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Industria y Comercio
SUPERINTENDENCIA

11-11-07

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División de Nuevas Creaciones

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SOLICITUD

PATENTE DE INVENCION

07-29888

21. EXPEDIENTE No.

54. TITULO: "MEZCLAS FUNGICIDAS DE DERIVADOS DE TIOFENO"

51. CLASIFICACIÓN INTERNACIONAL: A01N 43/56

71. SOLICITANTE: E.I. DUPONT DE NEMOURS AND COMPANY

DOMICILIO: DELAWARE, ESTADOS UNIDOS DE AMÉRICA

74. APODERADO: ALVARO CORREA ORDOÑEZ
T.P.No.20.281

22. BOGOTA, D.C., 26 de marzo de 2007

F.W. 27-04-2007

Pake, Ceron

PETITORIO

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Industria y Comercio
SUPERINTENDENCIA

SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO

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Fec: 2007-05-25 18:44:40 Dep. 2020 NUECREACIONES

Tra. 11 PATENTE/VI

Ene. 378 FASE NACIONAL

Act. 411 PRESENTACION

Fec. 05-25

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FORMULARIO ÚNICO DE SOLICITUD DE PATENTE
PCT
ENTRADA EN FASE NACIONAL

①

SOLICITUD DE :

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Patente de Invención

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Patente de Modelo de Utilidad

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Capítulo I.

☐

Capítulo II.

②

SOLICITANTE
(71)

Nombre: EL DUPONT DE NEMOURS AND COMPANY

Dirección: 1007 MARKET STREET, WILMINGTON, Delaware 19858
Estados Unidos de América

Nacionalidad o Domicilio: Delaware, Estados Unidos de América

Lugar de Constitución: Delaware, Estados Unidos de América

Teléfono:

Fax:

E-Mail:

IDENTIFICACION

C.C.

☐

NIT

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C.E.

☐

Otro

☐

Cual

Número

③

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APODERADO

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IDENTIFICACION

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C.E.

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Otro

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Cual

Número

79.143.365 de
Usaquén

TP 20.281

④

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(72)

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⑤

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⑥

Título (54)

MEZCLAS FUNGICIDAS DE DERIVADOS DE TIOFENO

⑦

Clasificación Internacional (51)

A01N 43/56, A01N 37/50, A01N 43/53, A01N 43/76

⑧

Prioridad

SI

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27-SEP-2004

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Fecha 3/11/06; 3/11/06; 30/01/07; 12/01/07

Instrucciones para el diligenciamiento del presente formulario, ver reverso de esta página

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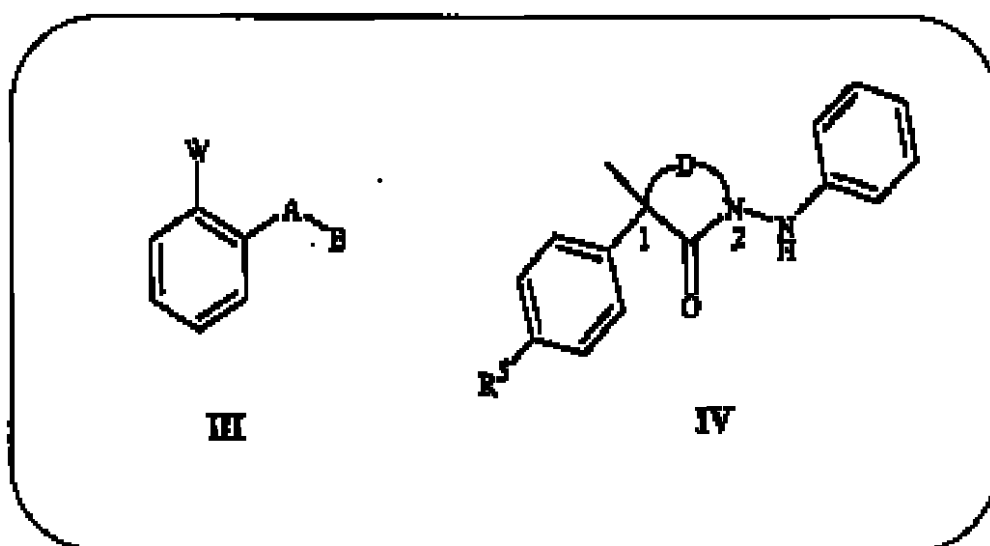
ANEXOS

00002 2

- ☒ Comprobante de pago de la tasa de presentación de la solicitud
- ☐ Documento que acredite la existencia y representación legal cuando el solicitante sea persona jurídica
- ☐ Poderes, si fuere el caso
- ☐ Documento de cesión del invento al solicitante o a su cesante
- ☐ Declaraciones
- ☒ Copia de la solicitud internacional. (Resumen, descripción, reivindicaciones, dibujos)
- ☒ Traducción de la solicitud internacional al castellano. (Resumen, descripción, reivindicaciones, dibujos)
- ☐ Copia del examen preliminar internacional efectuado por la Autoridad Internacional de Examen preliminar (IPEA)
- ☐ Traducción del examen preliminar internacional efectuado por la IPEA
- ☐ De ser el caso, copia del contrato de acceso
- ☐ De ser el caso, documento que acredite la licencia o autorización de uso de conocimientos tradicionales de las comunidades indígenas
- ☐ De ser el caso, certificado de depósito del material biológico
- ☐ Arto lineal 12 x 12
- ☐ De ser el caso, información sobre otras solicitudes de patente o tipos obtenidos en el extranjero por el mismo titular o su causante, relacionadas parcial o totalmente con la invención de esta solicitud
- ☐ De ser el caso, modificaciones efectuadas a la solicitud internacional.
- ☒ Otros: Extractos, Carta que sustenta modificación por el artículo 18, reporte de búsqueda, opinión escrita, notificación de recepción de documento de prioridad, modificación de la página 3 por la fórmula IV.

11

FIGURA CARACTERISTICA



12

Solicitud la concesión de la patente.

NOMBRE: ALVARO CORREA GARCIA

FIRMA:

G.E. 79.145.356 de Usaquén

T.P. 21.281

Instrucciones para la presentación de los anexos, ver reverso de esta página

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MAA-3

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SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO
Rad: 07-029888-00000-0000
Pec: 2007-03-26 15:44:40 Dep. 2000 NUECREACIONES
Tm. 11 PATENTEEYE
Ere: 379 FASENACIONAL
Act 411 PRESENTACION P0808:95

SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO

NIT : 000174087-2

RECIBO OFICIAL DE CAJA : 04 - 05,248
FECHA : NOVIEMBRE 3 DE 2004

0000 CONSIGNACION 0000

DEPOSITANTE	TIPO PAGO	BANCO	CUENTA	No. PAGO	FECHA PAGO	Gr. PAGO
ORLANDO LINARES	CONSIGNACION	BANCO POPULAR	050-00110-6	54663178	03/11/2004	126,285,300.00

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1 50003-01-06 REGISTRO DE CESION, CAMBIO MON	13 AJUROS Y ENSENAS COMERCIALES,MM	234,000.00
	TOTAL :	\$ 234,000.00

SON: DOSCIENTOS TREINTA Y CUATRO MIL PESOS

RESPONSABLE : _____

RECIBO DE CAJA APLICADO AL EXPEDIENTE No. _____

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SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO
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TR. 11 PATENTE Y
EVI. 378 FASE NACIONAL
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Fono: 95

SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO

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RECIBO OFICIAL DE CAJA : 04 - 85,249
FECHA : NOVIEMBRE 3 DE 2006

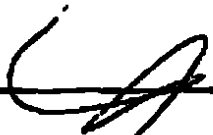
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ORLANDO LINARES	CONSIGNACION	BANCO POPULAR	050-00110-6	54663:78	03/11/2006	124,285,000.00

CONCEPTO

CANT. HISTORICO	CONCEPTO	TOTAL CONCEPTO
50005-01-04	REGISTRO DE CESION, CAMBIO NOM	
	13 MONEDAS Y ENSEÑAS COMERCIALES, MAR	234,000.00
	TOTAL :	234,000.00

SOM: DOSCIENTOS TREINTA Y CUATRO MIL PESOS

RESPONSABLE : 

RECIBO DE CAJA APLICADO AL EXPEDIENTE No. _____

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SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO
Rad: 07-029888-00000-0000
Per 2007-03-26 16:44:40 Dep. 2020 MUESTRACIONES
Tm 11 PATENTE/YS
Eve: 370 FASENACIONAL
Act 411 PRESENTACION
Pag 1 de 50

SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO

NIT : 630176707-2

RECIBO OFICIAL DE CAJA : 07 8.734
VA : DERO 10 DE 2007

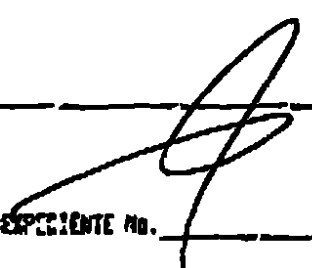
CONSOLIDACION

DEFICIENTE	TIPO PAGO	VALOR	CLIENTE	NO. PAGO	FECHA PAGO	Dr. PAGO
DELANO LITIGES	CONSOLIDACION	BANCO PARTICULAR	059 50150-3	55136719	30/01/2007	58,675.000

CONCEPTO

CONCEPTO	CONCEPTO	TOTAL CONCEPTO
1 50000-01-09 OTROS SERVICIOS DE PROPIEDADES	21 CERTIFICACIONES	11,000.00
TOTAL :		11,000.00

ENQUE MIL PESOS

RESPONSABLE : 
RECIBO DE CASH APLICADO AL EXPEDIENTE No. _____

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SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO
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NIT : 600176089-2

RECIBO OFICIAL DE CAJA : 7 - 2,799
 FECHA : ENERO 12 DE 2007

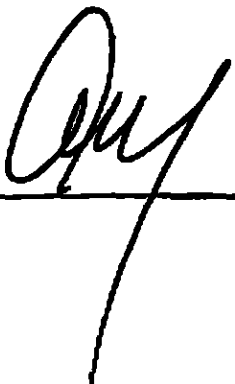
CONSISTENCIA

DEPOSITANTE	TIPO PAGO	BANCO	CUENTA	No. PAGO	FECHA PEG	Vr. PAGO
CRANCO LINARES	CONSISTENCIA	BANCO POPULAR	650-06110-6	64321256	11/01/2007	71,059,000.00

CONCEPTO

CANT. REENTISTICO	CONCEPTO	TOTAL CONCEPTO
1	50005-01-07 OTROS SERVICIOS DE PROPIEDAD I	3,000.00
	998 VALOR COMPLEMENTO TARIFA ADE ANTE	3,000.00
	TOTAL :	6,000.00

EN: TRES MIL PESOS

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6 April 2006 (06.04.2006)(10) International Publication Number
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(51) International Patent Classification:

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19898 (US).

(72) Inventors and

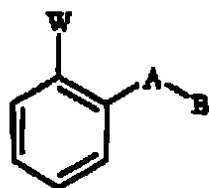
(73) Inventor/Applicant (for US only): FOXM, Stephen, Ray
(US/US); 508 Hemingway Drive, Hockessin, Delaware
19707 (US).(74) Agent: HEISKR, David, E.; E. I. DU PONT DE
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RECORDS CENTER, 4417 Lancaster Pike, Wilmington,
Delaware 19805 (US).(81) Designated States (unless otherwise indicated, for every
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AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN,
CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI,
GB, GD, GH, GM, GR, HU, ID, IL, IN, IS, JP, KH,
KG, KM, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, LY,
MA, MD, MG, MK, MN, MW, MX, MY, NA, NG, NI, NO,
NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK,
SI, SM, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ,
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RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA,
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Published:

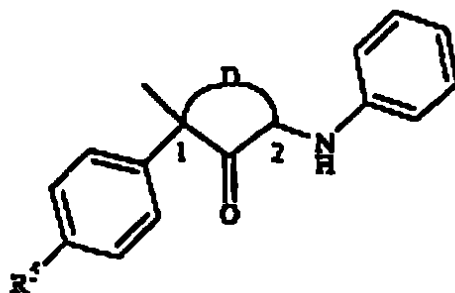
- with international search report
- with amended claims

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: FUNGICIDAL MIXTURES OF THIOPHENE DERIVATIVE



(III)



(IV)

(57) Abstract: Disclosed are fungicidal mixtures, compositions and methods for controlling plant diseases relating to combinations comprising (a) N-[2-(1,3-dimethylbutyl)-3-ethyl]-1-methyl-3-(trifluoromethyl)-1H-pyrazole-4-carboxamide (including all stereoisomers) or an agriculturally suitable salt thereof, and (b) at least one compound selected from the group consisting of compounds of Formula III or Formula IV which

act as the b:l complex of the fungal mitochondrial respiratory electron transfer site; (INSERT FORMULA III HERE) (INSERT FORMULA IV HERE) wherein W, A, B, D and R5 are disclosed in this specification, and agriculturally suitable salts thereof; and optionally (c) at least one compound selected from the group of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof.

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TITLE

FUNGICIDAL MIXTURES OF THIOPHENE DERIVATIVE

FIELD OF THE INVENTION

This invention relates to fungicidal mixtures of a certain thiophene derivative and to compositions comprising such mixtures and methods for using such mixtures as fungicides.

BACKGROUND OF THE INVENTION

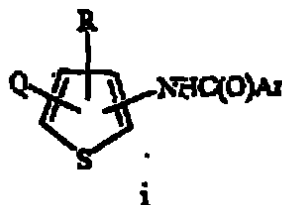
The control of plant diseases caused by fungal plant pathogens is extremely important in achieving high crop efficiency. Plant disease damage to ornamental, vegetable, field, cereal, and fruit crops can cause significant reduction in productivity and thereby result in increased costs to the consumer. Many products are commercially available for these purposes, but the need continues for new mixtures and compositions that are more effective, less costly, less toxic, environmentally safer or have different modes of action.

Fungicides that effectively control plant fungi are in constant demand by growers. Combinations of fungicides are often used to facilitate disease control and to retard resistance development. It is desirable to enhance the activity spectrum and the efficacy of disease control by using mixtures of active ingredients that provide a combination of curative, systemic and preventative control of plant pathogens. Also desirable are combinations that provide greater residual control to allow for extended spray intervals. It is also very desirable to combine fungicidal agents that inhibit different biochemical pathways in the fungal pathogens to retard development of resistance to any one particular plant disease control agent.

Being able to reduce the quantity of chemical agents released in the environment while ensuring effective protection of crops from diseases caused by plant pathogens is always desirable. Mixtures of fungicides may provide significantly better disease control than could be predicted based on the activity of the individual components. This synergism has been described as "the cooperative action of two components of a mixture, such that the total effect is greater or more prolonged than the sum of the effects of the two (or more) taken independently" (see P. M. L. Tamcs, *Neth. J. Plant Pathology* 1964, 70, 73-80).

New fungicidal agents that are particularly advantageous in achieving one or more of the preceding objectives continue to be needed.

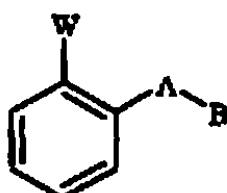
United States Patent No. 5,747,518 discloses certain thiophene derivative compounds of Formula i as new fungicidal active ingredients.



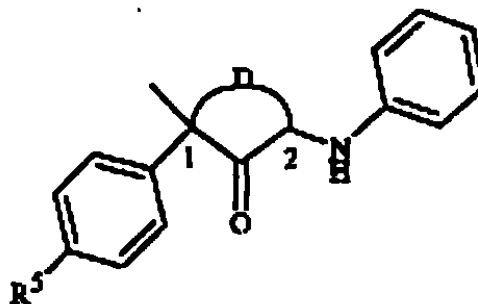
SUMMARY OF THE INVENTION

This invention relates to a fungicidal mixture comprising

- (a) *N*-[2-(1,3-dimethylbutyl)-3-thienyl]-1-methyl-3-(trifluoromethyl)-1*H*-pyrazole-4-carboxamide (including all stereoisomers) or an agriculturally suitable salt thereof; and
- (b) at least one compound selected from the group consisting of compounds of Formula III or Formula IV which act at the *bc*₁ complex of the fungal mitochondrial respiratory electron transfer site;



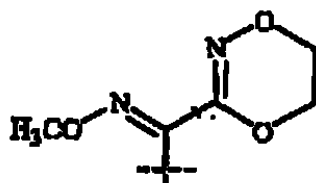
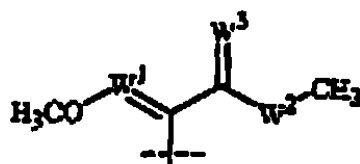
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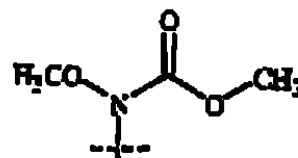
IV

10 wherein

W is one of the following group



or



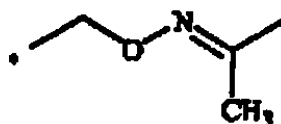
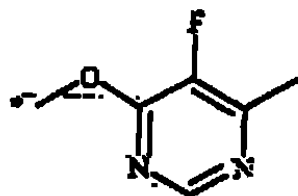
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W¹ is N or CH;

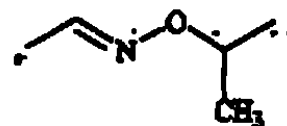
W² is NH, O or CH₂;

W³ is O or CH₂;

A is one of the following groups (wherein asterisk (*) is attached to the phenyl ring of the Formula III),



or



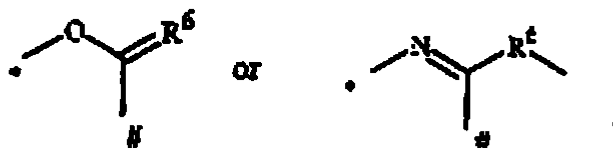
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B is 1,2-propanediol-bis(*O*-methyloxim)-1-yl; or phenyl, phenoxy or pyridinyl, each optionally substituted with 1 or 2 substituents selected from Cl, CN, methyl or trifluoromethyl; or

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A and B together are -CH₂O(1-[4-chlorophenyl]-pyrazol-3-yl) or -CH₂O(6-trifluoromethyl-2-pyridinyl).

D is one of the following groups (wherein asterisk (*) is attached to the carbon next to the phenyl ring of the Formula IV (carbon 1) and pound sign (#) is attached to the carbon next to the nitrogen of the Formula IV (carbon 2)),



R⁵ is H or phenoxy; and

R⁶ is O or S;

and agriculturally suitable salts thereof; and optionally

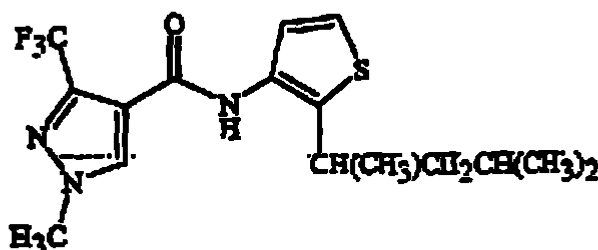
(c) at least one compound selected from the group consisting of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof.

This invention also relates to a fungicidal composition comprising a fungicidally effective amount of a mixture of the invention and at least one additional component selected from the group consisting of surfactants, solid diluents and liquid diluents.

This invention also relates to a method for controlling a plant disease caused by a fungal plant pathogen comprising applying to the plant or portion thereof a fungicidally effective amount of a mixture of the invention (e.g., as a composition described herein).

DETAILS OF THE INVENTION

The compound *N*-[2-(1,3-dimethylbutyl)-3-thienyl]-1-methyl-3-(trifluoromethyl)-1*H*-pyrazole-4-carboxamide, alternatively named 3-(trifluoromethyl)-1-methyl-*N*-(2-(4-methylpentan-2-yl)thiophen-3-yl)-1*H*-pyrazole-4-carboxamide and *N*-[2-(1,3-dimethylbutyl)-3-thienyl]-3-trifluoromethyl-1-methylpyrazole-4-carboxamide, can be represented by Formula I:



I

Many compounds in the mixtures of this invention (e.g., the compound of Formula I) can exist as one or more stereoisomers. Depending on the compounds, various stereoisomers can include enantiomers, diastereomers, atropisomers and geometric isomers. One skilled in the art will appreciate that one stereoisomer may be more active and/or may exhibit beneficial effects when enriched relative to the other stereoisomer(s) or when separated from the other stereoisomer(s). Additionally, the skilled artisan knows how to

separate, enrich, and/or to selectively prepare said stereoisomers. The compounds in the mixtures of this invention may be present as a mixture of stereoisomers, individual stereoisomers, or as an optically active form.

Agriculturally suitable salts of the compounds in the mixtures of the present invention include acid-addition salts with inorganic or organic acids such as hydrobromic, hydrochloric, nitric, phosphoric, sulfuric, acetic, butyric, fumaric, lactic, malic, malonic, oxalic, propionic, salicylic, tartaric, 4-toluenesulfonic or valeric acids. Agriculturally suitable salts of the compounds in the mixtures of the present invention also include those formed with organic bases (pyridine, ammonia, or triethylamine) or inorganic bases (hydrides, hydroxides, or carbonates of sodium, potassium, lithium, calcium, magnesium or barium) when the compound contains an acidic group such as a carboxylic acid or phenol.

Embodiments of the present invention include:

Embodiment 1. A mixture wherein component (b) is pyraclostrobin.

Embodiment 2. A mixture wherein component (b) is trifloxystrobin.

Embodiment 3. A mixture wherein component (b) is famoxadone.

Embodiment 4. A mixture also comprising at least one compound selected from the group consisting of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof.

Also noteworthy as embodiments are fungicidal compositions of the present invention comprising a fungicidally effective amount of a mixture of Embodiments 1 to 4 and at least one additional component selected from the group consisting of surfactants, solid diluents and liquid diluents. Embodiments of the invention further include methods for controlling plant diseases caused by fungal plant pathogens comprising applying to the plant or portion thereof, or to the plant seed or seedling, a fungicidally effective amount of a mixture of Embodiments 1 to 4 (e.g., as a composition described herein).

The compound *N*-[2-(1,3-dimethylbutyl)-3-thienyl]-1-methyl-3-(trifluoromethyl)-1*H*-pyrazole-4-carboxamide can be prepared by one or more of the methods and variations thereof as described in United States Patent No. 5,747,518 (see e.g., Example 14).

The fungicidal compounds of components (b) and (c) have been described in published patents and scientific journal papers. Many of these compounds are commercially available as active ingredients in fungicidal products. These compounds are described in compendia such as *The Pesticide Manual*, 13th edition, C. D. S. Thomlin (Ed.), British Crop Protection Council, Surrey, UK, 2003. These groups are further described below.

bc₁ Complex Fungicides (component (b))

Strobilurin fungicides such as fluoxastrobin, oryzaastrobin, picoxystrobin, pyraclostrobin and trifloxystrobin are known to have a fungicidal mode of action which inhibits the *bc₁* complex in the mitochondrial respiration chain (*Angew. Chem. Int. Ed.* 1999,

38, 1328-1349). Other strobilurin fungicides suitable for component (b) include (2*E*)-2-(2-
 {[6-(3-chloro-2-methylphenoxy)-5-fluor-4-pyrimidinyl]oxy}phenyl)-2-(methoxyimino)-*N*-
 methyl-ethanamide, (2*E*)-2-(methoxyimino)-*N*-methyl-2-(2-[[[(1*E*)-1-[3-
 (trifluoromethyl)phenyl]ethylidene]amino]oxy]methyl)phenyl)ethanamide, (2*E*)-2-
 5 methoxyimino)-*N*-methyl-2-(2-[(*E*)-{(1-[3-(trifluoromethyl)phenyl]ethoxy)imino]methyl]
 phenyl)ethanamide. Other compounds that inhibit the *bc*₁ complex in the mitochondrial
 respiration chain include fenoxadone and fenamidone. The *bc*₁ complex is sometimes
 referred to by other names in the biochemical literature, including complex III of the electron
 transfer chain, and ubiquinone:cytochrome *c* oxidoreductase. It is uniquely identified
 10 by the Enzyme Commission number EC1.10.2.2. The *bc*₁ complex is described in, for
 example, *J. Biol. Chem.* 1989, 264, 14543-48; *Methods Enzymol.* 1986, 126, 253-71; and
 references cited therein.

Inhibitors of Demethylase in Sterol Biosynthesis (component (c))

Sterol biosynthesis inhibitors control fungi by inhibiting enzymes in the sterol
 15 biosynthesis pathway. Demethylase-inhibiting fungicides have a common site of action
 within the fungal sterol biosynthesis pathway; that is inhibition of demethylation at position
 14 of lanosterol or 24-methylene dihydrolanosterol, which are precursors to sterols in fungi.
 Compounds acting at this site are often referred to as demethylase inhibitors, DMI
 fungicides, or DMIs. The demethylase enzyme is sometimes referred to by other names in
 20 the biochemical literature, including cytochrome P-450 (14DM). The demethylase enzyme
 is described in, for example, *J. Biol. Chem.* 1992, 267, 13175-79 and references cited
 therein. DMI fungicides fall into several chemical classes: azoles (including triazoles and
 imidazoles), pyrimidines, piperazines and pyridines. The triazoles include azaconazole,
 bromuconazole, cyproconazole, difenoconazole, diniconazole (including diniconazole-M),
 25 epoxiconazole, etaconazole, fenbuconazole, fluquinconazole, flusilazole, flutriafol,
 hexaconazole, imibenconazole, ipconazole, metconazole, myclobutanil, penconazole,
 propiconazole, prothioconazole, quinconazole, simeconazole, tebuconazole, tetraconazole,
 triadimefon, triadimenol, triticonazole and uniconazole. The imidazoles include
 clotrimazole, econazole, imazalil, isoconazole, miconazole, oxpoconazole, prochloraz and
 30 triflumizole. The pyrimidines include fenarimol, nuarimol and triarimol. The piperazines
 include triforine. The pyridines include buttiobate and pyrifenoxy. Biochemical
 investigations have shown that all of the above mentioned fungicides are DMI fungicides as
 described by K. H. Knock, et al. in *Modern Selective Fungicides - Properties, Applications
 and Mechanisms of Action*, H. Lyr (Ed.), Gustav Fischer Verlag: New York, 1995, 205-258.

35 Descriptions of the commercially available compounds listed above may be found in
The Pesticide Manual, Thirteenth Edition, C. D. S. Tomlin (Ed.), British Crop Protection
 Council, 2003.

This invention provides combinations of fungicides that have different biochemical modes of action. Such combinations can be particularly advantageous for resistance management, especially where the fungicides of the combination control the same or similar diseases. Examples include combinations of the compound of Formula I with strobilurins such as fluoxastrobin, picoxystrobin, pyraclostrobin and trifloxystrobin; and optionally, DMIs such as cyproconazole, epoxiconazole, fluquinconazole, flusilazole, hexaconazole, metconazole, propiconazole, prothioconazole and tebuconazole.

This invention also provides combinations of fungicides that provide an expanded spectrum of disease control or enhanced efficacy, including enhanced residual, curative, or preventive control. Examples include combinations of the compound of Formula I with strobilurins such as fluoxastrobin, picoxystrobin, pyraclostrobin and trifloxystrobin; and optionally, DMIs such as bromuconazole, cyproconazole, epoxiconazole, fluquinconazole, flusilazole, hexaconazole, metconazole, propiconazole, prothioconazole and tebuconazole.

This invention also provides combinations of fungicides that are particularly useful for controlling cereal diseases (e.g., *Erysiphe graminis*, *Septoria nodorum*, *Septoria tritici*, *Puccinia recondita* and *Pyrenophora teres*). Examples include combinations of the compound of Formula I with strobilurins such as fluoxastrobin, picoxystrobin, pyraclostrobin and trifloxystrobin; and optionally, DMIs such as bromuconazole, cyproconazole, epoxiconazole, fluquinconazole, flusilazole, hexaconazole, metconazole, propiconazole, prothioconazole and tebuconazole. Of particular note is the use of these combinations for controlling barley diseases (e.g., *Pyrenophora teres*).

This invention also provides combinations of fungicides that are particularly useful for controlling diseases of fruits and vegetables (*Alternaria solani*, *Botrytis cinerea*, *Rhizoctonia solani*, *Uncinula necator* and *Venteria inaequalis*). Examples include combinations of the compound of Formula I with strobilurins such as picoxystrobin, pyraclostrobin and trifloxystrobin; and optionally, DMIs such as bromuconazole, cyproconazole, epoxiconazole, fluquinconazole, flusilazole, hexaconazole, metconazole, propiconazole, prothioconazole and tebuconazole.

The weight ratios of component (b) to component (a) in the mixtures and compositions of the present invention are typically from 100:1 to 1:100, preferably from 25:1 to 1:25, and more preferably from 10:1 to 1:10. Of note are mixtures and compositions wherein the weight ratio of component (b) to component (a) is from 5:1 to 1:1. Examples of these compositions include compositions comprising a mixture of the compound of Formula I with trifloxystrobin, a mixture of the compound of Formula I with fluoxastrobin, a mixture of the compound of Formula I with picoxystrobin, or a mixture of the compound of Formula I with pyraclostrobin.

Of note are compositions wherein component (b) and component (c) are both present. Examples are compositions comprising pyraclostrobin or trifloxystrobin as component (b),

and at least one component (c) compound. Of note are such compositions wherein the overall weight ratio of components (b) and (c) to component (a) is from 100:1 to 1:100 and the weight ratio of component (b) to component (a) is from 25:1 to 1:25. Included are compositions wherein the weight ratio of component (b) to component (a) is from 5:1 to 1:1.

- 5 Examples of these compositions include compositions comprising mixtures of component (a) with trifloxystrobin and a compound selected from the group consisting of epoxiconazole and flusilazole.

Formulation/Utility

- Mixtures of this invention will generally be used as a formulation or composition comprising at least one carrier selected from agriculturally suitable liquid diluents, solid diluents and surfactants. The formulation or composition ingredients are selected to be consistent with the physical properties of the active ingredient, mode of application and environmental factors such as soil type, moisture and temperature. Useful formulations include liquids such as solutions (including emulsifiable concentrates), suspensions, 15 emulsions (including microemulsions and/or suspoemulsions) and the like which optionally can be thickened into gels. Useful formulations further include solids such as dusts, powders, granules, pellets, tablets, films, and the like which can be water-dispersible ("wetable") or water-soluble. The active ingredients can be (micro)encapsulated and further formed into a suspension or solid formulation; alternatively the entire formulation of active 20 ingredient can be encapsulated (or "overcoated"). Encapsulation can control or delay release of the active ingredient. Sprayable formulations can be extended in suitable media and used at spray volumes from about one to several hundred liters per hectare. High-strength compositions are primarily used as intermediates for further formulation.

- The formulations will typically contain effective amounts (e.g., from 0.01-99.99 weight percent) of active ingredients together with diluent and/or surfactant within the following approximate ranges which add up to 100 percent by weight.

	Weight Percent		
	<u>Active Ingredients</u>	<u>Diluent</u>	<u>Surfactant</u>
Water-Dispersible and Water-soluble Granules, Tablets and Powders.	5-90	0-94	1-15
Suspensions, Emulsions, Solutions (including Emulsifiable Concentrates)	5-50	40-95	0-25
Dusts	1-25	70-99	0-5
Granules and Pellets	0.01-99	5-99.99	0-15
High Strength Compositions	90-99	0-10	0-2

Typical solid diluents are described in Watkins, et al., *Handbook of Insecticide Dust Diluents and Carriers*, 2nd edition, Dorland Books, Caldwell, New Jersey. Typical liquid diluents are described in Marsden, *Solvents Guide*, 2nd edition, Interscience, New York, 1950. *McCutcheon's Detergents and Emulsifiers Annual*, Allured Publ. Corp., Ridgewood, New Jersey, as well as Sisely and Wood, *Encyclopedia of Surface Active Agents*, Chemical Publ. Co., Inc., New York, 1964, list surfactants and recommended uses. All formulations can contain minor amounts of additives to reduce foam, caking, corrosion, microbiological growth and the like, or thickeners to increase viscosity.

Surfactants include, for example, polyethoxylated alcohols, polyethoxylated alkylphenols, polyethoxylated sorbitan fatty acid esters, dialkyl sulfosuccinates, alkyl sulfates, alkylbenzene sulfonates, organosilicones, *N,N*-dialkyltaurates, lignin sulfonates, naphthalene sulfonate formaldehyde condensates, polycarboxylates, and polyoxyethylene/polyoxypropylene block copolymers. Solid diluents include, for example, clays such as bentonite, montmorillonite, attapulgite and kaolin, starch, sugar, silica, talc, diatomaceous earth, urea, calcium carbonate, sodium carbonate and bicarbonate, and sodium sulfate. Liquid diluents include, for example, water, *N,N*-dimethylformamide, dimethyl sulfoxide, *N*-alkylpyrrolidone, ethylene glycol, polypropylene glycol, paraffins, alkylbenzenes, alkylnaphthalenes, oils of olive, castor, linseed, tung, sesame, corn, peanut, cotton-seed, soybean, rape-seed and coconut, fatty acid esters, ketones such as cyclohexanone, 2-heptanone, isophorone and 4-hydroxy-4-methyl-2-pentanone, and alcohols such as methanol, cyclohexanol, decanol and tetrahydrofurfuryl alcohol.

Solutions, including emulsifiable concentrates, can be prepared by simply mixing the ingredients. Dusts and powders can be prepared by blending and, usually, grinding as in a hammer mill or fluid-energy mill. Suspensions are usually prepared by wet-milling; see, for example, U.S. 3,060,084. Preferred suspension concentrates include those containing, in addition to the active ingredient, from 5 to 20% nonionic surfactant (for example, polyethoxylated fatty alcohols) optionally combined with 50-65% liquid diluents and up to 5% anionic surfactants. Granules and pellets can be prepared by spraying the active material upon preformed granular carriers or by agglomeration techniques. See Browning, "Agglomeration", *Chemical Engineering*, December 4, 1967, pp 147-48, *Perry's Chemical Engineer's Handbook*, 4th edition, McGraw-Hill, New York, 1963, pages 8-57 and following, and WO 91/13546. Pellets can be prepared as described in U.S. 4,172,714. Water-dispersible and water-soluble granules can be prepared as taught in U.S. 4,144,050, U.S. 3,920,442 and DE 3,246,493. Tablets can be prepared as taught in U.S. 5,180,587, U.S. 5,232,701 and U.S. 5,208,030. Films can be prepared as taught in GB 2,095,558 and U.S. 3,299,566.

For further information regarding the art of formulation, see U.S. 3,235,361, Col. 6, line 16 through Col. 7, line 19 and Examples 10-41; U.S. 3,309,192, Col. 5, line 43 through

Col. 7, line 62 and Examples 8, 12, 15, 39, 41, 52, 53, 58, 132, 138-140, 162-164, 166, 167 and 169-182; U.S. 2,891,855, Col. 3, line 66 through Col. 5, line 17 and Examples 1-4; Klingman, *Weed Control as a Science*, John Wiley and Sons, Inc., New York, 1961, pp 81-96; and Hance et al., *Weed Control Handbook*, 8th edition., Blackwell Scientific Publications, Oxford, 1989.

In the following Examples, all percentages are by weight and all formulations are prepared in conventional ways. The term "active ingredients" as used in Example A through E refers to the combination of compounds from group (a) and group (b), together with any other active ingredient(s) present (e.g., any compound from group (c)). Without further elaboration, it is believed that one skilled in the art using the preceding description can utilize the present invention to its fullest extent. The following Examples are, therefore, to be construed as merely illustrative, and not limiting of the disclosure in any way whatsoever. Percentages are by weight except where otherwise indicated.

Example A

15 Wettable Powder

active ingredients	65.0%
dodecylphenol polyethylene glycol ether	2.0%
sodium ligninsulfonate	4.0%
sodium silicoaluminate	6.0%
montmorillonite (calcined)	23.0%

Example B

Granule

active ingredients	10.0%
attapulgate granules (low volatile matter, 0.71/0.30 mm; U.S.S. No. 25-50 sieves)	90.0%

Example C

Extruded Pellet

active ingredients	25.0%
anhydrous sodium sulfate	10.0%
crude calcium ligninsulfonate	5.0%
sodium alkyl naphthalenesulfonate	1.0%
calcium/magnesium bentonite	59.0%

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Example D

Emulsifiable Concentrate

	active ingredients	20.0%
	blend of oil soluble sulfonates	
5	and polyoxyethylene ethers	10.0%
	isophorone	70.0%

Example E

Suspension Concentrate

	active ingredients	20.0%
10	polyethoxylated fatty alcohol	15.0%
	ester derivative of montan wax	3.0%
	calcium lignosulfonate	2.0%
	polyethoxylated/polypropoxylated	
	polyglycol block copolymer	1.0%
15	propylene glycol	6.4%
	poly(dimethylsiloxane)	0.6%
	antimicrobial agent	0.1%
	water	51.9%

Compositions of this invention can also include one or more other insecticides,
 20 fungicides, nematocides, bactericides, acaricides, growth regulators, chemosterilants,
 semiochemicals, repellents, attractants, pheromones, feeding stimulants or other biologically
 active compounds to form a multi-component pesticide giving an even broader spectrum of
 agricultural protection. Examples of such agricultural protectants with which compositions
 of this invention can be formulated are: insecticides such as abamectin, acephate,
 25 azinphos-methyl, bifenthrin, buprofezin, carbofuran, chlorfenspyr, chlorpyrifos,
 chlorpyrifos-methyl, cyfluthrin, beta-cyfluthrin, cyhalothrin, lambda-cyhalothrin,
 deltamethrin, diafenthiuron, diazinon, disubenzuron, dimethoate, esfenvalerate, ethiprole,
 fenoxycarb, fenpropathrin, fenvalerate, fipronil, flucythrinate, tau-fluvalinate, fenopbos,
 imidacloprid, indoxacarb, isofenphos, malathion, metaldehyde, methamidophos,
 30 methidathion, methomyl, methoprene, methoxychlor, monocrotophos, oxamyl, parathion,
 parathion-methyl, permethrin, phorate, phosalone, phosmet, phosphamidon, pirimicarb,
 profenofos, rotenone, sulprofos, tebufenozide, terbufos, tetrachlorvinphos,
 thiodicarb, tralomethrin, trichlorfon and triflunuron; fungicides besides those listed for
 components (b) and component (c), such as acibenzolar-S-methyl, benalaxyl (including
 35 benalaxyl-M), benthiavalicarb, benomyl, blastocidin-S, Bordeaux mixture (tribasic copper
 sulfate), boscalid, buthiobata, carpropamid, captan, captan, carbendazim, chloronch,
 chlorothalonil, copper oxychloride, copper salts, cymoxanil, cyazofamid, cyflufenamid,

cyprodinil, diclofymet, diclomezline, dicloran, dimethomorph, doctus, edifenphos,
 ethaboxam, fenhexamid, fenoxanil, fenpiclonil, fenpropiidin, fenpropimorph, fenlin acetate,
 fenlin hydroxide, flumazuron, fludioxonil, flumorph, flumolanil, folpet, fosetyl-aluminum,
 furalaxyl, furametapyr, guazatine, hymexazol, iminoctadine, iprobenfos, iprodione,
 5 iprovalicarb, isoprothiolane, kasugamycin, mancozeb, maneb, mefenoxam, nepanapyrin,
 nepronil, metalaxyl, metrafenone, neo-asozin (ferric methanearsonate), oxadixyl,
 pencyuron, picobenzazoid, prothiazole, propanocarb, proquinazid, pyrimethanil,
 pyroquilon, quinoxifen, silthiofuran, spiromoxamine, sulfur, thiabendazole, thifluzamide,
 thiophanate-methyl, thiram, tiadinil, tolyfluanid, validamycin, vinclozolin and zoxamide;
 10 nematocides such as aldoxycarb and fenamiphos; bactericides such as streptomycin;
 acaricides such as amitraz, chloromethionat, chlorobenzilate, cyhexatin, dicofol, dinoclor,
 etoxazole, fenazaquin, fenbutatin oxide, fenpropathrin, fenpyroximate, hexythiazox,
 propargite, pyridaben and tebufenpyrad; and biological agents such as *Bacillus thuringiensis*
 including ssp. *aisawa* and *kaistaki*, *Bacillus thuringiensis* delta endotoxin, baculovirus, and
 15 entomopathogenic bacteria, virus and fungi. Descriptions of various commercially available
 compounds listed above may be found in *The Pesticide Manual, Twelfth Edition*, C.D.S.
 Tomlin, ed., British Crop Protection Council, 2000. For embodiments where one or more of
 these various mixing partners are used, the weight ratio of these various mixing partners (in
 total) to the total amount of component (a) and component (b) is typically between 100:1 and
 20 1:3000. Of note are weight ratios between 30:1 and 1:500 (for example ratios between 1:1
 and 1:30). It will be evident that including these additional components may expand the
 spectrum of diseases controlled beyond the spectrum controlled by the combination of
 component (a), component (b), and the optional component (c) alone.

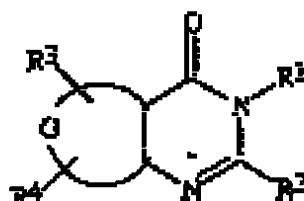
Of particular note are compositions which in addition to component (a), component
 25 (b), and the optional component (c), if present, include (d) at least one compound selected
 from the group consisting of

- (d1) alkylenebis(dithiocarbamate) fungicides;
- (d2) cyanoxanil;
- (d3) phenylamide fungicides;
- 30 (d4) pyrimidinone fungicides;
- (d5) chlorothalonil;
- (d6) carboxamides acting at complex II of the fungal mitochondrial respiratory electron
 transfer site;
- (d7) quinoxifen;
- 35 (d8) metrafenone;
- (d9) cythiuzenamid;
- (d10) cyprodinil;
- (d11) copper compounds;

- (d12) phthalimide fungicides;
 (d13) fosetyl-aluminum;
 (d14) benzimidazole fungicides;
 (d15) cyazofamid;
 5 (d16) fluazinam;
 (d17) iprodione;
 (d18) propiconazole;
 (d19) validamycin;
 (d20) dichlorophenyl dicarboximide fungicides;
 10 (d21) zoxamide; and
 (d22) dimethomorph;
 (d23) non-DMI sterol biosynthesis inhibitors; and
 agriculturally suitable salts of compounds of (d1) through (d23).

Pyrimidinone Fungicides (group (d4))

- 15 Pyrimidinone fungicides include compounds of Formula II



II

wherein

G forms a fused phenyl, thiophene or pyridine ring;

R¹ is C₁-C₆ alkyl;

20 R² is C₁-C₆ alkyl or C₁-C₆ alkoxy;

R³ is halogen; and

R⁴ is hydrogen or halogen.

Pyrimidinone fungicides are described in World Patent Application Publication
 WO94/26722, U.S. Patent No. 5,066,638, U.S. Patent No. 6,245,770, U.S. Patent No.
 25 6,262,058 and U.S. Patent No. 6,277,858.

Of note are pyrimidinone fungicides selected from the group:

6-bromo-3-propyl-2-propyloxy-4(3*H*)-quinazolinone,

6,8-difluoro-3-propyl-2-propyloxy-4(3*E*)-quinazolinone,

6-iodo-3-propyl-2-propyloxy-4(3*H*)-quinazolinone (proquinazid),

30 6-chloro-2-propoxy-3-propylthieno[2,3-*d*]pyrimidin-4(3*H*)-one,

6-bromo-2-propoxy-3-propylthieno[2,3-*d*]pyrimidin-4(3*E*)-one,

7-bromo-2-propoxy-3-propylthieno[3,2-*d*]pyrimidin-4(3*H*)-one,

6-bromo-2-propoxy-3-propylpyrido[2,3-*d*]pyrimidin-4(3*H*)-one,
 6,7-dibromo-2-propoxy-3-propylthieno[3,2-*d*]pyrimidin-4(3*H*)-one, and
 3-(cyclopropylmethyl)-6-iodo-2-(propylthio)pyrido[2,3-*d*]pyrimidin-4(3*H*)-one.

Other Fungicide Groups

5 Alkylenebis(dithiocarbamate)s (d1) include compounds such as mancozeb, maneb, propineb and zineb.

Phenylamides (d3) include compounds such as metalaxyl, benalaxyl, furalaxyl and oxadixyl.

10 Carboxanides (d6) include compounds such as boscalid, carboxin, fenfuram, flutolanil, furametpyr, mepronil, oxycarboxin and thifluzamide are known to inhibit mitochondrial function by disrupting complex II (succinate dehydrogenase) in the respiratory electron transport chain.

15 Copper compounds (d11) include compounds such as copper oxychloride, copper sulfate and copper hydroxide, including compositions such as Bordeaux mixture (tribasic copper sulfate).

Phthalimides (d12) include compounds such as folpet and captan.

Benzimidazole fungicides (d14) include benomyl and carbendazim.

Dichlorophenyl dicarboximide fungicides (d20) include chlorzoline, dichlozoline, iprodione, isovaldicione, myclozolin, procymidone and vinclozolin.

20 Non-DMI sterol biosynthesis inhibitors (d23) include morpholine and piperidine fungicides. The morpholines and piperidines are sterol biosynthesis inhibitors that have been shown to inhibit steps in the sterol biosynthesis pathway at a point later than the inhibitions achieved by the DMI sterol biosynthesis (i.e., component (c)). The morpholines include aldimorph, dodemorph, fenpropimorph, tridemorph and trimorphamide. The
 25 piperidines include fenpropidin.

Of note are such compositions wherein the overall weight ratio of components (b), (d) and, if present, (c) to component (a) is from 100:1 to 1:100 and the weight ratio of component (b) to component (a) is from 25:1 to 1:25. Included are compositions wherein the weight ratio of component (b) to component (a) is from 5:1 to 1:1.

30 The mixtures and compositions of this invention are useful as plant disease control agents. The present invention therefore further comprises a method for controlling plant diseases caused by fungal plant pathogens comprising applying to the plant or portion thereof to be protected, or to the plant seed or seedling to be protected, an effective amount of a mixture of the invention or a fungicidal composition containing said mixture.

35 The mixtures and compositions of this invention provide control of diseases caused by a broad spectrum of fungal plant pathogens in the Basidiomycete, Ascomycete, Oomycete and Deuteromycete classes. They are effective in controlling a broad spectrum of plant

diseases, particularly foliar pathogens of ornamental, vegetable, field, cereal, and fruit crops. These pathogens include:

Oomycetes, including *Phytophthora* diseases such as *Phytophthora infestans*, *Phytophthora megasperma*, *Phytophthora parasitica*, *Phytophthora cinnamomi* and *Phytophthora capsici*;

5 *Pythium* diseases such as *Pythium aphanidermatum*; and diseases in the Peronosporaceae family, such as *Plasmopara viticola*, *Peronospora* spp. (including *Peronospora tabacina* and *Peronospora parasitica*), *Pseudoperonospora* spp. (including *Pseudoperonospora cubensis*), and *Bremia lactucae*;

Ascomycetes, including *Alternaria* diseases such as *Alternaria solani* and *Alternaria*
10 *brassicarum*; *Guignardia* diseases such as *Guignardia bidwellii*; *Venturia* diseases such as *Venturia inaequalis*; *Septoria* diseases such as *Septoria nodorum* and *Septoria tritici*; powdery mildew diseases such as *Erysiphe* spp. (including *Erysiphe graminis* and *Erysiphe polygoni*), *Uncinula necator*, *Sphaeria fuliginea*, and *Podiumphaera leucotricha*; *Pseudocercospora herpotrichoides*; *Botrytis* diseases such as *Botrytis cinerea*; *Monilinia*
15 *fructicola*; *Sclerotinia* diseases such as *Sclerotinia sclerotiorum*; *Magnaporthe grisea*; *Phomopsis viticola*; *Helminthosporium* diseases such as *Helminthosporium tritici repentis*; *Pyrenophora teres*; anthracnose diseases such as *Glomerella* or *Colletotrichum* spp. (such as *Colletotrichum graminicola*); and *Gaeumannomyces graminis*;

Basidiomycetes, including rust diseases caused by *Puccinia* spp. (such as *Puccinia*
20 *recondita*, *Puccinia striiformis*, *Puccinia hordei*, *Puccinia graminis*, and *Puccinia arachidis*); *Hemileia vastatrix*; and *Phakopsora pachyrhizi*;

other pathogens including *Rhizoctonia* spp (such as *Rhizoctonia solani*); *Fusarium* diseases such as *Fusarium roseum*, *Fusarium graminearum* and *Fusarium oxysporum*; *Verticillium dahliae*; *Sclerotium rolfsii*; *Rhynchosporium secalis*; *Cercosporidium personatum*;

25 *Cercospora arachidicola* and *Cercospora beticola*;

and other genera and species closely related to these pathogens.

In addition to their fungicidal activity, the mixtures and compositions can also have activity against bacteria such as *Erwinia amylovora*, *Xanthomonas campestris*, *Pseudomonas syringae*, and other related species.

30 Of note is use of a mixture of this invention for controlling *Erysiphe graminis* (wheat powdery mildew), especially using a mixture wherein component (b) is trifloxystrobin.

Of note is use of a mixture of this invention for controlling *Septoria nodorum* (*Septoria glume blotch*), especially using a mixture wherein component (b) is trifloxystrobin.

Of note is use of a mixture of this invention for controlling *Pyrenophora teres* (barley
35 net blotch), especially using a mixture wherein component (b) is trifloxystrobin.

Of note is use of a mixture of this invention for controlling *Puccinia recondita* (wheat leaf rust), especially using a mixture wherein component (b) is trifloxystrobin.

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Also noteworthy is the use of a mixture or composition of this invention to provide control of diseases caused by a broad spectrum of fungal plant pathogens preventatively or curatively by applying an effective amount of the mixture or composition either pre- or post-infection.

Plant disease control is ordinarily accomplished by applying an effective amount of a mixture of this invention either pre- or post-infection, to the portion of the plant to be protected such as the roots, stems, foliage, fruit, seeds, tubers or bulbs, or to the media (soil or sand) in which the plants to be protected are growing. Application of the mixture to a seed can protect both the seed and the seedling grown from the seed. Typically the mixture is applied in the form of a composition comprising at least one additional component selected from the group consisting of surfactants, solid diluents and liquid diluents.

Rates of application for these compounds can be influenced by many factors of the environment and should be determined under actual use conditions. Foliage can normally be protected when treated at a rate of from less than 1 g/ha to 5,000 g/ha total of active ingredients of components (a) and (b) in the mixtures and compositions of the present invention. Seed and seedlings can normally be protected when seed is treated at a rate of from 0.1 to 10 g total of active ingredients of components (a) and (b) per kilogram of seed.

The mixture of this invention provides advantageous control of fungal plant diseases when compared to the control achieved by each of the component alone. Mixtures of this invention have been found to exhibit synergy, particularly in connection with controlling certain diseases such as barley net blotch caused by *Pyrenophora teres*.

The following Tests can be used to demonstrate the control efficacy of compositions of this invention on specific pathogens. The pathogen control protection afforded by the compounds is not limited, however, to these species.

BIOLOGICAL EXAMPLES OF THE INVENTION

Test suspensions comprising a single formulated active ingredient are sprayed to demonstrate the control efficacy of the active ingredient individually. To demonstrate the control efficacy of a combination, (a) the active ingredients can be combined in the appropriate amounts in a single test suspension, (b) stock solutions of individual active ingredients can be prepared and then combined in the appropriate ratio, and diluted to the final desired concentration to form a test suspension or (c) test suspensions comprising single active ingredients can be sprayed sequentially in the desired ratio.

Composition 1

<u>Ingredients</u>	<u>Wt. %</u>
Compound of Formula I, prepared as a 20 % suspension concentrate	200 grams active /liter

Composition 2

<u>Ingredients</u>	<u>Wt %</u>
trifloxystrobin (TWIST Fungicide), formulated as a suspension concentrate	125 grams active /liter

Composition 3

<u>Ingredients</u>	<u>Wt %</u>
epoxiconazole (OPUS Fungicide), formulated as a emulsifiable concentrate	125 grams active /liter

Test compositions are first mixed with water. The resulting test suspensions are then used in the following field tests. Test suspensions are sprayed at a volume of 200 liters per hectare. Application rates are 50, 100, and 200 grams per hectare.

TEST A

Established field plots of winter wheat (cv. 'Onvanlis') are sprayed when node 4 is at least 2 cm above node 3 in the wheat plants and again when the flag leaf is fully emerged. Four replications are used. Plots are evaluated visually for control of symptoms of wheat leaf blotch caused by *Septoria tritici*. The results are reported as the mean average of the four replications.

TEST B

Established field plots of winter barley (cv. 'Esterel') are sprayed when the flag leaf of the barley plant is fully emerged. Three replications are used. Plots are evaluated visually for control of symptoms of barley net blotch caused by *Pyrenophora teres*. The results are reported as the average mean percent disease control of the three replications.

Table A lists results for Tests A and B. In Table A, a rating of 100 indicates 100% disease control and a rating of 0 indicates no disease control (relative to the controls). Columns labeled "Avg" indicates the average of the three or four replications.

000000 10

Table A
Test Results

Composition	Application Rate (g/ha)	Test A		Test B	
		Avg	Exp	Avg	Exp
1	50	21	-	12	-
1	100	48	-	28	-
1	200	56	-	31	-
2	100	53	-	66	-
3	100	63	-	31	-
1 + 2	100 + 200	73	76	86	75
1 + 3	100 + 100	86	80	66	50
2 + 3	200 + 100	80	53	67	76

Table A shows mixtures and compositions of the present invention demonstrating advantageous control.

5 The presence of a synergistic effect between two active ingredients is established with the aid of the Colby equation (see S. R. Colby, "Calculating Synergistic and Antagonistic Responses of Herbicide Combinations", *Weeds*, 1967, 15, 20-22):

$$p = A + B - \left[\frac{A \times B}{100} \right]$$

10 Using the method of Colby, the presence of a synergistic interaction between two active ingredients is established by first calculating the predicted activity, p, of the mixture based on activities of the two components applied alone. If p is lower than the experimentally observed effect, synergism has occurred. In the equation above, A is the fungicidal activity in percentage control of one component applied alone at rate x. The B term is the fungicidal activity in percentage control of the second component applied at
15 rate y. The equation estimates p, the fungicidal activity of the mixture of A at rate x with B at rate y if their effects are strictly additive and no interaction has occurred.

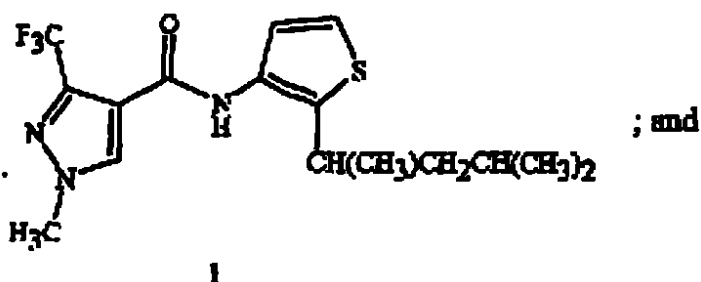
 Columns labeled "Exp" in Table A indicate the expected value for each treatment mixture using the Colby equation. As is shown in Table A, application of the Colby equation reveals that the observed control is higher than expected for the mixture of
20 Composition 1 with Composition 2 (100 g/ha + 200 g/ha) for Test B and for the mixture of Composition 1 with Composition 3 (100 g/ha + 100 g/ha) for both Test A and Test B.

CLAIMS

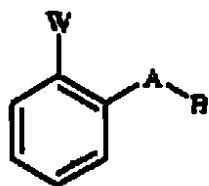
What is claimed is:

1. A fungicidal mixture comprising:

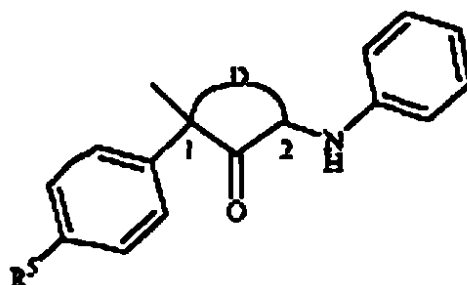
5 (a) the thiophene derivative of Formula I or an agriculturally suitable salt thereof



(b) at least one compound selected from the group consisting of compounds of Formula III or Formula IV which act at the bc_1 complex of the fungal mitochondrial respiratory electron transfer site;



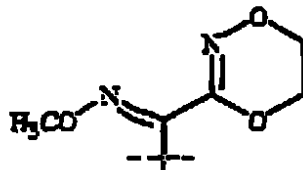
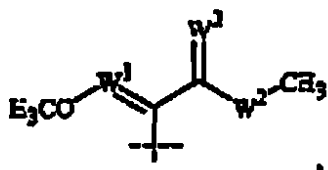
III



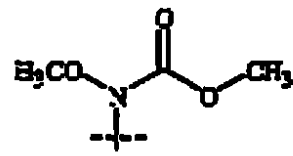
IV

10 wherein

W is one of the following group



or



15

W¹ is N or CH;

W² is NH, O or CH₂;

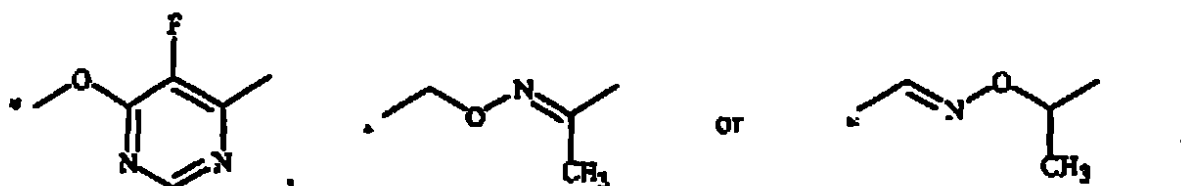
W³ is O or CH₂;

A is one of the following groups (wherein asterisk (*) is attached to the phenyl ring of the Formula III),

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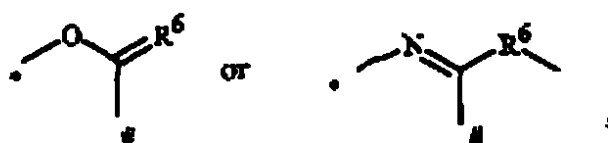
17



B is 1,2-propandion-bis(O-methyloxim)-1-yl; or phenyl, phenoxy or pyridinyl, each optionally substituted with 1 or 2 substituents selected from Cl, CN, methyl or trifluoromethyl; or

A and B together are -CH₂O(1-[4-chlorophenyl]-pyrazol-3-yl) or -CH₂O(6-trifluoromethyl-2-pyridinyl).

D is one of the following groups (wherein asterisk (*) is attached to the carbon next to the phenyl ring of the Formula IV (carbon 1) and pound sign (#) is attached to the carbon next to the nitrogen of the Formula IV (carbon 2)),



R⁵ is H or phenoxy; and

R⁶ is O or S;

and agriculturally suitable salts thereof; and optionally

(c) at least one compound selected from the group consisting of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof.

2. A mixture of Claim 1 wherein component (b) is trifloxystrobin.

3. A mixture of Claim 2 wherein component (c) is present and is epoxiconazole.

4. A fungicidal composition comprising a fungicidally effective amount of the mixture of any one of Claims 1 to 3 and at least one additional component selected from the group consisting of surfactants, solid diluents and liquid diluents.

5. A composition of Claim 4 wherein the weight ratio of component (b) to component (a) is from 100:1 to 1:100.

6. A composition of Claim 5 wherein the weight ratio of component (b) to component (a) is from 10:1 to 1:10.

7. A composition of Claim 6 wherein the weight ratio of component (b) to component (a) is from 5:1 to 1:1.

8. A method for controlling a plant disease caused by a fungal plant pathogen comprising applying to the plant or portion thereof a fungicidally effective amount of the mixture of any one of Claims 1 to 3.

9. A method of Claim 8 wherein the fungal plant pathogen is *Pyrenophora teres*.

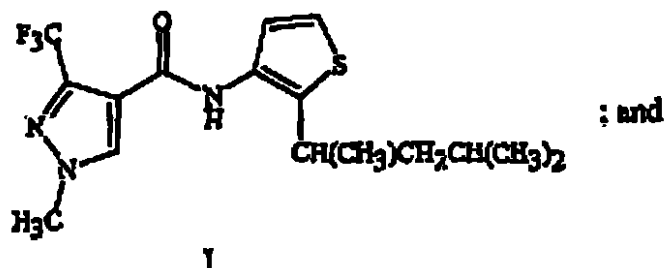
5 10. A method of Claim 8 wherein the fungal plant pathogen is *Septoria tritici*.

AMENDED CLAIMS

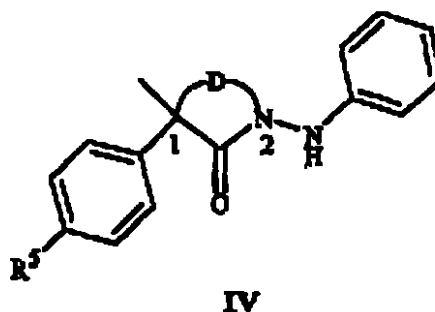
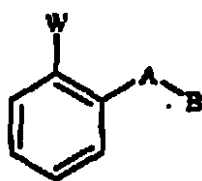
[received by the International Bureau on 16 February 2006 (16.02.06);
original claim 1 amended; remaining claims unchanged]

What is claimed is:

1. A fungicidal mixture comprising:
(a) the thiophene derivative of Formula I or an agriculturally suitable salt thereof

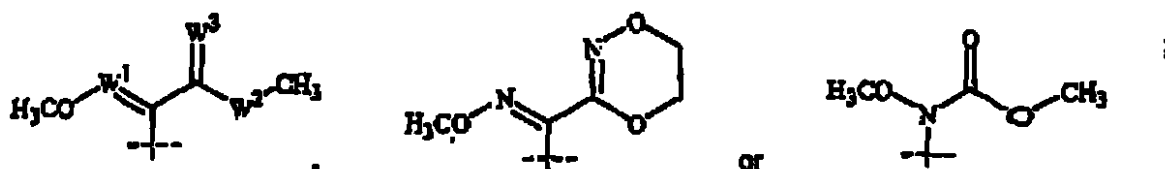


- (b) at least one compound selected from the group consisting of compounds of Formula III or Formula IV which act at the bc_1 complex of the fungal mitochondrial respiratory electron transfer site;



wherein

W is one of the following group



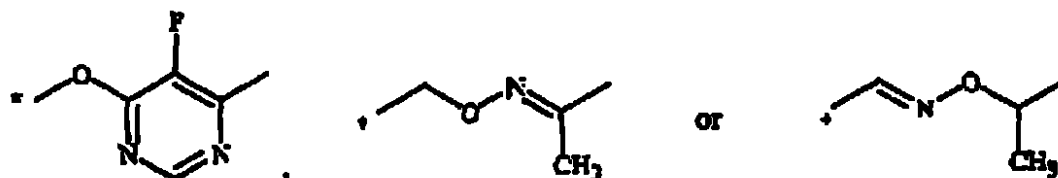
W^1 is N or CH;

W^2 is NH, O or CH_2 ;

W^3 is O or CH_2 ;

A is one of the following groups (wherein asterisk (*) is attached to the phenyl ring of the Formula III),

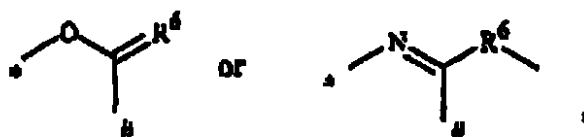
AMENDED SHEET (ARTICLE 19)



B is 1,2-propandion-bis(*O*-methyloxim)-1-yl; or phenyl, phenoxy or pyridinyl, each optionally substituted with 1 or 2 substituents selected from Cl, CN, methyl or trifluoromethyl; or

A and B together are $-\text{CH}_2\text{O}(1\text{-[4-chlorophenyl]-pyrazol-3-yl})$ or $-\text{CH}_2\text{O}(6\text{-trifluoromethyl-2-pyridinyl})$.

D is one of the following groups (wherein asterisk (*) is attached to the carbon next to the phenyl ring of the Formula IV (carbon 1) and pound sign (#) is attached to the ring nitrogen of the Formula IV (nitrogen 2)),



R^5 is H or phenoxy; and

R^6 is O or S;

and agriculturally suitable salts thereof; and optionally

(c) at least one compound selected from the group consisting of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof.

2. A mixture of Claim 1 wherein component (b) is trifloxystrobin.

3. A mixture of Claim 2 wherein component (c) is present and is epoxiconazole.

4. A fungicidal composition comprising a fungicidally effective amount of the mixture of any one of Claims 1 to 3 and at least one additional component selected from the group consisting of surfactants, solid diluents and liquid diluents.

5. A composition of Claim 4 wherein the weight ratio of component (b) to component (a) is from 100:1 to 1:100.

6. A composition of Claim 5 wherein the weight ratio of component (b) to component (a) is from 10:1 to 1:10.

AMENDED SHEET (ARTICLE 15)

7. A composition of Claim 6 wherein the weight ratio of component (b) to component (a) is from 5:1 to 1:1.

8. A method for controlling a plant disease caused by a fungal plant pathogen comprising applying to the plant or portion thereof a fungicidally effective amount of the mixture of any one of Claims 1 to 3.

9. A method of Claim 8 wherein the fungal plant pathogen is *Pyrenophora teres*.

10. A method of Claim 8 wherein the fungal plant pathogen is *Septoria tritici*.

AMENDED SHEET (ARTICLE 19)

TÍTULO~~nnn~~

20

MEZCLAS FUNGICIDAS DE DERIVADOS DE TIOFENOCAMPO DE LA INVENCIÓN

Esta invención se relaciona con mezclas fungicidas de un cierto derivado de tiofeno y a composiciones que comprenden tales mezclas y métodos para utilizar tales mezclas como fungicidas.

ANTECEDENTES DE LA INVENCIÓN

El control de las enfermedades de plantas causadas por patógenos de plantas de hongos es extremadamente importante para lograr una alta eficiencia en las cosechas. El daño de enfermedades de plantas a las plantas ornamentales, vegetales, plantas de campo, de cereales, y árboles frutales puede causar la reducción significativa en la productividad y por lo tanto resultar en costos incrementados para el consumidor. Muchos productos están disponibles para estos comercialmente propósitos, pero continúa la necesidad de tener nuevas mezclas y composiciones que sean más efectivas, menos costosas, menos tóxicas, más seguras para el ambiente, o que tengan diferentes modos de acción.

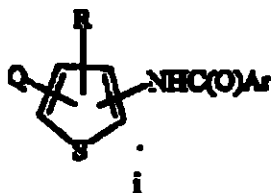
Los fungicidas que controlan de manera efectiva los hongos en las plantas están en constante demanda por los agricultores. Las combinaciones de fungicidas también son frecuentemente usadas para facilitar el control de las enfermedades y para retardar el

desarrollo de resistencia. Es deseable mejorar la actividad del espectro y la eficiencia del control de la enfermedad mediante usar mezclas de ingredientes activos que suministren una combinación de control curativo, sistémico y preventivo de patógenos de las plantas. También es deseable tener combinaciones que suministren un control residual mayor para permitir los intervalos de rociado más extendidos. También es muy deseable combinar agentes fungicidas que inhiban diferentes caminos bioquímicos en los patógenos fúngicos para retardar el desarrollo de la resistencia de cualquier agente de control de enfermedad de planta particular. 21

Siendo capaz de reducir la cantidad de agentes químicos liberados en el ambiente mientras que se asegura una protección efectiva de las cosechas de las enfermedades causadas por patógenos de plantas es siempre lo deseable. Las mezclas de fungicidas pueden suministrar un control de enfermedades significativamente mejor que lo que podría predecirse con base en la actividad de los componentes individuales. Este sinergismo ha sido descrito como "la acción cooperativa de dos componentes de una mezcla, de modo que el efecto total es mayor o más prolongado que la suma de los efectos de los dos tomados independientemente" (ver P. M. L. Tames, *Neth. J. Plant Pathology* 1964, 70, 73-80).

Nuevos agentes fungicidas que son particularmente ventajosos para lograr uno o más de los objetivos anteriores continúan siendo una necesidad.

La Patente de Estados Unidos No. 5,747,518 describe ciertos compuestos derivados de tiofeno de Fórmula I como nuevos ingredientes activos fungicidas. v

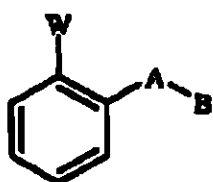


RESUMEN DE LA INVENCIÓN

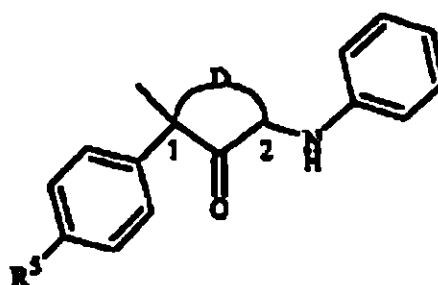
Esta invención se relaciona con una mezcla fungicida que comprende

(a) N-[2-(1,3-dimetilbutil)-3-tienil]-1-metil-3-(trifluorometil)-1H-pirazol-4-carboxamida (incluye todos los estereoisómeros) o una sal agrícolamente adecuada de ellos; y

(b) por lo menos un compuesto seleccionado del grupo que consiste de compuestos de Fórmula III o Fórmula IV que actúan en el complejo bc₁ del sitio de transferencia de electrón respiratorio mitocondrial fúngico;



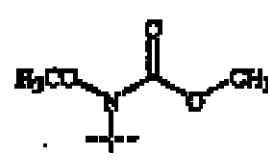
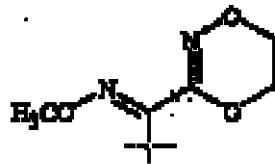
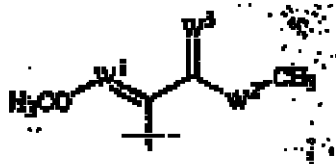
III



IV

en donde

W es uno del siguiente grupo



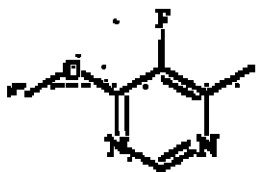
B

W^1 es N o CH_2 ;

W^2 es NH , O o CH_2 ;

W^3 is O o CH_2 ;

A es uno de los siguientes grupos (en donde el asterisco (*) está ligado al anillo fenilo de la Fórmula III),



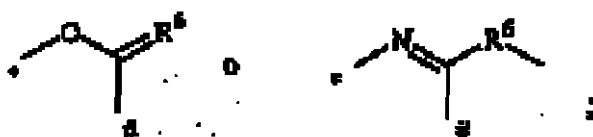
o



B es 1,2-propandion-bis(O-metiloxim)-1-ilo; o fenilo, fenoxi o piridinilo cada uno sustituido opcionalmente con 1 o 2 sustituyentes seleccionados entre Cl, CN, metilo o trifluorometilo; o

A y B juntos son $-CH_2O(1-[4\text{-clorofenil}]-\text{pirazol-3-ilo})$ o $-CH_2O(0\text{-trifluorometil-2-piridinil})$.

D es uno de los siguientes grupos (en donde el asterisco (*) está ligado al carbono siguiente al anillo fenilo de la Fórmula IV (carbono 1) y el signo número (#) está sujetado al carbono siguiente al nitrógeno de la Fórmula IV (carbono 2)).



24

R^4 es H o fenoxi; y

R^5 es O o S;

y sales agrícolaemente adecuadas de ellos; y opcionalmente

(c) por lo menos un compuesto seleccionado del grupo que consiste de compuestos que actúan entre la enzima desmetilasa del trayecto de la biosíntesis del esteroide y sales agrícolaemente adecuadas de ellos.

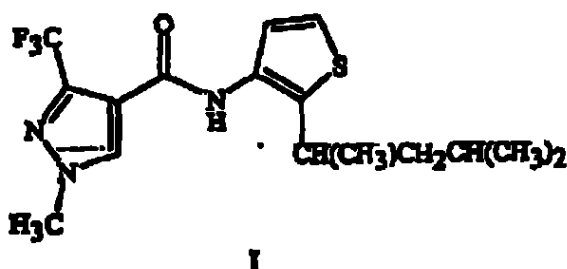
Esta invención también se relaciona con una composición fungicida que comprende una cantidad fungicidamente efectiva de una mezcla la invención y por lo menos un componente adicional seleccionado del grupo que consiste de tensioactivos, diluyentes sólidos y diluyentes líquidos.

Esta invención también se relaciona con un método para controlar enfermedades de plantas causadas por un patógeno de plantas fúngico que comprenden aplicar a la planta una porción de ella una cantidad fungicidamente efectiva de una mezcla de la invención (por ejemplo, como una composición descrita aquí).

DETALLES DE LA INVENCION

El compuesto N-[2-(1,3-dimetilbutil)-3-tienil]-1-metil-3-(trifluorometil)-1H-pirazol-4-carboxamida, alternativamente llamado 3-(trifluorometil)-1-metil-N-(2-(4-metilpentan-2-

lio)tiofen-3-il)-1H-pirazol-4-carboxamida y N-(2-(1,3-dimetilbutil)-3-tienil)-3-trifluorometil-1-metilpirazol-4-carboxamida, pueden ser representados por la Fórmula I:



Muchos compuestos en la mezcla de esta invención (por ejemplo el compuesto de fórmula I) pueden existir como uno o más estereoisómeros. Dependiendo de los compuestos varios estereoisómeros pueden incluir enantiómeros, diastereómeros, atropisómeros e isómeros geométricos. Una persona versada en la técnica podrá apreciar que un estereoisómero puede ser más activo y/o puede exhibir efectos benéficos cuando es enriquecido con relación a otro u otros estereoisómeros o cuando está separado de otros estereoisómeros. Adicionalmente, el artesano versado conoce como separar, enriquecer, y/o preparar de manera selectiva dichos estereoisómeros. Los compuestos de la invención pueden estar presentes en una mezcla de estereoisómeros, estereoisómeros individuales, o como una forma ópticamente activa.

Las sales agricolamente adecuadas de los compuestos en las mezclas de la presente invención incluyen las sales de adición de ácido con ácido inorgánicos u orgánicos tales como ácido bromhídrico, clorhídrico, nítrico, fosfórico, sulfúrico, acético, butírico, fumárico, láctico, maléico, malónico, oxálico, propiónico, salicílico, tartárico, 4-toluenosulfónico, o

valérico. Las sales agrícolaemente adecuadas de los compuestos en las mezclas de la presente invención también incluyen aquellas formadas con bases orgánicas (piridina, amonio o trietilamina) o bases inorgánicas (hidruros, hidróxidos, o carbonatos de potasio, litio, calcio, magnesio, o bario) cuando el compuesto contiene un grupo ácido tal como ácido carboxílico o fenol. 26

Modalidades de la presente invención incluyen:

Modalidad 1. Una mezcla en donde el componente (b) es piraclostrobina.

Modalidad 2. Una mezcla en donde el componente (b) es trifloxistrobina.

Modalidad 3. Una mezcla en donde el componente (b) es famoxadona.

Modalidad 4. Una mezcla que comprende por lo menos un compuesto seleccionado del grupo que consiste de compuestos que actúan en la enzima de desmetilasa del trayecto de la biosíntesis del esteroide y sales agrícolaemente adecuadas de ellos.

También son notorias como modalidades las composiciones fungicidas de la presente invención que comprenden una cantidad fungicidamente efectiva de una mezcla de las modalidades 1 a 4 y por lo menos un componente adicional seleccionado del grupo que consiste de tensoactivos, diluyentes sólidos y diluyentes líquidos. Las modalidades de la invención incluyen adicionalmente métodos para controlar enfermedades de plantas causados por

patógenos fúngicos de plantas que comprenden aplicar a la planta o porción de esta, o a la semilla de la planta o a plántula una cantidad fungicidamente efectiva de una mezcla de las modalidades 1 a 4 (por ejemplo, como una composición descrita aquí).

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El compuesto N-[2-(1,3-dimetilbutil)-3-tienil]-1-metil-3-(trifluorometil)-1H-pirazol-4-carboxamida puede ser preparado mediante uno o más de los métodos y variaciones de ellos según se describe en la Patente Estadounidense No. 5,747,518 (ver por ejemplo, Ejemplo 14).

Los compuestos herbicidas de los componentes (b) y (c) han descrito en patentes publicadas y en revistas y periódicos científicos. Muchos de estos compuestos están comercialmente disponibles como ingredientes activos en productos fungicidas. Estos compuestos se describen en compendios tales como *The Pesticide Manual*, 13^{ava} edición., C. D. S. Thomlin (Ed.), British Crop Protection Council, Surrey, UK, 2003. Estos grupos se describen adicionalmente más adelante.

Complejos Fungicidas bc₁ (componente (b))

Los fungicidas de estrobilurina tales como la fluoxastrobina, piraclostrobina y trifloxistrobina son conocidos por tener un modo de acción fungicida que inhibe el complejo bc₁ en la cadena de respiración mitocondrial (*Angew. Chem. Int. Ed.*, 1999, 38, 1328-1349). Otros fungicidas de estrobilurina adecuados como componentes (b) incluyen (2E)-2-(2-([6-(3-cloro-2-metilfenoxi)-5-fluor-4-pirimidinil]oxi)fenil)-2-(metoxiimino)-N-

metiletanamida, (2E)-2-(metoxoimino)-N-metil-2-(2-([{(1E)-1-(3-(trifluorometil)fenil)etiliden}amino)oxi]metil]fenil)etanamida, (2E)-2-metoxiimino)-N-metil-2-(2-[(E)-{(1-(3-(trifluorometil)fenil)etoxi]imino)metil]fenil)etanamida. Otros compuestos que inhiben el complejo bc₁ en la cadena de respiración mitocondrial incluyen la famoxadona y la fenamidona. Los complejos de bc₁ son llamados algunas veces con otros nombres en la literatura bioquímica, incluyendo complejo III de cadena de transferencia de electrón, y ubihidroquinona: citocromo c oxidoreductasa. Es identificado por el Número de Comisión de Enzima EC1.10.2.2. El complejo bc₁ está describe en, por ejemplo, J. Biol. Chem. 1989, 264, 14543-48; Methods Enzymol. 1986, 126, 253-71; y las referencias citadas allí.

Inhibidores de Desmetilasa en la Biosíntesis de Esterol
(componente (c))

Los inhibidores de la biosíntesis del esterol en el control de los hongos mediante inhibir las enzimas en el trayecto de la biosíntesis del esterol. Los fungicidas que inhiben la desmetilasa tienen un sitio común de acción dentro del trayecto de biosíntesis de esterol fúngico; esto es, la inhibición de la demetilación en la posición 14 de lanosterol o 24-metileno dihidrolanosterol, que son precursores de los esteroides en los hongos. Los compuestos que actúan en este sitio frecuentemente son llamados como inhibidores de desmetilasa, fungicidas DMI, o simplemente DMI. La enzima desmetilasa de llama algunas veces por otros nombres en la literatura bioquímica, incluyendo citocromo P-450 (14DM). La enzima desmetilasa está descrita en, por

ejemplo, *J. Biol. Chem.* 1992, 267, 13175-79 y las referencias citadas allí. Los fungicidas DMI caen en varias clases químicas: azoles (que incluyen triazoles e imidazoles), pirimidinas, piperazinas y piridinas. Los triazoles incluyen azaconazol, bromuconazol, ciproconazol, difenoconazol, diniconazol, (incluyendo el diniconazol-M), epoxiconazol, eteconazol, fenbuconazol, fluquinconazol, flusilazol, flutriafol, hexaconazol, imibenconazol, ipconazol, metconazol, miclebutenil, penconazol, propiconazol, prothioconazol, quiconazol, sineconazol, rebuconazol, tetraconazol, triadimefon, triadimenol, triticonazol y uniconazol. Los imidazoles incluyen clotrimazol, econazol, imazalil, isaconazol, miconazol, oxpoconazol, procloraz y triflumizol. Las pirimidinas incluyen fenacimol, nuarimol y triarimol. Las piperazinas incluyen triforina. Las piridinas incluyen butiobato y pirifenox. Las investigaciones bioquímicas han mostrados que todos los anteriores fungicidas mencionados son fungicidas de DMI como se describió por K. H. Kuck, et al. En *Modern Selective Fungicides - Properties, Applications and Mechanisms of Action*, H. Lyr (Ed.), Gustav Fischer Verlag: New York, 1995, 205-258.

Las descripciones de los compuestos disponibles comercialmente listados anteriormente pueden encontrarse en *The Pesticide Manual, Thirteenth Edition*, C. D. S. Tomlin (Ed.), British Crop Protection Council, 2003.

Esta invención suministra combinaciones de fungicidas que tienen diferentes modos bioquímicos de acción. Tales combinaciones pueden ser ventajosas particularmente en cuanto al manejo de la

resistencia, especialmente en donde los fungicidas de la combinación controlan a la misma o enfermedades diferentes. Ejemplos incluyen combinaciones del compuesto de Fórmula I con estrobilurinas tales como fluxastrobina, picoxistrobina, piraclostrobina y trifloxistrobina; y opcionalmente, DMIs tales como ciproconazol, epoxiconazol, fluquinconazol, flusilazol, hexaconazol, metconazol, propiconazol, protioconazol y tebuconazol.

La invención también suministra combinaciones de fungicidas que suministran un espectro expandido del control de la enfermedad o de la eficacia mejorada, incluyendo el control residual, curativo o preventivo mejorado. Ejemplos incluyen las combinaciones de los compuestos de Fórmula I con estrobilurinas tales como fluxastrobina, picoxistrobina, piraclostrobina y trifloxistrobina; y opcionalmente, DMIs tales como bromuconazol, ciproconazol, epoxiconazol, fluquinconazol, flusilazol, hexaconazol, metconazol, propiconazol, protioconazol y tebuconazol.

Esta invención también suministra combinaciones de fungicidas que son particularmente útiles para controlar enfermedades en los cereales (por ejemplo *Erysiphe graminis*, *Septoria nodorum*, *Septoria tritici*, *Puccinia recondita* y *Pyrenophora teres*). Ejemplos incluyen combinaciones del compuesto de Fórmula I con estrobilurinas tales como fluxastrobina, picoxistrobina, piraclostrobina y trifloxistrobina; y opcionalmente, DMIs tales como bromuconazol, ciproconazol, epoxiconazol, fluquinconazol, flusilazol, hexaconazol, metconazol, propiconazol, protioconazol

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y tebuconazol. De particular nota es el uso de estas combinaciones para controlar las enfermedades de la cebada (por ejemplo, *Pyrenophora teres*). 3

Esta invención también suministra combinaciones de fungicidas que son particularmente útiles para controlar enfermedades de los frutos y vegetales (*Alternaria solani*, *Botrytis cinerea*, *Rhizoctonia solani*, *Uncinula necator* y *Venturia inaequalis*). Ejemplos incluyen combinaciones del compuesto de Fórmula I con estrobilurinas tales como picoxistrobina, piraclostrobina y trifloxistrobina; y opcionalmente, DMIs tales como bromuconazol, ciproconazol, epoxiconazol, fluquinconazol, flusilazol, hexaconazol, metconazol, propiconazol, protioconazol y tebuconazol.

Las proporciones en peso del componente (b) al componente (a) en las mezclas y composiciones de la presente invención están típicamente entre 100:1 a 1:100, preferiblemente entre 25:1 a 1:25, y más preferiblemente entre 10:1 a 1:10. de nota son las mezclas y composiciones en donde la proporción en peso del componente (b) al componente (a) es de 5:1 a 1:1. Ejemplos de estas composiciones incluyen composiciones que comprende una mezcla del compuesto de Fórmula I con trifloxistrobina, una mezcla del compuesto de Fórmula I con fluoxastrobina, una mezcla del compuesto de Fórmula I con picoxistrobina, o una mezcla del compuesto de Fórmula I con piraclostrobina.

Se debe notar composiciones en donde el componente (b) y componente (c) están ambos presentes. Ejemplos son composiciones

que comprenden piraclostrobina o trifloxistrobina como componente (b), y por lo menos un componente compuesto (c). Se hace notar que tales composiciones en donde la proporción de peso general de los componentes (b) y (c) al componente (a) es de 100:1 a 1:100 y la proporción en peso del componente (b) al componente (a) es de 25 : 1 a 1:25. Se incluyen también las composiciones en donde la proporción en peso del componente (b) al componente (a) es de 5: 1 a 1: 1. Ejemplos de estas composiciones incluyen composiciones que comprenden mezclas del componente (a) con trifloxistrobina y un compuesto seleccionado del grupo que consiste de epoxiconazol y flusilazol.

Formulación/Utilidad

Mezclas de esta invención serán generalmente como una formulación o composición que comprende por lo menos un vehículo seleccionado de diluyentes líquidos, diluyentes sólidos y tens activos agricolamente adecuados. La formulación o los ingredientes de la composición son seleccionados para ser consistentes con las propiedades físicas del ingrediente activo, modo de aplicación y los factores ambientales tales como tipo de suelo, la humedad y temperatura. Las formulaciones útiles incluyen líquidos tales como soluciones (que incluyen concentrados emulsificables), suspensiones, emulsiones (que incluyen microemulsiones y/o suspoemulsiones) y similares que opcionalmente se pueden ser espesados para convertirse en geles. Las formulaciones útiles incluyen también sólidos tal como polvos, gránulos, glóbulos, tabletas, películas, y similares que pueden ser dispersables en agua ("humectables") o solubles en

agua. Los ingredientes activos pueden ser (micro) encapsulados y adicionalmente formados en una suspensión o formulación sólida; alternativamente la formulación completa del ingrediente activo puede ser encapsulada (o "sobrecubierta"). La encapsulación puede controlar o retardar la liberación del ingrediente activo. Formulaciones en rocío pueden ser extendidas a medios adecuados y usadas en volúmenes de rociado de entre uno a varios cientos de litros por hectárea. Las composiciones de alta fuerza son principalmente utilizadas como intermedios para las formulaciones adicionales.

Las formulaciones típicamente contendrán cantidades efectivas (por ejemplo de 0.01-99.99 por ciento en peso) de ingredientes activos junto con diluyentes y/o tensioactivos dentro de los siguientes rangos aproximados que agregan hasta el 100 por ciento en peso.

PORCENTAJE EN PESO

	<u>Ingrediente</u>	<u>Diluyente</u>	<u>Tensioactivo</u>
	<u>Activo</u>		
Gránulos, Tabletas u Polvos en			
Dispersables Agua y Solubles en	5-90	0-94	1-15
Agua, Tabletas y Polvo.			
Suspensiones, Emulsiones,			
Soluciones (incluyendo	5-50	40-95	0-25
Concentrados Emulsificables)			
Polvos	1-25	70-99	0-5
Gránulos y Glóbulos	0.01-99	5-99.99	0-15
Composiciones de Alta Fuerza	90-99	0-10	0-2

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Diluyentes sólidos típicos se describen en Watkins, et al., *Handbook of Insecticide Dust Diluents y Carriers*, 2nd edition, Dorland Books, Caldwell, New Jersey. Los diluyentes líquidos típicos se describen en Marsden, *Solvents Guide*, 2nd edition., Interscience, Nueva York, 1950. McCutcheon's *Detergents and Emulsifiers Annual*, Allured Publ. Corp., Ridgewood, Nueva Jersey, lo mismo que Sisely and Wood, *Encyclopedia of Surface Active Agents*, Chemical Publ. Co., Inc., Nueva York, 1964, una lista de tensoactivos y sus usos recomendados. Todas las formulaciones pueden contener cantidades menores de aditivos para reducir la espuma, la formación de pegue, la corrosión, el crecimiento microbiológico y similar, o espesantes para incrementar la viscosidad.

Los tensoactivos incluyen, por ejemplo, alcoholes polietoxilados, alquifenoles polietoxilados, ésteres de ácido graso de sorbitan polietoxilado, sulfosuccinatos de dialquilo, sulfatos de alquilo, sulfonatos de alquilbenceno, organosiliconas, N,N-dialquiltauratos, sulfonatos de lignina, condensados de formaldehído sulfonato naftaleno, policarboxilatos, y copolímeros de bloque polioxietileno/polioxipropileno. Los diluyentes sólidos incluyen, por ejemplo, arcillas, tal como bentonita, montmorillonita, atapulgita y caolín, almidón, azúcar, sílice, talco, tierras diatomáceas, urea, carbonato de calcio, carbonato y bicarbonato de sodio, y sulfato de sodio. Diluyentes líquidos incluyen, por ejemplo, agua N,N-dimetilformamida, sulfóxido de dimetilo, N-alquilpirrolidona, etilenglicol, polipropilenglicol, parafinas,

alquilbencenos, alquilnaftalenos, aceites de oliva, de castor, linaza, alefita, sésamo, maíz, cacahuete, semilla de algodón, soya, colza, y coco, ésteres de ácidos grasos, cetonas tal como ciclohexanona, 2-heptanona, isoforona y 4-hidroxí-4-metil-2-pentanona, y alcoholes tal como metanol, ciclohexanol, decanol y alcohol tetrahidrofurfurilo.

Las soluciones que incluyen concentrados emulsificables, pueden ser preparadas simplemente mezclando los ingredientes. Los polvos u tierras pueden ser preparados mediante mezclado y, usualmente, molido a artillo o en un molino energizado con fluido. Las suspensiones son usualmente preparadas por molido en húmedo; ver, por ejemplo, Patente Estadounidense 3,060,084. Los concentrados de suspensión preferidos incluyen aquellos que contienen, en adición al ingrediente activo entre 5 a 20% de tensoactivo no iónico (por ejemplo, alcoholes grasos polietoxilados) opcionalmente combinados con diluyentes líquidos en 50-65% y hasta 5% de tensoactivos aniónicos. Los gránulos y glóbulos pueden ser preparados mediante el rociado del material activo y sobre el vehículo granular preformado o mediante técnicas de aglomeración. Ver Browning, "Agglomeration", *Chemical Engineering*, Diciembre 4, 1967, pp 147-48, *Perry's Chemical Engineer's Handbook*, 4ª edición, McGraw-Hill, Nueva York, 1963, páginas E-57 y siguientes y WO 91/13546. Los glóbulos pueden ser preparados como se describió en la Patente Estadounidense 4,172,714. Los gránulos dispersibles en agua y solubles en agua pueden ser preparados como se enseña en la Patente Estadounidense 4,144,050, la Patente Estadounidense 3,920,442 y la Patente Alemana 3,246,493. Las tabletas pueden ser preparadas como se

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enseña en la Patente Estadounidense 5,180,587, Patente Estadounidense 5,232,701 y la Patente Estadounidense 5,208,030. Las películas se pueden preparar como se enseña en la GB 2,095,558 y Patente Estadounidense 3,299,566.

Para mayor información con respecto a La técnica de la formulación, ver la patente U.S 3,235,361, columna 6 línea 16 hasta la columna 7, línea 19 y los ejemplos 10-41; la patente U.S 3,309,192, columna 5, línea 43 a columna 7, línea 62 y los ejemplos 8,12,15,39,41,52,53, 58, 132,138-140,162-164,166,167 y 169-182; la patente U.S 2,891,855, columna 3 línea 66 a columna 5, línea 17 y los ejemplos 1-4; Klingman, Weed Control as a Science, John Wiley y Sons, Inc., New York, 1961, pp 81- 96; y Hance et al., Weed Control Handbook, 8th edition., Blackwell Scientific Publications, Oxford, 1989.

En los siguientes ejemplos, todos los porcentajes se en peso y todas las formulaciones son preparadas de maneras convencionales. El término "ingredientes activos" como se usa en los ejemplos A a E se refiere a la combinación de compuestos del grupo (a) y grupo (b), junto con cualquier otro u otros ingredientes activos presentes (por ejemplo, cualquier compuesto del grupo (e)). Sin mayor elaboración adicional, se cree que un experto en la técnica que utiliza la descripción precedente puede utilizar la presente invención a su completo alcance. Los ejemplos siguientes deben, por lo tanto ser constituidos únicamente como ilustrativos, y no limitantes de la descripción en ninguna forma. Los porcentajes son en peso excepto en donde se indique lo contrario.

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Ejemplo APolvo Humectable

Ingredientes activos	65.0%
Dodecifenil polietilenglicol éter	2.0%
Ligninsulfonato de sodio	4.0%
Silicoaluminato de sodio	6.0%
Montmorillonita (calcínada)	23.0%

Ejemplo BGránulos

Ingredientes activos	10.0%
Gránulos de atapulgita (baja materia volátil 0.71/0.30 mm; U.S.S. No. 25-50 tamices)	90.0%

Ejemplo CGlóbulos Extruidos

Ingredientes activos	25.0%
Sulfato de sodio anhidroso	10.0%
Lignisulfonato de calcio crudo	5.0%
Alquilnaftalenosulfonato de sodio	1.0%
Bentonita de calcio/magnesio	59.0%

Ejemplo D

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Concentrado emulsificable

Ingredientes activos	20.0%
Mezcla de sulfonatos solubles en aceite y polioxietilen éteres	10.0%
isoforona	70.0%

Ejemplo EGlóbulos Extruidos

Ingredientes activos	20.0%
Alcoholes grasos polietoxilados	15.0%
Éster derivado de cera de montana	3.0%
Lignisulfonato de calcio	2.0%
Copolímero de bloque poliglicol polietoxilado/polipropoxilado	1.0%
Propilenglicol	6.4%
Poli(dimetilsiloxano)	0.6%
Agente antimicrobial	0.1%
Agua	51.9%

Las composiciones de esta invención también se pueden mezclar con uno o más insecticidas, fugicidas, nematocidas, bactericidas, acaricidas, reguladores de crecimiento, químico esterilizantes, semio químicos, repelentes, atrayentes, feromonas, estimuladores de alimentación u otros compuestos

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biológicamente activos para formar un pesticida multi componente que le da un amplio espectro de protección agrícola. Ejemplos de tal protección agrícola con tales composiciones de esta invención que pueden ser formuladas son: insecticidas tales como abamectina, acefato, azinfos-metilo, bifentrin, -buprofezin, carbofuran, clorfenapir, clorpirifos, clorpirifos-metilo, ciflutrin, beta-ciflutrin, cihalotrin, lambda-cihalotrin, deltametrin, diafenthiuron, diazinon, diflubenzuron, dimetoate, esfenvalerato, fenoxicarb, fenpropatrin, fenvalerato, fipronil, flucitrinato, tau-fluvalinato, fonofos, imidacloprid, indoxacarb, isofenfos, malatión, metaldehido, metamidofos, metidatión, metomil, metopreno, metoxiclor, monocrotofos, oxamil, paratión, paratión-metilo, permetrin, forato, fosadona, fosmet, fosdamidón, pirimicarb, profenofos, rotenona, sulprofos, tebufenozida, teflutrin, terbufos, tetraclorvinfos, tiodicarb, trialometrin, triclofon y triflumuron; los fungicidas a parte de aquellos listados en el componente (b) y el componente (c), tales como acibenzolar-S-metilo, benalapsil (incluyendo benalapsil-^{am}), bentisbalar, benovil, blastocidin-s, mezcla bordó (sulfato de cobre tribásico), bucalip, butiolato, capoxpremit, captafol, captano, carbendacin, cloronet, clorotalonil, oxicloruro de cobre, sales de cobre, simoxenil, ciasofamid, ciflufenamid, ciprodimil, diclosimed, diclomesina, dicloran, dimetofos morfo, domina, edifenfos, etafosam, fenxamid, fenoxanil, fenticlonil, fentopridin, fentreprimob, acetato de pentino, hidróxido de pentino, flucicinan, fludioxonil, flumorb, flutolanil, fosetil-aluminio, furalapsil, fluremetapir, vasatina, imesazol, imino ortadina, hiprovenfos, hiprodiona, hiprovalicar, isoprotiolano, mangaseb, manet, fenoxan, metanapirin, metronil, matalapsil,

metratenona, meo-asocin (metaearsonato férrico) oxadisil, pensicuron, tricobenzamida, provenasol, propamocarb, proquinasib, pririmetanil, piroquilon, quinoxifen, siltiofan, piroxamina, azufre, tiabendazol, trifusamida, tiofanato-metilo, tiran, tiarinil, tolil fluanil, validamicina, diclosolina, y soxamida.

tolilfluanid, validamicina, vinclozolin y zoxamida; nematocidas tales como aldoxicarb y fenamifos; bactericidas tales estreptomicina; acaricidas tales como amitraz, quinometionat, clorobenzilata, cihexatina, dicofol, dienoclor, etoxazol, fenazaquina, óxido de fenbutatina, fenpropatrin, fenpiroximato, hexitiazox, propargita, piridaben y tebufenpirad; y agentes biológicos tales como *Bacillus thuringiensis*, *Bacillus thuringiensis* delta endotoxina, baculovirus, y bacteria entomopatogénica, virus y hongos. Las descripciones de diversos compuestos comercialmente disponibles listados anteriormente pueden encontrarse en The Pesticide Manual, Twelfth Edition, CD. S. Tomlin, ed., British Crop Protection Council, 2000. Para modalidades en donde uno o más de estos espacios de mezclado diversos usados, la proporción de peso de estos varios socios de mezclado (en total) frente a la cantidad total del componente (a) y el componente (b) está típicamente entre 100:1 y 1:3000. Se debe notar que las proporciones de peso entre 30:1 y 1:300 (por ejemplo proporciones entre 1:1 y 1:30). Será evidente que incluir estos componentes adicionales puede expandir el espectro de enfermedades controladas más allá del espectro controlado por la combinación del componente (a) componente (b) y el componente opcional (c) solos.

De particular interés son las composiciones que en adición al componente (a), componente (b) y el componente opcional (c), si está presente incluye (d) por lo menos un compuesto seleccionado del grupo que consiste de

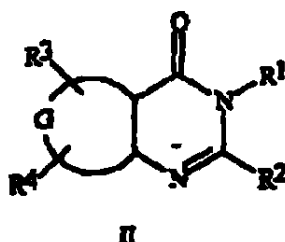
- (d1) fungicidas de alquilenobis(ditiocarbamato);
- (d2) cimoxanil;
- (d3) fungicidas de fenilamida;
- (d4) fungicidas de pirimidinona;
- (d5) clorotalonil;
- (d6) carboxamidas que actúan en el complejo II del sitio de transferencia de electrón respiratoria mitocondrial de hongos;
- (d7) quinoxifen;
- (d8) metrafenona;
- (d9) ciflufenamid;
- (d10) ciprodinil;
- (d11) compuestos de cobre;
- (d12) fungicidas de ftalimida;
- (d13) fosetil-aluminio;
- (d14) fungicidas de benzimidazol;
- (d15) ciazofamid;
- (d16) fluazinam;
- (d17) iprovalicarb;
- (d18) propamocarb;
- (d19) validamicina;
- (d20) fungicidas de dicarboximida de diclorofenilo;
- (d21) zoxamida; y
- (d22) dimetomorf; y

(d23) inhibidores de la biosíntesis del esteroI que no son DMI y

sales agricolamente adecuadas de los compuestos de (d1) a (d23)

Fungicidas de pirimidinona (grupo (d4))

Los fungicidas de pirimidinona incluyen compuestos de la Fórmula II



en donde

G forma un fenilo, tiofeno o anillo de piridina fundidos;

R¹ es alquilo con 1 a 6 carbonos;

R² es alquilo con 1 a 6 carbonos o alcoxi con 1 a 6 carbonos;

R³ es halógeno; y

R⁴ es hidrógeno o halógeno.

Los fungicidas de pirimidinona están descritos en la Solicitud de Publicación de Patente Mundial WO 94/26722, la Patente Estadounidense 6,066,638, la Patente Estadounidense 6,245,770, la Patente Estadounidense 6,262,058 y la Patente Estadounidense 6,277,858.

De notar los fungicidas pirimidinona seleccionados ^{NDA-44} del grupo:

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6-bromo-3-propil-2-propiloxi-4 (3H)-quinazolinona,
 6,8-di-yodo-3-propil-2-propiloxi-4 (3H)-quinazolinona,
 6-yodo-3-propil-2-propiloxi-4 (3H)-quinazolinona
 (proquinazid),
 6-cloro-2-propoxi-3-propiltieno[2,3-d]pirimidin-4 (3H)-ona,
 6-bromo-2-propoxi-3-propiltieno[2,3-d]pirimidin-4 (3H)-ona,
 7-bromo-2-propoxi-3-propiltieno[3,2-d]pirimidin-4 (3H)-ona,
 6-bromo-2-propoxi-3-propilpirido[2,3-d]pirimidin-4 (3H)-ona,
 6,7-dibromo-2-propoxi-3-propiltieno[3,2-d]pirimidin-4 (3H)-
 ona, y
 3-(ciclopropilmetil)-6-yodo-2-(propiltio)pirido[2,3-d]
 pirimidin-4 (3H)-ona.

Otros Grupos de Fungicidas

Los alquilenobis(ditiocarbamato)s (d1) incluyen los compuestos tales como mancozeb, maneb, propineb y zineb.

Las fenilamidas (d3) incluyen compuestos tales como metalaxil, benalaxil, furalaxil y oxadixil.

Las carboxamidas (d6) incluyen compuestos tales como boscalid, carboxin, fenfuram, flutolanil, furametpir, mepronil, oxicarboxin y tipluzamida que son conocidos por inhibir la función mitocondrial mediante romper el complejo II

(dehidrogenasa de succinato) en la cadena de transporte de electrones respiratorios.

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Los compuestos de cobre (d11) incluyen compuestos tales como oxiclорuro de cobre, sulfato de cobre e hidróxido de cobre, que incluye composiciones tales como las mezclas de Bordeaux (sulfato de cobre tribásico).

Ftalimidas (b15) incluyen compuestos tales como folpet y captan.

Fungicidas de benzimidazol (d14) incluyen benomil y carbendazim.

Fungicidas de dicarboxamida de diclorofenilo (d20) incluyen clozolinato, diclozolina, iprodiona, isovalediona, miclozolina, procimidona y vinclozolina.

Los inhibidores de biosíntesis del estero! que no son DMI (d23) incluyen la morfolina y los fungicidas de piperidina. Las morfolinás y piperidinás son inhibidores de la biosíntesis del estero! que han mostrado que inhiben las etapas en el trayecto de biosíntesis en el estero! en un punto más tarde que la inhibiciones logradas por la biosíntesis de estero! DMI (es decir, componente (c)). Las morfolinás incluyen aldimorf, dodemorf, fenpropimorf, tridemorf y trimorfamida. Las piperidinás incluyen el fenpropidin.

De notarse son tales composiciones en donde la proporción de peso general de los componentes (b), (d) y, si esta presente, (c)

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frente al componente (a) esta desde 100:1 a 1:100 y y la proporción de peso del componente (b) al componente (a) esta entre 25:1 a 1:25. Se incluyen las composiciones en donde la proporción de peso del componente (b) al componente (a) esta entre 5:1 a 1:1. 44

Las mezclas y composiciones de esta invención son útiles como agentes de control para enfermedades de plantas. La presente invención por lo tanto comprende adicionalmente un método para controlar las enfermedades de plantas causadas por patógenos de plantas fúngicos que comprenden aplicar a la planta o a una porción de ellas a ser protegidas o a la semilla de la planta o a las plantas a ser protegidas una cantidad efectiva de una mezcla de la invención o una composición fungicida que contiene dicha mezcla.

Las mezclas y composiciones de esta invención suministran control de enfermedades causadas por un amplio espectro de patógenos de fúngicos de plantas en las clases Basidiomiceto, Ascomiceto, Oomiceto y Deuteromiceto. Ellos son efectivos para controlar un amplio espectro de enfermedades de plantas, particularmente patógenos foliales de cosechas ornamentales, vegetales, de campo, cereales, y frutos.

Estos patógenos incluyen:

Oomicetos, que incluyen enfermedades de *Phytophthora* tales como *Phytophthora infestans*, *Phytophthora megasperma*, *Phytophthora parasitica*, *Phytophthora cinnamoni* y *Phytophthora capsici*;

enfermedades de *Pythium* tales como *Pythium aphanidermatum*; y enfermedades en la familia de los Peronosporaceae, tales como *Plasmopara viticola*, *Peronospora* spp. (incluyendo *Peronospora tabacina* y *Peronospora parasitica*), *Pseudoperonospora* spp. (incluye *Pseudoperonospora cubensis*), y *Bremia lactucae*; 45

Ascomycetes, incluyen enfermedades de *Alternaria* tal como *Alternaria solani* y *Alternaria brassicae*; enfermedades de *Guignardia* tal como *Guignardia bidwelli*; enfermedades de *Venturia* tal como *Venturia inaequalis*; enfermedades de *Septoria* tal como *Septoria nodorum* y *Septoria tritici*; enfermedad de moho tal como *Erysiphe* spp. (incluye *Erysiphe graminis* y *Erysiphe polygoni*), *Uncinula necator*, *Sphaerotheca fuliginea*, y *Podosphaera leucotricha*; *Pseudocercospora herpotrichoides*; enfermedad de *Botrytis* tal como *Botrytis cinerea*; *Monilinia fructicola*; enfermedad de *Sclerotinia* tal como *Sclerotinia sclerotiorum*; *Magnaporthe grisea*; *Phomopsis viticola*; enfermedad de *Helminthosporium* tal como *Helminthosporium tritici repens*; *Pyrenophora teres*; enfermedad de antracnosis tal como *Glomerella* o *Colletotrichum* spp. (tal como *Colletotrichum graminicola*); y *Gaeumannomyces graminis*;

Basidiomycetes, incluye enfermedades de la roya causadas por *Puccinia* spp. (tal como *Puccinia recondita*, *Puccinia striiformis*, *Puccinia hordei*, *Puccinia graminis*, y *Puccinia arachidis*); *Hemileia vastatrix*; y *Phakopsora pachyrhizi*;

otros patógenos incluyen *Rhizoctonia* spp (tal como *Rhizoctonia solani*); enfermedad de *Fusarium* tal como *Fusarium roseum*,

Fusarium graminearum, *Fusarium oxysporum*; *Verticillium dahliae*; *Sclerotium rolfsii*; *Rhynchosporium secalis*; *Cercosporidium personatum*, *Cercospora arachidicola* y *Cercospora batricola*; 46

y otros géneros y especies cercanamente relacionados con estos patógenos.

Adicionalmente a su actividad fungicida, la mezclas y composiciones también pueden tener actividad contra bacterias tales como *Erwinia amylovora*, *Xanthomonas campestris*, *Pseudomonas syringae*, y otras especies relacionadas.

Es de notar el uso de una mezcla de esta invención para controlar *Erysiphe graminis* (moho del trigo) especialmente utilizando una mezcla en donde el componente (b) es trifloxistrobina.

Se destacan también el uso de una mezcla de esta invención para controlar la *Septoria nodorum* (Septoriosis del trigo), especialmente utilizando una mezcla en donde el componente (b) trifloxistrobina.

Se nota también el uso de una mezcla de esta invención para controlar la *Pyrenophora teres* (borrón de la malla de la cebada), especialmente usando una mezcla en donde el componente (b) es trifloxistrobina.

Se nota también el uso de una mezcla de esta invención para controlar la *Puccinia recondita* (rolla de la hoja del trigo),

especialmente usando una mezcla en donde el componente (b) es trifloxistrobina.

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También se debe el notar el uso de una mezclas o composición de esta invención para suministrar el control de enfermedades causadas por un amplio espectro de patógenos fúngicos de plantas preventivamente o curativamente mediante aplicar una cantidad efectiva de la mezcla o composición sea pre o post infección.

El control de enfermedades de plantas se logra ordinariamente al aplicar una cantidad efectiva de una mezcla de esta invención sea pre o post-infección, a la porción de la planta que va a ser protegida tal como las raíces, tallo, follaje, frutos, semillas, tubérculos o bulbos, o al medio (tierra o arena) en el que la planta a ser protegida crece. La mezcla también se puede aplicar a la semilla para proteger la semilla y la plántula. Típicamente la mezcla se aplica en la forma de una composición que comprende al menos un componente adicional seleccionado del grupo que consiste de tensoactivos, diluyentes sólidos y diluyentes líquidos.

Los índices de aplicación para estos compuestos se pueden influenciar por muchos factores del ambiente y se deben determinar bajo condiciones de uso actual. El follaje normalmente se puede proteger cuando se trata en una proporción de menos de 1 g/ha a 5,000 g/ha total de ingrediente activo de grupos (a) y (b) en las mezclas y composiciones de la presente invención. Las semillas y plántulas normalmente se pueden proteger cuando la semilla se trata en una proporción de 0.1 a 10 g total de

ingredientes activos de los grupos (a) y (b) por kilogramo de semilla. 48

La mezcla de esta invención suministre un control ventajoso contra enfermedades fúngicas de las plantas cuando se compara al control logrado por cada uno de los componentes solos. Las mezclas de esta invención se ha encontrado que exhiben sinergia, particularmente en condición con el control de ciertas enfermedades tales como el borron de la mala de la cebada causada por *Pyrenophora teres*.

Se pueden utilizar los siguientes ensayos para demostrar la eficacia del control de las composiciones de esta invención en patógenos específicos. La protección de control de patógenos producidas por los compuestos no se limitan sin embargo a estas especies.

EJEMPLOS BIOLÓGICOS DE LA INVENCION

Las suspensiones de ensayo que comprendan un ingrediente activo sencillo se rocian para demostrar la eficacia de control del ingrediente activo individualmente. Para demostrar la eficacia de control de una combinación, (a) los ingredientes activos se pueden combinar en las cantidades apropiadas en una suspensión de ensayo sencilla, (b) las soluciones de depósito de ingredientes activos individuales se pueden preparar y luego combinar en la proporción apropiada, y diluir a la concentración final deseada para formar una suspensión de ensayo o (c) ensayar

00061

suspensiones que comprenden ingredientes activos sencillos que se pueden rociar secuencialmente en la proporción deseada

49

Composición 1

Ingredientes	Por ciento en peso
El compuesto de fórmula 1 preparado como un 20% de concentrado de suspensión	200 gramos de activo/litro

Composición 2

Ingredientes	Por ciento en peso
Trifloxistrobina (fungicida TWIST), formulado como un concentrado en suspensión	125 gramos de activo/litro

Composición 3

Ingredientes	Por ciento en peso
Epoxiconazol (fungicida OPUS), formulado como un concentrado emulsificable	125 gramos de activo/litro

Las composiciones de ensayo se mezclan con agua. Las suspensiones de ensayo resultantes se utilizan luego en los siguientes ensayos de campo. Las suspensiones de ensayo se rocían en un volumen de 200 litros por hectárea. Las tasa de aplicación son de 50, 100, y 200 gramos por hectárea.

ENSAYO A

Gráficos de campo establecidos de trigo de invierno (cv. 'Onvanlis') son rociados cuando el nodo 4 esta por lo menos 2 cm

110062

por encima del nodo 3 en las plantas de trigo y nuevamente cuando la hoja bandera esta completamente emergida. Cuatro replicas son usadas. Los gráficos son evaluados mensualmente para controlar los síntomas del borron de la hoja del trigo causados por la *Septoria tritici*. Los resultados son reportados en promedio de la media de las cuatro replicas.

ENSAYO B

Los gráficos de campo establecidos para la cebada de invierno (cv. 'Esterel') son rociados cuando la hoja bandera de la planta de cebada esta emergida completamente. Tres replicas son usadas. Los gráficos son evaluados usualmente para el control de los síntomas del borron de malta de la cebada causado por la *Pyrenophora teres*. Los resultados son reportados como la media promedio en porcentaje de control de enfermedad de las tres replicas

La tabla A lista los resultados para los ensayos A y B. En la tabla A, una proporción de 100 indicados al 100% de control de enfermedad y una marcación de 0 indica que no hay control de enfermedad (controlación a los controles). Las columnas marcadas "Avg" indican el promedio de tres o de cuatro replicas.

Tabla A

Resultados de ensayo

Composición	Tasa de aplicación	Ensayo A		Ensayo B	
		Avg	Exp	Avg	Exp

	(g/ha)				
1	50	31	-	12	-
1	100	48	-	20	-
1	200	56	-	31	-
2	100	53	-	66	-
3	100	63	-	31	-
1+2	100+200	73	76	96	75
1+3	100+100	96	80	66	50
2+3	200+100	80	83	67	76

La tabla A muestra las mezclas y composiciones de la presente invención que demuestran un control ventajoso.

La presencia del efecto sinérgico entre dos ingredientes activos se ha establecido con la ayuda de la ecuación Colby (ver S. R. Colby, "Calculating Synergistic y Antagonistic Responses of Herbicide Combinations", Weeds, 1967, 15, 20-22):

$$p=A+B-\left[\frac{A \times B}{100}\right]$$

Utilizando el método de Colby, la presencia de una interacción sinérgica entre dos ingredientes activos se establece al calcular primero la actividad predicha, p, de la mezcla basada en actividades de los dos componentes aplicados solos. Si p es menor que el efecto establecido experimentalmente, ha ocurrido sinérgismo. En la ecuación anterior, A es la actividad fungicida en porcentaje de control de un componente aplicado solo en la proporción x. El término B es la actividad fungicida en porcentaje de control del segundo componente

aplicado en la proporción y. La ecuación estima p, la actividad fungicida de la mezcla de A en proporción x con B en proporción y si sus efectos son estrictamente aditivos y no ha ocurrido interacción. 52

Las columnas "Exp" en la tabla A indican el valor esperado para cada mezcla de tratamiento usando la expresión Colby. La muestra en la tabla A, la aplicación de la ecuación Colby revela que el control observado es mayor que el esperado para la mezcla de la composición 1 con la composición 2 (100 g/ha + 200 g/ha) para el ensayo B y para la mezcla de la composición 1 con la composición 3 (100 g/ha + 100 g/ha) tanto para el ensayo A como el ensayo B.

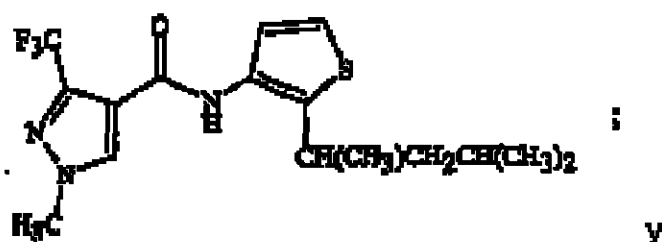
REIVINDICACIONES

53

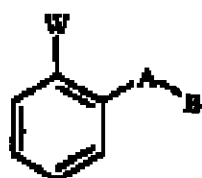
Lo que se reivindica es:

1. una mezcla fungicida que comprende:

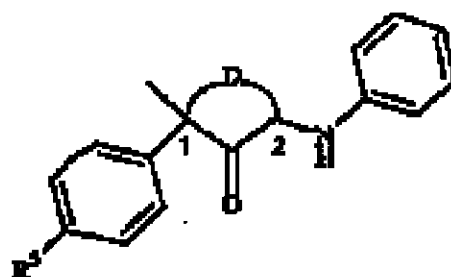
(a) el derivado de tiofeno de fórmula I o una sal agrícolamente adecuada del mismo



(b) Por lo menos un compuesto seleccionado del grupo que consiste de compuestos de Fórmula III o Fórmula IV que actúan en el complejo bc₁ del sitio de transferencia de electrón respiratorio mitocondrial fúngico;



III

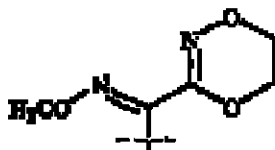
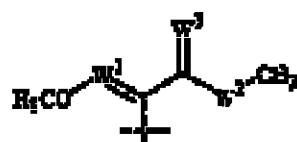


IV

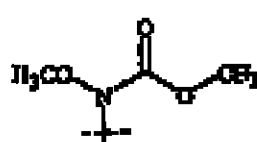
En donde

W es uno de los siguientes grupos

... (1111111111)



or



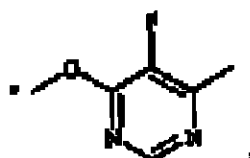
64

W¹ es N O CH;

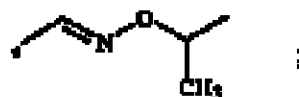
W² es NH, O o CH₂;

W³ es O o CH₂;

A es uno de los siguientes grupos (en donde el asterisco (*) está ligado al anillo fenilo de la Fórmula III),



or



B es 1,2-propanodiol-bis(0-metiloim)-1-il; o fenilo, fenoxi o piridinilo, cada uno se substituyó opcionalmente con 1 o 2 substituyentes seleccionados entre Cl, CN, metilo o trifluorometilo; o

A y B juntos son -CH₂O(I -[4-clorofenil]-pirazol-3-il) o -CH₂O(6- trifluorometil-2-piridinil) .

D es uno de los siguientes grupos (en donde el asterisco (*) está ligado al carbono próximo al anillo fenilo de la Fórmula IV (carbono 1) y el signo número (#) está sujetado al carbono próximo de la Fórmula IV (carbono 2)),

n99c7



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R^5 es a H o fenoxi:

y R^6 is C o S;

y sales agrícolamente adecuadas de las mismas; y opcionalmente

(c) al menos un compuesto seleccionado del grupo que consiste de compuestos que actúan en la enzima de desmetilasa del trayecto de la biosíntesis del esteroi y sus sales agrícolamente adecuadas.

2. Una mezcla de la reivindicación 1 en donde el componente (b) es trifloxistrobina

3. Una mezcla de la reivindicación 2 en donde el componente (c) está presente y es epoxiconazol.

4. Una composición fungicida que comprende una cantidad fungicidamente efectiva de la mezcla de cualquiera una de las reivindicaciones 1 a 3 y por lo menos un componente adicional seleccionado del grupo que consiste de tensoactivos, diluyentes sólidos y diluyentes líquidos.

5. Una composición de la reivindicación 4 en donde la proporción en peso del componente (b) al componente (a) es de 100:1 a 1:100.

6. Una composición de la reivindicación 5 en donde la proporción en peso del componente (b) al componente (a) es de 10:1 a 1:10.
7. Una composición de la reivindicación 6 en donde la proporción en peso del componente (b) al componente (a) es de 5:1 a 1:1.
8. Un método para controlar una enfermedad de las plantas causada por un patógeno fúngico de plantas que comprende aplicar a la planta o a la porción de la misma una cantidad fungicidamente efectiva de la mezcla de cualquiera de una de las reivindicaciones 1 a 5
9. Un método de la reivindicación 8 en donde el patógeno Un método de la reivindicación 8 en donde el patógeno de la planta es *Pyrenophora teres*.
10. Un método de la reivindicación 9 en donde el patógeno fúngico de la planta es *Septoria tritici*.
- 56

TO THE INTERNATIONAL BUREAU

Applicant(s):

E.I. DU PONT DE NEMOURS AND COMPANY

Attorney Docket No.:

BA9341PCT

International Application No.:

PCT/US2005/034254

International Filing Date:

SEPTEMBER 27, 2005

For:

FUNGICIDAL MIXTURES OF THIOPHENE DERIVATIVE

International Bureau of WIPO

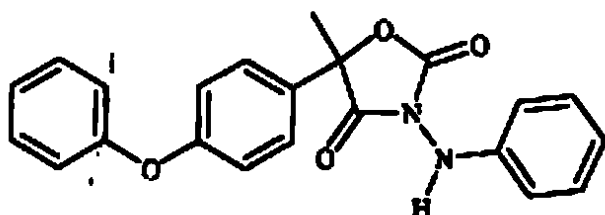
34 chemin des Colombettes

121 Geneva 20 Switzerland

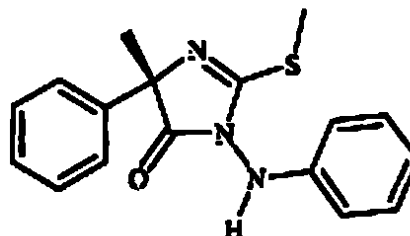
Fax (41-22) 740-14-35

Letter Accompanying Article 19 Amendment

The amendment requested is the substitution of application claim pages 18 through 20 filed herewith for application pages 18 through 20 as originally filed. Claim 1 is replaced with an amended Claim 1. It is noted that Formula IV in (b) of amended Claim 1 has been corrected on substitute page 18, to include a nitrogen atom ("N") at the position identified by "2" (i.e., nitrogen 2) and the Claim 1 recital of "D" on page 19 has been correspondingly corrected. These corrections are supported by the structures of famoxadone and fenamidone



Famoxadone



Fenamidonc

The other claims are unchanged with regard to wording.

Respectfully submitted,

David E. Heiser

E.I. du Pont de Nemours and Company

Legal, BMP25/1354

4417 Lancaster Pike

Wilmington, DE 19805

Telephone: (302) 892-1926

Fax: (302) 892-7949

Attachments

Date: February 16, 2006

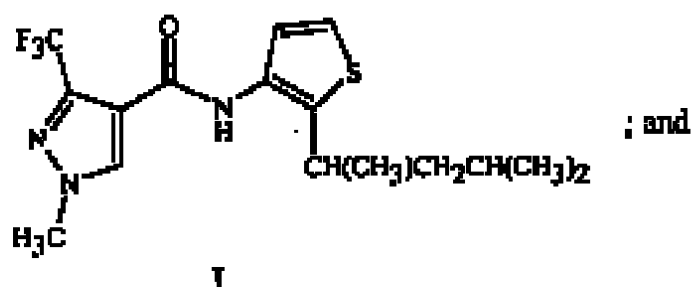
T:\Patent Documents\Ag Products\ADA-93\cc-BA9341\Article 19.doc

CLAIMS

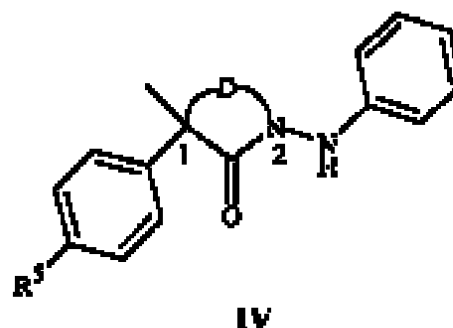
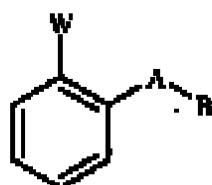
What is claimed is:

1. A fungicidal mixture comprising:

(a) the thiophene derivative of Formula I or an agriculturally suitable salt thereof

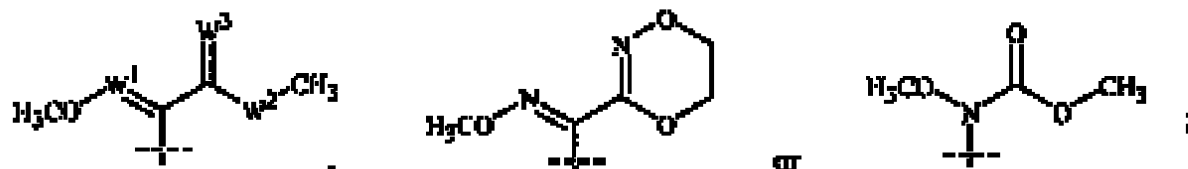


(b) at least one compound selected from the group consisting of compounds of Formula III or Formula IV which act at the bc_1 complex of the fungal mitochondrial respiratory electron transfer site;



wherein

W is one of the following group



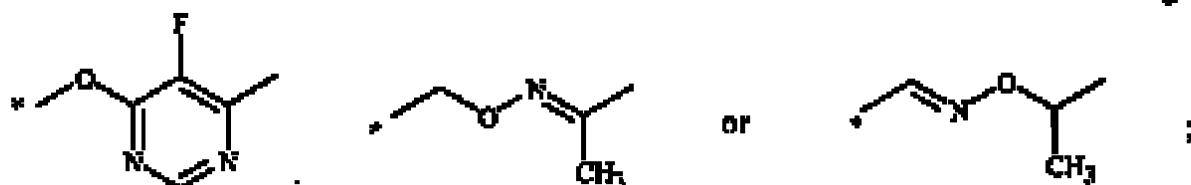
W¹ is N or CH;

W² is NH, O or CH₂;

W³ is O or CH₂;

A is one of the following groups (wherein asterisk (*) is attached to the phenyl ring of the Formula III),

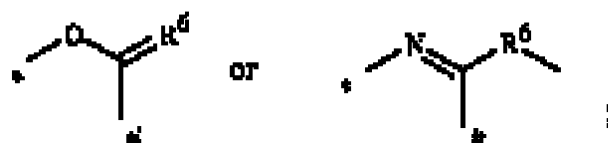
00071 



B is 1,2-propandion-bis(O-methyloxim)-1-yl; or phenyl, phenoxy or pyridinyl, each optionally substituted with 1 or 2 substituents selected from Cl, CN, methyl or trifluoromethyl; or

A and B together are $-\text{CH}_2\text{O}(1\text{-}[4\text{-chlorophenyl}]\text{-pyrazol-3-yl})$ or $-\text{CH}_2\text{O}(6\text{-trifluoromethyl-2-pyridinyl})$.

D is one of the following groups (wherein asterisk (*) is attached to the carbon next to the phenyl ring of the Formula IV (carbon 1) and pound sign (#) is attached to the ring nitrogen of the Formula IV (nitrogen 2)).



R^5 is H or phenoxy; and

R^6 is O or S;

and agriculturally suitable salts thereof; and optionally

(c) at least one compound selected from the group consisting of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof.

2. A mixture of Claim 1 wherein component (b) is trioxystrobin.

3. A mixture of Claim 2 wherein component (c) is present and is epoxiconazole.

4. A fungicidal composition comprising a fungicidally effective amount of the mixture of any one of Claims 1 to 3 and at least one additional component selected from the group consisting of surfactants, solid diluents and liquid diluents.

5. A composition of Claim 4 wherein the weight ratio of component (b) to component (a) is from 100:1 to 1:100.

6. A composition of Claim 5 wherein the weight ratio of component (b) to component (a) is from 10:1 to 1:10.

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7. A composition of Claim 6 wherein the weight ratio of component (b) to component (a) is from 5:1 to 1:1.
8. A method for controlling a plant disease caused by a fungal plant pathogen comprising applying to the plant or portion thereof a fungicidally effective amount of the mixture of any one of Claims 1 to 3.
9. A method of Claim 8 wherein the fungal plant pathogen is *Pyrenophora teres*.
10. A method of Claim 8 wherein the fungal plant pathogen is *Septoria tritici*.

. ~~041173~~**REIVINDICACIONES MODIFICADAS**

[(Recibido por la oficina Internacional el 16 de Febrero del 2006 (16.02.06); reivindicación original 1 modificada; el resto de reivindicaciones sin cambio]

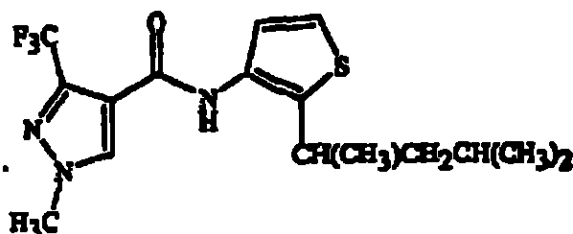
Lo que se reivindica es:

REIVINDICACIONES

Lo que se reivindica es:

1. una mezcla fungicida que comprende:

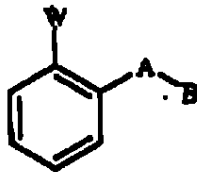
(a) el derivado de tiofeno de fórmula 1 o una sal agricolamente adecuada del mismo



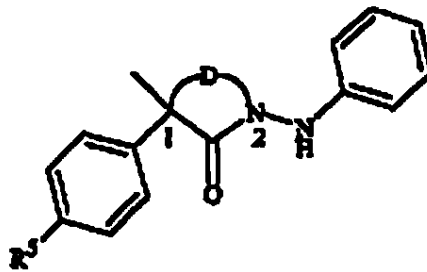
y

(b) Por lo menos un compuesto seleccionado del grupo que consiste de compuestos de Fórmula III o Fórmula IV que actúan en el complejo bc₁ del sitio de transferencia de electrón respiratorio mitocondrial fúngico;

HOJA CORREGIDA (ARTÍCULO 19)



III



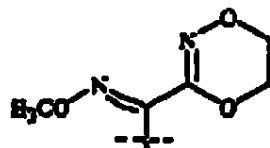
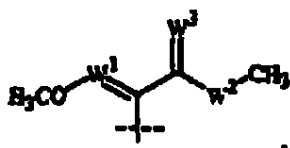
IV

no 74

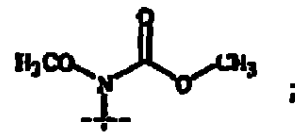
62

En donde

W es uno de los siguientes grupos



or

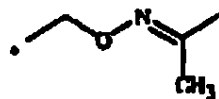
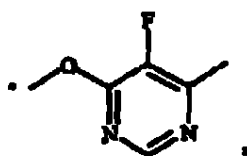


W¹ es N O CH₂;

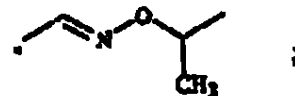
W² es NH, O o CH₂;

W³ es O o CH₂;

A es uno de los siguientes grupos (en donde el asterisco (*) está ligado al anillo fenilo de la Fórmula III),



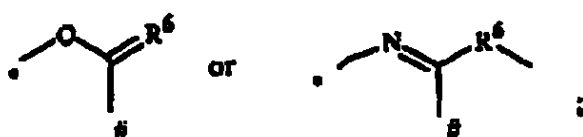
or



B es 1,2-propanodiol-bis(0-metiloxim)-1-il; o fenilo, fenoxi o piridinilo, cada uno se substituyó opcionalmente con 1 o 2 substituyentes seleccionados entre Cl, CN, metilo o trifluorometilo; o

A y B juntos son $-\text{CH}_2\text{O}(1-[4\text{-clorofenil}]\text{-piraz 1-3-il})$ o $-\text{CH}_2\text{O}(6\text{-trifluorometil-2-piridinil})$.

D es uno de los siguientes grupos (en donde el asterisco (*) está ligado al carbono próximo al anillo fenilo de la Fórmula IV (carbono 1) y el signo número (#) está sujetado al anillo de nitrógeno de la Fórmula IV (nitrógeno 2)),



R^5 es S H o fenoxi;

y R^6 es O o S;

y sales agrícolas adecuadas de las mismas; y opcionalmente

(c) al menos un compuesto seleccionado del grupo que consiste de compuestos que actúan en la enzima de desmetilasa del trayecto de la biosíntesis del esteroide y sus sales agrícolas adecuadas.

2. Una mezcla de la reivindicación 1 en donde el componente (b) es trifloxistrobina

3. Una mezcla de la reivindicación 2 en donde el componente (c) está presente y es epoxiconazol.

4. Una composición fungicida que comprende una cantidad fungicidamente efectiva de la mezcla de cualquiera una de

HOJA CORREGIDA (ARTÍCULO 19)

las reivindicaciones 1 a 3 y por lo menos un componente adicional seleccionado del grupo que consiste de tensoactivos, diluyentes sólidos y diluyentes líquidos.

5. Una composición de la reivindicación 4 en donde la proporción en peso del componente (b) al componente (a) es de 100:1 a 1:100.
6. Una composición de la reivindicación 5 en donde la proporción en peso del componente (b) al componente (a) es de 10:1 a 1:10.
7. Una composición de la reivindicación 6 en donde la proporción en peso del componente (b) al componente (a) es de 5:1 a 1:1.
8. Un método para controlar una enfermedad de las plantas causada por un patógeno fúngico de plantas que comprende aplicar a la planta o a la porción de la misma una cantidad fungicidamente efectiva de la mezcla de cualquiera de una de las reivindicaciones 1 a 3
9. Un método de la reivindicación 8 en donde el patógeno Un método de la reivindicación 8 en donde el patógeno de la planta es *Pyrenophora teres*.
10. Un método de la reivindicación 8 en donde el patógeno fúngico de la planta es *Septoria tritici*.

TENT COOPERATION TREATY

SPP
W/RTV
NAA29 5

From the INTERNATIONAL SEARCHING AUTHORITY

PCT

NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL SEARCH REPORT AND
THE WRITTEN OPINION OF THE INTERNATIONAL
SEARCHING AUTHORITY, OR THE DECLARATION

To:

E.I. DU PONT DE NEMOURS AND COMPANY
Attn. Heiser, David E.
Legal Patent Records Center
4417 Lancaster Pike
Wilmington, DE 19805
UNITED STATES OF AMERICA

RECEIVED
PATENT RECORDS CENTER

(PCT Rule 44.1)

Applicant's or agent's file reference FEB 07 2006 BA9241PCT	Date of mailing (day/month/year) 03/02/2006
International application No. PCT/US2005/034254	International filing date (day/month/year) 27/09/2005
Applicant E.I. DUPONT DE NEMOURS AND COMPANY	

1. ☒ The applicant is hereby notified that the international search report and the written opinion of the International Searching Authority have been established and are transmitted herewith.

Filing of amendments and statement under Article 19:
 The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 48):
 When? The time limit for filing such amendments is normally two months from the date of transmittal of the International Search Report.
 Where? Directly to the International Bureau of WIPO, 34 chemin des Colombettes
 1211 Geneva 20, Switzerland, Facsimile No.: (41-22) 338.82.70
 For more detailed instructions, see the notes on the accompanying sheet.

2. ☐ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect and the written opinion of the International Searching Authority are transmitted herewith.

3. ☐ With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

☐ no protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.
☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Reminders**

Shortly after the expiration of 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.

The applicant may submit comments on an informal basis on the written opinion of the International Searching Authority to the International Bureau. The International Bureau will send a copy of such comments to all designated Offices unless an international preliminary examination report has been or is to be established. These comments would also be made available to the public but not before the expiration of 30 months from the priority date.

Within 18 months from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later); otherwise, the applicant must, within 20 months from the priority date, perform the prescribed acts for entry into the national phase before those designated Offices.

In respect of other designated Offices, the time limit of 30 months (or later) will apply even if no demand is filed within 18 months.

See the Annex to Form PCT/IB/301 and, for details about the applicable time limits, Office by Office, see the PCT Applicant's Guide, Volume II, National Chapters and the WIPO Internet site.

Name and mailing address of the International Searching Authority European Patent Office, P.O. 5818 Postfach 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 eps nl Fax (+31-70) 340-3018	Authorized officer Sandrine Parriche
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KEY NOTED

These Notes are intended to give the basic instructions concerning the filing of amendments under article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article", "Rule", and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions respectively.

INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, when all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the letter to be published for the purposes of provisional protection or has another reason for amending the claims before international publication. Furthermore, it should be emphasized that provisional protection is available in some States only.

What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 20 or, where applicable, Article 41.

When?

Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 48.1).

Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 48.2).

Where a demand for international preliminary examination has been filed, see below.

How?

Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

777779 62

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]:
"Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
2. [Where originally there were 15 claims and after amendment of all claims there are 11]:
"Claims 1 to 15 replaced by amended claims 1 to 11."
3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:
"Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or
"Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
4. [Where various kinds of amendments are made]:
"Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 16, 16 and 17; new claims 20 and 21 added."

"Statement under article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. References to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

Consequences if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the same time of filing the amendments with the international Bureau, also file a copy of such amendments with the International Preliminary Examining Authority (see Rule 62.2(a), first sentence).

Consequences with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, where upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/selected Office, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/selected Office, see Volume II of the PCT Applicant's Guide.

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference BA9341PCT	FOR FURTHER ACTION see Form PCT/ISA/220 as well as, where applicable, Item 5 below.	
International application No. PCT/US2005/034254	International filing date (day/month/year) 27/09/2005	(Earliest) Priority Date (day/month/year) 27/09/2004
Applicant E. I. DUPONT DE NEMOURS AND COMPANY		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 4 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

- a. With regard to the language, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ The international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(e)).

- b. ☐ With regard to any nucleotide and/or amino acid sequence disclosed in the international application, see Box No. I.

2. ☐ Certain claims were found unsearchable (See Box II).

3. ☐ Unity of invention is lacking (see Box III).

4. With regard to the title,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the abstract,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 35.2(b), by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the drawings,

- a. the figure of the drawings to be published with the abstract is Figure No. _____

☐ as suggested by the applicant.

☐ as selected by this Authority, because the applicant failed to suggest a figure.

☐ as selected by this Authority, because this figure better characterizes the invention.

- b. ☐ none of the figures is to be published with the abstract.

A. CLASSIFICATION OF SUBJECT MATTER A01N43/56 //(A01N43/56,A01N37:50,A01N43:653),(A01N43/56,A01N43:76),(A01N43/56 A01N37:50) According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) A01N Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, WPI Data, PAJ, BIOSIS, CHEM ABS Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 737 682 A (MITSUI TOATSU CHEMICALS, INCORPORATED; MITSUI CHEMICALS, INC) 16 October 1996 (1996-10-16) cited in the application page 26; example 14 page 28; compound 1.13	1-10
A	TOMIYA, K.; YANASE, Y.: "MTF-753: a novel fungicide" CONGRESS PROCEEDINGS - BCPC INTERNATIONAL CONGRESS: CROP SCIENCE & TECHNOLOGY, GLASGOW, UNITED KINGDOM, NOV. 10-12, 2003, vol. 1, 2003, pages 99-104, XP009060687 Bracknell, UK page 99 page 102	1-10
-/-		
☒ Further documents are listed in the continuation of part C. ☒ Patent family members are listed in annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "I" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or after special reason (as specified) "C" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step upon the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "A" document member of the same patent family		
Date of the actual completion of the international search 30 January 2006		Date of mailing of the international search report 03/02/2006
Name and mailing address of the ISA European Patent Office, P.O. Box 5616 Patentstrasse 2 AL - 7200 HV Rijswijk Tel (+31-70) 340-2040, Tx 31 651 epo nl, Fax (+31-70) 340-2010		Authorized officer Romano-Götsch, R

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,A	WO 2005/077901 A (BAYER CROPSCIENCE SA; GOUOT, JEAN-MARIE; GROSJEAN-COURNOYER, MARIE-CLA) 25 August 2005 (2005-08-25) page 3, line 30 - page 4, line 5 page 6, lines 29,30 page 7, lines 6,7 - lines 20,21 page 28 - page 29; example 14	1-10
P,A	WO 2005/041653 A (BAYER CROPSCIENCE AKTIENGESELLSCHAFT; WACHENDORFF-NEUMANN, ULRIKE; DAH) 12 May 2005 (2005-05-12) page 21; compounds 6-4 page 95; table f	1-10
P,A	WO 2005/034628 A (BAYER CROPSCIENCE AKTIENGESELLSCHAFT; WACHENDORFF-NEUMANN, ULRIKE; DAH) 21 April 2005 (2005-04-21) page 53; examples E-5; table 5	1-10
E	WO 2005/122770 A (BAYER CROPSCIENCE AKTIENGESELLSCHAFT; KNEEN, GEOFF; SUTY-HEINZE, ANNE;) 29 December 2005 (2005-12-29) M=M2 and A=A1 page 1, line 8 - page 2, line 19 page 11; compounds 1-22 claim 3	1-10

INTERNATIONAL SEARCH REPORT

In no patent family members

Serial Application No

PC 1/US2005/034254

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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0737682	A	16-10-1996	CA 2173788 A1 12-10-1996
			CN 1146993 A 09-04-1997
			DE 69618370 D1 14-02-2002
			DE 69618370 T2 26-09-2002
			ES 2169773 T3 16-07-2002
			KR 201426 B1 15-06-1999
			US 5747518 A 05-05-1998
WO 2005077901	A	25-08-2005	NONE
WO 2005041653	A	12-05-2005	DE 10349501 A1 25-05-2005
WO 2005034628	A	21-04-2005	DE 10347090 A1 04-05-2005
WO 2005122770	A	29-12-2005	DE 102004029972 A1 05-01-2006

PCT COOPERATION TREATY

From the
INTERNATIONAL SEARCHING AUTHORITY

noted
72

To:

see form PCT/ISA/220

PCT

WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY (PCT Rule 43bis.1)

Date of mailing
(day/month/year) see form PCT/ISA/210 (second sheet)

Applicant's or agent's file reference
see form PCT/ISA/220

FOR FURTHER ACTION
See paragraph 2 below

International application No.
PCT/US2005/034254

International filing date (day/month/year)
27.09.2005

Priority date (day/month/year)
27.09.2004

International Patent Classification (IPC) or both national classification and IPC
A01N43/56

Applicant
E.I. DUPONT DE NEMOURS AND COMPANY

1. This opinion contains indications relating to the following items:

- ☒ Box No. I Basis of the opinion
- ☐ Box No. II Priority
- ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- ☐ Box No. IV Lack of unity of invention
- ☒ Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- ☒ Box No. VI Certain documents cited
- ☐ Box No. VII Certain defects in the international application
- ☐ Box No. VIII Certain observations on the international application

2. FURTHER ACTION

If a demand for international preliminary examination is made, this opinion will usually be considered to be a written opinion of the International Preliminary Examining Authority ("IPEA"). However, this does not apply where the applicant chooses an Authority other than this one to be the IPEA and the chosen IPEA has notified the International Bureau under Rule 68.1bis(b) that written opinions of this International Searching Authority will not be so considered.

If this opinion is, as provided above, considered to be a written opinion of the IPEA, the applicant is invited to submit to the IPEA a written reply together, where appropriate, with amendments, before the expiration of three months from the date of mailing of Form PCT/ISA/220 or before the expiration of 22 months from the priority date, whichever expires later.

For further options, see Form PCT/ISA/220.

3. For further details, see notes to Form PCT/ISA/220.

Name and mailing address of the ISA:



European Patent Office
D-80298 Munich
Tel. +49 89 2339 - 0 Tlx 523656 epmr d
Fax: +49 89 2339 - 4465

Authorized Officer

Romano-Götsch, R
Telephone No. +49 89 2339-8874



**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY**

International application No.
PCT/US2005/034254

Box No. I Basis of the opinion

1. With regard to the language, this opinion has been established on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.
☐ This opinion has been established on the basis of a translation from the original language into the following language , which is the language of a translation furnished for the purposes of international search (under Rules 12.3 and 23.1(b)).
2. With regard to any nucleotide and/or amino acid sequence disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:
 - a. type of material:
☐ a sequence listing
☐ table(s) related to the sequence listing
 - b. format of material:
☐ in written format
☐ in computer readable form
 - c. time of filing/furnishing:
☐ contained in the international application as filed.
☐ filed together with the international application in computer readable form.
☐ furnished subsequently to this Authority for the purposes of search.
3. ☐ In addition, in the case that more than one version or copy of a sequence listing and/or table relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.
4. Additional comments:

**WRITTEN OPINION OF THE
INTERNATIONAL SEARCHING AUTHORITY**

ncas 74
International application No.
PCT/US2005/034254

Box No. V Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-10
	No: Claims	
Inventive step (IS)	Yes: Claims	1-10
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-10
	No: Claims	

2. Citations and explanations

see separate sheet

Box No. VI Certain documents cited

1. Certain published documents (Rules 43bis.1 and 70.10)

and /or

2. Non-written disclosures (Rules 43bis.1 and 70.9)

see form 210

Re Item V.

Reference is made to the following documents:

D1 : EP 0 737 682 A (MITSUI TOATSU CHEMICALS, INC.) (1998-10-16)

D2 : Tomya, K.; Yanase, Y., Congress Proceedings - BCPC Int. Congress: Crop Science & Technology, Glasgow, UK, Nov.10-12,2003, vol. 1, 2003, pages 99-104,

The present application is directed to fungicidal mixtures comprising (a) penthiopyrad or an agriculturally suitable salt thereof and (b) at least one compound of Formula III or Formula IV which act at the bc1 complex of the fungal mitochondrial respiratory electron transfer site and optionally (c) at least one compound selected from the group of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof (claim 1). The application is also directed to a method for controlling a plant disease using said composition (claim 8).

Novelty

None of D1 or D2 is prejudicial to the novelty of present claims 1-10.

Document D1 discloses thiophene derivatives, among which penthiopyrad (see example 14), and their use as fungicide.

Document D2 is a report on the fungicidal activity of penthiopyrad and the identification of the complex II (succinate dehydrogenase) as target site. The subject-matter of independent claim 1 differs from D1 and D2 in that it is directed to mixtures comprising, in addition to penthiopyrad, the component (b) and optionally (c).

It follows that the composition and the method using said compositions, i.e the subject-matter of claims 1 and 8 and their respective dependent claims, meets the requirements of Art. 33(2)PCT.

Inventive Step

Departing from D2 as closest prior art, the problem to be solved is regarded as the provision of fungicidal mixtures containing penthiopyrad with enhanced efficacy at lower dosages (synergy).

The solution proposed in claim 1 the application is to combine penthiopyrad at least one compound of Formula III or Formula IV which act at the bc1 complex of the fungal mitochondrial respiratory electron transfer site, and optionally, at least one compound selected from the group of compounds acting at the demethylase enzyme of the sterol biosynthesis pathway and agriculturally suitable salts thereof, and use this combination for controlling fungal plant diseases.

There is no teaching in either D1 or D2 that renders obvious the combinations on file, because none of D1 or D2 suggest combining penthiopyrad with compounds which act at the bc1 complex of the fungal mitochondrial respiratory electron transfer site. Therefore, the solution proposed in present claims 1 and 8 (and their respective dependent claims) is considered as involving an inventive step (Article 33(3) PCT).

From the INTERNATIONAL BUREAU

PCT**NOTIFICATION CONCERNING
SUBMISSION OR TRANSMITTAL
OF PRIORITY DOCUMENT**

The

HEISER, David, E.
E. I. DU PONT DE NEMOURS AND COMPANY
LEGAL PATENT RECORDS CENTER
4417 Lancaster Pike
Wilmington, Delaware 19805
ETATS-UNIS D'AMERIQUE

(PCT Administrative Instructions, Section 411)

Date of mailing (day/month/year) 05 December 2005 (06.12.2005)	
Applicant's or agent's file reference BA8341PCT	IMPORTANT NOTIFICATION
International application No. PCT/US2005/034254	International filing date (day/month/year) 27 September 2005 (27.09.2005)
International publication date (day/month/year)	Priority date (day/month/year) 27 September 2004 (27.09.2004)
Applicant E.I. DUPONT DE NEMOURS AND COMPANY et al	

1. By means of this Form, which replaces any previously issued notification concerning submission or transmittal of priority documents, the applicant is hereby notified of the date of receipt by the International Bureau of the priority document(s) relating to all earlier application(s) whose priority is claimed. Unless otherwise indicated by the letters "NK" in the right-hand column or by an asterisk appearing next to a date of receipt, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).

2. (If applicable) The letters "NK" appearing in the right-hand column denote a priority document which, on the date of mailing of this Form, had not yet been received by the International Bureau under Rule 17.1(a) or (b). Where, under Rule 17.1(a), the priority document must be submitted by the applicant to the receiving Office or the International Bureau, but the applicant fails to submit the priority document within the applicable time limit under that Rule, the attention of the applicant is directed to Rule 17.1(d) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

3. (If applicable) An asterisk (*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b) (the priority document was received after the time limit prescribed in Rule 17.1(a) or the request to prepare and transmit the priority document was submitted to the receiving Office after the applicable time limit under Rule 17.1(b)). Even though the priority document was not furnished in compliance with Rule 17.1(a) or (b), the International Bureau will nevertheless transmit a copy of the document to the designated Offices, for their consideration. In case such a copy is not accepted by the designated Office as the priority document, Rule 17.1(c) provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

Priority date	Priority application No.	Country or regional Office or PCT receiving Office	Date of receipt of priority document
27 September 2004 (27.09.2004)	008113,430	US	16 November 2005 (16.11.2005)

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Authorized officer

Françoise HENNEQUIN (Fax 338 7010)

Facsimile No. (41-22) 338.70.10

Telephone No. +41 22 338 9935

Facsimile No. +41 22 338 82 70

Form PCT/IB/204 (January 2004)

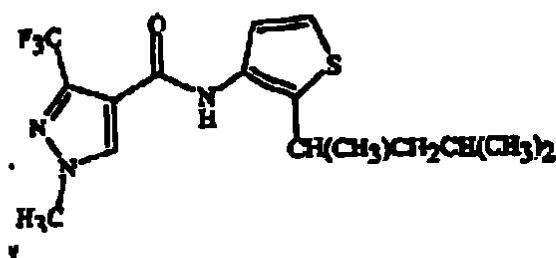
CNISVURY

 Industria y Comercio SUPERINTENDENCIA		SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO DIVISION DE NUEVAS CREACIONES EXTRACTO PARA PUBLICACIÓN SOLICITUD DE PATENTE DE INVENCION PCT		10-11-09
(21) N° de solicitud: (22) Fecha de solicitud: (30) Prioridad (31) No. 32) Fecha (33) País 80613,430 27/08/2004 US		(51) Int Cl: A01N 43/56, A01N 37/50, A01N 43/853, (71) Solicitante(s) E.I. DUPONT DE NEMOURS AND COMPANY (72) Inventor(es) FOUR, Stephen Ray (74) Apoderado: ALVARO CORREA ORDOÑEZ		
(85) Fecha límite inicio fase nacional: 27/04/07 (86) Datos relativos a la presentación de la solicitud PCT Fecha de presentación de la solicitud: 27/08/2005 No. de solicitud PCT/US2005/034254		(87) Publicación internacional Fecha: 08/04/2006 N° publicación: WO2006/036827		
(54) Título: MEZCLAS FUNGICIDAS DE DERIVADOS DE TIOFENO				

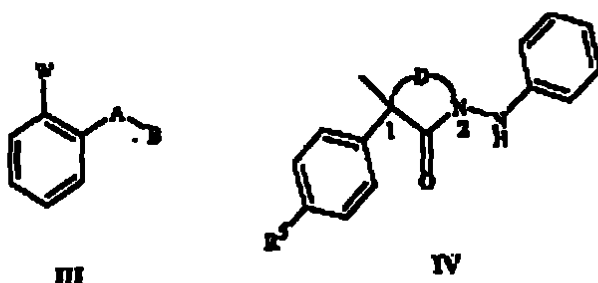
Reivindicación(es):

1. una mezcla fungicida que comprende:

(a) el derivado de tiofeno de fórmula 1 o una sal agrícolamente adecuada del mismo

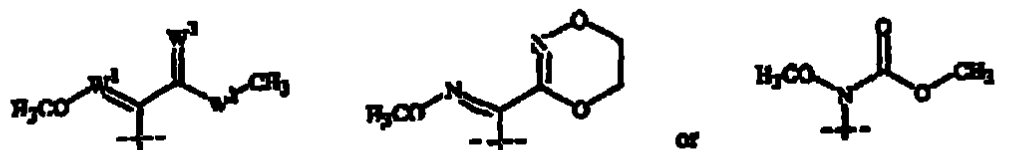


(b) Por lo menos un compuesto seleccionado del grupo que consiste de compuestos de Fórmula III o Fórmula IV que actúan en el complejo bc1 del sitio de transferencia de electrón respiratorio mitocondrial fúngico;



En donde

W es uno de los siguientes grupos

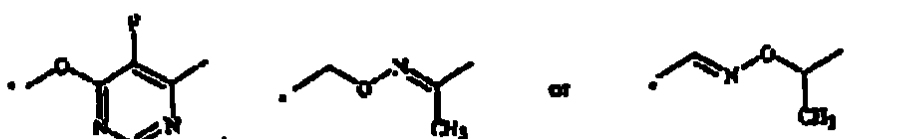


W1 es N O CH1

W2 es NH, O o CH2

W3 es O o CH2

A es uno de los siguientes grupos (en donde el asterisco (*) está ligado al anillo fenilo de la Fórmula III),



nnnyy

70

B es 1,2-propanodiol-bis(0-metoxim)-1-It; o fenilo, fenoxi o piridinilo, cada uno se sustituyó opcionalmente con 1 o 2 sustituyentes seleccionados entre Cl, CN, metilo o trifluorometilo; o

A y B juntos son -CH₂O(1-(4-clorofenil)-pirazol-3-il) o -CH₂O(6-trifluorometil-2-piridinil) .

C es uno de los siguientes grupos (en donde el asterisco (*) está ligado al carbono próximo al anillo fenilo de la Fórmula IV (carbono 1) y el signo número (#) está sujetado al anillo de nitrógeno de la Fórmula IV (nitrógeno 2)),



R⁶ es s H o fenoxi;

y R⁶ es O o S;

y sales agrícolamente adecuadas de las mismas; y opcionalmente

(c) al menos un compuesto seleccionado del grupo que consiste de compuestos que actúan en la enzima de desmetilasa del trayecto de la biosíntesis del esterol y sus sales agrícolamente adecuadas.

4. Una composición fungicida que comprende una cantidad fungicidamente efectiva de la mezcla de cualquiera una de las reivindicaciones 1 a 3 y por lo menos un componente adicional seleccionado del grupo que consiste de tensioactivos, diluyentes sólidos y diluyentes líquidos.

8. Un método para controlar una enfermedad de las plantas causada por un patógeno fúngico de plantas que comprende aplicar a la planta o a la porción de la misma una cantidad fungicidamente efectiva de la mezcla de cualquiera de una de las reivindicaciones 1 a 3

Reverso Folios 91 y 92 'Tayrtas'



Industria y Comercio
SUPERINTENDENCIA

Espacio reservado para el adhesivo de radicación

FORMULARIO UNICO DE CORRECCIONES Y MODIFICACIONES

①

SOLICITUD DE

☐

Corrección de Error Material

☒

Modificación de una solicitud

②

SOLICITANTE

Nombre: E.I. DUPONT DE NEMOURS AND COMPANY

Dirección: 1007 MARKET STREET, WILMINGTON, Delaware
19896

Nacionalidad o Domicilio: Delaware, Estados Unidos de América

Lugar de Constitución:

Teléfono:

Fax:

E-Mail:

IDENTIFICACION

C.C.

☐

NIT

☐

C.E.

☐

Otro

☐

Cual

Número

③

REPRESENTANTE O APODERADO

Nombre: ALVARO CORREA ORDOÑEZ

Dirección: Avenida E2 No.10-42 Piso 4to.

Teléfono: 634 1500

Fax: 376 2211

E-Mail: patents.bogota@bahasa.com

IDENTIFICACION

C.C.

☒

NIT

☐

C.E.

☐

Otro

☐

Cual

Número 79.143.366

TP 20.281

④

Número de expediente

⑤

Descripción de la corrección o modificación solicitada

Se presenta modificación voluntaria de la Fórmula IV

⑥

Comprobante de Pago No.

07-15147

Fecha FEB-20-2007

⑦

Anexos

☒

Comprobante de pago de la tasa.

☐

Podares, si teara al caso.

☐

Documento que acredite la existencia y representación legal de los partes, cuando sean personas jurídicas.

⑧

NOMBRE ALVARO CORREA ORDOÑEZ

FIRMA

C.C. 79.143.366

T.P. 20.281

Instrucciones para el diligenciamiento del presente formulario, ver reverso de esta página.

2000-F03

153 784
111111 10

SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO
Red: 07-029888-00000-0000
Pag: 2007-03-26 05:44:40 Cap. 2820 NUECREACIONES
Tel: 62 PATENTARIO
Bv: 373 FASENACIONAL
Act: 411 PRESENTACION Folio: 55

NIT: 000176009-2

RECIBO OFICIAL DE CASA : 7 - 19,047
FECHA : FEBRERO 20 DE 2007

***** COMBINACION *****

DEPOSITANTE	TIPO PAGO	BANCO	CUENTA	No. PAGH	FECHA PGO	Vr. PAGO
RAJSECK LARA ANDRIGUEZ	CONSIGNACION	BANCO POPULAR	390-00110-6	54016978	20/02/2007	34,823,000.00

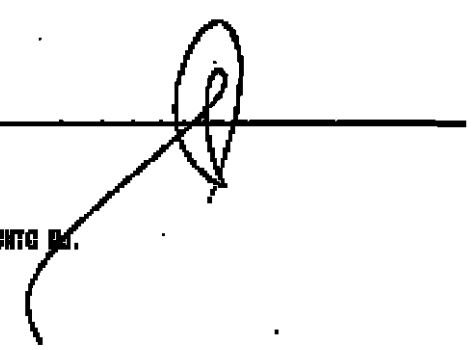
***** CONCEPTO *****

CONT. DETALLE	CONCEPTO	TOTAL CONCEPTO
1 36045-01-03 CAMBIO DE HUELADEO Y NOTIFICACION	12 MARCAS Y LEHAS COMERCIALES	99,000.00
	TOTAL :	99,000.00

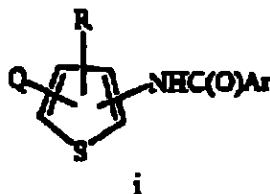
SEN: NOVENTA Y NUEVE MIL PESOS

RESPONSABLE :

RECIBO DE CASA APLICADO AL ASUNTO DE:



La Patente de Estados Unidos No. 5,747,518 describe ciertos compuestos derivados de tiofeno de Fórmula i como nuevos ingredientes activos fungicidas.

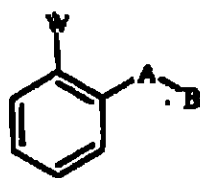


RESUMEN DE LA INVENCION

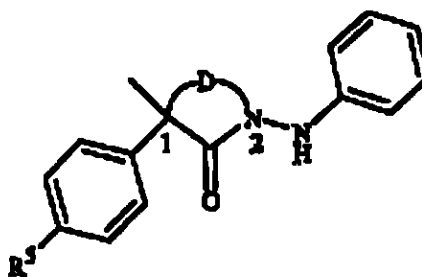
Esta invención se relaciona con una mezcla fungicida que comprende

(a) N-[2-(1,3-dimetilbutil)-3-tienil]-i-metil-3-(trifluorometil)-1H-pirazol-4-carboxamida (incluye todos los estereoisómeros) o una sal agrícolamente adecuada de ellos; y

(b) por lo menos un compuesto seleccionado del grupo que consiste de compuestos de Fórmula III o Fórmula IV que actúan en el complejo bc₁ del sitio de transferencia de electrón respiratorio mitocondrial fúngico;



III



IV

en donde

W es uno del siguiente grupo

(82)

REPÚBLICA DE COLOMBIA
SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO

Bogotá,
2020

Doctor(a)
CARMEN ORDÓÑEZ ALVARO

REFERENCIA	Radicación No.	7 29888
	Solicitud internacional	PCT/US2006/034254
	Fecha inicio fase nacional	27/04/2007
	Trámite	11
	Evento	378
	Actuación	430
	Oficio No.	000-5768

Como resultado del examen de forma, realizado con fundamento en el artículo 38 de la Decisión 486 de la Comisión de la Comunidad Andina, artículo 27 del Tratado de Cooperación en Materia de Patentes (PCT), regla 51 bis del Reglamento y Circular Unica de la Superintendencia de Industria y Comercio, se le comunica que:

1. La solicitud no contiene la copia del documento en que consta la cesión del derecho a la patente del inventor al solicitante o a su causante. (Art. 26-k) Decisión 486.

2. La solicitud no contiene el poder debidamente otorgado, con el lleno de los requisitos exigidos por los artículos 63 y subsiguientes del Código de Procedimiento Civil. Adicionalmente, deberá presentar el documento que acredita la existencia y representación legal de la sociedad solicitante, cuando ésta fuere persona jurídica.

En cumplimiento de lo dispuesto por el artículo 39 de la Decisión 486, se le requiere para que dentro del término de dos meses siguientes a la fecha de notificación del presente oficio, complete los requisitos anteriormente enunciados. En caso de no dar cumplimiento al presente requerimiento, la solicitud se considerará abandonada y perderá su prelación.

Notifíquese el presente oficio conforme a lo dispuesto en el numeral 5.2, literal a), del capítulo quinto del título primero de la Circular Unica No.10 de 2001.


ALEX CARMONA CRESPO DE VENGE
Jefe de la división de Nuevas Creaciones

El anterior requerimiento fue notificado por fijación en lista de fecha 11 Mayo 2007
adpall

