素環基、置換されていてもよいアミノカルボニル、または置換されていてもよい非芳香族含窒素複素環カルボニル)から選択される置換基で置換されていてもよいアルキル、または置換基群B(置換基群B:アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、オキソ、アミノ、カルボキシ、シアノ、シアノアルキル、ヒドロキシアルキル、アルキルカルボニルアミノ、アルキルスルホニルアミノ、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基である、(1)または(2)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(4)  $R^1$ が、置換基群C(置換基群C:アルキル、アルケニル、アルキニル、置換されていてもよいアリール、アラルキル、アルキルオキシ、ヒドロキシアルキル、ヒドロキシアルキルオキシアルキル、ハロアルキル、1または2のアルキルで置換されていてもよいアミノアルキル、アルキルスルホニル、アルキルスルホニルアルキル、ハロゲンもしくはアルキルオキシで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、置換されていてもよいシクロアルキル、カルボキシアルキル、置換されていてもよいアミノカルボニルアルキル、置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、アルキルチオアルキル、アルキルカルボニルアミノアルキル、アルキルスルホニルアミノアルキル、アルキルスルホニル(アルキル)アミノアルキル、アルキルオキシカルボニルアルキル、アルキルチオ(ヒドロキシ)アルキル、シクロアルキルアルキル、シアノアルキル、置換されていてもよいアミノアルキルカルボニル、置換されていてもよいヘテロアリール、ヘテロアリールアルキル、ヒドロキシアルキルオキシアルキル、置換されていてもよい非芳香族複素環基、置換されていてもよい非芳香族複素環アルキル)から選択される置換基で置換されていてもよいアミノで置換されたアルキル、置換基群D(置換基群D:ハロゲン、アリール、ヒドロキシ、オキソ、置換されていてもよいアミノカルボニル、アルキルオキシカルボニル、置換基群H(置換基群H:置換されていてもよいアミノカルボニル、シアノ、アルキルオキシ、アルキルスルホニルアミノ、アミノ、カルボキシ、アルキルオキシカルボニル、およびヒドロキシ)から選択される置換基で置換されていてもよいアルキル、アルキルアミノカルボニル、カルボキシ、シアノ、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル、アルキルオキシアルキルカルボニル、アルキルカルボニルアミノ、アミノカルボニルオキシ、非芳香族含窒素複素環基、および非芳香族複素環カルボニル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキル、または置換基群Bから選択される置換基で置換されていてもよい非芳香族含窒素複素環基である(1)~(3)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(5)  $Z$ が $-O-$ である、(1)~(4)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

## 【0006】

(6)  $R^{19}$ が、置換基群Aから選択される置換基で置換されていてもよいアルキル、置換基群I(置換基群I:アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、シアノアルキル、ヒドロキシアルキル、およびアミノカルボニルアルキル)

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)から選択される置換基で置換されていてもよいアリール、または置換基群Iから選択される置換基で置換されていてもよいヘテロアリールである、(1)または(2)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(7)  $R^{19}$ が、置換基群Cから選択される置換基で置換されていてもよいアミノで置換されたアルキル、または置換基群Dから選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキルである、(1)、(2)、または(6)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(8)  $W^1$ が、置換基群J(置換基群J:アルキル、シアノアルキル、ヒドロキシアルキル、アミノカルボニルアルキル、アルキルオキシ、アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、およびシアノ)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で表される基または置換基群Jから選択される置換基で置換されていてもよいヘテロアリールで表される基である、(1)または(2)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(9)  $R^2$ が、水素原子、ハロゲンで置換されていてもよいC1-C6アルキル、C2-C6アルケニル、C2-C6アルキニル、ハロゲン、またはフェニルである、(1)~(8)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(10)  $R^2$ が、C2-C4アルキニルである、(1)~(9)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

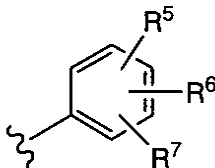
(11)  $R^3$ および $R^4$ の一方が水素原子、他方が水素原子、置換されていてもよいアルキルオキシまたは置換されていてもよいアルケニルオキシである、(1)~(10)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(12) Xが、-NH-である、(1)~(11)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【0007】

(13) Aが、式:

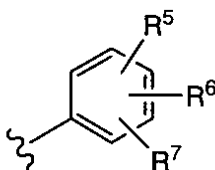
【化6】



(式中、 $R^5$ は、水素原子またはハロゲン； $R^6$ は、ハロゲンまたはアルキニル；および $R^7$ は水素原子)で表される基である、(1)~(12)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(14) Aが、式:

【化7】



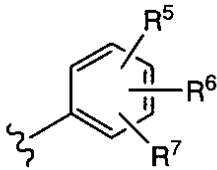
[式中、 $R^5$ は、置換基群E(置換基群E:カルボキシ、アルキルオキシカルボニル、シクロアルキル、および置換されていてもよいアミノカルボニル)から選択される置換基で置換されていてもよいアルキルオキシ、置換基群Eから選択される置換基で置換されていてもよいアルケニルオキシ、または置換基群Eから選択される置換基で置換されていてもよいアルキニルオキシ； $R^6$ は、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；および $R^7$ は水素原子]で表される基である、(1

) ~ (12) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【0008】

(15) A が、式：

【化8】



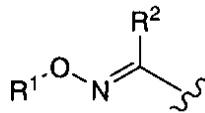
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[式中、 $R^5$  は、式： $-Y-R^8$  (式中、 $Y$ は、 $-O-$ を介在してもよいアルキレン； $R^8$  は、置換基群 F (置換基群 F：ハロゲン、カルボキシ、アルキル、ハロアルキル、ヒドロキシアルキル、アルキルオキシ、アルキルオキシカルボニル、および置換されていてもよいアミノ) から選択される置換基で置換されていてもよいフェニル、置換基群 F から選択される置換基で置換されていてもよいピリジル、置換基群 F から選択される置換基で置換されていてもよいフリル、置換基群 F から選択される置換基で置換されていてもよいチエニル、置換基群 F から選択される置換基で置換されていてもよいチアゾリル、または置換基群 F から選択される置換基で置換されていてもよいオキサゾリル) で表される基； $R^6$  が、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；および  $R^7$  が水素原子] で表される基である、(1) ~ (12) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(16)  $R^X$  が、式：

【化9】



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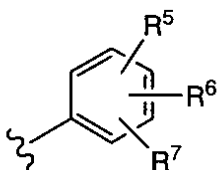
(式中、 $R^1$  はアルキル、ヒドロキシアルキル、アルキルカルボニルアミノアルキル、もしくは非芳香族複素環で置換されていてもよいアミノで置換されたアルキル；アルキル、ヒドロキシアルキル、もしくはアルキルカルボニルアミノで置換されていてもよい非芳香族含窒素複素環基で置換されたアルキル；またはヒドロキシアルキルで置換された非芳香族含窒素複素環基、 $R^2$  は C2 ~ C4 アルキニル) で表される基；

$R^3$  および  $R^4$  はいずれも水素原子；

$X$  は  $-NH-$ ；および

A は、式：

【化10】



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[式中、 $R^5$  は、式： $-Y-R^8$  (式中、 $Y$ は、 $-O-$ を介在してもよい C1 ~ C3 アルキレン； $R^8$  は、ハロゲンで置換されていてもよいフェニルまたはハロゲンで置換されていてもよいピリジル) で表される基； $R^6$  がハロゲン；および  $R^7$  が水素原子] で表される基である、(1) に記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(17)(1) ~ (16) のいずれかに記載の化合物を有効成分として含有する医薬組成物。

(18)(1) ~ (16) のいずれかに記載の化合物を有効成分として含有する抗癌剤。

(19)(1) ~ (16) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物を投与することを特徴とする癌の予防および/または治療方法。

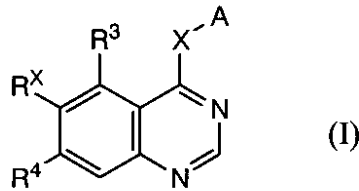
(20) 癌の予防および/または治療剤を製造するための(1) ~ (16) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物の使用。

【0009】

また、本発明は別の態様として以下の発明を包含する。

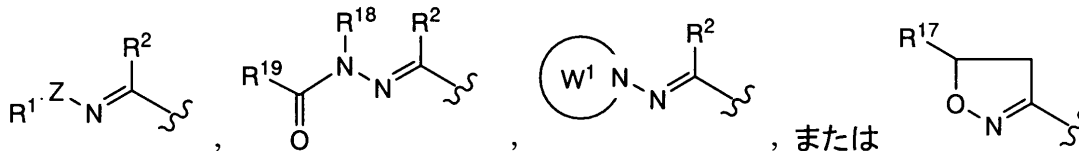
(I) 一般式(I) :

【化11】



[式中、R<sup>x</sup>は、式：

【化12】



(式中、R<sup>1</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよい非芳香族含窒素複素環基；

Zは、-O-もしくは-N(R<sup>10</sup>)-、または-O-もしくは-N(R<sup>11</sup>)-を介在してもよいアルキレン；R<sup>10</sup>およびR<sup>11</sup>は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル；

R<sup>2</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアリール、または置換されていてもよいヘテロアリール；

R<sup>19</sup>は、置換されていてもよいアルキル、置換されていてもよいアリール、置換されていてもよいヘテロアリール、または置換されていてもよい非芳香族複素環基；

R<sup>18</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよいアルキニル；

W<sup>1</sup>は、置換されていてもよい非芳香族含窒素複素環基または置換されていてもよいヘテロアリール；

R<sup>17</sup>は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよい非芳香族含窒素複素環基)で表される基；

R<sup>3</sup>およびR<sup>4</sup>は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、ハロゲン、ヒドロキシ、メルカプト、または置換されていてもよいアミノ；

Xは、-O-、-S-、-N(R<sup>12</sup>)-、または-O-、-S-、もしくは-N(R<sup>13</sup>)-を介在してもよいアルキレン；R<sup>12</sup>およびR<sup>13</sup>は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、また

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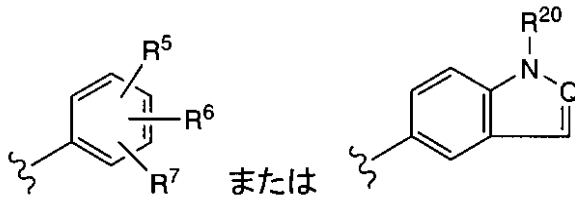
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はアラルキルオキシカルボニル；および

Aは、式：

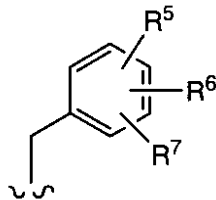
【化13】



(式中、 $R^5$  は、水素原子、ハロゲン、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、または式： $-Y-R^8$  (式中、 $Y$ は $-O-$ 、 $-S-$ 、または $-N(R^9)-$ を介在してもよいアルキレン； $R^8$ は置換されていてもよいアリールまたは置換されていてもよいヘテロアリール； $R^9$ は水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル)で表される基；

$R^6$ および $R^7$ は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、ハロゲン、ヒドロキシ、メルカプト、または置換されていてもよいアミノ；  
 $R^{20}$ は、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、または式：

【化14】



(式中、 $R^5$ 、 $R^6$ 、および $R^7$ は、前記と同意義)で表される基)；

QはNまたは $CH_2$ )で表される基]

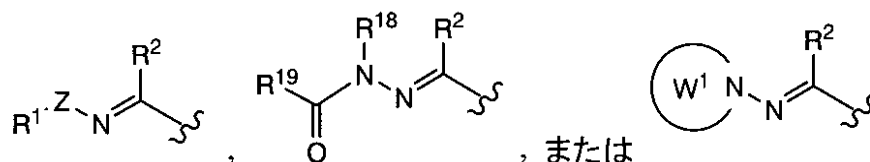
で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物、に関する。

【0010】

さらに詳しくは、以下の(II)~(XV)に関する。

(II)  $R^x$ が、式：

【化15】



(式中、 $R^1$ 、 $R^2$ 、 $R^{18}$ 、 $R^{19}$ 、 $Z$ 、および $W^1$ は、(I)と同意義)で表される基である、(I)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(III)  $R^1$ が、置換基群A' (置換基群A'：ヒドロキシ、置換されていてもよいアミノ、置換されていてもよい非芳香族含窒素複素環基、置換されていてもよいアミノカルボニル、または置換されていてもよい非芳香族含窒素複素環カルボニル)から選択される置換基で置換されていてもよいアルキル、または置換基群B' (置換基群B'：アルキル

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オキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、シアノアルキル、ヒドロキシアルキル、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基である、(I)または(II)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(IV)  $R^1$  が、置換基群 C' (置換基群 C' : アルキル、アルキニル、アルキルスルホニルアルキル、ヒドロキシアルキル、ハロゲンで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、シクロアルキル、置換されていてもよいアミノカルボニルアルキル、置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、シクロアルキルアルキル、シアノアルキル、および置換されていてもよいアミノアルキルカルボニル)から選択される置換基で置換されていてもよいアミノで置換されたアルキル、または置換基群 D' (置換基群 D' : ヒドロキシ、オキソ、置換されていてもよいアミノカルボニル、アルキルオキシカルボニル、置換基群 H' (置換基群 H' : 置換されていてもよいアミノカルボニル、シアノ、アルキルオキシ、およびヒドロキシ)から選択される置換基で置換されていてもよいアルキル、アルキルアミノカルボニル、シアノ、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル、アルキルオキシアルキルカルボニル、アルキルカルボニルアミノおよび非芳香族複素環カルボニル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキルである、(I) ~ (III)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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【0011】

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(V)  $R^1$  が、置換基群 A' から選択される置換基で置換されていてもよいアルキル、置換基群 I' (置換基群 I' : アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、シアノアルキル、ヒドロキシアルキル、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよいアリール、または置換基群 I' から選択される置換基で置換されていてもよいヘテロアリールである、(I)または(II)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(VI)  $R^1$  が、置換基群 C' から選択される置換基で置換されていてもよいアミノで置換されたアルキル、または置換基群 D' から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキルである、(I)、(II)、または(V)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(VII)  $W^1$  が、置換基群 J' (置換基群 J' : アルキル、シアノアルキル、ヒドロキシアルキル、アミノカルボニルアルキル、アルキルオキシ、アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、およびシアノ)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で表される基または置換基群 J' から選択される置換基で置換されていてもよいヘテロアリールで表される基である、(I)または(II)記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(VIII)  $R^2$  が、水素原子、ハロゲンで置換されていてもよい C1 - C6 アルキル、C2 - C6 アルケニル、C2 - C6 アルキニル、ハロゲン、またはフェニルである、(I) ~ (VI)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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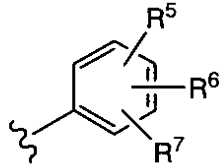
(IX)  $R^3$  および  $R^4$  の一方が水素原子、他方が水素原子、置換されていてもよいアルキルオキシまたは置換されていてもよいアルケニルオキシである、(I) ~ (VIII)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(X) X が、-NH- である、(I) ~ (IX)のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

【0012】

(XI) A が、式：

## 【化16】

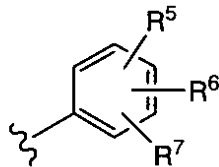


(式中、 $R^5$  は、水素原子またはハロゲン； $R^6$  は、ハロゲンまたはアルキニル；および  $R^7$  は水素原子) で表される基である、(I) ~ (IX) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(XII) Aが、式：

## 【化17】



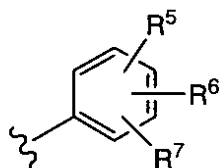
[式中、 $R^5$  は、置換基群 E' (置換基群 E' : カルボキシ、アルキルオキシカルボニル、シクロアルキル、および置換されていてもよいアミノカルボニル) から選択される置換基で置換されていてもよいアルキルオキシ、置換基群 E' から選択される置換基で置換されていてもよいアルケニルオキシ、または置換基群 E' から選択される置換基で置換されていてもよいアルキニルオキシ； $R^6$  は、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；および  $R^7$  は水素原子] で表される基である、(I) ~ (IX) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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## 【0013】

(XIII) Aが、式：

## 【化18】



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[式中、 $R^5$  は、式： $-Y-R^8$  (式中、Yは、 $-O-$  を介在してもよいアルキレン； $R^8$  は、置換基群 F' (置換基群 F' : ハロゲン、カルボキシ、アルキル、ハロアルキル、ヒドロキシアルキル、アルキルオキシ、アルキルオキシカルボニル、および置換されていてもよいアミノ) から選択される置換基で置換されていてもよいフェニル、置換基群 F' から選択される置換基で置換されていてもよいピリジル、置換基群 F' から選択される置換基で置換されていてもよいフリル、置換基群 F' から選択される置換基で置換されていてもよいチエニル、置換基群 F' から選択される置換基で置換されていてもよいチアゾリル、または置換基群 F' から選択される置換基で置換されていてもよいオキサゾリル) で表される基； $R^6$  が、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；および  $R^7$  が水素原子] で表される基である、(I) ~ (IX) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。(XIV) (I) ~ (XIII) のいずれかに記載の化合物を有効成分として含有する医薬組成物。

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(XV) (I) ~ (XIII) のいずれかに記載の化合物を有効成分として含有する抗癌剤。

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(XVI) (I) ~ (XIII) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物を投与することを特徴とする癌の予防および/または治療方法。

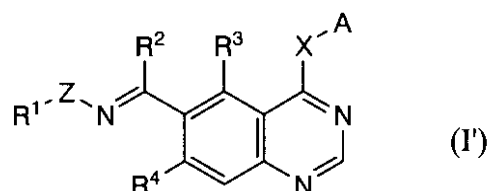
(XVII) 癌の予防および/または治療剤を製造するための (I) ~ (XIII) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物の使用。

【0014】

本発明のさらなる別の態様として、以下の発明を包含する。

(i) 一般式 (I') :

【化19】



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【式中、 $R^1$  は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよい非芳香族含窒素複素環基；

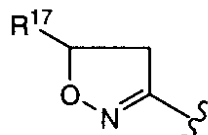
Z は、 $-O-$  もしくは  $-N(R^{10})-$ 、または  $-O-$  もしくは  $-N(R^{11})-$  を介してもよいアルキレン； $R^{10}$  および  $R^{11}$  は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル；

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$R^2$  は、水素原子、置換されていてもよいアルキル、アルケニル、アルキニル、置換されていてもよいアリール、または置換されていてもよいヘテロアリール；

または、 $-C(R^2)=N-Z-R^1$  で表される基が、式：

【化20】



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(式中、 $R^{17}$  は、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、または置換されていてもよい非芳香族含窒素複素環基) で表される基；

$R^3$  および  $R^4$  は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、ハロゲン、ヒドロキシ、メルカプト、または置換されていてもよいアミノ；

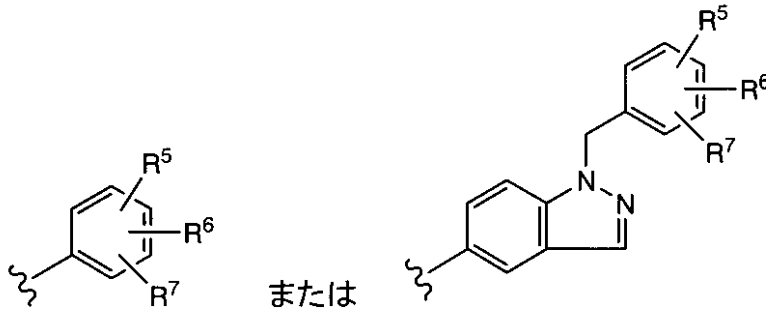
X は、 $-O-$ 、 $-S-$ 、もしくは  $-N(R^{12})-$ 、または  $-O-$ 、 $-S-$ 、もしくは  $-N(R^{13})-$  を介してもよいアルキレン； $R^{12}$  および  $R^{13}$  は、それぞれ独立して、水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル；および

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A は、式：



## 【化 2 1】



(式中、 $R^5$  は、水素原子、ハロゲン、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、または式： $-Y-R^8$  (式中、 $Y$  は  $-O-$ 、 $-S-$ 、または  $-N(R^9)-$  を介在してもよいアルキレン； $R^8$  は置換されていてもよいアリールまたは置換されていてもよいヘテロアリール； $R^9$  は水素原子、アルキル、アシル、アルキルオキシカルボニル、アルケニルオキシカルボニル、またはアラルキルオキシカルボニル) で表される基；

$R^6$  および  $R^7$  は、それぞれ独立して、水素原子、置換されていてもよいアルキル、置換されていてもよいアルケニル、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、置換されていてもよいアルケニルオキシ、置換されていてもよいアルキニルオキシ、ハロゲン、ヒドロキシ、メルカプト、または置換されていてもよいアミノ) で表される基]

で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(ii)  $R^1$  が、置換基群  $A'$  (置換基群  $A'$  : 置換されていてもよいアミノおよび置換されていてもよい非芳香族含窒素複素環基) から選択される置換基で置換されていてもよいアルキル、または置換基群  $B'$  (置換基群  $B'$  : アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、およびシアノ) から選択される置換基で置換されていてもよい非芳香族含窒素複素環基である、(i) 記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(iii)  $R^1$  が、置換基群  $C'$  (置換基群  $C'$  : アルキル、アルキルスルホニルアルキル、ハロゲンで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、シクロアルキル、置換されていてもよいアミノカルボニルアルキル、置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、および置換されていてもよいアミノアルキルカルボニル) から選択される置換基で置換されていてもよいアミノで置換されたアルキル、または置換基群  $D'$  (置換基群  $D'$  : オキシ、置換されていてもよいアミノカルボニル、アルキルオキシカルボニル、シアノもしくはアルキルオキシで置換されていてもよいアルキル、シアノ、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、およびアルキルスルホニルアルキルカルボニル) から選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキルである、(i) または (ii) 記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(iv)  $R^3$  および  $R^4$  の一方が水素原子、他方が水素原子、置換されていてもよいアルキルオキシまたは置換されていてもよいアルケニルオキシである、(i) ~ (iii) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(v)  $X$  が、 $-NH-$  である、(i) ~ (iv) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

(vi)  $A$  が、式：

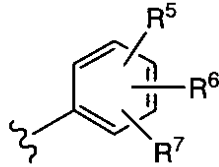
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## 【化 2 2】

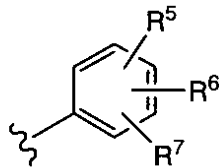


(式中、 $R^5$  は、水素原子またはハロゲン； $R^6$  は、ハロゲンまたはアルキニル；および  $R^7$  は水素原子) で表される基である、(i) ~ (v) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(vii) A が、式：

## 【化 2 3】

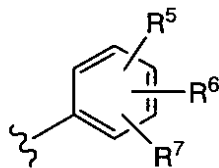


[式中、 $R^5$  は、置換基群 E' (置換基群 E' : カルボキシ、アルキルオキシカルボニル、および置換されていてもよいアミノカルボニル) から選択される置換基で置換されていてもよいアルキルオキシ、置換基群 E' から選択される置換基で置換されていてもよいアルケニルオキシ、または置換基群 E' から選択される置換基で置換されていてもよいアルキニルオキシ； $R^6$  は、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；および  $R^7$  は水素原子] で表される基である、(i) ~ (vi) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(viii) A が、式：

## 【化 2 4】



[式中、 $R^5$  は、式： $-Y-R^8$  (式中、Y は、 $-O-$  を介在してもよいアルキレン； $R^8$  は、置換基群 F' (置換基群 F' : ハロゲン、カルボキシ、アルキル、ハロアルキル、ヒドロキシアルキル、アルキルオキシ、アルキルオキシカルボニル、および置換されていてもよいアミノ) から選択される置換基で置換されていてもよいフェニル、置換基群 F' から選択される置換基で置換されていてもよいピリジル、置換基群 F' から選択される置換基で置換されていてもよいフリル、置換基群 F' から選択される置換基で置換されていてもよいチエニル、置換基群 F' から選択される置換基で置換されていてもよいチアゾリル、または置換基群 F' から選択される置換基で置換されていてもよいオキサゾリル) で表される基； $R^6$  が、置換されていてもよいアルキニル、置換されていてもよいアルキルオキシ、またはハロゲン；および  $R^7$  が水素原子] で表される基である、(i) ~ (vii) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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(ix) (i) ~ (viii) のいずれかに記載の化合物を有効成分として含有する医薬組成物。

(x) (i) ~ (viii) のいずれかに記載の化合物を有効成分として含有する抗癌剤。

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( x i ) ( i ) ~ ( v i i i ) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物を投与することを特徴とする癌の予防および/または治療方法。

( x i i ) 癌の予防および/または治療剤を製造するための ( i ) ~ ( v i i i ) のいずれかに記載の化合物もしくはその製薬上許容される塩またはそれらの溶媒和物の使用。

【 0 0 1 5 】

本明細書中、「ハロゲン」とは、フッ素、塩素、臭素、ヨウ素を意味する。フッ素、塩素、および臭素が好ましい。

【 0 0 1 6 】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「アルキル」とは、炭素原子数 1 ~ 1 0 の直鎖または分枝鎖の 1 価の炭化水素基を包含する。例えば、メチル、エチル、n - プロピル、イソプロピル、n - ブチル、イソブチル、s e c - ブチル、t e r t - ブチル、n - ペンチル、イソペンチル、n e o - ペンチル、n - ヘキシル、イソヘキシル、n - ヘプチル、n - オクチル、n - ノナニル、n - デカニル等が挙げられる。好ましくは、C 1 ~ C 1 0 アルキルが挙げられる。さらに好ましくは、C 1 ~ C 6 アルキルが挙げられる。最も好ましくは、C 1 ~ C 4 アルキルが挙げられる。

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「ハロアルキル」なる用語は、前記「ハロゲン」によって 1 ~ 8 箇所、好ましくは 1 ~ 5 箇所置換された前記「アルキル」を包含する。例えば、トリフルオロメチル、トリクロロメチル、ジフルオロエチル、トリフルオロエチル、ジクロロエチル、トリクロロエチル等が挙げられる。好ましくは、前記「ハロゲン」によって 1 ~ 5 箇所置換された C 1 ~ C 6 アルキルが挙げられる。特に、前記「ハロゲン」によって 1 ~ 3 箇所置換された C 1 ~ C 3 アルキルが好ましい。最も好ましくは、トリフルオロメチルが挙げられる。

本明細書中、「アルキルスルホニル」としては、メチルスルホニル、エチルスルホニル、プロピルスルホニル等が挙げられる。好ましくは、C 1 ~ C 6 アルキルスルホニルが挙げられる。特に、C 1 ~ C 3 アルキルスルホニルが好ましい。

本明細書中、「アルキルスルホニルアルキル」としては、メチルスルホニルメチル、メチルスルホニルエチル、メチルスルホニルプロピル、メチルスルホニルブチル、メチルスルホニルプロピル等が挙げられる。好ましくは、C 1 ~ C 6 アルキルスルホニル C 1 ~ C 6 アルキルが挙げられる。特に、C 1 ~ C 3 アルキルスルホニル C 1 ~ C 4 アルキルが好ましい。

本明細書中、「アルキルオキシ」としては、メチルオキシ、エチルオキシ、n - プロピルオキシ、イソプロピルオキシ、n - ブチルオキシ、イソブチルオキシ、s e c - ブチルオキシ、t e r t - ブチルオキシ、n - ペンチルオキシ、n - ヘキシルオキシ、n - ヘプチルオキシ、n - オクチルオキシ、n - ノナニルオキシ、n - デカニルオキシ等が挙げられる。好ましくは、C 1 ~ C 6 アルキルオキシが挙げられる。特に、C 1 ~ C 3 アルキルオキシが好ましい。

本明細書中、「アルキルオキシアルキル」としては、メチルオキシメチル、メチルオキシエチル、メチルオキシプロピル、エチルオキシメチル、エチルオキシエチル、エチルオキシプロピル等が挙げられる。好ましくは、C 1 ~ C 6 アルキルオキシ C 1 ~ C 6 アルキルが挙げられる。特に、C 1 ~ C 3 アルキルオキシ C 1 ~ C 4 アルキルが好ましい。

本明細書中、「アルキルオキシカルボニル」としては、メチルオキシカルボニル、エチルオキシカルボニル、n - プロピルオキシカルボニル、イソプロピルオキシカルボニル、n - ブチルオキシカルボニル、t - ブチルオキシカルボニル、n - ペンチルオキシカルボニル等が挙げられる。好ましくは、C 1 ~ C 6 アルキルオキシカルボニルが挙げられる。特に、C 1 ~ C 3 アルキルオキシカルボニルが好ましい。

本明細書中、「アルキルカルボニル」としては、メチルカルボニル、エチルカルボニル、n - プロピルカルボニル、イソプロピルカルボニル、n - ブチルカルボニル、t - ブチルカルボニル、n - ペンチルカルボニル等が挙げられる。好ましくは、C 1 ~ C 6 アルキルカルボニルが挙げられる。特に、C 1 ~ C 3 アルキルカルボニルが好ましい。

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本明細書中、「ハロアルキルオキシ」としては、トリフルオロメチルオキシ、トリクロロメチルオキシ、ジフルオロエチルオキシ、トリフルオロエチルオキシ、ジクロロエチルオキシ、トリクロロエチルオキシ等が挙げられる。好ましくは、ハロC<sub>1</sub>~C<sub>6</sub>アルキルオキシが挙げられる。特に、好ましくは、ハロC<sub>1</sub>~C<sub>3</sub>アルキルオキシが好ましい。最も好ましくは、トリフルオロメチルオキシが挙げられる。

本明細書中、「アミノカルボニルアルキル」としては、アミノカルボニルメチル、アミノカルボニルエチル、アミノカルボニルプロピル、アミノカルボニルブチル等が挙げられる。好ましくは、アミノカルボニルC<sub>1</sub>~C<sub>6</sub>アルキルが挙げられる。特に、アミノカルボニルC<sub>1</sub>~C<sub>3</sub>アルキルが好ましい。

本明細書中、「アミノカルボニルアルキルオキシ」としては、アミノカルボニルメチルオキシ、アミノカルボニルエチルオキシ、アミノカルボニルプロピルオキシ、アミノカルボニルブチルオキシ等が挙げられる。好ましくは、アミノカルボニルC<sub>1</sub>~C<sub>6</sub>アルキルオキシが挙げられる。特に、アミノカルボニルC<sub>1</sub>~C<sub>3</sub>アルキルオキシが好ましい。

本明細書中、「ヒドロキシアルキル」としては、ヒドロキシメチル、ヒドロキシエチル、ヒドロキシプロピル、ヒドロキシブチル等が挙げられる。好ましくは、ヒドロキシC<sub>1</sub>~C<sub>6</sub>アルキルが挙げられる。特に、ヒドロキシC<sub>1</sub>~C<sub>3</sub>アルキルが好ましい。

本明細書中、「アルキルチオ」としては、メチルチオ、エチルチオ、プロピルチオ、ブチルチオ等が挙げられる。好ましくは、C<sub>1</sub>~C<sub>6</sub>アルキルチオが挙げられる。特に、C<sub>1</sub>~C<sub>3</sub>アルキルチオが好ましい。

#### 【0017】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「アルキレン」とは、炭素原子数1~4の直鎖または分枝鎖の2価の炭化水素基を包含する。例えば、メチレン、エチレン、プロピレン、ブチレン等が挙げられる。好ましくは、C<sub>1</sub>~C<sub>3</sub>アルキレンが挙げられる。特に、C<sub>1</sub>~C<sub>2</sub>アルキレンが好ましい。

Zにおける「-O-もしくは-N(R<sup>1</sup>)<sub>1</sub>-を介在してもよいアルキレン」としては、-CH<sub>2</sub>-O-、-O-CH<sub>2</sub>-、-CH<sub>2</sub>-N(R<sup>1</sup>)<sub>1</sub>-、-N(R<sup>1</sup>)<sub>1</sub>-CH<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-O-、-O-(CH<sub>2</sub>)<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-N(R<sup>1</sup>)<sub>1</sub>-、-N(R<sup>1</sup>)<sub>1</sub>-(CH<sub>2</sub>)<sub>2</sub>-等を包含する。特に、-O-CH<sub>2</sub>-、-N(R<sup>1</sup>)<sub>1</sub>-CH<sub>2</sub>-、-O-(CH<sub>2</sub>)<sub>2</sub>-、-N(R<sup>1</sup>)<sub>1</sub>-(CH<sub>2</sub>)<sub>2</sub>-等が好ましい。

Xにおける「-O-、-S-、または-N(R<sup>1</sup>)<sub>2</sub>-を介在してもよいアルキレン」としては、-CH<sub>2</sub>-O-、-O-CH<sub>2</sub>-、-CH<sub>2</sub>-S-、-S-CH<sub>2</sub>-、-CH<sub>2</sub>-N(R<sup>1</sup>)<sub>2</sub>-、-N(R<sup>1</sup>)<sub>2</sub>-CH<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-O-、-O-(CH<sub>2</sub>)<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-S-、-S-(CH<sub>2</sub>)<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-N(R<sup>1</sup>)<sub>2</sub>-、-N(R<sup>1</sup>)<sub>2</sub>-(CH<sub>2</sub>)<sub>2</sub>-等を包含する。

Yにおける「-O-、-S-、または-N(R<sup>9</sup>)-を介在してもよいアルキレン」としては、-CH<sub>2</sub>-O-、-O-CH<sub>2</sub>-、-CH<sub>2</sub>-S-、-S-CH<sub>2</sub>-、-CH<sub>2</sub>-N(R<sup>9</sup>)-、-N(R<sup>9</sup>)-CH<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-O-、-O-(CH<sub>2</sub>)<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-S-、-S-(CH<sub>2</sub>)<sub>2</sub>-、-(CH<sub>2</sub>)<sub>2</sub>-N(R<sup>9</sup>)-、-N(R<sup>9</sup>)-(CH<sub>2</sub>)<sub>2</sub>-、-O-CH(CH<sub>3</sub>)-等を包含する。

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「シクロアルキル」とは、炭素原子数が3~8個であるシクロアルキルを包含する。例えば、シクロプロピル、シクロブチル、シクロペンチル、シクロヘキシル、シクロヘプチル、シクロオクチルが挙げられる。好ましくはC<sub>5</sub>~C<sub>6</sub>シクロアルキルが挙げられる。

#### 【0018】

本明細書中、「アルケニル」とは、炭素原子数が2~8個であり、1個もしくは2個以上の二重結合を有する、直鎖または分枝鎖の1価の炭化水素基を包含する。三重結合を鎖内に有していてもよい。例えば、ビニル、アリル、1-プロペニル、2-プロペニル、種々のブテニル異性体等が挙げられる。好ましくは、C<sub>2</sub>~C<sub>6</sub>アルケニルが挙げられる。さらに好ましくは、C<sub>2</sub>~C<sub>4</sub>アルケニルが挙げられる。

本明細書中、「アルケニルオキシ」としては、ビニルオキシ、アリルオキシ、1-プロ

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ペニルオキシ、2 - プロペニルオキシ、種々のブテニルオキシが挙げられる。好ましくは、C 2 ~ C 6 アルケニルオキシが挙げられる。特に、C 2 ~ C 4 アルケニルオキシが好ましい。

本明細書中、「アルケニルオキシカルボニル」としては、ビニルオキシカルボニル、アリルオキシカルボニル、1 - プロペニルオキシカルボニル、2 - プロペニルオキシカルボニル、種々のブテニルオキシカルボニルが挙げられる。好ましくは、C 2 ~ C 6 アルケニルオキシカルボニルが挙げられる。特に、C 2 ~ C 4 アルケニルオキシカルボニルが好ましい。

本明細書中、「アルキニル」とは、炭素原子数が2 ~ 8個であり、1個もしくは2個以上の三重結合を有する、直鎖または分枝鎖の1価の炭化水素基を包含する。例えば、エチニル、1 - プロピニル、2 - プロピニル、1 - ブチニル、2 - ブチニル、3 - ブチニル、1 - ペンチニル、2 - ペンチニル、種々のペンチニル異性体等が挙げられる。好ましくは、C 2 ~ C 6 アルキニルが挙げられる。さらに好ましくは、C 2 ~ C 4 アルキニルが挙げられる。

本明細書中、「アルキニルオキシ」としては、エチニルオキシ、プロピニルオキシ、ブチニルオキシ、ペンチニルオキシ等が挙げられる。好ましくは、C 2 ~ C 6 アルキニルオキシが挙げられる。特に、C 2 ~ C 4 アルキニルオキシが好ましい。

#### 【0019】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「アリアル」とは、単環状もしくは縮合環状芳香族炭化水素を包含する。例えば、フェニル、1 - ナフチル、2 - ナフチル、アントリル等が挙げられる。フェニル、1 - ナフチル、2 - ナフチルが好ましい。特にフェニルが好ましい。

本明細書中、「アラルキル」とは、前記「アルキル」に前記「アリール」が1または2以上置換したものを包含し、これらは可能な全ての位置で置換しうる。例えば、ベンジル、フェニルエチル（例えば、2 - フェニルエチル等）、フェニルプロピル（例えば、3 - フェニルプロピル等）、ナフチルメチル（例えば、1 - ナフチルメチル、2 - ナフチルメチル等）、アントリルメチル（例えば、9 - アントリルメチル等）等が挙げられる。好ましくは、ベンジル、フェニルエチルが挙げられる。

本明細書中、「アラルキルオキシ」としては、ベンジルオキシ、フェニルエチルオキシ（例えば、2 - フェニルエチルオキシ等）、フェニルプロピルオキシ（例えば、3 - フェニルプロピルオキシ等）、ナフチルメチルオキシ（例えば、1 - ナフチルメチル、2 - ナフチルメチルオキシ等）、アントリルメチルオキシ（例えば、9 - アントリルメチルオキシ等）等が挙げられる。好ましくは、ベンジルオキシ、フェニルエチルオキシが挙げられる。

本明細書中、「アラルキルオキシカルボニル」としては、ベンジルオキシカルボニル、フェニルエチルオキシカルボニル（例えば、2 - フェニルエチルオキシカルボニル等）、フェニルプロピルオキシカルボニル（例えば、3 - フェニルプロピルオキシカルボニル等）、ナフチルメチルオキシカルボニル（例えば、1 - ナフチルメチル、2 - ナフチルメチルオキシカルボニル等）、アントリルメチルオキシカルボニル（例えば、9 - アントリルメチルオキシカルボニル等）等が挙げられる。好ましくは、ベンジルオキシカルボニル、フェニルエチルオキシカルボニルが挙げられる。

#### 【0020】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「非芳香族複素環基」なる用語は、任意に選ばれる、酸素原子、硫黄原子又は窒素原子を環内に1個以上含む非芳香族の5 ~ 7員環またはそれに別の1以上の「非芳香族複素環基」もしくは「ヘテロアリアル」が縮合した環から誘導される基を包含する。例えば、ピロリジニル（例えば、1 - ピロリジニル、2 - ピロリジニル）、ピロリニル（例えば、3 - ピロリニル）、イミダゾリジニル（例えば、2 - イミダゾリジニル）、イミダゾリニル（例えば、イミダゾリニル）、ピラゾリジニル（例えば、1 - ピラゾリジニル、2 - ピラゾリジニル）、ピラゾリニル（例えば、ピラゾリニル）、ピペリジニル（例えば、ピペリジニル、2 - ピペリジニル）、ピ

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ペラジニル（例えば、1 - ピペラジニル、2 - ピペラジニル）、インドリニル（例えば、1 - インドリニル）、イソインドリニル（例えば、イソインドリニル）、モルホリニル（例えば、モルホリノ、2 - モルホリニル、3 - モルホリニル）、テトラヒドロフラニル、ジヒドロピラニル、テトラヒドロピラニル、ジオキサニル、テトラヒドロチエニル、ジヒドロチオピラニル、テトラヒドロチオフラニル、デカヒドロイソキノリル、アゼピニル、オキセピニル、ジヒドロオキセピニル、テトラヒドロオキセピニル、オキセパニル、4, 5, 6, 7 - テトラヒドロチエノ [ 3, 2 ] ピリジル、2 - オキサ - 5 - アザ - ビシクロ [ 2 . 2 . 1 ] ヘプタ - 5 - イル、ヘキサヒドロピラジル [ 2 . 1 - b ] [ 1, 3 ] オキサジン - 8 - イル等が挙げられる。

R<sup>19</sup>における「非芳香族複素環基」としては、ピロリジニル（例えば、1 - ピロリジニル、2 - ピロリジニル）、ピペリジル（例えば、ピペリジノ、2 - ピペリジル）、ピペラジニル（例えば、1 - ピペラジニル）、モルホリニル（例えば、モルホリノ、3 - モルホリニル）、テトラヒドロフラニルが好ましい。

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「非芳香族含窒素複素環基」なる用語は、窒素原子を少なくとも1つ環内に含み、さらに酸素原子および硫黄原子から任意に選ばれる原子を環内に1個以上含んでもよい非芳香族の4～7員環またはそれらが2個以上縮合した環から誘導される基を包含する。例えば、アゼチジニル（例えば、1 - アゼチジニル、2 - アゼチジニル、3 - アゼチジニル）、ピロリジニル（例えば、1 - ピロリジニル、2 - ピロリジニル、3 - ピロリジニル）、ピロリニル（例えば、3 - ピロリニル）、イミダゾリジニル（例えば、2 - イミダゾリジニル）、イミダゾリニル（例えば、イミダゾリニル）、ピラゾリジニル（例えば、1 - ピラゾリジニル、2 - ピラゾリジニル）、ピラゾリニル（例えば、ピラゾリニル）、ピペリジル（例えば、ピペリジノ、2 - ピペリジル、3 - ピペリジル、4 - ピペリジル）、ピペラジニル（例えば、1 - ピペラジニル、2 - ピペラジニル）、インドリニル（例えば、1 - インドリニル）、イソインドリニル（例えば、イソインドリニル）、モルホリニル（例えば、モルホリノ、2 - モルホリニル、3 - モルホリニル）、1, 4 - チアジニル（例えば、1, 4 - チアジン - 1 - イル、1, 4 - チアジン - 2 - イル、）、チオモルホリニル（例えば、1 - チオモルホリニル、2 - チオモルホリニル）、デカヒドロイソキノリル（例えば2 - デカヒドロイソキノリル）、アゼピニル（例えば1 - アゼピニル）、ジアゼピニル、4, 5, 6, 7 - テトラヒドロチエノ [ 3, 2 ] ピリジル、2 - オキサ - 5 - アザ - ビシクロ [ 2 . 2 . 1 ] ヘプタ - 5 - イル、ヘキサヒドロピラジル [ 2 . 1 - b ] [ 1, 3 ] オキサジン - 8 - イル等が挙げられる。

R<sup>1</sup>およびR<sup>17</sup>における「非芳香族含窒素複素環基」としては、アゼチジニル（例えば、1 - アゼチジニル、2 - アゼチジニル、3 - アゼチジニル）、ピロリジニル（例えば、1 - ピロリジニル、2 - ピロリジニル、3 - ピロリジニル）、ピペリジル（例えば、ピペリジノ、2 - ピペリジル、3 - ピペリジル、4 - ピペリジル）が好ましい。

W<sup>1</sup>における「非芳香族含窒素複素環基」としては、アゼチジニル（例えば、1 - アゼチジニル、2 - アゼチジニル、3 - アゼチジニル）、ピロリジニル（例えば、1 - ピロリジニル、2 - ピロリジニル、3 - ピロリジニル）、ピペリジル（例えば、ピペリジノ、2 - ピペリジル、3 - ピペリジル、4 - ピペリジル）、ピペラジニル（例えば、1 - ピペラジニル）、モルホリニル（例えば、モルホリノ、3 - モルホリニル）が好ましい。

置換基群Aにおける「非芳香族含窒素複素環基」としては、ピロリジニル（例えば、1 - ピロリジニル、2 - ピロリジニル、3 - ピロリジニル）、ピペリジル（例えば、ピペリジノ、2 - ピペリジル、3 - ピペリジル、4 - ピペリジル）、ピペラジニル（例えば、1 - ピペラジニル、2 - ピペラジニル）、モルホリニル（例えば、モルホリノ、2 - モルホリニル、3 - モルホリニル）、チオモルホリニル（例えば、1 - チオモルホリニル、2 - チオモルホリニル）、デカヒドロイソキノリル（例えば2 - デカヒドロイソキノリル）、アゼピニル（例えば1 - アゼピニル）、ジアゼピニル、4, 5, 6, 7 - テトラヒドロチエノ [ 3, 2 ] ピリジル、2 - オキサ - 5 - アザ - ビシクロ [ 2 . 2 . 1 ] ヘプタ - 5 - イル、ヘキサヒドロピラジル [ 2 . 1 - b ] [ 1, 3 ] オキサジン - 8 - イルが好ましい

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## 【 0 0 2 1 】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「ヘテロアリアル」とは、任意に選ばれる、酸素原子、硫黄原子又は窒素原子を環内に1個以上含む5～6員の芳香環基を包含する。これは前記「シクロアルキル」、前記「アリアル」、前記「非芳香族複素環基」、もしくは他のヘテロアリアルと可能な全ての位置で縮合していてもよい。ヘテロアリアルが単環および縮合環のいずれである場合も、すべての可能な位置で結合しうる。例えば、ピロリル（例えば、1 - ピロリル、2 - ピロリル、3 - ピロリル）、フリル（例えば、2 - フリル、3 - フリル）、チエニル（例えば、2 - チエニル、3 - チエニル）、イミダゾリル（例えば、2 - イミダゾリル、4 - イミダゾリル）、ピラゾリル（例えば、1 - ピラゾリル、3 - ピラゾリル）、イソチアゾリル（例えば、3 - イソチアゾリル）、イソオキサゾリル（例えば、3 - イソオキサゾリル）、オキサゾリル（例えば、2 - オキサゾリル）、チアゾリル（例えば、2 - チアゾリル、5 - チアゾリル）、ピリジル（例えば、2 - ピリジル、3 - ピリジル、4 - ピリジル）、ピラジニル（例えば、2 - ピラジニル）、ピリミジニル（例えば、2 - ピリミジニル、4 - ピリミジニル）、ピリダジニル（例えば、3 - ピリダジニル）、トリアゾリル、テトラゾリル（例えば、1 H - テトラゾリル）、オキサジアゾリル（例えば、1, 3, 4 - オキサジアゾリル）、チアジアゾリル（例えば、1, 3, 4 - チアジアゾリル）、インドリジニル（例えば、2 - インドリジニル、6 - インドリジニル）、イソインドリル（例えば、2 - イソインドリル）、インドリル（例えば、1 - インドリル、2 - インドリル、3 - インドリル）、インダゾリル（例えば、3 - インダゾリル）、プリニル（例えば、8 - プリニル）、キノリジニル（例えば、2 - キノリジニル）、イソキノリル（例えば、3 - イソキノリル）、キノリル（例えば、2 - キノリル、5 - キノリル）、フタラジニル（例えば、1 - フタラジニル）、ナフチリジニル（例えば、2 - ナフチリジニル）、キノラニル（例えば、2 - キノラニル）、キナゾリニル（例えば、2 - キナゾリニル）、シンノリニル（例えば、3 - シンノリニル）、プテリジニル（例えば、2 - プテリジニル）、カルバゾリル（例えば、2 - カルバゾリル、4 - カルバゾリル）、フェナントリジニル（例えば、2 - フェナントリジニル、3 - フェナントリジニル）、アクリジニル（例えば、1 - アクリジニル、2 - アクリジニル）、ジベンゾフラニル（例えば、1 - ジベンゾフラニル、2 - ジベンゾフラニル）、ベンゾイミダゾリル（例えば、2 - ベンゾイミダゾリル）、ベンゾイソオキサゾリル（例えば、3 - ベンゾイソオキサゾリル）、ベンゾオキサゾリル（例えば、2 - ベンゾオキサゾリル）、ベンゾオキサジアゾリル（例えば、4 - ベンゾオキサジアゾリル）、ベンゾイソチアゾリル（例えば、3 - ベンゾイソチアゾリル）、ベンゾチアゾリル（例えば、2 - ベンゾチアゾリル）、ベンゾフリル（例えば、3 - ベンゾフリル）、ベンゾチエニル（例えば、2 - ベンゾチエニル）、4, 5 - ジヒドロナフト [ 1, 2 - d ] チアゾリル、4 H - クロメノ [ 4, 3 - d ] チアゾリル、4 H - チオクロメノ [ 4, 3 - d ] チアゾリル、4, 5 - ジヒドロチアゾロ [ 5, 4 - c ] キノリル、8 H - インデノ [ 1, 2 - d ] チアゾリル、5, 6 - ジヒドロ - 4 H - 3 - チア - 1 - アザ - ベンゾ [ e ] アズレニル等が挙げられる。

R<sup>2</sup>における「ヘテロアリアル」としては、ピリジル、フリル、チエニル、チアゾリル、オキサゾリル、チアジアゾリル、オキサジアゾリル、トリアゾリル等が好ましい。さらに好ましくは、ピリジル、フリル、チエニル、チアゾリル等が挙げられる。

R<sup>8</sup>における「ヘテロアリアル」としては、ピリジル、フリル、チエニル、チアゾリル、オキサゾリル、チアジアゾリル、オキサジアゾリル、トリアゾリル等が好ましい。さらに好ましくは、ピリジル、フリル、チエニル、チアゾリル等が挙げられる。

R<sup>19</sup>における「ヘテロアリアル」としては、ピリジル、フリル、チエニル等が好ましい。

W<sup>1</sup>における「ヘテロアリアル」としては、ピロリル、イミダゾリル、ピラゾリル、トリアゾリル、テトラゾリル等が好ましい。

本明細書中、「ヘテロアリアルアルキル」とは、前記「アルキル」の任意の位置に前記「

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「ヘテロアリール」が1または2以上置換したものを包含し、これらは可能な全ての位置で置換しうる。例えば、チエニルメチル（例えば、2-チエニルメチル）、チエニルエチル（例えば、2-(チオフェン-2-イル)エチル）、フリルメチル（例えば、2-フリルメチル）、フリルエチル（例えば2-(フラン-2-イル)エチル）、ピロリルメチル（例えば、2-ピロリルメチル）、ピロリルエチル（例えば、2-(ピロール-2-イル)エチル）、イミダゾリルメチル（例えば、2-イミダゾリルメチル、4-イミダゾリルメチル）、イミダゾリエチル（例えば、2-(イミダゾール-2-イル)エチル）、ピラゾリルメチル（例えば、3-ピラゾリルメチル）、ピラゾリエチル（例えば、2-(ピラゾール-3-イル)エチル）、チアゾリルメチル（例えば、2-チアゾリルメチル）、チアゾリエチル（例えば、2-(チアゾール-2-イル)エチル）、イソチアゾリルメチル（例えば、3-イソチアゾリルメチル）、イソキサゾリルメチル（例えば、3-イソキサゾリルメチル）、オキサゾリルメチル（例えば、2-オキサゾリルメチル）、オキサゾリエチル（例えば、2-(オキサゾール-2-イル)エチル）、ピリジルメチル（例えば、2-ピリジルメチル、3-ピリジルメチル、4-ピリジルメチル）、ピリジリエチル（例えば、2-ピリジリエチル）等が挙げられる。

#### 【0022】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「アシル」なる用語は、アルキル部分が前記「アルキル」であるアルキルカルボニル、ハロアルキル部分が前記「ハロアルキル」であるハロアルキルカルボニル、アルケニル部分が前記「アルケニル」であるアルケニルカルボニル、アラルキル部分が前記「アラルキル」であるアラルキルカルボニル、またはアリアル部分が前記「アリアル」であるアリアルカルボニルを包含する。例えば、アセチル、プロピオニル、ブチロイル、トリフルオロメチルカルボニル、ビニルカルボニル、フェニルアセチル、ベンゾイル等が挙げられる。「アルキル」、「アルケニル」、および「アリアル」は後述のそれぞれの置換基によって置換されていてもよい。

R<sup>10</sup> および R<sup>11</sup> における、「アシル」としては、アセチル、ベンゾイル、フェニルアセチル等が好ましい。

R<sup>12</sup> および R<sup>13</sup> における、「アシル」としては、アセチル、ベンゾイル、フェニルアセチル等が好ましい。

R<sup>9</sup> における、「アシル」としては、アセチル、ベンゾイル、フェニルアセチル等が好ましい。

本明細書中、「アシルオキシ」としては、アセチルオキシ、プロピオニルオキシ、ベンゾイルオキシ等が挙げられる。

#### 【0023】

本明細書中、単独でもしくは他の用語と組み合わせて用いられる「置換されていてもよいアミノ」なる用語は、置換基群Aから選択される置換基で置換されていてもよい前記「アルキル」、前記「ハロアルキル」、前記「アルケニル」、前記「アルキニル」、前記「アルキルスルホニル」、前記「アルキルスルホニルアルキル」、前記「アルキルオキシカルボニル」、前記「アミノカルボニルオキシアルキル」、前記「アルキルオキシ」、置換基群Aから選択される置換基で置換されていてもよい前記「アルキルオキシアルキル」、置換基群Aから選択される置換基で置換されていてもよい前記「シクロアルキル」、置換基群Gから選択される置換基で置換されていてもよい前記「アリアル」、前記「アラルキル」、前記「ヘテロアリアル」、前記「ヘテロアリアルアルキル」、前記「アシル」、置換基群Gから選択される置換基で置換されていてもよい前記「アミノカルボニルアルキル」、または置換基群Gから選択される置換基で置換されていてもよい前記「アミノアルキルカルボニル」、アルケニルオキシ部分が前記「アルケニルオキシ」である「アルケニルオキシカルボニル」、アラルキルオキシ部分が前記「アラルキルオキシ」である「アラルキルオキシカルボニル」で1または2箇所置換されていてもよいアミノを包含する（置換基群G：アルキル、アルケニル、およびアラルキル）。例えば、アミノ、メチルアミノ、ジメチルアミノ、エチルメチルアミノ、ジエチルアミノ、エチルメチルアミノ、ジイソプロピルアミノ、ベンジルアミノ、アセチルアミノ、トリフルオロメチルカルボニル、ベン



ゾイルアミノ、メチルオキシカルボニルアミノ、エチルオキシカルボニルアミノ、*n*-プロピルオキシカルボニルアミノ、イソプロピルオキシカルボニルアミノ、*n*-ブチルオキシ、イソブチルオキシカルボニルアミノ、*sec*-ブチルオキシカルボニルアミノ、*tert*-ブチルオキシカルボニルアミノ、アリルオキシカルボニルアミノ、ベンジルオキシカルボニルアミノ、シクロペンチルアミノ、シクロヘキシルアミノ等が挙げられる。

R<sup>3</sup>およびR<sup>4</sup>における、「置換されていてもよいアミノ」の置換基としては、アルキル、アシル等が好ましい。

R<sup>6</sup>およびR<sup>7</sup>における、「置換されていてもよいアミノ」の置換基としては、アルキル等が好ましい。

置換基群Aにおける、「置換されていてもよいアミノ」および「置換されていてもよいアミノカルボニル」の置換基としては、アルキル、アルケニル、アルキニル、置換基群Iから選択される置換基で置換されていてもよいアリール、アラルキル、アルキルオキシ、ヒドロキシアルキル、ヒドロキシアルキルオキシアルキル、ハロアルキル、1または2のアルキルで置換されていてもよいアミノアルキル、アルキルスルホニル、アルキルスルホニルアルキル、ハロゲンもしくはアルキルオキシで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、置換基群Iから選択される置換基で置換されていてもよいシクロアルキル、カルボキシアルキル、置換基群Gから選択される置換基で置換されていてもよいアミノカルボニルアルキル、置換基群Gから選択される置換基で置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、アルキルチオアルキル、アルキルカルボニルアミノアルキル、アルキルスルホニルアミノアルキル、アルキルスルホニル(アルキル)アミノアルキル、アルキルオキシカルボニルアルキル、アルキルチオ(ヒドロキシ)アルキル、シクロアルキルアルキル、シアノアルキル、置換基群Gから選択される置換基で置換されていてもよいアミノアルキルカルボニル、置換基群Gから選択される置換基で置換されていてもよいヘテロアリール、ヘテロアリールアルキル、ヒドロキシアルキルオキシアルキル、置換基群Iから選択される置換基で置換されていてもよい非芳香族複素環基、置換基群Iから選択される置換基で置換されていてもよい非芳香族複素環アルキル等が好ましい。

本明細書中、「置換されていてもよいアミノカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。特に、アルキルが好ましい。

置換基群Bにおける、「置換されていてもよいアミノカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。

置換基群Dにおける、「置換されていてもよいアミノカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。

置換基群Eにおける、「置換されていてもよいアミノカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。

置換基群Hにおける、「置換されていてもよいアミノカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。

本明細書中、「置換されていてもよいアミノカルボニルアルキル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。特に、アルキルが好ましい。

置換基群Cにおける、「置換されていてもよいアミノカルボニルアルキル」における置換基としては、アルキルアルケニル、アラルキル等が好ましい。

本明細書中、「置換されていてもよいアミノカルボニルオキシアルキル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。特に、アルキルが好ましい。

置換基群Cにおける、「置換されていてもよいアミノカルボニルオキシアルキル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。

本明細書中、「置換されていてもよいアミノアルキルカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。特に、アルキルが好ましい。

置換基群Cにおける、「置換されていてもよいアミノアルキルカルボニル」における置換基としては、アルキル、アルケニル、アラルキル等が好ましい。

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本明細書中、「置換されていてもよいウレイド」なる用語は、前記「アルキル」、前記「シクロアルキル」、前記「アリール」、前記「アラルキル」、前記「ヘテロアリール」、前記「ヘテロアリールアルキル」、前記「アシル」、シアノアルキル、またはアルキルオキシアルキルで1または2箇所以上置換されていてもよいウレイドを包含する。

本明細書中、「置換されていてもよいグアニジノ」なる用語は、前記「アルキル」、前記「シクロアルキル」、前記「アリール」、前記「アラルキル」、前記「ヘテロアリール」、前記「ヘテロアリールアルキル」、前記「アシル」、シアノ、シアノアルキル、またはアルキルオキシアルキルで1または2箇所以上置換されていてもよいグアニジノを包含する。

#### 【0024】

本明細書中、「置換されていてもよいアルキル」における置換基としては、置換されていてもよいアミノ、置換されていてもよい非芳香族含窒素複素環基、置換されていてもよい非芳香族含窒素複素環カルボニル、アルキル、アルキルオキシ、シアノ、アルキルスルホニル、シクロアルキル、アルケニル、ヘテロアリール、ヘテロアリールオキシ、ヒドロキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、カルボキシ、アルキルオキシカルボニル、ヒドロキシアルキル、ハロアルキル、ハロアルキルオキシ、アルキルカルボニルオキシ、カルバモイルオキシ、置換されていてもよいアミノカルボニル、アミノカルボニルアルキルオキシ、アシル、アシルオキシ、アリールオキシ、アラルキル、アラルキルオキシ、置換されていてもよいグアニジノ、アゾ基、置換されていてもよいウレイド等が挙げられる。これらは、全ての可能な位置で1個以上置換しうる。

$R^1$ 、 $R^{1'}$ および $R^{1''}$ における「置換されていてもよいアルキル」の置換基としては、置換基群Aから選択される置換基、アルキルオキシ、シアノ、ハロゲン、ヘテロアリール等が好ましい。特に、置換基群Cから選択される置換基で置換されていてもよいアミノ、置換基群Dから選択される置換基で置換されていてもよい非芳香族含窒素複素環基が好ましい。

$R^2$ における「置換されていてもよいアルキル」の置換基としては、置換基群Aから選択される置換基、アルキルオキシ、アルキルカルボニル、およびハロゲンが好ましい。特に、ヒドロキシ、アルキルオキシ、およびハロゲンが好ましい。

$R^3$ および $R^4$ における、「置換されていてもよいアルキル」の置換基としては、アルキルオキシ、アルキルオシキカルボニル、アミノカルボニル、シアノ等が好ましい。

$R^6$ および $R^7$ における、「置換されていてもよいアルキル」の置換基としては、アルキルオキシ、アルキルオシキカルボニル、アミノカルボニル、シアノ、ヒドロキシ等が好ましい。

$R^{1'}$ における、「置換されていてもよいアルキル」の置換基としては、ヒドロキシ、アルキルオキシ、ハロゲン、シアノ等が好ましい。

$R^{2'}$ における、「置換されていてもよいアルキル」の置換基としては、ヒドロキシ、アルキルオキシ、ハロゲン、シアノ等が好ましい。

#### 【0025】

本明細書中、「置換されていてもよいアルケニル」および「置換されていてもよいアルキニル」における置換基としては、置換されていてもよいアミノ、置換されていてもよい非芳香族含窒素複素環基、シクロアルキル、ヒドロキシ、アルキルオキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、シアノ、カルボキシ、アルキルオキシカルボニル、ハロアルキル、ハロアルキルオキシ、置換されていてもよいアミノカルボニル、アシル、アシルオキシ、アリール、アリールオキシ、アラルキル、アラルキルオキシ、アルキルスルホニル、グアニジノ、アゾ基、置換されていてもよいウレイド等が挙げられる。これらは、全ての可能な位置で1個以上置換しうる。

$R^1$ および $R^{1'}$ における「置換されていてもよいアルケニル」の置換基としては、置換基群Aから選択される置換基が好ましい。特に、置換基群Cから選択される置換基で置換されていてもよいアミノ、置換基群Dから選択される置換基で置換されていてもよい非芳香族含窒素複素環基が好ましい。

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R<sup>2</sup>における、「置換されていてもよいアルケニル」および「置換されていてもよいアルキニル」の置換基としては、置換基群Aから選択される置換基、シクロアルキル、アルキルオキシ、およびハロゲンが好ましい。特に、ヒドロキシ、シクロアルキル、アルキルオキシ、およびハロゲンが好ましい。

R<sup>3</sup>およびR<sup>4</sup>における、「置換されていてもよいアルケニル」および「置換されていてもよいアルキニル」の置換基としては、アルキルオキシ、アルキルオキシカルボニル、アミノカルボニル、シアノ等で置換されていてもよいアルキル、アルキルオキシ、アルキルオキシカルボニル、アミノカルボニル、シアノ等で置換されていてもよいアルキルオキシカルボニル、置換されていてもよいアミノカルボニル等が好ましい。

R<sup>6</sup>およびR<sup>7</sup>における、「置換されていてもよいアルケニル」および「置換されていてもよいアルキニル」の置換基としては、アルキルオキシ、アルキルオキシカルボニル、アミノカルボニル、シアノ等で置換されていてもよいアルキル、アルキルオキシ、アルキルオキシカルボニル、アミノカルボニル、シアノ等で置換されていてもよいアルキルオキシカルボニル、置換されていてもよいアミノカルボニル等が好ましい。

R<sup>1 8</sup>における、「置換されていてもよいアルケニル」および「置換されていてもよいアルキニル」の置換基としては、ヒドロキシ、アルキルオキシ、ハロゲン、シアノ等が好ましい。

R<sup>2 0</sup>における、「置換されていてもよいアルケニル」および「置換されていてもよいアルキニル」の置換基としては、ヒドロキシ、アルキルオキシ、ハロゲン、シアノ等が好ましい。

#### 【0026】

本明細書中、「置換されていてもよいシクロアルキル」における置換基としては、アルキルオキシカルボニル、置換基群Gから選択される置換基で置換されていてもよいアミノカルボニル、シアノ、置換基群M（置換基群M：シアノ、ヒドロキシ、カルボキシ、アミノカルボニル、アルキルオキシカルボニル、およびアルキルオキシ）から選択される置換基で置換されていてもよいアルキル、アルキルスルホニル、アルキルスルホニルアルキルカルボニル、シクロアルキル、ヒドロキシ、アルキルオキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、カルボキシ、ハロアルキル、ハロアルキルオキシ、アシル、アシルオキシ、アリール、アリールオキシ（例えば、フェニルオキシ）、アラルキル、アラルキルオキシ（例えば、ベンジルオキシ）、非芳香族含窒素複素環基、アルキルカルボニルアミノ、アミノカルボニルオキシ、アミノ、オキソ、グアニジノ、アゾ基、置換されていてもよいウレイド等が挙げられる。

置換基群Cにおける、「置換されていてもよいシクロアルキル」の置換基としては、置換基群Iで挙げられる置換基が好ましい。

本明細書中、「置換されていてもよいアルキルオキシ」、「置換されていてもよいアルケニルオキシ」、および「置換されていてもよいアルキニルオキシ」における置換基としては、シクロアルキル、アルケニル、ヒドロキシ、アルキルオキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、シアノ、カルボキシ、アルキルオキシカルボニル、ハロアルキル、ハロアルキルオキシ、置換されていてもよいアミノ、置換されていてもよいアミノカルボニル、アシル、アシルオキシ、置換されていてもよい非芳香族複素環基、アリールオキシ、アラルキルオキシ、アルキルスルホニル、グアニジノ、アゾ基、置換されていてもよいウレイド等が挙げられる。これらは、全ての可能な位置で1個以上置換しうる。好ましくは、ハロゲン等が挙げられる。

R<sup>3</sup>およびR<sup>4</sup>における、「置換されていてもよいアルキルオキシ」および「置換されていてもよいアルケニルオキシ」の置換基としては、アルキルオキシ、置換されていてもよいアミノ、置換基群Dから選択される置換基で置換されていてもよい非芳香族複素環基、シアノ等が好ましい。

R<sup>5</sup>における、「置換されていてもよいアルキルオキシ」、「置換されていてもよいアルケニルオキシ」、および「置換されていてもよいアルキニルオキシ」の置換基としては、置換基群Eから選択される置換基が好ましい。

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R<sup>6</sup> および R<sup>7</sup> における、「置換されていてもよいアルキルオキシ」、「置換されていてもよいアルケニルオキシ」、および「置換されていてもよいアルキニルオキシ」の置換基としては、アルキルオキシ、シアノ、アルキルオキシカルボニル、置換されていてもよいアミノカルボニル等が好ましい。

【0027】

本明細書中、「置換されていてもよい非芳香族複素環基」における置換基としては、アルキルオキシカルボニル、置換基群 G から選択される置換基で置換されていてもよいアミノカルボニル、シアノ、置換基群 M (置換基群 M : シアノ、ヒドロキシ、カルボキシ、アミノカルボニル、アルキルオキシカルボニル、およびアルキルオキシ) から選択される置換基で置換されていてもよいアルキル、アルキルスルホニル、アルキルスルホニルアルキルカルボニル、シクロアルキル、ヒドロキシ、アルキルオキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、カルボキシ、ハロアルキル、ハロアルキルオキシ、アシル、アシルオキシ、アリール、アリールオキシ (例えば、フェニルオキシ)、アラルキル、アラルキルオキシ (例えば、ベンジルオキシ)、非芳香族含窒素複素環基、アルキルカルボニルアミノ、アミノカルボニルオキシ、アミノ、オキソ、グアニジノ、アゾ基、置換されていてもよいウレイド等が挙げられる。

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本明細書中、「置換されていてもよい非芳香族含窒素複素環基」における置換基としては、アルキルオキシカルボニル、置換基群 G から選択される置換基で置換されていてもよいアミノカルボニル、シアノ、置換基群 M (置換基群 M : シアノ、ヒドロキシ、カルボキシ、アミノカルボニル、アルキルオキシカルボニル、およびアルキルオキシ) から選択される置換基で置換されていてもよいアルキル、アルキルスルホニル、アルキルスルホニルアルキルカルボニル、シクロアルキル、ヒドロキシ、アルキルオキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、カルボキシ、ハロアルキル、ハロアルキルオキシ、アシル、アシルオキシ、アリール、アリールオキシ (例えば、フェニルオキシ)、アラルキル、アラルキルオキシ (例えば、ベンジルオキシ)、非芳香族含窒素複素環基、アルキルカルボニルアミノ、アミノカルボニルオキシ、置換されていてもよいアミノ、オキソ、グアニジノ、アゾ基、置換されていてもよいウレイド等が挙げられる。

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R<sup>1</sup> および R<sup>17</sup> における「置換されていてもよい非芳香族含窒素複素環基」の置換基としては、アルキル、アルキルオキシカルボニル、置換基群 G から選択される置換基で置換されていてもよいアミノカルボニル、シアノ、オキソ、アミノ、カルボキシル、ヒドロキシアルキル、シアノアルキル、アルキルオキシアルキル、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル、アルキルカルボニルアミノ、アルキルスルホニルアミノ、カルボキシアルキル、アルキルオキシカルボニルアルキル等が好ましい。特に、置換基群 B で挙げられる置換基が好ましい。

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W<sup>1</sup> における「置換されていてもよい非芳香族含窒素複素環基」の置換基としては、アルキル、アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、ヒドロキシアルキル、シアノアルキル、アルキルオキシアルキル、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル等が好ましい。特に、置換基群 J で挙げられる置換基が好ましい。

置換基群 A における「置換されていてもよい非芳香族含窒素複素環基」および「置換されていてもよい非芳香族含窒素複素環カルボニル」の置換基としては、置換基群 D から選択される置換基が好ましい。

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【0028】

本明細書中、「置換されていてもよいアリール」、「置換されていてもよいフェニル」、「置換されていてもよいヘテロアリール」、「置換されていてもよいピリジル」、「置換されていてもよいフリル」、「置換されていてもよいチエニル」、「置換されていてもよいチアゾリル」、「置換されていてもよいオキサゾリル」、「置換されていてもよい非芳香族複素環基」、「置換されていてもよい非芳香族複素環アルキル」、「置換されていてもよいシクロアルキル」、「置換されていてもよいアラルキル」、および「置換されていてもよいヘテロアリールアルキル」における置換基としては、置換されていてもよいア

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ルキル（置換基としては、ハロゲン、ヒドロキシ、ニトロ、シアノ、カルボキシ、アルキルオキシ、アルキルオキシカルボニル、アシル等）、シクロアルキル、アルケニル、アルキニル、ヒドロキシ、アルキルオキシ、アラルキルオキシ、メルカプト、アルキルチオ、ハロゲン、ニトロ、シアノ、カルボキシ、アルキルオキシカルボニル、アリールオキシカルボニル、ハロアルキル、ハロアルキルオキシ、置換されていてもよいアミノ、置換されていてもよいアミノカルボニル、アシル、アシルオキシ、置換されていてもよいアリール（置換基としては、ハロゲン、ヒドロキシ、ニトロ、シアノ、カルボキシ、アルキルオキシ、アルキルオキシカルボニル、アシル等）、置換されていてもよいヘテロアリール（置換基としては、ハロゲン、ヒドロキシ、ニトロ、シアノ、カルボキシ、アルキルオキシ、アルキルオキシカルボニル、アシル等）、置換されていてもよい非芳香族複素環基（置換基としては、ハロゲン、ヒドロキシ、ニトロ、シアノ、カルボキシ、アルキルオキシ、アルキルオキシカルボニル、アシル等）、置換されていてもよいアラルキル（置換基としては、ハロゲン、ヒドロキシ、ニトロ、シアノ、カルボキシ、アルキルオキシ、アルキルオキシカルボニル、アシル等）、アルキルスルホニル、グアニジノ、アゾ基、 $-N=N-$ （置換されていてもよいフェニル）、または置換されていてもよいウレイド等が挙げられる。これらは、全ての可能な位置で1個以上置換しうる。

$R^1$ における「置換されていてもよいアリール」の置換基としては、置換基群Iで挙げられる置換基が好ましい。

$R^2$ における「置換されていてもよいアリール」および「置換されていてもよいヘテロアリール」の置換基としては、ヒドロキシまたは置換されていてもよいアミノで置換されていてもよいアルキル、置換されていてもよいアミノ、ハロゲン等が好ましい。特に、ヒドロキシまたは置換されていてもよいアミノで置換されていてもよいアルキルが好ましい。

$R^8$ における「置換されていてもよいアリール」、「置換されていてもよいヘテロアリール」、および「置換されていてもよい非芳香族複素環基」の置換基としては、アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、ハロゲン、アルキル、アルキルオキシ、アルキルチオ、ハロアルキル等が好ましい。特に、置換基群Fで挙げられる置換基が好ましい。

$R^{19}$ における「置換されていてもよいアリール」および「置換されていてもよいヘテロアリール」の置換基としては、アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、シアノ、シアノアルキル、ヒドロキシアルキル、およびアミノカルボニルアルキル等が好ましい。特に、置換基群Iで挙げられる置換基が好ましい。

$W^1$ における「置換されていてもよいヘテロアリール」の置換基としては、アルキル、ヒドロキシ、アルキルオキシ等が好ましい。特に、置換基群Jで挙げられる置換基が好ましい。

置換基群Cにおける「置換されていてもよいアリール」、「置換されていてもよいヘテロアリール」、「置換されていてもよい非芳香族複素環基」、および「置換されていてもよい非芳香族複素環アルキル」の置換基としては、置換基群Iで挙げられる置換基が好ましい。

#### 【0029】

本明細書中、「抗癌剤」および「癌の治療剤」とは、脳腫瘍（例えば神経膠芽細胞種）、泌尿器癌（例えば膀胱癌、腎臓癌）、生殖器癌（例えば前立腺癌、卵巣癌、子宮癌）、リンパ系腫瘍、消化器癌（例えば胃癌、大腸癌）、咽喉癌、肺癌（例えば肺腺癌、小細胞肺癌、非小細胞肺癌）、膵臓癌、乳癌、頭頸部癌、食道癌、甲状腺癌の治療剤を包含する。特に、乳癌、脳腫瘍、膀胱癌、腎臓癌、前立腺癌、卵巣癌、子宮癌、肺癌、膵臓癌、大腸癌、および頭頸部癌の治療剤として好ましく使用される。

本発明は、癌を治療又は予防する必要がある哺乳動物において癌を治療又は予防する方法を包含し、当該方法は、前記哺乳動物に治療有効量の式(I)の化合物を投与することからなる。治療するのに好ましい癌は、脳腫瘍（例えば神経膠芽細胞種）、泌尿器癌（例えば膀胱癌、腎臓癌）、生殖器癌（例えば前立腺癌、卵巣癌、子宮癌）、リンパ系腫瘍、消

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化器癌（例えば胃癌）、咽喉癌、肺癌（例えば肺腺癌、小細胞肺癌、非小細胞肺癌）、膵臓癌、乳癌、大腸癌、頭頸部癌、食道癌、甲状腺癌から選択される。より好ましくは、乳癌、脳腫瘍、膀胱癌、腎臓癌、前立腺癌、卵巣癌、子宮癌、肺癌、膵臓癌、大腸癌、および頭頸部癌である。

【発明の効果】

【0030】

本発明化合物は、優れたEGF受容体およびHER2の阻害作用を有し、安全性も高い（CYP阻害作用が軽減された）ことから、医薬品、とりわけ癌の治療剤として有用である。

【発明を実施するための最良の形態】

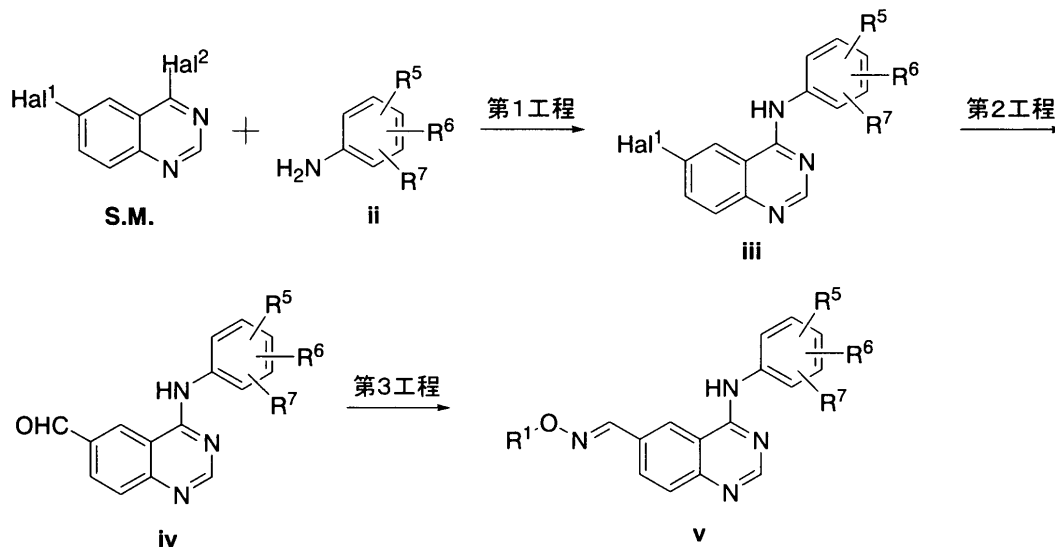
【0031】

本発明化合物（I）は、以下に記載のA法～E法のいずれかにより合成することができる。また、キナゾリン誘導体の合成法が、WO96/09294号、WO98/02434等に記載されているので、これらに従って合成することもできる。化合物が各反応に影響を与える基を有する場合は、適切な保護基で保護し、適切な段階で脱保護することができる。また、各工程の原料、試薬、中間体、および反応生成物が塩を形成し得る場合は、その塩も包含する。

【0032】

（A法）

【化25】



（式中、Hal<sup>1</sup> および Hal<sup>2</sup> はそれぞれ独立してハロゲン；R<sup>1</sup>、R<sup>5</sup>、R<sup>6</sup>、および R<sup>7</sup> は前記（1）と同意義）

（第1工程）

WO96/09294号等に記載の方法に従って合成できるまたは市販品として入手可能な出発原料（S.M.）とWO98/02434等に記載の方法に従って合成できるまたは市販品として入手できる化合物（ii）を反応させることにより、化合物（iii）を合成する工程である。

出発原料（S.M.）と化合物（ii）を、テトラヒドロフラン、アセトニトリル等の溶媒中、20～加熱還流下、好ましくは60～加熱還流下で反応させることにより化合物（iii）を得ることができる。

（第2工程）

化合物（iii）をパラジウム触媒の存在下で一酸化炭素を挿入することにより、化合物（iv）を得る工程である。

化合物（iii）を、N,N-ジメチルホルムアミド、N,N-ジメチルアセタミド、N,N'-ジメチルイミダゾリジノン、ジメチルスルホキシド等の溶媒中、ギ酸ナトリウ

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ム、水素化トリブチルスズ等の水素源、ジクロロビス(トリフェニルホスフィン)パラジウム、テトラキス(トリフェニルホスフィン)パラジウム、トリス(ジベンジリデンアセトン)ジパラジウム、[1,1'-ビス(ジフェニルホスフィノ)フェロセン]ジクロロパラジウム、酢酸パラジウム+トリフェニルホスフィン等のパラジウム触媒、およびトリエチルアミン、ジ(イソプロピル)エチルアミン等の塩基の存在下、一酸化炭素雰囲気下で、50 ~ 150 °C、好ましくは70 ~ 90 °Cで反応させることにより化合物(v)を得ることができる。

(第3工程)

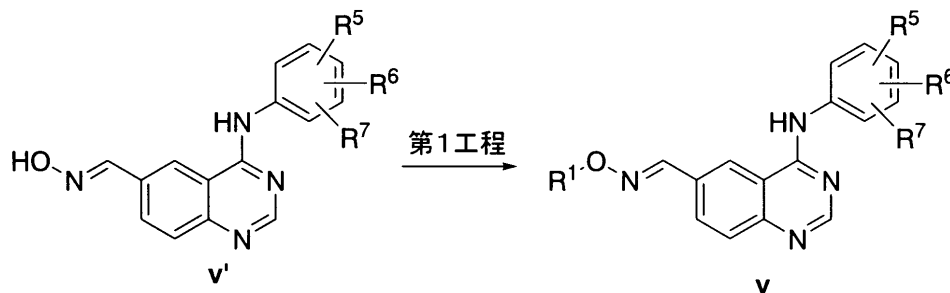
化合物(v)と式： $R^1-O-NH_2$  (式中、 $R^1$ は上記(1)と同意義)で表される化合物を反応させることにより、目的とする化合物(v)を合成する工程である。

出発原料(v)と式： $R^1-O-NH_2$  (式中、 $R^1$ は上記(1)と同意義)で表される化合物またはその塩を、テトラヒドロフラン、ジオキサン、メタノール等の溶媒中、またはそれらと水の混合溶媒中、要すれば塩酸、硫酸、p-トルエンスルホン酸、メタンスルホン酸、ギ酸、酢酸、トリフルオロ酢酸等の酸の存在下に、0 ~ 加熱還流下、好ましくは20 ~ 70 °Cで反応させることにより化合物(v)を得ることができる。

【0033】

(B法)

【化26】



( $R^1$ 、 $R^5$ 、 $R^6$ 、および $R^7$ は前記(1)と同意義)

(第1工程)

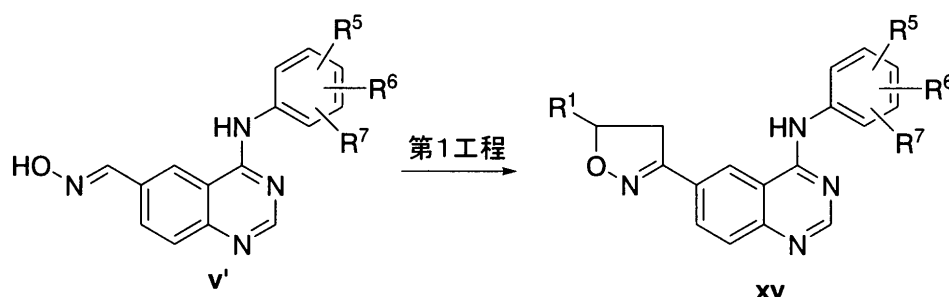
A法で得られた化合物(v)にヒドロキシルアミンを反応させて得られた化合物(v')と式： $R^1-Hal^3$  (式中、 $Hal^3$ はハロゲン； $R^1$ は前記(1)と同意義)で表される化合物を反応させることにより、目的とする化合物(v)を合成する工程である。

化合物(v')と式： $R^1-Hal^3$  (式中、 $Hal^3$ はハロゲン； $R^1$ は前記(1)と同意義)で表される化合物をN,N-ジメチルアセトアミド、N,N-ジメチルアセタミド、N,N'-ジメチルイミダゾリジノン、ジメチルスルホキシド等の溶媒に溶解し、炭酸カリウム、炭酸ナトリウム等の塩基の存在下、50 ~ 150 °C、好ましくは60 ~ 80 °Cで反応させることにより、化合物(v)を得ることができる。

【0034】

(C法)

【化27】



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( $R^1$ 、 $R^5$ 、 $R^6$ 、および $R^7$ は前記(1)と同意義)

(第1工程)

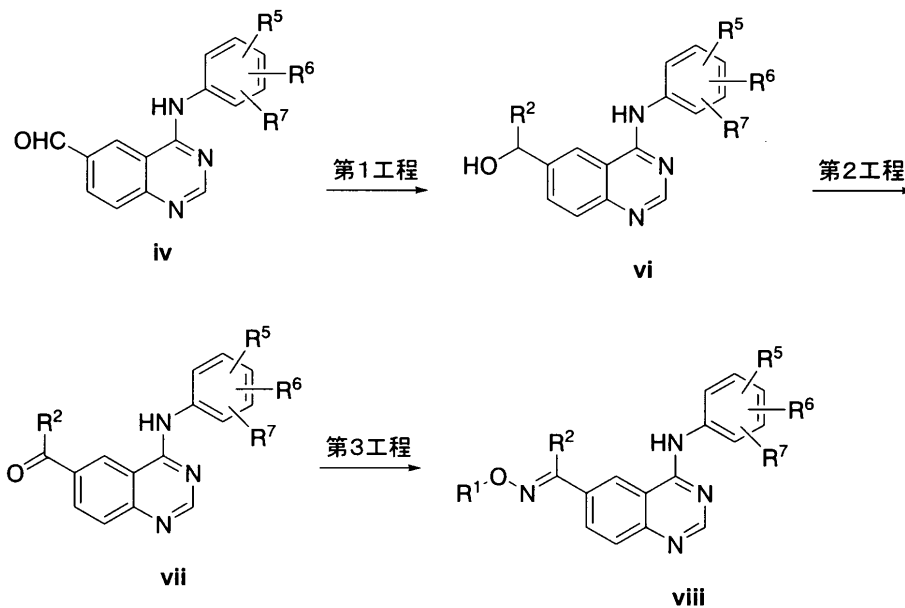
A法で得られた化合物(iv)にヒドロキシルアミンを反応させて得られた化合物(v')と式： $R^1 - CH = CH_2$ (式中、 $R^1$ は前記(1)と同意義)で表される化合物を反応させることにより、目的とする化合物(xv)を合成する工程である。

化合物(v')をクロロホルム、ジクロロメタン、1,2-ジクロロエタン等の溶媒に溶解し、ピリジン、4-ジメチルアミノピリジン等の触媒量の塩基存在下、N-クロロスクシンイミド、N-クロロフタルイミド等の塩素系酸化剤と20 ~ 100、好ましくは50 ~ 60で反応させた後、続けて、式： $R^1 - CH = CH_2$ (式中、 $R^1$ は前記(1)と同意義)で表される化合物、トリエチルアミン、ジ(イソプロピル)エチルアミン等の塩基を加えて、0 ~ 100、好ましくは10 ~ 30で反応させることにより、化合物(xv)を得ることができる。

【0035】

(D法)

【化28】



( $R^1$ 、 $R^2$ 、 $R^5$ 、 $R^6$ 、および $R^7$ は前記(1)と同意義)

(第1工程)

A法で得られた化合物(iv)に式： $R^2 MgHal^4$ または $R^2 Li$ (式中 $Hal^4$ はハロゲン； $R^2$ は前記(1)と同意義)で表される試薬を反応させて、化合物(vi)を得る工程である。

化合物(iv)をテトラヒドロフラン、ジエチルエーテル、トルエン、n-ヘキサン等の溶媒中、-100 ~ 50、好ましくは-78 ~ 10で、式： $R^2 MgHal^4$ または $R^2 Li$ (式中 $Hal^4$ はハロゲン； $R^2$ は前記(1)と同意義)で表される試薬と反応させることにより、化合物(vi)を得ることができる。

(第2工程)

化合物(vi)を酸化して、化合物(vii)を合成する工程である。

化合物(vi)を実験化学講座23巻、第4版(日本化学会編)等に記載の一般的な2級アルコールの酸化方法で酸化することにより、化合物(vii)を得ることができる。

(第3工程)

A法の第3工程と同様の方法により、化合物(vii)から化合物(viii)を得ることができる。

【0036】

(E法)

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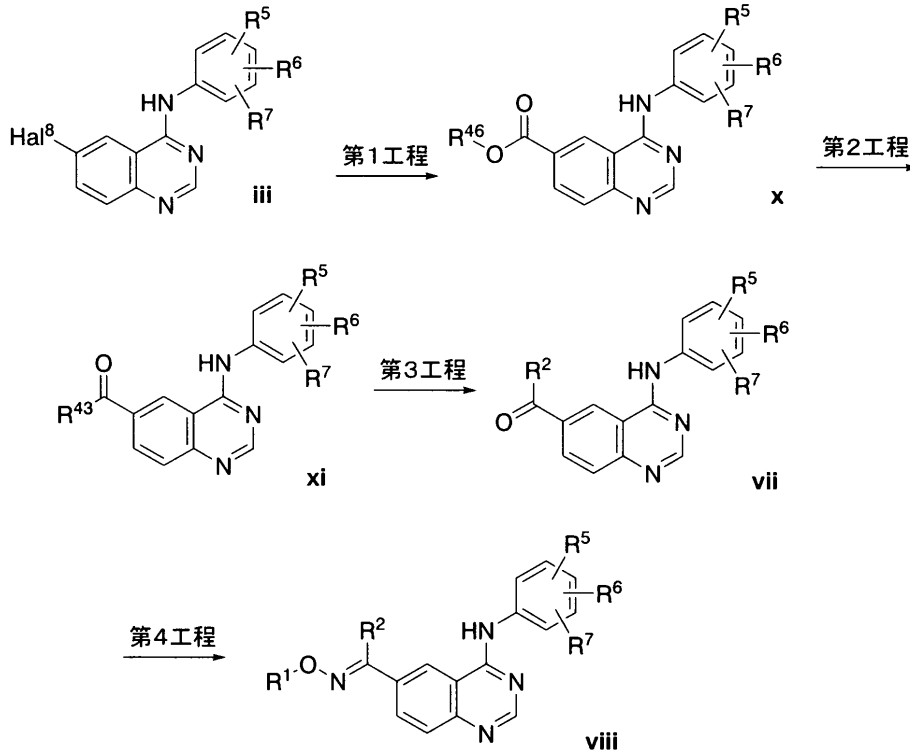
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## 【化29】

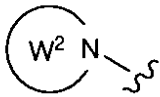


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【Hal<sup>8</sup>はハロゲン；R<sup>43</sup>は式：R<sup>44</sup>(R<sup>45</sup>O)N(式中R<sup>44</sup>、R<sup>45</sup>は互いに独立してC1～C3アルキル)で表される基または式：

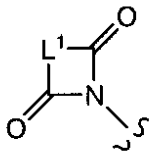
## 【化30】



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(式中W<sup>2</sup>は非芳香族含窒素複素環基)で表される基；R<sup>46</sup>はC1～C3アルキルまたは式：

## 【化31】

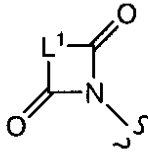


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(式中L<sup>1</sup>はC2～C3アルキレン)で表される基；  
R<sup>1</sup>、R<sup>2</sup>、R<sup>5</sup>、R<sup>6</sup>、およびR<sup>7</sup>は前記(1)と同意義]  
(第1工程)

A法で得られた化合物(iii)を、パラジウム触媒の存在下で一酸化炭素を挿入しながらアルコールまたは式：

## 【化32】



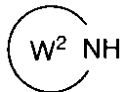
(式中  $L^1$  は  $C_2 \sim C_3$  アルキレン) で表される試薬と反応させることにより、化合物 (x) を得る工程である。

化合物 (iii) を、 $N, N$ -ジメチルホルムアミド、 $N, N$ -ジメチルアセタミド、 $N, N'$ -ジメチルイミダゾリジノン、ジメチルスルホキシド等の溶媒中、ジクロロビス(トリフェニルホスフィン)パラジウム、テトラキス(トリフェニルホスフィン)パラジウム、トリス(ジベンジリデンアセトン)ジパラジウム、[1,1'-ビス(ジフェニルホスフィノ)フェロセン]ジクロロパラジウム、酢酸パラジウム+トリフェニルホスフィン、酢酸パラジウム+1,3-ビスジフェニルホスフィノプロパン等のパラジウム触媒、およびトリエチルアミン、ジ(イソプロピル)エチルアミン等の塩基の存在下、メタノール、エタノール、 $n$ -プロパノール、イソプロパノール等のアルコール、または $N$ -ヒドロキシコハク酸イミド等の試薬と一酸化炭素雰囲気下で、 $50 \sim 150$ 、好ましくは $70 \sim 90$  で反応させることにより化合物 (x) を得ることができる。

(第2工程)

化合物 (x) を式： $R^{44} (R^{45} O) NH$  (式中  $R^{44}$ 、 $R^{45}$  は互いに独立して  $C_1 \sim C_3$  アルキル) で表される試薬または式：

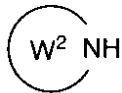
## 【化33】



(式中  $W^2$  は非芳香族含窒素複素環基) で表される試薬と反応させることにより、化合物 (xi) を得る工程である。

化合物 (x) と式： $R^{44} (R^{45} O) NH$  (式中  $R^{44}$ 、 $R^{45}$  は互いに独立して  $C_1 \sim C_3$  アルキル) で表される試薬または式：

## 【化34】



(式中  $W^2$  は非芳香族含窒素複素環基) で表される試薬をテトラヒドロフラン、 $N, N$ -ジメチルホルムアミド、クロロホルム、ジクロロメタン等の溶媒中で混合し、 $-100 \sim 50$ 、好ましくは $-78 \sim 20$  で塩化メチルマグネシウム、臭化メチルマグネシウム、塩化イソプロピルマグネシウム、メチルリチウム等の試薬を加え反応させることにより化合物 (xi) を得ることができる。

(第3工程)

化合物 (xi) に式： $R^2 M$  [式中  $M$  は  $Li$ 、 $Na$ 、 $K$ 、または式： $MgHal^5$  (式中  $Hal^5$  はハロゲン) で表される基； $R^2$  は前記(1)と同意義] で表される試薬を反応させて、化合物 (vii) を得る工程である。

化合物 (xi) をテトラヒドロフラン、ジエチルエーテル、トルエン、 $n$ -ヘキサン等の

溶媒中、 $-100 \sim 50$ 、好ましくは $-78 \sim 20$ で、式： $R^2 M$  [式中MはLi、Na、K、または式： $MgHal^5$  (式中 $Hal^5$ はハロゲン)で表される基； $R^2$ は前記(1)と同意義]で表される試薬と反応させることにより、化合物(vii)を得ることができる。

(第4工程)

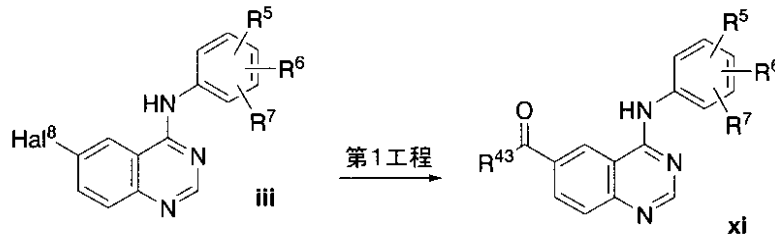
A法の第3工程と同様の方法により、化合物(vii)から化合物(viii)を得ることができる。

【0037】

E法の第1工程、第2工程の別の方法として、以下に示すE'法が挙げられる。

(E'法)

【化35】



( $Hal^8$  および  $R^{43}$  は前記と同意義； $R^5$ 、 $R^6$ 、および  $R^7$  は前記(1)と同意義)

(第1工程)

化合物(iii)を、パラジウム触媒の存在下で一酸化炭素を挿入しながらと式： $R^{43}H$  ( $R^{43}$ は前記と同意義)で表される試薬と反応させることにより、化合物(xi)を得る工程である。

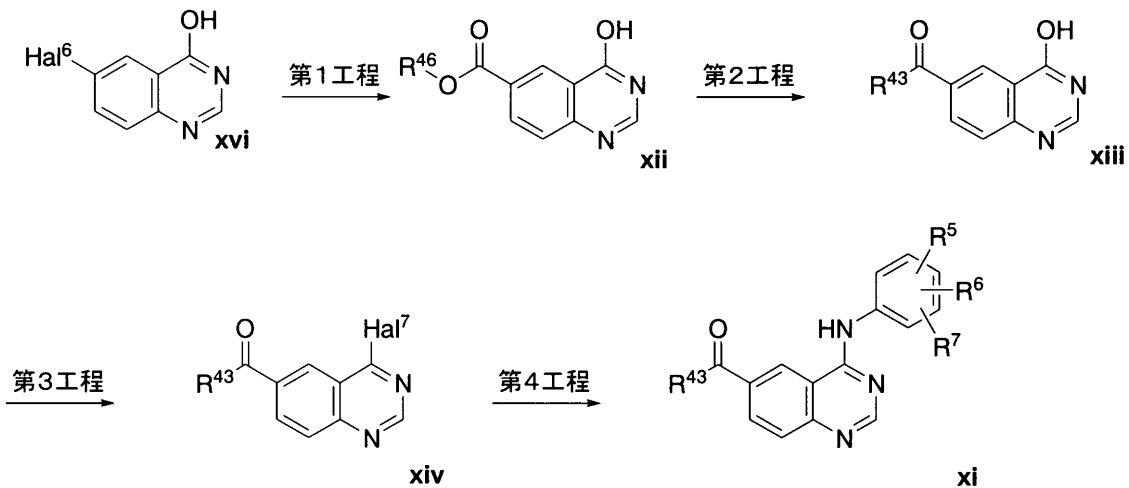
E法の第1工程と同様の方法でアルコール等の代わりに $R^{43}H$  ( $R^{43}$ は前記と同意義)で表される試薬を反応させることにより、化合物(iii)から化合物(xi)を得ることができる。

【0038】

E法のアミド誘導体(xi)を得る別の方法として以下に示すE''法が挙げられる。

(E''法)

【化36】



( $Hal^6$  はハロゲン； $Hal^7$  はハロゲン； $R^{43}$  および  $R^{46}$  は前記と同意義； $R^5$ 、 $R^6$ 、および  $R^7$  は前記(1)と同意義)

(第1工程)

E法の第1工程と同様の方法により、化合物(xvi)から化合物(xi)を得ること

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ができる。

【0039】

(第2工程)

E法の第2工程と同様の方法により、化合物(xiii)から化合物(xiiii)を得ることができる。

【0040】

(第3工程)

化合物(xiiii)の水酸基をハロゲンに変換して、化合物(xiv)を得る工程である。

化合物(xiiii)をトルエン、テトラヒドロフラン等の溶媒中、トリエチルアミン、ジイソプロピルエチルアミン、ピリジン等の塩基の存在下に、オキシ塩化リン、オキシ臭化リン等のハロゲン化剤と0 ~ 200 °C、好ましくは20 ~ 150 °Cで反応させることにより、化合物(xiv)を得ることができる。

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【0041】

(第4工程)

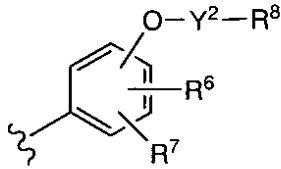
A法の第1工程と同様の方法により、化合物(xiv)から化合物(xi)を得ることができる。

【0042】

Aが式：

【化37】

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(式中Y<sup>2</sup>は単結合またはC<sub>1</sub>~C<sub>3</sub>アルキレン；R<sup>6</sup>、R<sup>7</sup>、およびR<sup>8</sup>は前記(1)と同意義)

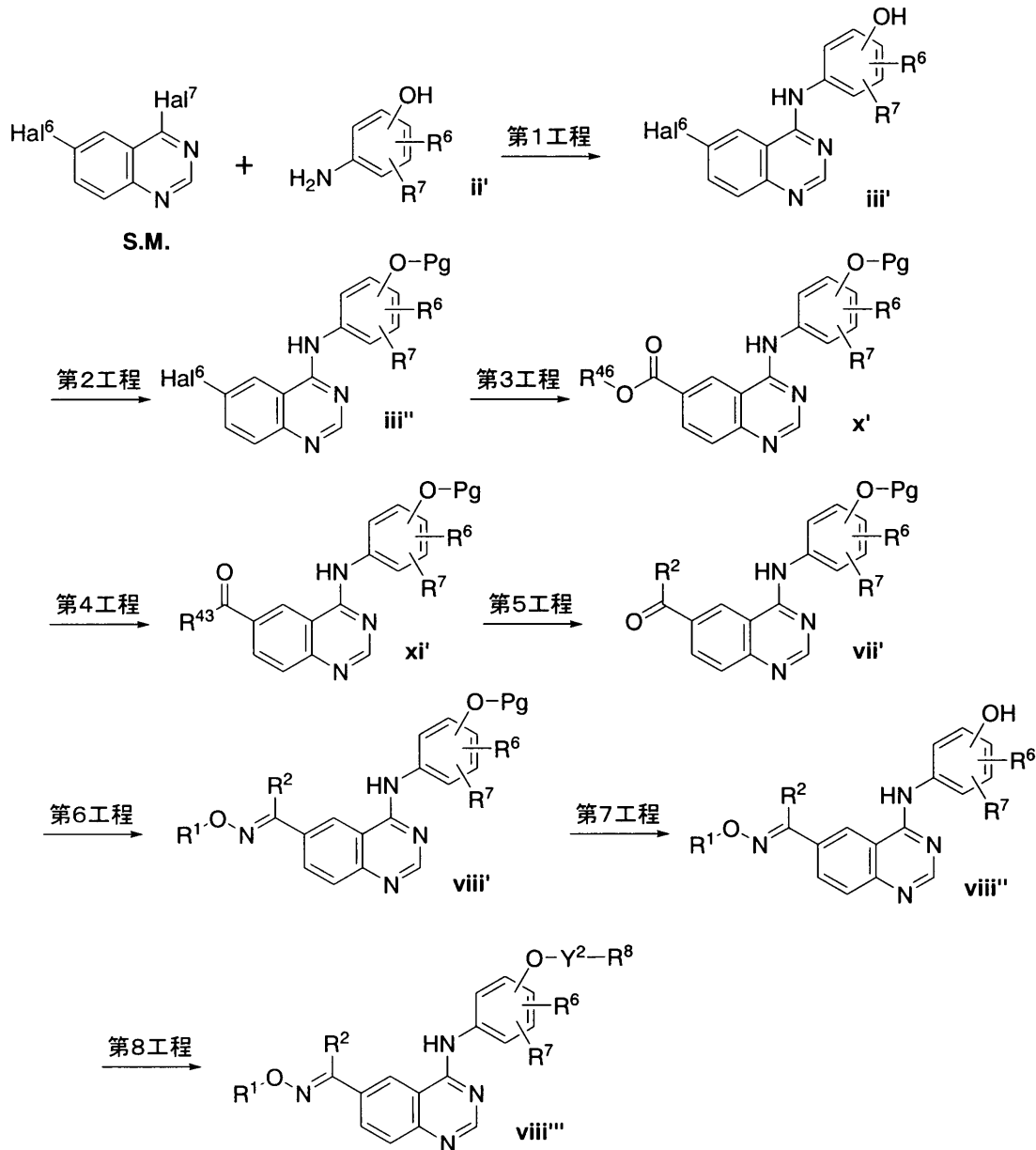
で表される基である本発明化合物(I)は下記F法によっても合成することができる。

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【0043】

(F法)

## 【化38】



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(Hal<sup>6</sup>およびHal<sup>7</sup>はハロゲン; Y<sup>2</sup>は単結合またはC1~C3アルキレン; Pgは水酸基の適切な保護基; R<sup>1</sup>、R<sup>2</sup>、R<sup>6</sup>、R<sup>7</sup>、およびR<sup>8</sup>は前記(1)と同意義; R<sup>43</sup>およびR<sup>46</sup>は前記と同意義)

(第1工程)

A法の第1工程と同様の方法により、出発原料(S.M.)と化合物(ii')から化合物(iii')を得ることができる。

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(第2工程)

化合物(iii')の水酸基に保護基を結合させて、化合物(iii'')を合成する工程である。

Protective Groups in Organic Synthesis, Theodora W Green (John Wiley & Sons)等に記載の方法で、適切な保護基を結合させればよい。

(第3工程)

E法の第1工程と同様の方法により、化合物(iii'')から化合物(x')を得ることができる。

(第4工程)

E法の第2工程と同様の方法により、化合物(x')から化合物(xi')を得ることが

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できる。

(第5工程)

E法の第3工程と同様の方法により、化合物(x i ' )から化合物(v i i ' )を得ることができる。

(第6工程)

A法の第3工程と同様の方法により、化合物(v i i ' )から化合物(v i i i ' )を得ることができる。

(第7工程)

化合物(v i i i ' )の保護基の脱保護を行い、化合物(v i i i ' ' )を合成する工程である。

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Protective Groups in Organic Synthesis, Theodora W Green (John Wiley & Sons)等に記載の方法で、適切な条件により脱保護を行えばよい。

(第8工程)

化合物(v i i i ' ' )に式： $R^8 - Y^2 - Hal^8$  (式中 $Hal^8$ はハロゲン； $Y^2$ は単結合またはC1~C3アルキレン； $R^8$ は前記(1)と同意義)で表される試薬を反応させて、化合物(v i i i ' ' ' )を得る工程である。

化合物(v i i i ' ' )をN、N-ジメチルホルムアミド、ジメチルスルホキシド、テトラヒドロフラン、ジエチルエーテル、トルエン、n-ヘキサン等の溶媒中、炭酸セシウム、炭酸カリウム、炭酸ナトリウム、炭酸リチウム、炭酸水素カリウム、炭酸水素ナトリウム、水酸化カリウム、水酸化ナトリウム、トリエチルアミン、ピリジン等の塩基の存在下

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-200~200、好ましくは0~100で $R^8 - Y^2 - Hal^8$  (式中 $Hal^8$ はハロゲン； $Y^2$ は単結合またはC1~C3アルキレン； $R^8$ は前記(1)と同意義)で表される試薬と反応させることにより、化合物(v i i i ' ' ' )を得ることができる。

F法における $Y^2$ としてはC1~C3アルキレンが好ましい。特にメチレンが好ましい。

F法における $R^8$ としてはハロゲンで置換されていてもよいフェニルまたはハロゲンで置換されていてもよいピリジルが好ましい。

【0044】

A法の第3工程で使用する式： $R^1 - O - NH_2$  (式中、 $R^1$ は上記(1)と同意義)で表されるアルコキシアミン試薬は、市販品として入手可能であるかJ. Med. Chem., 1999, 42 : 2007-2020、Liebigs Ann. Chem., 1988 : 1149-1153、Organic Letters, 2004, 6

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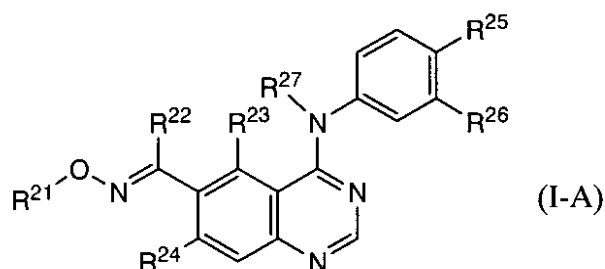
: 4069-4072、Organic Letters, 2005, 7 : 937-939、WO 0 4 / 0 5 4 5 1 4号等に記載の方法に従って合成できるアルコール体を出発原料としてWO 0 2 / 0 6 2 1 3号等に記載の方法に従って合成できる。

【0045】

式(I)で表される化合物において、以下に示す式(I-A)~式(I-P)で示す化合物が好ましい。

式(I-A) :

【化39】



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[式中、 $R^{21}$ は、置換基群C (置換基群C : アルキル、アルケニル、アルキニル、置換されていてもよいアリール、アラールキル、アルキルオキシ、ヒドロキシアルキル、ヒドロキシアルキルオキシアルキル、ハロアルキル、1または2のアルキルで置換されていても

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よいアミノアルキル、アルキルスルホニル、アルキルスルホニルアルキル、ハロゲンもしくはアルキルオキシで置換されていてもよいアルキルカルボニル、アルキルオキシカルボニル、置換されていてもよいシクロアルキル、カルボキシアルキル、置換されていてもよいアミノカルボニルアルキル、置換されていてもよいアミノカルボニルオキシアルキル、アルキルオキシアルキル、アルキルチオアルキル、アルキルカルボニルアミノアルキル、アルキルスルホニルアミノアルキル、アルキルスルホニル(アルキル)アミノアルキル、アルキルオキシカルボニルアルキル、アルキルチオ(ヒドロキシ)アルキル、シクロアルキルアルキル、シアノアルキル、置換されていてもよいアミノアルキルカルボニル、置換されていてもよいヘテロアリール、ヘテロアリールアルキル、ヒドロキシアルキルオキシアルキル、置換されていてもよい非芳香族複素環基、置換されていてもよい非芳香族複素環アルキル)から選択される置換基で置換されていてもよいアミノで置換されたアルキル、置換基群D(置換基群D:ハロゲン、アリール、ヒドロキシ、オキソ、置換されていてもよいアミノカルボニル、アルキルオキシカルボニル、置換基群H(置換基群H:置換されていてもよいアミノカルボニル、シアノ、アルキルオキシ、アルキルスルホニルアミノ、アミノ、カルボキシ、アルキルオキシカルボニル、およびヒドロキシ)から選択される置換基で置換されていてもよいアルキル、アルキルアミノカルボニル、カルボキシ、シアノ、アルキルスルホニル、アルキルカルボニル、アルケニルカルボニル、アルキルスルホニルアルキルカルボニル、アルキルオキシアルキルカルボニル、アルキルカルボニルアミノ、アミノカルボニルオキシ、非芳香族含窒素複素環基、および非芳香族複素環カルボニル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基(ここで非芳香族含窒素複素環基は、ピロリジニル、ピペリジニル、ピペラジニル、モルホリニル、およびチオモルホリニルから選択される)で置換されたアルキル、または置換基群B(置換基群B:アルキルオキシカルボニル、置換されていてもよいアミノカルボニル、オキソ、アミノ、カルボキシ、シアノ、シアノアルキル、ヒドロキシアルキル、アルキルカルボニルアミノ、アルキルスルホニルアミノ、およびアミノカルボニルアルキル)から選択される置換基で置換されていてもよい非芳香族含窒素複素環基(ここで非芳香族含窒素複素環基は、アゼチジニル、ピロリジニル、およびピペリジニルから選択される);

$R^{22}$ は、置換基群K(置換基群K:アルキルで置換されていてもよいアミノ、ヒドロキシ、アルキルオキシ、およびハロゲン)から選択される置換基で置換されていてもよいアルキル、置換基群Kから選択される置換基で置換されていてもよいアルケニル、置換基群Kから選択される置換基で置換されていてもよいアルキニル、ヒドロキシ、ハロゲン、フェニル、ピリジニル、フリル、チエニル、またはチアゾリル;

$R^{23}$ および $R^{24}$ の一方が水素原子、他方が水素原子、アルキルオキシ、アルキルオキシアルキルオキシ、ハロゲン、またはアルキルで置換されていてもよいアミノアルキル;

$R^{27}$ は、水素原子、アルキル、またはアシル;および

$R^{25}$ および $R^{26}$ は、それぞれ独立して、水素原子、ハロゲン、アルキニル、ヒドロキシ、アルキルオキシ、アルケニルオキシ、アルキニルオキシ、またはヘテロアリールアルキルオキシ]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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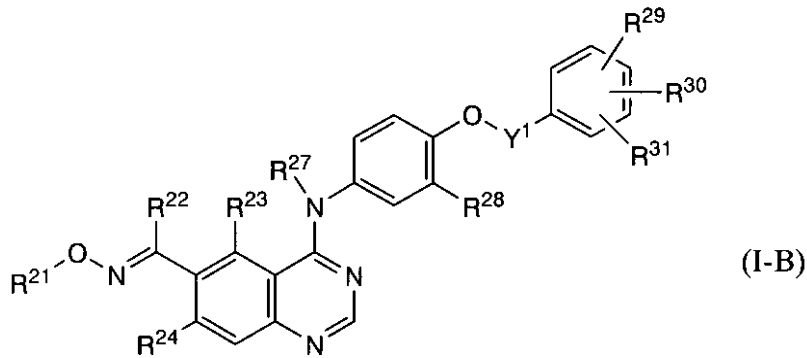
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式(I-B):

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【化40】



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[式中、 $R^{21}$ 、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$  および  $R^{27}$  は前記と同意義；

$Y^1$  は、C1～C3アルキレン；

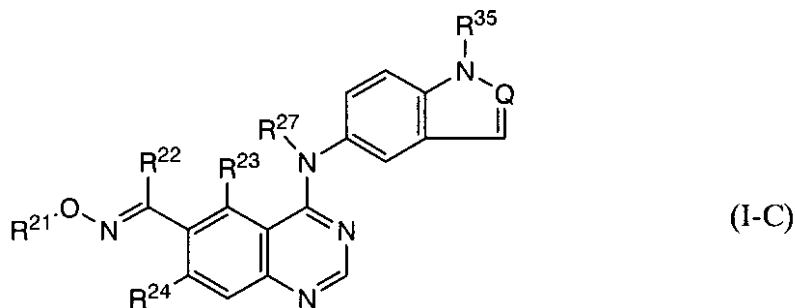
$R^{28}$  は、アルキル、アルキルオキシ、アルキニル、またはハロゲン；および

$R^{29}$ 、 $R^{30}$ 、および  $R^{31}$  は、それぞれ独立して、水素原子または置換基群F（置換基群F：ハロゲン、カルボキシ、アルキル、ハロアルキル、ヒドロキシアルキル、アルキルオキシ、アルキルオキシカルボニル、および置換されていてもよいアミノ）から選択される置換基]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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式(I-C)：

【化41】

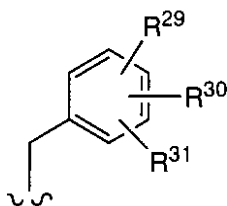


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[式中、 $R^{21}$ 、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、および  $R^{27}$  は前記と同意義；

$R^{35}$  は、置換基群L（置換基群L：ヒドロキシ、アルキルオキシ、ハロゲン、およびシアノ）から選択される置換基で置換されていてもよいアルキル、置換基群Lから選択される置換基で置換されていてもよいアルケニル、置換基群Lから選択される置換基で置換されていてもよいアルキニル、または式：

【化42】



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(式中、 $R^{29}$ 、 $R^{30}$ 、および  $R^{31}$  は前記と同意義)で表される基；

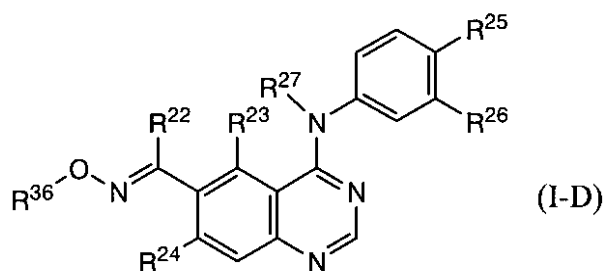
QはNまたは $CH_2$ ]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-D)：

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## 【化 4 3】



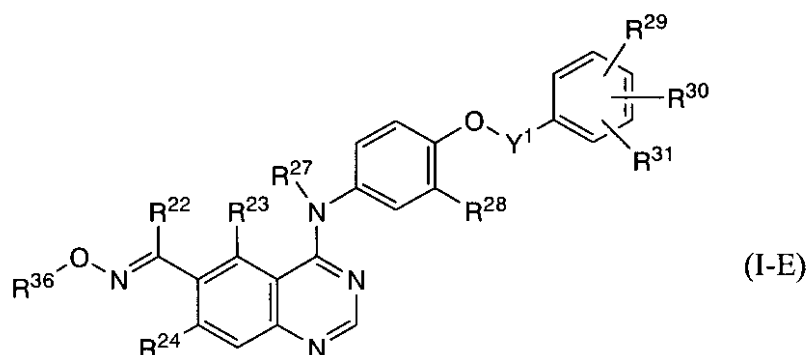
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[式中、 $R^{21}$ 、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{25}$ 、 $R^{26}$ 、および $R^{27}$ は前記と同意義；および

$R^{36}$ は、置換基群Cから選択される置換基で置換されていてもよいアミノカルボニルで置換されたアルキル、または置換基群Dから選択される置換基で置換されていてもよい非芳香族複素環カルボニル（ここで非芳香族含窒素複素環は、ピロリジニル、ピペリジル、ピペラジニル、モルホリニル、およびチオモルホリニルから選択される）で置換されたアルキル]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-E)：

## 【化 4 4】

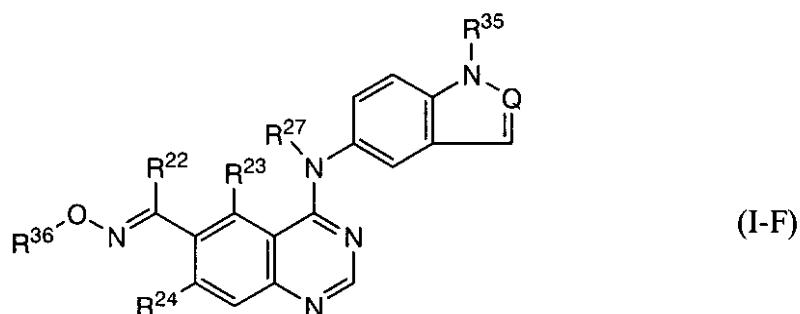


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[式中、 $R^{21}$ 、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{28}$ 、 $R^{29}$ 、 $R^{30}$ 、 $R^{31}$ 、 $R^{36}$ 、および $Y^1$ は前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-F)：

## 【化 4 5】



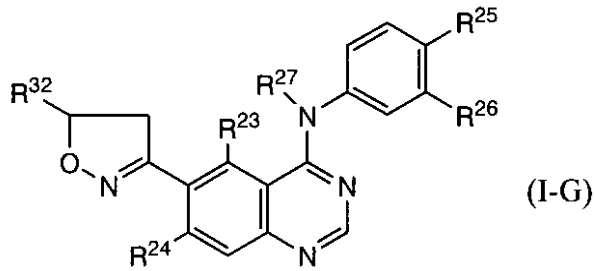
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[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{36}$ 、およびQは前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-G)：

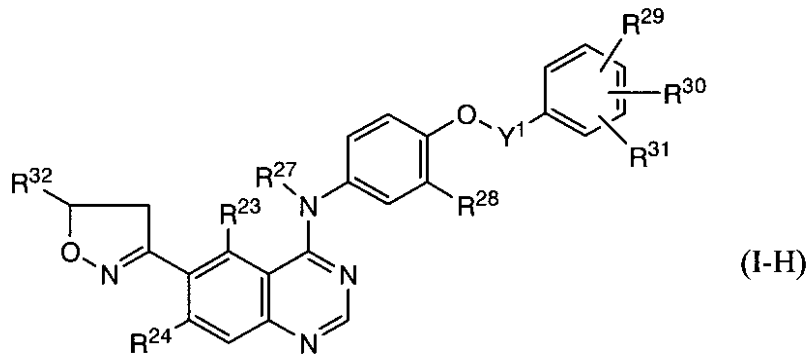
## 【化46】



[ 式中、 $R^{23}$ 、 $R^{24}$ 、 $R^{25}$ 、 $R^{26}$ 、および $R^{27}$ は前記と同意義；  
 $R^{32}$ は、置換基群Cから選択される置換基で置換されていてもよいアミノで置換されたアルキル、置換基群Dから選択される置換基で置換されていてもよい非芳香族含窒素複素環基（ここで非芳香族含窒素複素環基は、ピロリジニル、ピペリジル、ピペラジニル、モルホリニル、およびチオモルホリニルから選択される）で置換されたアルキル、または置換基群Bから選択される置換基で置換されていてもよい非芳香族含窒素複素環基（ここで非芳香族含窒素複素環基は、アゼチジニル、ピロリジニル、およびピペリジルから選択される）]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-H)：

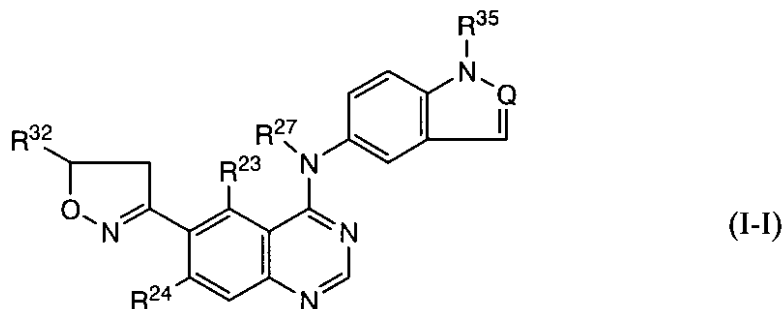
## 【化47】



[ 式中、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{28}$ 、 $R^{29}$ 、 $R^{30}$ 、 $R^{31}$ 、 $R^{32}$ 、および $Y^1$ は、前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-I)：

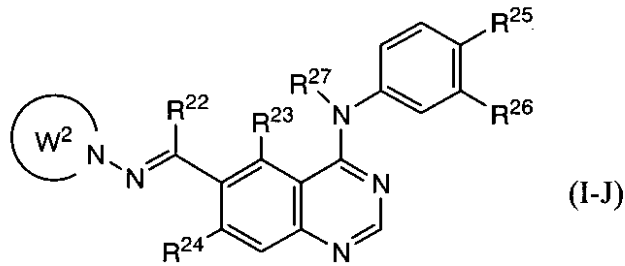
## 【化48】



[ 式中、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{32}$ 、 $R^{35}$ 、およびQは前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-J)：

【化49】

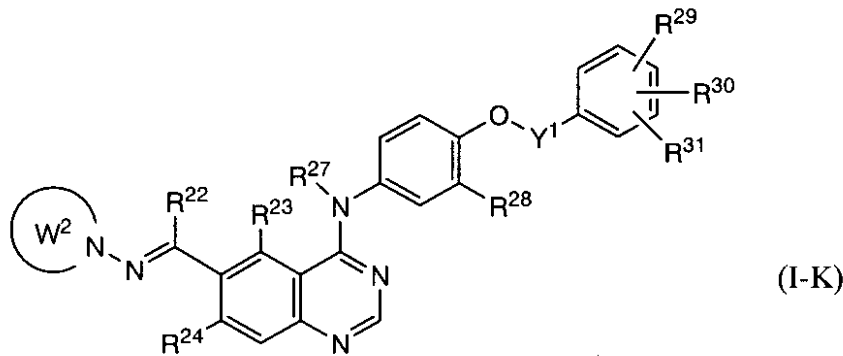


[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{25}$ 、 $R^{26}$ 、および $R^{27}$ は前記と同意義；  
 $W^2$ は、置換基群Jから選択される置換基で置換されていてもよいピロリジニル、置換基群Jから選択される置換基で置換されていてもよいピペリジル、置換基群Jから選択される置換基で置換されていてもよいピペラジニル、置換基群Jから選択される置換基で置換されていてもよいモルホリニル、または置換基群Jから選択される置換基で置換されていてもよいトリアゾリル]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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式(I-K)：

【化50】



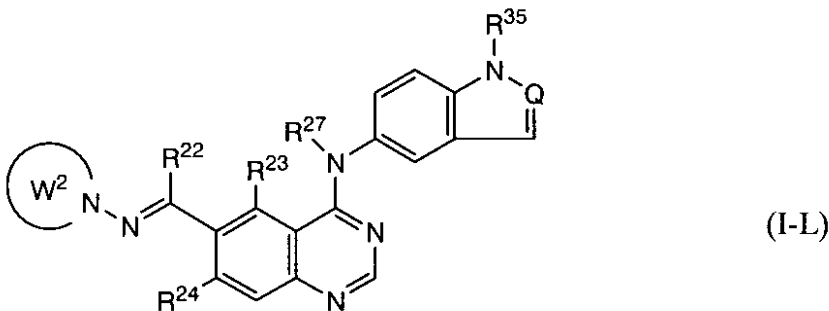
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[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{28}$ 、 $R^{29}$ 、 $R^{30}$ 、 $R^{31}$ 、 $Y^1$ 、および $W^2$ は、前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

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式(I-L)：

【化51】

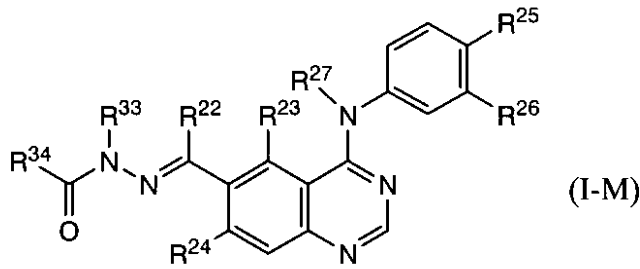


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[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{35}$ 、 $W^2$ 、およびQは前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-M)：

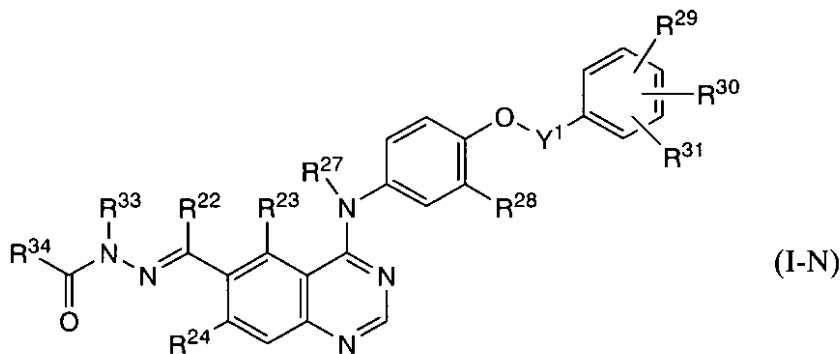
## 【化52】



[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{25}$ 、 $R^{26}$ 、および $R^{27}$ は前記と同意義；  
 $R^{33}$ は、水素原子、置換基群Lから選択される置換基で置換されていてもよいC1-C6アルキル、置換基群Lから選択される置換基で置換されていてもよいC2-C6置換されていてもよいアルケニル、または置換基群Lから選択される置換基で置換されていてもよいC2-C6置換されていてもよいアルキニル；および  
 $R^{34}$ は、置換基群Cから選択される置換基で置換されていてもよいアミノで置換されたアルキル、または置換基群Dから選択される置換基で置換されていてもよい非芳香族含窒素複素環基で置換されたアルキル、置換基群Fから選択される置換基で置換されていてもよいアリール、置換基群Fから選択される置換基で置換されていてもよいヘテロアリール、または置換基群Fから選択される置換基で置換されていてもよい非芳香族複素環基]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-N)：

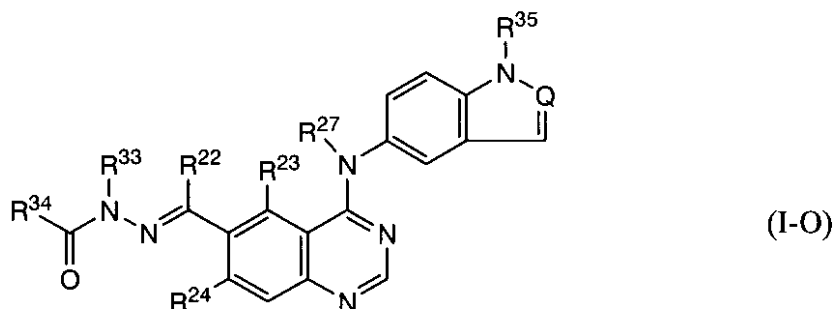
## 【化53】



[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{28}$ 、 $R^{29}$ 、 $R^{30}$ 、 $R^{31}$ 、 $R^{33}$ 、 $R^{34}$ 、および $Y^1$ は前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-O)：

## 【化54】



[式中、 $R^{22}$ 、 $R^{23}$ 、 $R^{24}$ 、 $R^{27}$ 、 $R^{34}$ 、 $R^{35}$ 、およびQは前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-P)：

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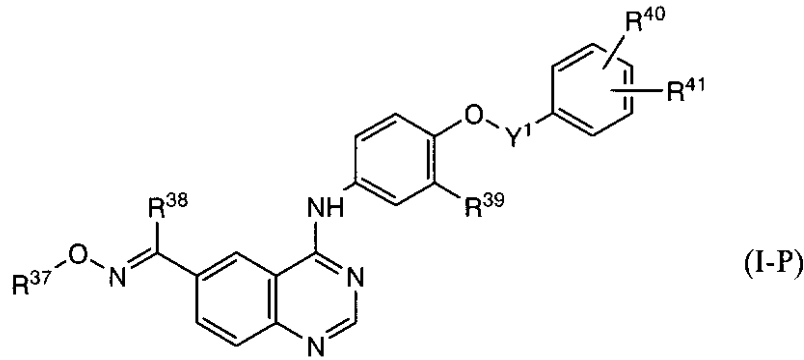
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## 【化55】



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[式中、 $R^{37}$ は、アルキル、ヒドロキシアルキル、アルキルカルボニルアミノアルキル、もしくは非芳香族複素環で置換されていてもよいアミノで置換されたアルキル、アルキル、ヒドロキシアルキル、もしくはアルキルカルボニルアミノで置換されていてもよい非芳香族含窒素複素環基で置換されたアルキル、またはヒドロキシアルキルで置換された非芳香族含窒素複素環基；

$R^{38}$ はC2～C4アルキニル；

$R^{39}$ はハロゲン；

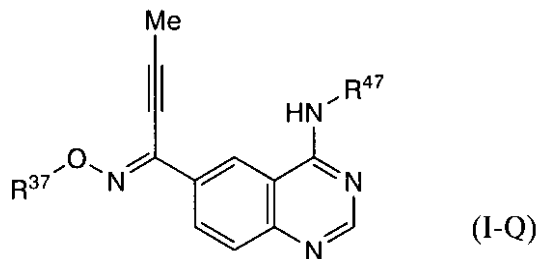
$R^{40}$ は、式： $-Y^3-R^{42}$ （式中、 $Y^3$ は、 $-O-$ を介在してもよいアルキレン； $R^{42}$ は、ハロゲンで置換されていてもよいフェニルまたはハロゲンで置換されていてもよいピリジル）で表される基；

$R^{41}$ はハロゲン；

$Y^1$ は、前記と同意義]で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式(I-Q)：

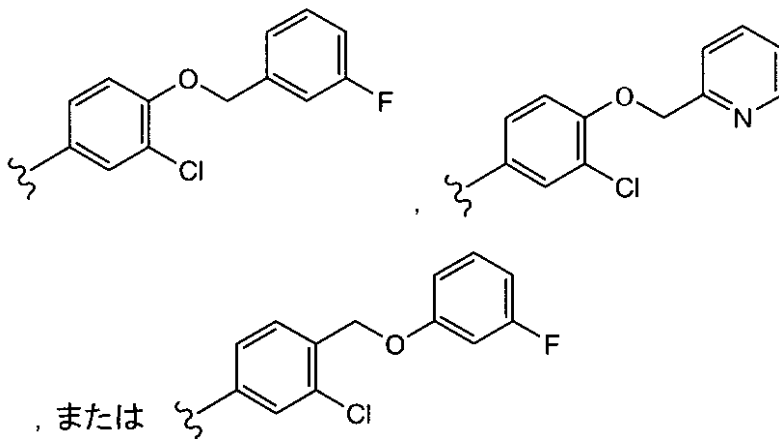
## 【化56】



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(式中、 $R^{47}$ は式：

## 【化57】



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で表される基； $R^{37}$ は前記と同意義）で表される化合物もしくはその製薬上許容される塩またはそれらの溶媒和物。

式（I - A）～式（I - F）および式（I - J）～式（I - O）で示される化合物における $R^{22}$ としては、メチル、エチル、*n*-プロピル、イソプロピル、プロピニル、ブチニルまたはフェニルが好ましい。

【0046】

本明細書中、「溶媒和物」とは、例えば有機溶媒との溶媒和物、水和物等を包含する。水和物を形成する時は、任意の数の水分子を配位していてもよい。

「本発明化合物」という場合には、製薬上許容される塩、またはその水和物も包含される。例えば、アルカリ金属（リチウム、ナトリウム、カリウム等）、アルカリ土類金属（マグネシウム、カルシウム等）、アンモニウム、有機塩基およびアミノ酸との塩、または無機酸（塩酸、臭化水素酸、リン酸、硫酸等）、および有機酸（酢酸、クエン酸、マレイン酸、フマル酸、ベンゼンスルホン酸、*p*-トルエンスルホン酸等）との塩が挙げられる。これらの塩は、通常行われる方法によって形成させることができる。

また、本発明化合物は特定の異性体に限定するものではなく、全ての可能な異性体やラセミ体を含むものである。

本発明化合物は後述する実験例の記載の通り、優れた細胞増殖阻害作用を示し、「抗癌剤」および「癌の治療剤」として使用しうる。

本発明化合物を、上記の疾患の治療を目的としてヒトに投与する場合は、散剤、顆粒剤、錠剤、カプセル剤、丸剤、液剤等として経口的に、または注射剤、坐剤、経皮吸収剤、吸入剤等として非経口的に投与することができる。また、本化合物の有効量にその剤型に適した賦形剤、結合剤、湿潤剤、崩壊剤、滑沢剤等の医薬用添加剤を必要に応じて混合し、医薬製剤とすることができる。注射剤の場合には、適当な担体と共に滅菌処理を行って製剤とする。

投与量は疾患の状態、投与ルート、患者の年齢、または体重によっても異なるが、成人に経口で投与する場合、通常0.1～100 mg/kg/日であり、好ましくは1～20 mg/kg/日である。

【0047】

以下に実施例および試験例を挙げて本発明をさらに詳しく説明するが、本発明はこれらにより限定されるものではない。

実施例中、以下の略号を使用する。

Me：メチル

Et：エチル

nPr：*n*-プロピル

iPr：イソプロピル

Ph：フェニル

【実施例】

【0048】

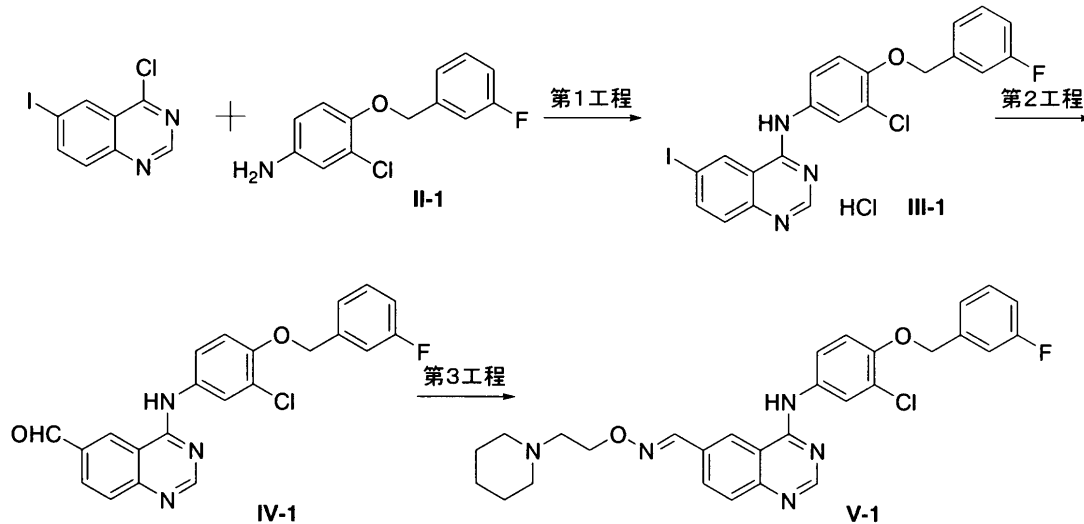
実施例1

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## 【化58】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ヨードキナゾリン(III-1)塩酸塩の合成

4-クロロ-6-ヨードキナゾリン(6.0g)と3-クロロ-4-(3-フルオロベンジルオキシ)アニリン(II-1, 5.2g)をテトラヒドロフラン180mlに溶解し、3時間加熱還流した。析出物を濾取し、テトラヒドロフランで洗浄することにより、4-(4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ヨードキナゾリン塩酸塩(III-1, 10.5g)を黄色固体として得た。

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$^1\text{H NMR}$ ( $d_6$ -DMSO): 11.47(1H, brs), 9.25(1H, s), 8.94(1H, s), 8.36(1H, dd,  $J=1.8\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 7.92(1H, d,  $J=2.7\text{Hz}$ ), 7.71(1H, d,  $J=8.7\text{Hz}$ ), 7.66(1H, dd,  $J=2.7\text{Hz}$ ,  $J=8.7\text{Hz}$ ),

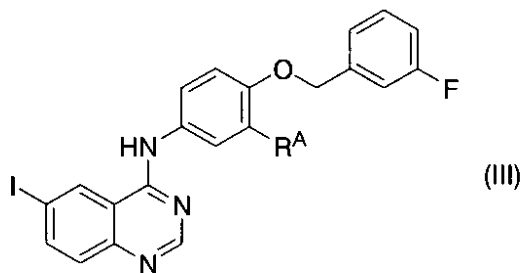
7.48(1H, dt,  $J=6.0\text{Hz}$ ,  $J=7.8\text{Hz}$ ), 7.37-7.30(3H, m), 7.19(1H, dt,  $J=2.7\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 5.30(2H, s).

## 【0049】

上記第1工程と同様の方法で下記の化合物を合成した。

## 【化59】

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【表 1】

化合物 No.	R <sup>A</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
III-2	-F	9.86(1H, s), 8.95(1H, d, J=1.5Hz), 8.61(1H, s), 8.11(1H, dd, J=1.8Hz, J=8.7Hz), 7.89(1H, J=2.4Hz, J=13.5Hz), 7.60-7.49(2H, m), 7.45(1H, dd, J=6.3Hz, J=8.1Hz), 7.36-7.23(3H, m), 7.19(1H, dt, J=8.4Hz, J=2.1Hz), 5.22(2H, s).
III-3	-OMe	11.26(1H, brs), 9.18(1H, s), 8.89(1H, s), 8.34(1H, dd, J=1.5Hz, J=8.7Hz), 7.66(1H, d, J=8.7Hz), 7.46(1H, dt, J=6.3Hz, J=8.1Hz), 7.39(1H, d, J=2.1Hz), 7.34-7.25(3H, m), 7.22-7.13(1H, m), 7.13(1H, d, J=8.7Hz), 5.17(2H, s), 3.82(3H, s).

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## 【 0 0 5 0 】

(第 2 工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ホルミルキナゾリン(IV-1)の合成

4-(4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ヨードキナゾリン塩酸塩(III-1, 5.0g)をジメチルホルムアミド100mlに溶かし、ギ酸ナトリウム1.25g、ジクロロビス(トリフェニルホスフィン)パラジウム(324mg)、トリエチルアミン(3.2ml)を加えた。反応液を脱気した後、一酸化炭素雰囲気下、80度で3時間加熱撹拌した。その後、反応液を氷水中に注ぎ、酢酸エチルで抽出した。抽出液を水及び食塩水で洗浄後、硫酸マグネシウムで乾燥した。乾燥した溶液をセライト濾過、濃縮後、酢酸エチル-ヘキサン混液(1:2)で粉末化し、4-(4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ホルミルキナゾリン(IV-1, 2.86g)を黄色固体として得た。

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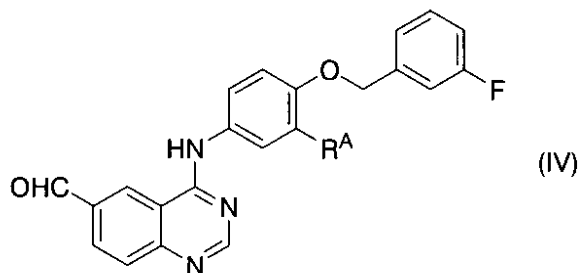
<sup>1</sup>H NMR(d<sub>6</sub>-DMSO): 10.24(1H, s), 10.13(1H, s), 9.16(1H, s), 8.69(1H, s), 8.27(1H, br d, J=8.7Hz), 8.03(1H, d, J=2.1Hz), 7.90(1H, d, J=9.0Hz), 7.74(1H, dd, J=0.7Hz, J=8.7Hz), 7.48(1H, dt, J=6.0Hz, J=8.4Hz), 7.36-7.30(2H, m), 7.30(1H, d, J=8.7Hz), 7.19(1H, br t, J=8.7Hz), 5.27(2H, s).

## 【 0 0 5 1 】

上記第 2 工程と同様の方法で下記の化合物を合成した。

## 【化 6 0】

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【表 2】

化合物 No.	RA	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
IV-2	-F	10.24(1H, s), 10.14(1H, s), 9.16(1H, s), 8.69(1H, s), 8.26(1H, dd, J=1.5Hz, J=8.7Hz), 7.94-7.85(2H, m), 7.55(1H, br d, J=9.0Hz), 7.47(1H, dt, J=6.3Hz, J=8.1Hz), 7.36-7.26(3H, m), 7.23-7.15(1H, m), 5.23(2H, s).
IV-3	-OMe	10.13(2H, s), 9.18(1H, s), 8.64(1H, s), 8.25(1H, dd, J=1.5Hz, J=8.7Hz), 7.88(1H, d, J=8.4Hz), 7.52-7.38(3H, m), 7.34-7.26(2H, m), 7.22-7.12(1H, m), 7.07(1H, d, J=8.7Hz), 5.14(2H, s), 3.83(3H, s).

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## 【 0 0 5 2 】

(第3工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(1-ピペリジル)エトキシイミノ)キナゾリン(V-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ホルミルキナゾリン(I V-1, 50mg)と2-(1-ピペリジル)エトキシアミン 2 塩酸塩(36.5mg)をテトラヒドロフラン(2 ml)と水(0.1ml)の混液に溶かし、室温で終夜反応した。反応液を酢酸エチルで薄め、飽和炭酸水素ナトリウム水溶液、食塩水で順に洗浄後、硫酸マグネシウム上で乾燥した。濃縮後、アミノカラムを用いたクロマトグラフィー(ヘキサン:酢酸エチル=2:3で溶出)により精製、さらに薄層クロマトグラフィーにより再精製し、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(1-ピペリジル)エトキシイミノ)キナゾリン(V-1, 13mg)を得た。

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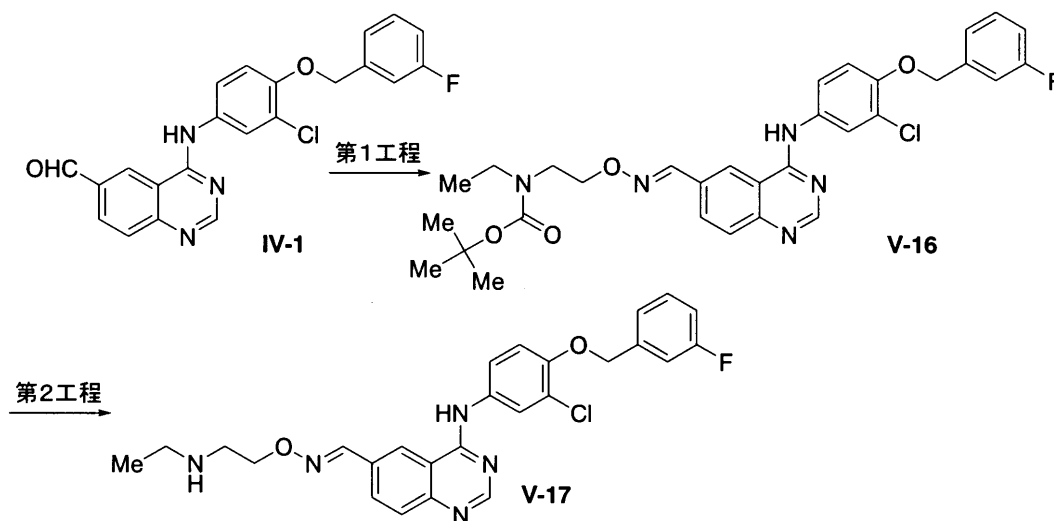
<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 9.95(1H, s), 8.63(1H, s), 8.59(1H, s), 8.34(1H, s), 8.10(1H, d, J=9.3Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz, J=2.1Hz), 7.51-7.44(1H, m), 7.36-7.31(2H, m), 7.28(1H, d, J=9.0Hz), 7.19(1H, br t, J=9.0Hz), 5.26(2H, s), 4.28(2H, t, J=6.0 Hz), 2.63(2H, t, J=6.0Hz), 2.41(4H, t, J=4.8Hz), 1.50(4H, quin, J=5.3Hz), 1.45-1.35(2H, m).

## 【 0 0 5 3 】

実施例 2

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## 【化 6 1】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(N-(t-ブトキシカルボニル)N-エチルアミノ)エトキシイミノ)キナゾリン(V-16)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ホルミルキナゾリン(I V-1, 120mg)とN-(t-ブトキシカルボニル)エチルアミノエトキシアミン(110mg)をテトラヒ

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ドロフラン(1.5ml)に溶かし、2mol/L塩酸134 $\mu$ lを加えて、室温で4時間反応した。反応終了後、氷冷した飽和炭酸水素ナトリウム水溶液中に注ぎ、酢酸エチルで抽出した。有機層を水及び食塩水で洗浄後、硫酸マグネシウム上で乾燥、濃縮した。得られた残渣をアミノカラムクロマトグラフィー(ヘキサン：酢酸エチル=1：1で溶出)により精製、さらにシリカゲルクロマトグラフィー(ヘキサン：酢酸エチル=1：1で溶出)により再精製し、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(N-(t-ブトキシカルボニル)-N-エチルアミノ)エチルオキシイミノ)キナゾリン(V-16, 96mg)を黄色粘性油状物質として得た。

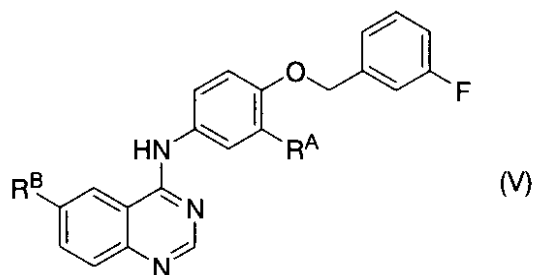
$^1\text{H NMR}$ ( $d_6$ -DMSO,  $\delta$ ): 9.93(1H, s), 8.63(1H, brs), 8.60(1H, s), 8.34(1H, s), 8.12(1H, dd,  $J=1.2\text{Hz}$ ,  $J=9.0\text{Hz}$ ), 8.01(1H, d,  $J=2.4\text{Hz}$ ), 7.79(1H, d,  $J=8.4\text{Hz}$ ), 7.72(1H, dd,  $J=2.4\text{Hz}$ ,  $J=8.4\text{Hz}$ ), 7.48(1H, dt,  $J=6.0\text{Hz}$ ,  $J=8.1\text{Hz}$ ), 7.36-7.30(2H, m), 7.28(1H, d,  $J=9.0\text{Hz}$ ), 7.23-7.14(1H, m), 5.26(2H, s), 4.26(2H, t,  $J=5.7\text{Hz}$ ), 3.50(2H, t,  $J=6.3\text{Hz}$ ), 3.23(2H, q,  $J=6.9\text{Hz}$ ), 1.38(9H, s), 1.12-1.00(3H, m).

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【0054】

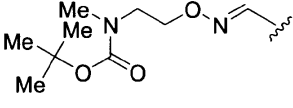
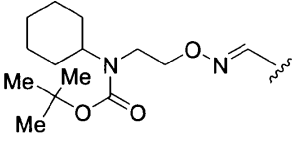
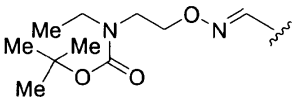
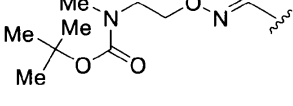
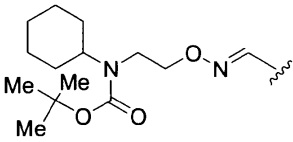
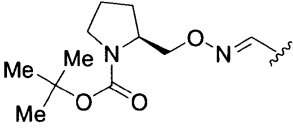
上記第1工程と同様の方法で下記の化合物を合成した。

【化62】



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【表 3】

化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-18	Cl		9.92(1H, s), 8.62(1H, s), 8.60(1H, s), 8.34(1H, s), 8.12(1H, d, J=8.4Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.4Hz, J=8.7Hz), 7.47(1H, dt, J=6.0Hz, J=7.8Hz), 7.36-7.30(2H, m), 7.28(1H, d, J=9.0Hz), 7.23-7.14(1H, m), 5.26(2H, s), 4.28(2H, t, J=5.7Hz), 3.57-3.48(2H, m), 2.84(3H, brs), 1.36(9H, s).
V-19	Cl		9.92(1H, s), 8.63(1H, s), 8.60(1H, s), 8.33(1H, s), 8.12(1H, dd, J=1.5Hz, J=9.0Hz), 8.01(1H, d, J=2.4Hz), 7.80(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.7Hz, J=9.0Hz), 7.47(1H, dt, J=6.0Hz, J=7.8Hz), 7.36-7.30(2H, m), 7.28(1H, d, J=9.0Hz), 7.22-7.14(1H, m), 5.26(2H, s), 4.21(2H, t, J=6.0Hz), 3.8-3.5(1H, brs), 3.48-3.37(2H, m), 1.8-1.4(6H, m), 1.35-1.0(4H, m).
V-20	OMe		9.83(1H, s), 8.64(1H, s), 8.57(1H, s), 8.34(1H, s), 8.12(1H, dd, J=1.5Hz, J=8.7Hz), 7.78(1H, d, J=8.7Hz), 7.50-7.36(3H, m), 7.34-7.26(2H, m), 7.21-7.13(1H, m), 7.06(1H, d, J=8.7Hz), 5.13(2H, s), 4.26(2H, t, J=5.7Hz), 3.82(3H, s), 3.50(2H, t, J=5.7Hz), 3.23(2H, q, J=7.2Hz), 1.38(9H, s), 1.12-1.02(3H, m).
V-21	OMe		9.84(1H, s), 8.63(1H, s), 8.56(1H, s), 8.36(1H, s), 8.12(1H, dd, J=1.5Hz, J=8.7Hz), 7.77(1H, d, J=8.7Hz), 7.50-7.35(3H, m), 7.33-7.25(2H, m), 7.20-7.12(1H, m), 7.05(1H, d, J=8.7Hz), 5.13(2H, s), 4.27(2H, t, J=5.7Hz), 3.82(3H, s), 3.55-3.47(2H, m), 2.84(3H, brs), 1.36(9H, s).
V-22	OMe		9.82(1H, s), 8.64(1H, s), 8.56(1H, s), 8.33(1H, s), 8.11(1H, dd, J=1.5Hz, J=8.7Hz), 7.78(1H, d, J=8.7Hz), 7.50-7.36(3H, m), 7.33-7.26(2H, m), 7.21-7.12(1H, m), 7.05(1H, d, J=8.7Hz), 5.13(2H, s), 4.21(2H, t, J=6.6Hz), 3.82(3H, s), 3.8-3.5(1H, m), 3.47-3.35(2H, m), 1.8-1.35(6H, m), 1.41(9H, s), 1.35-0.95(4H, m).
V-28	Cl		9.91(1H, s), 8.62(1H, brs), 8.60(1H, s), 8.36(1H, s), 8.10(1H, dd, J=8.7Hz), 8.01(1H, brs), 7.79(1H, d, J=8.4Hz), 7.73(1H, d, J=8.1Hz), 7.52-7.44(1H, m), 7.35-7.27(3H, m), 7.19(1H, dt, J=8.1Hz, J=2.4Hz), 5.26(2H, s), 4.28(1H, brs), 4.17(1H, dd, J=10.5Hz, J=6.9Hz), 4.00(1H, brs), 3.25(3H, br), 1.92(3H, br), 1.80(1H, br), 1.43(9H, s).

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## 【 0 0 5 5 】

(第 2 工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(N-エチルアミノ)エチルオキシイミノ)キナゾリン(V-17)の合成

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4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(N-(t-ブトキシカルボニル)-N-エチルアミノ)エチルオキシイミノ)キナゾリン(V-16, 54mg)をクロロホルム(1.8ml)に溶かし、トリフルオロ酢酸(1.8ml)を加え、室温で1時間攪拌した。反応終了後、反応液を濃縮し、酢酸エチルで希釈した。飽和炭酸水素ナトリウム水溶液、水、食塩水で順次洗浄後、硫酸マグネシウム上で乾燥した。濃縮後、結晶性残渣をジクロロメタン-ヘキサン(1:4)の混液で洗い、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(N-エチルアミノ)エチルオキシイミノ)キナゾリン(V-17, 28mg)を白色固体として得た。

<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 9.96(1H, s), 8.65(1H, brs), 8.60(1H, s), 8.35(1H, s), 8.11

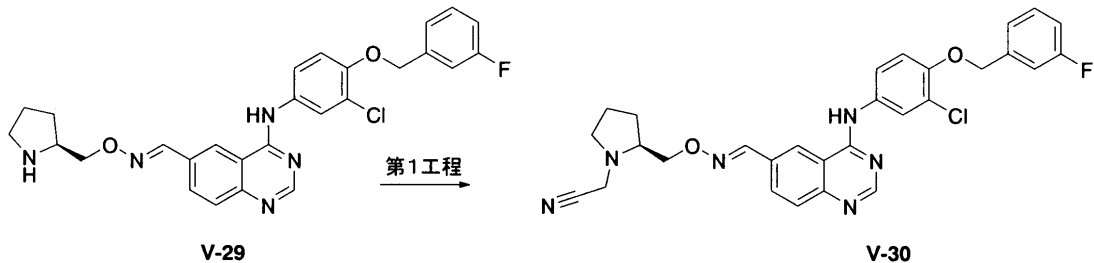
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(1H, d, J=9.0Hz), 8.01(1H, d, J=3.0Hz), 7.80(1H, d, J=9.0Hz), 7.75-7.70(1H, m), 7.52-7.43(1H, m), 7.36-7.31(2H, m), 7.28(1H, d, J=9.3Hz), 7.23-7.14(1H, m), 5.26(2H, s), 4.26(2H, t, J=6.0Hz), 2.93(2H, t, J=6.0Hz), 2.66(2H, q, J=7.2Hz), 1.05(3H, t, J=7.2Hz).

【 0 0 5 6 】

実施例 3

【 化 6 3 】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(N-シアノメチルピロリジン-2-イル)メトキシイミノ)メチルキナゾリン(V-30)の合成

化合物V-29 40mgおよびヨードアセトニトリル8 $\mu$ LをN,N-ジメチルアセタミド0.4mLに溶かし、炭酸カリウム15mgを加えて、室温で1時間攪拌した。反応後、飽和食塩水溶液を加え、酢酸エチルで抽出した。有機層をプレセップ(登録商標)を通して脱水し、ろ液を濃縮した。濃縮残渣をシリカゲルクロマトグラフィー(ヘキサン:酢酸エチル=2:1で溶出)により精製し、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(N-

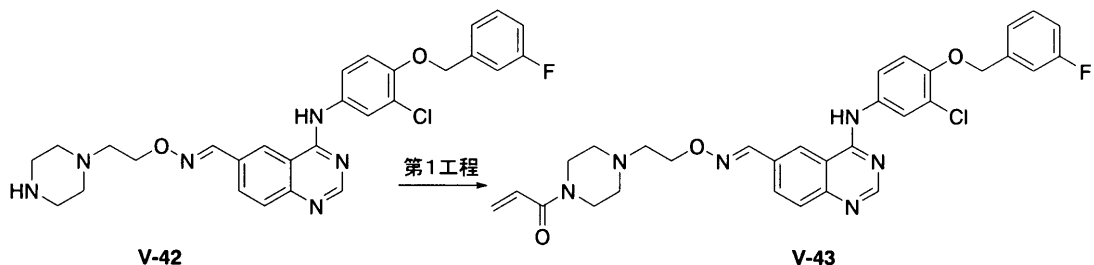
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シアノメチルピロリジン-2-イル)メトキシイミノ)メチルキナゾリン(V-30)を35mg得た。  
 $^1\text{H NMR}$ ( $d_6$ -DMSO, ): 9.92(1H, s), 8.62(1H, d, J=1.5Hz), 8.58(1H, s), 8.36(1H, s), 8.09(1H, dd, J=8.7Hz, J=1.5Hz), 7.99(1H, d, J=2.6Hz), 7.78(1H, d, J=8.7Hz), 7.71(1H, dd, J=9.0Hz, J=2.6Hz), 7.49-7.42(1H, m), 7.33-7.25(3H, m), 7.16(1H, dt, J=8.1Hz, J=1.8Hz), 5.24(2H, s), 4.17(2H, d, J=5.7Hz), 3.97(1H, d, J=17Hz), 3.90(1H, d, J=17Hz), 3.00-2.89(2H, m), 2.01-1.89(1H, m), 1.78-1.54(3H, m).

【 0 0 5 7 】

実施例 4

【 化 6 4 】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(4-アクリロイル-1-ピペラジン-1-イル)エトキシイミノ)メチルキナゾリン(V-43)の合成

化合物V-42 40mgおよび塩化アクリロイル6 $\mu$ Lをテトラヒドロフラン1.5mLに溶かし、室温で2時間攪拌した。反応後、飽和食塩水溶液を加え、酢酸エチルで抽出した。有機層をCHEM ELUT(商品名)を通して脱水し、ろ液を濃縮した。濃縮残渣を、アミノカラムを用いたクロマトグラフィー(1%メタノール含有酢酸エチルで溶出)により精製し、得られた残渣をヘキサン-酢酸エチル(6:1)にて固体化させ、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(4-アクリロイル-1-ピペラジン-1-イル)エトキシイミノ)メチルキナゾリン(V-43)を20mg得た。

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$^1\text{H NMR}$ ( $d_6$ -DMSO, ): 9.95(1H, s), 8.64(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, d, J=8.7Hz), 8.00(1H, d, J=2.4Hz), 7.78(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz

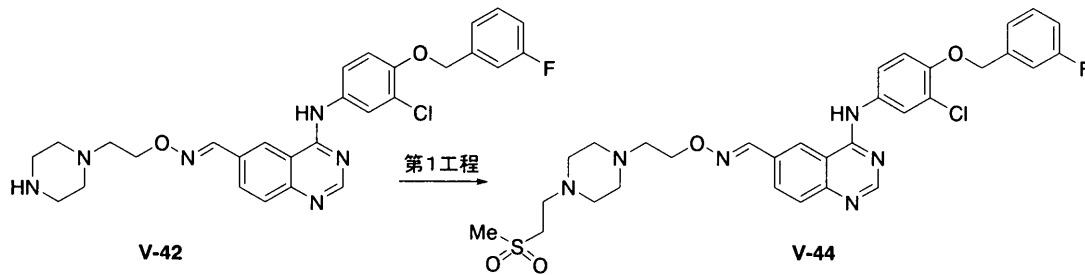
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, 2.7Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.22(1H, m), 6.79(1H, dd, J=16.5Hz, 10.2Hz), 6.10(1H, dd, J=16.5Hz, 2.4Hz), 6.17(1H, dd, J=10.5Hz, 2.4Hz), 5.26(2H, s), 4.32(2H, t, J=5.7Hz), 3.55(4H, m), 2.72(2H, t, J=5.7Hz), 2.50(4H, m)

【 0 0 5 8 】

実施例 5

【 化 6 5 】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(4-(2-メタンスルホニルエチル)-1-ピペラジン-1-イル)エトキシイミノ)メチルキナゾリン(V-44)の合成

化合物V-42 25mgおよびメチルビニルスルホン4 $\mu$ Lをテトラヒドロフラン1mLに溶かし、室温で21時間攪拌した。さらにメチルビニルスルホン4 $\mu$ Lを加えた後、室温で29時間攪拌した。反応後、飽和食塩水溶液を加え、酢酸エチルで抽出した。有機層を水、飽和食塩水溶液の順で洗浄後、硫酸マグネシウム上で乾燥した。濃縮残渣をアミノカラムを用いたクロマトグラフィー(酢酸エチルで溶出)により精製し、得られた残渣をヘキサン-酢酸エチル(6:1)にて固体化させ、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(2-(4-(2-メタンスルホニルエチル)-1-ピペラジン-1-イル)エトキシイミノ)メチルキナゾリン(V-44)を15mg得た。

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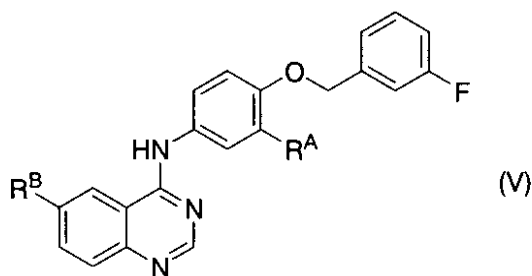
<sup>1</sup>H NMR( $d_6$ -DMSO, ): 9.95(1H, s), 8.63(1H, s), 8.59(1H, s), 8.34(1H, s), 8.10(1H, d, J=9.6Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.4Hz), 7.71(1H, dd, J=8.7Hz, 2.7Hz), 7.44-7.51(1H, m), 7.26-7.34(3H, m), 7.15-7.22(1H, m), 5.26(2H, s), 4.29(2H, t, J=6.0Hz), 3.27(2H, t, J=6.6Hz), 3.02(3H, s), 2.67-2.70(4H, m), 2.46(8H, m)

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【 0 0 5 9 】

実施例 1 ~ 5 と同様の方法で下記の化合物を合成した。

【 化 6 6 】



(V)

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【 0 0 6 0 】

【表 4】

化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-2	Cl		11.69(1H, s), 11.11(1H, brs), 8.84(1H, s), 8.83(1H, s), 8.29(1H, s), 8.26(1H, d, J=8.7Hz), 7.95(1H, d, J=2.7Hz), 7.88(1H, d, J=8.7Hz), 7.67(1H, dd, J=2.3Hz, J=9.2Hz), 7.48(1H, dt, J=6.0Hz, J=7.8Hz), 7.37-7.29(3H, m), 7.20(1H, br t, J=9.2Hz), 5.30(2H, s).
V-3	Cl		9.96(1H, s), 8.65(1H, s), 8.60(1H, s), 8.34(1H, s), 8.10(1H, br d, J=8.7Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.4Hz), 7.73(1H, dd, J=2.7Hz, J=9.0Hz), 7.48(1H, dt, J=6.0Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.0Hz), 7.19(1H, br t, J=8.7Hz), 5.26(2H, s), 3.98(3H, s).
V-4	Cl		9.95(1H, s), 8.63(1H, s), 8.60(1H, s), 8.36(1H, s), 8.10(1H, dd, J=8.7Hz, J=1.2Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.4Hz), 7.72(1H, dd, J=2.6Hz, J=8.9Hz), 7.48(1H, dt, J=6.0Hz, J=8.1Hz), 7.36-7.29(2H, m), 7.28(1H, d, J=9.0Hz), 7.19(1H, dt, J=2.4Hz, J=8.6Hz), 5.27(2H, s), 3.97(2H, d, J=6.9Hz), 2.05(1H, m), 0.95(6H, d, J=6.9Hz).
V-5	Cl		9.96(1H, s), 8.64(1H, s), 8.60(1H, s), 8.39(1H, s), 8.11(1H, dd, J=1.5Hz, J=8.7Hz), 8.01(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.7Hz, J=9.0Hz), 7.48(1H, dt, J=6.3Hz, J=7.8Hz), 7.36-7.29(2H, m), 7.28(1H, d, J=9.0Hz), 7.19(1H, dt, J=2.2Hz, J=8.6Hz), 6.08(1H, ddt, J=17.4Hz, J=10.5Hz, J=5.7Hz), 5.38(1H, ddd, J=17.4Hz, J=3.6Hz, J=1.8Hz), 5.30-5.24(1H, m), 5.26(2H, s), 4.72(2H, dt, J=5.7Hz, J=1.5Hz).
V-6	Cl		9.95(1H, s), 8.64(1H, s), 8.60(1H, s), 8.35(1H, s), 8.11(1H, br d, J=8.7Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.7Hz, J=9.0Hz), 7.51-7.44(1H, m), 7.36-7.31(2H, m), 7.28(1H, d, J=9.3Hz), 7.19(1H, br t, J=8.4Hz), 5.26(2H, s), 4.29(2H, t, J=6.3Hz), 2.810(2H, t, J=4.8Hz), 2.6-2.5(4H, m), 1.75-1.65(4H, m).
V-7	Cl		9.95(1H, s), 8.64(1H, s), 8.60(1H, s), 8.35(1H, s), 8.11(1H, dd, J=8.7Hz, J=1.5Hz), 8.01(1H, d, J=2.7Hz), 7.80(1H, d, J=9.0Hz), 7.72(1H, dd, J=9.0Hz, J=2.7Hz), 7.48(1H, dt, J=6.0Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.3Hz), 7.19(1H, br t, J=9.0Hz), 5.27(2H, s), 4.31(2H, t, J=5.7Hz), 3.58(4H, t, J=5.1Hz), 2.67(2H, t, J=5.7Hz), 2.46(4H, t, J=4.5Hz).

【表 5】

化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-8	-Cl		9.94(1H, s), 8.63(1H, s), 8.59(1H, s), 8.33(1H, s), 8.10(1H, br d, J=7.2Hz), 8.00(1H, d, J=2.4Hz), 7.78(1H, d, J=8.7Hz), 7.71(1H, dd, J=8.7Hz, J=2.1Hz), 7.47(1H, dt, J=6.3Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.0Hz), 7.18(1H, br t, J=9.0Hz), 5.26(2H, s), 4.23(2H, t, J=6.3Hz), 3.57(4H, t, J=4.5Hz), 2.45-2.35(6H, m), 1.87(2H, quin, J=6.8Hz).
V-9	-Cl		9.95(1H, s), 8.63(1H, s), 8.60(1H, s), 8.34(1H, s), 8.11(1H, dd, J=8.7Hz, J=1.5Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=8.7Hz, J=2.7Hz), 7.48(1H, dt, J=6.0Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.0Hz), 7.18(1H, dt, J=2.4Hz, J=8.4Hz), 5.26(2H, s), 4.28(2H, t, J=5.7Hz), 2.63(2H, t, J=5.7Hz), 2.23(6H, s).
V-10	-Cl		9.93(1H, s), 8.63(1H, s), 8.60(1H, s), 8.33(1H, s), 8.11(1H, dd, J=8.7Hz, J=1.5Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.7Hz, J=8.7Hz), 7.47(1H, dt, J=5.7Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.3Hz), 7.18(1H, dt, J=8.7Hz, J=2.7Hz), 5.26(2H, s), 4.25(2H, t, J=6.2Hz), 2.79(2H, brs), 2.57(4H, q, J=6.6Hz), 0.99(6H, t, J=7.1Hz).
V-11	-Cl		9.91(1H, s), 8.61(1H, d, J=1.5Hz), 8.60(1H, s), 8.31(1H, s), 8.12(1H, dd, J=1.5Hz, J=8.7Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.4Hz, J=8.7Hz), 7.47(1H, dt, J=5.7Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.3Hz), 7.18(1H, dt, J=8.7Hz, J=2.7Hz), 5.26(2H, s), 4.10(2H, t, J=6.9Hz), 3.01(2H, quin, J=6.3Hz), 2.74(2H, t, J=6.9Hz), 1.99(12H, d, 6.3Hz).
V-12	-F		9.94(1H, s), 8.64(1H, s), 8.60(1H, s), 8.34(1H, s), 8.11(1H, d, J=8.8Hz), 7.88(1H, d, J=13.2Hz), 7.79(1H, d, J=8.8Hz), 7.56-7.44(2H, m), 7.36-7.24(3H, m), 7.19(1H, br t, J=8.8Hz), 5.18(2H, s), 4.32-4.27(2H, m), 2.82-2.74(2H, m), 2.55-2.48(4H, m), 1.73-1.65(4H, m).
V-13	-OMe		9.84(1H, s), 8.64(1H, d, J=1.5Hz), 8.55(1H, s), 8.34(1H, s), 8.09(1H, dd, J=1.5Hz, J=8.7Hz), 7.77(1H, d, J=8.7Hz), 7.49-7.36(3H, m), 7.33-7.26(2H, m), 7.20-7.12(1H, m), 7.05(1H, d, J=9.0Hz), 5.13(2H, s), 4.28(2H, t, J=6.0Hz), 3.82(3H, s), 2.76(2H, t, J=6.0Hz), 2.54-2.46(4H, m), 1.76-1.62(4H, m).

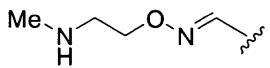
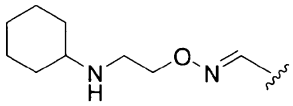
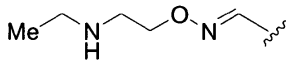
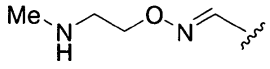
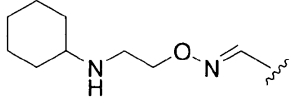
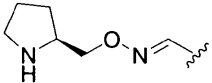
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【表 6】

化合物 No.	RA	RB	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-23	Cl		9.96(1H, s), 8.64(1H, s), 8.60(1H, s), 8.34(1H, s), 8.11(1H, dd, J=1.2Hz, J=8.4Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=2.4Hz, J=8.7Hz), 7.48(1H, dt, J=6.0Hz, J=8.4Hz), 7.36-7.30(2H, m), 7.28(1H, d, J=9.0Hz), 7.23-7.15(1H, m), 5.27(2H, s), 4.24(2H, t, J=5.7Hz), 2.81(2H, t, J=5.7Hz), 2.33(3H, s).
V-24	Cl		9.95(1H, s), 8.65(1H, d, J=1.2Hz), 8.60(1H, s), 8.35(1H, s), 8.11(1H, dd, J=1.5Hz, J=8.7Hz), 8.01(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.73(1H, dd, J=2.7Hz, J=9.0Hz), 7.47(1H, dt, J=6.0Hz, J=8.1Hz), 7.36-7.30(2H, m), 7.28(1H, d, J=9.0Hz), 7.23-7.14(1H, m), 5.26(2H, s), 4.25(2H, t, J=5.7Hz), 2.95(2H, t, J=5.7Hz), 2.55-2.4(1H, m), 1.9-1.8(2H, m), 1.75-1.62(2H, m), 1.6-1.5(1H, m), 1.3-0.97(5H, m).
V-25	OMe		9.86(1H, s), 8.64(1H, s), 8.56(1H, s), 8.34(1H, s), 8.10(1H, d, J=8.4Hz), 7.77(1H, d, J=8.7Hz), 7.5-7.35(3H, m), 7.34-7.26(2H, m), 7.21-7.13(1H, m), 7.06(1H, d, J=8.7Hz), 5.13(2H, s), 4.23(2H, t, J=6.0Hz), 3.82(3H, s), 2.86(2H, t, J=6.0Hz), 2.59(2H, q, J=7.2Hz), 1.02(3H, t, J=7.2Hz).
V-26	OMe		9.86(1H, s), 8.64(1H, s), 8.56(1H, s), 8.33(1H, s), 8.10(1H, d, J=8.7Hz), 7.77(1H, d, J=9.0Hz), 7.50-7.35(3H, m), 7.33-7.26(2H, m), 7.21-7.13(1H, m), 7.05(1H, d, J=8.4Hz), 5.13(2H, s), 4.23(2H, t, J=5.7Hz), 3.82(3H, s), 2.81(2H, d, J=6.0Hz), 2.33(3H, s).
V-27	OMe		9.86(1H, s), 8.64(1H, s), 8.56(1H, s), 8.34(1H, s), 8.09(1H, dd, J=1.5Hz, J=8.7Hz), 7.77(1H, d, J=8.7Hz), 7.50-7.35(3H, m), 7.33-7.26(2H, m), 7.21-7.13(1H, m), 7.05(1H, d, J=8.7Hz), 5.13(2H, s), 4.22(2H, t, J=5.7Hz), 3.82(3H, s), 2.88(2H, t, J=5.7Hz), 2.45-2.35(1H, m), 1.88-1.76(2H, m), 1.72-1.62(2H, m), 1.6-1.5(2H, m), 1.3-0.93(4H, m).
V-29	Cl		9.94(1H, s), 8.65(1H, brs), 8.60(1H, s), 8.35(1H, s), 8.11(1H, dd, J=8.7Hz, J=1.2Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz, J=2.1Hz), 7.51-7.44(1H, m), 7.35-7.27(3H, m), 7.18(1H, dt, J=8.1Hz, J=1.5Hz), 5.26(2H, s), 4.12(2H, d, J=6.3Hz), 3.53-3.44(1H, m), 2.96-2.82(2H, m), 1.94-1.64(3H, m), 1.53-1.41(1H, m).

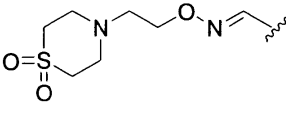
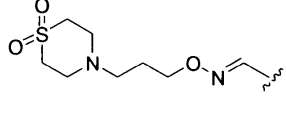
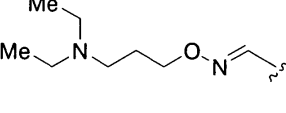
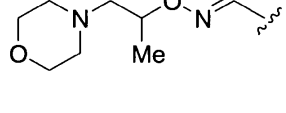
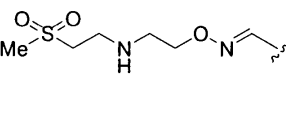
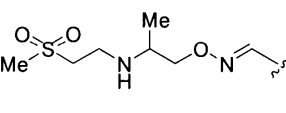
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【表 7】

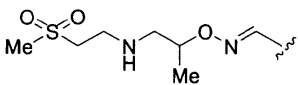
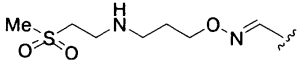
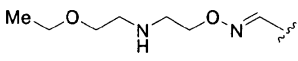
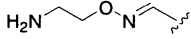
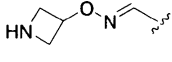
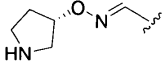
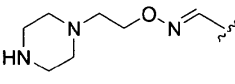
化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-30	-Cl		9.95(1H, s), 8.64(1H, s), 8.59(1H, s), 8.35(1H, s), 8.11(1H, d, J=8.7Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.71(1H, dd, J=8.4Hz, J=1.8Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.22(1H, m), 5.26(2H, s), 4.31(2H, t, J=5.7Hz), 3.08-3.10(4H, m), 3.02-3.04(4H, m), 2.91(2H, t, J=5.7Hz)
V-31	-Cl		9.95(1H, s), 8.63(1H, s), 8.60(1H, s), 8.34(1H, s), 8.11(1H, d, J=9.3Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0 Hz, J=2.4Hz), 7.51-7.44(1H, m), 7.35-7.27(3H, m), 7.22-7.15(1H, m), 5.26(2H, s), 4.23(2H, t, J=6.3Hz), 3.09-3.07(4H, m), 2.92-2.90(4H, m), 2.59(2H, t, J=6.9Hz), 1.90-1.85(2H, m)
V-32	-Cl		9.94(1H, s), 8.64(1H, s), 8.59(1H, s), 8.33(1H, s), 8.10(1H, d, J=8.7Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=8.7Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.22(1H, m), 5.26(2H, s), 4.22(2H, t, J=6.3Hz), 2.49-2.51(6H, m), 1.78-1.83(2H, m), 0.95(6H, t, J=7.2Hz)
V-33	-Cl		9.94(1H, s), 8.61(1H, s), 8.59(1H, s), 8.31(1H, s), 8.11(1H, dd, J=8.4Hz, 1.5Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.71(1H, dd, J=9.3Hz, 2.7Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.22(1H, m), 5.26(2H, s), 4.50-4.56(1H, m), 3.57(4H, t, J=4.8Hz), 2.59-2.65(2H, m), 2.46(4H, m), 1.31(3H, d, J=6.0Hz)
V-34	-Cl		9.95(1H, s), 8.65(1H, s), 8.60(1H, s), 8.34(1H, s), 8.12(1H, dd, J=9.0Hz, 1.8Hz), 8.01(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=8.7Hz, J=2.4Hz), 7.44-7.52(1H, m), 7.27-7.35(3H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.25(2H, t, J=5.4Hz), 3.25(2H, t, J=6.9Hz), 3.02(3H, s), 2.99(2H, t, J=6.9Hz), 2.89(2H, t, J=5.7Hz)
V-35	-Cl		9.95(1H, s), 8.65(1H, s), 8.60(1H, s), 8.36(1H, s), 8.12(1H, dd, J=8.7Hz, 1.8Hz), 8.01(1H, d, J=2.7Hz), 7.80(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz, 2.4Hz), 7.44-7.52(1H, m), 7.27-7.35(3H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.04-4.09(2H, m), 3.20-3.26(2H, m), 2.98-3.08(6H, m), 1.06(3H, d, J=6.3Hz)

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【表 8】

化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-36	-Cl		9.95(1H, s), 8.63(1H, s), 8.60(1H, s), 8.32(1H, s), 8.13(1H, d, J=9.0Hz), 8.00(1H, d, J=2.7Hz), 7.80(1H, d, J=8.4Hz), 7.72(1H, dd, J=9.3Hz, 2.7Hz), 7.44-7.52(1H, m), 7.27-7.35(3H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.40-4.46(1H, m), 3.27(2H, t, J=6.9Hz), 2.99-3.02(5H, m), 2.74-2.90(2H, m), 1.30(3H, d, J=6.6Hz)
V-37	-Cl		9.95(1H, s), 8.64(1H, s), 8.60(1H, s), 8.34(1H, s), 8.11(1H, dd, J=9.0Hz, 1.2Hz), 8.01(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=8.7Hz, 2.7Hz), 7.44-7.51(1H, m), 7.26-7.34(3H, m), 7.16-7.22(1H, m), 5.26(2H, s), 4.25(2H, t, J=6.3Hz), 3.25(2H, t, J=7.2Hz), 3.03(3H, s), 2.98(2H, t, J=6.9Hz), 2.70(2H, t, J=6.9Hz), 1.84-1.91(2H, m)
V-38	-Cl		9.96(1H, s), 8.64(1H, s), 8.60(1H, s), 8.35(1H, s), 8.11(1H, dd, J=8.7Hz, 1.5Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.73(1H, dd, J=9.0Hz, 2.7Hz), 7.44-7.52(1H, m), 7.27-7.35(3H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.25(2H, t, J=5.4Hz), 3.38-3.46(4H, m), 2.91(2H, t, J=5.7Hz), 2.73(2H, t, J=5.4Hz), 1.09(3H, t, J=6.9Hz)
V-39	-Cl		9.96(1H, s), 8.63(1H, brs), 8.60(1H, s), 8.34(1H, s), 8.11(1H, dd, J=8.7Hz, J=1.5Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz, J=2.4Hz), 7.52-7.44(1H, m), 7.35-7.27(3H, m), 7.20(1H, dt, J=9.3Hz, J=2.7Hz), 5.27(2H, s), 4.37(2H, t, J=6.0Hz), 3.51(2H, t, J=6.0Hz)
V-40	-Cl		9.95(1H, s), 8.64(1H, brs), 8.59(1H, s), 8.40(1H, s), 8.09(1H, d, J=6.6Hz), 8.01(1H, brs), 7.78(1H, d, J=6.3Hz), 7.72(1H, d, J=6.3Hz), 7.50-7.45(1H, m), 7.35-7.26(3H, m), 7.19(1H, t, J=6.3Hz), 5.26(2H, s), 5.09(1H, m), 3.69(2H, t-like, J=5.1Hz), 3.60(2H, t-like, J=5.1Hz)
V-41	-Cl		9.93(1H, s), 8.62(1H, brs), 8.59(1H, s), 8.30(1H, s), 8.11(1H, d, J=6.3Hz), 8.00(1H, d, J=1.5Hz), 7.79(1H, d, J=6.6Hz), 7.72(1H, dd, J=6.6Hz, J=1.8Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=5.7Hz), 5.26(2H, s), 4.88(1H, brs), 3.01-2.88(3H, m), 2.79-2.73(1H, m), 1.99-1.85(2H, m)
V-42	-Cl		9.95(1H, s), 8.63(1H, brs), 8.60(1H, s), 8.35(1H, s), 8.11(1H, dd, J=8.7Hz, J=1.5Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.3Hz, J=2.7Hz), 7.52-7.44(1H, m), 7.35-7.27(3H, m), 7.19(1H, dt, J=9.3Hz, J=2.7Hz), 5.27(2H, s), 4.29(2H, t, J=6.0Hz), 2.69(4H, t, J=4.8Hz), 2.63(2H, t, J=6.0Hz), 2.39(4H, brs)

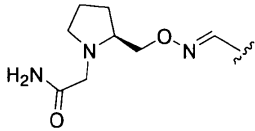
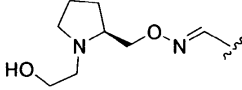
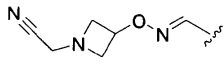
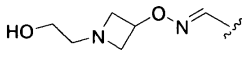
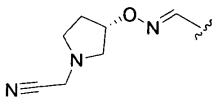
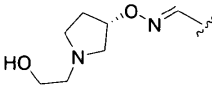
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【表 9】

化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-45	-Cl		9.93(1H, s), 8.63(1H, brs), 8.60(1H, s), 8.33(1H, s), 8.10(1H, d, J=6.6Hz), 8.02(1H, d, J=1.5Hz), 7.79(1H, d, J=6.6Hz), 7.73(1H, d, J=6.9Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(2H, m), 7.08(1H, brs), 5.26(2H, s), 4.20(1H, dd, J=8.1Hz, J=4.2Hz), 4.13(1H, dd, J=8.1Hz, J=4.2Hz), 3.38(1H, d, J=12Hz), 3.08-2.93(3H, m), 2.38(1H, m), 1.99-1.89(1H, m), 1.76-1.57(3H, m)
V-46	-Cl		9.93(1H, s), 8.63(1H, brs), 8.60(1H, s), 8.34(1H, s), 8.11(1H, d, J=6.6Hz), 8.01(1H, d, J=2.1Hz), 7.80(1H, d, J=6.6Hz), 7.73(1H, d, J=8.1Hz), 7.51-7.44(1H, m), 7.35-7.27(3H, m), 7.19(1H, m), 5.27(2H, s), 4.38(1H, brs), 4.19(1H, dd, J=8.1Hz, J=4.2Hz), 4.03(1H, dd, J=8.1Hz, J=4.2Hz), 3.49(2H, dd, J=9.3Hz, J=4.5Hz), 3.09-3.05(1H, m), 2.95-2.90(1H, m), 2.84-2.82(1H, m), 2.47-2.40(1H, m), 2.29-2.22(1H, m), 1.91-1.85(1H, m), 1.73-1.57(3H, m)
V-47	-Cl		9.94(1H, s), 8.66(1H, brs), 8.60(1H, s), 8.43(1H, s), 8.12(1H, d, J=6.6Hz), 8.01(1H, d, J=1.2Hz), 7.79(1H, d, J=6.9Hz), 7.72(1H, d, J=6.6Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=6.0Hz), 5.27(2H, s), 4.97(1H, m), 3.74(2H, s), 3.68(2H, t-like, J=5.1Hz), 3.37(2H, t-like, J=5.1Hz)
V-48	-Cl		9.95(1H, s), 8.64(1H, brs), 8.60(1H, s), 8.39(1H, s), 8.09(1H, d, J=6.6Hz), 8.01(1H, brs), 7.79(1H, d, J=6.3Hz), 7.73(1H, d, J=6.6Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=6.6Hz), 5.26(2H, s), 4.92(1H, m), 4.42(1H, brs), 3.62(2H, t, J=5.4Hz), 3.14(2H, t, J=5.4Hz), 2.54(2H, t, J=4.5Hz)
V-49	-Cl		9.94(1H, s), 8.63(1H, brs), 8.60(1H, s), 8.32(1H, s), 8.11(1H, d, J=6.6Hz), 8.01(1H, d, J=1.8Hz), 7.79(1H, d, J=6.6Hz), 7.72(1H, dd, J=6.9Hz, J=1.8Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=6.3Hz), 5.26(2H, s), 4.95(1H, brs), 3.87(2H, s), 2.91-2.81(3H, m), 2.57-2.51(1H, m), 2.30-2.21(1H, m), 2.00-1.91(1H, m)
V-50	-Cl		9.95(1H, s), 8.62(1H, brs), 8.59(1H, s), 8.32(1H, s), 8.10(1H, d, J=6.3Hz), 8.00(1H, brs), 7.79(1H, d, J=6.3Hz), 7.72(1H, d, J=6.3Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=6.0Hz), 5.26(2H, s), 4.89(1H, brs), 4.45(1H, brs), 3.49(3H, m), 2.82-2.75(3H, m), 2.41-2.39(1H, m), 2.17(1H, m), 1.86(1H, brs)

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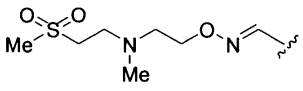
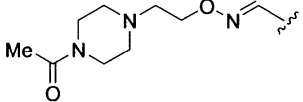
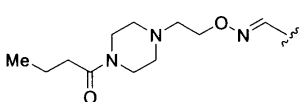
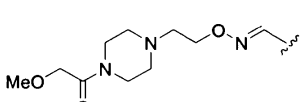
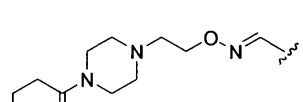
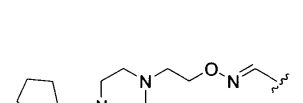
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【 0 0 6 6 】

【表 10】

化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-51	-Cl		9.94(1H, s), 8.64(1H, s), 8.60(1H, s), 8.33(1H, s), 8.12(1H, d, J=10.2Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, dd, J=8.7Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.34(3H, m), 7.16-7.22(1H, m), 5.26(2H, s), 4.29(2H, t, J=5.7Hz), 3.27(2H, t, J=7.2Hz), 3.02(3H, s), 2.84(2H, t, J=6.9Hz), 2.75(2H, t, J=5.7Hz), 2.29(3H, s)
V-52	-Cl		9.94(1H, s), 8.64(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, dd, J=8.7Hz, 1.5Hz), 8.00(1H, d, J=2.7Hz), 7.78(1H, d, J=8.7Hz), 7.71(1H, dd, J=8.7Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.21(1H, m), 5.26(2H, s), 4.32(2H, t, J=5.7Hz), 3.42-3.43(4H, m), 2.71(2H, t, J=5.7Hz), 2.41(4H, t, J=5.1Hz), 1.98(3H, s)
V-53	-Cl		9.95(1H, s), 8.63(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, dd, J=8.4Hz, 1.2Hz), 8.00(d, 1H, J=2.4Hz), 7.79(1H, d, J=8.4Hz), 7.71(1H, dd, J=9.3Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.34(3H, m), 7.15-7.22(1H, m), 5.26(2H, s), 4.32(2H, t, J=5.4Hz), 3.43-3.45(4H, m), 2.70(2H, t, J=5.7Hz), 2.41-2.46(4H, m), 2.26(2H, t, J=7.2Hz), 1.43-1.56(2H, m), 0.87(3H, t, J=7.5Hz)
V-54	-Cl		9.94(1H, s), 8.63(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, dd, J=8.7Hz, 1.5Hz), 8.00(1H, d, J=2.4Hz), 7.78(1H, d, J=8.7Hz), 7.71(1H, dd, J=9.0Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.21(1H, m), 5.26(2H, s), 4.32(2H, t, J=5.7Hz), 4.06(3H, s), 3.37-3.42(4H, m), 3.27(2H, s), 2.71(2H, t, J=5.7Hz), 2.46(4H, m)
V-55	-Cl		9.94(1H, s), 8.63(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, dd, J=8.7Hz, 1.5Hz), 8.00(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.71(1H, dd, J=9.0Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.21(1H, m), 5.26(2H, s), 4.32(2H, t, J=5.7Hz), 3.53(2H, t, J=6.6Hz), 3.43-3.47(4H, m), 3.21(3H, s), 2.71(2H, t, J=6.0Hz), 2.55(2H, t, J=6.6Hz), 2.42-2.50(4H, m)
V-56	-Cl		9.94(1H, s), 8.64(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, dd, J=8.7Hz, 1.5Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=9.0Hz), 7.71(1H, dd, J=8.7Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.15-7.22(1H, m), 5.26(2H, s), 4.61-4.66(1H, m), 4.32(2H, t, J=5.7Hz), 3.68-3.80(2H, m), 3.44-3.54(4H, m), 2.71(2H, 5.7Hz), 2.44-2.50(4H, m), 1.95-2.04(2H, m), 1.75-1.86(2H, m)

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【 0 0 6 7 】

【表 1 1】

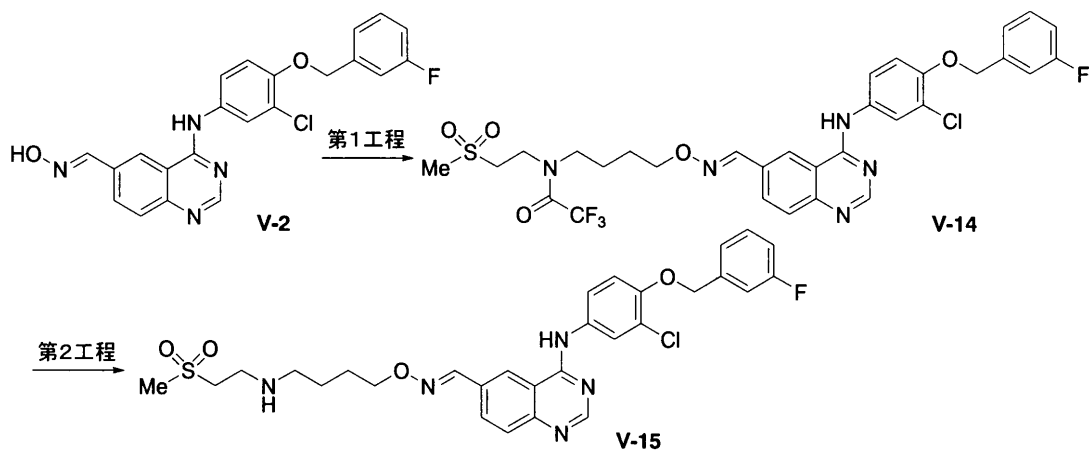
化合物 No.	R <sup>A</sup>	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
V-57	-Cl		9.94(1H, s), 8.64(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, dd, J=8.7Hz, 1.2Hz), 8.00(1H, d, J=2.7Hz), 7.79(1H, d, J=8.7Hz), 7.71(1H, dd, J=9.0Hz, 2.4Hz), 7.44-7.51(1H, m), 7.26-7.35(3H, m), 7.16-7.21(1H, m), 5.26(2H, s), 4.45(2H, s), 4.32(2H, t, J=5.4Hz), 3.49-3.53(4H, m), 3.09(3H, s), 2.72(2H, t, J=5.7Hz), 2.44-2.50(4H, m)
V-58	-Cl		9.95(1H, s), 8.64(1H, s), 8.59(1H, s), 8.35(1H, s), 8.10(1H, d, J=8.7Hz), 8.00(1H, d, J=2.7Hz), 7.78(1H, d, J=8.7Hz), 7.69-7.73(1H, m), 7.44-7.51(1H, m), 7.26-7.34(3H, m), 7.16-7.22(1H, m), 5.26(2H, s), 4.31(2H, t, J=5.7Hz), 3.12(4H, t, J=5.1Hz), 2.87(3H, s), 2.75(2H, t, J=5.4Hz), 2.58(4H, t, J=5.3Hz)

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【 0 0 6 8 】

実施例 6

【化 6 7】



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(第 1 工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(4-(N-(2-(メタンシルホニル)エチル)-N-(2,2,2-トリフルオロアセチル)アミノ)ブチルオキシイミノ)キナゾリン(V-14)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(ヒドロキシイミノ)キナゾリン(V-2, 84mg)およびN-(4-プロモブチル)-N-(2-メタンシルホニルエチル)-2,2,2-トリフルオロアセタミド(71mg)をN,N-ジメチルアセタミド(1.5ml)に溶かし、炭酸カリウム(41mg)を加えて、60 で13時間加熱攪拌した。その後、反応液を0.5Mクエン酸中に注ぎ、酢酸エチルで抽出した。有機層を水、食塩水で洗浄後、硫酸マグネシウム上で乾燥した。粗成績体をシリカゲルクロマトグラフィー(ヘキサン:酢酸エチル=1:4で溶出)により精製し、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(4-(N-(2-(メタンシルホニル)エチル)-2,2,2-トリフルオロアセタミド)ブチルオキシイミノ)キナゾリン(V-14, 41mg)を淡黄色固体として得た。

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<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 9.94(1H, s), 8.63(1H, s), 8.59(1H, s), 8.33(1H, s), 8.10(1H, dd, J=8.4Hz, J=1.5Hz), 8.00(1H, d, J=2.4Hz), 7.78(1H, d, J=8.7Hz), 7.71(1H, dd, J=2.3Hz, J=8.7Hz), 7.47(1H, dt, J=6.0Hz, J=8.1Hz), 7.36-7.31(2H, m), 7.28(1H, d, J=9.0Hz), 7.18(1H, dt, J=2.0Hz, J=8.6Hz), 5.26(2H, s), 4.26-4.18(2H, m), 3.79(2H, t, J=7.1Hz), 3.60-3.44(4H, m), 3.05(3H, s), 1.78-1.66(4H, m).

【 0 0 6 9 】

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(第2工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(4-(2-(メタンシルホニル)エチルアミノ)ブチルオキシイミノ)キナゾリン(V-15)の合成

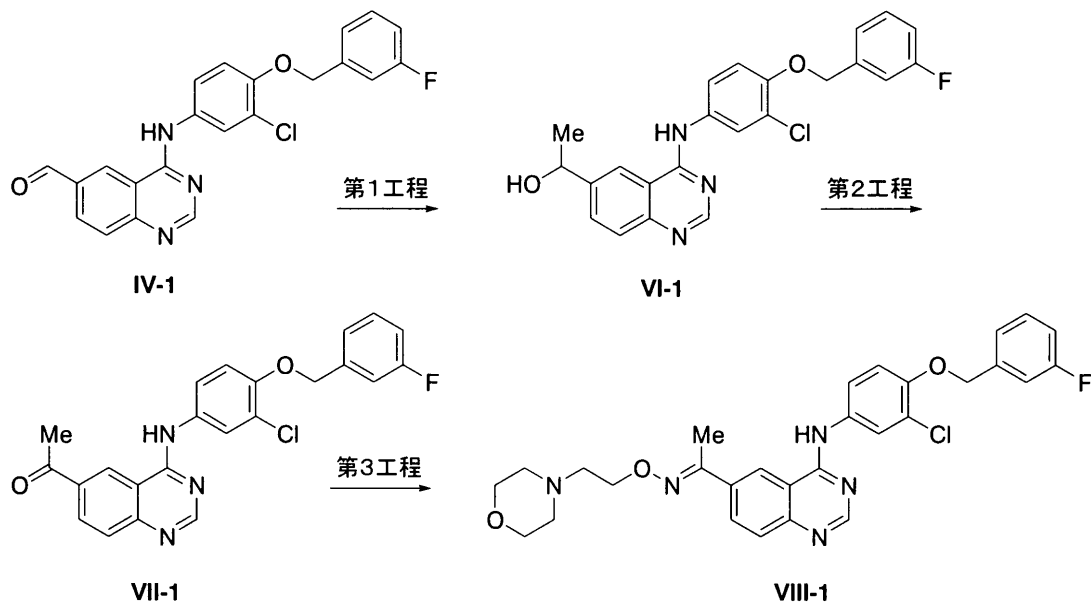
4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(4-(N-(2-(メタンシルホニル)エチル)-2,2,2-トリフルオロアセタミド)ブチルオキシイミノ)キナゾリン(V-14, 40mg)をテトラヒドロフランとメタノールの混液(1:1)2.6mlに溶かし、1mol/L水酸化ナトリウム水溶液86 $\mu$ lを加えて、室温で14時間、40 $^{\circ}$ Cで1時間攪拌した。反応後、酢酸エチルで抽出し、有機層を脱水し、濾液を濃縮した。濃縮残渣を、アミノカラムを用いたクロマトグラフィー(0~3%メタノール含有酢酸エチルで溶出)により精製、得られた淡黄色の油状物質をヘキサンと酢酸エチルの混液で粉末化し、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(4-(2-(メタンシルホニル)エチルアミノ)ブチルオキシイミノ)キナゾリン(V-15, 12mg)を淡黄色固体として得た。

$^1\text{H NMR}$ ( $d_6$ -DM SO,  $\delta$ ): 9.95(1H, s), 8.63(1H, s), 8.59(1H, s), 8.33(1H, s), 8.11(1H, d, J=8.7Hz), 8.01(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.72(1H, br d, J=9.0Hz), 7.51-7.44(1H, m), 7.36-7.31(2H, m), 7.28(1H, d, J=9.0Hz), 7.19(1H, br t, J=9Hz), 5.26(2H, s), 4.20(2H, t, J=6.6Hz), 3.21(2H, t, J=6.9Hz), 2.91(2H, t, J=6.6Hz), 2.56(2H, t, J=6.9Hz), 1.73(2H, quin, J=7Hz), 1.51(2H, quin, J=7Hz).

【0070】

実施例7

【化68】



(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-ヒドロキシエタン-1-イル)キナゾリン(VI-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ホルミルキナゾリン(V-1) 620mgをテトラヒドロフラン30mLに溶解後、窒素雰囲気下0 $^{\circ}$ Cに冷却し、臭化メチルマグネシウム(0.93M テトラヒドロフラン溶液)を4.9mL滴下した。窒素雰囲気下0 $^{\circ}$ Cで1時間反応後、水と酢酸エチルを加え分液操作を行った。有機層を水、飽和食塩水の順で洗浄後、硫酸マグネシウム上で乾燥した。濃縮後、得られた結晶をクロロホルムで洗浄して4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-ヒドロキシエタン-1-イル)キナゾリン(VI-1)330mgを得た。

$^1\text{H NMR}$ ( $d_6$ -DMSO,  $\delta$ ): 9.81(1H, s), 8.55(1H, s), 8.44(1H, s), 8.03(1H, d, J=2.4Hz), 7.86(1H, dd, J=8.7Hz, J=1.2Hz), 7.78-7.73(2H, m), 7.51-7.44(1H, m), 7.35-7.25(3H, m), 7.18(1H, dt, J=8.4Hz, J=2.4Hz), 5.44(1H, d, J=3.9Hz), 5.26(2H, s), 4.93(1H, m), 1.45(3H, d, J=6.6Hz)

【0071】

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(第2工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(エタン-1-オン)キナゾリン(VII-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-ヒドロキシエタン-1-イル)キナゾリン(VI-1) 360mgをテトラヒドロフラン150mLに溶かし、二酸化マンガン3.7gを加え室温で終夜攪拌した。反応後、無機物をろ去してろ液を濃縮した。得られた結晶をクロロホルムで洗浄して4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(エタン-1-オン)キナゾリン(VII-1) 320mgを得た。

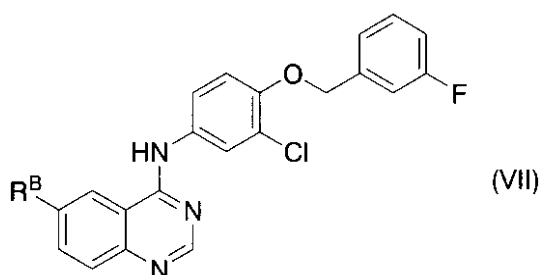
$^1\text{H NMR}$  ( $d_6$ -DMSO): 10.18(1H, s), 9.16(1H, d,  $J=1.5\text{Hz}$ ), 8.64(1H, s), 8.31(1H, d,  $J=8.4\text{Hz}$ ,  $J=2.4\text{Hz}$ ), 7.97(1H, d,  $J=2.4\text{Hz}$ ), 7.83(1H, d,  $J=8.7\text{Hz}$ ), 7.71(1H, dd,  $J=8.7\text{Hz}$ ,  $J=2.4\text{Hz}$ ), 7.51-7.44(1H, m), 7.35-7.29(3H, m), 7.19(1H, dt,  $J=8.1\text{Hz}$ ,  $J=2.7\text{Hz}$ ), 5.27(2H, s), 2.74(3H, s)

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【0072】

上記第1工程および第2工程と同様の方法で下記の化合物を合成した。

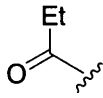
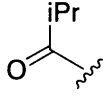
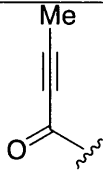
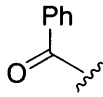
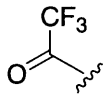
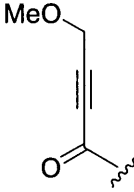
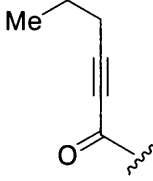
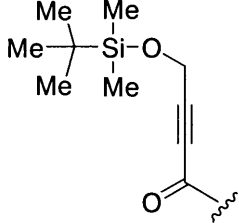
【化69】



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【0073】

【表 1 2】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VII-2		10.17(1H, s), 9.17(1H, d, J=1.5Hz), 8.64(1H, s), 8.32(1H, dd, J=8.7Hz, J=1.5Hz), 7.97(1H, d, J=2.7Hz), 7.84(1H, d, J=8.7Hz), 7.72(1H, dd, J=8.7Hz, J=2.7Hz), 7.52-7.44(1H, m), 7.35-7.29(3H, m), 7.20(1H, dt, J=8.7Hz, J=2.7Hz), 5.28(2H, s), 3.22(2H, q, J=7.2Hz), 1.17(3H, t, J=7.2Hz)
VII-3		(CDCl <sub>3</sub> ) 8.76(1H, s), 8.65(1H, brs), 8.29(1H, d, J=9.0Hz), 7.97(1H, d, J=8.4Hz), 7.88(1H, brs), 7.52(1H, d, J=9.0Hz), 7.40-7.33(1H, m), 7.25-7.19(3H, m), 7.05-6.97(2H, m), 5.16(2H, s), 3.72-3.63(1H, m), 1.28(3H, d, J=1.8Hz), 1.26(3H, d, J=1.8Hz)
VII-4		10.34(1H, s), 9.23(1H, s), 8.66(1H, s), 8.44(1H, dd, J=9 Hz, J=1.5Hz), 7.99(1H, d, J=2.7Hz), 7.88(1H, d, J=8.7Hz), 7.72(1H, dd, J=9 Hz, J=2.4Hz), 7.43-7.52(1H, m), 7.14-7.36(4H, m), 5.28(2H, s), 2.28(3H, s)
VII-5		10.14(1H, s), 8.98(1H, s), 8.67(1H, s), 8.11(1H, dd, J=8.7 Hz, J=0.9Hz), 7.98-7.82(4H, m), 7.78-7.58(4H, m), 7.52-7.43(1H, m), 7.36-7.14(4H, m), 5.26(2H, s)
VII-6		10.50(1H, s), 9.27(1H, s), 8.67(1H, s), 8.35(1H, d, J=9.3Hz), 7.99(1H, d, J=2.7Hz), 7.88(1H, brs), 7.70(1H, d, J=7.5Hz), 7.52-7.43(1H, m), 7.36-7.14(4H, m), 5.28(2H, s)
VII-8		10.36(1H, s), 9.26(1H, s), 8.67(1H, s), 8.44(1H, dd, J=8.7Hz, J=1.8Hz), 8.00(1H, d, J=2.4Hz), 7.91(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz, J=2.4Hz), 7.51-7.44(1H, m), 7.35-7.28(3H, m), 7.22-7.15(1H, m), 5.27(2H, s), 4.55(2H, s).
VII-9		10.4(1H, s), 9.24(1H, s), 8.67(1H, s), 8.46(1H, d, J=8.7Hz), 8.01(1H, d, J=2.7Hz), 7.90(1H, d, J=8.7Hz), 7.70-7.80(1H, m), 7.44-7.54(1H, m), 7.15-7.40(4H, m), 5.27(2H, s), 2.64(2H, t, J=6.9Hz), 1.63-1.75(2H, m), 1.05(3H, t, J=7.5Hz)
VII-10		10.34(1H, s), 9.24(1H, s), 8.67(1H, s), 8.42(1H, d, J=8.4Hz), 7.99(1H, s), 7.88(1H, d, J=8.7Hz), 7.73(1H, d, J=9.0Hz), 7.48-7.44(1H, m), 7.34-7.27(3H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.76(2H, s), 0.94(9H, s), 0.16(6H, s).

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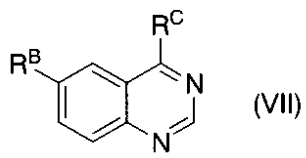
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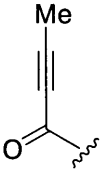
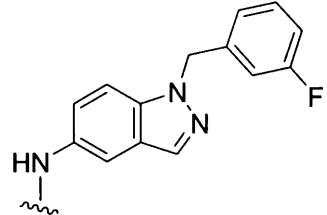
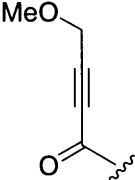
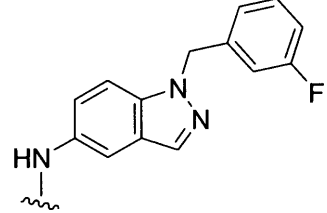
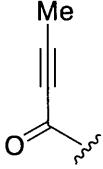
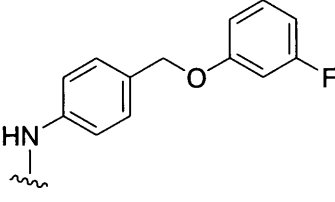
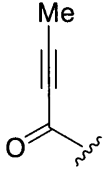
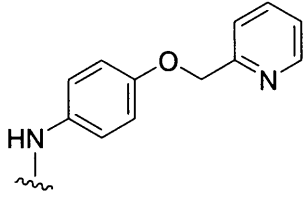
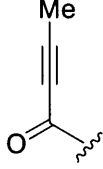
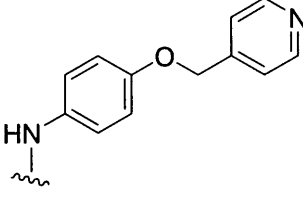
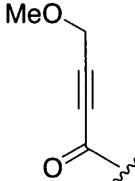
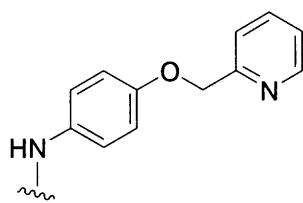


【化 7 0】



【 0 0 7 5】

【表 1 3】

化合物 No.	R <sup>B</sup>	R <sup>C</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VII-11			10.44(1H, s), 9.24(1H, s), 8.60(1H, s), 8.43(1H, dd, J=2.4, 11.6Hz), 8.17(2H, s), 7.87(1H, d, J=12.0Hz), 7.78-7.67(2H, m), 7.41-7.34(1H, m), 7.14-7.04(3H, m), 5.72(2H, s), 2.28(3H, s)
VII-12			10.47(1H, s), 9.29(1H, d, J=1.8Hz), 8.62(1H, s), 8.43(1H, dd, J=1.5Hz, J=9.0Hz), 8.21-8.17(2H, m), 7.89(1H, d, J=8.7Hz), 7.78-7.68(2H, m), 7.41-7.33(1H, m), 7.15-7.04(3H, m), 5.71(2H, s), 4.55(2H, s), 3.42(3H, s).
VII-13			10.44(1H, s), 9.25(1H, s), 8.73(1H, s), 8.44(1H, d, J=8.8Hz), 8.16(1H, s), 7.89(1H, d, J=8.4Hz), 7.62(1H, d, J=8.4, 16.0Hz), 6.95-6.86(2H, m), 6.82-6.77(1H, m), 5.15(2H, s), 2.27(3H, s).
VII-14			10.34(1H, s), 9.22(1H, s), 8.64-8.59(2H, m), 8.43-8.41(1H, m), 7.98(1H, s), 7.87-7.85(2H, m), 7.71-7.68(1H, m), 7.59-7.58(1H, m), 7.40-7.34(1H, m), 7.29-7.27(1H, m), 5.30(2H, s).
VII-15			9.21(1H, s), 8.64-8.58(2H, m), 8.45-8.39(1H, m), 7.97(1H, s), 7.88-7.84(1H, m), 7.71-7.66(1H, m), 7.45-7.51(1H, m), 7.27-7.23(1H, m), 7.29-7.27(1H, m), 5.30(2H, s).
VII-16			10.35(1H, s), 9.24(1H, s), 8.67(1H, s), 8.60(1H, d, J=4.4Hz), 8.42(1H, d, J=8.8Hz), 8.01(1H, s), 7.91-7.87(2H, m), 7.73-7.71(1H, m), 7.59(1H, d, J=8.0Hz), 7.39-7.36(1H, m), 7.29(1H, d, J=8.8Hz), 5.31(2H, s), 4.55(2H, s), 3.42(3H, s).

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【表 1 4】

化合物 No.	R <sup>B</sup>	R <sup>C</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VII-17			10.32 (1H, s), 8.85(1H, s), 8.74(1H, s), 8.20(2H, d, J=6.0Hz), 8.01-7.96(4H, m), 7.87(1H, d, J=6.0Hz), 7.68-7.64 (3H, m), 3.56(3H, s).
VII-18			10.52(1H, s), 9.14(1H, s), 8.60(1H, s), 8.31(1H, d, J=8.4Hz), 8.40(2H, d, J=8.4Hz), 7.84-7.78(5H, m), 7.52-7.43(3H, m), 4.37(2H, s), 3.24(3H, s).
VII-19			10.44(1H, s), 9.28(1H, s), 8.70(1H, s), 8.45(1H, d, J=8.8Hz), 7.92(3H, t, J=8.8Hz), 7.46(2H, d, J=8.8Hz), 7.37 (2H, t, J=7.6Hz), 7.30-7.26(3H, m), 4.24-4.20(2H, m), 2.28 (3H, s).

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## 【 0 0 7 7 】

(第3工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-(1-モルホリノ)エトキシイミノ)エチル)キナゾリン(VIII-1)の合成

化合物VII-1 100mgと2-(1-モルホリノ)エトキシアミン 2 塩酸塩78mgをテトラヒドロフランと水の混液6mL(20:1)に溶かし、室温で終夜反応した。反応液を酢酸エチルで薄め、飽和炭酸水素ナトリウム水溶液で洗浄後、硫酸マグネシウム上で乾燥した。濃縮後、ヘキサンと酢酸エチルの混液(2:1)で固体を洗浄して、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-(1-モルホリノ)エトキシイミノ)エチル)キナゾリン(VIII-1)を72mg得た。

<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 9.93(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.22(1H, d, J=6.6 Hz), 7.96(1H, d, J=1.8Hz), 7.76(1H, d, J=6.6Hz), 7.70(1H, dd, J=6.6Hz, J=1.8Hz), 7.51-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.0Hz), 5.27(2H, s), 4.33(2H, t, J=6.0Hz), 3.59(4H, t, J=4.5Hz), 2.69(2H, t, J=6.0Hz), 2.47(4H, t, J=4.5Hz), 2.36(3H, s)

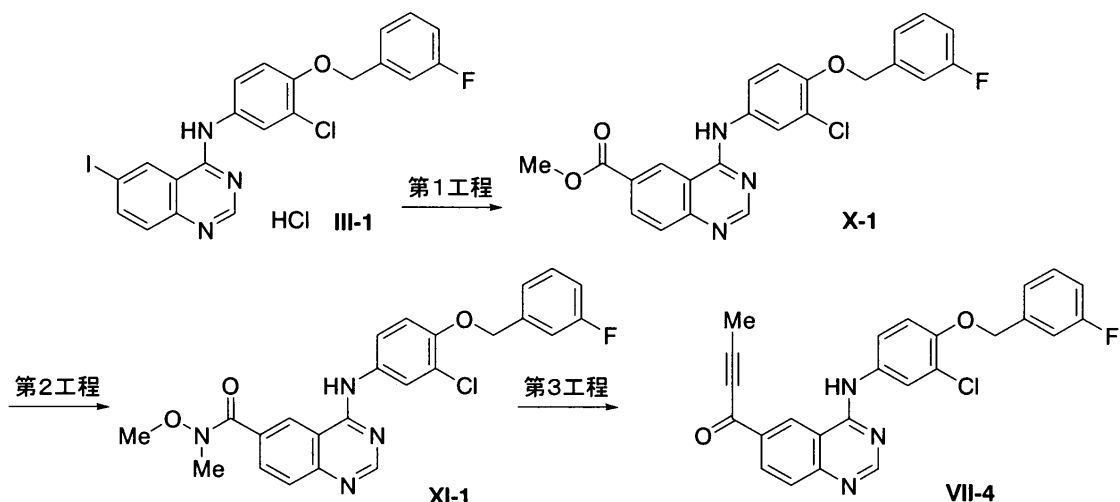
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## 【 0 0 7 8 】

## 参考例 1

ケトン誘導体(VII)を得る方法として、実施例7の第1工程、第2工程の他に以下の方法が挙げられる。

## 【化71】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メチルエステル(X-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ヨードキナゾリン塩酸塩(III-1, 20g)とジクロロビス(トリフェニルホスフィン)パラジウム1.29gをジメチルホルムアミド200mlおよびメタノール200mlの混合液に溶かし、トリエチルアミン25.7mlを加えた。反応容器内を十分に一酸化炭素ガスで置換した後、一酸化炭素雰囲気下80℃で15時間反応した。反応液をセライトでろ過し、ろ液を攪拌しながら0℃に冷却した。析出物をろ取して冷メタノールで洗浄後乾燥することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メチルエステル(X-1, 13.5g)を無色固体として得た。

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$^1\text{H NMR}$ ( $d_6$ -DMSO,  $\delta$ ): 10.26(1H, s), 9.25(1H, d,  $J=1.5\text{Hz}$ ), 8.65(1H, s), 8.31(1H, d,  $J=1.5\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 8.02(1H, d,  $J=2.7\text{Hz}$ ), 7.85(1H, d,  $J=2.7\text{Hz}$ ), 7.77(1H, dd,  $J=2.7\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 7.49-7.44(1H, m), 7.35-7.27(3H, m), 7.20-7.16(1H, m), 5.27(2H, s), 3.96(3H, s)。

## 【0079】

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(第2工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(XI-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メチルエステル(X-1, 11.6g)をテトラヒドロフラン(230ml)に溶かし、N,N-ジメチルヒドロキシルアミン塩酸塩(7.8g)を加え、氷冷下2 mol/L  $i\text{PrMgCl}$ テトラヒドロフラン溶液(79.8ml)を30分かけて滴下した。滴下終了後20分間、氷冷下で攪拌し、反応終了後に飽和塩化アンモニウム水溶液200mlを加えた。混合液に水100mlを加えた後、酢酸エチルで抽出し、有機層を硫酸ナトリウム上で乾燥した。ろ液を濃縮することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(XI-1, 12.7g)を淡黄色固体として得た。

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$^1\text{H NMR}$ ( $d_6$ -DMSO,  $\delta$ ): 10.01(1H, s), 8.79(1H, d,  $J=1.5\text{Hz}$ ), 8.64(1H, s), 8.05(1H, d,  $J=2.7\text{Hz}$ ), 8.00(1H, dd,  $J=1.5\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 7.81(1H, d,  $J=8.7\text{Hz}$ ), 7.76(1H, dd,  $J=2.7\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 7.52-7.44(1H, m), 7.35-7.26(3H, m), 7.22-7.16(1H, m), 5.27(2H, s), 3.57(3H, s), 3.34(3H, s)。

## 【0080】

(第3工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン(VII-4)の合成

-78℃に冷却したN,N-ジイソプロピルアミン4.22mlのテトラヒドロフラン(25ml)溶液に1.6 mol/L  $n\text{-BuLi}$ ヘキサン溶液18.8mlを滴下した。滴下終了後0℃まで昇温して0℃で20分間攪拌して再び-78℃まで冷却した。2-ブロモ-1-プロペン1.33mlのテトラヒドロフラン(25ml)

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1) 溶液を加え、-78℃で2時間攪拌した。この溶液に4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド (XI-1, 2.33g) のテトラヒドロフラン (50ml) 溶液を滴下し、滴下終了後に0℃まで昇温し3時間攪拌した。反応終了後に反応液を氷水中に注ぎ2mol/L 塩酸水溶液で中和後、酢酸エチルで抽出した。有機層を硫酸ナトリウム上で乾燥し、ろ液を濃縮して得られた結晶性残渣を塩化メチレンで洗浄することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン (VII-4, 1.71g) を黄色固体として得た。

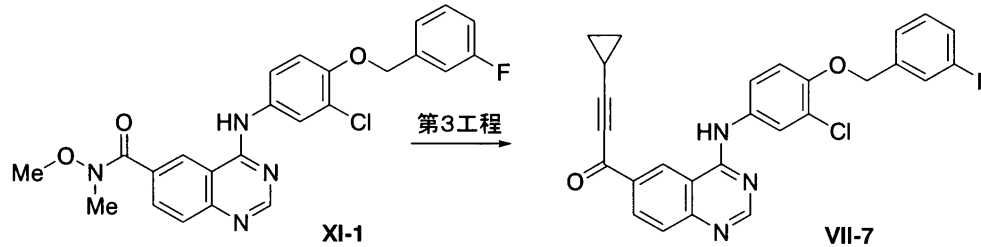
【0081】

参考例 2

参考例 1 の第 3 工程の他の方法として、以下に示す方法が挙げられる。

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【化 7 2】



(第 3 工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-オキソ-2-プロピン-3-シクロプロピル-1-イル)キナゾリン (VII-7) の合成

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-78℃に冷却したエチニルシクロプロパン0.16mlのテトラヒドロフラン(3ml)溶液に1.54mol/L n-BuLiヘキサン溶液1.25mlを滴下した。滴下終了後、-78℃で1時間攪拌した。この溶液に4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド (XI-1, 300mg) のテトラヒドロフラン(3ml)溶液を滴下し、滴下終了後に0℃まで昇温し3時間攪拌した。反応終了後に反応液に水10ml、酢酸エチル10mlを加え、分液操作を行った。水層を酢酸エチル10mlで2回再抽出後、すべての有機層を合わせて硫酸ナトリウム上で乾燥し、ろ液を濃縮した。残渣をシリカゲルカラムクロマトグラフィーで精製することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-オキソ-2-プロピン-3-シクロプロピル-1-イル)キナゾリン (VII-7, 265mg) を黄色固体として得た。

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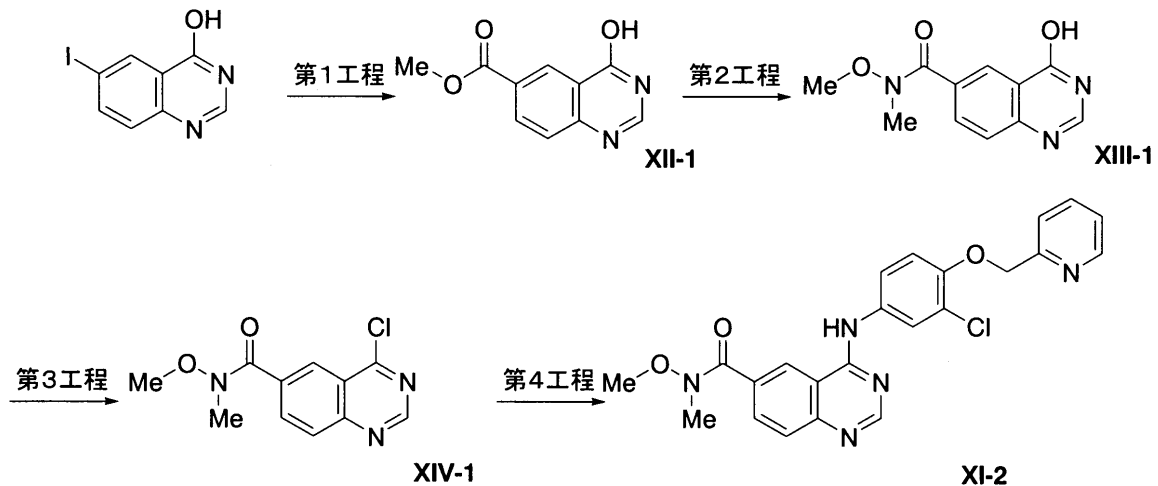
$^1\text{H NMR}$ ( $d_6$ -DMSO,  $\delta$ ): 10.4(1H, s), 9.21(1H, s), 8.67(1H, s), 8.41(1H, d,  $J=8.7\text{Hz}$ ), 8.01(1H, d,  $J=2.7\text{Hz}$ ), 7.88(1H, d,  $J=8.7\text{Hz}$ ), 7.73(1H, dd,  $J=8.7\text{Hz}$ ,  $J=3\text{Hz}$ ), 7.40-7.52(1H, m), 7.15-7.4(4H, m), 5.28(2H, s), 1.77-1.87(1H, m), 1.05-1.20(3H, m).

【0082】

参考例 3

アミド誘導体 (XI) を得る方法として、参考例 1 の第 1 工程、第 2 工程の他に以下の方法が挙げられる。

## 【化73】



(第1工程) 4-ヒドロキシキナゾリン-6-カルボン酸メチルエステル(XII-1)の合成  
 4-ヒドロキシ-6-ヨードキナゾリン(250mg)と触媒量の酢酸パラジウム(II)および1,3-ビス(ジフェニルホスフィノ)プロパンをN,N-ジメチルホルムアミド(3.8ml)、メタノール(1.5ml)に溶かし、トリエチルアミン1.5mlを加えた。反応容器内を十分に一酸化炭素ガスで置換した後、一酸化炭素雰囲気下80℃で20時間反応した。反応終了後、10%クエン酸水溶液に注ぎ、酢酸エチルで抽出した。有機層を水で洗浄後、硫酸ナトリウム上で乾燥し、ろ液を濃縮した。得られた残渣をシリカゲルカラムクロマトグラフィー(酢酸エチルで溶出)により精製し、4-ヒドロキシキナゾリン-6-カルボン酸メチルエステル(XII-1, 157mg)を褐色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \delta): 12.48(1\text{H}, \text{s}), 8.63(1\text{H}, \text{s}), 8.26(1\text{H}, \text{d}, J=8.4\text{Hz}), 8.19(1\text{H}, \text{s}), 7.73(1\text{H}, \text{d}, J=8.4\text{Hz})$ .

## 【0083】

(第2工程) 4-ヒドロキシキナゾリン-6-カルボン酸メトキシメチルアミド(XIII-1)の合成

4-ヒドロキシキナゾリン-6-カルボン酸メチルエステル(XII-1, 4.0g)をテトラヒドロフラン(120ml)に溶かし、N,O-ジメチルヒドロキシルアミン塩酸塩(5.7g)を加え、氷冷下2mol/L iPrMgClテトラヒドロフラン溶液(59ml)を30分かけて滴下した。滴下終了後30分間、氷冷下で攪拌し、反応終了後に2mol/L塩酸40mlを滴下した。混合液を飽和炭酸水素ナトリウム水溶液で中和後、クロロホルムで抽出し、有機層を食塩水で洗浄後に硫酸ナトリウム上で乾燥した。濃縮後、結晶性残渣をシリカゲルカラムクロマトグラフィー(酢酸エチル:メタノール=9:1で溶出)により精製して4-ヒドロキシキナゾリン-6-カルボン酸メトキシメチルアミド(XIII-1, 2.9g)を白色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \delta): 12.42(1\text{H}, \text{s}), 8.36(1\text{H}, \text{s}), 8.18(1\text{H}, \text{s}), 8.02(1\text{H}, \text{d}, J=8.4\text{Hz}), 7.73(1\text{H}, \text{d}, J=8.4\text{Hz}), 3.57(3\text{H}, \text{s}), 3.32(3\text{H}, \text{s})$ .

## 【0084】

(第3工程) 4-クロロキナゾリン-6-カルボン酸メトキシメチルアミド(XIV-1)の合成

4-ヒドロキシキナゾリン-6-カルボン酸メトキシメチルアミド(XIII-1, 2.9g)をトルエン(57ml)に懸濁させ、トリエチルアミン6.8mlおよびオキシ塩化リン3.4mlを加え、130℃で30分間攪拌した。反応終了後に室温まで冷却後、飽和炭酸水素ナトリウム水溶液50mlおよび酢酸エチルを加えた。水層がpH8を示すまで炭酸水素ナトリウムを加え、酢酸エチルで抽出した。有機層を飽和炭酸水素ナトリウム水溶液で洗浄後に硫酸ナトリウム上で乾燥した。濃縮後、結晶性残渣をジエチルエーテルにより洗浄後、乾燥して4-クロロキナゾリン-6-カルボン酸メトキシメチルアミド(XIV-1, 2.3g)を白色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \delta): 8.75(1\text{H}, \text{s}), 8.36(1\text{H}, \text{s}), 8.11(1\text{H}, \text{dd}, J=2.0\text{Hz}, J=8.4\text{Hz}),$

7.87(1H, d, J=8.4Hz), 3.56(3H, s), 3.32(3H, s).

【 0 0 8 5 】

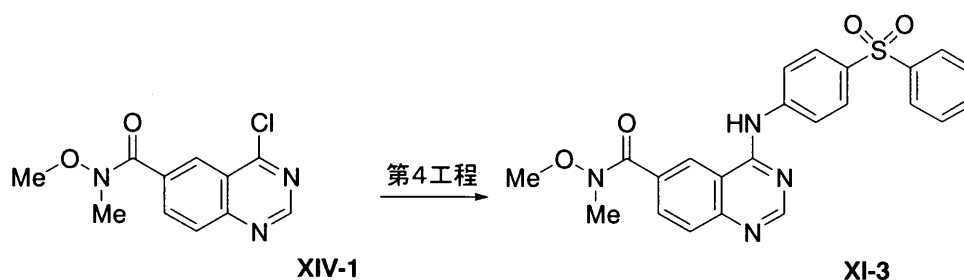
(第4工程) 4-(3-クロロ-4-(ピリジン-2-イルメトキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド (X I - 2) の合成

4-クロロナゾリン-6-カルボン酸メトキシメチルアミド(X IV - 1, 5.78g)をテトラヒドロフラン(100ml)に溶解させ、3-クロロ-4-(ピリジン-2-イルメトキシ)アニリン5.39gを加え、60 で1時間攪拌した。析出物を濾取し、テトラヒドロフランで洗浄することにより4-(3-クロロ-4-(ピリジン-2-イルメトキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(X I - 2, 7.87g)を白色固体として得た。濾液を室温で静置して得られた析出物を濾取することにより新たにX I - 2を2.21g得た。

<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 10.02(1H, s), 8.79(1H, s), 8.62-8.59(2H, m), 8.04(1H, s), 8.00-7.98(1H, m), 7.90-7.86(1H, s), 7.80-7.78(1H, m), 7.75-7.73(1H, m), 7.59-7.57(1H, m), 7.37-7.35(1H, m), 7.28-7.26(1H, m), 5.29(2H, s), 3.57(3H, s).

【 0 0 8 6 】

【化74】



参考例 4

参考例 3 の第 4 工程の他の方法として、以下に示す方法が挙げられる。

(第4工程) 4-(4-ベンゼンスルホニルフェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド (X I - 3) の合成

4-クロロナゾリン-6-カルボン酸メトキシメチルアミド(X IV - 1, 550mg)、4-ベンゼンスルホニルフェニルアミン611mg、酢酸パラジウム(II) 49mg、Xant phos 190mgおよび炭酸セシウム997mgを1,4-ジオキサン(30ml)に加え、反応容器内を窒素ガスで置換後、100 で2.5時間攪拌した。反応溶液をセライトろ過して、ろ液に水を加えて酢酸エチルで抽出した。有機層を飽和食塩水で洗浄後に硫酸ナトリウム上で乾燥した。ろ液を濃縮後、残渣をシリカゲルカラムクロマトグラフィー(酢酸エチル:メタノール=95:5で溶出)により精製して4-(4-ベンゼンスルホニルフェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(X I - 3, 651mg)を淡黄色固体として得た。

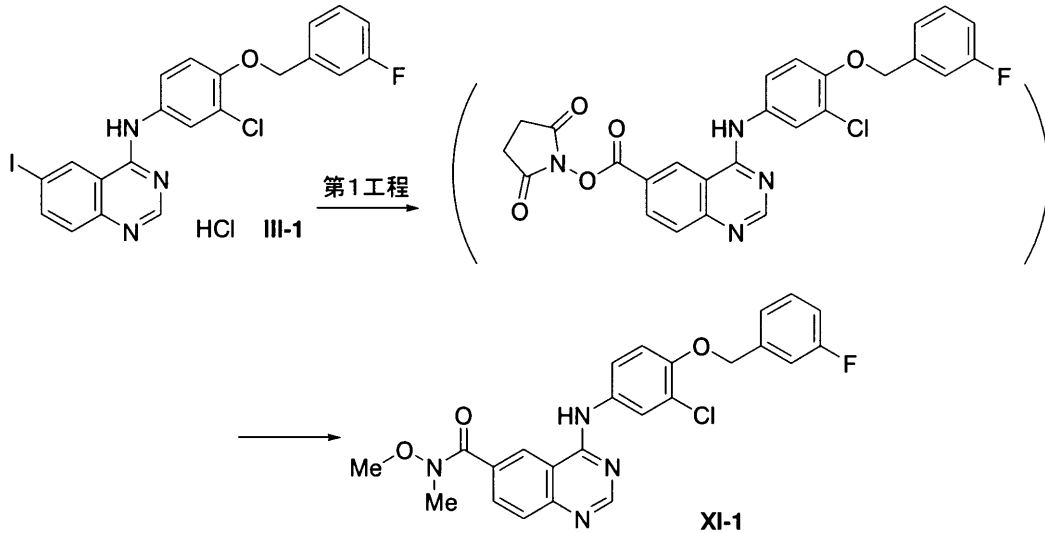
<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 10.66(1H, s), 9.29(1H, s), 8.80(1H, s), 8.48(1H, d, J=7.7Hz), 8.16(2H, d, J=7.7Hz), 7.99-7.94(5H, m), 7.80-7.61 (3H, m), 2.27 (3H, s).

【 0 0 8 7 】

参考例 5

アミド誘導体 (X I) を得る方法として、参考例 1 の第 1 工程、第 2 工程および参考例 3 の他にさらに以下の方法が挙げられる。

## 【化75】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(XI-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ヨードキナゾリン塩酸塩(III-1, 500mg)とジクロロビス(トリフェニルホスフィン)パラジウム32mg、N-ヒドロキシスクシンイミド191mgをジメチルホルムアミド10mlに溶かし、トリエチルアミン360 $\mu$ lを加えた。反応容器内を十分に一酸化炭素ガスで置換した後、一酸化炭素雰囲気下80 $^{\circ}$ Cで3時間反応した。反応液の五分の一に40%N, O-ジメチルヒドロキシルアミン水溶液0.3mlを加え室温で一夜攪拌した。反応終了後、反応液に水を加えて酢酸エチルで抽出した。有機層を水、飽和食塩水の順で洗浄し、硫酸ナトリウム上で乾燥した。ろ液の濃縮残渣をシリカゲルカラムクロマトグラフィー(酢酸エチルで溶出)により精製することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(XI-1, 74mg)を淡黄色固体として得た。

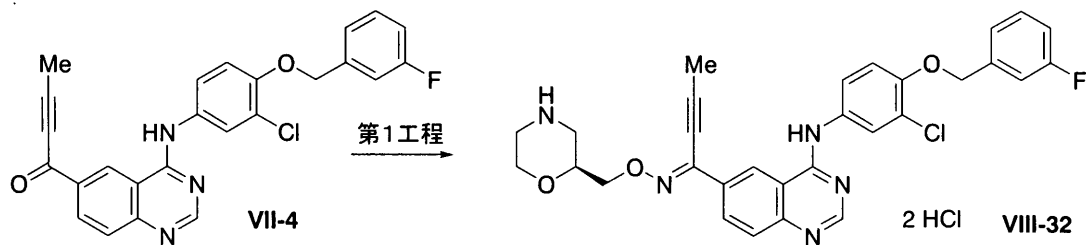
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## 【0088】

実施例8

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## 【化76】



(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-((S)-モルホリン-2-イルメトキシイミノ)-2-ブチン-1-イル)キナゾリン 二塩酸塩(VIII-32)の合成

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4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン(VII-4, 786mg)と(S)-2-アミノオキシメチル-モルホリン-4-カルボン酸 tert-ブチルエステル614mgの1,4-ジオキサン31ml懸濁液に、2mol/Lメタンスルホン酸水溶液2.21mlを加えて80 $^{\circ}$ Cで22時間攪拌した。2mol/Lメタンスルホン酸水溶液1.32mlを追加してさらに5.5時間攪拌した。反応終了後、反応液を氷-炭酸水素ナトリウム水溶液中に注ぎ、酢酸エチルで抽出した。水層を酢酸エチルで再抽出後、すべての有機層を合わせて水洗後、無水硫酸マグネシウム上で乾燥した。ろ液を濃縮後、残渣をシリカゲルカラムクロマトグラフィー(クロロホルム:メタノール=9:1で溶出)で精製することにより黄色油

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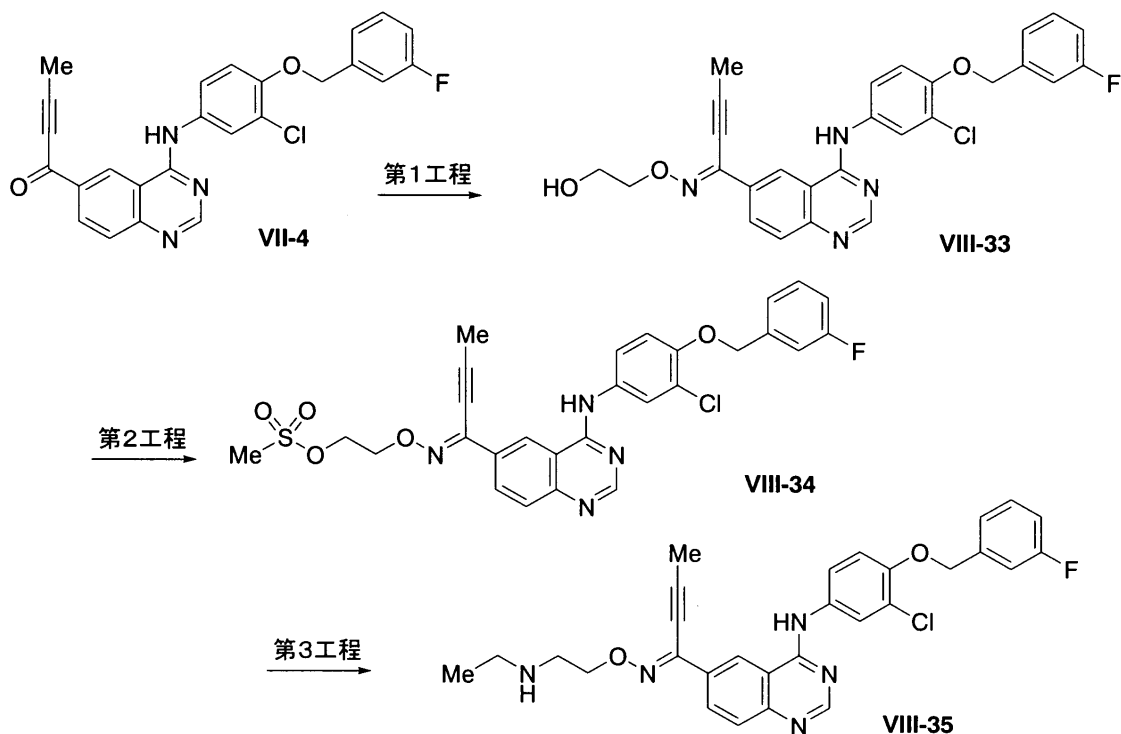
状物を得た。この油状物を酢酸エチル50mlに溶解させてる過後、攪拌しながら4mol/L塩酸-酢酸エチル溶液0.95mlを加え、室温で1時間攪拌した。析出物をろ取後、酢酸エチル、ヘキサンの順で洗浄した。ろ取物をメタノール-酢酸エチルにより再結晶をすることにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-((S)-モルホリン-2-イルメトキシイミノ)-2-ブチン-1-イル)キナゾリン 二塩酸塩 (VI II-32, 839mg)を黄色結晶として得た。

$^1\text{H NMR}$ ( $d_6$ -DMSO, ): 11.69(1H, bs), 9.49-9.37(2H, m), 9.05(1H, s), 8.88(1H, s), 8.38(1H, dd,  $J=1.5\text{Hz}$ ,  $J=8.7\text{Hz}$ ), 7.96(1H, d,  $J=8.7\text{Hz}$ ), 7.89(1H, d,  $J=2.7\text{Hz}$ ), 7.64(1H, dd,  $J=2.4\text{Hz}$ ,  $J=9.0\text{Hz}$ ), 7.52-7.45(1H, m), 7.36-7.30(3H, m), 7.23-7.16(1H, m), 5.31(2H, s), 4.36-4.34(1H, m), 4.25-4.22(1H, m), 4.04-3.98(1H, m), 3.84-3.77(1H, m), 3.04-2.85(3H, m), 2.28(3H, s).

【 0 0 8 9 】

実施例 9

【 化 7 7 】



(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-ヒドロキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン (VIII-33)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン(VII-4, 10g)を1,4-ジオキサン300mlに溶解させ、1.5当量の2-(アセトキシ)エトシアンミンを加えた後、2mol/Lメタンスルホン酸水溶液28mlを加えて60℃で17時間攪拌した。反応液を飽和炭酸水素ナトリウム水溶液に注ぎ、混合液を酢酸エチルで抽出した。有機層を水洗後、硫酸ナトリウム上で乾燥した。ろ液を濃縮した残渣を含水エタノール-水より再結晶後、ろ取、乾燥することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-ヒドロキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-33, 7.6g)を無色固体として得た。

$^1\text{H NMR}$ ( $d_6$ -DMSO, ): 10.07(1H, s), 8.74(1H, s), 8.58(1H, s), 8.22(1H, d,  $J=8.8\text{Hz}$ ), 7.96(1H, d,  $J=2.4\text{Hz}$ ), 7.80(1H, d,  $J=8.8\text{Hz}$ ), 7.69(1H, dd,  $J=2.4\text{Hz}$ ,  $J=8.8\text{Hz}$ ), 7.50-7.45(1H, m), 7.35-7.24(3H, m), 7.20-7.16(1H, m), 5.27(2H, s), 4.79(1H, t,  $J=5.6\text{Hz}$ ), 4.29(2H, t,  $J=5.6\text{Hz}$ ), 3.75(2H, dd,  $J=5.2\text{Hz}$ ,  $J=10.4\text{Hz}$ ), 2.26(3H, s).

【 0 0 9 0 】

(第2工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-ス

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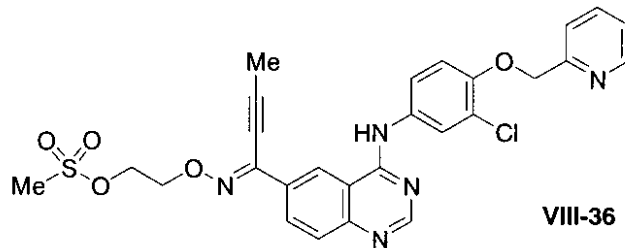
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ルホニルオキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-34)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-ヒドロキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-33, 7.6g)をテトラヒドロフラン150mlに溶かし、トリエチルアミン4.19mlと塩化メタンスルホニル2.33mlを加え3.5時間攪拌した。反応終了後に反応液を水に注ぎ、飽和炭酸水素ナトリウム水溶液を加えて、酢酸エチルで抽出した。有機層を硫酸ナトリウム上で乾燥し、ろ液を濃縮した。濃縮残渣に酢酸エチルを加え室温で静置すると結晶が析出したのでヘキサンで希釈後、結晶をろ取することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-スルホニルオキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-34, 7.66g)を淡黄色結晶として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 10.07(1\text{H}, \text{s}), 8.77(1\text{H}, \text{s}), 8.60(1\text{H}, \text{s}), 8.24(1\text{H}, \text{d}, \text{J}=8.8\text{Hz}), 7.97(1\text{H}, \text{d}, \text{J}=2.4\text{Hz}), 7.81(1\text{H}, \text{d}, \text{J}=8.8\text{Hz}), 7.69(1\text{H}, \text{dd}, \text{J}=2.4\text{Hz}, \text{J}=8.8\text{Hz}), 7.51\text{-}7.45(1\text{H}, \text{m}), 7.35\text{-}7.27(3\text{H}, \text{m}), 7.21\text{-}7.17(1\text{H}, \text{m}), 5.27(2\text{H}, \text{s}), 4.58(2\text{H}, \text{t}, \text{J}=4.8\text{Hz}), 4.54(2\text{H}, \text{t}, \text{J}=4.8\text{Hz}), 3.24(3\text{H}, \text{s}), 2.27(3\text{H}, \text{s}).$

【化78】

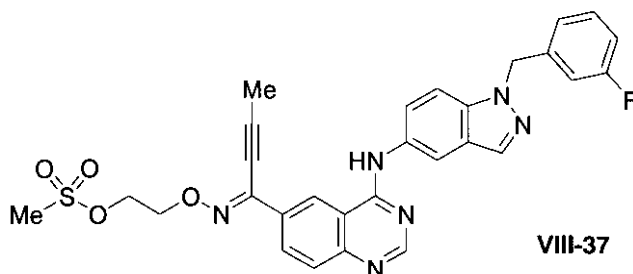


4-(3-クロロ-4-(ピリジン-2-イルメトキシ)フェニルアミノ)-6-(1-(2-スルホニルオキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-36)

第1工程および第2工程と同様の方法で合成した。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 10.09(1\text{H}, \text{s}), 8.77(1\text{H}, \text{s}), 8.61(1\text{H}, \text{s}), 8.59(1\text{H}, \text{s}), 8.24(1\text{H}, \text{d}, \text{J}=8.1\text{Hz}), 7.97(1\text{H}, \text{d}, \text{J}=2.4\text{Hz}), 7.89(1\text{H}, \text{t}, \text{J}=7.5\text{Hz}), 7.81(1\text{H}, \text{d}, \text{J}=9.0\text{Hz}), 7.68(1\text{H}, \text{dd}, \text{J}=2.4\text{Hz}, \text{J}=9.0\text{Hz}), 7.59(1\text{H}, \text{d}, \text{J}=8.1\text{Hz}), 7.40\text{-}7.35(1\text{H}, \text{m}), 7.29(1\text{H}, \text{d}, \text{J}=9.0\text{Hz}), 5.31(2\text{H}, \text{s}), 4.57(2\text{H}, \text{bs}), 4.55(2\text{H}, \text{bs}), 3.24(3\text{H}, \text{s}), 2.27(3\text{H}, \text{s}).$

【化79】



4-(1-(3-フルオロベンジル)-1H-インダゾール-5-イルアミノ)-6-(1-(2-スルホニルオキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-37)

第1工程および第2工程と同様の方法で合成した。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 10.20(1\text{H}, \text{s}), 8.81(1\text{H}, \text{s}), 8.53(1\text{H}, \text{s}), 8.24(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 8.17(1\text{H}, \text{s}), 8.13(1\text{H}, \text{s}), 7.80(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 7.75(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 7.67(1\text{H}, \text{d}, \text{J}=9.3\text{Hz}), 7.41\text{-}7.36(1\text{H}, \text{m}), 7.14\text{-}7.04(3\text{H}, \text{m}), 5.72(2\text{H}, \text{s}), 4.58(2\text{H}, \text{t}, \text{J}=5.4\text{Hz}), 4.54(2\text{H}, \text{t}, \text{J}=5.4\text{Hz}), 3.24(3\text{H}, \text{s}), 2.26(3\text{H}, \text{s}).$

(第3工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-エ

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チルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン( VIII-35)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-スルホニルオキシエトキシイミノ)-2-ブチン-1-イル)キナゾリン( VIII-34, 100mg)をN,N-ジメチルホルムアミド3mlに溶解し、70%エチルアミン水溶液を160  $\mu$ l加え、60 で14時間攪拌した。反応液に水を加え、酢酸エチルで抽出した。有機層を硫酸マグネシウム上で乾燥し、ろ液の濃縮残渣をアミノカラムを用いたクロマトグラフィー(酢酸エチルで溶出)により精製することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(1-(2-エチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン( VIII-35, 53mg)を無色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad )$ : 10.08(1H, s), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d,  $J=8.4\text{Hz}$ ), 7.96(1H, s), 7.80(1H, d,  $J=8.8\text{Hz}$ ), 7.69(1H, d,  $J=8.0\text{Hz}$ ), 7.51-7.45(1H, m), 7.35-7.27(3H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.31(2H, t,  $J=5.6\text{Hz}$ ), 2.89(2H, t,  $J=6.0\text{Hz}$ ), 2.61(2H, q,  $J=7.2\text{Hz}$ ), 2.26(3H, s), 1.02(3H, t,  $J=7.6\text{Hz}$ ).

【 0 0 9 1 】

第3工程のアミノ化では市販品として入手可能であるアミンまたはJ. Syn. Org. Chem., Jpn., 2001, 59 : 779 -789., Tetrahedron Lett., 1995, 36 : 6373-6374., Synlett, 1999 : 1301-1303., Tetrahedron, 2002, 58 : 6267-6276.に記載の方法に従って合成できるアミンまたはその塩を使用できる。

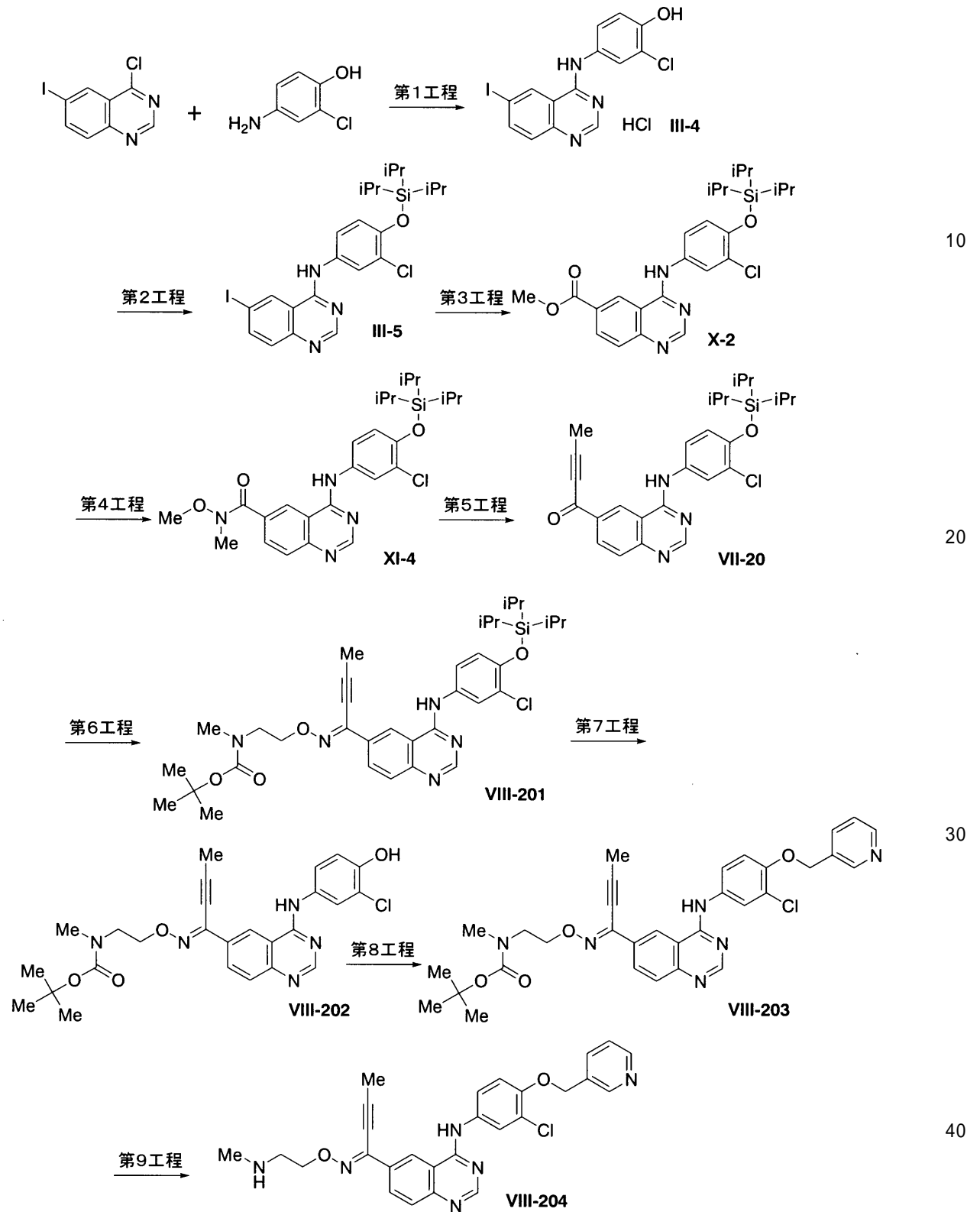
【 0 0 9 2 】

実施例 10

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【化 8 0】



(第1工程) 4-(3-クロロ-4-ヒドロキシフェニルアミノ)-6-ヨードキナゾリン (III-4) の合成

4-クロロ-6-ヨードキナゾリン20.3gおよび3-クロロ-4-ヒドロキシアニリン10gを出発原料として実施例1の第1工程と同様にして4-(3-クロロ-4-ヒドロキシフェニルアミノ)-6-ヨードキナゾリン塩酸塩(III-4, 26.6g)を黄色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad )$ : 11.4(1H, s), 10.5(1H, s), 9.21(1H, s), 8.93(1H, s), 8.35(1H

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, d, J=8.4Hz), 7.79(1H, s), 7.69(1H, d, J=9.0Hz), 7.48(1H, d, J=9.0Hz), 7.10(1H, d, J=8.4Hz).

【 0 0 9 3 】

(第2工程) 4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-ヨードキナゾリン ( III - 5 )の合成

4-(3-クロロ-4-ヒドロキシフェニルアミノ)-6-ヨードキナゾリン塩酸塩( III-4, 20g)、塩化トリイソプロピルシラン13.3g、イミダゾール12.6gをN,N-ジメチルホルムアミド200mlに溶解させ、室温で4時間攪拌した。反応終了後、飽和炭酸水素ナトリウム水溶液200mlおよび酢酸エチル200mlを加えて分液操作を行った。水層を酢酸エチル200mlで再抽出を2回繰り返した後、すべての有機層を合わせて水、飽和食塩水の順で洗浄し、無水硫酸マグネシウム上で乾燥した。ろ液を濃縮後、残渣をヘキサン-酢酸エチル混合液(2:1)で洗浄することにより4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-ヨードキナゾリン ( III-5, 25.3g)を無色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 9.8(1\text{H}, \text{s}), 8.96(1\text{H}, \text{s}), 8.62(1\text{H}, \text{s}), 8.12(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 8.02(1\text{H}, \text{d}, \text{J}=2.7\text{Hz}), 7.70(1\text{H}, \text{dd}, \text{J}=8.7\text{Hz}, \text{J}=2.7\text{Hz}), 7.56(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 7.04(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 1.25\text{-}1.40(3\text{H}, \text{m}), 1.11(18\text{H}, \text{d}, \text{J}=7.2\text{Hz}).$

(第3工程) 4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-キナゾリン-6-カルボン酸メチルエステル (X-2)の合成

4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-ヨードキナゾリン( III-5, 21.8g)より参考例1の第1工程と同様にして4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-キナゾリン-6-カルボン酸メチルエステル(X-2, 20.4g)を黄色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 10.2(1\text{H}, \text{s}), 9.26(1\text{H}, \text{s}), 8.66(1\text{H}, \text{s}), 8.30(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 8.01(1\text{H}, \text{d}, \text{J}=2.4\text{Hz}), 7.84(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 7.71(1\text{H}, \text{dd}, \text{J}=9\text{Hz}, \text{J}=2.1\text{Hz}), 7.04(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 3.96(3\text{H}, \text{s}), 1.29\text{-}1.41(3\text{H}, \text{m}), 1.11(18\text{H}, \text{d}, \text{J}=7.2\text{Hz}).$

(第4工程) 4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド (X1-4)の合成

4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-キナゾリン-6-カルボン酸メチルエステル(X-2, 10g)より参考例1の第2工程と同様にして4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(X1-4, 4.1g)を無色固体として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 10.0(1\text{H}, \text{s}), 8.80(1\text{H}, \text{s}), 8.65(1\text{H}, \text{s}), 7.97\text{-}8.08(2\text{H}, \text{m}), 7.81(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 7.70\text{-}7.75(1\text{H}, \text{m}), 7.04(1\text{H}, \text{d}, \text{J}=9.0\text{Hz}), 3.58(3\text{H}, \text{s}), 3.35(3\text{H}, \text{s}), 1.29\text{-}1.41(3\text{H}, \text{m}), 1.11(18\text{H}, \text{d}, \text{J}=7.5\text{Hz}).$

(第5工程) 4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン( VII - 20 )の合成

4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-キナゾリン-6-カルボン酸メトキシメチルアミド(X1-4, 4.1g)より参考例1の第3工程と同様にして4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン( VII - 20, 1.3g)を黄色油状物として得た。

$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad): 10.3(1\text{H}, \text{s}), 9.24(1\text{H}, \text{s}), 8.67(1\text{H}, \text{s}), 8.45(1\text{H}, \text{d}, \text{J}=8.7\text{Hz}), 7.99(1\text{H}, \text{s}), 7.88(1\text{H}, \text{s}, \text{J}=8.7\text{Hz}), 7.68(1\text{H}, \text{d}, \text{J}=9.0\text{Hz}), 7.06(1\text{H}, \text{d}, \text{J}=9.0\text{Hz}), 2.28(3\text{H}, \text{s}), 1.29\text{-}1.41(3\text{H}, \text{m}), 1.11(18\text{H}, \text{d}, \text{J}=6.9\text{Hz}).$

(第6工程) 4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン( VIII-201 )の合成

4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-(1-オキソ-2-ブチン-1-イル)キナゾリン( VII - 20, 1.3g)より実施例2の第2工程と同様にして4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン( VIII-201, 1.1g)を

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黄色固体として得た。

ESI-MS(m/z): (M+)=666.71, (M+H)=667.73

(第7工程) 4-(3-クロロ-4-ヒドロキシフェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-202)の合成

4-(3-クロロ-4-トリイソプロピルシリルオキシフェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-201, 1.1g)をテトラヒドロフラン20mlに溶解させ、1mol/Lフッ化テトラブチルアンモニウムテトラヒドロフラン溶液を6.7ml加え室温で30分撹拌した。反応終了後、水10mlおよび酢酸エチル10mlを加えて分液操作を行った。水層を酢酸エチル10mlで再抽出を2回繰り返した後、すべての有機層を合わせて水、飽和食塩水の順で洗浄し、無水硫酸マグネシウム上で乾燥した。ろ液を濃縮後、残渣をヘキサン-酢酸エチル混合液(1:1)で洗浄することにより4-(3-クロロ-4-ヒドロキシフェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-202, 0.7g)を黄色固体として得た。

<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 9.96(1H, s), 8.73(1H, s), 8.55(1H, s), 8.21(1H, d, J=8.7Hz), 7.75-7.86(2H, m), 7.50(1H, d, J=7.8Hz), 7.00(1H, d, J=8.7Hz), 4.36(2H, bs), 3.56(2H, bs), 2.86(3H, bs), 2.25(3H, bs), 1.36(9H, s).

(第8工程) 4-(3-クロロ-4-(ピリジン-3-イルメトキシ)フェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-203)の合成

4-(3-クロロ-4-ヒドロキシフェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-202, 70mg)、3-プロモメチルピリジン臭化水素酸塩87mg、炭酸セシウム223mgをN,N-ジメチルホルムアミド-水混合液(10:1)1.1mlに溶解させ、室温で3時間撹拌させた。反応終了後、水5mlおよび酢酸エチル5mlを加えて分液操作を行った。水層を酢酸エチル5mlで再抽出を2回繰り返した後、すべての有機層を合わせて水、飽和食塩水の順で洗浄し、無水硫酸マグネシウム上で乾燥した。ろ液を濃縮後、残渣をシリカゲルカラムクロマトグラフィ(ヘキサン:酢酸エチル=1:1で溶出)で精製することにより4-(3-クロロ-4-(ピリジン-3-イルメトキシ)フェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-203, 70mg)を黄色油状物として得た。

<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 10.0(1H, bs), 8.74(1H, bs), 8.72(1H, bs), 8.60(1H, s), 8.57(1H, d, J=4.4Hz), 8.23(1H, d, J=8.8Hz), 7.97(1H, bs), 7.91(1H, d, J=8.0Hz), 7.80(1H, d, J=8.8Hz), 7.72(1H, d, J=8.8Hz), 7.46(1H, dd, J=7.6Hz, J=4.4Hz), 7.33(1H, d, J=8.8Hz), 5.29(2H, s), 4.37(2H, bs), 3.56(2H, bs), 2.86(3H, bs), 2.25(3H, bs), 1.36(9H, s).

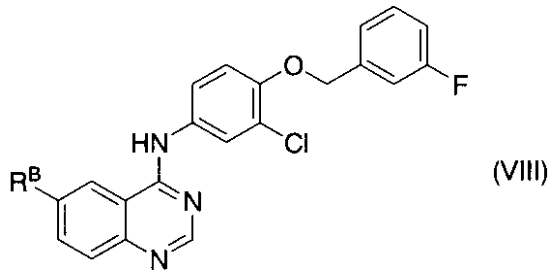
(第9工程) 4-(3-クロロ-4-(ピリジン-3-イルメトキシ)フェニルアミノ)-6-(1-(2-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-204)の合成

4-(3-クロロ-4-(ピリジン-3-イルメトキシ)フェニルアミノ)-6-(1-(2-(N-(tert-ブトキシカルボニル)-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-203, 70mg)より実施例2の第2工程と同様にして反応させ、ヘキサン-酢酸エチルから再結晶で精製することにより4-(3-クロロ-4-(ピリジン-3-イルメトキシ)フェニルアミノ)-6-(1-(2-N-メチルアミノエトキシイミノ)-2-ブチン-1-イル)キナゾリン(VIII-204, 7mg)を黄色固体として得た。

<sup>1</sup>H NMR(d<sub>6</sub>-DMSO, ): 10.3(1H, s), 8.84(1H, s), 8.72(1H, s), 8.42-8.65(2H, m), 8.26(1H, d, J=7.6Hz), 7.98(1H, s), 7.91(1H, brs), 7.81(1H, brs), 7.71(1H, brs), 7.47(1H, brs), 7.34(1H, d, J=8.0Hz), 5.30(2H, s), 4.53(2H, brs), 3.38(2H, brs), 2.68(3H, s), 2.26(3H, s).

【0094】

実施例 7 ~ 10 と同様の方法で下記の化合物を合成した。  
【化 8 1】



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【 0 0 9 5 】

【表 15】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)	
VIII-2		9.92(1H, s), 8.65(1H, brs), 8.62(1H, s), 8.22(1H, d, J=6.6Hz), 7.96(1H, d, J=2.1Hz), 7.76(1H, d, J=6.6Hz), 7.70(1H, dd, J=6.6Hz, J=1.8Hz), 7.53-7.45(1H, m), 7.35-7.28(3H, m), 7.18(1H, t, J=5.7Hz), 5.27(2H, s), 4.30(2H, t, J=4.5Hz), 2.64(2H, t, J=4.5Hz), 2.43(4H, brs), 2.35(3H, s), 1.53-1.47(4H, m), 1.40-1.36(2H, m).	10
VIII-3		9.92(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.23(1H, d, J=6.6Hz), 7.96(1H, d, J=1.8Hz), 7.76(1H, d, J=6.6Hz), 7.70(1H, d, J=6.9Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.18(1H, t, J=6.9Hz), 5.27(2H, s), 4.26(2H, t, J=4.5Hz), 2.77(2H, t, J=4.5Hz), 2.55(4H, q, J=5.4Hz), 2.36(3H, s), 0.96(2H, t, J=5.4Hz)	
VIII-4		(一塩酸塩) 11.4(1H, brs), 10.82(1H, brs), 9.07(1H, s), 8.82(1H, s), 8.40(1H, d, J=10.2Hz), 7.94(1H, d, J=2.7Hz), 7.88(1H, d, J=9.0Hz), 7.70(1H, dd, J=8.7Hz, 2.7Hz), 7.45-7.52(1H, m), 7.31-7.36(3H, m), 7.17-7.23(1H, m), 5.30(2H, s), 4.94(1H, m), 3.85(4H, m), 3.36-3.60(6H, m), 2.44(3H, s), 1.38(3H, d, J=6.3Hz)	20
VIII-5		9.93(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.22(1H, d, J=6.6Hz), 7.96(1H, d, J=1.8Hz), 7.76(1H, d, J=6.6Hz), 7.70(1H, dd, J=6.6Hz, J=1.8Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.6Hz), 5.27(2H, s), 4.31(2H, t, J=4.5Hz), 2.78(2H, t, J=4.5Hz), 2.50(4H, br), 2.36(3H, s), 1.69(4H, br)	
VIII-6		9.95(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.21(1H, d, J=6.9Hz), 7.94(1H, d, J=1.5Hz), 7.77(1H, d, J=6.6Hz), 7.69(1H, d, J=6.9Hz), 7.51-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.3Hz), 5.28(2H, s), 4.32(2H, t, J=4.2Hz), 3.09(4H, brd, J=4.5Hz), 3.05(4H, brd, J=4.5Hz), 2.97-2.89(4H, m), 1.13(3H, t, J=5.4Hz)	30
VIII-7		9.95(1H, s), 8.61(1H, brs), 8.58(1H, s), 8.20(1H, d, J=6.6Hz), 7.94(1H, brs), 7.77(1H, d, J=6.6Hz), 7.69(1H, d, J=6.9Hz), 7.51-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.0Hz), 5.27(2H, s), 4.32(2H, t, J=4.2Hz), 3.59(4H, t, J=3.3Hz), 2.91(2H, q, J=5.4Hz), 2.68(2H, t, J=4.2Hz), 1.13(3H, t, J=5.4Hz)	40

【 0 0 9 6 】



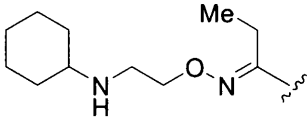
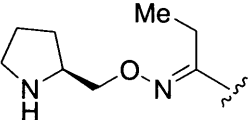
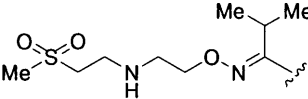
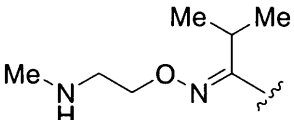
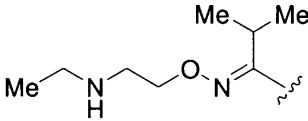
【表 16】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)	
VIII-8		(E/Z mixture) 8.61 (0.5H, s), 8.58 (0.5H, s), 8.48 (0.5H, s), 8.25 (0.5H, s), 7.99 (0.5H, d, J = 2.4Hz), 7.97 (0.5H, d, J = 2.4 Hz), 7.90 (0.5H, d, J = 7.9 Hz), 7.65-7.73 (2.5H, m), 7.50-7.55 (0.5H, m), 7.26-7.34 (3.5H, m), 7.16 (1H, d, t J = 7.9, 2.4 Hz), 5.26 (2H, s), 4.29 (1H, t, J = 7.0 Hz), 4.15 (1H, t, J = 7.0 Hz), 3.50-3.55 (1H, m), 3.28-3.35 (2H, m), 3.20-3.28 (2H, m), 3.10-3.20 (2H, m), 2.99-3.07 (2H, m), 2.90-2.96 (1H, m), 2.87-2.95 (1H, m), 1.21 (3H, d, J = 7.0Hz), 1.16 (3H, d, J = 7.0Hz)	10
VIII-9		(E/Z mixture) 8.58 (0.5H, s), 8.56 (0.5H, s), 8.44 (0.5H, s), 8.36 (0.5H, s), 7.99 (0.5H, d, J = 2.4Hz), 7.97 (0.5H, d, J = 2.4 Hz), 7.90 (0.5H, d, J = 7.9 Hz), 7.65-7.73 (2.5H, m), 7.50-7.55 (0.5H, m), 7.26-7.34 (3.5H, m), 7.16 (1H, d, t J = 7.9, 2.4 Hz), 5.24 (2H, s), 4.21 (1H, t, J = 7.0 Hz), 4.05 (1H, t, J = 7.0 Hz), 2.73 (1H, t, J = 7.2Hz), 2.57 (1H, t, J = 7.2Hz), 2.45-2.55 (2H, m), 2.30-2.35 (2H, m), 1.64-1.70 (2H, m), 1.50-1.58 (2H, m), 1.21 (3H, d, J = 7.0Hz), 1.16 (3H, d, J = 7.0Hz)	20
VIII-10		(E/Z mixture) 10.09(1H, s, major), 10.00(1H, s, minor), 8.74(1H, s, major), 8.59(1H, s), 8.22(1H, d, J=9Hz, major), 7.94-8.02(1H, m), 7.64-7.82(2H, m), 7.44-7.52(1H, m), 7.16-7.36(4H, m), 5.27(2H, s), 4.32-4.43(2H, m), 3.56-3.62(4H, m), 2.66-2.74(2H, m), 2.34(3H, s, minor), 2.25(3H, s, major)	30
VIII-11		10.08(1H, s), 8.74(1H, s), 8.58(1H, s), 8.22(1H, d, J=8.4Hz), 7.96(1H, brs), 7.80(1H, d, J=8.4Hz), 7.68(1H, d, J=9.6Hz), 7.43-7.52(1H, m), 7.14-7.36(4H, m), 5.27(2H, s), 4.37(2H, t, J=6.0Hz), 2.80(2H, t, J=6.0Hz), 2.25(3H, s), 1.69(4H, brs), 1.24(4H, brs)	30
VIII-12		(E/Z mixture) 9.95(1H, s, major), 9.80(1H, s, minor), 8.65(1H, s, minor), 8.58(1H, s, major), 8.54(1H, s, major), 8.44(1H, s, minor), 7.99(1H, d, J=1.5Hz, minor), 7.91(1H, d, J=1.8Hz, major), 7.15-7.92(14H, m), 5.26(2H, s), 5.25(2H, s), 4.32(2H, d, J=3.9Hz, major), 4.27(2H, t, J=4.2Hz, minor), 2.82-3.06(10H, m)	40
VIII-13		(E/Z mixture) 9.96(1H, s, major), 9.82(1H, s, minor), 8.64(1H, s, minor), 8.57(1H, s, major), 8.53(1H, brs, major), 8.44(1H, brs, minor), 7.99(1H, d, J=2.7Hz, minor), 7.90(1H, d, J=2.4Hz, major), 7.14-7.88(13H, m), 5.26(2H, s, major), 5.24(2H, s, minor), 4.32(2H, t, J=5.7Hz, major), 4.27(2H, t, J=5.4Hz, minor), 3.53-3.58(4H, m, major), 3.44-3.50(4H, m, minor), 2.56-2.70(2H, m), 2.32-2.46(4H, m)	40

【表 17】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)	
VIII-14		9.99(1H, s), 8.65(s, 1H), 8.61(1H, s), 7.98(1H, brs), 7.90(1H, d, J=6.9Hz), 7.87(1H, d, J=6.9Hz), 7.70(1H, d, J=6.3Hz), 7.40-7.51(1H, m), 7.27-7.35(3H, m), 7.15-7.21(1H, m), 5.27(2H, s), 4.38(2H, t, J=3.9Hz), 3.47-3.51(4H, m), 2.57-2.65(2H, m), 2.33-2.40(4H, m)	
VIII-15		9.94(1H, s), 8.63(1H, s), 8.58(1H, s), 8.23(1H, dd, J=8.7Hz, 1.2Hz), 7.96(1H, d, J=2.4Hz), 7.76(1H, d, J=9.0Hz), 7.70(1H, dd, J=8.4Hz, 2.4Hz), 7.44-7.52(1H, m), 7.28-7.35(3H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.07-4.10(2H, m), 3.23(2H, t, J=6.9Hz), 3.01-3.04(6H, m), 2.38(3H, s), 1.05(3H, d, J=6.3Hz)	10
VIII-16		9.94(1H, s), 8.62(1H, s), 8.58(1H, s), 8.25(1H, dd, J=9.0Hz, 1.5Hz), 7.95(1H, d, J=2.7Hz), 7.76(1H, d, J=8.7Hz), 7.69(1H, dd, J=8.7Hz, 2.4Hz), 7.44-7.52(1H, m), 7.28-7.35(3H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.39-4.45(1H, m), 3.25(2H, t, J=6.9Hz), 2.95-3.01(5H, m), 2.73-2.92(2H, m), 2.36(3H, s), 1.30(3H, d, J=6.3Hz)	20
VIII-17		(二塩酸塩) 11.09(1H, brs), 9.00(1H, s), 8.90(2H, brs), 8.75(1H, s), 8.34(1H, d, J=9.3Hz), 7.96(1H, d, J=2.7Hz), 7.85(1H, d, J=8.7Hz), 7.72(1H, dd, J=9.0Hz, 2.7Hz), 7.45-7.52(1H, m), 7.31-7.35(3H, m), 7.17-7.22(1H, m), 5.29(2H, s), 4.49(2H, t, J=5.4Hz), 3.67(2H, t, J=5.4Hz), 3.49(2H, q, J=6.9Hz), 3.21-3.23(4H, m), 2.45(3H, s), 1.13(3H, t, J=6.9Hz)	
VIII-18		9.93(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.22(1H, d, J=6.9Hz), 7.95(1H, brs), 7.76(1H, d, J=6.0Hz), 7.70(1H, d, J=6.3Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.18(1H, t, J=5.4Hz), 5.27(2H, s), 4.11-4.03(2H, m), 2.87-2.67(2H, m), 2.37(3H, s), 1.85-1.76(1H, m), 1.73-1.59(2H, m), 1.48-1.39(1H, m)	30
VIII-19		(二トリフルオロ酢酸塩) 11.40(1H, s), 9.10(2H, s), 8.89(1H, s), 8.81(1H, s), 8.42(1H, d, J=6.0Hz), 7.90-7.87(2H, m), 7.62(1H, d, J=6.6Hz), 7.51-7.46(1H, m), 7.38-7.31(3H, m), 7.20(1H, t, J=6.3Hz), 5.32(2H, s), 4.48(2H, brs), 3.61-3.46(6H, m), 3.13(3H, s), 2.97(2H, q, J=5.7Hz), 1.14(3H, t, J=5.7Hz).	40
VIII-20		(二トリフルオロ酢酸塩) 11.07(1H, s), 8.82(1H, s), 8.78(3H, brs), 8.37(1H, d, J=6.6Hz), 7.89-7.86(2H, m), 7.64(1H, d, J=6.6Hz), 7.51-7.46(1H, m), 7.36-7.32(3H, m), 7.20(1H, t, J=6.3Hz), 5.31(2H, s), 4.45(2H, brs), 2.97(2H, q, J=5.4Hz), 2.67(3H, brs), 1.13(3H, t, J=5.4Hz).	

【表 18】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-21		(ニトリフルオロ酢酸塩) 11.03(1H, s), 8.81(1H, s), 8.78(1H, s), 8.71(2H, s), 8.37(1H, d, J=6.6Hz), 7.88-7.86(2H, m), 7.64(1H, d, J=6.6Hz), 7.51-7.46(1H, m), 7.36-7.31(3H, m), 7.20(1H, t, J=6.3Hz), 5.31(2H, s), 4.46(2H, brs), 3.39(2H, brs), 3.10(1H, brs), 2.97(2H, q, J=6.3Hz), 2.06(2H, brd, J=8.4Hz), 1.78(2H, brd, J=8.4Hz), 1.37-1.04(9H, m).
VIII-22		9.95(1H, s), 8.61(1H, brs), 8.58(1H, s), 8.19(1H, d, J=6.3Hz), 7.94(1H, brs), 7.77(1H, d, J=6.6Hz), 7.69(1H, d, J=6.6Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.0Hz), 5.27(2H, s), 4.14-4.07(2H, m), 3.43(1H, br), 2.95-2.82(4H, m), 1.86-1.80(1H, m), 1.69(2H, m), 1.46(1H, m), 1.14(3H, t, J=5.4Hz)
VIII-23		(E/Z mixture) 8.60 (0.5H, s), 8.58 (0.5H, s), 8.47 (0.5H, s), 8.39 (0.5H, s), 7.96 (0.5H, d, J = 2.4Hz), 7.93 (0.5H, d, J = 2.4 Hz), 7.88 (0.5H, d, J = 7.9 Hz), 7.65-7.73 (2.5H, m), 7.50-7.55 (0.5H, m), 7.26-7.34 (3.5H, m), 7.16 (1H, d, t J = 7.9, 2.4 Hz), 5.26 (2H, s), 4.19 (1H, t, J = 7.0 Hz), 4.03 (1H, t, J = 7.0 Hz), 3.50-3.55 (1H, m), 3.22-3.30 (1H, m), 3.16-3.23 (1H, m), 2.96-3.03 (1H, m), 3.04 (1.5H, s), 2.90-2.95 (2H, m), 2.93 (1.5H, s), 2.70-2.80 (1H, m), 1.21 (3H, d, J = 7.0Hz), 1.16 (3H, d, J = 7.0Hz)
VIII-24		(E/Z mixture) 8.61 (0.5H, s), 8.59 (0.5H, s), 8.47 (0.5H, s), 8.41 (0.5H, s), 7.99 (0.5H, d, J = 2.4Hz), 7.97 (0.5H, d, J = 2.4 Hz), 7.90 (0.5H, d, J = 7.9 Hz), 7.65-7.73 (2.5H, m), 7.50-7.55 (0.5H, m), 7.26-7.34 (3.5H, m), 7.16 (1H, d, t J = 7.9, 2.4 Hz), 5.26 (2H, s), 4.29 (1H, t, J = 7.0 Hz), 4.13 (1H, t, J = 7.0 Hz), 3.50-3.55 (1H, m), 3.14-3.30 (1H, m), 2.96-3.04 (1H, m), 2.54 (1.5H, s), 2.49 (1.5H, s), 1.21 (3H, d, J = 7.0Hz), 1.16 (3H, d, J = 7.0Hz), 1.00 (3H, t, J = 7.2Hz)
VIII-25		(E/Z mixture) 8.61 (0.5H, s), 8.58 (0.5H, s), 8.48 (0.5H, s), 8.31 (0.5H, s), 7.99 (0.5H, d, J = 2.4Hz), 7.97 (0.5H, d, J = 2.4 Hz), 7.90 (0.5H, d, J = 7.9 Hz), 7.65-7.73 (2.5H, m), 7.50-7.55 (0.5H, m), 7.26-7.34 (3.5H, m), 7.16 (1H, d, t J = 7.9, 2.4 Hz), 5.26 (2H, s), 4.29 (1H, t, J = 7.0 Hz), 4.15 (1H, t, J = 7.0 Hz), 3.50-3.55 (1H, m), 3.24-3.30 (1H, m), 3.10-3.14 (1H, m), 2.99 (1H, q, J = 7.2Hz), 2.89 (1H, q, J = 7.2Hz), 1.21 (3H, d, J = 7.0Hz), 1.16 (3H, d, J = 7.0Hz), 1.00 (3H, t, J = 7.2Hz)

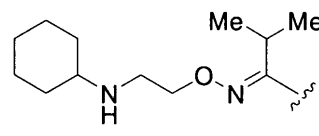
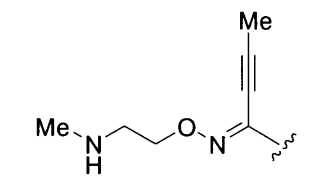
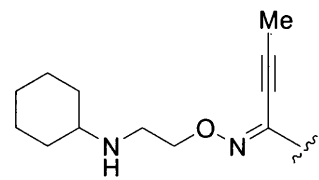
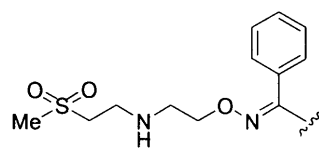
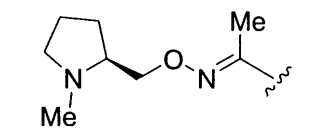
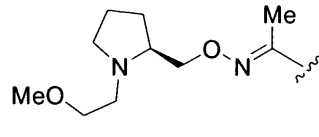
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【表 19】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-26		(E/Z mixture) 8.61 (0.5H, s), 8.58 (0.5H, s), 8.48 (0.5H, s), 8.31 (0.5H, s), 7.99 (0.5H, d, J = 2.4Hz), 7.94 (0.5H, d, J = 2.4 Hz), 7.90 (0.5H, d, J = 7.9 Hz), 7.65-7.73 (2.5H, m), 7.50-7.55 (0.5H, m), 7.26-7.34 (3.5H, m), 7.16 (1H, d, t J = 7.9, 2.4 Hz), 5.26 (2H, s), 4.29 (1H, t, J = 7.0 Hz), 4.11 (1H, t, J = 7.0 Hz), 3.50-3.55 (1H, m), 3.14-3.17 (1H, m), 3.00-3.04 (1H, m), 2.00-2.10 (2H, m), 1.70-1.85 (2H, m), 1.00-1.30 (6H, m), 1.23 (3H, d, J = 7.0Hz), 1.16 (3H, d, J = 7.0Hz)
VIII-27		10.10(1H, s), 8.76(1H, s), 8.60(1H, s), 8.25(1H, d, J=8.7Hz), 7.96(1H, brs), 7.82(1H, d, J=9.0Hz), 7.69(1H, d, J=9.0Hz), 7.43-7.52(1H, m), 7.14-7.36(4H, m), 5.28(2H, s), 4.43(2H, brs), 2.98(2H, brs), 2.54(3H, s), 2.27(3H, brs), 1.60(1H, s)
VIII-28		10.10(1H, s), 8.77(1H, s), 8.60(1H, s), 8.25(1H, d, J=8.7Hz), 7.95(1H, brs), 7.82(1H, d, J=9.6Hz), 7.69(1H, d, J=9.3Hz), 7.43-7.52(1H, m), 7.14-7.36(4H, m), 5.28(2H, s), 4.45(2H, brs), 2.91(2H, brs), 2.27(3H, s), 2.00(2H, brs), 1.73(2H, brs), 1.60(1H, brs), 1.23(7H, brs)
VIII-29		(E/Z mixture) 9.94(1H, s, major), 9.78(1H, s, minor), 8.62(1H, s, minor), 8.56(1H, s, major), 8.51(1H, brs, major), 8.42(1H, brs, minor), 7.12-7.53(14H, m), 5.24(2H, s), 4.15-4.28(2H, m), 3.13-3.25(2H, m), 2.80-3.00(7H, m)
VIII-30		9.93(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.22(1H, d, J=6.3Hz), 7.96(1H, d, J=1.2Hz), 7.76(1H, d, J=6.6Hz), 7.70(1H, d, J=6.6Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.6Hz), 5.27(2H, s), 4.23(1H, dd, J=8.1Hz, J=4.2Hz), 4.11(1H, dd, J=8.1Hz, J=4.2Hz), 2.98-2.93(1H, m), 2.59-2.54(1H, m), 2.36(6H, s), 2.17(1H, dd, J=13Hz, J=6.6Hz), 1.97-1.88(1H, m), 1.71-1.54(3H, m)
VIII-31		9.92(1H, s), 8.62(1H, brs), 8.58(1H, s), 8.22(1H, d, J=6.9Hz), 7.96(1H, brs), 7.76(1H, d, J=6.6Hz), 7.70(1H, d, J=6.6Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.6Hz), 5.27(2H, s), 4.18(1H, dd, J=7.8Hz, J=4.5Hz), 4.06(1H, dd, J=7.8Hz, J=4.5Hz), 3.43(1H, t, J=4.8Hz), 3.24(3H, s), 3.08-2.99(2H, m), 2.88-2.81(1H, m), 2.36(3H, s), 2.27(1H, dd, J=13Hz, J=6.6Hz), 1.93-1.84(1H, m), 1.72-1.54(3H, m)

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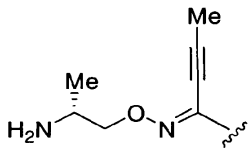
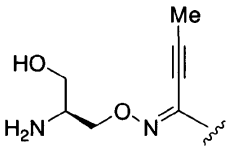
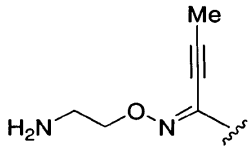
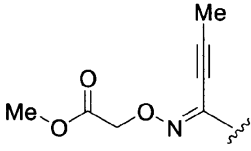
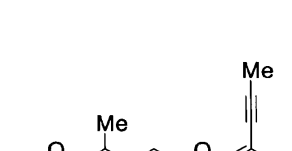
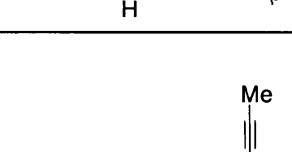
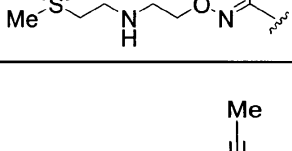
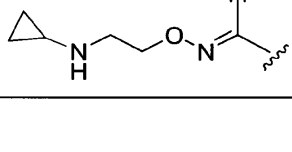
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【 0 1 0 0 】

【表 20】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-40		10.08(1H, bs), 8.74(1H, s), 8.58(1H, s), 8.21(1H, d, J=8.8Hz), 7.96(1H, s), 7.80(1H, d, J=8.8Hz), 7.51-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=8.0Hz), 5.27(2H, s), 4.04(2H, t, J=5.6Hz), 3.08-3.05(1H, m), 2.22(3H, s), 1.04(3H, d, J=6.4Hz).
VIII-41		10.13-10.09(1H, m), 8.82(1H, s), 8.74(1H, s), 8.21(1H, d, J=14.0Hz), 7.69-7.62(1H, m), 7.50-7.45(1H, m), 7.35-7.26(3H, m), 7.18(1H, t, J=8.2Hz), 5.27(2H, s), 4.69(1H, bs), 4.25-4.21(1H, m), 4.13-4.09(1H, m), 3.46-3.44(2H, m), 3.09(1H, t, J=5.6Hz), 2.26(3H, s)
VIII-42		10.2(1H, s), 8.78(1H, s), 8.59(1H, s), 8.24(1H, d, J=8.8Hz), 7.96(1H, s), 7.81(1H, d, J=8.8Hz), 7.66-7.73(1H, m), 7.43-7.50(1H, m), 7.23-7.36(3H, m), 7.15-7.22(1H, m), 5.27(2H, s), 4.31-4.48(2H, m), 3.07-3.15(2H, m), 2.27(3H, s)
VIII-43		10.10(1H, brs), 8.75(1H, d, J=2.0Hz), 8.59(1H, s), 8.17(1H, dd, J=2.4, 11.6Hz), 7.96(1H, d, J=3.2Hz), 7.80(1H, d, J=12.0Hz), 7.68(1H, dd, J=3.6, 12.0Hz), 7.51-7.44(1H, m), 7.34-7.26(3H, m), 7.22-7.15(1H, m), 5.27(2H, s), 4.94(2H, s), 3.71(3H, s), 2.27(3H, s)
VIII-44		(CDCl <sub>3</sub> ) 8.71(1H, s), 8.53(1H, s), 8.39(1H, s), 8.23(1H, d, J=9.0Hz), 7.88(1H, d, J=9.0Hz), 7.81(1H, d, J=2.4Hz), 7.57(1H, dd, J=2.4, 9.0Hz), 7.40-7.33(1H, m), 7.26-7.21(2H, m), 7.06-6.97(2H, m), 5.17(2H, s), 4.49(2H, t, J=5.4Hz), 3.40(2H, d, J=6.0Hz), 3.25(3H, s), 3.14-2.98(3H, m), 2.25(3H, s), 1.06(3H, d, J=6.6Hz).
VIII-45		(二塩酸塩) 12.2(1H, s), 9.61(2H, brs), 9.38(1H, s), 8.93(1H, s), 8.46(1H, d, J=8.8Hz), 8.03(1H, d, J=8.4Hz), 7.93(1H, d, J=2.4Hz), 7.69(1H, dd, J=8.8Hz, J=2.4Hz), 7.44-7.52(1H, m), 7.30-7.36(3H, m), 7.15-7.23(1H, m), 5.32(2H, s), 4.61(2H, brs), 3.65-3.70(2H, m), 3.45-3.56(5H, m), 3.12(3H, s), 2.28(3H, s)
VIII-46		10.07(1H, brs), 8.73(1H, s), 8.58(1H, s), 8.21(1H, d, 6.6Hz), 7.96(1H, s), 7.79(1H, d, J=6.6Hz), 7.68(1H, d, J=6.6Hz), 7.49-7.44(1H, m), 7.34-7.25(3H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.31(2H, t, 4.5 Hz), 2.95(2H, t, 4.5 Hz), 2.25(3H, s), 2.16(1H, m), 0.36(2H, m), 0.25(2H, m)
VIII-47		(二塩酸塩) 12.26(1H, brs), 9.36(1H, s), 8.91(1H, s), 8.45(1H, d, 6.6Hz), 8.29(1H, m), 8.00(1H, d, J=6.6Hz), 7.91(1H, s), 7.68(1H, d, J=6.6Hz), 7.51-7.45(1H, m), 7.36-7.31(3H, m), 7.23-7.17(1H, m), 5.32(2H, s), 4.59(2H, m), 3.48(2H, m), 3.41(2H, m), 3.10(2H, m), 2.28(3H, s), 1.82(3H, s)

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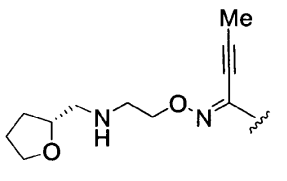
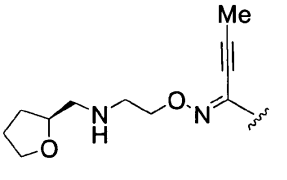
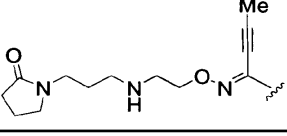
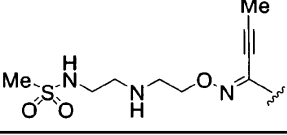
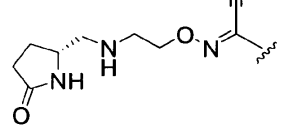
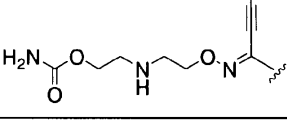
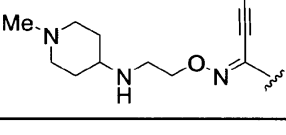
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【 0 1 0 1 】

【表 2 1】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-48		(二塩酸塩) 12.49(1H, brs), 9.43(1H, s), 8.93(1H, s), 8.43(1H, d, 6.6Hz), 8.09(1H, d, J=6.6Hz), 7.93(1H, s), 7.70(1H, d, J=6.6Hz), 7.51-7.45(1H, m), 7.36-7.31(3H, m), 7.23-7.17(1H, m), 5.32(2H, s), 4.61(2H, m), 4.24(1H, m), 3.80(1H, m), 3.68(1H, m), 3.49(2H, m), 3.20(1H, m), 3.05(1H, m), 2.28(3H, s), 1.99(1H, m), 1.83(2H, m), 1.58(1H, m)
VIII-49		(二塩酸塩) 12.44(1H, brs), 9.41(1H, s), 8.92(1H, s), 8.43(1H, d, 6.6Hz), 8.08(1H, d, J=6.6Hz), 7.92(1H, s), 7.70(1H, d, J=6.6Hz), 7.51-7.45(1H, m), 7.36-7.31(3H, m), 7.23-7.16(1H, m), 5.31(2H, s), 4.61(2H, m), 4.24(1H, m), 3.80(1H, m), 3.68(1H, m), 3.48(2H, m), 3.19(1H, m), 3.05(1H, m), 2.28(3H, s), 1.99(1H, m), 1.84(2H, m), 1.58(1H, m).
VIII-50		(二塩酸塩) 12.37(1H, brs), 9.41(1H, s), 8.91(1H, s), 8.41(1H, d, 6.0Hz), 8.04(1H, d, J=6.0Hz), 7.91(1H, s), 7.68(1H, d, J=6.0Hz), 7.45(1H, m), 7.36-7.31(3H, m), 7.20-7.14(1H, m), 5.29(2H, s), 4.58(2H, m), 3.44(2H, m), 3.31(2H, m), 3.22(2H, m), 2.95(2H, m), 2.27-1.55(9H, m)
VIII-51		(二塩酸塩) 10.07(1H, brs), 8.74(1H, s), 8.58(1H, s), 8.20(1H, d, 6.6Hz), 7.96(1H, s), 7.79(1H, d, J=6.6Hz) 7.69(1H, d, J=6.6Hz), 7.66(1H, s), 7.49-7.43(1H, m), 7.34-7.24(3H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.32(2H, m), 3.61(1H, m), 2.93(2H, m), 2.61(2H, d, 4.5Hz), 2.25(3H, s), 2.09(3H, m), 1.70(1H, m)
VIII-52		10.07(1H, brs), 8.74(1H, s), 8.58(1H, s), 8.20(1H, d, 6.6Hz), 7.96(1H, s), 7.79(1H, d, J=6.6Hz) 7.69(1H, d, J=6.6Hz), 7.66(1H, s), 7.49-7.43(1H, m), 7.34-7.24(3H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.32(2H, m), 3.61(1H, m), 2.93(2H, m), 2.61(2H, d, 4.5Hz), 2.25(3H, s), 2.09(3H, m), 1.70(1H, m)
VIII-53		(二塩酸塩) 12.04(1H, brs), 9.26(1H, s), 8.91(1H, s), 8.45(1H, d, 10.5Hz), 7.99(1H, d, 9.0Hz), 7.91(1H, s), 7.67(1H, d, J=9.0Hz), 7.53-7.45(1H, m), 7.37-7.31(3H, m), 7.23-7.17(1H, m), 6.65(2H, s), 5.32(2H, s), 4.58(2H, m), 4.23(2H, m), 3.50(2H, m), 3.30(2H, m), 2.28(3H, s)
VIII-54		(三塩酸塩) 10.88(1H, bs), 9.68(1H, bs), 9.33(1H, s), 8.83(1H, s), 8.61(1H, bs), 8.54(2H, d, J=5.2Hz), 8.00(1H, d, J=7.96Hz), 7.95(1H, s), 7.71(1H, d, J=8.4Hz), 7.49-7.45(1H, m), 7.33(2H, d, J=5.6Hz), 7.24-7.17(1H, m), 5.31(2H, s), 4.61(2H, bs), 2.91(2H, bs), 2.68(3H, s), 2.37-2.33(2H, m), 2.29(3H, s), 2.05-1.79(5H, m), 1.24(2H, s).

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【表 2 2】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-55		10.07(1H, bs), 8.74(1H, s), 8.59(1H, s), 8.31(1H, s), 8.21(1H, d, J=8.8Hz), 7.96(1H, s), 7.80(1H, d, J=8.4Hz), 7.69(1H, d, J=9.2Hz), 7.50-7.45(3H, m), 7.18(1H, t, J=6.4Hz), 5.27(2H, s), 4.33-4.30(2H, m), 3.81(2H, d, J=7.2Hz), 2.93(2H, t, J=5.9Hz), 2.67-2.65(1H, m), 2.33(3H, s), 1.76(2H, d, J=11.6Hz), 1.24-1.16(4H, m).
VIII-56		10.08(1H, s), 8.75(1H, s), 8.59(1H, s), 8.21(1H, d, J=10.0Hz), 7.80(1H, d, J=10.0Hz), 7.69(1H, d, J=8.4Hz), 7.47(1H, t, J=6.8Hz), 7.35-7.17(4H, m), 5.27(2H, s), 3.29(3H, s), 3.08-2.93(1H, m), 2.41(3H, s), 2.26(3H, s).
VIII-57		10.1(1H, s), 8.75(1H, s), 8.59(1H, s), 8.22(1H, d, J=8.4Hz), 7.96(1H, s), 7.80(1H, d, J=8.8Hz), 7.69(1H, d, J=8.8Hz), 7.43-7.51(1H, m), 7.23-7.36(3H, m), 7.18(1H, t, J=6.4Hz), 5.27(2H, s), 4.33(2H, t, J=5.6Hz), 3.30-3.33(2H, m), 2.98-3.05(2H, m), 2.25(3H, s)
VIII-58		10.1(1H, s), 8.75(1H, s), 8.59(1H, s), 8.23(1H, d, J=8.8Hz), 7.96(1H, s), 7.80(1H, d, J=8.8Hz), 7.74(1H, d, J=8.8Hz), 7.43-7.50(1H, m), 7.25-7.35(3H, m), 7.18(1H, t, J=7.6Hz), 6.99-7.08(2H, m), 5.27(2H, s), 4.35(2H, t, J=5.6Hz), 3.48-3.53(2H, m), 2.68(3H, d, J=4.4Hz), 2.26(3H, s)
VIII-59		(二塩酸塩) 11.83(1H, brs), 9.02(1H, s), 8.93(1H, s), 8.45(1H, d, J=8.7Hz), 7.89(1H, d, 8.7Hz), 7.87(1H, d, J=2.4Hz), 7.62(1H, dd, J=2.4Hz, J=9.0Hz), 7.53-7.45(1H, m), 7.38-7.31(3H, m), 7.82-7.20(1H, m), 5.32(2H, s), 4.37(2H, m), 3.56-3.25(10H, m), 2.26(3H, s).
VIII-60		10.08(1H, s), 8.74(1H, s), 8.59(1H, s), 8.22(1H, d, J=8.8Hz), 7.76(1H, s), 7.81(1H, d, J=8.8Hz), 7.69(1H, d, J=7.2Hz), 7.47(1H, t, J=7.2Hz), 7.35-7.27(2H, m), 7.22-7.17(1H, m), 5.27(2H, s), 4.31-4.26(2H, m), 3.51-3.44(2H, m), 2.93-2.88(2H, m), 2.83(3H, s), 2.63-2.59(1H, m), 2.26(3H, s), 1.91-1.88(2H, m), 1.33-1.24(2H, m).
VIII-61		10.07(1H, s), 8.75(1H, s), 8.59(1H, s), 8.21(1H, d, J=7.2Hz), 7.96(1H, s), 7.80(1H, d, J=7.2Hz), 7.69(1H, d, J=8.8Hz), 7.48(1H, dd, J=8.4Hz, J=7.2Hz), 7.34-7.27(3H, m), 7.18(1H, t, J=7.2Hz), 5.27(2H, s), 4.32(2H, t, J=6.8Hz), 4.15(1H, bs), 3.72(1H, d, J=12.8Hz), 3.05(1H, t, J=10.8Hz), 2.94(1H, t, J=5.6Hz), 2.26(3H, s), 1.97(3H, s), 1.24(2H, bs).

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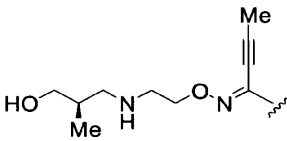
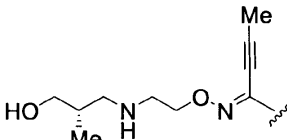
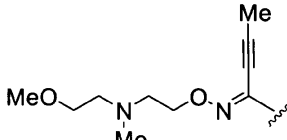
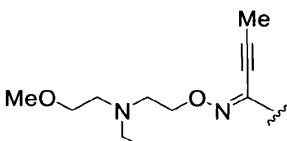
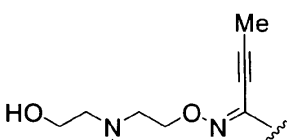
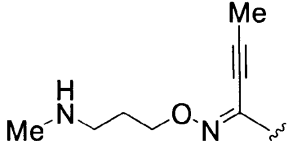
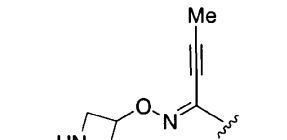
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【 0 1 0 3 】

【表 2 3】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-62		(二塩酸塩) 12.14(1H, bs), 9.16-9.08(1H, m), 8.91(2H, s), 8.44(1H, d, J=8.0Hz), 8.91(2H, s), 8.44(1H, d, J=8.0Hz), 8.02(1H, d, J=8.8Hz), 7.91(1H, s), 7.68(1H, d, J=8.4Hz), 7.51-7.46(1H, m), 7.37-7.31(2H, m), 7.21-7.18(1H, m), 5.32(2H, s), 4.62(2H, bs), 3.44-3.38(2H, m), 3.32-3.21(2H, m), 3.08-3.07(1H, m), 2.99-2.91(1H, m), 2.28(3H, s), 2.08-2.02(1H, m), 1.24-1.19(1H, m), 0.91(3H, d, J=6.4Hz).
VIII-63		10.08(1H, s), 8.75(1H, s), 8.68(1H, s), 8.22(1H, d, J=8.8Hz), 7.79(1H, s), 7.80(1H, d, J=7.2Hz), 7.70(1H, d, J=8.8Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.18(1H, t, J=8.8Hz), 5.27(2H, s), 4.34(2H, t, J=5.6Hz), 3.35-3.32(2H, m), 2.92(2H, t, J=5.6Hz), 2.65-2.61(1H, m), 2.26(3H, s), 1.75-1.70(1H, bs), 0.84(3H, d, J=5.6Hz).
VIII-64		10.09(1H, s), 8.58(1H, s), 8.22(1H, dd, J=8.7Hz, 1.8Hz), 7.95(1H, d, J=2.7Hz), 7.80(1H, d, J=9.0Hz), 7.68(1H, dd, J=8.7Hz, 2.7Hz), 7.51-7.44 (1H, m), 7.35-7.16(4H, m), 3.22(3H, s), 2.77(2H, t, J=5.7Hz), 2.60(2H, t, J=5.7Hz), 2.29(3H, s), 2.24 (3H, s).
VIII-65		10.09(1H, bs), 8.74(1H, s), 8.58(1H, s), 7.96(1H, d, J=2.1Hz), 7.80(1H, d, J=8.7Hz), 7.68(1H, d, J=9.0Hz), 7.48 (1H, dd, J=8.1Hz), 7.35-7.26(3H, m), 7.19(1H, t, J=8.1Hz), 5.27(2H, s), 4.32(2H, t, J=6.0Hz), 3.23(3H, s), 2.85(2H, t, J=6.0Hz), 2.87-2.59(4H, m), 2.24(3H, s), 0.98(3H, t, J=7.2Hz).
VIII-66		(二塩酸塩) 10.70(1H, brs), 9.92(1H, brs), 8.94(1H, s), 8.69(1H, s), 8.30(1H, d, J=12), 7.95-7.94(1H, m), 7.86(1H, d, J=12), 7.71-7.68(1H, m), 7.52-7.44(1H, m), 7.35-7.28(3H, m), 7.22-7.15(1H, m), 5.40(1H, br), 5.30(2H, s), 4.74-4.60(2H, m), 3.83-3.75(2H, m), 3.70-3.55(2H, m), 2.92(3H, s), 2.28(3H, s)
VIII-67		10.09(1H, s), 8.75(1H, s), 8.59(1H, s), 8.22(1H, d, J=8.7Hz), 7.95(1H, d, J=2.1Hz), 7.81 (1H, d, J=9.0Hz), 7.69(1H, d, J=8.7Hz), 7.51-7.34 (1H, m), 7.34-7.26(2H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.34(2H, t, J=6.3Hz), 2.96(2H, t, J=6.9Hz), 2.56(3H, s), 2.26(3H, s).
VIII-68		10.4(1H, s), 9.06(1H, s), 8.59(1H, s), 8.30(1H, d J=8Hz), 8.01(1H, s), 7.81(1H, d, J=8.8Hz), 7.74(1H, d, J=8.8Hz), 7.44-7.50(1H, m), 7.24-7.36(3H, m), 7.18(1H, t, J=7.2Hz), 5.27(2H, s), 4.21-4.36(4H, m), 3.98-4.04(1H, m), 2.25(3H, s)

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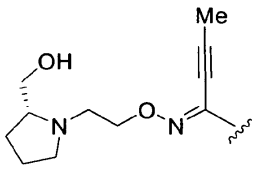
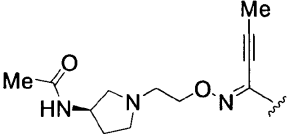
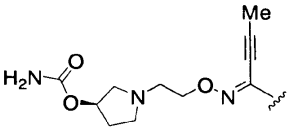
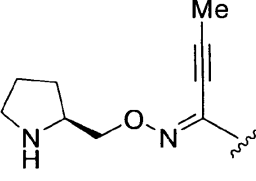
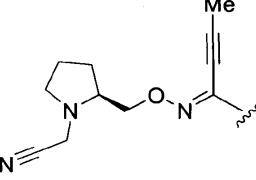
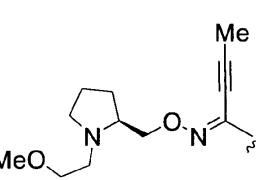
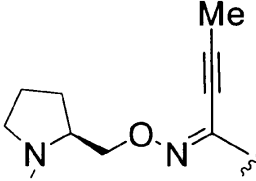
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【 0 1 0 4 】



【表 2 4】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-69		(二塩酸塩) 9.37(1H, s), 8.91(1H, s), 8.42(1H, d, 7.8Hz), 8.00(1H, d, J=6.6Hz), 7.92(1H, s), 7.69(1H, d, J=7.8Hz), 7.51-7.45(1H, m), 7.36-7.31(3H, m), 7.22-7.17(1H, m), 5.32(2H, s), 4.75-4.60(2H, m), 3.85-3.10(7H, m), 2.27(3H, s), 2.15-1.74(4H, m)
VIII-70		10.07(1H, bs), 8.74(1H, bs), 8.59(1H, s), 8.22(1H, d, J=8.8Hz), 7.98-7.96(2H, m), 7.80(1H, d, J=8.8Hz), 7.68(1H, d, J=8.8Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.20-7.16(1H, m), 5.27(2H, s), 4.37(2H, t, J=5.6Hz), 4.16(1H, bs), 2.79-2.66(4H, m), 2.46-2.42(3H, m), 2.25(3H, s), 2.06(1H, bs), 1.78(3H, s), 1.56-1.51(1H, m).
VIII-71		(二塩酸塩) 9.29(1H, m), 8.91(1H, s), 8.42(1H, m), 7.99(1H, d, 8.7Hz), 7.92(1H, s), 7.67(1H, d, J=9.0Hz), 7.53-7.45(1H, m), 7.37-7.31(3H, m), 7.23-7.17(1H, m), 6.68(2H, brs), 5.32(2H, s), 5.11(1H, m), 4.66(3H, m), 3.72-3.70(5H, m), 3.51(1H, m), 3.26(2H, m), 2.28(3H, s)
VIII-72		10.08(1H, bs), 8.74(1H, s), 8.56(1H, s), 8.21(1H, d, J=8.0Hz), 7.93(1H, s), 7.79(1H, d, J=8.8Hz), 7.67-7.65(1H, m), 7.50-7.40(1H, m), 7.33-7.24(2H, m), 7.20-7.12(1H, m), 5.25(2H, s), 4.36-4.30(1H, m), 4.22-4.17(1H, m), 4.08-3.92(2H, m), 2.98(2H, bs), 2.24(3H, s), 2.00-1.90(1H, m), 1.80-1.70(2H, m), 1.62-1.52(1H, m)
VIII-73		10.08(1H, bs), 8.80(1H, s), 8.57(1H, s), 8.21(1H, d, J=8.8Hz), 7.97(1H, s), 7.79(1H, d, J=8.8Hz), 7.71-7.68(1H, m), 7.46(1H, dd, J=8.0, J=14.4), 7.34-7.25(2H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.22-4.20(2H, m), 2.98-2.84(2H, m), 2.25(2H, s), 1.92-1.82(1H, m), 1.79-1.62(2H, m), 1.54-1.45(1H, m).
VIII-74		10.07(1H, bs), 8.74(1H, s), 8.58(1H, s), 8.21(1H, d, J=8.4Hz), 7.95(1H, s), 7.79(1H, d, J=8.8Hz), 7.69-7.67(1H, m), 7.47(1H, dd, J=8.4, J=15.2), 7.34-7.26(2H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.23-4.13(2H, m), 3.43(2H, t, J=6.0), 3.23(3H, s), 3.11-3.03(2H, m), 2.91-2.86(1H, m), 2.53-2.47(1H, m), 2.30-2.24(1H, m), 2.24(3H, s), 1.90-1.83(1H, m), 1.72-1.67(2H, m), 1.59-1.53(1H, m).
VIII-75		10.07(1H, s), 8.73(1H, s), 8.57(1H, s), 8.20(1H, d, J=8.8Hz), 7.95-7.94(1H, m), 7.79(1H, d, J=8.8Hz), 7.68-7.63(1H, m), 7.46(1H, dd, J=8.0, J=14.0), 7.33-7.25(2H, m), 7.19-7.14(1H, m), 5.26(2H, s), 4.27-4.18(2H, m), 2.95-2.92(1H, m), 2.62-2.57(1H, m), 2.41(3H, s), 2.23(3H, s), 2.19-2.15(1H, m), 1.96-1.87(1H, m), 1.74-1.61(2H, m), 1.59-1.53(1H, m).

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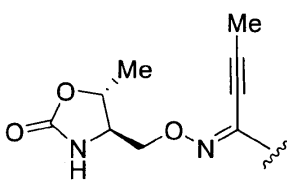
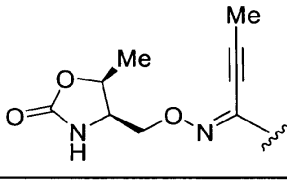
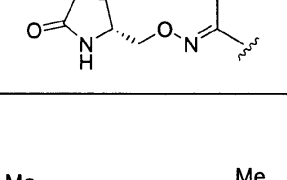
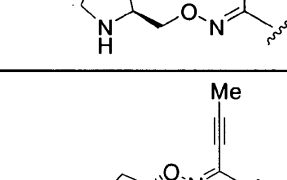
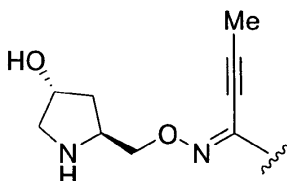
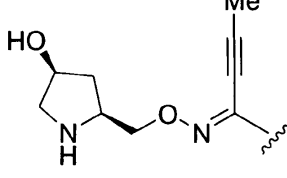

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【 0 1 0 5 】

【表 25】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-76		10.09(1H, bs), 8.76(1H, d, J=1.8Hz), 8.58(1H, s), 8.22(1H, dd, J=1.8Hz, J=8.7Hz), 7.96(1H, d, J=2.4Hz), 7.87-7.78(2H, m), 7.68(1H, dd, J=2.1Hz, J=9.0Hz), 7.52-7.44(1H, m), 7.35-7.26(3H, m), 7.22-7.16(1H, m), 5.27(2H, s), 4.57-4.49(1H, m), 4.35-4.27(2H, m), 3.77-3.71(1H, m), 2.26(3H, s), 1.36(3H, d, J=6.3Hz).
VIII-77		10.08(1H, bs), 8.75(1H, d, J=1.8Hz), 8.58(1H, s), 8.22(1H, dd, J=1.8Hz, J=9.0Hz), 7.96(1H, d, J=2.4Hz), 7.80(1H, d, J=9.0Hz), 7.76(1H, dd, J=2.4Hz, J=9.0Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.21-7.15(1H, m), 5.26(2H, s), 4.85-4.76(1H, m), 4.39(1H, dd, J=5.7Hz, J=11.4Hz), 4.30(1H, dd, J=4.5Hz, J=11.4Hz), 4.11-4.07(1H, m), 2.25(3H, s), 1.40(3H, d, J=6.6Hz).
VIII-78		10.07(1H, brs), 8.74(1H, s), 8.58(1H, s), 8.21(1H, d, 9.0Hz), 7.97(1H, s), 7.82-7.78(2H, m), 7.69(1H, d, 9.0Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.22-7.15(1H, m), 5.27(2H, s), 4.24(2H, d, 4.8 Hz), 3.91(1H, m), 2.31-2.06(6H, m), 1.95-1.87(1H, m)
VIII-79		10.09(1H, s), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d, J=8.8Hz), 7.97(1H, s), 7.91-7.89(1H, m), 7.80(1H, d, J=8.4Hz), 7.69(1H, d, J=8.8Hz), 7.51-7.45(1H, m), 7.35-7.27(3H, m), 7.20-7.17(1H, m), 5.27(2H, s), 4.26-4.06(3H, m), 3.46(1H, t, J=6.8Hz), 3.02-2.98(1H, m), 2.58(1H, dd, J=12.0Hz, J=5.6Hz), 2.26(3H, s), 2.20-2.10(1H, m), 1.77(3H, m), 1.38-1.31(1H, m).
VIII-80		10.3(1H, s), 8.95(1H, s), 8.57(1H, s), 8.22(1H, d, J=9.6), 8.01(1H, s), 7.80-7.73(2H, m), 7.47-7.41(1H, m), 7.34-7.25(2H, m), 7.20-7.12(1H, m), 5.37(1H, br), 5.26(2H, s), 5.10(1H, brs), 3.78-3.48(5H, m), 2.45-2.35(1H, m), 2.26(3H, s), 2.05-1.96(1H, m).
VIII-81		10.45(1H, s), 8.92(1H, s), 8.65(1H, s), 8.31(1H, d, J=8.4Hz), 7.97(1H, s), 7.77(1H, d, J=8.8Hz), 7.71(1H, d, J=9.2Hz), 7.51-7.45(1H, m), 7.35-7.28(2H, m), 7.19(1H, t, J=8.4Hz), 5.28(2H, s), 4.56-4.47(3H, m), 4.15(1H, bs), 3.60(1H, bs), 3.12(1H, bs), 2.28(3H, s), 2.09(1H, dd, J=13.2Hz, J=6.0Hz), 1.862-1.81(1H, m).
VIII-82		10.08(1H, s), 8.75(1H, s), 8.59(1H, s), 8.22(1H, d, J=8.8Hz), 7.97(1H, s), 7.80(1H, d, J=8.8Hz), 7.69(1H, d, J=8.0Hz), 7.51-7.45(1H, m), 7.35-7.27(m, 3H), 7.20-7.17(1H, m), 5.27(2H, s), 4.67(1H, bs), 4.29-4.19(3H, m), 3.45-4.40(1H, m), 2.88(1H, dd, J=11.2Hz, J=5.6Hz), 2.70(1H, dd, J=11.0, J=3.6Hz), 2.27(3H, s), 2.10-2.02(1H, m), 1.46-1.39(1H, m).

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【 0 1 0 6 】

【表 26】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-83		10.07(1H, brs), 8.74(1H, s), 8.58(1H, s), 8.21(1H, d, 8.7Hz), 7.97(1H, s), 7.82-7.78(2H, m), 7.69(1H, d, 9.0Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.22-7.15(1H, m), 5.27(2H, s), 4.24(2H, d, 4.8 Hz), 3.91(1H, m), 2.31-2.06(6H, m), 1.95-1.87(1H, m)
VIII-84		10.35(1H, s), 9.08(1H, s), 8.60(1H, s), 8.40(1H, s), 8.22(1H, d, J=8.4Hz), 8.09(1H, s), 7.83-7.82(2H, m), 7.49-7.47(1H, m), 7.33-7.19(4H, m), 5.27(2H, s), 4.43-4.16(4H, m), 3.01-2.98(2H, m), 2.28(3H, s), 1.92(2H, bs), 1.83(3H, s).
VIII-85		10.1(1H, brs), 8.82(1H, s), 8.58(1H, s), 8.21(1H, d, J=8.8), 7.98(1H, s), 7.80(1H, d, J=8.4), 7.72-7.70(1H, m), 7.50-7.44(1H, m), 7.34-7.26(3H, m), 7.20-7.15(1H, m), 5.26(2H, s), 4.22(2H, s), 3.57(1H, br), 2.96-2.89(2H, m), 2.26(3H, s), 1.98-1.62(3H, m), 1.58-1.45(1H, m).
VIII-86		10.07(1H, bs), 8.75(1H, s), 8.57(1H, s), 8.21(1H, d, J=8.8Hz), 7.95(1H, s), 7.79(1H, d, J=8.8Hz), 7.71-7.68(1H, m), 7.50-7.44(1H, m), 7.34-7.26(2H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.22-4.20(2H, m), 3.03-2.95(2H, m), 2.26(3H, s), 2.02-1.93(1H, m), 1.80-1.73(2H, m), 1.62-1.56(1H, m)
VIII-87		10.0(1H, brs), 8.73(1H, s), 8.57(1H, s), 8.19(1H, d, J=9.2), 7.95(1H, s), 7.79(1H, d, J=8.8), 7.69-7.67(1H, m), 7.50-7.44(1H, m), 7.34-7.26(3H, m), 7.20-7.16(1H, m), 5.26(2H, s), 4.24-4.17(2H, m), 3.77(1H, d, J=17.2), 3.58(1H, s), 3.50(1H, d, J=17.6), 3.22-3.12(1H, m), 3.10-3.00(1H, m), 2.58-2.52(1H, m), 2.23(3H, s), 1.98-1.89(1H, m), 1.77-1.70(2H, m), 1.59-1.52(1H, m).
VIII-88		10.8(1H, s), 8.98(1H, s), 8.58(1H, s), 8.20-8.16(2H, m), 7.91-7.88(1H, m), 7.80(1H, d, J=7.6), 7.50-7.44(1H, m), 7.34-7.30(2H, m), 7.23(1H, d, J=8.8), 7.20-7.15(1H, m), 5.26(2H, s), 4.23-4.41(2H, m), 3.76(1H, d, J=16.4), 3.52(1H, d, J=16.4), 3.61-3.52(1H, m), 3.45-3.36(1H, m), 2.91-2.85(1H, m), 2.25(3H, s), 2.09-2.02(1H, m), 1.91-1.79(2H, m), 1.73-1.66(1H, m).
VIII-89		10.87(1H, s), 8.97(1H, s), 8.58(1H, s), 8.20-8.15(2H, m), 7.91-7.88(1H, m), 7.80(1H, d, J=8.8), 7.50-7.44(1H, m), 7.34-7.30(2H, m), 7.23(1H, d, J=8.8), 7.20-7.15(1H, m), 5.26(2H, s), 4.22-4.41(2H, m), 3.76(1H, d, J=16.4), 3.52(1H, d, J=16.4), 3.61-3.52(1H, m), 3.45-3.36(1H, m), 2.91-2.85(1H, m), 2.25(3H, s), 2.09-2.02(1H, m), 1.91-1.79(2H, m), 1.73-1.66(1H, m).

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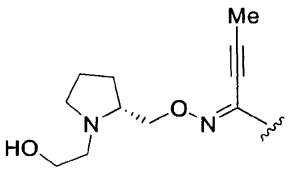
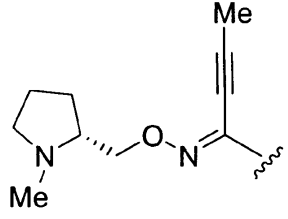
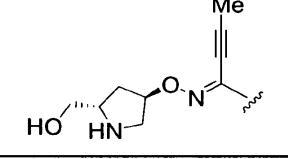
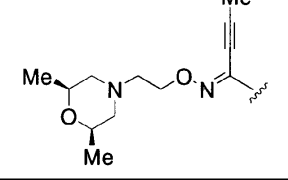
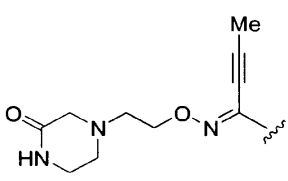
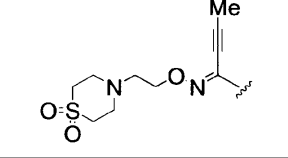
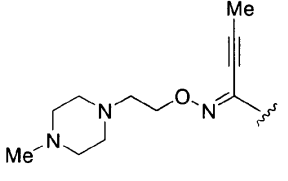
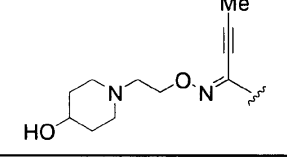
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【 0 1 0 7 】

【表 27】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-90		10.0(1H, s), 8.72(1H, s), 8.56(1H, s), 8.21-8.19(1H, m), 7.93(1H, s), 7.80-7.78(1H, m), 7.67-7.65(1H, m), 7.49-7.42(1H, m), 7.33-7.25(3H, m), 7.19-7.14(1H, m), 5.26(2H, s), 4.40(1H, brs), 4.27-4.19(1H, m), 4.16-4.09(1H, m), 3.10-2.94(2H, m), 2.90-2.80(1H, m), 2.23(3H, s), 1.93-1.82(1H, m), 1.75-1.62(2H, m), 1.61-1.52(1H, m).
VIII-91		10.08(1H, s), 8.74(1H, s), 8.56(1H, s), 8.21(1H, d, J=8.8Hz), 7.93(1H, s), 7.89(1H, d, J=8.8Hz), 7.68-7.63(1H, m), 7.46(1H, dd, J=8.0, J=14.0), 7.33-7.24(2H, m), 7.19-7.14(1H, m), 5.25(2H, s), 4.27-4.18(2H, m), 2.95-2.92(1H, m), 2.62-2.57(1H, m), 2.41(3H, s), 2.23(3H, s), 2.46-2.39(1H, m), 2.04-1.95(1H, m), 1.78-1.73(2H, m), 1.65-1.56(1H, m)
VIII-92		10.0(1H, s), 8.72(1H, s), 8.57(1H, s), 8.21(1H, d, J=8.4), 7.94(1H, s), 7.79(1H, d, J=8.8), 7.68-7.66(1H, m), 7.49-7.44(1H, m), 7.34-7.25(3H, m), 7.19-7.15(1H, m), 5.26(2H, s), 4.93(1H, brs), 3.02(1H, d, J=10.7), 2.25(3H, s), 2.09-2.01(1H, m), 1.77-1.70(1H, m)
VIII-93		10.07(1H, bs), 8.74(1H, bs), 8.59(1H, s), 8.22(1H, d, J=7.6Hz), 7.96(1H, d, J=2.4Hz), 7.80(1H, d, J=8.8Hz), 7.69(1H, dd, J=2.4Hz, J=8.8Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.20-7.17(1H, m), 5.27(2H, s), 4.40(2H, t, J=5.6Hz), 3.56(2H, t, J=8.0Hz), 2.85(2H, t, J=8.4Hz), 2.69(2H, t, J=5.6Hz), 2.25(3H, s), 1.04(6H, d, J=5.6Hz).
VIII-94		10.07(1H, bs), 8.74(1H, bs), 8.59(1H, s), 8.22(1H, d, J=8.8Hz), 7.97(1H, d, J=2.4Hz), 7.80(1H, d, J=8.8Hz), 7.72(1H, bs), 7.69(1H, dd, J=2.4Hz, J=8.8Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.41(2H, t, J=5.6Hz), 3.17(2H, bs), 3.10(2H, s), 2.80(2H, t, J=5.6Hz), 2.70(2H, t, J=5.6Hz), 2.26(3H, s).
VIII-95		10.27(1H, brs), 8.74(1H, s), 8.58(1H, s), 8.23-8.21(1H, m), 7.95(1H, s), 7.80(1H, d, J=8.8), 7.69-7.67(1H, m), 7.50-7.44(1H, m), 7.34-7.26(2H, m), 7.20-7.16(1H, m), 5.27(2H, s), 4.39(2H, t, J=5.6Hz), 3.18(8H, brs), 2.96(2H, t, J=5.6Hz), 2.55(3H, s)
VIII-96		10.1(1H, s), 8.79(1H, s), 8.59(1H, s), 8.21(1H, d, J=8Hz), 7.96(1H, s), 7.79(1H, d, J=8.8Hz), 7.68(1H, d, J=8.8Hz), 7.44-7.50(1H, m), 7.24-7.36(3H, m), 7.18(1H, t, J=7.2Hz), 5.27(2H, s), 4.37(2H, t, J=10Hz), 3.29-3.34(2H, m), 2.70(2H, t, J=6.4Hz), 2.26-2.35(4H, m), 2.25(3H, s), 2.14(3H, s)
VIII-97		10.1(1H, s), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d, J=8.8Hz), 7.96(1H, s), 7.80(1H, s), 7.69(1H, s), 7.42-7.50(1H, m), 7.13-7.36(4H, m), 5.27(2H, s), 4.52(1H, s), 4.36(2H, brs), 3.40-3.50(1H, brs), 2.80(2H, brs), 2.69(2H, brs), 2.10-2.27(5H, m), 1.69(2H, brs), 1.40(2H, brs)

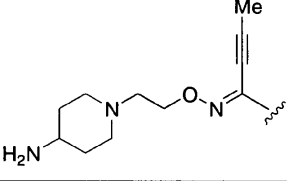
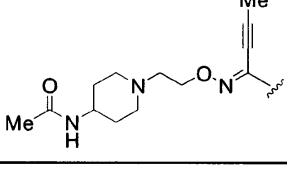
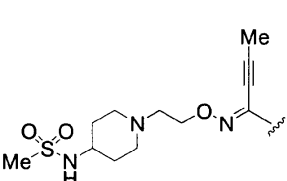
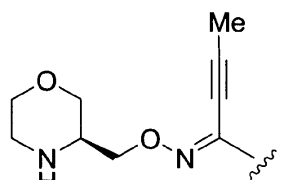
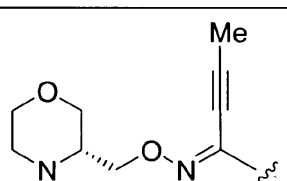
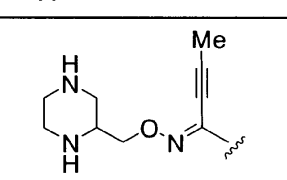
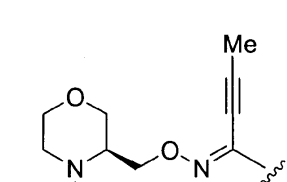
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【表 28】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-98		8.74(1H, s), 8.58(1H, s), 8.21(1H, d, J=9.2Hz), 7.96(1H, s), 7.79(1H, d, J=9.2Hz), 7.68(1H, d, J=9.2Hz), 7.48(1H, dd, J=5.8Hz, J=7.2Hz), 7.35-7.26(3H, m), 7.18(1H, t, J=9.2Hz), 5.27(2H, s), 4.36(2H, t, J=9.2Hz), 3.38(1H, dd, J=5.8Hz, J=7.2Hz), 2.86(2H, d, J=11.6Hz), 2.68(2H, t, J=6.0Hz), 2.25(3H, s), 2.06(2H, t, J=11.2Hz), 1.66(2H, d, J=11.7Hz), 1.09(2H, t, J=7.2Hz).
VIII-99		10.1(1H, bs), 8.78(1H, bs), 8.59(1H, s), 8.22(1H, d, J=8.0Hz), 7.97(1H, s), 7.81(1H, d, J=8.8Hz), 7.70(1H, d, J=6.4Hz), 7.49-7.45(1H, m), 5.27(2H, s), 4.49-4.12(2H, m), 3.41-3.38(1H, m), 2.82-2.65(2H, m), 2.25(3H, s), 1.79(3H, s), 1.09(2H, t, J=7.2Hz).
VIII-100		10.07(1H, s), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d, J=9.2Hz), 8.59(1H, s), 8.21(1H, d, J=9.2Hz), 7.96(1H, s), 7.80(1H, d, J=8.4Hz), 7.68(1H, d, J=8.4Hz), 7.50-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=8.0Hz), 7.04(1H, t, J=6.0Hz), 5.20(2H, s), 4.30(2H, bs), 3.10(1H, bs), 2.91(3H, s), 2.63(2H, d, J=2.4Hz), 2.55(3H, s), 2.03-1.99(2H, m), 1.77(2H, d, J=22.0Hz), 1.48-1.38(2H, m), 1.24(2H, bs).
VIII-101		10.09(1H, bs), 8.75(1H, s), 8.58(1H, s), 8.20(1H, d, J=8.7Hz), 7.95(1H, d, J=2.1Hz), 7.80(1H, d, J=8.7Hz), 7.68(1H, d, J=9.0Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.20-4.09(2H, m), 3.79(1H, dd, J=2.7Hz, J=10.8Hz), 3.68-3.64(1H, m), 3.13-3.08(1H, m), 2.82-2.71(3H, m), 2.26(3H, s).
VIII-102		10.11(1H, bs), 8.74(1H, s), 8.57(1H, s), 8.22-8.19(1H, m), 7.96(1H, m), 7.80(1H, d, J=9.0Hz), 7.69-7.66(1H, m), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.20-4.09(2H, m), 3.79(1H, dd, J=3.0Hz, J=10.8Hz), 3.68-3.64(1H, m), 3.25-3.18(1H, m), 3.13-3.05(1H, m), 2.82-2.71(2H, m), 2.26(3H, s).
VIII-103		10.09(1H, brs), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d, 9.0Hz), 7.96(1H, s), 7.79(1H, d, J=9.0Hz), 7.68(1H, d, J=9.0Hz), 7.49-7.44(1H, m), 7.34-7.22(3H, m), 7.19-7.15(1H, m), 5.27(2H, s), 4.11(2H, d, J=4.8Hz), 2.95-2.50(6H, m), 2.34-2.26(4H, m)
VIII-104		10.07(1H, s), 8.75(1H, d, J=1.8Hz), 8.58(1H, s), 8.22(1H, dd, J=1.8Hz, J=9.0Hz), 7.96(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.68(1H, dd, J=1.8Hz, J=9.3Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.21-7.15(1H, m), 5.27(2H, s), 4.35(1H, dd, J=2.7Hz, J=11.7Hz), 4.24(1H, dd, J=3.9Hz, J=11.7Hz), 4.12(1H, d, J=17.4Hz), 3.90-3.76(3H, m), 3.50(1H, dt, J=2.4Hz, J=11.1Hz), 2.83-2.71(2H, m), 2.26(3H, s).

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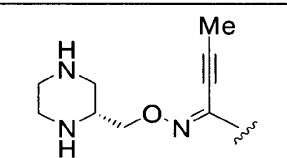
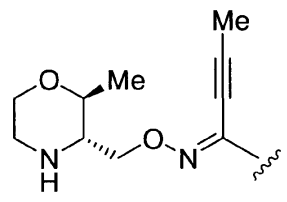
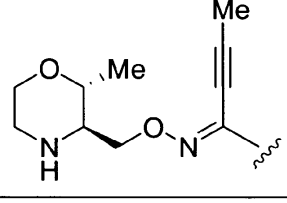
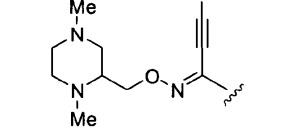
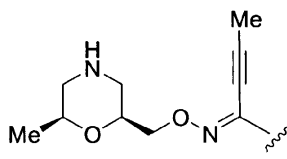
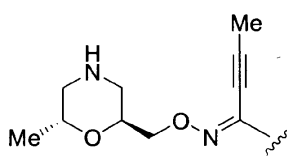
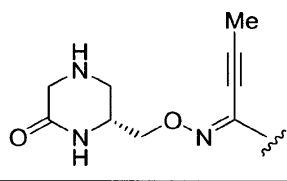
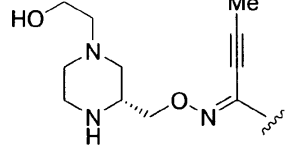
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【 0 1 0 9 】

【表 29】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-105		10.09(1H, brs), 8.73(1H, s), 8.58(1H, s), 8.20(1H, d, J=7.8Hz), 7.96(1H, s), 7.79(1H, d, J=9.0Hz), 7.69(1H, d, J=8.4Hz), 7.51-7.42(1H, m), 7.35-7.25(3H, m), 7.22-7.16(1H, m), 5.27(2H, s), 4.31(2H, d, 4.5 Hz), 2.95-2.53(5H, m), 2.34-2.10(5H, m)
VIII-106		10.1(1H, s), 8.75(1H, s), 8.59(1H, s), 8.21(1H, d, J=8.8Hz), 7.97(1H, s), 7.80(1H, d, J=8.4Hz), 7.69(1H, d, J=8.8Hz), 7.43-7.52(1H, m), 7.15-7.37(4H, m), 5.27(2H, s), 4.23-4.29(1H, m), 4.11-4.17(1H, m), 3.69(1H, d, J=12Hz), 3.37-3.45(1H, m), 3.23-3.32(1H, m), 2.66-2.80(3H, m), 2.28-2.40(1H, m), 2.26(3H, s), 1.16(3H, d, J=6Hz)
VIII-107		10.1(1H, s), 8.75(1H, s), 8.59(1H, s), 8.21(1H, d, J=8.8Hz), 7.97(1H, s), 7.80(1H, d, J=8.4Hz), 7.69(1H, d, J=8.8Hz), 7.43-7.52(1H, m), 7.15-7.37(4H, m), 5.27(2H, s), 4.23-4.29(1H, m), 4.11-4.17(1H, m), 3.69(1H, d, J=12Hz), 3.37-3.45(1H, m), 3.23-3.32(1H, m), 2.66-2.80(3H, m), 2.28-2.40(1H, m), 2.26(3H, s), 1.16(3H, d, J=6Hz)
VIII-108		10.09(1H, brs), 8.75(1H, s), 8.59(1H, s), 8.21(1H, d, J=10.5Hz), 7.96(1H, s), 7.80(1H, d, J=9.0Hz), 7.68(1H, d, J=9.0Hz), 7.52-7.44(1H, m), 7.35-7.25(3H, m), 7.22-7.16(1H, m), 5.27(2H, s), 4.44(1H, m), 4.18(1H, m), 2.80-2.60(4H, m), 2.32-2.00(12H, m)
VIII-109		10.1(1H, s), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d, J=8.0Hz), 7.96(1H, s), 7.80(1H, d, J=8.8Hz), 7.66-7.70(1H, m), 7.43-7.50(1H, m), 7.15-7.36(4H, m), 5.72(2H, s), 4.10-4.27(2H, m), 3.75-3.82(1H, m), 3.45-3.55(1H, m), 2.85(1H, d, J=10Hz), 2.74(1H, d, J=11Hz), 2.37(1H, t, J=11Hz), 2.26(4H, brs), 1.03(3H, d, J=6.4Hz).
VIII-110		10.1(1H, s), 8.74(1H, s), 8.58(1H, s), 8.21(1H, d, J=8.8Hz), 7.96(1H, s), 7.80(1H, d, J=8.4Hz), 7.68(1H, s, J=9.6Hz), 7.42-7.52(1H, m), 7.14-7.37(4H, m), 5.27(2H, s), 4.38-4.55(2H, m), 3.80-4.05(2H, m), 2.75-2.85(2H, m), 2.65-2.72(1H, m), 2.30-2.40(1H, m), 2.26(3H, s), 1.06(3H, d, J=5Hz)
VIII-111		10.08(1H, s), 8.74(1H, s), 8.59(1H, s), 8.20(1H, d, J=8.7Hz), 7.98(1H, d, J=0.9Hz), 7.83-7.72(2H, m), 7.10(1H, dd, J=2.7Hz, J=9.0Hz), 7.51-7.44(1H, m), 7.34-7.26(3H, m), 7.22-7.16(1H, m), 5.27(2H, s), 4.32(2H, m), 3.65(1H, m), 3.17(2H, s), 3.10-2.84(3H, m), 2.26(3H, s).
VIII-112		(二塩酸塩) 12.03(1H, brs), 8.93(1H, s), 8.46(1H, d, J=9.0Hz), 8.02(1H, d, J=8.7Hz), 7.92(1H, d, J=2.4Hz), 7.68-7.67(1H, m), 7.52-7.36(1H, m), 7.35-7.31(3H, m), 7.23-7.19(1H, m), 5.31(2H, s), 4.58(2H, m), 4.33(1H, brs), 3.99-3.23(12H, m), 2.29(3H, s).

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【 0 1 1 0 】

【表 30】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-113		10.09(1H, brs), 8.74(1H, s), 8.58(1H, s), 8.20(1H, d, J=8.7Hz), 7.96(1H, d, J=2.4Hz), 7.79(1H, d, J=8.7Hz), 7.68(1H, dd, J=2.7Hz, J=9.0Hz), 7.51-7.44(1H, m), 7.36-7.16(4H, m), 5.27(2H, s), 4.15(2H, m), 3.07(1H, m), 2.87-2.55(4H, m), 2.26(3H, s), 2.15(3H, s), 1.90(1H, m), 1.71(1H, m).
VIII-114		(二塩酸塩) 11.65(1H, bs), 9.46-9.35(2H, m), 9.04(1H, s), 8.88(1H, s), 7.95(1H, d, J=9.0Hz), 7.89(1H, d, J=2.4Hz), 7.66-7.62(1H, m), 7.52-7.45(1H, m), 7.36-7.31(3H, m), 7.23-7.17(1H, m), 5.31(2H, s), 4.35-4.34(2H, m), 4.24-4.21(1H, m), 4.04-3.98(1H, m), 3.85-3.77(1H, m), 3.05-2.85(3H, m), 2.28(3H, s).
VIII-115		
VIII-116		10.1(1H, s), 8.74(1H, s), 8.59(1H, s), 8.21(1H, d, J=8.0Hz), 7.96(1H, s), 7.80(1H, d, J=8.8Hz), 7.66-7.70(1H, m), 7.43-7.50(1H, m), 7.15-7.36(4H, m), 5.72(2H, s), 4.10-4.27(2H, m), 3.75-3.82(1H, m), 3.45-3.55(1H, m), 2.85(1H, d, J=10Hz), 2.74(1H, d, J=11Hz), 2.37(1H, t, J=11Hz), 2.26(4H, brs), 1.03(3H, d, J=6.4Hz).
VIII-117		10.1(1H, s), 8.74(1H, s), 8.58(1H, s), 8.21(1H, d, J=8.8Hz), 7.96(1H, s), 7.80(1H, d, J=8.4Hz), 7.68(1H, s, J=9.6Hz), 7.42-7.52(1H, m), 7.14-7.37(4H, m), 5.27(2H, s), 4.38-4.55(2H, m), 3.80-4.05(2H, m), 2.75-2.85(2H, m), 2.65-2.72(1H, m), 2.30-2.40(1H, m), 2.26(3H, s), 1.06(3H, d, J=5Hz)
VIII-118		10.10(1H, s), 8.76(1H, s), 8.59(1H, s), 8.22(1H, d, J=8.7Hz), 7.96(1H, d, J=2.1Hz), 7.81(2H, d, J=8.7Hz), 7.69(1H, dd, J=2.1Hz, J=8.7Hz), 7.44-7.52(1H, m), 7.27-7.35(2H, m), 7.16-7.22(1H, m), 5.27(2H, s), 4.20-4.35(2H, m), 3.84-3.98(1H, m), 2.85-3.15(6H, m), 2.28(3H, m).
VIII-119		(二塩酸塩) 9.14(1H, s), 8.91(1H, s), 8.61-8.55(1H, m), 8.41(1H, d, J=9.0Hz), 8.22(1H, d, J=9.0Hz), 8.00-7.97(1H, m), 7.91(1H, s), 7.66(1H, d, J=9.0Hz), 7.52-7.45(1H, m), 7.37-7.30(3H, m), 7.23-7.17(1H, m), 5.32(2H, s), 4.40(2H, m), 4.18(1H, m), 3.65-3.20(4H, m), 2.287(3H, s)
VIII-120		10.1(1H, s), 8.71(1H, s), 8.59(1H, s), 8.16(1H, d, J=8.8Hz), 7.98(1H, s), 7.80(1H, d, J=9.2Hz), 7.69(1H, d, J=9.2Hz), 7.44-7.51(1H, m), 7.14-7.36(4H, m), 5.27(2H, s), 4.31(2H, t, J=5.6Hz), 2.85(2H, t, J=5.6Hz), 2.35(3H, s), 1.70-1.78(1H, m), 1.02-1.07(2H, m), 0.90-0.95(2H, m)

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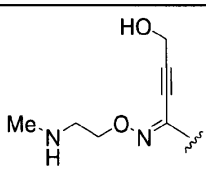
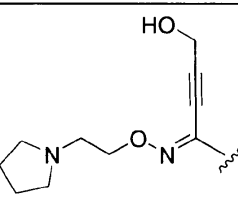
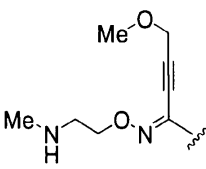
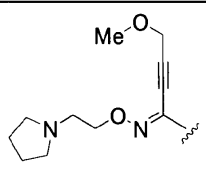
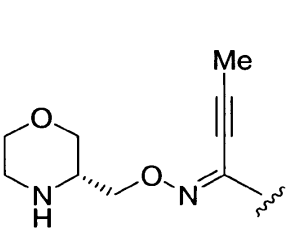
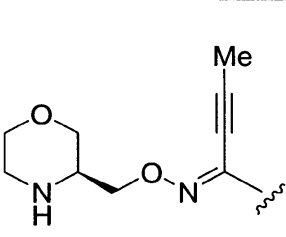
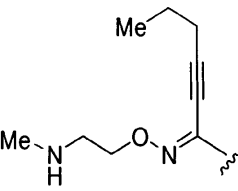
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【 0 1 1 1 】

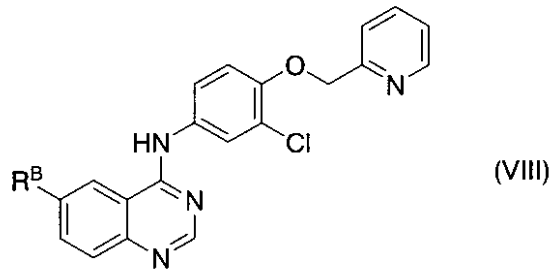
【表 3 1】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)	
VIII-121		10.1(1H, s), 8.82(1H, s), 8.61(1H, s), 8.27(1H, d, J=7.6Hz), 7.97(1H, s), 7.84(1H, d, J=9.6Hz), 7.70(1H, d, J=7.6Hz), 7.43-7.51(1H, m), 7.25-7.36(3H, m), 7.15-7.22(1H, m), 5.62(1H, s), 5.72(2H, s), 4.49-4.55(4H, m), 3.29-3.40(2H, m), 2.66(3H, s)	
VIII-122		10.05(1H, s), 8.76(1H, s), 8.60(1H, s), 8.20(1H, d, J=9.2Hz), 7.97(1H, s), 7.82(1H, d, J=8.8Hz), 7.68(1H, d, J=9.2Hz), 7.53-7.45(1H, m), 7.37-7.27(3H, m), 7.19(1H, t, J=8.4Hz), 5.63(1H, t, J=5.2Hz), 5.27(2H, s), 4.70(2H, d, J=5.2Hz), 4.40(2H, t, J=6.0Hz), 2.81(2H, t, J=6.0Hz), 2.53(4H, bs), 1.68(4H, bs).	10
VIII-123		10.02 (1H, s), 8.89 (1H, s), 8.60(1H, s), 8.22(1H, d, J=9.3Hz), 8.00(1H, s), 7.83(1H, d, J=8.7Hz), 7.73(1H, d, J=9.6Hz), 7.51-7.44(1H, m), 7.35-7.29(2H, m), 7.22-7.16(1H, m), 5.27(2H, s), 4.53(2H, s), 3.40(5H, bs), 2.51(5H, bs).	
VIII-124		10.13(1H, s), 8.80(1H, s), 8.61(1H, s), 8.21(1H, d, J=8.4Hz), 7.99(1H, s), 7.83(1H, d, J=8.8Hz), 7.71(1H, d, J=8.8Hz), 7.51-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=8.4Hz), 5.27(2H, s), 4.53(2H, s), 4.46(2H, t, J=5.6Hz), 2.93(2H, bs), 2.65(4H, bs), 1.72(4H, bs).	20
VIII-125		10.09(1H, s), 8.76(1H, bs), 8.59(1H, s), 8.19(1H, d, J=9.0Hz), 7.96(1H, bs), 7.82(1H, d, J=8.7Hz), 7.67(1H, bd, J=7.5Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.21-7.16(1H, m), 5.27(2H, s), 4.52(2H, s), 4.23(1H, dd, J=5.7Hz, J=10.8Hz), 4.17(1H, dd, J=6.3Hz, J=10.8Hz), 3.79(1H, dd, J=3.0Hz, J=10.8Hz), 3.65(1H, bd, J=7.8Hz), 3.40(3H, s), 3.27-3.20(1H, m), 3.13-3.04(1H, m).	
VIII-126		10.10(1H, bs), 8.76(1H, d, J=1.5Hz), 8.59(1H, s), 8.19(1H, dd, J=1.8Hz, J=8.7Hz), 7.96(1H, d, J=2.1Hz), 7.83(1H, d, J=9.0Hz), 7.71-7.64(1H, m), 7.52-7.44(1H, m), 7.35-7.27(3H, m), 7.22-7.16(1H, m), 5.27(2H, s), 4.53(2H, s), 4.23(1H, dd, J=6.3Hz, J=11.4Hz), 4.17(1H, dd, J=6.3Hz, J=10.8Hz), 3.80(1H, dd, J=3.0Hz, J=10.8Hz), 3.68-3.64(1H, m), 3.12-3.07(1H, m), 2.82-2.70(2H, m).	30
VIII-127		10.1(1H, s), 8.76(1H, s), 8.59(1H, t, J=5.6Hz), 8.20(1H, d, J=8.8Hz), 7.98(1H, s), 7.81(1H, d, J=8.8Hz), 7.70(1H, d, J=8.8Hz), 7.44-7.51(1H, m), 7.15-7.37(4H, m), 5.27(2H, s), 4.34(2H, t, J=6Hz), 2.86(2H, t, J=5.6Hz), 2.61(2H, t, J=7.2Hz), 2.36(3H, s), 1.67(2H, q, J=7.2Hz), 1.05(3H, t, J=7.2Hz)	40

【 0 1 1 2 】



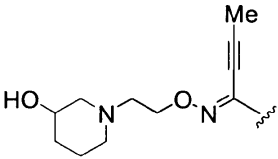
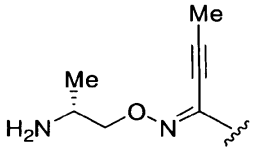
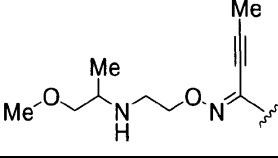
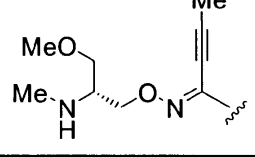
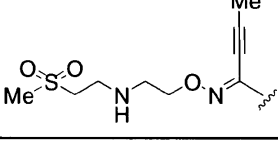
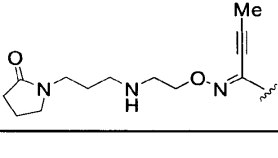
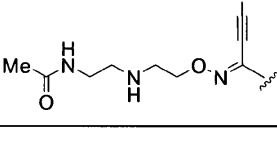
【化 8 2】



(VIII)

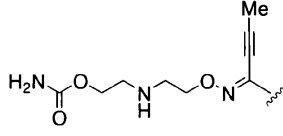
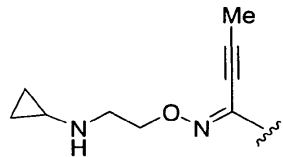
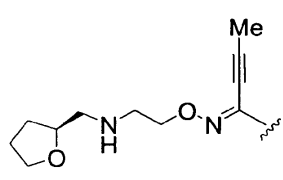
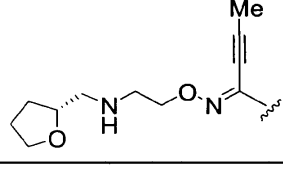
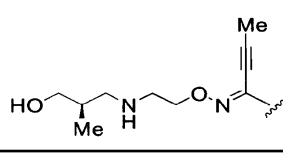
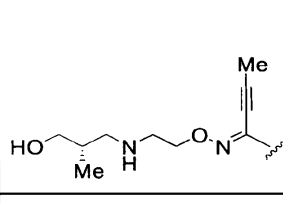
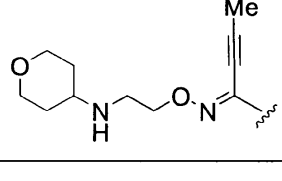
【 0 1 1 3】

【表 3 2】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)	
VIII-128		10.0(1H, s), 8.74(1H, s), 8.60-8.58(2H, m), 8.21(1H, d, J=8.8), 7.96(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.8), 7.68-7.66(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=8.8), 5.30(2H, s), 4.61-4.60(1H, m), 4.37-4.35(2H, m), 3.52-3.38(1H, m), 2.93-2.92(1H, m), 2.76-2.66(2H, m), 2.24(3H, s), 2.00-1.95(1H, m), 1.89-1.84(1H, m), 1.79-1.77(1H, m), 1.62-1.59(1H, m), 1.45-1.36(1H, m), 1.10-1.02(1H, m).	10
VIII-129		10.09(1H, s), 8.61-8.59(2H, m), 8.21(1H, d, J=9.2Hz), 8.07(1H, s), 7.89(1H, d, J=7.6Hz), 7.80(1H, d, J=8.4Hz), 7.68(1H, d, J=9.2Hz), 7.37(1H, t, J=7.6Hz), 7.28(1H, d, J=8.8Hz), 5.31(2H, s), 4.60(2H, d, J=6.0Hz), 2.27(3H, s), 0.89(3H, d, J=7.6Hz).	
VIII-130		(三塩酸塩) 12.58(1H, s), 9.44(1H, s), 9.33(1H, bs), 9.09(1H, bs), 8.95(1H, s), 8.78(1H, d, J=4.4Hz), 8.46(1H, d, J=8.8Hz), 8.23(1H, t, J=6.8Hz), 8.09(1H, d, J=8.4Hz), 7.94(1H, d, J=2.4Hz), 7.86(1H, d, J=8.0Hz), 7.72-7.67(2H, m), 7.39(1H, d, J=9.2Hz), 5.51(2H, s), 4.61(2H, bs), 3.61-3.47(5H, m), 2.28(3H, s), 1.28(3H, d, J=6.4Hz).	20
VIII-131		10.0(1H, s), 8.74(1H, s), 8.60-8.58(2H, m), 8.20(1H, d, J=8.8), 7.97(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.4), 7.69-7.67(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=9.2), 5.30(2H, s), 4.24-4.23(2H, m), 3.44-3.34(2H, m), 2.97-2.92(1H, m), 2.38(3H, s), 2.26(3H, s).	
VIII-132		10.08(1H, s), 8.98(1H, s), 8.61-8.59(2H, m), 8.23(1H, d, J=8.8Hz), 7.98(1H, s), 7.89(1H, t, J=7.2Hz), 7.80(1H, d, J=8.8Hz), 7.69(1H, d, J=9.2Hz), 7.59(1H, t, J=8.0Hz), 7.37(1H, d, J=7.6Hz), 7.28(1H, d, J=8.8Hz), 5.31(2H, s), 4.33(2H, t, J=4.8Hz), 3.26(2H, t, J=6.4Hz), 3.03(5H, bs), 2.94(2H, bs), 2.26(3H, s).	30
VIII-133		10.0(1H, s), 8.74(1H, s), 8.60-8.58(2H, m), 8.21(1H, d, J=8.0), 7.97(1H, m), 7.90-7.86(1H, m), 7.80(1H, d, J=8.8), 7.69-7.67(1H, m), 7.59(1H, d, J=8.0), 7.38-7.35(1H, m), 7.28(1H, d, J=9.2), 5.30(2H, s), 4.32-4.29(2H, m), 3.22-3.18(2H, m), 2.90-2.87(2H, m), 2.57-2.53(2H, m), 2.25(3H, s), 2.20-2.16(2H, m), 1.91-1.84(2H, m), 1.62-1.55(1H, m)	
VIII-134		10.11(1H, brs), 8.75(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.91-7.78(3H, m), 7.67(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 5.31(2H, s), 4.32(2H, m), 3.13(2H, m), 2.93(2H, m), 2.66(2H, m), 2.26(3H, s), 1.78(3H, s)	40

【 0 1 1 4 】

【表 3 3】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-135		10.09(1H, brs), 8.75(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.81(1H, d, J=9.0Hz), 7.69(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 6.47(2H, brs), 5.31(2H, s), 4.34 (2H, m), 3.99(2H, m), 2.99(2H, m), 2.84(2H, m), 2.26(3H, s)
VIII-136		10.10(1H, brs), 8.75(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.81(1H, d, J=9.0Hz), 7.69(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 5.31(2H, s), 4.33 (2H, t, 6.0Hz), 3.00(2H, t, 6.0Hz), 2.25(3H, s), 2.23(1H, m), 0.408(2H, m), 0.29(2H, m)
VIII-137		(三塩酸塩) 12.14(1H, brs), 9.28(1H, s), 8.93(1H, s), 8.64(1H, d, J=4.2Hz), 8.44(1H, d, J=10.5Hz), 8.02-7.91(3H, m), 7.69-7.62(1H, m), 7.46-7.42(1H, m), 7.37(1H, d, 9.0Hz), 5.38(2H, s), 4.59(2H, m), 4.19(1H, m), 3.81(1H, m), 3.71(1H, m), 3.47(2H, m), 3.21(1H, m), 3.02(1H, m), 2.28(3H, s), 2.02(1H, m), 1.84(2H, m), 1.56(1H, m)
VIII-138		(三塩酸塩) 9.28(1H, s), 8.93(1H, s), 8.64(1H, d, J=4.2Hz), 8.44(1H, d, J=10.5Hz), 8.02-7.91(3H, m), 7.69-7.62(1H, m), 7.46-7.42(1H, m), 7.37(1H, d, 9.0Hz), 5.38(2H, s), 4.59(2H, m), 4.19(1H, m), 3.81(1H, m), 3.71(1H, m), 3.47(2H, m), 3.21(1H, m), 3.02(1H, m), 2.28(3H, s), 2.02(1H, m), 1.84(2H, m), 1.56(1H, m)
VIII-139		10.08(1H, s), 8.75(1H, s), 8.61-8.58(2H, m), 8.22(1H, d, J=8.8Hz), 7.98(1H, s), 7.89(1H, t, J=8.0Hz), 7.68 (1H, d, J=8.4Hz), 7.37(1H, t, J=6.4Hz), 7.37(1H, t, J=6.4Hz), 7.28(1H, d, J=8.4Hz), 5.31(2H, s), 4.34(2H, t, J=6.4Hz), 3.33(1H, t, J=6.4Hz), 2.92(2H, t, J=5.2Hz), 2.65-2.61(1H, m), 2.26(3H, s), 1.75-1.70(1H, m), 0.83(3H, d, J=6.8Hz).
VIII-140		10.09(1H, s), 8.75(1H, s), 8.59(2H, bs), 8.22(1H, d, J=8.8Hz), 7.98(1H, s), 7.89(1H, t, J=6.4Hz), 7.80 (1H, d, J=8.8Hz), 7.69(1H, t, J=8.4Hz), 7.59(1H, t, J=8.4Hz), 7.37(1H, d, J=5.2Hz), 7.28(1H, d, J=8.8Hz), 5.31(2H, s), 4.34(2H, t, J=6.4Hz), 3.32(2H, t, J=6.4Hz), 2.92(2H, t, J=5.6Hz), 2.65-2.61(1H, m), 2.26(3H, s), 1.75-1.68(1H, m), 1.24(1H, bs), 0.83(3H, d, J=6.8Hz).
VIII-141		(三塩酸塩) 12.48(1H, s), 9.48(1H, s), 9.42(1H, bs), 9.29(2H, bs), 8.94(1H, s), 8.72(1H, s), 8.47(1H, s), 8.13-8.06 (1H, m), 7.92(1H, s), 7.76(1H, bs), 7.68(1H, bs), 7.59(1H, bs), 7.39(1H, bs), 5.46(2H, s), 4.62 (2H, bs), 3.88(4H, bs), 2.28(3H, s), 2.01-1.94(4H, m), 1.69-1.60(4H, m).

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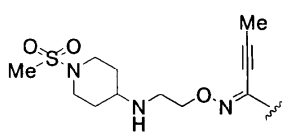
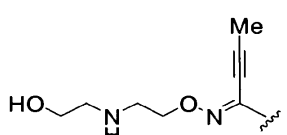
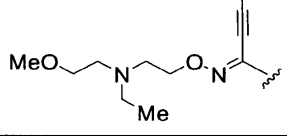
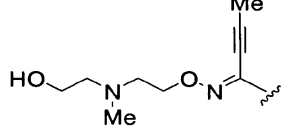
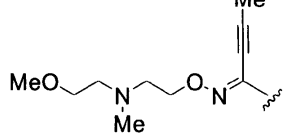
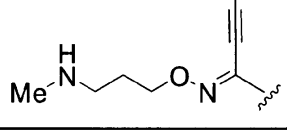
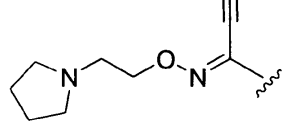
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【 0 1 1 5 】

【表 3 4】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-142		10.07(1H, s), 8.74(1H, s), 8.59(2H, s), 8.20(1H, bs), 7.97(1H, s), 7.89(1H, bs), 7.80(1H, d, J=8.0Hz), 7.68 (1H, bs), 7.58(1H, d, J=8.8Hz), 7.38(1H, bs), 7.28(1H, d, J=9.2Hz), 5.31(2H, s), 4.31(2H, bs), 3.45(2H, bs), 2.94(2H, bs), 2.85-2.81(5H, m), 2.26(3H, s), 1.89-1.85(2H, m), 1.36-1.28(2H, m).
VIII-143		10.09(1H, brs), 8.74(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.81(1H, d, J=9.0Hz), 7.69(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 5.31(2H, s), 4.23(2H, t, 5.4Hz), 3.47(2H, t, 5.4Hz), 2.91(2H, t, 5.4Hz), 2.65(2H, m), 2.25(3H, s)
VIII-144		(三塩酸塩) 12.4(1H, s), 10.5(1H, s), 9.34(1H, s), 8.93(1H, s), 8.68-8.67(1H, m), 8.45(1H, d, J=9.2), 8.05-8.02(2H, m), 7.92-7.91(1H, m), 7.71-7.66(2H, m), 7.52-7.49(1H, m), 7.37(1H, d, J=8.8), 5.41(2H, s), 4.73-4.67(1H, m), 3.30(6H, s), 2.27 (3H, s), 1.29-1.25(1H, m).
VIII-145		10.08(1H, s), 8.75(1H, s), 8.59(2H, bs), 8.22(1H, d, J=8.4Hz), 7.98(1H, bs), 7.89(1H, t, J=8.0Hz), 7.80(1H, d, J=9.2Hz), 7.68 (1H, d, J=8.8Hz), 7.38(1H, d, J=6.4Hz), 7.28(1H, d, J=8.8Hz), 5.31(2H, s), 4.36(4H, t J=6.4Hz), 3.49(2H, bs), 2.79(2H, t, J=6.0Hz), 2.30(3H, s), 2.25(3H, s).
VIII-146		(三塩酸塩) 12.70(1H, s), 10.83(1H, bs), 9.49(1H, s), 8.95(1H, s), 8.77(1H, bs), 8.44(1H, d, J=8.84Hz), 8.23 (1H, bs), 8.10(1H, d, J=8.8Hz), 7.95(1H, s), 7.85(1H, d, J=6.8Hz), 7.74-7.68(2H, m), 7.39(1H, d, J=8.4Hz), 5.51(2H, s), 4.70 (2H, bs), 7.75-7.42(4H, m), 3.48-3.44(1H, m), 3.40(1H, bs), 3.30(3H, s), 2.90(3H, s), 2.28(3H, s).
VIII-147		10.28-10.03(1H, m), 8.75(1H, s), 8.61-8.58(2H, m), 8.21(1H, d, J=8.8Hz), 7.98(1H, s), 7.89(1H, t, J=8.0Hz), 7.79(1H, d, J=7.2Hz), 7.68(1H, d, J=8.0Hz), 7.37(1H, t, J=6.0Hz), 7.28(1H, t, J=8.8Hz), 5.31(2H, s), 4.33(2H, t J=6.4Hz), 2.31(3H, s), 2.06(3H, s), 1.88(2H, t, J=6.4Hz).
VIII-148		10.09(1H, brs), 8.75(1H, s), 8.61-8.57(2H, m), 8.22(1H, d, J=9.0Hz), 7.98(1H, s), 7.92-7.86(1H, m), 7.80(1H, d, J=9.0Hz), 7.68(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.28(1H, d, J=9.0Hz), 5.31(2H, s), 4.37(2H, m), 2.82(2H, m), 2.54(4H, m), 2.25(3H, s), 1.69(4H, m)

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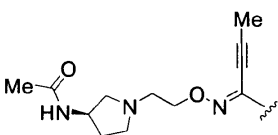
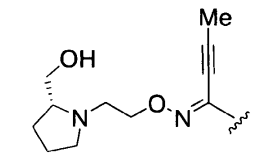
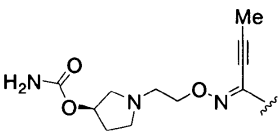
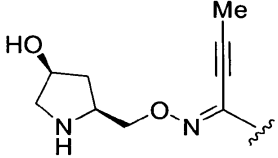
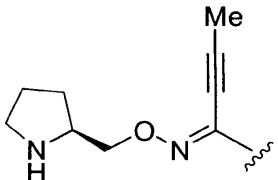
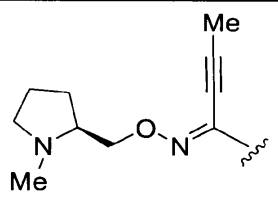
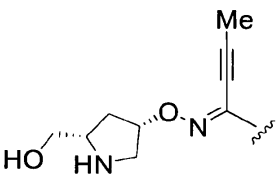
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【 0 1 1 6 】

【表 3 5】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-149		10.09(1H, s), 8.75(1H, s), 8.70(2H, s), 8.20(1H, d, J=8.0Hz), 8.07(2H, s), 7.98(1H, t, J=4.8Hz), 7.88(1H, t, J=8.0Hz), 7.68(1H, d, J=8.0Hz), 7.38(1H, s), 7.28(1H, d, J=9.6Hz), 5.31(2H, s), 4.42(2H, s), 4.29-4.24(1H, m), 2.91-2.76(2H, m), 2.25(3H, s), 2.17-2.12(1H, m), 1.75(3H, s), 1.53-1.34(1H, m), 1.11(2H, d, J=4.4Hz).
VIII-150		10.09(1H, s), 8.75(1H, s), 8.59(2H, s), 8.21(1H, bs), 7.98(1H, s), 7.89(1H, s), 7.79(1H, s), 7.68(1H, s), 7.58(1H, s), 7.37(1H, s), 7.28(1H, s), 5.31(2H, s), 4.36(2H, bs), 2.71(2H, bs), 2.25(3H, s), 1.81(1H, bs), 1.66(2H, bs), 1.52(1H, bs), 1.24(2H, bs).
VIII-151		10.09(1H, brs), 8.75(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.80(1H, d, J=9.0Hz), 7.69(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.28(1H, d, J=9.0Hz), 6.48(2H, brs), 5.31(2H, s), 4.94(1H, m), 4.37(2H, m), 2.81-2.63(5H, m), 2.44(1H, m), 2.25(3H, s), 2.14(1H, m), 1.69(1H, m)
VIII-152		10.13(1H, brs), 8.79(1H, s), 8.61-8.58(2H, m), 8.23(1H, dd, J=1.8Hz, J=8.7Hz), 7.98(1H, d, J=2.7Hz), 7.92-7.86(1H, m), 8.80(1H, d, J=8.7Hz), 7.79(1H, dd, J=2.7Hz, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.39-7.35(1H, m), 7.28(1H, d, J=9.0Hz), 5.31(2H, s), 4.91(1H, brs), 4.36-4.24(3H, m), 3.59(1H, m), 2.99-2.81(2H, m), 2.26(3H, s), 2.13(1H, m), 1.50(1H, m).
VIII-153		10.0(1H, br), 8.64(1H, s), 8.62-8.55(2H, m), 8.20(1H, d, J=8.8), 7.96(1H, s), 7.90-7.87(1H, m), 7.79(1H, d, J=8.4), 7.68-7.66(1H, m), 7.58(1H, d, J=8.4), 7.38-7.35(1H, m), 7.27(1H, d, J=8.8), 5.30(2H, s), 4.18-4.08(2H, m), 3.43-3.40(1H, m), 3.69(3H, s), 2.84-2.77(2H, m), 2.25(3H, s), 1.88-1.55(3H, m), 1.50-1.38(1H, m).
VIII-154		10.08(1H, br), 8.75(1H, s), 8.60-8.58(2H, m), 8.21-8.19(1H, m), 7.98-7.97(1H, m), 7.93-7.86(1H, m), 7.79(1H, d, J=8.8), 7.70-7.67(1H, m), 7.58(1H, d, J=8.0), 7.38-7.35(1H, m), 7.27(1H, d, J=9.2), 5.30(2H, s), 4.30-4.20(2H, m), 3.01-2.96(1H, m), 2.68(1H, br), 2.41(3H, s), 2.24(3H, s), 1.99-1.89(1H, m), 1.73-1.66(2H, m), 1.62-1.53(1H, m).
VIII-155		10.0(1H, brs), 8.73(1H, s), 8.60-8.58(2H, m), 8.31(1H, s), 8.22-8.20(1H, m), 7.96(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.8), 7.68-7.66(1H, m), 7.59(1H, d, J=7.6), 7.38-7.35(1H, m), 7.28(1H, d, J=9.2), 5.30(2H, s), 4.93(1H, brs), 3.02(1H, d, J=11.6), 2.24(3H, s), 2.06-2.01(1H, m), 1.77-1.70(1H, m).

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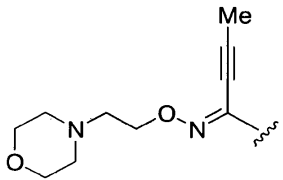
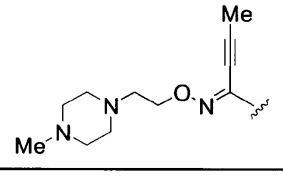
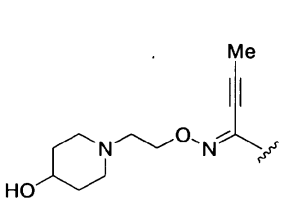
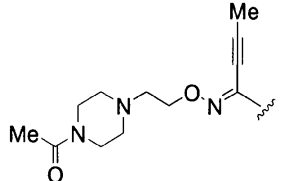
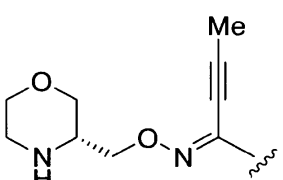
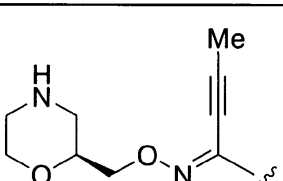
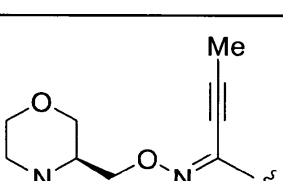
【 0 1 1 7 】

【表 3 6】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)	
VIII-156		10.13(1H, brs), 8.81(1H, s), 8.61-8.59(2H, m), 8.24(1H, d, J=10.5Hz), 7.99(1H, s), 7.92-7.88(1H, m), 7.81(1H, d, J=9.0Hz), 7.70(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.38-7.36(1H, m), 7.28(1H, d, J=9.0Hz), 5.31(2H, s), 4.92(1H, s), 4.28(3H, m), 3.82(1H, m), 3.09(1H, d, 11.4Hz), 3.83(1H, d, 11.4Hz), 2.27(3H, s), 1.87(1H, m), 1.65(1H, m)	
VIII-157		10.03(1H, br), 8.73(1H, s), 8.59-8.58(1H, m), 8.55(1H, s), 8.20(1H, d, J=8.8), 7.94(1H, s), 7.89-7.85(1H, m), 7.78(1H, d, J=8.4), 7.66-7.64(1H, m), 7.57(1H, d, J=7.6), 7.38-7.35(1H, m), 7.25(1H, d, J=8.8), 5.28(2H, s), 4.20-4.08(2H, m), 2.85-2.74(2H, m), 2.24(3H, s), 1.82-1.76(1H, m), 1.73-1.58(2H, m), 1.48-1.39(1H, m)	10
VIII-158		10.07(1H, br), 8.74(1H, s), 8.60-8.58(2H, m), 8.22-8.19(1H, m), 7.98-7.97(1H, m), 7.93-7.86(1H, m), 7.80(1H, d, J=9.2), 7.70-7.67(1H, m), 7.59(1H, d, J=7.6), 7.38-7.35(1H, m), 7.28(1H, d, J=9.2), 5.30(2H, s), 4.30-4.20(2H, m), 3.01-2.96(1H, m), 2.65(1H, br), 2.40(3H, s), 2.24(3H, s), 1.99-1.89(1H, m), 1.73-1.66(2H, m), 1.62-1.53(1H, m).	20
VIII-159		10.08(1H, s), 8.73(1H, s), 8.59-8.56(2H, m), 8.21-8.19(1H, m), 7.94(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.8), 7.66-7.64(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.26(1H, d, J=9.2), 5.29(2H, s), 4.44(1H, brs), 4.25-4.21(1H, m), 4.15-4.11(1H, m), 3.05-2.96(2H, m), 2.46-2.39(1H, m), 2.23(2H, s), 1.92-1.82(1H, m), 1.73-1.63(2H, m), 1.59-1.54(1H, m).	
VIII-160		10.0(1H, s), 8.72(1H, s), 8.61-8.53(2H, m), 8.21-8.19(1H, m), 7.94(1H, s), 7.89-7.86(1H, m), 7.79(1H, d, J=8.8), 7.66-7.64(1H, m), 7.58(1H, d, J=8.0), 7.38-7.35(1H, m), 7.26(1H, d, J=9.2), 5.29(2H, s), 4.44(1H, brs), 4.25-4.21(1H, m), 4.15-4.11(1H, m), 3.05-2.96(2H, m), 2.45-2.39(1H, m), 2.23(3H, s), 1.92-1.82(1H, m), 1.73-1.63(2H, m), 1.60-1.53(2H, m).	30
VIII-161		10.0(1H, brs), 8.73(1H, s), 8.60-8.57(2H, m), 8.21(1H, d, J=8.4), 7.95(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.8), 7.67-7.65(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=8.8), 5.30(2H, s), 4.93(1H, brs), 3.02(1H, d, J=11.6), 2.24(3H, s), 2.06-2.01(1H, m), 1.77-1.70(1H, m)	
VIII-162		10.07(1H, s), 8.75(1H, s), 8.61-8.59(2H, m), 8.23(1H, d, J=8.8Hz), 7.98(1H, d, J=2.4Hz), 7.89(1H, t, J=8.4Hz), 7.80(1H, d, J=8.8Hz), 7.68(1H, d, J=6.4Hz), 7.57(1H, t, J=11.8Hz), 7.28(1H, d, J=8.8Hz), 5.31(2H, s), 4.40(2H, t, J=5.2Hz), 3.32(4H, s), 3.09(4H, bs), 2.96(2H, t, J=5.6Hz), 2.26(3H, s).	40

【 0 1 1 8 】

【表 3 7】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-163		10.07(1H, s), 8.75(1H, s), 8.59(2H, bs), 8.21(1H, d, J=4.8Hz), 7.98(1H, s), 7.89(1H, t, J=7.6Hz), 7.80(1H, d, J=8.4Hz), 7.68(1H, d, J=8.4Hz), 7.60(1H, t, J=8.4Hz), 7.37(1H, t, J=7.6Hz), 7.28(1H, d, J=8.8Hz), 5.31(2H, s), 4.40(2H, t, J=4.8Hz), 3.60(4H, bs), 2.72(4H, t, J=4.8Hz), 2.25(3H, s).
VIII-164		10.0(1H, s), 8.73(1H, s), 8.60-8.57(2H, m), 8.20(1H, d, J=8.8), 7.96(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.4), 7.67-7.65(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=8.8), 5.30(2H, s), 4.38-4.35(2H, m), 2.71-2.68(2H, m), 2.31(1H, br), 2.24(3H, s), 2.13(3H, s).
VIII-165		10.0(1H, s), 8.73(1H, s), 8.62-8.54(2H, m), 8.21-8.19(1H, m), 7.96(1H, s), 7.90-7.86(1H, m), 7.80-7.78(1H, m), 7.68-7.66(1H, m), 7.59(1H, d, J=8.0), 7.38-7.36(1H, m), 7.27(1H, d, J=8.8), 5.30(2H, s), 4.54(1H, brs), 4.37-4.34(2H, m), 2.83-2.75(2H, m), 2.69-2.66(2H, m), 2.24(3H, s), 2.17-2.11(2H, m), 1.71-1.69(2H, m), 1.71-1.69(2H, m), 1.44-1.32(2H, m).
VIII-166		10.08(1H, s), 8.75(1H, s), 8.59(2H, bs), 8.22(1H, d, J=8.8Hz), 7.80(1H, s), 7.89(1H, t, J=6.4Hz), 7.80(1H, d, J=8.4Hz), 7.70-7.67(1H, m), 7.59(1H, d, J=8.4Hz), 7.37(1H, bs), 7.28(1H, d, J=8.0Hz), 5.31(2H, s), 4.41(2H, t, J=4.4Hz), 3.43(2H, bs), 2.76(2H, bs), 2.26(3H, s), 1.99(3H, s).
VIII-167		10.10(1H, s), 8.73(1H, s), 8.60-8.56(2H, m), 8.20(1H, d, J=8.8), 7.94(1H, s), 7.90-7.86(1H, m), 7.79(1H, d, J=8.8), 7.68-7.64(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=9.2), 5.29(2H, s), 4.20-4.08(2H, m), 3.79(1H, d, J=10.4), 3.65(1H, d, J=11.2), 3.25-3.20(1H, m), 3.09(1H, br), 2.83-2.70(2H, m), 2.25(3H, s).
VIII-168		10.0(1H, s), 8.73(1H, s), 8.59-8.57(2H, m), 8.19(1H, d, J=8.8), 7.95(1H, s), 7.89-7.85(1H, m), 7.78(1H, d, J=8.8), 7.68-7.65(1H, m), 7.58(1H, d, J=7.6), 7.37-7.34(1H, m), 7.26(1H, d, J=8.8), 5.29(2H, s), 4.26-4.14(2H, m), 3.75-3.72(2H, m), 2.85(1H, d, J=12.4), 2.69-2.61(2H, m), 2.45(1H, d, J=10.8), 2.25(3H, s).
VIII-169		10.0(1H, s), 8.74(1H, s), 8.60-8.58(2H, m), 8.20(1H, d, J=8.8), 7.96(1H, s), 7.90-7.86(1H, m), 7.80(1H, d, J=8.8), 7.68-7.66(1H, m), 7.59(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=9.2), 5.30(2H, s), 4.20-4.08(2H, m), 3.79(1H, d, J=10.4), 3.65(1H, d, J=11.2), 3.25-3.20(1H, m), 3.09(1H, br), 2.83-2.70(2H, m), 2.26(2H, s).

【 0 1 1 9 】

【表 3 8】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-170		10.09(1H, brs), 8.74(1H, s), 8.61-8.58(2H, m), 8.22(1H, d, J=10.5Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.80(1H, d, J=9.0Hz), 7.68(1H, d, J=10.5Hz), 7.59(1H, d, J=8.1Hz), 7.39-7.35(1H, m), 7.28(1H, d, J=9.0Hz), 5.31(2H, s), 4.12(2H, d, 4.5Hz), 2.95-2.53(5H, m), 2.34-2.09(5H, m)
VIII-171		10.08(1H, brs), 8.74(1H, s), 8.61-8.59(2H, m), 8.21(1H, d, J=9.0Hz), 7.98(1H, s), 7.92-7.86(1H, m), 7.81(1H, d, J=9.0Hz), 7.75(1H, s), 7.67(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 5.31(2H, s), 4.31(2H, m), 3.66(1H, m), 3.27-3.10(2H, m), 2.99-2.80(2H, m)
VIII-172		10.09(1H, brs), 8.74(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.81(1H, d, J=9.0Hz), 7.68(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 5.31(2H, s), 4.44(1H, m), 4.16(1H, m), 2.70-2.51(4H, m), 2.30-2.16(10H, m), 2.09-1.88(2H, m)
VIII-173		10.0(1H, br), 8.74(1H, s), 8.60-8.58(2H, m), 8.20(1H, d, J=8.8), 7.97(1H, s), 7.90-7.86(1H, m), 7.80(1H, d, J=8.4), 7.68-7.66(1H, m), 7.59(1H, d, J=8.0), 7.38-7.35(1H, m), 7.28(1H, d, J=9.2), 5.30(2H, s), 4.26-4.14(2H, m), 3.75-3.72(2H, m), 3.48-3.42(2H, m), 2.85(1H, d, J=12.4), 2.69-2.60(2H, m), 2.45(1H, d, J=10.4), 2.26(3H, s)
VIII-174		(三塩酸塩) 12.31(1H, brs), 9.41(1H, s), 8.97(1H, s), 8.73(1H, d, J=1.8Hz), 8.49(1H, d, 8.7Hz), 8.17-8.05(2H, m), 7.94(1H, s), 7.77(1H, d, J=7.5Hz), 7.69(1H, d, 8.7Hz), 7.59(1H, t, J=5.7Hz), 7.38(1H, d, J=8.7Hz), 5.46(2H, s), 4.60(2H, m), 4.35-3.31(12H, m), 2.30(3H, s).
VIII-175		
VIII-176		(三塩酸塩) 12.31(1H, brs), 9.41(1H, s), 8.97(1H, s), 8.73(1H, d, J=5.4Hz), 8.49(1H, d, J=8.7Hz), 8.13(1H, s), 8.07(1H, d, J=9.0Hz), 7.94(1H, s), 7.72(1H, d, J=8.7Hz), 7.02(1H, d, J=9.0Hz), 7.59(1H, s), 7.38(1H, d, J=9.0Hz), 5.46(2H, s), 4.65-3.62(10H, m), 2.89(3H, s), 2.30(3H, s).

【 0 1 2 0 】

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【表 3 9】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-177		10.1(1H, s), 8.74(1H, s), 8.59(2H, brs), 8.21(1H, d, J=8.8Hz), 7.97(1H, s), 7.85-7.92(1H, m), 7.80(1H, d, J=8.4Hz), 7.67(1H, d, J=9.6Hz), 7.59(1H, d, J=6.4Hz), 7.34-7.40(1H, m), 7.29(1H, d, J=8.8Hz), 5.31(2H, m), 4.10-4.35(2H, m), 3.75-3.82(1H, m), 3.48(1H, m), 2.85(1H, d, J=12Hz), 2.74(1H, d, J=13Hz), 2.34-2.41(1H, m), 2.26(4H, brs), 1.03(3H, d, J=6.0Hz)
VIII-178		10.1(1H, s), 8.74(1H, s), 8.59(2H, brs), 8.21(1H, d, J=8.8Hz), 7.97(1H, s), 7.79-7.93(2H, m), 7.66-7.70(1H, m), 7.59(1H, d, J=6.4Hz), 7.38(1H, brs), 7.29(1H, d, J=8.8Hz), 5.31(2H, m), 4.37-4.56(2H, m), 3.95-4.02(1H, m), 3.80-3.88(1H, m), 2.62-2.85(3H, m), 2.31-2.4(1H, m), 2.26(3H, s), 1.06(3H, d, J=5.2Hz)
VIII-179		10.06(1H, brs), 8.75(1H, s), 8.61-8.59(2H, m), 8.22(1H, d, J=9.0Hz), 7.97(1H, s), 7.92-7.86(1H, m), 7.81(1H, d, J=9.0Hz), 7.76(1H, s), 7.69(1H, d, J=9.0Hz), 7.59(1H, d, J=7.8Hz), 7.40-7.36(1H, m), 7.29(1H, d, J=9.0Hz), 5.31(2H, s), 4.30(2H, m), 3.65(1H, m), 3.16(2H, m), 2.99-2.80(2H, m), 2.26(3H, s)
VIII-180		10.0(1H, brs), 8.76(1H, s), 8.60-8.58(2H, m), 8.19(1H, d, J=8.8), 7.97(1H, s), 7.89-7.86(1H, m), 7.81(1H, d, J=8.8), 7.68-7.66(1H, m), 7.58(1H, d, J=7.6), 7.38-7.35(1H, m), 7.27(1H, d, J=8.8), 5.30(2H, s), 4.51(2H, s), 4.38-4.35(2H, m), 3.40(3H, s), 2.87-2.84(2H, m), 2.34(3H, s)
VIII-181		10.0(1H, s), 8.76(1H, s), 8.59(2H, s), 8.19(1H, d, J=8.8), 7.97(1H, s), 7.90-7.86(1H, m), 7.82(1H, d, J=8.8), 7.69-7.66(1H, m), 7.59(1H, d, J=8.0), 7.38-7.35(1H, m), 7.28(1H, d, J=8.8), 5.30(2H, s), 4.52(2H, s), 4.25-4.15(2H, m), 3.82-3.79(1H, m), 3.67-3.64(1H, m), 3.40(3H, s), 3.26-3.21(2H, m), 3.14-3.05(1H, m), 2.81-2.72(2H, m)
VIII-182		10.1(1H, s), 8.79(1H, s), 8.60(2H, s), 8.25(1H, d, J=7.6Hz), 7.98(1H, s), 7.89(1H, brs), 7.81(1H, d, J=7.6Hz), 7.69(1H, d, J=7.6Hz), 7.60(1H, d, J=7.2Hz), 7.38(1H, brs), 7.29(1H, d, J=8.4Hz), 5.31(2H, s), 4.43(2H, brs), 3.14(2H, brs), 2.52(3H, s), 2.27(3H, s)

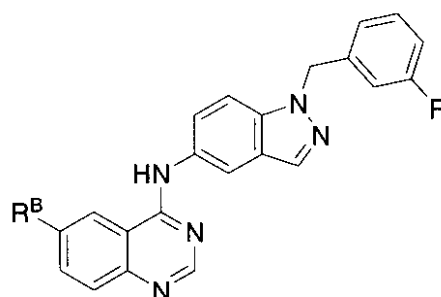
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【 0 1 2 1 】

【 化 8 3 】



(VIII)

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【 0 1 2 2 】

【表 40】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-183		10.19(1H, s), 8.78(1H, s), 8.53(1H, s), 8.23(1H, dd, J=8.4Hz, J=4.0Hz), 8.17(1H, s), 8.14(1H, s), 7.80-7.65(3H, m), 7.40-7.35(1H, m), 7.13-7.04(3H, m), 5.72(2H, s), 4.32(2H, t, J=6.0Hz), 2.86(2H, t, J=6.0Hz), 2.35(3H, s), 2.25(3H, s).
VIII-184		(二塩酸塩) 12.46(1H, s), 9.37(1H, s), 8.87(1H, s), 8.48(1H, d, J=8.7Hz), 8.27(1H, t, J=5.4Hz), 8.24(1H, s), 8.03(2H, m), 7.85(1H, d, J=9.0Hz), 7.68(1H, d, 9.0Hz), 7.35(1H, s), 7.13-7.05(3H, m), 5.75(2H, s), 4.58(2H, m), 7.13-7.05(3H, m), 5.75(2H, s), 4.58(2H, m), 3.51-3.37(4H, m), 3.11(2H, m), 2.27(3H, s), 1.01(1H, s).
VIII-185		(二塩酸塩) 10.12(1Hs), 8.82(1H, d, J=1.5Hz), 8.52(1H, s), 8.22(1H, dd, J=1.8Hz, J=8.7Hz), 8.14(1H, s), 7.80(1H, s), 7.80(1H, s), 7.77-7.68(3H, m), 7.40-7.33(1H, m), 7.12-7.04(3H, m), 5.71(2H, s), 4.86(1H, brs), 4.28(3H, m), 3.58(1H, m), 2.95-2.78(2H, m), 2.25(3H, s), 2.11(1H, m), 1.47(1H, m).
VIII-186		10.18(1H, s), 8.78(1H, s), 8.52(1H, s), 8.23-8.12(3H, m), 7.79-7.65(3H, m), 7.41-7.32(1H, m), 7.18(7.02)(3H, m), 5.71(2H, s), 4.31(2H, m), 3.14(2H, m), 2.29(2H, m), 2.87(3H, s), 2.77(6H, m), 2.24(3H, s).
VIII-187		10.19(1Hs), 8.78(1H, s), 8.52(1H, s), 8.23-8.12(3H, m), 7.80-7.65(3H, m), 7.38-7.33(1H, m), 7.13-7.03(3H, m), 6.49(2H, brs), 5.71(2H, s), 4.31(2H, m), 4.04-3.95(2H, m), 2.92(2H, m), 2.78(2H, m), 2.25(3H, s).
VIII-188		(二塩酸塩) 12.09(1H, s), 8.96(1H, s), 8.83(1H, s), 8.43(1H, d, J=8.7Hz), 8.06(1H, s), 7.97(1H, d, J=8.4Hz), 7.83(1H, d, J=9.0Hz), 7.67(1H, d, J=1.8Hz), 7.65(1H, dd, J=2.1Hz, J=9.0Hz), 7.41-7.34(1H, m), 7.14-7.05(3H, m), 5.71(2H, s), 4.54(2H, m), 4.19(1H, m), 3.83-3.70(2H, m), 3.68-2.95(5H, m), 2.27(3H, s), 2.04(1H, m), 1.93(1H, m), 1.56(1H, m).
VIII-189		(二塩酸塩) 12.21(1H, brs), 8.97(1H, brs), 8.84(1H, s), 8.43(1H, dd, J=1.5Hz, J=9.0Hz), 8.07(1H, s), 8.01(1H, s), 7.99(1H, d, J=9.0Hz), 7.84(1H, d, J=9.0Hz), 7.67(1H, dd, J=1.8Hz, J=9.0Hz), 7.41-7.32(1H, m), 7.14-7.05(3H, m), 5.74(2H, s), 4.59(2H, m), 4.21(1H, m), 3.82-3.65(2H, m), 3.62-3.05(8H, m), 2.28(3H, s), 2.02(1H, m), 1.83(1H, M), 1.55(1H, m).

【 0 1 2 3 】

【表 4 1】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-190		(二塩酸塩) 12.18(1H, s), 9.29(1H, s), 8.45(1H, dd, J=1.2, J=9.0Hz), 8.23(1H, s), 8.06(1H, s), 8.06(1H, d, J=1.2Hz), 7.99(1H, d, J=8.7Hz), 7.87-7.82(1H, m), 7.67(1H, dd, J=1.2Hz, J=9.0Hz), 7.39-7.14(1H, m), 7.14-7.05(3H, m), 5.74(2H, s), 4.61(2H, m), 3.95(1H, m), 8.23(2H, m), 3.13(3H, m), 2.28(3H, s), 2.17(1H, m), 1.78(1H, m).
VIII-191		(二塩酸塩) 12.18(1H, s), 9.12(1H, s), 8.87(1H, s), 8.46(1H, d, J=8.7Hz), 8.43(1H, s), 8.23(1H, s), 8.04-7.97(2H, m), 7.87-7.82(1H, m), 7.64(1H, dd, J=1.2Hz, J=9.0Hz), 7.37-7.14(1H, m), 7.11-7.05(3H, m), 5.74(2H, s), 4.61(2H, m), 4.09-3.26(7H, m), 2.26(3H, s), 2.17(1H, m), 1.79(1H, s).
VIII-192		10.17(1H, s), 8.78(1H, s), 8.51(1H, s), 8.20(1H, dd, J=1.8Hz, J=8.7Hz), 8.14(1H, s), 7.80-7.65(3H, m), 7.40-7.33(1H, m), 7.13-7.03(3H, m), 5.71(2H, s), 4.31(2H, t, J=5.7Hz), 3.41(2H, t, J=6.3Hz), 3.22(3H, s), 2.85(2H, m), 2.70-2.57(4H, m), 2.23(3H, s), 0.99(2H, t, J=7.2Hz).
VIII-193		10.19(1H, s), 8.78(1H, s), 8.52(1H, s), 8.21(1H, d, J=8.4Hz), 8.16(1H, s), 8.13(1H, s), 7.78-7.65(3H, m), 7.49-7.33(1H, m), 7.13-7.04(3H, m), 6.48(2H, brs), 5.71(2H, s), 4.93(1H, m), 4.36(2H, t, J=5.7Hz), 2.80(3H, m), 2.71(2H, m), 2.42(1H, m), 2.24(3H, s), 2.10(1H, m), 1.66(1H, m).
VIII-194		10.21(1H, s), 8.82(1H, d, J=1.5Hz), 8.52(1H, s), 8.22(1H, dd, J=1.8Hz, J=8.7Hz), 8.14(1H, s), 7.80(1H, s), 7.77-7.66(3H, m), 7.40-7.33(1H, m), 7.12-7.04(3H, m), 5.71(2H, s), 4.86(1H, brs), 4.28(3H, m), 3.58(1H, m), 2.95-2.78(2H, m), 2.25(3H, s), 2.11(1H, m), 1.47(1H, m).
VIII-195		10.17(1H, s), 8.78(1H, d, J=1.8Hz), 8.52(1H, s), 8.19(1H, dd, J=1.8Hz, J=10.5Hz), 8.13(1H, s), 7.80(1H, s), 7.77-7.33(1H, m), 7.12-7.03(3H, m), 5.71(2H, s), 4.39(2H, t, J=5.7Hz), 3.58(4H, t, J=4.8Hz), 2.71(2H, t, J=6.0Hz), 2.50(2H, m), 2.24(3H, s).
VIII-196		10.19(1H, s), 8.78(1H, d, J=1.5Hz), 8.19(1H, dd, J=1.8Hz, J=10.5Hz), 8.16(1H, s), 8.13(1H, d, J=1.5Hz), 7.80(1H, s), 7.80-7.66(4H, m), 7.40-7.33(1H, m), 7.13-7.06(3H, m), 5.71(2H, s), 4.41(2H, t, J=5.2Hz), 3.16(2H, m), 3.10(2H, s), 2.80(1H, t, J=5.7Hz), 2.67(3H, s), 2.25(3H, s).

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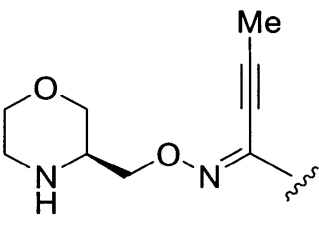
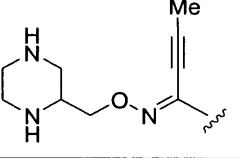
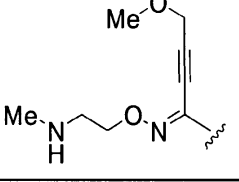
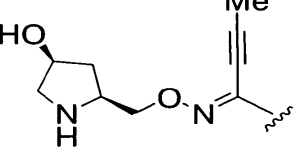
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【 0 1 2 4 】

【表 4 2】

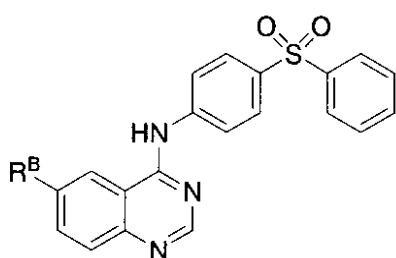
化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-197		10.21(1H, s), 8.79(1H, d, J=1.8Hz), 8.53(1H, s), 8.21(1H, dd, J=1.8Hz, J=8.7Hz), 8.17(1H, s), 8.13(1H, d, J=1.5Hz), 7.79(1H, d, J=13.2Hz), 7.76(1H, d, J=13.2Hz), 7.67(1H, dd, J=1.8Hz, J=9.0Hz), 7.41-7.34(1H, m), 7.16-7.04(3H, m), 5.72(2H, s), 4.20-4.09(2H, m), 3.79(1H, dd, J=2.7Hz, J=10.8Hz), 3.68-3.64(1H, m), 3.30-3.19(1H, m), 3.13-3.10(1H, m), 2.78-2.74(3H, m), 2.26(3H, s).
VIII-198		10.20(1H, brs), 8.79(1H, s), 8.53(1H, s), 8.22-8.13(3H, m), 7.81-7.66(3H, m), 7.41-7.33(1H, m), 7.13-7.04(3H, m), 5.72(2H, s), 4.13(2H, d, J=5.2Hz), 3.00-2.51(6H, m), 2.39-2.26(4H, m)
VIII-199		(二塩酸塩) 12.69(1Hbrs), 9.44(1H, s), 8.69(1H, s), 8.47(1H, d, J=8.1Hz), 8.23(1H, d, J=2.1Hz), 7.38(1H, s), 7.19-7.01(1H, m), 5.74(2H, s), 4.64(5H, m), 4.01(1H, m), 3.39-3.21(4H, m), 2.59(3H, s).
VIII-200		10.19(1H, s), 8.81(1H, s), 8.53(1H, s), 8.21-8.14(3H, m), 7.82(7.82-7.65(3H, m), 7.41(1H, m), 7.15-7.04(13H, m), 5.71(2H, s), 4.68(1H, brs), 4.51(2H, s), 4.27(2H, m), 4.18(1H, m), 3.42(2H, m), 3.45(3H, s), 2.87(1H, m), 2.69(1H, m), 2.04(1H, m), 1.41(1H, m).

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【 0 1 2 5】

【化 8 4】

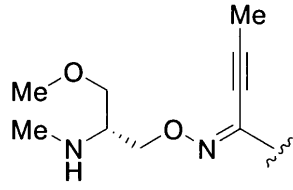
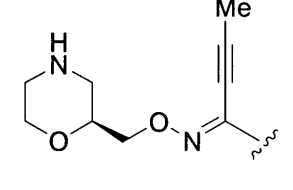
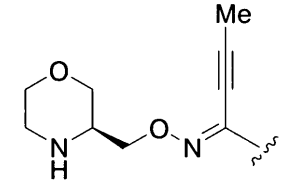
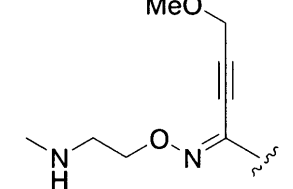
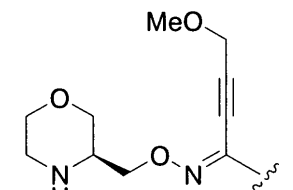


(VIII)

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【 0 1 2 6】

【表 4 3】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-205		10.43-10.40(1H, m), 8.79(1H, s), 8.70(1H, s), 8.25(1H, d, J=8.4Hz), 8.11(2H, bs), 7.98-7.96(4H, m), 7.85(1H, d, J=7.2Hz), 7.69-7.62(3H, m), 4.24-4.20(2H, m), 3.28(3H, s), 2.94(1H, t, J=6.4Hz), 2.38(3H, s), 2.25(3H, s).
VIII-206		10.44(1H, bs), 8.78(1H, d, J=1.5Hz), 8.67(1H, bs), 8.24(1H, d, J=9.0Hz), 8.16-8.05(2H, m), 7.99-7.94(4H, m), 7.84(1H, d, J=8.7Hz), 7.71-7.60(3H, m), 4.24(1H, dd, J=6.3Hz, J=11.7Hz), 4.16(1H, dd, J=4.8Hz, J=11.7Hz), 3.75-3.71(2H, m), 3.51-3.37(1H, m), 2.84(1H, dd, J=2.4Hz, J=12.3Hz), 2.73-2.58(2H, m), 2.50-2.42(1H, m), 2.25(3H, s).
VIII-207		10.45(1H, bs), 8.78(1H, d, J=1.5Hz), 8.67(1H, bs), 8.24(1H, dd, J=1.5Hz, J=8.7Hz), 8.12-8.09(2H, m), 7.99-7.95(4H, m), 7.85(1H, d, J=8.7Hz), 7.71-7.61(3H, m), 4.18(1H, dd, J=6.0Hz, J=11.1Hz), 4.12(1H, dd, J=6.3Hz, J=11.1Hz), 3.78(1H, dd, J=2.7Hz, J=10.5Hz), 3.65(1H, dt, J=11.1Hz, J=2.7Hz), 3.11-3.07(1H, m), 2.81-2.69(3H, m), 2.25(3H, s).
VIII-208		10.42(1H, s), 8.84(1H, s), 8.69(1H, s), 8.25(1H, d, J=8.8Hz), 8.13(1H, d, J=8.4Hz), 8.00-7.97(3H, m), 7.87(1H, d, J=8.8Hz), 7.97-7.62(5H, m), 4.51(2H, s), 4.42(2H, t, J=5.2Hz), 3.37(3H, s), 2.97(2H, t, J=4.8Hz), 2.41(3H, s).
VIII-209		10.45(1H, bs), 8.80(1H, bs), 8.68(1H, bs), 8.23(1H, d, J=7.5Hz), 8.14-8.06(2H, m), 7.99-7.95(4H, m), 7.87(1H, d, J=9.0Hz), 7.69-7.60(3H, m), 4.52(2H, s), 4.23(1H, dd, J=6.3Hz, J=10.8Hz), 4.17(1H, dd, J=6.3Hz, J=10.8Hz), 3.80(1H, dd, J=2.7Hz, J=11.1Hz), 3.67-3.64(1H, m), 3.40(3H, s), 3.12-3.07(1H, m).

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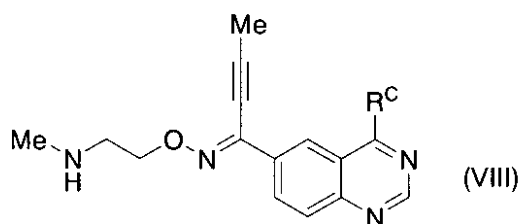
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【 0 1 2 7 】

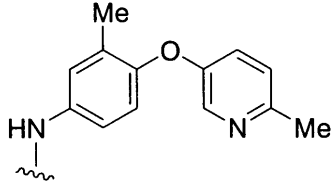
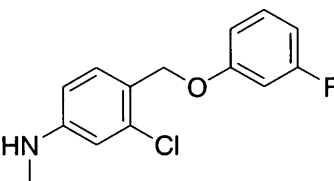
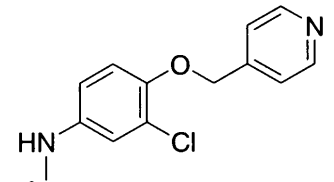
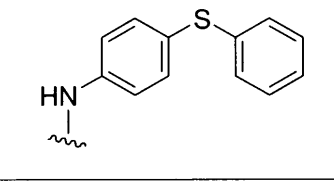
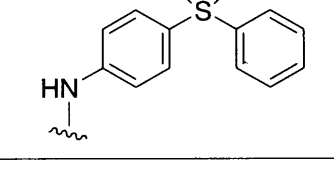
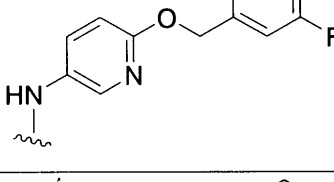
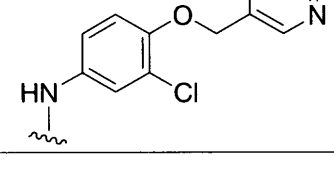
【 化 8 5 】

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【 0 1 2 8 】

【表 4 4】

化合物 No.	R <sup>c</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-210		10.13(1H, s), 8.82(1H, s), 8.60(1H, s), 8.26(1H, d, J=9.2Hz), 8.18(1H, s), 7.82(1H, d, J=8.8Hz), 7.74(1H, s), 7.64(1H, d, J=9.2Hz), 7.27-7.22(2H, m), 6.99(1H, d, J=8.4Hz), 4.50(2H, t, J=5.2Hz), 2.64(3H, s), 2.50(2H, bs), 2.45(3H, s), 2.28(3H, s), 2.23(3H, s).
VIII-211		10.26(1H, brs), 8.80(1H, s), 8.66(1H, s), 8.26-8.23(1H, m), 8.14(1H, s), 7.88-7.79(2H, m), 7.62(1H, d, J=8.4), 7.34(1H, dd, J=8.4, 15.2), 6.95-6.88(2H, m), 6.82-6.78(1H, m), 5.16(2H, s), 4.35(2H, t, J=6.0Hz), 2.92(2H, t, J=6.0Hz), 2.38(3H, s), 2.26(3H, s), 2.26(3H, s).
VIII-212		10.0(1H, brs), 8.74(1H, s), 8.62-8.58(3H, m), 8.21(1H, d, J=8.8), 7.97(1H, s), 7.80(1H, d, J=8.8), 7.69-7.67(1H, m), 7.48-7.47(2H, m), 7.25(1H, d, J=9.2), 5.32(2H, s), 4.34-4.31(2H, s), 2.89-2.86(2H, m), 2.36(3H, s), 2.25(3H, s).
VIII-213		10.43(1H, s), 9.23(1H, s), 9.21(1H, s), 8.64(1H, s), 8.26(1H, d, J=6.6Hz), 7.93(2H, d, J=3.9Hz), 7.84(1H, d, J=6.9Hz), 7.45(2H, d, J=5.7Hz), 7.35(1H, d, J=5.4Hz), 7.27(2H, t, J=5.7Hz), 4.56(2H, d, J=3.9Hz), 2.64(3H, s), 2.28(3H, s), 1.99(3H, s).
VIII-214		8.85(1H, s), 8.80(1H, s), 8.26(1H, d, J=6.6Hz), 8.12(1H, d, J=6.3Hz), 7.99-7.96(4H, m), 7.85(1H, d, J=6.6Hz), 7.71-7.62(3H, m), 4.33(2H, t, J=4.2Hz), 2.86(2H, t, J=4.5Hz), 2.35(3H, s), 2.25(3H, s).
VIII-215		10.15(1H, s), 8.77(1H, s), 8.55(1H, s), 8.47(1H, s), 8.24(1H, d, J=9.2Hz), 8.07(1H, d, J=9.2Hz), 7.81(1H, d, J=9.2Hz), 7.47-7.39(1H, m), 7.32-7.28(2H, m), 7.16(1H, t, J=7.6Hz), 7.00(1H, d, J=8.8Hz), 5.41(1H, s), 4.41(2H, t, J=5.6Hz), 3.10(2H, t, J=5.2Hz), 2.49(3H, s), 2.26(3H, s).
VIII-216		10.3(1H, s), 8.84(1H, s), 8.72(1H, s), 8.42-8.65(2H, m), 8.26(1H, d, J=7.6Hz), 7.98(1H, s), 7.91(1H, brs), 7.81(1H, brs), 7.71(1H, brs), 7.47(1H, brs), 7.34(1H, d, J=8.0Hz), 5.30(2H, s), 4.53(2H, brs), 3.38(2H, brs), 2.68(3H, s), 2.26(3H, s).

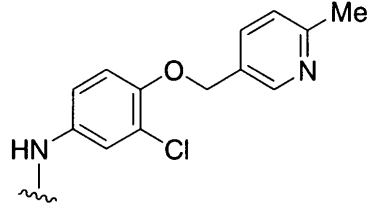
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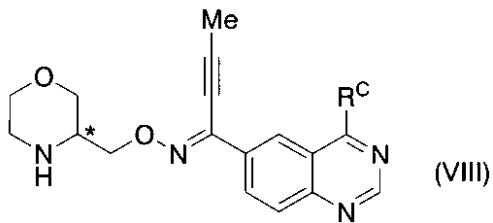
【表 4 5】

化合物 No.	R <sup>C</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-217		10.1(1H, s), 8.76(1H, s), 8.59(1H, s), 8.57(1H, s), 8.24(1H, d, J=10Hz), 7.94(1H, bs), 7.67-7.73(3H, m), 7.29-7.35(2H, m), 5.24(2H, s), 4.40(2H, t, J=5.6Hz), 3.06(2H, t, J=5.2Hz), 2.49(3H, s), 2.48(3H, s), 2.26(3H, s)

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【 0 1 3 0 】

【化 8 6】



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(式中、\*を付した原子は不斉炭素原子。)

【 0 1 3 1 】

【表 4 6】

化合物 No.	*	R <sup>c</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-218	S		(三塩酸塩) 12.18(1H, bs), 9.78(1H, m), 9.61(1H, m), 9.36(1H, s), 8.94(1H, s), 8.48(1H, dd, J=1.8Hz, J=9.0Hz), 8.41(1H, d, J=3Hz), 8.03(1H, d, J=9Hz), 7.76-7.73(2H, m), 7.64-7.60(2H, m), 7.13(1H, d, J=8.7Hz), 4.51-4.49(2H, m), 2.60(3H, s), 2.29(3H, s), 2.27(3H, s).
VIII-219	R		10.46(1H, bs), 8.78(1H, d, J=1.5Hz), 8.67(1H, s), 8.24(1H, dd, J=1.5Hz, J=8.7Hz), 8.12-8.09(2H, m), 7.99-7.95(4H, m), 7.85(1H, d, J=8.7Hz), 7.71-7.60(3H, m), 4.17(1H, dd, J=5.7Hz, J=11.1Hz), 4.12(1H, dd, J=6.3Hz, J=11.1Hz), 3.78(1H, dd, J=2.7Hz, J=10.5Hz), 3.65(1H, dt, J=10.8Hz, J=2.7Hz), 3.26-3.18(1H, m), 3.13-3.05(1H, m), 2.82-2.70(2H, m), 2.25(3H, s).
VIII-220	S		10.26(1H, bs), 8.79(1H, s), 8.67(1H, s), 8.23(1H, d, J=8.8Hz), 8.14(1H, s), 7.84(1H, d, J=8.8Hz), 7.62(1H, d, J=8.8Hz), 7.35(1H, dd, J=8.0, J=15.6), 6.96-6.86(2H, m), 6.82-6.78(1H, m), 5.16(2H, s), 4.20-4.11(2H, m), 3.79(1H, d, J=10.8Hz), 3.66(1H, d, J=11.2Hz), 3.22(2H, t, d=10.4), 3.10-3.02(1H, m), 2.81-2.68(2H, m), 2.26(3H, s).
VIII-221	R		10.26(1H, bs), 8.79(1H, s), 8.67(1H, s), 8.23(1H, d, J=9.6Hz), 8.14(1H, s), 7.85-7.83(1H, m), 7.62(1H, d, J=8.8Hz), 7.35(1H, dd, J=8.0, J=15.6), 6.95-6.88(2H, m), 6.82-6.78(1H, m), 5.16(2H, s), 4.20-4.11(2H, m), 3.79(1H, d, J=10.8Hz), 3.66(1H, d, J=11.2Hz), 3.22(2H, t, d=10.4), 3.10-3.02(1H, m), 2.81-2.68(2H, m), 2.26(3H, s)

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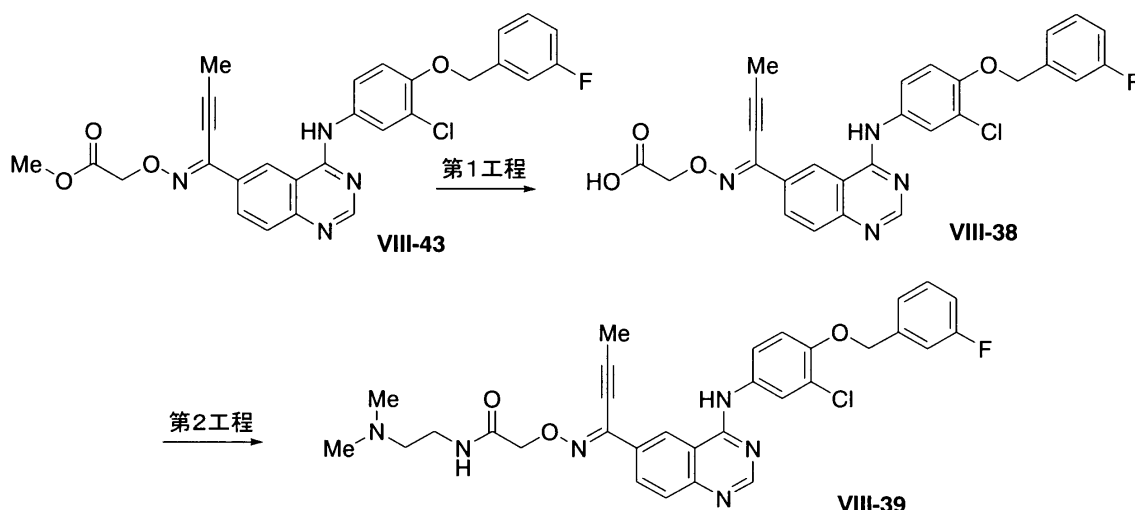
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【 0 1 3 2 】

実施例 1 1



## 【化 8 7】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(カルボキシメトキシイミノ)-2-プチン-1-イル)キナゾリン (VIII-38)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(メトキシカルボニルメトキシイミノ)-2-プチン-1-イル)キナゾリン(VIII-43, 1.2g)をテトラヒドロフラン12mlとメタノール12mlの混合液に溶解させ、1.5mlの2mol/L水酸化ナトリウム水溶液を加えて室温で1.5時間攪拌した。反応液を酢酸エチル20mlで希釈後、2mol/L塩酸を1.6ml加え、水を加えると目的物が析出した。析出物をろ取後、水、酢酸エチルで洗浄し乾燥することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(カルボキシメトキシイミノ)-2-プチン-1-イル)キナゾリン(VIII-38, 0.9g)を黄色固体として得た。

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$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad )$ : 10.18(1H, brs), 8.77(1H, s), 8.60(1H, s), 8.19(1H, dd,  $J=2.0, 11.6\text{Hz}$ ), 7.95(1H, d,  $J=3.2\text{Hz}$ ), 7.80(1H, d,  $J=11.6\text{Hz}$ ), 7.67(1H, dd,  $J=3.2, 11.6\text{Hz}$ ), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.22-7.15(1H, m), 5.27(2H, s), 4.83(2H, s), 2.27(3H, s).

## 【 0 1 3 3】

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(第2工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-((2-ジメチルアミノエチルカルバモイル)メトキシイミノ)-2-プチン-1-イル)キナゾリン (VIII-39)の合成

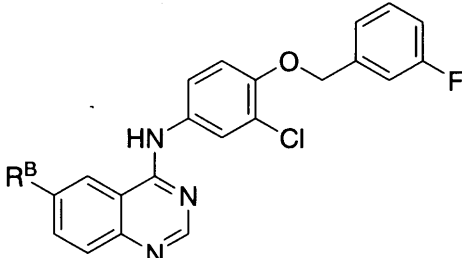
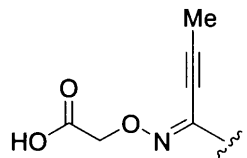
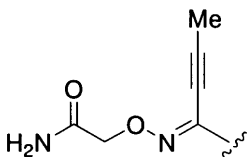
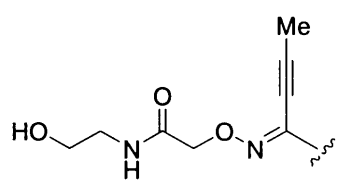
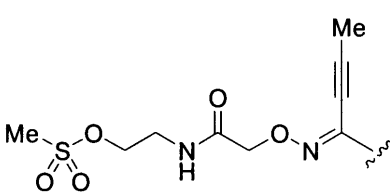
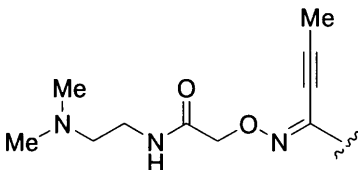
4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(カルボキシメトキシイミノ)-2-プチン-1-イル)キナゾリン(VIII-38, 70mg)、1-ヒドロキシベンゾトリアゾール24mg、1-エチル3-(3-ジメチルアミノプロピル)カルボジイミド塩酸塩27mgをN,N-ジメチルホルムアミド1.4mlに溶かし、室温で5分攪拌後、N,N-ジメチルエチレンジアミン22 $\mu\text{l}$ を加え室温で2時間攪拌した。反応終了後に酢酸エチルで希釈し、水と飽和炭酸水素ナトリウム水溶液を加えて、分液操作を行った。有機層を水洗後、硫酸ナトリウム上で乾燥し、ろ液を濃縮した。濃縮残渣に酢酸エチルを加えて固体化させ、固体をろ取、乾燥することにより4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-((2-ジメチルアミノエチルカルバモイル)メトキシイミノ)-2-プチン-1-イル)キナゾリン(VIII-39, 30mg)を無色結晶として得た。

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$^1\text{H NMR}(\text{d}_6\text{-DMSO}, \quad )$ : 10.12(1H, s), 8.77(1H, s), 8.59(1H, s), 8.19(1H, dd,  $J=2.0, 12.0\text{Hz}$ ), 8.08(1H, brs), 7.95(1H, d,  $J=3.2\text{Hz}$ ), 7.80(1H, d,  $J=11.6\text{Hz}$ ), 7.70-7.62(2H, m), 7.51-7.44(1H, m), 7.34-7.26(3H, m), 7.21-7.15(1H, s), 5.27(2H, s), 4.69(2H, s), 3.22(2H, dd,  $J=8.4, 16.0\text{Hz}$ ), 2.31(1H, t,  $J=8.4\text{Hz}$ ), 2.28(3H, s), 2.11(6H, s).

実施例 11と同様の方法で下記の化合物を合成した。

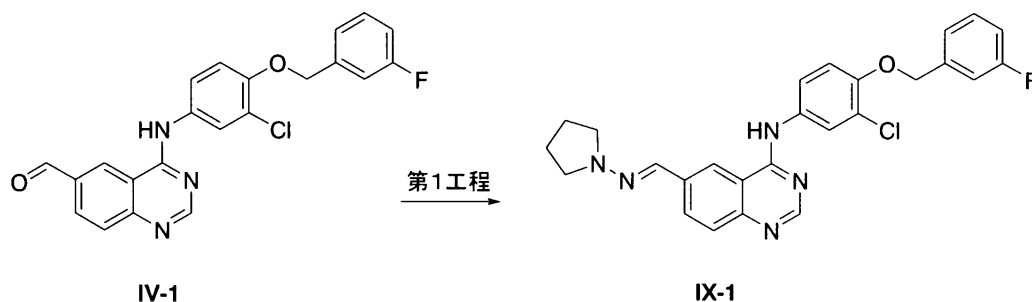
【表 4 7】

		
化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
VIII-222		10.18(1H, brs), 8.77(1H, s), 8.60(1H, s), 8.19(1H, dd, J=2.0, 11.6Hz), 7.95(1H, d, J=3.2Hz), 7.80(1H, d, J=11.6Hz), 7.67(1H, dd, J=3.2, 11.6Hz), 7.51-7.44(1H, m), 7.35-7.26(3H, m), 7.22-7.15(1H, m), 5.27(2H, s), 4.83(2H, s), 2.27(3H, s).
VIII-223		10.12(1H, bs), 8.76(1H, d, J=1.5Hz), 8.59(1H, s), 8.20(1H, dd, J=1.8Hz, J=8.7Hz), 7.96(1H, d, J=2.4Hz), 7.81(1H, d, J=8.7Hz), 7.68(1H, dd, J=2.7Hz, J=9.0Hz), 7.52-7.44(1H, m), 7.35-7.16(6H, m), 5.27(2H, s), 4.65(2H, s), 2.28(3H, s).
VIII-224		10.10(1H, s), 8.59(1H, s), 8.19(1H, dd, J=2.4, 11.6Hz), 7.95(1H, d, J=3.2Hz), 7.80(1H, d, J=11.6), 7.76-7.73(1H, m), 7.68(1H, dd, J=3.6, 12Hz), 7.48-7.44(1H, m), 7.35-7.26(3H, m), 7.21-7.15(1H, m), 5.27(2H, s), 4.73(1H, t, J=7.2Hz), 4.70(2H, s), 3.44(2H, dd, J=8.0, 15.6Hz), 3.21(2H, dd, J=8.0, 15.6Hz), 2.28(3H, s).
VIII-225		10.10(1H, s), 8.76(1H, s), 8.59(1H, s), 8.19(1H, d, J=12.0Hz), 8.08(1H, brs), 7.95(1H, s), 7.80(1H, d, J=11.6Hz), 7.69-7.66(1H, m), 7.51-7.44(1H, m), 7.34-7.26(3H, m), 7.21-7.16(1H, s), 5.27(2H, s), 4.71(2H, s), 3.60-3.53(1H, m), 2.99(3H, s), 2.28(3H, s).
VIII-226		10.12(1H, s), 8.77(1H, s), 8.59(1H, s), 8.19(1H, dd, J=2.0, 12.0Hz), 8.08(1H, brs), 7.95(1H, d, J=3.2Hz), 7.80(1H, d, J=11.6Hz), 7.70-7.62(2H, m), 7.51-7.44(1H, m), 7.34-7.26(3H, m), 7.21-7.15(1H, s), 5.27(2H, s), 4.69(2H, s), 3.22(2H, dd, J=8.4, 16.0Hz), 2.31(1H, t, J=8.4Hz), 2.28(3H, s), 2.11(6H, s).

【 0 1 3 4 】

実施例 1 2

## 【化 8 8】



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(第1工程) 4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(ピロリジン-1-イルイミノメチル)キナゾリン(IX-1)の合成

4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-ホルミルキナゾリン(IV-1) 40mgと1-アミノ-ピロリジン塩酸塩 13mgをテトラヒドロフラン1mL、水0.1mLの混液に溶かし、室温で終夜反応した。反応後、飽和炭酸水素ナトリウム水溶液を加え酢酸エチルで抽出した。有機層をプレセップ(登録商標)を通して脱水し、ろ液を濃縮した。濃縮残渣を、アミノカラムを用いたクロマトグラフィー(ヘキサン:酢酸エチル=2:1 1:1で溶出)により精製し、4-(3-クロロ-4-(3-フルオロベンジルオキシ)フェニルアミノ)-6-(ピロリジン-1-イルイミノ)メチルキナゾリン(IX-1)38mgを黄色固体として得た。

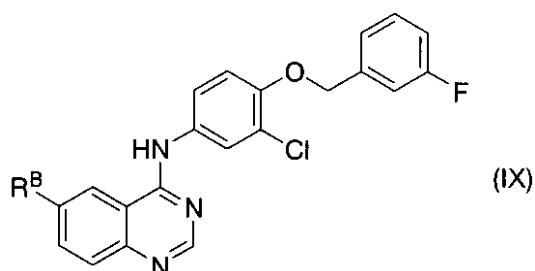
$^1\text{H NMR}$ ( $d_6$ -DMSO,  $d$ ): 9.76(1H, s), 8.52(1H, s), 8.40(1H, brs), 8.08(1H, d,  $J=6.6\text{Hz}$ ), 8.06(1H, d,  $J=1.8\text{Hz}$ ), 7.76(1H, dd,  $J=6.6\text{Hz}$ ,  $J=1.8\text{Hz}$ ), 7.69(1H, dd,  $J=6.6\text{Hz}$ ), 7.50-7.45(1H, m), 7.35-7.26(3H, m), 7.19(1H, t,  $J=6.6\text{Hz}$ ), 5.26(2H, s), 3.39(4H, br), 1.94(4H, br)

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## 【 0 1 3 5】

上記実施例 1 2 と同様の方法で下記の化合物を合成した。

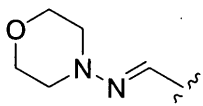
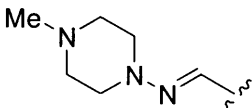
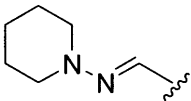
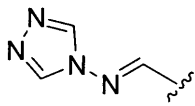
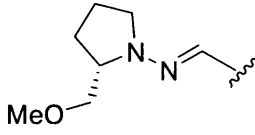
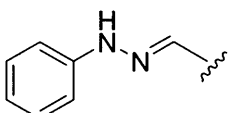
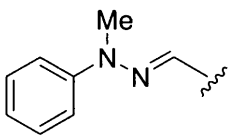
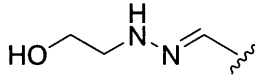
## 【化 8 9】



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## 【 0 1 3 6】

【表 4 8】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
IX-2		9.86(1H, s), 8.56(1H, s), 8.55(1H, brs), 8.14(1H, d, J=6.6Hz), 8.05(1H, d, J=1.8Hz), 7.78-7.74(3H, m), 7.50-7.45(1H, m), 7.35-7.26(3H, m), 7.19(1H, t, J=6.3Hz), 5.26(2H, s), 3.18(4H, t-like, J=3.6Hz), 3.20(4H, t-like, J=3.6Hz)
IX-3		9.83(1H, s), 8.56(1H, s), 8.52(1H, brs), 8.13(1H, d, J=6.6Hz), 8.05(1H, brs), 7.77-7.73(3H, m), 7.50-7.45(1H, m), 7.35-7.26(3H, m), 7.19(1H, t, J=6.3Hz), 5.26(2H, s), 3.21(4H, brs), 2.53(4H, brs), 2.25(3H, s)
IX-4		9.83(1H, s), 8.54(1H, s), 8.50(1H, brs), 8.13(1H, d, J=8.7Hz), 8.04(1H, d, J=2.7Hz), 7.76-7.70(3H, m), 7.52-7.43(1H, m), 7.36-7.25(3H, m), 7.22-7.15(1H, m), 5.26(2H, s), 3.25-3.17(4H, m), 1.76-1.65(4H, m), 1.58-1.49(2H, m)
IX-5		9.19(3H, brs), 9.16(1H, s), 8.95(1H, brs), 8.52(1H, brs), 8.20(1H, d, J=8.4Hz), 7.95(1H, s), 7.79(1H, d, J=6.9Hz), 7.66-7.60(1H, m), 7.52-7.43(1H, m), 7.36-7.14(4H, m), 5.23(2H, s)
IX-6		9.73(1H, s), 8.52(1H, s), 8.38(1H, brs), 8.13(1H, d, J=6.6Hz), 8.05(1H, d, J=1.8Hz), 7.75(1H, dd, J=6.9Hz, J=1.8Hz), 7.70(1H, dd, J=6.6Hz, J=1.8Hz), 7.50-7.45(1H, m), 7.33-7.26(3H, m), 7.18(1H, t, J=6.6Hz), 5.26(2H, s), 3.72(1H, brs), 3.64(1H, dd, J=6.9Hz, J=2.7Hz), 3.46(2H, dd, J=7.2Hz, J=5.1Hz), 3.13(1H, br), 1.99(3H, br), 1.80(1H, br)
IX-7		10.59(1H, s), 9.84(1H, s), 8.56(1H, s), 8.53(1H, brs), 8.29(1H, d, J=6.9Hz), 8.03(2H, brs), 7.78-7.73(2H, m), 7.51-7.45(1H, m), 7.35-7.16(8H, m), 6.79(1H, t, J=5.4Hz), 5.27(2H, s)
IX-8		9.86(1H, s), 8.61(1H, s), 8.57(1H, brs), 8.32(1H, d, J=7.2Hz), 8.05(2H, J=2.1Hz), 7.80-7.73(3H, m), 7.52-7.45(3H, m), 7.37-7.28(5H, m), 7.19(1H, t, J=6.3Hz), 6.94(1H, t, J=5.4Hz), 5.27(2H, s), 3.51(3H, s)
IX-9		9.77(1H, s), 8.53(1H, s), 8.39(1H, brs), 8.09(1H, d, J=6.3Hz), 8.04(1H, s), 7.75-7.69(3H, m), 7.55-7.45(2H, m), 7.34-7.26(3H, m), 7.18(1H, t, J=6.3Hz), 5.21(2H, s), 4.69(1H, t, J=3.6Hz), 3.61(2H, dd, J=8.4Hz, J=4.5Hz), 3.29(2H, m)

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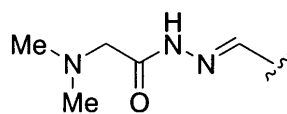
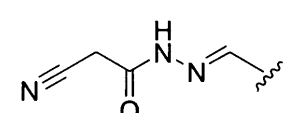
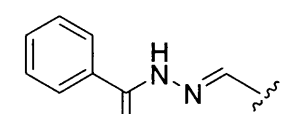
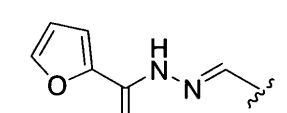
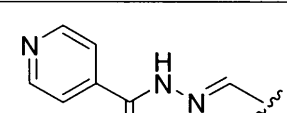
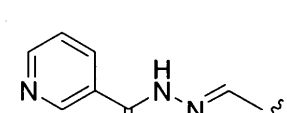
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【 0 1 3 7 】

【表 4 9】

化合物 No.	R <sup>B</sup>	<sup>1</sup> H-NMR(d <sub>6</sub> -DMSO)
IX -10		(E/Z mixture) 11.51(1H, s, minor), 11.45(1H, s, major), 10.00(1H, s, major), 9.89(1H, s, minor), 8.65(1H, brs, minor), 8.60(1H, brs), 8.53(1H, brs, major), 8.26(1H, d, J=6.6Hz, minor), 8.22(1H, d, J=6.6Hz, major), 8.04(1H, brs, minor), 8.00(1H, d, J=1.8Hz, major), 7.80(1H, d, J=6.6Hz), 7.73(1H, d, J=6.6Hz), 7.51-7.45(1H, m), 7.35-7.27(3H, m), 7.19(1H, t, J=6.6Hz), 5.27(2H, s), 3.58(2H, s, minor), 3.08(2H, s, major), 2.34(3H, s, minor), 2.28(3H, s, major)
IX -11		12.04(1H, s), 9.88(1H, s), 8.64(1H, brs), 8.61(1H, s), 8.31(1H, d, J=6.3Hz), 8.17(1H, s), 8.01(1H, d, J=2.1Hz), 7.80(1H, d, J=6.6Hz), 7.72(1H, d, J=6.6Hz), 7.50-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=5.4Hz), 5.27(2H, s), 4.31(2H, s)
IX -12		12.02(1H, s), 10.00(1H, s), 8.70(1H, brs), 8.65(1H, brs), 8.61(1H, s), 8.31(1H, d, J=6.0Hz), 8.02(1H, d, J=1.8Hz), 7.96-7.94(2H, m), 7.84(1H, d, J=6.0Hz), 7.72(1H, d, J=13.8Hz), 7.65-7.55(3H, m), 7.51-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.3Hz), 5.27(2H, s)
IX -13		12.04(1H, s), 10.01(1H, s), 8.68(1H, brs), 8.65(1H, brs), 8.61(1H, s), 8.28(1H, d, J=6.9Hz), 8.02(1H, d, J=1.8Hz), 7.99(1H, s), 7.83(1H, d, J=6.6Hz), 7.74(1H, dd, J=6.6Hz, J=1.8Hz), 7.51-7.45(1H, m), 7.35-7.28(4H, m), 7.19(1H, t, J=6.3Hz), 6.74(1H, brs), 5.27(2H, s)
IX -14		12.22(1H, s), 10.02(1H, s), 8.82(2H, m), 8.73(1H, brs), 8.65(1H, s), 8.62(1H, s), 8.31(1H, d, J=7.2Hz), 8.01(1H, d, J=1.8Hz), 7.86-7.84(2H, m), 7.73-7.72(2H, m), 7.51-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.9Hz), 5.27(2H, s)
IX -15		12.18(1H, s), 10.01(1H, s), 9.11(1H, s), 8.80(1H, d, J=3.3Hz), 8.72(1H, brs), 8.64-8.62(2H, m), 8.32-8.28(2H, m), 8.02(1H, d, J=1.8Hz), 7.85(1H, d, J=6.3Hz), 7.73(1H, dd, J=6.6Hz, J=1.8Hz), 7.62-7.59(1H, m), 7.51-7.45(1H, m), 7.35-7.28(3H, m), 7.19(1H, t, J=6.9Hz), 5.27(2H, s)

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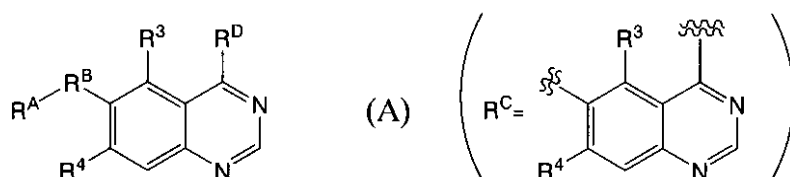
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## 【 0 1 3 8】

上記実施例と同様の方法によって以下に示す化合物を合成することができる。すなわち、以下の一般式 (A) :

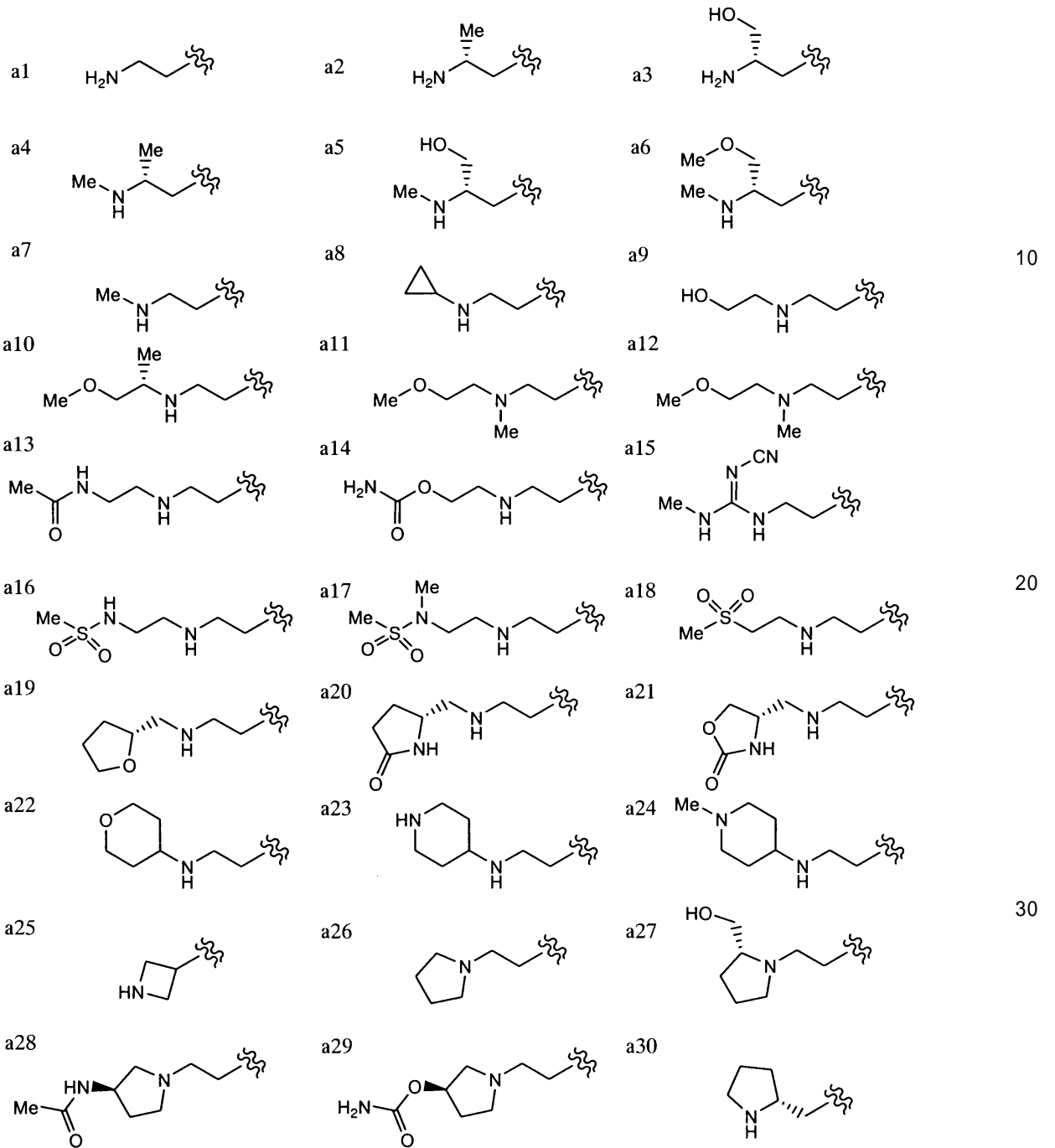
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## 【化 9 0】

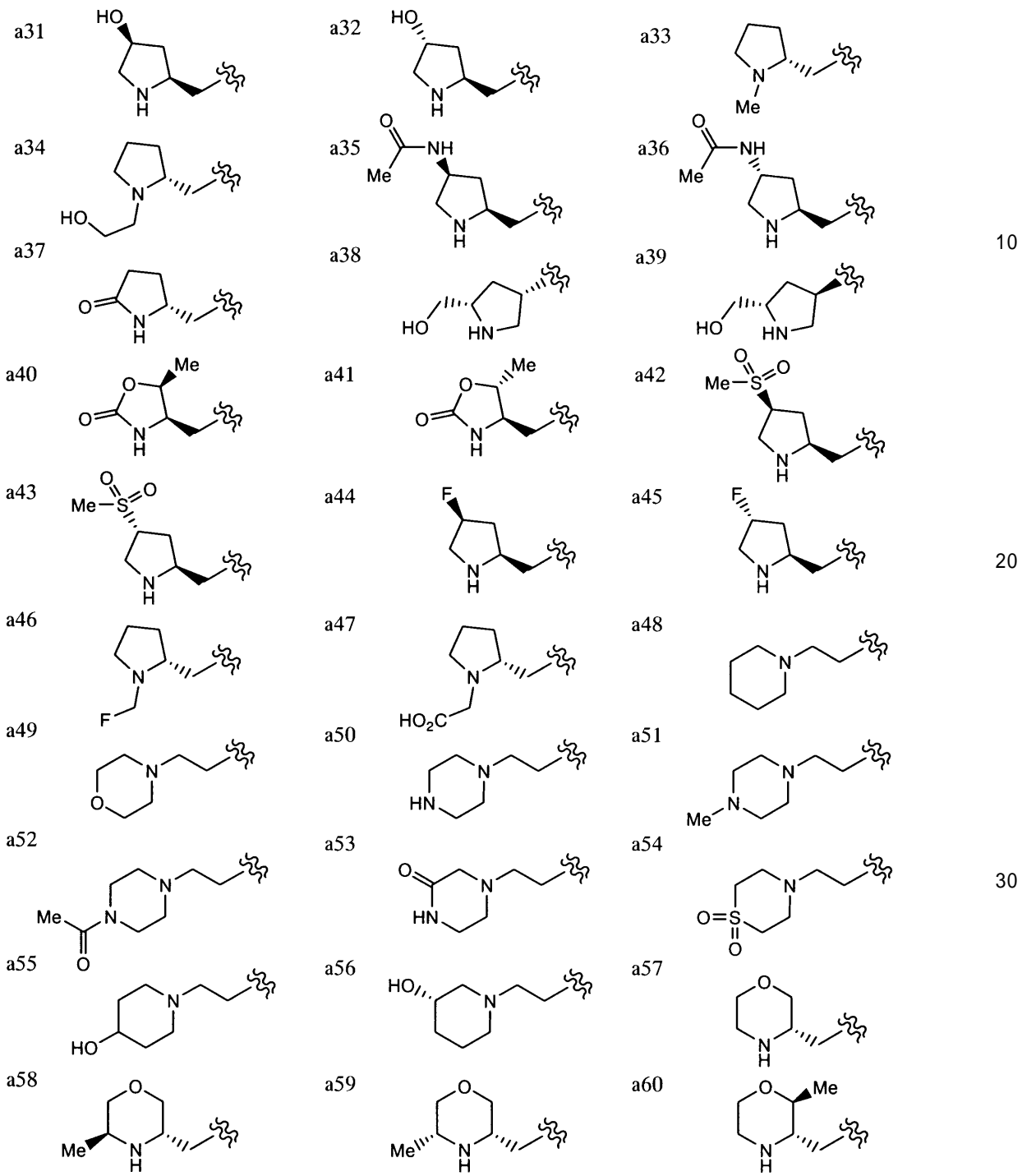


で表される化合物において、R<sup>A</sup>が以下の a 1 ~ a 9 0 :

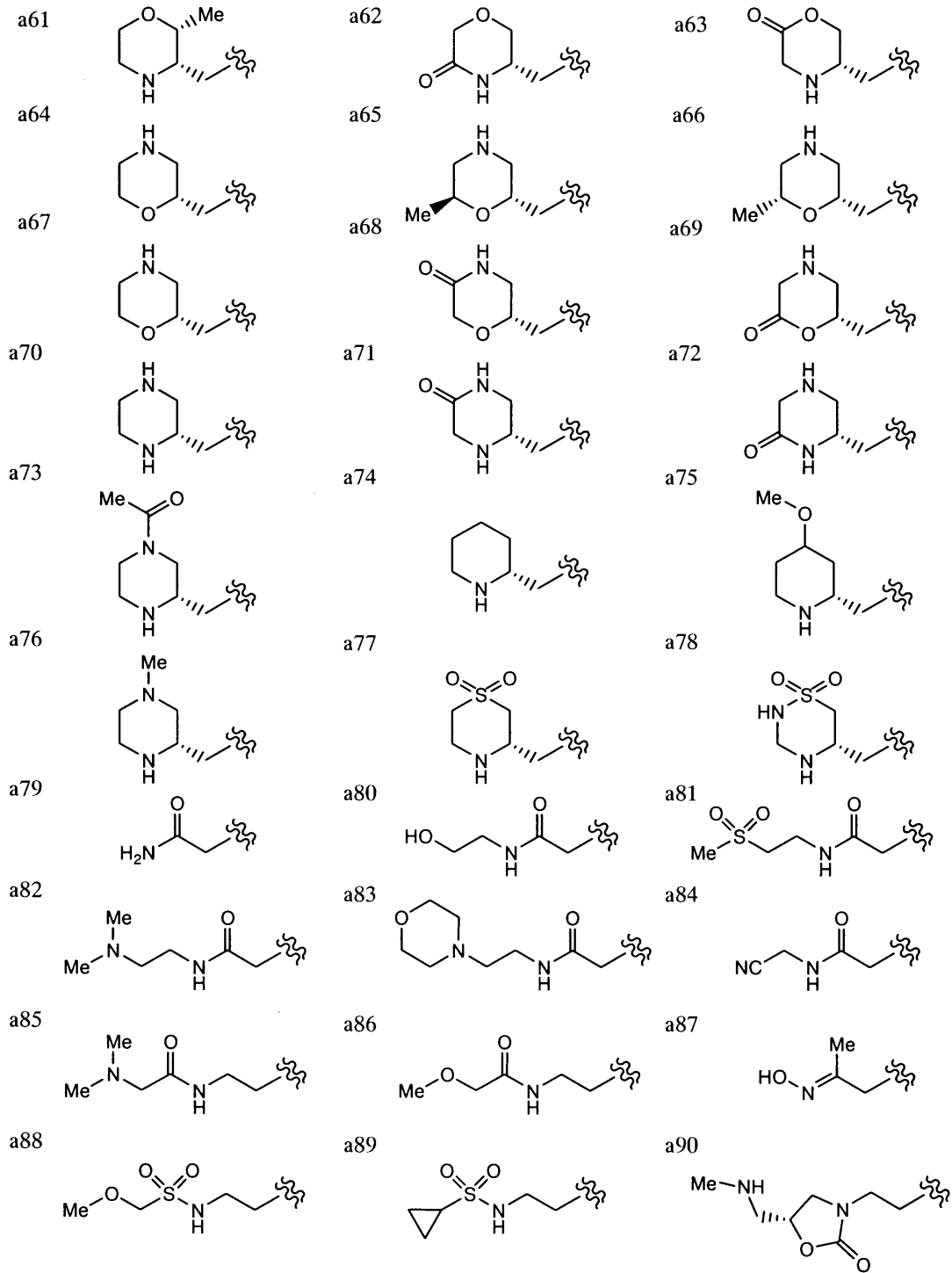
## 【化 9 1】



【化92】



## 【化93】



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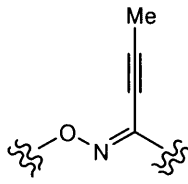
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(式中、Meはメチル)から選択される基、  
R<sup>B</sup>が以下のb1~b6:

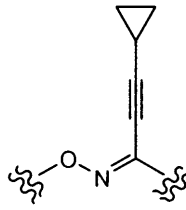


## 【化94】

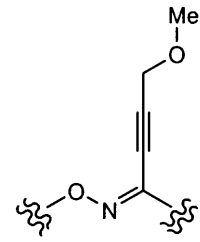
b1



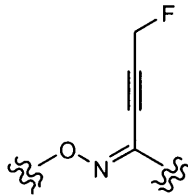
b2



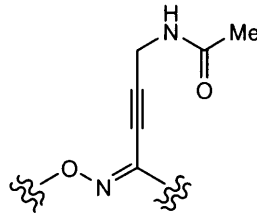
b3



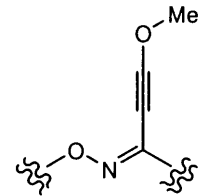
b4



b5



b6



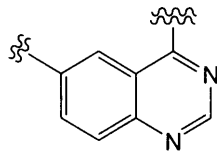
10

(式中、Meはメチル)から選択される基、

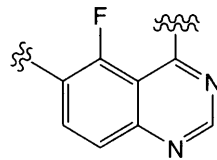
R<sup>c</sup>が以下のc1~c3:

## 【化95】

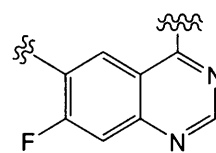
c1



c2



c3

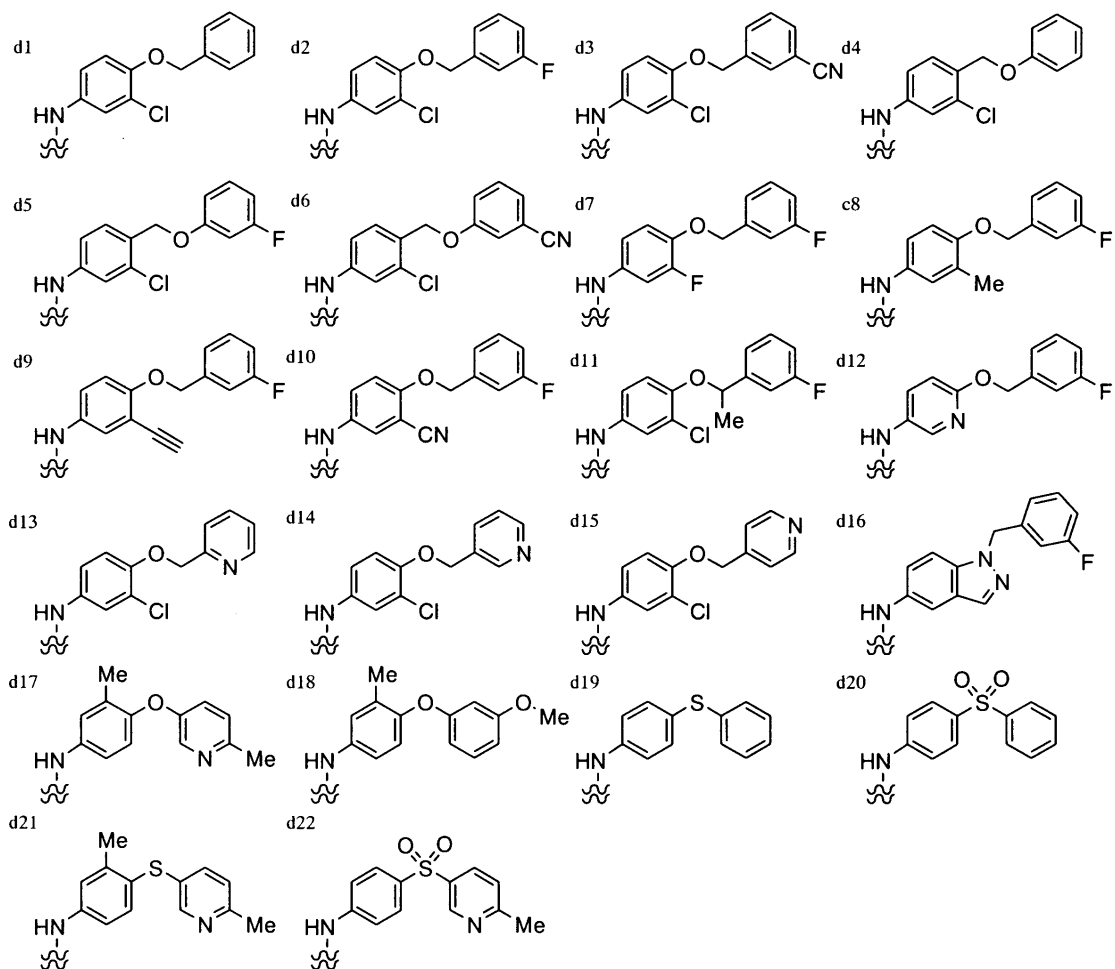


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から選択される基、

R<sup>d</sup>が以下のd1~d22:

## 【化 9 6】



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(式中、Me はメチル)

から選択される基の組み合わせで表される、以下の化合物を合成することができる。

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## 【0139】

(a1, b1, c1, d1), (a1, b1, c1, d2), (a1, b1, c1, d3), (a1, b1, c1, d4), (a1, b1, c1, d5), (a1, b1, c1, d6), (a1, b1, c1, d7), (a1, b1, c1, d8), (a1, b1, c1, d9), (a1, b1, c1, d10), (a1, b1, c1, d11), (a1, b1, c1, d12), (a1, b1, c1, d13), (a1, b1, c1, d14), (a1, b1, c1, d15), (a1, b1, c1, d16), (a1, b1, c1, d17), (a1, b1, c1, d18), (a1, b1, c1, d19), (a1, b1, c1, d20), (a1, b1, c1, d21), (a1, b1, c1, d22), (a1, b1, c2, d1), (a1, b1, c2, d2), (a1, b1, c2, d3), (a1, b1, c2, d4), (a1, b1, c2, d5), (a1, b1, c2, d6), (a1, b1, c2, d7), (a1, b1, c2, d8), (a1, b1, c2, d9), (a1, b1, c2, d10), (a1, b1, c2, d11), (a1, b1, c2, d12), (a1, b1, c2, d13), (a1, b1, c2, d14), (a1, b1, c2, d15), (a1, b1, c2, d16), (a1, b1, c2, d17), (a1, b1, c2, d18), (a1, b1, c2, d19), (a1, b1, c2, d20), (a1, b1, c2, d21), (a1, b1, c2, d22), (a1, b1, c3, d1), (a1, b1, c3, d2), (a1, b1, c3, d3), (a1, b1, c3, d4), (a1, b1, c3, d5), (a1, b1, c3, d6), (a1, b1, c3, d7), (a1, b1, c3, d8), (a1, b1, c3, d9), (a1, b1, c3, d10), (a1, b1, c3, d11), (a1, b1, c3, d12), (a1, b1, c3, d13), (a1, b1, c3, d14), (a1, b1, c3, d15), (a1, b1, c3, d16), (a1, b1, c3, d17), (a1, b1, c3, d18), (a1, b1, c3, d19), (a1, b1, c3, d20), (a1, b1, c3, d21), (a1, b1, c3, d22), (a1, b2, c1, d1), (a1, b2, c1, d2), (a1, b2, c1, d3), (a1, b2, c1, d4), (a1, b2, c1, d5), (a1, b2, c1, d6), (a1, b2, c1, d7), (a1, b2, c1, d8), (a1, b2, c1, d9), (a1, b2, c1, d10), (a1, b2, c1, d11), (a1, b2, c1, d12), (a1, b2, c1, d13), (a1, b2, c1, d14), (a1, b2, c1, d15), (a1, b2, c1, d16), (a1, b2, c1, d17), (a1, b2, c1, d18), (a1, b2, c1, d19), (a1, b2, c1, d20), (a1, b2, c1, d21), (a1, b2, c1, d22)

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d16), (a1, b2, c1, d17), (a1, b2, c1, d18), (a1, b2, c1, d19), (a1, b2, c1, d20)  
 , (a1, b2, c1, d21), (a1, b2, c1, d22), (a1, b2, c2, d1), (a1, b2, c2, d2), (a1  
 , b2, c2, d3), (a1, b2, c2, d4), (a1, b2, c2, d5), (a1, b2, c2, d6), (a1, b2, c2  
 , d7), (a1, b2, c2, d8), (a1, b2, c2, d9), (a1, b2, c2, d10), (a1, b2, c2, d11),  
 (a1, b2, c2, d12), (a1, b2, c2, d13), (a1, b2, c2, d14), (a1, b2, c2, d15), (a1  
 , b2, c2, d16), (a1, b2, c2, d17), (a1, b2, c2, d18), (a1, b2, c2, d19), (a1, b2  
 , c2, d20), (a1, b2, c2, d21), (a1, b2, c2, d22), (a1, b2, c3, d1), (a1, b2, c3,  
 d2), (a1, b2, c3, d3), (a1, b2, c3, d4), (a1, b2, c3, d5), (a1, b2, c3, d6), (a  
 1, b2, c3, d7), (a1, b2, c3, d8), (a1, b2, c3, d9), (a1, b2, c3, d10), (a1, b2,  
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b6, c1, d2), (a10, b6, c1, d3), (a10, b6, c1, d4), (a10, b6, c1, d5), (a10, b6, c1, d6), (a10, b6, c1, d7), (a10, b6, c1, d8), (a10, b6, c1, d9), (a10, b6, c1, d10), (a10, b6, c1, d11), (a10, b6, c1, d12), (a10, b6, c1, d13), (a10, b6, c1, d14), (a10, b6, c1, d15), (a10, b6, c1, d16), (a10, b6, c1, d17), (a10, b6, c1, d18), (a10, b6, c1, d19), (a10, b6, c1, d20), (a10, b6, c1, d21), (a10, b6, c1, d22), (a10, b6, c2, d1), (a10, b6, c2, d2), (a10, b6, c2, d3), (a10, b6, c2, d4), (a10, b6, c2, d5), (a10, b6, c2, d6), (a10, b6, c2, d7), (a10, b6, c2, d8), (a10, b6, c2, d9), (a10, b6, c2, d10), (a10, b6, c2, d11), (a10, b6, c2, d12), (a10, b6, c2, d13), (a10, b6, c2, d14), (a10, b6, c2, d15), (a10, b6, c2, d16), (a10, b6, c2, d17), (a10, b6, c2, d18), (a10, b6, c2, d19), (a10, b6, c2, d20), (a10, b6, c2, d21), (a10, b6, c2, d22), (a10, b6, c3, d1), (a10, b6, c3, d2), (a10, b6, c3, d3), (a10, b6, c3, d4), (a10, b6, c3, d5), (a10, b6, c3, d6), (a10, b6, c3, d7), (a10, b6, c3, d8), (a10, b6, c3, d9), (a10, b6, c3, d10), (a10, b6, c3, d11), (a10, b6, c3, d12), (a10, b6, c3, d13), (a10, b6, c3, d14), (a10, b6, c3, d15), (a10, b6, c3, d16), (a10, b6, c3, d17), (a10, b6, c3, d18), (a10, b6, c3, d19), (a10, b6, c3, d20), (a10, b6, c3, d21), (a10, b6, c3, d22), (a11, b1, c1, d1), (a11, b1, c1, d2), (a11, b1, c1, d3), (a11, b1, c1, d4), (a11, b1, c1, d5), (a11, b1, c1, d6), (a11, b1, c1, d7), (a11, b1, c1, d8), (a11, b1, c1, d9), (a11, b1, c1, d10), (a11, b1, c1, d11), (a11, b1, c1, d12), (a11, b1, c1, d13), (a11, b1, c1, d14), (a11, b1, c1, d15), (a11, b1, c1, d16), (a11, b1, c1, d17), (a11, b1, c1, d18), (a11, b1, c1, d19), (a11, b1, c1, d20), (a11, b1, c1, d21), (a11, b1, c1, d22), (a11, b1, c2, d1), (a11, b1, c2, d2), (a11, b1, c2, d3), (a11, b1, c2, d4), (a11, b1, c2, d5), (a11, b1, c2, d6), (a11, b1, c2, d7), (a11, b1, c2, d8), (a11, b1, c2, d9), (a11, b1, c2, d10), (a11, b1, c2, d11), (a11, b1, c2, d12), (a11, b1, c2, d13), (a11, b1, c2, d14), (a11, b1, c2, d15), (a11, b1, c2, d16), (a11, b1, c2, d17), (a11, b1, c2, d18), (a11, b1, c2, d19), (a11, b1, c2, d20), (a11, b1, c2, d21), (a11, b1, c2, d22), (a11, b1, c3, d1), (a11, b1, c3, d2), (a11, b1, c3, d3), (a11, b1, c3, d4), (a11, b1, c3, d5), (a11, b1, c3, d6), (a11, b1, c3, d7), (a11, b1, c3, d8), (a11, b1, c3, d9), (a11, b1, c3, d10), (a11, b1, c3, d11), (a11, b1, c3, d12), (a11, b1, c3, d13), (a11, b1, c3, d14), (a11, b1, c3, d15), (a11, b1, c3, d16), (a11, b1, c3, d17), (a11, b1, c3, d18), (a11, b1, c3, d19), (a11, b1, c3, d20), (a11, b1, c3, d21), (a11, b1, c3, d22), (a11, b2, c1, d1), (a11, b2, c1, d2), (a11, b2, c1, d3), (a11, b2, c1, d4), (a11, b2, c1, d5), (a11, b2, c1, d6), (a11, b2, c1, d7), (a11, b2, c1, d8), (a11, b2, c1, d9), (a11, b2, c1, d10), (a11, b2, c1, d11), (a11, b2, c1, d12), (a11, b2, c1, d13), (a11, b2, c1, d14), (a11, b2, c1, d15), (a11, b2, c1, d16), (a11, b2, c1, d17), (a11, b2, c1, d18), (a11, b2, c1, d19), (a11, b2, c1, d20), (a11, b2, c1, d21), (a11, b2, c1, d22), (a11, b2, c2, d1), (a11, b2, c2, d2), (a11, b2, c2, d3), (a11, b2, c2, d4), (a11, b2, c2, d5), (a11, b2, c2, d6), (a11, b2, c2, d7), (a11, b2, c2, d8), (a11, b2, c2, d9), (a11, b2, c2, d10), (a11, b2, c2, d11), (a11, b2, c2, d12), (a11, b2, c2, d13), (a11, b2, c2, d14), (a11, b2, c2, d15), (a11, b2, c2, d16), (a11, b2, c2, d17), (a11, b2, c2, d18), (a11, b2, c2, d19), (a11, b2, c2, d20), (a11, b2, c2, d21), (a11, b2, c2, d22), (a11, b2, c3, d1), (a11, b2, c3, d2), (a11, b2, c3, d3), (a11, b2, c3, d4), (a11, b2, c3, d5), (a11, b2, c3, d6), (a11, b2, c3, d7), (a11, b2, c3, d8), (a11, b2, c3, d9), (a11, b2, c3, d10), (a11, b2, c3, d11), (a11, b2, c3, d12), (a11, b2, c3, d13), (a11, b2, c3, d14), (a11, b2, c3, d15), (a11, b2, c3, d16), (a11, b2, c3, d17), (a11, b2, c3, d18), (a11, b2, c3, d19), (a11, b2, c3, d20), (a11, b2, c3, d21), (a11, b2, c3, d22), (a11, b3, c1, d1), (a11, b3, c1, d2), (a11, b3, c1, d3), (a11, b3, c1, d4), (a11, b3, c1, d5), (a11, b3, c1, d6), (a11, b3, c1, d7),

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(a12, b3, c1, d15), (a12, b3, c1, d16), (a12, b3, c1, d17), (a12, b3, c1, d18),  
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c1, d20), (a12, b6, c1, d21), (a12, b6, c1, d22), (a12, b6, c2, d1), (a12, b6, c2, d2 ), (a12, b6, c2, d3), (a12, b6, c2, d4), (a12, b6, c2, d5), (a12, b6, c2, d6), (a12, b6, c2, d7), (a12, b6, c2, d8), (a12, b6, c2, d9), (a12, b6, c2, d10 ), (a12, b6, c2, d11), (a12, b6, c2, d12), (a12, b6, c2, d13), (a12, b6, c2, d14 ), (a12, b6, c2, d15), (a12, b6, c2, d16), (a12, b6, c2, d17), (a12, b6, c2, d18 ), (a12, b6, c2, d19), (a12, b6, c2, d20), (a12, b6, c2, d21), (a12, b6, c2, d22 ), (a12, b6, c3, d1), (a12, b6, c3, d2), (a12, b6, c3, d3), (a12, b6, c3, d4), ( a12, b6, c3, d5), (a12, b6, c3, d6), ( a 12, b6, c3, d7), (a12, b6, c3, d8), (a12 , b6, c3, d9), (a12, b6, c3, d10), (a12, b6, c3, d11), (a12, b6, c3, d12), (a12, b6, c3, d13), (a12, b6, c3, d14), (a12, b6, c3, d15), (a12, b6, c3, d16), (a12, b6, c3, d17), (a12, b6, c3, d18), (a12, b6, c3, d19), ( a12, b6, c3, d20), (a12 , b6, c3, d21), (a12, b6, c3, d22), (a13, b1, c1, d1), (a13, b1, c1, d2), (a13, b1, c1, d3), (a13, b1, c1, d4), (a13, b1, c1, d5), (a13, b1, c1, d6), (a13, b1, c1, d7), (a13, b1, c1, d8), (a13, b1, c1, d9), (a13, b1, c1, d10), (a13, b1, c1, d11), (a13, b1, c1, d12), (a13, b1, c1, d13), (a13, b1, c1, d14), (a13, b1, c1, d15), (a13, b1, c1, d16), (a13, b1, c1, d17), (a13, b1, c1, d18), (a13, b1, c1, d19), (a13, b1, c1, d20), (a13, b1, c1, d21), (a13, b1, c1, d22), (a13, b1, c2, d1), (a1 3, b1, c2, d2), (a13, b1, c2, d3), (a13, b1, c2, d4), (a13, b1, c2, d5 ), (a13, b1, c2, d6), (a13, b1, c2, d7), (a13, b1, c2, d8), (a13, b1, c2, d9), ( a13, b1, c2, d10), (a13, b1, c2, d11), (a13, b1, c2, d12), (a13, b1, c2, d13), ( a13, b1, c2, d14), (a13, b 1, c2, d15), (a13, b1, c2, d16), (a13, b1, c2, d17), (a13, b1, c2, d18), (a13, b1, c2, d19), (a13, b1, c2, d20), (a13, b1, c2, d21), (a13, b1, c2, d22), (a13, b1, c3, d1), (a13, b1, c3, d2), (a13, b1, c3, d3), (a1 3, b1, c3, d4), (a13, b1, c3, d5), (a13, b1 , c3, d6), (a13, b1, c3, d7), (a13, b1, c3, d8), (a13, b1, c3, d9), (a13, b1, c3, d10), (a13, b1, c3, d11), (a13, b1 , c3, d12), (a13, b1, c3, d13), (a13, b1, c3, d14), (a13, b1, c3, d15), (a13, b1 , c3, d16), (a13, b1, c3, d17), (a13, b1, c3, d18), (a13, b1 , c3, d19), (a13, b 1, c3, d20), (a13, b1, c3, d21), (a13, b1, c3, d22), (a13, b2, c1, d1), (a13, b2 , c1, d2), (a13, b2, c1, d3), (a13, b2, c1, d4), (a13, b2, c1, d5), (a13, b2, c1 , d6), (a13, b2, c1, d7), (a13, b2, c1, d8), (a13, b2, c1, d9), (a13, b2, c1, d 10), (a13, b2, c1, d11), (a13, b2, c1, d12), (a13, b2, c1, d13), (a13, b2, c1, d 14), (a13, b2, c1, d15), (a13, b2, c1, d16), (a13, b2, c1, d17), (a13, b2, c1, d 18), (a13, b2, c1, d19), (a13, b2, c1, d20), (a13, b2, c1, d21), (a13, b2, c1, d 22), (a13, b2, c2, d1), (a13, b2, c2, d2), (a13, b2, c2, d3), (a13, b2, c2, d4) , (a13, b2, c2, d5), (a13, b2, c2, d6), (a13, b2, c2, d7), (a13, b2, c2, d8), (a 13, b2, c2, d9), (a13, b2, c2, d10), (a13, b2, c2, d11), (a13, b2, c2, d12), (a1 3, b2, c2, d13), (a13, b2, c2, d14), (a13, b2, c2, d15), (a13, b2, c2, d16), (a1 3, b2, c2, d17), (a13, b2, c2, d18), (a13, b2, c2, d19), (a13, b2, c2, d20), (a1 3, b2, c2, d21), (a13, b2, c2, d22), (a13, b2, c3, d1), (a13, b2, c3, d2), (a13, b2, c3, d3), (a13, b2, c3, d4), (a13, b2, c3, d5), (a13, b2, c3, d6), (a13, b2, c3, d7), (a13, b2, c3, d8), (a13, b2, c3, d9), (a13, b2, c3, d10), (a13, b2, c3, d11), (a13, b2, c3, d12), (a13, b2, c3, d13), (a13, b2, c3, d14), (a13, b2, c3, d15), (a13, b2, c3, d16), (a13, b2, c3, d17), (a13, b2, c3, d18), (a13, b2, c3, d19), (a13, b2, c3, d20), (a13, b2, c3, d21), ( a13, b2, c3, d22), (a13, b3, c1, d1), (a13, b3, c1, d2), (a13, b3, c1, d3), (a13, b3, c1, d4), (a13, b3, c1, d5 ), (a13, b3, c1, d6), (a13, b3, c1, d7), (a13, b3, c1, d8), (a13, b3, c1, d9), ( a13, b3, c1, d10), (a13, b3, c1, d11), (a13, b3, c1, d12), (a13, b3, c1, d13), ( a13, b3, c1, d14), (a13, b3, c1, d15), (a13, b3, c1, d16), (a13, b3, c1, d17), ( a13, b3, c1, d18), (a13, b3, c1, d19), (a13, b3, c1, d20), (a13, b3, c1, d21), (

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d12), (a14, b6, c2, d13), (a14, b6, c2, d14), (a14, b6, c2, d15), (a14, b6, c2, d16), (a14, b6, c2, d17), (a14, b6, c2, d18), (a14, b6, c2, d19), (a14, b6, c2, d20), (a14, b6, c2, d21), (a14, b6, c2, d22), (a14, b6, c3, d1), (a14, b6, c3, d2), (a14, b6, c3, d3), (a14, b6, c3, d4), (a14, b6, c3, d5), (a14, b6, c3, d6), (a14, b6, c3, d7), (a14, b6, c3, d8), (a14, b6, c3, d9), (a14, b6, c3, d10), (a14, b6, c3, d11), (a14, b6, c3, d12), (a14, b6, c3, d13), (a14, b6, c3, d14), (a14, b6, c3, d15), (a14, b6, c3, d16), (a14, b6, c3, d17), (a14, b6, c3, d18), (a14, b6, c3, d19), (a14, b6, c3, d20), (a14, b6, c3, d21), (a14, b6, c3, d22), (a15, b1, c1, d1), (a15, b1, c1, d2), (a15, b1, c1, d3), (a15, b1, c1, d4), (a15, b1, c1, d5), (a15, b1, c1, d6), (a15, b1, c1, d7), (a15, b1, c1, d8), (a15, b1, c1, d9), (a15, b1, c1, d10), (a15, b1, c1, d11), (a15, b1, c1, d12), (a15, b1, c1, d13), (a15, b1, c1, d14), (a15, b1, c1, d15), (a15, b1, c1, d16), (a15, b1, c1, d17), (a15, b1, c1, d18), (a15, b1, c1, d19), (a15, b1, c1, d20), (a15, b1, c1, d21), (a15, b1, c1, d22), (a15, b1, c2, d1), (a15, b1, c2, d2), (a15, b1, c2, d3), (a15, b1, c2, d4), (a15, b1, c2, d5), (a15, b1, c2, d6), (a15, b1, c2, d7), (a15, b1, c2, d8), (a15, b1, c2, d9), (a15, b1, c2, d10), (a15, b1, c2, d11), (a15, b1, c2, d12), (a15, b1, c2, d13), (a15, b1, c2, d14), (a15, b1, c2, d15), (a15, b1, c2, d16), (a15, b1, c2, d17), (a15, b1, c2, d18), (a15, b1, c2, d19), (a15, b1, c2, d20), (a15, b1, c2, d21), (a15, b1, c2, d22), (a15, b1, c3, d1), (a15, b1, c3, d2), (a15, b1, c3, d3), (a15, b1, c3, d4), (a15, b1, c3, d5), (a15, b1, c3, d6), (a15, b1, c3, d7), (a15, b1, c3, d8), (a15, b1, c3, d9), (a15, b1, c3, d10), (a15, b1, c3, d11), (a15, b1, c3, d12), (a15, b1, c3, d13), (a15, b1, c3, d14), (a15, b1, c3, d15), (a15, b1, c3, d16), (a15, b1, c3, d17), (a15, b1, c3, d18), (a15, b1, c3, d19), (a15, b1, c3, d20), (a15, b1, c3, d21), (a15, b1, c3, d22), (a15, b2, c1, d1), (a15, b2, c1, d2), (a15, b2, c1, d3), (a15, b2, c1, d4), (a15, b2, c1, d5), (a15, b2, c1, d6), (a15, b2, c1, d7), (a15, b2, c1, d8), (a15, b2, c1, d9), (a15, b2, c1, d10), (a15, b2, c1, d11), (a15, b2, c1, d12), (a15, b2, c1, d13), (a15, b2, c1, d14), (a15, b2, c1, d15), (a15, b2, c1, d16), (a15, b2, c1, d17), (a15, b2, c1, d18), (a15, b2, c1, d19), (a15, b2, c1, d20), (a15, b2, c1, d21), (a15, b2, c1, d22), (a15, b2, c2, d1), (a15, b2, c2, d2), (a15, b2, c2, d3), (a15, b2, c2, d4), (a15, b2, c2, d5), (a15, b2, c2, d6), (a15, b2, c2, d7), (a15, b2, c2, d8), (a15, b2, c2, d9), (a15, b2, c2, d10), (a15, b2, c2, d11), (a15, b2, c2, d12), (a15, b2, c2, d13), (a15, b2, c2, d14), (a15, b2, c2, d15), (a15, b2, c2, d16), (a15, b2, c2, d17), (a15, b2, c2, d18), (a15, b2, c2, d19), (a15, b2, c2, d20), (a15, b2, c2, d21), (a15, b2, c2, d22), (a15, b2, c3, d1), (a15, b2, c3, d2), (a15, b2, c3, d3), (a15, b2, c3, d4), (a15, b2, c3, d5), (a15, b2, c3, d6), (a15, b2, c3, d7), (a15, b2, c3, d8), (a15, b2, c3, d9), (a15, b2, c3, d10), (a15, b2, c3, d11), (a15, b2, c3, d12), (a15, b2, c3, d13), (a15, b2, c3, d14), (a15, b2, c3, d15), (a15, b2, c3, d16), (a15, b2, c3, d17), (a15, b2, c3, d18), (a15, b2, c3, d19), (a15, b2, c3, d20), (a15, b2, c3, d21), (a15, b2, c3, d22), (a15, b3, c1, d1), (a15, b3, c1, d2), (a15, b3, c1, d3), (a15, b3, c1, d4), (a15, b3, c1, d5), (a15, b3, c1, d6), (a15, b3, c1, d7), (a15, b3, c1, d8), (a15, b3, c1, d9), (a15, b3, c1, d10), (a15, b3, c1, d11), (a15, b3, c1, d12), (a15, b3, c1, d13), (a15, b3, c1, d14), (a15, b3, c1, d15), (a15, b3, c1, d16), (a15, b3, c1, d17), (a15, b3, c1, d18), (a15, b3, c1, d19), (a15, b3, c1, d20), (a15, b3, c1, d21), (a15, b3, c1, d22), (a15, b3, c2, d1), (a15, b3, c2, d2), (a15, b3, c2, d3), (a15, b3, c2, d4), (a15, b3, c2, d5), (a15, b3, c2, d6), (a15, b3, c2, d7), (a15, b3, c2, d8), (a15, b3, c2, d9), (a15, b3, c2, d10), (a15, b3, c2, d11), (a15, b3, c2, d12), (a15, b3, c2, d13), (a15, b3, c2, d14), (a15, b3, c2, d15), (a15, b3, c2, d16), (a15, b3, c2, d17), (a15,

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d19), (a15, b6, c2, d20), (a15, b6, c2, d21), (a15, b6, c2, d22), (a15, b6, c3, d1), (a15, b6, c3, d2), (a15, b6, c3, d3), (a15, b6, c3, d4), (a15, b6, c3, d5), (a15, b6, c3, d6), (a15, b6, c3, d7), (a15, b6, c3, d8), (a15, b6, c3, d9), (a15, b6, c3, d10), (a15, b6, c3, d11), (a15, b6, c3, d12), (a15, b6, c3, d13), (a15, b6, c3, d14), (a15, b6, c3, d15), (a15, b6, c3, d16), (a15, b6, c3, d17), (a15, b6, c3, d18), (a15, b6, c3, d19), (a15, b6, c3, d20), (a15, b6, c3, d21), (a15, b6, c3, d22), (a16, b1, c1, d1), (a16, b1, c1, d2), (a16, b1, c1, d3), (a16, b1, c1, d4), (a16, b1, c1, d5), (a16, b1, c1, d6), (a16, b1, c1, d7), (a16, b1, c1, d8), (a16, b1, c1, d9), (a16, b1, c1, d10), (a16, b1, c1, d11), (a16, b1, c1, d12), (a16, b1, c1, d13), (a16, b1, c1, d14), (a16, b1, c1, d15), (a16, b1, c1, d16), (a16, b1, c1, d17), (a16, b1, c1, d18), (a16, b1, c1, d19), (a16, b1, c1, d20), (a16, b1, c1, d21), (a16, b1, c1, d22), (a16, b1, c2, d1), (a16, b1, c2, d2), (a16, b1, c2, d3), (a16, b1, c2, d4), (a16, b1, c2, d5), (a16, b1, c2, d6), (a16, b1, c2, d7), (a16, b1, c2, d8), (a16, b1, c2, d9), (a16, b1, c2, d10), (a16, b1, c2, d11), (a16, b1, c2, d12), (a16, b1, c2, d13), (a16, b1, c2, d14), (a16, b1, c2, d15), (a16, b1, c2, d16), (a16, b1, c2, d17), (a16, b1, c2, d18), (a16, b1, c2, d19), (a16, b1, c2, d20), (a16, b1, c2, d21), (a16, b1, c2, d22), (a16, b1, c3, d1), (a16, b1, c3, d2), (a16, b1, c3, d3), (a16, b1, c3, d4), (a16, b1, c3, d5), (a16, b1, c3, d6), (a16, b1, c3, d7), (a16, b1, c3, d8), (a16, b1, c3, d9), (a16, b1, c3, d10), (a16, b1, c3, d11), (a16, b1, c3, d12), (a16, b1, c3, d13), (a16, b1, c3, d14), (a16, b1, c3, d15), (a16, b1, c3, d16), (a16, b1, c3, d17), (a16, b1, c3, d18), (a16, b1, c3, d19), (a16, b1, c3, d20), (a16, b1, c3, d21), (a16, b1, c3, d22), (a16, b2, c1, d1), (a16, b2, c1, d2), (a16, b2, c1, d3), (a16, b2, c1, d4), (a16, b2, c1, d5), (a16, b2, c1, d6), (a16, b2, c1, d7), (a16, b2, c1, d8), (a16, b2, c1, d9), (a16, b2, c1, d10), (a16, b2, c1, d11), (a16, b2, c1, d12), (a16, b2, c1, d13), (a16, b2, c1, d14), (a16, b2, c1, d15), (a16, b2, c1, d16), (a16, b2, c1, d17), (a16, b2, c1, d18), (a16, b2, c1, d19), (a16, b2, c1, d20), (a16, b2, c1, d21), (a16, b2, c1, d22), (a16, b2, c2, d1), (a16, b2, c2, d2), (a16, b2, c2, d3), (a16, b2, c2, d4), (a16, b2, c2, d5), (a16, b2, c2, d6), (a16, b2, c2, d7), (a16, b2, c2, d8), (a16, b2, c2, d9), (a16, b2, c2, d10), (a16, b2, c2, d11), (a16, b2, c2, d12), (a16, b2, c2, d13), (a16, b2, c2, d14), (a16, b2, c2, d15), (a16, b2, c2, d16), (a16, b2, c2, d17), (a16, b2, c2, d18), (a16, b2, c2, d19), (a16, b2, c2, d20), (a16, b2, c2, d21), (a16, b2, c2, d22), (a16, b2, c3, d1), (a16, b2, c3, d2), (a16, b2, c3, d3), (a16, b2, c3, d4), (a16, b2, c3, d5), (a16, b2, c3, d6), (a16, b2, c3, d7), (a16, b2, c3, d8), (a16, b2, c3, d9), (a16, b2, c3, d10), (a16, b2, c3, d11), (a16, b2, c3, d12), (a16, b2, c3, d13), (a16, b2, c3, d14), (a16, b2, c3, d15), (a16, b2, c3, d16), (a16, b2, c3, d17), (a16, b2, c3, d18), (a16, b2, c3, d19), (a16, b2, c3, d20), (a16, b2, c3, d21), (a16, b2, c3, d22), (a16, b3, c1, d1), (a16, b3, c1, d2), (a16, b3, c1, d3), (a16, b3, c1, d4), (a16, b3, c1, d5), (a16, b3, c1, d6), (a16, b3, c1, d7), (a16, b3, c1, d8), (a16, b3, c1, d9), (a16, b3, c1, d10), (a16, b3, c1, d11), (a16, b3, c1, d12), (a16, b3, c1, d13), (a16, b3, c1, d14), (a16, b3, c1, d15), (a16, b3, c1, d16), (a16, b3, c1, d17), (a16, b3, c1, d18), (a16, b3, c1, d19), (a16, b3, c1, d20), (a16, b3, c1, d21), (a16, b3, c1, d22), (a16, b3, c2, d1), (a16, b3, c2, d2), (a16, b3, c2, d3), (a16, b3, c2, d4), (a16, b3, c2, d5), (a16, b3, c2, d6), (a16, b3, c2, d7), (a16, b3, c2, d8), (a16, b3, c2, d9), (a16, b3, c2, d10), (a16, b3, c2, d11), (a16, b3, c2, d12), (a16, b3, c2, d13), (a16, b3, c2, d14), (a16, b3, c2, d15), (a16, b3, c2, d16), (a16, b3, c2, d17), (a16, b3, c2, d18), (a16, b3, c2, d19), (a16, b3, c2, d20), (a16, b3, c2, d21), (a16, b3, c2, d22), (a16, b3, c3, d1), (a16, b3, c3, d2), (a16

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b1, c2, d11), (a23, b1, c2, d12), (a23, b1, c2, d13), (a23, b1, c2, d 14), (a23, b1, c2, d15), (a23, b1, c2, d16), (a23, b1, c2, d17), (a23, b1, c2, d18), (a23, b1, c2, d19), (a23, b1, c2, d20), (a23, b1, c2, d21), (a23, b1, c2, d22), (a23, b1, c3, d1), (a23, b1, c3, d2), (a23, b1, c3, d3), (a23, b1, c3, d4), (a23, b1, c3, d 5), (a23, b1, c3, d6), (a23, b1, c3, d7), (a23, b1, c3, d8), (a23, b1, c3, d9), (a23, b1, c3, d10), (a23, b1, c3, d11), (a23, b1, c3, d12), (a23, b1, c3, d13), (a23, b1, c3, d14), (a23, b1, c3, d15), (a23, b1, c3, d16), (a23, b1, c3, d17), (a23, b1, c3, d1 8), (a23, b1, c3, d19), (a23, b1, c3, d20), (a23, b1, c3, d21), (a23, b1, c3, d22), (a23, b2, c1, d1), (a23, b2, c1, d2), (a23, b2, c1, d3), (a23, b2, c1, d4), (a23, b2, c1, d5), (a23, b2, c1, d6), (a23, b2, c1, d7), (a23, b2, c1, d8), (a23, b2, c1, d9), ( a23, b2, c1, d10), (a23, b2, c1, d11), (a23, b2, c1, d12), (a23, b2, c1, d13), (a23, b2, c1, d14), (a23, b2, c1, d15), (a23, b2, c1, d16), (a23, b2, c1, d17), (a23, b2, c1, d18), (a23, b2, c1, d19), (a23, b2, c1, d20), (a23, b2, c1, d21), (a23, b2, c1, d22 ), (a23, b2, c2, d1), (a23, b2, c2, d2), (a23, b2, c2, d3), (a23, b2, c2, d4), (a23, b2, c2, d5), (a23, b2, c2, d6), (a23, b2, c2, d7), (a23, b2, c2, d8), (a23, b2, c2, d9), (a23, b2, c2, d10), (a23, b2, c2, d11), (a23, b2, c2, d12), (a23, b2, c2, d13), (a 23, b2, c2, d14), (a23, b2, c2, d15), (a23, b2, c2, d16), (a23, b2, c2, d17), (a23, b2, c2, d18), (a23, b2, c2, d19), (a23, b2, c2, d20), (a23, b2, c2, d21), (a23, b2, c2, d22), (a23, b2, c3, d1), (a23, b2, c3, d2), (a23, b2, c3, d3), (a23, b2, c3, d4), (a 23, b2, c3, d5), (a23, b2, c3, d6), (a23, b2, c3, d7), (a23, b2, c3, d8), (a23, b2, c3, d9), (a23, b2, c3, d10), (a23, b2, c3, d11), (a23, b2, c3, d12), (a23, b2, c3, d13), (a23, b2, c3, d14), (a23, b2, c3, d15), (a23, b2, c3, d16), (a23, b2, c3, d17), (a2 3, b2, c3, d18), (a23, b2, c3, d19), (a23, b2, c3, d20), (a23, b2, c3, d21), (a23, b2, c3, d22), (a23, b3, c1, d1), (a23, b3, c1, d 2), (a23, b3, c1, d3), (a23, b3, c1, d4), (a23, b3, c1, d5), (a23, b3, c1, d6), (a23, b3, c1, d7), (a23, b3, c1, d8), (a23, b 3, c1, d9), (a23, b3, c1, d10), (a23, b3, c1, d11), (a23, b3, c1, d12), (a23, b3, c1, d13), (a23, b3, c1, d14), (a23, b3, c1, d15), (a23, b3, c1, d16), (a23, b3, c1, d17), (a23, b3, c1, d18), (a23, b3, c1, d19), (a23, b3, c1, d20), (a23, b3, c1, d21), (a23, b3, c1, d22), ( a23, b3, c2, d1), (a23, b3, c2, d2), (a23, b3, c2, d3), (a23, b3, c2, d4), (a23, b3, c2, d5), (a23, b3, c2, d6), (a23, b3, c2, d7), (a23, b3, c2, d8), (a23, b3, c2, d9), (a23, b3, c2, d10), (a23, b3, c2, d11), (a23, b3, c2, d12), (a23, b3, c2, d13), (a23, b3, c2, d14), (a23, b3, c2, d15), (a23, b3, c2, d16), (a23, b3, c2, d17), (a23, b3, c2, d18), (a23, b3, c2, d19), (a23, b3, c2, d20), (a23, b3, c2, d21), (a23, b3, c2, d22), (a23, b3, c3, d1), (a23, b3, c3, d2), (a23, b3, c3, d3), (a23, b3, c3, d4), (a23, b3, c3, d5), (a23, b3, c3, d6), (a23, b3, c3, d7), (a23, b3, c3, d8), (a23, b3, c3, d9), (a23, b3, c3, d10), (a23, b3, c3, d11 ), (a23, b3, c3, d12), (a23, b3, c3, d13), (a23, b3, c3, d14), (a23, b3, c3, d15 ), (a23, b3, c3, d16), (a23, b3, c3, d17), (a23, b3, c3, d18), (a23, b3, c3, d19 ), (a23, b3, c3, d20), (a23, b3, c3, d21), (a23, b3, c3, d22), (a23, b4, c1, d1), (a23, b4, c1, d2), (a23, b4, c1, d3), (a23, b4, c1, d4), (a23, b4, c1, d5), (a23, b4, c1, d6), (a23, b4, c1, d7), (a23, b4, c1, d8), (a23, b4, c1, d9), (a23, b4, c1, d10), (a23, b4, c1, d11), (a23, b4, c1, d12), (a23, b4, c1, d13), (a23, b4, c1, d14), (a23, b4, c1, d15), (a23, b4, c1, d16), (a23, b4, c1, d17), (a23, b4, c1, d18), (a23, b4, c1, d19), (a23, b4, c1, d20), (a23, b4, c 1, d21), (a23, b4, c1, d22), (a23, b4, c2, d1), (a23, b4, c2, d2), (a23, b4, c2, d3), (a23, b4, c2, d4), (a23, b4, c2, d5), (a23, b4, c2, d6), (a23, b4, c2, d7), (a23, b4, c2, d8), (a23, b4, c2, d9), (a23, b4, c2, d10), (a23, b4, c2, d11), (a23, b4, c2, d1 2), (a23, b4, c2, d13), (a23, b4, c2, d14), (a23, b4, c2, d15), (a23, b4, c2

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4, b1, c2, d18), (a24, b1, c2, d19), (a24, b1, c2, d20), (a24, b1, c2, d21), (a24, b1, c2, d22), (a24, b1, c3, d1), (a24, b1, c3, d2), (a24, b1, c3, d3), (a24, b1, c3, d4), (a24, b1, c3, d5), (a24, b1, c3, d6), (a24, b1, c3, d7), (a24, b1, c3, d8), (a24, b1, c3, d9), (a24, b1, c3, d10), (a24, b1, c3, d11), (a24, b1, c3, d12), (a24, b1, c3, d13), (a24, b1, c3, d14), (a24, b1, c3, d15), (a24, b1, c3, d16), (a24, b1, c3, d17), (a24, b1, c3, d18), (a24, b1, c3, d19), (a24, b1, c3, d20), (a24, b1, c3, d21), (a24, b1, c3, d22), (a24, b2, c1, d1), (a24, b2, c1, d2), (a24, b2, c1, d3), (a24, b2, c1, d4), (a24, b2, c1, d5), (a24, b2, c1, d6), (a24, b2, c1, d7), (a24, b2, c1, d8), (a24, b2, c1, d9), (a24, b2, c1, d10), (a24, b2, c1, d11), (a24, b2, c1, d12), (a24, b2, c1, d13), (a24, b2, c1, d14), (a24, b2, c1, d15), (a24, b2, c1, d16), (a24, b2, c1, d17), (a24, b2, c1, d18), (a24, b2, c1, d19), (a24, b2, c1, d20), (a24, b2, c1, d21), (a24, b2, c1, d22), (a24, b2, c2, d1), (a24, b2, c2, d2), (a24, b2, c2, d3), (a24, b2, c2, d4), (a24, b2, c2, d5), (a24, b2, c2, d6), (a24, b2, c2, d7), (a24, b2, c2, d8), (a24, b2, c2, d9), (a24, b2, c2, d10), (a24, b2, c2, d11), (a24, b2, c2, d12), (a24, b2, c2, d13), (a24, b2, c2, d14), (a24, b2, c2, d15), (a24, b2, c2, d16), (a24, b2, c2, d17), (a24, b2, c2, d18), (a24, b2, c2, d19), (a24, b2, c2, d20), (a24, b2, c2, d21), (a24, b2, c2, d22), (a24, b2, c3, d1), (a24, b2, c3, d2), (a24, b2, c3, d3), (a24, b2, c3, d4), (a24, b2, c3, d5), (a24, b2, c3, d6), (a24, b2, c3, d7), (a24, b2, c3, d8), (a24, b2, c3, d9), (a24, b2, c3, d10), (a24, b2, c3, d11), (a24, b2, c3, d12), (a24, b2, c3, d13), (a24, b2, c3, d14), (a24, b2, c3, d15), (a24, b2, c3, d16), (a24, b2, c3, d17), (a24, b2, c3, d18), (a24, b2, c3, d19), (a24, b2, c3, d20), (a24, b2, c3, d21), (a24, b2, c3, d22), (a24, b3, c1, d1), (a24, b3, c1, d2), (a24, b3, c1, d3), (a24, b3, c1, d4), (a24, b3, c1, d5), (a24, b3, c1, d6), (a24, b3, c1, d7), (a24, b3, c1, d8), (a24, b3, c1, d9), (a24, b3, c1, d10), (a24, b3, c1, d11), (a24, b3, c1, d12), (a24, b3, c1, d13), (a24, b3, c1, d14), (a24, b3, c1, d15), (a24, b3, c1, d16), (a24, b3, c1, d17), (a24, b3, c1, d18), (a24, b3, c1, d19), (a24, b3, c1, d20), (a24, b3, c1, d21), (a24, b3, c1, d22), (a24, b3, c2, d1), (a24, b3, c2, d2), (a24, b3, c2, d3), (a24, b3, c2, d4), (a24, b3, c2, d5), (a24, b3, c2, d6), (a24, b3, c2, d7), (a24, b3, c2, d8), (a24, b3, c2, d9), (a24, b3, c2, d10), (a24, b3, c2, d11), (a24, b3, c2, d12), (a24, b3, c2, d13), (a24, b3, c2, d14), (a24, b3, c2, d15), (a24, b3, c2, d16), (a24, b3, c2, d17), (a24, b3, c2, d18), (a24, b3, c2, d19), (a24, b3, c2, d20), (a24, b3, c2, d21), (a24, b3, c2, d22), (a24, b3, c3, d1), (a24, b3, c3, d2), (a24, b3, c3, d3), (a24, b3, c3, d4), (a24, b3, c3, d5), (a24, b3, c3, d6), (a24, b3, c3, d7), (a24, b3, c3, d8), (a24, b3, c3, d9), (a24, b3, c3, d10), (a24, b3, c3, d11), (a24, b3, c3, d12), (a24, b3, c3, d13), (a24, b3, c3, d14), (a24, b3, c3, d15), (a24, b3, c3, d16), (a24, b3, c3, d17), (a24, b3, c3, d18), (a24, b3, c3, d19), (a24, b3, c3, d20), (a24, b3, c3, d21), (a24, b3, c3, d22), (a24, b4, c1, d1), (a24, b4, c1, d2), (a24, b4, c1, d3), (a24, b4, c1, d4), (a24, b4, c1, d5), (a24, b4, c1, d6), (a24, b4, c1, d7), (a24, b4, c1, d8), (a24, b4, c1, d9), (a24, b4, c1, d10), (a24, b4, c1, d11), (a24, b4, c1, d12), (a24, b4, c1, d13), (a24, b4, c1, d14), (a24, b4, c1, d15), (a24, b4, c1, d16), (a24, b4, c1, d17), (a24, b4, c1, d18), (a24, b4, c1, d19), (a24, b4, c1, d20), (a24, b4, c1, d21), (a24, b4, c1, d22), (a24, b4, c2, d1), (a24, b4, c2, d2), (a24, b4, c2, d3), (a24, b4, c2, d4), (a24, b4, c2, d5), (a24, b4, c2, d6), (a24, b4, c2, d7), (a24, b4, c2, d8), (a24, b4, c2, d9), (a24, b4, c2, d10), (a24, b4, c2, d11), (a24, b4, c2, d12), (a24, b4, c2, d13), (a24, b4, c2, d14), (a24, b4, c2, d15), (a24, b4, c2, d16), (a24, b4, c2, d17), (a24, b4, c2, d18), (a24, b4, c2, d19), (a24, b4, c2, d20), (a24, b4, c2, d21), (a24, b4, c2, d22), (a24, b4, c3

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c3, d21), (a27, b1, c3, d22), (a27, b2, c1, d1), (a27, b2, c1, d2), (a27, b2, c1, d3), (a27, b2, c1, d4), (a27, b2, c1, d5), (a27, b2, c1, d6), (a 27, b2, c1, d7), (a27, b2, c1, d8), (a27, b2, c1, d9), (a27, b2, c1, d10), (a27, b2, c1, d11), (a27, b2, c1, d12), (a27, b2, c1, d13), (a27, b2, c1, d14), (a27, b2, c1, d15), (a27, b2, c1, d16), (a27, b2, c1, d17), (a27, b2, c1, d18), (a27, b2, c1, d19), ( a27, b2, c1, d20), (a27, b2, c1, d21), (a27, b2, c1, d22), (a27, b2, c2, d1), (a27, b2, c2, d2), (a27, b2, c2, d3), (a27, b2, c2, d4), (a27, b2, c2, d5), ( a27, b2, c2, d6), (a27, b2, c2, d7), (a27, b2, c2, d8), (a27, b2, c2, d9), (a27, b2, c2, d10), (a27, b2, c2, d11), (a27, b2, c2, d12), (a27, b2, c2, d13), (a27, b2, c2, d14), (a27, b2, c2, d15), (a27, b2, c2, d16), (a27, b2, c2, d17), (a27, b2, c2, d18), (a27, b2, c2, d19), (a27, b2, c2, d20), (a27, b2, c2, d21), (a27, b2, c2, d22), (a27, b2, c3, d1), (a2 7, b2, c3, d2), (a27, b2, c3, d3), (a27, b2, c3, d4), (a27, b2, c3, d5), (a27, b2, c3, d6), (a27, b2, c3, d7), (a27, b2, c3, d8), (a27, b2, c3, d9), (a27, b2, c3, d10), (a27, b2, c3, d11), (a27, b2, c3, d12), (a27, b2, c3, d13), (a27, b2, c3, d14), (a27, b 2, c3, d15), (a27, b2, c3, d16), (a27, b2, c3, d17), (a27, b2, c3, d18), (a27, b2, c3, d19), (a27, b2, c3, d20), (a27, b2, c3, d21), (a27, b2, c3, d22), (a27, b3, c1, d1), (a27, b3, c1, d2), (a27, b3, c1, d3), (a27, b3, c1, d4), (a27, b3, c1, d5), (a27, b3, c1, d6), (a27, b3, c1, d7), (a27, b3, c1, d8), (a27, b3, c1, d9), (a27, b3, c1, d10), (a27, b3, c1, d11), (a27, b3, c1, d12), (a27, b3, c1, d13), (a27, b3, c1, d14), (a27, b3, c1, d15), (a27, b3, c1, d16), (a27, b3, c1, d17), (a27, b3, c1, d18), (a27, b3, c1, d19), (a27, b3, c1, d20), (a27, b3, c1, d21), (a27, b3, c1, d22), (a27, b3, c2, d1), (a27, b3, c2, d2), (a27, b3, c2, d3), (a27, b3, c2, d4), (a2 7, b3, c2, d5), (a27, b3, c2, d6), (a27, b3, c2, d7), (a27, b3, c2, d8), (a27, b3, c2, d9), (a27, b3, c2, d10), (a27, b3, c2, d11), (a27, b3, c2, d12), (a27, b3, c2, d13), (a27, b3, c2, d14), (a27, b3, c2, d15), (a27, b3, c2, d16), (a27, b3, c2, d17), (a27, b3, c2, d18), (a27, b3, c2, d19), (a27, b3, c2, d20), (a27, b3, c2, d21), (a27, b3, c2, d22), (a27, b3, c3, d1), (a27, b3, c3, d2), (a27, b3, c3, d3), (a27, b3, c3, d4), (a27, b3, c3, d5), (a27, b3, c3, d6), (a27, b3, c3, d7), (a27, b3, c3, d8), (a27, b3, c3, d9), (a27, b3, c3, d10), (a27, b3, c3, d 11), (a27, b3, c3, d12), (a27, b3, c3, d13), (a27, b3, c3, d14), (a27, b3, c3, d15), (a27, b3, c3, d16), (a27, b3, c3, d17), (a27, b3, c3, d18), (a27, b3, c3, d19), (a27, b3, c3, d20), (a27, b3, c3, d21), (a27, b3, c3, d22), (a27, b4, c1, d 1), (a27, b4, c1, d2), (a27, b4, c1, d3), (a27, b4, c1, d4), (a27, b4, c1, d5), (a27, b4, c1, d6), (a27, b4, c1, d7), (a27, b4, c1, d8), (a27, b4, c1, d9), (a27, b4, c1, d10), (a27, b4, c1, d11), (a27, b4, c1, d12), (a27, b4, c1, d13), (a27, b4, c1, d14), (a27, b4, c1, d15), (a27, b4, c1, d16), (a27, b4, c1, d17), (a27, b4, c1, d 18), (a27, b4, c1, d19), (a27, b4, c1, d20), (a27, b4, c1, d21), (a2 7, b4, c1, d22), (a27, b4, c2, d1), (a27, b4, c2, d2), (a27, b4, c2, d3), (a27, b4, c2, d4), (a27, b4, c2, d5), (a27, b4, c2, d6), (a27, b4, c2, d7), (a27, b4, c2, d8), (a27, b4, c2, d9), (a27, b4, c2, d10), (a27, b4, c2, d11), (a27, b4, c2, d12), (a27, b4, c2, d13), (a27, b4, c2, d14), (a27, b4, c2, d15), (a27, b4, c2, d16), (a27, b4, c2, d17), (a27, b4, c2, d18), (a27, b4, c2, d19), (a27, b4, c2, d20), (a27, b4, c2, d21), (a27, b4, c2, d2 2), (a27, b4, c3, d1), (a27, b4, c3, d2), (a27, b4, c3, d3), (a27, b4, c3, d4), (a27, b4, c3, d5), (a27, b4, c3, d6), (a27, b4, c3, d7), (a27, b4, c3, d8), (a27, b4, c3, d9), (a27, b4, c3, d10), (a27, b4, c3, d11), (a27, b4, c3, d12), (a27, b4, c3, d13), ( a27, b4, c3, d14), (a27, b4, c3, d15), (a27, b4, c3, d16), (a27, b4, c3, d17), (a27, b4, c3, d18), (a27, b4, c3, d19), (a27, b4, c3, d20), (a27, b4, c3, d21), (a27, b4, c3, d22), (a27, b5, c1, d1), (a27, b5, c1, d2), (a27, b5, c1, d3), (a27, b5, c1, d4), ( a

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27, b5, c1, d5), (a27, b5, c1, d6), (a27, b5, c1, d7), (a27, b5, c1, d8), (a27, b5, c1, d9), (a27, b5, c1, d10), (a27, b5, c1, d11), (a27, b5, c1, d12), (a27, b5, c1, d13), (a27, b5, c1, d14), (a27, b5, c1, d15), (a27, b5, c1, d16), (a27, b5, c1, d17), (a 27, b5, c1, d18), (a27, b5, c1, d19), (a27, b5, c1, d20), (a27, b5, c1, d21), (a27, b5, c1, d22), (a27, b5, c2, d1), (a27, b5, c2, d2), (a27, b5, c2, d3), (a27, b5, c2, d4), (a27, b5, c2, d5), (a27, b5, c2, d6), (a27, b5, c2, d7), (a27, b5, c2, d8), (a27, b5, c2, d9), (a27, b5, c2, d10), (a27, b5, c2, d11), (a27, b5, c2, d12), (a27, b5, c2, d13), (a27, b5, c2, d14), (a27, b5, c2, d15), (a27, b5, c2, d16), (a27, b5, c2, d17), (a27, b5, c2, d18), (a27, b5, c2, d19), (a27, b5, c2, d20), (a27, b5, c2, d21), (a2 7, b5, c2, d22), (a27, b5, c3, d1), (a27, b5, c3, d2), (a27, b5, c3, d3), (a27, b5, c3, d4), (a27, b5, c3, d5), (a27, b5, c3, d6), (a27, b5, c3, d7), (a27, b5, c3, d8), (a27, b5, c3, d9), (a27, b5, c3, d10), (a27, b5, c3, d11), (a27, b5, c3, d12), (a27, b5, c3, d13), (a27, b5, c3, d14), (a27, b5, c3, d15), (a27, b5, c3, d16), (a27, b5, c3, d17), (a27, b5, c3, d18), (a27, b5, c3, d19), (a27, b5, c3, d20), (a27, b5, c3, d21), (a27, b5, c3, d22), (a27, b6, c1, d1), (a27, b6, c1, d2), (a27, b6, c1, d3), (a27, b6, c1, d4), (a27, b6, c1, d5), (a27, b6, c1, d6), (a27, b6, c1, d7), (a27, b6, c1, d8), (a27, b6, c1, d9), (a27, b6, c1, d10), (a27, b6, c1, d11), (a27, b6, c1, d12), (a27, b6, c1, d13), (a27, b6, c1, d14), (a27, b6, c1, d15), (a27, b6, c1, d16), (a27, b6, c1, d17), (a27, b6, c1, d18), (a27, b6, c1, d19), (a27, b6, c1, d20), (a27, b6, c1, d21), (a27, b6, c1, d22), (a27, b6, c2, d1), (a27, b6, c2, d2), (a27, b6, c2, d3), (a27, b6, c2, d4), (a27, b6, c2, d5), (a27, b6, c2, d6), (a27, b6, c2, d7), (a27, b6, c2, d8), (a27, b6, c2, d9), (a27, b6, c2, d10), (a27, b6, c2, d11), (a27, b6, c2, d12), (a27, b6, c2, d13), (a27, b6, c2, d14), (a27, b6, c2, d15), (a27, b6, c2, d16), (a27, b6, c2, d17), (a27, b6, c2, d18), (a27, b6, c2, d19), (a27, b6, c2, d20), (a27, b6, c2, d21), (a27, b6, c2, d22), (a27, b6, c3, d1), (a27, b6, c3, d2), (a27, b6, c3, d3), (a27, b6, c3, d4), (a27, b6, c3, d5), (a27, b6, c3, d6), (a27, b6, c3, d7), (a27, b6, c3, d8), (a27, b6, c3, d9), (a27, b6, c3, d10), (a27, b6, c3, d11), (a27, b6, c3, d12), (a27, b6, c3, d13), (a27, b6, c3, d14), (a27, b6, c3, d15), (a27, b6, c3, d16), (a27, b6, c3, d17), (a27, b6, c3, d18), (a27, b6, c3, d19), (a27, b6, c3, d20), (a27, b6, c3, d21), (a27, b6, c3, d22), (a28, b1, c1, d1), (a28, b1, c1, d2), (a28, b1, c1, d3), (a28, b1, c1, d4), (a28, b1, c1, d5), (a28, b1, c1, d6), (a28, b1, c1, d7), (a28, b1, c1, d8), (a28, b1, c1, d9), (a28, b1, c1, d10), (a28, b1, c1, d11), (a28, b1, c1, d12), (a28, b1, c1, d13), (a28, b1, c1, d14), (a28, b1, c1, d15), (a28, b1, c1, d16), (a28, b1, c1, d17), (a28, b1, c1, d18), (a28, b1, c1, d19), (a28, b1, c1, d20), (a28, b1, c1, d21), (a28, b1, c1, d22), (a28, b1, c2, d1), (a28, b1, c2, d2), (a28, b1, c2, d3), (a28, b1, c2, d4), (a28, b1, c2, d5), (a28, b1, c2, d6), (a28, b1, c2, d7), (a28, b1, c2, d8), (a28, b1, c2, d9), (a28, b1, c2, d10), (a28, b1, c2, d11), (a28, b1, c2, d12), (a28, b1, c2, d13), (a28, b1, c2, d14), (a28, b1, c2, d15), (a28, b1, c2, d16), (a28, b1, c2, d17), (a28, b1, c2, d18), (a28, b1, c2, d19), (a28, b1, c2, d20), (a28, b1, c2, d21), (a28, b1, c2, d22), (a28, b1, c3, d1), (a28, b1, c3, d2), (a28, b1, c3, d3), (a28, b1, c3, d4), (a28, b1, c3, d5), (a28, b1, c3, d6), (a28, b1, c3, d7), (a28, b1, c3, d8), (a28, b1, c3, d9), (a28, b1, c3, d10), (a28, b1, c3, d11), (a28, b1, c3, d12), (a28, b1, c3, d13), (a28, b1, c3, d14), (a28, b1, c3, d15), (a28, b1, c3, d16), (a28, b1, c3, d17), (a28, b1, c3, d18), (a28, b1, c3, d19), (a28, b1, c3, d20), (a28, b1, c3, d21), (a28, b1, c3, d22), (a28, b2, c1, d1), (a28, b2, c1, d2), (a28, b2, c1, d3), (a28, b2, c1, d4), (a28, b2, c1, d5), (a28, b2, c1,

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d13), (a29, b2, c1, d14), (a29, b2, c1, d15), (a29, b2, c1, d16), (a29, b2, c1, d17), (a29, b2, c1, d18), (a29, b2, c1, d19), (a29, b2, c1, d20), (a29, b2, c1, d21), (a29, b2, c1, d22), (a29, b2, c2, d1), (a29, b2, c2, d2), (a29, b2, c2, d3), (a29, b2, c2, d4), (a29, b2, c2, d5), (a29, b2, c2, d6), (a29, b2, c2, d7), (a29, b2, c2, d8), (a29, b2, c2, d9), (a29, b2, c2, d10), (a29, b2, c2, d11), (a29, b2, c2, d12), (a29, b2, c2, d13), (a29, b2, c2, d14), (a29, b2, c2, d15), (a29, b2, c2, d16), (a29, b2, c2, d17), (a29, b2, c2, d18), (a29, b2, c2, d19), (a29, b2, c2, d20), (a29, b2, c2, d21), (a29, b2, c2, d22), (a29, b2, c3, d1), (a29, b2, c3, d2), (a29, b2, c3, d3), (a29, b2, c3, d4), (a29, b2, c3, d5), (a29, b2, c3, d6), (a29, b2, c3, d7), (a29, b2, c3, d8), (a29, b2, c3, d9), (a29, b2, c3, d10), (a29, b2, c3, d11), (a29, b2, c3, d12), (a29, b2, c3, d13), (a29, b2, c3, d14), (a29, b2, c3, d15), (a29, b2, c3, d16), (a29, b2, c3, d17), (a29, b2, c3, d18), (a29, b2, c3, d19), (a29, b2, c3, d20), (a29, b2, c3, d21), (a29, b2, c3, d22), (a29, b3, c1, d1), (a29, b3, c1, d2), (a29, b3, c1, d3), (a29, b3, c1, d4), (a29, b3, c1, d5), (a29, b3, c1, d6), (a29, b3, c1, d7), (a29, b3, c1, d8), (a29, b3, c1, d9), (a29, b3, c1, d10), (a29, b3, c1, d11), (a29, b3, c1, d12), (a29, b3, c1, d13), (a29, b3, c1, d14), (a29, b3, c1, d15), (a29, b3, c1, d16), (a29, b3, c1, d17), (a29, b3, c1, d18), (a29, b3, c1, d19), (a29, b3, c1, d20), (a29, b3, c1, d21), (a29, b3, c1, d22), (a29, b3, c2, d1), (a29, b3, c2, d2), (a29, b3, c2, d3), (a29, b3, c2, d4), (a29, b3, c2, d5), (a29, b3, c2, d6), (a29, b3, c2, d7), (a29, b3, c2, d8), (a29, b3, c2, d9), (a29, b3, c2, d10), (a29, b3, c2, d11), (a29, b3, c2, d12), (a29, b3, c2, d13), (a29, b3, c2, d14), (a29, b3, c2, d15), (a29, b3, c2, d16), (a29, b3, c2, d17), (a29, b3, c2, d18), (a29, b3, c2, d19), (a29, b3, c2, d20), (a29, b3, c2, d21), (a29, b3, c2, d22), (a29, b3, c3, d1), (a29, b3, c3, d2), (a29, b3, c3, d3), (a29, b3, c3, d4), (a29, b3, c3, d5), (a29, b3, c3, d6), (a29, b3, c3, d7), (a29, b3, c3, d8), (a29, b3, c3, d9), (a29, b3, c3, d10), (a29, b3, c3, d11), (a29, b3, c3, d12), (a29, b3, c3, d13), (a29, b3, c3, d14), (a29, b3, c3, d15), (a29, b3, c3, d16), (a29, b3, c3, d17), (a29, b3, c3, d18), (a29, b3, c3, d19), (a29, b3, c3, d20), (a29, b3, c3, d21), (a29, b3, c3, d22), (a29, b4, c1, d1), (a29, b4, c1, d2), (a29, b4, c1, d3), (a29, b4, c1, d4), (a29, b4, c1, d5), (a29, b4, c1, d6), (a29, b4, c1, d7), (a29, b4, c1, d8), (a29, b4, c1, d9), (a29, b4, c1, d10), (a29, b4, c1, d11), (a29, b4, c1, d12), (a29, b4, c1, d13), (a29, b4, c1, d14), (a29, b4, c1, d15), (a29, b4, c1, d16), (a29, b4, c1, d17), (a29, b4, c1, d18), (a29, b4, c1, d19), (a29, b4, c1, d20), (a29, b4, c1, d21), (a29, b4, c1, d22), (a29, b4, c2, d1), (a29, b4, c2, d2), (a29, b4, c2, d3), (a29, b4, c2, d4), (a29, b4, c2, d5), (a29, b4, c2, d6), (a29, b4, c2, d7), (a29, b4, c2, d8), (a29, b4, c2, d9), (a29, b4, c2, d10), (a29, b4, c2, d11), (a29, b4, c2, d12), (a29, b4, c2, d13), (a29, b4, c2, d14), (a29, b4, c2, d15), (a29, b4, c2, d16), (a29, b4, c2, d17), (a29, b4, c2, d18), (a29, b4, c2, d19), (a29, b4, c2, d20), (a29, b4, c2, d21), (a29, b4, c2, d22), (a29, b4, c3, d1), (a29, b4, c3, d2), (a29, b4, c3, d3), (a29, b4, c3, d4), (a29, b4, c3, d5), (a29, b4, c3, d6), (a29, b4, c3, d7), (a29, b4, c3, d8), (a29, b4, c3, d9), (a29, b4, c3, d10), (a29, b4, c3, d11), (a29, b4, c3, d12), (a29, b4, c3, d13), (a29, b4, c3, d14), (a29, b4, c3, d15), (a29, b4, c3, d16), (a29, b4, c3, d17), (a29, b4, c3, d18), (a29, b4, c3, d19), (a29, b4, c3, d20), (a29, b4, c3, d21), (a29, b4, c3, d22), (a29, b5, c1, d1), (a29, b5, c1, d2), (a29, b5, c1, d3), (a29, b5, c1, d4), (a29, b5, c1, d5), (a29, b5, c1, d6), (a29, b5, c1, d7), (a29, b5, c1, d8), (a29, b5, c1, d9), (a29, b5, c1, d10), (a29, b5, c1, d11), (a29, b5, c1, d12), (a29, b5, c1, d13), (a29, b5, c1, d14), (a29, b5, c1, d15), (a29, b5, c1, d16), (a29, b5, c1, d17), (a29, b5, c1, d18), (a29,

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d18), (a43, b3, c3, d19), (a43, b3, c3, d20), (a43, b3, c3, d21), (a43, b3, c3, d22), (a43, b4, c1, d1), (a43, b4, c1, d2), (a43, b4, c1, d3), (a43, b4, c1, d4), (a43, b4, c1, d5), (a43, b4, c1, d6), (a43, b4, c1, d7), (a43, b4, c1, d8), (a43, b4, c1, d9), (a43, b4, c1, d10), (a43, b4, c1, d11), (a43, b4, c1, d12), (a43, b4, c1, d13), (a43, b4, c1, d14), (a43, b4, c1, d15), (a43, b4, c1, d16), (a43, b4, c1, d17), (a43, b4, c1, d18), (a43, b4, c1, d19), (a43, b4, c1, d20), (a43, b4, c1, d21), (a43, b4, c1, d22), (a43, b4, c2, d1), (a43, b4, c2, d2), (a43, b4, c2, d3), (a43, b4, c2, d4), (a43, b4, c2, d5), (a43, b4, c2, d6), (a43, b4, c2, d7), (a43, b4, c2, d8), (a43, b4, c2, d9), (a43, b4, c2, d10), (a43, b4, c2, d11), (a43, b4, c2, d12), (a43, b4, c2, d13), (a43, b4, c2, d14), (a43, b4, c2, d15), (a43, b4, c2, d16), (a43, b4, c2, d17), (a43, b4, c2, d18), (a43, b4, c2, d19), (a43, b4, c2, d20), (a43, b4, c2, d21), (a43, b4, c2, d22), (a43, b4, c3, d1), (a43, b4, c3, d2), (a43, b4, c3, d3), (a43, b4, c3, d4), (a43, b4, c3, d5), (a43, b4, c3, d6), (a43, b4, c3, d7), (a43, b4, c3, d8), (a43, b4, c3, d9), (a43, b4, c3, d10), (a43, b4, c3, d11), (a43, b4, c3, d12), (a43, b4, c3, d13), (a43, b4, c3, d14), (a43, b4, c3, d15), (a43, b4, c3, d16), (a43, b4, c3, d17), (a43, b4, c3, d18), (a43, b4, c3, d19), (a43, b4, c3, d20), (a43, b4, c3, d21), (a43, b4, c3, d22), (a43, b5, c1, d1), (a43, b5, c1, d2), (a43, b5, c1, d3), (a43, b5, c1, d4), (a43, b5, c1, d5), (a43, b5, c1, d6), (a43, b5, c1, d7), (a43, b5, c1, d8), (a43, b5, c1, d9), (a43, b5, c1, d10), (a43, b5, c1, d11), (a43, b5, c1, d12), (a43, b5, c1, d13), (a43, b5, c1, d14), (a43, b5, c1, d15), (a43, b5, c1, d16), (a43, b5, c1, d17), (a43, b5, c1, d18), (a43, b5, c1, d19), (a43, b5, c1, d20), (a43, b5, c1, d21), (a43, b5, c1, d22), (a43, b5, c2, d1), (a43, b5, c2, d2), (a43, b5, c2, d3), (a43, b5, c2, d4), (a43, b5, c2, d5), (a43, b5, c2, d6), (a43, b5, c2, d7), (a43, b5, c2, d8), (a43, b5, c2, d9), (a43, b5, c2, d10), (a43, b5, c2, d11), (a43, b5, c2, d12), (a43, b5, c2, d13), (a43, b5, c2, d14), (a43, b5, c2, d15), (a43, b5, c2, d16), (a43, b5, c2, d17), (a43, b5, c2, d18), (a43, b5, c2, d19), (a43, b5, c2, d20), (a43, b5, c2, d21), (a43, b5, c2, d22), (a43, b5, c3, d1), (a43, b5, c3, d2), (a43, b5, c3, d3), (a43, b5, c3, d4), (a43, b5, c3, d5), (a43, b5, c3, d6), (a43, b5, c3, d7), (a43, b5, c3, d8), (a43, b5, c3, d9), (a43, b5, c3, d10), (a43, b5, c3, d11), (a43, b5, c3, d12), (a43, b5, c3, d13), (a43, b5, c3, d14), (a43, b5, c3, d15), (a43, b5, c3, d16), (a43, b5, c3, d17), (a43, b5, c3, d18), (a43, b5, c3, d19), (a43, b5, c3, d20), (a43, b5, c3, d21), (a43, b5, c3, d22), (a43, b6, c1, d1), (a43, b6, c1, d2), (a43, b6, c1, d3), (a43, b6, c1, d4), (a43, b6, c1, d5), (a43, b6, c1, d6), (a43, b6, c1, d7), (a43, b6, c1, d8), (a43, b6, c1, d9), (a43, b6, c1, d10), (a43, b6, c1, d11), (a43, b6, c1, d12), (a43, b6, c1, d13), (a43, b6, c1, d14), (a43, b6, c1, d15), (a43, b6, c1, d16), (a43, b6, c1, d17), (a43, b6, c1, d18), (a43, b6, c1, d19), (a43, b6, c1, d20), (a43, b6, c1, d21), (a43, b6, c1, d22), (a43, b6, c2, d1), (a43, b6, c2, d2), (a43, b6, c2, d3), (a43, b6, c2, d4), (a43, b6, c2, d5), (a43, b6, c2, d6), (a43, b6, c2, d7), (a43, b6, c2, d8), (a43, b6, c2, d9), (a43, b6, c2, d10), (a43, b6, c2, d11), (a43, b6, c2, d12), (a43, b6, c2, d13), (a43, b6, c2, d14), (a43, b6, c2, d15), (a43, b6, c2, d16), (a43, b6, c2, d17), (a43, b6, c2, d18), (a43, b6, c2, d19), (a43, b6, c2, d20), (a43, b6, c2, d21), (a43, b6, c2, d22), (a43, b6, c3, d1), (a43, b6, c3, d2), (a43, b6, c3, d3), (a43, b6, c3, d4), (a43, b6, c3, d5), (a43, b6, c3, d6), (a43, b6, c3, d7), (a43, b6, c3, d8), (a43, b6, c3, d9), (a43, b6, c3, d10), (a43, b6, c3, d11), (a43, b6, c3, d12), (a43, b6, c3, d13), (a43, b6, c3, d14), (a43, b6, c3, d15), (a43, b6, c3, d16), (a43, b6, c3, d17), (a43, b6, c3, d18), (a43, b6, c3, d19), (a43, b6, c3, d20), (a43, b6, c3, d21), (a43, b6, c3, d22)

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d3), (a44, b4, c1, d4), (a44, b4, c1, d5), (a44, b4, c1, d6), (a44, b4, c1, d7), (a44, b4, c1, d8), (a44, b4, c1, d9), (a44, b4, c1, d10), (a44, b4, c1, d11), (a44, b4, c1, d12), (a44, b4, c1, d13), (a44, b4, c1, d14), (a44, b4, c1, d15), (a44, b4, c1, d16), (a44, b4, c1, d17), (a44, b4, c1, d18), (a44, b4, c1, d19), (a44, b4, c1, d20), (a44, b4, c1, d21), (a44, b4, c1, d22), (a44, b4, c2, d1), (a44, b4, c2, d2), (a44, b4, c2, d3), (a44, b4, c2, d4), (a44, b4, c2, d5), (a44, b4, c2, d6), (a44, b4, c2, d7), (a44, b4, c2, d8), (a44, b4, c2, d9), (a44, b4, c2, d10), (a44, b4, c2, d11), (a44, b4, c2, d12), (a44, b4, c2, d13), (a44, b4, c2, d14), (a44, b4, c2, d15), (a44, b4, c2, d16), (a44, b4, c2, d17), (a44, b4, c2, d18), (a44, b4, c2, d19), (a44, b4, c2, d20), (a44, b4, c2, d21), (a44, b4, c2, d22), (a44, b4, c3, d1), (a44, b4, c3, d2), (a44, b4, c3, d3), (a44, b4, c3, d4), (a44, b4, c3, d5), (a44, b4, c3, d6), (a44, b4, c3, d7), (a44, b4, c3, d8), (a44, b4, c3, d9), (a44, b4, c3, d10), (a44, b4, c3, d11), (a44, b4, c3, d12), (a44, b4, c3, d13), (a44, b4, c3, d14), (a44, b4, c3, d15), (a44, b4, c3, d16), (a44, b4, c3, d17), (a44, b4, c3, d18), (a44, b4, c3, d19), (a44, b4, c3, d20), (a44, b4, c3, d21), (a44, b4, c3, d22), (a44, b5, c1, d1), (a44, b5, c1, d2), (a44, b5, c1, d3), (a44, b5, c1, d4), (a44, b5, c1, d5), (a44, b5, c1, d6), (a44, b5, c1, d7), (a44, b5, c1, d8), (a44, b5, c1, d9), (a44, b5, c1, d10), (a44, b5, c1, d11), (a44, b5, c1, d12), (a44, b5, c1, d13), (a44, b5, c1, d14), (a44, b5, c1, d15), (a44, b5, c1, d16), (a44, b5, c1, d17), (a44, b5, c1, d18), (a44, b5, c1, d19), (a44, b5, c1, d20), (a44, b5, c1, d21), (a44, b5, c1, d22), (a44, b5, c2, d1), (a44, b5, c2, d2), (a44, b5, c2, d3), (a44, b5, c2, d4), (a44, b5, c2, d5), (a44, b5, c2, d6), (a44, b5, c2, d7), (a44, b5, c2, d8), (a44, b5, c2, d9), (a44, b5, c2, d10), (a44, b5, c2, d11), (a44, b5, c2, d12), (a44, b5, c2, d13), (a44, b5, c2, d14), (a44, b5, c2, d15), (a44, b5, c2, d16), (a44, b5, c2, d17), (a44, b5, c2, d18), (a44, b5, c2, d19), (a44, b5, c2, d20), (a44, b5, c2, d21), (a44, b5, c2, d22), (a44, b5, c3, d1), (a44, b5, c3, d2), (a44, b5, c3, d3), (a44, b5, c3, d4), (a44, b5, c3, d5), (a44, b5, c3, d6), (a44, b5, c3, d7), (a44, b5, c3, d8), (a44, b5, c3, d9), (a44, b5, c3, d10), (a44, b5, c3, d11), (a44, b5, c3, d12), (a44, b5, c3, d13), (a44, b5, c3, d14), (a44, b5, c3, d15), (a44, b5, c3, d16), (a44, b5, c3, d17), (a44, b5, c3, d18), (a44, b5, c3, d19), (a44, b5, c3, d20), (a44, b5, c3, d21), (a44, b5, c3, d22), (a44, b6, c1, d1), (a44, b6, c1, d2), (a44, b6, c1, d3), (a44, b6, c1, d4), (a44, b6, c1, d5), (a44, b6, c1, d6), (a44, b6, c1, d7), (a44, b6, c1, d8), (a44, b6, c1, d9), (a44, b6, c1, d10), (a44, b6, c1, d11), (a44, b6, c1, d12), (a44, b6, c1, d13), (a44, b6, c1, d14), (a44, b6, c1, d15), (a44, b6, c1, d16), (a44, b6, c1, d17), (a44, b6, c1, d18), (a44, b6, c1, d19), (a44, b6, c1, d20), (a44, b6, c1, d21), (a44, b6, c1, d22), (a44, b6, c2, d1), (a44, b6, c2, d2), (a44, b6, c2, d3), (a44, b6, c2, d4), (a44, b6, c2, d5), (a44, b6, c2, d6), (a44, b6, c2, d7), (a44, b6, c2, d8), (a44, b6, c2, d9), (a44, b6, c2, d10), (a44, b6, c2, d11), (a44, b6, c2, d12), (a44, b6, c2, d13), (a44, b6, c2, d14), (a44, b6, c2, d15), (a44, b6, c2, d16), (a44, b6, c2, d17), (a44, b6, c2, d18), (a44, b6, c2, d19), (a44, b6, c2, d20), (a44, b6, c2, d21), (a44, b6, c2, d22), (a44, b6, c3, d1), (a44, b6, c3, d2), (a44, b6, c3, d3), (a44, b6, c3, d4), (a44, b6, c3, d5), (a44, b6, c3, d6), (a44, b6, c3, d7), (a44, b6, c3, d8), (a44, b6, c3, d9), (a44, b6, c3, d10), (a44, b6, c3, d11), (a44, b6, c3, d12), (a44, b6, c3, d13), (a44, b6, c3, d14), (a44, b6, c3, d15), (a44, b6, c3, d16), (a44, b6, c3, d17), (a44, b6, c3, d18), (a44, b6, c3, d19), (a44, b6, c3, d20), (a44, b6, c3, d21), (a44, b6, c3, d22), (a45, b1, c1, d1), (a45, b1, c1, d2), (a45, b1, c1, d3), (a45, b1, c1, d4), (a45

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d18), (a52, b1, c3, d19), (a52, b1, c3, d20) , (a52, b1, c3, d21), (a52, b1, c3, d22), (a52, b2, c1, d1), (a52, b2, c1, d2), (a52, b2, c1, d3), (a52, b2, c1, d4), (a52, b2, c1, d5), (a52, b2, c1, d6), (a52, b2, c1, d7), (a52, b2, c1, d8), (a52, b2, c1, d9), (a52, b2, c1, d10), (a52, b2, c1, d11), (a5 2, b2, c1, d12), (a52, b2, c1, d13), (a52, b2, c1, d14), (a52, b2, c1, d15), (a52, b2, c1, d16), (a52, b2, c1, d17), (a52, b2, c1, d18), (a52, b2, c1, d19), (a52, b2, c1, d20), (a52, b2, c1, d21), (a52, b2, c1, d22), (a52, b2, c2, d1), (a52, b2, c2, d2), (a52, b2, c2, d3), (a52, b2, c2, d4), (a52, b2, c2, d5), (a52, b2, c2, d6), (a52, b2, c2, d7), (a52, b2, c2, d8), (a52, b2, c2, d9), (a52, b2, c2, d10), (a52, b2, c2, d11), (a52, b2, c2, d12), (a52, b2, c2, d13), (a52, b2, c2, d14), (a52, b2, c2, d15), (a52, b2, c2, d16), (a52, b2, c2, d17), (a52, b2, c2, d18), (a52, b2, c2, d19), (a52, b2, c2, d20), (a52, b2, c2, d21), (a52, b2, c2, d22), (a52, b2, c3, d1), (a52, b2, c3, d2), (a52, b2, c3, d3), (a52, b2, c3, d4), (a52, b2, c3, d5), (a52, b2, c3, d6), (a52, b2, c3, d7), (a52, b2, c3, d8), (a52, b2, c3, d9), (a52, b2, c3, d10), (a52, b2, c3, d11), (a52, b2, c3, d12), (a52, b2, c3, d13), (a52, b2, c3, d14), (a52, b2, c3, d15), (a52, b2, c3, d16), (a52, b2, c3, d17), (a52, b2, c3, d18), (a52, b2, c3, d19), (a52, b2, c3, d20), (a52, b2, c3, d21), (a52, b2, c3, d22), (a52, b3, c1, d1), (a52, b3, c1, d2), (a52, b3, c1, d3), (a52, b3, c1, d4), (a52, b3, c1, d5), (a52, b3, c1, d6), (a52, b3, c1, d7), (a52, b3, c1, d8), (a52, b3, c1, d9), (a52, b3, c1, d10), (a52, b3, c1, d11), (a52, b3, c1, d12), (a52, b3, c1, d13), (a52, b3, c1, d14), (a52, b3, c1, d15), (a52, b3, c1, d16), (a52, b3, c1, d17), (a52, b3, c1, d18), (a52, b3, c1, d19), (a52, b3, c1, d20), (a52, b3, c1, d21), (a52, b3, c1, d22), (a52, b3, c2, d1), (a52, b3, c2, d2), (a52, b3, c2, d3), (a52, b3, c2, d4), (a52, b3, c2, d5), (a52, b3, c2, d6), (a52, b3, c2, d7), (a52, b3, c2, d8), (a52, b3, c2, d9), (a52, b3, c2, d10), (a52, b3, c2, d11), (a52, b3, c2, d12), (a52, b3, c2, d13), (a52, b3, c2, d14), (a52, b3, c2, d15), (a52, b3, c2, d16), (a52, b3, c2, d17), (a52, b3, c2, d18), (a52, b3, c2, d19), (a52, b3, c2, d20), (a52, b3, c2, d21), (a52, b3, c2, d22), (a52, b3, c3, d1), (a52, b3, c3, d2), (a52, b3, c3, d3), (a52, b3, c3, d4), (a52, b3, c3, d5), (a52, b3, c3, d6), (a52, b3, c3, d7), (a52, b3, c3, d8), (a52, b3, c3, d9), (a52, b3, c3, d10), (a52, b3, c3, d11), (a52, b3, c3, d12), (a52, b3, c3, d13), (a52, b3, c3, d14), (a52, b3, c3, d15), (a52, b3, c3, d16), (a52, b3, c3, d17), (a52, b3, c3, d18), (a52, b3, c3, d19), (a52, b3, c3, d20), (a52, b3, c3, d21), (a52, b3, c3, d22), (a52, b4, c1, d1), (a52, b4, c1, d2), (a52, b4, c1, d3), (a52, b4, c1, d4), (a52, b4, c1, d5), (a52, b4, c1, d6), (a52, b4, c1, d7), (a52, b4, c1, d8), (a52, b4, c1, d9), (a52, b4, c1, d10), (a52, b4, c1, d11), (a52, b4, c1, d12), (a52, b4, c1, d13), (a52, b4, c1, d14), (a52, b4, c1, d15), (a52, b4, c1, d16), (a52, b4, c1, d17), (a52, b4, c1, d18), (a52, b4, c1, d19), (a52, b4, c1, d20), (a52, b4, c1, d21), (a52, b4, c1, d22), (a52, b4, c2, d1), (a52, b4, c2, d2), (a52, b4, c2, d3), (a52, b4, c2, d4), (a52, b4, c2, d5), (a52, b4, c2, d6), (a52, b4, c2, d7), (a52, b4, c2, d8), (a52, b4, c2, d9), (a52, b4, c2, d10), (a52, b4, c2, d11), (a52, b4, c2, d12), (a52, b4, c2, d13), (a52, b4, c2, d14), (a52, b4, c2, d15), (a52, b4, c2, d16), (a52, b4, c2, d17), (a52, b4, c2, d18), (a52, b4, c2, d19), (a52, b4, c2, d20), (a52, b4, c2, d21), (a52, b4, c2, d22), (a52, b4, c3, d1), (a52, b4, c3, d2), (a52, b4, c3, d3), (a52, b4, c3, d4), (a52, b4, c3, d5), (a52, b4, c3, d6), (a52, b4, c3, d7), (a52, b4, c3, d8), (a52, b4, c3, d9), (a52, b4, c3, d10), (a52, b4, c3, d11), (a52, b4, c3, d12), (a52, b4, c3, d13), (a52, b4, c3, d14), (a52, b4, c3, d15), (a52, b4, c3, d16), (a52, b4, c3, d17), (a52, b4, c3, d18), (a52, b4, c3, d19), (a52, b4, c3, d20), (a52, b4, c3, d21), (a52, b4, c3, d22), (a52, b5, c1, d1), (

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d3), (a53, b2, c1, d4), (a53, b2, c1, d5), (a53, b2, c1, d6), (a53, b2, c1, d7),  
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c1, d9), (a53, b5, c1, d10), (a53, b5, c1, d11), (a53, b5, c1, d12), (a53, b5, c1, d13), (a53, b5, c1, d14), (a53, b5, c1, d15), (a53, b5, c1, d16), (a53, b5, c1, d17), (a53, b5, c1, d18), (a 53, b5, c1, d19), (a53, b5, c1, d20), (a53, b5, c1, d21), (a53, b5, c1, d22), (a53, b5, c2, d1), (a53, b5, c2, d2), (a53, b5, c2, d3), (a53, b5, c2, d4), (a53, b5, c2, d5), (a53, b5, c2, d6), (a53, b5, c2, d7), (a53, b5, c2, d8), (a53, b5, c2, d9), (a53, b 5, c2, d10), (a53, b5, c2, d11), (a53, b5, c2, d12), (a53, b5, c2, d13), (a53, b5, c2, d14), (a53, b5, c2, d15), (a53, b5, c2, d16), (a53, b5, c2, d17), (a53, b5, c2, d18), (a53, b5, c2, d19), (a53, b5, c2, d20), (a53, b5, c2, d21), (a53, b5, c2, d22), (a5 3, b5, c3, d1), (a53, b5, c3, d2), (a53, b5, c3, d3), (a53, b5, c3, d4), (a53, b5, c3, d5), ( a53, b5, c3, d6), (a53, b5, c3, d7), (a53, b5, c3, d8), (a53, b5, c3, d9), (a53, b5, c3, d10), (a53, b5, c3, d11), (a53, b5, c3, d12), (a53, b5, c3, d13), (a53, b5, c3, d14), (a53, b5, c3, d15), (a53, b5, c3, d16), (a53, b5, c3, d17), (a53, b5, c3, d18), (a53, b5, c3, d19), (a53, b5, c3, d20), (a53, b5, c3, d21), (a53, b5, c3, d22), (a53, b6, c1, d1), (a53, b6, c1, d2), (a53, b6, c1, d3), (a53, b6, c1, d4), (a53, b6, c1, d5), (a53, b6, c1, d6), (a53, b6, c1, d7), (a53, b6, c1, d8), (a53, b6, c1, d9), (a53, b6, c1, d10), (a53, b6, c1, d11), (a53, b6, c1, d12), (a53, b6, c1, d13), (a53, b6, c1, d14), (a53, b6, c1, d15), (a53, b6, c1, d16), (a53, b6, c1, d17), (a53, b6, c1, d18), (a53, b6, c1, d19), (a53, b6, c1, d20), (a53, b6, c1, d21), (a53, b6, c1, d22), (a53, b6, c2, d1), (a53, b6, c2, d2), (a53, b6, c2, d3), (a53, b6, c2, d4), (a53, b6, c2, d5), (a53, b6, c2, d6), (a53, b6, c2, d7), (a53, b6, c2, d8), (a53, b6, c2, d9), (a53, b6, c2, d10), (a53, b6, c2, d11), (a53, b6, c2, d12), (a53, b6, c2, d13), (a53, b6, c2, d14), (a53, b6, c2, d15), (a53, b6, c2, d16), (a53, b6, c2, d17), (a53, b6, c2, d18), (a53, b6, c2, d19), (a53, b6, c2, d20), (a53, b6, c2, d21), (a53, b6, c2, d22), (a53, b6, c3, d1), (a53, b6, c3, d2), (a53, b6, c3, d3), (a53, b6, c3, d4), (a5 3, b6, c3, d5), (a53, b6, c3, d6), (a53, b6, c3, d7), (a53, b6, c3, d8), (a53, b6, c3, d9), (a53, b6, c3, d10), (a53, b6, c3, d11), (a53, b6, c3, d12), (a53, b6, c3, d13), (a53, b6, c3, d14), (a53, b6, c3, d15), (a53, b6, c3, d16), (a53, b6, c3, d17), (a53, b6, c3, d18), (a53, b6, c3, d19), (a53, b6, c3, d20), (a53, b6, c3, d21), (a53, b6, c3, d22), (a54, b1, c1, d1), (a54, b1, c1, d2), (a54, b1, c1, d3), (a54, b1, c1, d4), (a54, b1, c1, d5), (a54, b1, c1, d6), (a54, b1, c1, d7), (a54, b1, c1, d8), (a54, b1, c1, d9), (a54, b1, c1, d10), (a54, b1, c1, d11), (a54, b1, c1, d12), (a54, b1, c1, d13), (a54, b1, c1, d14), (a54, b1, c1, d15), (a54, b1, c1, d16), (a54, b1, c1, d17), (a54, b1, c1, d18), (a54, b1, c1, d19), (a54, b1, c1, d20), (a54, b1, c1, d21), (a54, b1, c1, d22), (a54, b1, c2, d1), (a54, b1, c2, d2), (a54, b1, c2, d3), (a54, b1, c2, d4), (a54, b1, c2, d5), (a54, b1, c2, d6), (a54, b1, c2, d7), (a54, b1, c2, d8), (a54, b1, c2, d9), (a54, b1, c2, d10), (a54, b1, c2, d11), (a54, b1, c2, d12), (a54, b1, c2, d13), (a54, b1, c2, d14), (a54, b1, c2, d15), (a54, b1, c2, d16), (a54, b1, c2, d17), (a54, b1, c2, d18), (a54, b1, c2, d19), (a54, b1, c2, d20), (a54, b1, c2, d21), (a54, b1, c2, d22), (a54, b2, c1, d1), (a54, b2, c1, d2), (a54, b2, c1, d3), ( a54, b2, c1, d4), (a54, b2, c1, d5), (a54, b2, c1, d6), (a54, b2, c1, d7), (a54, b2, c1, d8), (a54, b2, c1, d9), (a54, b2, c1, d10),

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(a54, b2, c1, d11), (a54, b2, c1, d12), (a54, b2, c1, d13), (a54, b2, c1, d14),  
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(a62, b6, c1, d15), (a62, b6, c1, d16), (a62, b6, c1, d17), (a62, b6, c1, d18),  
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b4, c3, d3), (a75, b4, c3, d4), (a75, b4, c3, d5 ), (a75, b4, c3, d6), (a75, b4, c3, d7), (a75, b4, c3, d8), (a75, b4, c3, d9), (a75, b4, c3, d10), (a75, b4, c3, d11), (a75, b4, c3, d12), (a75, b4, c3, d13), (a75, b4, c3, d14), (a75, b4, c3, d15), (a75, b4, c3, d16), (a75, b4, c3, d17), (a75, b4, c3, d18 ), (a75, b4, c3, d19), (a75, b4, c3, d20), (a75, b4, c3, d21), (a75, b4, c3, d22), (a75, b5, c1, d1), (a75, b5, c1, d2), (a75, b5, c1, d3), (a75, b5, c1, d4), (a75, b5, c1, d5), (a75, b5, c1, d6), (a75, b5, c1, d7), (a75, b5, c1, d8), (a75, b5, c1, d9), (a75, b5, c1, d10), (a75, b5, c1, d11), (a75, b5, c1, d12), (a75, b5, c1, d13), (a75, b5, c1, d14), (a75, b5, c1, d15), (a75, b5, c1, d16), (a75, b5, c1, d17), (a75, b5, c1, d18), (a75, b5, c1, d19), (a75, b5, c1, d20), (a75, b5, c1, d21), (a75, b5, c1, d22) , (a75, b5, c2, d1), (a75, b5, c2, d2), (a75, b5, c2, d3), (a75, b5, c2, d4), (a75, b5, c2, d5), (a75, b5, c2, d6), (a75, b5, c2, d7), (a75, b5, c2, d8), (a75, b5, c2, d9), (a75, b5, c2, d10), (a75, b5, c2, d11), (a75, b5, c2, d12), (a75, b5, c2, d13), (a75, b5, c2, d14), (a75, b5, c2, d15), (a75, b5, c2, d16), (a75, b5, c2, d17), (a75, b5, c2, d18), (a75, b5, c2, d19), (a75, b5, c2, d20), (a75, b5, c2, d21), (a75, b5, c2, d22), (a75, b5, c3, d1), (a75, b5, c3, d2), (a75, b5, c3, d3), (a75, b5, c3, d4), (a75, b5, c3, d5), (a75, b5, c3, d6), (a75, b5, c3, d7), (a75, b5, c3, d8), (a75, b5, c3, d9), (a75, b5, c3, d10), (a75, b5, c3, d11), (a75, b5, c3, d12), (a75, b5, c3, d13), (a75, b5, c3, d14), (a75, b5, c3, d15), (a75, b5, c3, d16), (a75, b5, c3, d17), (a75, b5, c3, d18), (a75, b5, c3, d19), (a75, b5, c3, d20), (a75, b5, c3, d21), (a75, b5, c3, d22), (a75, b6, c1, d1), (a75, b6, c1, d2), (a75, b6, c1, d3), (a75, b6, c1, d4), (a75, b6, c1, d5), (a75, b6, c1, d6), (a75, b6, c1, d7), (a75, b6, c1, d8), (a75, b6, c1, d9), (a75, b6, c1, d10), (a75, b6, c1, d11), (a75, b6, c1, d12), (a75, b6, c1, d13), (a75, b6, c1, d14), (a75, b6, c1, d15), (a75, b6, c1, d16), (a75, b6, c1, d17), (a75, b6, c1, d18), (a75, b6, c1, d19), (a75, b6, c1, d20), (a75, b6, c1, d21), (a75, b6, c1, d22), (a75, b6, c2, d1), (a75, b6, c2, d2), (a75, b6, c2, d3), (a75, b6, c2, d4), (a75, b6, c2, d5), (a75, b6, c2, d6), (a75, b6, c2, d7), (a75, b6, c2, d8), (a75, b6, c2, d9), (a75, b6, c2, d10), (a75, b6, c2, d11), (a75, b6, c2, d12), (a75, b6, c2, d13), (a75, b6, c2, d14), (a75, b6, c2, d15), (a75, b6, c2, d16), (a75, b6, c2, d17), (a75, b6, c2, d18), (a75, b6, c2, d19), (a75, b6, c2, d20), (a75, b6, c2, d21), (a75, b6, c2, d22), (a75, b6, c3, d1), (a75, b6, c3, d2), (a75, b6, c3, d3), (a75, b6, c3, d4), (a75, b6, c3, d5), (a75, b6, c3, d6), (a75, b6, c3, d7), (a75, b6, c3, d8), (a75, b6, c3, d9), (a75, b6, c3, d10), (a75, b6, c3, d11), (a75, b6, c3, d12), (a75, b6, c3, d13), (a75, b6, c3, d14), (a75, b6, c3, d15), (a75, b6, c3, d16), (a75, b6, c3, d17), (a75, b6, c3, d18), (a75, b6, c3, d19), (a75, b6, c3, d20), (a75, b6, c3, d21), (a75, b6, c3, d22), (a76, b1, c1, d1), (a76, b1, c1, d2), (a76, b1, c1, d3), (a76, b1, c1, d4), (a76, b1, c1, d5), (a76, b1, c1, d6), (a76, b1, c1, d7), (a76, b1, c1, d8), (a76, b1, c1, d9), (a76, b1, c1, d10), (a76, b1, c1, d11), (a76, b1, c1, d12), (a76, b1, c1, d13), (a76, b1, c1, d14), (a76, b1, c1, d15), (a76, b1, c1, d16), (a76, b1, c1, d17), (a76, b1, c1, d18), (a76, b1, c1, d19), (a76, b1, c1, d20), (a76, b1, c1, d21), (a76, b1, c1, d22), (a76, b1, c2, d1), (a76, b1, c2, d2), (a76, b1, c2, d3), (a76, b1, c2, d4), (a76, b1, c2, d5), (a76, b1, c2, d6), (a76, b1, c2, d7), (a76, b1, c2, d8), (a76, b1, c2, d9), (a76, b1, c2, d10), (a76, b1, c2, d11), (a76, b1, c2, d12), (a76, b1, c2, d13), (a76, b1, c2, d14), (a76, b1, c2, d15), (a76, b1, c2, d16), (a76, b1, c2, d17), (a76, b1, c2, d18), (a76, b1, c2, d19), (a76, b1, c2, d20), (a76, b1, c2, d21), (a76, b1, c2, d22), (a76, b1, c3, d1), (a76, b1, c3, d2), (a76, b1, c3, d3), (a76, b1, c3, d4), (a76, b1, c3, d5), (a76, b1, c3, d6), (a76, b1, c3, d7), (a76, b1, c3, d8),

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(a76, b1, c3, d9), (a76, b1, c3, d10), (a76, b1, c3, d11), (a76, b1, c3, d12),  
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2, c1, d15), (a76, b2, c1, d16), (a76, b2, c1, d17), (a76, b2, c1, d18), (a76, b  
2, c1, d19), (a76, b2, c1, d20), (a76, b2, c1, d21), (a76, b2, c1, d22), (a76,  
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d9), (a76, b2, c2, d10), (a76, b2, c2, d11), (a76 , b2, c2, d12), (a76, b2, c2,  
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), (a76, b3, c2, d13), (a76, b3, c2, d14), (a76, b3, c2, d15), (a76, b3, c2, d16  
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), (a76, b3, c2, d21), (a76, b3, c2, d22), (a76, b3, c3, d1), (a76, b3 , c3, d2)  
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b4, c3, d6), (a76, b4, c3, d7), (a76, b4, c3, d8), (a76, b4, c3, d9), (a76, b4, 50

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, c3, d21), (a77, b4, c3, d22), (a77, b5, c1, d1) , (a77, b5, c1, d2), (a77, b5, c1, d3), (a77, b5, c1, d4), (a77, b5, c1, d5), (a77, b5, c1, d6), (a77, b5, c1, d7), (a77, b5, c1, d8), (a77, b5, c1, d9), (a77, b5, c1, d10), (a77, b5, c1, d11), (a77, b5, c1, d12), (a77, b5, c1, d13), (a77, b5, c1, d14), (a 77, b5, c1, d15), (a77, b5, c1, d16), (a77, b5, c1, d17), (a77, b5, c1, d18), (a77, b5, c1, d19), (a77, b5, c1, d20), (a77, b5, c1, d21), (a77, b5, c1, d22), (a77, b5, c2, d1), (a77, b5, c2, d2), (a77, b5, c2, d3), (a77, b5, c2, d4), (a77, b5, c2, d5), (a7 7, b5, c2, d6), (a77, b5, c2, d7), (a77, b5, c2, d8), (a77, b5, c2, d9), (a7 7, b5, c2, d10), (a77, b5, c2, d11), (a77, b5, c2, d12), (a77, b5, c2, d13), (a7 7, b5, c2, d14), (a77, b5, c2, d15), (a77, b5, c2, d16), (a77, b5, c2, d17), (a7 7, b5, c2, d18), (a7 7, b5, c2, d19), (a77, b5, c2, d20), (a77, b5, c2, d21), (a 77, b5, c2, d22), (a77, b5, c3, d1), (a77, b5, c3, d2), (a77, b5, c3, d3), (a77,

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b5, c3, d4), (a77, b5, c3, d5), (a77, b5, c3, d6), (a77, b5, c3, d7), (a77, b5, c3, d8), (a77, b5, c3, d9), (a77, b5 , c3, d10), (a77, b5, c3, d11), (a77, b5, c3, d12), (a77, b5, c3, d13), (a77, b5, c3, d14), (a77, b5, c3, d15), (a77, b5, c3, d16), (a77, b5, c3, d17), (a77, b5, c3, d18), (a77, b5, c3, d19), (a77, b5, c3, d20), (a77, b5, c3, d21), (a77, b5, c3, d22), (a77 , b6, c1, d1), (a77, b6, c1, d2), (a77, b6, c1, d3), (a77, b6, c1, d4), (a77, b6, c1, d5), (a77, b6, c1, d6), (a77, b6, c1, d7), (a77, b6, c1, d8), (a77, b6, c1, d9), (a77, b6, c1, d10),

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, c3, d3), (a77, b6, c3, d4), (a77, b6, c3, d5), (a77, b6, c3, d6), (a77, b6, c3 , d7), (a77, b6, c3, d8), (a77, b6, c3, d9), (a77, b6, c3, d10), (a77, b6, c3, d11), (a77, b6, c3, d12), (a77, b6, c3, d13), (a77, b6, c3, d14), (a77, b6, c3, d15), (a77, b6, c3, d16), (a77, b6, c3, d17), (a77, b6, c3, d18), (a77, b6, c3, d19), (a77, b6, c3, d20), (a77, b6, c3, d21), (a77, b6, c 3, d22), (a78, b1, c1, d1), (a78, b1, c1, d2), (a78, b1, c1, d3), (a78, b1, c1, d4), (a78, b1, c1, d5),

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b1, c2, d 4), (a78, b1, c2, d5), (a78, b1, c2, d6), (a78, b1, c2, d7), (a78, b1 , c2, d8), (a78, b1, c2, d9), (a78, b1, c2, d10), (a78, b1, c2, d11), (a78, b1, c2, d12), (a78, b1, c2, d13), (a78, b1, c2, d14), (a78, b1, c2, d15), (a78, b1, c2, d16), (a78, b1, c2, d17 ), (a78, b1, c2, d18), (a78, b1, c2, d19), (a78, b1, c2, d20), (a78, b1, c2, d21), (a78, b1, c2, d22), (a78, b1, c3, d1), (a78, b1, c3, d2), (a78, b1, c3, d3), (a78, b1, c3, d4), (a78, b1, c3, d5), (a78, b1, c3, d6), (a78, b1, c3, d7), (a78, b1, c3, d8), ( a78, b1, c3, d9), (a78, b1, c3, d10

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 , c2, d14), (a83, b2, c2, d15), (a83, b2, c2, d16), (a83, b2, c2, d17), (a83, b2

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b6, c2, d4), (a89, b6, c2, d5), (a89, b6, c2, d6), (a89, b6, c2, d7), (a89, b6, c2, d8), (a89, b6, c2, d9), (a 89, b6, c2, d10), (a89, b6, c2, d11), (a89, b6, c2, d12), (a89, b6, c2, d13), (a89, b6, c2, d14), (a89, b6, c2, d15), (a89, b6, c2, d16), (a89, b6, c2, d17), (a89, b6, c2, d18), (a89, b6, c2, d19), (a89, b6, c2, d20), (a89, b6, c2, d21), (a89, b6, c2, d22) , (a89, b6, c3, d1), (a89, b6, c3, d2), (a89, b6, c3, d3), (a89, b6, c3, d4), (a89, b6, c3, d5), (a89, b6, c3, d6), (a89, b6, c3, d7), (a89, b6, c3, d8), (a89, b6, c3, d9), (a89, b6, c3, d10), (a89, b6, c3, d11), (a89, b6, c3, d12), (a89, b6, c3, d13), (a8 9, b6, c3, d14) , (a89, b6, c3, d15), (a89, b6, c3, d16), (a89, b6, c3, d17), (a89, b6, c3, d18) , (a89, b6, c3, d19), (a89, b6, c3, d20), (a89, b6, c3, d21), (a89, b6, c3, d22) 10 , (a90, b1, c1, d1), (a90, b1, c1, d2), (a90, b1, c1, d3), (a90, b1, c1, d4), (a 9 0, b1, c1, d5), (a90, b1, c1, d6), (a90, b1, c1, d7), (a90, b1, c1, d8), (a90, b1, c1, d9), (a90, b1, c1, d10), (a90, b1, c1, d11), (a90, b1, c1, d12), (a90, b1, c1, d13), (a90, b1, c1, d14), (a90, b1, c1, d15), (a90, b1, c1, d16), (a90, b1, c1, d17), (a90 , b1, c1, d18), (a90, b1, c1, d19), (a90, b1, c1, d20), (a90, b1, c1, d21), (a90, b1, c1, d22), (a90, b1, c2, d1), (a90, b1, c2, d2), (a90, b1, c2, d3), (a90, b1, c2, d4), (a90, b1, c2, d5), (a90, b1, c2, d6), (a90, b1, c2, d7), (a90, b1, c2, d8), (a90, b1 , c2, d9), (a90, b1, c2, d10), (a90, b1, c2, d11), (a90, b1, c2, d12), (a90, b1, c2, d13), (a90, b1, c2, d14), (a90, b1, c2, d15), (a90, b1, c2, d16), (a90, b1, c2, d17), (a90, b1, c2, d18), (a90, b1, c2, 20 d19), (a90, b1, c2, d20), (a90, b1, c2, d21), (a90, b1, c2, d22), (a90, b1, c3, d1), (a90, b1, c3, d2), (a90, b1, c3, d3), (a90, b1, c3, d4), (a90, b1, c3, d5 ) , (a90, b1, c3, d6), (a90, b1, c3, d7), (a90, b1, c3, d8), (a90, b1, c3, d9), ( a90, b1, c3, d10), (a90, b1, c3, d11), (a90, b1, c3, d12), (a90, b1, c3, d13), ( a90, b1, c3, d14), (a90, b1, c3, d15), (a90, b1, c3, d16), (a90, b1, c3, d17), ( a90, b1, c3, d18), (a90, b1, c3, d19), (a90, b1, c3, d20), (a90, b1, c3, d21), ( a90, b1, c3, d22), (a90, b2, c1, d1), (a90, b2, c1, d2), (a90, b2, c1, d3), (a90 , b2, c1, d4), (a90, b2, c1, d5), (a90, b2, c1, d6), (a90, b2, c1, d7), (a90, b 2, c1, d8), (a90, b2, c1, d9), (a90, b2, c1, d10), (a90, b2, c1, d11), (a90, b2, c1, d12), (a90, b2, c1, d13), (a90, b2, c1, d14), (a90, b2, c1, d15), (a90, b2, 30 c1, d16), (a90, b2, c 1, d17), (a90, b2, c1, d18), (a90, b2, c1, d19), (a90, b2 , c1, d20), (a90, b2, c1, d21), (a90, b2, c1, d22), (a90, b2, c2, d1), (a90, b2, c2, d2), (a90, b2, c2, d3), (a90, b2, c2, d4), (a90, b2, c2, d5), (a90, b2, c2, d6), (a90, b2, c2, d7), (a90, b2, c2, d8), (a90, b2, c2, d9), (a90, b2, c2, d10 ) , (a90, b2, c2, d11), (a90, b2, c2, d12), (a90, b2, c2, d13), (a90, b2, c2, d14 ) , (a90, b2, c2, d15), (a90, b2, c2, d16), (a90, b2, c2, d17), (a90, b2, c2, d18 ) , (a90, b2, c2, d19), (a90, b2, c2, d20), (a90, b2, c2 , d21), (a90, b2, c2, d2 2), (a90, b2, c3, d1), (a90, b2, c3, d2), (a90, b2, c3, d3), (a90, b2, c3, d4) , (a90, b2, c3, d5), (a90, b2, c3, d6), (a90, b2, c3, d7), (a90, b2, c3, d8), (a90 , b2, c3, d9), (a90, b2, c3, d10), (a90, b2, c3, d11), (a90, b2, c3, d12 ) , (a90 , b2, c3, d13), (a90, b2, c3, d14), (a90, b2, c3, d15), (a90, b2, c3, d16), (a90 , b2, c3, d17), (a90, b2, c3, d18), (a90, b2, c3, d19), (a90, b2, c3, d20), (a90 , b2, c3, d21), (a90, b2, c3, d22), (a90, b3, c1, d1), (a90, b3, c1, d2), (a90, b3, c1, d 3), (a90, b3, c1, d4), (a90, b3, c1, d5), (a90, b3, c1, d6), (a90, b3, c1, d7), (a90, b3, c1, d8), (a90, b3, c1, d9), (a90, b3, c1, d10), (a90, b3, c1 , d11), (a90, b3, c1, d12), (a90, b3, c1, d13), (a90, b3, c1, d14), (a90, b3, c1 , d15), (a90, b3, c1, d16) , (a90, b3, c1, d17), (a90, b3, c1, d18), (a90, b3, c 1, d19), (a90, b3, c1, d20), (a90, b3, c1, d21), (a90, b3, c1, d22), (a90, b3, c 2, d1), (a90, b3, c2, d2), (a90, b3, c2, d3), (a90, b3, c2, d4), (a90, b3, c2, d 5), (a90, b3, c2, d6), (a90, b3, c2, d7), ( a90, b3, c2, d8), (a90, b3, c2, d9), 40 50



(a90, b3, c2, d10), (a90, b3, c2, d11), (a90, b3, c2, d12), (a90, b3, c2, d13),  
(a90, b3, c2, d14), (a90, b3, c2, d15), (a90, b3, c2, d16), (a90, b3, c2, d17),  
(a90, b3, c2, d18), (a90, b3, c2, d19), (a90, b3, c2, d20), (a90, b3, c2, d21)  
, (a90, b3, c2, d22), (a90, b3, c3, d1), (a90, b3, c3, d2), (a90, b3, c3, d3), (  
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b3, c3, d8), (a90, b3, c3, d9), (a90, b3, c3, d10), (a90, b3, c3, d11), (a90 ,  
b3, c3, d12), (a90, b3, c3, d13), (a90, b3, c3, d14), (a90, b3, c3, d15), (a90,  
b3, c3, d16), (a90, b3, c3, d17), (a90, b3, c3, d18), (a90, b3, c3, d19), (a90,  
b3, c3, d20), (a90, b3, c3, d21), (a90, b3, c3, d22), (a90, b4, c1, d1), (a90, b  
4, c1, d2), (a 90, b4, c1, d3), (a90, b4, c1, d4), (a90, b4, c1, d5), (a90, b4, 10  
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d10), (a90, b4, c1, d11), (a90, b4, c1, d12), (a90, b4, c1, d13), (a90, b4, c1,  
d14), (a90, b4, c1, d15), (a90, b4, c1, d16), (a90, b4, c1, d17), (a90, b4, c1,  
d18), (a90, b4, c1, d19), (a90, b4, c1, d20), (a90, b4, c1, d21), (a90, b4, c1,  
d22), (a90, b4, c2, d1), (a90, b4, c2, d2), (a90, b4, c2, d3), (a90, b4, c2, d4  
), (a90, b4, c2, d5), (a90, b4, c2, d6), (a90, b 4, c2, d7), (a90, b4, c2, d8),  
(a90, b4, c2, d9), (a90, b4, c2, d10), (a90, b4, c2, d11), (a90, b4, c2, d12), (  
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a90, b4, c2, d17), (a90, b4, c2, d18), (a90, b4, c2, d19), (a90, b4, c2, d20), (  
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c 3, d11), (a90, b4, c3, d12), (a90, b4, c3, d13), (a90, b4, c3, d14), (a90, b4,  
c3, d15), (a90, b4, c3, d16), (a90, b4, c3, d17), (a90, b4, c3, d18), (a90, b4,  
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), (a90, b5, c1, d10), (a90, b5, c1, d11), (a90, b5, c1, d12), (a90, b5, c1, d13  
), (a90, b5, c1, d14), (a90, b5, c1 , d15), (a90, b5, c1, d16), (a90, b5, c1, d1  
7), (a90, b5, c1, d18), (a90, b5, c1, d19), (a90, b5, c1, d20), (a90, b5, c1, d2  
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, b5, c2, d12), (a90, b5, c2, d13), (a90, b5, c2, d14), (a90, b5, c2, d15), (a90  
, b5, c2, d16), (a90, b5, c2, d17), (a90, b5, c2, d18), (a90, b5, c2, d19), (a9  
0, b5, c2, d20), (a90, b5, c2, d21), (a90, b5, c2, d22), (a90, b5, c3, d1), (a90  
, b5, c3, d2), (a90, b5, c3, d3), (a90, b5, c3, d4), (a90, b5, c3, d5), (a90, b5  
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3, d14), (a90, b5, c3, d15), (a90, b5, c3, d16), (a90, b5, c3, d17), (a90, b5, c  
3, d18), (a90, b5, c3, d19), (a90, b5, c3, d20), (a90, b5, c3, d21), (a90, b5, c  
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d4), (a90, b6, c1, d5), (a90, b6, c1, d6), (a90, b6, c1, d7), (a90, b6, c1, d8),  
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(a90, b6, c1, d13), (a90, b6, c1, d14), (a90, b6, c1, d15), (a90, b6, c1, d16),  
(a90, b6, c1, d17), (a90, b6, c1, d18), (a90, b6, c1, d19), (a90, b6, c1, d20),  
(a90, b6, c1, d21), (a90, b6, c1, d22), (a90, b6, c2, d1), (a90, b6, c2, d2), (  
a90, b6, c2, d3), (a90, b6, c2, d4), (a90, b6, c2, d5), (a90, b6, c2, d6), (a90,  
b6, c2, d7), (a90, b6, c2, d8), (a90, b6, c2, d9), (a90, b6, c2, d10), (a90, b6  
, c2, d11), (a90, b6, c2, d12), (a90, b6, c2, d13), (a90, b6, c2, d14), (a90, b6

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, c2, d15), (a90, b6, c2, d16), (a90, b6, c2, d17), (a90, b6, c2, d18), (a90, b6, c2, d19), (a90, b6, c2, d20), (a90, b6, c2, d21), (a90, b6, c2, d22), (a90, b6, c3, d1), (a90, b6, c3, d2), (a90, b6, c3, d3), (a90, b6, c3, d4), (a90, b6, c3, d5), (a90, b6, c3, d6), (a90, b6, c3, d7), (a90, b6, c3, d8), (a90, b6, c3, d9), (a90, b6, c3, d10), (a90, b6, c3, d11), (a90, b6, c3, d12), (a90, b6, c3, d13), (a90, b6, c3, d14), (a90, b6, c3, d15), (a90, b6, c3, d16), (a90, b6, c3, d17), (a90, b6, c3, d18), (a90, b6, c3, d19), (a90, b6, c3, d20), (a90, b6, c3, d21), (a90, b6, c3, d22)

一般式(A)で表される化合物においては、その光学異性体およびラセミ体も包含する。

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#### 【0140】

試験例1 EGFR及びHER2に対する被験化合物の酵素阻害活性

昆虫細胞発現系から精製されたEGFR、HER2および、ビオチン化基質(グルタミン酸:チロシン=4:1の合成ペプチド)を、ATPが添加された384穴プレートに加え、室温にて酵素反応を行なった。酵素反応終了後に、ユーロピウム標識した抗リン酸化チロシン抗体とアルフィコシアニン標識したストレプトアビジンを添加し、リン酸化チロシン残基を時間分解蛍光法で定量した。

(被験化合物の調製)

被験化合物はDMSOを用いて10mMに調製した。

さらに10%DMSO溶液を用いて1mM溶液を調製し、これを最高濃度ポイントとした。

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希釈系列は4倍希釈系列を10段階まで作製した。

(試薬調製)

バッファの調製:

バッファは、10mM MgCl<sub>2</sub>, 5mM MnCl<sub>2</sub>, 0.2mM Na<sub>3</sub>VO<sub>4</sub>, 0.1%BSAを含むTBSを用いた。

酵素:

EGFR、HER2酵素は、昆虫細胞発現系から精製したものをカルナバイオサイセンス社より購入した。

EGFR酵素溶液は、バッファを用いて250ng/mlに調製した。

HER2キナーゼは、バッファを用いて1000ng/mlに調製した。

基質溶液:

基質には、ビオチン化poly-Glu-Tyr(4:1, CISbio社製)を用いた。濃度は、上記バッファを用いて3.125 μg/mlとした。

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ATP溶液:

EGFR酵素の検討でのATP濃度は、上記バッファを用いて125 μMに調製した。

HER2酵素の検討でのATP濃度は、290 μMに調製した。

検出試薬:

検出試薬の調製には、30mM EDTA, 0.8M KF, 0.1% BSAを含むTBSを用いた。

ユーロピウム標識した抗リン酸化チロシン抗体、PT-66K(CISbio社)は195ng/mlにアルフィコシアニン標識したストレプトアビジン、SA-XL(CISbio社)は5 μg/mlに調製した。

(アッセイ方法)

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アッセイプレートには、384穴プレートを用いた。1ウェルあたり被験化合物を1.1 μl、基質溶液4 μl、酵素溶液4 μl添加した後、ATP溶液2 μlを加え酵素反応を開始した。酵素反応は室温にて60分を行い、検出試薬を10 μl添加して反応を終了した。反応終了の1時間後に時間分解蛍光測定を行なった。

(評価方法)

被験化合物の代わりに10%DMSOを用い、ATP添加時のシグナルを0%阻害(酵素活性100%)、ATP非存在下のシグナルを100%阻害(酵素活性0%)として、被験化合物添加時の阻害活性を算出した。

実施例に記載のEGFR及びHER2に対する阻害活性(IC<sub>50</sub>値、nM)を以下に示す。

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【 0 1 4 1 】

【表 5 0】

化合物 No.	EGFR(nM)	HER2(nM)	化合物 No.	EGFR(nM)	HER2(nM)
V-6	6.5	3.6	VIII-74	164	150
V-7	36	76	VIII-75	50	34
V-43	186	151	VIII-76	162	216
VIII-7	103	348	VIII-77	217	246
VIII-10	409	484	VIII-78	111	410
VIII-11	43	45	VIII-80	23	29
VIII-12	209	227	VIII-81	9.8	17.7
VIII-13	300	888	VIII-82	7.6	7.5
VIII-19	89	115	VIII-83	164	121
VIII-20	29	31	VIII-84	44	47
VIII-27	33	43	VIII-85	32	33
VIII-28	162	167	VIII-86	244	304
VIII-29	102	513	VIII-87	642	476
VIII-32	50	46	VIII-88	47	56
VIII-35	11	25	VIII-89	27	24
VIII-40	9.3	19.2	VIII-90	93	97
VIII-41	12	15	VIII-91	74	83
VIII-42	10	11	VIII-92	32	36
VIII-44	26	6.3	VIII-93	453	321
VIII-45	58	152	VIII-94	213	150
VIII-46	82	104	VIII-95	209	176
VIII-47	12	23	VIII-96	20	20
VIII-48	44	15	VIII-97	19	22
VIII-49	28	31	VIII-98	21	21
VIII-50	19	15	VIII-99	84	84
VIII-51	45	64	VIII-100	208	203
VIII-52	19	28	VIII-101	56	129
VIII-53	79	80	VIII-102	72	121
VIII-55	19	68	VIII-103	3.9	5.6
VIII-56	7.9	13.8	VIII-104	238	202
VIII-57	301	303	VIII-105	14	14
VIII-58	207	236	VIII-106	160	148
VIII-59	115	278	VIII-107	150	135
VIII-60	54	30	VIII-108	53	56
VIII-61	30	24	VIII-109	23	18
VIII-62	22	22	VIII-110	29	35
VIII-63	9.6	9.2	VIII-111	37	55
VIII-64	52	88	VIII-112	11	13
VIII-65	97	98	VIII-113	9.7	12
VIII-66	27	43	VIII-114	57	69
VIII-67	16	14	VIII-116	37	25
VIII-68	19	25	VIII-117	47	53
VIII-69	43	28	VIII-118	197	223
VIII-70	63	31	VIII-119	81	77
VIII-71	130	199	VIII-120	33	29
VIII-72	6.3	11	VIII-121	17	18
VIII-73	275	199	VIII-122	29	24

【 0 1 4 2 】

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【表 5 1】

化合物 No.	EGFR(nM)	HER2(nM)	化合物 No.	EGFR(nM)	HER2(nM)
VIII-123	24	38	VIII-174	25	28
VIII-124	40	38	VIII-176	23	26
VIII-125	118	103	VIII-177	7.3	5.8
VIII-126	101	123	VIII-178	8.1	8.2
VIII-127	37	35	VIII-179	4.6	5
VIII-128	15	17	VIII-180	7.8	6
VIII-129	7.1	6.3	VIII-181	6.2	5.9
VIII-130	8.1	5.2	VIII-182	4.7	7.3
VIII-131	5.8	4.4	VIII-183	7.1	8.3
VIII-132	14	14	VIII-184	10	7
VIII-133	8.4	7.1	VIII-185	6.8	12.5
VIII-134	7.1	7.4	VIII-186	19	26
VIII-135	7.6	8.1	VIII-187	10	9
VIII-136	21	21	VIII-188	20	30
VIII-137	13	14	VIII-189	17	27
VIII-139	5	3.8	VIII-190	8.9	11.1
VIII-140	5.5	4.3	VIII-191	17	42
VIII-141	10	7.9	VIII-192	6.2	14.2
VIII-142	5.5	4.4	VIII-193	14	15
VIII-143	3.5	3.6	VIII-194	8.6	13
VIII-145	9.4	8.9	VIII-195	45	43
VIII-146	12	10	VIII-196	11	20
VIII-147	13	13	VIII-197	9	21
VIII-148	6.1	5.8	VIII-198	3.6	4.6
VIII-149	13	14	VIII-199	6.3	5.2
VIII-150	12	11	VIII-200	9.7	9.6
VIII-151	20	19	VIII-205	17	13
VIII-152	2.4	4.3	VIII-207	21	17
VIII-153	10	11	VIII-208	28	23
VIII-154	9.5	9	VIII-209	38	30
VIII-155	10	8	VIII-210	42	14
VIII-156	7	6.4	VIII-211	23	18
VIII-157	4.1	4	VIII-213	18	25
VIII-158	3.8	3.8	VIII-214	4.5	3.3
VIII-159	16	19	VIII-216	22	7
VIII-160	17	18	VIII-218	76	22
VIII-161	14	15	VIII-219	26	20
VIII-162	51	62	VIII-220	263	449
VIII-163	23	27	VIII-221	143	168
VIII-164	7.3	7.6	VIII-223	253	217
VIII-165	11	11	VIII-224	122	118
VIII-167	4.8	4.2	VIII-225	47	70
VIII-168	8	9	VIII-226	20	10
VIII-169	13	15	IX-1	138	203
VIII-170	5	5.6	IX-4	193	334
VIII-171	9.9	11.7	IX-6	344	326
VIII-172	14	16	IX-8	426	549
VIII-173	4.8	3.9			

【 0 1 4 3 】

試験例 2 被験化合物のin vitro細胞増殖抑制試験

ヒト由来がん細胞株として、BT- 474 を用いた。細胞はすべてDulbecco ' s Modified Ess

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ential Medium (DMEM, ナカライテスク) に10%牛胎児血清 (Fetal Bovine Serum, ICN) と50 units/mlペニシリン / 50  $\mu$ g/mlストレプトマイシン溶液 (GIBCO) を添加した培地で37 $^{\circ}$ C、5%CO<sub>2</sub>インキュベーター内にて培養した。96穴プレート (住友ベークライト) に細胞を3000個 / 穴ずつ播種し、翌日薬剤を最終濃度 $3 \times 10^{-10}$  -  $10^{-5}$  Mとなるように添加した。3日間培養した後WS T(water-soluble tetrazolium salt)アッセイを行い、IC<sub>50</sub>値を算出した。

実施例に記載のBT-474に対する増殖阻害活性(IC<sub>50</sub>値、nM)を以下に示す。

【 0 1 4 4 】

【表 5 2】

化合物 No.	BT-474(nM)	化合物 No.	BT-474(nM)	化合物 No.	BT-474(nM)
V-6	110	VIII-75	22	VIII-124	14
V-7	134	VIII-76	12	VIII-125	10
V-43	71	VIII-77	11	VIII-126	9
VIII-7	63	VIII-78	35	VIII-127	61
VIII-10	27	VIII-80	35	VIII-128	37
VIII-11	26	VIII-81	8	VIII-129	12
VIII-12	69	VIII-82	16	VIII-130	50
VIII-13	64	VIII-83	13	VIII-131	14
VIII-20	37	VIII-84	28	VIII-132	18
VIII-27	13	VIII-85	11	VIII-133	43
VIII-28	72	VIII-86	13	VIII-134	14
VIII-29	77	VIII-87	6	VIII-135	28
VIII-32	32	VIII-88	12	VIII-136	53
VIII-35	24	VIII-89	25	VIII-137	78
VIII-40	9	VIII-90	9	VIII-139	32
VIII-41	14	VIII-91	24	VIII-140	30
VIII-42	24	VIII-92	29	VIII-141	46
VIII-44	28	VIII-93	38	VIII-142	31
VIII-45	13	VIII-94	26	VIII-143	24
VIII-46	12	VIII-95	25	VIII-145	21
VIII-47	4	VIII-96	30	VIII-146	45
VIII-48	28	VIII-97	11	VIII-147	25
VIII-49	28	VIII-98	65	VIII-148	26
VIII-50	27	VIII-99	24	VIII-149	28
VIII-51	9	VIII-100	16	VIII-150	30
VIII-52	9	VIII-101	12	VIII-151	47
VIII-53	12	VIII-102	7	VIII-152	22
VIII-55	25	VIII-103	11	VIII-153	21
VIII-56	14	VIII-104	60	VIII-154	31
VIII-57	15	VIII-105	10	VIII-155	28
VIII-58	37	VIII-106	16	VIII-156	24
VIII-59	11	VIII-107	23	VIII-157	25
VIII-60	14	VIII-108	31	VIII-158	27
VIII-61	11	VIII-109	40	VIII-159	15
VIII-62	12	VIII-110	21	VIII-160	47
VIII-63	13	VIII-111	15	VIII-161	49
VIII-64	22	VIII-112	8	VIII-162	42
VIII-65	30	VIII-113	13	VIII-163	38
VIII-66	8	VIII-114	37	VIII-164	31
VIII-67	32	VIII-116	50	VIII-165	23
VIII-68	41	VIII-117	20	VIII-167	21
VIII-69	16	VIII-118	22	VIII-168	44
VIII-70	7	VIII-119	31	VIII-169	35
VIII-71	17	VIII-120	32	VIII-170	19
VIII-72	21	VIII-121	10	VIII-171	52
VIII-73	12	VIII-122	14	VIII-172	70
VIII-74	28	VIII-123	31	VIII-173	40

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【表 5 3】

化合物 No.	BT-474(nM)	化合物 No.	BT-474(nM)	化合物 No.	BT-474(nM)
VIII-174	14	VIII-190	14	VIII-209	19
VIII-176	26	VIII-191	33	VIII-210	31
VIII-177	79	VIII-192	24	VIII-211	16
VIII-178	36	VIII-193	25	VIII-213	33
VIII-179	48	VIII-194	25	VIII-214	25
VIII-180	5	VIII-195	29	VIII-216	32
VIII-181	13	VIII-196	28	VIII-218	41
VIII-182	20	VIII-197	14	VIII-219	45
VIII-183	10	VIII-198	10	VIII-220	23
VIII-184	9	VIII-199	33	VIII-221	15
VIII-185	12	VIII-200	8	VIII-223	6
VIII-186	29	VIII-205	31	VIII-224	1.2
VIII-187	24	VIII-207	68	VIII-225	7
VIII-188	40	VIII-208	27	VIII-226	5
VIII-189	25				

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## 【 0 1 4 6 】

試験例 3 被験化合物の *in vivo* 抗腫瘍評価試験

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NCI-N87細胞（ヒト胃癌細胞株）を雌性ヌードマウス(BALB/cA Jcl-nu/nu, 日本クレア)の乳腺部脂肪組織（mammary fat pad）に $5 \times 10^6$ 細胞/マウスで移植した(day 0)。Day 5に腫瘍の長径と短径をノギスで測定し、群間の腫瘍サイズが均等になるようマウスを群分けした(N=6)。0.5%メチルセルロース(MC)溶液に懸濁した被験化合物(20, 80 mg/kg)をday 5より15日間連日経口投与し、day 20における化合物投与群(T)とcontrol(C; 0.5%MC)群の腫瘍サイズの平均値の比(T/C%)によって抗腫瘍効果を表した。

化合物VIII-11の*in vivo*抗腫瘍評価の結果を表54に、化合物VII I-66の*in vivo*抗腫瘍評価の結果を表55に示す。

【表 5 4】

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腫瘍サイズ平均値(mm <sup>3</sup> )	Control 群	20 mg/kg 投与群	80 mg/kg 投与群
day 5	192	197	197
day 20	655	356	193
T/C%	100%	54%	29%

【表 5 5】

腫瘍サイズ平均値(mm <sup>3</sup> )	Control 群	20 mg/kg 投与群	80 mg/kg 投与群
day 5	150	134	150
day 20	483	154	17
T/C%	100%	32%	4%

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製剤例

## 【 0 1 4 7 】

製剤例 1

以下の成分を含有する顆粒剤を製造する。

成分	式(I)で表わされる化合物	10 mg
	乳糖	700 mg
	コーンスターチ	274 mg
	HPC-L	16 mg

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1000 mg

式(I)で表わされる化合物と乳糖を60メッシュのふるいに通す。コーンスターチを120メッシュのふるいに通す。これらをV型混合機にて混合する。混合末にHPC-L(低粘度ヒドロキシプロピルセルロース)水溶液を添加し、練合、造粒(押し出し造粒孔径0.5~1mm)したのち、乾燥する。得られた乾燥顆粒を振動ふるい(12/60メッシュ)で櫛過し顆粒剤を得る。

## 製剤例2

以下の成分を含有するカプセル充填用散剤を製造する。

成分	式(I)で表わされる化合物	10 mg	
	乳糖	79 mg	10
	コーンスターチ	10 mg	
	ステアリン酸マグネシウム	1 mg	
		<hr/>	
		100 mg	

式(I)で表わされる化合物、乳糖を60メッシュのふるいに通す。コーンスターチは120メッシュのふるいに通す。これらとステアリン酸マグネシウムをV型混合機にて混合する。10倍散100mgを5号硬ゼラチンカプセルに充填する。

## 製剤例3

以下の成分を含有するカプセル充填用顆粒剤を製造する。

成分	式(I)で表わされる化合物	15 mg	
	乳糖	90 mg	20
	コーンスターチ	42 mg	
	HPC-L	3 mg	
		<hr/>	
		150 mg	

式(I)で表わされる化合物、乳糖を60メッシュのふるいに通す。コーンスターチを120メッシュのふるいに通す。これらを混合し、混合末にHPC-L溶液を添加して練合、造粒、乾燥する。得られた乾燥顆粒を整粒後、その150mgを4号硬ゼラチンカプセルに充填する。

## 製剤例4

以下の成分を含有する錠剤を製造する。

成分	式(I)で表わされる化合物	10 mg	30
	乳糖	90 mg	
	微結晶セルロース	30 mg	
	CMC-Na	15 mg	
	ステアリン酸マグネシウム	5 mg	
		<hr/>	
		150 mg	

式(I)で表わされる化合物、乳糖、微結晶セルロース、CMC-Na(カルボキシメチルセルロース ナトリウム塩)を60メッシュのふるいに通し、混合する。混合末にステアリン酸マグネシウム混合し、製錠用混合末を得る。本混合末を直打し、150mgの錠剤を得る。

## 製剤例5

静脈用製剤は次のように製造する：

式(I)で表わされる化合物	100 mg
飽和脂肪酸グリセリド	1000 ml

上記成分の溶液は通常、1分間に1mlの速度で患者に静脈内投与される。

【産業上の利用可能性】

【0148】

本発明化合物は、優れたEGF受容体およびHER2の阻害作用を有し、安全性も高いことから、医薬品、とりわけ癌に対する治療剤として有用である。



## フロントページの続き

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- (72)発明者 大森 直樹  
大阪府大阪市福島区鷺洲5丁目12番4号 塩野義製薬株式会社内
- (72)発明者 高山 正己  
大阪府大阪市福島区鷺洲5丁目12番4号 塩野義製薬株式会社内
- (72)発明者 大森 愛子  
大阪府大阪市福島区鷺洲5丁目12番4号 塩野義製薬株式会社内
- (72)発明者 遠藤 毅  
大阪府大阪市福島区鷺洲5丁目12番4号 塩野義製薬株式会社内

審査官 伊藤 幸司

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