

PATENT SPECIFICATION

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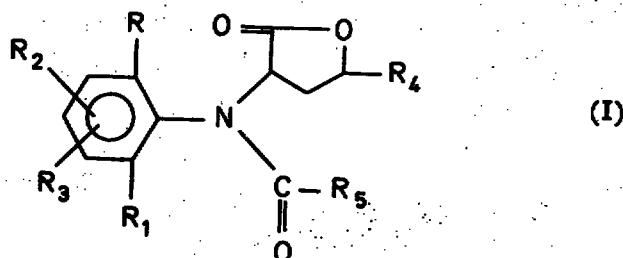
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(54) ACYLAMINO-TETRAHYDRO-2-FURANONE DERIVATIVES AND THEIR USE AS FUNGICIDES

(71) We, CIBA-GEIGY AG, a Swiss body corporate, of Basle, Switzerland, 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 acylaminotetrahydro-2-furanone derivatives and their use as fungicides.

According to the invention, there are provided compounds of the formula I



wherein

- R represents C_1-C_4 -alkyl, C_1-C_4 -alkoxy or halogen,
 R_1 represents C_1-C_4 -alkyl, C_1-C_4 -alkoxy or halogen,
 R_2 represents hydrogen, C_1-C_3 -alkyl, C_1-C_4 -alkoxy or halogen,
 R_3 represents hydrogen or methyl, with the total number of carbon atoms of the substituents R_1 , R_2 and R_3 in the phenyl ring not exceeding the number 8,
 R_4 represents hydrogen or methyl, whilst
 R_5 represents one of the following groups:
 an alkylthioalkyl or alkoxyalkyl group having 2 to 5 carbon atoms or an alkoxyalkoxy-methyl group having a maximum of 6 carbon atoms, a 2-furanyl or 2-tetrahydro-furanyl group optionally substituted by halogen, a 1,2,4-triazolylmethyl group, a 1,2-pyrazolylmethyl group, or a group $-CH_2-OR_6$ in which R_6 represents a 5- or 6-membered heterocycle having oxygen as the hetero atom.

By alkyl or as alkyl moiety of an alkylthio or alkoxy group are meant, depending on the given number of carbon atoms, the following groups: methyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec.-butyl or tert.-butyl. Alkoxyalkoxy groups are, in particular, methoxymethoxy, ethoxymethoxy, 1-methoxyethoxy and 1-ethoxyethoxy.

By halogen is meant fluorine, chlorine, bromine or iodine. R_6 preferably represents a tetrahydrofuranyl or pyranyl group, particularly one bound in the 2-position.

In U.S. Patent Specification No. 3,933,860 are described 3-(N-acyl-N-arylamino)-lactones and -lactams as fungicides which are derived from the structure of benzoyl-, alkanoyl- or haloalkanoyl-(particularly haloacetyl-)anilides. Anilides of this type are described in the literature in great numbers as herbicides, e.g. U.S. Patent Specifica-

tions Nos. 3,403,994, 3,442,945, 3,547,620, 3,637,847, 3,598,859 or 3,946,045.

It is therefore not surprising that the compounds given in U.S. Patent Specification No. 3,933,860 give rise in part, when applied in the amounts required in practice, to undesirable phytotoxicity in the plants to be protected from fungus infestation.

It has now been found that, surprisingly, compounds having the structure of the formula I have for practical requirements a very favourable microbicidal spectrum for the protection of cultivated plants, without affecting them disadvantageously by causing undesirable secondary effects. Within the scope of the present invention, cultivated plants are, for example, grain, maize, rice, vegetables, sugar beet, soya beans, peanuts, fruit trees and ornamental plants, especially however grape vines, hops, cucurbitaceae (cucumbers, pumpkins and melons), and solanaceae such as potatoes, tobacco and tomatoes, as well as bananas, cocoa plants and natural rubber plants. According to the invention there is provided a method of combating phytopathogenic fungi which method comprises applying to a locus infested with or liable to be infested with the fungi a compound of the formula I.

Fungi occurring on plants or on parts of plants (fruit, blossom, foliage, stalks, tubers or roots) in the above cultivated crops, or in related crops, can be inhibited or destroyed by application of the active substances of the formula I: and also parts of plants subsequently growing remain protected from such fungi. The active substances are effective against the phytopathogenic fungi belonging to the following classes: *Ascomycetes* (e.g. *Erysiphaceae*); *Basidiomycetes*, such as in particular rust fungi; *Fungi imperfecti* (e.g. *Moniliales*); but especially against *Oomycetes* belonging to the *Phycomycetes* class, such as *Phytophthora*, *Peronospora*, *Pseudoperonospora*, *Pythium* or *Plasmopara*. Furthermore, the compounds of the formula I have a systemic action. They can be used also as dressing agents for treating seeds (fruits, tubers and grain) and plant cuttings to preserve them from fungus infections; and they can be used also against phytopathogenic fungi occurring in the soil.

Formula I includes compounds wherein R_1 represents an alkylthioalkyl or alkoxyalkyl group having 2 to 5 carbon atoms, a 2-furanyl or 2-tetrahydrofuranyl group optionally substituted by halogen, or a 1,2,4-triazolylmethyl group or compounds wherein R_1 represents a 1,2-pyrazolylmethyl group.

Preferred compounds of the formula I as plant fungicides are those wherein R represents methyl; R_1 represents methyl, ethyl, chlorine or bromine; R_2 represents hydrogen, halogen or methyl, and R_3 represents hydrogen or methyl, while R_4 and R_5 have the meanings given for the formula I. These are to be referred to as compound group Ia.

An important subgroup of the compound group Ia is formed by those compounds wherein R_1 represents an alkylthioalkyl or alkoxyalkyl group having 2 to 5 carbon atoms, a 2-furanyl or 2-tetrahydrofuranyl group optionally substituted by halogen, or a 1,2,4-triazolylmethyl group. This subgroup is to be called compound group Ib.

Compounds to be emphasised among these compounds of the group Ib by virtue of their effectiveness are those wherein R_1 represents methoxymethyl or ethoxymethyl. These are to be called compound group Ic.

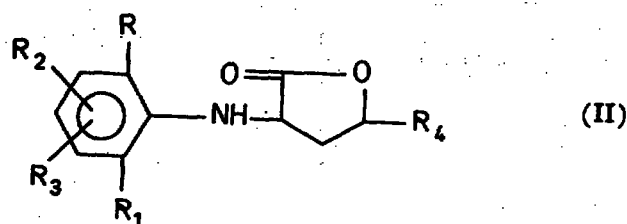
Particularly preferred active substances within the compound group Ia, on account of their advantageous action, are those wherein R_1 represents the 1,2,4-triazol-1-yl-methyl group. These are to be designated as compound group Id.

A further important subgroup of the compound group Ia is formed by those compounds wherein R_1 represents a 2-furanyl or 2-tetrahydrofuranyl group optionally substituted by halogen. This subgroup is to be termed compound group Ie.

Another important subgroup of the compound group Ia is made up of those compounds wherein R_1 represents a 1,2-pyrazolylmethyl group. This subgroup is to be referred to as compound group If.

Yet another important subgroup of the compound group Ia is composed of those compounds wherein R_1 represents an alkoxyalkoxymethyl group having a maximum of 5 carbon atoms. This subgroup is to be identified as compound group Ig.

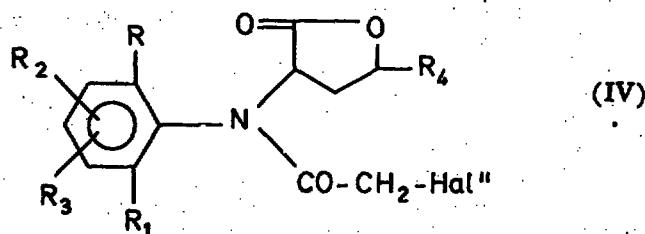
The compounds of the formula I may be prepared (Process A) by reacting a compound of the formula II



with a compound of the formula III

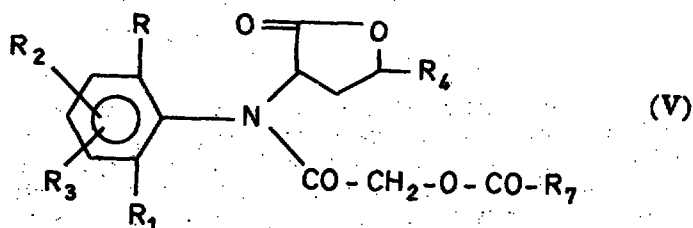


Compounds of the formula I wherein R_5 represents a 1,2,4-triazolylmethyl, 1,2-pyrazolylmethyl or alkylthio group may be prepared (Process B) by initial monohaloacetylation of a compound of the formula II to give a compound of the formula IV



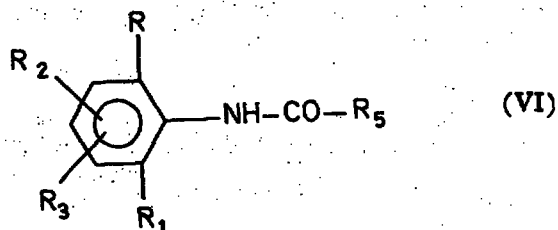
and reacting the compound of formula IV with 1,2,4-triazole, 1,2-pyrazole or an alkylmercaptan, or with the metal salt of one of these reactants, preferably with the alkali metal salt or alkaline-earth metal salt.

Compounds of the formula I wherein R_5 represents an alkoxyalkoxymethyl group or a group $-\text{CH}_2-\text{OR}_6$ in which R_6 represents a saturated 5- or 6-membered heterocycle having oxygen as the heteroatom may be prepared (Process C) by initial monohaloacetylation of a compound of the formula II to give a compound of the formula IV, and exchanging the reactive substituent Hal'' for an alkanecarboxylic acid containing less than 7 carbon atoms in the alkane moiety, or for one of the alkali metal salts or alkaline-earth metal salts thereof, to obtain an acyloxyacetylanilide of the formula V

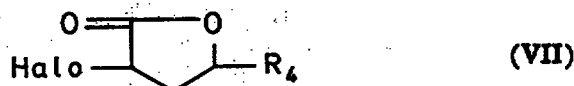


from which there is then obtained by mild alcoholysis with e.g. an alkali alcoholate of a lower alcohol, such as methanol or ethanol, and acidification of the corresponding hydroxyacetylanilide, which is activated with an alkali metal, an alkali metal hydride or p-toluenesulphonic acid, and reacted with an alkoxyalkyl halide, 2,3-dihydrofuran or 3,4-dihydro-2H-pyran.

Compounds of the formula I may also be prepared (Process D) by reacting an already acylated aniline of the formula VI



in the presence of a proton acceptor such as butyl-lithium or sodium hydride, with a 2-halo-4-butyrolactone of the formula VII



In the formulae II, III, IV, V, VI and VII, the symbols R to R₄ have the meanings given under the formula I, and Halo, Hal' and Hal'' represent halogen, preferably chlorine or bromine. R₄ is an alkyl group having less than 7 carbon atoms.

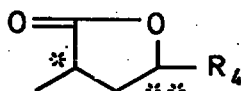
The reactions can be performed in the presence or absence of solvents or diluents which are inert to the reactants. The following are for example suitable: aliphatic or aromatic hydrocarbons such as benzene, toluene, xylenes or petroleum ether; halogenated hydrocarbons such as chlorobenzene, methylene chloride, ethylene chloride or chloroform; ethers and ethereal compounds such as dialkyl ether, dioxane or tetrahydrofuran; nitriles such as acetonitrile; N,N-dialkylated amides such as dimethylformamide; dimethylsulphoxide, ketones such as methyl ethyl ketone, and mixtures of such solvents with each other.

For acylation according to process A or C, or for monohaloacetylation according to process B, it is possible to use the corresponding carboxylic acids themselves and also the esters thereof, advantageously however the acid anhydrides or, as given in the case of formula III, the acid halides, preferably the acid chlorides or acid bromides.

The reaction temperatures are generally between 0° and 180°C, preferably between 20° and 120°. The use of acid-binding agents or condensation agents is advantageous in some cases. Suitable as such are tertiary amines such as trialkylamines (e.g. triethylamine), pyridine and pyridine bases, or inorganic bases, such as the oxides and hydroxides, hydrogen carbonates and carbonates of alkali metals and alkaline-earth metals, and also sodium acetate.

The production process A starting with compounds of the formula II, and likewise the acylation stage leading to compounds of the formula IV, can be performed also without acid-binding agents; in some cases, however, the passing through of nitrogen to expel the formed hydrogen halide is advisable. In other cases, the addition of dimethylformamide as a reaction catalyst is very advantageous.

Details regarding the production of the intermediates of the formula II are known. The compounds of the formula I possess in the lactone radical



a (*) center of asymmetry and, where R₄=CH₃, a second (**) center of asymmetry, and can be resolved in the customary manner (e.g. fractional crystallisation or chromatographical separation) into optical antipodes. In this respect, the different configurations have a varying strong microbicidal action. Centers of asymmetry can occur also in the case of some substituents R₅. Provided that no specific synthesis for the isolation of the pure isomers of the formula I or of the employed butyrolactone is performed, a product is usually obtained as an isomeric mixture.

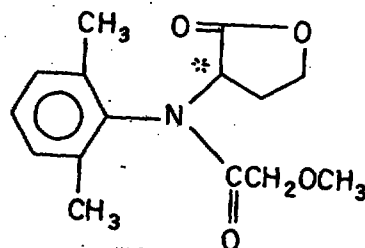
The following compounds belong to a class of particularly effective plant fungicides:

- 3-[N-(methoxyacetyl)-N-(2,6-dimethylphenyl)]-aminotetrahydro-2-furanone,
- 3-[N-(ethoxyacetyl)-N-(2,6-dimethylphenyl)]-aminotetrahydro-2-furanone,
- 3-[N-(methoxyacetyl)-N-(2,6-dimethyl-3-chlorophenyl)]-aminotetrahydro-2-furanone,
- 3-[N-(ethoxyacetyl)-N-(2,6-dimethyl-3-chlorophenyl)]-aminotetrahydro-2-furanone,
- 3-[N-(methoxyacetyl)-N-(2,6-dimethylphenyl)]-amino-5-methyltetrahydro-2-furanone,
- 2,6-dimethyl-N-(3-tetrahydrofuran-2-one)-2-tetrahydrofuroyl-anilide,
- 2,3,6-trimethyl-N-(3-tetrahydrofuran-2-one)-2-tetrahydrofuroyl-anilide,
- 2,6-dimethyl-4-chloro-N-(3-tetrahydrofuran-2-one)-2-tetrahydrofuroyl-anilide,
- 3-[N-(methoxyacetyl)-N-(2,3,5,6-tetramethylphenyl)]-aminotetrahydro-2-furanone, and

2,3,5,6-tetramethyl-N-(3-tetrahydrofuran-2-one)-2-furoyl-anide.
The following Examples serve further to illustrate the invention. The temperatures are in degrees Centigrade.

Example 1

Production of



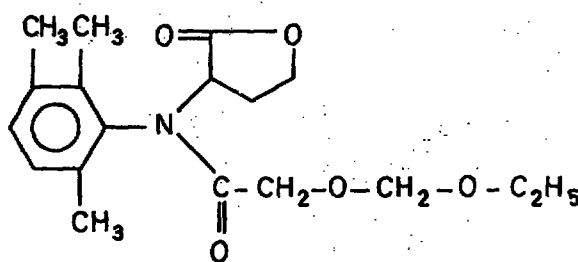
3-[N-(methoxyacetyl)-N-(2,6-dimethylphenyl)]-amino-tetrahydro-2-furanone (compound No. 2).

a) 165 g of 2-bromo-4-butyrolactone is slowly added to 121 g of 2,6-dimethylaniline and 106 g of sodium carbonate in 750 ml of dimethylformamide, and then stirred for 30 hours at 160°. After cooling, the unreacted aniline and the solvent are distilled off in vacuo; the residue is added to water and extracted three times with methylene chloride. The extracts are washed with water, dried over sodium sulphate, and then freed from solvent. After recrystallisation from petroleum ether/diethyl ether (1:10), the residue melts at 79—83°.

b) 11.9 g of methoxyacetyl chloride in 20 ml of toluene is added, with stirring, to 20.5 g of the intermediate product, obtained according to a), in 80 ml of abs. toluene, whereupon the temperature rises from 25° to 35°. The reaction mixture is refluxed for six hours and, after the addition of active charcoal, filtered through Hyflo; it is then concentrated by evaporation and recrystallised from ethyl acetate/petroleum ether, m.p. 124—127°.

Example 2

Production of



N-(3'-tetrahydrofuranone-2')-N-ethoxymethoxyacetyl-2,3,6-trimethylaniline (compound No. 14).

a) 0.5 g of sodium is dissolved in 10 ml of absolute methanol. There is then added dropwise, with stirring, a solution of 21.4 g of 3-[N-acetoxyacetyl]-N-(2,3,6-trimethylphenyl)]-aminotetrahydro-2-furanone in 60 ml of abs. methanol at 20—25°. Stirring is continued overnight at room temperature; the reaction mixture is subsequently concentrated in a rotary evaporator, and 500 ml of ice water is added. The suspension is then adjusted with 1% hydrochloric acid, with ice cooling, to pH 2 (acidified to Congo red); it is filtered with suction and then washed with water until neutral. The 3-[N-hydroxyacetyl]-N-(2,3,6-trimethylphenyl)]-aminotetrahydrofuranone - 2, occurring as a mixture of diastereoisomers, has a melting point of 151—155° after being recrystallised from isopropanol.

b) In a nitrogen atmosphere, 1.3 g of sodium hydride (as a 55% oil dispersion) is suspended in 50 ml of abs. tetrahydrofuran. There is then added dropwise at 0—10° 13.8 g of the intermediate product obtained under a), dissolved in 50 ml of abs. tetra-

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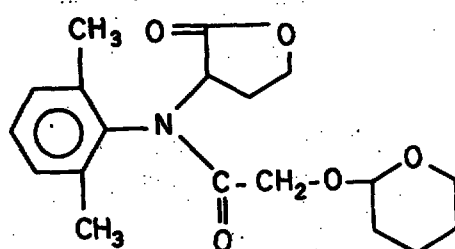
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hydrofuran. After completion of the generation of hydrogen (3 hours), a solution of 4.7 g of chloromethylethyl ether in 10 ml of tetrahydrofuran is added dropwise at 0°, and stirring is maintained for 20 hours at room temperature. Water is then carefully added, and extraction with ether is performed four times. The extracts are washed with water and dried over sodium sulphate. Concentration by evaporation yields a resin which, for the removal of paraffin oil, is digested four times with a small amount of petroleum ether; the elementary analysis of the resin obtained gives the following values:

found: C 64.1% H 7.7% N 4.0%
calculated: C 64.4% H 7.5% N 4.2%

Example 3

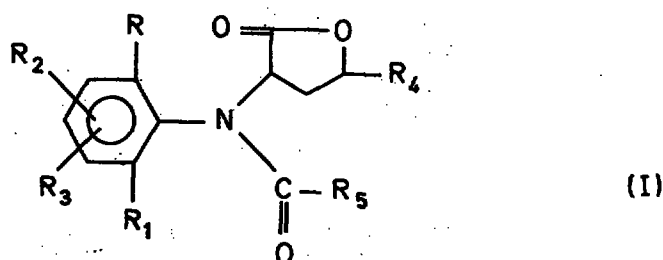


Production of

N-(3''-tetrahydrofuranone-2'')-N-2'-pyranyloxyacetyl-2,6-dimethylaniline (compound No. 43).

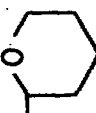
13.2 g of the 3-[N-hydroxyacetyl-N-(2,6-dimethyl-phenyl)]-amino-tetrahydrofuranone-2 obtained in a manner analogous to that described in Example 2a is suspended in 50 ml of abs. ethyl acetate, and a trace of p-toluenesulphonic acid is added. There is then added dropwise at room temperature in the course of 10 minutes, with stirring, a solution of 5.9 g of 3,4-dihydro-2-H-pyran in 10 ml of abs. ethyl acetate. The reaction mixture is then stirred for a further 1 1/4 hours at room temperature; it is subsequently cooled to 0° and 15 g of solid potassium carbonate is added. After stirring at 0° for 15 minutes, the mixture is filtered; thorough washing is then carried out with ethyl acetate, and the filtrate is concentrated by evaporation. On trituration with petroleum ether, the residue crystallises, and melts at 106—112° (diastereoisomeric mixture).

There are produced in an analogous manner the following compounds of the formula



(R=CH₃)
(R₁=6-position)

Compound No.	R ₁	R ₂	R ₃	R ₄	R ₅	Physical constants
1	CH ₃	H	H	CH ₃	-CH ₂ OCH ₃	m.p. 109-110°
2	CH ₃	H	H	H	-CH ₂ OCH ₃	m.p. 124-127°
5	C ₂ H ₅	H	H	H	-CH ₂ OCH ₃	
4	CH ₃	H	H	CH ₃	-CH ₂ OC ₂ H ₅	
5	CH ₃	H	H	H	-CH ₂ OC ₂ H ₅	oil
6	Cl	H	H	H	-CH ₂ OCH ₃	
7	CH ₃	3-CH ₃	H	H	-CH ₂ OCH(CH ₃)OC ₂ H ₅	resin
8	CH ₃	H	H	H	-CH ₂ OC ₃ H ₇ (n)	
9	CH ₃	3-CH ₃	H	H	-CH ₂ OCH ₃	resin
10	Cl	H	H	H	-CH ₂ OC ₂ H ₅	
11	Br	4-Cl	H	H	-CH ₂ OCH ₃	
12	CH ₃	3-CH ₃	H	H	-CH ₂ OC ₂ H ₅	resin
13	CH ₃	4-Cl	H	H	-CH ₂ CH ₂ OCH ₃	
14	CH ₃	3-CH ₃	H	H	-CH ₂ OCH ₂ OC ₂ H ₅	resin
15	CH ₃	3-CH ₃	H	CH ₃	-CH ₂ OCH ₃	
16	CH ₃	H	H	H	-CH ₂ OC ₃ H ₇ (i)	
17	CH ₃	H	H	H	-CH ₂ CH ₂ OCH ₃	b.p. 178-185°/0.1 Torr
18	CH ₃	3-CH ₃	5-CH ₃	H	-CH ₂ OC ₃ H ₇ (i)	

Compound No.	R ₁	R ₂	R ₃	R ₄	R ₅	Physical constants
19	CH ₃	3-CH ₃	5-CH ₃	H	-CH ₂ OCH ₃	m.p. 111-114°
20	CH ₃	4-Cl	H	H	-CH ₂ OC ₂ H ₅	m.p. 105-106°
21	CH ₃	3-CH ₃	H	CH ₃	-CH ₂ CH ₂ OCH ₃	oil
22	CH ₃	H	H	H	-CH ₂ OCH ₂ OCH ₃	
23	CH ₃	3-CH ₃	H	CH ₃	-CH ₂ OC ₂ H ₅	
24	CH ₃	4-Cl	H	H	-CH ₂ OCH ₃	m.p. 111-113°
25	CH ₃	H	H	H	-CH ₂ CH ₂ OC ₂ H ₅	b.p. 160-165°/0.2 Torr
26	CH ₃	3-CH ₃	5-CH ₃	H	-CH ₂ OC ₂ H ₅	m.p. 81-83°
27	CH ₃	3-CH ₃	H	H	-CH ₂ -S-CH ₃	m.p. 90-94°
28	CH ₃	H	H	H	-CH ₂ -S-CH ₃	m.p. 72-73°
29	CH ₃	3-CH ₃	H	H	-CH ₂ -O- 	m.p. 88-91°
30	CH ₃	3-Cl	H	H	-CH ₂ OCH ₃	b.p. 190-200°/0.1 Torr
31	CH ₃	4-Cl	H	CH ₃	-CH ₂ OCH ₃	
32	CH ₃	H	H	H	-CH ₂ CH ₂ OC ₃ H ₇ (i)	
33	CH ₃	3-Cl	H	H	-CH ₂ OC ₂ H ₅	b.p. 120-135°/0.06 Torr
34	CH ₃	3-CH ₃	5-CH ₃	H	-CH ₂ CH ₂ OCH ₃	
35	CH ₃	3-Cl	H	H	-CH ₂ OC ₃ H ₇ (n)	

Compound No.	R ₁	R ₂	R ₃	R ₄	R ₅	Physical constants
36	CH ₃	4-Cl	H	CH ₃	-CH ₂ OC ₂ H ₅	
37	CH ₃	H	H	H	-CH ₂ OCH ₂ CH ₂ OCH ₃	b.p. 186-195°/0.1 Torr
38	CH ₃	3-Cl	H	H	-CH ₂ OC ₂ H ₅ (i)	
39	CH ₃	3-CH ₃	H	H	-CH ₂ OCH ₂ CH ₂ OCH ₃	oil
40	CH ₃	3-CH ₃	H	H	-CH ₂ CH ₂ OC ₂ H ₅	b.p. 190-193°/0.15 Torr
41	CH ₃	3-Cl	H	H	-CH ₂ CH ₂ OCH ₃	
42	CH ₃	3-Cl	H	H	-CH ₂ CH ₂ OC ₂ H ₅	
43	CH ₃	H	H	H	-CH ₂ -O- 	b.p. 106-112°
44	CH ₃	H	H	H	-CH ₂ O-CH(CH ₃)-OC ₂ H ₅	oil
45	CH ₃	3-CH ₃	3-CH ₃	H	-CH ₂ O-CH(CH ₃)-OC ₂ H ₅	oil
46	CH ₃	3-CH ₃	H	H	-CH ₂ CH ₂ OCH ₃	b.p. 182-185°/0.08 Torr
47	CH ₃	3-CH ₃	5-CH ₃	H	-CH ₂ -O- 	m.p. 93-97°

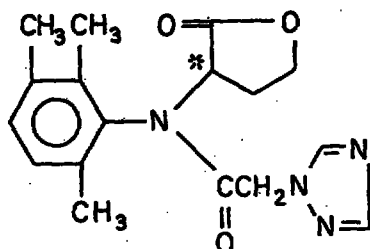
and also the following derivatives with R = -OCH₃ and R₄ = H:

Compound No.	R ₁	R ₂	R ₃	R ₄	Physical constants
48	CH ₃	H	H	-CH ₂ OCH ₃	m.p. 88-90°
49	Cl	H	H	-CH ₂ OCH ₃	m.p. 79-83°
50	Br	4-Cl	H	-CH ₂ OCH ₃	

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Example 4

Production of



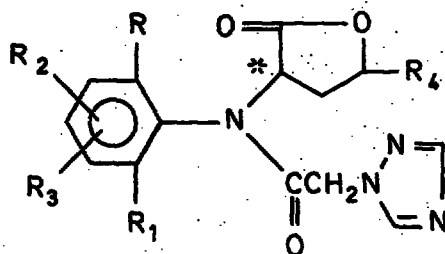
3-[N-(1,2,4-triazol-1-yl-acetyl)-N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone (compound No. 61).

a) 165 g of 2-bromo-4-butyrolactone is slowly added at 10° to 135 g of 2,3,6-trimethylaniline and 106 g of sodium carbonate in 750 ml of dimethylformamide, and the mixture is subsequently stirred for 16 hours at 70° and for 24 hours at 100°. After cooling to +10°, there is added, with cooling, 1200 ml of ice water. Stirring is maintained at 10° for 2 hours; the precipitate is filtered off and subsequently washed with water and petroleum ether to leave 3-[N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone, m.p. 108—110°.

b) 11 g of the intermediate product obtained according to a) is refluxed in 60 ml of abs. toluene with 6.2 g of chloroacetyl chloride for 4 hours; the solvent is distilled off after cooling, and the brownish-coloured residue is recrystallised from ethyl acetate/petroleum ether (b.p. 40—60°) to yield 3-[N-chloroacetyl-N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone, m.p. 92—94°.

c) 5.6 g of 1,2,4-triazole is added portionwise, in a nitrogen atmosphere, to 2.4 g of 50% sodium hydride dispersion in 100 ml of abs. dioxane, and refluxing is maintained until the generation of hydrogen has ceased. After cooling to +10°, there is added dropwise, with stirring, 8 g of the 3-[N-chloroacetyl-N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone in 100 ml of dioxane; the reaction mixture is refluxed for 16 hours in a nitrogen atmosphere and, after cooling, 100 ml of water is carefully added. The mixture is poured into ice water, and repeatedly extracted with methylene chloride. The combined extracts are washed with water, dried over sodium sulphate, filtered, and freed from solvent. After dissolving the residue in ethyl acetate, the solution is boiled with active charcoal and filtered hot, and petroleum ether (b.p. 40—60°) is added to the still hot filtrate until it starts to become cloudy; the filtrate is then cooled and the final product is filtered off. On being heated, it commences to slowly decompose from 70°.

There are produced in an analogous manner the following compounds of the formula

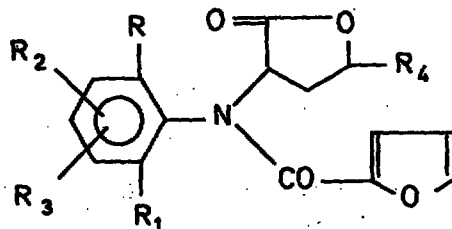


(R₁=6-position)

Compound No.	R	R ₁	R ₂	R ₃	R ₄	Physical data
51	CH ₃	CH ₃	H	H	H	m.p. 133 --135°
52	CH ₃	Cl	H	H	CH ₃	
53	CH ₃	C ₂ H ₅	H	H	H	
54	CH ₃	Cl	H	H	H	
55	CH ₃	CH ₃	3-CH ₃	5-CH ₃	H	oil
56	CH ₃	C ₂ H ₅	H	H	CH ₃	
57	CH ₃	CH ₃	H	H	CH ₃	
58	CH ₃	CH ₃	3-CH ₃	5-CH ₃	CH ₃	
59	CH ₃	CH ₃	3-Cl	H	H	m.p. 48° (decomp. °)
60	CH ₃	CH ₃	3-Br	H	CH ₃	
61	CH ₃	CH ₃	3-CH ₃	H	H	decomposition from 70°
62	CH ₃	CH ₃	3-Cl	H	CH ₃	
63	CH ₃	CH ₃	3-Br	H	H	
64	CH ₃	CH ₃	4-Cl	H	CH ₃	
65	CH ₃	CH ₃	3-CH ₃	H	CH ₃	
66	CH ₃	CH ₃	4-Cl	H	H	
67	CH ₃	Br	4-Cl	H	H	
68	CH ₃ O	CH ₃	H	H	H	oil
69	CH ₃ O	Cl	H	H	H	

20

There are produced in an analogous manner the following compounds of the formula



(R=CH₃; R₁=6-position)

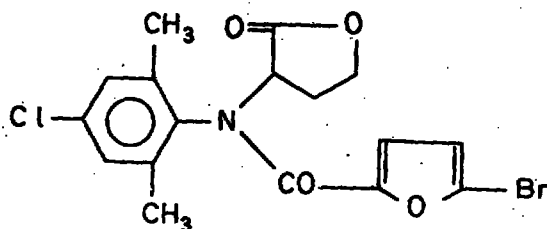
Compound No.	R ₁	R ₂	R ₃	R ₄	Physical constants
70	CH ₃	H	H	H	m.p. 143–145°
71	CH ₃	4-Cl	H	H	
72	CH ₃	H	H	CH ₃	m.p. 176–180°C
73	C ₂ H ₅	H	H	H	
74	Cl	H	H	H	
75	Br	4-Cl	H	H	
76	CH ₃	3-Br	H	H	
77	CH ₃	3-CH ₃	H	CH ₃	
78	CH ₃	3-CH ₃	H	H	m.p. 132–133°
79	CH ₃	3-Cl	H	H	m.p. 65° (decomp.)
80	CH ₃	3-CH ₃	5-CH ₃	H	m.p. 207–210°

and also the following derivatives with R = -OCH₃

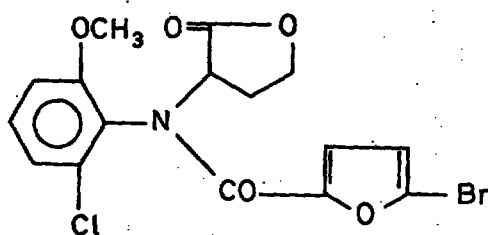
Compound No.	R ₁	R ₂	R ₃	R ₄	Physical constants
81	CH ₃	H	H	H	m.p. 164–165°
82	Cl	H	H	H	m.p. 152–153°
83	Cl	4-Cl	H	H	
84	Br	4-Br	H	H	
85	Br	H	H	H	
86	Cl	H	H	CH ₃	

and the compounds.

No. 87

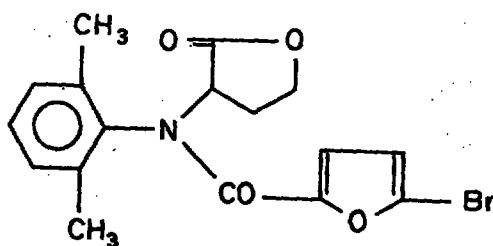


No. 88



m.p. 164—165°

No. 89

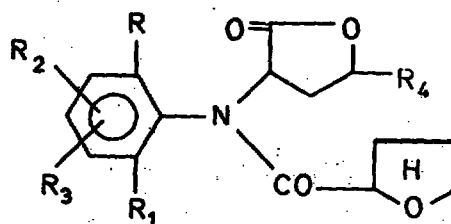


m.p. 158—160°

5

There are produced in an analogous manner the following compounds of the formula

5



(R=CH₃; R₁=6-position)

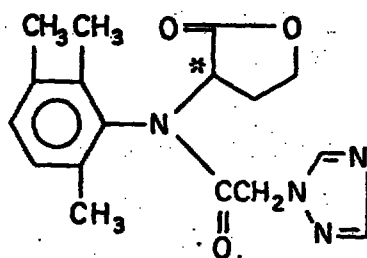
Compound No.	R ₁	R ₂	R ₃	R ₄	Physical constants
90	CH ₃	H	H	H	m.p. 142–146°
91	CH ₃	H	H	CH ₃	
92	C ₂ H ₅	H	H	H	
93	Cl	H	H	H	
94	CH ₃	4-Cl	H	H	m.p. 137–139°
95	CH ₃	3-CH ₃	H	H	m.p. 112–113°
96	CH ₃	4-Cl	H	CH ₃	
97	CH ₃	3-CH ₃	5-CH ₃	H	oil

as well as the following derivatives with R = -OCH₃:

Compound No.	R ₁	R ₂	R ₃	R ₄	Physical constants
98	CH ₃	H	H	H	oil
99	Cl	H	H	H	
100	Cl	4-Cl	H	H	
101	Br	4-Br	H	H	
102	Br	H	H	H	
103	Cl	H	H	CH ₃	

Example 5

Production of



5 3-[N-(1,2-pyrazol-1-yl-acetyl)-N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone (compound No. 111). 5

a) Production of 2-(1-pyrazolyl)-acetic acid

50 g of pyrazole in 250 ml of abs. tetrahydrofuran is added dropwise in a nitrogen atmosphere within 1/2 hour at 20–30°, with stirring and cooling, to 38.7 g of sodium hydride in 100 ml of abs. tetrahydrofuran. The reaction mixture is stirred for a further 3 hours at 40°; it is then cooled towards 5° and, with good cooling, 160.7 g of bromoacetic acid ethyl ester in 100 ml of abs. tetrahydrofuran is added dropwise at 0–10° during 1 hour. Stirring is maintained overnight at room temperature; there is then added dropwise 150 ml of ethanol; stirring is continued for 1 hour, and the suspension is subsequently concentrated by evaporation. To the residue is added a solution of 74 g of NaOH tablets in 600 ml of 60% methanol (aqueous), and refluxing is carried out

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10

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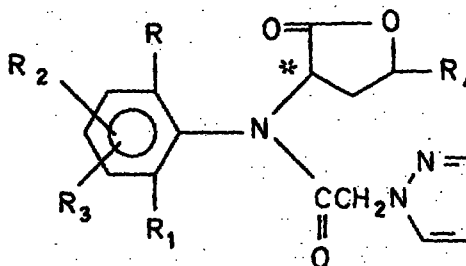
15

for 40 minutes. The solution is then cooled, and washed twice with 200 ml of ether. The aqueous phase is made acid to a congo-red indicator (about pH 2) with conc. hydrochloric acid at 5° with cooling, and the solution is continuously extracted during 24 hours with methylene chloride. The extract is concentrated by evaporation, and recrystallised from ether/tetrahydrofuran. The crystals melt at 167—169°.

b) 165 g of 2-bromo-4-butyrolactone is slowly added at 10°C to 135 g of 2,3,6-trimethylaniline and 106 g of sodium carbonate in 750 ml of dimethylformamide, and the mixture is subsequently stirred for 16 hours at 70° and for 24 hours at 100°. After cooling to +10°, there is added, with cooling, 1200 ml of ice water. Stirring is continued at 10° for 2 hours; the precipitate is filtered off, and washed with water and petroleum ether to yield 3-[N-2,3,6-trimethylphenyl]-amino-tetrahydro-2-furanone, m.p. 108—110°.

c) 6.3 g of 2-(1-pyrazolyl)-acetic acid is placed into 200 ml of toluene; 6.2 g of thionyl chloride is added, and the mixture is refluxed for 1 hour. It is then cooled to 20°, and 11 g of the intermediate obtained according to a) and 5.8 g of sodium carbonate are added. After stirring overnight at room temperature, ice water is added, and extraction is performed with methylene chloride; the extracts are washed with water, dried over sodium sulphate and concentrated by evaporation. The resin remaining is purified through a silica gel column, and finally frozen out from ether at -50°. The vitreous diastereoisomeric mixture of 3-[N-(1,2-pyrazol-1-yl-acetyl)-N-2,3,6-trimethylphenyl]-amino-tetrahydrofuranone melts at 50—60°.

There are produced in an analogous manner the following compounds of the formula



(R₁ = 6-position)

Compound	R	R ₁	R ₂	R ₃	R ₄
104	CH ₃	CH ₃	H	H	H
105	CH ₃	C ₂ H ₅	H	H	H
106	CH ₃	CH ₃	3-CH ₃	5-CH ₃	H
107	CH ₃	C ₂ H ₅	H	H	CH ₃
108	CH ₃	CH ₃	H	H	CH ₃
109	CH ₃	CH ₃	3-CH ₃	5-CH ₃	CH ₃
110	CH ₃	CH ₃	3-Cl	H	H
111	CH ₃	CH ₃	3-CH ₃	H	H
112	CH ₃	CH ₃	3-CH ₃	H	CH ₃
113	CH ₃	CH ₃	4-Cl	H	H
114	CH ₃ O	CH ₃	H	H	H

The compounds of the formula I can be used on their own or in composition form together with one or more inert carriers and/or other additives. Suitable carriers and additives can be solid or liquid and they correspond to the substances common in formulation practice, such as natural or regenerated mineral substances, solvents, dispersing agents, wetting agents, adhesives, thickeners, binders and/or fertilisers.

Suitable compositions according to the invention contain, together with the active ingredient, a solid extender and optionally a surface active agent, or a liquid diluent and a surface active agent.

The content of active substance in commercial compositions is generally between 0.1 and 90% by weight.

For application, the compounds of the formula I can be in the following forms (the weight-percentage figures in brackets signify advantageous amounts of active substance):

solid preparations:

dusts and scattering agents (up to 10%), granulates [coated granules, impregnated granules and homogeneous granules] (1 to 80%);

liquid preparations:

a) water-dispersible concentrates of active substance: wettable powders and pastes (25 to 90% in the commercial packing, 0.01 to 15% in ready-for-use solutions);

emulsion concentrates and solution concentrates (10 to 50%, 0.01 to 15% in ready-for-use solutions);

b) solutions (0.1 to 20%).

The active substances of the formula I of the present invention can be formulated for example as follows (parts and percentages are by weight).

Dust:

The following substances are used to produce a) a 5% dust and b) a 2% dust:

- | | | |
|----|----|--|
| a) | 5 | parts of active substance, |
| | 95 | parts of talcum; |
| b) | 2 | parts of active substance, |
| | 1 | part of highly dispersed silicic acid, and |
| | 97 | parts of talcum. |

The active substances are mixed and ground with the carriers, and in this form they can be applied by dusting.

Granulate:

The following substances are used to produce a 5% granulate:

- | | |
|------|---|
| 5 | parts of active substance, |
| 0.25 | part of epichlorohydrin, |
| 0.25 | part of cetyl polyglycol ether, |
| 3.50 | parts of polyethylene glycol, and |
| 91 | parts of kaolin (particle size 0.3—0.8 mm). |

The active substance is mixed with epichlorohydrin and dissolved in 6 parts of acetone; the polyethylene glycol and cetyl polyglycol ether are then added. The solution thus obtained is sprayed onto kaolin, and the acetone is subsequently evaporated off in vacuo. A microgranulate of this kind is particularly suitable for soil application.

Wettable powder:

The following constituents are used to produce a) a 70% wettable powder, b) a 40% wettable powder, c) and d) a 25% wettable powder, and e) a 10% wettable powder:

- | | | |
|----|----|--|
| a) | 70 | parts of active substance, |
| | 5 | parts of sodium dibutyl-naphthylsulphonate, |
| | 3 | parts of naphthalenesulphonic acid/phenolsulphonic acid/formaldehyde condensate 3:2:1, |
| | 10 | parts of kaolin, and |
| | 12 | parts of Champagne chalk; |

	b)	40	parts of active substance,	
		5	parts of sodium lignin sulphonate,	
		1	part of sodium dibutyl-naphthalenesulphonate, and	
		54	parts of silicic acid;	
5	c)	25	parts of active substance,	5
		4.5	parts of calcium lignin sulphonate,	
		1.9	parts of Champagne chalk/hydroxyethylcellulose mixture (1:1),	
		1.5	parts of sodium dibutyl-naphthalenesulphonate,	
		19.5	parts of silicic acid,	
10		19.5	parts of Champagne chalk, and	10
		28.1	parts of kaolin;	
	d)	25	parts of active substance,	
		2.5	parts of isooctylphenoxy-polyoxyethylene-ethanol,	
		1.7	parts of Champagne chalk/hydroxyethylcellulose mixture (1:1),	
15		8.3	parts of sodium aluminium silicate,	15
		16.5	parts of kieselguhr, and	
		46	parts of kaolin; and	
	e)	10	parts of active substance,	
		3	parts of a mixture of the sodium salts of saturated fatty alcohol	
20			sulphates,	20
		5	parts of naphthalenesulphonic acid/formaldehyde condensate, and	
		82	parts of kaolin.	

25 The active substances are intimately mixed in suitable mixers with the additives, and the mixture is then ground in the appropriate mills and rollers. There are obtained wettable powders which have excellent wetting and suspension properties, which can be diluted with water to give suspensions of the desired concentration, and which can be used in particular for leaf application. 25

Emulsifiable concentrate:

The following substances are used to produce a 25% emulsifiable concentrate:

30	25	parts of active substance,	30
	2.5	parts of epoxidised vegetable oil,	
	10	parts of an alkylarylsulphonate/fatty alcohol polyglycol ether mixture,	
	5	parts of dimethylformamide, and	
	57.5	parts of xylene.	

35 Emulsions of the desired concentration can be prepared from these concentrates by dilution with water; and these emulsions are particularly suitable for leaf application. 35

40 In order to adapt them to suit prevailing circumstances, the compounds of the formula I can be used, for widening their sphere of action, together with other suitable pesticides, such as fungicides, insecticides, acaricides, nematocides, rodenticides or herbicides, or with active substances affecting plant growth and also with fertilisers. 40

Example 6

Action against *Phytophthora infestans* on tomatoes

I) Curative action

45 Tomato plants of the variety "Roter Gnom" are sprayed, after three weeks' cultivation, with a zoospore suspension of the fungus, and incubated in a chamber at 18° to 20° with saturated atmospheric humidity. The incubation treatment is interrupted after 24 hours; the plants are dried and then sprayed with a liquor containing the active substance in the form of a wettable powder at a concentration of 0.06% and 0.02%. 45

50 The applied coating is allowed to dry, and the plants are subsequently returned to the moist-atmosphere chamber for 4 days. The number and size of the typical leaf spots appearing after this time serve as a criterion for an assessment of the effectiveness of the substances tested. Compounds of the formula I consistently produced, at an active-substance concentration of 0.06%, a reduction of fungus infestation to less than 20%. 50

55 The compounds Nos. 1, 2, 5, 12, 19, 24, 28, 30, 33, 51, 61, 70, 79, 80, 82, 89, 90, 95, 98, 111 and others brought about to this concentration a reduction of fungus infestation to 0—5%. At the concentration of active substance of 0.02%, a reduction 55

of fungus infestation to 0—5% was produced by, inter alia, the compounds Nos. 1, 2, 5, 12, 19, 28, 30, 33, 70, 79, 80, 90 and 95. No phytotoxicity was observed.

II) Preventitive systemic action

The active substance in the form of a wettable powder is applied in a concentration of 0.006% (relative to the volume of soil) to the surface of the soil around 3-week old potted tomato plants of the "Roter Gnom" variety. After a period of three days, the under side of the leaves of the plants is sprayed with a zoospore suspension of *Phytophthora infestans*. The plants are then stored for 5 days in a spray chamber at 18°—20° with a saturated atmosphere. The typical leaf spots appear after this time; on the basis of their number and size, an evaluation is then made of the effectiveness of the substances tested.

The tomato plants treated with compounds of the formula I showed consistently less than 20% fungus infestation, compared with that exhibited by control plants (=100% infestation). The compounds Nos. 1, 2, 5, 12, 19, 24, 28, 30, 33, 51, 61, 70, 78, 79, 80, 82, 88, 89, 90, 95, 98, 104, 106, 111 and others prevented fungus infestation completely or almost completely (0—5% infestation).

Example 7

Action against *Pythium debaryanum* on sugar beet

a) Action after soil application

The fungus is cultivated on sterile oat seeds and thus added to a soil/sand mixture. The soil infested in this way is filled into flower pots and sown with sugar-beet seeds. Immediately after sowing, the test preparations formulated as wettable powder are poured as aqueous suspensions over the soil (20 ppm of active substance relative to the volume of soil). The pots are then placed in a greenhouse at 20—24° for 2—3 weeks. The soil during this time is kept uniformly moist by a light spraying with water. In the assessment of the tests, the sprouting of the sugar-beet plants and also the proportion of healthy plants and of diseased plants is determined. After the treatment with the active substances of the formula I, more than 80% of the sugar-beet plants were sprouting and had a healthy appearance. This result was obtained also with an active-substance concentration in the soil of 6 ppm with use of the compounds Nos. 1, 2, 4, 5, 9, 12, 13, 15, 16, 19, 30, 33, 37, 59, 61, 79, 95, 104, 111 and others.

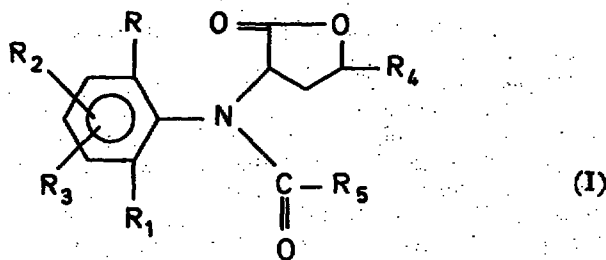
b) Action after application as dressing

The fungus is cultivated on sterile oat seeds and thus added to a soil/sand mixture. The soil infested in this manner is filled into flower pots, and sown with sugar-beet seeds which have been dressed with the test preparations formulated as dressing powder (1000 ppm of active substance relative to the weight of seed). The sown pots are placed in a greenhouse at 20—24° for 2—3 weeks. The soil during this time is kept uniformly moist by a light spraying with water. In the assessment of the tests, the sprouting of the sugar-beet plants and also the proportion of healthy plants and of diseased plants is determined.

After the treatment with active substances of the formula I, more than 80% of the sugar-beet plants were sprouting and had a healthy appearance.

WHAT WE CLAIM IS:—

1. A compound of the formula I



wherein

- R represents C₁—C₄ alkyl, C₁—C₄ alkoxy or halogen,
 R₁ represents C₁—C₃ alkyl, C₁—C₄ alkoxy or halogen,
 R₂ represents hydrogen, C₁—C₃ alkyl, C₁—C₄ alkoxy or halogen,

II) Preventitive systemic action

The tomato plants treated with compounds of the formula I showed consistently less than 20% fungus infestation, compared with that exhibited by control plants (=100% infestation). The compounds Nos. 1, 2, 5, 12, 19, 24, 28, 30, 33, 51, 61, 70, 78, 79, 80, 82, 88, 89, 90, 95, 98, 104, 106, 111 and others prevented fungus infestation completely or almost completely (0—5% infestation).

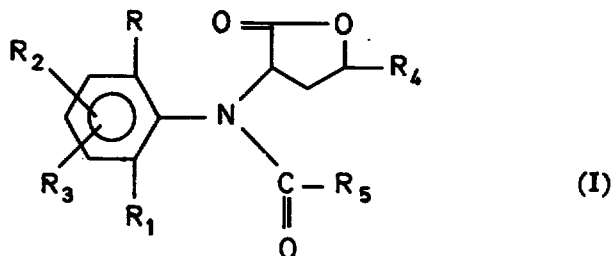
Action against *Pythium debaryanum* on sugar beet

The fungus is cultivated on sterile oat seeds and thus added to a soil/sand mixture. The soil infested in this way is filled into flower pots and sown with sugar-beet seeds. Immediately after sowing, the test preparations formulated as wettable powder are poured as aqueous suspensions over the soil (20 ppm of active substance relative to the volume of soil). The pots are then placed in a greenhouse at 20—24° for 2—3 weeks. The soil during this time is kept uniformly moist by a light spraying with water. In the assessment of the tests, the sprouting of the sugar-beet plants and also the proportion of healthy plants and of diseased plants is determined. After the treatment with the active substances of the formula I, more than 80% of the sugar-beet plants were sprouting and had a healthy appearance. This result was obtained also with an active-substance concentration in the soil of 6 ppm with use of the compounds Nos. 1, 2, 4, 5, 9, 12, 13, 15, 16, 19, 30, 33, 37, 59, 61, 79, 95, 104, 111 and others.

The fungus is cultivated on sterile oat seeds and thus added to a soil/sand mixture. The soil infested in this manner is filled into flower pots, and sown with sugar-beet seeds which have been dressed with the test preparations formulated as dressing powder (1000 ppm of active substance relative to the weight of seed). The sown pots are placed in a greenhouse at 20—24° for 2—3 weeks. The soil during this time is kept uniformly moist by a light spraying with water. In the assessment of the tests, the sprouting of the sugar-beet plants and also the proportion of healthy plants and of diseased plants is determined.

After the treatment with active substances of the formula I, more than 80% of the sugar-beet plants were sprouting and had a healthy appearance.

1. A compound of the formula I



wherein

R represents C₁—C₄ alkyl, C₁—C₄ alkoxy or halogen,
R₁ represents C₁—C₃ alkyl, C₁—C₄ alkoxy or halogen,
R₂ represents hydrogen, C₁—C₃ alkyl, C₁—C₄ alkoxy or halogen,



R_2 represents hydrogen or methyl, with the total number of carbon atoms of the substituents R , R_1 , R_2 and R_3 in the phenyl ring not exceeding the number 8,

R_4 represents hydrogen or methyl, and

R_5 represents one of the following groups:

an alkylthioalkyl or alkoxyalkyl group having 2 to 5 carbon atoms or an alkoxyalkoxy-methyl group having a maximum of 6 carbon atoms, a 2-furanyl or 2-tetrahydrofuranyl group optionally substituted by halogen, a 1,2,4-triazolylmethyl group, a 1,2-pyrazolyl-methyl group, or a group $-\text{CH}_2-\text{OR}_6$ in which R_6 represents a 5- or 6-membered heterocycle having oxygen as the hetero atom.

2. A compound according to claim 1 wherein R represents methyl; R_1 represents methyl, ethyl, chlorine or bromine; R_2 represents hydrogen, halogen or methyl; R_3 represents hydrogen or methyl; and R_4 and R_5 are as defined in claim 1.

3. A compound according to Claim 1 or 2, wherein R_5 represents an alkylthioalkyl or alkoxyalkyl group having 2 to 5 carbon atoms, a 2-furanyl or 2-tetrahydrofuranyl group optionally substituted by halogen, or a 1,2,4-triazolylmethyl group.

4. A compound according to Claim 3, wherein R_5 represents methoxymethyl or ethoxymethyl.

5. A compound according to Claim 1 or 2, wherein R_5 represents a 1,2-pyrazolyl-methyl group.

6. 3-[N-(Methoxyacetyl)-N-(2,6-dimethylphenyl)]-aminotetrahydro-2-furanone.

7. 3-[N-(Ethoxyacetyl)-N-(2,6-dimethylphenyl)]-amino-tetrahydro-2-furanone.

8. 3-[N-(Methoxyacetyl)-N-(2,6-dimethyl-3-chlorophenyl)]-amino-tetrahydro-2-furanone.

9. 3-[N-(Ethoxyacetyl)-N-(2,6-dimethyl-3-chlorophenyl)]-amino-tetrahydro-2-furanone.

10. 3-[N-(Methoxyacetyl)-N-(2,6-dimethylphenyl)]-amino-5-methyl-tetrahydro-2-furanone.

11. 2,6-Dimethyl-N-(3-tetrahydrofuran-2-one)-2-tetrahydrofuroyl-anilide.

12. 2,3,6-Trimethyl-N-(3-tetrahydrofuran-2-one)-2-tetrahydrofuroyl-anilide.

13. 2,6-Dimethyl-4-chloro-N-(3-tetrahydrofuran-2-one)-2-tetrahydrofuroyl-anilide.

14. 3-[N-(Methoxyacetyl)-N-(2,3,5,6-tetramethylphenyl)]-amino-tetrahydro-2-furanone.

15. 2,3,5,6-Tetramethyl-N-(3-tetrahydrofuran-2-one)-2-furoyl-anilide.

16. 3-[N-(Methoxyacetyl)-N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone.

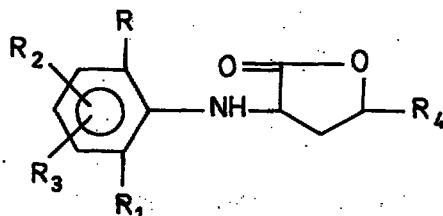
17. 3-[N-(Methoxyacetyl)-N-(2,3,5,6-tetramethylphenyl)]-amino-tetrahydro-2-furanone.

18. 3-[N-(Methoxyacetyl)-N-(2,6-dimethyl-4-chlorophenyl)]-amino-tetrahydro-2-furanone.

19. 3-[N-(2-Methoxyethoxy-acetyl)-N-(2,3,6-trimethylphenyl)]-amino-tetrahydro-2-furanone.

20. 3-[N-(Methoxyacetyl)-N-(2-methoxy-6-chlorophenyl)]-amino-tetrahydrofuran-2-one.

21. A process for preparing a compound of the formula I as claimed in claim 1 which process comprises reacting a compound of the formula II



(II)

(wherein R , R_1 , R_2 , R_3 and R_4 are as defined in claim 1) with a compound of the formula III



(III).

(wherein R_5 is as defined in claim 1 and Hal' represents a halogen atom).

22. A process for preparing a compound of the formula I as claimed in claim 1 wherein R_5 represents a 1,2,4-triazolylmethyl, 1,2-pyrazolylmethyl or alkylthio group,

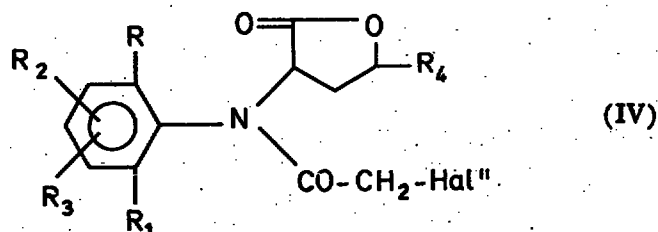
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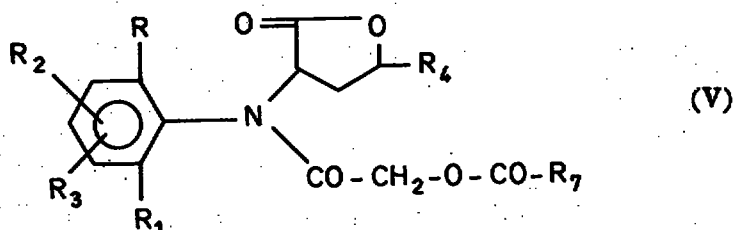
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which process comprises monohaloacetylating a compound of the formula II as defined in claim 21 to give a compound of the formula IV



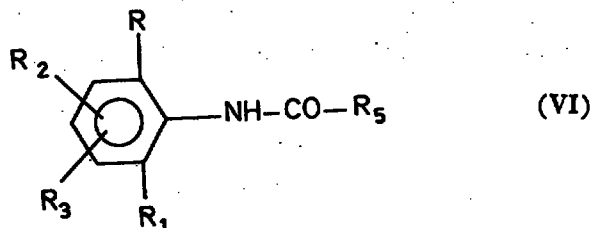
(wherein R, R₁, R₂, R₃ and R₄ are as defined in claim 1 and Hal'' represents a halogen atom) and reacting the compound of the formula IV with 1,2,4-triazole, 1,2-pyrazole or an alkylmercaptan, or with the metal salt of one of these reactants.

23. A process for preparing a compound of the formula I as claimed in claim 1 wherein R₄ represents an alkoxyalkoxymethyl group or a group —CH₂—OR₄ in which R₄ represents a saturated 5- or 6-membered heterocycle having oxygen as the hetero atom which process comprises monohaloacetylating a compound of the formula II as defined in claim 21 to give a compound of the formula IV as defined in claim 22, exchanging the Hal'' substituent by reaction with an alkanecarboxylic acid containing less than 7 carbon atoms in the alkane moiety or an alkali metal or alkaline earth metal salt thereof to give a compound of the formula V

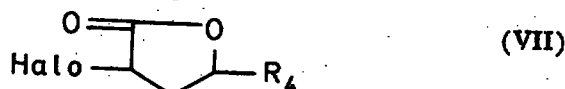


(wherein R, R₁, R₂, R₃ and R₄ are as defined in claim 1 and R₇ represents an alkyl group containing less than 7 carbon atoms), subjecting the compound of the formula V to mild alcoholysis to give the corresponding hydroxyacetanilide, acidifying the hydroxyacetanilide which is activated with an alkali metal, an alkali metal hydride or p-toluene sulphonic acid, and reacting with an alkoxyalkyl halide, 2,3-dihydrofuran or 3,4-dihydro-2H-pyrene.

24. A process for preparing a compound of the formula I as claimed in claim 1 which process comprises reacting a compound of the formula VI



(wherein R, R₁, R₂, R₃ and R₅ are as defined in claim 1) in the presence of a proton acceptor with a compound of the formula VII



(wherein R₄ is as defined in claim 1 and Halo represents a halogen atom).

25. A process for preparing a compound of the formula I as claimed in claim 1 substantially as described with reference to any one of Examples 1 to 5.



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26. A compound according to claim 1 prepared by the process claimed in any one of claims 21 to 25.
27. A method of combating phytopathogenic fungi which method comprises applying to a locus infested with or liable to be infested with the fungi a compound as claimed in claim 1. 5
28. A method according to claim 27 wherein a compound as claimed in claim 2 or 19 is applied.
29. A method according to claim 27, wherein a compound as claimed in any one of claims 3, 4, 6 to 18 and 20 is applied.
- 10 30. A method according to claim 27 wherein a compound as claimed in claim 5 is applied. 10
31. A fungicidal composition which contains, as active ingredient, a compound as claimed in claim 1, together with one or more inert carriers and/or other additives.
- 15 32. A fungicidal composition according to claim 31 containing, as active ingredient, a compound as claimed in claim 2 or 19. 15
33. A fungicidal composition according to claim 31 containing, as active ingredient, a compound as claimed in any one of claims 3, 4, 6 to 18 and 20.
34. A fungicidal composition according to claim 31 containing as active ingredient, a compound as claimed in claim 5.
- 20 35. A composition according to any one of claims 31 to 34 which contains, together with the active ingredient, a solid extender and optionally a surface active agent, or a liquid diluent and a surface active agent. 20
36. Seeds or plant cuttings dressed with a compound as claimed in claim 1.

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