

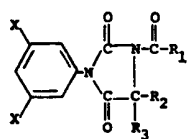
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(54) 1-CYCLOALKYLCARBONYL-3-(3,5-DIHALOPHENYL)
 IMIDAZOLIDINE-2,4-DIONES

(71) We, SUMITOMO CHEMICAL COMPANY LIMITED, a Japanese Body Corporate, of No 15, Kitshama 5-chome, Higashi-ku, Osaka-shi, Osaka-fu, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to 1-cycloalkylcarbonyl-3-(3,5-dihalophenyl)-imidazolidine-2,4-diones (hereinafter referred to as "1-cycloalkylcarbonyl-imidazolidinedione(s)") of the formula:



[I]

wherein each X is independently a chlorine or bromine atom, R₁ is a C₃—C₈ cycloalkyl, tetramethylcyclopropyl, 2,2-dimethyl-3-isobutenylcyclopropyl or 2,2-dimethyl-3-dichlorovinylcyclopropyl group and R₂ and R₃ are individually a hydrogen atom or a methyl group, and their preparation and use.

It is known that some 3-(3,5-dihalophenyl)imidazolidine-2,4-dione derivatives, of which 1- and 5-positions may be optionally substituted with various substituents, have a fungicidal activity on certain microorganisms (U.S. patent Nos. 3,668,217 and 3,716,552). We have now found that the above compounds [I] having a cycloalkylcarbonyl group at the 1-position exhibit a wide fungicidal activity which is markedly superior as compared with their homologues. These compounds show no material phytotoxicity to plants and low mammalian toxicity.

The 1-cycloalkylcarbonyl-imidazolidinediones [I], are especially effective in controlling the diseases of agricultural and horticultural crops caused by the phytopathogenic fungi belonging to *Alternaria* genus, such as pear black spot (*Alternaria kikuchiana*), Alternaria apple leaf spot (*Alternaria mali*), and tomato blight (*Alternaria solani*). In addition to these diseases, several plant diseases which cause serious damage in agriculture and horticulture can be effectively controlled by the application of the compounds of the invention. Those diseases include brown peach rot (*Sclerotinia cinerea*), citrus melanose (*Diaporthe citri*), common green citrus fruit mold (*Penicillium digitatum*), blue citrus fruit mold (*Penicillium italicum*), gray gape mold (*Botrytis cinerea*), gray mold and Sclerotinia rot of beans and vegetables (*Botrytis cinerea* and *Sclerotinia sclerotiorum*) and the like.

Recently, the emergence of plant pathogens resistant to fungicides has been often noticed in fields, becoming a serious practical problem in crop protection with fungicide application. 1-Cycloalkylcarbonylimidazolidinediones [I] have been found to exhibit a strong fungitoxicity towards those fungicide-resistant pathogens. For example, they have the same fungitoxic activity on the Polyoxin-resistant strain of *Alternaria kikuchiana* and the thiophanate-methyl[1,2-bis(3-methoxycarbonyl-2-thioureido)benzene]-resistant strain of *Botrytis cinerea* as on the respective wild

strains (susceptible strains). It can be therefore expected that 1-cycloalkyl-carbonylimidazolidinediones [I] exert prominent controlling effectiveness on plant diseases in the fields where fungicide-resistant pathogens have already emerged.

Furthermore, 1-cycloalkylcarbonylimidazolidinediones [I] are systemic in plants. The compounds can penetrate from leaf surfaces into leaf tissues, and can be absorbed by roots and translocated to leaves. Due to this property, the compounds can effectively suppress propagation of the pathogens invading leaf tissues, in addition to protecting plants from infection.

The 1-cycloalkylcarbonylimidazolidinediones [I] of the present invention are structurally similar to some of the compounds disclosed in U.S. patent 3,716,552, but their effectiveness in controlling the said diseases are superior to those of the latter compounds, and the 1-cycloalkylcarbonylimidazolidinediones [I] are effective at lower dosages. This indicates that substitution with a cycloalkylcarbonyl group at the 1-position of 3-(3,5-dihalophenyl)imidazolidine-2,4-dione derivatives resulted in marked increase in their fungitoxic activities. The present inventors are the first to point out this unexpected increase of activity.

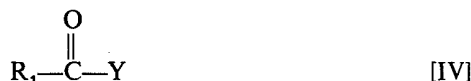
The 1-cycloalkylcarbonylimidazolidinediones [I] can be prepared by reacting the corresponding 1-unsubstituted compound of the formula:



wherein X, R₂ and R₃ are each as defined above, with a cycloalkanecarboxylic acid of the formula:



wherein R₁ is as defined above, or a reactive derivative thereof. Usually, the reaction is effected between the said 1-unsubstituted compound [II] and either a cycloalkanecarbonyl halide of the formula:



wherein R₁ is as defined above and Y is a halogen atom, or a cycloalkylcarboxylic anhydride of the formula:



wherein R₁ is as defined above.

Examples of typical procedures for carrying out the preparation so as to obtain the desired 1-cycloalkylcarbonylimidazolidinediones [I] in a good yield are as follows:

Procedure A

The starting 1-unsubstituted compound [II] is reacted with an equivalent or molar excess of the cycloalkylcarbonyl halide [IV] at room temperature (0–35°C) in the presence of a suitable solvent (e.g. tetrahydrofuran, methyl isobutyl ketone, benzene, toluene, xylene, chlorobenzene, chloroform, carbon tetrachloride, or nitrobenzene) or in the absence of a solvent and, if necessary, in the presence of a dehydrohalogenating agent (e.g. triethylamine, N-methylmorpholine, pyridine, dimethylaniline, diethylaniline) and/or with heating (up to reflux) to give the 1-cycloalkylcarbonylimidazolidinedione [I].

Procedure B

The starting 1-unsubstituted compound [II] is reacted with an equivalent amount of molar excess of the cycloalkylcarboxylic anhydride [V] at room temperature (0—35°C) in the presence of a suitable solvent (e.g. chloroform, carbon tetrachloride, benzene, toluene, xylene, ligroin) or in the absence of solvent, with heating if necessary to give the 1-cycloalkylcarbonylimidazolidinedione [I].

The 1-cycloalkylcarbonylimidazolidinedione [I] thus produced may be purified, if necessary, by a *per se* conventional procedure such as recrystallization.

The starting 1-unsubstituted compound [II] is obtainable, for instance, by the process described in U.S. patent 3,668,217.

For use as fungicides, the 1-cycloalkylcarbonylimidazolidinediones [I] may be used alone or, for easier application, as fungicidal compositions together with carriers or diluents, for example solid carriers or diluents such as talc or a clay or liquid carriers or diluents such as organic solvent. Such fungicidal compositions can be formulated into any of the ordinarily adopted forms such as, for example, dusts, wettable powders, oil sprays, aerosols, tablets, emulsifiable concentrates and granules.

The foregoing preparations generally contain from 0.1 to 95.0 % by weight, preferably from 0.2 to 90.0 % by weight of the active ingredient (including other ingredients mixed). Such preparations are generally applied at a rate of from 10 g to 1000 g/10 are, and the concentration of the active ingredients in preparations applied is preferably from 0.001 to 0.1 % by weight. These amounts and concentrations will of course depend upon the type of preparation, time and method of application, application site, and the diseases and crops being treated. Accordingly the foregoing ranges are intended only as a general guide.

The 1-cycloalkylcarbonylimidazolidinediones [I] may be used in admixture with other fungicides such as, for example, N-(3,5-dichlorophenyl)-1,2-dimethylcyclopropane-1,2-dicarboximide, S-n-butyl-S'-p-tert-butylbenzyl-N-3-pyridylidithiocarbonylimidate, O,O-dimethyl-O-2,6-dichloro-4-methylphenylphosphorothioate, methyl N-benzimidazol-2-yl-N-(butylcarbamoyl)carbamate, N-trichloromethylthio-4-cyclohexene-1,2-dicarboximide, cis-N-(1,1,2,2-tetrachloroethylthio)-4-cyclohexene-1,2-dicarboximide, Polyoxin, Streptomycin, zinc ethylene-bis(dithiocarbamate), zinc dimethylthiocarbamate, manganese ethylene-bis(dithiocarbamate), bis(dimethylthiocarbamoyl)disulfide, tetrachloroisophthalonitrile, 8-hydroxyquinoline, dodecylguanidine acetate, 5,6-dihydro-2-methyl-1,4-oxathiine-3-carboxanilide, N'-dichlorofluoromethylthio-N,N-dimethyl-N'-phenylsulfamide, 1-(4-chlorophenoxy)-3,3-dimethyl-1-(1,2,4-triazol-1-yl)-2-butanone, or 1,2-bis(3-methoxycarbonyl-2-thioureido)benzene or with insecticides such as, for example, O,O-dimethyl-O-(4-nitro-m-tolyl)phosphorothioate, O-p-cyanophenyl-O,O-dimethylphosphorothioate, O-p-cyanophenyl-O-ethylphenylphosphonothioate, O,O-dimethyl-S-(N-methylcarbamoylmethyl)phosphorodithioate, 2-methoxy-4H-1,3,2-benzodioxaphosphorine-2-sulfide, O,O-dimethyl-S'-(1-ethoxycarbonyl-1-phenylmethyl)phosphorodithioate, α -cyano-3-phenoxybenzyl-2-(4-chlorophenyl)isovalerate, 3-phenoxybenzyl-2,2-dimethyl-3-(2,2-dichlorovinyl)cyclopropanecarboxylate, or 3-phenoxybenzyl chrysanthemate. In none of these cases is the efficiency of any individual compound decreased by such admixture. Accordingly, simultaneous control of two or more pests and injurious insects is possible. They may also be used in admixture with such agricultural chemicals as nematocides and acaricides and with fertilizers.

Practical and presently preferred embodiments of the present invention are illustratively shown in the following examples, wherein parts and % are by weight.

Example 1.

Preparation of the 1-cycloalkylcarbonylimidazolidinediones [I]:—

Procedure A

0.05 mole of a 3-(3,5-dihalophenyl)imidazolidine-2,4-dione derivative of the formula [II] and 0.06 mole of triethylamine are dissolved in 150 ml of toluene, and 0.06 mole of an acid chloride of the formula [IV] is added thereto dropwise slowly at room temperature with stirring. After the addition is finished, the mixture is heated under reflux for 7 hours. The reaction mixture is filtered, and the filtrate is concentrated under reduced pressure. The residue obtained is well washed with water, and the water-insoluble portion is collected by filtration and dried to obtain the objective 1-cycloalkylcarbonyl-3-(3,5-dihalophenyl)imidazolidine-2,4-dione

derivative of the formula [I] in a high yield. The crude product is recrystallized from ethanol to obtain a pure product.

Procedure B

5 0.05 mole of a 3-(3,5-dihalophenyl)imidazolidine-2,4-dione derivative of the
formula [II] and 0.15 mole of an acid anhydride of the formula [V] are 5
dissolved in 50 ml of toluene. The mixture is heated under reflux for
15 hours. After the reaction is finished, the reaction solution is cooled to room
temperature and poured into water saturated with sodium hydrogen carbonate,
followed by stirring for some time. The solid material obtained is collected by
10 filtration, washed with water several times and dried to obtain the objective 1- 10
cycloalkylcarbonyl-3-(3,5-dihalophenyl)imidazolidine-2,4-dione derivative of the
formula [I] in a high yield. The crude product is recrystallized from ethanol to
obtain a pure product.

15 According to either one of the above procedures, the 1-cycloalkylcarbonyl-
imidazolidinediones [I] as shown in Table I are prepared. 15

Table 1

Procedure	Starting materials		1-Cycloalkylcarbonyl-3-(3,5-dihalophenyl)imidazolidine-2,4-dione compound [I]							
	3-(3,5-Dihalo-phenyl)imidazolidine-2,4-dione compound [II]	Acid chloride [IV] or acid anhydride [V]	Compound No.	Chemical structure	Melting point (°C)	Yield (%)	Elementary analysis (%)			
							C	H	N	Halogen
A			1		154.5- 156.0	90	49.86 (49.68)	3.22 (3.13)	8.95 (8.82)	(Cl) 22.64 (22.61)
A			2		101.5- 104.5	72	51.40 (51.23)	3.70 (3.52)	8.56 (8.39)	(Cl) 21.67 (21.86)
A			3		102.0- 103.0	83	52.80 (52.65)	4.14 (4.11)	8.21 (8.38)	(Cl) 20.78 (20.91)
A			4		159.5- 161.0	87	54.10 (53.97)	4.54 (4.34)	7.89 (7.80)	(Cl) 19.96 (19.90)

(Continued)

Procedure	Starting materials		Chemical structure	Melting point (°C)	Yield (%)	Elementary analysis (%)			
	3-(3,5-Dihalo-phenyl)imidazole-2,4-dione compound [II]	Acid chloride [IV] or acid anhydride [V]				Comp. No.	C	H	N
A				128.5- 131.0	89	55.30 (55.51)	4.91 (5.00)	7.59 (7.43)	(Cl) 19.20 (18.86)
A				122.0- 125.0	78	57.73 (57.96)	5.10 (4.98)	7.09 (7.21)	(Cl) 17.94 (18.00)
A				119.0- 144.5	81	46.82 (46.70)	3.24 (3.20)	6.42 (6.33)	(Cl) 32.52 (32.31)
A				121- 122	85	51.40 (51.29)	3.70 (3.71)	8.56 (8.42)	(Cl) 21.67 (21.62)

(Continued)

Procedure	Starting materials		1-Cycloalkylcarbonyl-3-(3,5-dihalophenyl)imidazolide-2,4-dione compound [I]							
	3-(3,5-Dihalo-phenyl)imidazo-2,4-dione compound [II]	Acid chloride [IV] or acid anhydride [V]	Compound No.	Chemical structure	Melting point (°C)	Yield (%)	Elementary analysis (%)			
							C	H	N	Halogen
B			9		110.5-113.5	83	55.30 (55.49)	4.91 (5.08)	7.59 (7.36)	(Cl) 19.20 (19.31)
A			10		96.5-97.5	92	52.80 (52.64)	4.14 (4.08)	8.21 (8.10)	(Cl) 20.78 (20.72)
A			11		140.5-143.5	77	38.84 (38.61)	2.51 (2.35)	6.97 (7.11)	(Cl) 39.75 (39.88)

Note: In the elemental analysis, the values as calculated are unparenthesized, and the values as found are parenthesized.

Example 2.

Formulation of compositions:—

a) *Dust*

2 Parts of the compound (8) and 98 parts of clay were thoroughly pulverized and mixed together to obtain a dust containing 2% of the active ingredient. The dust was suitable for direct application.

b) *Dust*

3 Parts of the compound (4) and 97 parts of talc were thoroughly pulverized and mixed together to obtain a dust containing 3% of the active ingredient. The dust was suitable for direct application.

c) *Wettable powder*

50 Parts of the compound (1), 5 parts of a wetting agent of the alkylbenzene-sulfonate type and 45 parts of diatomaceous earth were thoroughly pulverized and mixed together to obtain a wettable powder containing 50% of the active ingredient. For application, the wettable powder was diluted with water, and the resulting solution was sprayed.

d) *Emulsifiable concentrate*

10 Parts of the compound (2), 40 parts of dimethyl sulfoxide, 40 parts of xylene and 10 parts of an emulsifier of the polyoxyethylene dodecylphenol ether type were mixed together to obtain an emulsifiable concentrate containing 10% of the active ingredient. For application, the emulsifiable concentrate was diluted with water, and the resulting emulsion was sprayed.

e) *Granules*

5 Parts of the compound (10), 93.5 parts of clay and 1.5 parts of a binder of the polyvinyl alcohol type were thoroughly pulverized and mixed together, kneaded with water and then granulated and dried to obtain granules containing 5% of the active ingredient.

The following examples show some typical test data supporting the excellent activity of the 1-cycloalkylcarbonylimidazolidinediones [I]. In these examples, the compound numbers correspond to those in Table 1.

Example 3.

Fungicidal activity test on fungicide-resistant pathogens:—

Ten milliliters of a potato sucrose agar medium was turned into a solution by heating, and each of the emulsifiable concentrates containing the test compounds was added to the solution and well mixed. The mixture was poured into a glass Petri dish of 9 cm in diameter to make an agar plate. After the agar was hardened, mycelial discs (5 mm in diameter) of a fungicide-resistant strain of gray mold fungus (*Botrytis cinerea*) resistant to 1,2-bis(3-methoxycarbonyl-2-thioureido)benzene which was isolated from grapes and a wild (susceptible) strain of *Botrytis cinerea* were each placed at the center of the agar plate and incubated at 20°C for 3 days. After the incubation, the diameter of the grown colony was measured, and the percentage of growth inhibition was calculated from the following equation in comparison with an untreated plot:

$$\text{Percentage of growth inhibition (\%)} = 100 - \frac{\text{Diameter of colony in treated plot}}{\text{Diameter of colony in untreated plot}} \times 100$$

As the result, the compounds of the present invention showed a strong antimicrobial activity on a fungicide-resistant strain of *Botrytis cinerea* as well as on a susceptible wild strain of *Botrytis cinerea*, as shown in Table 2.

TABLE 2

Compound No.	Fungicidal effect on a wild (susceptible) strain of <i>Botrytis cinerea</i>		Fungicidal effect on a fungicide-resistant strain of <i>Botrytis cinerea</i> * 1)	
	Concentration of pesticide in medium (concentration of active ingredient) (ppm)	Percentage of growth inhibition (%)	Concentration of pesticide in medium (concentration of active ingredient) (ppm)	Percentage of growth inhibition (%)
1	10	94	10	94
	1	94	1	94
2	10	94	10	94
	1	92	1	90
4	10	94	10	94
	1	90	1	92
5	10	94	10	94
	1	90	1	88
6	10	94	10	94
	1	85	1	83
7	10	94	10	94
	1	78	1	80
8	10	94	10	94
	1	92	1	90
9	10	94	10	94
	1	88	1	90
10	10	94	10	94
	1	89	1	85
11	10	94	10	94
	1	94	1	94
Thiophanate-*2) methyl	10	94	100	32
	1	70	10	10
			1	0

Footnotes to Table 2:

*) Strain resistant to 1,2 - bis(3 - methoxycarbonyl - 2 - thioureido)benzene (isolated from grapes)

5 *2) Commercially available fungicide, 1-2-bis(3-methoxycarbonyl-2-thio-
ureido)benzene. 5

Example 4.

Protective activity test on black spot of pear (*Alternaria kikuchiana*):—

10 A 3-year old pear tree (var.: 20-Seiki) cultivated in a flowerpot of 30 cm in
diameter was used as a test plant. Each of the emulsifiable concentrates containing
the test compounds was diluted with water to a required concentration. When each
15 plant shot out three to four young branches having 10 to 20 leaves thereon, the
prepared aqueous solution was sprayed thereon in a rate of 30 ml/seedling. The
plant was cultivated in a green-house for 7 days after spraying. The whole body of
the plant was then inoculated by spraying the spore suspension of *Alternaria*
kikuchiana cultured in a vegetable juice agar medium for 10 days. The plant was
then placed in a humid chamber for 24 hours after inoculation and then in a green-
house for 2 days. The disease severity was examined as follows using a disease index
(0, 1, 2, 3, 4, 5): 15

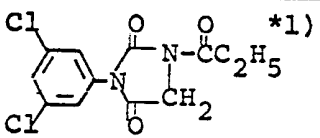
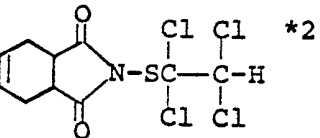
	Disease severity	Disease index	
20	No infected area	0	20
	Infected leaf area of less than 10%	1	
	Infected leaf area of 10 to less than 20%	2	
	Infected leaf area of 20 to less than 40%	3	
	Infected leaf area of 40 to less than 60%	4	
25	Infected leaf area of 60% or more	5	25

The disease severity was calculated according to the following equation:

$$\text{Disease severity (\%)} = \frac{\Sigma (\text{Disease index}) \times (\text{Number of leaves})}{5 \times (\text{Total number of leaves examined})} \times 100$$

30 As a result, the compounds of the present invention were much superior in the
protective activity to the control compounds tested at the same time, as shown in
Table 3. 30

Table 3

Compound No.	Concentration of active ingredient (ppm)	Disease severity (%)
1	1000	0.0
	100	1.0
2	1000	0.5
	100	3.2
3	1000	0.8
	100	3.0
4	1000	1.0
	100	2.8
5	1000	1.0
	100	4.5
6	1000	1.6
	100	5.0
7	1000	0.5
	100	2.5
8	1000	0.8
	100	4.0
9	1000	2.0
	100	5.5
10	1000	2.5
	100	6.8
11	1000	0.0
	100	2.5
	1000 100	4.5 58.0
	1000 100	5.0 63.2
No treatment	-	72.5

Note: *1) Compound disclosed in U.S. patent 3,716,552.

*2) Commercially available fungicide, generic name "Captafol".

Example 4.

Protective activity test on stem rot of kidney bean (*Sclerotinia sclerotiorum*):—

Kidney beans (var.: Taishō-kintoki) were grown up to the two-leaf stage in a flower pot of 15 cm in diameter. Each of the emulsifiable concentrates containing the test compounds was diluted with water and sprayed on the kidney beans in a

rate of 10 cm/pot. After the kidney beans were air-dried for 4 hours, they were inoculated with the mycelial disc (5 mm in diameter) of *Sclerotinia sclerotiorum*. After the inoculation, the test plants were infected by placing them under a highly humid, dark condition at 20°C for 4 days. The disease severity was calculated on the basis of the following standard for evaluating infection:

5

5

Disease index	Evaluation of infection		
0	No infection		
1	Slight infection around the inoculum		
10	2	Infected area of about 1/6 of the inoculated leaf	10
	3	Infected area of about 2/5 of the inoculated leaf	
	4	Infected area of about 3/5 of the inoculated leaf	
	5	Infected area of 3/5 or more of the inoculated leaf	

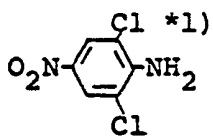
$$\text{Disease severity (\%)} = \frac{\Sigma (\text{Disease index}) \times (\text{Number of leaves})}{5 \times (\text{Total number of leaves examined})} \times 100$$

15

As the result, the compounds of the present invention were superior in the protective activity to the control compound tested at the same time, as shown in Table 4.

15

Table 4

Compound No.	Concentration of active ingredient (ppm)	Disease severity (%)
1	100	0.0
2	100	0.0
4	100	2.5
5	100	0.0
6	100	0.0
7	100	2.5
8	100	0.0
9	100	0.0
10	100	2.5
11	100	0.0
 Cl *1)	100	47.5
No treatment	-	100.0

Note: *1) Commercially available fungicide, generic name "Dcna".

Example 5.

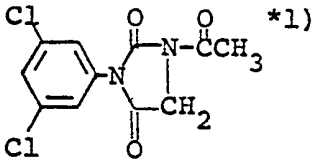
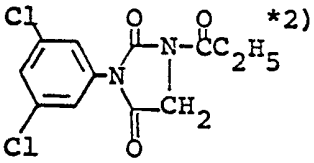
Protective activity test on blue mold of orange (*Penicillium italicum*):—

5 Orange fruits (var.: Unshū) were well washed with water and air-dried. Each of
the emulsifiable concentrates containing the test compounds was diluted with
water to a required concentration and the air-dried orange fruits were dipped in the
aqueous solution for 1 minute. After air-drying, the surface of the fruit was
inoculated by spraying a spore suspension of *Penicillium italicum* cultured in a
10 potato agar medium for 5 days. After the fruits were placed in a humid chamber for
7 days after inoculation, the disease severity was examined using a disease index (0,
1, 2, 3, 4, 5). The results were shown in the mean value of disease index. 10

Disease severity	Disease index	
No infected area	0	
Infected surface area of less than 20%	1	
Infected surface area of 20 to less than 40%	2	
15 Infected surface area of 40 to less than 60%	3	15
Infected surface area of 60 to less than 80%	4	
Infected surface area of 80% or more	5	

20 As the result, the compounds of the present invention were superior in the
protective activity to the control compounds tested at the same time, as shown in
Table 5. 20

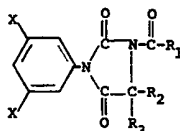
Table 5

Compound No.	Concentration of active ingredient (ppm)	Mean value of disease index
1	100	0.0
2	100	0.0
4	100	0.0
5	100	0.8
6	100	0.5
7	100	1.3
8	100	0.0
9	100	0.0
10	100	0.2
11	100	0.0
 Cl *1) Cl	200	2.6
 Cl *2) Cl	200	3.1
No treatment	-	5.0

Note: *1) & *2) Compounds disclosed in U.S. patent 3,716,552.

WHAT WE CLAIM IS:—

1. A compound of the formula:



[r]

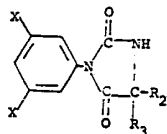
- 5 wherein each X is independently a chlorine or bromine atom, R₁ is a C₃—C₆ cycloalkyl, tetramethylcyclopropyl, 2,2-dimethyl-3-isobutenylcyclopropyl or 2,2-dimethyl-3-dichlorovinylcyclopropyl group and R₂ and R₃ are each independently a hydrogen atom or a methyl group. 5
- 10 2. A compound as claimed in claim 1, wherein X is a chlorine atom, R₁ is a C₃—C₆ cycloalkyl group and R₂ and R₃ are each a hydrogen atom. 10
3. A compound as claimed in claim 2, wherein R₁ is a cyclopropyl group.
4. A compound as claimed in claim 2, wherein R₁ is a cyclobutyl group.
5. A compound as claimed in claim 2, wherein R₁ is a cyclopentyl group.

6. A compound as claimed in claim 2, wherein R_1 is a cyclohexyl group.

7. A compound as claimed in claim 1, wherein R_1 is a 2,2,3,3-tetramethyl-cyclopropyl group, or a 2,2-dimethyl-3'-(2',2-dichlorovinyl)cyclopropyl group.

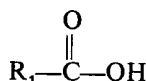
8. A compound as claimed in claim 1 and substantially as hereinbefore described.

9. A process for preparing a compound as claimed in claim 1, which comprises reacting the 1-unsubstituted compound of the formula:



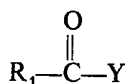
[III]

with a cycloalkanecarboxylic acid of the formula:

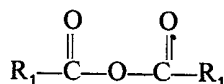


or a reactive derivative thereof wherein R_1 , R_2 , R_3 and X are each as defined in claim 1.

10. A process as claimed in claim 9, wherein the reaction is carried out using a reactive derivative of the cycloalkanecarboxylic acid, which is an acid halide of the formula:



or an acid anhydride of the formula:



wherein R_1 is as defined in claim 1 and Y is a halogen atom.

11. A process for preparing a compound as claimed in claim 1 and substantially as hereinbefore described in Example 1.

12. A compound as claimed in claim 1, when prepared by a method as claimed in any one of claims 9 to 11.

13. A fungicidal composition which comprises a compound as claimed in any one of claims 1 to 8, or claim 12, together with an inert diluent or carrier.

14. A composition as claimed in claim 13, in the form of a dust, a wettable powder, an emulsifiable concentrate, or granules or tablets.

15. A composition as claimed in claim 14, and substantially as hereinbefore described in Example 2.

16. A method of controlling or preventing the growth of a fungus, which comprises applying to the fungus, or to a substrate on which the growth of fungus is to be prevented, a compound as claimed in any one of claims 1 to 8 or claim 12, or a composition as claimed in any one of claims 13 to 15.

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