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3,436,207

# CONTROL OF UNDESIRABLE VEGETATION

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Int. Cl. A01n 9/22, 7/00; C07d 51/34
U.S. Cl. 71—92 9 Claims

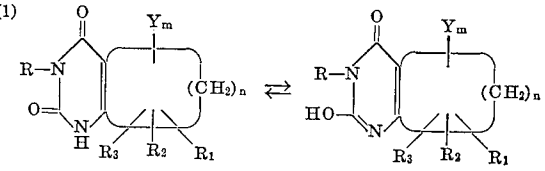
## ABSTRACT OF THE DISCLOSURE

The invention relates to the use of 3-substituted-5,6-substituted - 2,4 - pyrimidinediones as herbicides for the control of undesired vegetation. Typical compounds useful in the method of this invention are 3-cyclohexyl-5,6-dihydro - 7 - methyl-5-cyclopenta[D]pyrimidine-2,4(1H, 3H)-dione and 3-sec-butyl-5,6-dihydro-7-methyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione.

This invention relates to novel 3-substituted-5,6-substituted alkylene-2,4-pyrimidinediones and to compositions and methods employing these compounds as herbicides.

More particularly, this invention is directed to novel pyrimidinediones of the formula:

(1)



where:

R is alkyl of 1 through 10 carbon atoms,

substituted alkyl of 1 through 10 carbon atoms,
aryl of 6 through 14 carbon atoms,
substituted aryl of 6 through 14 carbon atoms,
aralkyl of 7 through 15 carbon atoms,
substituted aralkyl of 7 through 15 carbon atoms,
alkenyl of 3 through 10 carbon atoms,
alkynyl of 3 through 10 carbon atoms,
cycloalkyl of 3 through 12 carbon atoms,
cycloalkenyl of 4 through 12 carbon atoms,
cycloalkyl alkyl of 4 through 13 carbon atoms,
cycloalkenyl alkyl of 5 through 13 carbon atoms,
(substituted cycloalkyl)alkyl of 5 through 14 carbon atoms,
(substituted cycloalkenyl)alkyl of 5 through 14 carbon atoms,
cyano or



which is a cycloimino group of 2 through 14 carbon atoms having a maximum of 3 hetero atoms.

$R_1$, $R_2$ and $R_3$ are the same or different and are selected from the group consisting of hydrogen, alkyl of $C_1$–$C_4$, haloalkyl of $C_1$–$C_4$, alkoxy of $C_1$–$C_4$, alkylthio of $C_1$–$C_4$, and alkenyl of $C_2$–$C_4$;

Y is halogen including chlorine, bromine, fluorine and iodine;

*n* is 3, 4, or 5; and

*m* is 0 or 1; with the proviso that when $R_1$, $R_2$, and $R_3$ are hydrogen, *m* is 1.

As is well known in the art, herbicidal compounds frequently are utilized in the form of a herbicidal equivalent salt, addition compound, or complex derivative



thereof for some formulating or handling advantage the derivative may provide over the compound per se. Similarly, it is to be understood that also included within the scope of this invention are salts, addition compounds, and complexes of the pyrimidinediones of Formula 1 which can be formed from Lewis bases and acids and which exert a herbicidal effect similar to that of the uracils per se. Lewis bases and acids are generally described in the following texts: (1) J. Hine, Physical Organic Chemistry, McGraw-Hill Book Company, Inc., New York, 1956, chapter 2; (2) W. F. Luder and S. Zuffanti, The Electronic Theory of Acids and Bases, John Wiley and Sons, Inc., New York, 1946; (3) E. S. Gould, Mechanism and Structure in Organic Chemistry, Holt, Rinehart and Winston, New York, 1959, Chapter 4; (4) E. M. Arnett, Progress in Physical Organic Chemistry, vol. 1, edited by S. G. Cohen, A Streitwieser, Jr., and R. W. Taft, Interscience Publishers, Inc., New York, 1963, chapter entitled "Quantitative Comparisons of Weak Organic Bases;" and (5) G. A. Olah, editor, Friedel-Crafts and Related Reactions, volumes 1 and 2, Interscience Publishers, Inc., New York, 1963.

Non-limiting illustrations of compounds shown in the above texts which can be used to form salts, addition compounds, or complexes with the pyrimidinediones of Formula 1 for purposes of the present invention are:

Bases

sodium hydroxide
potassium hydroxide
lithium hydroxide
calcium hydroxide
magnesium hydroxide
barium hydroxide
strontium hydroxide
ferric hydroxide
manganese hydroxide
quaternary ammonium hydroxides
primary, secondary and tertiary amines
heterocyclic amines
guanidines
amidines
phenols
cresols
naphthols

Acids

Friedel-Crafts catalysts
halogenated benzoic acids
mineral acids
organic sulfonic acids
halogenated aliphatic acids
halogenated aryloxyaliphatic acids
organic aliphatic and aromatic acids

In Formula 1 the term "substituted alkyl" is intended to include such radicals as

bromoalkyl of 1 through 10 carbon atoms
chloroalkyl of 1 through 10 carbon atoms
hydroxyalkyl of 1 through 10 carbon atoms
alkoxyalkyl of 2 through 12 carbon atoms
alkoxy carbonyl alkyl of 3 through 12 carbon atoms
dialkyl amino alkyl of 3 through 12 carbon atoms and
cyanoalkyl of 2 through 10 carbon atoms

Similarly, the term "aryl" and "substituted aryl" embrace radicals such as

phenyl
naphthyl
o-biphenylyl
pyridyl
chlorophenyl
bromophenyl

alkoxyphenyl
dichlorophenyl
dibromophenyl
fluorophenyl
trichlorophenyl
alkylphenyl of 7 through 11 carbon atoms
dialkylphenyl of 8 through 12 carbon atoms
chloroalkylphenyl of 7 through 10 carbon atoms
nitrochlorophenyl
nitrophenyl
dichloronitrophenyl
chloroalkoxyphenyl of 7 through 11 carbon atoms
trifluoromethylphenyl
alkylnaphthyl of 11 through 16 carbon atoms
chloronaphthyl
tetrahydronaphthyl and
indenyl

The terms "aralkyl" and "substituted aralkyl" are intended to include such radicals as

furfuryl
benzyl
phenylalkyl of 8 through 11 carbon atoms (total)
chlorobenzyl
dichlorobenzyl
alkylbenzyl of 8 through 11 carbon atoms (total)
dialkylbenzyl of 9 through 13 carbon atoms (total)
nitrobenzyl
alkoxybenzyl of 8 through 11 carbon atoms (total) and
naphthylmethyl

The terms "cycloalkyl," "cycloalkenyl," "cycloalkyl alkyl," and "cycloalkenyl alkyl" will include

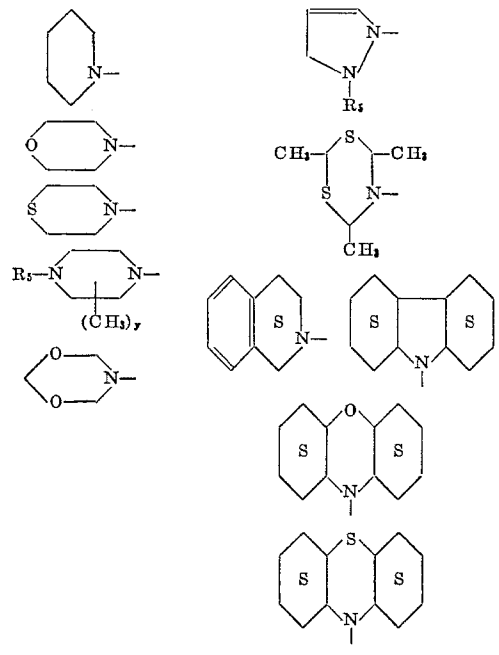
cyclopropyl
cyclohexyl
cyclohexenyl
cyclohexylalkyl
cyclohexenylalkyl
cyclopentyl
cyclopentenyl
cyclopentylalkyl
cyclopentenylalkyl
norbornyl
norbornenyl
norbornylalkyl
norbornenylalkyl
bicyclo(2,2,2)octyl
bicyclo(2,2,2)octenyl
bicyclo(2,2,2)octylalkyl
bicyclo(2,2,2)octenylalkyl
cyclopropyl
cyclobutyl
cyclobutylalkyl
cyclobutenyl
cyclobutenylalkyl
hexahydroindanyl
tetrahydroindanyl
hexahydroindenyl
hexahydroindenyl alkyl
tetrahydroindanyl alkyl
hexahydroindanyl alkyl
hexahydro-4,7-methanoindenyl
tetrahydro-4,7-methanoindanyl
hexahydro-4,7-methanoindanyl
hexahydro-4,7-methanoindenyl alkyl
tetrahydro-4,7-methanoindanyl alkyl
hexahydro-4,7-methanoindanyl alkyl
decahydronaphthyl
decahydronaphthyl alkyl
tetrahydronaphthyl
tetrahydronaphthyl alkyl
decahydro-1,4-methanonaphthyl
decahydro-1,4-methanonaphthyl alkyl
octahydro-1,4-methanonaphthyl
octahydro-1,4-methanonaphthyl alkyl
decahydro-1,4,5,8-dimethanonaphthyl



decahydro-1,4-5,8-dimethanonaphthyl alkyl
octahydro-1,4-5,8-dimethanonaphthyl and
octahydro-1,4-5,8-dimethanonaphthyl alkyl

These cyclic substituents can be further substituted with alkyl groups containing 1 through 4 carbon atoms, methoxy, chlorine and bromine.

The term "cycloimino" is intended to include such radicals as



where $R_5$ is alkyl of 1 through 4 carbon atoms or

$$-CH_2CH_2OH$$

and $y$ is 0–4.

It is obvious from the above that the term "cycloimino" as used herein includes hetero atoms but it is intended in accordance with the invention that the number of hereto atoms in the



group of the 3-position of the pyrimidinedione ring does not exceed three.

## Utility

The pyrimidinediones of Formula 1 and their various salt, complex, and addition compound derivatives are novel herbicides for the control of undesirable vegetation and quite surprisingly exhibit the unusual capability of having a highly potent activity in general weed control sites such as railroad rights-of-way, roadsides, fence rows, and areas adjacent cropland and industrial plants and in addition having a selective herbicidal activity in crops such as in sugar cane so that weed control can be accomplished without damage to the crop.

The concentrations at which the compounds of this invention are to be used will vary according to the result desired, the type of vegetation, the formulation used, the mode of application, weather conditions, foliage density, and other similar factors. Since so many factors play a role, it is not possible to indicate a concentration suitable for all situations. Generally, when they are used for pre-emergence treatment in crops, these pyrimidinediones are used at concentrations of at least about 0.5 pound of active ingredient per acre. Concentrations of from 0.5 to 3 pounds per acre are preferred.

When used in non-crop applications, the pyrimidinediones are used at concentrations of from 10 to 30 pounds of active ingredient per acre. The optimum concentrations to be used in any particular application will be readily apparent to one skilled in the art.

The preferred compounds for use according to this invention are the compounds of Formula 1 where

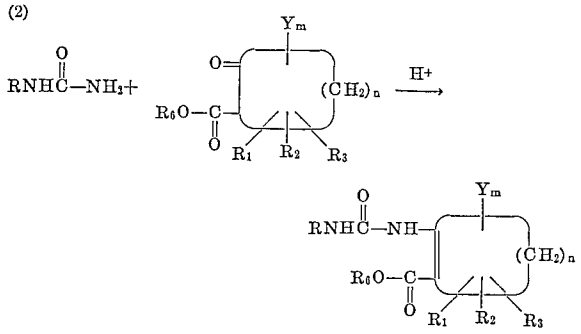
R is an alkyl or alkenyl radical containing 3 through 6 carbon atoms, phenyl, substituted phenyl, cycloalkyl radical of 3 through 10 carbon atoms or cycloalkenyl radical of 4 through 10 carbon atoms,
*n* is 3, 4, or 5,
$R_1$ is methyl or ethyl
$R_2$ and $R_3$ are H

and their sodium, and potassium salts, nitrogenous base addition compounds, phenol complexes and acid addition compounds.

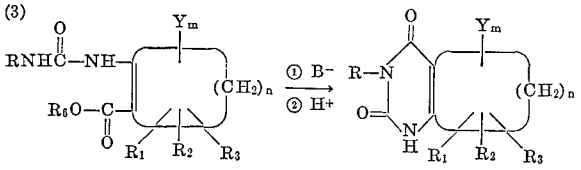
## PREPARATION OF COMPOUNDS

The pyrimidinediones of Formula 1 can be prepared according to the following equations:

(2)



(3)



In Equations 2 and 3, R, $R_1$, $R_2$, $R_3$, Y, *m* and *n* have the same meaning as in Formula 1; $R_6$ is an alkyl radical of 1–6 carbon atoms, $H^+$ is an acid, and $B^-$ is a base.

The reaction sequence consists of two steps: first, the acid catalyzed condensation of a 1-substituted urea with a substituted alkyl 2-cycloalkanone-1-carboxylate to give the ureido intermediate; and, second, the alkaline ring closure of this intermediate to the salt of the pyrimidinedione product, followed by treatment with acid to liberate the free pyrimidinedione.

The first step, illustrated by Equation 2, is best carried out by dissolving or suspending the 1-substituted urea in a solvent such as xylene, benzene or a mixture of benzene and dioxane, and heating to reflux. To this solution is then added a mixture of the acid catalyst in slight excess of the alkyl 2-cycloalkanone-1-carboxylate. Suitable acid catalysts are p-toluenesulfonic, sulfuric, benzenesulfonic, phosphoric, polyphosphoric, formic, chloroacetic, or Lewis acids such as $BF_3$ and $AlCl_3$. The mixture is stirred and heated to reflux until the water, split out during the reaction and collected in a suitable apparatus, ceases to be given off. The clear solution is decanted or filtered and then concentrated to give the intermediate ureido derivative.

In the second step, illustrated by Equation 3 the ureido intermediate is dissolved in alcohol containing a slight excess of base, preferably an alkali metal alkoxide or hydroxide, and the solution is refluxed for a short time. This step effects the ring closure. The salt of the pyrimidinedione is obtained by concentrating this solution to dryness. The free pyrimidinedione is prepared by acidifying an aqueous solution of the salt, and then isolating the solid. For the most part the compounds prepared by this route will consist of a mixture of optical isomers.

Many of the cyclic keto esters needed to prepare the novel pyrimidinediones of this invention may be obtained by Dieckmann cyclization of substituted adipic, pimelic, or suberic acids. All of the ring sizes encompassed by this invention may be included according to this process. The diacids in turn may be purchased or prepared according to classical methods of organic chemistry from such readily available starting materials as substituted phenols or butadiene dimers. Cyclic keto esters having ring sizes of 5 and 7 carbon atoms are most easily prepared by these processes.

According to the above, substituted phenols are hydrogenated to the corresponding cyclohexanols, which are then oxidized to the corresponding adipic acids. Details for carrying out these oxidations may be found in J.A.C.S., 52, 3235 (1930); U.S. 1,921,101; Ger. 854,507; J. Chem. Ed., 10 114 (1933); and C.A., 31 3875 (1937). The butadiene dimers may be further substituted and then likewise oxidized to the substituted suberic acids.

Another source of these cyclic keto esters is the carbonylation of available cyclic ketones according to the following references.

(1) J.A.C.S., 70, 2287 (1948)
(2) J.A.C.S., 70, 497 (1948)
(3) J.A.C.S., 66, 1768 (1944)
(4) J.A.C.S., 69, 2677
(5) J.A.C.S., 70, 2884

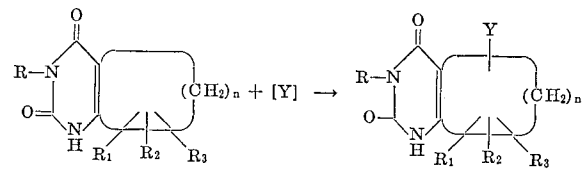
These procedures are especially useful, though not restricted to, cyclic keto esters having a six membered ring. Alkyl substituted cyclic ketones possessing a six numbered ring are readily available from both synthetic and natural sources. A wide variety of cyclic keto esters is, therefore, automatically made available by this technique.

Another source of substituted cyclic keto esters arises by carrying out one or more reactions on those available cyclic keto esters which are either unsubstituted or not as highly substituted as desired. 3-alkyl substituted cyclic keto esters are made especially available by this procedure. For further details see:

(1) J.O.C., 29, 2781 (1964), and
(2) J.C.S., 1942, 559.

Still another source of these intermediates is from the Kolbe or modified Kolbe reactions which are used to produce substituted salicylic acids. These acids are esterified by routine procedures. They are reduced by hydrogenation to the corresponding cyclohexanol carboxylic esters and then mildly oxidized to the intermediate cyclic keto esters.

Finally many halogenated compounds within the scope of this invention may also be prepared by halogenating a previously prepared substituted pyrimidinedione. For example:



Substitution by the halogen takes place most easily in the positions allylic to the double bond.

Chlorination may be carried out either with chlorine per se or sulfuryl chloride. When direct chlorination is desired solvents such as acetic acid are useful for promoting and moderating the reaction. The reaction may be carried out most conveniently from room temperature to the boiling point of acetic acid. A higher percentage of allylic chlorides is obtained at the higher temperatures. Chlorination with sulfuryl chloride is less vigorous than direct chlorination, but may be desirable under certain circumstances.

Bromination to form the substituted pyrimidinediones of this invention may be carried out directly with elemental bromine or with N-bromosuccinimide.

Additional information pertinent to the halogenations described here may be found in "Synthetic Organic Chemistry" R. B. Wagner and H. D. Zook, pp. 98, 104 (1956).

Metal salts of the pyrimidinediones of Formula 1 are prepared by conventional methods such as dissolving the free pyrimidinedione in an aqueous or nonaqueous solution of at least an equimolar amount of a base or basic salt containing the desired cation. For example, a sodium salt can be prepared by dissolving the pyrimidinedione in water containing an equilmolar amount of sodium hydroxide. The salt can then be isolated from the solution by removal of the water. The pyrimidinedione salts which are not soluble in water can be best prepared by treating an aqueous solution of an alkali metal salt of the pyrimidinedione with an aqueous solution of a water-soluble salt of the metal.

Quaternary ammonium salts of the pyrimidinediones of Formula 1 are prepared by the reaction of the substituted pyrimidinedione with an appropriate quaternary ammonium hydroxide. Since these hydroxides are generally available in solution, the reaction is most conveniently carried out in the same solvent. If the solvent-free salt is desired, it can be easily prepared by removing the solvent.

Alternately, the quarternary ammonium salts can be prepared from the corresponding sodium salts in a dry inert solvent such as toluene or xylene. The appropriate quarternary ammonium halide is then added with stirring and, if necessary, mild heating. The sodium halide which forms is removed by filtration, leaving the quaternary ammonium salt of the pyrimidinediones in solution. If desired, the solvent-free salt can be prepared by removing the solvent, preferably in vacuum.

Additional compounds of the pyrimidinediones of Formula 1 with Lewis bases such as amines can be prepared by mixing together equimolar quantities of an appropriate pyrimidinedione and a nitrogenous base. The mixture is gradually heated, with stirring, until a clear melt is formed. On cooling, the addition compound crystallizes. This product can then be recrystallized from a solvent such as benzene, cyclohexane, nitromethane, or acetonitrile.

It is sometimes advantageous to use an inert solvent medium to carry out the reaction. Such a solvent moderates the reaction by acting as a heat sink, and allows better control of the reaction, especially if it is being carried out on a large scale. Suitable inert solvents are benzene, cyclohexane, nitromethane, acetonitrile and dioxane.

When an inert solvent is used, the addition compounds are prepared by dissolving the amine in the solvent and then adding the pyrimidinedione gradually, with stirring. Stirring is continued for from ten minutes to two hours. Mild heating may be necessary. Some addition compounds precipitate and can be removed by filtration. Other addition compounds are isolated by evaporation of the solvent. The addition compounds prepared in this way are suitable for use without further purification, but can be purified by recrystallization if desired.

In some instances, the pyrimidinedione and amine are highly soluble in the inert solvent, but the addition compound is not, and so it can be filtered off pure when the reaction is complete.

Complexes of the pyrimidinediones of Formula 1 with a Lewis base such as phenol can be formed by comelting the pyrimidinediones and phenol in a 1:1 to 2:1 (pyrimidinediones:phenol) ratio. They can also be formed by codissolving the reactants, in the same ratio, in a solvent such as nitromethane or a mixture of nitromethane and cyclohexane. Process conditions and isolation procedures are the same as those described above for the additional compounds.

## HERBICIDIAL COMPOSITIONS

The compounds of the invention can be prepared for use by incorporating them with adjuvants.

The amount of herbicide in such preparations can vary over a wide range according to need. Generally speaking, they will contain from about 0.5% to 95% by weight of a pyrimidinedione.

Powder and dust preparations can be made by mixing pyrimidinediones of the invention with finely-divided solids such as talcs, natural clays, pyrophillite, diatomaceous earth; flours such as walnut shell, wheat, redwood, soyabean and cotton seed; or inorganic substances such as magnesium carbonate, calcium carbonate, calcium phosphate, sulfur and lime. These preparations are made by thoroughly blending the active ingredient and the solid. The particles in such preparations should be less than 50 microns in average diameter, preferably about 20 microns.

Water-soluble powders can be prepared by blending a suitable pyrimidinedione with such water-soluble alkaline powders as silicates, carbonates, phosphates, or hydroxides, and optionally with water-soluble diluents such as urea or dextrose.

Granules and pellets can be made by mixing a finely-divided pyrimidinedione with a suitable clay such as kaolinite, montmorillonite or attapulgite, moistening this mixture with from 15 to 20% by weight of water, and then extruding the mass through a suitable die under pressure. The extrusions are cut into pre-determined lengths and then dried. These pellets can be granulated if desired.

Granules or pellets can also be prepared by spraying a suspension or solution of a pyrimidinedione onto the surface of a preformed granule of clay, vermiculite or other suitable granular material. If the pyrimidinedione is in solution, it will penetrate into the pores of the granule and so will adhere without the aid of a binding agent when the active material is insoluble in the liquid and is carried as a suspension, it is preferable that a binding agent such as goulac, dextrin, swollen starch, glue or polyvinyl alcohol be added. In either case, the granule is then dried and ready for use.

The pyrimidinediones can also be prepared in liquids. Water and aliphatic and aromatic hydrocarbons, especially those derived from petroleum and having boiling points of from 125° C. to 400° C. are preferred. Hydrocarbons having lower boiling points should not be used because of their undesirable volatilization characteristics and inflammability. These liquid preparations are made by dissolving the active in the liquid, or, if the active is insoluble in the liquid, by milling the components in a mill such as a pebble mill until the particles have average diameters of from 1 to 50 microns, preferably 5 to 20 microns.

The herbicidal preparations, whatever physical form they take, can also contain a surface-active agent. The surfactant renders the preparations readily dispersible in liquids and improves their action on waxy leaves and the like. For general application, surface-active agents are used in the preparations at concentrations of from about 1 to 10% by weight. Levels of from 0.5 to 6 parts of surfactant for each part of pyrimidinedione, however, give unusual and unexpected results. Preparations having these higher levels of surfactants show greater herbicidal effectiveness than can be expected from a consideration of activity of the components used separately.

The term "surface-active agent" is intended to include wetting agents, dispersing agents, suspending agents and emulsifying agents. Surface-active agents suitable for use are set forth in "Detergents and Emulsifiers . . . up-to-date," 1962, John W. McCutcheon, Inc., Morristown, N.J. Other surface-active agents which can be used in these preparations are listed in U.S. Patents 2,139,276; 2,412,510; 2,426,417; 2,655,447; and Bulletin E–607 of the Bureau of Entomology and Plant Quarantine of the U.S. Department of Agriculture.

The preparations can also contain adhesives, corrosion inhibitors, antifoam agents, dyes and pigments, anticaking agents, and hard water stabilizers.

The salts of the compounds of Formula 1 are especially advantageous for use as herbicides because many are soluble in water and can be applied as aqueous solutions.

## FORMULATION WITH OTHER HERBICIDES

The herbicidal compositions of this invention can be formulated to contain two or more of the pyrimidinediones. They can also be formulated to contain other known herbicides in addition to the pyrimidinediones to give compositions which have advantages over the individual components.

Among the known herbicides which can be combined with the pyrimidinediones are:

### Substituted ureas

3-(3,4-dichlorophenyl)-1,1-dimethylurea
3-(4-chlorophenyl)-1,1-dimethylurea
3-phenyl-1,1-dimethylurea
3-(3,4-dichlorophenyl)-3-methoxy - 1,1 - dimethylurea
3-(4-chlorophenyl)-3-methoxy-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-(3,4-dichlorophenyl)-1,1-diethylurea
3-(p-chlorophenoxyphenyl)-1,1-dimethylurea

These ureas can be mixed with the pyrimidinediones 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-ethylamino-4-(2-methoxyethylamine) - 6 - chloro-s-triazine
2-isopropylamino - 4 - methoxyethylamino - 6 - methylmercapto-s-triazine
2-methylmercapto-4,6-bis(isopropylamino)-s-triazine
2-methylmercapto-4,6-bis(ethylamino)-s-triazine
2-methylmercapto - 4 - ethylamino - 6 - isopropylamino-s-triazine
2-methoxy-4,6-bis(ethylamino)-s-triazine
2-methoxy - 4 - ethylamino - 6 - isopropylamino-s-triazine
2-chloro-4,6-bis(isopropylamino)-s-triazine
2-tert-butylamino - 4 - ethylamino - 6 - methylmercapto-s-triazine
2-isopropylamino - 4 - methoxypropylamino - 6 - methylmercapto-s-triazine

These triazines can be mixed with the pyrimidinediones in proportions of from 1:4 to 4:1, respectively, the preferred ratio being 1:2 to 2:1.

### Phenols

4-cyano-2,6-diiodophenol and its salts
4-cyano-2,6-dibromophenol and its salts
dinitro-o-sec-butylphenol and its salts
pentachlorophenol and its salts

These phenols can be mixed with the pyrimidinediones in the proportions 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 mixed with the pyrimidinediones in the listed respective proportions:

A.

2,3,6-trichlorobenzoic acid and its salts
2,3,5,6-tetrachlorobenzoic acid and its salts
2-methoxy-3,5,6-trichlorobenzoic acid and its salts
2-methoxy-3,6-dichlorobenzoic acid and its salts
3-amino-2,5-dichlorobenzoic acid and its salts
3-nitro-2,5-dichlorobenzoic acid and its salts
2-methyl-3,6-dichlorobenzoic acid and its salts
2,3-dichloro-6-methylbenzoic 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 and 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)butyric acid and its salts and esters
2,3,6-trichlorobenzyloxypropanol

Mixed in a 1:20 to 8:1 ratio, preferably a 1:4 to 4:1 ratio.

B.

2,6-dichlorobenzonitrile

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

C.

trichloroacetic acid and its salts

Mixed in a 1:4 to 25:1 ratio, preferably a 1:2 to 10:1 ratio.

D.

2,2-dichloropropionic acid and its salts

Mixed in a 1:4 to 10:1 ratio, preferably a 1:2 to 5:1 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, 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 1:2 to 12:1 ratio.

G.

2,3,6-trichlorophenylacetic acid and its salts

Mixed in a 1:4 to 10:1 ratio, preferably a 1:2 to 5:1 ratio.

H.

2-chloro-N,N-diallylacetamide
2-methyl-6-tert-butyl-N-methoxymethyl-2'-bromo-acetanilide
N-isopropyl-N-phenyl chloroacetamide

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 uracils in the listed proportions:

A.

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 1:10 to 40:1 ratio, preferably a 1:5 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
sodium pentaborate
polychlorborate
unrefined borate ore, such as borascu

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

E.

ammonium thiocyanate

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

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

Other organic herbicides

The following herbicides can be mixed with the pyrimidinediones in the listed respective proportions:

A.

5,6-dihydro-(4A,6A)-dipyrido-(1,2-A,2',1'-C) pyrazinium dibromide
1,1'-dimethyl-4,4'-dipyridinium di-(methyl sulfate)

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 5:1 ratio.

C.

3,6-endoxohexahydrophthalic acid

Mixed in a 1:4 to 20:1 ratio, preferably a 1:2 to 10:1 ratio.

D.

diphenylacetonitrile
N,N-dimethyl-α,α-diphenylacetamide
N,N-di-(n-propyl)-2,6-dinitro-4-methylaniline
N-methyl-N-n-butyl-2,6-dinitro-4-trifluoromethylaniline

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

E.

O-(2,4-dichlorophenyl)-O-methyl-isopropyl-phosphoramidothioate
2,3,5,6-tetrachloroterephthalic acid, dimethyl ester
thiomethyl ester of 2,3,5,6-tetrachloro-4-carbomethoxy benzoic acid

Mixed in a 1:4 to 20:1 ratio, preferably a 1:3 to 15:1 ratio.

F.

2,4-dichloro-4'-nitrodiphenyl ether

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

G.

1-(3,4-dichlorophenyl)-3,5-dimethylhexahydro-1,3,5-triazinone-2
1-phenyl-3-methyl-5-allyl-hexahydro-1,3,5-triazinone-2

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

Substituted uracils

The pyrimidinediones of this invention can be mixed with substituted uracils, in the respective proportions listed below. Methods for the preparation of the listed uracils which are novel can be found in copending applications Ser. Nos. 233,952 filed Oct. 29, 1962 and 237,311 filed Oct. 22, 1963.

5-bromo-3-sec-butyl-6-methyluracil
5-bromo-3-sec-butyl-6-methyluracil, sodium salt
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-bromo-3-cyclohexylmethyl-6-methyluracil
5-chloro-3-cyclohexylmethyl-6-methyluracil
3-cyclohexyl-5-methoxy-6-methyluracil
3-sec-butyl-5,6-dimethyluracil
5-bromo-6-methyl-3-norbornylmethyluracil
3-sec-butyl-6-methyl-5-nitrouracil
3-cyclohexyl-5,6-dimethyluracil
5-bromo-3-cyclohexyl-6-methyluracil
5-chloro-3-phenyl-6-methyluracil
5-bromo-6-methyl-3-(1-piperidino)uracil
5-chloro-6-methyl-3-(1-piperidino)uracil
5-bromo-6-methyl-3-(1-pyrrolidinyl)uracil
5-chloro-6-methyl-3-(1-pyrrolidinyl)uracil
5-bromo-6-methyl-3-(hexahydro-1-azepinyl)uracil
5-chloro-6-methyl-3-(hexahydro-1-azepinyl)uracil
6-methyl-3-(hexahydro-1-azepinyl)uracil
5-bromo-6-methyl-3-(4-morpholino)uracil

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

3-cyclohexyl-6-methyluracil
3-cyclohexyl-6-ethyluracil
3-cyclohexyl-6-sec-butyluracil
3-cyclohexyl-6-propyluracil
3-cyclophentyl-6-methyluracil
3-cyclohexyluracil

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

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

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

3-isopropyl-1-trichloromethylthio-5-bromo-6-methyluracil
3-cyclohexyl-1-trichloromethylthio-5-bromo-6-methyluracil
3-sec-butyl-1-acetyl-5-bromo-6-methyluracil
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.

## EXAMPLES

This invention will be better understood by referring to the following illustrative examples:

### Preparation of compounds

EXAMPLE 1.—PREPARATION OF 3 - CYCLOHEXYL-5,6-DIHYDRO-7-METHYL-5-CYCLOPENTA[D]PYRIMIDINE-2,4(1H,3H)-DIONE

A mixture of 142 parts by weight of cyclohexylurea, 170 parts by weight of ethyl 3-methyl-2-cyclopentanone-1-carboxylate, 10 parts by weight of p-toluene sulfonic acid and 879 parts by weight of benzene is stirred at reflux for 8 hours. During this time, the water given off by the reaction is trapped out. The solvent is stripped at reduced pressure and the residue is dissolved in 1000 parts by weight of absolute alcohol containing 80 parts by weight of sodium methoxide, and is refluxed for 30 minutes. The solvent is stripped at reduced pressure and the residue dissolved in water, cooled, and acidified. The solid is filtered off, dried, and recrystallized from ethanol to give the desired 3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4-(1H,3H)-dione, melting at 201–203° C.

EXAMPLE 2.—PREPARATION OF 3(2-METHYLCYCLOHEXYL)-5,6-DIHYDRO-6-ETHYL - 5 - CYCLOPENTA[D] PYRIMIDINE-2,4(1H,3H)-DIONE

A mixture of 184 parts by weight of ethyl 4-ethyl-2-cyclopentanone-1-carboxylate, 156 parts by weight of 2-methylcyclohexylurea, 10 parts by weight of p-toluene sulfonic acid, and 1000 parts by weight of toluene is stirred at reflux for 6 hours. During this time the water given off by the reaction is trapped out. The solvent is stripped from the resulting solution at reduced pressure. The residue is then dissolved in 793 parts by weight of absolute ethyl alcohol.

To this solution is added a mixture of 80 parts by weight of sodium methoxide in 300 parts by weight of absolute ethyl alcohol. The mixture is then refluxed for 20 minutes. The solvent is stripped from this mixture and the resulting solid is dissolved in 3000 parts by weight of water.

This solution is cooled, acidified with excess hydrochloric acid and the solid is filtered off. Recrystallization of this solid from ethanol or acetonitrile gives pure 3(2-methylcyclohexyl - 5,6 - dihydro-6-ethyl - 5 - cyclopenta-[D]-pyrimidine-2,4(1H,3H)-dione.

The compounds listed in the following table can be prepared according to the procedures of Example 1 or 2 by substituting equivalent amounts of the listed ureas, and alkyl substituted-2-cycloalkanone-1-carboxylates for those used in the examples:

TABLE I.—PREPARED ACCORDING TO PROCEDURES OF EXAMPLE 1

| Substituted urea | Parts by weight | Ester of the cycloalkanone carboxylate | Parts by weight | Substituted[D]pyrimidine-2,4(1H,3H)-dione product |
|---|---|---|---|---|
| 1-ethylpropylurea | 130 | Ethyl-3-methyl-2-cyclopentanone-1-carboxylate | 176 | 3-(1-ethylpropyl)-5,6-dihydro-7-methyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione. |
| n-Butylurea | 116 | Ethyl-3-ethyl-cyclopentanone-1-carboxylate | 190 | 3-n-butyl-5,6-dihydro-7-ethyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione. |
| n-Hexylurea | 144 | Ethyl-3-n-propyl-2-cyclopentanone-1-carboxylate. | 204 | 5,6-dihydro-3-n-hexyl-7-n-propyl-5-cyclopenta [D]pryimidine-2,4(1H,3H)-dione. |
| Isooctylurea | 172 | Ethyl-3-n-butyl-2-cyclopentanone-1-carboxylate | | 7-n-butyl-5,7-dihydro-3-isooctyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| α-Naphthylurea | 204 | Ethyl-3-methyl-2-cyclopentanone-1-carboxylate | 176 | 7-methyl-5,6-dihydro-3-(α-napthyl)-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione. |
| Tert-butylurea | 116 | Ethyl-3-methyl-2-cyclohexanone-1-carboxylate | 201 | 3-tert-butyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione. |
| n-Octylurea | 172 | Ethyl-3-ethyl-2-cyclohexanone-1-carboxylate | 215 | 7,8-diethyl-3-n-octyl-5,6,7,8-tetrahydro-5-cyclo-hexa[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclooctylurea | 170 | Methyl-3-methyl-2-cyclopentanone-1-carboxylate. | 170 | 3-cyclooctyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclopentenylurea | 126 | do | 170 | 3-cyclopentenyl-5,6-dihydro-7-methyl-5-cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione. |
| Fenchylurea | 216 | do | 170 | 3-fenchyl-5,6-dihydro-7-methyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| Methylurea | 74 | Ethyl-3-sec-butyl-2-cyclohexanone-1-carboxylate | 218 | 7-sec-butyl-3-methyl-5,6,7,8-tetrahydro-5-cyclo-hexa[D]pyrimidine-2,4(1H,3H)-dione. |
| 2-methylcyclohexylurea | 156 | Ethyl-3-methyl-2-cyclopentanone-1-carboxylate | 176 | 5,6-dihydro-7-methyl-3-(2-methylcyclohexyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione. |
| Sec-butylurea | 116 | do | 176 | 3-sec-butyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4-(1H,3H)-dione. |
| Isopropylurea | 102 | do | 176 | 5,6-dihydro-3-isopropyl-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione. |
| Tert-butylurea | 116 | do | 176 | 3-tert-butyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclohexylurea | 142 | Ethyl-3,5,5-trimethyl-2-cyclopentanone-1-carboxylate. | 204 | 3-tert-butyl-8-methyl-5,6,7,8-tetrahydro-5-cyclo-h xa[D]pyrimidine-2,4(1H,3H)-dione. |
| Sec-butylurea | 116 | do | 204 | 7,8-diethyl-3-n-octyl-5,6,7,8-tetrahydro-5-cyclo-hexa[D]pyrimidine-2,4(1H,3H)-dione. |
| Chloronorbornylurea | 188.5 | Methyl-3-ethyl-2-cyclopentanone-1-carboxylate | 184 | 3-chloronorbornyl-5,6-dihydro-7-ethyl-5-cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione. |
| Bornylurea | 216 | do | 184 | 3-bronyl-5,6-dihydro-7-ethyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| Norbornylurea | 154 | do | 190 | 5,6-dihydro-7-ethyl-3-norbornyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| Cyclopropylmethylurea | 115 | Ethyl-3-methyl-2-cycloheptanone-1-carboxylate | 216 | 3-cyclopropylmethyl-9-methyl-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione. |
| 2-methoxyethylurea | 118 | Ethyl-3-n-butyl-2-cycloheptanone-1-carboxylate | 258 | 9-n-butyl-3-(2-methoxy-ethyl)-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione. |
| Allylurea | 100 | Ethyl-3-methyl-2-cyclohexanone-1-carboxylate | 201 | 3-allyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa-[D]pyrimidine-2,4(1H,3H)-dione. |
| Decenylurea | 200 | Ethyl-3-ethyl-2-cycloheptanone-1-carboxylate | 230 | 3-decenyl-9-ethyl-5,6,7,8-tetrahydro-5-cyclohepta-[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclohexylurea | 142 | Ethyl-3-methyl-2-cyclohexanone-1-carboxylate | 201 | 3-cyclohexyl-8-methyl-5,6,7,8-tetrahydro-5-cyclo-hexa[D]pyrimidine-2,4(1H,3H)-dione. |
| 2-methylcyclohexylurea | 156 | Ethyl-3-ethyl-2-cyclohexanone-1-carboxylate | 215 | 8-ethyl-3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione. |
| Sec-butylurea | 116 | Ethyl-3-methyl-2-cyclohexanone-1-carboxylate | 201 | 3-sec-butyl-8-methyl-5,6,7,8-tetrahydro-5-cyclo-hexa[D]pyrimidine-2,4(1H,3H)-dione. |
| Isopropylurea | 102 | do | 201 | 3-isopropyl-8-methyl-5,6,7,8-tetrahydro-5-cyclo-hexa[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclohexylurea | 142 | Ethyl-3-methyl-2-cycloheptanone-1-carboxylate | 201 | 3-cyclohexyl-9-methyl-5,6,7,8-tetrahydro-5-cyclo-hepta[D]pyrimidine-2,4(1H,3H)-dione. |
| 2-methylcyclohexylurea | 156 | do | 201 | 9-methyl-3-(2-methylcyclohexyl)-5,6,7,8-tetra-hydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione. |
| Sec-butylurea | 116 | do | 201 | 3-sec-butyl-5,6,7,8-tetrahydro-9-methyl-5-cyclo-hepta[D]pyrimidine-2,4(1H,3H)-dione. |
| Isopropylurea | 102 | do | 201 | 3-isopropyl-9-methyl-5,6,7,8-tetrahydro-5-cyclo-hepta[D]pyrimidine-2,4(1H,3H)-dione. |

TABLE I.—PREPARED ACCORDING TO PROCEDURES OF EXAMPLE 1.—Continued

| Substituted urea | Parts by weight | Ester of the cycloalkanone carboxylate | Parts by weight | Substituted[D]pyrimidine-2,4(1H,3H)-dione product |
|---|---|---|---|---|
| Tert-butylurea | 116 | do | 201 | 3-tert-butyl-9-methyl-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclopentylurea | 128 | Ethyl-4-methyl-2-cyclopentanone-1-carboxylate | 187 | 3-cyclopentyl-5,6-dihydro-6-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione. |
| 1-(4-methoxycyclo-hexyl)-urea. | 172 | Ethyl-4-ethyl-2-cyclopentanone-1-carboxylate | 201 | 5,6-dihydro-6-ethyl-3(4-methoxycyclohexyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione. |
| PREPARED ACCORDING TO PROCEDURES OF EXAMPLES 1 AND 2 | | | | |
| 1-(3-4-dichloro-cyclo-hexyl)-urea. | 211 | Ethyl-4-n-propyl-2-cyclopentanone-1-carboxyl-ate. | 229 | 5,6-dihydro-3-(3,4-dichlorocyclohexyl)-6-n-propyl 5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclooctenylurea | 168 | Methyl-4-iospropyl-2-cyclopentanone-1-carboxyl-ate. | 190 | 3-cycloctenyl-5,6-dihydro-6-isopropyl-5-cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione. |
| Carvacrylurea | 191 | Methyl-4-ethyl-2-cyclopentanone-1-carboxylate | 187 | 3-carvacryl-5,6-dihydro-6-ethyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| Isopropylurea | 102 | Methyl-4-n-butyl-2-cyclopentanone-1-carboxyl-ate. | 190 | 6-n-butyl-5,6-dihydro-3-isopropyl-5-cyclopenta [D]pyrimidine-2,4(1H,3H)-dione. |
| n-Amylurea | 130 | Ethyl-4-methyl-2-cyclopentanone-1-carboxylate | 187 | 3-n-amyl-5,6-dihydro-6-methyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| Bromonorbornylurea | 231 | do | 187 | 3-bromonorbornyl-5,6-dihydro-6-methyl-5-cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione. |
| Cyclohetenylurea | 156 | do | 202 | 3-cycloheptenyl-5,6-dihydro-6-methyl-5-cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione. |
| Decahydronaphthylurea | 196 | Ethyl-5-methyl-2-cyclopentanone-1-carboxylate | 176 | 3-decahydronaphthyl-5,6-dihydro-5-methyl-5-cyclopenta[D]pyrimidine-2,4-(1H,3H)-dione. |
| 1-(4-isopropylcyclohexyl)-urea. | 185 | Ethyl-5-ethyl-2-cyclopentanone-1-carboxylate | 190 | 5,6-dihydro-5-ethyl-3-(4-isopropylcyclohexyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione. |
| 1-(1-ethylhexyl)-urea | 158 | Ethyl-5-isopropyl-2-cyclopentanone-1-carboxyl-ate. | 204 | 5,6-dihydro-3-(1-ethylhexyl)-5-isopropyl-5-cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione. |
| 1-(1,1-dimethylbutyl)-urea. | 144 | Ethyl-5-n-butyl-2-cyclopentanone-1-carboxyl-ate. | 230 | 5-n-butyl-5,6-dihydro-3-(1,1-dimethylbutyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione. |
| 1-(3a,4,5,6,7,7a-hexahydro-4,7-methano-5-indenyl)-urea. | 196 | Ethyl-5-methyl-2-cyclopentanone-1-carboxylate | 176 | 5,6-dihydro-3-(3a,4,5,6,7,7a-hexahydro-4,7-methano-5-indenyl)-5-methyl-5-cyclopenta[D]-pyrimidine-2,4(1H,3H)-dione. |
| 1-(3a,4,5,6,7,7a-hexahydro-5-indenyl)-urea. | 196 | do | 187 | 5,6-dihydro-3(3a,4,5,6,7,7a-hexanhydro-5-indenyl)-5-methyl-5-cyclopenta[D]pyrimidine-2-4(1H,3H)-dione. |

EXAMPLE 3.—PREPARATION OF 3-n-BUTYL-5,6-DIHYDRO - 5 - VINYL-5-VINYL-5-CYCLOPENTA[D]PYRIMIDINE-2,4(1H,3H)-DIONE

To a warm stirred slurry of 104 parts by weight of n-butylurea and 1000 parts by weight of benzene is added 10 parts by weight of p-toluenesulfonic acid and 180 parts by weight of ethyl 2-cyclopentanone-5-vinyl-1-carboxylate. The slurry is refluxed for 16 hours during which the water given off by the reaction is trapped out. The resulting solution is decanted and concentrated to an oil.

A solution of 90 parts by weight of sodium methoxide in 1400 parts by weight of ethyl alcohol is added to this oil and the mixture is refluxed for 20 minutes. The solution is then concentrated to dryness and the residue dissolved in 2000 parts by weight of water.

The aqueous basic solution is extracted free of neutral impurities with 1500 parts by weight of ether. It is then cooled and acidified to a pH of 5 with hydrochloric acid. Solid, white 3-n-butyl-5,6-dihydro-5-vinyl-5-cpclopenta-[D]pyrimidine-2,4(1H,3H)-dione precipitates, which is filtered and dried.

EXAMPLE 4.—PREPARATION OF 3-n-BUTYL-5,6-DIHYDRO - 5 - (1,2-DIBROMOETHYL) - 5 - CYCLOPENTA[D]PYRIMIDINE-2,4(1H,3H)-DIONE

A mixture of 46.8 parts of 3-n-butyl-5,6-dihydro-5-vinyl - 5 - cyclopenta[D]pyrimidine-2,4(1H,3H)-dione is stirred at room temperature in 600 parts of methylene chloride. To this is gradually added 32 parts of bromine. At the end of the addition the reaction mixture is stripped to dryness giving essentially pure 3-n-butyl-5,6-dihydro-5-(1,2 - dibromoethyl) - 5 - cyclopenta[D]pyrimidine-2,4(1H,3H)-dione.

Compounds similarly prepared are:

3-cyclohexyl-5,6-dihydro-5-(1,2-dibromoethyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-5,6-dihydro-5-(1,2-dichloroethyl)-5-cyclopenta[D]pyrimidine-2,4-(1H,3H)-dione
3-m-chlorophenyl-5,6-dihydro-5-(1,2-dichloroethyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

Additional compounds which may be made and are within the scope described for this invention are:

3-hexahydroazepinyl-7-methyl-5,6-dihydro-5-cyclopenta [D]pyrimidine-2,4(1H,3H)-dione
8-ethyl-3-piperidino-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-6,7-dimethoxy-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-6,7-dichloro-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione
3-n-butyl-5,6,7-trichloro-5,6,7,8-tetrahydro-5-cyclohepta [D]pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-5,7-dibromo-5,6-dihydro-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
5-bromo-3-cyclohexyl-5,6-dihydro-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
7-bromo-3-cyclohexyl-5,6-dihydro-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-6-ethyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-6-propyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-6-sec-butyl-5,6,7,8-tetrahydro-5-cyclohexa [D]pyrimidine-2,4(1H,3H)-dione
3-sec-butyl-6-sec-butyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-6,8-diisopropyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-8-methoxy-5,6,7,8-tetrahydro-5-cyclohexa [D]pyrimidine-2,4(1H,3H)-dione
3-sec-butyl-6-methoxy-7-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione
3-cyclopropyl-6,7,8-trimethyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione
6-tert-butyl-8-methyl-3(2-cyclohexylmethyl)-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-9-isopropyl-6-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione
8-methoxy-6-methyl-3-phenyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione
6-isobutyl-3-cyclohexyl-5,6,7,8-tetrahydro-5-cyclohexa [D]pyrimidine-2,4(1H,3H)-dione
3-tert-butyl-6,7-dimethyl-5,6,7,8-tetrahydro-5-cyclohexa [D]pyrimidine-2,4(1H,3H)-dione
3-sec-butyl-5,6-dihydro-5,7-dimethyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-5,6-dihydro-5,7-dimethyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-5,6-dihydro-5,7-trimethyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione

3-m-chlorophenyl-5,6-dihydro-5,7-trimethyl-5-cyclopenta [D]pyrimidine-2,4(1H,3H)-dione
3-n-butyl-5,6-dihydro-6-methylthio-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
3-iso-butyl-5,6-dihydro-6-ethylthio-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
5,6-dihydro-6-methylthio-3(2-ethylpropyl)-5-cyclopenta [D]pyrimidine-2,4(1H,3H)-dione
3-cyclopentyl-5,6-dihydro-6-n-propylthio-5-cyclopenta [D]pyrimidine-2,4(1H,3H)-dione
3-cyclohexyl-5,6-dihydro-6-n-butylthio-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
3-cycloheptyl-5,6-dihydro-6-methylthio-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione
5,6-dihydro-6-methyl-3-phenyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione
5,6-dihydro-6-methyl-3(3-methylphenyl)-5-cyclopenta [D]pyrimidine-2,4(1H,3H)-dione

EXAMPLE 5.—PREPARATION OF 3-CYCLOHEXYL-5,6-DIHYDRO - 7 - METHYL-5-CYCLOPENTA[D]PYRIMIDINE-2,4(1H,3H)-DIONE

Into a solution of 12 parts by weight of sodium hydroxide, 50 parts of ethanol and 100 parts of water is added 62.5 parts by weight of 3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta[D]pyrimidine - 2,4(1H,3H) - dione. This slurry is heated and stirred until the solids are completely dissolved. The sodium salt of the pyrimidinedione separates on cooling. More of the solid can be forced out of solution by dilution with acetonitrile. The salt is filtered off and dried.

The following salts are prepared according to this method by substituting equivalent quantities of an appropriate substituted pyrimidinedione for the 3-cyclohexyl - 5,6 - dihydro - 7 - methyl - 5 - cyclopenta[D]pyrimidine-2,4(1H,3H)-dione and the proper hydroxides for sodium hydroxide:

5,6-dihydro-3-isopropyl-7-methyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione potassium salt
3-cyclohexyl-5,6-dihydro-8-methyl-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione sodium salt
3-tert-butyl-5,6-dihydro-8-methyl-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione sodium salt
3-cyclohexyl-5,6-dihydro-9-methyl-5-cyclohepta[D] pyrimidine-2,4(1H,3H)-dione potassium salt

EXAMPLE 6.—PREPARATION OF 3-CYCLOHEXYL-5,6-DIHYDRO - 7 - METHYL-5-CYCLOPENTA[D]PYRIMIDINE-2,4(1H,3H)-DIONE, TETRABUTYLAMMONIUM SALT

A mixture containing 250 parts of 3-cyclohexyl-5,6-dihydro - 7 - methyl - 5 - cyclopenta[D]pyrimidine - 2,4(1H,3H-dione) and 865.8 parts of a one-molar solution of tetrabutylammonium hydroxide in methanol is stirred and heated slightly until solution is complete. The solvent is removed by distillation under reduced pressure. The resulting white solid is essentially pure 3-cyclohexyl-5,6-dihydro - 7 - methyl - 5 - cyclopenta[D]pyrimidine - 2,4(1H,3H)-dione, tetrabutylammonium salt.

Other quaternary ammonium salts can be similarly prepared by reacting equivalent amounts of an appropriately substituted pyrimidinedione and a suitable quaternary ammonium hydroxide. The following compounds can be prepared in this fashion:

3-cyclopentenyl-5,6-dihydro-8-methyl-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione, tertamethylammonium salt
3-bornyl-5,6-dihydro-8-ethyl-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione, trimethylbenzylammonium salt
3-isopropyl-5,6-dihydro-9-methyl-3-cyclohepta[D] pyrimidine-2,4(1H,3H)-dione, trimethyldodecylammonium salt

EXAMPLE 7.—PREPARATION OF 3-SEC-BUTYL-5,6-DIHYDRO - 8 - METHYL-5-CYCLOHEXA[D]PYRIMIDINE - 2,4(1H,3H)-DIONE, PIPERIDINE ADDITION COMPOUND

A mixture containing one part of 3-sec-butyl-5,6-dihydro - 8 - methyl - 5 - cyclohexa[D]pyrimidine - 2,4(1H,3H)-dione and 4 parts of piperidine is stirred and gently heated until a clear solution is obtained.

By similarly employing equivalent amounts of appropriate pyrimidinedione and nitrogenous bases in place of the 3 - sec-butyl-5,6-dihydro-8-methyl-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione and piperidine, the following pyrimidinedione-nitrogenous-base-addition products can be prepared:

3-sec-butyl-7-ethyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione, triethylenediamine addition compound
3-sec-butyl-6-ethoxy-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione, ethylenediamine
3-sec-butyl-6-ethyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione, octylamine addition compound
3-sec-butyl-6-isopropyl-5,6,7,8-tetrahydro-5-cyclohexa [D]pyrimidine-2,4(1H,3H)-dione, dodecylamine addition compound
6-ethyl-3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione, piperidine addition compound

EXAMPLE 8.—PREPARATION OF 7-METHYL-3-(2-METHYLCYCLOHEXYL) - 5,6,7,8 - TETRAHYDRO - 5 - CYCLOHEXA[D]PYRIMIDINE - 2,4(1H,3H) - DIONE, PIPERIDINE ADDITION COMPOUND

A mixture containing one part 7-methyl-3-(2-methylcyclohexyl) - 5,6,7,8 - tetrahydro - 5 - cyclohexa[D]pyrimidine-2,4(1H,3H)-dione, and 50 parts of piperidine is stirred and gently heated until a clear solution is obtained.

The following can be prepared in the same fashion:

3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione, butylamine addition compound
3-cyclopentyl-5,6-dihydro-8-methyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione, piperidine addition compound
5,6-dihydro-7-methyl-3-phenyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione, piperidine addition compound

EXAMPLE 9.—PREPARATION OF THE 1:1 COMPLEX OF 3 - CYCLOHEXYL - 5,6 - DIHYRO - 7 - METHYL - 5 - CYCLOPENTA[D]PYRIMIDINE - 2,4(1H,3H)-DIONE WITH PHENOL

A mixture of 248 parts of 3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione and 94 parts of phenol is gradually heated until a clear melt forms. The liquid is stirred to assure complete mixing and is then allowed to cool.

EXAMPLE 10.—PREPARATION OF THE COMPLEX OF 3-CYCLOHEXYL - 5,6 - DIHYDRO - 7 - METHYL - 5 - CYCLOPENTA[D]PYRIMIDINE - 2,4(1H,3H)-DIONE WITH p-CHLOROPHENOL

A mixture of 248 parts of 3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione and 128 parts of p-chlorophenol is gradually heated until a clear melt forms. The liquid is stirred and allowed to cool.

The following complexes can be prepared in a similar fashion by replacing the 3-cyclohexyl-5,6-dihydro-7-methyl - 5 - cyclopenta[D]pyrimidine - 2,4(1H,3H)-dione and p-chlorophenol with equivalent amounts of appropriate pyrimidinediones and phenols:

5,6-dihydro-3-isopropyl-7-methyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione, pentachlorophenol complex

3-n-butyl-5,6-dihydro-7-methyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione, 2,4-dichlorophenol complex

3-cyclohexyl-7-methyl-5,6,7,8-tetrahydro-5-cyclohexa [D]pyrimidine-2,4(1H,3H)-dione, p-nitrophenol complex

EXAMPLE 11.—AQUEOUS DISPERSION

The following ingredients (except the water) are mixed, micropulverized, blended with the water and then ground until the particles of active material are less than 5 microns in diameter:

| | Percent |
|---|---|
| 3 - cyclohexyl - 5,6 - dihydro - 7 - methyl - 5 - cyclopenta[D]pyrimidine-2-,4(1H,3H)-dione | 30.0 |
| Sodium lignin sulfonate | 5.0 |
| Hydrated attapulgite | 1.5 |
| Water | 63.5 |

This stable suspension, when diluted with water and when applied at the rate of 25 pounds of active ingredient per acre in 100 gallons of water, gives excellent control of such annual weeds as crab grass, foxtail, wild rye, wild barley, bachelor buttons, and lamb's quarters growing in a parking lot.

The following substituted pyrimidinediones can be formulated similarly. When applied in a like fashion, in equivalent amounts, they also give good herbicidal results:

3-tert-butyl-9-methyl-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione

3-chloronorbornyl-5,6-dihydro-7-ethyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

3-bornyl-5,6-dihydro-7-ethyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

5,6-dihydro-7-ethyl-3-norbornyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

3-cyclopropylmethyl-9-methyl-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione

9-n-butyl-3-(2-methoxyethyl)-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione

3-allyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione

3-decenyl-9-ethyl-5,6,7,8-tetrahydro-5-cyclohepta[D]pyrimidine-2,4(1H,3H)-dione

3-cyclohexyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione

8-ethyl-3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione

EXAMPLE 12.—WETTABLE POWDER

The following components are blended, then micropulverized until substantially all the solids are under 50 microns in particle size, then reblended to homogeneity. They are then passed through an air attrition mill, such as an air reductionizer, to reduce the particle size to under 10 microns:

| | Percent |
|---|---|
| 3-tert-butyl-6-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione | 80.0 |
| Dioctyl sodium sulfosuccinate concreted with sodium benzoate (Aerosol OTB) | 2.0 |
| Partially desulfonated sodium lignin sulfonate | 1.0 |
| Calcined, non-swelling montmorillonoid type clay (Pikes Peak clay) | 17.0 |

This composition is used for pre-emergence application in sugar cane. It is dispersed in 40 gallons of water and sprayed with a pressure sprayer. At 0.5 to 2.0 pounds of active ingredient per acre, excellent control of lamb's quarters, purslane, mustard, crab grass, foxtail, and water grass is obtained.

The following compound can also be used as indicated above and gives good weed control:

3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione

EXAMPLE 13.—PELLETS

| | Percent |
|---|---|
| 3-cyclohexyl-5,6-dihydro-7-methyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione | 6.67 |
| 3-sec-butyl-5-bromo-6-methyluracil | 3.33 |
| Sodium sulfate, anhydrous | 10.00 |
| California sub-bentonite clay | 80.00 |

These ingredients are blended, micropulverized, pugmilled with 15–20% water, and extruded through ⅛ inch holes. The extrusions are cut into ⅛ inch lengths, dried, and screened.

This formulation keeps lumber yards free of weeds. The pellets are distributed in the spring with a seeder at 20 pounds (actives) per acre. The lumber-yard stacking area is kept free of such pernicious weeds as crab grass, ragweed, plantain, pigweed, Russian thistle, fall panicum, goat weed, cheat, and seedling Johnson grass.

EXAMPLE 14.—GRANULES

| | Percent |
|---|---|
| 3-isopropyl-7-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D]pyrimidine-2,4(1H,3H)-dione | 5 |
| 8–15 mesh granular, expanded vermiculite | 95 |

This granular composition is prepared by dissolving the active material in water and spraying it upon the granules while they are being tumbled. The granules are then dried.

These granules are applied at 25 pounds of active ingredient per acre to mixed annual and perennial vegetation, including quackgrass, plantain, crab grass, barnyard grass, volunteer wheat, bachelor buttons and chickweed, growing under cyclone fences, around highway markers, along fence rows, and around oil tank installations. Excellent weed control is obtained.

The following compounds can be formulated in a like manner, and when used in herbicidally equivalent amounts, will give good weed control:

3-cyclooctenyl-5,6-dihydro-6-isopropyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

3-carvacryl-5,6-dihydro-6-ethyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

6-n-butyl-5,6-dihydro-3-isopropyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

3-n-amyl-5,6-dihydro-6-methyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

5,6-dihydro-5-ethyl-3-(4-isopropylcyclohexyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

5,6-dihydro-3-(1-ethylhexyl)-5-isopropyl-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

5-n-butyl-5,6-dihydro-3-(1,1-dimethylbutyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

EXAMPLE 15.—EMULSIFIABLE OIL SUSPENSION

The following components are mixed together and milled in a roller mill, pebble mill, or sand mill until substantially all the particles of the active component are under 10 microns in size. The resulting suspension may be emulsified in water or diluted further with weed oils for spray application.

| | Percent |
|---|---|
| 5,6 - dihydro - 7 - methyl-3-phenyl-5-cyclopenta[D] pyrimidine-2,4(1H,3H)-dione | 25 |
| Blend of polyalcohol carboxylic esters and oil soluble petroleum sulfonates | 6 |
| Isophorone | 69 |

This formulation is diluted with 80 gallon of Lion Herbicidal Oil—No. 6 and applied at 20 to 30 pounds of active ingredient per acre for the control of morning glory, yarrow, ragweed, wild carrot, water grass, barnyard grass, crab grass, foxtail and pigweed growing along fence rows. Excellent control is obtained.

Similarly formulated and used in equivalent amounts, with excellent results are:

3-n-butyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa[D] pyrimidine-2,4(1H,3H)-dione
3-isopropyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa-[D]-pyrimidine-2,4(1H,3H)-dione

EXAMPLE 16.—WATER-SOLUBLE POWDER

| | Percent |
|---|---|
| 3-cyclohexyl-5,6-dihydro-7-methyl - 5 - cyclopenta[D] pyrimidine-2,4(1H,3H)-dione | 25 |
| Sodium silicate | 74 |
| Sodium dioctyl sulfosuccinate | 1 |

These components are blended and micropulverized. Ten pounds of this formulation are dissolved in 100 gallons of water containing 0.4% of trimethylnonyl ether of polyethylene glycol. Three hundred gallons of this solution per acre are applied to a germinating stand of crab grass, black grass (*Alopec Urus* sp.), annual bluegrass, Kentucky bluegrass, foxtail, barnyard grass, chickweed and wild mustard. Good control of these weeds is obtained.

The following compounds when similarly formulated give good weed control.

3-(1-ethylpropyl)-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
3-n-butyl-5,6-dihydro-7-ethyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
5,6-dihydro-3-n-hexyl-7-n-propyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
7-n-butyl-5,6-dihydro-3-isooctyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
7-methyl-5,6-dihydro-3-(α-naphthyl)-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
3-tert-butyl-8-methyl-5,6,7,8-tetrahydro-5-cyclohexa-[D]pyrimidine-2,4(1H,3H)-dione
3-cyclooctyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
3-cyclopentenyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
3-fenchyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
7-sec-butyl-3-methyl-5,6,7,8-tetrahydro-5-cyclohexa-[D]pyrimidine-2,4(1H,3H)-dione
5,6-dihydro-7-methyl-3-(2-methylcyclohexyl)-5-cyclopenta[D]pyrimidine-2,4(1H,3H)-dione

EXAMPLE 17.—WETTABLE POWDER

| | Percent |
|---|---|
| 3-cyclohexyl-8-methyl-5,6,7,8-tetrahydro - 5 - cyclo-hexa[D]pyrimidine-2,4-(1H,3H)-dione | 40.0 |
| 3-(3,4-dichlorophenyl)1,1-dimethylurea | 20.0 |
| Alkyl naphthalene sulfonate, Na salt | 1.5 |
| Low viscosity methyl cellulose | 0.5 |
| Attapugite clay | 38.0 |

A wettable powder is prepared by blending these components and then micropulverizing the mixture until the particles are below 50 microns in size.

This formulation controls weeds along railroad rights-of-way. Applied in the spring in about 100 gallons of water and at the rate of 25 pounds of active ingredients per acre, the formulation gives good control of annual ryegrass, bromegrass, ragweed, henbit, goldenrod, field daisy, buckhorn plantain, water grass, and volunteer rye.

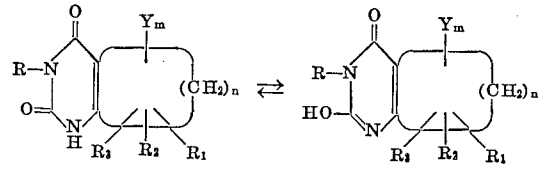
The following compounds when similarly formulated give good weed control:

3-sec-butyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
5,6-dihydro-3-isopropyl-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione
3-tert-butyl-5,6-dihydro-7-methyl-5-cyclopenta-[D]pyrimidine-2,4(1H,3H)-dione



What is claimed is:

1. A method for the control of undesirable vegetation, said method comprising applying to a locus to be protected from such vegetation a herbicidally effective amount of a compound selected from the compounds of the formula



where:

R is selected from the group consisting of
- alkyl of 1 through 10 carbon atoms,
- substituted alkyl of 1 through 10 carbon atoms, wherein said substituent is selected from the group consisting of bromine, chlorine, hydroxy, alkoxy, alkoxycarbonyl, dialkylamino, and cyano,
- aryl of 6 through 14 carbon atoms,
- substituted aryl of 6 through 14 carbon atoms, wherein said substituent is selected from the group consisting of chlorine, bromine, fluorine, phenyl, alkoxy, alkyl, nitro, trifluoromethyl, 1,2-tetramethylene, and 1,2-trimethylenylene,
- aralkyl of 7 through 15 carbon atoms,
- substituted aralkyl of 7 through 15 carbon atoms, wherein said substituent is selected from the group consisting of chlorine, alkyl, nitro, and alkoxy,
- tetrahydronaphthylalkyl,
- alkenyl of 3 through 10 carbon atoms,
- alkynyl of 3 through 10 carbon atoms,
- substituted cycloalkyl of 3 through 12 carbon atoms, wherein said substituent is selected from the group consisting of bromine, chlorine, methoxy, and alkyl,
- cycloalkenyl of 4 through 12 carbon atoms,
- substituted cycloalkenyl of 4 through 12 carbon atoms, wherein said substituent is selected from the group consisting of bromine, chlorine, methoxy, and alkyl,
- cycloalkyl alkyl of 4 through 13 carbon atoms,
- cycloalkenyl alkyl of 5 through 13 carbon atoms,
- (substituted cycloalkyl) alkyl of 5 through 14 carbon atoms, wherein said substituent is selected from the group consisting of bromine, chlorine, methoxy, and alkyl,
- (substituted cycloalkenyl)alkyl of 5 through 14 carbon atoms, wherein said substituent is selected from the group consisting of bromine, chlorine, methoxy, and alkyl,
- cyano,
- piperidino,
- morpholino,
- tetrahydro-4H-1,4-thiazin-4-yl,
- 4-alkyl-1-piperazinyl, wherein said alkyl portion contains from 1 through 4 carbon atoms,
- substituted 4-alkyl-1-piperazinyl, wherein said alkyl portion contains from 1 through 4 carbon atoms and said substituent is 1 through 4 methyl groups on said ring,
- dihydro-4H-1,3,5-dioxazin-5-yl,
- 1-alkyl-3-pyrazolin-2-yl, wherein said alkyl portion contains from 1 through 4 carbon atoms,
- 2,4,6-trimethyldihydro-4H-1,3,5 - diathiazin -5 -yl,
- 1,2,3,4-tetrahydro-2-isoquinolinyl,
- 9-carbazolyl,
- 10-phenoxazinyl,
- 10-phenothiazinyl;

$R_1$, $R_2$, and $R_3$ are each selected from the group consisting of
- hydrogen,

alkyl of 1 through 4 carbon atoms,
haloalkyl of 1 through 4 carbon atoms,
alkoxy of 1 through 4 carbon atoms,
alkylthio of 1 through 4 carbon atoms, and
alkenyl of 2 through 4 carbon atoms;

Y is halogen;

*n* is a number 3 through 5;

*m* is selected from the group consisting of 0 and 1; with the proviso that when $R_1$, $R_2$, and $R_3$ are hydrogen, *m* is 1.

**2.** The method of claim **1** wherein the active compound is 3-cyclohexyl-5,6-dihydro - 7 - methyl - 5 - cyclopenta - [D]pyrimidine-2,4(IH,3H)-dione.

**3.** The method of claim **1** wherein the active compound is 3 - sec-butyl - 5,6, - dihydro - 7 - methyl - 5 - cyclopen - ta[D]pyrimidine-2,4(1H,3H)-dione.

**4.** The method of claim **1** wherein the active compound is 5,6 - dihydro - 3 - isopropyl - 7 -methyl - 5 -cyclopenta - [D]pyrimidine-2,4(1H,3H)-dione.

**5.** The method of claim **1** wherein the active compound is 3 - tert - butyl - 5,6 - dihydro - 7 - methyl - 5 - cyclo-penta[D]pyrimidine-2,4(1H,3H)-dione.

**6.** The method of claim **1** wherein the active compound is 3 - cyclohexyl - 8 - methyl - 5,6,7,8 - tetrahydro - 5 - cyclohexa[D]pyrimidine-2,4(1H,3H)-dione.

**7.** The method of claim **1** wherein the active compound is 3 - sec - butyl - 8 - methyl - 5,6,7,8 - tetrahydro - 5 - cyclohexa[D]pyrimidine-2,4(1H,3H)-dione.



**8.** The method of claim **1** wherein the active compound is 3 - cyclohexyl - 6 - methyl - 5,6,7,8 - tetrahydro - 5 - cyclohexa[D]pyrimidine-2,4(1H,3H)-dione.

**9.** The method of claim **1** wherein the active compound is 3 - sec - butyl - 3 - cyclohexyl - 6 - methyl - 5,6,7,8 - tetrahydro - 5 - cyclohexa[D]pyrimidine - 2,4(1H,3H) - dione.

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