

PATENT SPECIFICATION

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(54) PROCESS FOR SELECTIVELY COMBATING WEEDS IN CROPS

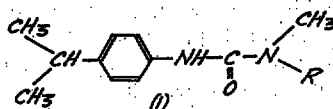
(71) We, FARBWERKE HOECHST AKTIENGESSELLSCHAFT, vormal's Meister Lucius & Brüning, a Body Corporate recognised under German Law, of 6230 Frankfurt (M)-Hoechst, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to the selective combating of broad-leaf weeds and weed grasses in cereal, cotton, peanut and leguminous plant crops.

Known herbicides, for example triazine, urea or phenoxycarboxylic acid derivatives, have had good results in a great number of agricultural crops in relation to the combating of broad-leaf weeds. However, there are difficulties in killing weed grasses, for example *Avena fatua* (wild oat) or *Alopecurus myosuroides* (foxtail grass), which grasses have spread to an extraordinary extent during recent years on account of the increase in cereal cultivation and the decrease in the cultivation of root crops.

Until now, the destruction of these and other weed grasses in cereals, i.e. in crop grasses, has been rather difficult. Though there are many herbicides, for example triallate or 2-chloro-3-(4'-chlorophenyl)-methylpropionate, which, as post- or pre-emergence agents, are effective especially against wild oat without substantially damaging wheat or barley, these agents are ineffective against broad-leaf weeds.

The present invention provides a process for selectively combating broad-leaf weeds and weed grasses in cereal, cotton, peanut and leguminous plant crops, which comprises treating the crop area with one or more herbicidal preparations which comprise a compound of the formula (I)



in which R represents a hydrogen atom or a methyl group, in admixture or conjunction with one or more formulation additives, the amount of the compound(s) of the formula (I) being sufficient to destroy the weeds while sparing the crop plants.

The above compounds of the formula (I) are both known compounds. United States Patent Specification No. 2,655,447 discloses a very large number of di- and tri-substituted urea compounds which possess herbicidal properties, the compounds used in the process of the present invention being mentioned therein.

The present invention is based on the discovery that the compounds of the formula (I) possess extremely advantageous properties. It has been found that these compounds can be used in amounts which, while exhibiting good herbicidal actions against weed-grasses and a series of broad-leaf weeds, spare a number of crop plants, especially cereals, for example wheat, barley, corn and rice, cotton, peanuts and leguminous plants, for example soybeans and dwarf-bush beans. The process of the present invention may be used to cause the complete destruction of undesired weed grasses even when the compound(s) of the formula (I) are applied in amounts which injure cereal crops to only an insignificant extent, if at all. This very important property of the herbicidal compounds of the formula (I) means that weeds and weed

grasses can be eliminated during crop cultivation, in both pre-emergence and post-emergence stages.

The herbicidal compounds of the formula (I) used in the process of the present invention may be applied in the form of wettable powders, emulsifiable concentrates, spray solutions, dusting powders or granules. Wettable powders are preparations which are homogeneously dispersible in water, and which, in addition to the active ingredient and a diluent or inert substance, contain also one or more wetting agents, for example polyoxyethylated alkylphenols, polyoxethylated oleyl or stearyl amines, or alkyl- or alkylphenyl-sulphonates, and one or more dispersing agents, for example the sodium salts of lignin-sulphonic acid, 2,2'-dinaphthyl-methane-6,6'-disulphonic acid, dibutyl-naphthalene-sulphonic acid, or oleylmethylaurine.

Emulsifiable concentrates may be obtained by dissolving the active ingredient in an organic solvent, for example butanol, cyclohexanone, dimethylformamide, xylene, or higher boiling aromatics. In order to obtain a good suspension or emulsion in water, one or more further wetting agents from the above series should be used.

Dusting powders are obtained by grinding the active ingredient with one or more finely divided solid substances, for example talcum or natural aluminates, for example kaolin, bentonite, pyrophyllite or diatomaceous earth.

Spraying solutions, commercially available as aerosol sprays, contain the active ingredient dissolved in an organic solvent, and a propellant, for example a mixture of hydrocarbon fluorides.

Granules may be obtained either by atomizing the active ingredient through a nozzle onto an absorptive granulated inert material, or by applying a concentrate of the active ingredient by means of an adhesive, for example polyvinyl alcohol, the sodium salt of polyacrylic acid or mineral oils, onto the surface of a carrier, for example, sand, kaolinities or granulated inert materials. The active substances may also be granulated by the methods used in the preparation of fertilizer granulated material, if desired in admixture with one or more fertilizers.

The concentration of the active ingredient(s) in the preparations may vary considerably. For example, in wettable powders the concentration of active ingredient may vary within the range of from 10% to 95% by weight, the remaining amount consisting of additives. In emulsifiable concentrates, the concentration of active ingredient may vary in the range of from 10% to 80% by weight. Dusting powders generally contain from 5 to 20% by weight, spray solutions from 2 to 20% by weight, of active ingredient.

In the case of granules, the content of active ingredient partially depends on the fact whether the active compound is liquid or solid and on what granulating agents, fillers and other additives are used.

For practical applications, the concentrates are usually diluted in usual manner, for example, in the case of wettable powders and emulsifiable concentrates, by means of water. Dust formulations, granulated preparations and spray solutions are not diluted with further inert substances before their application. The amount of active ingredient required for application varies in accordance with the external conditions, for example temperature and moisture; generally from 0.015 to 0.25 g/m², preferably from 0.03 to 0.12 g/m², of active substance is used. Furthermore, the herbicidal compounds of the formula (I) used in the process of the invention may be combined with other herbicides and/or soil insecticides. Known herbicides which may be suitable for combination with the compounds used in the process of the invention are, for example:

Urea derivatives:

linuron, chloroxuron, monolinuron, fluometuron, diuron;

Triazine derivatives:

simazine, atrazine, ametryne, prometryne, desmetryne, methoprotetryne;

Uracil derivatives:

lenacil, bromacil;

Pyrazon derivatives:

1-phenyl-4-amino-5-chloropyrazidone-(6);

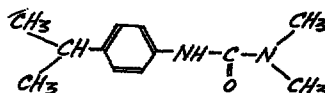
Growth-promoting preparations:

2,4-dichloro-phenoxy-acetic acid, 4-chloro-2-methyl-phenoxy-acetic acid, 2,4,5-trichloro-phenoxy-acetic acid, 4-chloro-2-methyl-phenoxy-butyric acid, 2,3,6-trichlorobenzoic acid;

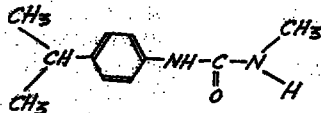
Carbamic acid derivatives:

barban, phenmedipham, triallate, diallate, vernolate and 2-chloro-allyl-N,N-diethyl-dithiocarbamate, Swep;

	Dinitrophenol derivatives:	dinitro- <i>o</i> -cresol, dinitro-sec.-butyl-phenol or dinoseb, dinoseb-acetate;	
	Chlorinated aliphatic acids:	sodium trichloro-acetate;	
5	Amides:	diphenamide, N,N-diallyl-chloro-acetamide;	5
	Dipyridilium derivatives:	paraquat, diquat, morfamquate;	
	Anilides:	N-(3,4-dichlorophenyl) - methacryl-amide, propanil, pentanochlor, monalide, 2-chloro-2' - 6' - diethyl - N - (methoxymethyl)-acetanilide, propachlor;	
10	Nitriles:	dichlobenil, ioxynil;	10
	Other preparations:	flurenol, 3,4-dichloropropionanilide, tri-fluralin, bensulide, 4-trifluoromethyl-2,4'-dinitrodiphenyl ether, monosodiummethyl-arsonate.	
15	Another application form for the active substances used in the process of the invention is in a mixture with one or more fertilizers, thus obtaining fertilizing agents which simultaneously have a herbicidal action.		
20	The compounds of the formula (I) show a very good activity against weed grasses which hitherto could be destroyed only with difficulty, and they are superior to known herbicides, for example fluometuron, monuron, diuron, terbutryne or triallate, with respect to their properties of sparing a number of important crop plants, for example wheat, barley, corn, rice, cotton, peanuts, leguminous plants and cabbage.		
	The following Preparations and Examples illustrate the invention; parts and percentages being by weight unless otherwise stated.		
25	Preparation 1		
	A wettable powder which is easily dispersible in water is obtained by mixing		
	25 parts of N-4-isopropylphenyl-N'-methyl-urea as active ingredient		
	64 parts of quartz containing kaolin as inert substance		
	10 parts of the potassium salt of ligninsulphonic acid and		
30	1 part of the sodium salt of oleylmethyltaurine as wetting and dispersing agent and by grinding the mixture in a disk attrition mill.		30
	Preparation 2		
	A dusting powder which is suitable for application as a herbicide is obtained by mixing		
35	10 parts of N-4-isopropylphenyl-N',N'-dimethyl-urea as active ingredient and 90 parts of talcum as the inert substance and by grinding the mixture in a cross beater mill.		35
	Preparation 3		
	An emulsifiable concentrate contains		
40	15 parts of N-4-isopropylphenyl-N'-methyl-urea		40
	75 parts of cyclohexanone as solvent and		
	10 parts of nonylphenol (10 EO) as emulsifier.		
	Example 1.		
45	Pre-emergence application		45
	Seeds of different weeds and crop plants each—i.e. separated by species—were sowed in pots filled with loam earth and then covered with a layer of earth of about 1 cm depth.		
50	On the day of sowing, aqueous suspensions of a wettable powder of the different preparations suspended in water according to Preparation 1 were sprayed on the surface of the soil. The concentrations are listed in Table I. The following products were used:		50
	(A)		

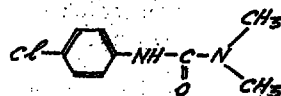


(B)



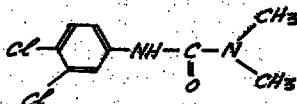
As comparative materials, suspensions of the following substances, prepared in an analogous manner, were used:

(C)



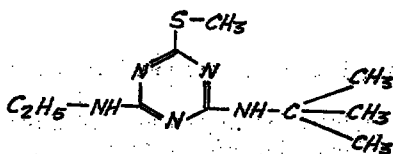
monuron

(D)



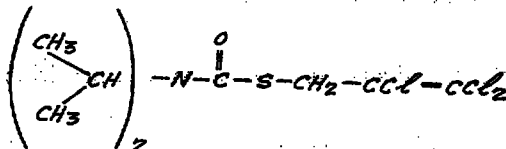
diuron

(E)



terbutryne

(F)



triallate

Triallate in the form of an aqueous emulsion was mixed with the top layer of earth.

Subsequently, the pots were put into a greenhouse. After 4 weeks, the effect of the agents on the weeds and crop plants was noted, expressed as the degree of damage in per cent, i.e. 100=complete destruction of the plants, 0=no damage. Details are listed in Table I.

TABLE I

Effect against weeds and crop plants in pre-emergence application (dosage in g of active substance per m² of soil surface)

Species of plants	Agents of invention		Comparative agents					
	A	B	C	D	E	F		
Preparation Dosage	0.015 0.03	0.015 0.03	0.015 0.03	0.015 0.03	0.015 0.03	0.015 0.03		
<u>Weeds (grasses)</u>								
<i>Avena fatua</i>	100 100	89 95	80 90	20 65	20 40	95 100		
<i>Alopecurus myosuroides</i>	100 100	95 100	75 80	90 100	80 100	40 70		
<i>Apera spicae-venti</i>	100 100	95 100	95 100	85 100	100 100	50 75		
<i>Poa annua</i>	100 100	100 100	100 100	100 100	90 100	20 30		
<u>Broad-leaf weeds</u>								
Mixture of the species listed immediately hereafter	85 95	75 90	95 100	90 100	90 100	0 0		
<u>Crop plants</u>								
<i>Triticum aestivum</i> (wheat)	10 30	0 10	60 70	45 90	15 25	10 10		
<i>Hordeum distichum</i> (barley)	0 20	0 10	36 65	40 80	10 20	0 0		

The values of Table I show that, with respect to the herbicidal activity against broad-leaf weeds or dicotyledons, in Table I a mixture of *Sinapis arvensis*, *Chrysanthemum segetum*, *Chenopodium album*, *Viola tricolor* and *Anthemis arvensis*, the compounds of the invention (A and B) give similarly good results as the chemically related comparative agents C and D or as the product E which is often used in cereals.

They preserve wheat and barley nearly as well as does the product E, while the preparations C and D cause too much damage to the cereals. Both the weed grasses *Poa* and *Apera* are destroyed by the products of the invention as they are by the comparative agents C, D and E. Especially in *Alopecurus myosuroides*, preparation A showed an excellent effect; also product B, at an amount of 0.03 g of active ingredient per m², was similar in effect to the comparative agents D and E, and superior to preparation C. Furthermore, A showed an excellent effect and B a good effect in *Avena fatua*; they were thus far superior to the comparative agents D and E. Comparative agent C had a good effect, but it caused damage to the crop plants. Comparative agent F is a special preparation for the destruction of *Avena* and *Alopecurus* in cereals, so its effect with respect to *Avena* was very good also in this case; however, with respect to *Alopecurus*, its effect at 0.03 g/m² was barely sufficient, and much too feeble with respect to most of the other weeds.

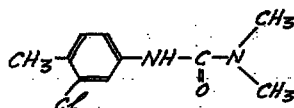
Table I shows that, in the pre-emergence application on cereals, the substances A and B of the invention, in addition to killing broad-leaf weeds or dicotyledons, sufficiently destroy weed grasses (gramineae) such as *Avena fatua* and *Alopecurus myosuroides*, the combating of which is more difficult, while the comparative agents either do not have a sufficient efficiency range or cause too much damage to the crop plants. In comparison with compound F, an advantage with respect to application technique residues in the fact that the substances used according to the invention need only be sprayed on the soil, while preparation F must be worked into the soil by mechanical means, in practice, for example, by means of a harrow, which means that an additional operation is necessary.

Example 2.

Pre-emergence application

In a similar manner as described in Example 1, seeds of *Alopecurus*, *Triticum* and *Hordeum* were sown in pots and treated on the day of sowing. In this case, the substances A and B (see Example 1), E (terbutryne) and the following product

(G)



chlortoluron

were used. Both the preparations terbutryne and chlortoluron were used as specific comparative agents for the destruction of *Alopecurus* in cereal crops.

The results after four weeks were as follows (see Table II):

TABLE II

Effect against weeds in crop plants in pre-emergence application
(dosage in g of active substance per m² of soil surface)

Species of plants	Preparation	Degree of damage in %			DL95 in Alopecurus
	Dosage	0.0075	0.015	0.03	
Alopecurus myosuroides	A	95	100	100	0.0075
	B	75	95	100	0.015
	E	25	85	100	0.0195
	G	65	90	95	0.023
Triticum aestivum	A	0	10	35	
	B	0	0	20	
	E	0	5	20	
	G	0	0	15	
Hordeum distichum	A	0	5	30	
	B	0	5	20	
	E	0	10	15	
	G	0	0	20	

A comparison of the substances A and B according to the invention with known standard products proved that A and B clearly are more effective against Alopecurus than the comparative products. When, for an exact comparison, reference is made to the dosage necessary for a 95% killing of the weeds, the values indicated under DL 95 in the last column of Table II are obtained (partially interpolated by means of logarithmic diagrams). When the value of the comparative agent G (chlortoluron) is set at 100 and the values of the other substances are brought into a corresponding relation to this value, the following relative numbers are obtained:

DL 95 in Alopecurus G (chlortoluron) = 100
 E (terbutryne) = 85
 preparation A = 35
 preparation B = 65

Thus, the substances A and B used according to the invention are applied in a dose of only 35% and 65%, respectively, of that necessary in the case of the comparative agent chlortoluron in order to achieve a sufficient destruction of the Alopecurus weed.

The data indicated in Table IV below for the damage of crop plants furthermore show that the preparations of the invention, in comparison with their efficiency against weeds, preserve wheat and barley to the same extent as do the comparative agents.

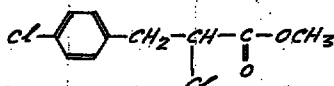
Example 3.

Post-emergence application

Seeds of the broad-leaf weeds *Sinapis arvensis*, *Chrysanthemum segetum*, *Chenopodium album*, *Viola tricolor* and *Anthemum arvensis*, seeds of the weed grass *Avena fatua* as well as of the crop plant *Triticum aestivum* (wheat) were sown in pots, the plants were brought to germination and, after 3 weeks, sprayed with aqueous suspensions of the preparations A and B according to the invention.

As comparative agent, a product emulsified in water was used containing the active ingredient 2-chloro-3-(4'-chlorophenyl)-methylpropionate:

(H)



Four weeks after the treatment, the test gave the results listed in the following Table III:

TABLE III

Preparation: Dosage: g/m ²	A		B	H	
	0.03	0.06	0.06	0.06	0.5
Weed:					
<i>Avena fatua</i>	95	100	95	10	95
Broad-leaf weed species:					
Mixture of different species	96	100	93	0	10
Crop plant:					
<i>Triticum aestivum</i>	0	10	0	0	0

These values show that the preparations of the invention are efficient also in post-emergence application; product A, at 0.03 g/cm² already being of good efficiency and, at 0.06 g/m², of excellent efficiency against broad-leaf weeds and wild oat. B showed nearly the same effects; the wheat was not damaged in either case.

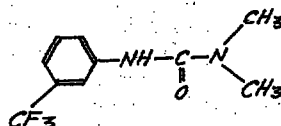
The comparative agent H, however, a special preparation for the destruction of wild oat after emergence, practically had no effect as a herbicide at 0.06 g/m². An eight-fold dosage of this agent was necessary to sufficiently combat the wild oat, and, even then, it had no effect on broad-leaf weeds.

Example 4.

Pre-emergence application

In an analogous manner as described in Example 1, weeds and crop plants were sown and treated. Again, preparations A and B of the invention were used. As comparative agent, the following product, which is often applied in cotton, was used:

(J)



fluometuron

The results, recorded four weeks after the treatment, are listed in the following Table IV:

TABLE IV

Effect against weeds and crop plants in pre-emergence application dosage in g of active substance per m² of soil surface

Preparation: Dosage:	A		B		J	
	0.03	0.12	0.03	0.12	0.03	0.12
Weeds:						
<i>Xanthium pungens</i>	95		100		40	
<i>Bidens pilosa</i>	100		95		30	
<i>Amaranthus retroflexus</i>	100		100		100	
<i>Chenopodium album</i>	100		95		85	
<i>Setaria faberii</i>	95		95		85	
<i>Eleusine indica</i>	100		100		85	
Crop plants:						
<i>Oryza sativa</i> (rice)	0	20	10	50	40	85
<i>Zea mais</i> (corn)	0	40	0	25	30	90
<i>Gossypium hirsutum</i> (cotton)	0	0	0	20	0	40
<i>Arachis hypogaea</i> (peanut)	0	10	10	50	30	80
<i>Glycine soja</i> (soybean)	0	50	0	60	60	90
<i>Phaseolus vulgaris</i> (dwarf-bush bean)	10	60	0	10	20	90

It is observed from the values given in Table IV that the substances A and B according to the invention have a good to very good effect in the cited weeds at a dose of 0.03 g/m². The effect of the comparative preparation J (fluometuron) in Xanthium and Bidens was too feeble. Also in this case it proved that the substances of the invention have a selective herbicidal effect against those weeds in cereals which hitherto could be killed only with difficulty, here in rice and corn and other crops.

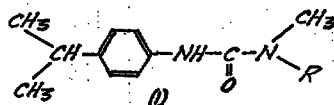
At a dose four times as high (0.12 g/m²), all products had a 100% effect against all weeds. Therefore, in this case, only the effect against crop plants is indicated in order to show how these plants react to an overdose of the corresponding preparations, since overdosing occurs often in daily practice.

From this test it is observed that the substances of the invention as well as fluometuron do not cause any damage, for example, to cotton when normally dosed (0.03 g/m²), but there is a substantial difference at an overdose: fluometuron clearly causes more damage to cotton than the substances of the invention.

These substances can also be applied in peanuts, soybeans and dwarf-bush beans, i.e. they do not cause damage to the cited crop plants when applied at a dose of 0.03 g/m², which dose is of good herbicidal efficiency. Even at an overdose, part of the crop plants remain practically undamaged. The substances may be applied with success also in peas, cabbage and tobacco, while the comparative agent caused too much damage to nearly all cited crops.

WHAT WE CLAIM IS:—

1. A process for selectively combating broad-leaf weeds and weed grasses in cereal, cotton, peanut and leguminous plant crops, which comprises treating the crop area with one or more herbicidal preparations which comprise a compound of the formula (I)



(I)

in which R represents a hydrogen atom or a methyl group, in admixture or conjunction with one or more formulation additives, the amount of the compound(s) of the formula (I) being sufficient to destroy the weeds while sparing the crop plants.

2. A process as claimed in claim 1, wherein the amount of compound of the formula (I) used is in the range of from 0.015 to 0.25 g/m².

3. A process as claimed in claim 2, wherein the amount of compound of the formula (I) used is in the range of from 0.03 to 0.12 g/m².

4. A process as claimed in any one of claims 1 to 3, wherein the or each herbicidal preparation also comprises one or more other herbicides and/or soil insecticides.

5. A process as claimed in any one of claims 1 to 4, wherein the or each herbicidal preparation also comprises one or more fertilizers.

6. A process for selectively combating broad-leaf weeds and weed grasses in cereal, cotton, peanut and leguminous plant crops as claimed in claim 1, conducted substantially as described herein.

7. A process for selectively combating broad-leaf weeds and weed grasses in cereal, cotton, peanut and leguminous plant crops as claimed in claim 1, conducted substantially as described in any one of the Examples.

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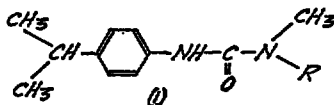
of the invention have a selective herbicidal effect against those weeds in cereals which hitherto could be killed only with difficulty, here in rice and corn and other crops. At a dose four times as high (0.12 g/m²), all products had a 100% effect against all weeds. Therefore, in this case, only the effect against crop plants is indicated in order to show how these plants react to an overdose of the corresponding preparations, since overdosing occurs often in daily practice.

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(I)

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3. A process as claimed in claim 2, wherein the amount of compound of the formula (I) used is in the range of from 0.03 to 0.12 g/m².

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