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## 3,385,692 Patented May 28, 1968

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# 3,385,692 METHOD FOR CONTROLLING THE

GROWTH OF WEEDS Richard N. Knowles, Hockessin, Del., assignor to E. I. du Pont de Nemours and Company, Wilmington, Del., a 5 corporation of Delaware No Drawing, Filed Aug. 26, 1964, Ser. No. 392,305 3 Claims. (Cl. 71-120)

ABSTRACT OF THE DISCLOSURE

Controlling the growth of weeds with 3-(o-fluorophenyl)-1-methylurea.

This invention relates to methods of destroying weeds with substituted phenylurea compounds. More particularly, this invention refers to a method of controlling the growth of weeds by applying to an area of weed growth 3-(o-fluorophenyl)-1-methylurea.

Tri-substituted urea compounds are well known for their herbicidal properties; see U.S. Patents 2,655,445 and 2,655,446. Well-known commercial herbicides included within the scope of these patents include monuron, 3-(p-25 chlorophenyl)-1,1-dimethylurea; and diuron, 3-(3,4-di-chlorophenyl)-1,1-dimethylurea. I have now discovered another outstanding urea herbicide characterized as a disubstituted urea molecule.

The compound in my invention has a desirable charac- 30 teristic that makes it more attractive than known herbicides for certain types of herbicidal usage. I have found that 3-(o-fluorophenyl)-1-methylurea is substantially more soluble in water than either of the aforementioned commercial herbicides. Accordingly, it is dissipated much 35 methylurea can preferably be in the form of wettable more rapidly in the soil.

The compound used in my invention is formulated in conventional herbicidal compositions. Usually the compositions will comprise the 3-(o-fluorophenyl-1-methylurea and one or more surface-active agents.

40 The surface-active agent used in this invention can be a wetting, dispersing or an emulsifying agent which will assist dispersion of the compound. The surface-active agent or surfactant can include such anionic, cationic and non-ionic agents as have heretofore been generally em- 45 ployed in plant control compositions of similar type. Suitable surface-active agents are set out, for example, in Searle U.S. Patent 2,426,417; Todd U.S. Patent 2,655,447; Jones U.S. Patent 2,412,510; or Lenher U.S. Patent 2,139,276. A detailed list of such agents is set forth in 50 "Detergents and Emulsifiers Annual" (1964) by John W. McCutcheon, Inc.

Suitable surface-active agents for use in compositions of the present invention are: polyethylene glycol fatty esters and fatty alkylol amide condensates, alkylaryl 55 sulfonates, fatty alcohol sulfates, dialkyl esters of sodium sulfosuccinate, fatty acid esters of sodium isethionate, polyoxyethylene ethers and thioethers, and long chain quaternary ammonium chloride compounds.

Surface-active dispersing agents such as sodium, mag- 60 nesium or calcium lignin sulfonates, low viscosity methyl cellulose, polymerized sodium salts of alkylnaphthalene sulfonic acids are also suitable in the herbicidal compositions of this invention.

Among the more preferred surfactants are the anionic 65 and non-ionic type. Among the anionic surface-active agents, preferred ones are alkali metal or amine salts of alkylbenzene sulfonic acids, such as dodecylbenzenesulfonic acid, sodium lauryl sulfate, alkylnaphthalene sulfonates, sodium N-methyl-N-oleoyltaurate, oleic acid 70 ester of sodium isethionate, dioctyl sodium sulfosuccinate, sodium dodecyldiphenyloxide disulfonate. Among the

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non-ionic compounds, preferred members are alkylphenoxy poly(ethyleneoxy)ethanols such as nonylphenol adducts with ethylene oxide; trimethylnonyl polyethylene glycol ethers, polyethylene oxide adducts of fatty and rosin acids, long chain alkyl mercaptan adducts with ethylene oxide and polyethylene oxide adducts with sorbitan fatty acid esters.

In general, less than 10% by weight of the surfaceactive agent will be used in compositions of this invention and ordinarily the amount of surface-active agents 10 will range from 1-5% but may even be less than 1% by weight.

Additional surface-active agents can be added to the above formulation to increase the ratio of surface-active agent: active agent up to as high as 5:1 by weight. Normally the purpose of adding higher amounts of surfactant is to increase the herbicidal effect of the active compound. When used at higher rates it is preferred that the surfactant be present in the range of one fifth to five parts surfactant for each one part of 3-(o-fluorophenyl)-1-

methylurea. The herbicidal compositions of this invention can contain in addition to the surfactant, finely divided diluents such as talc, natural clay including attapulgite and kaolinite clay, pyrophyllite, diatomaceous earths, synthetic fine silicas, calcium silicate, carbonates, calcium phosphates, sulfur, lime, such flours as walnut shell, wheat, redwood, soybeans and cottonseed.

The amount of finely divided inert solid diluent can vary widely but will generally range from 10 to 98% by weight of the herbicidal composition. The particle size can vary considerably but will ordinarily be somewhat under 50 microns in the finished formulation.

Compositions containing the 3-(o-fluorophenyl)-1powders. In addition to the active ingredient these contain one or more surfactants and one or more finely divided inert solid diluent, as described above. Other ingredients, such as corrosion inhibitors, stickers, antifoam agents, pigments and the like can be optionally included.

These wettable powders are prepared by blending the ingredients and then grinding the composition in a hammer mill, air attrition mill or similar device. Such wettable powders contain from 10 to 90% of the active ingredient, from 1 to 5% surfactant and the remainder finely divided inert solid diluent together with one or more of the optional additives. The particle size of the composition is preferably below 50 microns.

Other compositions commonly used in the herbicide art, such as emulsifiable oil solutions, granules, pellets and aqueous suspension concentrates can also be used with the compound used in this invention.

The 3-(o-fluorophenyl)-1-methylurea can also be mixed with other herbicides to form a mixture having extremely broad herbicidal activity. The following compounds are mixed in the proportions set forth below.

#### Substituted ureas

- 3-(3,4-dichlorophenyl)-1,1-dimethylurea
- 3-(4-chlorophenyl)-1,1-dimethylurea
- 3-phenyl-1,1-dimethylurea
- 3-(3,4-dichlorophenyl)-1-n-butyl-1-methylurea
- 3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea
- 3-(4-chlorophenyl)-1-methoxy-1-methylurea
- 3-(3,4-dichlorophenyl)-1,1,3-trimethylurea
- 3-(p-chlorophenoxyphenyl)-1,1-dimethylurea
- 1-(2-methylcycylohexyl)-3-phenylurea

The above ureas can be mixed with the 3-(o-fluorophenyl)-1-methylurea used in this invention in proportions of from 1:4 to 4:1, respectively, the preferred ratio being 1:2 to 2:1.

### Substituted triazines

2-chloro-4,6-bis(ethylamino)-s-triazine

2-chloro-4-ethylamino-6-isopropylamino-s-triazine

2-chloro-4,6-bis(methoxypropylamino)-s-triazine

2-methoxy-4,6-bis(isopropylamino)-s-triazine

2-methylmercapto-4,6-bis(isopropylamino)-s-triazine

2-methylmercapto-4,6-bis(ethylamino)-s-triazine

2-methylmercapto-4-ethylamino-6-isopropylamino-striazine

2-methoxy-4,6-bis(ethylamino)-s-triazine

2-methoxy-4-ethylamino-6-isopropylamino-s-triazine 2-chloro-4,6-bis(isopropylamino)-s-triazine

These triazines can be mixed with the 3-(o-fluorophenyl)-1-methylurea used in this invention in proportions of from 1:4 to 4:1 respectively, the preferred ratio being 1:2 to 2:1.

#### Phenols

dinitro-o-sec-butylphenol and its salts pentachlorophenol and its salts

These phenols can be mixed with the 3-(o-fluorophenyl)-1-methylurea used in this invention in the propor-25 tions of 1:10 to 20:1, respectively, the preferred ratio being 1:5 to 5:1.

#### Carboxylic acids and derivatives

The following carboxylic acids and derivatives can be  $_{30}$ mixed with the 3-(o-fluorophenyl)-1-methylurea used in this invention in the listed proportions:

Α.

2,3,6-trichlorobenzoic acid and its salts 35 2,3,5,6-tetrachlorbenzoic acid and its salts 2-methoxy-3,5,6-trichlorobenzoic acid and its salts 2-methoxy-3,6-dichlorbenzoic acid and its salts 3-ammino-2,5-dichlorobenzoic acid and its salts 3-nitro,2,5-dichlorobenzoic acid and its salts 40 2-methyl-3,6-dichlorobenzoic acid and its salts 2,4-dichlorophenoxyacetic acid and its salts and esters 2,4,5-trichlorophenoxyacetic acid and its salts and esters (2-methyl-4-chlorophenoxy) acetic acid and its salts and esters 2-(2,4,5-trichlorophenoxy) propionic acid and its salts 45and esters 2-(2,4,5-trichlorophenoxy)ethyl-2,2-dichloropropionate 4-(2,4-dichlorophenoxy) butyric acid and its salts and esters 4-(2-methyl-4-chlorophenoxy)butric acid and its salts 50 and esters 2,3,6-trichlorobenzyloxypropanol Mixed in a 1:16 to 8:1 ratio, preferably a 1:4 to 4:1 ratio. B. 2,6-dichlorobenzontrile Mixed in a 1:4 to 4:1 ratio, preferably a 1:3 to 3:1 ratio. 60 C. trichloroacetic acid and its salts Mixed in a 1:2 to 25:1 ratio, preferably a 1:1 to 8:1

ratio. D.

2,2-dichloropropionic acid and its salts

Mixed in a 1:4 to 8:1 ratio, preferably a 1:2 to 4:1 ratio.

E.

N.N-di(n-propyl)thiolcarbamic acid, ethyl ester N,N-di(n-propyl)thiolcarbamic acid, N-propyl ester N-ethyl-N-(n-butyl)thiolcarbamic acid, ethyl ester N-ethyl-N-(n-butyl)thiolcarbamic acid, n-propyl ester

Mixed in a 1:2 to 24:1 ratio, preferably a 1:1 to 12:1 ratio.

F.

N-phenylcarbamic acid, 2-methylcyclohexyl ester  $\mathbf{5}$ 

- N-phenylcarbamic acid, isopropyl ester N-(m-chlorophenyl)carbamic acid, isopropyl ester N-(m-chlorophenyl)carbamic acid, 4-chloro-2-butynyl ester
- Mixed in a 1:2 to 24:1 ratio, preferably a 1:1 to 12:1 10 ratio.

G.

2,3,6-trichlorophenylacetic acid and its salts

Mixed in a 1:12 to 8:1 ratio, preferably a 1:4 to 4:1 15ratio.

H.

2-chloro-N,N-diallylacetamide

maleic hydrazide 20

Mixed in a 1:2 to 10:1 ratio, preferably a 1:1 to 5:1 ratio.

Inorganic and mixed inorganic-organic salts

The following salts can be mixed with the 3-(o-fluorophenyl)-1-methylurea used in this invention in the listed proportions. Α.

calcium propylarsonate

disodium monomethylarsonate octyl-dodecylammoniummethylarsonate

dimethylarsinic acid

Mixed in a 1:4 to 4:1 ratio, preferably a 1:2 to 2:1 ratio. B.

sodium arsenite

Mixed in a 1:5 to 40:1 ratio, preferably a 1:4 to 25:1 ratio. C.

lead arsenate calcium arsenate

> Mixed in a 150:1 to 600:1 ratio, preferably a 100:1 to 400:1 ratio.

#### D.

sodium tetraborate hydrated, granulated

sodium metaborate

polyborchlorate

ratio.

Mixed in a 1:10 to 20:1 ratio, preferably a 1:5 to 5:1 ratio. F.

sodium chlorate

Mixed in a 1:1 to 40:1 ratio, preferably a 2:1 to 20:1 ratio. G.

ammonium sulfamate

65 Mixed in a 1:1 to 100:1 ratio, preferably a 1:1 to 50:1 ratio.

Other organic herbicides

These organic herbicides can be mixed with the 3-(o-70 fluorophenyl)-1-methylurea used in this invention in the listed proportions:

А.

5,6-dihydro-(4A,6A)-dipyrido-(1,2-A,2',1'-C) 75 pyrazinium dibromide

sodium pentaborate

unrefined borate or such as borascu

Mixed in a 3:1 to 1500:1, preferably a 6:1 to 1000:1 E.

55 ammonium thiocyanate

 $\mathbf{20}$ 

Mixed in a 1:20 to 16:1 ratio, preferably a 1:5 to 5:1 ratio. B.

3-amino-1,2,4-triazole

Mixed in a 1:20 to 20:1 ratio, preferably a 1:5 to  $\mathbf{5}$ 5:1 ratio. C.

3,6-endooxohexahydrophthalic acid

Mixed in a 1:3 to 20:1 ratio, preferably a 1:2 to 10:1 10 ratio. D.

hexachloroacetone

Mixed in a 1:2 to 16:1 ratio, preferably a 1:1 to 15 3-sec-butyl-1-acetyl-5-bromo-6-methyluracil 8:1 ratio.

E.

diphenylacetonitrile

N,N-dimethyl- $\alpha$ , $\alpha$ -diphenylacetamide

N,N-di-n-propyl-2,6-dinitro-4-trifluoromethylaniline N,N-di-n-propyl-2,6-dinitro-4-methylaniline

Mixed in a 1:10 to 30:1 ratio, preferably a 1:5 to 20:1 ratio.

F.

O-(2,4-dichlorophenyl)-O-methyl-isopropylphosphoramidothiate

2,3,5,6-tetrachloroterephthalic acid, dimethyl ester

Mixed in a 1:4 to 20:1 ratio, preferably a 1:3 to 51:1 ratio. G.

2,4-dichloro-4'-nitrodiphenyl ether

Mixed in a 1:10 to 30:1 ratio, preferably a 1:5 to  $_{35}$ 20:1 ratio.

Substituted uracils

3-(o-fluorophenyl)-1-methylurea can be mixed with substituted uracils, in the proportions listed below.

Α.

3-cyclohexyl-6-methyluracil

3-cyclohexyl-6-ethyluracil 3-cyclohexyl-6-sec-butyluracil

3-norbornyl-6-methyluracil

3-cyclopentyl-6-methyluracil

3-cyclohexyl-6-isopropyluracil

5-bromo-3-sec-butyl-6-methyluracil 5-chloro-3-sec-butyl-6-methyluracil

5-bromo-3-tert-butyl-6-methyluracil

5-chloro-3-tert-butyl-6-methyluracil

5-bromo-3-(1-ethylpropyl)-6-methyluracil

5-chloro-3-(1-ethylpropyl)-6-methyluracil

5-bromo-3-isopropyl-6-methyluracil 5-chloro-3-isopropyl-6-methyluracil

5-chloro-3-cyclohexylmethyl-6-methyluracil

5-methoxy-3-cyclohexyl-6-methyluracil

3-sec-butyl-5,6-dimethyluracil

5-bromo-3-norbornylmethyl-6-methyluracil

5-nitro-3-sec-butyl-6-methyluracil

3-cyclohexyl-5,6-dimethyluracil 5-bromo-3-cyclohexyl-6-methyluracil

5-chloro-3-phenyl-6-methyluracil

Mixed in a 1:4 to 4:1 ratio, preferably a 1:2 to 2:1 65 ratio.

B.

3-cyclohexyl-5,6-trimethyleneuracil

3-sec-butyl-5,6-trimethyleneuracil

3-isopropyl-5,6-trimethyleneuracil

3-isopropyl-5,6-tetramethyleneuracil

3-isopropyl-5,6-pentamethyleneuracil

Mixed in a 1:6 to 6:1 ratio, preferably a 1:4 to 4:1 ratio.

6 C.

3-cyclohexyl-5-bromouracil 3-cyclohexyl-5-chlorouracil 3-isopropyl-5-bromouracil 3-sec-butyl-5-bromouracil

3-sec-butyl-5-chlorouracil

Mixed in a 1:6 to 6:1 ratio, preferably a 1:2 to 2:1 ratio.

D.

3-isopropyl-1-trichloromethylthio-5-bromo-6methyluracil

3-cyclohexyl-1-trichloromethylthio-5-bromo-6methyluracil

3-isopropyl-1-acetyl-5-bromo-6-methyluracil

3-isopropyl-1-trichloromethylthio-5-chloro-6-

methyluracil

Mixed in a 1:4 to 4:1 ratio, preferably a 1:2 to 2:1 ratio.

All of the foregoing ratios are weight ratios.

The compound used in this invention is applied in a herbicidally effective amount directly to the soil as a 25 pre-emergence or a post-emergence treatment or it can be mixed intimately with the soil. One half to two pounds per acre of active ingredient will be applied to agricultural soils in order to remove weeds pre-emergence or early post-emergence. When soil sterilization is desired  $_{30}$  the herbicidally effective amount is four to forty pounds per acre depending upon the type of soil.

The following additional examples are set forth to provide a still clearer understanding of the invention.

#### Example 1

Fifteen grams of o-fluoroaniline is admixed with 7.67 grams of methylisocyanate and 150 milliliters of dioxane. After three hours at 25° C. the solution is evaporated in vacuum leaving a mixture of an oil and some white crys-

40 tals. The white crystals are filtered and washed with chloroform and then ether. The remaining solid, 3-(ofluorophenyl)-1-methylurea, melts at 155.7° C. to 156.7° C.

Calculated for: C<sub>8</sub>H<sub>9</sub>FN<sub>2</sub>O: C, 57.13; H, 5.40; F, 11.30; N, 16.66; O, 9.51. Found: C, 56.97; H, 5.75; <sup>45</sup> N, 16.86.

The composition is formulated as follows:

Percent 3-(o-fluorophenyl)-1-methylurea \_\_\_\_\_ 50 Attapulgite clay \_\_\_\_\_ 48 50 Dioctyl sodium sulfosuccinate \_\_\_\_\_ 1 Sodium lignin sulfonate \_\_\_\_\_ 1

The above composition is applied at the rate of 1.5 pounds of active ingredient to a field of cotton which is 55 in the process of being planted. Good pre-emergence control of weeds is obtained for a period of six to eight weeks. The weeds present are crabgrass, Johnsongrass seedlings, Florida pursley and annual morning glory.

Example 2

The crop shows normal growth.

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Percent

	3-(o-fluorophenyl)-1-methylurea	
5	Alkylnaphthalene sulfonate, Na salt	2
	Partially desulfonated sodium lignin sulfonate	2
	Synthetic fine silica	8
	Kaolin clay	8

The above composition is blended and micropulverized, then remilled through an air attrition mill until substan-70 tially all particles are below ten microns.

The above described formulation is applied pre-emergence by means of a tractor mounted sprayer at the rate of 1.5 pounds of active ingredient per acre to a plot of newly-planted potatoes. The potatoes emerge to a good 75 stand and develop an excellent crop. Weeds such as giant Percent

foxtail, barnyard grass, crabgrass, velvet leaf and pigweed germinate normally but within a few days succumb to the action of the chemical.

#### Example 3

1010			
3-(o-fluorophenyl)-1-methylurea	80		
Attapulgite clay	18		
Dioctyl sodium sulfosuccinate			
Sodium lignin sulfonate	1	17	

Twenty five pounds of the above formulation is suspended in forty gallons of water and applied at the beginning of the growing season to an industrial type outdoor storage area which is one acre in size. A mixed 15 stand of weedy vegetation including crabgrass, yellow foxtail, goosegrass, wild mustard, black-eyed susan, smartweed and morning glory germinate but die within ten days.

The invention claimed is:

1. Method for controlling the growth of weeds com-

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prising applying to an area of weed growth a herbicidally effective amount of 3-(o-fluorophenyl)-1-methylurea.

2. Method for controlling the growth of weeds growing among cotton plants comprising applying to an area 5 of weed growth within a cotton field a herbicidally effective amount of 3-(o-fluorophenyl)-1-methylurea.

3. Method for controlling the growth of weeds growing among potato plants comprising applying to an area of weed growth within potato field a herbicidally effective amount of 3-(o-fluorophenyl)-1-methylurea.

#### **References Cited**

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<sup>20</sup> JAMES O. THOMAS, JR., Primary Examiner.