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None
(58) Field of search
C2C
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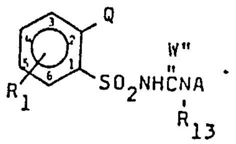
(54) Herbicidal sulfonamides

(57) Novel benzenesulfonamide derivatives of formula



wherein Q is selected from various five and six-membered heterocyclic groups which are optionally substituted;

R_1 is H, F, Cl, Br, CH_3, CF_3 or OCH_3;

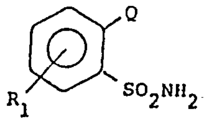
R_{13} is H or CH_3;

W″ is O or S; and

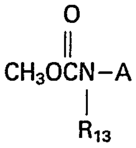
A is a substituted heterocyclic moiety;

and their agriculturally suitable salts, exhibit potent herbicidal activity. Some are also of interest as plant growth regulants.

The novel compounds may be formulated for use in conventional manner. They may be made by a variety of methods, e.g. by reacting a sulfonamide of formula



with a methyl carbamate of formula



$$CH_3O\overset{\overset{\displaystyle O}{\|}}{C}\underset{\underset{\displaystyle R_{13}}{|}}{N}-A$$

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SPECIFICATION

Herbicidal sulfonamides

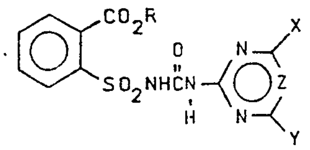
Background of the invention

This invention relates to *ortho*-heterocyclicbenzenesulfonamides and, more particularly, to *ortho*-(isoxazol-3, or 4-, or 5-yl)benzenesulfonamides, *ortho*-(isothiazol-3, or 4-, or 5-yl)benzenesulfonamides, *ortho*-(1H-pyrazol-1, or 3- or 4-, or 5-yl)benzenesulfonamides, *ortho*-(1,3,4-oxadiazol-2-yl)benzenesulfonamides, *ortho*-(1,2,4-oxadiazol-3, or 5-yl)benzenesulfonamides, *ortho*-(1,2,5-oxadiazol-3-yl)benzenesulfonamides, *ortho*-(1,3,4-thiadiazol-2-yl)benzenesulfonamides, *ortho*-(1,2,4-thiadiazol-3-, or 5-yl)benzenesulfonamides, *ortho*-(1,2,5-thiadiazol-3-yl)benzenesulfonamides, *ortho*-(1,2,3-thiadiazol-4-, or 5-yl)benzenesulfonamides, *ortho*-(1H-1,3,4-triazol-1-, or 2-yl)benzenesulfonamides, *ortho*-(1H-1-methyl-1,2,4-triazol-3-, or 5-yl)benzenesulfonamides, *ortho*-(1H-1,2,4-triazol-1-yl)benzenesulfonamides, *ortho*-(oxazol-2, or 4-, or 5-yl)benzenesulfonamides, *ortho*-(thiazol-2, or 4-, or 5-yl)benzenesulfonamides, *ortho*-(1H-imidazol-1, or 2-, or 4-, or 5-yl)benzenesulfonamides, *ortho*-(oxazolin-2-yl)benzenesulfonamides, *ortho*-(thiazolin-2-yl)benzenesulfonamides, *ortho*-(1H-imidazolin-2-yl)benzenesulfonamides, *ortho*-(oxazin-2-yl)benzenesulfonamides, *ortho*-(thiazin-2-yl)benzenesulfonamides, or *ortho*-(tetrahydropyrimidin-2-yl)benzenesulfonamides and their use in agriculturally suitable compositions as pre-emergence and/or post-emergence herbicides and as plant growth regulants.

Description of the prior art

U.S. Patents 4,127,405 and 4,169,719 disclose herbicidal methoxymethylpyrimidine sulfonylurea compounds of the type which contain a $-CH_2OCH_3$ heterocyclic substituent.

European Patent No. 7687 discloses herbicidal sulfonylurea compounds such as, among others,

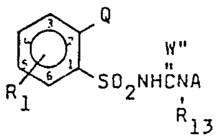


where
X is CH_3 or OCH_3;
Z is CH or N; and
Y is C_1-C_4 alkyl substituted with OCH_3, OC_2H_5, CN, C(O)L, or 1-3 atoms of F, Cl, or Br, where L is NH_2, OH, $N(OCH_3)CH_3$, $NH(C_1$-C_4 alkyl), $N(C_1$-C_4 alkyl) or C_1-C_6 alkoxy.

Our EP-A-0,044,211 discloses herbicidal *o*-phenylbenzenesulfonylureas.

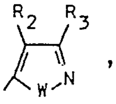
Summary of the invention

This invention relates to compounds of Formula I, suitable agricultural compositions containing them, and their method of use as general and/or selective pre-emergence and/or post-emergence herbicides, and as plant growth regulants.

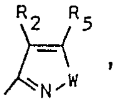


I

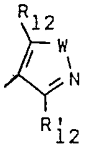
wherein
Q is



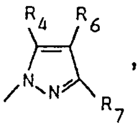
Q1



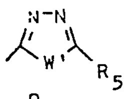
Q2



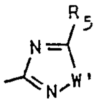
Q3



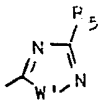
Q4



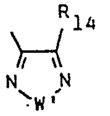
Q_5 ,



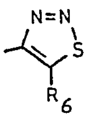
Q_6 ,



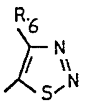
Q_7 ,



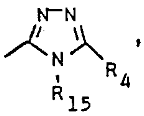
Q_8 ,



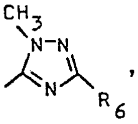
Q_9 ,



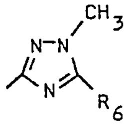
Q_{10} ,



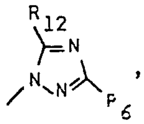
Q_{11} ,



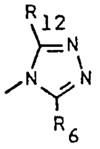
Q_{12} ,



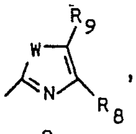
Q_{13} ,



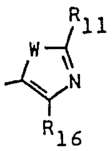
Q_{14} ,



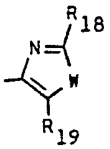
Q_{15} ,



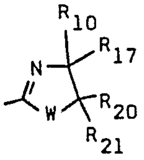
Q_{16} ,



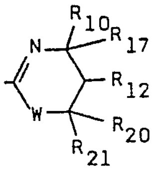
Q_{17} ,



Q_{18} ,

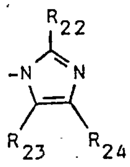


Q_{19} ,



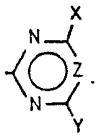
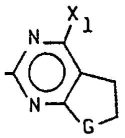
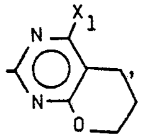
Q_{20}

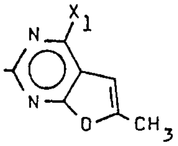
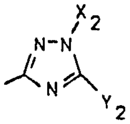
or

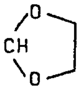


Q_{21} ;

W is O, S or NR;
W' is O or S;
W'' is O or S;
R is H or C_1-C_4 alkyl;
R_1 is H, F, Cl, Br, CH_3, CF_3 or OCH_3;
R_2 is H, CH_3, C_2H_5, Cl or Br;
R_3 is H, CH_3, C_2H_5, Cl, Br, OCH_3, OC_2H_5 or SCH_3;
R_4 is H or C_1-C_4 alkyl;
R_5 is H, CH_3, C_2H_5, Cl, Br, OCH_3, OC_2H_5, or SCH_3;
R_6 is H, CH_3, or C_2H_5;
R_7 is H or C_1-C_4 alkyl;
R_8 is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_9 is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_{10}, R_{17}, R_{20} and R_{21} are independently H or C_1-C_4 alky;
R_{11} is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_{12} is H or CH_3;
R'_{12} is H or CH_3;
R_{13} is H or CH_3;
R_{14} is H, CH_3, C_2H_5, Cl, OCH_3, OC_2H_5 or SCH_3;
R_{15} is C_1-C_3 alkyl;
R_{16} is H, CH_3 or C_2H_5;
R_{18} is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_{19} is H, CH_3 or C_2H_5;
R_{22} is H, CH_3, C_2H_5, Cl, Br, CH_3O, C_2H_5O or CH_3S;
R_{23} is H, CH_3 or C_2H_5;
R_{24} is H, CH_3 or C_2H_5;

A is  ,  ,  ,

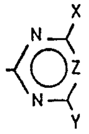
 or  ;

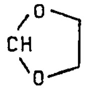
X is CH_3, OCH_3 or Cl;
X_1 is CH_3, OCH_3 or Cl;
X_2 is C_1-C_3 alkyl or CH_2CF_3;
Y is CH_3, C_2H_5, CH_2OCH_3, OCH_3, OC_2H_5, $CH(OCH_3)_2$, NH_2, $NHCH_3$, $N(CH_3)_2$, $OCH_2CH_2OCH_3$, OCH_2CF_3, SCH_3, CF_3 or  ;

Y_2 is CH_3O, C_2H_5O, CH_3S or C_2H_5S;
Z is CH or N;
G is O or CH_2;

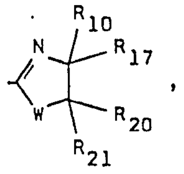
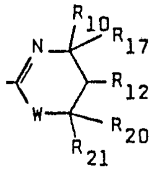
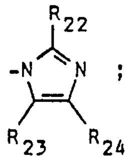
and their agriculturally suitable salts;
provided that

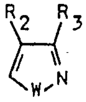
(a) when R_2 is Cl or Br, then W is O or S;
(b) when X is Cl, then Z is CH and Y is OCH_3, OC_2H_5, NH_2, $NHCH_3$ or $N(CH_3)_2$;
(c) when W″ is S, then R_{13} is H, A is

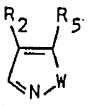
A is  ,

Y is CH_3, OCH_3, C_2H_5, OC_2H_5, CH_2OCH_3, $CH(OCH_3)_2$ or  ; and

Q is not

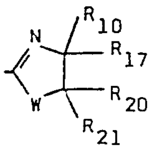
 ,  or  ;

(d) when Q is  , then one of R_2 or R_3 must be H, CH_3 or C_2H_5;

(e) when Q is  , then one of R_2 or R_5 must be H, CH_3 or C_2H_5;

(f) the total number of carbon atoms of Q must be less than or equal to 8;
(g) when R_{18} is other than H, CH_3, C_2H_5, or CH_3S, then W is S or O;
(h) the total number of carbon atoms of R_{10}, R_{17}, R_{20} and R_{21} is less than or equal to 4; and

(i) when Q is



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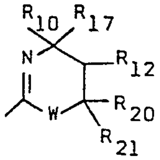

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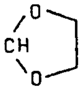
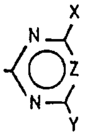
then R_{13} is H, R is other than H, X is Cl or OCH_3 and Y is OCH_3 or OC_2H_5.

Preferred for their higher herbicidal activity and/or their most favourable ease of synthesis are:

(1) Compounds of Formula I where
R and R_{15} are independently CH_3 or C_2H_5;
R_2, R_3, R_4, R_5, R_6, R_7 and R_{14} are independently H or CH_3;
R_8, R_9, R_{11}, R_{16}, R_{18}, R_{19}, R_{22}, R_{23} and R_{24} are independently H, CH_3 or C_2H_5;
R_{10}, R_{17}, R_{20} and R_{21} are independently H or CH_3; and
W″ is O.
(2) Compounds of the Preferred (1) where R_1 and R_{13} are H.
(3) Compounds of Preferred (2) where Y is CH_3, CH_2OCH_3, OCH_3, OC_2H_5, $CH(OCH_3)_2$ or



, and A is



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(4) Compounds of Preferred (3) where W = O.
(5) Compounds of Preferred (3) where W = S.
(6) Compounds of Preferred (3) where W = NR.
(7) Compounds of Preferred (3) where W′ = O.
(8) Compounds of Preferred (3) where W′ = S.
(9) Compounds of Formula I where W″ is O; R and R_{15} are CH_3;
R_1, R_2, R_3, R_4, R_5, R_6, R_7, R_8, R_9, R_{10}, R_{11}, R_{12}, R_{13}, R_{14}, R_{16}, R_{17}, R_{18}, R_{19}, R_{20}, R_{21}, R_{22}, R_{23} and R_{24} are H;
X is CH_3, CH_3O or Cl;
Y is CH_3, CH_3O or CH_2OCH_3; and
Q is Q_1, Q_2, Q_3, Q_4, Q_5, Q_6, Q_7, Q_8, Q_9, Q_{10}, Q_{11}, Q_{12}, Q_{13}, Q_{14}, Q_{15}, Q_{16}, Q_{17}, Q_{18} and Q_{21}.

Specifically Preferred are:

- 2-(isoxazol-5-yl)-N-[(4,6-dimethyoxypyrimidin-2-yl)aminocarbonyl]benzenesulfonamide;
- 2-(isoxazol-5-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide;
- 2-(isoxazol-4-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide;
- 2-(isoxazol-4-yl)-N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide;
- N-[(4-methyl-6-methoxypyrimidin-2-yl)aminiocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide;
- N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl]benzensulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide;
- N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide;

- N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(1-methyl-1H-imidazol-2-yl)benzenesulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide;
- N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide;
- 2-(4,5-dihydro-5,5-dimethyloxazol-2-yl)-N-[(4,6-dimethoxy-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide;
- N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide;
- N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide;
- N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide;
- N-[(4,6-dimethoxy-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide; and
- N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide.

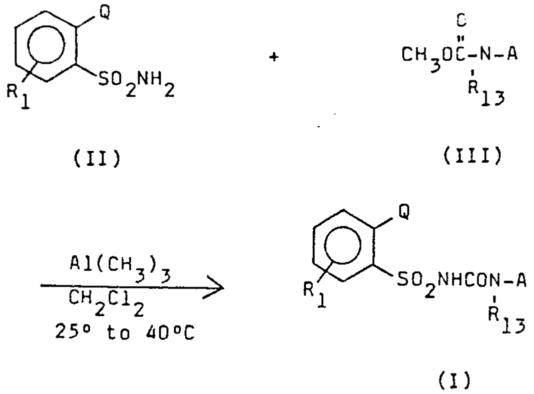
Detailed description of the invention

Synthesis

The compounds of Formula (I) can be prepared by one or more of the methods described below in Equations 1 to 4d. The preferred reaction conditions are given for guidance but may be varied within the discretion and knowledge of those skilled in the art.

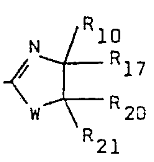
As shown in Equation 1 below, many compounds of Formula (I), where W″ is O, can be prepared by reacting a sulfonamide of Formula (II) with an appropriate methyl carbamate of Formula (III) in the presence of an equimolar amount of trimethylaluminum. The reactions are carried out at 25° to 40°C in an inert solvent such as methylene chloride for 10 to 96 hours under an inert atmosphere as taught in our EP-A-82305498.6, and in U.S. Application Serial No. 337,934, laid open as a priority document in support of the present Application.

Equation 1

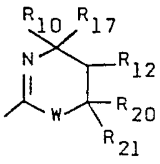


wherein

Q is other than



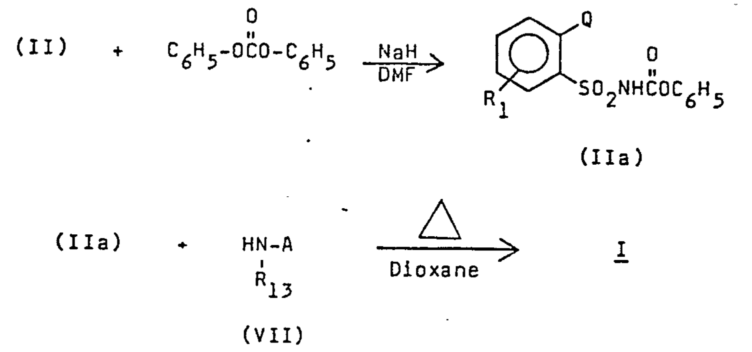
or



,

As shown in Equation 1a, many compounds of Formula (I), where W″ is O, can also be prepared by reacting a sulfonylcarbamate of Formula (IIa) with an appropriate amine of Formula (VII).

Equation 1a



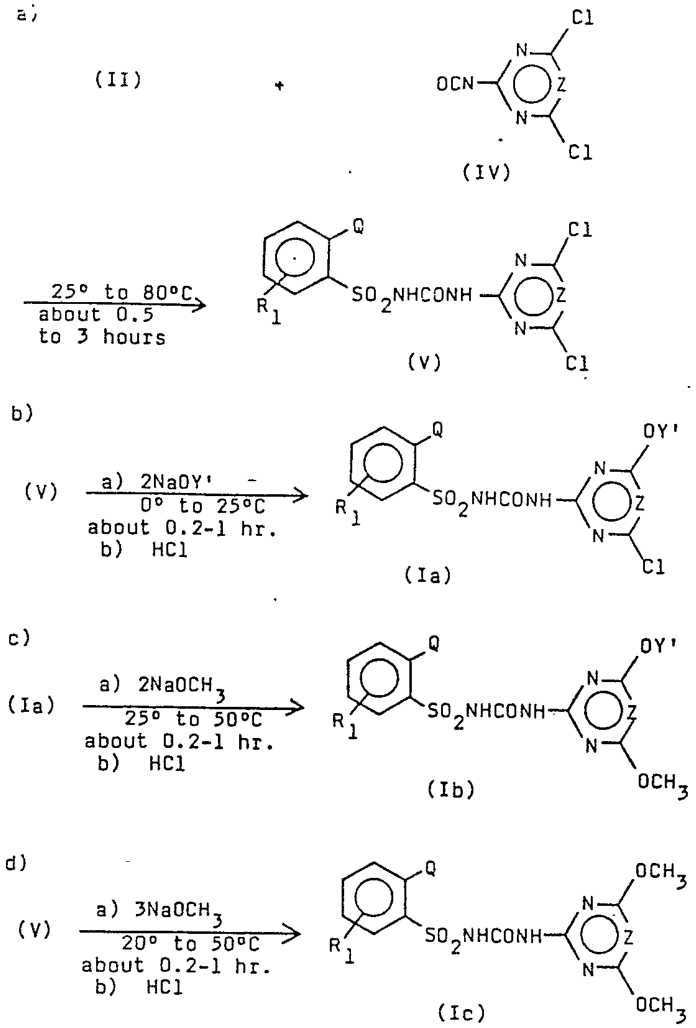
wherein

R_1, R_{13}, A and Q are as defined above;
except R_3 and R_{14} are H or C_1-C_2 alkyl,
R_5 is C_1-C_2 alkyl, and R is C_1-C_4 alkyl.

The reaction is carried out at 50-100°C in a solvent such as dioxane for 0.5 to 24 hours as taught in EP-A-44807. The required carbamates *IIa* are prepared by reacting the corresponding sulfonamide II with diphenylcarbonate in the presence of a strong base.

Some of the compounds of Formula (I) can also be prepared as shown in Equation 2 below.

Equation 2



wherein

Y' is CH_3 or C_2H_5 and R_1, Q and Z are as originally defined, except R_3 and R_5 are not Cl or Br and R_{14} is not Cl.

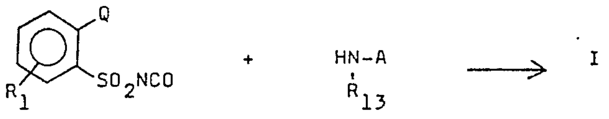
The reactions of Equation 2 are run according to similar procedures taught in European Patent 30,140.

Thus, in reaction 2a, a sulfonamide of Formula (II) is reacted with dichloropyrimidinyl isocyanate or dichlorotriazinyl isocyanate of Formula (IV) in an inert solvent such as acetonitrile at reflux for 0.5 to 3 hours to form a sulfonylurea of Formula (V). The product is isolated by filtration. In reaction 2b, V is reacted with two mole equivalents of sodium methoxide or sodium ethoxide in tetrahydrofuran at 0° to 25°C for about one hour, followed by acidification with hydrochloric acid to a pH of about 1, to form a sulfonylurea of Formula (Ia). The product is isolated by filtration. In reaction 2c, Ia is reacted with two mole equivalents of sodium methoxide in methanol at 25° to about 50°C for about 1 hour, followed by acidification with hydrochloric acid to a pH of about 1, to form a sulfonylurea of Formula (1b). Alternatively, as shown in reaction 2d, V can be reacted with at least three mole equivalents of sodium methoxide at 20° to 50°C for about 1 hour, followed by acidification with hydrochloric acid to a pH of about 1, to provide Ic directly, where Y′ of Ib is OCH_3. The products of reactions 2c and 2d are isolated by addition of water and filtration.

The heterocyclic isocyanates of Formula (IV) in Equation 2 above can be prepared by methods described in Swiss 579,062, U.S. 3,919,228, U.S. 3,732,223 and *Angew Chem. Int. Ed., 10,* 402 (1976), to which the reader is referred for further information.

As shown in Equation 3 below, some of the compounds of Formula (I), where W″ is O, can also be prepared by reacting a sulfonyl isocyanate of Formula (VI) with an amine of Formula (VII).

Equation 3



(VI) (VII)

wherein

R_1, R_{13} and A are as originally defined; and

Q is as defined in Equation 1 above.

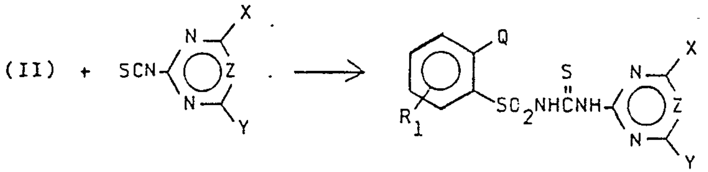
The reaction of Equation 3 above can best be carried out in an inert, aprotic, organic solvent such as methylene chloride, tetrahydrofuran or acetonitrile at ambient pressure and temperature. The mode of addition is not critical; however, it is often convenient to add the sulfonyl isocyanate to a stirred suspension of amine VII. The reaction is generally exothermic. In some cases, tho desired product is insoluble in the warm reaction medium and crystallizes from it in pure form. Products soluble in the reaction medium are isolated by evaporation of the solvent, trituration of the solid residue with solvents such as 1-chlorobutane, ethyl ether, ethyl acetate or pentane and filtration. Impure products may be purified by column chromatography on silica gel.

Many of the intermediate sulfonyl isocyanates of Formula (VI) in Equation 3 above can be prepared, although often times in low yields, from sulfonamides by methods described in U.S. 4,238,621. The method requires reacting sulfonamides with phosgene, in the presence of *n*-butyl isocyanate and a tertiary amine catalyst, at reflux in a solvent such as xylene. A preferred catalyst is 1,4-diazabicyclo[2.2.2]octane (DABCO).

Alternatively, many of the sulfonyl isocyanates VI can be prepared, although again in low yields, from sulfonamides by a two-step procedure. This consists of (1) reacting the sulfonamide with *n*-butyl isocyanate and a base such as potassium carbonate at reflux in an inert solvent such as 2-butanone to form a *n*-butyl sulfonylurea; and (2) reacting this compound with phosgene and DABCO catalyst at reflux in xylene solvent. This method is similar to a procedure taught by Ulrich and Sayigh, *New Methods of Preparative Organic Chemistry*, Vol. VI, p. 223-241, Academic Press, New York and London, W. Foerst Ed.

As shown in Equation 3a below, compounds of Formula (I), where W″ is S, can be prepared by reacting sulfonamide *II* with an appropriate triazine or pyrimidine isothiocyanate of Formula (VIIj).

Equation 3a



(VIIj) (Id)

wherein

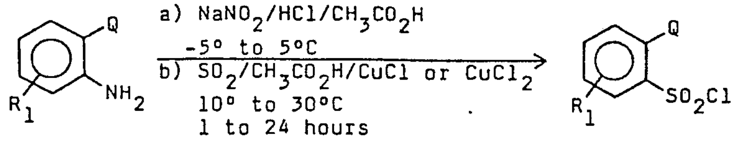
R_1, Q, X, Y and Z are as originally defined.

The reaction of Equation 3a is carried out by dissolving or suspending the sulfonamide and isothiocyanate in a polar solvent such as acetone, acetonitrile, ethyl acetate or methyl ethyl ketone, adding an equivalent of a base such as potassium carbonate and stirring the mixture at ambient temperature up to the reflux temperature for one to twenty-four hours as taught in our EP-A-35,893. The required isothiocyanates *VIIj* are prepared according to the method of JP-A-51-143686, dated June 5, 1976, or that of W. Abraham and G. Barnikow, *Tetrahedron*, *29*, 691 (1973).

As shown in Equation 4 below, intermediate sulfonamides of Formula (II) described above can be prepared from amines of Formula (VIII) by a two-step procedure. This consists of (4a) diazotizing VIII and coupling the diazonium salt with sulfur dioxide to form a sulfonyl chloride of Formula (IX); and (4b) aminating IX with ammonium hydroxide or anhydrous ammonia to form II.

Equation 4

a)



(VIII) (IX)

b)

$$(IX) \xrightarrow[\substack{-20° \text{ to } 40°C \\ 0.5 \text{ to } 10 \text{ hours}}]{NH_4OH \text{ or } NH_3} \qquad (II)$$

wherein

R_1 is as originally defined; and

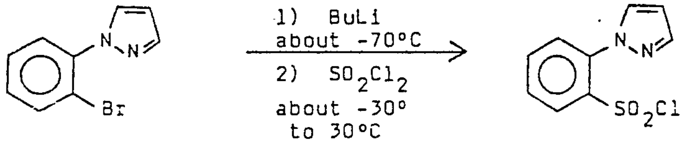
Q is as defined in Equation 1 above, provided that when Q is an imidazole ring, R is C_1-C_4 alkyl.

The reaction of Equation 4a is accomplished by treating a solution of amine VIII in a mixture of concentrated hydrochloric acid and glacial acetic acid with a solution of sodium nitrite in water at −5° to 50°C. After stirring for 10-30 minutes at about 0°C to insure complete diazotization, the solution is added to a mixture of an excess of sulfur dioxide and a catalytic amount of cuprous chloride or cupric chloride in glacial acetic acid at about 10°C. The temperature is kept at about 10°C for 1/4 to 1 hour, then raised to 20° to 30°C and held at that temperature for 2 to about 24 hours. This solution is then poured into a large excess of ice water. The sulfonyl chloride IX can be isolated by filtration or by extraction into a solvent such as ethyl ether, methylene chloride or preferably, 1-chlorobutane, followed by evaporation of the solvent.

The amination described in the reaction of Equation 4b above is conveniently carried out by treating a solution of the sulfonyl chloride IX with at least two mole equivalents of anhydrous ammonia in a solvent such as ethyl ether or methylene chloride at −20° to 30°C. If the sulfonamide product II is insoluble, it may be isolated by filtration followed by washing out the salts with water. If product II is soluble in the reaction solution, it may be isolated by filtering off the precipitated ammonium chloride and evaporation of the solvent. Alternatively, many sulfonamides II can be prepared by reaction of corresponding sulfonyl chlorides IX with excess aqueous ammonium hydroxide in tetrahydrofuran at 0° to about 40°C for 0.5 to 10 hours. The sulfonamide product II is isolated by evaporation of the tretrahydrofuran solvent, addition of water to the residue and filtration.

Alternatively, the intermediate sulfonyl chloride *IXa* can be prepared as shown below in Equation 4c.

Equation 4c



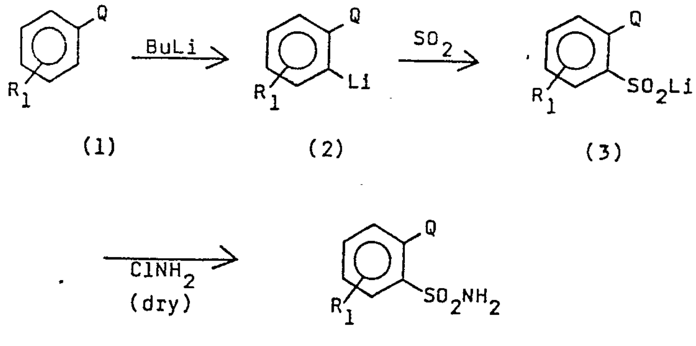
(IXa)

According to Equation 4c, a lithium salt, prepared by reaction of 1-(2-bromophenyl)pyrazole with butyl lithium in ether at about −70°C, is added to sulfuryl chloride in hexane at about −30° to −20°C and stirred for 0.5 to 10 hours at −30° to 30°C to yield sulfonyl chloride *IXa*, according to teachings of S. N. Bhattacharya et al., *J. Chem. Soc. (C)*, 1265 (1968). Subsequent reaction of IXa with ammonia or ammonium hydroxide as described above provides the corresponding sulfonamide.

Starting with appropriate *ortho*-(heterocyclic)bromobenzenes, and carrying out the procedures described in Equation 4c, or simple modifications thereof, one skilled in the art may prepare some of the other sulfonyl chlorides of Formula (IX) described above. Of necessity, the reactions are limited to those cases in which the *ortho*-hetero group, *Q*, is inert to lithium reagents under the conditions of the reactions, which will be obvious to one skilled in the art. For a general review of metalation with lithium reagents, see H. W. Gschwend and H. R. Rodriguez, *Org. Reactions, 26,* 1 (1979).

Also sulfonamides *IIb* are best prepared by the procedure of Equation 4d below.

Equation 4d

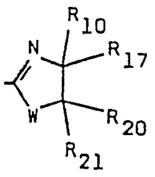


(1) (2) (3)

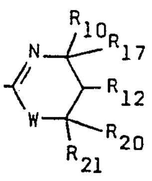
(IIb)

wherein

Q is



or

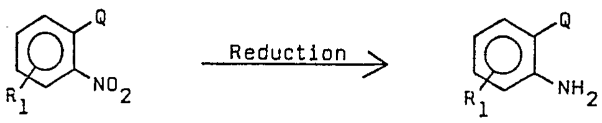


The preparation of sulfinic acid salts (3) by the procedure of Equation 4d is well known in the art. See U.S. 4,127,405 and *Organic Reactions, Vol. 26,* 1979, J. Wiley and Sons, Inc., N.Y. Sulfonamides (IIb) are best prepared by treatment of sulfinic acid salts with chloramine. In this procedure an ethereal solution or suspension of the salt (3) is treated at low temperature (25 to −30°) with a dry ethereal solution of chloramine. The reaction is stirred for a period of several minutes to several hours. After filtration, the reaction mixture is washed with aqueous bisulfite and then dried and the solvent removed on a rotary evaporator. The crude product is further purified by usual methods such as crystallization or chromatography.

Some of the amines of Formula (VIII) in Equation 4 above are known. For instance, 4-(2-aminophenyl)-isothiazole may be prepared by the procedure of J. H. Finley, *J. Heterocycl. Chem., 6,* 841 (1969); 2-(2-aminophenyl)-1,3,4-thiadiazole by the procedure of M. Ohta, *J. Pharm. Soc. Japan, 73,* 701 (1953); 2-(2-aminophenyl)-5-methyl-1,3,4-thiadiazole by the procedure of S. Leistner and G. Wagner, *Z. Chem., 14,* 305 (1974); 2-(2-aminophenyl)-1,3,4-oxadiazole by the procedure of M. Vincent et al., *Bull. Soc. Chim. France,* 1580 (1962); 3-(2-aminophenyl)-5-methyl-1,2,4-oxadiazole by the procedure of H. Goncalves et al., *Bull. Soc. Chim. France,* 2599 (1970); 4-(2-aminophenyl)-1,2,4-triazole by the procedure of M. Khan and J. Polya, *J. Chem. Soc. C,* 85 (1970); and 3-methyl-4-(2-aminophenyl)-1,2,4-triazole and 3,5-dimethyl-4-(2-aminophenyl)-1,2,4-triazole by the procedure of W. Ried and H. Lohwasser, *Justus Liebigs Ann. Chem., 699,* 88 (1966).

As shown in Equation 5 below, other amines of Formula (VIII) can be prepared by reduction of corresponding nitrobenzenes of Formula (X) with reagents described below.

Equation 5



(X) (VIII)

wherein

R_1 and Q are as originally defined.

The reduction reactions of Equation 5 above can be run by methods known in the literature by one skilled in the art. For instance, many of the reductions can be run by one or more of the following methods:

(a) with stannous chloride or tin and hydrochloric acid, either neat or in an inert solvent such as methanol, at about 25° to 80°C for 0.5 to 10 hours. For details refer to similar procedures described in G. Corsi et al., *Boll. Chim. Farm., 103,* 115 (1964); J. H. Finley, *J. Heterocycl. Chem., 6,* 841 (1969); A. Quilico et al., *Gazz. Chim. Ital., 76,* 87 (1946); and M. Khan and J. Polya, *J. Chem. Soc. C.,* 85 (1970).

(b) with ferrous sulfate heptahydrate and 28% ammonium hydroxide in an inert solvent such as aqueous ethanol at about 40° to 80°C for about 1 to 2 hours. For details refer to similar procedures described in T. Naito et al., *Chem. Pharm. Bull., 16,* 160 (1968); Neth. Appl. 6,608,094; and U.S. 3,341,518;

(c) with ammonium chloride and iron powder in an inert solvent such as water at 50° to about 80°C for 1 to 3 hours. For details refer to a similar procedure described in M. Ohta et al., *J. Pharm. Soc. Japan, 73,* 701 (1953);

(d) with sodium hydrogen sulfide in an inert solvent such as methanol at about 40° to 70°C for about 0.5 to 1 hour. For details refer to similar procedures described in G. Corsi et al., *Boll. Chim. Farm., 103,* 115 (1964); and U.S. 3,270,029;

(e) by catalytic reduction with 5% palladium-on-charcoal, in the presence of 2 to 5 equivalents of aqueous hydrochloric acid, in an inert solvent such as ethanol at 25° to 45°C at 1 to 3 atmospheres of hydrogen. For details refer to a similar procedure described in U.S. 3,910,942; and Ger. Offen. 2,415,978;

(f) by catalytic reduction with 5% Raney Nickel in an inert solvent such as ethanol or dioxane at 25° to 45°C at 1 to 3 atmospheres of hydrogen. For details refer to similar procedures described in U.S. 3,270,029 and Neth. Appl. 6,513,932;

(g) by catalytic reduction with 5% palladium-on-charcoal in an inert solvent such as methanol at 25° to 45°C at 1 to 3 atmospheres of hydrogen for short reaction times, i.e., less than 1 hour. For details refer to a similar procedure described in M. Vincent et al., *Bull. Soc. Chim. France,* 1580 (1962); and

(h) by reduction with Raney Nickel catalyst and hydrazine hydrate in 95% ethanol at 25° to 80°C for 0.2 to about 1 hour. For details refer to a similar procedure described in C. Ainsworth et al., *J. Med. Pharm. Chem., 5,* 383 (1962).

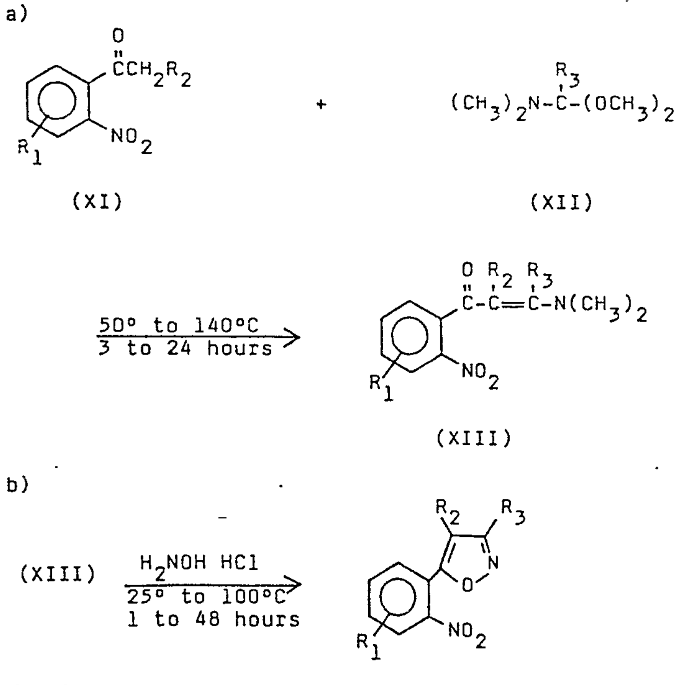
(i) with sodium sulfide in 50% aqueous *p*-dioxane at about 25 to 80°C for 0.25 to 1 hour, or with sodium sulfide and sodium bicarbonate in refluxing methanol for 1 to 10 hours. For details refer to Y. Lin and S. Lang, Jr., *J. Heterocycl. Chem., 17,* 1273 (1980) and P. Smith and J. Boyer, *J. Am. Chem. Soc., 73,* 2626 (1951) respectively; and,

(j) with sodium hydrosulfite in ethanol-water at about 25° to 60°C for 0.25 to 1 hour at a pH of less than 7. For details refer to U.S. 4,229,343.

The *ortho*-heteroaromatic nitrobenzenes of Formula (X) in Equation 5 above are important starting compounds for preparing the compounds I of this invention, which can be prepared by the following methods.

As shown in Equation 6 below, certain 5-(2-nitrophenyl)isoxazoles of Formula (Xa) can be prepared by reacting a 2-nitrophenyl alkyl ketone of Formula (XI) with an appropriate dimethylalkanamide dimethyl acetal of Formula (XII) to form a 3-dimethylamino-1-(2-nitrophenyl)-2-propen-1-one of Formula (XIII). Subsequent reaction of XIII with hydroxylamine hydrochloride provides Xa.

Equation 6



wherein

R_1 is as originally defined; and

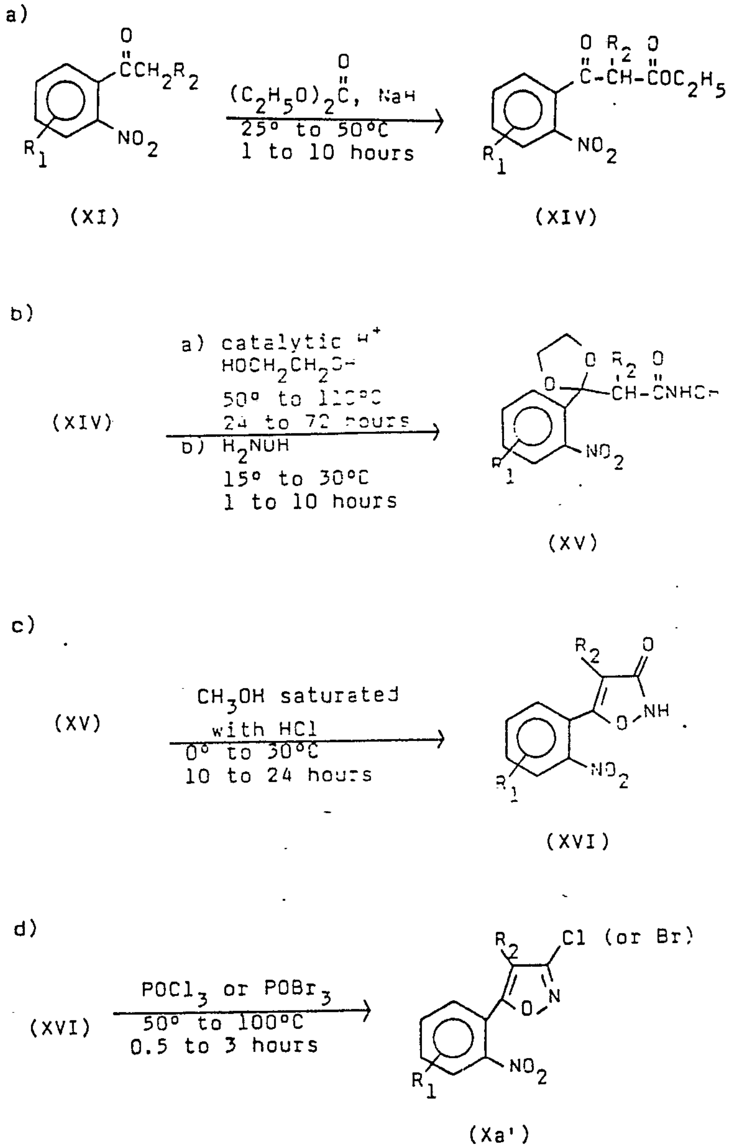
R_2 and R_3 are H, CH_3 or C_2H_5.

The reaction of Equation 6a is run at 50° to 140°C for 3 to 24 hours in a solvent such as toluene or dimethylformamide or excess dimethyl alkanamide dimethyl acetal. The product can be isolated by evaporating the solvent. For more details, refer to similar procedures described in *Technical Information Bulletin*, "DMF Acetals", Aldrich Chemical, December 1973, and Lin and Lang, *J. Org. Chem., 45,* 4857 (1980). The preparation of dimethyl alkanamide dialkyl acetals is reviewed in Abdulla and Brinkmeyer, *Tetrahedron, 35,* 1675 (1979).

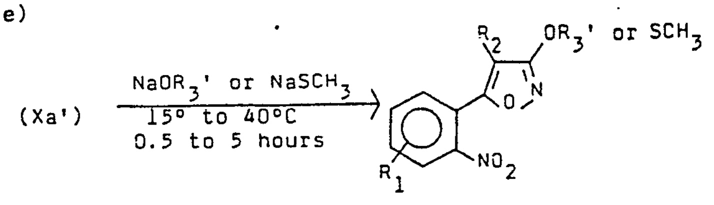
The reaction of Equation 6b above is run in an inert solvent such as ethanol or aqueous dioxane at 25° to 100°C for 1 to 48 hours. The product is isolated by addition of water and extraction with methylene chloride. For more details refer to similar procedures described in Lin and Lang, *J. Heterocycl. Chem., 14,* 345 (1977).

Another method for preparing some 5-(2-nitrophenyl)isoxazoles is shown in Equation 7 below. The method requires transforming a 2-nitrophenyl alkyl ketone of Formula (XI) by a series of procedures to a 5-(2-nitrophenyl)isoxazolin-3-one of Formula (XVI). Subsequent reaction of XVI with phosphorus oxychloride or phosphorus oxybromide provides 3-halo-5-(2-nitrophenyl)isoxazoles of Formula (Xa'). Reaction of Xa' with a sodium methoxide, sodium ethoxide or sodium methylmercaptide then provides 3-alkoxy or 3-methylthio-5-(2-nitrophenyl)isoxazoles of Formula (Xa").

Equation 7



e)



(Xa")

wherein

R_1 is as originally defined;

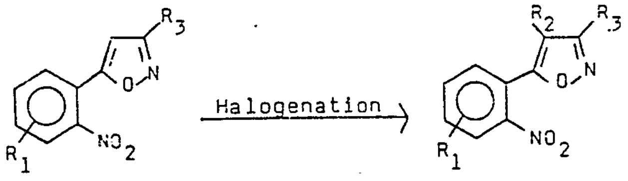
R_2 is H, CH_3 or C_2H_5; and

R_3' is CH_3 or C_2H_5.

The 5-(2-nitrophenyl)isoxazolin-3-one of Formula (XVI) in Equation 7 above can be prepared by a series of reactions similar to those described in the art for transforming acetophenone to 5-phenylisoxazolin-3-one, e.g., R. Jacquier et al., *Bull. Soc. Chim.*, 3694 (1969) and *ibid.*, 1978 (1970). Thus, by substituting 2-nitrophenyl alkyl ketone XI for acetophenone and carrying out the appropriate reactions in the cited art, and which are illustrated in reactions of Equations 7a to 7c, one skilled in the art can prepare XVI. The reaction of Equation 7d is run in a solvent such as toluene at 50° to 100°C for 0.5 to 3 hours. The product is isolated by evaporation of the solvent and purified by column chromatography on silica gel. Reaction 7e is run in a solvent such as methanol or tetrahydrofuran at about 15° to 40°C for 0.5 to 5 hours. The product is isolated by addition of water and extraction with methylene chloride. The product is purified by column chromatography on silica gel.

As shown in Equation 8 below, 4-halo-5-(2-nitrophenyl)isoxazoles of Formula (Xa"") can be prepared by halogenating 5-(2-nitrophenyl)isoxazoles of Formula (Xa''') with halogenating reagents described below.

Equation 8



(Xa''') (Xa"")

wherein

R_1 and R_3 are as originally defined; and

R_2 is Cl or Br.

The reaction of Equation 8 can be run by one or more of the following methods known in the art for halogenating phenylisoxazoles in the 4-position of the isoxazole ring in preference to the phenyl ring or other positions of the isoxazole ring:

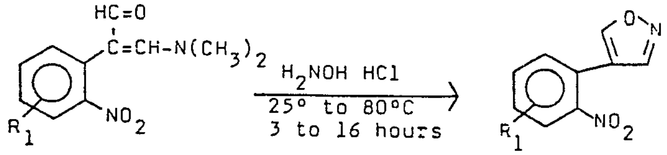
(a) reacting Xa''' with sulfuryl chloride or sulfuryl bromide at 15° to 80°C for 0.5 to 3 hours, either neat or in a solvent such as methylene chloride or carbon tetrachloride, according to the teachings of J. Carr et al., *J. Heterocycl. Chem., 20,* 934 (1977);

(b) reacting Xa''' with chlorine or bromine at 15° to 60°C for 0.5 to 5 hours in methylene chloride or acetic acid, according to the teachings of *ibid*; or

(c) reacting Xa''' with bromine or chlorine and iron powder catalyst in a solvent such as carbon tetrachloride at 25° to 80°C for 0.5 to 3 hours, according to the teachings of N. Kochetkov et al., *Zhur. Obshchei. Khim., 28,* 359 (1958). The preparation of Xa''' is described in Equations 6 and 7 above.

As shown in Equation 9 below, 4-(2-nitrophenyl)isoxazoles of Formula (Xb) can be prepared by reacting a 3-(dimethylamino)-2-(2-nitrophenyl)acrolein of Formula (XVII) with hydroxylamine hydrochloride.

Equation 9



(XVII) (Xb)

wherein

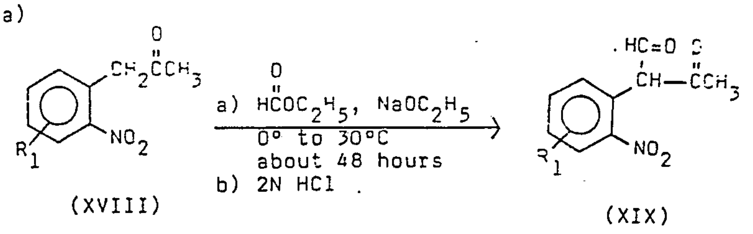
R_1 is as originally defined.

The reaction of Equation 9 is run in ethanol at 25° to 80°C for 3 to 16 hours. The product is isolated by addition of water and extraction with methylene chloride. The product is purified by recrystallization or column chromatography on silica gel. The starting material XVII is prepared by known methods, e.g., U. Hengartner et al., *J. Org. Chem., 44,* 3748 (1979).

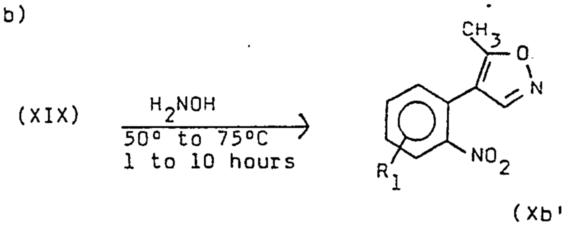
5-Methyl-4-(2-nitrophenyl)isoxazoles of Formula (Xb′) can be prepared as shown in Equation 10 below. The method requires reacting a 2-nitrophenylpropanone of Formula (XVIII) with ethyl formate and sodium ethoxide to form a 3-oxo-2-(2-nitrophenyl)butyraldehyde of Formula (XIX). Subsequent reaction of XIX with hydroxylamine provides Xb′.

Equation 10

a)



b)



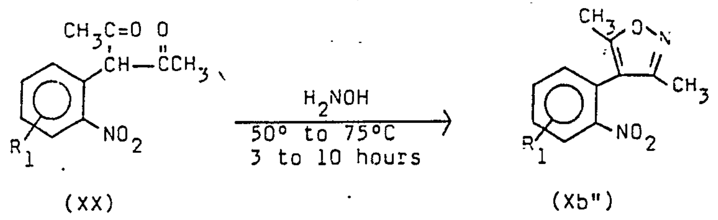
wherein

R_1 is as originally defined.

The reaction of Equation 10a is run in ethanol at 0° to about 30°C for about 48 hours. The product is isolated by addition of water and 2N HCl and extraction with methylene chloride. The reaction of Equation 10b is also run in ethanol at reflux for about 1 to 10 hours. The product is isolated by addition of water and extraction with methylene chloride. For more details refer to similar procedures described in H. Yasuda, *Yakugaku Zasshi, 79,* 623 (1959).

As shown in Equation 11 below, 3,5-dimethyl-4-(2-nitrophenyl)isoxazoles of Formula (Xb″) can be prepared by reacting a 3-(2-nitrophenyl)pentan-2,4-dione of Formula (XX) with hydroxylamine.

Equation 11



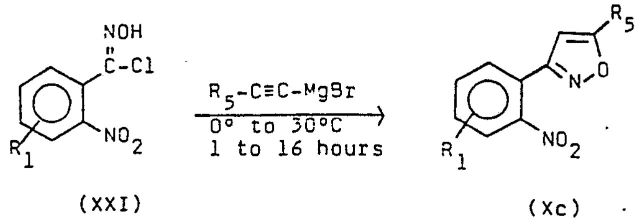
wherein

R_1 is as originally defined.

The reaction of Equation 11 is run in ethanol at 50 to 75°C for about 3 to 10 hours. The product is isolated by addition of water and extraction with methylene chloride. For more details refer to similar procedures described in Bobranski and Wojtowski, *Roczniki Chem., 38,* 1327 (1964). The starting compounds XX can be prepared by reacting an appropriate 2-halonitrobenzene with the sodium salt of pentan-2,4-dione by methods obvious to one skilled in the art.

Equation 12 below illustrates a method for preparing 3-(2-nitrophenyl)isoxazoles of Formula (Xc).

Equation 12



wherein

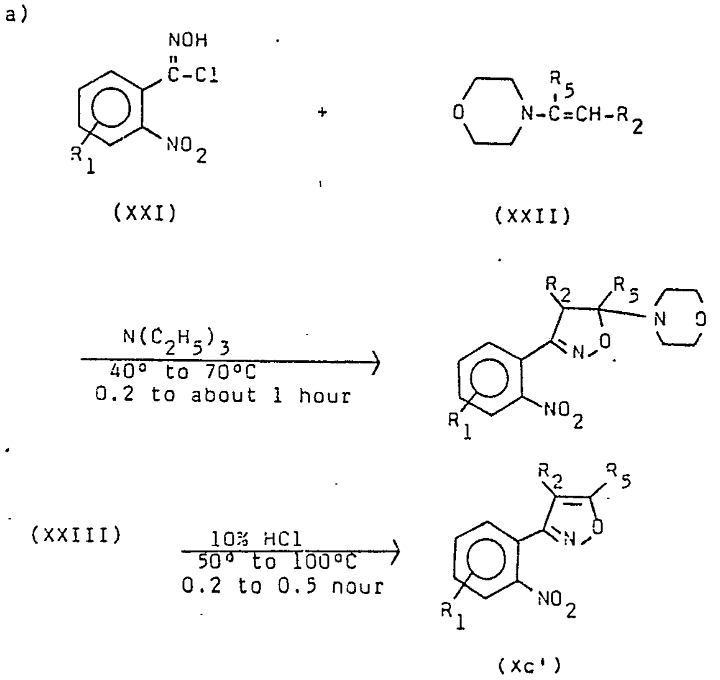
R_1 is as originally defined; and

R_5 is H, CH_3, C_2H_5, OCH_3 or OC_2H_5.

The reaction of Equation 12 is run by procedures similar to those taught by M. Langella et al., *Chim. Ind. (Milan), 47,* 996 (1965) for the preparation of 3-(2-nitrophenyl)isoxazole, and by G. Gaudiano et al., *Gazz. Chim. Ital., 89,* 2466 (1959) for the preparation of 5-ethoxy-3-(2-nitrophenyl)isoxazole. Thus, a 2-nitrophenylhydroxamic acid chloride of Formula (XXI) is reacted with an appropriate acetylenic Grignard reagent in tetrahydrofuran at 0° to 30°C for 1 to about 16 hours. The product is isolated by addition of water and ammonium chloride and extraction with methylene chloride. The acetylenic Grignard reagents are prepared from substituted acetylenes by procedures described in the cited references.

Equation 13 below illustrates a method for preparing 3-(2-nitrophenyl)isoxazoles of Formula (Xc′).

Equation 13



wherein

R_1 is as originally defined;

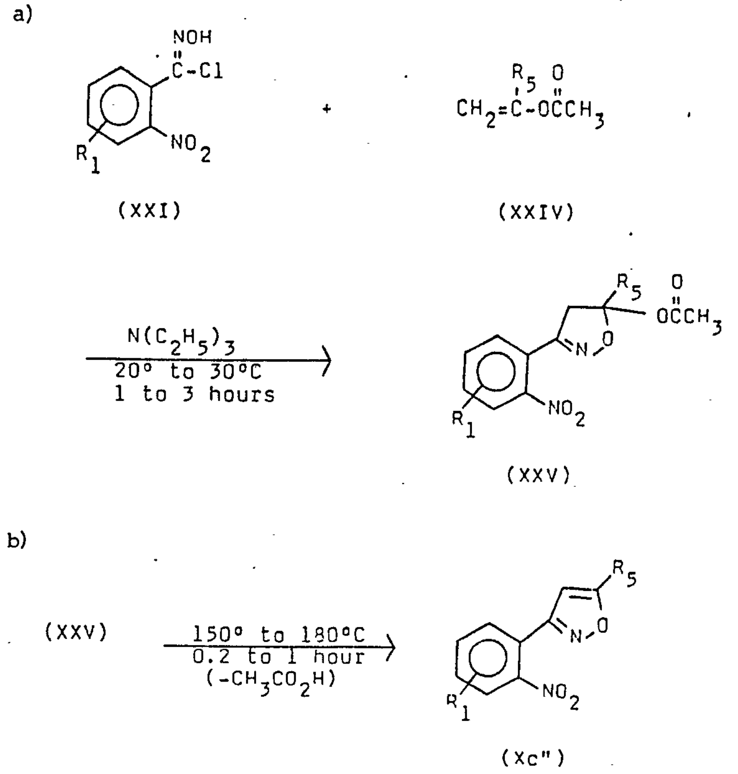
R_2 is CH_3 or C_2H_5; and

R_5 is H, CH_3 or C_2H_5.

The reactions of Equation 13 above can be run by procedures similar to those described in G. Bianchetti et al., *Gazz. Chim. Ital., 93,* 1714 (1963) for the preparation of various 3-phenylisoxazoles. Thus, in reaction 13a, a 2-nitrophenylhydroxamic acid chloride of Formula (XXI) is reacted with an equimolar amount of triethylamine and a N-alkenylmorpholine of Formula (XXII) in chloroform at reflux for 0.2 to about 1 hour to form a 5-(N-morpholinyl)-3-(2-nitrophenyl)isoxazoline of Formula (XXIII). In reaction 13b, XXIII is reacted with 10% hydrochloric acid at reflux for about 0.2 to 0.5 hour to form Xc′. The product Xc′ is isolated by extraction with methylene chloride.

Equation 4 below illustrates a method for preparing 3-(2-nitrophenyl)isoxazoles of Formula (Xc″).

Equation 14



wherein

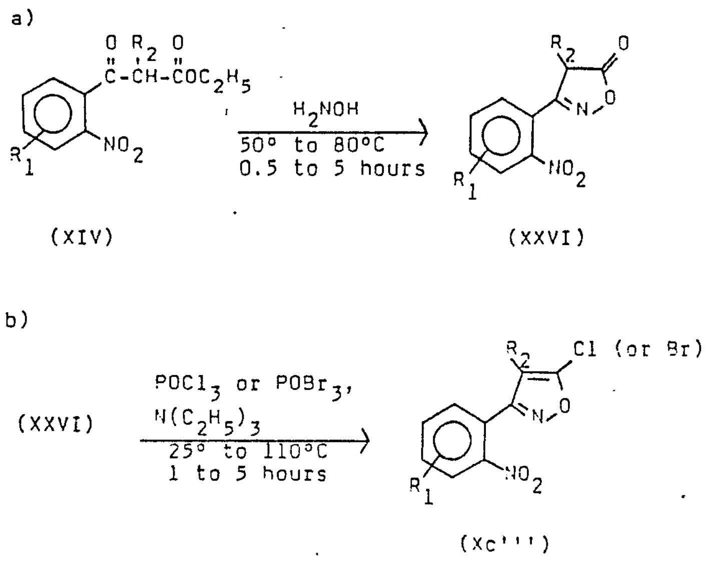
R$_1$ is as originally defined; and

R$_5$ is H, CH$_3$ or C$_2$H$_5$.

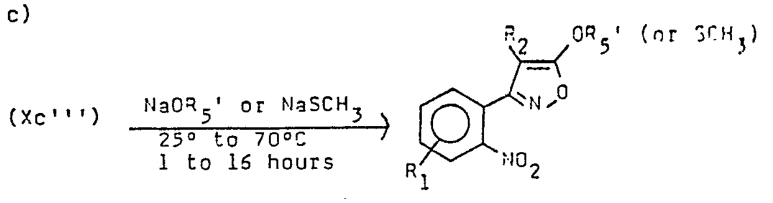
The reactions of Equation 14 above can be run by procedures similar to those described in R. Micetich, *Can. J. Chem.*, *48*, 467 (1970) for the preparation of various 3-phenylisoxazoles. Thus, in reaction 14a, a 2-nitrophenylhydroxamic acid chloride XXI is reacted with equimolar amounts of vinyl acetate of Formula (XXIV) and triethylamine in a solvent such as ether or tetrahydrofuran at about 30°C for 1 to 3 hours to form a 5-acetoxy-3-(2-nitrophenyl)isoxazoline of Formula (XXV). In reaction 14b, XXV is heated at about 150° to 180°C for a short period to form Xc″.

Equation 15 below illustrates a method for preparing 5-halo-3-(2-nitrophenyl)isoxazoles of Formula (Xc‴) and 5-alkoxy- or 5-methylthio-3-(2-nitrophenyl)isoxazoles of Formula (Xc″″).

Equation 15



c)



(Xc'''')

wherein

R_1 is as originally defined;

R_2 is H, CH_3 or C_2H_5; and

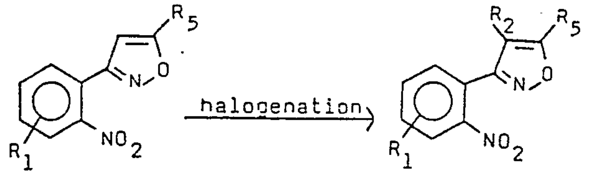
R_5' is CH_3 or C_2H_5.

The reactions of Equations 15a and 15b above can be run by procedures similar to those described in U.S. 3,781,438 for the preparation of 5-halo-3-phenylisoxazoles. Thus, in reaction 15a, an ethyl 2-(2-nitrobenzoyl)acetate, propionate or butyrate of Formula (XIV) is reacted with hydroxylamine hydrochloride and sodium acetate in ethanol at reflux for 0.5 to 5 hours to form a 3-(2-nitrophenyl)isoxazolin-5-one of Formula (XXVI). In reaction 15b, XXVI is reacted with an equimolar amount of triethylamine and excess phosphorus oxychloride or phosphorus oxybromide in toluene at 25° to 110°C for 1 to 5 hours to form Xc'''.

The reaction of Equation 15c above can be run by procedures similar to those described in J. Carr et al., *J. Med. Chem., 20,* 934 (1977) and R. Micetich et al., *Can. J. Chem., 48,* 1371 (1970). Thus, Xc''' is reacted with sodium methoxide, sodium ethoxide or sodium methylmercaptide in tetrahydrofuran at 25° to 70°C for 1 to 16 hours to form (Xc'''').

As shown in Equation 16 below, 4-halo-3-(2-nitrophenyl)isoxazoles of Formula Xc^{VI}) can be prepared by halogenating 3-(2-nitrophenyl)isoxazoles of Formula (Xc^{V}). The reaction is run using reagents and procedures described above in Equation 8. The preparation of Xc^{V} is described above in Equations 12, 14 and 15.

Equation 16



(Xc^{V}) (Xc^{VI})

wherein

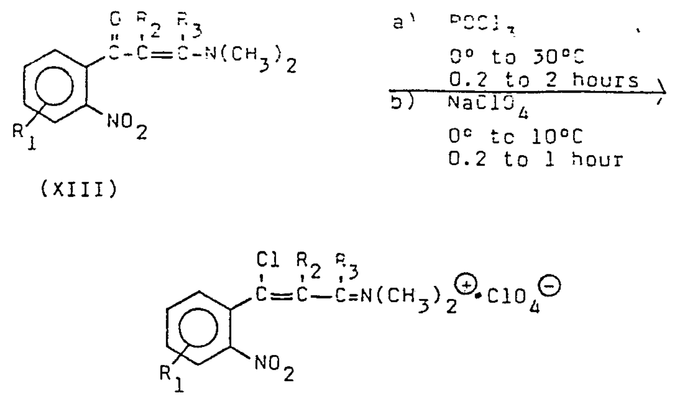
R_1 and R_5 are as originally defined; and

R_2 is Cl or Br.

The 5-(2-nitrophenyl)isothiazoles of Formula (Xd) in Equation 17 below can be prepared by methods analogous to those described in Yang-i Lin and S. A. Lang, *J. Org. Chem., 45,* 4857 (1980) for the preparation of 5-phenylisothiazole.

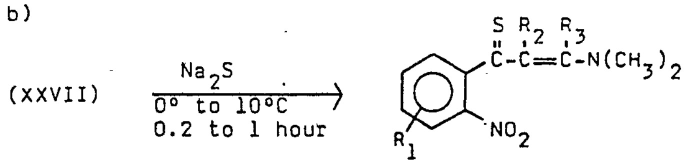
Equation 17

a)



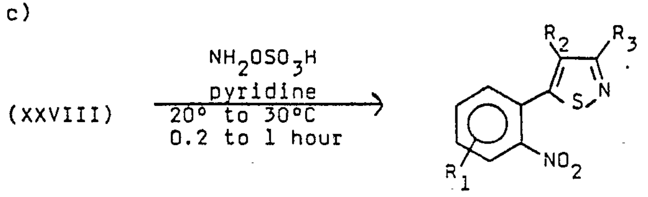
(XXVII)

b)



(XXVIII) .

c)



(Xd)

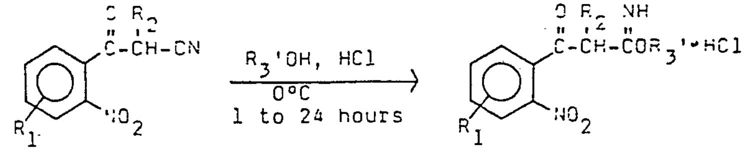
wherein
R_1 is as originally defined; and
R_2 and R_3 are H, CH_3 or C_2H_5.

According to Equation 17 above, in reaction 17a a 3-dimethylamino-1-(2-nitrophenyl)-2-propen-1-one of Formula (XIII) is reacted with phosphorus oxychloride in methylene chloride at 0° to 30°C for 0.2 to about 2 hours, followed by treatment with sodium perchlorate in water at 0° to 10°C for 0.2 to about 1 hour to form a perchlorate salt of Formula (XXVII). In reaction 17b, XXVII is reacted with sodium sulfide nonahydrate in dimethylformamide and water at 0° to 10°C for 0.2 to about 1 hour to form a 3-dimethylamino-1-(2-nitrophenyl)-2-propene-1-thione of Formula (XXVIII). And in reaction 17c, XXVIII is reacted with hydroxylamine-O-sulfonic acid (HSA) and two mole equivalents of pyridine in methanol at 20° to 30°C for 0.2 to about 1 hour to form Xd. The preparation of the starting compounds XIII is described in Equation 6 above.

3-Alkoxy-5-(2-nitrophenyl)isothiazoles of Formula (Xd′) in Equation 18 below can be prepared by methods similar to those described in *Ber., 96,* 944 (1963); German 1,193,050 and German 1,197,088. The cited references describe the preparation of other 3-alkoxy-5-phenylisothiazoles.

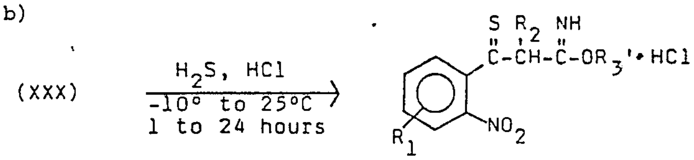
Equation 18

a)



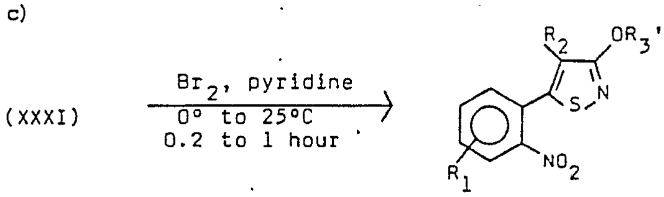
(XXIX) (XXX)

b)



(XXXI)

c)



(Xd′)

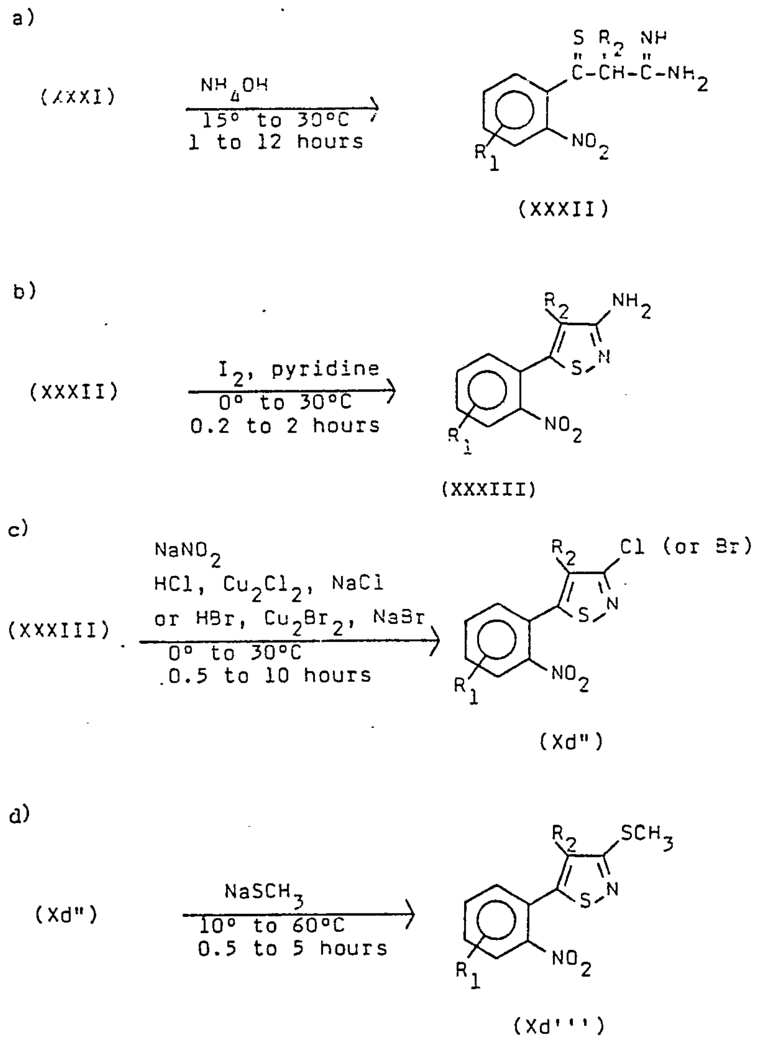
wherein
R_1 is as originally defined;
R_2 is H, CH_3 or C_2H_5; and

R_3' is CH_3 or C_2H_5.

According to Equation 18, in reaction 18a a 2′-cyano-2-nitroacetophenone of Formula (XXIX) is reacted with methanol or ethanol in a solvent such as ethyl ether or toluene saturated with hydrogen chloride gas at about 0°C for 1 to 24 hours to form a 2-nitrobenzoylacetamido alkyl ester of Formula (XXX). In reaction 18b, XXX is reacted with hydrogen sulfide in absolute methanol or ethanol saturated with hydrogen chloride gas at about −10° to 25°C for 1 to 24 hours to form a 2-nitrothiobenzoylacetamido alkyl ester of Formula (XXXI). And in reaction 18c, XXXI is reacted with bromine in ethyl acetate containing pyridine at 0° to about 25°C for about 0.2 to 1 hour to form Xd′.

Equation 19 below illustrates a method for preparing 3-halo-5-(2-nitrophenyl)isothiazoles of Formula (Xd″) and 3-methylthio-5-(2-nitrophenyl)isothiazoles of Formula (Xd‴).

Equation 19



wherein

R_1 is as originally defined; and

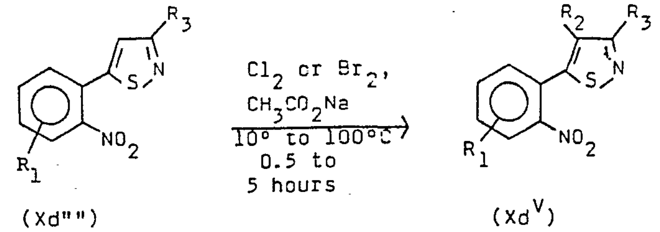
R_2 is H, CH_3 or C_2H_5.

The reactions of Equations 19a and 19b above can be run by procedures similar to those described in *Ber., 96,* 944 (1963); German 1,193,050; and German 1,197,088 for the preparation of other 3-amino-5-phenylisothiazoles. Thus, in reaction 19a, a 2-nitrothiobenzoylacetamido alkyl ester of Formula (XXXI) is reacted with aqueous 24% NH_4OH at 15° to 30°C for 1 to about 12 hours to form a 2-nitrothiobenzoylacetamidine of Formula (XXXII). In reaction 19b, XXXII is reacted with iodine in methanol containing pyridine at 0° to about 30°C for about 0.2 to 2 hours to form a 3-amino-5-(2-nitrophenyl)isothiazole of Formula (XXXIII).

In the reaction of Equation 19c above, Xd″ is prepared from XXXIII via Sandmeyer reactions, according to the teachings of J. Goerdeler and M. Roegler, *Chem. Ber., 103,* 112 (1970). Thus, XXXIII is reacted with sodium nitrite in concentrated HCl containing Cu_2Cl_2 and NaCl or in concentrated HBr containing Cu_2Br_2 and NaBr at 0° to 30°C for 0.5 to 10 hours to give Xd″. And in reaction 19d, Xd″ is reacted with sodium methylmercaptide in tetrahydrofuran at 10° to 60°C for 0.5 to 5 hours to form Xd‴. The product is isolated by addition of water and extraction with methylene chloride.

The 4-halo-5-(2-nitrophenyl)isothiazoles of Formula Xd^V) in Equation 20 below are prepared by halogenating 5-(2-nitrophenyl)isothiazoles of Formula (Xd'''') with chlorine or bromine in the presence of a base such as sodium acetate. The reaction is run in acetic acid at 10° to 100°C for 0.5 to 5 hours. For more details refer to similar procedures described in the art for halogenating other 5-phenylisothiazoles, e.g., D. Buttimore et al., *J. Chem. Soc.,* 2032 (1963); T. Naito et al., *Chem. Pharm. Bull., 16,* 148 (1968); and J. Goerdeler and W. Mittler, *Ber., 96,* 944 (1963).

Equation 20



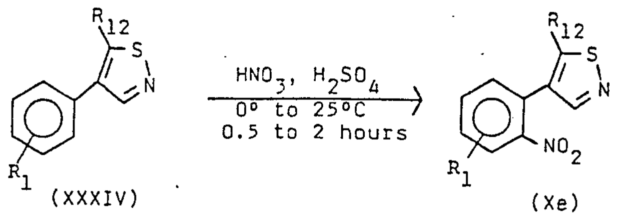
wherein

R_1 and R_3 are as originally defined; and

R_2 is Cl or Br.

The 4-(2-nitrophenyl)isothiazoles of Formula (Xe) in Equation 21 below can be prepared by nitrating 4-phenylisothiazoles of Formula (XXXIV) with concentrated nitric acid in concentrated sulfuric acid, according to the teachings of J. H. Finley and G. P. Volpp, *J. Heterocycl. Chem., 6,* 841 (1969).

Equation 21



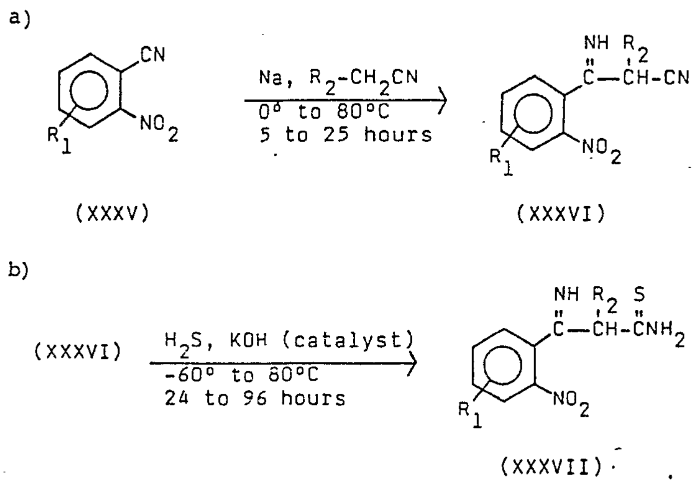
wherein

R_{12} is H or CH_3.

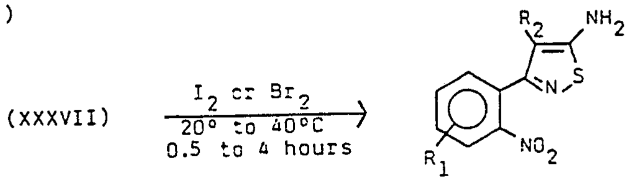
The reaction of Equation 21 above is run at 0° to 25°C for 0.5 to 2 hours. Following usual work-up the product Xe is purified by column chromatography on silica gel. The starting compounds XXXIV can be prepared by known methods. Several such methods are described in M. Muehlstaedt, *J. Prakt. Chem., 318,* 507 (1976); M. Ohashi et al., *J. Chem. Soc.,* 1148 (1970); R. A. Olofson et al., *Tetrahedron, 22,* 2119 (1966); and F. Huebenett et al., *Angew Chem., 75,* 1189 (1963).

As shown in Equation 22 below, the 3-(2-nitrophenyl)isothiazoles of Formula (Xf) and (Xf') can be prepared by a series of procedures starting from a 2-nitrobenzonitrile of Formula (XXXV).

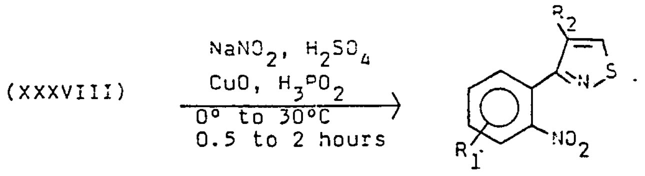
Equation 22



c)

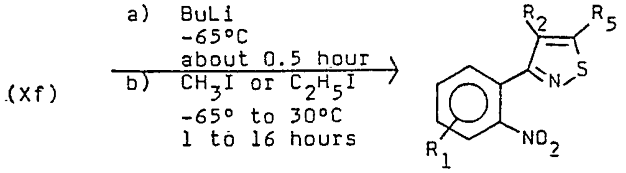


d)



(Xf)

e)



(Xf')

wherein

R_1 is as originally defined; and

R_2 and R_5 are H, CH_3 or C_2H_5.

The reactions of Equation 22 above can be run by procedures known in the art. Thus, in reaction 22a, 2-nitrobenzonitrile XXXV can be reacted with an appropriate alkyl nitrile and sodium metal in a solvent such as ether or toluene at 0° to 80°C for about 5 to 25 hours to form a 2-imino-2-(2-nitrophenyl)propionitrile of Formula (XXXVI), according to the teachings of U.S. 3,479,365; Netherlands 6,608,094; and T. Naito et al., *Bull. Chem. Soc. Japan, 41,* 965 (1968).

In the reaction of Equation 22b, XXXVI can be reacted with hydrogen sulfide and potassium hydroxide catalyst in methylene chloride at −60° to 80°C in a sealed tube for 24 to 96 hours to form a 2-imino-2-(2-nitrophenyl)thiopropionamide of Formula (XXXVII), according to the teachings of T. Naito et al., *Chem. Pharm. Bull., 16,* 148 (1968) and J. Goerdeler and H. Pohland, *Chem. Ber., 94,* 2950 (1961).

In the reaction of Equation 22c above, XXXVII can be cyclized by reaction with iodine or bromine in a solvent such as ether, chloroform or ethanol containing potassium carbonate at 20° to 40°C for 0.5 to 4 hours to form a 5-amino-3-(2-nitrophenyl)isothiazole of Formula (XXXVIII), according to the teachings of *ibid.,* Netherlands 6,608,094 and J. Goerdeler and H. Pohland, *Angew Chem., 72,* 77 (1962).

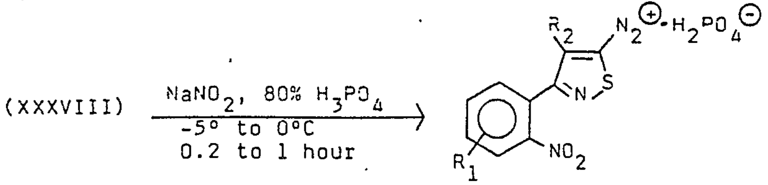
In the reaction of Equation 22d above, a diazonium salt, prepared from XXXVIII and sodium nitrite in sulfuric acid at 0°C for 0.5 hour, can be reacted with cuprous oxide and 50% hypophosphorous acid at 0° to 30°C for about 2 hours to form a 3-(2-nitrophenyl)isothiazole of Formula (Xf), according to the teachings of M. Beringer et al., *Helv. Chim. Acta., 49,* 2466 (1966).

And in the reaction of Equation 22e above, Xf can be reacted with butyl lithium in tetrahydrofuran at −65°C for about 0.5 hour to form a 5-lithio-3-(2-nitrophenyl)isothiazole reagent, according to the teachings of T. Naito et al., *Chem. Pharm. Bull., 16,* 148 (1968). Subsequent reaction of this reagent with methyl or ethyl iodide, at −65° to 30°C for 1 to 16 hours, can provide Xf', according to the teachings of *ibid.*

The 5-halo-3-(2-nitrophenyl)isothiazoles of Formula (Xf") in Equation 23 below can be prepared from 5-amino-3-(2-nitrophenyl)isothiazoles of Formula (XXXVIII) by Sandmeyer reactions.

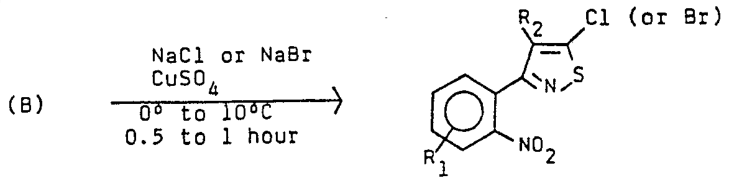
Equation 23

a)



(B)

b)



(Xf")

wherein

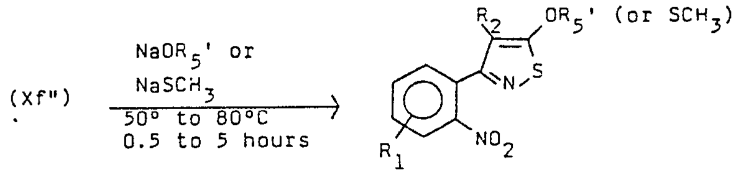
R_1 is as originally defined; and

R_2 is H, CH_3 or C_2H_5.

The reactions of Equation 23 above can be run by procedures similar to those described in J. Goerdeler and H. Pohland, *Chem. Ber., 94,* 2950 (1961) for the preparation of 5-chloro-3-phenylisothiazole. Thus, in reaction 23a, XXXVIII is diazotized with sodium nitrite in 80% phosphoric acid at −5° to 0°C for about 0.5 hour. In reaction 23b, the diazonium salt *B* is reacted with sodium chloride or sodium bromide and copper sulfate catalyst at 0° to 10°C for about 1 hour to form Xf". The preparation of XXXVIII is described in Equation 22 above.

The 5-alkoxy- and 5-methylthio-3-(2-nitrophenyl)isothiazoles of Formula (Xf''') in Equation 24 below are prepared by reacting a 5-halo-3-(2-nitrophenyl)isothiazole of Formula (Xf") with sodium methoxide, sodium ethoxide or sodium methylmercaptide.

Equation 24



(Xf''')

wherein

R_1 is as originally defined;

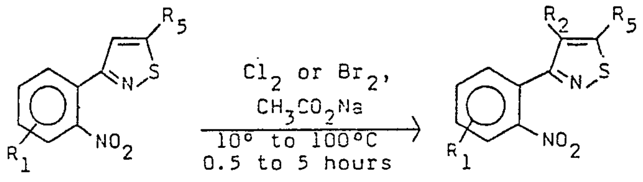
R_2 is H, CH_3 or C_2H_5; and

R_5' is CH_3 or C_2H_5.

The reaction of Equation 24 is run in a solvent such as methanol, ethanol or tetrahydrofuran at reflux for about 0.5 to 5 hours. The product is isolated by evaporation of solvent, addition of water and filtration. The reaction of 5-haloisothiazoles with alkoxides or thioalkoxides to form 5-alkoxy- or 5-alkylthioisothiazoles is known in the art, e.g., K. R. H. Wooldrige, *Adv. in Heterocycl. Chem., 14,* p. 24 (1972).

The 4-halo-3-(2-nitrophenyl)isothiazoles of Formula (Xf^V) in Equation 24 below are prepared by halogenating 3-(2-nitrophenyl)isothiazoles of Formula (Xf'''') according to procedures described above in Equation 20. The preparation of Xf'''' is described above in Equations 22-24.

Equation 25



(Xf'''') (Xf^V)

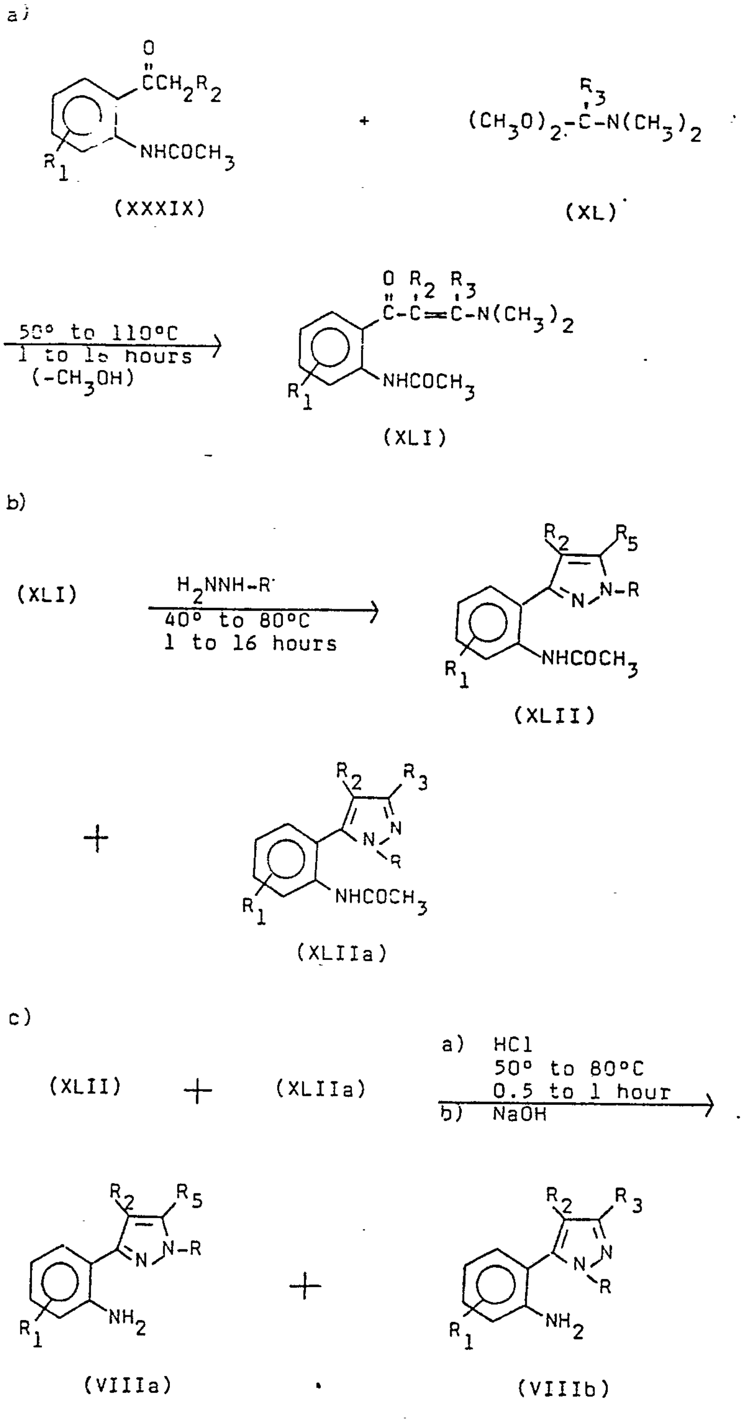
wherein

R_1 and R_5 are as originally defined; and

R_2 is Cl or Br.

Equation 26 below illustrates a method for preparing 3-(2-aminophenyl)-1H-pyrazoles of Formula (VIIIa) and 5-(2-aminophenyl)-1H-pyrazoles of Formula (VIIIb).

Equation 26



wherein

R and R_1 are as originally defined; and

R_2, R_3 and R_5 are H, CH_3 or C_2H_5.

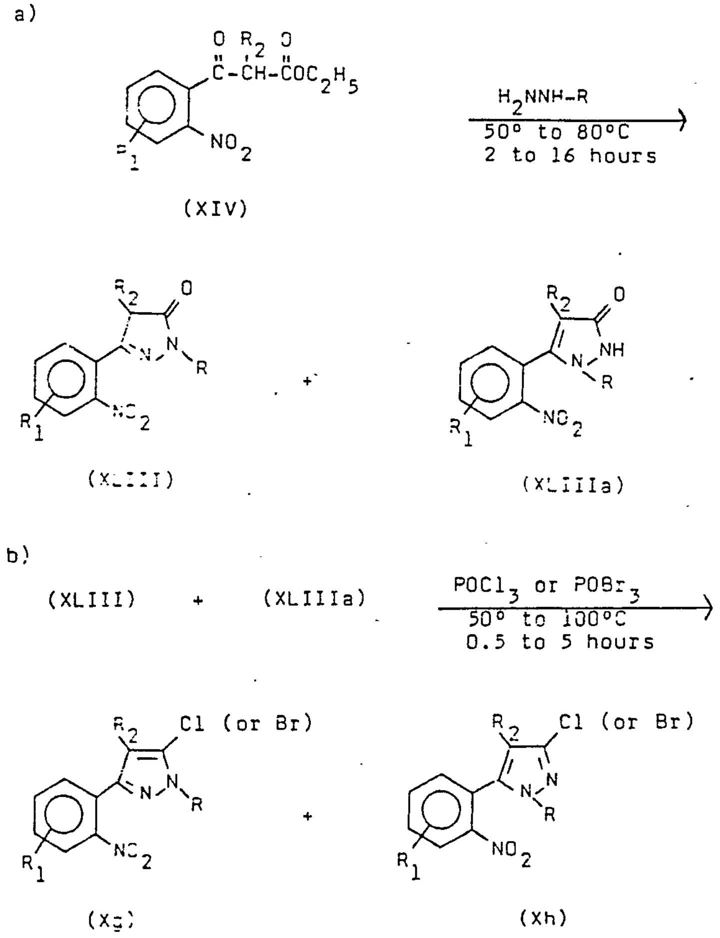
According to Equation 26 above, in reaction 26a, a 2-acetamidophenyl alkyl ketone of Formula (XXXIX) is reacted with a dimethylalkanamide dimethyl acetal of Formula (XL) to form a 3-dimethylamino-1-(2-acetamidophenyl)-2-propen-1-one of Formula (XLI). The reaction can be run by procedures described above for the reaction of Equation 6a.

In the reaction of Equation 26b, XLI is reacted with an appropriate hydrazine to form a mixture containing 3-(2-acetamidophenyl)-1H-pyrazole of Formula (XLII) and 5-(2-acetamidophenyl)-1H-pyrazole of Formula (XLIIa). The reaction is run in ethanol at reflux for 1 to 16 hours. The product mixture is isolated by evaporation of the solvent.

And in the reaction of Equation 26c, amines VIIIa and VIIIb are obtained by acid hydrolysis of acetamides XLII and XLIIa in the following manner. A mixture containing XLII and XLIIa in concentrated hydrochloric acid is heated at reflux for about 1 hour, cooled and filtered. The solid, composed of hydrochloride salts of VIIIa and VIIIb, is neutralized in water with 50% NaOH. A mixture containing amines VIIIa and VIIIb is isolated by extraction with methylene chloride. Amines VIIIa and VIIIb may be separated by high pressure liquid chromatography by one skilled in the art. More preferably, the mixture is reacted directly by procedures described in Equation 4 and 1 or 2 above to provide corresponding compounds I of the invention as a mixture.

Equation 27 below illustrates a method for preparing 5-halo-3-(2-nitrophenyl)-1H-pyrazoles of Formula (Xg) and 3-halo-5-(2-nitrophenyl)-1H-pyrazoles of Formula (Xh).

Equation 27



wherein

R is C_1-C_3 alkyl;

R_1 is as originally defined; and

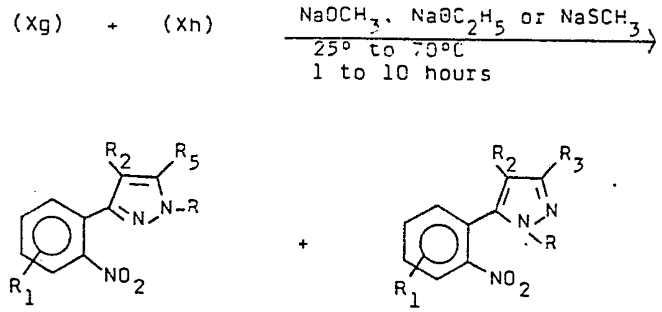
R_2 is H, CH_3 or C_2H_5.

According to Equation 27 above, in reaction 27a, an ethyl 3-(2-nitrophenyl)-3-oxopropanoate of Formula (XIV) is reacted with an alkylhydrazine to form a mixture containing a 3-(2-nitrophenyl)pyrazolin-5-one of Formula (XLIII) and a 5-(2-nitrophenyl)pyrazolin-3-one of Formula (XLIIIa). The reaction is run in ethanol at reflux for 2 to 16 hours. The product is isolated by addition of water and extraction with methylene chloride.

And in the reaction of Equation 27b, the mixture containing XLIII and XLIIIa is reacted with phosphorus oxychloride or phosphorus oxybromide to form a mixture containing Xg and Xh. The reaction is run in toluene at 50° to 100°C for 0.5 to 5 hours. The product mixture is isolated by evaporation of the solvent and may be purified by column chromatography on silica gel. The mixture may be separated by high pressure liquid chromatography by one skilled in the art.

Equation 28 below illustrates a method for preparing 5-alkoxy- and 5-methylthio-3-(2-nitrophenyl)-1H-pyrazoles of Formula (Xg′) and 3-alkoxy- and 3-methylthio-5-(2-nitrophenyl)-1H-pyrazoles of Formula (Xh′).

Equation 28



(Xg′) (Xh′)

wherein

R is C_1-C_3 alkyl;

R_1 is as originally defined;

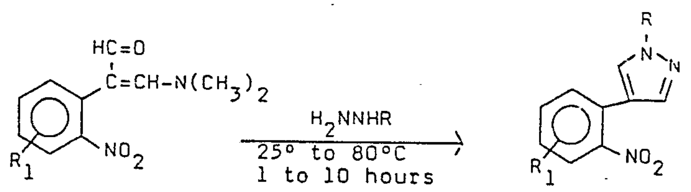
R_2 and H, CH_3 or C_2H_5; and

R_3 and R_5 are OCH_3, OC_2H_5 or SCH_3.

According to Equation 28 above, a mixture containing a 5-halo-3-(2-nitrophenyl-1H-pyrazole of Formula (Xg) and a 3-halo-5-(2-nitrophenyl)-1H-pyrazole of Formula (Xh) is reacted with sodium methoxide, sodium ethoxide or sodium methylmercaptide to form a mixture containing Xg′ and Xh′. The reaction is run in a solvent such as methanol or tetrahydrofuran at 25° to 70°C for 1 to 10 hours. The product mixture is isolated by addition of water followed by extraction with methylene chloride. The mixture may be purified by column chromatography on silica gel. The mixture may be separated by high pressure liquid chromatography by one skilled in the art.

As shown in Equation 29 below, 4-(2-nitrophenyl)-1H-pyrazoles of Formula (Xi) are prepared by reacting a 3-dimethylamino-2-(nitrophenyl)acrolein of Formula (XVII) with an appropriate hydrazine.

Equation 29



(XVII) (Xi)

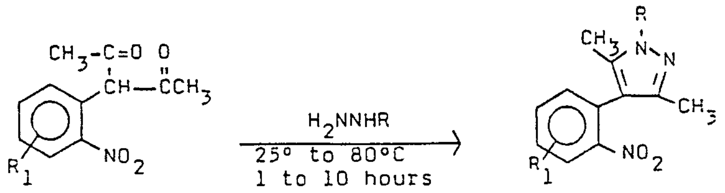
wherein

R and R_1 are as originally defined.

The reaction of Equation 29 above is run in ethanol at 25° to 80°C for 1 to 10 hours. The product is isolated by evaporation of the solvent and purified by recrystallization procedures.

As shown in Equation 30 below, a 3,5-dimethyl-4-(2-nitrophenyl)-1H-pyrazole of Formula (Xi′) is prepared by reacting a 3-(2-nitrophenyl)pentan-2,4-dione of Formula (XX) with an appropriate hydrazine. The reaction can be run by procedures described above in Equation 29.

Equation 30



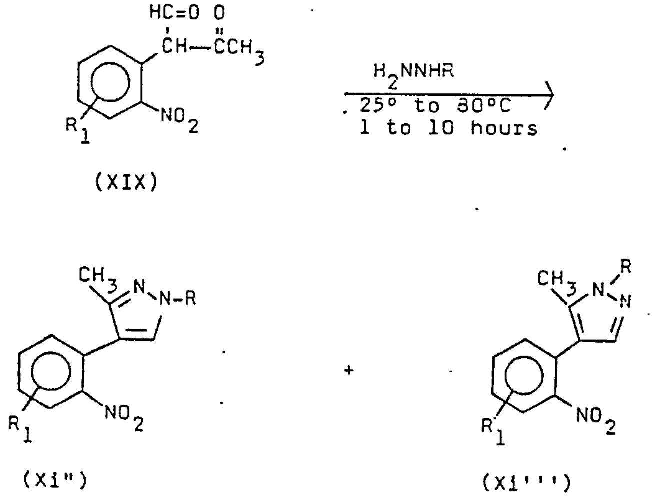
(XX) (Xi′)

wherein

R and R_1 are as originally defined.

A mixture containing 4-(2-nitrophenyl)-1H-pyrazoles of Formula (Xi″) and (Xi‴) can be prepared by reacting a 3-oxo-2-(2-nitrophenyl)butyraldehyde of Formula (XIX) with an appropriate hydrazine, as shown in Equation 31 below.

Equation 31



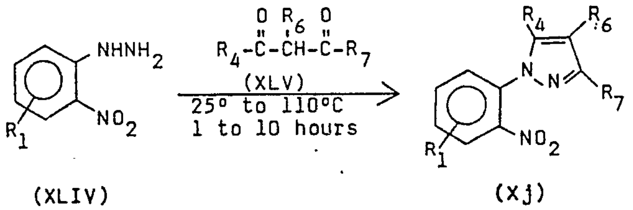
wherein

R and R_1 are as originally defined.

The reaction of Equation 31 above can be run by procedures described in Equation 29 above. The product mixture can be transformed to a mixture of corresponding compounds I of the invention by a sequence of reactions described above in Equations 5, 4 and 1 or 2, respectively.

As shown in Equation 32 below, a 1-(2-nitrophenyl)-1H-pyrazole of Formula (Xj) can be prepared by reacting a 2-nitrophenylhydrazine of Formula (XLIV) with a 1,3-diketone of Formula (XLV).

Equation 32



wherein

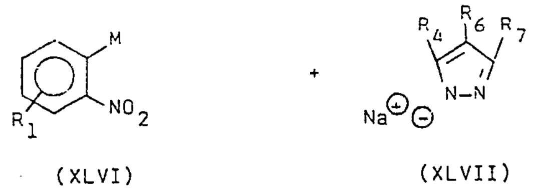
R_1 and R_6 are as originally defined; and

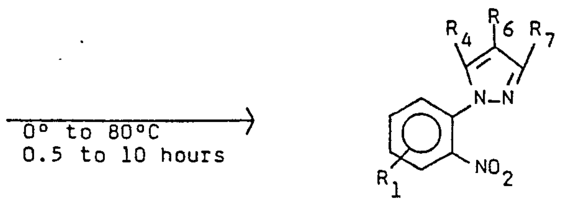
R_4 and R_7 are C_1-C_3 alkyl.

The reaction of Equation 32 is run in a solvent such as tetrahydrofuran or toluene at 25° to 110°C for 1 to 10 hours. The product is isolated by evaporation of the solvent and purified by recrystallization or chromatography procedures in the usual manner. 1-(2-Nitrophenyl)-1H-pyrazoles of Formula Xj above can also be prepared, where R_4 to R_7 are H, by reacting an appropriate 2-nitrophenylhydrazine with 1,1,3,3-tetraethoxypropane in ethanol at reflux for about 0.5 to 3 hours, according to the teachings of I. Finar and R. Hurlock, *J. Chem. Soc.,* 3024 (1957).

Another method for preparing 1-(2-nitrophenyl)-1H-pyrazoles is illustrated in Equation 33 below, where R_4 and R_7 can be H as well as C_1-C_3 alkyl.

Equation 33





(Xj')

wherein

M is Cl, Br or F; and

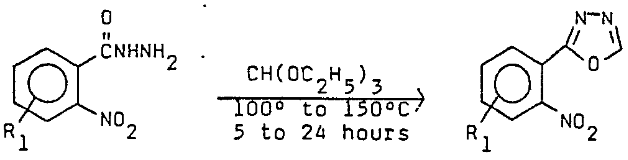
R_1 to R_7 are as originally defined.

According to Equation 33, a pyrazole sodium salt of Formula (XLVII) is reacted with a 2-halo-1-nitrobenzene of Formula (XLVI) to form Xj'. The reaction can be run in an aprotic solvent such as tetrahydrofuran or dimethylformamide at about 0° to 80°C for 0.5 to 10 hours. The product is isolated in the usual manner by addition of water and extraction with methylene chloride. The product is purified by recrystallization or chromatography procedures. The sodium salt XLVII is formed by reacting an appropriate pyrazole with sodium hydride *in situ* by methods known in the art.

Many 1-(2-nitrophenyl)-1H-pyrazoles of Formula (Xj') above can also be prepared by the Ullmann reaction, according to the teachings of M. Khan and J. Polya, *J. Chem. Soc. C.,* 85 (1970). This requires the reaction of a 2-halonitrobenzene, such as XLVI above, with an appropriately substituted pyrazole, copper (II) oxide catalyst and potassium carbonate in pyridine at reflux for 0.5 to several hours. The product is purified by column chromatography.

The 2-(2-nitrophenyl)-1,3,4-oxadiazoles of Formula (Xk) in Equation 34 below can be prepared by reacting a 2-nitrobenzhydrazide of Formula (XLVIII) with excess triethylorthoformate at 100° to 150°C for 5 to 24 hours, according to the procedures described in U.S. 3,808,223.

Equation 34



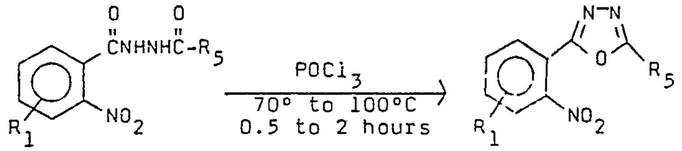
(XLVIII) (Xk)

wherein

R_1 is as originally defined.

The 2-alkyl-5-(2-nitrophenyl)-1,3,4-oxadiazoles of Formula (Xk') in Equation 35 below can be prepared by heating a 2-nitrobenzhydrazide of Formula (XLIX) in excess phosphorus oxychloride at 70° to 100°C for 0.5 to 2 hours, according to procedures described in *ibid.*

Equation 35



(XLIX) (Xk')

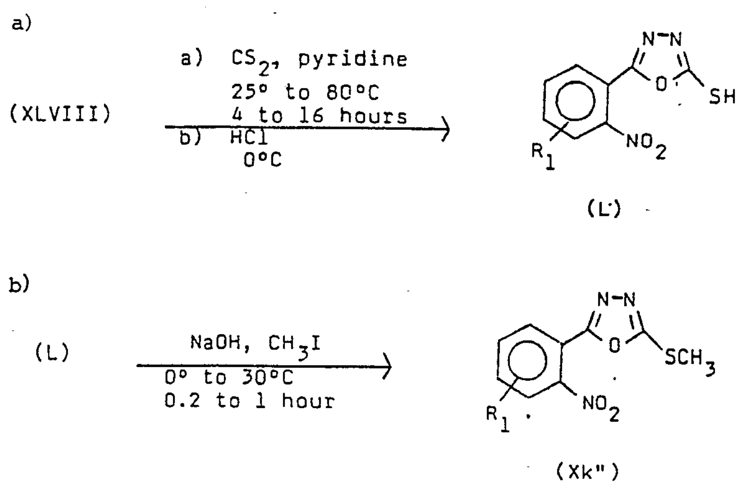
wherein

R_1 is as originally defined; and

R_5 is CH_3 or C_2H_5.

Equation 36 below illustrates a method for preparing 2-methylthio-5-(2-nitrophenyl)-1,3,4-oxadiazoles of Formula (Xk'').

Equation 36

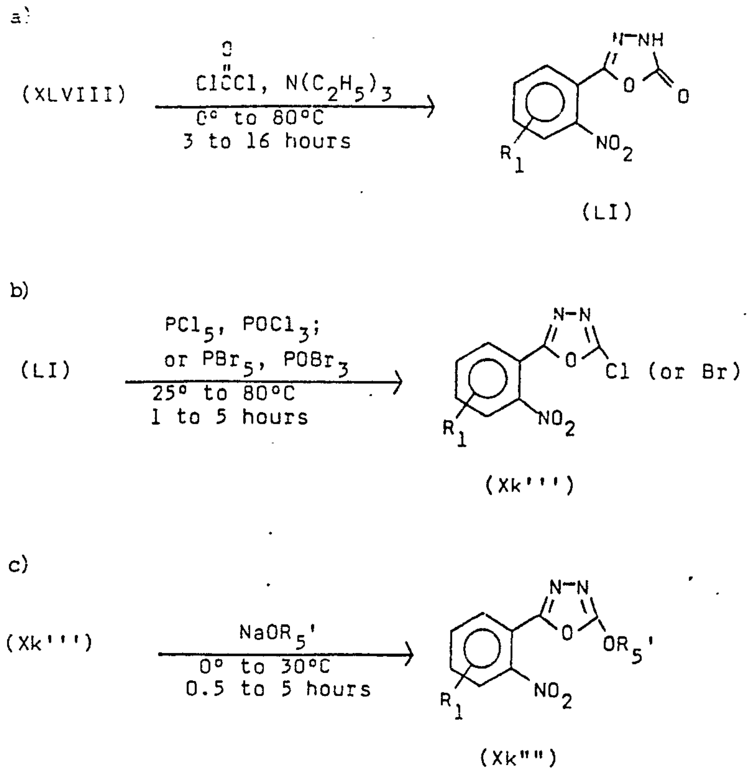


wherein
R_1 is as originally defined.

The reactions of Equation 36 above can be run according to similar procedures described in E. Hoggarth, *J. Chem. Soc.,* 4811 (1952). Thus, in reaction 36a, 2-nitrobenzhydrazide XLVIII is reacted with carbon disulfide in pyridine solvent at 25° to 80°C for about 4 to 16 hours, followed by addition of water and acidification with hydrochloric acid to form a 2-mercapto-5-(2-nitrophenyl)-1,3,4-oxadiazole of Formula (L). In reaction 36b, L is reacted with sodium hydroxide and methyl iodide in water at 0° to 30°C for 0.2 to 1 hour to form Xk''.

Equation 37 below illustrates a method for preparing 2-halo-5-(2-nitrophenyl)-1,3,4-oxadiazoles of Formula (Xk''') and 2-alkoxy-5-(2-nitrophenyl)-1,3,4-oxadiazoles of Formula (Xk'''').

Equation 37



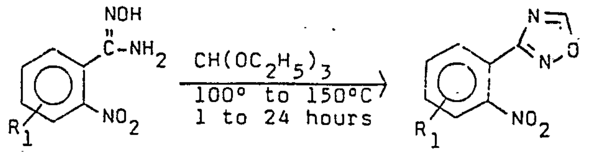
wherein
R_1 is as originally defined; and

R_5' is CH_3 or C_2H_5.

The reactions of Equation 37 above can be run according to similar procedures described in U.S. 4,259,104; Golfier and Milcent, *J. Heterocycl. Chem., 10,* 989 (1973); and R. Madhavan and V. Srinivasan, *Indian J. Chem., 7,* 760 (1969). Thus, in reaction 37a, a 2-nitrobenzhydrazide XLVIII is reacted with phosgene in an aprotic solvent such as benzene at ambient temperature for about 10 hours, followed by addition of two mole equivalents of triethylamine and heating at reflux for about 2 hours to form 2-(2-nitrophenyl)-1,3,4-oxadiazolin-5-one of Formula (LI). In reaction 37b, LI is reacted with phosphorus pentachloride in phosphorus oxychloride or with phosphorus pentabromide in phosphorus oxybromide at 25° to 80°C for about 5 hours to form (Xk'''). And in reaction 37c, Xk''' is reacted with sodium methoxide or sodium ethoxide in a solvent such as methanol or tetrahydrofuran at 0° to 30°C for about 0.5 to 5 hours to form Xk''''.

The 3-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula (XI) in Equation 38 below can be prepared by reacting a 2-nitrobenzamidoxime of Formula (LII) with excess triethylorthoformate at 100° to 150°C for about 1 to 24 hours, according to the teachings of U.S. 3,910,942.

Equation 38



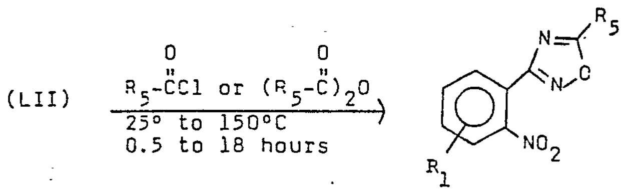
(LII) (XI)

wherein

R_1 is as originally defined.

The 5-alkyl-3-(2-nitropheny)-1,2,4-oxadiazoles of Formula (XI') in Equation 39 below can be prepared by reacting 2-nitrobenzamidoxime LII with an appropriate acid chloride in dioxane, with $BF_3 \cdot (C_2H_5)_2O$ catalyst, at 25° to 100°C for about 1 to 18 hours, according to the teachings of *ibid.*, or by reacting LII with acid chloride and pyridine in xylene at 25° to 130°C for 0.5 to 5 hours, according to the teachings of U.S. 3,270,029. Also, XI' can be prepared by reacting LII with excess anhydride at 100° to 150°C for 0.5 to 5 hours, according to the teachings of *ibid.*

Equation 39



(XI')

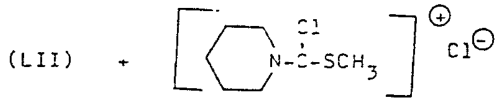
wherein

R_1 is as originally defined; and

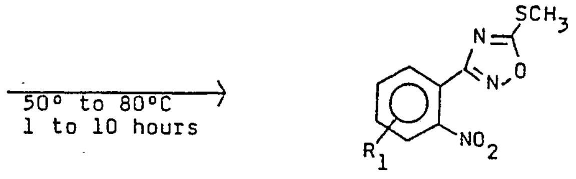
R_5 is CH_3 or C_2H_5.

The 5-methylthio-3-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula (XI'') in Equation 40 below can be prepared by reacting 2-nitrobenzamidoxime LII with a N,N-pentamethylen-methylmercapto-formamide chloride of Formula (LIII) in N-methylpyrrolidinone at 50° to 80°C for about 1 to 10 hours, according to the teachings of H. Eilingsfeld and L. Moebius, *Chem. Ber., 98,* 1293 (1965).

Equation 40



(LIII)



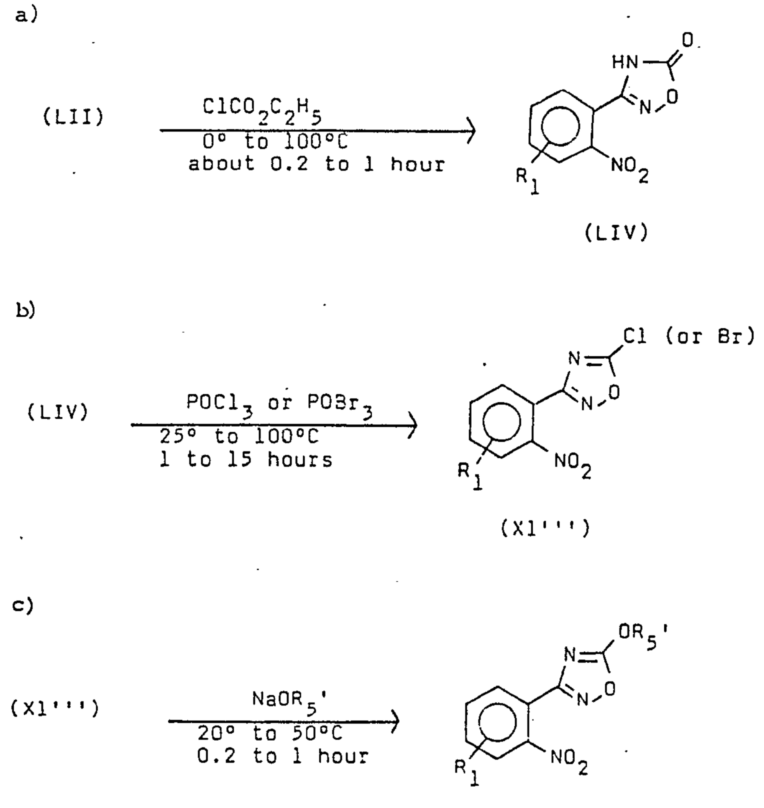
(XI")

wherein

R_1 is as originally defined.

Equation 41 below illustrates a method for preparing 5-halo-3-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula (XI''') and 5-alkoxy-3-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula (XI'''').

Equation 41



wherein

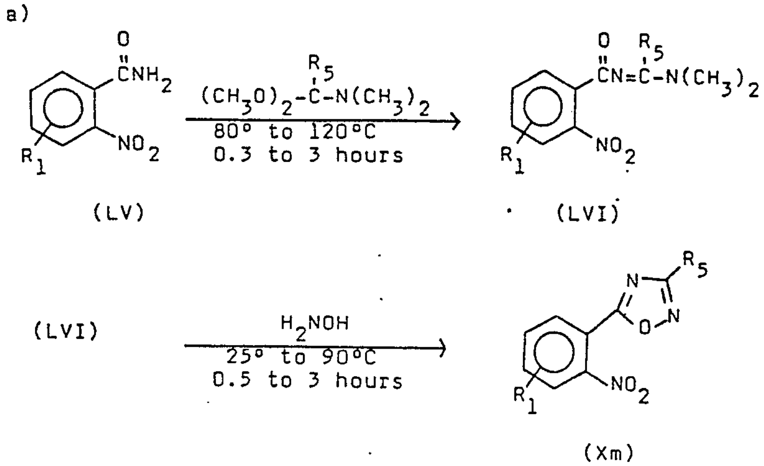
R_1 is as originally defined; and

R_5' is CH_3 or C_2H_5.

The reactions of Equation 41 above can be run by methods known in the art. Thus, in reaction 41a, 2-nitrobenzamidoxime LII is reacted with ethyl chloroformate in excess pyridine at 0° to 100°C for about 1 hour to form a 3-(2-nitrophenyl)-1,2,4-oxadiazolin-5-one of Formula (LIV), according to the teachings of A. R. Katritzky et al., *Tetrahedron, 21,* 1961 (1965). In reaction 41b, LIV is reacted with excess phosphorous oxychloride or phosphorus oxybromide with pyridine catalyst at 25° to 100°C for 1 to 15 hours to form XI''', according to the teachings of T. Fujita et al., *Yakugaku Zasshi, 84,* 1061 (1964). And in reaction 41c, XI''' is reacted with sodium methoxide or sodium ethoxide in a solvent such as methanol, ethanol or tetrahydrofuran at 20° to 50°C for about 0.2 to 1 hour to form XI'''', according to the teachings of F. Eloy et al., *Bull. Soc. Chim. Belg., 78,* 47 (1969).

The 5-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula (Xm) in Equation 42 below are prepared according to the teachings of Y. Lin et al., *J. Org. Chem.*, *44*, 4160 (1979).

Equation 42



(LV)

(LVI)

(Xm)

wherein

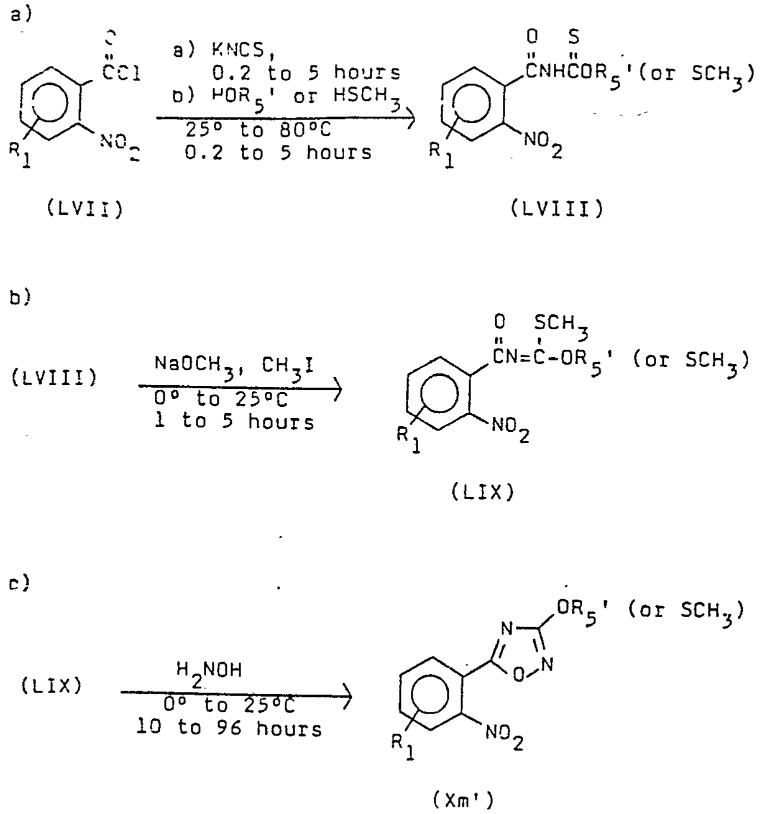
R_1 is as originally defined; and

R_5 is H, CH_3 or C_2H_5.

In the reaction of Equation 42a above, 2-nitrobenzamide LV is reacted with excess dimethylalkanamide dimethyl acetal at 80° to 120°C for about 0.3 to 3 hours to form a N-[(dimethylamino)methylene]benzamide of Formula (LVI). In reaction 42b, LVI is reacted with hydroxylamine in aqueous dioxane-acetic acid at 25° to 90°C for 0.5 to 3 hours to form Xm.

The 3-alkoxy- and 3-methylthio-5-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula (Xm′) in Equation 43 below are prepared according to the teachings of B. Nash et al., *J. Chem. Soc.*, 2794 (1969).

Equation 43



(LVII)

(LVIII)

(LIX)

(Xm′)

wherein

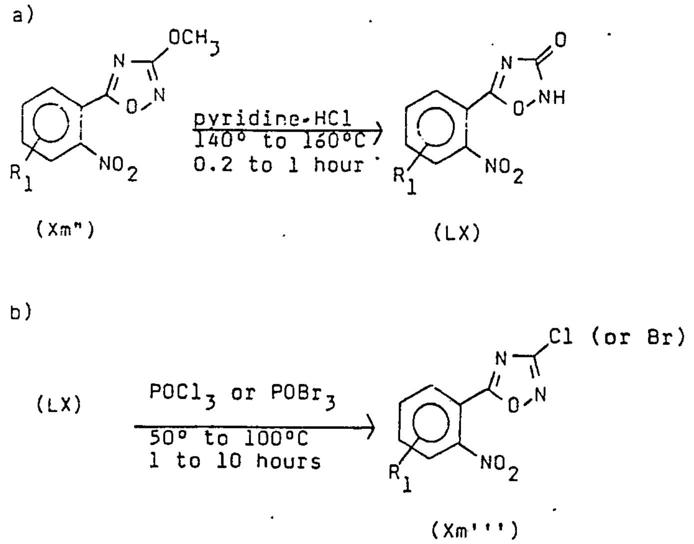
R_1 is as originally defined; and

R_5' is CH_3 or C_2H_5.

Thus, in the reaction of Equation 43a, 2-nitrobenzoyl chloride LVII is reacted with potassium thiocyanate in a solvent such as toluene or acetonitrile at reflux for 0.2 to 5 hours; the resultant benzoyl thiocyanate is then reacted with excess methanol, ethanol or methyl mercaptan at 25° to 80°C for 0.2 to 5 hours to form an O-alkyl benzoylthiocarbamate or methyl benzoyldithiocarbamate of Formula (LVIII). In reaction 43b, LVIII is reacted with sodium methoxide and methyl iodide in methanol at 0° to 25°C for 1 to 5 hours to form a dialkyl benzoyliminothiocarbonate or dimethyl benzoyliminodithiocarbonate of Formula (LIX). And in reaction 43c, LIX is reacted with hydroxylamine in methanol or ethanol at 0° to 25°C for about 10 to 96 hours to form Xm'.

Equation 44 below illustrates a method for preparing 3-halo-5-(2-nitrophenyl)-1,2,4-oxadiazoles of Formula Xm'''.

Equation 44



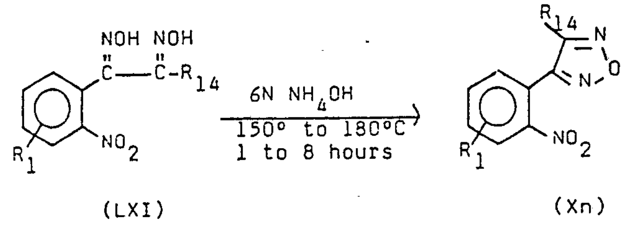
wherein

R_1 is as originally defined, except $R_1 \neq OCH_3$.

The reactions of Equation 44 can be run by methods known in the art. Thus, in reaction 44a, 3-methoxy-1,2,4-oxadiazole Xm" is demethylated by reaction with excess pyridine·HCl neat at 140° to 160°C for 0.2 to 1 hour under a nitrogen atmosphere to form a 5-(2-nitrophenyl)-1,2,4-oxadiazolin-3-one of Formula (LX), according to the teachings of A. Katritzky et al., *Tetrahedron, 21,* 1961 (1965). In reaction 44b, LX is reacted with phosphorus oxychloride or phosphorus oxybromide, with pyridine catalyst, at 50° to 100°C for 1 to 10 hours to form Xm''', according to the teachings of Eloy and Deryckere, *Bull. Soc. Chem. Belg., 78,* 41 (1969).

The 3-(2-nitrophenyl)-1,2,5-oxadiazoles of Formula (Xn) in Equation 45 below can be prepared by heating a 2-nitrophenylglyoxime of Formula (LXI) with 6N NH_4OH in an autoclave at 150-180°C for 1 to 8 hours, according to the teachings of M. Milone, *Gazz. Chim. Ital., 63,* 456 (1933).

Equation 45



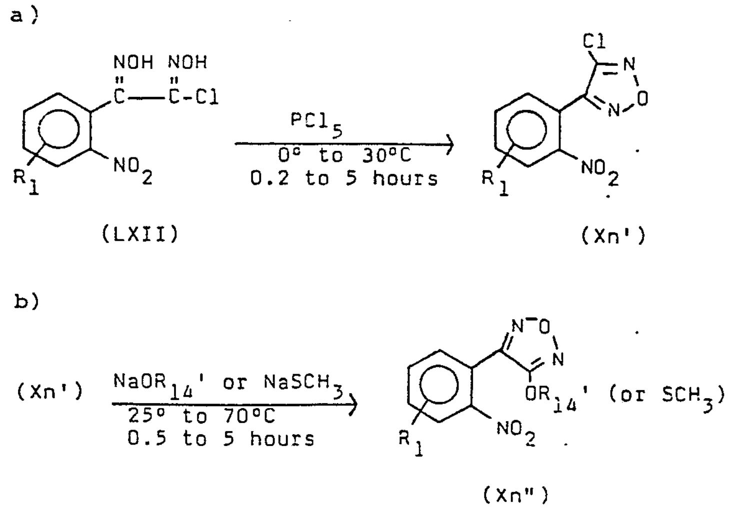
wherein

R_1 is as originally defined; and

R_{14} is H, CH_3 or C_2H_5.

Equation 46 below illustrates a method for preparing 3-halo-4-(2-nitrophenyl)-1,2,5-oxadiazoles of Formula (Xn′) and 3-alkoxy- and 3-methylthio-4-(2-nitrophenyl)-1,2,5-oxadiazoles of Formula (Xn″).

Equation 46



wherein

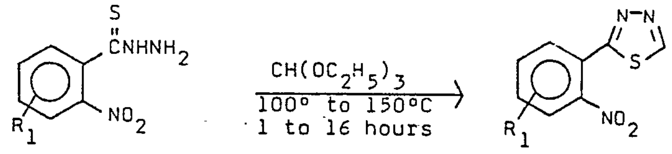
R_1 is as originally defined; and

R_{14}' is CH_3 or C_2H_5.

The reactions of Equation 46 above can be run according to similar procedures described in B. Nash et al., *J. Chem. Soc.,* 2794 (1969). Thus, in reaction 46a, a ω-chloro-(2-nitrophenyl)glyoxime of Formula (LXII) is reacted with phosphorus pentachloride in a solvent such as ether or toluene at about 0° to 30°C for 0.2 to 5 hours to form Xn′. In reaction 46b, Xn′ is reacted with sodium methoxide, sodium ethoxide or sodium methylmercaptide in a solvent such as methanol, ethanol or tetrahydrofuran at 25° to 70°C for 0.5 to 5 hours to form Xn″.

The 2-(2-nitrophenyl)-1,3,4-thiadiazoles of Formula (Xo) in Equation 47 below can be prepared by reacting 2-nitrothiobenzhydrazide LXIII with excess triethylorthoformate at reflux for 1 to 16 hours, according to the teachings of C. Ainsworth, *J. Am. Chem. Soc., 77,* 1150 (1955).

Equation 47



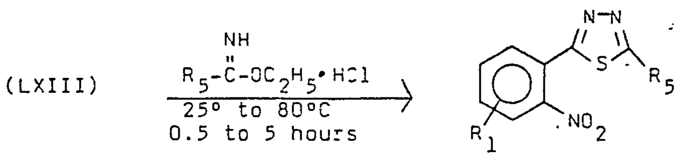
(LXIII) (Xo)

wherein

R_1 is as originally defined.

The 2-alkyl-5-(2-nitrophenyl)-1,3,4-thiadiazoles of Formula (Xo′) in Equation 48 below can be prepared by reacting 2-nitrothiobenzhydrazide LXIII with an appropriate alkylimidate ester HCl in a solvent such as ethanol at 25° to 80°C for 0.5 to 5 hours, according to the teachings of H. Weidinger and J. Kranz, *Ber., 96,* 1059 (1963).

Equation 48



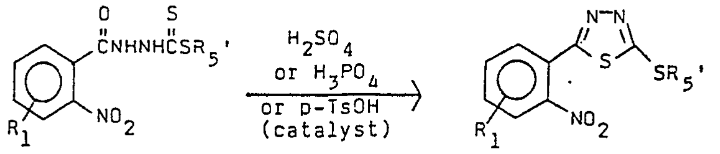
(Xo′)

wherein

R_1 is as originally defined; and
R_5 is CH_3 or C_2H_5.

The 2-methylthio-5-(2-nitrophenyl)-1,3,4-thiadiazoles of Formula (Xo") in Equation 49 below can be prepared by cyclizing a methyl 3-(2-nitrobenzoyl)dithiocarbazate of Formula (LXIV) in sulfuric acid, polyphosphoric acid, or in benzene with *p*-toluenesulfonic acid catalyst (p-TsOH), according to the teachings of R. Young and K. Wood, *J. Am. Chem. Soc., 77,* 400 (1955).

Equation 49



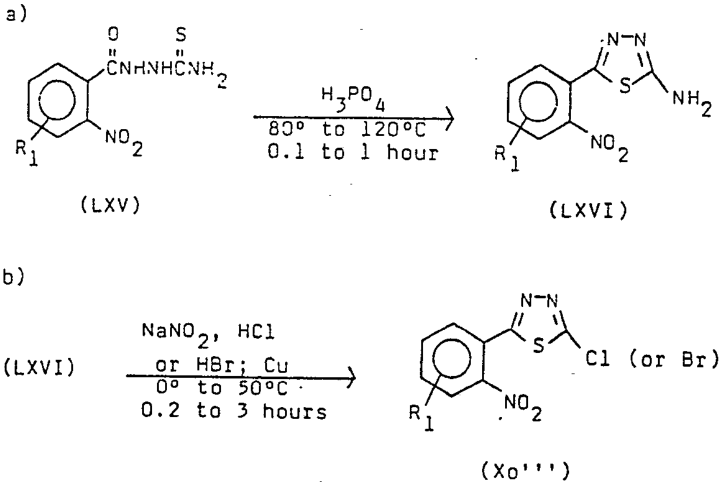
(LXIV) (Xo")

wherein
R_1 is as originally defined; and
R_5' is CH_3.

Thus, the reaction of Equation 49 above is run in concentrated sulfuric acid at 0° to 30°C for 0.1 to 0.5 hour. In polyphosphoric acid, the reaction is run at 50° to 90°C for 1 to 24 hours. And in benzene with p-TsOH catalyst, the reaction is run at reflux for 1 to 24 hours. The starting compound LXIV is prepared by alkylation with alkyl iodide of the salt formed from an appropriate 2-nitrobenzhydrazide and carbon disulfide in alcoholic potassium hydroxide, according to the method of M. Busch and M. Starke, *J. Prakt. Chem., 93,* 49 (1916).

Equation 50 below illustrates a method for preparing 2-halo-5-(2-nitrophenyl)-1,3,4-thiadiazoles of Formula (Xo‴).

Equation 50



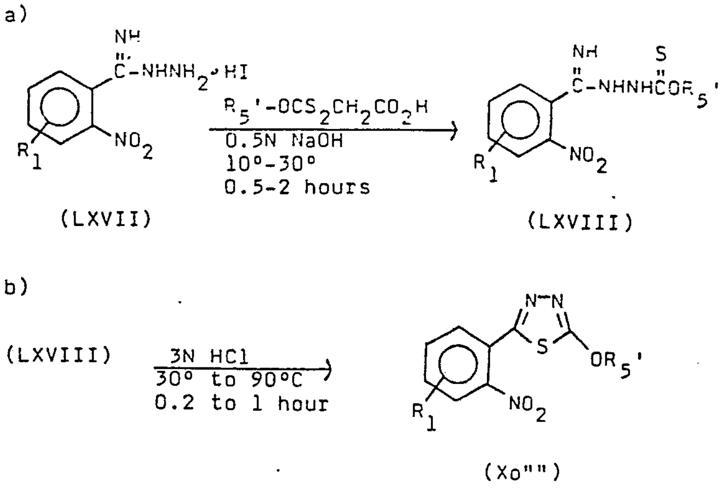
(Xo''')

wherein
R_1 is as originally defined

The reactions of Equation 50 are run by methods known in the art. Thus, in reaction 50a, LXV is cyclized in polyphosphoric acid at 80° to 120°C for about 0.1 to 1 hour to form a 2-amino-5-(2-nitrophenyl)-1,3,4-thiadiazole of Formula (LXVI), according to the teachings of E. Hoggarth, *J. Chem. Soc.,* 1163 (1949). In reaction 50b, LXVI is transformed to Xo‴ by Sandmeyer reactions according to methods described in J. Goerdeler et al., *Chem. Ber., 89,* 1534 (1956) and A. Alemagna and T. Bacchetta, *Tetrahedron, 24,* 3209 (1968). This requires reacting LXVI with sodium nitrite in hydrochloric or hydrobromic acid in the presence of copper powder at 0° to 50°C for 0.2 to 3 hours.

Equation 51 below illustrates a method for preparing 2-alkoxy-5-(2-nitrophenyl)-1,3,4-thiadiazoles of Formula (Xo'''').

Equation 51

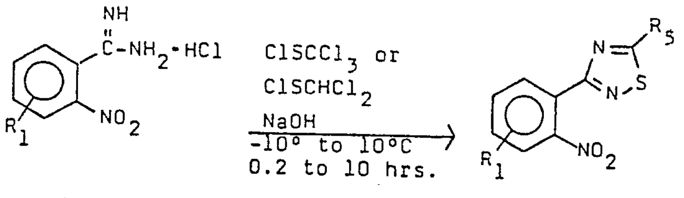


wherein
R_1 is as originally defined; and
R_5' is CH_3 or C_2H_5.

The reactions of Equation 51 above can be run according to similar procedures described in K. Doyle and F. Kurzer, *Tetrahedron, 32,* 1031 (1976). Thus, in reaction 51a, a 2-nitrobenzamidrazone·HI of Formula (LXVII) is reacted with an appropriate alkoxythiocarbonylthioacetic acid in 0.5N NaOH at 10° to 30°C for 0.5 to 2 hours to form a N-(2-nitrobenzimidoyl)-N'-alkoxythiocarbonylhydrazine of Formula (LXVIII). In reaction 51b, LXVIII is cyclized by stirring in 3N HCl at 30° to 90°C for 0.2 to 2 hours to form Xo''''. The alkoxythiocarbonylthioacetic acids are prepared by known methods, e.g., K. Jensen et al., *Acta Chemica. Scand., 23,* 1916 (1969).

Equation 52 below illustrates a method for preparing 3-(2-nitrophenyl)-1,2,4-thiadiazoles and 5-chloro-3-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (Xp).

Equation 52

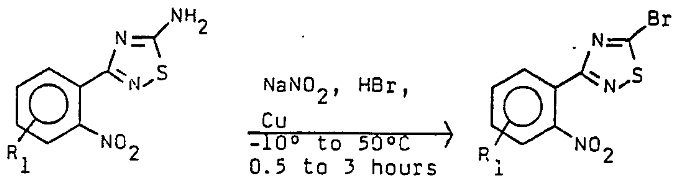


wherein
R_1 is as originally defined; and
R_5 is H or Cl.

The reactions of Equation 52 can be run according to similar procedures described in J. Goerdeler et al., *Chem. Ber., 53,* 8166 (1959); and J. Goerdeler and M. Budnowski, *Chem. Ber., 94,* 1682 (1961). Thus, a 2-nitrobenzamidine·HCl of Formula (LXIX) is reacted with perchloromethylmercaptan or dichloromethanesulfenyl chloride and sodium hydroxide in a solvent such as water-methylene chloride or aqueous dioxane at about −10° to 10°C for 0.2 to 10 hours to form Xp.

The 5-bromo-3-(2-nitrophenyl)-1,2,4,-thiadiazoles of Formula (Xp') in Equation 53 below can be prepared from 5-amino-3-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (LXX) by Sandmeyer reactions according to the teachings of J. Goerdeler et al., *Chem. Ber.,* 1534 (1956).

Equation 53



(LXX) (Xp')

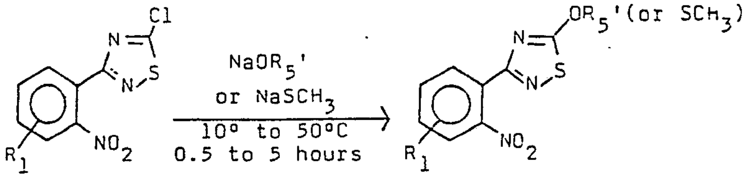
wherein

R_1 is as originally defined.

Thus, according to Equation 53, LXX is reacted with sodium nitrite in hydrobromic acid in the presence of copper powder at −10° to 50°C for 0.5 to 3 hours to form Xp′. The starting amine LXX can be prepared by reaction of 5-chloro-3-(2-nitrophenyl)-1,2,4-thiadiazoles Xp, described above in Equation 52, with ammonia by standard methods, e.g., F. Kurzer, *Adv. in Heterocycl. Chem., 5,* 159 (1965).

The 5-alkoxy- and 5-methylthio-3-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (Xp″) in Equation 54 below can be prepared by reacting 5-chloro-3-(2-nitrophenyl)-1,2,4-thiadiazole Xp with sodium methoxide, sodium ethoxide or sodium methylmercaptide in a solvent such as methanol, ethanol or tetrahydrofuran at 10° to 50°C for 0.5 to 5 hours, according to the teachings of J. Goerdeler et al., *Chem. Ber., 90,* 182 (1957).

Equation 54



(Xp) (Xp")

wherein

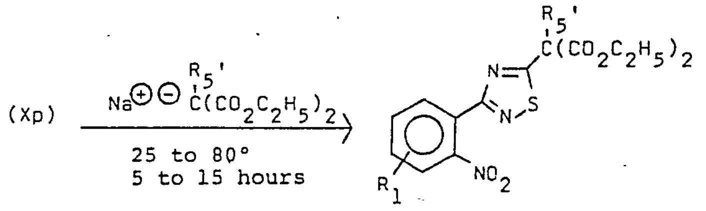
R_1 is as originally defined; and

R_5' is CH_3 or C_2H_5.

Equation 55 below illustrates a method for preparing 5-alkyl-3-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (Xp‴).

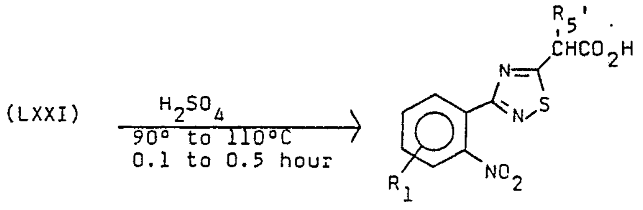
Equation 55

a)



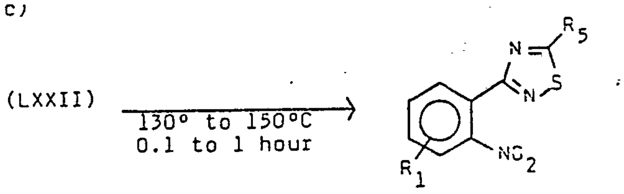
(LXXI)

b)



(LXXII)

c)



(Xp''')

wherein

R_1 is as originally defined;

R_5' is H or CH_3; and

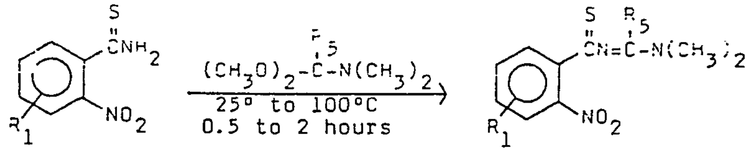
R_5 is CH_3 or C_2H_5.

The reactions of Equation 55 above can be run according to similar procedures described in G. Goerdeler and H. Hammer, *Ber., 97,* 1134 (1964). Thus, in reaction 55a, 5-chloro-3-(2-nitrophenyl)-1,2,4-thiadiazole Xp is reacted with an appropriate diethyl sodiomalonate in a solvent such as benzene or tetrahydrofuran at reflux for about 5 to 15 hours to form a 5-(substituted)-3-(2-nitrophenyl)-1,2,4-thiadiazole of Formula (LXXI). In reaction 55b, LXXI is deesterified by heating it in aqueous sulfuric acid at 90° to 110°C for about 0.1 to 0.5 hour to form a 5-carboxymethylene-3-(2-nitrophenyl)-1,2,4-thiadiazole of Formula (LXXII). And in reaction 55, LXXII is decarboxylated by heating under nitrogen at about 130° to 150°C for 0.1 to 1 hour to form Xp'''.

Equation 56 below illustrates a method for preparing 5-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (Xq).

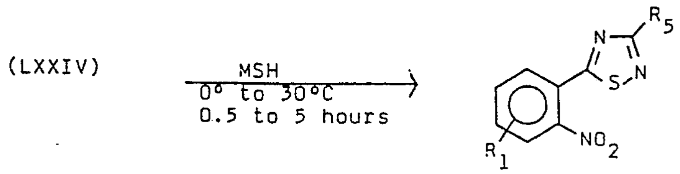
Equation 56

a)



(LXXIII) (LXXIV)

b)



(Xq)

wherein

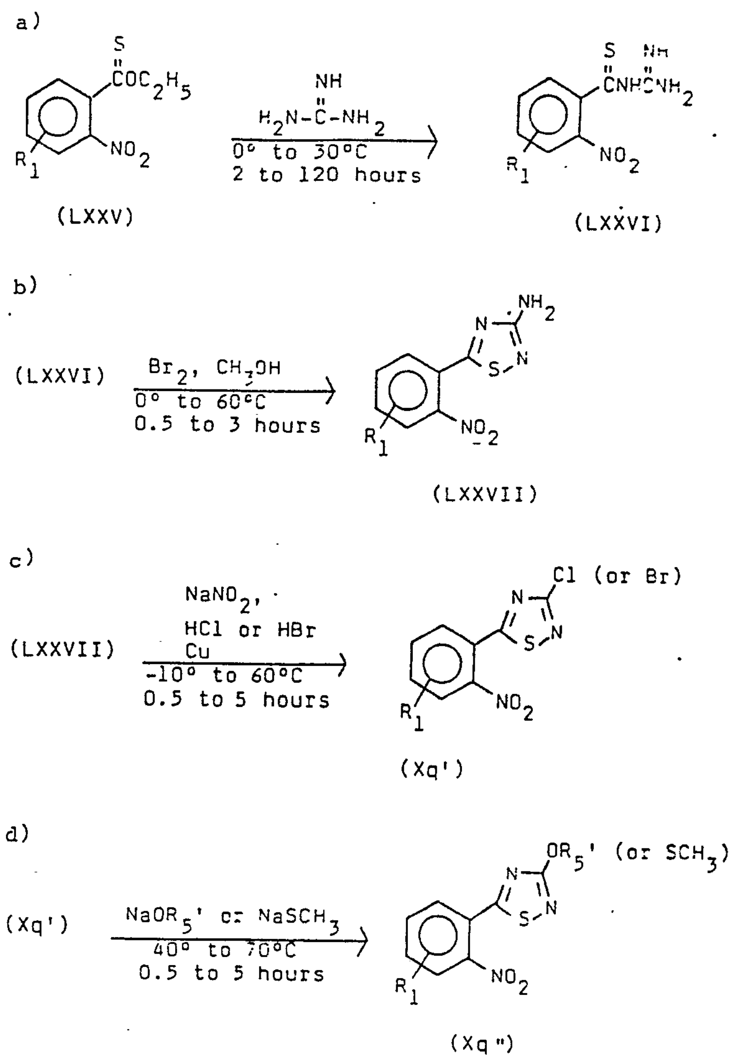
R_1 is as originally defined; and

R_5 is H, CH_3 or C_2H_5.

The reactions of Equation 56 above can be run according to similar procedures described in Yang-i Lin et al., *J. Org. Chem., 45,* 3750 (1980). Thus, in reaction 56a, a 2-nitrobenzothioamide of Formula (LXXIII) is reacted with an appropriate N,N-dimethylalkanamide dimethyl acetal at 25° to 100°C for 0.5 to 2 hours to form N-[(dimethylamino)methylene]-2-nitrobenzothioamide of Formula (LXXIV). In the second reaction, LXXIV is reacted with O-(mesitylenesulfonyl)-hydroxylamine (MSH) in a solvent such as methylene chloride at 0° to 30°C for 0.5 to 5 hours to form Xq.

Equation 57 below illustrates a method for preparing 3-halo-5-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (Xq′) and 3-alkoxy- and 3-methylthio-5-(2-nitrophenyl)-1,2,4-thiadiazoles of Formula (Xq″).

Equation 57



wherein

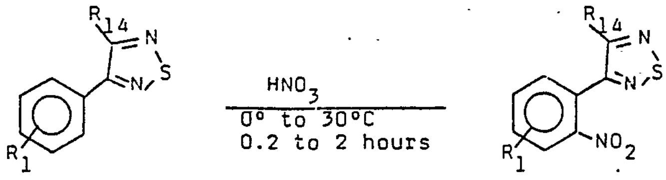
R_1 is as originally defined; and

R_5' is CH_3 or C_2H_5.

The reactions of Equation 57 above can be run by methods known in the art. Thus, in reaction 57a, a 2-nitrothiobenzoate of Formula (LXXV) is reacted with guanidine in a solvent such as ethanol or tetrahydrofuran at 0° to about 30°C for 2 to 120 hours to form a 2-nitrothiobenzoylguanidine of Formula (LXXVI), according to the teachings of J. Goerdeler and A. Fincke, *Chem. Ber., 89,* 1033 (1956). In reaction 57b, LXXVI is cyclized by reaction with bromine in methanol at 0° to 60°C for 0.5 to 3 hours to form a 3-amino-5-(2-nitrophenyl)-1,2,4-thiadiazole of Formula (LXXVII), according to the teachings of *ibid.* In reaction 57c, LXXVII is reacted with sodium nitrite in hydrochloric acid or hydrobromic acid, in the presence of copper powder, at −10° to 60°C for 0.5 to 5 hours to form Xq′ via Sandmeyer reaction, according to the teachings of F. Kurzer and S. Taylor, *J. Chem. Soc.,* 3234 (1960). And in the last reaction, Xq′ is reacted with sodium methoxide, sodium ethoxide or sodium methylmercaptide in a solvent such as methanol, ethanol or tetrahydrofuran at reflux for 0.5 to 5 hours to form Xq″, according to the teachings of *ibid.*

The 3-(2-nitrophenyl)-1,2,5-thiadiazoles of Formula (Xr) in Equation 58 below can be prepared by nitrating 3-phenyl-1,2,5-thiadiazoles of Formula (LXXVIII) with nitric acid at 0° to 30°C for 0.2 to 2 hours, according to the teachings of A. de Munno et al., *Int. J. Sulfur Chem.*, Part A, *2*, 25 (1972).

Equation 58



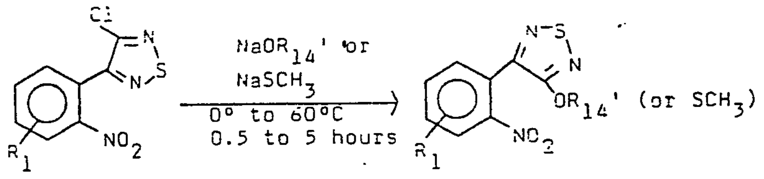
(LXXVIII) (Xr)

wherein
R_1 is as originally defined; and
R_{14} is H, Cl or CH_3.

The starting compounds LXXVIII in Equation 58 above can be prepared by known methods. Several such methods described in L. Weinstock et al., *J. Org. Chem.*, *32*, 2823 (1967); S. Mataka et al., *Synthesis*, *7*, 524 (1979); V. Bertini and P. Pino, *Agew. Chem. Internat. Edit.*, *5*, 514 (1966); and V. Bertini and P. Pino, *Corsi. Semin. Chim.*, *10*, 82 (1968).

The 3-alkoxy- and 3-methylthio-4-(2-nitrophenyl)-1,2,5-thiadiazoles of Formula (Xr') in Equation 59 below can be prepared by reacting 3-chloro-4-(2-nitrophenyl)-1,2,5-thiazoles of Formula (Xr) with sodium methoxide, sodium ethoxide or sodium methylmercaptide in a solvent such as methanol, ethanol or tetrahydrofuran at about 0° to 60°C for 0.5 to 5 hours, according to the teachings of German 1,175,683 and L. Weinstock and P. Pollak, *Adv. in Het. Chem.*, *9*, 107 (1968).

Equation 59

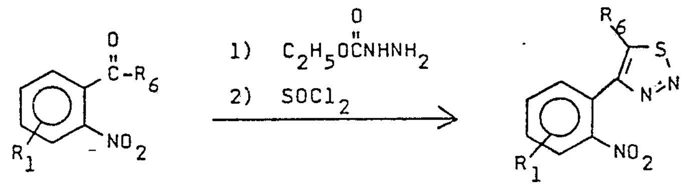


(Xr) (Xr')

wherein
R_1 is as originally defined; and
R_{14}' is CH_3 or C_2H_5.

The 4-(2-nitrophenyl)-1,2,3-thiadiazoles of Formula (Xs) in Equation 60 below can be prepared according to the teachings in U.S. 3,940,407. The method requires reacting an appropriate 2-nitrophenyl alkyl ketone with ethyl carbazate to form the corresponding hydrazide. Subsequent reaction of the hydrazide with thionyl chloride yields Xs.

Equation 60

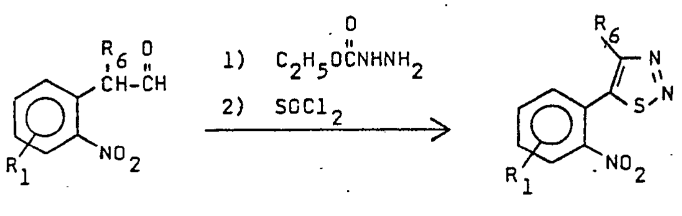


(Xs)

wherein
R^1 and R^6 are as originally defined.

Also, the 5-(2-nitrophenyl)-1,2,3-thiadiazoles of Formula (Xs') in Equation 60a below can be prepared by reacting an appropriate 2-nitrophenylacetaldehyde with ethyl carbazate, followed by cyclizing the hydrazone thus obtained with thionyl chloride according to teachings in U.S. 3,940,407.

Equation 60a



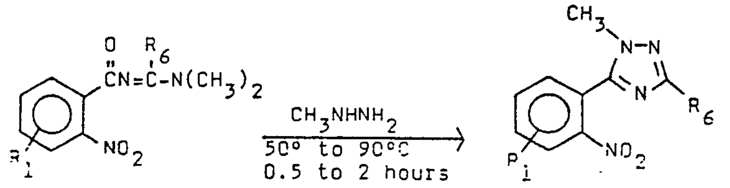
(Xs')

wherein

R_1 and R_6 are as originally defined.

The 1-methyl-5-(2-nitrophenyl)-1H-1,2,4-triazoles of Formula (Xt) in Equation 61 below can be prepared by reacting a N-[(dimethylamino)methylene]-2-nitrobenzamide of Formula (LXXIX) with methylhydrazine in acetic acid at 50° to 90°C for 0.5 to 2 hours, according to the teachings of Lin et al., *J. Org. Chem.*, *44*, 4160 (1979). The starting material LXXIX can be prepared by procedures described above in the first step of Equation 42.

Equation 61



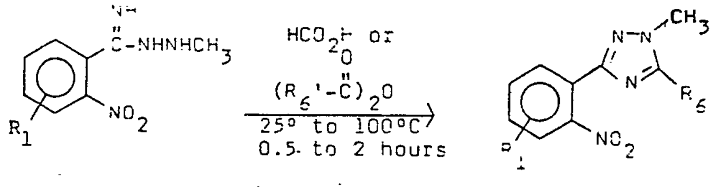
(LXXIX) (Xt)

wherein

R_1 and R_6 are as originally defined.

Equation 62 below illustrates a method for preparing 1-methyl-3-(2-nitrophenyl)-1H-1,2,4-triazoles of Formula (Xu).

Equation 62



(LXXX) (Xu)

wherein

R_1 is as originally defined;

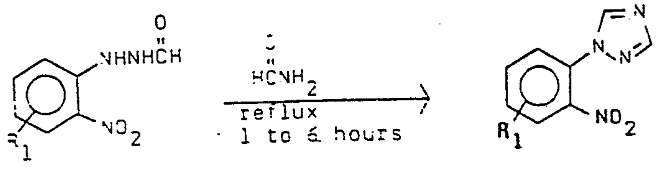
R_6' is CH_3 or C_2H_5; and

R_6 is H, CH_3 or C_2H_5.

The reaction of Equation 62 above can be run according to similar procedures described in M. Atkinson and J. Polya, *J. Chem. Soc.*, 3319 (1954). Thus, Xu is prepared by reacting a N-2-nitrobenzimidoyl-N'-methylhydrazine of Formula (LXXX) with formic acid, acetic anhydride or propionic anhyride at about 25° to 100°C for 0.5 to 1 hour. The starting material LXXX is prepared by reacting an appropriate 2-nitrobenzimidoate HCl with methylhydrazine in pyridine at ambient temperature according to the teachings of *ibid.*

The 1-(2-nitrophenyl)-1H-1,2,4-pyrazoles of Formula Xv in Equation 63 below can be prepared by reacting a 1-formyl-2-(2-nitrophenyl)hydrazine of Formula (LXXXI) with excess formamide at reflux for about 1 to 6 hours, according to the procedures described in C. Ainsworth et al., *J. Med. Pharm. Chem., 5,* 383 (1962).

Equation 63



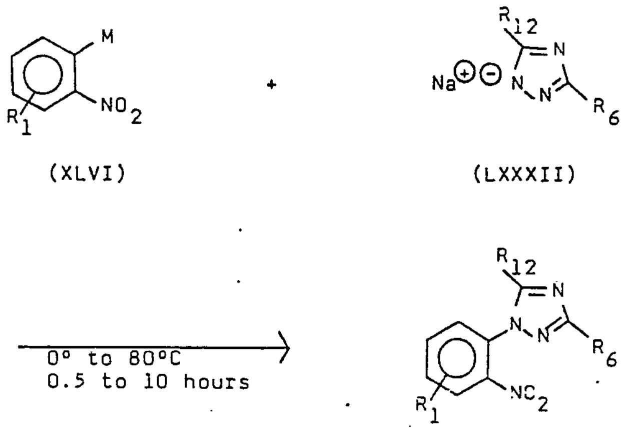
(LXXXI) (Xv)

wherein

R_1 is as originally defined.

The 1-(2-nitrophenyl)-1H-1,2,4-triazoles of Formula (Xv') in Equation 64 below can be prepared by reacting a 2-halo-1-nitrobenzene of Formula (XLVI) with a sodium 1,2,4-triazole salt of Formula (LXXXII). The reaction can be run by procedures described above in Equation 33 by one skilled in the art.

Equation 64



(XLVI) (LXXXII) (Xv')

wherein

M is Cl, Br or F; and

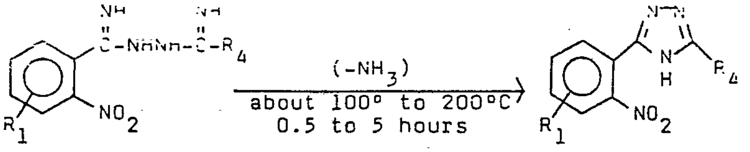
R_1-R_{12} are as originally defined.

Many 1-(2-nitrophenyl)-1H-1,2,4-triazoles of Formula (Xv') above can also be prepared by the Ullman reaction, according to the teachings of M. Khan and J. Polya, *J. Chem. Soc. C.,* 85 (1970). This requires reacting a 2-halonitrobenzene, such as XLVI above, with an appropriately substituted 1,2,4-triazole, copper (II) oxide catalyst and potassium carbonate in pyridine at reflux for 0.5 to several hours. The product is purified by column chromatography.

Equation 65 below illustrates a method for preparing 4-alkyl-3-(2-nitrophenyl)-4H-1,2,4-triazoles of Formula (Xw).

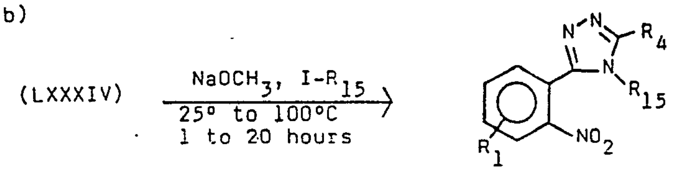
Equation 65

a)



(LXXXIII) (LXXXIV)

b)



(Xw)

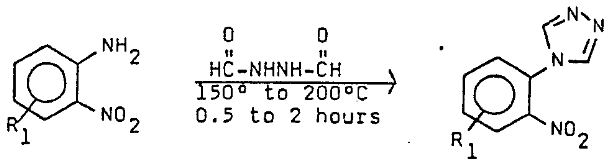
wherein

R_1, R_4 and R_{15} are as originally defined.

In the reaction of Equation 65a above, a 2-nitrophenyldihydrazidine of Formula (LXXXIII) can be heated at elevated temperatures, i.e. 100° to 200°C, in a solvent such as N-methyl-2-pyrrolidinone to cause cyclization to form a 3-(2-nitrophenyl)-4H-1,2,4-triazole of Formula (LXXXIV), according to methods known in the art, e.g., A. Rusanov, *Russ. Chem. Rev., 43,* 795 (1974). In reaction 65b, LXXXIV can be alkylated to form Xw. This requires reacting LXXXIV with sodium methoxide followed by an appropriate alkyl iodide in methanol at 25° to 100°C in a sealed tube for 1 to 20 hours. The product is purified by chromatography procedures.

The 4-(2-nitrophenyl)-4H-1,2,4-triazoles of Formula (Xx) in Equation 66 below can be prepared by reacting a 2-nitroaniline of Formula (LXXXV) with N,N′-diformylhydrazine at 150° to 200°C for about 0.5 to 2 hours, according to methods known in the art, e.g., C. Ainsworth et al., *J. Med. Pharm. Chem., 5,* 383 (1962).

Equation 66



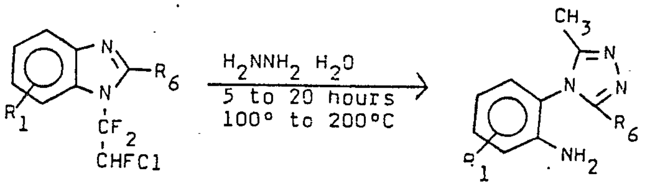
(LXXXV) (Xx)

wherein

R_1 is as originally defined.

Equation 67 below illustrates a method for preparing 4-(2-aminophenyl)-4H-1,2,4-triazoles of Formula (VIIIc).

Equation 67



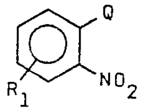
(LXXXVI) (VIIIc)

wherein

R_1 and R_6 are as originally defined.

The reaction of Equation 67 above is run according to similar procedures described in W. Ried and H. Lohwasser, *Justus Liebigs Ann. Chem., 699,* 88 (1966) and *Angew Chem. Int. Ed. Engl., 5,* 835 (1966). Thus, N-(1,1,2-trifluoro-2-chloroethyl)benzimidazole of Formula (LXXXVI) is reacted with excess hydrazine hydrate in ethylene glycol at reflux for 5 to 20 hours to form VIIIc.

Methods for preparing *ortho*-heteroaromatic nitrobenzenes of Formula (Xy) below are also known in the art. For suitable references, see Elderfield, "Heterocyclic Compounds", Vol. 5, Chapters 4, 5 and 8, 1957, J. Wiley and Sons, Inc., New York and R. Lakham and B. Ternai, *Advances in Heterocyclic Chemistry, 17,* 99-212 (1975).

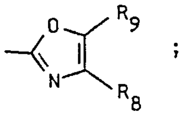


(Xy)

wherein Q is an

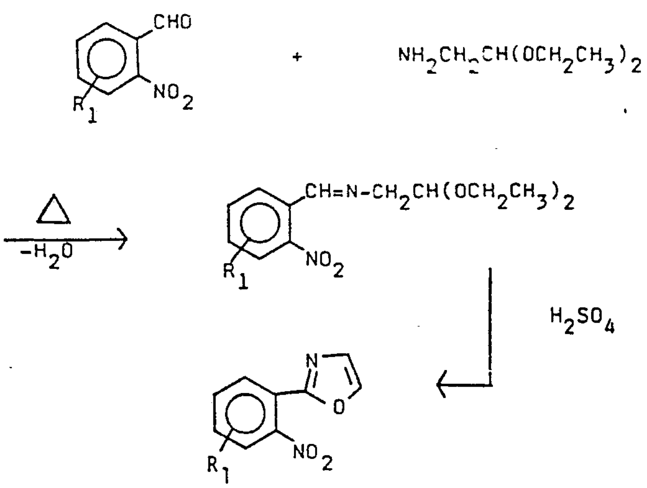
imidazole, thiazole, oxazole, imidazoline, thiazoline, oxazoline, oxazine, thiazine or tetrahydropyrimidine ring originally defined.

Nitro compounds of Formula (Xy) in which Q is



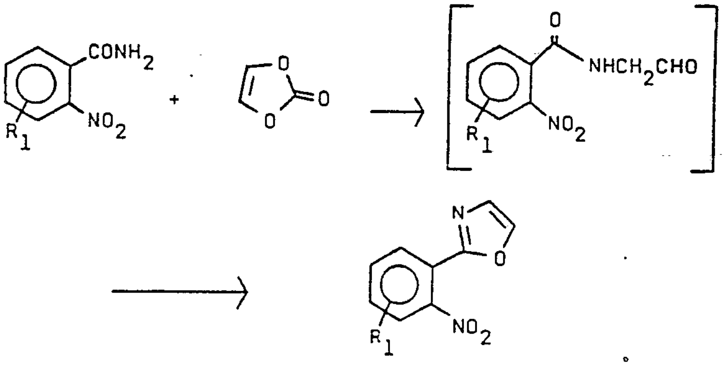
and R_8 and R_9 are H are prepared by the procedures shown in Equations 68 and 69.

Equation 68



(Xy-a)

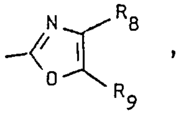
Equation 69



(Xy-a)

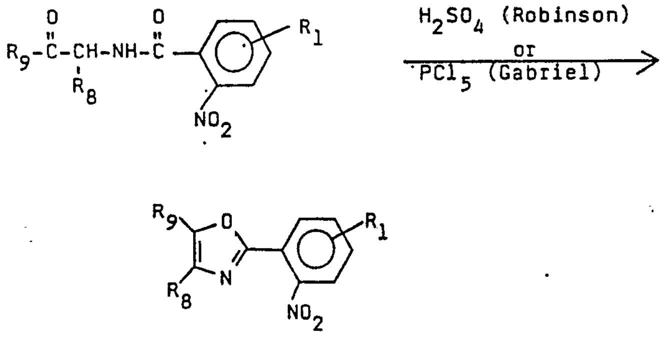
Details of the procedures of Equations 68 and 69 can be found in W. E. Cass, *J. Am. Chem. Soc.*, *64,* 785 (1942).

Compounds of Formula (Xy) in which Q is



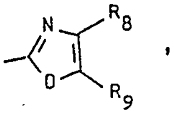
and R_8 and R_9 are H, CH_3 or CH_3CH_2 and both R_8 and R_9 are not simultaneously H, are prepared by Robinson-Gabriel synthesis of oxazoles from the corresponding acylaminoketones as described in Equation 70. See Elderfield, "Heterocyclic Compounds", Vol. 5, Chapter 5, 1957, J. Wiley and Sons, New York and references therein.

Equation 70



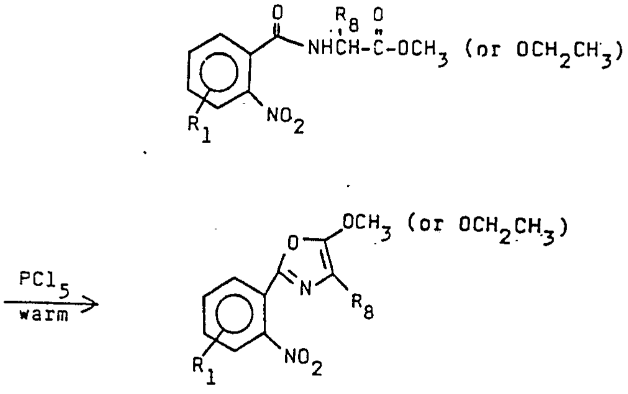
(Xy-b)

Compounds of Formula (Xy) in which Q is



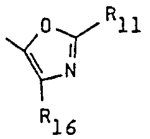
and R_9 is CH_3O or CH_3CH_2O and R_8 is H, CH_3 or CH_2CH_2 are prepared as shown in Equation 71. For further details, see Elderfield, location cited above.

Equation 71



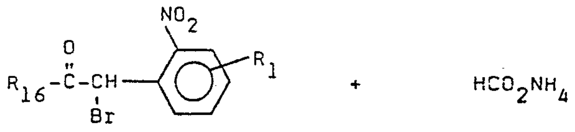
(Xy-c)

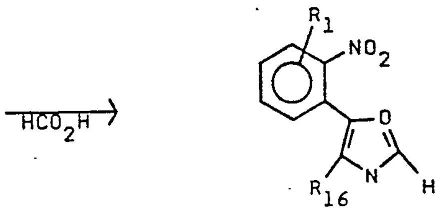
Compounds of Formula (Xy) in which Q is



R_{11} is H and R_{16} is CH_3 or CH_3CH_2 are prepared as shown in Equation 72.

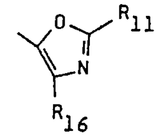
Equation 72





(Xy-d)

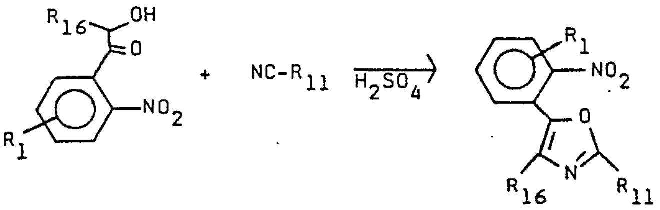
Compounds of Formula (Xy) in which Q is



,

R_{11} is CH_3 or CH_3CH_2 and R_{16} is CH_3 or CH_3CH_2 are prepared as shown in Equation 73.

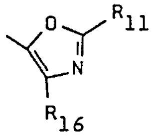
Equation 73



(Xy-e)

For details of the procedure of Equations 72 and 73, see the cited reference on page 95, line 11.

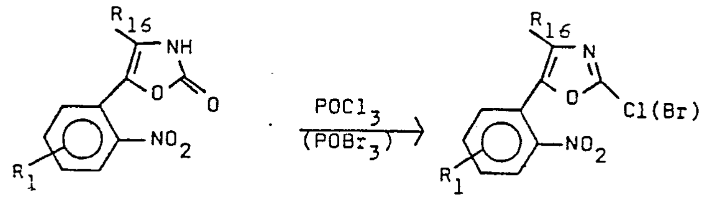
Compounds of Formula (Xy) in which Q is



,

R_{11} is Cl or Br and R_{16} is H, CH_3 or CH_3CH_2 are prepared from the corresponding oxazolone and $POCl_3$ or $POBr_3$ as shown in Equation 74. For details of this procedure, see I. J. Turche, M. J. S. Dewan, *Chem. Rev., 75,* 391 (1975) page 411 and related references therein.

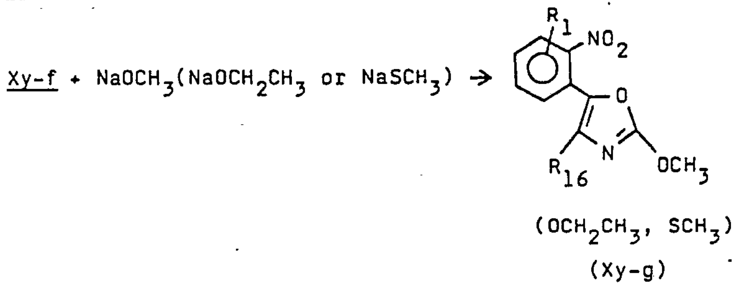
Equation 74



(Xy-f)

Compounds of Formula (Xy) where R_{16} is H, CH_3 or CH_3CH_2 and R_{11} is CH_3O, CH_3CH_2O or CH_3S are prepared from (Xy-f) by displacement with the appropriate nucleophile as shown in Equation 75.

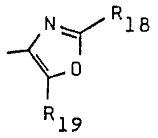
Equation 75



(Xy-g)

For details of this reaction see the cited reference on page 98, section IIo, page 402 and references cited therein.

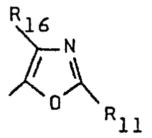
The oxazoles in which Q is



,

are prepared by analogous procedures to those described in Equations 72, 73, 74 and 75. Substituent limitations are the same.

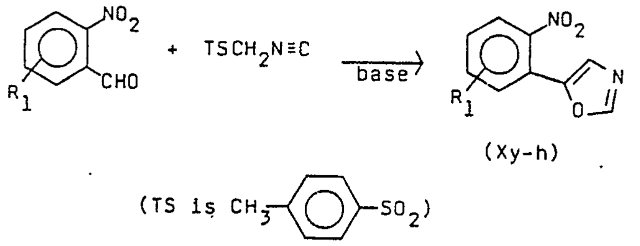
Compounds of Formula (Xy) in which Q is



,

and R_{11} and R_{16} are both H are best prepared by the procedures of Equations 76 or 77.

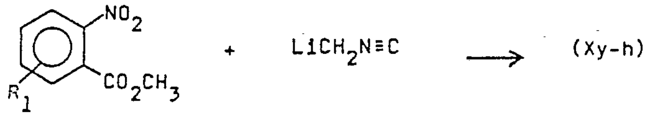
Equation 76



(Xy-h)

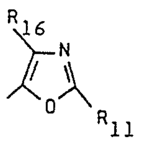
For details of this procedure, see A. M. Van Leusen, B. E. Hoogenboom and H. Sederius, *Tet. Let.* 2369 (1972).

Equation 77



For details of this procedure, see Y. Koyama, K. Yokose and L. Dolby, *Agric. Biol. Chem., 45,* 1285 (1981).

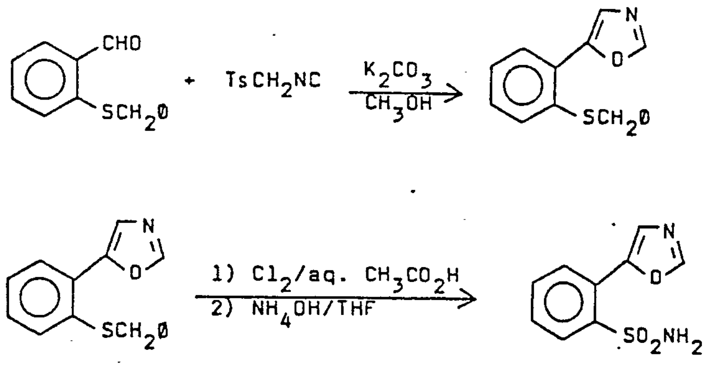
Alternatively, sulfonamide of Formula II where Q is



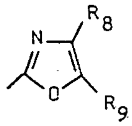
,

and R_{11} and R_{16} are both H can be prepared by the procedure of Equation 77a.

Equation 77a

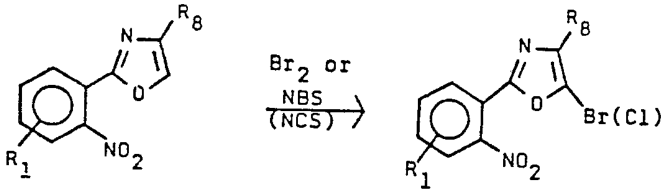


Compounds of Formula (Xy) in which Q is



R_9 is Cl or Br and R_8 is H, CH_3, CH_3CH_2, OCH_3 or OCH_2CH_3 are prepared by the procedure of Equation 78.

Equation 78



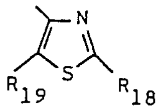
(Xy-i)

Details and references can be found in I. J. Turchi and M. J. S. Dewar, *Chem. Rev., 75,* 409 (1975).

Compounds of Formula (Xy-i) in which Br or Cl has been replaced by CH_3O, CH_3CH_2O or CH_3S are prepared by a displacement reaction with the corresponding nucleophile, using procedures known to those skilled in the art.

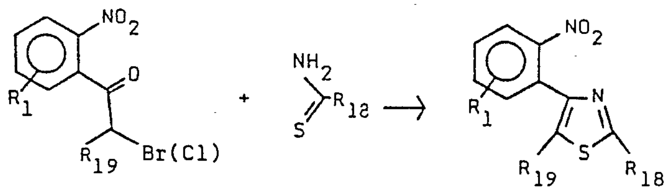
Thiazoles

Compounds of Formula (Xy) in which Q is



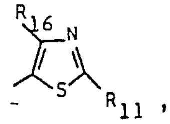
and R_{18} is H, CH_3 or CH_3CH_2 and R_{19} is H, CH_3 or CH_3CH_2 are prepared by the procedure of Equation 79.

Equation 79



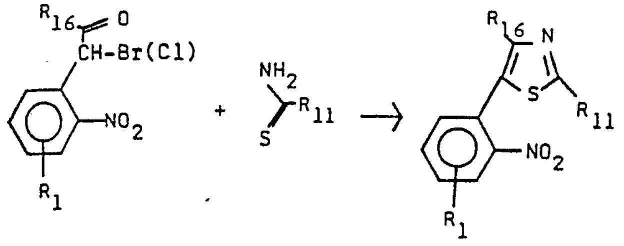
(Xy-j)

Similarly, compounds of Formula (Xy) in which Q is



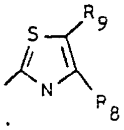
R_{16} is H, CH_3, CH_3CH_2 and R_{11} is H, CH_3 or CH_3CH_2 are prepared by the procedure of Equation 80.

Equation 80



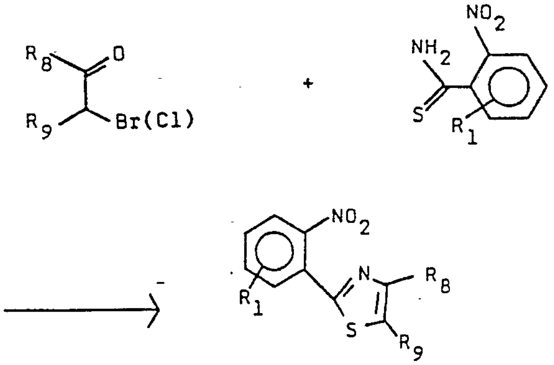
(4) (5) (Xy-k)

In like fashion to the above, compounds of formula (Xy) in which Q is



and R_8 is CH_3 or CH_3CH_2, and R_9 is H, CH_3 or CH_3CH_2 are prepared by the procedure of Equation 81.

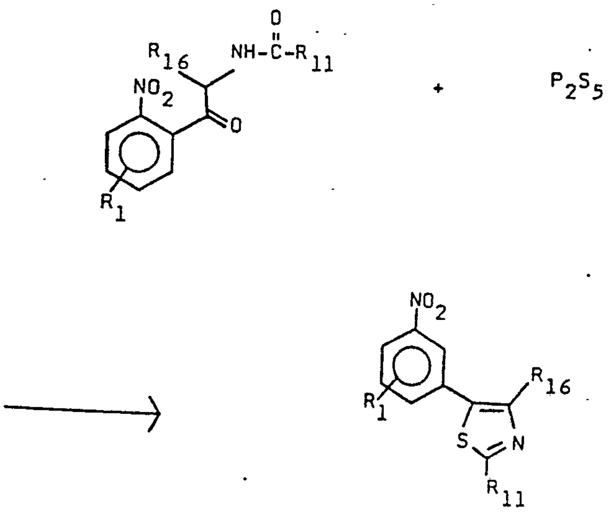
Equation 81



(Xy-1)

Routes to compounds of Formulas (Xy-m), (Xy-n) and (Xy-p) are available by reaction of the corresponding acylaminocarbonyl compounds with P_2S_5 (Gabriel Synthesis) as shown in Equations 82, 83 and 84 respectively.

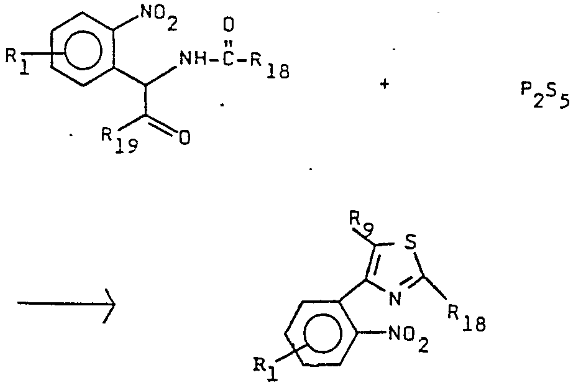
Equation 82



(Xy-m)

where
R_{11} is H, CH_3, CH_3CH_2, CH_3O or CH_3CH_2O;
R_{16} is CH_3, CH_3CH_2.

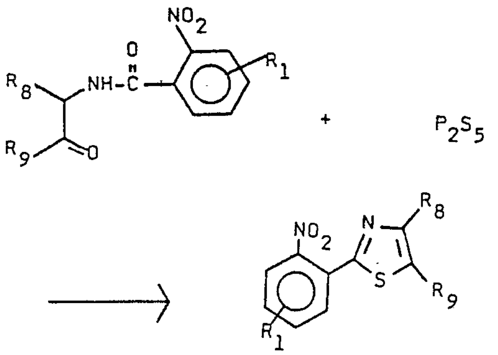
Equation 83



(Xy-n)

where
R_{18} is H, CH_3, CH_3CH_2, CH_3O or CH_3CH_2O;
R_{19} is CH_3 or CH_3CH_2.

Equation 84



(Xy-p)

where

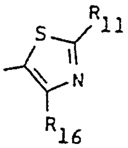
R_8 is H, CH_3 or Et;
R_9 is CH_3, CH_3CH_2, CH_3O or CH_3CH_2O.

For additional details on the procedures of Equations 79-84 see:

(1) J. V. Metzger (ed.), *Chem. Heterocyclic Compounds, 34,* parts (1-3) (1978-1979).

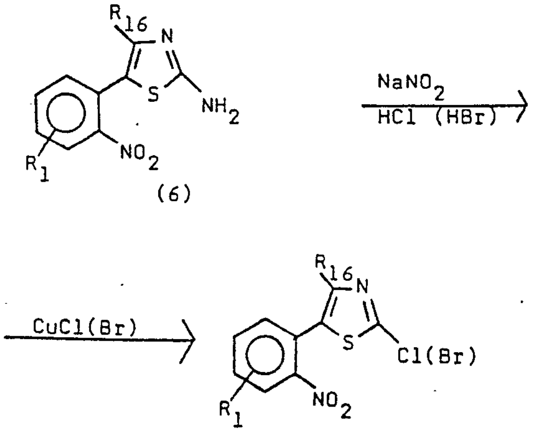
(2) J. M. Sprague and A. H. Land, "Heterocyclic Compounds", (R. C. Elderfield, ed.) *V,* 484-722. Wiley, N.Y.

Compounds of Formula (Xy) in which Q is



and R_{16} is H, CH_3 or CH_3CH_2, and R_{11} is Cl or Br are prepared via the Sandmeyer reaction on the corresponding amino derivative as shown in Equation 85.

Equation 85

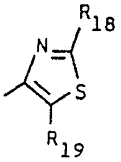


(6)

(Xy-q)

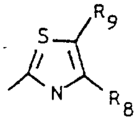
The amino derivatives (16) are prepared by substitution of the thioamide (5) with thiourea (Equation 80). Treatment of (Xy-q) with $NaOCH_3$, $NaOCH_2CH_3$ or $NaSCH_3$ by procedures known to one skilled in the art yields the products in which R_{11} is OCH_3, OCH_2CH_3 or SCH_3 in Formula (Xy-q).

In like fashion to the above, compounds of Formula (Xy) in which Q is



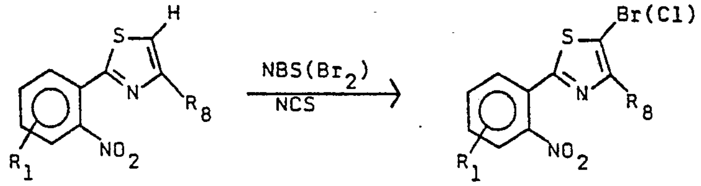
R_{19} is H, CH_3 or CH_3CH_2 and R_{18} is Cl, Br, OCH_3, OCH_2CH_3 or SCH_3 can be prepared.

Compounds of the Formula (Xy-t) in which Q in (Xy) is



and R_8 is H, CH_3 or CH_3CH_2 and R_9 is Cl or Br are prepared by the procedure of Equation 86.

Equation 86



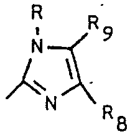
(Xy-t)

Compounds (Xy-t) are in turn used to prepare the corresponding analogs in which Br is replaced by OCH_3, OCH_2CH_3 or SCH_3 via displacement.

While OCH_3, OCH_2CH_3 and SCH_3 are best introduced at the nitro stage, Cl or Br are best introduced at the sulfonamide stage in order to avoid possible loss of halogen during reduction.

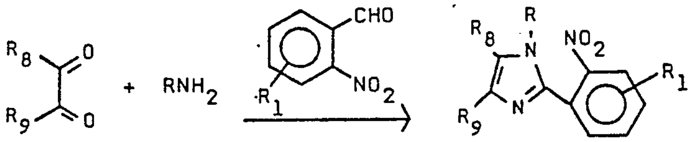
Imidazoles

Compounds of Formula (Xy-u) in which Q in (Xy) is



and R is H or C_1-C_4 alkyl, R_8 is H, CH_3 or CH_3CH_2 and R_9 is H, CH_3 or CH_3CH_2 are prepared by the procedure of Equation 87

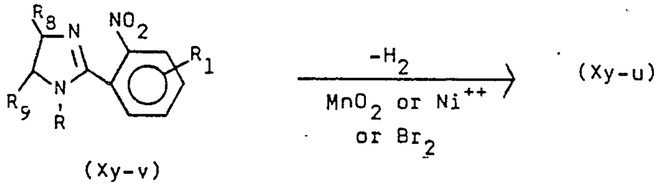
Equation 87



(Xy-u)

Additionally, compound (Xy-u) may be prepared from the corresponding imidazoline (Xy-v) by dehydrogenation with a suitable catalyst (Equation 88).

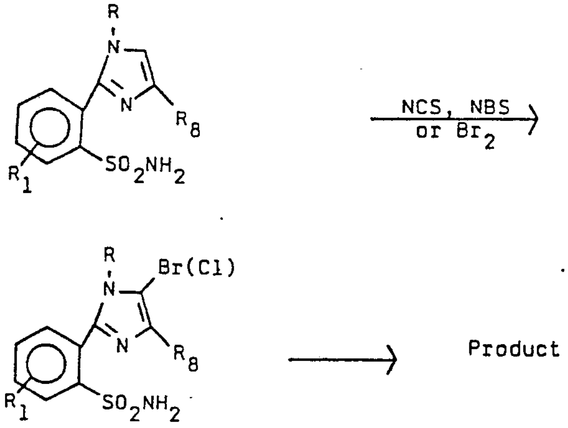
Equation 88



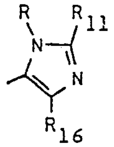
(Xy-v)

For compounds in which R_9 is OCH_3, OCH_2CH_3 or SCH_3 in the final product, these substituents are best introduced into the nitro compound (Xy-u) and are prepared by the displacement of compounds in which R_9 is Cl or Br in (Xy-u); halogen is introduced by treatment of the compounds of Formula (Xy-u) (R_9 is H) with NBS or NCS. To avoid loss of halogen during reduction, halogen is best introduced into the final product at the sulfonamide stage (Equation 89).

Equation 89

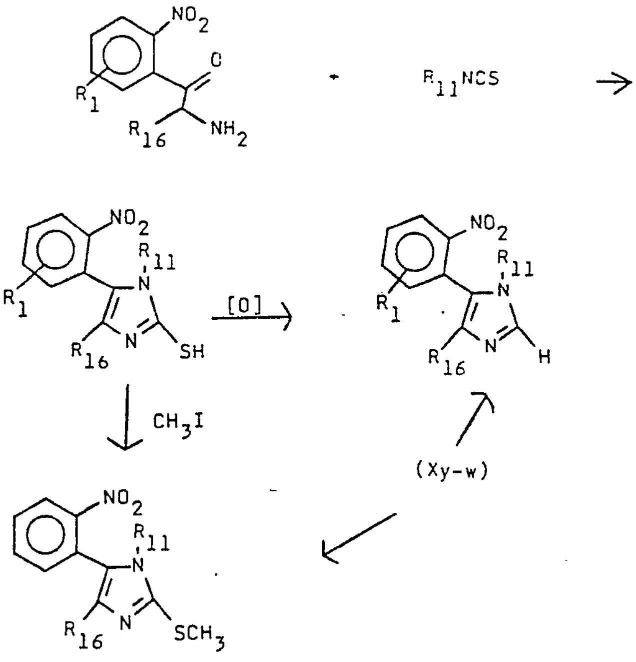


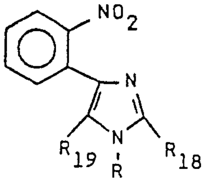
Compounds of Formula (Xy-w) in which Q is



in (Xy), R_{11} is H or CH_3S and R_{16} is H, CH_3 or CH_3CH_2 are prepared by treatment of the corresponding α-aminoketone with the appropriate isothiocyanate $R_{11}NCS$ as shown in Equation 90. Similarly prepared are compounds (Xy-x) in which R_{18} is H or SCH_3 and R_{19} is H, CH_3 or CH_3CH_2.

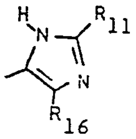
Equation 90





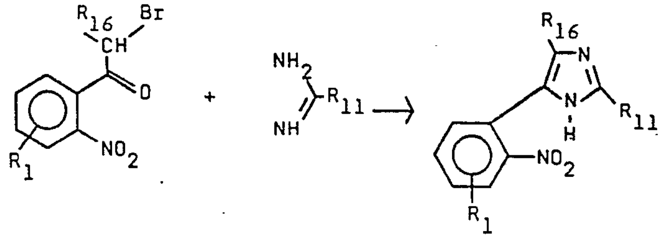
(Xy-x)

Contacting the appropriate α-haloketone with an amidine provides compounds of Formula Xy in which Q is



and R_{11} is H, CH_3 or CH_3CH_2 as shown in Equation 90-A.

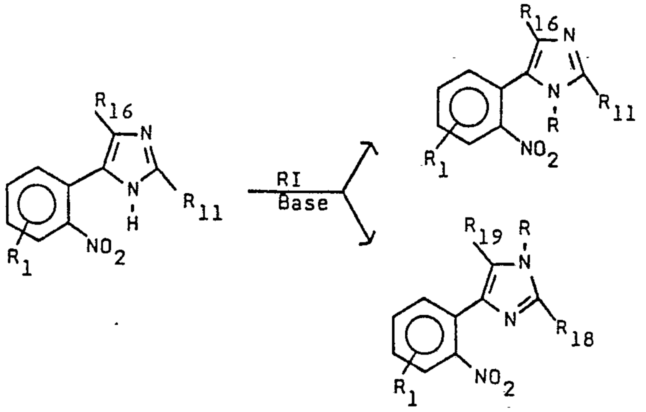
Equation 90-A



Alkylation of this product with a C_1-C_4 alkyl iodide using procedures known to those skilled in the art leads to products in which R_{11} is H, CH_3 or CH_3CH_2 and R is C_1-C_4 alkyl.

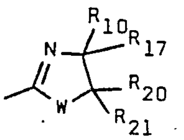
Also produced in the alkylation are compounds in which R is C_1-C_4 alkyl and R_{18} is H, CH_3 or CH_3CH_2 and R_{19} is H, CH_3 or CH_3CH_2 as shown in Equation 90-B.

Equation 90-B

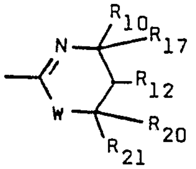


Oxazolines, thiazolines and imidazolines

The intermediate oxazolines, thiazolines and imidazolines (1), are required for use in preparing compounds of Formula I in which Q is



or



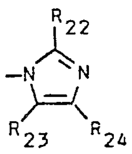
For methods see:

"Heterocyclic Compounds", *V*, R. C. Elderfield, Editor, John Wiley and Sons, Inc., New York, 1957.

R. J. Fern and J. C. Riebsomer, *Chem. Rev., 54,* 543-613 (1954).

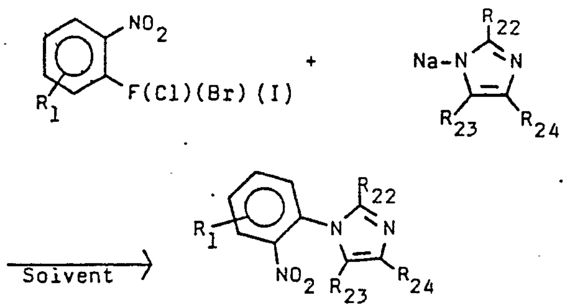
R. H. Wiley and L. L. Bennett, Jr., *Chem. Rev., 44,* 447-476 (1964).

Compounds of Formula (Xy) in which Q is



are best prepared by displacement of halogen from the corresponding halo aromatic nitro compound with the imidazole salt as shown in Equation 91.

Equation 91



(Xy-y)

The reaction is conducted in the presence of an appropriate solvent such as dimethylformamide, N-methylpyrolidone or THF at temperatures from ambient to reflux and for a period of one to twenty-four hours. The procedure is especially useful when R_{22} is H, CH_3 or CH_3CH_2. When R_{22} is CH_3S, CH_3O or CH_3CH_2O in (Xy-y), these substituents are best introduced by displacement of the corresponding iodo, bromo or chloro derivatives of (Xy-y).

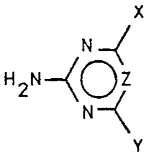
For the preparation of 2-haloimidazoles see:

"Heterocyclic Compounds" *V*, R. C. Elderfield, Editor, John Wiley and Sons, New York, 1957.

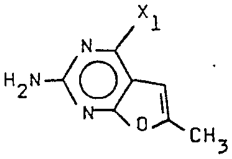
K. Hofmann, "The Chemistry of Heterocyclic Compounds," (A. Weissberger, ed.), Vol. 6, pp. 1-420. Wiley (Interscience), New York, 1953.

The heterocyclic amines of Formula (VII) in Equations 1a and 3 are also important intermediates for the preparation of the compounds of this invention, which can be prepared by the following methods.

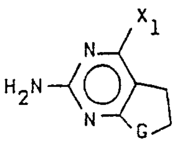
The pyrimidines and triazines of Formula (VIIa) to (VIId) below are either known or can be prepared by obvious methods by one skilled in the art. For instance, the synthesis of pyrimidines and triazines of the general formula VIIa has been reviewed in "The Chemistry of Heterocyclic Compounds", a series published by Interscience Publishers, Inc., New York and London. 2-Aminopyrimidines are described by D. J. Brown in "The Pyrimidines", Vol. 16 of this series. 2-Amino-1,3,5-triazines are reviewed by E. M. Smolin and L. Rapaport in "*s*-Triazines and Derivatives", Vol. 13 of the same series. The synthesis of triazines is also described by F. C. Schaefer, U.S. 3,154,547 and by K. R. Huffman and F. C. Schaefer, *J. Org. Chem., 28,* 1812 (1963). The synthesis of the bicyclic amines VIIc and VIId are described in EPO Publication No. 15,683, and that of VIIb in European Patent Publication No. 46,677.



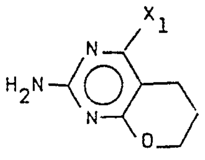
(VIIa)



(VIIb)



(VIIc)

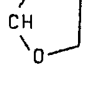


(VIId)

wherein

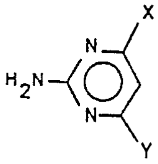
G, X, X_1, Y and Z are as originally defined

except Y $\neq$ $CH(OCH_3)_2$ or



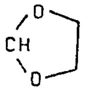
.

Pyrimidines below of Formula (VIIe), where Y is $CH(OC_2H_5)_2$, are described by W. Braker et al., *J. Am. Chem. Soc., 69,* 3072 (1947), the disclosure of which is herein incorporated by reference. Using techniques taught by Braker, or suitable modifications that would be obvious to one skilled in the art, the pyrimidines VIIe can be prepared.



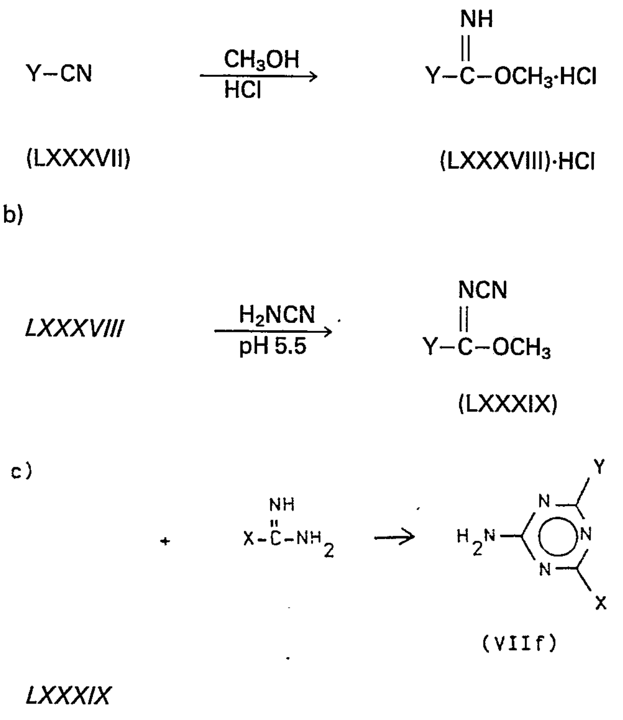
(VIIe)

wherein
X is CH_3, OCH_3 or Cl; and
Y is $CH(OCH_3)_2$ or

 ;

Triazines of Formula (VIIf) may be prepared according to the methods outlined in Equations 92 and 93.

Equation 92



a)

$$Y{-}CN \xrightarrow[HCl]{CH_3OH} Y{-}\overset{\overset{NH}{\|}}{C}{-}OCH_3 \cdot HCl$$

(LXXXVII) (LXXXVIII)·HCl

b)

$$LXXXVIII \xrightarrow[pH\ 5.5]{H_2NCN} Y{-}\overset{\overset{NCN}{\|}}{C}{-}OCH_3$$

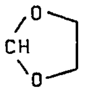
(LXXXIX)

c)

$$LXXXIX + X{-}\overset{\overset{NH}{\|}}{C}{-}NH_2 \longrightarrow$$

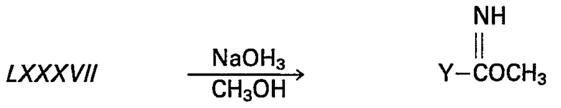
(VIIf)

wherein
X is CH_3 or OCH_3; and
Y is $CH(OCH_3)_2$ or

 ;

Equation 93

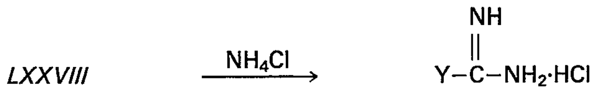
a)



$$LXXXVII \xrightarrow[CH_3OH]{NaOH_3} Y-\overset{\overset{NH}{\|}}{C}OCH_3$$

(LXXXVIII)

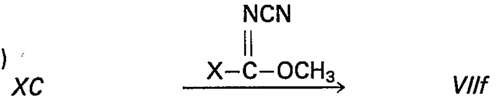
b)



$$LXXVIII \xrightarrow{NH_4Cl} Y-\overset{\overset{NH}{\|}}{C}-NH_2{\cdot}HCl$$

(XC)

c)



$$XC \xrightarrow{X-\overset{\overset{NCN}{\|}}{C}-OCH_3} VIIf$$

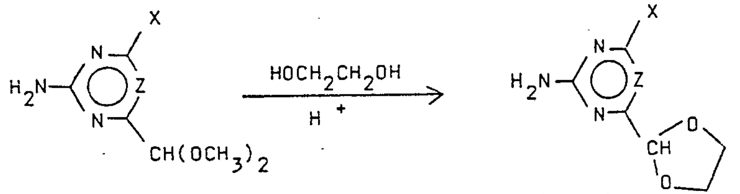
wherein

X and Y are as defined in Equation 68.

The reaction of Equation 92a is carried out according to the teachings of J. M. McElvain and R. L. Clarke, *J. Amer. Chem. Soc., 69,* 2657 (1947), in which the preparation of ethyl diethoxyiminoacetate is described. The intermediate N-cyanoimidate of Formula (LXXXIX) may be prepared according to the teaching of D. Lwowski in *Synthesis, 1971,* 263, by reacting LXXXVIII with cyanamide at pH 5.5, and this may be condensed according to reaction 92c with either acetamidine or O-methyl isourea in an alcoholic solvent at 25 to 80°C to provide the appropriate triazines. Alternatively, the reaction of Equation 93a, described for substituted acetonitriles by F. C. Schaefer and G. A. Peters in *J. Org. Chem., 26,* 412 (1961), may be used to convert nitrile of Formula (LXXXVII) to the corresponding iminoester. The free base may be carried on through reactions 93b and 93c, or, alternatively, converted to the amidinium hydrochloride salt (XC) as described in the aforementioned reference, and condensed with either methyl N-cyanoacetimidate or with dimethyl N-cyano imidocarbonate in the presence of one equivalent of sodium methoxide to provide the triazines of Formula (VIIf).

Cyclic acetals of Formula (VIIh) may also be prepared from compounds of Formula (VIIg) according to Equation 94 by acetal exchange.

Equation 94



(VIIg) (VIIh)

wherein

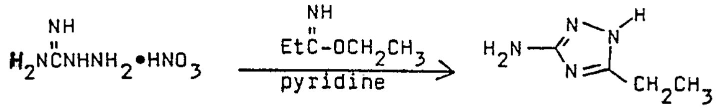
X is CH_3 or OCH_3; and

Z is CH or N.

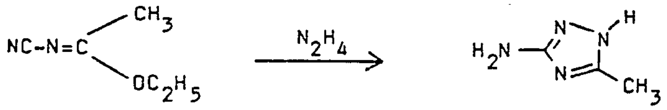
The reaction of Equation 94 is carried out by heating the acyclic acetal in an inert solvent in the presence of one equivalent ethylene glycol and slightly more than one equivalent of a strong acid, such as *p*-toluenesulfonic acid with removal of the methanol or ethanol formed in the reaction by distillation. The product is isolated by treatment with aqueous base, and extraction with an organic solvent, and purified by crystallization or column chromatography.

Preparations of 3-amino-1,2,4-triazoles of Formula (VII) in Equations 1a and 3 are known in the art and 1,2,4-triazoles are reviewed in *The Chemistry of Heterocyclic compounds* "Triazoles 1,2,4" (John Wiley and Sons, New York, 1981). Commonly used starting materials containing nitrogen are N-aminoguanidine, hydrazine, alkylhydrazines, cyanamide, ethyl cyanoacetimidate, dimethyl cyanodithioimidocarbonate, dimethyl cyanoimidocarbonate, ethoxymethylenecyanamide, and acylhydrazines. Some literature synthesis are illustrated below. Using these techniques or suitable modifications that would be apparent to one skilled in the art, the 3-amino-1,2,4-triazole intermediates can be readily prepared.

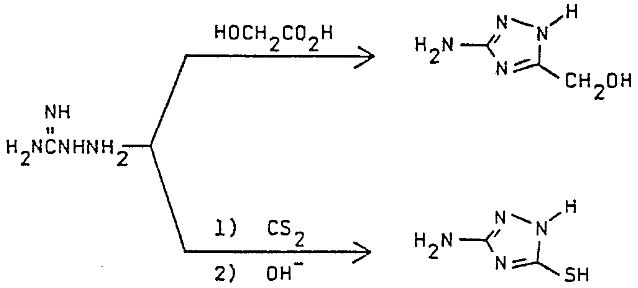
Heating equimolar amounts of ethyl propionimidate hydrochloride and N-aminoguanidine nitrate in pyridine gives 3-amino-5-ethyltriazole; *German Patent* 1,073,499 (1960); *Berichte, 96,* 1064 (1963).



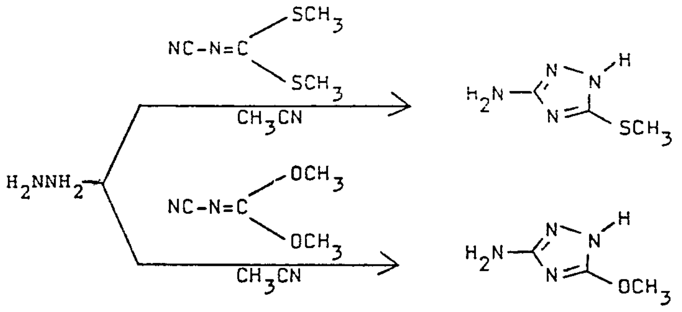
Condensation of hydrazine with ethyl N-cyanoacetimidate yields 3-amino-5-methyltriazole; *Journal of Organic Chemistry, 28,* 1816 (1963).



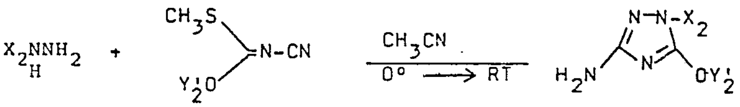
U.S. Patent 2,835,581 (1958) teaches the preparation of 3-amino-5-(hydroxymethyl)triazole from N-aminoguanidine and glycolic acid and *British Patent* 736,568 (1955) describes the synthesis of 3-amino-5-mercaptotriazole.



Condensing hydrazine with dimethyl cyanodithioimidocarbonate in acetonitrile gives 3-amino-5-methylthio-1,2,4-triazole while reaction of hydrazine with dimethyl N-cyanoimidocarbonate produces 3-amino-5-methoxy-1,2,4-triazole; *Journal of Organic Chemistry, 39,* 1522 (1974).



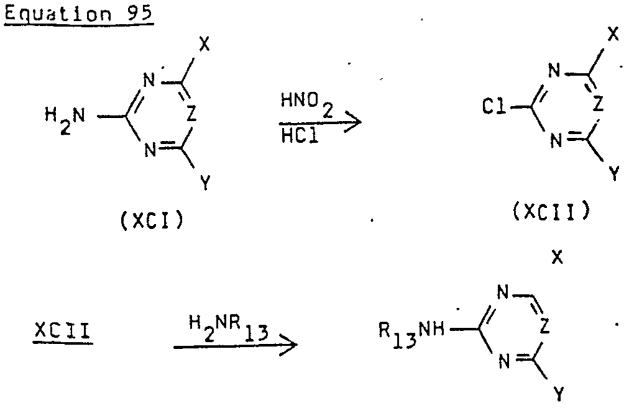
Reaction of substituted hydrazines with N-cyanothioimidocarbonates (prepared according to the procedure given in D. M. Wieland, Ph.D. Thesis, 1971, pp. 123-124) yields disubstituted aminotriazoles as shown below.



$(Y_2' = CH_3 \text{ or } C_2H_5)$

Many of the aminoheterocyclic intermediates of Formula (VII) where R_{13} is methyl may be prepared by a two-step procedure as described for VIIi in Equation 95.

Equation 95



(XCI) (XCII)

(VIIi)

wherein

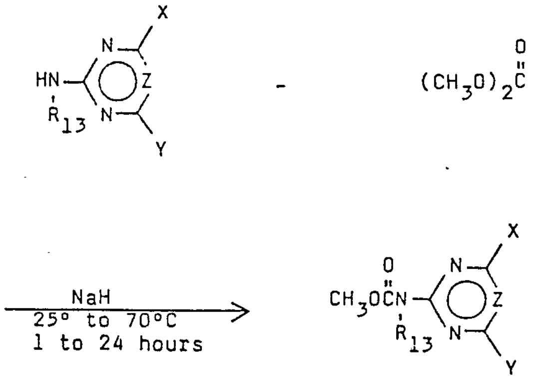
X, Y and Z are as originally defined and

R_{13} is CH_3.

A solution of the amine (XCI) in concentrated hydrochloric acid is treated with sodium nitrite solution and the chloro compound (XCII) is isolated in the usual manner by filtration of the acidic solution. A representative procedure is described by Bee and Rose in *J. Chem. Soc. C,* 2031 (1966), for the case in which Z = CH, and X = Y = OCH_3. Displacement of the chlorine of (XCII) may be accomplished by heating with an excess of methylamine in water to obtain the methylamino heterocycle (VIIi).

Equation 96 below illustrates the preparation of the required methyl pyrimidinyl carbamates and methyl triazinyl carbamates of Formula (III) in Equation 1. By obvious modifications, other methyl carbamates of Formula (III) may be prepared by this method by one skilled in the art.

Equation 96



(IIIa)

wherein

X, Y, Z and R_{13} are as originally defined.

According to Equation 96, a heterocyclic amine is reacted with two equivalents of sodium hydride and excess dimethyl carbonate to form IIIa. The reaction is run in an inert solvent such as tetrahydrofuran at 25°C to reflux for 1 to 24 hours. The product is isolated by (a) adding about two equivalents of concentrated hydrochloric acid under nitrogen at 0° to 30°C; (b) filtering; and (c) separating out the organic phase, then drying (sodium sulfate and/or magnesium sulfate) and concentrating to dryness *in vacuo*. The product IIIa may be purified further by recrystallization or chromatography procedures.

Agriculturally suitable salts of compounds of Formula I are also useful herbicides and can be prepared in a number of ways known to the art. For example, metal salts can be made by contacting compounds of Formula I with a solution of an alkali or alkaline earth metal salt having a sufficiently basic anion (e.g., hydroxide, alkoxide, carbonate or hydroxide). Quaternary amine salts can be made by similar techniques.

Salts of compounds of Formula I can also be prepared by exchange of one cation to another. Cationic exchange can be effected by direct treatment of an aqueous solution of a salt of a compound of Formula I (e.g., alkali metal or quaternary amine salt) with a solution containing the cation to be exchanged. This method is most effective when the desired salt containing the exchanged cation is insoluble in water, e.g., a copper salt, and can be separated by filtration.

Exchange may also be effected by passing an aqueous solution of a salt of a compound of Formula I (e.g., an alkali metal or quaternary amine salt) through a column packed with a cation exchange resin containing the cation to be exchanged. In this method, the cation of the resin is exchanged for that of the original salt and the desired product is eluted from the column. This method is particularly useful when the desired salt is water-insoluble, e.g., a potassium, sodium or calcium salt.

Acid addition salts, useful in this invention, can be obtained by reacting a compound of Formula I with a suitable acid, e.g., *p*-toluenesulfonic acid, trichloroacetic acid or the like.

The compounds of this invention and their preparation are further illustrated by the following examples wherein temperatures are given in degrees centigrade and all parts are by weight unless otherwise indicated.

EXAMPLE 1

3-(Dimethylamino)-1-(2-nitrophenyl)-2-propen-1-one

To a solution of 126 g of 2-nitroacetophenone in 125 ml of toluene was slowly added 119 g of N,N-dimethylformamide dimethyl acetal. The solution was refluxed for 16 hours, then concentrated to dryness *in vacuo*. The solid residue was washed 1 × 200 ml of 2-propanol and 2 × 200 ml of hexane to yield 147 g of the title compound; m.p. 118-121°C.

Anal. Calcd. for $C_{11}H_{12}N_2O_3$:
C, 59.9; H, 5.5; N, 12.7.
Found:
C, 59.4; H, 5.5; N, 12.6.

EXAMPLE 2

5-(2-Nitrophenyl)isoxazole

A suspension containing 99 g of 3-(dimethylamino)-1-(2-nitrophenyl)-2-propen-1-one, prepared in Example 1, and 63 g of hydroxylamine hydrochloride in 250 ml of ethanol was refluxed for 16 hours, then concentrated to dryness *in vacuo*. Water (400 ml) was added to the residue, and the resulting suspension was filtered. The solid was recrystallized from 2-propanol to give 60 g of the title compound; m.p. 66-69°C.

Anal. Calcd. for $C_9H_6N_2O_3$:
C, 56.8; H, 3.2; N, 14.7.
Found:
C, 56.3; H, 3.3; N, 14.6.

EXAMPLE 3

5-(2-Aminophenyl)isoxazole

To a suspension containing 206 g of stannous chloride dihydrate in 520 ml of concentrated hydrochloric acid was cautiously added 56 g of 5-(2-nitrophenyl)isoxazole, prepared in Example 2. The resulting suspension was refluxed on a steam bath for about 1 hour, then cooled to 10°C and filtered. The solid was added to about 600 ml of ice-water, and the suspension was made basic to a pH of about 10 with addition of 50% NaOH. The aqueous mixture was extracted with methylene chloride. After drying the methylene chloride extract over sodium sulfate, the solvent was evaporated under reduced pressure to give 32 g of the title compound as an oil.

Anal. Calcd. for $C_9H_8N_2O$:
C, 67.5; H, 5.1; N, 17.5.
Found:
C, 67.1; H, 5.1; N, 17.3.

EXAMPLE 4

2-(Isoxazol-5-yl)benzenesulfonyl chloride

A diazonium salt was prepared by adding a solution of 14.5 g of sodium nitrate in 30 ml of water to a suspension of 32 g of 5-(2-aminophenyl)isoxazole, prepared in Example 3, and 72 ml of concentrated hydrochloric acid in 210 ml of glacial acetic acid cooled at 0° to 5°C. After stirring about 0.4 hour, the diazonium salt suspension was poured in one portion into a mixture consisting of 150 ml of acetic acid, 8.4 g of cupric chloride dihydrate and 60 ml of sulfur dioxide and cooled at 10°C by an ice-water bath. The mixture was stirred at 10° to 15°C for 0.2 hour then at about 20° to 30°C for 3 hours. The suspension was poured into ice-water (about 700 ml) and stirred to form a solid. The mixture was filtered and the solid was washed 3 × 100 ml of water and suction dried to give 37 g of crude 2-(isoxazol-5-yl)benzenesulfonyl chloride; m.p. 63-65°C.

EXAMPLE 5

2-(Isoxazol-5-yl)benzenesulfonamide

A solution of 37 g of 2-(isoxazol-5-yl)benzenesulfonyl chloride, prepared in Example 4, in 200 ml of tetrahydrofuran, was cooled in an ice-water bath while about 40 ml of concentrated aqueous ammonium hydroxide was added slowly at 10° to 30°C. The resulting suspension was stirred at room temperature for 4 hours, then the solvent was evaporated under reduced pressure. The residue was stirred in 150 ml of water for 0.5 hour, then filtered. The crude, wet solid was dissolved in tetrahydrofuran and dried over sodium sulfate. The solid was recrystallized from 2-propanol to give 20 g of the title compound; m.p. 132-135°C.

Anal. Calcd. for $C_9H_8N_2O_3S$:

C, 48.1; H, 3.6; N, 12.5.

Found:

C, 47.9; H, 3.6; N, 12.5.

EXAMPLE 6

N-(Butylaminocarbonyl)-2-(isoxazol-5-yl)-benzenesulfonamide

A solution of 8.7 g of 2-(isoxazol-5-yl)benzenesulfonamide prepared in Example 5, 4.7 g of *n*-butyl isocyanate and 5.4 g of potassium carbonate in 125 ml of 2-butanone was refluxed for 7 hours. The resulting suspension was concentrated to dryness *in vacuo*. The residue was taken up in 200 ml of water and extracted once with 100 ml of ethyl ether. The aqueous layer was acidified with 2N HCl and the resulting suspension was extracted with methylene chloride. After drying the methylene chloride extract over sodium sulfate, the solvent was evaporated off under reduced pressure. The solid residue was recrystallized from acetonitrile to give 10 g of the title compound; m.p. 110-113°C.

Anal. Calcd. for $C_{14}H_{17}N_3O_4S$:

C, 51.9; H, 5.3; N, 12.9.

Found:

C, 51.2; H, 5.3; N, 12.6.

EXAMPLE 7

2-(Isoxazol-5-yl)benzenesulfonyl isocyanate

A suspension of 6 g of N-(butylaminocarbonyl)-2-(isoxazol-5-yl)benzenesulfonamide, prepared in Example 6, in 50 ml of xylene containing 0.2 g of DABCO was heated at 130-135°C while 1.6 ml of phosgene was added portionwise at a rate to maintain a reflux temperature of 130-135°C. The mixture was refluxed for an additional 2 hours, cooled under nitrogen to room temperature, filtered, and the filtrate was concentrated to dryness *in vacuo*. A sample of the crude oil product displayed a characteristic sulfonyl isocyanate band in the IR at 2200 cm^{-1}.

EXAMPLE 8

N-[(4,6-Dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(isoxazol-5-yl)benzenesulfonamide

To a suspension of 2.5 g of 2-amino-4,6-dimethoxypyrimidine in 25 ml of acetonitrile was added 4 g of crude 2-(isoxazol-5-yl)benzenesulfonyl isocyanate prepared in Example 7. The resulting suspension was warmed at 40°C for about 3 minutes to form a solution. The solution was stirred at room temperature for 10 hours, then concentrated *in vacuo* to give a viscous oil. The oil was chromatographed in a dry column of silica gel with ethyl acetate as eluant. The first fraction from the column was concentrated *in vacuo* to yield a solid. The solid was washed 1 × 10 ml of acetonitrile and suction dried to yield 0.5 g of the title compound; m.p. 183-187°C. The IR spectrum showed a carbonyl absorption at 1710 cm^{-1} indicative of a sulfonylurea.

Anal. Calcd. for $C_{16}H_{15}N_5O_6S$:

C, 47.4; H, 3.7.

Found:

C, 47.1; H, 3.9.

EXAMPLE 9

Methyl (4,6-dimethoxypyrimidin-2-yl)carbamate

2-Amino-4,6-dimethoxypyrimidine (56 g) was added portionwise to 50% sodium hydride (42.8 g) in 1000 ml of dry tetrahydrofuran. After stirring for 0.5 hour, dimethylcarbonate (58.5 g) was added dropwise with cooling. The mixture was stirred under nitrogen for about 16 hours at ambient temperature. Concentrated HCl (80 ml) was added slowly as external cooling was used to maintain a pot temperature of about 25°C. Saturated aqueous sodium chloride (80 ml) was then added. The solvents were decanted from the precipitated solids and dried over sodium sulfate. Filtering and evaporating the solvents afforded the crude material which was recrystallized from hexane to yield 54 g of the title compound, m.p. 81-83°C. The IR spectrum showed characteristic absorption bands at 3400 and 1760 cm^{-1}.

EXAMPLE 10

4-(2-Nitrophenyl)isoxazole

A suspension of 75 g of 3-(dimethylamino)-2-(2-nitrophenyl)acrolein [prepared by the procedure of U. Hengartner et al., *J. Org. Chem., 44,* 3748 (1979)] and 47.3 g of hydroxylamine hydrochloride in 300 ml of ethanol was refluxed for 10 hours, then concentrated to dryness *in vacuo*. Water (400 ml) was added to the residue and the suspension filtered. The isolated solid was recrystallized from 2-propanol to yield 56 g of the title compound; m.p. 51-54°C.

Anal. Calcd. for $C_9H_6N_2O_3$:
C, 56.8; H, 3.2; N, 14.7.
Found:
C, 56.6, H, 3.1; N, 14.4.

EXAMPLE 11

4-(2-Aminophenyl)isoxazole

To a suspension of 108 g of stannous chloride dihydrate in 270 ml of concentrated hydrochloric acid was added portionwise 30 g of 4-(2-nitrophenyl)isoxazole prepared in Example 10 at less than 30°C. The suspension was stirred at about 20°-35°C for 10 hours, then cooled to 10°C and filtered. The solid was added to ice-water (400 ml), 50% NaOH was added to make the suspension basic to a pH of about 10, and the resulting suspension filtered. The solid was washed 3 × 100 ml of water and suction dried to give 18 g of the title compound; m.p. 48-51°C.

Anal. Calcd. for $C_9H_8N_2O$:
C, 67.5; H, 5.1; N, 17.5.
Found:
C, 67.1; H, 5.1; N, 17.2.

EXAMPLE 12

2-(Isoxazol-4-yl)benzenesulfonyl chloride

By the procedure of Example 4, a diazonium salt was prepared by reacting 12 g of 4-(2-aminophenyl)isoxazole prepared in Example 11 with 5.5 g of sodium nitrite and 27 ml of concentrated hydrochloric acid in 80 ml of glacial acetic acid. The diazonium salt suspension was added to a suspension of 3.2 g cupric chloride dihydrate and 23 ml of sulfur dioxide in 57 ml of glacial acetic acid to give, after addition of excess water and filtration and suction drying, 15 g of the title compound as a crude solid; m.p. 100-104°C.

EXAMPLE 13

2-(Isoxazol-4-yl)benzenesulfonamide

By the procedure of Example 5, 15 g of 2-(isoxazol-4-yl)benzenesulfonyl chloride prepared in Example 12 was reacted with 30 ml of concentrated ammonium hydroxide in 150 ml of tetrahydrofuran to give, after recrystallization from 2-propanol, 11 g of the title compound; m.p. 165-167°C.

Anal. Calcd. for $C_9H_8N_2O_3S$:
C, 48.2; H, 3.6; N, 12.5.
Found:
C, 47.8; H, 3.7; N, 12.7.

EXAMPLE 14

N-[(4,6-Dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide

A solution of 2 g of 2-(isoxazol-4-yl)benzenesulfonamide, prepared in Example 13, in 100 ml of methylene chloride was purged with nitrogen. To the solution was added carefully 0.8 g of trimethylaluminum (5.5 ml of 2M solution in toluene) while cooling the flask at 15° to 30°C. After stirring about 0.2 hour, 1.9 g of methyl (4,6-dimethoxypyrimidin-2-yl)carbamate, prepared in Example 9, was added and the suspension was refluxed for 20 hours under a nitrogen atmosphere. The suspension was cooled in an ice-water bath while 50 ml of 25% aqueous acetic acid was carefully added. Excess water was added and the organic phase was separated and dried over sodium sulfate. After evaporation of the solvent *in vacuo*, the residue was triturated with 25 ml of ethyl acetate to give 0.5 g of the title compound; m.p. 175-178°C.

Anal. Calcd. for $C_{16}H_{15}N_5O_6S$:
C, 47.4; H, 3.7; N, 17.3.
Found:
C, 46.8; H, 3.7; N, 17.2.

EXAMPLE 15

N-[(4,6-Dichloro-1,3,5-triazin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide

To 4.5 g of 2-(isoxazol-4-yl)benzenesulfonamide prepared in Example 13 in 20 ml of acetonitrile is added 4 g of dichloro-5-triazinyl isocyanate. The mixture was refluxed for 2 hours, then is cooled to room temperature and filtered. The resulting solid is washed with diethyl ether to give N-[(4,6-dichloro-1,3,5-triazin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide.

EXAMPLE 16

N-[(4,6-Dimethyoxy-1,3,5-triazin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide

To 1 g of sulfonamide prepared in Example 15 in 10 ml of methanol is added portionwise 0.41 g of sodium methoxide. After an initial exotherm, the suspension is stirred at 25°C to 50°C for about 1 hour, then cooled to room temperature and diluted with excess water (about 30 ml). The solution is acidified to a pH of about 1 with concentrated HCl. The resulting mixture is filtered and suction dried to give the title compound.

EXAMPLE 17

1-Methyl-4-(2-nitrophenyl)-1H-pyrazole

A solution of 60 g of 3-(dimethylamino)-2(2-nitrophenyl)acrolein (see Example 10) and 16.1 g of methylhydrazine in 200 ml of toluene was refluxed for 10 hours, then concentrated to dryness *in vacuo*. The residue was recrystallized from 1-chlorobutane/hexane (about 1:1) to yield 51 g of the title compound; m.p. 41-44°C.

EXAMPLE 18

1-Methyl-4-(2-aminophenyl)-1H-pyrazole

By the procedure of Example 3, 25 g of 1-methyl-4-(2-nitrophenyl)-1H-pyrazole prepared in Example 17 was reacted with 81.2 g of stannous chloride dihydrate in 200 ml concentrated HCl at reflux for 1 hour to yield, after similar work up, 18 g of the title compound as an oil.

Anal. Calcd. for: $C_{10}H_{11}N_3$:
C, 69.3, H, 6.4; N, 24.2.
Found:
C, 68.8; H, 6.2; N, 23.9.

EXAMPLE 19

2-(1-Methyl-1H-pyrazol-4-yl)benzenesulfonamide

By the procedure of Example 4, 17.3 g of 1-methyl-4-(2-aminophenyl)-1H-pyrazole prepared in Example 18 was diazotized with 7.2 g of sodium nitrite and 36 ml of concentrated HCl in 104 ml of glacial acetic acid. The diazonium salt suspension was added to a suspension containing 4.3 g of cupric chloride dihydrate, 30 ml of sulfur dioxide and 76 ml of acetic acid. After completion of the reaction and addition of water, the suspension was extracted with 1-chlorobutane. The extraction was dried over sodium sulfate and concentrated *in vacuo* at less than 40°C to yield 22 g of crude 2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonyl chloride as an oil.

By the procedure of Example 5, 21 g of the above oil was reacted with 30 ml of concentrated NH_4OH in 150 ml of tetrahydrofuran to yield a solid. The solid was recrystallized from acetonitrile to yield 17 g of 2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide; m.p. 183-186°C.

Anal. Calcd. for $C_{10}H_{11}N_3O_2S$:
C, 50.6; H, 4.7; N, 17.7.
Found:
C, 50.8; H, 4.7; N, 18.0.

EXAMPLE 20

N-[(4-Methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide

By the procedure of Example 14, 2 g of 2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide, prepared in Example 19, was reacted in 100 ml of methylene chloride with 0.7 g of trimethylaluminum (5 ml of 2M toluene solution) followed by 1.7 g of methyl (4-methoxy-6-methyl-1,3,5-triazine-2-yl)carbamate. After work up and evaporation of the methylene chloride solvent, the residue was triturated with ethyl acetate to yield 0.6 g of the title compound; m.p. 221-224°C. The IR spectrum showed a carbonyl absorption at 1700 cm^{-1} indicative for a sulfonylurea.

EXAMPLE 21

N-[2-[3-(Dimethylamino)-1-oxo-2-propenyl]phenyl]acetamide

A suspension of 48 g of N-[2-(1-oxo-1-ethanyl)phenyl]acetamide and 38 g of N,N-dimethylformamide dimethyl acetal in 150 ml of toluene was heated at reflux for about 16 hours, then concentrated *in vacuo* to a solid. The solid was recrystallized from 1-chlorobutane to yield 40 g of the title compound: m.p. 90-95°C.

Anal. Calcd. for $C_{13}H_{16}N_2O_2$:
C, 67.2; H, 6.9; N, 12.0.
Found:
C, 66.8; H, 6.8; N, 11.7.

EXAMPLE 22

N-[2-(1- and/or 2-methyl-1H-pyrazol-3-yl)phenyl]acetamide

A suspension of 40 g of N-[2-[3-(dimethylamino)-1-oxo-2-propenyl]phenyl]acetamide prepared in Example 21 and 9.2 g of methylhydrazine in 130 ml of ethanol was heated at reflux for 10 hours, then concentrated *in vacuo*. The residue was recrystallized from 1-chlorobutane to yield 30 g of the title compound; m.p. 102-106°C.

Anal. Calcd. for $C_{12}H_{13}N_3O$:
C, 66.9; H, 6.1; N, 19.5.
Found:
C, 66.0; H, 6.0; N, 18.5.

EXAMPLE 23

1- and/or 2-Methyl-1H-3-(2-aminophenyl)pyrazole

A suspension of 30 g of the acetamide prepared in Example 22 in 75 ml of concentrated HCl was stirred and heated at reflux for 1 hour, then cooled to 10°C and filtered. The solid isolated was added to about 300 ml of ice-water, and the solution was made basic to a pH of about 8 by addition of 50% NaOH to yield an oil. After extracting the aqueous suspension with diethyl ether, the ether extract was concentrated *in vacuo* to yield an oil that solidified on cooling. The solid was recrystallized from 1-chlorobutane to yield 20 g of the title compound; m.p. 82-86°C.

Anal. Calcd. for $C_{10}H_{11}N_3$:
C, 69.3; H, 6.4; N, 24.2
Found:
C, 69.0; H, 6.3; N, 23.9.

EXAMPLE 24

2-(1- and/or 2-Methyl-1H-pyrazol-3-yl)benzenesulfonamide

By the procedure of Example 4, 20 g of amine prepared in Example 23 was diazotized with 8.4 g of sodium nitrite and 42 ml of concentrated HCl in 118 ml of glacial acetic acid. The diazonium salt suspension was added to a suspension containing 4.9 g of cupric chloride dihydrate, 36 ml of sulfur dioxide and 89 ml of acetic acid. After completion of reaction and addition of water, a precipitate formed. The mixture was filtered and the solid obtained was dissolved in methylene chloride and dried over sodium sulfate. Concentration of the methylene chloride solution *in vacuo* yielded 27 g of crude 2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonyl chloride; m.p. 108-115°C.

By the procedure of Example 5, 27 g of the above solid was reacted with about 40 ml of concentrated NH_4OH in 200 ml of tetrahydrofuran to yield a solid after work up. The solid was recrystallized from 2-propanol to yield 17 g of the title compound; m.p. 174-177°C.

Anal. Calcd. for $C_{10}H_{11}N_3O_2S$:
C, 50.6; H, 4.7; N, 17.7.
Found:
C, 50.5; H, 4.6; N, 17.6.

EXAMPLE 25

N-[(4,6-Dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide

By the procedure of Example 14, 2 g of the sulfonamide prepared in Example 24, was reacted with 0.7 g of trimethylaluminum (4.8 ml of 2M toluene solution) and 1.6 g of methyl (4,6-dimethoxypyrimidin-2-yl)carbamate in 100 ml of methylene chloride under a nitrogen atmosphere. After evaporation of the methylene chloride solvent *in vacuo*, the residue was recrystallized from acetonitrile to yield 0.8 g of the title compound; m.p. 210-212°C.

Anal. Calcd. for $C_{17}H_{18}N_6O_5S$:
Found, 48.8; H, 4.3; N, 20.1.
Found:
C, 49.4; H, 4.5; N, 21.4.

EXAMPLE 26

2-(1H-Pyrazol-1-yl)benzenesulfonamide

A. To a solution of 52 g of 1-(2-bromophenyl)pyrazole in 230 ml of diethyl ether under a nitrogen atmosphere and cooled at −70°C was added dropwise 175 ml of a 1.6M solution of *n*-butyl lithium in hexane. The suspension was allowed to warm from −70°C to −25°C on its own, then a solution of 37 ml of sulfuryl chloride in 69 ml of hexane was added dropwise at −25° to −20°C. After allowing the suspension to warm to room temperature, the suspension was stirred for 5 hours, then filtered to yield 38 g of crude 2-(1H-pyrazol-1-yl)benzenesulfonyl chloride which was contaminated with inorganic salts.

B. To a suspension of the above solid in 200 ml of tetrahydrofuran cooled at 10-20°C with an ice bath was added dropwise 50 ml of concentrated aqueous ammonium hydroxide. After adding 25 ml of water to dissolve the salts present, the suspension was stirred at 25°C for 10 hours, then concentrated *in vacuo*. After adding water to the residue, the mixture was filtered and the residue was recrystallized from 2-propanol to yield 11.2 g of the title compound; m.p. 167-170°C.

Anal. Calcd. for $C_9H_9N_3O_2S$:
C, 48.4; H, 4.1; N, 18.8.
Found:
C, 48.1; H, 4.1; N, 18.9.

NMR ($CDCl_3$ - DMSO)δ: 6.5 (t, 1H, NC=CHC=N);
7.3 (br, 2H, SO_2NH_2); and
7.3-8.2 (m, 6H, arom -
NCH =C—CHN).

EXAMPLE 27

N-[(4-Methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(1H-pyrazol-1-yl)benzenesulfonamide

By the procedure of Example 14, 1.3 g of the sulfonamide prepared in Example 26 was reacted in 100 ml of methylene chloride with 0.5 g of trimethylaluminum (3.5 ml of 2M toluene solution) followed by 1.3 g of methyl (4-methoxy-6-methylpyrimidin-2-yl)carbamate. After work-up and evaporation of the methylene chloride solvent, the residue was triturated with warm ethyl acetate to yield 1 g of the title compound; m.p. 200-204°C.

Anal. Calcd. for $C_{16}H_{16}N_6O_4S$:
C, 49.5; H, 4.2; N, 21.6.
Found:
C, 48.8; H, 4.2; N, 21.3.

EXAMPLE 28

Preparation of 2-(2-oxazolyl)benzenamine

To 19.0 g of 2-(2-nitrophenyl)oxazole, (prepared by the procedure of W. E. Cass, *J. Am. Chem. Soc., 64,* 785 [1942]) 3.0 g of Raney-Nickel catalyst (purchased from Aldrich Chemical Company, Milwaukee, Wisc. 53201) and 200 ml ethanol were combined and reduced at ambient temperature and at a H_2 pressure of 5-50 psi on a Paar Hydrogenator until the hydrogen uptake ceased. The catalyst was filtered through celite under a blanket of nitrogen. The solvent was removed on a rotary evaporator. 17.4 g of crude product with m.p. 31-32° was obtained and used in the next step.

EXAMPLE 29

Preparation of 2-(2-oxazolyl)benzenesulfonamide

Diazotization

To 100 ml H_2O and 180 ml of concentrated HCl was added 40.0 g of 2-(2-oxazolyl)benzenamine. The mixture was cooled to 0-5°. To this mixture was added dropwise a solution of 18.6 g $NaNO_2$ in 30 ml of water, keeping the temperature in the range of 0-5° during the addition. The reaction mixture was stirred for 15 minutes after the nitrite addition was complete.

Coupling (sulfonyl chloride formation)

A second flask was charged with 100 ml glacial acetic acid, 100 ml concentrated HCl and 4.3 g $CuCl_2$. The mixture was cooled to 0-10°C.

The diazonium salt was added dropwise to the coupling mixture during a period of 15 minutes while maintaining the temperature between 0-10°C. The progress of the reaction was monitored by nitrogen evolution via. a bubbler attached to the reaction vessel. When nitrogen evolution ceased (approximately 2 hours reaction time), the reaction mixture was poured into 1500 ml ice-water. The crude solid sulfonyl chloride was filtered and converted directly to the sulfonamide in the next step. A small dried sample had a m.p. of 88-89°C.

Amination (sulfonamide formation)

The crude solid product from the previous step was added to a solution of 400 ml tetrahydrofuran containing excess aqueous ammonia and cooled to 10-15°. The reaction mixture was stirred for 30 minutes. 300-400 ml of water was added to the reaction mixture, which was then extracted several times with CH_2Cl_2. The CH_2Cl_2 extracts were combined and back extracted with water, then dried with anhydrous $MgSO_4$. The solution of the product was filtered and the solvent removed on a rotary evaporator yielding 36.4 g of crude sulfonamide with m.p. 139-140°C.

EXAMPLE 30

Preparation of N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide

Under N_2, 5.5 ml of 2M$(CH_3)_3Al$ in toluene (purchased from Aldrich Chemical Co., Milwaukee, Wisc.) was added to 2.2 g (.01m) of 2-(2-oxazolyl)benzenesulfonamide dissolved in 80 ml dry CH_2Cl_2. The reaction was stirred for 15 minutes after the addition. 2.1 g of solid 4-methoxy-6-methylpyrimidin-2-yl)carbamic acid, methyl ester, (prepared by procedures described previously) was added. The reaction was stirred at ambient temperature for one hour and then refluxed for approximately 16 hours. After cooling to 10°C, 50 ml of water was added cautiously, dropwise, followed by enough glacial HOAc to lower the pH of the reaction mixture to 3.0. The crude reaction mixture was extracted several times with CH_2Cl_2. The extracts were dried with anhydrous Na_2SO_4, filtered, and the solvent stripped on a rotary evaporator. The crude product was triturated with *n*-butyl chloride-ethyl acetate 1:1. The resulting solid was filtered yielding 1.6 g product with m.p. 208-209°(d).

EXAMPLE 31

Preparation of 1-Methyl-2-(2-nitrophenyl)-1H-imidazole

Under N_2, 9.5 g of 2-(2-nitrophenyl)-1H-imidazole in a solution of 100 ml dimethylformamide and 25 ml of anhydrous tetrahydrofuran cooled to 0-5°C, was treated with 2.3 g of a 50% mineral oil dispersion of NaH. After the N_2 evolution ceased, 8.5 g of CH_3I was added dropwise over a period of 15 minutes maintaining the temperature of 0-5°. The reaction mixture was allowed to warm to ambient temperature and then stirred for two hours. The reaction mixture was then poured into water. The aqueous suspension of the product was extracted several times with CH_2Cl_2. The extracts were combined and dried with anhydrous Na_2SO_4. The solvent was removed on a rotary evaporator. The last traces of dimethylformamide were removed under high vacuum; a waxy solid remained. The crude product was crystallized from CH_3CN. Two crops totaling 3.6 g were obtained. The m.p. was 89-90°.

EXAMPLE 32

Preparation of 2-(1-methyl-1H-imidazol-2-yl)benzenamine

28.0 g Nitro compound from the preceding step was added to 120 ml of concentrated HCl and cooled to 0-10°C. A solution of 96.6 g of $SnCl_2 \cdot 2H_2O$ in 120 ml concentrated HCl (cooled to 0-10°C) was added to the nitro compound. The reaction mixture was warmed to 50° on a steam bath to complete the reduction.

The reaction mixture was poured onto ice-water and the pH raised to 10-12 with 50% NaOH solution. The precipitated product was filtered and taken up in CH_2Cl_2. The organic solution of the crude product was dried with anhydrous Na_2SO_4. The solvent was removed on a rotary evaporator yielding 16.9 g tan product with m.p. 152-153°.

EXAMPLE 33

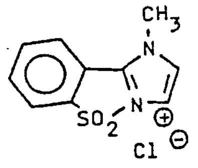
Preparation of 2-(1-methyl-1H-imidazol-2-yl)benzenesulfonamide

Diazotization

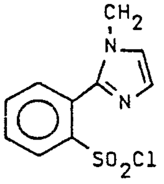
16.0 g of Amino compound from the preceding step was added to 100 ml of concentrated HCl and cooled to 0-5°. A solution of 6.7 g $NaNO_2$ in 15 ml of water was added dropwise maintaining the temperature at 0-5°. The reaction mixture was stirred 10 minutes after the addition was completed.

Coupling

Into a vessel cooled to 0-5°C were charged 50.0 ml HCl, .16 g $CuCl_2$ and 21.5 ml SO_2. The cooled diazonium salt from the preceding step was added dropwise over a period of 10-15 minutes keeping the temperature between 0-5°C. The reaction was stirred for 1 hour at 0-5°C. A yellow precipitate formed which was filtered. A small sample of this product was crystallized from CH_3CN and had a m.p. of 215-218°C. The spectral data and color are more consistent with the tricyclic salt



(1H-imidazol[1,2-β][1,2]benzisothizol-1-ium, 1-methyl-5,5-dioxide chloride) rather than the sulfonyl chloride



The salt was then aminated as described below.

Amination

The crude yellow solid from the preceding step was dissolved in water and enough concentrated aqueous NH_3 was added to raise the pH to 9.0. The reaction mixture was stirred at ambient temperature for about 30 minutes. The reaction mixture was poured into H_2O and then extracted with CH_2Cl_2, dried with anhydrous Na_2SO_4 and the solvent removed on a rotary evaporator. 4.5 g of crude product was obtained. The product was purified by dry column chromatography on silica gel with 1% EtOH/EtOAc. The purified product was washed from the silica with MeOH. The MeOH was removed on a rotary evaporator yielding 3.6 g of product sulfonamide with a m.p. of 158-163°.

EXAMPLE 34

Preparation of N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1-methyl-1H-imidazol-2-yl)benzenesulfonamide

Under N_2, 2.3 ml of a 2M solution of $(CH_3)_3Al$ was added dropwise to a solution of 1.0 g of sulfonamide from the preceding step in 40 ml of CH_2Cl_2. The mixture was stirred at ambient temperature for 15 minutes after which .89 g of (4,6-dimethoxypyrimidin-2-yl)carbamic acid, methyl ester was then added. The reaction was stirred at ambient temperature for 15 minutes and then heated to reflux for 16 hours.

The reaction mixture was cooled to 10° and 50 ml of water was added dropwise. The crude reaction mixture was extracted with CH_2Cl_2 several times. The organic extract of the product was dried with anhydrous Na_2SO_4, filtered and the solvent removed on a rotary evaporator. The crude product was triturated with EtOAc and filtered. .5 g of product with m.p. 182-183° was obtained.

EXAMPLE 35

Preparation of 2-(4,5-dihydro-4,4-dimethyloxazole-2-yl)benzenesulfonic acid, lithium salt

To 200 ml of dry tetrahydrofuran was added 16.3 g (.1M) of 2-phenyl-4,4-dimethyl-4,5-dihydrooxazole (prepared by the procedure taught in *J. Org. Chem., 28,* 2759 [1963]). The solution was cooled to −30° to −40° and 65 ml of 1.6 M solution (.105M) of butyl lithium was added dropwise while maintaining the temperature. After the addition the reaction was allowed to warm to 0°. The reaction was stirred for one hour at 0° and then cooled to −40°. While maintaining the temperature, 4.7 ml of sulfur dioxide was added dropwise. When the last drop of SO_2 was added, the reddish yellow solution of the lithium agent turned lemon yellow. The reaction was stirred for one hour after the addition and allowed to warm to room temperature. The solvent was stripped on a rotary evaporator leaving a solid residue which was triturated with anhydrous ether. The product was filtered and dried in a vacuum oven at 70° for 16 hours.

EXAMPLE 36

Preparation of 2-(4,5-dihydro-4,4-dimethyloxazol-2-yl)benzenesulfonamide

21.0 g of sulfinic acid from the preceding step was dissolved in 150 ml of anhydrous tetrahydrofuran and cooled to −5°. To this mixture was added a solution containing approximately an equivalent amount of chloramine prepared as follows: (see Coleman and Hauser, *J. Am. Chem. Soc., 50,* 1193 (1928). To 12.0 gms of aqueous NH_3 and 300 ml of ether cooled to 0-5°C was added 286 g of aqueous NaOCl1 (5.25%) also cooled to 0-5°C. The mixture was stirred for 15 minutes. The ether layer was separated, the aqueous phase was extracted with 100 ml of ether cooled to 0°. The ether extracts were cooled to −30° and dried with anhydrous $CaCl_2$. The resulting cold ethereal chloramine solution was added to the sulfinic acid maintaining the temperature at −30 to −50°. The reaction mixture was allowed to warm to 0 to −10°C and stirred for 30 minutes. The reaction mixture was then allowed to warm to ambient temperature over a period of one hour. The reaction mixture was extracted once with an aqueous $NaHSO_3$ solution and then dried and filtered. The ether was stripped on a rotary evaporator to yield an oil. Trituration with CH_2Cl_2 yielded 1.1 g of product with m.p. 142-146°. Dry column chromatography of the filtrate on silica gel with ethyl acetate yielded an additional 1.2 g of product with m.p. 138-142°.

EXAMPLE 37

2-(4,5-dihydro-4,4-dimethyloxazol-2-yl)-N-[(4,6-dichloro-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide

To 1.1 g of sulfonamide from the preceding step dissolved in 15 ml of anhydrous tetrahydrofuran under N_2 at ambient temperature was added a solution of .78 g of 4,6-dichloro-1,3,5-triazine isocyanate dissolved in 5 ml of anhydrous tetrahydrofuran. The reaction mixture was refluxed for 30 minutes. After cooling to ambient temperature, the solvent was removed on a rotary evaporator. The crude product was triturated with hexane and filtered yielding 1.7 g which was used in the following step without further purification.

EXAMPLE 38

Preparation of 2-(4,5-dihydro-4,4-dimethyloxazol-2-yl)-N-[(4,6-dimethoxy-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide

To 5 ml of CH_3OH was added .4 g of urea from the preceding step. Solid $NaOCH_3$ (.157 g) was added and the reaction mixture refluxed for 30 minutes. The reaction mixture was poured into 50 ml H_2O. The aqueous solution was extracted with CH_2Cl_2 and the organic extract discarded. The pH was lowered to 3.0 with HOAc. The aqueous solution was then extracted with CH_2Cl_2 and the extracts were combined, dried with anhydrous Na_2SO_4, filtered and the solvent was removed on a rotary evaporator yielding 300 mg of crude product, i.r. (nujol) 5.82μ (C=O).

EXAMPLE 39

Preparation of 5-[(2-phenylmethylthio)phenyl]oxazole

A mixture of 22.8 g 2-(phenylmethylthio)benzaldehyde (prepared by the procedure of G. W. Stacy, et al., *J. Org. Chem., 35,* 3495 (1970)), 19.5 g of *p*-toluenesulfonylmethylisocyanide, 13.8 g of anhydrous potassium carbonate and 150 ml methanol was heated at reflux for 3.75 hours. The reaction solution was cooled and the solvent evaporated under reduced pressure. The resulting oily solid was partitioned between water and ether. The organic phase was dried over anhydrous magnesium sulfate, filtered, and the solvent evaporated under reduced pressure. Distillation of the crude oil gave 16.4 g of the title compound as a viscous yellow-orange oil, bp 180-194° (0.7-1.0 mm); NMR ($CDCl_3$) δ 4.05 (s, 2H, CH_2), 7.2-7.6 (m, 9H, aromatic), 7.8 (s, 1H, CH) and 7.95 (s, 1H, CH).

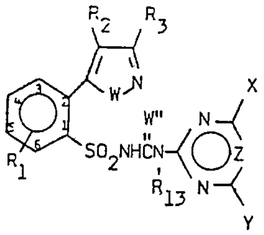
EXAMPLE 40

Preparation of 2-(oxazol-5-yl)benzenesulfonamide

A solution of 16.0 g of 5-[(2-phenylmethylthio)phenyl]oxazole in a mixture of 100 ml of acetic acid and 10 ml of water was cooled to 5° and 9.1 ml of chlorine was added portionwise while maintaining the temperature below 15°. When addition of chlorine was complete, the yellow-brown solution was stirred 15 minutes at 5-10° then poured into 600 ml of ice water. The aqueous mixture was extracted with ether. The organic solution was washed with water (3 × 250 ml) followed by brine (50 ml), then dried over anhydrous magnesium sulfate, filtered and the solvent removed under reduced pressure. The crude reaction product was dissolved in 150 ml of tetrahydrofuran and cooled to 0-5°. Ammonium hydroxide (8.0 ml) was added dropwise. The red-orange suspension was stirred 20 minutes at 0-5° then allowed to warm to ambient temperature and stirred until TLC (Hexane/ethyl acetate-1:1) showed no sulfenyl chloride remained. The solvent was removed under reduced pressure and the resulting oily solid triturated with 250 ml of cold water. The resulting oily solid was collected and washed sequentially with water, butyl chloride/hexane (1:1), isopropyl alcohol and butyl chloride giving 7.1 g of the title compound as a slightly yellow solid, m.p. 157-163°; ir (mull) 3420 and 3210 cm^{-1} (NH_2).

Using the techniques described in Equations 1 to 96 and Examples 1 to 40, or simple modifications thereof, the following compounds in Tables 1-15 may be made by one skilled in the art.

TABLE 1



R_1	R_2	R_3	R_{13}	X	Y	Z	W	W''	m.p.(°C)
H	H	H	H	CH_3	CH_3	CH	O	O	180-185°
H	H	H	H	CH_3	OCH_3	CH	O	O	185-189°
H	H	H	H	CH_3	CH_3	N	O	O	183-185°
H	H	H	H	CH_3	OCH_3	N	O	O	164-172°
H	H	H	H	OCH_3	OCH_3	N	O	O	168-173°
H	H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	H	OCH_3	CH_3	CH	S	O	
H	H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	H	CH_3	CH_3	N	S	O	
H	H	H	H	OCH_3	CH_3	N	S	O	
H	H	H	H	OCH_3	OCH_3	N	S	O	
5-F	H	H	H	OCH_3	OCH_3	CH	O	O	
6-Cl	H	H	H	OCH_3	OCH_3	CH	O	O	
4-Br	H	H	H	OCH_3	OCH_3	CH	O	O	
3-CH_3	H	H	H	CH_3	OCH_3	N	O	O	
5-CF_3	H	H	H	OCH_3	OCH_3	N	O	O	
5-OCH_3	H	H	H	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	OCH_3	CH_3	N	O	O	
H	CH_3	H	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	H	H	CH_3	OCH_3	CH	O	O	
H	CH_3	H	H	CH_3	CH_3	CH	O	O	
H	CH_3	H	H	CH_3	OCH_3	N	O	O	

TABLE 1 (continued)

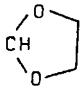
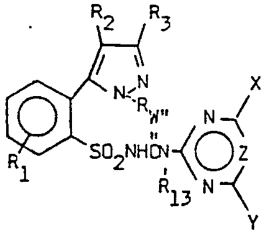
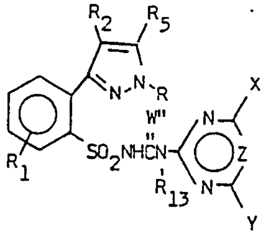
R_1	R_2	R_3	R_{13}	X	Y	Z	W	W''	m.p.(°C)
H	C_2H_5	H	H	OCH_3	OCH_3	CH	O	O	
H	C_2H_5	H	H	CH_3	OCH_3	CH	O	O	
H	C_2H_5	H	H	CH_3	CH_3	CH	O	O	
H	C_2H_5	H	H	OCH_3	CH_3	N	O	O	
H	Cl	H	H	OCH_3	OCH_3	CH	O	O	
H	Br	H	H	OCH_3	OCH_3	CH	O	O	
H	H	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	H	CH_3	H	CH_3	OCH_3	CH	O	O	
H	H	CH_3	H	CH_3	CH_3	CH	O	O	
H	H	CH_3	H	OCH_3	CH_3	N	O	O	
H	H	C_2H_5	H	OCH_3	OCH_3	CH	O	O	
H	H	C_2H_5	H	CH_3	OCH_3	CH	O	O	
H	H	C_2H_5	H	CH_3	CH_3	CH	O	O	
H	H	C_2H_5	H	CH_3	OCH_3	N	O	O	
H	H	Cl	H	OCH_3	OCH_3	CH	S	O	
H	H	Br	H	OCH_3	CH_3	CH	S	O	
H	H	OCH_3	H	OCH_3	OCH_3	CH	S	O	
H	H	OC_2H_5	H	CH_3	OCH_3	CH	S	O	
H	H	SCH_3	H	CH_3	CH_3	CH	S	O	
H	H	H	H	Cl	NH_2	CH	O	O	
H	H	H	H	Cl	OCH_3	CH	O	O	
H	H	H	H	Cl	$NHCH_3$	CH	O	O	
H	H	H	H	Cl	$N(CH_3)_2$	CH	O	O	
H	H	H	H	CH_3	CH_2OCH_3	CH	O	O	
H	H	H	H	CH_3	C_2H_5	CH	O	O	
H	H	H	H	CH_3	OC_2H_5	CH	O	O	
H	H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	CH_3	H	OCH_3	CH_3	N	O	O	
H	CH_3	CH_3	H	OCH_3	CH_3	CH	O	O	
H	H	CH_3	H	OCH_3	OCH_3	N	O	O	
H	H	CH_3	H	CH_3	CH_3	N	O	O	
H	CH_3	H	H	OCH_3	OCH_3	N	O	O	
H	CH_3	H	H	CH_3	CH_3	N	O	O	
H	CH_3	CH_3	H	CH_3	CH_3	CH	O	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	N	O	O	
H	CH_3	CH_3	H	CH_3	CH_3	N	O	O	
H	H	C_2H_5	H	OCH_3	OCH_3	N	O	O	
H	H	C_2H_5	H	CH_3	CH_3	N	O	O	
H	C_2H_5	H	H	OCH_3	OCH_3	N	O	O	
H	C_2H_5	H	H	CH_3	CH_3	N	O	O	
H	H	H	CH_3	CH_3	OCH_3	CH	O	O	
H	H	H	CH_3	CH_3	CH_3	CH	O	O	
H	H	H	CH_3	OCH_3	OCH_3	N	O	O	
H	H	H	CH_3	CH_3	CH_3	N	O	O	
H	H	H	H	OCH_3	OCH_3	CH	O	O	183-187°
H	H	H	H	OCH_3	OCH_3	CH	O	S	
H	H	H	H	OCH_3	CH_3	N	O	S	
H	H	H	H	CH_3	OC_2H_5	CH	O	S	
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N	O	O	
H	H	H	H	CH_3	OCH_2CF_3	N	O	O	
H	H	H	H	OCH_3	SCH_3	N	O	O	
H	H	H	H	CH_3	CF_3	CH	O	O	
H	H	H	H	CH_3	 158	CH	O	O	
H	H	H	H	Cl	OC_2H_5	CH	O	O	

TABLE 1a



and/or



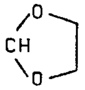
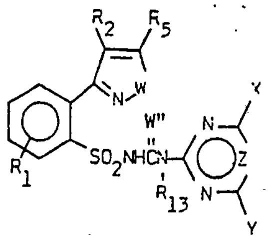
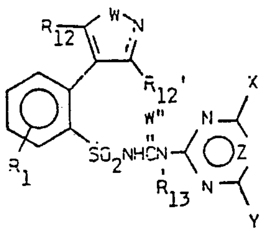
R	R_1	R_2	R_3	R_5	R_{13}	X	Y	Z	W''	m.p.(°C)
CH_3	H	H	H	H	H	CH_3	CH_3	CH	O	212-216°
CH_3	H	H	H	H	H	CH_3	OCH_3	CH	O	207-211°
CH_3	H	H	H	H	H	OCH_3	OCH_3	N	O	190-195°
CH_3	H	H	H	H	H	CH_3	CH_3	N	O	192-197°
CH_3	H	H	H	H	H	CH_3	OCH_3	N	O	202-205°
H	H	H	H	H	H	OCH_3	OCH_3	CH	O	
C_2H_5	H	H	H	H	H	CH_3	OCH_3	CH	O	
n-propyl	H	H	H	H	H	OCH_3	OCH_3	CH	O	
$CH(CH_3)_2$	H	H	H	H	H	OCH_3	OCH_3	CH	O	
CH_3	H	H	H	H	CH_3	OCH_3	OCH_3	CH	O	
CH_3	H	CH_3	H	H	H	OCH_3	OCH_3	CH	O	
CH_3	H	CH_3	H	H	H	OCH_3	CH_3	CH	O	
CH_3	H	CH_3	H	H	H	OCH_3	CH_3	N	O	
CH_3	H	H	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	
CH_3	H	H	CH_3	CH_3	H	CH_3	OCH_3	CH	O	
CH_3	H	H	CH_3	CH_3	H	OCH_3	CH_3	N	O	
CH_3	H	CH_3	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	
CH_3	H	CH_3	CH_3	CH_3	H	OCH_3	CH_3	N	O	
CH_3	H	C_2H_5	H	H	H	OCH_3	OCH_3	CH	O	
CH_3	H	H	CH_3	CH_3	H	CH_3	CH_3	CH	O	
CH_3	H	H	CH_3	CH_3	H	OCH_3	OCH_3	N	O	
CH_3	H	H	CH_3	CH_3	H	CH_3	CH_3	N	O	
CH_3	H	CH_3	H	H	H	CH_3	CH_3	CH	O	
CH_3	H	CH_3	H	H	H	OCH_3	OCH_3	N	O	
CH_3	H	CH_3	H	H	H	CH_3	CH_3	N	O	
CH_3	H	H	C_2H_5	C_2H_5	H	OCH_3	OCH_3	CH	O	
CH_3	H	H	C_2H_5	C_2H_5	H	OCH_3	CH_3	CH	O	
CH_3	H	H	C_2H_5	C_2H_5	H	OCH_3	CH_3	N	O	
CH_3	H	H	C_2H_5	C_2H_5	H	CH_3	CH_3	CH	O	
CH_3	H	H	C_2H_5	C_2H_5	H	OCH_3	OCH_3	N	O	
CH_3	H	H	H	H	H	OCH_3	OCH_3	CH	O	210-212°
CH_3	H	H	H	H	H	Cl	OCH_3	CH	O	
$CH_2(CH_2)_2CH_3$	H	H	H	H	H	OCH_3	OCH_3	CH	O	
CH_3	H	H	H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	
CH_3	H	H	H	H	H	OCH_3	CH_2OCH_3	CH	O	
CH_3	H	H	H	H	H	CH_3	OC_2H_5	CH	O	
CH_3	H	H	H	H	H	OCH_3		CH	O	
CH_3	H	H	H	H	H	OCH_3	OCH_3	CH	S	

TABLE 1b



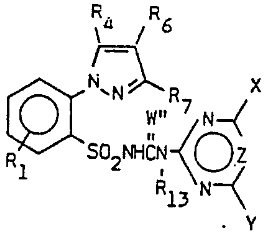
R_1	R_2	R_5	R_{13}	X	Y	Z	W	W"	m.p.(°C)
H	H	H	H	CH_3	OCH_3	CH	O	O	
H	H	H	H	CH_3	CH_3	CH	O	O	
H	H	H	H	OCH_3	OCH_3	CH	O	O	189-193°
H	H	H	H	CH_3	CH_3	N	O	O	
H	H	H	H	OCH_3	OCH_3	N	O	O	
H	H	H	H	CH_3	OCH_3	N	O	O	
H	H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	H	OCH_3	CH_3	CH	S	O	
H	H	H	H	CH_3	CH_3	N	S	O	
H	H	H	H	OCH_3	OCH_3	N	S	O	
H	H	H	H	CH_3	OCH_3	N	S	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	O	O	
H	H	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	H	CH_3	H	OCH_3	CH_3	CH	O	O	
H	H	CH_3	H	CH_3	CH_3	CH	O	O	
H	H	CH_3	H	OCH_3	CH_3	N	O	O	
H	H	C_2H_5	H	OCH_3	OCH_3	CH	O	O	
H	H	C_2H_5	H	OCH_3	CH_3	CH	O	O	
H	H	C_2H_5	H	CH_3	CH_3	CH	O	O	
H	H	C_2H_5	H	OCH_3	CH_3	N	O	O	
H	H	OCH_3	H	OCH_3	OCH_3	CH	O	O	
H	H	OCH_3	H	CH_3	OCH_3	CH	O	O	
H	H	OCH_3	H	CH_3	CH_3	CH	O	O	
H	H	OCH_3	H	OCH_3	CH_3	N	O	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	CH_3	H	OCH_3	CH_3	CH	O	O	
H	H	Cl	H	OCH_3	OCH_3	CH	S	O	
H	H	Br	H	OCH_3	CH_3	CH	S	O	
H	H	OCH_3	H	OCH_3	OCH_3	CH	S	O	
H	H	OC_2H_5	H	OCH_3	CH_3	CH	S	O	
H	H	SCH_3	H	CH_3	CH_3	CH	S	O	
H	H	CH_3	H	OCH_3	OCH_3	N	O	O	
H	H	CH_3	H	CH_3	CH_3	N	O	O	
H	H	OCH_3	H	OCH_3	OCH_3	N	O	O	
H	H	OCH_3	H	CH_3	CH_3	N	O	O	
H	H	C_2H_5	H	OCH_3	OCH_3	N	O	O	
H	H	H	H	Cl	OCH_3	CH	O	O	
H	H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	O	
H	H	H	H	OCH_3	OCH_3	CH	O	S	

TABLE 1c



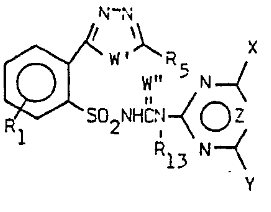
R_1	R_{12}	R_{12}'	R_{13}	X	Y	Z	W	W"	m.p.(°C)
H	H	H	H	CH_3	CH_3	CH	O	O	197-202°
H	H	H	H	OCH_3	CH_3	CH	O	O	181-185°
H	H	H	H	CH_3	CH_3	N	O	O	187-192°
H	H	H	H	OCH_3	OCH_3	N	O	O	180-185°
H	H	H	H	CH_3	OCH_3	N	O	O	178-181°
H	CH_3	H	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	H	H	OCH_3	CH_3	CH	O	O	
H	CH_3	H	H	OCH_3	CH_3	N	O	O	
H	CH_3	H	H	CH_3	CH_3	CH	O	O	
H	CH_3	CH_3	H	CH_3	CH_3	CH	O	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	CH_3	H	OCH_3	CH_3	CH	O	O	
H	CH_3	CH_3	H	OCH_3	CH_3	N	O	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	CH_3	OCH_3	CH	O	O	
H	H	H	H	CH_3	CH_2OCH_3	CH	O	O	
H	H	H	H	CH_3	OC_2H_5	CH	O	O	
H	H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	O	
H	H	H	H	CH_3	C_2H_5	CH	O	O	
H	H	H	H	Cl	OCH_3	CH	O	O	
H	H	H	H	Cl	NH_2	CH	O	O	
H	H	H	H	Cl	$NHCH_3$	CH	O	O	
H	H	H	H	Cl	$N(CH_3)_2$	CH	O	O	
H	H	H	H	CH_3	CH_3	CH	NCH_3	O	218-222°
H	H	H	H	OCH_3	CH_3	CH	NCH_3	O	222-226°
H	H	H	H	OCH_3	OCH_3	CH	NCH_3	O	220-226°
H	H	H	H	CH_3	CH_3	N	NCH_3	O	232-237°
H	H	H	H	OCH_3	OCH_3	N	NCH_3	O	195-200°
H	CH_3	H	H	OCH_3	OCH_3	CH	NCH_3	O	
H	CH_3	H	H	OCH_3	CH_3	CH	NCH_3	O	
H	CH_3	H	H	CH_3	CH_3	CH	NCH_3	O	
H	CH_3	H	H	OCH_3	CH_3	N	NCH_3	O	
H	CH_3	CH_3	H	CH_3	CH_3	CH	NCH_3	O	
H	CH_3	CH_3	H	OCH_3	CH_3	CH	NCH_3	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	CH	NCH_3	O	
H	CH_3	CH_3	H	CH_3	OCH_3	N	NCH_3	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	N	NCH_3	O	
H	H	H	H	CH_3	CH_2OCH_3	CH	NCH_3	O	
H	H	H	H	CH_3	OC_2H_5	CH	NCH_3	O	
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH	NCH_3	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	NCH_3	O	
H	H	H	CH_3	CH_3	OCH_3	CH	NCH_3	O	
H	H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	H	OCH_3	CH_3	CH	S	O	
H	H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	H	CH_3	CH_3	N	S	O	
H	H	H	H	CH_3	OCH_3	N	S	O	
H	H	H	H	OCH_3	OCH_3	N	S	O	
H	CH_3	H	H	OCH_3	OCH_3	N	O	O	
H	CH_3	H	H	CH_3	CH_3	N	O	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	N	O	O	
H	CH_3	CH_3	H	CH_3	CH_3	N	O	O	
H	H	H	H	OCH_3	OCH_3	CH	O	O	175-178°
H	H	H	H	OCH_3	CH_3	N	NCH_3	O	221-224°

TABLE 1d



R_1	R_4	R_6	R_7	R_{13}	X	Y	Z	W''	m.p.(°C)
H	H	H	H	H	CH_3	CH_3	CH	O	198-202°
H	H	H	H	H	OCH_3	OCH_3	CH	O	194-198°
H	H	H	H	H	OCH_3	CH_3	CH	O	200-204°
H	H	H	H	H	OCH_3	OCH_3	N	O	188-192°
H	H	H	H	H	OCH_3	CH_3	N	O	184-188°
H	CH_3	H	CH_3	H	CH_3	CH_3	CH	O	
H	CH_3	H	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	H	OCH_3	CH_3	CH	O	
H	CH_3	H	CH_3	H	OCH_3	OCH_3	N	O	
H	CH_3	H	CH_3	H	OCH_3	CH_3	N	O	
H	CH_3	H	CH_3	H	CH_3	CH_3	N	O	
H	H	H	H	CH_3	OCH_3	OCH_3	CH	O	
H	CH_3	C_2H_5	CH_3	H	OCH_3	OCH_3	CH	O	
H	H	C_2H_5	H	H	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	CH_3	H	OCH_3	CH_3	CH	O	
H	CH_3	CH_3	CH_3	H	OCH_3	CH_3	N	O	
H	$CH_2CH_2CH_3$	H	H	H	OCH_3	OCH_3	CH	O	
H	H	H	$CH(CH_3)_2$	H	OCH_3	CH_3	N	O	
H	H	H	H	H	CH_3	CH_3	N	O	201-205°
H	C_2H_5	H	C_2H_5	H	OCH_3	OCH_3	CH	O	
H	$CH_2(CH_2)_2CH_3$	H	H	H	OCH_3	OCH_3	CH	O	
H	H	H	$CH_2(CH_2)_2CH_3$	H	OCH_3	OCH_3	CH	O	
H	H	H	H	H	Cl	OCH_3	CH	O	
H	H	H	H	H	OCH_3	C_2H_5	CH	O	
H	H	H	H	H	CH_3	OC_2H_5	CH	O	
H	H	H	H	H	OCH_3	CH_2OCH_3	CH	O	
H	H	H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	
H	H	H	H	H	CH_3	OC_2H_5	N	O	
H	H	H	H	H	OCH_3	OCH_3	CH	S	
H	H	H	H	H	OCH_3	SCH_3	CH	O	
H	H	H	H	H	CH_3	CF_3	CH	O	

TABLE 2



R_1	R_5	R_{13}	X	Y	Z	W'	W''	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	O	
H	H	H	OCH_3	OCH_3	CH	O	O	
H	H	H	OCH_3	CH_3	CH	O	O	
H	H	H	CH_3	CH_3	N	O	O	
H	H	H	OCH_3	OCH_3	N	O	O	
H	H	H	OCH_3	CH_3	N	O	O	

TABLE 2 (continued)

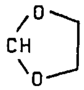
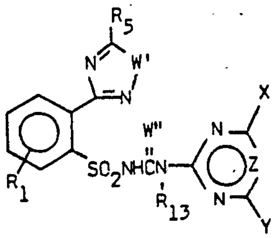
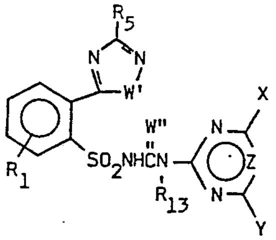
R_1	R_5	R_{13}	X	Y	Z	W'	W''	m.p.(°C)
H	CH_3	H	CH_3	CH_3	CH	O	O	209-212°
H	CH_3	H	OCH_3	OCH_3	CH	O	O	214-218°
H	CH_3	H	CH_3	OCH_3	CH	O	O	196-200°
H	CH_3	H	CH_3	CH_3	N	O	O	182-192°
H	CH_3	H	OCH_3	OCH_3	N	O	O	
H	CH_3	H	OCH_3	CH_3	N	O	O	186-190°
H	C_2H_5	H	CH_3	CH_3	CH	O	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	O	
H	C_2H_5	H	CH_3	OCH_3	CH	O	O	
H	C_2H_5	H	CH_3	CH_3	N	O	O	
H	C_2H_5	H	OCH_3	OCH_3	N	O	O	
H	C_2H_5	H	CH_3	OCH_3	N	O	O	
H	CH_3	H	Cl	NH_2	CH	O	O	
H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	OCH_3	CH_3	CH	S	O	
H	H	H	CH_3	CH_3	N	S	O	
H	H	H	OCH_3	OCH_3	N	S	O	
H	H	H	OCH_3	CH_3	N	S	O	
H	CH_3	H	CH_3	CH_3	CH	S	O	
H	CH_3	H	OCH_3	OCH_3	CH	S	O	227-230°
H	CH_3	H	OCH_3	CH_3	CH	S	O	
H	CH_3	H	CH_3	CH_3	N	S	O	
H	CH_3	H	OCH_3	OCH_3	N	S	O	
H	CH_3	H	CH_3	OCH_3	N	S	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	S	O	
H	C_2H_5	H	OCH_3	CH_3	CH	S	O	
H	C_2H_5	H	CH_3	OCH_3	N	S	O	
H	H	CH_3	OCH_3	OCH_3	CH	O	O	
5-F	CH_3	H	OCH_3	OCH_3	CH	O	O	
6-Cl	CH_3	H	OCH_3	OCH_3	CH	O	O	
4-Br	CH_3	H	OCH_3	CH_3	CH	O	O	
3-CH_3	CH_3	H	OCH_3	OCH_3	CH	O	O	
5-CF_3	CH_3	H	OCH_3	OCH_3	CH	O	O	
5-OCH_3	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	H	Cl	OCH_3	CH	O	O	
H	CH_3	H	Cl	$NHCH_3$	CH	O	O	
H	CH_3	H	Cl	$N(CH_3)_2$	CH	O	O	
H	CH_3	H	CH_3	CH_2OCH_3	CH	O	O	
H	CH_3	H	CH_3	C_2H_5	CH	O	O	
H	CH_3	H	CH_3	OC_2H_5	CH	O	O	
H	CH_3	H	OCH_3	$CH(OCH_3)_2$	CH	O	O	
H	C_2H_5	H	CH_3	CH_3	CH	S	O	
H	C_2H_5	H	OCH_3	OCH_3	N	S	O	
H	C_2H_5	H	CH_3	CH_3	N	S	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	S	
H	CH_3	H	OCH_3	CH_3	N	O	S	
H	CH_3	H	OCH_3		CH	O	O	

TABLE 2a



R_1	R_5	R_{13}	X	Y	Z	W'	W''	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	O	
H	H	H	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	OCH_3	CH	O	O	
H	H	H	CH_3	CH_3	N	O	O	
H	H	H	OCH_3	OCH_3	N	O	O	
H	H	H	CH_3	OCH_3	N	O	O	
H	CH_3	H	CH_3	CH_3	CH	O	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	CH_3	H	CH_3	OCH_3	CH	O	O	
H	CH_3	H	CH_3	CH_3	N	O	O	
H	CH_3	H	OCH_3	OCH_3	N	O	O	
H	CH_3	H	CH_3	OCH_3	N	O	O	
H	C_2H_5	H	CH_3	CH_3	CH	O	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	O	
H	C_2H_5	H	CH_3	OCH_3	CH	O	O	
H	C_2H_5	H	CH_3	OCH_3	N	O	O	
H	C_2H_5	H	OCH_3	OCH_3	N	O	O	
H	SCH_3	H	OCH_3	OCH_3	CH	S	O	
H	OCH_3	H	OCH_3	OCH_3	CH	S	O	
H	OC_2H_5	H	OCH_3	OCH_3	CH	S	O	
H	Cl	H	OCH_3	OCH_3	CH	S	O	
H	Br	H	OCH_3	CH_3	CH	S	O	
H	H	CH_3	OCH_3	CH_3	CH	O	O	
H	C_2H_5	H	CH_3	CH_3	N	O	O	
H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	CH_3	OCH_3	CH	S	O	
H	H	H	CH_3	CH_3	N	S	O	
H	H	H	OCH_3	OCH_3	N	S	O	
H	H	H	CH_3	OCH_3	N	S	O	
H	Cl	H	CH_3	CH_3	CH	S	O	
H	Cl	H	CH_3	OCH_3	CH	S	O	
H	Cl	H	CH_3	CH_3	N	S	O	
H	Cl	H	OCH_3	OCH_3	N	S	O	
H	Cl	H	CH_3	OCH_3	N	S	O	

TABLE 2b



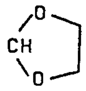
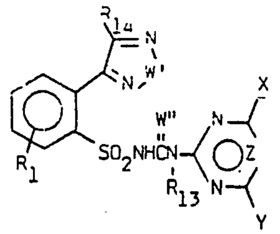
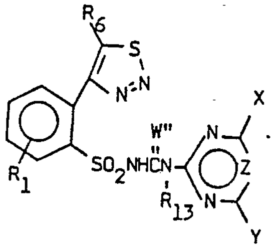
R_1	R_5	R_{13}	X	Y	Z	W'	W"	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	O	
H	H	H	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	OCH_3	CH	O	O	
H	H	H	CH_3	CH_3	N	O	O	
H	H	H	OCH_3	OCH_3	N	O	O	
H	H	H	OCH_3	CH_3	N	O	O	
N	CH_3	H	CH_3	CH_3	CH	O	O	202-206°
H	CH_3	H	OCH_3	OCH_3	CH	O	O	230-234°
H	CH_3	H	CH_3	OCH_3	CH	O	O	189-193°
H	CH_3	H	CH_3	CH_3	N	O	O	211-214°
H	CH_3	H	OCH_3	OCH_3	N	O	O	194-198°
H	CH_3	H	CH_3	OCH_3	N	O	O	188-192°
H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	CH_3	OCH_3	CH	S	O	
H	H	H	CH_3	CH_3	N	S	O	
H	H	H	OCH_3	OCH_3	N	S	O	
H	H	H	OCH_3	CH_3	N	S	O	
H	H	CH_3	OCH_3	OCH_3	CH	O	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	O	
H	C_2H_5	H	OCH_3	CH_3	CH	O	O	
H	C_2H_5	H	OCH_3	CH_3	N	O	O	
H	CH_3	H	OCH_3	OCH_3	CH	S	O	
H	CH_3	H	OCH_3	CH_3	CH	S	O	
H	CH_3	H	OCH_3	CH_3	N	S	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	S	
H	CH_3	H	CH_3	$OCH_2CH_2OCH_3$	N	O	O	
H	CH_3	H	CH_3	OCH_2CF_3	N	O	O	
H	CH_3	H	OCH_3	SCH_3	N	O	O	
H	CH_3	H	CH_3	CF_3	CH	O	O	
H	CH_3	H	OCH_3		CH	O	O	
H	CH_3	H	OCH_3	CH_2OCH_3	CH	O	O	
H	CH_3	H	CH_3	OC_2H_5	N	O	O	
H	CH_3	H	OCH_3	$CH(OCH_3)_2$	CH	O	O	

TABLE 2c



R_1	R_{13}	R_{14}	X	Y	Z	W'	W''	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	O	
H	H	H	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	OCH_3	CH	O	O	
H	H	H	CH_3	CH_3	N	O	O	
H	H	H	OCH_3	OCH_3	N	O	O	
H	H	H	OCH_3	CH_3	N	O	O	
H	H	Cl	OCH_3	OCH_3	CH	O	O	
H	H	Cl	CH_3	OCH_3	CH	O	O	
H	H	OCH_3	CH_3	CH_3	CH	S	O	
H	H	Cl	OCH_3	OCH_3	N	O	O	
H	H	Cl	CH_3	OCH_3	N	O	O	
H	H	OCH_3	OCH_3	OCH_3	CH	O	O	
H	H	OC_2H_5	OCH_3	OCH_3	CH	O	O	
H	H	SCH_3	OCH_3	OCH_3	CH	O	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	O	
H	H	H	CH_3	CH_3	CH	S	O	
H	H	H	CH_3	OCH_3	CH	S	O	
H	H	H	OCH_3	OCH_3	CH	S	O	
H	H	H	CH_3	CH_3	N	S	O	
H	H	H	CH_3	OCH_3	N	S	O	
H	H	H	OCH_3	OCH_3	N	S	O	
H	H	Cl	OCH_3	OCH_3	CH	S	O	
H	H	Cl	OCH_3	CH_3	CH	S	O	
H	H	Cl	OCH_3	CH_3	N	S	O	
H	H	OC_2H_5	OCH_3	OCH_3	CH	S	O	
H	H	SCH_3	OCH_3	OCH_3	CH	S	O	
H	H	CH_3	OCH_3	OCH_3	CH	O	O	
H	H	C_2H_5	OCH_3	OCH_3	CH	O	O	

TABLE 2d



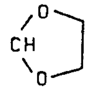
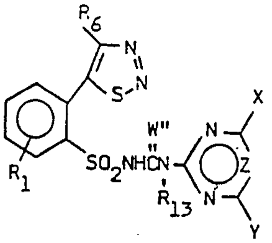
R_1	R_6	R_{13}	X	Y	Z	W''	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	189-192°
H	H	H	CH_3	OCH_3	CH	O	190-193°
H	H	H	OCH_3	OCH_3	CH	O	199-203°
H	H	H	CH_3	CH_3	N	O	190-194°
H	H	H	OCH_3	CH_3	N	O	174-177°
H	H	H	OCH_3	OCH_3	N	O	167-172°
H	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	OCH_3	N	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	OCH_3	N	O	
H	H	CH_3	OCH_3	OCH_3	CH	O	
H	H	H	OCH_3	OCH_3	CH	S	
H	H	H	Cl	OCH_3	CH	O	
H	H	H	OCH_3	CH_2OCH_3	CH	O	
H	H	H	CH_3	OC_2H_5	N	O	
H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	
H	H	H	OCH_3		CH	O	
H	H	H	OCH_3	SCH_3	N	O	
H	H	H	CH_3	CF_3	CH	O	
H	H	H	OCH_3	C_2H_5	CH	O	
H	H	H	Cl	NH_2	CH	O	

TABLE 2e



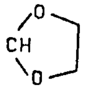
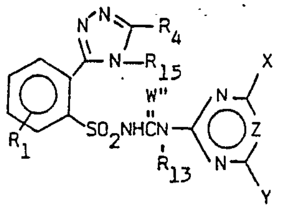
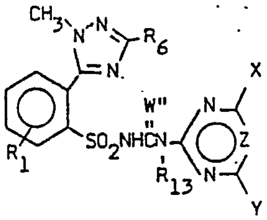
R_1	R_6	R_{13}	X	Y	Z	W''	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	
H	H	H	CH_3	OCH_3	CH	O	
H	H	H	CH_3	OCH_3	CH	O	
H	H	H	CH_3	CH_3	N	O	
H	H	H	OCH_3	CH_3	N	O	
H	H	H	OCH_3	OCH_3	N	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	OCH_3	N	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	OCH_3	N	O	
H	H	CH_3	OCH_3	OCH_3	CH	O	
H	H	H	OCH_3	OCH_3	CH	S	
H	H	H	Cl	OCH_3	CH	O	
H	H	H	OCH_3	CH_2OCH_3	CH	O	
H	H	H	CH_3	OC_2H_5	N	O	
H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	
H	H	H	OCH_3		CH	O	
H	H	H	OCH_3	SCH_3	N	O	
H	H	H	CH_3	CF_3	CH	O	
H	H	H	OCH_3	C_2H_5	CH	O	
H	H	H	Cl	NH_2	CH	O	

TABLE 3



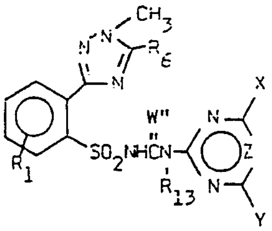
R_1	R_4	R_{13}	R_{15}	X	Y	Z	W''	m.p.(°C)
H	CH_3	H	CH_3	CH_3	CH_3	CH	O	
H	CH_3	H	CH_3	OCH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	CH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	CH_3	CH_3	N	O	
H	CH_3	H	CH_3	OCH_3	OCH_3	N	O	
H	CH_3	H	CH_3	CH_3	OCH_3	N	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	OCH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	OCH_3	CH_3	N	O	
H	$CH_2CH_2CH_3$	H	CH_3	OCH_3	OCH_3	CH	O	
H	$CH_2(CH_2)_2CH_3$	H	CH_3	OCH_3	CH_3	N	O	
H	CH_3	H	C_2H_5	OCH_3	OCH_3	CH	O	
H	CH_3	H	$CH_2CH_2CH_3$	OCH_3	OCH_3	CH	O	
H	CH_3	H	$CH(CH_3)_2$	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	CH_3	OCH_3	OCH_3	CH	O	

TABLE 3a



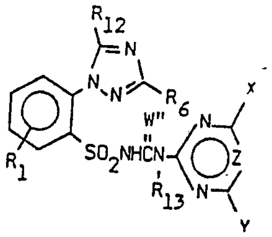
R_1	R_6	R_{13}	X	Y	Z	W''	m.p.(°C)
H	H	H	CH_3	CH_3	CH	O	
H	H	H	CH_3	OCH_3	CH	O	
H	H	H	OCH_3	OCH_3	CH	O	
H	H	H	CH_3	CH_3	N	O	
H	H	H	CH_3	OCH_3	N	O	
H	H	H	OCH_3	OCH_3	N	O	
H	CH_3	H	CH_3	CH_3	CH	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	CH_3	N	O	
H	CH_3	H	CH_3	OCH_3	N	O	
H	CH_3	H	OCH_3	OCH_3	N	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	OCH_3	CH	O	
H	C_2H_5	H	OCH_3	CH_3	N	O	
5-F	CH_3	H	OCH_3	OCH_3	CH	O	
6-Cl	CH_3	H	OCH_3	CH_3	CH	O	
4-Br	CH_3	H	OCH_3	OCH_3	CH	O	
3-CH_3	CH_3	H	OCH_3	OCH_3	CH	O	
5-CF_3	CH_3	H	OCH_3	OCH_3	CH	O	
5-OCH_3	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	OCH_3	OCH_3	CH	O	

TABLE 3b



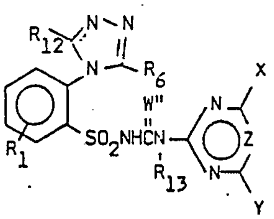
R_1	R_6	R_{13}	X	Y	Z	W''	m.p.(°C)
H	CH_3	H	CH_3	CH_3	CH	O	
H	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	OCH_3	CH	O	
H	CH_3	H	CH_3	CH_3	N	O	
H	CH_3	H	OCH_3	OCH_3	N	O	
H	CH_3	H	OCH_3	CH_3	N	O	
H	C_2H_5	H	OCH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	OCH_3	CH	O	
H	C_2H_5	H	CH_3	CH_3	CH	O	
H	C_2H_5	H	OCH_3	OCH_3	N	O	
H	C_2H_5	H	CH_3	OCH_3	N	O	
H	H	H	OCH_3	OCH_3	CH	O	
H	H	H	OCH_3	CH_3	CH	O	
H	H	H	CH_3	CH_3	CH	O	
H	H	H	OCH_3	OCH_3	N	O	
H	H	H	OCH_3	CH_3	N	O	
H	CH_3	CH_3	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	CH_3	OCH_3	N	O	

TABLE 3c



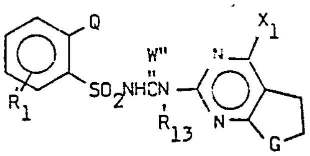
R_1	R_6	R_{12}	R_{13}	X	Y	Z	W''	m.p.(°C)
H	H	H	H	CH_3	CH_3	CH	O	198-202°
H	H	H	H	CH_3	OCH_3	CH	O	214-217°
H	H	H	H	OCH_3	OCH_3	CH	O	228-231°
H	H	H	H	CH_3	CH_3	N	O	220-224°
H	H	H	H	OCH_3	OCH_3	N	O	202-210°
H	H	H	H	CH_3	OCH_3	N	O	200-204°
H	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	H	OCH_3	CH_3	CH	O	
H	CH_3	CH_3	H	CH_3	CH_3	CH	O	
H	CH_3	CH_3	H	CH_3	OCH_3	N	O	
H	C_2H_5	CH_3	H	CH_3	OCH_3	CH	O	
H	C_2H_5	CH_3	H	OCH_3	OCH_3	CH	O	
H	C_2H_5	CH_3	H	OCH_3	CH_3	N	O	
H	C_2H_5	CH_3	H	CH_3	CH_3	CH	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	O	
H	H	H	H	OCH_3	CH_2OCH_3	CH	O	
H	H	H	H	CH_3	OC_2H_5	N	O	
H	H	H	H	OCH_3	$CH(OCH_3)_2$	CH	O	
H	H	H	H	CH_3	C_2H_5	CH	O	
H	H	H	H	Cl	NH_2	CH	O	
H	H	H	H	Cl	$NHCH_3$	CH	O	
H	H	H	H	Cl	$N(CH_3)_2$	CH	O	
H	H	H	H	Cl	OCH_3	CH	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	N	O	
H	H	H	CH_3	OCH_3	CH_3	N	O	
H	H	H	CH_3	OCH_3	CH_3	CH	O	
H	CH_3	CH_3	H	CH_3	CH_3	N	O	

TABLE 3d



R_1	R_6	R_{12}	R_{13}	X	Y	Z	W''	m.p.(°C)
H	H	H	H	CH_3	CH_3	CH	O	
H	H	H	H	OCH_3	OCH_3	CH	O	
H	H	H	H	CH_3	OCH_3	CH	O	
H	H	H	H	CH_3	CH_3	N	O	
H	H	H	H	OCH_3	OCH_3	N	O	
H	H	H	H	CH_3	OCH_3	N	O	
H	H	H	CH_3	OCH_3	OCH_3	CH	O	
H	H	CH_3	H	CH_3	CH_3	CH	O	
H	H	CH_3	H	OCH_3	CH_3	CH	O	
H	H	CH_3	H	OCH_3	OCH_3	CH	O	
H	H	CH_3	H	CH_3	CH_3	N	O	
H	H	CH_3	H	OCH_3	OCH_3	N	O	
H	H	CH_3	H	OCH_3	CH_3	N	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	CH	O	
H	CH_3	CH_3	H	OCH_3	CH_3	CH	O	
H	CH_3	CH_3	H	CH_3	CH_3	CH	O	
H	CH_3	CH_3	H	OCH_3	OCH_3	N	O	
H	CH_3	CH_3	H	OCH_3	CH_3	N	O	
H	C_2H_5	CH_3	H	OCH_3	OCH_3	CH	O	
H	C_2H_5	CH_3	H	OCH_3	CH_3	N	O	
H	C_2H_5	CH_3	H	OCH_3	CH_3	CH	O	
5-F	H	H	H	OCH_3	OCH_3	CH	O	
6-Cl	H	H	H	OCH_3	OCH_3	CH	O	
4-Br	H	H	H	OCH_3	CH_3	CH	O	
3-CH_3	H	H	H	OCH_3	CH_3	CH	O	
H	H	H	CH_3	OCH_3	CH_3	CH	O	
H	H	H	CH_3	OCH_3	CH_3	N	O	
H	CH_3	CH_3	H	CH_3	CH_3	N	O	

TABLE 4



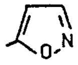
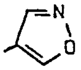
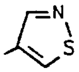
Q	R_1	R_{13}	W''	X_1	G	m.p.(°C)
	H	H	O	CH_3	O	
	H	H	O	CH_3	O	
	H	H	O	Cl	O	

TABLE 4 (continued)

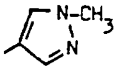
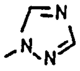
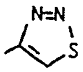
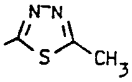
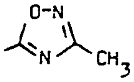
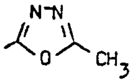
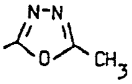
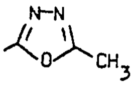
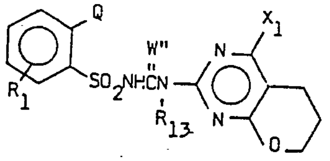
Q	R_1	R_{13}	W"	X_1	G	m.p.(°C)
	H	H	O	CH_3	O	
	H	H	O	OCH_3	O	
	H	H	O	CH_3	O	
	H	H	O	CH_3	O	
	H	H	O	CH_3	O	
	H	H	O	OCH_3	CH_2	
	H	H	O	CH_3	O	
	H	H	O	CH_3	CH_2	
	H	H	O	Cl	CH_2	

TABLE 5



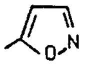
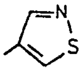
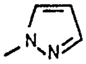
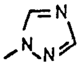
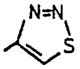
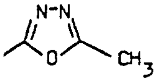
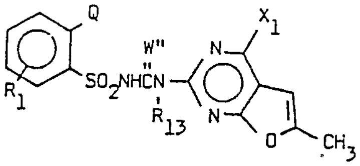
Q	R1	R13	W"	X1	m.p.(°C)
	H	H	O	CH_3	
	H	H	O	Cl	
	H	H	O	OCH_3	
	H	H	O	CH_3	
	H	H	O	CH_3	
	H	H	O	CH_3	

TABLE 6



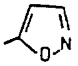
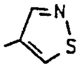
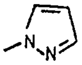
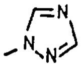
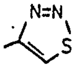
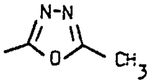
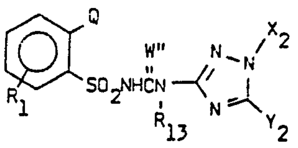
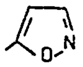
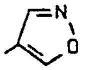
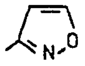
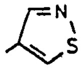
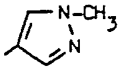
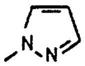
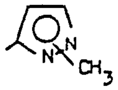
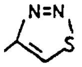
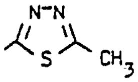
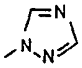
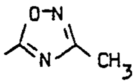
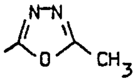
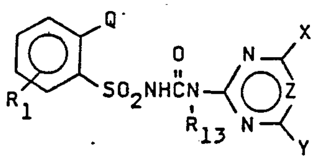
Q	R1	R13	W"	X1	m.p.(°C)
	H	H	O	CH_3	
	H	H	O	Cl	
	H	H	O	OCH_3	
	H	H	O	CH_3	
	H	H	O	CH_3	
	H	H	O	CH_3	

TABLE 7



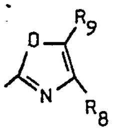
Q	R_1	R_{13}	W"	X_2	Y_2	m.p.(°C)
	H	H	O	CH_3	OCH_3	
	H	H	O	C_2H_5	OCH_3	
	H	H	O	$CH_2CH_2CH_3$	OCH_3	
	H	H	O	CH_2CF_3	OCH_3	
	H	H	O	CH_3	OC_2H_5	
	H	H	O	CH_3	OCH_3	
	H	H	O	CH_3	SCH_3	
	H	H	O	CH_3	OCH_3	
	H	H	O	CH_3	SC_2H_5	
	H	H	O	CH_3	OCH_3	
	H	H	O	CH_2CF_3	SCH_3	
	H	H	O	CH_3	OCH_3	

In Tables 8-15, the headings refer to the following structure.



(I)

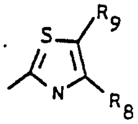
TABLE 8

[In Formula I] Q is 

R_1	R_{13}	R_8	R_9	X	Y	Z	m.p.(°C)
5-F	H	H	H	CH_3	CH_3	N	
6-Cl	H	H	H	CH_3	OCH_3	N	
4-Br	H	H	H	OCH_3	OCH_3	N	
3-CH_3	H	H	H	CH_3	CH_3	CH	
5-OCH_3	H	H	H	CH_3	OCH_3	CH	
5-CF_3	H	H	H	OCH_3	OCH_3	CH	
H	CH_3	H	H	CH_3	CH_3	N	
H	CH_3	H	H	CH_3	OCH_3	N	
H	CH_3	H	H	OCH_3	OCH_3	N	
H	CH_3	H	H	CH_3	CH_3	CH	
H	CH_3	H	H	OCH_3	CH_3	CH	
H	CH_3	H	H	OCH_3	OCH_3	CH	
H	H	H	H	Cl	OCH_3	CH	
H	H	H	H	Cl	NH_2	CH	
H	H	H	H	Cl	$NHCH_3$	CH	
H	H	H	H	Cl	$N(CH_3)_2$	CH	
H	H	H	H	CH_3	CH_2CH_3	CH	
H	H	H	H	OCH_3	CH_2OCH_3	N	
H	H	H	H	CH_3	OCH_2CH_3	N	
H	H	H	H	OCH_3	OCH_2CH_3	CH	
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH	
H	H	H	H	CH_3	CH_3	N	207-209°
H	H	H	H	OCH_3	CH_3	N	199-201°
H	H	H	H	OCH_3	OCH_3	N	191-192°
H	H	H	H	CH_3	CH_3	CH	213-214°
H	H	H	H	OCH_3	CH_3	CH	208-209°
H	H	H	H	OCH_3	OCH_3	CH	197-200°
H	H	CH_3	H	CH_3	CH_3	N	
H	H	CH_3CH_2	H	OCH_3	CH_3	N	
H	H	H	CH_3	OCH_3	OCH_3	N	
H	H	H	CH_3CH_2	CH_3	CH_3	CH	
H	H	H	OCH_3	OCH_3	CH_3	CH	
H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH	
H	H	H	SCH_3	CH_3	OCH_3	N	
H	H	H	Cl	OCH_3	OCH_3	N	
H	H	H	Br	OCH_3	CH_3	CH	
H	H	H	H	OCH_3	SCH_3	N	
H	H	H	H	CH_3	CF_3	CH	
H	H	H	H	CH_3	OCH_2CF_3	CH	
H	H	H	H	CH_3	OCF_2CF_3	N	
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N	
H	H	H	H	CH_3	CH_2OCH_3	CH	

TABLE 8a

[In Formula I] Q is

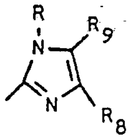


,

R_1	*R_{13}*	*R_8*	*R_9*	*X*	*Y*	*Z*
5-F	H	H	H	CH_3	CH_3	N
6-Cl	H	H	H	CH_3	OCH_3	N
4-Br	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	CH_3	CH_3	CH
5-OCH_3	H	H	H	CH_3	OCH_3	CH
5-CF_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	H	H	CH_3	CH_3	N
H	CH_3	H	H	CH_3	OCH_3	N
H	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	CH_3	CH_3	CH
H	CH_3	H	H	OCH_3	CH_3	CH
H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	Cl	OCH_3	CH
H	H	H	H	Cl	NH_2	CH
H	H	H	H	Cl	$NHCH_3$	CH
H	H	H	H	CH_3	CH_2CH_3	CH
H	H	H	H	OCH_3	CH_2OCH_3	N
H	H	H	H	CH_3	OCH_2CH_3	N
H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
H	H	H	H	CH_3	CH_3	N
H	H	H	H	OCH_3	CH_3	N
H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	CH_3	CH_3	CH
H	H	H	H	OCH_3	CH_3	CH
H	H	H	H	OCH_3	OCH_3	CH
H	H	CH_3	H	CH_3	CH_3	N
H	H	CH_3CH_2	H	OCH_3	CH_3	N
H	H	H	CH_3	OCH_3	OCH_3	N
H	H	H	CH_3CH_2	CH_3	CH_3	CH
H	H	H	OCH_3	OCH_3	CH_3	CH
H	H	H	OCH_2	CH_3	OCH_3	CH
H	H	H	SCH_3	CH_3	OCH_3	N
H	H	H	Cl	OCH_3	OCH_3	N
H	H	H	Br	OCH_3	CH_3	CH
H	H	H	H	OCH_3	SCH_3	N
H	H	H	H	CH_3	CF_3	CH
H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 8b

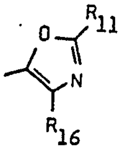
[In Formula I] Q is



R	*R₁*	*R₁₃*	*R₈*	*R₉*	*X*	*Y*	*Z*	*m.p.(°C)*
CH_3	H	H	H	H	CH_3	CH_3	N	
CH_3CH_2	H	H	H	H	OCH_3	CH_3	N	
$CH_3CH_2CH_2$	H	H	H	H	OCH_3	OCH_3	N	
$(CH_3)_2CH$	H	H	H	H	CH_3	CH_3	CH	
$CH_3CH_2CH_2CH_2$	H	H	H	H	OCH_3	CH_3	CH	
$(CH_3)_2CHCH_2-$	H	H	H	H	OCH_3	OCH_3	CH	
$CH_3CHCH_2CH_3$	H	H	H	H	CH_3	CH_3	N	
$(CH_3)_3C$	H	H	H	H	OCH_3	CH_3	N	
H	H	H	H	H	OCH_3	OCH_3	N	
CH_3	5-F	H	H	H	CH_3	CH_3	N	
CH_3	6-Cl	H	H	H	CH_3	OCH_3	N	
CH_3	4-Br	H	H	H	OCH_3	OCH_3	N	
CH_3	3-CH_3	H	H	H	CH_3	CH_3	CH	
CH_3	5-OCH_3	H	H	H	CH_3	OCH_3	CH	
CH_3	5-CF_3	H	H	H	OCH_3	OCH_3	CH	
CH_3	H	CH_3	H	H	CH_3	CH_3	N	
CH_3	H	CH_3	H	H	CH_3	OCH_3	N	
CH_3	H	CH_3	H	H	OCH_3	OCH_3	N	
CH_3	H	CH_3	H	H	CH_3	CH_3	CH	
CH_3	H	CH_3	H	H	OCH_3	CH_3	CH	
CH_3	H	CH_3	H	H	OCH_3	OCH_3	CH	
CH_3	H	H	H	H	Cl	OCH_3	CH	
CH_3	H	H	H	H	Cl	NH_2	CH	
CH_3	H	H	H	H	Cl	$NHCH_3$	CH	
CH_3	H	H	H	H	Cl	$N(CH_3)_2$	CH	
CH_3	H	H	H	H	CH_3	CH_2CH_3	CH	
CH_3	H	H	H	H	OCH_3	CH_2OCH_3	N	
CH_3	H	H	H	H	CH_3	OCH_2CH_3	N	
CH_3	H	H	H	H	OCH_3	OCH_2CH_3	CH	
CH_3	H	H	H	H	CH_3	$CH(OCH_3)_2$	CH	
CH_3	H	H	H	H	CH_3	CH_3	N	
CH_3	H	H	H	H	OCH_3	CH_3	N	
CH_3	H	H	H	H	OCH_3	OCH_3	N	
CH_3	H	H	H	H	CH_3	CH_3	CH	175-177°
CH_3	H	H	H	H	OCH_3	CH_3	CH	200-205°
CH_3	H	H	H	H	OCH_3	OCH_3	CH	182-183°
CH_3	H	H	CH_3	H	CH_3	CH_3	N	
CH_3	H	H	CH_3CH_2	H	OCH_3	CH_3	N	
CH_3	H	H	H	CH_3	OCH_3	OCH_3	N	
CH_3	H	H	H	CH_3CH_2	CH_3	CH_3	CH	
CH_3	H	H	H	OCH_3	OCH_3	CH_3	CH	
CH_3	H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH	
CH_3	H	H	H	SCH_3	CH_3	OCH_3	N	
CH_3	H	H	H	Cl	OCH_3	OCH_3	N	
CH_3	H	H	H	Br	OCH_3	CH_3	CH	
H	H	H	H	H	OCH_3	SCH_3	N	
H	H	H	H	H	CH_3	CF_3	CH	
H	H	H	H	H	CH_3	OCH_2CF_3	CH	
H	H	H	H	H	CH_3	OCF_2CF_3	N	
H	H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N	
H	H	H	H	H	CH_3	CH_2OCH_3	CH	

TABLE 9

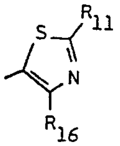
[In Formula I] Q is



R_1	R_{13}	R_{16}	R_{11}	X	Y	Z
5-F	H	H	H	CH_3	CH_3	N
6-Cl	H	H	H	CH_3	OCH_3	N
4-Br	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	CH_3	CH_3	CH
5-OCH_3	H	H	H	CH_3	OCH_3	CH
5-CF_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	H	H	CH_3	CH_3	N
H	CH_3	H	H	CH_3	OCH_3	N
H	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	CH_3	CH_3	CH
H	CH_3	H	H	OCH_3	CH_3	CH
H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	Cl	OCH_3	CH
H	H	H	H	Cl	NH_2	CH
H	H	H	H	Cl	$NHCH_3$	CH
H	H	H	H	Cl	$N(CH_3)_2$	CH
H	H	H	H	CH_3	CH_2CH_3	CH
H	H	H	H	OCH_3	CH_2OCH_3	N
H	H	H	H	CH_3	OCH_2CH_3	N
H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
H	H	H	H	CH_3	CH_3	N
H	H	H	H	OCH_3	CH_3	N
H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	CH_3	CH_3	CH
H	H	H	H	OCH_3	CH_3	CH
H	H	H	H	OCH_3	OCH_3	CH
H	H	CH_3	H	CH_3	CH_3	N
H	H	CH_3CH_2	H	OCH_3	CH_3	N
H	H	H	CH_3	OCH_3	OCH_3	N
H	H	H	CH_3CH_2	CH_3	CH_3	CH
H	H	H	OCH_3	OCH_3	CH_3	CH
H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH
H	H	H	SCH_3	CH_3	OCH_3	N
H	H	H	Cl	OCH_3	OCH_3	N
H	H	H	Br	OCH_3	CH_3	CH
H	H	H	H	OCH_3	SCH_3	N
H	H	H	H	CH_3	CF_3	CH
H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 9a

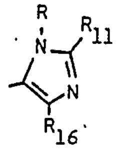
[In Formula I] Q is



R_1	R_{13}	R_{16}	R_{11}	X	Y	Z
5-F	H	H	H	CH_3	CH_3	N
6-Cl	H	H	H	CH_3	OCH_3	N
4-Br	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	CH_3	CH_3	CH
5-OCH_3	H	H	H	CH_3	OCH_3	CH
5-CF_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	H	H	CH_3	CH_3	N
H	CH_3	H	H	CH_3	OCH_3	N
H	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	CH_3	CH_3	CH
H	CH_3	H	H	OCH_3	CH_3	CH
H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	Cl	OCH_3	CH
H	H	H	H	Cl	NH_2	CH
H	H	H	H	CL	$NHCH_3$	CH
H	H	H	H	Cl	$N(CH_3)_2$	CH
H	H	H	H	CH_3	CH_2CH_3	CH
H	H	H	H	OCH_3	CH_2OCH_3	N
H	H	H	H	CH_3	OCH_2CH_3	N
H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
H	H	H	H	CH_3	CH_3	N
H	H	H	H	OCH_3	CH_3	N
H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	CH_3	CH_3	CH
H	H	H	H	OCH_3	CH_3	CH
H	H	H	H	OCH_3	OCH_3	CH
H	H	CH_3	H	CH_3	CH_3	N
H	H	CH_3CH_2	H	OCH_3	CH_3	N
H	H	H	CH_3	OCH_3	OCH_3	N
H	H	H	CH_3CH_2	CH_3	CH_3	CH
H	H	H	OCH_3	OCH_3	CH_3	CH
H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH
H	H	H	SCH_3	CH_3	OCH_3	N
H	H	H	Cl	OCH_3	OCH_3	N
H	H	H	Br	OCH_3	CH_3	CH
H	H	H	H	OCH_3	SCH_3	N
H	H	H	H	CH_3	CF_3	CH
H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 9b

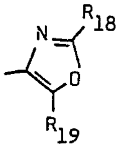
[In Formula I] Q is



R	R_1	R_{13}	R_{16}	R_{11}	X	Y	Z
CH_3	H	H	H	H	CH_3	CH_3	N
CH_3CH_2	H	H	H	H	OCH_3	CH_3	N
$CH_3CH_2CH_2$	H	H	H	H	OCH_3	OCH_3	N
$(CH_3)_2CH$	H	H	H	H	CH_3	CH_3	CH
$CH_3CH_2CH_2CH_2$	H	H	H	H	OCH_3	CH_3	CH
$(CH_3)_2CHCH_2-$	H	H	H	H	OCH_3	OCH_3	CH
$CH_3CHCH_2CH_3$	H	H	H	H	CH_3	CH_3	N
$(CH_3)_3C$	H	H	H	H	OCH_3	CH_3	N
H	H	H	H	H	OCH_3	OCH_3	N
CH_3	5-F	H	H	H	CH_3	CH_3	N
CH_3	6-Cl	H	H	H	CH_3	OCH_3	N
CH_3	4-Br	H	H	H	OCH_3	OCH_3	N
CH_3	3-CH_3	H	H	H	CH_3	CH_3	CH
CH_3	5-OCH_3	H	H	H	CH_3	OCH_3	CH
CH_3	5-CF_3	H	H	H	OCH_3	OCH_3	CH
CH_3	H	CH_3	H	H	CH_3	CH_3	N
CH_3	H	CH_3	H	H	CH_3	OCH_3	N
CH_3	H	CH_3	H	H	OCH_3	OCH_3	N
CH_3	H	CH_3	H	H	CH_3	CH_3	CH
CH_3	H	CH_3	H	H	OCH_3	CH_3	CH
CH_3	H	CH_3	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	Cl	OCH_3	CH
CH_3	H	H	H	H	Cl	NH_2	CH
CH_3	H	H	H	H	Cl	$NHCH_3$	CH
CH_3	H	H	H	H	Cl	$N(CH_3)_2$	CH
CH_3	H	H	H	H	CH_3	CH_2CH_3	CH
CH_3	H	H	H	H	OCH_3	CH_2OCH_3	N
CH_3	H	H	H	H	CH_3	OCH_2CH_3	N
CH_3	H	H	H	H	OCH_3	OCH_2CH_3	CH
CH_3	H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
CH_3	H	H	H	H	CH_3	CH_3	N
CH_3	H	H	H	H	OCH_3	CH_3	N
CH_3	H	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	H	CH_3	CH_3	CH
CH_3	H	H	H	H	OCH_3	CH_3	CH
CH_3	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	CH_3	H	CH_3	CH_3	N
CH_3	H	H	CH_3CH_2	H	OCH_3	CH_3	N
CH_3	H	H	H	CH_3	OCH_3	OCH_3	N
CH_3	H	H	H	CH_3CH_2	CH_3	CH_3	CH
CH_3	H	H	H	OCH_3	OCH_3	CH_3	CH
CH_3	H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH
CH_3	H	H	H	SCH_3	CH_3	OCH_3	N
CH_3	H	H	H	Cl	OCH_3	OCH_3	N
CH_3	H	H	H	Br	OCH_3	CH_3	CH
H	H	H	H	H	OCH_3	SCH_3	N
H	H	H	H	H	CH_3	CF_3	CH
H	H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 10

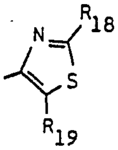
[In Formula I] Q is



R_1	R_{13}	R_{19}	R_{18}	X	Y	Z
5-F	H	H	H	CH_3	CH_3	N
6-Cl	H	H	H	CH_3	OCH_3	N
4-Br	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	CH_3	CH_3	CH
5-OCH_3	H	H	H	CH_3	OCH_3	CH
5-CF_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	H	H	CH_3	CH_3	N
H	CH_3	H	H	CH_3	OCH_3	N
H	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	CH_3	CH_3	CH
H	CH_3	H	H	OCH_3	CH_3	CH
H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	Cl	OCH_3	CH
H	H	H	H	Cl	NH_2	CH
H	H	H	H	Cl	$NHCH_3$	CH
H	H	H	H	Cl	$N(CH_3)_2$	CH
H	H	H	H	CH_3	CH_2CH_3	CH
H	H	H	H	OCH_3	CH_2OCH_3	N
H	H	H	H	CH_3	OCH_2CH_3	N
H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
H	H	H	H	CH_3	CH_3	N
H	H	H	H	OCH_3	CH_3	N
H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	CH_3	CH_3	CH
H	H	H	H	OCH_3	CH_3	CH
H	H	H	H	OCH_3	OCH_3	CH
H	H	CH_3	H	CH_3	CH_3	N
H	H	CH_3CH_2	H	OCH_3	CH_3	N
H	H	H	CH_3	OCH_3	OCH_3	N
H	H	H	CH_3CH_2	CH_3	CH_3	CH
H	H	H	OCH_3	OCH_3	CH_3	CH
H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH
H	H	H	SCH_3	CH_3	OCH_3	N
H	H	H	Cl	OCH_3	OCH_3	N
H	H	H	Br	OCH_3	CH_3	CH
H	H	H	H	OCH_3	SCH_3	N
H	H	H	H	CH_3	CF_3	CH
H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 10a

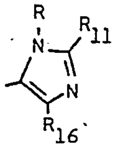
[In Formula I] Q is



R_1	R_{13}	R_{19}	R_{18}	X	Y	Z
5-F	H	H	H	CH_3	CH_3	N
6-Cl	H	H	H	CH_3	OCH_3	N
4-Br	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	CH_3	CH_3	CH
5-OCH_3	H	H	H	CH_3	OCH_3	CH
5-CF_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	H	H	CH_3	CH_3	N
H	CH_3	H	H	CH_3	OCH_3	N
H	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	CH_3	CH_3	CH
H	CH_3	H	H	OCH_3	CH_3	CH
H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	Cl	OCH_3	CH
H	H	H	H	Cl	NH_2	CH
H	H	H	H	Cl	$NHCH_3$	CH
H	H	H	H	CH_3	CH_2CH_3	CH
H	H	H	H	OCH_3	CH_2OCH_3	N
H	H	H	H	CH_3	OCH_2CH_3	N
H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
H	H	H	H	CH_3	CH_3	N
H	H	H	H	OCH_3	CH_3	N
H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	CH_3	CH_3	CH
H	H	H	H	OCH_3	CH_3	CH
H	H	H	H	OCH_3	OCH_3	CH
H	H	CH_3	H	CH_3	CH_3	N
H	H	CH_3CH_2	H	OCH_3	CH_3	N
H	H	H	CH_3	OCH_3	OCH_3	N
H	H	H	CH_3CH_2	CH_3	CH_3	CH
H	H	H	OCH_3	OCH_3	CH_3	CH
H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH
H	H	H	SCH_3	CH_3	OCH_3	N
H	H	H	Cl	OCH_3	OCH_3	N
H	H	H	Br	OCH_3	CH_3	CH
H	H	H	H	OCH_3	SCH_3	N
H	H	H	H	CH_3	CF_3	CH
H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 10b

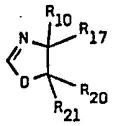
[In Formula I] Q is



R	R1	R13	R16	R11	X	Y	Z
CH_3	H	H	H	H	CH_3	CH_3	N
CH_3CH_2	H	H	H	H	OCH_3	CH_3	N
$CH_3CH_2CH_2$	H	H	H	H	OCH_3	OCH_3	N
$(CH_3)_2CH$	H	H	H	H	CH_3	CH_3	CH
$CH_3CH_2CH_2CH_2$	H	H	H	H	OCH_3	CH_3	CH
$(CH_3)_2CHCH_2$–	H	H	H	H	OCH_3	OCH_3	CH
$CH_3CHCH_2CH_3$	H	H	H	H	CH_3	CH_3	N
$(CH_3)_3C$	H	H	H	H	OCH_3	CH_3	N
H	H	H	H	H	OCH_3	OCH_3	N
CH_3	5-F	H	H	H	CH_3	CH_3	N
CH_3	6-Cl	H	H	H	CH_3	OCH_3	N
CH_3	4-Br	H	H	H	OCH_3	OCH_3	N
CH_3	3-CH_3	H	H	H	CH_3	CH_3	CH
CH_3	5-OCH_3	H	H	H	CH_3	OCH_3	CH
CH_3	5-CF_3	H	H	H	OCH_3	OCH_3	CH
CH_3	H	CH_3	H	H	CH_3	CH_3	N
CH_3	H	CH_3	H	H	CH_3	OCH_3	N
CH_3	H	CH_3	H	H	OCH_3	OCH_3	N
CH_3	H	CH_3	H	H	CH_3	CH_3	CH
CH_3	H	CH_3	H	H	OCH_3	CH_3	CH
CH_3	H	CH_3	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	Cl	OCH_3	CH
CH_3	H	H	H	H	Cl	NH_2	CH
CH_3	H	H	H	H	Cl	$NHCH_3$	CH
CH_3	H	H	H	H	Cl	$NH(CH_3)_2$	CH
CH_3	H	H	H	H	CH_3	CH_2CH_3	CH
CH_3	H	H	H	H	OCH_3	CH_2OCH_3	N
CH_3	H	H	H	H	CH_3	OCH_2CH_3	N
CH_3	H	H	H	H	OCH_3	OCH_2CH_3	CH
CH_3	H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
CH_3	H	H	H	H	CH_3	CH_3	N
CH_3	H	H	H	H	OCH_3	CH_3	N
CH_3	H	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	H	CH_3	CH_3	CH
CH_3	H	H	H	H	OCH_3	CH_3	CH
CH_3	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	CH_3	H	CH_3	CH_3	N
CH_3	H	H	CH_3CH_2	H	OCH_3	CH_3	N
CH_3	H	H	H	CH_3	OCH_3	OCH_3	N
CH_3	H	H	H	CH_3CH_2	CH_3	CH_3	CH
CH_3	H	H	H	OCH_3	OCH_3	CH_3	CH
CH_3	H	H	H	OCH_2CH_3	OCH_3	OCH_3	CH
CH_3	H	H	H	SCH_3	CH_3	OCH_3	N
H	H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	H	CH_3	CF_3	CH
H	H	H	H	H	CH_3	OCH_2CF_3	CH
H	H	H	H	H	CH_3	OCF_2CF_3	N
H	H	H	H	H	CH_3	$OCH_2CH_2OCH_3$	N
H	H	H	H	H	CH_3	CH_2OCH_3	CH

TABLE 11

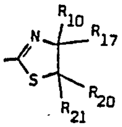
[In Formula I] Q is



R_1	R_{10}	R_{17}	R_{20}	R_{21}	X	Y	Z
5-F	H	H	H	H	OCH_3	OCH_3	N
6-Cl	H	H	H	H	OCH_3	OCH_3	N
4-Br	H	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	H	OCH_3	OCH_3	CH
5-OCH_3	H	H	H	H	OCH_3	OCH_3	CH
5-CF_3	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	H	Cl	OCH_3	CH
H	H	H	H	H	Cl	OCH_3	N
H	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	OCH_3	OCH_2CH_3	N
H	H	H	H	H	OCH_3	OCH_2CH_3	CH
H	CH_3	H	H	H	OCH_3	OCH_3	N
H	CH_3	CH_3	H	H	OCH_3	OCH_3	CH
H	CH_3	CH_3	CH_3	H	OCH_3	OCH_3	N
H	CH_3	CH_3	CH_3	CH_3	OCH_3	OCH_3	CH
H	CH_3CH_2	CH_3	CH_3	H	OCH_3	OCH_3	N
H	CH_3CH_2	CH_3CH_2	H	H	OCH_3	OCH_3	CH
H	H	H	CH_3CH_2	CH_3CH_2	OCH_3	OCH_3	N
H	$CH_3CH_2CH_2$	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3CH_2CH_2$	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3CH_2CH_2$	H	OCH_3	OCH_3	CH
H	H	H	H	$CH_3CH_2CH_2$	OCH_3	OCH_3	N
H	$CH_3(CH_2)_3$–	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3(CH_2)_3$–	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3(CH_2)_3$–	H	OCH_3	OCH_3	CH
H	H	H	H	$CH_3(CH_2)_3$–	OCH_3	OCH_3	N
H	$(CH_3)_3C$–	H	H	H	OCH_3	OCH_3	CH
H	H	$(CH_3)_3C$–	H	H	OCH_3	OCH_3	N
H	H	H	$(CH_3)_3C$–	H	OCH_3	OCH_3	CH
H	H	H	H	$(CH_3)_3C$–	OCH_3	OCH_3	N
H	$CH_3CHCH_2CH_3$	H	H	H	OCH_3	OCH_3	CH
H	H	$(CH_3)_2CHCH_2$–	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3CHCH_2CH_3$	H	OCH_3	OCH_3	CH
H	H	H	H	$(CH_3)_2CHCH_2$–	OCH_3	OCH_3	N
H	CH_3	CH_3	H	H	OCH_3	OCH_3	CH
H	CH_3	CH_3	H	H	OCH_3	OCH_3	N

TABLE 11a

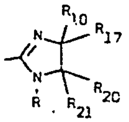
[In Formula I] Q is



R_1	R_{10}	R_{17}	R_{20}	R_{21}	X	Y	Z
5-F	H	H	H	H	OCH_3	OCH_3	N
6-Cl	H	H	H	H	OCH_3	OCH_3	N
4-Br	H	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	H	OCH_3	OCH_3	CH
5-OCH_3	H	H	H	H	OCH_3	OCH_3	CH
5-CF_3	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	H	Cl	OCH_3	CH
H	H	H	H	H	Cl	OCH_3	N
H	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	OCH_3	OCH_2CH_3	N
H	H	H	H	H	OCH_3	OCH_2CH_3	CH
H	CH_3	H	H	H	OCH_3	OCH_3	N
H	CH_3	CH_3	H	H	CH_3	CH_3	CH
H	CH_3	CH_3	CH_3	H	OCH_3	OCH_3	N
H	CH_3	CH_3	CH_3	CH_3	OCH_3	OCH_3	CH
H	CH_3CH_2	CH_3	CH_3	H	OCH_3	OCH_3	N
H	CH_3CH_2	CH_3CH_2	H	H	OCH_3	OCH_3	CH
H	H	H	CH_3CH_2	CH_3CH_2	OCH_3	OCH_3	N
H	$CH_3CH_2CH_2$	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3CH_2CH_2$	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3CH_2CH_2$	H	OCH_3	OCH_3	CH
H	H	H	H	$CH_3CH_2CH_2$	OCH_3	OCH_3	N
H	$CH_3(CH_2)_3-$	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3(CH_2)_3-$	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3(CH_2)_3-$	H	OCH_3	OCH_3	CH
H	H	H	H	$CH_3(CH_2)_3-$	OCH_3	OCH_3	N
H	$(CH_3)_3C-$	H	H	H	OCH_3	OCH_3	CH
H	H	$(CH_3)_3C-$	H	H	OCH_3	OCH_3	N
H	H	H	$(CH_3)_3C-$	H	OCH_3	OCH_3	CH
H	H	H	H	$(CH_3)_3C-$	OCH_3	OCH_3	N
H	$CH_3CHCH_2CH_3$	H	H	H	OCH_3	OCH_3	CH
H	H	$(CH_3)_2CHCH_2-$	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3CHCH_2CH_3$	H	OCH_3	OCH_3	CH
H	H	H	H	$(CH_3)_2CHCH_2-$	OCH_3	OCH_3	N
H	CH_3	CH_3	H	H	OCH_3	OCH_3	CH
H	CH_3	CH_3	H	H	OCH_3	OCH_3	N

TABLE 11b

[In Formula I] Q is



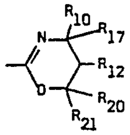
R	*R₁*	*R₁₀*	*R₁₇*	*R₂₀*	*R₂₁*	*X*	*Y*	*Z*
CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	H	OCH_3	OCH_3	N
CH_3CH_2	H	H	H	H	H	OCH_3	OCH_3	CH
$CH_3CH_2CH_2$	H	H	H	H	H	OCH_3	OCH_3	N
$CH_3(CH_2)_3$	H	H	H	H	H	OCH_3	OCH_3	CH
$(CH_3)_2CH$	H	H	H	H	H	OCH_3	OCH_3	N
$(CH_3)_2CHCH_2$–	H	H	H	H	H	OCH_3	OCH_3	CH
$CH_3CHCH_2CH_3$	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	5-F	H	H	H	H	OCH_3	OCH_3	N
CH_3	6-Cl	H	H	H	H	OCH_3	OCH_3	N
CH_3	4-Br	H	H	H	H	OCH_3	OCH_3	N
CH_3	3-CH_3	H	H	H	H	OCH_3	OCH_3	CH
CH_3	5-OCH_3	H	H	H	H	OCH_3	OCH_3	CH
CH_3	5-CF_3	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	H	H	Cl	OCH_3	CH
CH_3	H	H	H	H	H	Cl	OCH_3	N
CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	H	OCH_3	OCH_2CH_3	N
CH_3	H	H	H	H	H	OCH_3	OCH_2CH_3	CH
CH_3	H	CH_3	H	H	H	OCH_3	OCH_3	N
CH_3	H	CH_3	CH_3	H	H	OCH_3	OCH_3	CH
CH_3	H	CH_3	CH_3	CH_3	H	OCH_3	OCH_3	N
CH_3	H	CH_3	CH_3	CH_3	CH_3	OCH_3	OCH_3	CH
CH_3	H	CH_3CH_2	CH_3	CH_3	H	OCH_3	OCH_3	N
CH_3	H	CH_3CH_2	CH_3CH_2	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	CH_3CH_2	CH_3CH_2	OCH_3	OCH_3	N
CH_3	H	$CH_3CH_2CH_2$	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$CH_3CH_2CH_2$	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$CH_3CH_2CH_2$	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$CH_3CH_2CH_2$	OCH_3	OCH_3	N
CH_3	H	$CH_3(CH_2)_3$–	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$CH_3(CH_2)_3$–	H	H	OCH_3	OCH_3	N

TABLE 11b (continued)

R	*R₁*	*R₁₀*	*R₁₇*	*R₂₀*	*R₂₁*	*X*	*Y*	*Z*
CH_3	H	H	H	$CH_3(CH_2)_3-$	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$CH_3(CH_2)_3-$	OCH_3	OCH_3	N
CH_3	H	$(CH_3)_3C-$	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$(CH_3)_3C-$	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$(CH_3)_3C-$	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$(CH_3)_3C-$	OCH_3	OCH_3	N
CH_3	H	$CH_3CHCH_2CH_3$	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$(CH_3)_2CHCH_2-$	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$CH_3CHCH_2CH_3$	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$(CH_3)_2CHCH_2-$	OCH_3	OCH_3	N
CH_3	H	CH_3	CH_3	H	H	OCH_3	OCH_3	CH
CH_3	H	CH_3	CH_3	H	H	OCH_3	OCH_3	N

TABLE 12

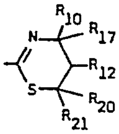
[In Formula I] Q is



R_1	R_{10}	R_{17}	R_{20}	R_{21}	R_{12}	X	Y	Z
5-F	H	H	H	H	H	OCH_3	OCH_3	N
6-Cl	H	H	H	H	H	OCH_3	OCH_3	N
4-Br	H	H	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
5-OCH_3	H	H	H	H	H	OCH_3	OCH_3	CH
5-CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	H	Cl	OCH_3	CH
H	H	H	H	H	H	Cl	OCH_3	N
H	H	H	H	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	CH_3	H	H	H	OCH_3	OCH_2CH_3	N
H	H	H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	CH_3	H	OCH_3	OCH_3	N
H	H	H	H	H	CH_3	OCH_3	OCH_3	CH
H	CH_3CH_2	CH_3	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3CH_2	CH_3CH_2	H	H	H	OCH_3	OCH_3	CH
H	H	H	CH_3CH_2	CH_3CH_2	H	OCH_3	OCH_3	N
H	$CH_3CH_2CH_2$	H	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3CH_2CH_2$	H	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3CH_2CH_2$	H	H	OCH_3	OCH_3	CH
H	H	H	H	$CH_3CH_2CH_2$	H	OCH_3	OCH_3	N
H	$CH_3(CH_2)_3$	H	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3CHCH_2CH_3$	H	H	H	OCH_3	OCH_3	N
H	H	H	$(CH_3)_2CHCH_2$	H	H	OCH_3	OCH_3	CH
H	H	H	H	$(CH_3)_3C-$	H	OCH_3	OCH_3	N
H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	N

TABLE 12a

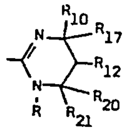
[In Formula I] Q is



R_1	R_{10}	R_{17}	R_{20}	R_{21}	R_{12}	X	Y	Z
5-F	H	H	H	H	H	OCH_3	OCH_3	N
6-Cl	H	H	H	H	H	OCH_3	OCH_3	N
4-Br	H	H	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
5-OCH_3	H	H	H	H	H	OCH_3	OCH_3	CH
5-CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	H	Cl	OCH_3	CH
H	H	H	H	H	H	Cl	OCH_3	N
H	H	H	H	H	H	OCH_3	OCH_3	N
H	CH_3	H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	CH_3	H	H	H	OCH_3	OCH_2CH_3	N
H	H	H	CH_3	H	H	OCH_3	OCH_3	CH
H	H	H	H	CH_3	H	OCH_3	OCH_3	N
H	H	H	H	H	CH_3	OCH_3	OCH_3	CH
H	CH_3CH_2	CH_3	CH_3	H	H	OCH_3	OCH_3	N
H	CH_3CH_2	CH_3CH_2	H	H	H	OCH_3	OCH_3	CH
H	H	H	CH_3CH_2	CH_3CH_2	H	OCH_3	OCH_3	N
H	$CH_3CH_2CH_2$	H	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3CH_2CH_2$	H	H	H	OCH_3	OCH_3	N
H	H	H	$CH_3CH_2CH_2$	H	H	OCH_3	OCH_3	CH
H	H	H	H	$CH_3CH_2CH_2$	H	OCH_3	OCH_3	N
H	$CH_3(CH_2)_3$	H	H	H	H	OCH_3	OCH_3	CH
H	H	$CH_3CHCH_2CH_3$	H	H	H	OCH_3	OCH_3	N
H	H	H	$(CH_3)_2CHCH_2$	H	H	OCH_3	OCH_3	CH
H	H	H	H	$(CH_3)_3C-$	H	OCH_3	OCH_3	N
H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	N

TABLE 12b

[In Formula I] Q is



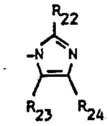
R	*R₁*	*R₁₀*	*R₁₇*	*R₂₀*	*R₂₁*	*X*	*Y*	*Z*	
CH_3	H	H	H	H	H	H	OCH_3	OCH_3	N
CH_3CH_2	H	H	H	H	H	H	OCH_3	OCH_3	CH
$CH_3CH_2CH_2$	H	H	H	H	H	H	OCH_3	OCH_3	N
$(CH_3)_2CH$	H	H	H	H	H	H	OCH_3	OCH_3	CH
$CH_3(CH_2)_3$	H	H	H	H	H	H	OCH_3	OCH_3	N
$(CH_3)_2CHCH_2-$	H	H	H	H	H	H	OCH_3	OCH_3	CH
$CH_3CHCH_2CH_3$	H	H	H	H	H	H	OCH_3	OCH_3	N
$(CH_3)_3C$	H	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	5-F	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	6-Cl	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	4-Br	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	3-CH_3	H	H	H	H	H	OCH_3	OCH_3	CH
CH_3	5-OCH_3	H	H	H	H	H	OCH_3	OCH_3	CH
CH_3	5-CF_3	H	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	H	H	H	Cl	OCH_3	CH
CH_3	H	H	H	H	H	H	Cl	OCH_3	N
CH_3	H	H	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	H	H	OCH_3	OCH_2CH_3	N
CH_3	H	H	H	H	H	H	OCH_3	OCH_2CH_3	CH
CH_3	H	CH_3	H	H	H	H	OCH_3	OCH_3	N
CH_3	H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	CH
CH_3	H	CH_3	CH_3	CH_3	H	H	OCH_3	OCH_3	N
CH_3	H	CH_3	CH_3	CH_3	CH_3	H	OCH_3	OCH_3	CH
CH_3	H	CH_3CH_2	CH_3	CH_3	H	H	OCH_3	OCH_3	N
CH_3	H	CH_3CH_2	CH_3CH_2	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	CH_3CH_2	CH_3CH_2	H	OCH_3	OCH_3	N
CH_3	H	$CH_3CH_2CH_2$	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$CH_3CH_2CH_2$	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$CH_3CH_2CH_2$	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$CH_3CH_2CH_2$	H	OCH_3	OCH_3	N
CH_3	H	$CH_3(CH_2)_3-$	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$CH_3(CH_2)_3-$	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$CH_3(CH_2)_3-$	H	H	OCH_3	OCH_3	CH

TABLE 12b (continued)

R	*R₁*	*R₁₀*	*R₁₇*	*R₂₀*	*R₂₁*	*X*	*Y*	*Z*	
CH_3	H	H	H	H	$CH_3(CH_2)_3-$	H	OCH_3	OCH_3	N
CH_3	H	$(CH_3)_3C-$	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$(CH_3)_3C-$	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$(CH_3)_3C-$	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$(CH_3)_3C-$	H	OCH_3	OCH_3	N
CH_3	H	$CH_3CHCH_2CH_3$	H	H	H	H	OCH_3	OCH_3	CH
CH_3	H	H	$(CH_3)_2CHCH_2-$	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	$CH_3CHCH_2CH_3$	H	H	OCH_3	OCH_3	CH
CH_3	H	H	H	H	$(CH_3)_2CHCH_2-$	H	OCH_3	OCH_3	N
CH_3	H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	CH
CH_3	H	CH_3	CH_3	H	H	H	OCH_3	OCH_3	N
CH_3	H	H	H	H	H	CH_3	OCH_3	OCH_3	CH

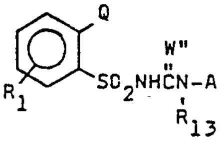
TABLE 13

[In Formula I] Q is



R_1	R_{13}	R_{22}	R_{23}	R_{24}	X	Y	Z
5-F	H	H	H	H	CH_3	CH_3	N
6-Cl	H	H	H	H	CH_3	OCH_3	N
4-Br	H	H	H	H	OCH_3	OCH_3	N
3-CH_3	H	H	H	H	CH_3	CH_3	CH
2-OCH_3	H	H	H	H	CH_3	OCH_3	CH
5-CF_3	H	H	H	H	OCH_3	OCH_3	CH
H	H	H	H	H	Cl	OCH_3	CH
H	H	H	H	H	Cl	NH_2	CH
H	H	H	H	H	Cl	$NHCH_3$	CH
H	H	H	H	H	Cl	$N(CH_3)_2$	CH
H	H	H	H	H	CH_3	CH_2CH_3	CH
H	H	H	H	H	OCH_3	CH_2OCH_3	N
H	H	H	H	H	CH_3	OCH_2CH_3	N
H	H	H	H	H	OCH_3	OCH_2CH_3	CH
H	H	H	H	H	CH_3	$CH(OCH_3)_2$	CH
H	CH_3	H	H	H	CH_3	CH_3	CH
H	CH_3	H	H	H	CH_3	OCH_3	CH
H	CH_3	H	H	H	OCH_3	OCH_3	CH
H	CH_3	H	H	H	CH_3	CH_3	N
H	CH_3	H	H	H	OCH_3	CH_3	N
H	CH_3	H	H	H	OCH_3	OCH_3	N
H	H	CH_3	H	H	CH_3	CH_3	CH
H	H	CH_3CH_2	H	H	CH_3	OCH_3	CH
H	H	CH_3O	H	H	OCH_3	OCH_3	CH
H	H	CH_3CH_2O	H	H	CH_3	OCH_3	N
H	H	CH_3S	H	H	CH_3	OCH_3	N
H	H	Cl	H	H	OCH_3	OCH_3	N
H	H	Br	H	H	OCH_3	OCH_3	N
H	H	H	CH_3	H	CH_3	CH_3	CH
H	H	H	H	CH_3	CH_3	OCH_3	CH
H	H	H	CH_3	CH_3	OCH_3	OCH_3	CH
H	H	H	CH_3CH_2	H	CH_3	OCH_3	N
H	H	H	H	CH_3CH_2	OCH_3	OCH_3	N
H	H	H	H	H	CH_3	CH_3	N
H	H	H	H	H	CH_3	OCH_3	N
H	H	H	H	H	OCH_3	OCH_3	N
H	H	H	H	H	CH_3	CH_3	CH
H	H	H	H	H	CH_3	OCH_3	CH
H	H	H	H	H	OCH_3	OCH_3	CH

TABLE 14



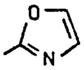
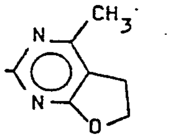
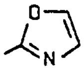
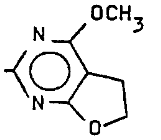
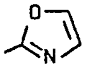
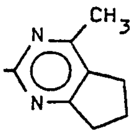
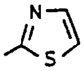
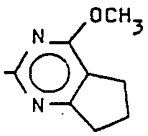
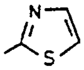
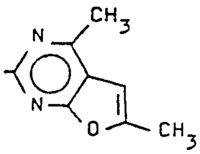
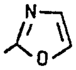
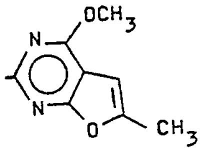
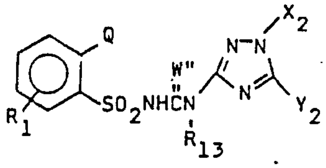
Q	W"	R_1	R_{13}	A
	O	H	H	
	O	H	H	
	O	H	H	
	O	H	H	
	O	H	H	
	O	H	H	

TABLE 15



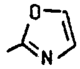
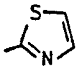
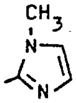
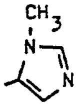
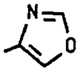
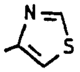
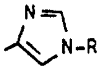
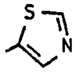
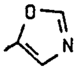
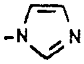
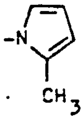
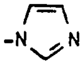
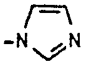
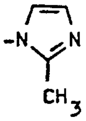
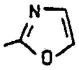
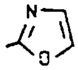
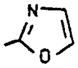
Q	W"	R_1	R_{13}	X_2	Y_2
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_3
	O	H	H	CH_3	OCH_2CH_3
	O	H	H	CH_3	CH_3S

TABLE 15 (continued)

Q	W"	R_1	R_{13}	X_2	Y_2
	O	H	H	CH_3	CH_3CH_2S
	O	H	H	CH_3CH_2	OCH_3
	O	H	H	$CH_3CH_2CH_3$	OCH_3
	O	H	H	$(CH_3)_2CH$	OCH_3
	O	H	H	CF_3CH_2	OCH_3

Formulations

Useful formulations of the compounds of Formula I can be prepared in conventional ways. They include dusts, granules, pellets, solutions, suspensions, emulsions, wettable powders, emulsifiable concentrates and the like. Many of these may be applied directly. Sprayable formulations can be extended in suitable media and used at spray volumes of from a few liters to several hundred liters per hectare. High strength compositions are primarily used as intermediates for further formulation. The formulations, broadly, contain about 0.1% to 99% by weight of active ingredient(s) and at least one of (a) about 0.1% to 20% surfactant(s) and (b) about 1% to 99.9% solid or liquid inert diluent(s). More specifically, they will contain these ingredients in the following approximate proportions:

TABLE 16

	*Weight Percent**		
	Active Ingredient	*Diluent(s)*	*Surfactant(s)*
Wettable Powders	20-90	0-74	1-10
Oil Suspensions, Emulsions, Solutions, (including Emulsifiable Concentrates)	3-50	40-95	0-15
Aqueous Suspension	10-50	40-84	1-20
Dusts	1-25	70-99	0-5
Granules and Pellets	0.1-95	5-99.9	0-15
High Strength Compositions	90-99	0-10	0-2

* Active ingredient plus at least one of a Surfactant or a Diluent equals 100 weight percent.

Lower or higher levels of active ingredient can, of course, be present depending on the intended use and the physical properties of the compound. Higher ratios of surfactant to active ingredient are sometimes desirable, and are achieved by incorporation into the formulation or by tank mixing.

Typical solid diluents are described in Watkins, et al., "Handbook of Insecticide Dust Diluents and Carriers" 2nd Ed., Dorland Books, Caldwell, New Jersey, but other solids, either mined or manufactured, may be used.

The more absorptive diluents are preferred for wettable powders and the denser ones for dusts. Typical liquid diluents and solvents are described in Marsden, "Solvents Guide, " 2nd Ed., Interscience, New York, 1950. Solubility under 0.1% is preferred for suspension concentrates; solution concentrates are preferably stable against phase separation at 0°C. "McCutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood, New Jersey, as well as Sisely and Wood, "Encyclopedia of Surface Active Agents", Chemical Publishing Co., Inc., New York, 1964, list surfactants and recommended uses. All formulations can contain minor amounts of additives to reduce foaming, caking, corrosion, microbiological growth, etc.

The methods of making such compositions are well known. Solutions are prepared by simply mixing the ingredients. Fine solid compositions are made by blending and, usually, grinding as in a hammer or fluid energy mill. Suspensions are prepared by wet milling (see, for example, Littler, U.S. Patent 3,060,084). Granules and pellets may be made by spraying the active material upon preformed granular carriers or by agglomeration techniques. See J. E. Browning, "Agglomeration", *Chemical Engineering*, December 4, 1967, pp. 147ff. and "Perry's Chemical Engineer's Handbook", 5th Ed., McGraw-Hill, New York, 1973, pp. 8-57ff.

For further information regarding the art of formulation, see for example:

H. M. Loux, U.S. Patent 3,235,361, February 15, 1966, Col. 6, line 16 through Col. 7, line 19 and Examples 10 through 41;

R. W. Luckenbaugh, U.S. Patent 3,309,192, March 14, 1967, Col. 5, Line 43 through Col. 7, line 62 and Examples 8, 12, 15, 39, 41, 52, 53, 58, 132, 138-140, 162-164, 166, 167 and 169-182;

H. Gysin and E. Knusli, U.S. Patent 2,891,855, June 23, 1959, Col. 3, line 66 through Col. 5, line 17 and Examples 1-4;

G. C. Klingman, "Weed Control as a Science", John Wiley and Sons, Inc., New York, 1961, pp. 81-96; and

J. D. Fryer and S. A. Evans, "Weed Control Handbook", 5th Ed., Blackwell Scientific Publications, Oxford, 1968, pp. 101-103.

In the following examples, all parts are by weight unless otherwise indicated.

EXAMPLE 41
Wettable Powder

2-(isoxazol-5-yl)-N-[(4,6-dimethoxypryimidin-2-yl)-aminocarbonyl]benzenesulfonamide	80%
sodium alkylnaphthalenesulfonate	2%
sodium ligninsulfonate	2%
synthetic amorphous silica	3%
kaolinite	13%

The ingredients are blended, hammer-milled until all the solids are essentially under 50 microns, re-blended, and packaged.

EXAMPLE 42
Wettable Powder

2-(isoxazol-5-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide	50%
sodium alkylnaphthalenesulfonate	2%
low viscosity methyl cellulose	2%
diatomaceous earth	46%

The ingredients are blended, coarsely hammer-milled and then air-milled to produce particles essentially all below 10 microns in diameter. The product is reblended before packaging.

EXAMPLE 43
Granule

Wettable Powder of Example 42	5%
attapulgite granules (U.S.S. 20-40 mesh; 0.84-0.42 mm)	95%

A slurry of wettable powder containing ≈25% solids is sprayed on the surface of attapulgite granules in a double-cone blender. The granules are dried and packaged:

EXAMPLE 44
Extruded pellet

N-[(4,6-dimethyoxypryimidine-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide	25%
anhydrous sodium sulfate	10%
crude calcium ligninsulfonate	5%
sodium alkylnaphthalenesulfonate	1%
calcium/magnesium bentonite	59%

The ingredients are blended, hammer-milled and then moistened with about 12% water. The mixture is extruded as cylinders about 3 mm diameter which are cut to produce pellets about 3 mm long. These may be used directly after drying, or the dried pellets may be crushed to pass a U.S.S. No. 20 sieve (0.84 mm openings). The granules held on a U.S.S. No. 40 sieve (0.42 mm openings) may be packaged for use and the fines recycled.

EXAMPLE 45
Oil suspension

N-[(4,6-dimethyoxypryimidine-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide	25%
polyoxyethylene sorbitol hexaoleate	5%
highly aliphatic hydrocarbon oil	70%

The ingredients are ground together in a sand mill until the solid particles have been reduced to under about 5 microns. The resulting thick suspension may be applied directly, but preferably after being extended with oils or emulsified in water.

EXAMPLE 46
Wettable powder

N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)amino-carbonyl-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)-benzenesulfonamide	20%
sodium alkylnaphthalenesulfonate	4%
sodium ligninsulfonate	4%
low viscosity methyl cellulose	3%
attapulgite	69%

The ingredients are thoroughly blended. After grinding in a hammer-mill to produce particles essentially all below 100 microns, the material is reblended and sifted through a U.S.S. No. 50 sieve (0.3 mm opening) and packaged.

EXAMPLE 47

Low strength granule

N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzene sulfonamide	1%
N,N-dimethylformamide	9%
attapulgite granules	90%
(U.S.S. 20-40 sieve)	

The active ingredient is dissolved in the solvent and the solution is sprayed upon dedusted granules in a double cone blender. After spraying of the solution has been completed, the blender is allowed to run for a short period and then the granules are packaged.

EXAMPLE 48

Aqueous suspension

N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide	40%
polyacrylic acid thickener	0.3%
dodecylphenol polyethylene glycol ether	0.5%
disodium phosphate	1%
monosodium phosphate	0.5%
polyvinyl alcohol	1.0%
water	56.7%

The ingredients are blended and ground together in a sand mill to produce particles essentially all under 5 microns in size.

EXAMPLE 49

Solution

2-(isoxazol-4-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide, sodium salt	5%
water	95%

The salt is added directly to the water with stirring to produce the solution, which may then be packaged for use.

EXAMPLE 50

Low strength granule

2-(isoxazol-4-yl)-N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide	0.1%
attapulgite granules	99.9%
(U.S.S. 20-40 mesh)	

The active ingredient is dissolved in a solvent and the solution is sprayed upon dedusted granules in a double-cone blender. After spraying of the solution has been completed, the material is warmed to evaporate the solvent. The material is allowed to cool and then packaged.

EXAMPLE 51
Granule

2-(isoxazol-5-yl)-N-[(4,6-dimethoxypyrimidin-2-yl)-aminocarbonyl]benzenesulfonamide	80%
wetting agent	1%
crude ligninsulfonate salt (containing 5-20% of the natural sugars)	10%
attapulgite clay	9%

The ingredients are blended and milled to pass through a 100 mesh screen. This material is then added to a fluid bed granulator, the air flow is adjusted to gently fluidize the material, and a fine spray of water is sprayed onto the fluidized material. The fluidization and spraying are continued until granules of the desired size range are made. The spraying is stopped, but fluidization is continued, optionally with heat, until the water content is reduced to the desired level, generally less than 1%. The material is then discharged, screened to the desired size range, generally 14-100 mesh (1410-149 microns), and packaged for use.

EXAMPLE 52
High strength concentrate

N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide	99%
silica aerogel	0.5%
synthetic amorphous silica	0.5%

The ingredients are blended and ground in a hammer-mill to produce a material essentially all passing a U.S.S. No. 50 screen (0.3 mm opening). The concentrate may be formulated further if necessary.

EXAMPLE 53
Wettable powder

N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide	90%
dioctyl sodium sulfosuccinate	0.1%
synthetic fine silica	9.9%

The ingredients are blended and ground in a hammer-mill to produce particles essentially all below 100 microns. The material is sifted through a U.S.S. No. 50 screen and then packaged.

EXAMPLE 54
Wettable powder

2-(isoxazol-5-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide	40%
sodium ligninsulfonate	20%
montmorillonite clay	40%

The ingredients are thoroughly blended, coarsely hammer-milled and then air-milled to produce particles essentially all below 10 microns in size. The material is reblended and then packaged.

EXAMPLE 55
Oil suspension

N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide	35%
blend of polyalcohol carboxylic esters and oil soluble petroleum sulfonates	6%
xylene	59%

The ingredients are combined and ground together in a sand mill to produce particles essentially all below 5 microns. The product can be used directly, extended with oils, or emulsified in water.

EXAMPLE 56
Dust

N-[(4-methoxy-6-methyl-1,3-5-triazin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide	10%
attapulgite	10%
Pyrophyllite	80%

The active ingredient is blended with attapulgite and then passed through a hammer-mill to produce particles substantially all below 200 microns. The ground concentrate is then blended with powdered pyrophyllite until homogeneous.

Utility
The compounds of the present invention are powerful herbicides. They have utility for broad-spectrum pre- and/or post-emergence weed control in areas where complete control of all vegetation is desired, such as around fuel storage tanks, ammunition depots, industrial storage areas, parking lots, drive-in theaters, around billboards, highway and railroad structures. Alternatively, the subject compounds are useful for the selective pre- or post-emergence weed control in crops, such as wheat, barley, rice, soybeans and corn.

The rates of application for the compounds of the invention are determined by a number of factors, including their use as selective or general herbicides, the crop species involved, the types of weeds to be controlled, weather and climate, formulations selected, mode of application, amount of foliage present, etc. In general terms, the subject compounds should be applied at levels in the range of about 0.01 to 10 kg/ha, the lower rates being suggested for use on lighter soils and/or those having a low organic matter content, for selective weed control or for situations where only short-term persistence is required.

The compounds of the invention may be used in combination with any other commercial herbicide examples of which are those of the triazine, triazole, uracil, urea, amide, diphenylether, carbamate and bipyridylium types.

The herbicidal properties of the subject compounds were discovered in a number of greenhouse tests. The test procedures and results follow.

Test A
Seeds of crabgrass (*Digitaria* sp.), barnyardgrass (*Echinochloa crusgalli*), wild oats (*Avena fatua*), sicklepod (*Cassia obtusifolia*), morningglory (*Ipomoea* sp.), cocklebur (*Xanthium* sp.), sorghum, corn, soybean, rice, wheat and nutsedge tubers (*Cyperus rotundus*) were planted in a growth medium and treated pre-emergence with a non-phytotoxic solvent solution of the compounds of Table A. At the same time, cotton having five leaves (including cotyledonary ones), bush beans with the third trifoliolate leaf expanding, crabgrass, barnyardgrass and wild oats with two leaves, sicklepod with three leaves (including cotyledonary ones), morningglory and cocklebur with four leaves (including the cotyledonary ones), sorghum and corn with four leaves, soybean with two cotyledonary leaves, rice with three leaves, wheat with one leaf, and nutsedge with three-five leaves were sprayed with a non-phytotoxic solvent solution of the compounds of Table A. Other containers of the above-mentioned weeds and crops were treated pre- or post-emergence with the same non-phytotoxic solvent so as to provide a solvent control. A set of untreated control plants was also included for comparison. Pre-emergence and post-emergence treated plants and controls were maintained in a greenhouse for sixteen days, then all treated plants were compared with their respective controls and rated visually for response to treatment.

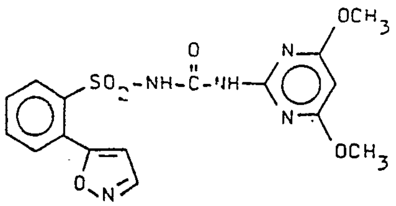
The following rating system was used:

0 = no effect;
10 = maximum effect;
C = chlorosis or necrosis;
D = defoliation;
E = emergence inhibition;
G = growth retardation;
H = formative effects;
I = increased chlorophyl;
P = terminal bud kill;
S = albinism;
U = unusual pigmentation;
X = axillary stimulation;
6F = delayed flowering; and
6Y = abscised buds or flowers.

The ratings are summarized in Table A. The compounds tested are highly active herbicides. Certain of the compounds have utility for weed control in wheat.

Compound Structures

Compound 1



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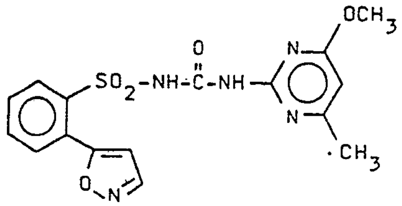

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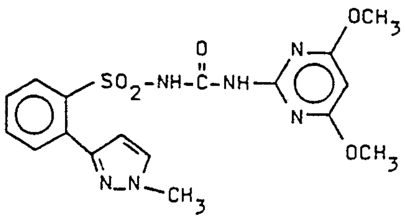

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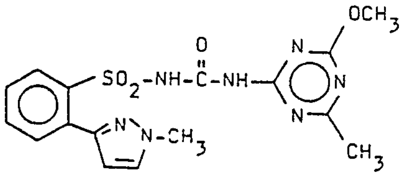

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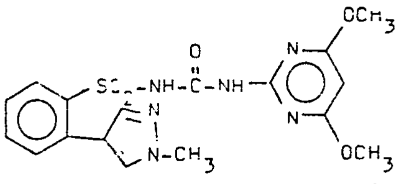

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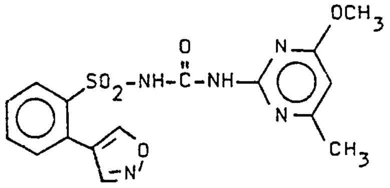

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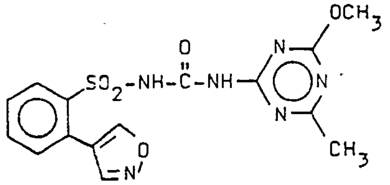

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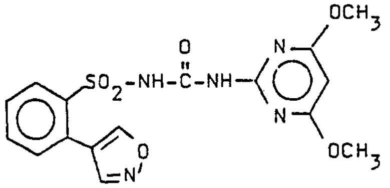

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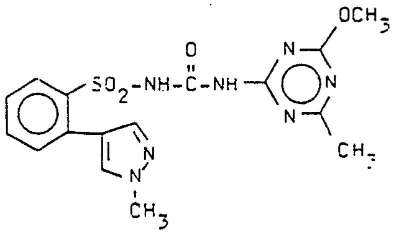

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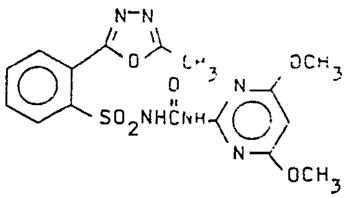

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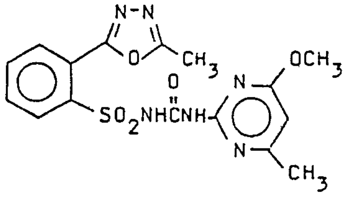

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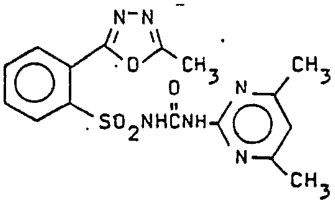

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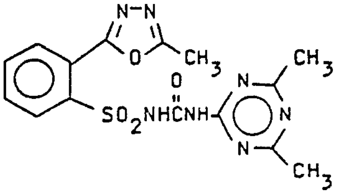

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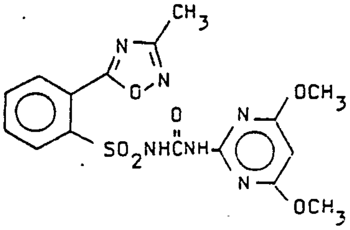

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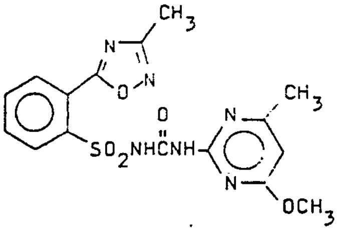

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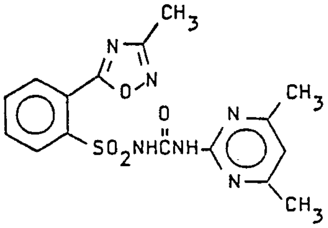

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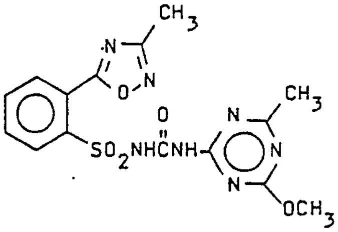

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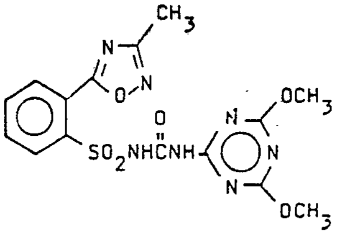

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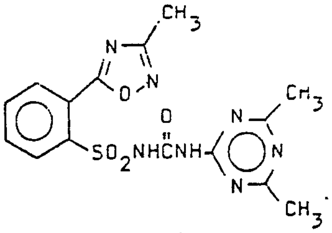

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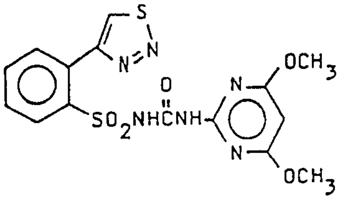

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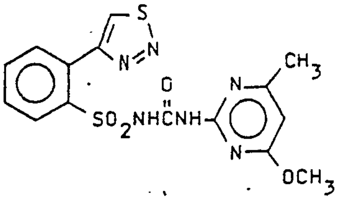

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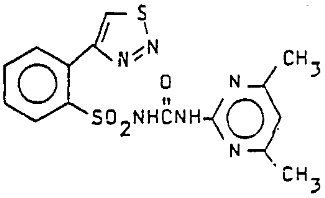

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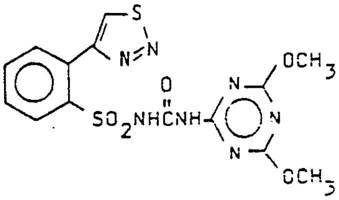

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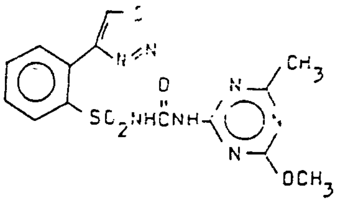

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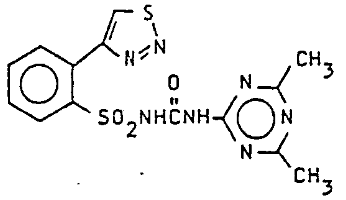

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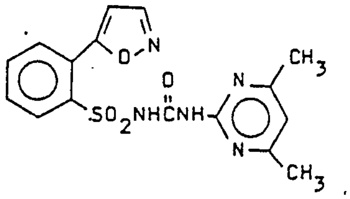

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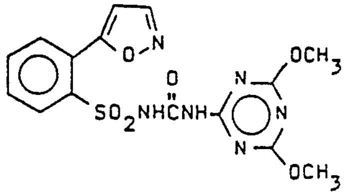

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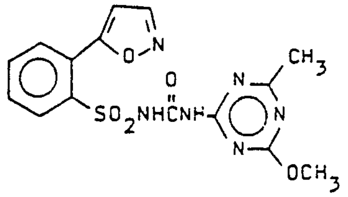

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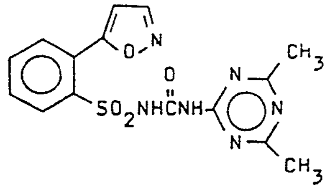

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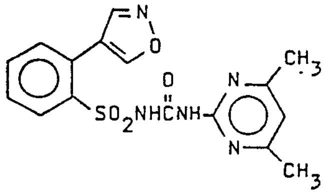

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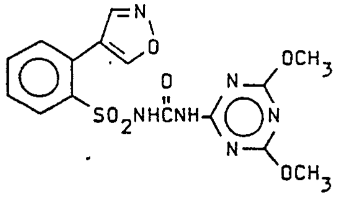

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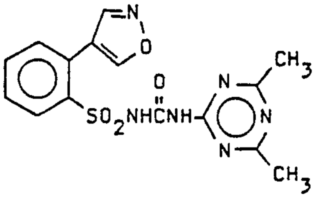

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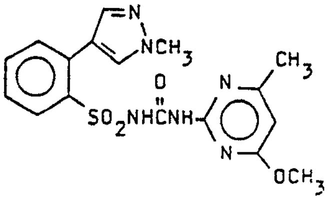

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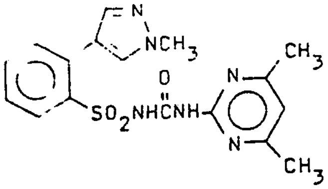

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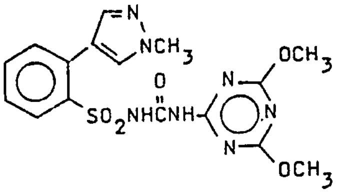

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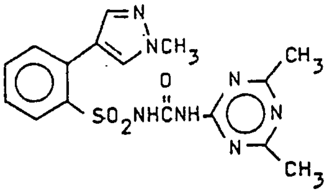

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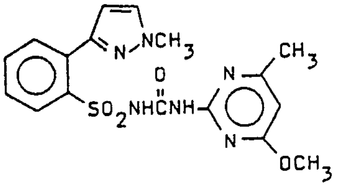

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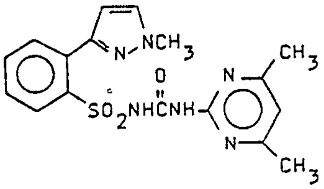

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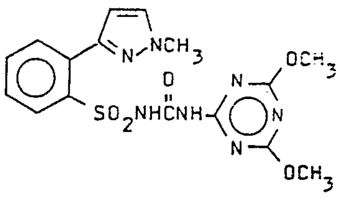

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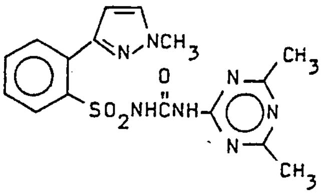

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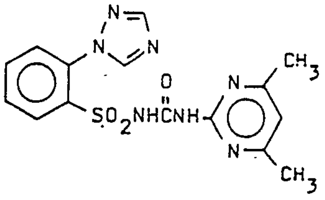

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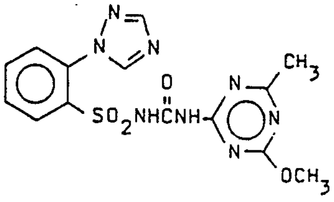

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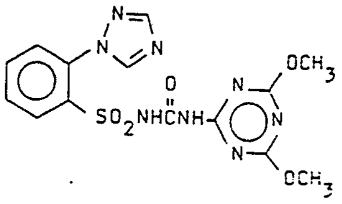

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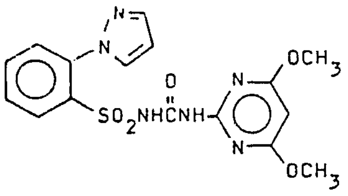

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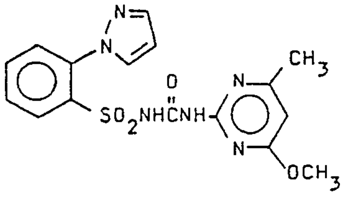

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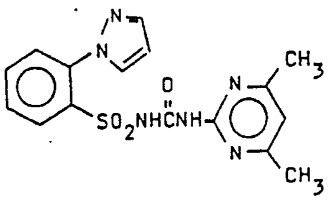

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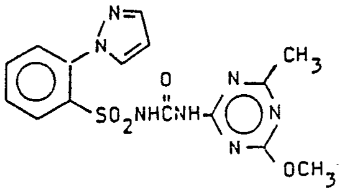

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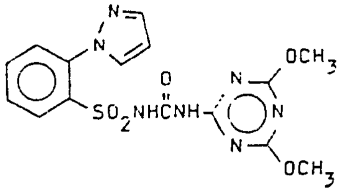

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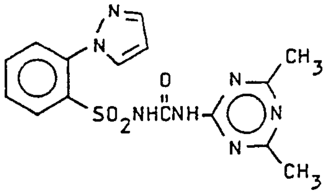

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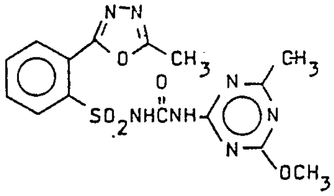

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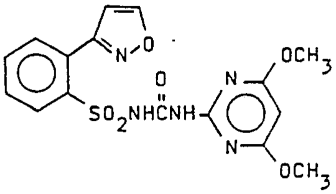

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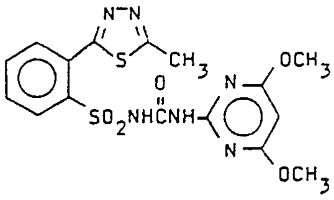

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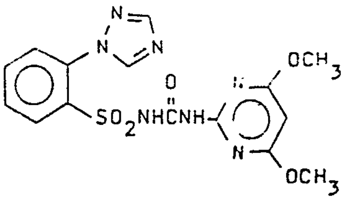

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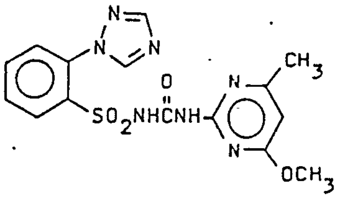

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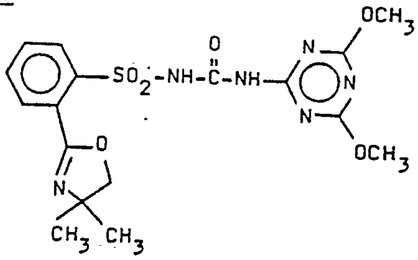

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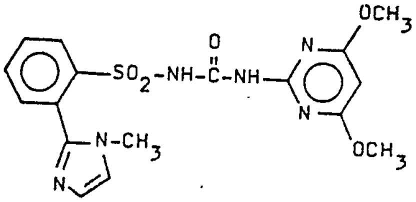

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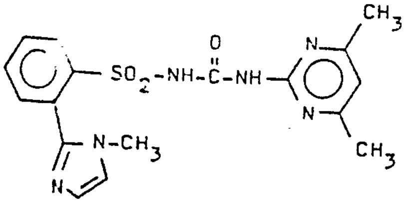
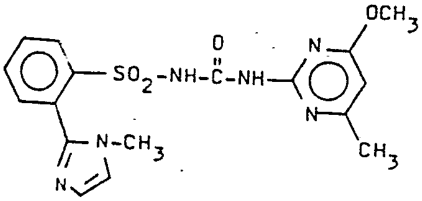
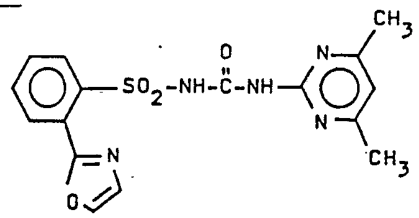


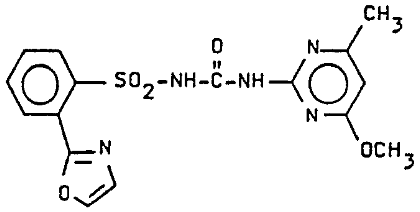
Compound 58



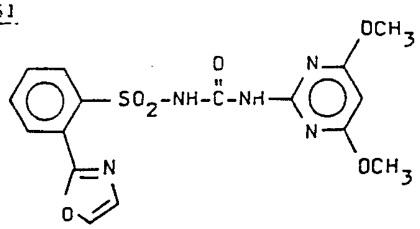
Compound 59



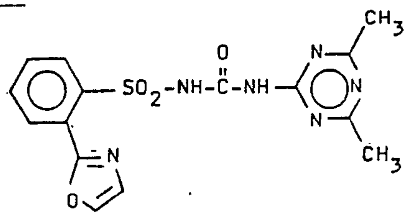
Compound 60



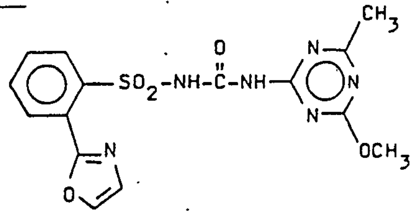
Compound 61



Compound 62



Compound 63



Compound 64

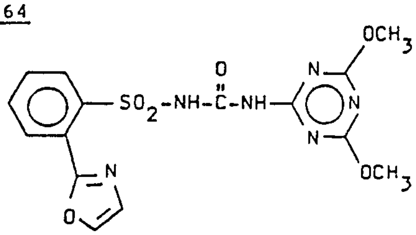


TABLE A

	Cmpd. 1	Cmpd. 2	Cmpd. 3	Cmpd. 3	Cmpd. 4	Cmpd. 4	Cmpd. 5	Cmpd. 5
Rate kg/ha	.05	.05	0.4	.05	0.4	.05	0.4	.05
POST-EMERGENCE								
Bush bean	9C	10C	9C	9C	9C	5C,9G,6Y	5C,9G,6Y	6C,9G,6Y
Cotton	9C	9C	6C,9G	6C,9G	6C,9G	5C,9G	6C,9G	6C,9G
Morningglory	4C,8G	10C	6C,9G	6C,9G	5C,9G	5C,9H	5C,9H	6C,9G
Cocklebur	9C	10C	9C	6C,9G	9C	5C,9H	5C,9H	10C
Sicklepod	9C	9C	6C,9G	6C,9G	3C,5H	4C,5H	4C,9G	6C,9G
Nutsedge	6C,9G	4C,8G	6C,9G	6C,9G	2G	0	6C,9G	5C,9G
Crabgrass	2C,6G	2C,8G	2C,9G	5G	2C,8G	1C	3C,8G	2C,5H
Barnyardgrass	9C	9C	9C	4C,9H	10C	9C	10C	10C
Wild Oats	8G,5X	3C,9G	2C,9G	2C,9G	2C	0	3C,9H	2C,5G
Wheat	2C,7G	3C,9G	2U,9G	3C,9G	0	0	2C,9G	2C,3G
Corn	5U,9G	5U,9G	5U,9G	3U,9G	7U,9C	5C,9G	5C,9G	5C,9G
Soybean	9C	9C	6C,9G	9C	3C,9G	3C,9G	5C,9G	6C,9G
Rice	5C,9G	6C,9G	5C,9G	5C,9G	1C,3G	0	6C,9G	5C,9G
Sorghum	3U,9G	3U,9G	4U,9G	1U,9G	3C,9H	2C,9G	1C,9H	9H
POST-EMERGENCE								
Morningglory	3C,9G	9C	3C,9G	9G	3C,9G	9G	2C,9H	3C,9G
Cocklebur	9H	9H	9H	9H	3C,9H	8H	9H	2C,9H
Sicklepod	3C,9G	3C,9G	3C,9G	5C,8G	3C,7H	2C,5H	2C,9G	3C,9H
Nutsedge	10E	10E	10E	10E	0	0	10E	10E
Crabgrass	2C,5G	2C,8G	2C,6G	1C,3G	1C	0	3C,9G	2C,6G
Barnyardgrass	5C,9H	6C,9H	9H	2C,9H	2C,9H	2C,6H	6C,9H	5C,9H
Wild Oats	2C,9G	5C,9G	2C,9G	2C,9H	1C	0	3C,9G	2C,9G
Wheat	2C,9G	2C,9G	9H	2C,9G	0	0	1C,9G	2C,9G
Corn	2U,9G	9G	2C,9G	2C,9G	2C,9G	9G	5C,9G	2C,9G
Soybean	9H	9H	9H	2C,8H	9H	2C,5H	9H	8H
Rice	10E	10E	10E	2C,9H	2C,5G	2C,3G	10E	5C,9H
Sorghum	2C,9G	9H	10H	2C,9H	2C,9H	2C,9H	2C,9H	2C,8H

TABLE A (continued)

	Cmpd. 6	*Cmpd. 7*	*Cmpd. 8*	*Cmpd. 8*	*Cmpd. 9*	*Cmpd. 9*
Rate kg/ha	.05	.05	0.4	.05	0.4	.05
POST-EMERGENCE						
Bush bean	9C	9C	9C	9C	9C	9C
Cotton	6C,9G	9C	9C	6C,9G	9C	6C,9G
Morningglory	6C,9G	10C	10C	9C	6C,9G	9C
Cocklebur	6C,9G	9C	9C	9C	9C	9C
Sicklepod	6C,9G	9C	10C	9C	6C,9G	6C,9G
Nutsedge	9C	8C	10C	10C	6C,9G	1C,5G
Crabgrass	2C,7H	2C,6G	3C,8G	3C,7H	3C,8G	2C,7G
Barnyardgrass	9C	6C,9H	9C	9C	9C	10C
Wild Oats	2C,9G	2C	3C,9G	2C,5G	5C,9G	2C,8G
Wheat	3C,9G	0	4C,9G	2C,8G	1C	0
Corn	9C	5U,9G	9C	5U,9G	7U,9C	5U,9C
Soybean	9C	9C	9C	9C	9C	2C,9G
Rice	5C,9G	2C,7G	6C,9G	4C,9G	4C,9G	3C,9G
Sorghum	4U,9C	3C,8G	4U,9C	4U,9G	6U,9G	2C,9G
PRE-EMERGENCE						
Morningglory	9C	10C	9C	1C,9H	9C	1C,9H
Cocklebur	9H	9H	9H	9H	-	-
Sickelpod	5C,9G	9C	9C	3C,9G	9C	7C,9G
Nutsedge	10E	1C,6G	10E	10E	5C,9G	6G
Crabgrass	2C,9G	1C,3G	2C,9G	2C,7H	4C,8G	2C,5G
Barnyardgrass	5C,9H	2C,7H	9C	5C,9H	9C	3C,9H
Wild Oats	3C,9G	2C,6G	2C,9H	2C,9G	3C,9H	3C,8G
Wheat	2C,9H	2C	9H	2C,9G	3G	0
Corn	5C,9G	1C,9G	9C	3C,9G	3C,9H	2C,8H
Soybean	9H	9H	9H	2C,8H	9H	3C,7H
Rice	10E	5G	10E	5C,9H	10E	3C,8G
Sorghum	7C,9H	1C,6H	7C,9H	2C,9H	2C,9H	9G

TABLE A (continued)

	Cmpd. 10	Cmpd. 11	Cmpd. 12	Cmpd. 13	Cmpd. 14	Cmpd. 15	Cmpd. 16	Cmpd. 17
Rate kg/ha	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
POST-EMERGENCE								
Bush bean	9C	9C	9C	6C,9G,6Y	9C	9C	9C	9C
Cotton	5C,9G	4C,9G	5C,9G	4C,8G	5C,9G	4C,9G	4C,9G	4C,9G
Sorghum	9C	10C	10C	5C,9G	2U,9G	5C,9G	2C,9G	9G
Corn	10C	10C	9C	5U,9G	5U,9H	6U,9G	4C,9G	3C,9H
Soybean	9C	9C	9C	1C,2G	5C,9G	5C,9G	4C,9G	5C,9G
Wheat	5C,9G	9C	9C	0	1C,6G	9C,9G	2C,8G	0
Wild Oats	9C	9C	9C	5C,9H	2C,9G	6C,9G	5C,9H	2C
Rice	6C,9G	6C,9G	6C,9G	5C,9G	5C,9G	5C,9G	5C,9G	6G
Barnyardgrass	9C	9C	9C	5C,9H	9C	9C	5C,9H	3C,6H
Crabgrass	9G	5C,9G	9C	2C,5G	5C,9G	2C,9G	3C,8G	1C
Morningglory	9C	10C	10C	4C,8H	5C,9G	4C,9G	3C,8G	9C
Cocklebur	9C	10C	10C	5C,9H	10C	5C,9G	3C,9H	9C
Sicklepod	9C	9C	9C	3H,5C	9C	5C,9G	3C,7H	4C,9G
Nutsedge	5C,9G	10C	6C,9G	3C,9G	10C	9C	4C,9G	5C,9G
Sugar beet	9C	9C	9C	5C,8G	9C	9C	5C,9G	9C
PRE-EMERGENCE								
Sorghum	10H	10H	6C,9H	2C,8H	10H	10H	3C,9H	4C,9H
Corn	5C,9H	10H	5C,9G	4C,7G	4C,9G	10E	3C,9H	3C,9H
Soybean	9H	9H	3C,6H	1C	9H	9H	8H	2C,9H
Wheat	3C,9H	5C,9H	4C,9G	2G	5C,9G	5C,9G	2C,9G	0
Wild Oats	4C,8G	6C,9H	5C,9H	6H	5C,9G	5C,9G	2C,9H	1C
Rice	10E	10E	10E	2C,5G	10E	10E	10E	3C,6G
Barnyardgrass	5C,9G	6C,9G	5C,9G	3C,7G	5C,9H	5c,9H	3C,8H	2G
Crabgrass	2C,8G	6C,9G	5C,9G	3G	2C,4G	3C,7G	1C,4G	1C
Morningglory	9C	9C	9G	2C,3H	9C	9C	9H	9C
Cocklebur	9H	9H	9G	9G	9H	9H	9H	9H
Sicklepod	9G	9G	9G	1C	5c,9G	9C	8H	5C,9G
Nutsedge	10E	10E	10E	0	10E	10E	10E	10E
Sugar beet	10E	10E	10E	5G	10E	10E	5C,9G	9C

TABLE A (continued)

	Cmpd. 18	Cmpd. 19	Cmpd. 20	Cmpd. 21	Cmpd. 22	Cmpd. 23	Cmpd. 24	Cmpd. 25
Rate kg/ha	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
POST-EMERGENCE								
Bush bean	9C	4C,9G,6Y	9C	6C,9G,6Y	9C	9C	9C	5C,9G,6Y
Cotton	4C,9G	3C,4G	6C,9G	4C,9G	4C,9G	5C,9G	6C,9G	4C,9G
Sorghum	1C,8H	2G	3U,9G	5C,9G	9C	1C,5H	2C,6H	2U,9G
Corn	3C,9H	1C,4G	2U,9G	5U,9G	9C	1U,9H	2U,9H	3U,9G
Soybean	5C,9G	4H	4C,8G	4C,9G	2C,8H	5C,9G	2C,8G	3C,9G,5X
Wheat	0	0	1C	1C,7G	1C,9G	1C	1C,3G	1C,2G
Wild Oats	0	0	2C	3C,9H	9C	1C	1C	1C,4G
Rice	1C,2G	3G	4C,8G	5C,9G	5C,9G	4G	1C,5G	5C,9G
Barnyardgrass	0	0	5C,8H	6C,9H	9C	2C,5H	1C,4H	3C,8H
Crabgrass	0	0	3C,8G	2C,8G	6C,9G	3G	0	2C,5G
Morningglory	5C,9G	3C,4H	5C,9G	5C,9G	5C,9G	5C,9G	9C	5C,9G
Cocklebur	10C	2C,8H	9C	10C	9C	10C	10C	10C
Sicklepod	5C,9G	1C	5C,9G	5C,9G	9C	3C,8G	6C,9G	3C,7G
Nutsedge	4C,9G	2C,7G	9C	9C	10C	2C,6G	4C,8G	5C,9G
Sugar beet	9C	4G	9C	9C	9C	9C	9C	9C
PRE-EMERGENCE								
Sorghum	2C,8H	3H	5C,9H	5C,9H	5C,9H	2C,7H	2C,8H	2C,9H
Corn	9H	2C,7H	2C,9G	2C,9G	10E	3C,8G	2C,9H	2C,9H
Soybean	9H	2C,2H	2C,2H	2C,6H	2C,5H	3C,4H	3C,4H	3C,3H
Wheat	0	0	5G	1C,9G	2C,9H	0	0	1C,7G
Wild Oats	0	0	7G	2C,9G	3C,9H	0	1C,3G	3C,8H
Rice	2C,4G	2C,4G	10E	5C,9H	10E	2C,6G	3C,7G	10E
Barnyardgrass	2C	0	5C,9H	5C,9H	5C,9H	3C,5H	2C,6G	2C,5H
Crabgrass	0	0	2C	2C,5G	2C,5G	1C	0	1C,2G
Morningglory	9C	2C,9H	9C	5C,9G	2C,9G	5C,9G	9C	9G
Cocklebur	9H	8H	-	9H	9H	8H	9H	-
Sicklepod	4C,9G	3H	3C,9G	2C,9G	2C,9G	3C,9G	3C,9G	5C,9G
Nutsedge	10E	0	10E	10E	10E	10E	7G	2C,7G
Sugar beet	5C,9G	7G	5C,9G	10E	5C,9G	9C	10E	6C,9G

TABLE A (continued)

	Cmpd. 26	*Cmpd. 27*	*Cmpd. 28*	*Cmpd. 29*	*Cmpd. 30*	*Cmpd. 31*	*Cmpd. 32*	*Cmpd. 33*
Rate kg/ha	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
POST-EMERGENCE								
Bush bean	9C	9C	9C	9C	9C	6C,9G,6Y	4C,9G,6Y	6C,9G,6Y
Cotton	5C,9G	5C,9G	9C	5C,9G	9C	5C,9G	4C,9G	6C,9G
Sorghum	5C,9G	2C,7H	2C	1C,9G	4C,9G	2C,9G	1C,9G	2C,9H
Corn	3U,9C	3C,9G	3U	4U,9C	2U,8G	9G	3U,9G	2C,8G
Soybean	4C,8G	9C	9C	9C	9C	9C	6C,9G	5C,9G
Wheat	6C,9G	0	4G	8G	6C,9G	9G	3G	2C,9G
Wild Oats	6C,9G	0	1C	2C,9H	9C	9G	2C,9G	4C,9G
Rice	5C,9G	1C,7G	4C	5C,9G	5C,9G	4C,9G	4C,9G	5C,9G
Barnyardgrass	10C	2C,9H	5C	3C,9H	9C	5C,9H	5C,9H	5C,9H
Crabgrass	4C,9G	1C,5G	6G	3C,8G	4C,8G	1C,5G	2C,6G	5C,9G
Morningglory	10C	10C	10C	10C	2C,8G	5C,9G	2C,7G	9C
Cocklebur	10C	3C,9G	10C	9C	9C	5C,9G	5C,9G	6C,9G
Sicklepod	9C	5C,9G	9C	5C,9G	5C,9G	4C,8H	4C,7H	4C,8H
Nutsedge	5C,9G	2C,6G	6G	1C,5G	9G	2C,8G	4G	8G
Sugar beet	-	-	-	-	5C,9G	5C,9G	4C,9G	4C,9G
PRE-EMERGENCE								
Sorghum	10H	1C,6G	4C,8H	2C,9H	5C,9H	4C,9H	3C,8G	5C,9H
Corn	10H	3C,9G	2C,9G	3U,9H	9H	3C,9G	1C,8G	9H
Soybean	9H	9H	8H	8H	9H	2C,8H	1C	8H
Wheat	5C,9H	0	4G	9H	1C,9H	8G	1C,5G	3C,9G
Wild Oats	5C,9H	2C	2C,6G	10H	4C,9H	8G	2C,7H	4C,8G
Rice	10E	4C,9H	5C,9H	10E	10E	3C,8H	5C,9H	4C,9H
Barnyardgrass	5C,9H,	2C,5H	2C,7H	10H	9H	3C,9H	3C,7H	5C,9H
Crabgrass	2C,8G	2C	2C	2C,5G	1C,7G	2C	0	3C,8G
Morningglory	9H	9H	9C	9G	7G	9G	1C	9G
Cocklebur	9H	9H	9H	9H	9H	2C,3H	1C	9H
Sicklepod	9G	9G	9G	9G	9G	2C,2H	4C,6H	2C,8G
Nutsedge	10E	10E	9G	9G	10E	9G	0	10E
Sugar beet	10E	10E	10E	10E	10E	4C,9G	10E	10E

TABLE A (continued)

	Cmpd. 34	*Cmpd. 35*	*Cmpd. 36*	*Cmpd. 37*	*Cmpd. 38*	*Cmpd. 39*	*Cmpd. 40*	*Cmpd. 40*
Rate kg/ha	0.05	0.05	0.05	0.05	0.05	0.05	0.4	0.05
POST-EMERGENCE								
Bush bean	5C,9G,6Y	3C,9G,6Y	4C,9G,6Y	5C,9G,6Y	4C,5G,6Y	6C,9G,6Y	6C,6G,6Y	1C
Cotton	4C,9G	3C,9G	4C,9G	4C,9G	2C,5G	3C,8G	4C	1C
Sorghum	4C,9G	2C,7H	9G	4C,9G	3C,7H	2C,5H	3C,8H	1C
Corn	2C,9G	3C,9H	2C,9H	2C,9H	0	2C,8H	2C,9H	1C
Soybean	9C	4C,9G	2C,8H	4C,9G	3C,8G	3C,8G	3C,7G,7X	1C
Wheat	9G	0	2C,9G	2C,9G	1C,5G	0	1C	0
Wild Oats	4C,9H	0	2C,9H	4C,9G	2C	0	2C	0
Rice	6C,9G	1C,4G	4C,9G	4C,8G	3C,8G	0	3C,9G	1C
Barnyardgrass	5C,9G	4C,9H	4C,9H	5C,8H	5C,8H	4C,7H	2C,8H	1C
Crabgrass	3C,9G	1C,3G	1C,6G	8G	1C,4G	1C	1C,3G	0
Morningglory	5C,9G	4C,9G	5C,9G	5C,9G	4C,8G	4C,8H	3C,7H	4C
Cocklebur	5C,9G	3C,8H	4C,9H	4C,9G	2C,2H	4C,8H	3C	1C
Sicklepod	5C,8H	4C,8H	4C,8H	4C,6H	2C,2H	5C	4C	1C
Nutsedge	9G	1C	1C,9G	2C,9G	5G	0	0	0
Sugar beet	2C,8G	4C,9G	3C,8H	2C,7G	2C,5G	3C,8G	4C,7G	1C
PRE-EMERGENCE								
Sorghum	4C,9H	1C,4G	2C,8H	5C,9H	2C,5G	3C,5G	2C,3G	1C
Corn	3C,9G	2C,5G	2C,5G	2C,9G	1C	4C,8G	3C,6H	1C
Soybean	3C,7H	0	0	3C,6H	0	4C,3H	2C	0
Wheat	3C,9G	0	1C	2C,9G	3G	1C,2G	0	0
Wild Oats	4C,8G	0	2C	4C,8G	2G	1C,3G	0	0
Rice	3C,9H	0	3C,8H	3C,9H	1C	0	2C	1C
Barnyardgrass	5C,9H	3C,8H	2C,5G	4C,9H	2C,4G	2C,5G	2C	1C
Crabgrass	2C,8g	2C	1C	2C,7G	2G	0	1G	3G
Morningglory	9C	9G	2C,7C	9C	2C,4G	3C,8H	3C,7H	0
Cocklebur	9H	9H	-	9H	0	3C,9H	0	2G
Sicklepod	3C,8G	2C,7G	1C	2C,5G	2C	3C	0	0
Nutsedge	10E	7G	7G	8G	4G	1C	0	3G
Sugar beet	4C,9G	10E	3C,6G	2C,7G	0	3C,7G	2G	0

TABLE A (continued)

	Cmpd. 41	*Cmpd. 42*	*Cmpd. 43*	*Cmpd. 44*	*Cmpd. 45*	*Cmpd. 46*	*Cmpd. 47*
Rate kg/ha	0.05	0.05	0.05	0.05	0.05	0.05	0.05
POST-EMERGENCE							
Bush bean	4C,9G,6Y	6C,9G,6Y	6C,9G,6Y	9C	9D,9G,6Y	2C,9H,6Y	9C
Cotton	5C,9G	4C,8H	4C,4H	5C,9G	6C,9G	6C,9G	5C,9H
Sorghum	5C,9G	2C	3C,4G	9H	5C,9G	3C,9G	2C,8H
Corn	2C,9G	2C,8H	2C	3U,9G	2U,9H	3C,8H	2C,8H
Soybean	4C,8H	2C,5H	2C,2H	5C,9G	5C,9G	5C,9G	5C,9G
Wheat	3C,9G	0	0	0	1C	1C,3G	0
Wild Oats	2C,9G	0	0	2C,6G	3C,9G	2C,9G	1C
Rice	5C,9G	2C	1C	2C,9G	5C,9G	5C,9G	4G
Barnyardgrass	4C,8H	2C	1C	9C	5C,9H	3C,7H	0
Crabgrass	1C,4G	1C	0	2C,7G	8G	3C,5H	0
Morningglory	5C,9G	3C	2C,2H	5C,9G	5C,9G	5C,9G	5C,9H
Cocklebur	5C,9G	3C	1C	5C,9G	5C,9G	4C,9G	10C
Sicklepod	5C,9G	2C	2C,5G	9C	5C,9G	4C,8H	5C,9G
Nutsedge	2C,5G	0	0	10C	5C,9G	4C,9G	5G
Sugar beet	-	-	-	5C,9G	9C	3C,8G	9G
PRE-EMERGENCE							
Sorghum	3C,6G	0	1C	9C	9C	4C,9H	2C,9H
Corn	4C,7G	1C	2C,5G	9H	5C,9H	3C,9H	2C,8H
Soybean	1C,1H	0	0	9H	9H	3C,7H	2C,8H
Wheat	4G	0	0	0	1C,7G	4G	0
Wild Oats	1C,3G	0	0	3C,5G	2C,9G	3C,9G	0
Rice	3C,5H	0	0	5C,9H	5C,9H	4C,9G	3C,4G
Barnyardgrass	1C	2H	1C	4C,9H	2C,8H	2C	0
Crabgrass	0	2G	2G	1C,3G	2C	1C	0
Morningglory	8H	0	0	9C	9C	9H	9C
Cocklebur	8H	0	0	9H	9H	9H	9H
Sicklepod	2C	0	0	5C,9G	5C,9G	2C,9G	9C
Nutsedge	0	0	0	10E	9G	2C,8G	0
Sugar beet	-	-	-	10E	5C,9G	3C,9H	9C

TABLE A (continued)

	Cmpd. 48	*Cmpd. 49*	*Cmpd. 50*	*Cmpd. 51*	*Cmpd. 52*	*Cmpd. 53*	*Cmpd. 54*
Rate kg/ha	0.05	0.05	0.05	0.05	0.05	0.05	0.05
POST-EMERGENCE							
Bush bean	9C	5C,9G,6Y	6C,9G,6Y	9C	9C	9C	5S,9G,6Y
Cotton	5C,9G	4C,8H	4C,8G	9C	5C,9G	6C,9G	5C,9G
Sorghum	4C	0	5U,9G	5U	3C,9G	2C,9G	5C,9G
Corn	6G	2C,2H	9C	4U	1U,9H	2U,9G	5C,9G
Soybean	4C,9G	2C,9G	4C,9G	5C	5C,9G	9C	6C,9G
Wheat	0	0	0	9G	1C,7G	4G	8G
Wild Oats	0	0	1C,3G	2C	2C,9G	5G,5X	2C,9H
Rice	0	6G	8G	5C	6C,9G	4C,9G	5C,9G
Barnyardgrass	0	0	5C,9H	9C	9C	5C,9H	10C
Crabgrass	0	0	1C,5G	6C	3C,7G	3C,8G	3C,7G
Morningglory	9C	4C,9H	2C,8G	9C	5C,9G	10C	9C
Cocklebur	8G	3C,9G	10C	9C	9C	9C	9C
Sicklepod	4C,9G	3C,4H	4C,8H	9C	5C,9G	9C	4C,6G
Nutsedge	9G	2C	6C,9G	10C	6C,9G	5C,9G	2C,8G
Sugar beet	9C	9C	9C	9C	9C	-	-
PRE-EMERGENCE							
Sorghum	2C,7H	3G	2C,9H	10H	5C,9H	10E	6C,9H
Corn	3C,8H	2C,4G	5C,9H	9H	2C,8H	2C,9H	5C,9H
Soybean	9H	2C,4H	4C,7H	9H	3C,5H	9H	9H
Wheat	0	0	1C	2C,8G	8G	6G	2C,9H
Wild Oats	0	0	1C	2C,9G	2C,8H	2C,6G	3C,9H
Rice	1C	2C,4G	5C,8G	10E	10E	10E	9H
Barnyardgrass	0	0	2C,7H	3C,9G	5C,9H	2C,9G	3C,9H
Crabgrass	0	0	0	2C,3G	1C,5G	2C,7G	3C,6G
Morningglory	9C	2C,8H	8G	9C	9G	9C	2C,9G
Cocklebur	2C,8H	2C,7G	9H	9H	9H	9H	9H
Sicklepod	5C,9G	2C,7G	8G	2C,9G	2C,9G	9G	2C,8G
Nutsedge	7G	3G	2C,9G	10E	10E	10E	10E
Sugar beet	5C,9G	2C,6H	4C,8G	4C,9G	10E	-	-

TABLE A (continued)

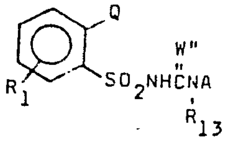
	Cmpd. 55	Cmpd. 56	Cmpd. 56	Cmpd. 57	Cmpd. 58	Cmpd. 58	Cmpd. 59
Rate kg/ha	0.4	0.4	0.05	0.4	0.4	0.05	0.05
POST-EMERGENCE							
Bush bean	9C	9D,9G,6Y	7C,9G,6Y	4C,5G	6C,8G,6Y	4C,8G,6Y	4C,9G,6Y
Cotton	5C,9G	7C,9G	5C,5H,9G	2C	4C,5H	2C	5C,9G
Morningglory	6C,9G	5C,9G	5C,9G	2C,3H	2C,7G	3C,7H	3C,9G
Cocklebur	2C,9G	2C,6G	2C,5G	1C	4C,8G	1C	5C,9G
Sicklepod	5C,9G	3C,7G	3C,7G	2C	4C,4H	1C	5C,8G
Nutsedge	5G	2C,7G	1C,8G	0	2C,8G	2G	3C,8G
Crabgrass	1C	3C,7G	3C,7G	2G	2C,5G	2G	1C,3H
Barnyardgrass	3C,8H	9C	5C,9H	2C,4G	4C,9H	3H	9C
Wild Oats	0	4C,9G	3C,8H	2C,6G	2C,9G	1H	2C,9H
Wheat	0	3C,9G	9G	1C,5G	1C,8G	1H	2C,9G
Corn	3C,9H	2C,7H	2C,5H	0	2C,3H	2C	2U,9G
Soybean	3C,9G	4C,9G	3C,8G	2C,3H	3C,8G	2C,4H	4C,9G
Rice	3C,8G	5C,9G	6C,9G	6C,8G	4C,9G	5C,7G	5C,9G
Sorghum	2C,9G	6C,9G	4C,9G	2C,5G	4C,8H	2C,5G	3U,9G
PRE-EMERGENCE							
Morningglory	9G	8H	2C,9G	3C,7H	2C,9G	4C	7G
Cocklebur	9G	9H	9H	2C,5G	2C,9H	7G	8H
Sicklepod	9G	3C,8G	2C,6G	3C	2C	3C	3C,7G
Nutsedge	9G	2C,8G	6G	0	2C,5G	2G	9G
Crabgrass	2G	7C,9G	5C,8G	1C	4C,7G	2C	2C
Barnyardgrass	4C,9H	7C,9H	7C,9H	3C	3C,6H	2C	2C,7H
Wild Oats	5G	6C,9H	6C,9H	0	3C,8H	2C,6G	2C,9G
Wheat	0	5C,9H	5C,9H	0	3C,8G	1C	2C,7G
Corn	2C,8G	5C,9H	6C,9H	1C,5G	3C,6G	2C	1C,8G
Soybean	2C,8H	4C,6H	4C,5H	1C	4C	1C	2C,6H
Rice	2C,8G	10E	9H	1C,3G	5C,9H	3C	2C,8H
Sorghum	2C,7G	5C,9H	6C,9H	1C	4C,8G	2C	2C,9H

TABLE A (continued)

	Cmpd. 60	*Cmpd. 61*	*Cmpd. 62*	*Cmpd. 63*	*Cmpd. 64*
Rate kg/ha	0.05	0.05	0.05	0.05	0.05
POST-EMERGENCE					
Bush bean	9C	9C	3C,9G,6Y	9C	9C
Cotton	6C,9G	5C,9H	3C,7H	6C,9G	5C,9G
Morningglory	9C	6C,9G	4C,9G	9C	9C
Cocklebur	9C	9C	2C,8H	9C	9C
Sicklepod	9C	9C	4C,5H	9C	9C
Nutsedge	9C	9C	2C,5G,5X	1C,5G	3C,8G
Crabgrass	2C,7G	2C,8G	1C	1C,5G	1C,2H
Barnyardgrass	9C	6C,9H	3C,7H	4C,6H	3C,9H
Wild Oats	9C	2C,9G	2C,7G	3C	1C,4G
Wheat	6U,9G	3C,9G	0	0	0
Corn	6U,9G	6U,9G	2U,8G	4U,9G	2C,8H
Soybean	9C	9C	3C,8G	6C,9G	9C
Rice	5C,9G	5C,9G	5C,9G	2C,8G	6G
Sorghum	5U,9H	6U,9C	2C,7H	1C,9H	1C,8H
PRE-EMERGENCE					
Morningglory	2C,9G	9H	8H	9H	9C
Cocklebur	9H	-	9H	8H	3C,9H
Sicklepod	2C,9G	3C,9G	2C,5H	5C,9G	2C,9G
Nutsedge	10E	10E	0	5G	8G
Crabgrass	2C,7G	2C,8G	0	1C,5H	1C
Barnyardgrass	6C,9H	6C,9H	1C	2C,8G	4C,7H
Wild Oats	5C,9H	2C,9G	2C,6G	2C	1C,8G
Wheat	2C,9H	1C,8G	0	0	1C
Corn	9H	2U,9H	2C	2C,8G	2C,8H
Soybean	9H	9H	2C	3C,8H	9H
Rice	10E	10E	2C	2C,7G	3C,8G
Sorghum	10H	10H	2C	2C,8H	2C,8H

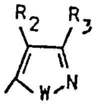
CLAIMS

1. A compound of the formula:



wherein

Q is



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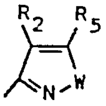

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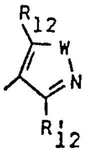

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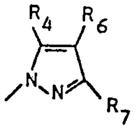

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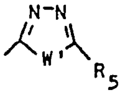

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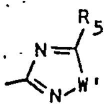

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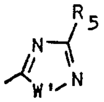

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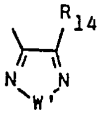

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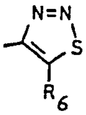

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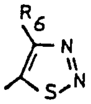

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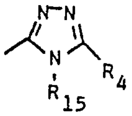

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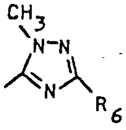

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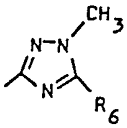

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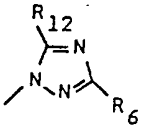

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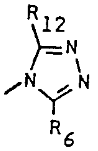

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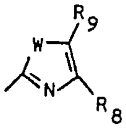

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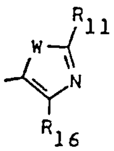

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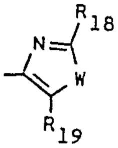

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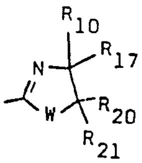

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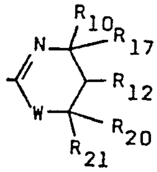
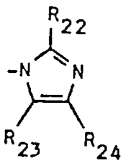


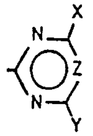
Q_{20}

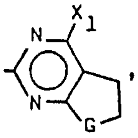
or

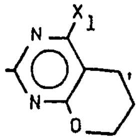


Q_{21} ;

W is O, S or NR;
W′ is O or S;
W″ is O or S;
R is H or C_1-C_4 alkyl;
R_1 is H, F, Cl, Br, CH_3, CF_3 or OCH_2;
R_2 is H, CH_3, C_2H_5, Cl or Br;
R_3 is H, CH_3, C_2H_5, Cl, Br, OCH_3, OC_2H_5 or SCH_3;
R_4 is H or C_1-C_4 alkyl;
R_5 is H, CH_3, C_2H_5, Cl, Br, OCH_3, OC_2H_5 or SCH_3;
R_6 is H, CH_3 or C_2H_5;
R_7 is H or C_1-C_4 alkyl;
R_8 is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_9 is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_{10}, R_{17}, R_{20} and R_{21} are independently H or C_1-C_4 alkyl;
R_{11} is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_{12} is H or CH_3;
R'_{12} is H or CH_3;
R_{13} is H or CH_3;
R_{14} is H, CH_3, C_2H_5, Cl, OCH_3, OC_2H_5 or SCH_3;
R_{15} is C_1-C_3 alkyl;
R_{16} is H, CH_3 or C_2H_5;
R_{18} is H, CH_3, C_2H_5, OCH_3, OC_2H_5, SCH_3, Cl or Br;
R_{19} is H, CH_3 or C_2H_5;
R_{22} is H, CH_3, C_2H_5, Cl, Br, CH_3O, C_2H_5O or CH_3S;
R_{23} is H, CH_3 or C_2H_5;
R_{24} is H, CH_3 or C_2H_5;

A is 

, 

, 

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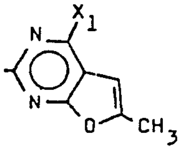
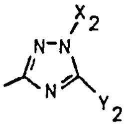
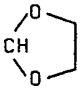


or 

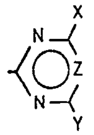
;

X is CH_3, OCH_3 or Cl;
X_1 is CH_3, OCH_3 or Cl;
X_2 is C_1-C_3 alkyl or CH_2CF_3;
Y is CH_3, C_2H_5, CH_2OCH_3, OCH_3, OC_2H_5, $CH(OCH_3)_2$, NH_2, $NHCH_3$, $N(CH_3)_2$, $OCH_2CH_2OCH_3$, OCH_2CF_3,

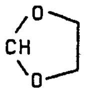
SCH_3, CF_3 or 

;

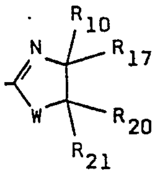
Y_2 is CH_3O, C_2H_5O, CH_3S or C_2H_5S;
Z is CH or N;
G is O or CH_2;
and their agriculturally suitable salts;
provided that
(a) when R_2 is Cl or Br, then W is O or S;
(b) when X is Cl, then Z is CH and Y is OCH_3, OC_2H_5, NH_2, $NHCH_3$ or $N(CH_3)_2$;
(c) when W″ is S, then R_{13} is H, A is

A is 

,

Y is CH_3, OCH_3, C_2H_5, OC_2H_5, CH_2OCH_3, $CH(OCH_3)_2$ or  ; and

Q is not



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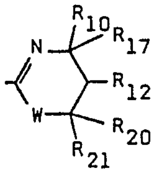
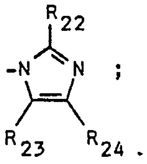
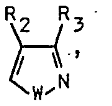
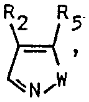


or

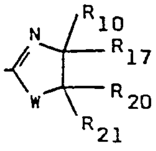


;

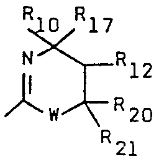
(d) when Q is  , then one of R_2 or R_3 must be H, CH_3 or C_2H_5;

(e) when Q is  , then one of R_2 or R_5 must be H, CH_3 or C_2H_5;

(f) The total number of carbon atoms of Q must be less than or equal to 8;
(g) when R_{18} is other than H, CH_3, C_2H_5 or CH_3S, then W is S or O;
(h) the total number of carbon atoms of R_{10}, R_{17}, R_{20} and R_{21} is less than or equal to 4; and
(i) when Q is



or



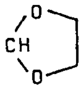
,

then R_{13} is H, R is other than H, X is Cl or OCH_3 and Y is OCH_3 or OC_2H_5.

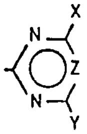
2. Compounds of Claim 1 where
R and R_{15} are independently CH_3 or C_2H_5;
R_2, R_3, R_4, R_5, R_6, R_7 and R_{14} are independently H or CH_3;
R_8, R_9, R_{11}, R_{16}, R_{18}, R_{19}, R_{22}, R_{23} and R_{24} are independently H, CH_3 or C_2H_5;
R_{10}, R_{17}, R_{20} and R_{21} are independently H or CH_3; and
W''' is O.

3. Compounds of Claim 2 where R_1 and R_{13} are H.

4. Compounds of Claim 3 where Y is CH_3, CH_2OCH_3, OCH_3, OC_2H_5, $CH(OCH_3)_2$ or



, and A is



.

5. Compounds of Claim 4 where W = O.
6. Compounds of Claim 4 where W = S.
7. Compounds of Claim 4 where W = NR.
8. Compounds of Claim 4 where W' = O.
9. Compounds of Claim 4 where W' = S.
10. A compound of Claim 1 where
W'' is O;
R and R_{15} are CH_3;
R_1, R_2, R_3, R_4, R_5, R_6, R_7, R_8, R_9, R_{10}, R_{11}, R_{12}, R_{13}, R_{14}, R_{16}, R_{17}, R_{18}, R_{19}, R_{20}, R_{21}, R_{22}, R_{23} and R_{24} are H;
X is CH_3, CH_3O or Cl;
Y is CH_3, CH_3O or CH_2OCH_3; and
Q is Q_1, Q_2, Q_3, Q_4, Q_5, Q_6, Q_7, Q_8, Q_9, Q_{10}, Q_{11}, Q_{12}, Q_{13}, Q_{14}, Q_{15}, Q_{16}, Q_{17}, Q_{18} or Q_{21}.

11. The compound of Claim 1 which is 2-(isoxazol-5-yl)-N-[(4,6-dimethoxypyrimidin-2-yl]aminocarbonyl]benzenesulfonamide.

12. The compound of Claim 1 which is 2-(isoxazol-5-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide.

13. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide.

14. The compound of Claim 1 which is N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1- and/or 2-methyl-1H-pyrazol-3-yl)benzenesulfonamide.

15. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide.

16. The compound of Claim 1 which is N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1-methyl-1H-pyrazol-4-yl)benzenesulfonamide.

17. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(isoxazol-4-yl)benzenesulfonamide.

18. The compound of Claim 1 which is 2-(isoxazol-4-yl)-N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]benzenesulfonamide.

19. The compound of Claim 1 which is 2-(isoxazole-4-yl)-N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide.

20. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide.

21. The compound of Claim 1 which is N-[(4-methyl-6-methoxypyrimidin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide.

22. The compound of Claim 1 which is N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide.

23. The compound of Claim 1 which is N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(5-methyl-1,3,4-oxidiazol-2-yl)benzenesulfonamide.

24. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide.

25. The compound of Claim 1 which is N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide.

26. The compound of Claim 1 which is N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide.

27. The compound of Claim 1 which is N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(3-methyl-1,2,4-oxadiazol-5-yl)benzenesulfonamide.

28. The compound of Claim 1 which is N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(1-methyl-1H-imidazol-2-yl)benzenesulfonamide.

29. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide.

30. The compound of Claim 1 which is N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide.

31. The compound of Claim 1 which is N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(oxazol-2-yl)benzenesulfonamide.

32. The compound of Claim 1 which is 2-(4,5-dihydro-5,5-dimethyloxazol-2-yl)-N-[(4,6-dimethoxy-1,3,5-triazin-2-yl)aminocarbonyl]benzenesulfonamide.

33. The compound of Claim 1 which is N-[(4,6-dimethoxypyrimidin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide.

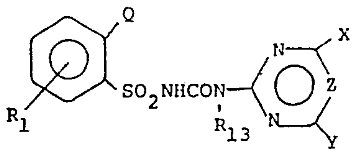
34. The compound of Claim 1 which is N-[(4-methoxy-6-methylpyrimidin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide.

35. The compound of Claim 1 which is N-[(4,6-dimethylpyrimidin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide.

36. The compound of Claim 1 which is N-[(4,6-dimethoxy-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide.

37. The compound of Claim 1 which is N-[(4-methoxy-6-methyl-1,3,5-triazin-2-yl)aminocarbonyl]-2-(1,2,3-thiadiazol-4-yl)benzenesulfonamide.

38. A compound of Claim 1 having the formula



wherein

Q is Q_{16}, Q_{17}, Q_{18}, Q_{19}, Q_{20} or Q_{21};

Y is CH_3, C_2H_5, CH_2OCH_3, OCH_3, OC_2H_5, $CH(OCH_3)_2$, NH_2, $NHCH_3$ or $N(CH_3)_2$;

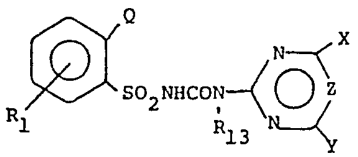
R_8 is H, CH_3 or C_2H_5; and

R_1, R_9, R_{10}, R_{11}, R_{13}, R_{16}, R_{17}, R_{18}, R_{19}, R_{20}, R_{21}, R_{22}, R_{23}, R_{24}, X and Z are defined in Claim 1;

and their agriculturally suitable salts.

39. A compound of Claim 1 wherein
Q is Q_{16}, Q_{17}, Q_{18}, Q_{19}, Q_{20} or Q_{21}

40. A compound of Claim 1 having the formula



wherein
Q is Q_1, Q_2, Q_3, Q_4, Q_5, Q_6, Q_7, Q_8, Q_{11}, Q_{12}, Q_{13}, Q_{14} or Q_{15},
R_4 is H or C_1-C_3 alkyl;
R_7 is H or C_1-C_3 alkyl;
Y is CH_3, C_2H_5, CH_2OCH_3, OCH_3, OC_2H_5, $CH(OCH_3)_2$, NH_2, $NHCH_3$ or $N(CH_3)_2$
and the remaining substituents are as defined in Claim 1;
and their agriculturally suitable salts.

41. Compounds of Claim 1, substantially as described herein with reference to Examples 1 to 40.

42. Compounds of Claim 1 as hereinbefore specifically disclosed in Tables 1 to 15.

43. A composition suitable for controlling the growth of undesired vegetation which comprises an effective amount of a compound or any of Claims 1 to 10 or 38 to 42 and at least one of the following: surfactant, solid or liquid diluent.

44. The composition of Claim 43 comprising a compound of any of claims 11 to 37.

45. The composition of Claim 43, substantially as described herein and as illustrated with reference to any of Examples 41 to 56.

46. A method for controlling the growth of undesired vegetation which comprises applying to the locus to be protected an effective amount of a compound of any of Claims 1 to 10 or 38 to 42.

47. The method of Claim 46 wherein a compound of any of Claims 11 to 37 is applied.

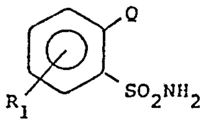
48. The method of Claim 46, substantially as hereinbefore desribed and as illustrated with reference with Table A herein.

49. A method for regulating the growth of plants which comprises applying to the locus of such plants an effective but substantially non-phytotoxic amount of a plant growth regulant compound according to any of Claims 1 to 10 or 38 to 42.

50. The method of Claim 49 wherein a compound of any of Claims 11 to 37 is applied.

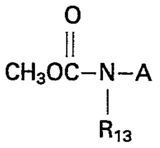
51. The method of Claim 49, substantially as illustrated in Table A herein.

52. A method for the preparation of a compound of Claim 1 which comprises reacting a sulfonamide of formula



(II)

wherein Q is as defined in Claim 1 other than Q_{19} or Q_{20} and R_1 is as defined in Claim 1, with a methyl carbamate of formulā



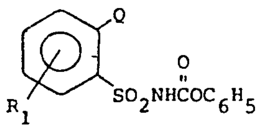
$$\underset{\displaystyle R_{13}}{\overset{\displaystyle O}{\overset{\|}{CH_3OC}}-\underset{|}{N}}-A$$

(III)

wherein A and R_{13} are as defined in Claim 1, under catalytic conditions.

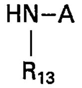
53. The method of Claim 52 wherein said reaction is performed in an inert solvent in the presence of trimethylaluminum.

54. A method for the preparation of a compound of Claim 1 which comprises reacting a sulfonyl carbamate of formula



(IIa)

with an appropriate amine of formula



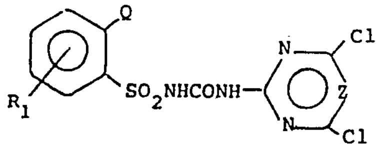
$$\begin{array}{l} HN-A \\ \;| \\ R_{13} \end{array}$$

(VII)

wherein R_1, R_{13}, A and Q are as defined in Claim 1 except that R_3 and R_{14} may only be H or C_1-C_2 alkyl, R_5 may only be C_1-C_2 alkyl and R may only be C_1-C_4 alkyl.

55. The method of Claim 54 wherein said process is carried out at 50-100°C in a solvent.

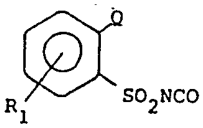
56. A method for preparation of a compound of Claim 1 which comprises displacing a chlorine atom in an intermediate of formula



(V)

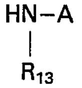
by OCH_3 or OC_2H_5 and if desired displacing the other chlorine atom by OCH_3; R_1, Q and Z being as defined in Claim 1 except that R_3 and R_5 may not be Cl or Br and R_{14} may not be Cl.

57. A method for the preparation of a compound of Claim 1 which comprises reacting a sulfonyl isocyanate of formula



(VI)

with an amine of formula

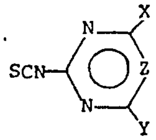


$$\begin{array}{l} HN-A \\ \;| \\ R_{13} \end{array}$$

(VII)

wherein R_1, R_{13} and A are as defined in Claim 1 and Q is as defined in Claim 52.

58. A method for the preparation of a compound of Claim 1 which comprises reacting the sulfonamide of formula (II) as defined in Claim 52 with an appropriate triazine or pyrimidine isothiocyanate of formula



wherein X, Y and Z are as defined in claim 1.

59. The method of any of Claims 52 to 58, substantially as described herein with reference to Examples 1 to 40.

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