

AMENDED SPECIFICATION

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PATENT SPECIFICATION

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(54) HERBICIDAL COMPOSITIONS CONTAINING ISOPROPYL PHENYL DIMETHYL UREA

(71) We, PEPRO, Societe pour le Development et la Vente de Specialites Chimiques, a French Corporate Body, of 14/20 Rue Pierre Baizet, 68 Lyon (9e), France, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to the application of a phenylurea to the selective weed-control of certain crops.

According to the present invention there is provided a herbicidal composition for selective control of weeds in crops of cotton, soybean, maize and rice, comprising N - (4 - isopropylphenyl) - N',N' - dimethylurea mixed with at least one carrier and/or additive, the composition being substantially free of 2,3,6- or 2,3,5-trichlorobenzoic acid or 2,3,5,6-tetrachlorobenzoic acid.

Some dimethylphenylureas have been used against noxious plants for a long time. Practical examples are:

- 25 N - (3,4 - dichloro - phenyl) - N',N' - dimethylurea ("Diuron")
- N - (4 - chloro - phenyl) - N',N' - dimethylurea ("Monuron")
- 30 N - (3,4 - dichloro - phenyl) - N' - methyl, N' - butyl urea ("Neburon").

These compounds are commonly used for crop weed-control. The first two products, however, have comparatively poor selective properties and cannot, in any event, be used for the control of adventitious plants present among cereals. On the other hand, Neburon has good selective properties, and it is widely used for cleaning cereal crops, in particular wheat.

As a result of our research we have found that N - (4 - isopropyl - phenyl) - N',N' -

dimethylurea (hereinafter referred to as I.P.U.) has an overall activity which is comparable to, or often higher, than that of the most active of the above-mentioned ureas, while having a surprising selectivity towards many crops, both graminaceae and dicotyledones. These include, in particular, rice, maize, cotton and soya.

Our work, which has included the synthesis of I.P.U., has shown that this substance, in contradistinction to other phenylurea compounds which might all be expected to have the same or similar properties offers unexpected advantages over these related compounds which cannot all be applied to the selective destruction of adventitious plants in certain crops, either because they lack herbicidal activity, or their activity is too marked towards the crop itself. I.P.U. is definitely different from its homologues, as will be shown by the following examples.

I.P.U. may be produced by allowing p-isopropylphenyl isocyanate to react with dimethylamine in a benzene solution. The isocyanate may be prepared by any of the following processes:

Acylation of cumene and oxidation of the p-isopropylacetophenone thus obtained, followed by cyanation of the resulting chloride of p-isopropyl benzyl;

Nitration of cumene, followed by catalytic reduction and the action of phosgene on the resulting p-isopropylaniline.

Example I: Synthesis of I.P.U.

Dimethylamine (0.1 mole) dissolved in 25 ml of anhydrous benzene is introduced into a flask containing 0.04 mole of p - isopropylphenylisocyanate dissolved in 100 ml of anhydrous benzene. The reaction mixture is then boiled for one hour. On cooling, I.P.U. precipitates out as white crystals, which are

washed and vacuum dried.

Reaction yield: 92%. The melting point of

the resulting crystals is 157° C., and analysis gives the following results:

	Carbon	Hydrogen	Nitrogen
Calculated	69.90%	8.73%	13.59%
Found	69.64%	9.18%	13.62%

The selective herbicidal properties of I.P.U. have been demonstrated by various tests, in which this compound has been compared with its homologues. These tests were carried out as follows:—

- 10 Pots (10 × 10 × 15 cm) were filled with earth which had never been subjected to herbicidal treatment. Seeds of various species of plants, the sensitivity of which to the herbicides was to be tested, were placed on the earth. The seeds were then covered with a layer of earth, the depth of which depended on the diameters of the seeds. The herbicidal composition to be tested was then sprayed onto the pots. The composition consisted of a wettable powder prepared by mixing the following ingredients for one minute in a cutter-mill:

- 25 Active material to be tested 20%
Defloculating agent
(calcium lignosulphate) 5%
Wetting agent
(sodium alkarylsulphonate) 1%

Carrier (aluminium silicate) 74%

This wettable powder was then mixed with a quantity of water, calculated for a spraying operation at the required level per hectare.

In every test, an untreated reference plant was kept, so as to enable a check on possible growth inhibition, and also a possible lack of germination, or a defective growth of the plants owing to particular conditions.

The pots treated in this way were then kept for some time in a greenhouse under constant moisture, temperature and lighting conditions. At the end of five weeks, the results were noted, in particular by estimating the percentage of destruction of each species with respect to the reference plant. It is to be understood that, in the case of post-emergence, the spraying operation was only carried out after the plants had germinated and reached the 2-leaf stage.

The various crops and adventitious plants on which tests were carried out are as follows:—

Species	Scientific name	Symbol
Wild oats	<i>Avena fatua</i>	AV
Wheat	<i>Triticum vulgare</i>	BL
Maize	<i>Zea mays</i>	MA
Barnyard grass	<i>Echinochloa crus galli</i>	PA
Rice	<i>Oryza sativa</i>	RI
Rye grass	<i>Lolium italicum</i>	RA
Foxtail grass	<i>Alopecurus myosuroides</i>	VU
Amaranth	<i>Amaranthus retroflexus</i>	AM
Lamb's quarter	<i>Chenopodium album</i>	CH
White mustard	<i>Sinapis alba</i>	MO
Buckwheat	<i>Polygonum fagopyrum</i>	SA
Cotton	<i>Gossypium hirsutum</i>	CO
Soybean	<i>Glycine max</i>	SO

In every test, I.P.U. was compared with a number of related compounds, as well as with the best known herbicidal ureas which are now commercially available.

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Example 2: Pre-emergence trials.

The results obtained are shown in the following Table, in which the destruction percentages on species for every compound tested are shown. The active materials are given the following symbols:

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Active material

Isopropyl phenyl dimethylurea

Diuron

Mometron

Neburon

N - (3,4 - dichlorophenyl) - N'-methylurea

N - (4 - methoxyphenyl) - N',N'-dimethylurea

N - (isopropoxyphenyl) - N',N'-dimethylurea

Symbol

I.P.U.

A

B

C

D

E

F

15

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Active Material	Amount per Hectare	BL	MA	RI	SO	CO	AV	RA	PA	VU	AM	CG	MO	SA
I.P.U.	4 kg	0	0	0	0	40	100	100	100	100	100	100	100	100
	2 kg	0	0	0	0	0	100	100	100	100	100	100	100	100
	1 kg	0	0	0	0	0	100	98	98	98	98	100	98	100
	0.5 kg	0	0	0	0	0	98	90	95	50	98	100	70	75
A	4 kg	98	100	100	100	98	100	100	100	—	—	100	100	100
	2 kg	98	98	100	100	20	98	100	100	—	—	100	100	100
	1 kg	80	80	98	98	10	60	98	98	—	—	100	98	85
B	4 kg	95	100	100	100	80	100	100	100	—	—	100	100	100
	2 kg	95	100	100	100	80	100	100	100	—	—	100	100	100
	1 kg	50	50	95	95	10	98	98	100	—	—	100	100	95
C	4 kg	0	0	10	10	0	0	0	95	25	100	100	100	98
	2 kg	0	0	0	0	0	0	0	75	10	98	98	98	95
D	4 kg	85	100	100	100	0	100	100	100	100	100	100	100	100
	2 kg	50	100	100	100	0	98	100	100	75	100	100	100	95
E	4 kg	0	10	90	90	60	30	55	10	60	95	90	90	100
	2 kg	0	0	20	20	0	10	10	0	20	90	80	70	85
	1 kg	0	0	20	20	0	0	5	0	10	0	80	25	60
F	8 kg	0	0	2	0	90	15	5	88	10	97	100	98	95
	2 kg	0	0	0	0	80	0	0	0	0	85	100	25	5

Example 3: Post-emergence treatments by species for every compound tested
 The results obtained are given in the following Table. The active materials are given in the preceding Table, in which the destruction percentages are the same symbols as in the preceding Table.

Active Material	Amount/ha	BL	MA	RI	SO	CO	AV	RA	PA	VU	AM	CH	MO	SA
I.P.U.	2 kg	0	0	0	0	10	100	100	100	100	100	100	98	100
	1 kg	0	0	0	0	0	100	98	100	100	98	100	98	100
	0.5 kg	0	0	0	0	0	98	95	100	90	98	85	95	98
A	4 kg	100	100	100	100	20	100	100	100	100	100	100	98	100
	2 kg	100	99	100	99	0	100	100	100	100	100	100	96	100
	1 kg	100	80	100	99	0	100	100	100	100	100	100	90	100
B	4 kg	100	100	100	100	60	100	100	100	100	100	100	99	100
	2 kg	99	100	100	100	10	100	100	100	100	100	100	85	100
	1 kg	98	70	100	95	0	100	100	100	100	100	100	70	100
E	4 kg	0	0	30	40	98	2	50	60	85	99	100	97	100
	2 kg	0	0	0	10	95	0	40	50	25	95	99	85	70
F	2 kg	0	0	0	0	55	0	15	65	0	98	97	95	96

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These were diluted in a sufficient amount of water to give the desired quantity per hectare of the active material the efficiency of which was to be ascertained.

(4c) *Pre-emergence trials*

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Herbicide spraying was carried out between the 20th and the 30th of October, on wheat sown between the 16th and the 23rd October, depending upon the plots. The quantities used ranged from 0.5 kg to 3 kg per hectare.

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On reaching a level of 0.5 kg/ha, very good results were obtained on black grass (percentage destruction higher than 85%).

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From 2 kg/ha upwards, the destruction of practically all the adventitious graminaceae present in the crop (wild oats, black grass, rye grass, darnel and agrostis) was obtained. In every test, and with every trial, the selectivity towards wheat was very good.

Example 4: Open-air trials

During winter and spring 1970-1971, I.P.U. formed the subject of numerous tests in open fields. These tests were intended as confirmation of its ability for use under practical conditions, including factors such as the nature of the ground and atmospheric conditions, which are likely to modify significantly the results obtained and which are not taken into account in tests carried out in green-houses.

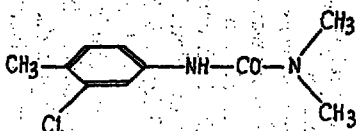
The tests were carried out on 20 square metre plots, both on pre- and post-emergence, using a wettable powder composed as follows:

I.P.U. 50%
Wetting agent (alkylnaphthalensulphonate) 1%
Defeculating agent (lignosulphite) 5%
Carrier (silica + china clay) 44%

(4b) Post-emergence trials

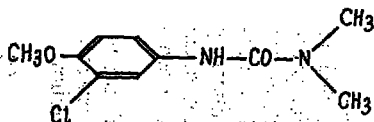
Spraying was carried out between the 23rd February and 1st April on crops sown between the 18th and the 28th October, at a time when the wheat growth was at the 2 to 4 tiller stage. Depending on the tests, amounts ranging from 1 to 3 kg/ha caused the complete destruction of the adventitious graminaceae (black grass, darnel, agrostis and wild oats) present among the wheat. The selectivity towards the wheat was always very good, except sometimes on chalky soil, where with an amount of 3 kg/ha a slight action on the wheat was noted.

In the above post-emergence tests, I.P.U. was compared with two related products commonly used for this type of application, viz.: N - (3 - chloro - 4 - methylphenyl) - N',N' - dimethylurea



and

N - (3 - chloro - 4 - methoxyphenyl) - N',N' - dimethylurea



In all tests, the amounts of I.P.U. required for a satisfactory destruction of the adventitious plants were 30% to 50% lower than those required for the two other compounds mentioned above.

It is clear from the above tests that I.P.U. is advantageous for herbicidal use, particularly with selected crops, in that their activity is always at least equal to that of the most active ureas tested, while their selectivity is at least equal to that of Neburon. If I.P.U. is compared more particularly with Neburon, it will be seen that I.P.U. has a much better activity on adventitious plants found very frequently among cereals, such as rye grass, wild oats, barnyard grass and black grass.

It should be noted that I.P.U. is characterized in that it can be used both before and after emergence.

To sum up, I.P.U. differs from the previously known ureas which have been used in that it has an outstanding selectivity towards some crops, such as rice, maize, cotton and soybean, besides which it has a very good activity on most of the adventitious plants

present in such crops. Under the same conditions, the other ureas are either highly herbicidal but not selective enough on such crops, or they are unsufficiently active on weeds.

In practice, I.P.U. will be used in amounts ranging from 0.2 to 5 kg/ha, depending on the nature of the treatment to be carried out, and the varieties and the growth of the adventitious plants.

In general, I.P.U. will be formulated according to the conventional methods used in the herbicide industry. The purpose of the formulation of the active materials is to provide easily-used compositions with maximum activity on the weeds to be destroyed. To this end, in general, a number of carriers and additives, depending on the type of formulation contemplated and the result desired, are added to the pure active material. The compositions thus obtained are either in liquid form (emulsions, true solutions, pastes, suspensions, and the like) ready to be used or to be diluted with water, or in solid form (wettable powders, granules and the like) to be used as such or to be diluted or dissolved in a liquid medium before use.

Such compositions include carriers, generally inert, and/or organic, mineral or mixed solvents, and/or emulsifying agents, adhesive agents, anti-caking agents, defloculating agents, and the like.

In particular, data concerning herbicidal formulations may be found in FRYER AND EVANS "Weed Control Handbook", 5th edition, page 101 *et seq.*

The Applicants make no claim to those solutions or suspensions of N - (4 - isopropylphenyl) - N',N' - dimethylurea in common solvents which are known in connection with the preparation or isolation of the compound.

Furthermore, although in some of the Examples and comparative tests described herein reference has been made to wheat and other crops, the present invention is limited to compositions for and methods of treating crops of cotton, soybean, maize and rice.

WHAT WE CLAIM IS:—

1. A herbicidal composition for selective control of weeds in crops of cotton, soybean, maize and rice, comprising N - (4 - isopropylphenyl) - N',N' - dimethylurea mixed with at least one carrier and/or additive, the composition being substantially free of 2, 3, 6- or 2, 3, 5-trichlorobenzoic acid or 2, 3, 5, 6-tetrachlorobenzoic acid.

2. A method for selectively controlling weeds in crops of cotton, soybean, maize or rice, comprising treating said crops, in pre- or post-emergence, with a herbicidal amount of a composition as defined in claim 1.

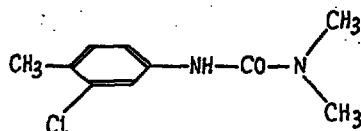
3. A method for the selective weed-control of crops of cotton, soybean, maize or rice, which comprises treating the crops either in



(4b) *Post-emergence trials*

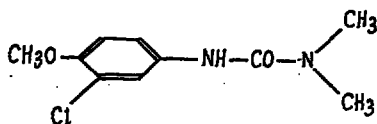
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The Applicants make no claim to those solutions or suspensions of N - (4 - isopropylphenyl) - N',N' - dimethylurea in common solvents which are known in connection with the preparation or isolation of the compound.

Furthermore, although in some of the Examples and comparative tests described herein reference has been made to wheat and other crops, the present invention is limited to compositions for and methods of treating crops of cotton, soybean, maize and rice.

WHAT WE CLAIM IS:—

1. A herbicidal composition for selective control of weeds in crops of cotton, soybean, maize and rice, comprising N - (4 - isopropylphenyl) - N',N' - dimethylurea mixed with at least one carrier and/or additive, the composition being substantially free of 2, 3, 6- or 2, 3, 5-trichlorobenzoic acid or 2, 3, 5, 6-tetrachlorobenzoic acid.

2. A method for selectively controlling weeds in crops of cotton, soybean, maize or rice, comprising treating said crops, in pre- or post-emergence, with a herbicidal amount of a composition as defined in claim 1.

3. A method for the selective weed-control of crops of cotton, soybean, maize or rice, which comprises treating the crops either in

- the pre-emergence or post-emergence stage or the land on which they are growing or are to grow with a herbicidal amount of a composition comprising N - (4 - isopropylphenyl)-N',N' - dimethylurea in a suitable carrier.
- 5 4. A method according to any of claims 1 to 3, wherein the crop is rice.
5. A method according to any of claims 1 to 3, wherein the crop is maize.
- 10 6. A method according to any of claims 1 to 3, wherein the crop is cotton.
7. A method according to any of claims 1 to 3, wherein the crop is soybean.
- 15 8. A cotton, soybean, maize or rice crop or crop area which has been treated by a method or with a composition as defined in any of the preceding claims.
9. The herbicidal composition according to claim 1 substantially as hereinbefore described.
10. A method for controlling weeds in crops as defined in any of the preceding claims substantially as herein described.
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