13318/88

COMMONWEALTH of AUSTRALIA

PATENTS ACT 1952

APPLICATION FOR A STANDARD PATENT

XX
I/We SCHERING AGROCHEMICALS LIMITED,
of Hauxton,
Cambridge CB2 5HU,
ENGLAND

601558

APPLICATION ACCEPTED AND AMENDMENTS
AS FILED 6-7-90

hereby apply for the grant of a Standard Patent for an invention entitled:

~~"FUNGICIDES"~~ Fungicidal imidazole and 1,2,4-triazole derivatives

which is described in the accompanying ~~provisional~~ complete specification.

Details of basic application(s):—

Number	Convention Country	Date
87/06779	UNITED KINGDOM	21st March 1987
87/22329	UNITED KINGDOM	22nd September 1987

LODGED AT SUB-OFFICE
2 1 MAR 1988
Melbourne

The address for service is care of DAVIES & COLLISON, Patent Attorneys, of 1 Little Collins Street, Melbourne, in the State of Victoria, Commonwealth of Australia.

Dated this 21st day of March 19 88

AUSTRALIAN PATENT OFFICE

To: THE COMMISSIONER OF PATENTS

H. M. Rimington
(a member of the firm of DAVIES & COLLISON for and on behalf of the Applicant).

Davies & Collison, Melbourne and Canberra.

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

DECLARATION IN SUPPORT OF CONVENTION OR NON-CONVENTION APPLICATION FOR A PATENT

In support of the Application made for a patent for an invention entitled : "FUNGICIDES"

Insert title of invention.

I / ~~We~~ DAVID LEONARD RICHER
SCHERING AGROCHEMICALS LIMITED
Hauxton, Cambridge CB2 5HU, England

Insert full name(s) and address(es) of declarant(s) being the applicant(s) or person(s) authorized to sign on behalf of an applicant company.

do solemnly and sincerely declare as follows :-

1. (a) ~~I am~~ / We are the applicant..... for the patent

or (b) I am authorized by

SCHERING AGROCHEMICALS LIMITED

the applicant..... for the patent to make this declaration on ~~its~~ / ~~xxxx~~ behalf.

Cross out whichever of paragraphs 1(a) or 1(b) does not apply

1(a) relates to application made by individual(s)
1(b) relates to application made by company; insert name of applicant company.

2. (a) ~~I am~~ / We are the actual inventor..... of the invention

or (b) ANTONY DAVID BUSS, a British Subject of
Ahornstrasse 31, 1000 Berlin 41,
Federal Republic of Germany
PHILIP JOHN DUDFIELD, a British Subject of
1 Hamilton Mews, Saffron Walden,
Essex, England
JOHN HENRY PARSONS, a British Subject of
7 Maypole Close, Saffron Walden
Essex, England

Cross out whichever of paragraphs 2(a) or 2(b) does not apply

2(a) relates to application made by inventor(s)
2(b) relates to application made by company(s) or person(s) who are not inventor(s); insert full name(s) and address(es) of inventors.

~~is~~ / are the actual inventor~~s~~..... of the invention and the facts upon which the applicant..... ~~is~~ / are entitled to make the application are as follows :-

The Applicant is the Assignee of the Inventors in respect of the invention.

State manner in which applicant(s) derive title from inventor(s)

3. The basic application..... as defined by Section 141 of the Act ~~was~~ / were made

in Great Britain on the 21st March 1987

by SCHERING AGROCHEMICALS LIMITED, Hauxton, Cambridge, England

in Great Britain on the 22nd September 1987

by SCHERING AGROCHEMICALS LIMITED, Hauxton, Cambridge, England

in ..... on the .....

by .....

Cross out paragraphs 3 and 4 for non-convention applications. For convention applications, insert basic country(s) followed by date(s) and basic applicant(s).

4. The basic application..... referred to in paragraph 3 of this Declaration ~~was~~ / were the first application..... made in a Convention country in respect of the invention the subject of the application.

Insert place and date of signature.

Declared at Cambridge, England this 15th day of February 1988

for SCHERING AGROCHEMICALS LIMITED

Signature of declarant(s) (no attestation required)

.....

D L Richer - By Power of Attorney

Note: Initial all alterations.

DAVIES & COLLISON, MELBOURNE and CANBERRA.

(12) PATENT ABRIDGMENT (11) Document No. AU-B-13318/88
(19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 601558

(54) Title
IMIDAZOLE AND 1,2,4 TRIAZOLE DERIVATIVES

International Patent Classification(s)
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C07D 233/84 C07D 249/12 C07D 409/04 C07D 417/04
C07F 009/65 A01N 043/653

(21) Application No. : 13318/88 (22) Application Date : 21.03.88

(30) Priority Data

(31) Number	(32) Date	(33) Country
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8722329	22.09.87	GB UNITED KINGDOM

(43) Publication Date : 22.09.88

(44) Publication Date of Accepted Application : 13.09.90

(71) Applicant(s)
SCHERING AGROCHEMICALS LIMITED

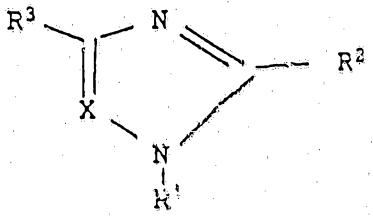
(72) Inventor(s)
ANTHONY DAVID BUSS; PHILIP JOHN DUDFIELD; JOHN HENRY PARSONS

(74) Attorney or Agent
DAVIES & COLLISON, MELBOURNE

(56) Prior Art Documents
AU 29660/77 C07D 233/90

(57) Claim

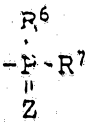
1) A compound of formula I

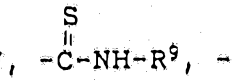


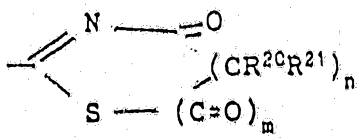
(I)

in which

X is CR^4 or N;

R^1 is $-SO_2R^5$,  or $-COR^8$

R^2 is CN,  , $-CH=N-OR^{10}$,

 or 

R^3 and R^4, may be the same or different and are alkyl,

.../2

cycloalkyl, cycloalkenyl, alkenyl, or alkynyl, ~~or amino,~~ (all of which are optionally substituted by halogen, hydroxy, alkoxy or aryl), hydrogen, halogen, hydroxy, cyano, nitro, acyl, $R^{11}SO_p$, $R^{12}O$ or aryl;
R^5 is aryl, optionally substituted alkyl or optionally substituted amino;
R^6 and R^7, may be the same or different and are amino, alkoxy or alkylthio, each of which is optionally substituted;
R^8 has the same meaning as R^5 or can be alkoxy, alkenyloxy, alkynyloxy or alkylthio, each of which is optionally substituted, or is aryloxy;
R^9 is hydrogen, optionally substituted alkyl, alkenyl, alkynyl, alkoxycarbonyl, acyl, aryl or cycloalkyl;
R^{10} is hydrogen, optionally substituted alkyl, alkenyl or alkynyl;
R^{11} is alkyl, cycloalkyl, cycloalkenyl, alkenyl, alkynyl, all of which are optionally substituted, or is aryl;
R^{12} has the same meaning as R^{11} or is acyl;
R^{17} is hydrogen, alkyl, alkoxycarbonyl, or aryl and R^{18} is hydrogen or alkyl or R^{17} and R^{18} together with the carbons to which they are attached, form a benzo ring;
R^{20} and R^{21} may be the same or different and are hydrogen or alkyl;
Z is oxygen or sulphur;
m is 0 and n is 1 or 2 or m is 1 and n is 0 or 1; and
p is 0 or 1,
with the proviso that R^3 and R^4 cannot both be chlorine, and in which

a) any substituent on any alkyl, cycloalkyl, cycloalkenyl, alkenyl, alkoxy or alkynyl containing group is selected from halogen, hydroxy, alkoxy and aryl,
b) any aryl group is phenyl, optionally substituted, by halogen, alkyl, alkoxy, nitro, cyano, $-COR^8$, optionally substituted sulphamoyl, optionally substituted amino, alkyl-SO_q, or aryl-SO_q, where q is 0, 1 or 2, and any alkyl or alkoxy group is

.../3

optionally substituted,

c) any acyl group is the residue of both carboxylic and sulphonic acids and includes the groups $R^{13}(O)_rCO$ and $R^{13}SO_2$, where R^{13} has the same meaning as R^{11}, or is optionally substituted amino and r is 0 or 1,

d) any amino or sulphamoyl group is optionally substituted by one or more of the groups R^{11}, acyl, optionally substituted amino, hydroxy or optionally substituted alkoxy.

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

COMPLETE SPECIFICATION

(Original)

FOR OFFICE USE

Class Int. Class

Application Number:
Lodged:

60 1558

Complete Specification Lodged:
Accepted:
Published:

This document contains the amendments made under Section 49 and is correct for printing.

Priority:

Related Art:

Name of Applicant: SCHERING AGROCHEMICALS LIMITED

Address of Applicant: Hauxton,
Cambridge CB2 5HU,
ENGLAND

Actual Inventor(s): Antony David **BUSS**
Philip John **DUDFIELD**
John Henry **PARSONS**

Address for Service: DAVIES & COLLISON, Patent Attorneys,
1 Little Collins Street, Melbourne, 3000.

Complete specification for the invention entitled:

~~"FUNGICIDES"~~

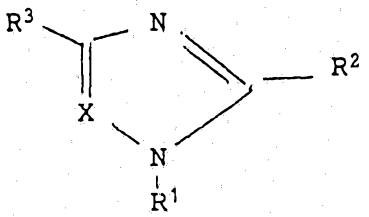
Fungicidal imidazole and 1,2,4-triazole derivatives

The following statement is a full description of this invention, including the best method of performing it known to us :-

AUSTRALIAN PATENT OFFICE

This invention relates to compounds having fungicidal activity.

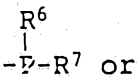
According to the invention there is provided a compound of formula I

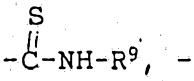


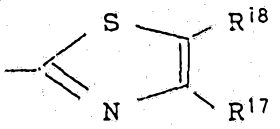
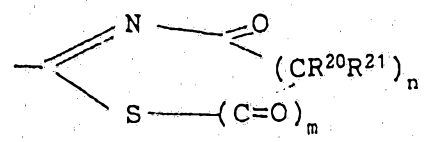
(I)

in which

X is CR^4 or N;

R^1 is $-SO_2R^5$,  or $-COR^8$

R^2 is CN,  , $-CH=N-OR^{10}$,

 or 

R^3 and R^4, may be the same or different and are alkyl, cycloalkyl, cycloalkenyl, alkenyl, or alkynyl, ~~or amino,~~ (all of which are optionally substituted by halogen, hydroxy, alkoxy or aryl), hydrogen, halogen, hydroxy, cyano, nitro, acyl, $R^{11}SO_p$, $R^{12}O$ or aryl;

R^5 is aryl, optionally substituted alkyl or optionally substituted amino;

R^6 and R^7, may be the same or different and are amino, alkoxy or alkylthio, each of which is optionally substituted;

R^8 has the same meaning as R^5 or can be alkoxy, alkenyloxy, alkynyloxy or alkylthio, each of which is optionally substituted, or is aryloxy;

R^9 is hydrogen, optionally substituted alkyl, alkenyl, alkynyl, alkoxycarbonyl, acyl, aryl or cycloalkyl;

R^{10} is hydrogen, optionally substituted alkyl, alkenyl or alkynyl;
R^{11} is alkyl, cycloalkyl, cycloalkenyl, alkenyl, alkynyl, all of which are optionally substituted, or is aryl;
R^{12} has the same meaning as R^{11} or is acyl;
R^{17} is hydrogen, alkyl, alkoxycarbonyl, or aryl and R^{18} is hydrogen or alkyl or R^{17} and R^{18} together with the carbons to which they are attached, form a benzo ring;
R^{20} and R^{21} may be the same or different and are hydrogen or alkyl;
Z is oxygen or sulphur;
m is 0 and n is 1 or 2 or m is 1 and n is 0 or 1; and
p is 0 or 1,
with the proviso that R^3 and R^4 cannot both be chlorine, and in which

a) any substituent on any alkyl, cycloalkyl, cycloalkenyl, alkenyl, alkoxy or alkynyl containing group is selected from halogen, hydroxy, alkoxy and aryl,

b) any aryl group is phenyl, optionally substituted, by halogen, alkyl, alkoxy, nitro, cyano, $-COR^8$, optionally substituted sulphamoyl, optionally substituted amino, alkyl-SO_q, or aryl-SO_q, where q is 0, 1 or 2, and any alkyl or alkoxy group is optionally substituted,

c) any acyl group is the residue of both carboxylic and sulphonic acids and includes the groups $R^{13}(O)_rCO$ and $R^{13}SO_2$, where R^{13} has the same meaning as R^{11}, or is optionally substituted amino and r is 0 or 1,

d) any amino or sulphamoyl group is optionally substituted by one or more of the groups R^{11}, acyl, optionally substituted amino, hydroxy or optionally substituted alkoxy.

Alkyl and alkoxy groups are preferably of 1 to 8 carbon atoms, especially methyl, and alkenyl and alkynyl groups are usually of 3 to 4 carbon atoms. Acyl groups are preferably alkanoyl, aroyl, alkylsulphonyl, arylsulphonyl, N,N-dialkylsulphamoyl or N-alkyl-N-arylsulphamoyl, in which the alkyl groups are e.g. of 1 to 4 carbon atoms, and the alkyl and phenyl can be substituted as previously mentioned. Cycloalkyl groups are usually of 3 to 8 carbon atoms especially cyclopentyl or cyclohexyl.

R^1 is generally a substituted sulphamoyl group, especially dimethylsulphamoyl. R^2 is preferably cyano or thiocarbamoyl. When X is CR^4, R^4 is preferably hydrogen, but may be cyano, methyl or ethoxyalkyl. R^3 is preferably phenyl or benzoyl, optionally substituted by up to three groups, selected from halogen, alkyl (especially methyl), trifluoromethyl, nitro and alkoxy (especially methoxy).

The compounds of the invention have activity as fungicides, especially against fungal diseases of plants, e.g. downy mildews, especially vine downy mildew (Plasmopara viticola), and late tomato blight and potato blight (Phytophthora infestans). They are also active against powdery mildews, such as barley powdery mildew (Erysiphe graminis), as well as being active against diseases such as rice blast (Pyricularia oryzae) and apple scab (Venturia inaequalis). They may also have activity against other fungi, such as Botrytis spp., Puccinia spp., Rhizoctonia spp., Fusarium spp. and Pythium spp..

The invention thus also provides a method of combating fungi at a locus infested or liable to be infested therewith, which comprises applying to the locus a

compound of formula I.

The invention also provides an agricultural composition comprising a compound of formula I in admixture with an agriculturally acceptable diluent or carrier.

The composition of the invention may of course include more than one compound of the invention.

In addition the composition can comprise one or more additional active ingredients, for example compounds known to possess plant-growth regulant, herbicidal, fungicidal, insecticidal or acaricidal properties. Alternatively the compounds of the invention can be used in sequence with the other active ingredient.

The diluent or carrier in the composition of the invention can be a solid or a liquid optionally in association with a surface-active agent, for example a dispersing agent, emulsifying agent or wetting agent. Suitable surface-active agents include anionic compounds such as a carboxylate, for example a metal carboxylate of a long chain fatty acid; an N-acylsarcosinate; mono- or di-esters of phosphoric acid with fatty alcohol ethoxylates or salts of such esters; fatty alcohol sulphates such as sodium dodecyl sulphate, sodium octadecyl sulphate or sodium cetyl sulphate; ethoxylated fatty alcohol sulphates; ethoxylated alkylphenol sulphates; lignin sulphonates; petroleum sulphonates; alkyl-aryl sulphonates such as alkyl-benzene sulphonates or lower alkylnaphthalene sulphonates, e.g. butyl-naphthalene sulphonate; salts of sulphonated naphthalene-formaldehyde condensates; salts of sulphonated phenol-formaldehyde condensates; or more complex sulphonates such as the amide sulphonates, e.g. the sulphonated condensation product of oleic acid and N-methyl taurine or the dialkyl sulphosuccinates, e.g. the sodium sulphonate of dioctyl succinate. Nonionic agents

include condensation products of fatty acid esters, fatty alcohols, fatty acid amides or fatty-alkyl- or alkenyl-substituted phenols with ethylene oxide, fatty esters of polyhydric alcohol ethers, e.g. sorbitan fatty acid esters, condensation products of such esters with ethylene oxide, e.g. polyoxyethylene sorbitan fatty acid esters, block copolymers of ethylene oxide and propylene oxide, acetylenic glycols such as 2,4,7,9-tetramethyl-5-decyn-4,7-diol, or ethoxylated acetylenic glycols.

Examples of a cationic surface-active agent include, for instance, an aliphatic mono-, di-, or polyamine as an acetate, naphthenate or oleate; an oxygen-containing amine such as an amine oxide or polyoxyethylene alkylamine; an amide-linked amine prepared by the condensation of a carboxylic acid with a di- or polyamine; or a quaternary ammonium salt.

The compositions of the invention can take any form known in the art for the formulation of agrochemicals, for example, a solution, a dispersion, an aqueous emulsion, a dusting powder, a seed dressing, a fumigant, a smoke, a dispersible powder, an emulsifiable concentrate or granules. Moreover it can be in a suitable form for direct application or as a concentrate or primary composition which requires dilution with a suitable quantity of water or other diluent before application.

An emulsifiable concentrate comprises a compound of the invention dissolved in a water-immiscible solvent which is formed into an emulsion with water in the presence of an emulsifying agent.

A dusting powder comprises a compound of the invention intimately mixed and ground with a solid pulverulent diluent, for example, kaolin.

A granular solid comprises a compound of the invention associated with similar diluents to those which may be employed in dusting powders, but the mixture is granulated

by known methods. Alternatively it comprises the active ingredient absorbed or adsorbed on a pre-granular diluent, for example, Fuller's earth, attapulgite or limestone grit.

Wettable powders, granules or grains usually comprise the active ingredient in admixture with a suitable surfactant and an inert powder diluent such as china clay.

Another suitable concentrate is a flowable suspension concentrate which is formed by grinding the compound with water or other liquid, a wetting agent and a suspending agent.

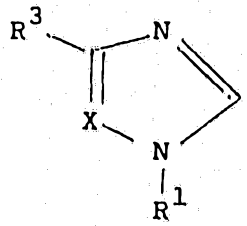
The concentration of the active ingredient in the composition of the present invention, as applied to plants is preferably within the range of 0.01 to 3.0 per cent by weight, especially 0.01 to 1.0 per cent by weight. In a primary composition the amount of active ingredient can vary widely and can be, for example, from 5 to 95 per cent by weight of the composition.

In the method of the invention the compound is generally applied to seeds, plants or their habitat. Thus, the compound can be applied directly to the soil before, at or after drilling so that the presence of active compound in the soil can control the growth of fungi which may attack seeds. When the soil is treated directly the active compound can be applied in any manner which allows it to be intimately mixed with the soil such as by spraying, by broadcasting a solid form of granules, or by applying the active ingredient at the same time as drilling by inserting it in the same drill as the seeds. A suitable application rate is within the range of from 0.05 to 20 kg per hectare, more preferably from 0.1 to 10 kg per hectare.

Alternatively the active compound can be applied directly to the plant by, for example, spraying or dusting either at the time when the fungus has begun to appear on the plant or before the appearance of fungus as a

protective measure. In both such cases the preferred mode of application is by foliar spraying. It is generally important to obtain good control of fungi in the early stages of plant growth as this is the time when the plant can be most severely damaged. The spray or dust can conveniently contain a pre- or post-emergence herbicide if this is thought necessary. Sometimes, it is practicable to treat the roots of a plant before or during planting, for example, by dipping the roots in a suitable liquid or solid composition. When the active compound is applied directly to the plant a suitable rate of application is from 0.01 to 10 kg. per hectare, preferably from 0.05 to 5 kg per hectare.

The compounds of the invention may be prepared, in known manner, in a variety of ways. Where R^2 is cyano, the compounds can be prepared by cyanation of a compound of formula II



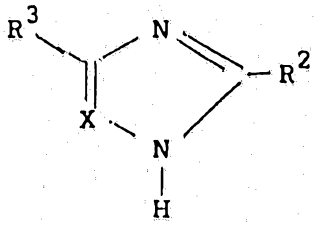
(II)

This can be achieved for instance by reacting the compound of formula II with a compound CN-Z, where Z is a leaving group, such as cyano or p-tosyl. This reaction is generally carried out in the presence of a stong base and preferably an alkyl metal, such as butyllithium. The cyanation can also be carried out by (i) formylating a compound of formula II, (e.g. using dimethylformamide in the presence of strong base, such as butyllithium) to give a compound of formula I in which R^2 is formyl, (ii) treating this compound with hydroxylamine and (iii) subsequently dehydrating the oxime so obtained to give the desired compound of formula I, in which R^2 is cyano. Dehydration may be achieved using a reagent such as

trifluoroacetic anhydride or a chloroformate ester, under alkaline conditions. In the latter case an ester group may be substituted onto the 1-position.

Compounds, where R^2 is $-CH=N-OR^{10}$, can be obtained in a similar manner to steps (i) and (ii) above, using compound of formula H_2N-OR^{10}.

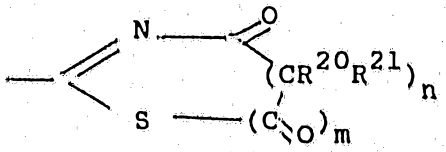
Compounds of the invention can also be obtained by reacting a compound of formula III



(III)

in which R^2 is as defined above, except for thiocarbamoyl, with a compound of formula R^1Q, where Q is a leaving group, such as halogen, especially chlorine.

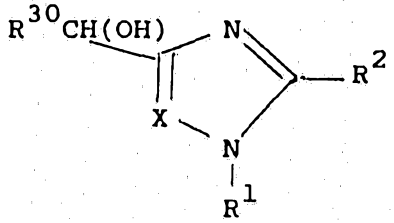
Compounds of formula I, where R^2 is cyano, can be modified in known manner to give compounds where R^2 is thiocarbamoyl, by reaction with hydrogen sulphide and if desired modifying this group in known manner to give compounds where R^9 is not hydrogen or to give compounds where R^2 is



These reactions are usually carried out using a suitable acyl halide or isocyanate, for instance as described in EP 219192.

Compounds of formula I, where X is CR^4, where R^4 is other than hydrogen may also be obtained in known manner by reacting the compound where R^4 is hydrogen with a compound, R^4-W, where W is a leaving group, such as halo (especially iodo or chloro) or p-tosyl. This reaction is generally carried out in the presence of a strong base, preferably an alkyl metal, such as butyllithium.

Compounds where R^3 is a alkanoyl or aroyl group can be prepared by oxidising a compound of formula IV

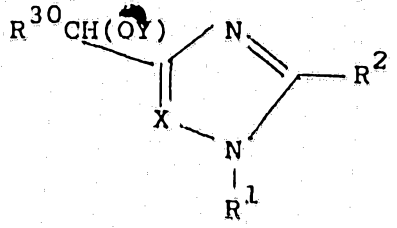


(IV)

in which R^{30} is the alkanoyl or aroyl group minus the carbonyl, e.g. using a dichromate, such as pyridinium dichromate.

Compounds of formula II or III are known or can be obtained in a variety of known ways, for instance using methods as described for the preparation of starting materials hereinafter.

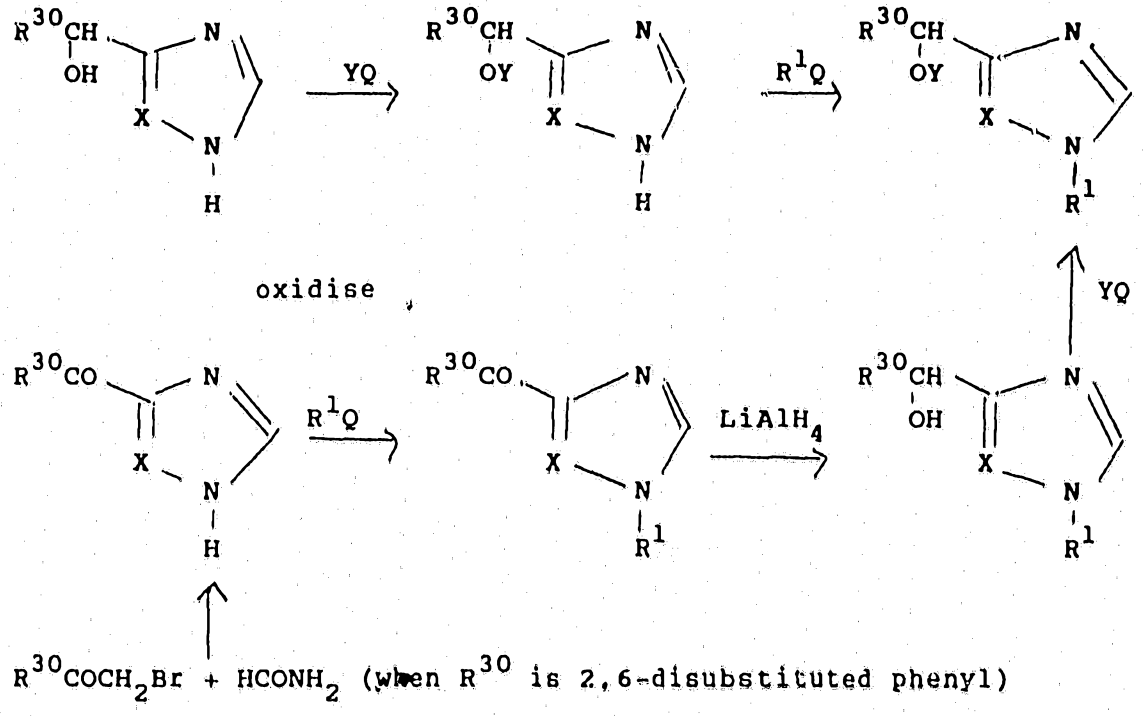
Compounds of formula IV can be obtained by removing the protecting group from a compound of formula V



(V)

where Y is a protecting group such as a trialkylsilyl.

Compounds of formula V can in their turn be prepared by cyanation etc, as described above, of the compounds of formula V in which R^2 is hydrogen. These compounds can in their turn be prepared according to the following reaction scheme.



The invention is illustrated in the following Examples. Structures of isolated novel compounds were confirmed by elemental and/or other appropriate analyses. Temperatures are in °C and are uncorrected.

Example 1

a) To 4-(2,4-Dichlorophenyl)-1-(dimethylsulphamoyl)-imidazole, (8.0 g) in tetrahydrofuran (125 ml), maintained under a dry nitrogen atmosphere at -60° was added 2.6M butyllithium in hexane (10.57 ml). The solution was stirred and after a short time dimethylformamide (2.9 ml) was added. The mixture was warmed slowly to room temperature and then added to dilute hydrochloric acid. The product was extracted with ethyl acetate and the extract washed with water, dried, the solvent evaporated and the solid residue recrystallised from toluene to give 4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)-2-formyl-imidazole, m.p. 148-157° (dec.).

b) This product (6.26g) was added to a mixture of

hydroxylamine hydrochloride (1.26 g) and sodium acetate (1.48 g) in 95% ethanol (45 ml), followed by addition of tetrahydrofuran (20 ml). The mixture was heated on a steam bath for two minutes, filtered and the solid remaining was washed with acetone. The filtrate and washings were combined and the solvent evaporated under reduced pressure. The residue was washed with water and dried over phosphorus pentoxide to give 4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)-2-(hydroxyiminomethyl)imidazole, m.p. 145-157° (dec.).

c) To this product (6.26g) in pyridine (4.24 ml) and dioxane (8 ml) was added trifluoroacetic anhydride (1.88 ml) with stirring and cooling on an ice bath. After stirring for 5 hours at room temperature the mixture was added to ice-water and the solid collected by filtration. This was recrystallised from toluene to give 2-cyano-4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)imidazole, m.p. 161-4° (dec.). (Compound 1)

Example 2

This example illustrates an alternative method of preparing compound 1.

To a solution of 4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)imidazole (1.62 g) in dry tetrahydrofuran (30 ml) at -70° under a dry nitrogen atmosphere was added 2.5M butyllithium in hexane (2 ml). After stirring for 15 mins, a solution of redistilled tosyl cyanide in dry tetrahydrofuran (7 mmol) was added and the reaction flask placed in an ice bath. The mixture was stirred for 30 mins, poured into water and the precipitate filtered and recrystallised to give compound 1.

Example 3

Hydrogen sulphide was bubbled through a stirred suspension of compound 1 (1.73 g) from Example 1 in triethylamine (0.7 ml) and pyridine (10 ml) with stirring and cooling in an ice bath for 30 minutes. The mixture was

poured into ice-water and the solid collected by filtration, then dried over phosphorus pentoxide to give 4-(2,4-dichlorophenyl)-1-dimethylsulphamoyl-2-thiocarbamoylimidazole, m.p. 184-7°. (Compound 2)

Example 4

In a similar manner to that described in Example 1, there was obtained, in turn

a) 1-(dimethylsulphamoyl)-2-formyl-4-phenylimidazole; and

b) 1-(dimethylsulphamoyl)-2-(hydroxyiminomethyl)-4-phenylimidazole, m.p. 165-170° (dec.). A slurry of this product (0.882 g) in dry tetrahydrofuran (20 ml) was added to sodium hydride (90 mg of 80% suspension in oil) in dry tetrahydrofuran. After 1.5 hours at room temperature ethyl chloroformate (0.285 ml) was added and the mixture stirred overnight. The mixture was then added to ethyl acetate and worked up in conventional manner to give a viscous oil. This was purified by flash column chromatography to give 2-cyano-1-ethoxycarbonyl-3-phenylimidazole, m.p. 109-111°. (Compound 3)

Example 5

Dimethylsulphamoyl chloride (0.54 ml) was added to a mixture of 2-cyano-4-(2-nitrophenyl)-1H-imidazole (0.865 g) and sodium hydride (150 mg of 80% in oil). The mixture was heated under reflux for one hour and then poured into ice/water and extracted with ethyl acetate. The extract was washed with brine, dried and evaporated. The residual oil slowly solidified and was washed with ether and filtered to give 2-cyano-1-(dimethylsulphamoyl)-4-(2-nitrophenyl)imidazole, m.p. 111-4°. (Compound 4)

Example 6

Butyllithium (2.5M; 2ml) was added to a solution of 1-(dimethylsulphamoyl)-4-(2,4,6-trichlorobenzoyl)imidazole (1.77 g) in tetrahydrofuran (25 ml), maintained at -78°. The solution was stirred for 15 minutes and cyanogen was bubbled in allowing the temperature to rise to 10°. The

mixture was evaporated and purified by column chromatography, followed by trituration with diisopropyl ether to give 2-cyano-1-(dimethylsulphamoyl)-4-(2,4,6-trichlorobenzoyl)imidazole, m.p. 142-50°. (Compound 5).

Example 7

A solution of 2-cyano-4-(2,4-dichlorophenyl)imidazole (1.0 g) in tetrahydrofuran (30 ml) was treated with sodium hydride (0.16 g of 80% dispersion in oil) at 0°. A solution of tetramethylphosphorodiamidic chloride (0.89 g) in tetrahydrofuran (10 ml) was added dropwise and the mixture stirred for 1½ hours under nitrogen. It was then poured into water, stirred, and the precipitate filtered, dried and recrystallised from toluene/hexane to give 1-[bis(dimethylamino)phosphinyl]-2-cyano-4-(2,4-dichlorophenyl)imidazole, m.p. 140.5-1°. (Compound 6).

Example 8

Oxalyl chloride (0.23 ml) was added to a stirred solution of compound 2 (0.95 g) in acetone (15 ml) kept cool in an ice-salt bath. The yellow precipitate, which formed, was filtered, washed with acetone and dried to give 2-[4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)-imidazol-2-yl]thiazole-4,5-dione, m.p. 158°, (dec.). (Compound 7)

Example 9

Butyllithium (1.67 ml of 2.5M solution in hexane) was added under a dry nitrogen atmosphere to a solution of compound 1 (1.38 g) in tetrahydrofuran (20 ml) maintained at -78°. After stirring at this temperature for 20 minutes, methyl iodide (0.8 ml) was added. The mixture was allowed to warm to room temperature and then poured into water. The aqueous phase was extracted with ethyl acetate and the extract dried and evaporated. The residual oil was purified by flash column chromatography using an ethyl

acetate/hexane mixture as eluent, to give 2-cyano-4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)-5-methylimidazole, m.p. 92-5°. (Compound 8)

Example 10

This example illustrates an alternative method of preparing compound 5.

2-Cyano-1-(dimethylsulphamoyl)-4-(2,4,6-trichloro-α-hydroxybenzyl)imidazole (15.7 g) was dissolved in dichloromethane and pyridinium dichromate (28.9 g) and silica gel (230-400 mesh; 58 g) added and the mixture stirred for 11 hours. The mixture was filtered and the precipitate washed with dichloromethane. The filtrate and washings were evaporated under reduced pressure and the residue washed with diisopropyl ether and dried under reduced pressure to give 2-cyano-1-(dimethylsulphamoyl)-4-(2,4,6-trichlorobenzoyl)imidazole, m.p. 142-151°. (Compound 5).

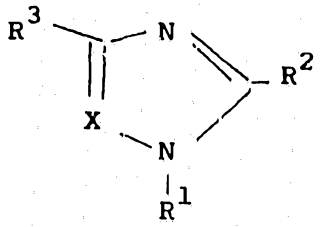
Example 11

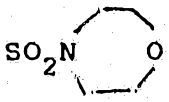
1-(Dimethylsulphamoyl)-2-formyl-4-(2,3,4-trichlorophenyl)imidazole was reacted with O-allylhydroxylamine hydrochloride in a similar manner to the reaction with hydroxylamine hydrochloride described in Example 1, to give 2-allyloxyiminomethyl-1-(dimethylsulphamoyl)-4-(2,3,4-trichlorophenyl)imidazole, m.p. 87-88°. (Compound 9)

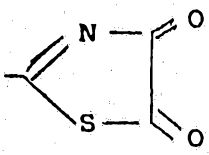
The starting material was prepared in a similar manner to that described in Example 1.

Example 12

In a similar manner to that described in one of the previous Examples, the following compounds were obtained: The Example method followed is given in the column headed "P". In the table Ph = phenyl, which may be substituted as indicated; e.g. 3,4-Cl_2-Ph- means 3,4-dichlorophenyl.



Cpd no.	R^3	R^1	X	R^2	P	m.p. (°)
10	4-Br-Ph-	SO_2NMe_2	CH	CN	2	158-74
11	4-MeO-Ph-	SO_2NMe_2	CH	CN	2	152-4
12	4-CF_3-Ph-	SO_2NMe_2	CH	CN	2	166-8
13	3,4-Cl_2-Ph-	SO_2NMe_2	CH	CN	2	145-6
14	2,4-Cl_2-3-CN-Ph-	SO_2NMe_2	CH	CN	2	197-201
15	4-Cl-Ph-	SO_2NMe_2	CH	CN	2	170-1
16	3,4-Cl_2-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	184-6
17	4-CF_3-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	183-5
18	4-F-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	191-3
19	4-Cl-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	194-5
20	2,3,4-Cl_3-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	161-3
21	4-Br-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	188-9
22	4-CF_3-	SO_2NMe_2	CH	CN	2	86-7
23	2,4-F_2-Ph-	SO_2NMe_2	CH	CN	2	161-3
24	3-CF_3-Ph-	SO_2NMe_2	CH	CN	2	144-5
25	2-CF_3-Ph-	SO_2NMe_2	CH	CN	2	76-77.5
26	2,4-Cl_2-Ph-		CH	CN	2	169-70
27	2,6-Cl_2-PhCO-	SO_2NMe_2	CH	CN	2	166-7
28	5-Cl-thien-2-yl	SO_2NMe_2	CH	CN	2	138-40
29	2,4,6-Me_3-PhCO-	SO_2NMe_2	CH	CN	2	149-51
30	4-F-Ph-	SO_2NMe_2	CH	CN	2	172-7
31	2,3,4-Cl_3-Ph-	SO_2NMe_2	CH	CN	2	143-8
32	Ph-	SO_2NMe_2	N	CN	1	107-11
33	2,5-Cl_2-thien-3-yl	SO_2NMe_2	CH	CN	2	118-9
34	Ph-	SO_2NMe_2	CH	CN	1	104-5

Cpd no.	R^3	R^1	X	R^2	P	m.p. (°)
35	4-NO_2-Ph-	SO_2NMe_2	CH	CN	5	202-8
36	2-CF_3-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	113-5
37	3-CF_3-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	193-5
38	2,4-F_2-Ph-	SO_2NMe_2	CH	$CSNH_2$	3	168-70
39	2,3,4-Cl_3-Ph-	$P(=O)(NMe_2)_2$	CH	CN	7	143-8
40	2,4,5-Cl_3-Ph-	SO_2NMe_2	CH	CN	2	182-4
41	2-Cl-Ph-	SO_2NMe_2	CH	CN	2	148-9
42	2,4-Cl_2-Ph-	SO_2NMe_2	CH	CN	2	172-3
43	2,4-Me_2-Ph-	SO_2NMe_2	CH	CN	2	100-1
44	2-F-Ph-	SO_2NMe_2	CH	CN	2	126-8
45	3-F-Ph-	SO_2NMe_2	CH	CN	2	122-3
46	Me	SO_2NMe_2	CH	CN	2	88-90
47	3-Cl-Ph-	SO_2NMe_2	CH	CN	2	118-20
48	H	SO_2NMe_2	C(Me)	CN	2	118-20
49	4-CN-Ph-	SO_2NMe_2	CH	CN	2	226-33
50	4-Cl-PhN(Me)SO_2-	SO_2NMe_2	CH	CN	2	194-7
51	Me_2NSO_2-	SO_2NMe_2	CH	CN	2	154-5
52	4-MeO-Ph-	SO_2NMe_2	N	CN	2	135-6
53	4-CN	SO_2NMe_2	CH	CN	2	110-2
54	2,3,4-Cl_3-Ph-	SO_2NMe_2	CH	CH=NOMe	11	182-4
55	2,4-Me_2-Ph-	SO_2NMe_2	N	CN	2	121-3
56	$Ph_2C(OEt)$-	SO_2NMe_2	C(Me)	CN	2	165-6
57	2,4-Cl_2-Ph-	SO_2NMe_2	C(CN)	CN	9[1]	105
58	4-Br-Ph-	SO_2NMe_2	C(COOEt)	CN	9[2]	110-3
59	4-Cl-Ph-CH=CH-	SO_2NMe_2	CH	CN	2	93-4
60	H	SO_2NMe_2	C(Ph)	CN	2	98-106
61	PhCO-	SO_2NMe_2	CH	CN	10	145-7
62	2,3,4-Cl_3-Ph-	SO_2NMe_2	CH		8	157-9
63	PhS-	SO_2NMe_2	CH	CN	2	83.5-84

Notes: [1] = reaction carried out using p-tosyl cyanide
[2] = reaction carried out using ethyl chloroformate

Example 13

Acetyl chloride (0.97 g) was added dropwise to a mixture of a solution of compound 20 (2.56 g) in acetone (20 ml) with stirring and cooling. The mixture was heated under reflux for 30 minutes and then evaporated. The residue was extracted with dichloromethane and the extract washed with water, dried, evaporated and purified by column chromatography to give 2-(N-acetylthiocarbamoyl)-1-(dimethylsulphamoyl)-4-(2,3,4-trichlorphenyl)imidazole, m.p. 132-134°. (Compound 64).

Example 14

A mixture of compound 63 (0.31 g) in dichloromethane (25 ml) and m-chloroperbenzoic acid (0.54 g of 80%) was stirred at room temperature for 3 hours. The mixture was extracted with water and then aqueous sodium carbonate, and the organic phase dried and evaporated. The residue was recrystallised from toluene/hexane to give 2-cyano-4-(phenylsulphonyl)-1-(dimethylsulphamoyl)imidazole, m.p. 164-165.5° (dec.). (Compound 65)

Preparation of starting materials

The following methods illustrate typical ways of preparing starting materials.

Method A

(as used for starting material of Example 1)

A mixture of 4-(2,4-dichlorophenyl)-1H-imidazole (12.14 g), triethylamine (13 ml) and dimethylsulphamoyl chloride (7.5 ml) in tetrahydrofuran (50 ml) was stirred for 18 hours at room temperature and then heated under reflux until almost no starting material remained. The organic phase was diluted with ethyl acetate (300 ml), washed with dilute hydrochloric acid, water and aqueous

sodium bicarbonate, dried and the solvent evaporated to give an oily solid which was treated to flash column chromatography and the product recrystallised to give 4-(2,4-dichlorophenyl)-1-(dimethylsulphamoyl)imidazole, m.p. 169-171°.

Method B

(as used for starting material of Compounds 4 and 35)

A mixture of compound 34 (6.72 g) and nitronium tetrafluoroborate (98 ml of 0.5M in sulpholane) in sulpholane 25 ml was stirred for 16 hours at room temperature. It was then added to ice/water and filtered. The solid from the filtration was washed with water and dried. This was treated to flash column chromatography (using ethyl acetate/hexane as eluent to give 2-cyano-4-(4-nitrophenyl)-1H-imidazole, m.p. 230° (dec.). The filtrate was extracted with ethyl acetate and the extract worked up in usual manner and the residue treated to flash column chromatography (using ethyl acetate/hexane as eluent to give 2-cyano-4-(2-nitrophenyl)-1H-imidazole, m.p. 166-170° (dec.).

Method C

(as used for starting material of Example 6)

A mixture of 2-bromo-2',4',6'-trichloroacetophenone (42 g) and formamide (200 ml) was heated under reflux for 2 hours, with stirring. It was cooled, poured into 2M hydrochloric acid, heated on a steam bath, filtered and the filtrate made basic with ammonia. The solid was collected and dried under reduced pressure to give crude 4-(2,4,6-trichlorobenzoyl)imidazole. This was then treated with dimethylsulphamoyl chloride in a similar manner to that described in Example 1, to give 1-(dimethylsulphamoyl)-4-(2,4,6-trichlorobenzoyl)imidazole.

Method D

(as used for starting material of Example 7)

A solution of compound 1 (4 g) in tetrahydrofuran (40 ml) and water (15 ml) was heated under reflux with in turn aqueous sodium hydroxide and methanolic potassium carbonate until tlc showed that the reaction was complete. The reaction mixture was neutralised with hydrochloric acid and the precipitate collected, washed with water, dried and purified by flash column chromatography (using light petroleum (b.p. 60-80°)/ethyl acetate (3:1) as eluent) to give 2-cyano-4-(2,4-dichlorophenyl)imidazole,

Method E

(as used for starting material of Example 10)

1-(Dimethylsulphamoyl)-4-(2,4,6-trichlorobenzoyl)imidazole (2.6 g) in dry tetrahydrofuran (15 ml) was added over 30 seconds to lithium aluminium hydride (110 mg) in dry tetrahydrofuran (15 ml) with cooling. The mixture was stirred at 5° for 30 minutes . Water (0.110 ml), aqueous sodium hydroxide (15%; 0.110 ml) and water (0.330 ml) were added and the mixture stirred for 15 minutes at room temperature. The mixture was filtered and the precipitate washed with tetrahydrofuran. The filtrate and washings were evaporated under reduced pressure and the residue washed with diisopropyl ether and dried under reduced pressure to give 1-(dimethylsulphamoyl)-4-(2,4,6-trichloro-α-hydroxybenzyl)imidazole, as an off white solid. To this product (1.92 g) in dry tetrahydrofuran (40 ml) was added sodium hydride (170 mg; 80% in oil), with stirring, under nitrogen. Trimethylsilyl chloride (0.7 ml) was addded to this mixture at 30° to give the trimethyl silyl ether. The mixture was then cooled to - 70° and butyllithium (2.2 ml; 2.5M in hexane) added. The mixture was stirred at -78° for 30 minutes, dimethylformamide (0.5 ml) added and the mixture allowed to warm to room temperature. The mixture was poured into ice/water,

stirred for 30 minutes and the solid collected and dried to give crude 1-(dimethylsulphamoyl)-2-formyl-4-[(2,4,6-trichloro-α-(trimethylsilyloxy)benzyl]imidazole. This was then then treated with hydroxylamine hydrochloride in a similar manner to step (b) in Example 1, to give crude 1-(dimethylsulphamoyl)-2-(hydroxyimino-methyl)-4-(2,4,6-trichloro-α-hydroxybenzyl)imidazole. This was then treated with trifluoroacetic anhydride in a similar manner to step (c) in Example 1, to give crude 2-cyano-1-(dimethylsulphamoyl)-4-(2,4,6-trichloro-α-hydroxybenzyl)imidazole, as an off white solid.

Method F

(as used for starting material of Compound 56)

Ethyl 5-methylimidazole-4-carboxylate was converted by a Grignard reaction, using phenylmagnesium bromide, to 4-[ethoxy(diphenyl)methyl]-5-methylimidazole, m.p. 205°. This was then treated with dimethylsulphamoyl chloride in a similar manner to that described in Example 1, to give 1-dimethylsulphamoyl-4-[ethoxy(diphenyl)methyl)-5-methyl-imidazole, m.p. 150°.

Method G

(as used for starting material of Compound 50)

Imidazole-4-sulphonyl chloride was reacted with 4-chloro-N-methylaniline to give 4-[N-(4-chlorophenyl)-N-methylsulphamoyl]imidazole, m.p. 217-9°. This was then treated with dimethylsulphamoyl chloride in a similar manner to that described in Example 1, to give 1-dimethylsulphamoyl-4-[N-(4-chlorophenyl)-N-methyl-sulphamoyl]imidazole, m.p. 131.5-132°.

Method H

(as used for starting material of Compound 60)

N-Benzylidene-N',N'-dimethylsulphamide (6.36 g) was stirred with p-toluenesulphonylmethyl isocyanide (9.75 g) and potassium carbonate (8.28 g) in methanol and dimethoxyethane for 18 hours. The mixture was filtered and

the filtrate evaporated and recrystallised from ethyl acetate/ hexane to give crude 1-dimethylsulphamoyl-5-phenylimidazole.

For Compound 14, the starting material was prepared by reacting the starting material used in Example 1 with two moles of butyllithium/dimethylformamide to give 4-(2,4-dichloro-3-formylphenyl)-1-(dimethylsulphamoyl)-2-formylimidazole which in turn was reacted with two moles of hydroxylamine hydrochloride to give 4-[2,4-dichloro-3-(hydroxyiminomethyl)phenyl]-1-(dimethylsulphamoyl)-2-(hydroxyiminomethyl)imidazole. This was reacted with two equivalents of trifluoroacetic anhydride to give the desired product.

For Compound 63, the starting material was prepared according to the method described in Tetrahedron, 1986, 42, 2351-8.

Other starting materials were either known or prepared by one of these or similar methods. Generally they were not purified so no physical data are given.

Test Example

The compounds of the invention were subjected to various tests.

a) Foliar tests

Compounds are assessed for activity against one or more of the following:

Erysiphe graminis: barley powdery mildew (EG)

Plasmopara viticola: vine downy mildew (PV)

Pyricularia oryzae: rice blast (PO)

Botrytis cinerea: grey mould of tomato (BC)

Venturia inaequalis: apple scab (VI)

Phytophthora infestans: late tomato blight (PI)

Aqueous solutions or dispersions of the compounds at the desired concentration, including a wetting agent, were sprayed onto the appropriate plant and then inoculated by spraying with spore suspensions of the fungi or by dusting or shaking diseased material over the treated plants for the Erysiphe spp.. Plants were then kept under controlled environment conditions suitable for maintaining plant growth and development of the disease. After an appropriate time, the degree of infection of the leaf surface was visually estimated.

Compounds were considered active if they gave greater than 50% control of the disease at a concentration of 125 ppm (w/v) or less.

(b) Soil pathogen test

In these tests compounds were assessed for activity against Rhizoctonia solani (RS)

Flasks containing maize meal/sand were inoculated with the test fungus and then incubated. The maize meal/sand cultures were used to infest potting compost which was then put into plastic pots. Aqueous solutions or dispersions of the compounds, including a wetting agent, were added to the pots to give a desired concentration of compound in each pot. Control pots were set up by adding

similar solutions or dispersions without the test compound Immediately after application of the test compound each pot was sown with a number of cabbage seeds. The seeds were covered with treated infested soil and the pots incubated under controlled environment conditions suitable for plant growth and development of the disease. The number of emerged cabbage seedlings is counted and percentage disease control calculated by comparison with the untreated infested pots.

Compounds were considered active if they gave greater than 50% control of the disease at a concentration of 100 parts by weight of compound or less per million parts by volume of soil.

Activities were demonstrated as follows (+ = active).

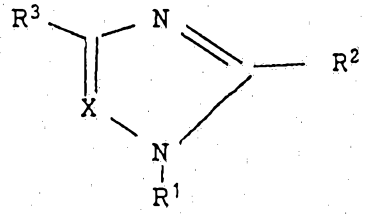
Compound No	EG	PO	PI	PV	RC	VI	RS
1	+		+	+			
2			+	+			
3				+	+		
4	+		+				
5			+	+			
6			+				
7	+			+			
8			+			+	
9			+				
10			+	+			
11						+	
12			+				
13			+				
14	+		+	+			
15		+	+				
16			+	+			
17			+				

Compound No	EG	PO	PI	PV	BC	VI	RS
18			+				
19		+	+				
20			+	+		+	
21		+	+				
22			+	+			+
23			+				
24			+	+		+	
25			+			+	
26					+		
27			+				
28		+	+		+		
29	+		+				
30		+	+	+			
31	+		+	+		+	
32				+			
33			+				
34			+				
35	+		+				
36			+				
37			+				
38			+				
39			+				
40			+				
41			+				
42			+				
43			+				
44			+				
45			+				
46		+					
47			+				
48		+					
49			+				

Compound No	EG	PO	PI	PV	BC	VI	RS
50			+				
51			+				
52			+				
53			+				
54			+			+	
55			+				
56			+				
57			+				
58			+				
59			+				
60			+				
63			+				
65			+				

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:-

1) A compound of formula I



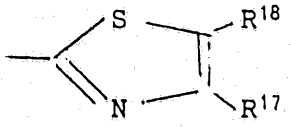
(I)

in which

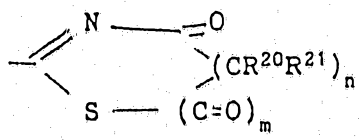
X is CR^4 or N;

R^1 is $-SO_2R^5$, $-\overset{\overset{R^6}{|}}{\underset{\underset{Z}{\|}}{P}}-R^7$ or $-COR^8$

R^2 is CN, $-\overset{\overset{S}{\|}}{C}-NH-R^9$, $-CH=N-OR^{10}$,



or



R^3 and R^4, may be the same or different and are alkyl, cycloalkyl, cycloalkenyl, alkenyl, or alkynyl, ~~or amino,~~ (all of which are optionally substituted by halogen, hydroxy, alkoxy or aryl), hydrogen, halogen, hydroxy, cyano, nitro, acyl, $R^{11}SO_p$, $R^{12}O$ or aryl;

R^5 is aryl, optionally substituted alkyl or optionally substituted amino;

R^6 and R^7, may be the same or different and are amino, alkoxy or alkylthio, each of which is optionally substituted;

R^8 has the same meaning as R^5 or can be alkoxy, alkenyloxy, alkynyloxy or alkylthio, each of which is optionally substituted, or is aryloxy;

R^9 is hydrogen, optionally substituted alkyl, alkenyl, alkynyl, alkoxycarbonyl, acyl, aryl or cycloalkyl;

R^{10} is hydrogen, optionally substituted alkyl, alkenyl or alkynyl;

R^{11} is alkyl, cycloalkyl, cycloalkenyl, alkenyl, alkynyl, all of which are optionally substituted, or is aryl;
R^{12} has the same meaning as R^{11} or is acyl;
R^{17} is hydrogen, alkyl, alkoxycarbonyl, or aryl and R^{18} is hydrogen or alkyl or R^{17} and R^{18} together with the carbons to which they are attached, form a benzo ring;
R^{20} and R^{21} may be the same or different and are hydrogen or alkyl;
Z is oxygen or sulphur;
m is 0 and n is 1 or 2 or m is 1 and n is 0 or 1; and
p is 0 or 1,
with the proviso that R^3 and R^4 cannot both be chlorine, and in which

a) any substituent on any alkyl, cycloalkyl, cycloalkenyl, alkenyl, alkoxy or alkynyl containing group is selected from halogen, hydroxy, alkoxy and aryl,

b) any aryl group is phenyl, optionally substituted, by halogen, alkyl, alkoxy, nitro, cyano, $-COR^8$, optionally substituted sulphamoyl, optionally substituted amino, alkyl-SO_q, or aryl-SO_q, where q is 0, 1 or 2, and any alkyl or alkoxy group is optionally substituted,

c) any acyl group is the residue of both carboxylic and sulphonic acids and includes the groups $R^{13}(O)_rCO$ and $R^{13}SO_2$, where R^{13} has the same meaning as R^{11}, or is optionally substituted amino and r is 0 or 1,

d) any amino or sulphamoyl group is optionally substituted by one or more of the groups R^{11}, acyl, optionally substituted amino, hydroxy or optionally substituted alkoxy.

2. A compound according to claim 1, in which R^1 is dimethylsulphamoyl.

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3. A compound according to claim 1 or 2, in which R^2 is cyano or thiocarbamoyl.

4. A compound according to any one of claims 1 to 3, in which R^3 is phenyl or benzoyl, optionally substituted by up to three groups, selected from halogen, alkyl (especially methyl), trifluoromethyl, nitro and alkoxy (especially methoxy).

5. A fungicidal composition comprising a compound as claimed in any one of the preceding claims, in admixture with an agriculturally acceptable diluent or carrier.

6. A method of combating phytopathogenic fungi which comprises applying to the fungus or its locus a compound claimed in any one of claims 1 to 5.

7. A compound according to claim 1 or a fungicidal method involving a said compound substantially as hereinbefore described with reference to the Examples.

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