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PATENT SPECIFICATION

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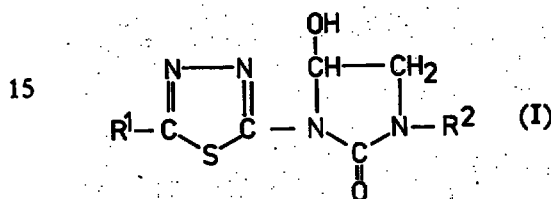
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(54) THIADIAZOLYLIMIDAZOLIDINONES

(71) We, VELSICOL CHEMICAL CORPORATION, a corporation organised and existing under the laws of the State of Delaware, United States of America, of 341 East Ohio Street, Chicago, Cook County, State of Illinois 60611, United States of America, (assignee of JOHN KRENZER), 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:—

This invention relates to new chemical compounds of the formula



wherein R¹ represents an alkyl of up to 4 carbon atoms, an alkenyl, chloroalkyl, trifluoromethyl, alkoxy, alkylthio, alkylsulfonyl or alkylsulfinyl group; and R² is an alkyl group.

The compounds of the present invention are unexpectedly useful as herbicides.

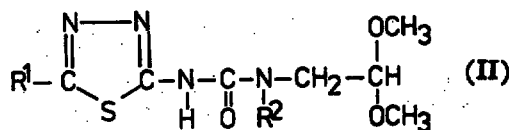
In a preferred embodiment of this invention R¹ represents an alkyl group of up to 4 carbon atoms, a lower alkenyl, lower chloroalkyl, trifluoromethyl, lower alkoxy, lower alkylthio, lower alkylsulfonyl or lower alkylsulfinyl group; and R² is a lower alkyl group.

Included within the scope of general formula I are compounds in which R¹ repre-

sents an alkyl group of up to 3 carbon atoms, an alkenyl, chloroalkyl, alkoxy, alkylthio, alkylsulfonyl or alkylsulfinyl group and R² is an alkyl group. Preferably R¹ represents an alkyl group of up to 3 carbon atoms, a lower alkenyl, lower chloroalkyl, lower alkoxy, lower alkylthio, lower alkylsulfonyl or lower alkylsulfinyl group and R² represents a lower alkyl group.

The term lower as used herein designates a straight or branched carbon chain of up to six carbon atoms.

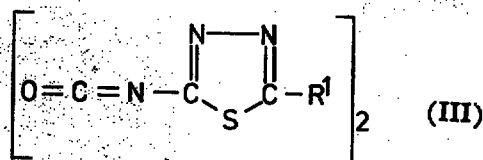
The compounds of this invention can be readily prepared by heating a compound of the formula



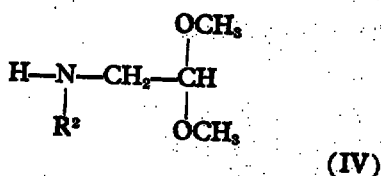
wherein R¹ and R² are as heretofore described, in a dilute, aqueous, acidic reaction medium for a period of about 10 to about 60 minutes. Temperatures of from about 70° C. to the reflux temperature of the reaction mixture can be utilized. The reaction medium can comprise a dilute aqueous inorganic acid such as hydrochloric acid at a concentration of from about 0.5 to about 5 percent. Upon completion of the reaction the desired product can be recovered as a precipitate by cooling the reaction mixture. This product can be used as such or can be further purified by conventional means such as recrystallization and the like.

The compounds of formula II can be pre-

pared by reacting a molar amount of an isocyanate dimer of the formula

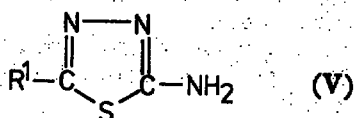


wherein R^1 is as heretofore described, with about two molar amounts of a dimethyl acetal of the formula



wherein R^2 is as heretofore described. This reaction can be effected by heating a mixture of the isocyanate dimer and the acetal in an inert organic reaction medium such as benzene at the reflux temperature of the reaction mixture. Heating at reflux can be continued for a period of from about 2 to about 30 minutes to ensure completion of the reaction. After this time the desired product can be recovered upon evaporation of the reaction medium and can be used as such or can be further purified by standard techniques in the art.

The isocyanate dimer of formula III can be prepared by reacting a thiadiazole of the formula



wherein R^1 is as heretofore described, with phosgene. This reaction can be effected by adding a slurry or solution of the thiadiazole, in a suitable organic solvent such as ethyl acetate, to a saturated solution of phosgene in an organic solvent such as ethyl acetate. The resulting mixture can be stirred at ambient temperatures for a period of from about 4 to about 24 hours. The reaction mixture can then be purged with nitrogen gas to remove unreacted phosgene. The desired product can then be recovered by filtration if formed as a precipitate or upon evaporation of the organic solvent used if soluble therein. This product can be used as such or can be further purified if desired.

Exemplary thiadiazoles of formula V useful for preparing the compounds of the present invention are:

5 - methyl - 2 - amino - 1,3,4 - thiadiazole, 5 - ethyl - 2 - amino - 1,3,4 - thiadiazole, 5 - propyl - 2 - amino - 1,3,4 - thiadiazole, 5 - allyl - 2 - amino - 1,3,4 - thiadiazole, 5 - pent - 3 - enyl - 2 - amino - 1,3,4 - thiadiazole, 5 - chloromethyl - 2 - amino - 1,3,4 - thiadiazole, 5 - β - chloroethyl - 2 - amino - 1,3,4 - thiadiazole, 5 - γ - chloropropyl - 2 - amino - 1,3,4 - thiadiazole, 5 - trichloromethyl - 2 - amino - 1,3,4 - thiadiazole, 5 - methoxy - 2 - amino - 1,3,4 - thiadiazole, 5 - ethoxy - 2 - amino - 1,3,4 - thiadiazole, 5 - propoxy - 2 - amino - 1,3,4 - thiadiazole, 5 - butyloxy - 2 - amino - 1,3,4 - thiadiazole, 5 - hexyloxy - 2 - amino - 1,3,4 - thiadiazole, 5 - methylthio - 2 - amino - 1,3,4 - thiadiazole, 5 - ethylthio - 2 - amino - 1,3,4 - thiadiazole, 5 - propylthio - 2 - amino - 1,3,4 - thiadiazole, 5 - butylthio - 2 - amino - 1,3,4 - thiadiazole, 5 - methylsulfonyl - 2 - amino - 1,3,4 - thiadiazole, 5 - butyl - 2 - amino - 1,3,4 - thiadiazole, 5 - methylsulfinyl - 2 - amino - 1,3,4 - thiadiazole, 5 - *t* - butyl - 2 - amino - 1,3,4 - thiadiazole and 5 - trifluoromethyl - 2 - amino - 1,3,4 - thiadiazole.

The manner in which the compounds of the present invention can be prepared is more specifically illustrated in the following examples.

Example 1.

Preparation of 5 - Methyl - 1,3,4 - thiadiazole-2 - yl Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) is charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - methyl - 2 - amino - 1,3,4 - thiadiazole (40 grams) in ethyl acetate (300 ml) is added to the reaction vessel and the resulting mixture is stirred for a period of about 16 hours, resulting in the formation of a precipitate. The reaction mixture is then purged with nitrogen gas to remove unreacted phosgene. The purged mixture is then filtered to recover the precipitate. The precipitate is then recrystallized to yield the desired product 5 - methyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer.

Example 2.

Preparation of the Dimethyl Acetal of 2 - [1 - Methyl - 3 - (5 - methyl - 1,3,4 - thiadiazole - 2 - yl)ureido]acetaldehyde

A mixture of 5 - methyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer (0.05 mole), the dimethyl acetal of 2 - methylaminoacetaldehyde (0.1 mole) and benzene (60 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. After this time the mixture is stripped of benzene under reduced pressure to yield a solid product as the residue. The residue is then recrystallized to yield the desired product dimethyl acetal of 2 - [1 - methyl - 3 - (5 - methyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde.

Example 3.

Preparation of 1 - (5 - Methyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - methyl - 3 - (5 - methyl - 1,3,4 - thiadiazol - 2 - yl) - ureido]acetaldehyde (15 grams), water (400 ml) and hydrochloric acid (4 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. The reaction mixture is then filtered while hot and the filtrate is cooled to form a precipitate. The precipitate is recovered by filtration, is dried and is recrystallized to yield the desired product 1 - (5 - methyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

Example 4.

Preparation of 5 - Methoxy - 1,3,4 - thiadiazol - 2 - yl Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) is charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - methoxy - 2 - amino - 1,3,4 - thiadiazole (40 grams) in ethyl acetate (300 ml) is added to the reaction vessel and the resulting mixture is stirred for a period of about 16 hours, resulting in the formation of a precipitate. The reaction mixture is then purged with nitrogen gas to remove unreacted phosgene. The purged mixture is then filtered to recover the precipitate. The precipitate is then recrystallized to yield the desired product 5 - methoxy - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer.

Example 5.

Preparation of the Dimethyl Acetal of 2 - [1 - Ethyl - 3 - (5 - methoxy - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde

A mixture of 5 - methoxy - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer (0.05 mole), the dimethyl acetal of 2 - ethylamino - acetaldehyde (0.1 mole) and benzene (60 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. After this time the mixture is stripped of benzene under reduced pressure to yield a solid product as the residue. The residue is then recrystallized to yield the desired product dimethyl acetal of 2 - [1 - ethyl - 3 - (5 - methoxy - 1,3,4 - thiadiazol - 2 - yl)ureido] - acetaldehyde.

Example 6.

Preparation of 1 - (5 - Methoxy - 1,3,4 - thiadiazol - 2 - yl) - 3 - ethyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - ethyl - 3 - (5 - methoxy - 1,3,4 - thiadiazol - 2 - yl) -

ureido]acetaldehyde (15 grams), water (400 ml) and hydrochloric acid (4 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. The reaction mixture is then filtered while hot and the filtrate is cooled to form a precipitate. The precipitate is recovered by filtration, is dried and is recrystallized to yield the desired product 1 - (5 - methoxy - 1,3,4 - thiadiazol - 2 - yl) - 3 - ethyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

Example 7.

Preparation of 5 - Methylthio - 1,3,4 - thiadiazol - 2 - yl Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) is charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - methylthio - 2 - amino - 1,3,4 - thiadiazole (45 grams) in ethyl acetate (300 ml) is added to the reaction vessel and the resulting mixture is stirred for a period of about 16 hours, resulting in the formation of a precipitate. The reaction mixture is then purged with nitrogen gas to remove unreacted phosgene. The purged mixture is then filtered to recover the precipitate. The precipitate is then recrystallized to yield the desired product 5 - methylthio - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer.

Example 8.

Preparation of the Dimethyl Acetal of 2 - [1 - Propyl - 3 - (5 - methylthio - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde

A mixture of 5 - methylthio - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer (0.05 mole), the dimethyl acetal of 2 - propylamino - acetaldehyde (0.1 mole) and benzene (60 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. After this time the mixture is stripped of benzene under reduced pressure to yield a solid product as the residue. The residue is then recrystallized to yield the desired product dimethyl acetal of 2 - [1 - propyl - 3 - (5 - methylthio - 1,3,4 - thiadiazol - 2 - yl) - ureido]acetaldehyde.

Example 9.

Preparation of 1 - (5 - Methylthio - 1,3,4 - thiadiazol - 2 - yl) - 3 - propyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - propyl - 3 - (5 - methylthio - 1,3,4 - thiadiazol - 2 - yl) - ureido]acetaldehyde (15 grams), water (400 ml) and hydrochloric acid (4 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. The

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reaction mixture is then filtered while hot and the filtrate is cooled to form a precipitate. The precipitate is recovered by filtration, is dried and is recrystallized to yield the desired product 1 - (5 - methylthio - 1,3,4 - thiadiazol - 2 - yl) - 3 - propyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

Example 10.

Preparation of 5 - Methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) is charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - methylsulfonyl - 2 - amino - 1,3,4 - thiadiazole (50 grams) in ethyl acetate (300 ml) is added to the reaction vessel and the resulting mixture is stirred for a period of about 16 hours, resulting in the formation of a precipitate. The reaction mixture is then purged with nitrogen gas to remove unreacted phosgene. The purged mixture is then filtered to recover the precipitate. The precipitate is then recrystallized to yield the desired product 5 - methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer.

Example 11.

Preparation of the Dimethyl Acetal of 2 - [1 - Methyl - 3 - (5 - methylsulfonyl) - 1,3,4 - thiadiazol - 2 - yl]ureido]acetaldehyde

A mixture of 5 - methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer (0.05 mole), the dimethyl acetal of 2 - methylaminoacetaldehyde (0.1 mole) and benzene (60 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. After this time the mixture is stripped of benzene under reduced pressure to yield a solid product as the residue. The residue is then recrystallized to yield the desired product dimethyl acetal of 2 - [1 - methyl - 3 - (5 - methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde.

Example 12.

Preparation of 1 - (5 - Methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - methyl - 3 - (5 - methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde (15 grams), water (400 ml) and hydrochloric acid (4 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. The reaction mixture is then filtered while hot and the filtrate is cooled to form a precipitate. The precipitate is recovered by filtration, is dried and is recrystallized to yield

the desired product 1 - (5 - methylsulfonyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

Example 13.

Preparation of 5 - Methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) is charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - methylsulfinyl - 2 - amino - 1,3,4 - thiadiazole (50 grams) in ethyl acetate (300 ml) is added to the reaction vessel and the resulting mixture is stirred for a period of about 16 hours, resulting in the formation of a precipitate. The reaction mixture is then purged with nitrogen gas to remove unreacted phosgene. The purged mixture is then filtered to recover the precipitate. The precipitate is then recrystallized to yield the desired product 5 - methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer.

Example 14.

Preparation of the Dimethyl Acetal of 2 - [1 - Methyl - 3 - (5 - methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde

A mixture of 5 - methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer (0.05 mole), the dimethyl acetal of 2 - methylaminoacetaldehyde (0.1 mole) and benzene (60 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. After this time the mixture is stripped of benzene under reduced pressure to yield a solid product as the residue. The residue is then recrystallized to yield the desired product dimethyl acetal of 2 - [1 - methyl - 3 - (5 - methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde.

Example 15.

Preparation of 1 - (5 - Methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - methyl - 3 - (5 - methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde (15 grams), water (400 ml) and hydrochloric acid (4 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. The reaction mixture is then filtered while hot and the filtrate is cooled to form a precipitate. The precipitate is recovered by filtration, is dried and is recrystallized to yield the desired product 1 - (5 - methylsulfinyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

Example 16.

Preparation of 5 - Trifluoromethyl - 1,3,4-thiadiazol - 2 - yl - Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) was charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - trifluoromethyl - 2-amino - 1,3,4 - thiadiazole (45 grams) in ethyl acetate (300 ml) was added to the reaction vessel and the resulting mixture was stirred for a period of about 16 hours resulting in the formation of a precipitate. The reaction mixture was then purged with nitrogen gas to remove unreacted phosgene. The purged mixture was filtered to recover 48 grams of a white solid. This solid was recrystallized from dimethyl formamide to yield the desired product 5 - trifluoromethyl - 1,3,4-thiadiazol - 2 - yl isocyanate dimer.

Example 17.

Preparation of the Dimethyl Acetal of 2 - [1-methyl - 3 - (5 - trifluoromethyl - 1,3,4-thiadiazol - 2 - yl)ureido]acetaldehyde

A mixture of 5 - trifluoromethyl - 1,3,4-thiadiazol - 2 - yl isocyanate dimer (9.5 grams), the dimethyl acetal of 2 - methylaminoacetaldehyde (5.8 grams) and benzene (60 ml) are charged into a glass reaction vessel equipped with a mechanical stirrer and reflux condenser. The reaction mixture is heated at reflux for a period of about 15 minutes. After this time the mixture is stripped of benzene under reduced pressure to yield a solid product as the residue. This product is recrystallized from heptane to yield the desired product the dimethyl acetal of 2 - [1 - methyl - 3 - (5 - trifluoromethyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde having a melting point of 101 to 102° C.

Example 18.

Preparation of 1 - (5 - Trifluoromethyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - methyl - 3 - (5 - trifluoromethyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde (15 grams), water (400 ml) and hydrochloric acid (4 ml) were charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture was heated at reflux for a period of about 15 minutes. The reaction mixture was then filtered while hot and the filtrate was cooled resulting in the formation of a precipitate. The precipitate was recovered by filtration, was dried and was recrystallized from an ethyl acetate-hexane mixture to yield the desired product 1 - (5 - trifluoromethyl - 1,3,4-thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one having a melting point of 136 to 138° C.

Example 19.

Preparation of 5 - t - Butyl - 1,3,4 - thiadiazol - 2 - yl Isocyanate Dimer

A saturated solution of phosgene in ethyl acetate (100 ml) was charged into a glass reaction vessel equipped with a mechanical stirrer. A slurry of 5 - t - butyl - 2 - amino - 1,3,4 - thiadiazole (10 grams) in ethyl acetate (300 ml) was added to the reaction vessel and the resulting mixture was stirred for a period of about 16 hours resulting in the formation of a precipitate. The reaction mixture was then purged with nitrogen gas to remove unreacted phosgene. The purged mixture was then filtered to recover the desired product 5 - t - butyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer as a solid having a melting point of 261 to 263° C.

Example 20.

Preparation of the Dimethyl Acetal of 2 - [1-methyl - 3 - (5 - t - butyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde

A mixture of 5 - t - butyl - 1,3,4 - thiadiazol - 2 - yl isocyanate dimer (6 grams), the dimethyl acetal of 2 - methylaminoacetaldehyde (3.9 grams) and benzene (50 ml) was charged into a glass reaction flask equipped with a mechanical stirrer and reflux condenser. The reaction mixture was heated at reflux, with stirring for a period of about 5 minutes. After this time the reaction mixture was stripped of benzene to yield an oil which solidified upon standing. The resulting solid was then recrystallized from pentane to yield the desired product the dimethyl acetal of 2 - [1 - methyl - 3 - (5 - t - butyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde having a melting point of 80—82° C.

Example 21.

Preparation of 1 - (5 - t - Butyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one

The dimethyl acetal of 2 - [1 - methyl - 3 - (5 - t - butyl - 1,3,4 - thiadiazol - 2 - yl)ureido]acetaldehyde (16 grams), concentrated hydrochloric acid (10 ml) and water (500 ml) were charged into a glass reaction vessel equipped with a mechanical stirrer, thermometer and reflux condenser. The reaction mixture was heated at reflux for a period of about 15 minutes. The reaction mixture was filtered while hot and the filtrate was then cooled, resulting in the formation of a precipitate. The precipitate was recovered by filtration, dried and was recrystallized from a benzene-hexane mixture to yield the desired product 1 - (5 - t - butyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one having a melting point of 133 to 134° C.

Additional compounds within the scope of the present invention which can be prepared

14

18

by the procedure of the foregoing examples are:

- 1 - (5 - ethyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - butyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - propyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - pentyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - butylthio - 1,3,4 - thiadiazol - 2 - yl) - 3 - hexyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - pentylthio - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - hexylthio - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - ethylthio - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - ethoxy - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - propoxy - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - butoxy - 1,3,4 - thiadiazole - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - hexyloxy - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - chloromethyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - β - chloroethyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - γ - chlorobutyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - allyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - pent - 3 - enyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - hex - 5 - enyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - ethylsulfonyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - propyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - butyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - hexyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one, 1 - (5 - ethylsulfonyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one and 1 - (5 - pentyl - 1,3,4 - thiadiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

For practical use as herbicides the compounds of this invention are generally incorporated into herbicidal compositions which comprise an inert carrier and a herbicidally toxic amount of such a compound. Such herbicidal compositions, which can also be called formulations, enable the active compound to be applied conveniently to the site of the weed infestation in any desired quantity. These compositions can be solids such as dusts, granules,

or wettable powders; or they can be liquids such as solutions, aerosols, or emulsifiable concentrates.

For example, dusts can be prepared by grinding and blending the active compound with a solid inert carrier such as the talcs, clays, silicas and pyrophyllite. Granular formulations can be prepared by impregnating the compound, usually dissolved in a suitable solvent, onto and into granulated carriers such as the attapulgites or the vermiculites, usually of a particle size range of from about 0.3 to 1.5 mm. Wettable powders, which can be dispersed in water or oil to any desired concentration of the active compound, can be prepared by incorporating wetting agents into concentrated dust compositions.

In some cases the active compounds are sufficiently soluble in common organic solvents such as kerosene or xylene so that they can be used directly as solutions in these solvents. Frequently, solutions of herbicides can be dispersed under superatmospheric pressure as aerosols. However, preferred liquid herbicidal compositions are emulsifiable concentrates, which comprise an active compound according to this invention and as the inert carrier, a solvent and an emulsifier. Such emulsifiable concentrates can be extended with water and/or oil to any desired concentration of active compound for application as sprays to the site of the weed infestation. The emulsifiers most commonly used in these concentrates are nonionic or mixtures of nonionic with anionic surface-active agents. With the use of some emulsifier systems an inverted emulsion (water in oil) can be prepared for direct application to weed infestations.

A typical herbicidal composition according to this invention is illustrated by the following example, in which the quantities are in parts by weight.

Example 22 A.		
Preparation of a Dust		
Product of Example 3	10	110
Powdered Talc	90	

The above ingredients are mixed in a mechanical grinder-blender and are ground until a homogeneous, free-flowing dust of the desired particle size is obtained. This dust is suitable for direct application to the site of the weed infestation.

Example 22 B.		
Preparation of a Dust		
Product of Example 21	10	120
Powdered Talc	90	

The above ingredients are mixed in a mechanical grinder-blender and are ground until a homogeneous, free-flowing dust of the desired particle size is obtained. This dust is

7
suitable for direct application to the site of the weed infestation.

Example 22 C.
Preparation of a Dust
Product of Example 18
Powdered Talc

10
90

5
10 The above ingredients are mixed in a mechanical grinder-blender and are ground until a homogeneous, free-flowing dust of the desired particle size is obtained. This dust is suitable for direct application to the site of the weed infestation.

15 The compounds of this invention can be applied as herbicides in any manner recognized by the art. One method for the control of weeds comprises contacting said weeds with a compound of the invention, in a quantity toxic to weeds, or with a herbicidal composition comprising an inert carrier and as an essential active ingredient in a quantity which is herbicidally toxic to said weeds, a compound of the present invention. One method of contacting the weeds with the compound of the invention is by applying the compound or composition containing it to the locus of the weeds. The concentration of the new compounds of this invention in the herbicidal compositions will vary greatly with the type of formulations and the purpose for which it is designed, but generally the herbicidal compositions will comprise from about 0.05 to about 95 percent by weight of the active compounds of this invention. In a preferred embodiment of this invention, the herbicidal compositions will comprise from about 5 to about 75 percent by weight of the active compound. The compositions can also comprise such additional substances as other pesticides, such as insecticides, nematocides, and fungicides; stabilisers, spreaders, deactivators, adhesives, stickers, fertilizers, activators and synergists.

40 Weeds are undesirable plants growing where they are not wanted, having no economic value, and interfering with the production of cultivated crops, with the growing of orna-

7
50 mental plants, or with the welfare of livestock. The new compounds of this invention are particularly valuable for weed control because they are toxic to many species and groups of weeds while they are relatively non-toxic to many beneficial plants. The exact amount of compound required will depend on a variety of factors, including the hardness of the particular weed species, weather, type of soil, method of application, the kind of beneficial plants in the same area, and the like. Thus, while the application of up to only about one or two ounces of active compound per acre may be sufficient for good control of a light infestation of weeds growing under adverse conditions, the application of ten pounds or more of active compounds per acre may be required for good control of a dense infestation of hardy perennial weeds growing under favorable conditions. 65

Examples 23 and 24.

The herbicidal toxicity of the new compounds of this invention was demonstrated by experiments carried out for the pre-emergence control of a variety of weeds. In these experiments small plastic greenhouse pots filled with dry soil were seeded with the weed seeds. Twenty-four hours or less after seeding the pots were sprayed with water until the soil was wet and the test compounds formulated as aqueous emulsions of acetone solutions containing emulsifiers were sprayed at the indicated concentrations on the surface of the soil. 70

After spraying, the soil containers were placed in the greenhouse and provided with supplementary heat as required and daily or more frequent watering. The plants were maintained under these conditions for a period of 27 days, at which time the condition of the plants and the degree of injury to the plants was rated on a scale of from 0 to 10, as follows: 0=no injury, 1,2=slight injury, 3,4=moderate injury, 5,6=moderately severe injury, 7,8,9=severe injury and 10=death. The effectiveness of these compounds is demonstrated by the data in Table I. 80 85 90

TABLE I

Injury Rating

		Weed Species											
		YNSG	WOAT	JSWD	VVLF	JSGR	PIGW	MUST	YFXT	BYGR	CRGR	CHGR	MNGL
Example 23	Product of Example 18												
	5	9	10	10	10	10	10	10	10	10	10	10	10
	2	9	10	10	10	10	10	10	10	10	5	10	10
Example 24	Product of Example 21												
	5	9	10	9	10	9	10	10	10	10	10	10	10
	2	9	10	9	10	9	9	10	10	10	10	10	10
	1	9	10	10	10	10	10	10	10	10	10	10	10
		YNSG =Yellow Nutsedge WOAT =Wild Oats JSWD =Jimsonweed VVLF =Velvetleaf JSGR =Johnsongrass PIGW =Pigweed MUST =Mustard YFXT =Yellow Foxtail BYGR =Barnyardgrass CRGR =Crabgrass CHGR =Cheatgrass MNGL =Morning glory											

Examples 25 and 26.

The herbicidal activity of the compounds of this invention was also demonstrated by experiments carried out for the post-emergence control of a variety of weeds. In these experiments the compounds of this invention were formulated as a solution in an inert solvent and sprayed at the indicated dosage on the foliage of the weeds that have attained

a prescribed size. After spraying, the plants were placed in a greenhouse and watered daily or more frequently. Water was not applied to the foliage of the treated plants. The severity of the injury was determined 17 days after treatment and was rated on the scale of from 0 to 10 heretofore described. The effectiveness of the compounds is demonstrated by the data in Table II.



TABLE II

Injury Rating

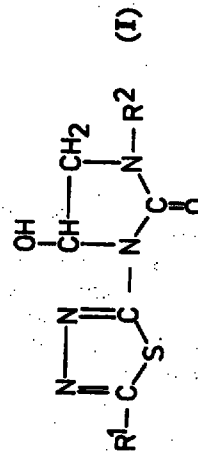
Concentration in Lbs/Acre			Weed Species											
Example	Product of		YNSG	WOAT	JSWD	VVLF	JSGR	PIGW	MUST	YFXT	BYGR	CRGR	CHGR	MNGL
25	Example 18	5	10	10	10	10	10	10	10	10	10	10	10	10
		2	10	10	10	10	10	10	10	10	10	10	10	10
		1	10	10	10	10	10	10	10	10	10	10	10	10
26	Example 21	5	10	10	10	10	10	10	10	10	10	10	10	10
		2	10	10	10	10	10	10	10	10	10	10	10	10
		1	10	10	10	10	10	10	10	10	10	10	10	10
			YNSG =Yellow Nutsedge	WOAT =Wild Oats	JSWD =Jimsonweed	VVLF =Velvetleaf	JSGR =Johnsongrass	PIGW =Pigweed	MUST =Mustard	YFXT =Yellow Foxtail	BYGR =Barnyardgrass	CRGR =Crabgrass	CHGR =Cheatgrass	MNGL =Morning glory

YNSG = Yellow Nutsedge
WOAT = Wild Oats
JSWD = Jimsonweed
VVLF = Velvetleaf
JSGR = Johnsongrass
PIGW = Pigweed

MUST = Mustard
YFXT = Yellow Foxtail
BYGR = Barnyardgrass
CRGR = Crabgrass
CHGR = Cheatgrass
MNGL = Morning glory

WHAT WE CLAIM IS:—

1. A compound of the formula I:



5 wherein R¹ represents an alkyl group of up to 4 carbon atoms, an alkenyl, chloroalkyl,

trifluoromethyl, alkoxy, alkylthio, alkylsulfonyl or alkylsulfinyl group; and R² is an alkyl group.

2. A compound as claimed in claim 1, in which R¹ represents an alkyl group of up to 4 carbon atoms, a lower alkenyl, lower chloroalkyl, trifluoromethyl, lower alkoxy, lower alkylthio, lower alkylsulfonyl or lower alkylsulfinyl group and R² represents a lower alkyl group.

3. A compound as claimed in claim 1, in which R¹ represents an alkyl group of up to 3 carbon atoms, an alkenyl, chloroalkyl,

8



alkoxy, alkylthio, alkylsulfonyl or alkylsulfinyl group and R² is an alkyl group.

4. A compound as claimed in claim 3, in which R¹ represents an alkyl group of up to 3 carbon atoms, a lower alkenyl, lower chloroalkyl, lower alkoxy, lower alkylthio, lower alkylsulfonyl or lower alkylsulfinyl group and R² represents a lower alkyl group.

5. 1 - (5 - methyl - 1,3,4 - thiadiazol-2 - yl) - 3 - methyl - 5 - hydroxy - 1,3-imidazolidin - 2 - one.

6. 1 - (5 - methoxy - 1,3,4 - thiadiazol-2 - yl) - 3 - ethyl - 5 - hydroxy - 1,3-imidazolidin - 2 - one.

7. 1 - (5 - methylthio - 1,3,4 - thiadiazol-2 - yl) - 3 - propyl - 5 - hydroxy - 1,3-imidazolidin - 2 - one.

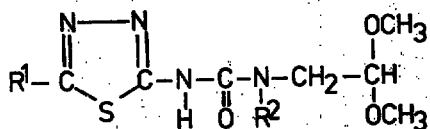
8. 1 - (5 - methylsulfonyl - 1,3,4 - thiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

9. 1 - (5 - methylsulfinyl - 1,3,4 - thia-
diazol - 2 - yl) - 3 - methyl - 5 - hydroxy-
1,3 - imidazolidin - 2 - one.

10. 1 - (5 - *t* - butyl - 1,3,4 - thiadiazol-2 - yl) - 3 - methyl - 5 - hydroxy - 1,3-imidazolidin - 2 - one.

11. 1 - (5 - trifluoromethyl - 1,3,4 - thiazol - 2 - yl) - 3 - methyl - 5 - hydroxy - 1,3 - imidazolidin - 2 - one.

12. A method of manufacturing a compound of the general formula I as set forth in claim 1, which comprises heating a compound of the general formula:



wherein R¹ and R² have the meanings given in claim 1, in a dilute aqueous, acidic reaction medium for about 10 to about 60 minutes.

13. A method as claimed in claim 12 in which R^1 and R^2 are as defined in claim 2.

14. A method as claimed in claim 12 in which R^1 and R^2 are as defined in claim 3.

15. A method as claimed in claim 12 in which R^1 and R^2 are as defined in claim 4.

16. A method as claimed in claim 12 in which R^1 is *t*-butyl and R^2 is methyl.

17. A method as claimed in claim 12, in which R^1 is trifluoromethyl and R^2 is methyl.

18. A herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in claim 1 or 2.

19. A herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in any one of claims 3 to 9.

20. A herbicidal composition comprising an inert carrier and, as an essential active

ingredient, in a quantity toxic to weeds, a compound as claimed in claim 10.

21. A herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in claim 11.

22. A method of controlling weeds which comprises contacting the weeds with a compound as claimed in claim 1 or 2 in a quantity toxic to weeds or with a herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in claim 1 or 2.

23. A method of controlling weeds which comprises contacting the weeds with a compound as claimed in any one of claims 3 to 9, in a quantity toxic to weeds, or with a herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in any one of claims 3 to 9.

24. A method of controlling weeds which comprises contacting the weeds with a compound as claimed in claim 10, in a quantity toxic to weeds, or with a herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in claim 10.

25. A method of controlling weeds which comprises contacting the weeds with a compound as claimed in claim 11, in a quantity toxic to weeds, or with a herbicidal composition comprising an inert carrier and, as an essential active ingredient, in a quantity toxic to weeds, a compound as claimed in claim 11.

26. A compound of the general formula I substantially as hereinbefore described in Example 18.

27. A method of manufacturing a compound of general formula I substantially as hereinbefore described in Example 18.

28. A compound of general formula I substantially as hereinbefore described in Example 21.

29. A method of manufacturing a compound of general formula I substantially as hereinbefore described in Example 21.

30. A compound of general formula I substantially as hereinbefore described in any one of Examples 3, 6, 9, 12 and 15.

31. A method of manufacturing a compound of general formula I substantially as hereinbefore described in any one of Examples 3, 6, 9, 12 and 15.

32. A herbicidal composition substantially as hereinbefore described in Example 22A.

33. A method of controlling weeds substantially as hereinbefore described in Example 23.

34. A method of controlling weeds substantially as hereinbefore described in Example 24.



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35. A method of controlling weeds substantially as hereinbefore described in Example 25.

38. A herbicidal composition substantially as hereinbefore described in Example 22C.

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36. A method of controlling weeds substantially as hereinbefore described in Example 26.

37. A herbicidal composition substantially as hereinbefore described in Example 22B.

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