

PATENT SPECIFICATION

L160.028



NO DRAWINGS

L160.028

Date of Application and filing Complete Specification: 4 April, 1967.

No. 15436/67.

Application made in United States of America (No. 539,673) on 4 April, 1966.

Application made in United States of America (No. 622,469) on 13 March, 1967.

Complete Specification Published: 30 July, 1969.

© Crown Copyright 1969.

Index at acceptance:—C2 C(1E6K4, 1E6K6, 1E6K7, 1E7C2, 1E7E1, 1E7F1, 1E7H1, 1E7H2, 1E7N5, 3A8A4, 3A8B2, 3A8C7, 3A8G1, 3A12A4A, 3A12B5, 3A12C6, 3A14C3, 3A14C8A, 3A19A4, 3A19B2, 3A19B3, 3A19C2, 3A19C3, 3A19D3, JA172, JA173, JA190, JA195, JA200, JA215, JA22Y, JA221, JA225, JA247, JA25Y, JA250, JA252, JA28X, JA281, JA285, JA31Y, JA315, JA351, JA352, JA355, JA36Y, JA364, JA366, JA368, JA679, JA79Y, JA791, 172—195—285, 173—190—281); A5 E(1C4B2, 1C4B3, 1C4B4)

Int. Cl.:—C 07 d 51/16

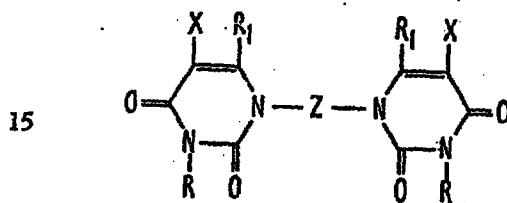
COMPLETE SPECIFICATION

Herbicidal Diuracils

We, E. I. DU PONT DE NEMOURS & COMPANY, a Corporation organised and existing under the laws of the State of Delaware, United States of America, of Wilmington 98, State of Delaware, United States of America, 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:—

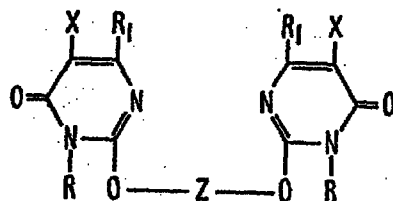
This invention relates to novel herbicidal diuracils.

The novel compounds of the invention have the formula:



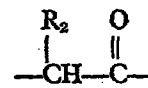
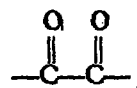
(1)

or the corresponding isomeric formula

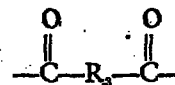


(2)

wherein each of the R radicals, which may be the same or different, represents an alkyl radical containing 3 to 8 carbon atoms, a phenyl radical, a benzyl radical, a cycloalkyl radical containing 5 to 8 carbon atoms or a substituted cycloalkyl radical containing 5 to 8 carbon atoms in which the or each substituent is a methyl or methoxy radical; each of the R₁ radicals, which may be the same or different, represents an alkyl radical containing 1 to 3 carbon atoms; each of the X radicals, which may be the same or different, represents a chlorine, bromine or iodine atom or a methyl radical or, when the R radical on the same ring is a cycloalkyl or substituted cycloalkyl radical, a hydrogen atom; or an X radical and an R₁ radical on the same ring together form a trimethylene or tetramethylene radical; and Z represents a 1,2,3,6-tetrahydrophthaloyl radical or a radical of the formula



wherein R₂ represents a hydrogen atom or an alkyl radical containing 1 to 4 carbon atoms, or

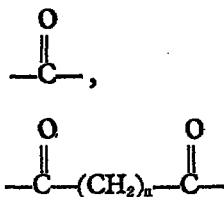


[Price 4s. 6d.]

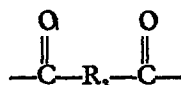
SEE ERRATA SLIP ATTACHED

wherein R_3 represents an alkylene radical containing 1 to 12 carbon atoms, an alkenylene radical containing 1 to 12 carbon atoms, a cyclohexylene radical or a phenylene radical.

- 5 A particular group of these compounds are those wherein Z represents a radical of the formula

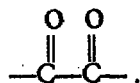


- 10 wherein n is 0 or an integer of 1 to 10, or



wherein R_3 represents a cyclohexylene radical or a phenylene radical.

- 15 The compounds may be symmetrical or unsymmetrical. Preferred compounds, however, are symmetrical and have Z equal to

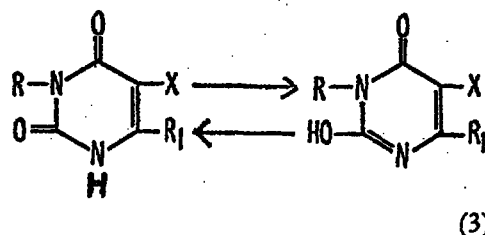


Exemplary of the preferred compounds are the following:

- 20 1,1' - oxalyl bis (3 - *sec.* - butyl - 5 - bromo-6 - methyluracil)
1,1' - oxalyl bis (3 - *iso.* - propyl - 5 - bromo-6 - methyluracil)
25 1,1' - oxalyl bis (3 - *iso.* - propyl - 5 - chloro-6 - methyluracil)
1,1' - oxalyl bis (3 - *tert.* - butyl - 5 - chloro-6 - methyluracil)
30 1,1' - oxalyl bis (3 - cyclohexyl - 6,7 - dihydro-5H - cyclopenta(d) pyrimidine - 2,4 - (1H, 3H) - dione)

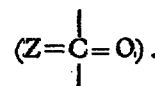
1,1' - oxalyl bis (3 - *tert.* - butyl) - 5 - bromo-6 - methyluracil).

The Z linkage may occur through the 2-positioned oxygens (structure (2)) or through the 1-positioned nitrogens (structure (1)) since uracils may react in their enol or keto form as:



(3)

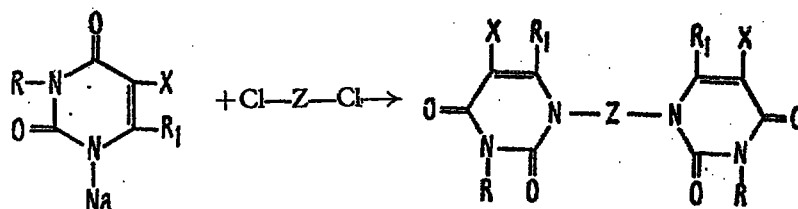
Absorption spectral evidence (infra-red) for the Z-oxygen linkage (structure (2)) is strongly suggested by the products of the invention where Z is derived from phosgene



Both isomers (Z-nitrogen or Z-oxygen linkage) or mixtures of these are considered within the scope of the invention. We believe that it is most likely that structure (1) predominates under ordinary conditions. For the sake of clarity and brevity, however, the compounds are herein named assuming the reaction to have occurred on the 1-nitrogen.

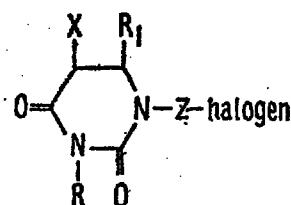
The present compounds are useful as herbicides. They are mainly soil sterilants but also are useful as selective herbicidal agents in certain crops.

The compounds of the invention can be prepared by reacting two equivalents of the sodio derivative of an appropriately substituted uracil with one equivalent of a bifunctional acid chloride, as illustrated by the following equation:



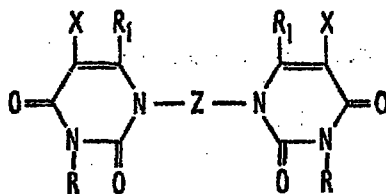
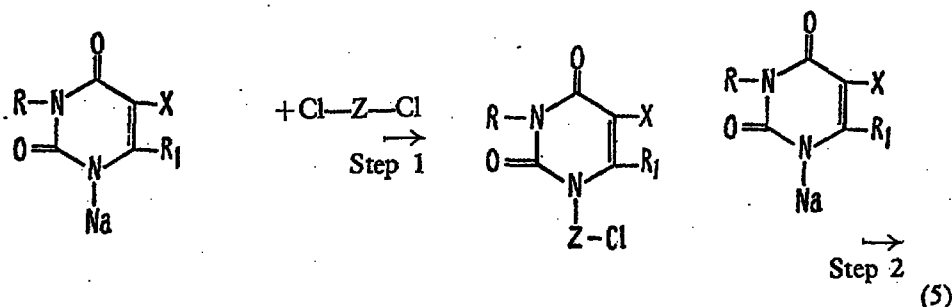
(4)

where R , R_1 , X and Z are as defined above. Alternatively, these compounds can be prepared by the reaction of a compound of formula



with the sodio derivative of a second uracil, in which R , R_1 , and X are not necessarily the same for the two uracil rings coupled

together. This is illustrated by the following equation:



in which R , R_1 , X , and Z are as defined above but where R , R_1 , and X for the sodio derivative of the substituted uracil added to step 2 can be different from the R , R_1 , and X for the sodio derivative of the substituted uracil employed in step 1.

Reaction (4) can be performed in the presence of any of several inert solvents, such as tetrahydrofuran, dioxane, dimethylformamide, chloroform or benzene to facilitate mixing of the reactants. The mixture of 2 molar equivalents of the sodio derivative of a uracil and one equivalent of a bifunctional acid chloride is usually stirred from 2 to 24 hours at temperatures from 20° to 75°C. The product can be isolated in any of several ways, such as (1) filtration, followed by a water wash to remove inorganic material leaving behind essentially pure product, or (2) vacuum stripping of the solvent, followed by treatment with base to remove uncoupled uracil, filtration and water wash as above. Purification, if necessary, can be accomplished by recrystallization from common organic solvents or by chemical extraction.

The first step of reaction (5) can be performed in the presence of any of several inert solvents, such as tetrahydrofuran, dioxane, dimethylformamide, chloroform or benzene to facilitate mixing of the reactants. The mixture of bifunctional acid chloride and one equivalent of a sodio derivative of an appropriately substituted uracil is usually stirred for $\frac{1}{2}$ to 24 hours at temperatures of from 0° to 75°. To the resulting stirred mixture of the uracil derivative can be added the second equivalent of an appropriately substituted uracil.

The second step of reaction (5) can be

effected by stirring for from 2 to 24 hours at temperatures or from 20° to 75°C. Product isolation can be performed as described above for reaction (4).

The present compounds and their preparation are illustrated by the following Examples. All parts in this specification are by weight unless otherwise indicated and temperatures are in °C.

EXAMPLE 1

1,1' - carbonyl bis(5 - bromo - 3 - *sec* - butyl-6 - methyluracil)

A total of 28 parts of 5-bromo-3-*sec*-butyl-6-methyluracil sodio derivative is suspended in 200 parts of tetrahydrofuran. Slowly with stirring 5 parts of phosgene are added. The temperature is allowed to rise autogenetically, then the reaction is stirred at room temperature for 16 hours. The solvent is removed under reduced pressure and the residue treated at 0° with 4 parts of sodium hydroxide dissolved in 100 parts of water. The solid is filtered, washed with water and dried and is essentially pure 1,1' - carbonyl bis(5 - bromo - 3 - *sec* - butyl-6-methyluracil), m.p. 220°.

EXAMPLE 2

1,1' - oxalyl bis (3 - *tert* - butyl - 5 - chloro-6 - methyluracil)

Two hundred parts of anhydrous tetrahydrofuran is placed in a 3-neck flask equipped with a stirrer, condenser and an additional funnel. Four and two tenths parts of a 60% suspension of sodium hydride (in mineral oil) is charged into the stirring flask followed by 21.6 parts of 3-*tert*-butyl-5-chloro-6-methyluracil. This mixture is stirred at room temperature for 16 hours.

Oxalyl chloride, 6.3 parts is added dropwise to the above vigorously stirred mixture. Stirring is continued for an additional four hours, then the reaction mixture is poured into 2000 parts of cold 2% aqueous sodium hydroxide solution. The precipitated solids collected by filtration, washed with water then recrystallized from ethanol gave essentially pure 1,1'-oxalyl bis (3-*tert*-butyl-5-chloro-6-methyluracil), m.p. 172—172.5°C.

EXAMPLE 3
The following products can be prepared by substituting the appropriate amounts of the following uracil sodio derivatives and polyfunctional acid chloride for 5-bromo-3-sec-

butyl 6-methyluracil sodium salt and phosgene cited in Example 1 or the 3-*tert.*-butyl-5-chloro-6-methyluracil sodium salt and oxalyl chloride of Example 2.

Substituted sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substituted uracil)
3- <i>sec</i> -butyl-5-bromo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	malonyldichloride	1,1'-malonylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	succinyldichloride	1,1'-succinylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	adipolydichloride	1,1'-adipoylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	sebacoyldichloride	1,1'-sebacoylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	phthaloyldichloride	1,1'-phthaloylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	isophthaloyldichloride	1,1'-isophthaloylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	terephthaloyldichloride	1,1'-terephthaloylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	perhydrophtaloyldichloride	1,1'-perhydrophtaloylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	perhydroisophthaloyldichloride	1,1'-perhydroisophthaloylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)
"	perhydroterephthaloyldichloride	1,1'-perhydroterephthaloylbis(3- <i>sec</i> -butyl-5-bromo-6-methyluracil)

Substituted uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substituted uracil)
3- <i>sec</i> -butyl-5-bromo-6-methyluracil sodio salt	fumaroyl chloride	1,1'-fumaroylbis(3- <i>sec</i> butyl-5-bromo-6-methyluracil)
3- <i>sec</i> -butyl-5-chloro-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>sec</i> -butyl-5-chloro-6-methyluracil)
3- <i>tert</i> -butyl-5-chloro-6-ethyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>tert</i> -butyl-5-chloro-6-ethyluracil)
3-(<i>iso</i> -propyl)-5-bromo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis-3-(<i>iso</i> -propyl)-5-bromo-6-methyluracil
3- <i>sec</i> -butyl-5-chloro-6-ethyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>sec</i> -butyl-5-chloro-6-ethyluracil)
3- <i>tert</i> -butyl-5-bromo-6- <i>n</i> -propyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>tert</i> -butyl-5-bromo-6- <i>n</i> -propyluracil)
3- <i>iso</i> -propyl-5-iodo-6- <i>iso</i> -propyluracil sodio salt	malonyldichloride	1,1'-malonylbis(3- <i>iso</i> -propyl-5-iodo-6- <i>iso</i> -propyluracil)
3-1-ethylpropyl-5-bromo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis[3-(1-ethylpropyl)-5-bromo-6-methyluracil]
3- <i>n</i> -butyl-5-methyl-6-methyluracil sodio salt	fumaroylchloride	1,1'-fumaroylbis(3- <i>n</i> -butyl-5,6-dimethyluracil)
3-(<i>n</i> -hexyl)-5-chloro-6-ethyluracil sodio salt	phthaloyldichloride	1,1'-phthaloylbis(3- <i>n</i> -hexyl-5-chloro-6-ethyluracil)
3-(1,1,3,3-tetramethylbutyl)-5-iodo-6-ethyluracil sodio salt	fumaroylchloride	1,1'-fumaroylbis(3-[1,1,3,3-tetramethylbutyl]-5-iodo-6-ethyluracil)

1,160,028

Substituted uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substituted uracil)
3- <i>n</i> -propyl-5-methyl-6-ethyluracil sodio salt	1,2,3,6 tetrahydrophthalyl dichloride	1,1'-1,2,3,6-tetrahydrophthalylbis(3- <i>n</i> -propyl-5-methyl-6-ethyluracil)
3- <i>sec</i> -butyl-5-iodo-6-methyluracil sodio salt	succinyl dichloride	1,1'-succinylbis(3- <i>sec</i> -butyl-5-iodo-6-methyluracil)
3-phenyl-5-bromo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3-phenyl-5-bromo-6-methyluracil)
3-benzyl-5-bromo-6-methyluracil sodio salt	itaconyl dichloride	1,1'-itaconylbis(3-benzyl-5-bromo-6-methyluracil)
3-phenyl-5-chloro-6-ethyluracil sodio salt	sebacoyl dichloride	1,1'-sebacoylbis(3-phenyl-5-chloro-6-ethyluracil)
3-cyclopentyl-5-chloro-6-methyluracil sodio salt	glutaconyl dichloride	1,1'-glutaconylbis(3-cyclopentyl-5-chloro-6-methyluracil)
3-cyclohexyl-5-bromo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3-cyclohexyl-5-bromo-6-methyluracil)
3-cycloheptyl-5-bromo-6-methyluracil sodio salt	perhydrotetraphthaloyl dichloride	1,1'-perhydrotetraphthaloylbis(3-cycloheptyl-5-bromo-6-methyluracil)
3-cyclooctyl-5-methyl-6- <i>n</i> -propyluracil sodio salt	succinyl dichloride	1,1'-succinylbis(3-cyclooctyl-5-methyl-6- <i>n</i> -propyluracil)

Substitued uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substitued uracil)
3-(2-methylcyclohexyl)-5-bromo-6-ethyluracil sodio salt	phosgene	1,1'-carbonylbis[3-(2-methylcyclohexyl)-5-bromo-6-ethyluracil]
3-(2-methoxycyclohexyl)-5-iodo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis[3-(2-methoxycyclohexyl)-5-iodo-6-methyluracil]
3-cyclohexyl-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3-cyclohexyl-6-methyluracil)
3-cyclooctyl-6-ethyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3-cyclooctyl-6-ethyluracil)
3-cyclopentyl-6-ethyluracil sodio salt	malonyldichloride	1,1'-malonylbis(3-cyclopentyl-6-ethyluracil)
3-(2-methylcyclohexyl)-6-methyluracil sodio salt	isophthaloyldichloride	1,1'-isophthaloylbis[3-(2-methylcyclohexyl)-6-methylmethyluracil]
3-(3-methoxycyclohexyl)-6-ethyluracil sodio salt	oxalylchloride	1,1'-oxalylbis[3-(3-methoxycyclohexyl)-6-ethyluracil]
3- <i>sec</i> -butyl-5-methyl-6- <i>iso</i> -propyluracil sodio salt	succinyldichloride	1,1'-succinylbis(3- <i>sec</i> -butyl-5-methyl-6- <i>iso</i> -propyluracil)
3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-2,4-tetrahydro-2,4-(1H,3H)-quinazolinedione sodio salt	phosgene	1,1'-carbonylbis[3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]

1,160,028

Substituted uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substituted uracil)
3-cyclohexyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H, 3H]-dione sodio salt	phosgene	1,1'-carbonylbis[3-cyclohexyl-6,7-dihydro 5H-cyclopenta(d)pyrimidine-2,4-(1H, 3H)-dione]
3- <i>tert</i> -butyl-5,6,7,8-tetrahydro-2,4-[1H, 3H]-[1H,3H]-quinazolinedione sodio salt	oxalylchloride	1,1'-oxalylbis[3- <i>tert</i> -butyl-5,6,7,8-tetrahydro-2,4-(1H, 3H)-quinazolinedione]
3- <i>iso</i> -propyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-[1H,3H]-quinazolinedione sodio salt	malonyldichloride	1,1'-malonylbis[3- <i>iso</i> -propyl-5,6,7,8-tetrahydro-2,4-2,4-(1H,3H)-quinazolinedione]
3-phenyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-2,4-[1H,3H]-quinazolinedione sodio salt	phthaloyldichloride	1,1'-phthaloylbis[3-phenyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3- <i>sec</i> -butyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	phosgene	1,1'-carbonylbis[3- <i>sec</i> -butyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3-cyclooctyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	adipoyldichloride	1,1'-adipoylbis[3-cyclooctyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3- <i>sec</i> -butyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	oxalylchloride	1,1'-oxalylbis[3- <i>sec</i> -butyl-6,7-dihydro-5H-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3- <i>iso</i> -propyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	citraconyldichloride	1,1'-citraconylbis[3- <i>iso</i> -propyl-6,7-dihydro-5H-cyclopenta pyrimidine-2,4-(1H,3H)-dione]

Substituted uracil sodil derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substituted uracil)
3-cyclohexyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	oxalylchloride	1,1'-oxalylbis[3-cyclohexyl-6,7-dihydro-5H-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3-(1-ethylpropyl)-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	phosgene	1,1'-carbonylbis[3-(1-ethylpropyl)-6,7-dihydro-5H-cyclopenta(d)-2,4-(1H,3H)-dione]
3- <i>sec</i> -butyl-5-iodo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>sec</i> -butyl-5-iodo-6-methyluracil)
3- <i>tert</i> -butyl-5-chloro-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>tert</i> -butyl-5-chloro-6-methyluracil)
3- <i>tert</i> -butyl-5-bromo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>tert</i> -butyl-5-bromo-6-methyluracil)
3-(1-methylbutyl)-5-bromo-6-methyluracil	oxalylchloride	1,1'-oxalylbis[3-1-methylbutyl)-5-bromo-6-methyluracil]
3- <i>sec</i> -butyl-5-chloro-6-ethyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>sec</i> -butyl-5-chloro-6-ethyluracil)
3- <i>tert</i> -butyl-5-bromo-6- <i>n</i> -propyluracil sodio salt	malonyldichloride	1,1'-malonylbis(3- <i>tert</i> -butyl-5-bromo-6- <i>n</i> -propyluracil)
3- <i>iso</i> -propyl-5-iodo-6- <i>iso</i> -propyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>iso</i> -propyl-5-iodo-6- <i>iso</i> -propyluracil)
3-(1-ethylpropyl)-5-bromo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis[3-(1-ethylpropyl)-5-bromo-6-methyluracil]

Substituted uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbabis(substituted uracil)
3- <i>n</i> -butyl-5-methyl-6-methyluracil sodio salt	glutaryldichloride	1,1'-glutarylbis(3- <i>n</i> -butyl-5,6-dimethyluracil)
3-(<i>n</i> -hexyl)-5-chloro-6-ethyluracil sodio salt	suberoyldichloride	1,1'-suberoylbis(3- <i>n</i> -hexyl-5-chloro-6-ethyluracil)
3-(1,1,3,3-tetramethylbutyl)-5-iodo-6-ethyluracil sodio salt	azelaoyldichloride	1,1'-azelaoylbis(3-[1,1,3,3-tetramethylbutyl]-5-iodo-6-ethyluracil)
3- <i>n</i> -propyl-5-methyl-6-ethyluracil sodio salt	pimelydichloride	1,1'-pimelylbis(3- <i>n</i> -propyl-5-methyl-6-ethyluracil)
3- <i>tert</i> -butyl-5-chloro-6-ethyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>tert</i> -butyl-5-chloro-6-ethyluracil)
3-(1-ethylpropyl)-5-chloro-6-methyluracil	phosgene	1,1'-carbonylbis[3-(1-ethylpropyl)-5-chloro-6-methyluracil]
3- <i>sec</i> -butyl-5-iodo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>sec</i> -butyl-5-iodo-6-methyluracil)
3-phenyl-5-bromo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3-phenyl-5-bromo-6-methyluracil)
3-benzyl-5-bromo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3-benzyl-5-bromo-6-methyluracil)
3-phenyl-5-chloro-6-ethyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3-phenyl-5-chloro-6-ethyluracil)
3-cyclopentyl-5-chloro-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3-cyclopentyl-5-chloro-6-methyluracil)

Substituted uracil sodio derivatives	Polyfunctional	1,1'-oxohydrocarbabis(substituted uracil)
3-cyclohexyl-5-bromo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3-cyclohexyl-5-bromo-6-methyluracil)
3-cycloheptyl-5-bromo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3-cycloheptyl-5-bromo-6-methyluracil)
3-cyclooctyl-5-methyl-6- <i>n</i> -propyluracil sodio salt	phosgene	1,1'-carbonylbis(3-cyclooctyl-5-methyl-6- <i>n</i> -propyluracil)
3-(2-methylcyclohexyl)-5-bromo-6-ethyluracil sodio salt	oxalylchloride	1,1'-oxalylbis[3-(2-methylcyclohexyl)-5-bromo-6-ethyluracil]
3-(2-methoxycyclohexyl)-5-iodo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis[3-(2-methoxycyclohexyl)-5-iodo-6-methyluracil]
3-(3-methylcyclohexyl)-5-chloro-6-methyluracil sodio salt	malonyldichloride	1,1'-malonylbis[3-(3-methylcyclohexyl)-5-chloro-6-methyluracil]
3-cyclohexyl-6-methyluracil sodio salt	pimeloyldichloride	1,1'-pimeloylbis(3-cyclohexyl-6-methyluracil)
3-cyclooctyl-6-ethyluracil sodio salt	succinyldichloride	1,1'-succinylbis(3-cyclooctyl-6-ethyluracil)
3-cyclopentyl-6-ethyluracil sodio salt	phthaloyldichloride	1,1'-phthaloylbis(3-cyclopentyl-6-ethyluracil)
3-(2-methylcyclohexyl)-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis[3-(2-methylcyclohexyl)-6-methyluracil]

Substituted uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbobis(substituted uracil)
3-(3-methoxycyclohexyl)-6-ethyluracil sodio salt	phosgene	1,1'-carbonylbis[3-(3-methoxycyclohexyl)-6-ethyluracil]
3- <i>sec</i> -butyl-5-methyl-6- <i>iso</i> -propyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>sec</i> -butyl-5-methyl-6- <i>iso</i> -propyluracil)
3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	oxalylchloride	1,1'-oxalylbis[3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3-cyclohexyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-(1H,3H)-dione sodio salt	sebacoyldichloride	1,1'-sebacoylbis[3-cyclohexyl-6,7-dihydro-5H-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3- <i>tert</i> -butyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	3,3-dimethylglutaryl-dichloride	1,1'-(3,3-dimethylglutaryl)bis[3- <i>tert</i> -butyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3- <i>iso</i> -propyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	phosgene	1,1'-carbonylbis[3- <i>iso</i> -propyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3-phenyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	phosgene	1,1'-carbonylbis[3-phenyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3- <i>sec</i> -butyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	isophthaloyldichloride	1,1'-isophthaloylbis[3- <i>sec</i> -butyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]
3-cyclooctyl-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	oxalylchloride	1,1'-oxalylbis[3-cyclooctyl-5,6,7,8-tetrahydro-2,4-(1H,3H)-quinazolinedione]

Substituted uracil sodio derivatives	Polyfunctional acid chloride	1,1'-oxohydrocarbobis(substituted uracil)
3- <i>sec</i> -butyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	phosgene	1,1'-carbonylbis[3- <i>sec</i> -butyl-6,7-dihydro-5H-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3- <i>iso</i> -propyl-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H, 3H.]dione sodio salt	perhydroterephthaloyl-dichloride	1,1'-perhydroterephthaloylbis[3- <i>iso</i> propyl-6,7-dihydro-5H-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3-benzyl-6,7-dihydro-5 cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	phosgene	1,1'-carbonylbis[3-benzyl-6,7-dihydro-5-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3-(1-ethylpropyl)-6,7-dihydro-5H-cyclopenta[d]pyrimidine-2,4-[1H,3H]-dione sodio salt	phthaloyldichloride	1,1'-phthaloylbis[3-(1-ethylpropyl)-6-7-dihydro-5H-cyclopenta(d)pyrimidine-2,4-(1H,3H)-dione]
3- <i>sec</i> -butyl-5-iodo-6-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>sec</i> -butyl-5-iodo-6-methyluracil)
3- <i>tert</i> -butyl-5-chloro-5-methyluracil sodio salt	oxalylchloride	1,1'-oxalylbis(3- <i>tert</i> -butyl-5-chloro-6-methyluracil)
3- <i>tert</i> -butyl-5-bromo-6-methyluracil sodio salt	phosgene	1,1'-carbonylbis(3- <i>tert</i> -butyl-5-bromo-6-methyluracil)
3-(1-methylbutyl)-5-bromo-6-methyluracil sodio salt	traumatyldichloride	1,1'-traumatylbis[3-(1-methylbutyl)-5-bromo-6-methyluracil]

EXAMPLE 4

5 - Bromo - 3 - *sec* - butyl - 1 - (5 - bromo - 3 - *tert* - butyl - 1,2,3,4, - tetrahydro - 6 - methyl - 2,4 - dioxo - 1 pyrimidinyl carbonyl) - 6 - methyluracil

A total of 14 parts of 5-bromo-3-*sec*-butyl-6-methyluracil sodium salt is suspended in 200 parts of tetrahydrofuran and stirred for a short time. Slowly, with stirring, is added 5 parts of phosgene at temperatures between 10° and 0°C, then the mixture is stirred at these temperatures for 24 hours. 14 Parts of 5-bromo-3-*tert*-butyl-6-methyluracil sodium salt is then added slowly, with stirring, and the temperature is allowed to rise to a temperature not greater than 75°C, usually 35°C as a maximum. This mixture is allowed to stir for 24 hours at 25°C. The solvent is removed at

reduced pressure and the residue treated at 0°C with 4 parts of sodium hydroxide dissolved in 100 parts of water. The solid is filtered off and washed with water to yield essentially pure 5-bromo-3-*sec*-butyl-1-(5-bromo - 3 - *tert* - butyl - 1,2,3,4 - tetrahydro - 6 - methyl - 2,4 - dioxo - pyrimidin - 1 - yl carbonyl)-6-methyluracil.

EXAMPLE 5

By an analogous method to that of Example 4, the following products are prepared by substituting the appropriate amount of the specified acid chloride for the *in situ* generated 3 - *sec* - butyl - 5 - bromo - 6 - methyluracil-1-carbonyl chloride and the appropriate amount of the specified 3,5,6-trisubstituted uracil sodio derivative for the 3-*tert*-butyl-5-bromo-6-methyl-uracil sodium salt of that Example.

Acid chloride	3,5,6-trisubstituted uracil, sodio derivative	Product
1-chlorocarbonyl-3- <i>tert</i> -butyl-5-bromo-6-methyluracil	3- <i>tert</i> -butyl-5-chloro-6-methyluracil sodio salt	3- <i>tert</i> -butyl-5-chloro-1-(5-bromo-3- <i>tert</i> -butyl-1,2,3,4-tetrahydro-6-methyl-2,4-dioxo-pyrimidinylcarbonyl)-6-methyluracil
1-chlorooxalyl-3- <i>sec</i> -butyl-5-bromo-6-methyluracil	3- <i>tert</i> -butyl-5-chloro-6-methyluracil sodio salt	3- <i>tert</i> -butyl-5-chloro-1-(3- <i>sec</i> -butyl-5-bromo-1,2,3,4-tetrahydro-6-methyl-2,4-dioxo-1-pyrimidinyl oxalyl)-6-methyluracil
1-chlorocarbonyl-3- <i>sec</i> -butyl-5-bromo-6-methyluracil	3-(2-methylcyclohexyl)-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione sodio salt	3-(2-methylcyclohexyl)-1-(3- <i>sec</i> -butyl-5-bromo-1,2,3,4-tetrahydro-6-methyl-2,4-dioxo-1-pyrimidinyl carbonyl)-5,6,7,8-tetrahydro-2,4-[1H,3H]-quinazolinedione
1-chloromalonyl-3- <i>tert</i> -butyl-5-chloro-6-methyluracil	3-(1-methylbutyl)-5-bromo-6-methyluracil sodio salt	3-(1-methylbutyl)-5-bromo-1-(3- <i>tert</i> -butyl-5-chloro-1,2,3,4-tetrahydro-6-methyl-2,4-dioxo-1-pyrimidinyl malonyl)-6-methyluracil
1-chlorooxalyl-3- <i>tert</i> -butyl-5-bromo-6-methyluracil	3- <i>sec</i> -butyl-5-bromo-6-methyluracil sodio salt	3- <i>sec</i> -butyl-5-bromo-1-(3- <i>tert</i> -butyl-5-bromo-1,2,3,4-tetrahydro-2,4-dioxo-1-pyrimidinyl oxalyl)-6-methyluracil
1-chlorocarbonyl-3- <i>sec</i> -butyl-5-chloro-6-methyluracil	3-cyclohexyl-6-methyluracil sodio salt	3-cyclohexyl-1-(3- <i>sec</i> -butyl-5-chloro-1,2,3,4-tetrahydro-2,4-dioxo-1-pyrimidinyl carbonyl)-6-methyluracil
1-chlorocarbonyl-3-phenyl-5-chloro-6-methyluracil	3-benzyl-5-bromo-6-methyluracil sodio salt	3-benzyl-5-bromo-1-(3-phenyl-5-chloro-1,2,3,4-tetrahydro-2,4-dioxo-1-pyrimidinyl carbonyl)-6-methyluracil

EXAMPLE 6

1,1' - (1 - oxoethylene) bis (5 - bromo - 3 - iso propyl - 6 - methyluracil)

Two hundred parts of dry tetrahydrofuran is placed in a reactor equipped with an agitator and a reflux condenser. A 60% suspension of sodium hydride (in mineral oil), 4.2 parts is charged into the stirring reactor followed by 24.7 parts of 5-bromo-3-iso propyl-6-methyluracil. This mixture is stirred at room temperature for 16 hours.

Chloroacetyl chloride 5.6 parts is added gradually to the above stirring mixture. After the addition is complete, the reaction mixture is stirred an additional 4 hours then poured into 2000 parts of ice-cold 2% aqueous sodium hydroxide solution. The precipitated solid collected by filtration, washed with water until alkali-free is 1,1'-(1-oxoethylene) bis (5-bromo-3-iso propyl-6-methyluracil) which can be purified by recrystallization from ethanol.

Equivalent amounts of the corresponding reagents (appropriate α -halogenated acid chlorides and uracils) can be substituted for the chloroacetyl chloride and 5-bromo-3-iso propyl-6-methyluracil of the procedure above and the following compounds of this invention can be prepared:

1, 1' - (1 - oxoethylene) bis - (5 - bromo - 3 - sec. - butyl - 6 - methyluracil)

1,1' - (1 - oxo - 2 - methylethylene) bis - (5 - bromo - 3 - sec. - butyl - 6 - methyluracil)

1,1' - (1 - oxo - 2 - methylethylene) bis - (5 - chloro - 3 - cyclohexyl - 6 - methyluracil)

1,1' - (1 - oxoethylene) bis - [5 - chloro - 3 - (1 - ethylpropyl) - 6 - methyluracil]

1,1' - (1 - oxo - 2 - methylethylene) bis - (5 - chloro - 3 - tert. - butyl - 6 - methyluracil)

1,1' - (1 - oxoethylene) bis - (5 - bromo - 3 - tert. - butyl - 6 - methyluracil)

1,1' - (1 - oxoethylene - bis - (5 - chloro - 3 - benzyl - 6 - methyluracil)

1,1' - (1 - oxoethylene) bis - (5 - bromo - 3 - phenyl - 6 - methyluracil)

1,1' - (1 - oxo - 2 - butylethylene) bis - (5 - chloro - 3 - isopropyl - 6 - methyluracil)

1,1' - (1 - oxo - 2 - ethylethylene) bis - (5 - bromo - 3 - isopropyl - 6 - methyluracil)

1,1' - (1 - oxo - 2 - methylethylene) bis - (5 - iodo - 3 - isopropyl - 6 - methyluracil)

1,1' - (1 - oxoethylene) bis - (3 - cyclohexyl - 6 - methyluracil)

1,1' - (1 - oxoethylene) bis (6 - methyl - 3 - norbornyluracil)

1,1' - (1 - oxoethylene) bis - (5,6 - dimethyl - 3 - cyclohexyluracil)

1,1' - (1 - oxoethylene) bis - [3 - cyclohexyl - 6,7 - dihydro - 5H - cyclopenta (d) pyrimidine - 2,4 - (1H,3H) - dione]

The compounds of the invention can be admixed with weed control adjuvants or modifiers to provide herbicidal compositions for example in the form of dusts, granules, pellets, water-dispersible powders, high-strength con-

centrates, and emulsions and solutions or dispersions in organic liquids.

Thus, the compounds of this invention can be used with a carrier e.g. a finely divided solid, an organic liquid, a wetting agent, a dispersing agent, or any suitable combination of these. The carrier may therefore be at least in part a solid or at least in part a liquid immiscible with water. Often, the carrier is at least in part water.

Compositions containing the compounds of the invention, especially liquids and wettable powders, may also contain surface-active agents, sometimes called surfactants, in amounts usually of course sufficient to render the composition readily dispersible in water or in oil.

The surface-active agent used can be a wetting, dispersing or an emulsifying agent which will assist dispersion of the compound. The surface-active agent or surfactant can include such anionic, cationic and non-ionic agents as have heretofore been generally employed in plant control compositions of similar type. Suitable surface-active agents are set forth, for example, in "Detergents and Emulsifiers" 1965 Annual by John W. McCutcheon, Inc. Of course, mixtures of surface active agents can be employed.

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

Additional surface-active agents can be added to the formulations to increase the ratio of surfactant:active ingredient up to as high for example as 5:1 by weight. Such compositions have a greater herbicidal effectiveness than can be expected from a consideration of the activity of the components used separately. When used at higher rates it is preferred that the surfactant be present in the range of one-fifth to five parts surfactant for each one part of active agent.

Wettable powders are water-dispersible compositions containing the active material, an inert solid extender, and a surfactant to provide rapid wetting and prevent heavy flocculation when suspended in water.

The classes of extenders suitable include the natural clays, diatomaceous earths, and synthetic mineral fillers derived from silica and silicates. Most preferred fillers are kaolinites, attapulgite clay, montmorillonite clays, synthetic silicas, synthetic magnesium silicate and calcium sulfate dihydrate.

Wettable powder formulations may contain from about 25 to 90 weight per cent active material, from 0.5 to 2.0 weight per cent wetting agent, from 0.25 to 5.0 weight per cent dispersant, and from 9.25 to 74.25 weight per cent inert extender, as these terms are described above.

High-strength compositions generally consist of 90 to 99.5% active ingredient and 0.5 to 10% of a liquid or solid surfactant such as those described by McCutcheon in "Detergents and Emulsifiers" 1965 Annual. Such high-strength compositions can often be used in a manner similar to the wettable powders but they are also suitable for further formulation.

The organic liquid suspension concentrates can be prepared by mixing together and sand-grinding a slurry of oil-insoluble active ingredient in the presence of dispersing agents.

These organic liquid suspension concentrates generally contain from 15 to 40% of active ingredient, from 45 to 70% organic liquid such as kerosene, mineral oil and certain weed oils with the remainder made up of surfactants, corrosion inhibitors, and suspending agents.

Dusts are dense powder compositions which are intended for application in dry form. They are characterized by their free-flowing and rapid settling properties so that they are not readily windborne to areas where their presence is not desired. They contain primarily an active material and a dense, free-flowing, solid extender but may also contain grinding aids and surfactants.

Thus, the dust compositions may comprise about 5 to 20 weight per cent active material, 5 to 50 weight per cent absorptive clay filler, 0 to 1.0 weight per cent wetting agent, and about 30 to 90 weight per cent dense, free-flowing dust diluent, as these terms are used herein.

Emulsifiable oils are usually solutions of active material in non-water miscible solvents together with a surfactant.

For the compounds of this invention, emulsifiable oils can be made by mixing the active ingredient with a solvent and surfactant. Suitable solvents for the compounds of this invention are aromatic hydrocarbons including many weed oils, chlorinated solvents, and non-water miscible ethers, esters, or ketones.

Thus, emulsifiable oil compositions may consist of from about 15 to 50 weight per cent active material, about 40 to 82 weight per cent solvent, and about 1 to 10 weight per cent emulsifier, as these terms are defined and used above.

Granules and pellets are physically stable, particulate compositions containing a compound of this invention which adheres to or is distributed through a basic matrix of a coherent, inert carrier with macroscopic dimensions. In order to aid leaching of the active ingredient from the granule or pellet, a surfactant can be present.

For the compounds of this invention, the inert carrier is preferably of mineral origin, and the surfactant is a compound known to the art as a wetting agent.

Two suitable types of carriers for the granule compositions containing compounds of the invention as an active material are as follows. The first type consists of porous, absorptive, preformed granules, such as preformed and screened granular attapulgite or heat expanded, granular, screened vermiculite. On either of these, a suspension of the active agent can be sprayed and can be absorbed at concentrations usually up to 25 weight per cent of the total weight. The second type, which is also suitable for pellets, consists of initially powdered kaolin clays, hydrated attapulgite, or bentonite clays in the form of sodium, calcium or magnesium bentonites. Water-soluble salts, such as sodium salts, may also be present to aid in the disintegration of granules or pellets in the presence of moisture. These ingredients can be blended with the active components to give mixtures that are granulated or pelleted, followed by drying, to yield formulations with the active components distributed evenly throughout the mass. Such granules and pellets can be made with for example 25 to 30 weight per cent active component, but more frequently a concentration of about 10 weight per cent is desired for optimum distribution. Granular compositions are most useful in a size range of 15—30 mesh.

Thus, the preferred granular or pelleted formulations comprise about 5 to 30 weight per cent active material, about 0 to 5 weight per cent wetting agent, and about 65 to 95 weight per cent inert mineral carrier, as these terms are used herein.

The present herbicides can be used in compositions with other herbicides.

The compounds of this invention exert herbicidal activity on a broad spectrum of plants. They are useful as general purpose weed killers or soil sterilants for control of vegetation on industrial sites and railroad ballast particularly in areas of high rainfall.

These compounds also exhibit selective herbicidal action in crops. By properly selecting a bis-uracil of the invention and a rate and time of application, weeds growing in such crops as blackberries, asparagus, sugar beets, mint, sisal, peaches and sugar cane can be controlled.

The amount of diuracil herbicide to be used in any instance will be governed by such factors as climate, the type of weed to be controlled and soil conditions. Generally speaking, one uses the diuracils at levels of 0.25 to 8 pounds per acre for selective control in crops. Rates of 5 to 40 pounds per acre are satisfactory for general weed control.

The following Examples illustrate the formulation and use of the present compounds. In this specification, gallons are United States gallons.

EXAMPLE 7

	1,1' - carbonylbis(5 - bromo - 3- sec-butyl-6-methyluracil)	25.0%
	attapulgitic clay	69.5%
5	synthetic silica	2.0%
	dioctyl sodium sulfosuccinate	2.0%
	sodium lignin sulfonate	1.5%

The above ingredients are blended, micro-pulverized to a particle size essentially below 50 microns and then re-blended.

The above material is suspended in water at the rate of 20 pounds of active ingredient per 50 gallons of suspension. This material is applied pre-emergence to areas in a tank farm at rates of 50 to 100 gallons per acre. The exact volume applied is predicated on the plant species to be controlled and the duration of control desired. Such weeds as crabgrass (*Digitaria* spp.), barnyard grass (*Echinochloa crusgalli*), lambsquarter (*Chenopodium album*), dog fennel (*Helenium tenuifolium*), Johnson grass seedlings (*Sorghum halepense*), blue-grass (*Poa* spp.), wild garlic (*Allium vineale*), dock (*Rumex* spp.), peppergrass (*Lepidium virginicum*) and poison ivy seedlings (*Rhus radicans*) are controlled for an extended period.

All of the other compounds of the invention can be formulated and applied in like manner.

EXAMPLE 8

	(a) 1,1' - oxalylbis(5 - bromo - 3- sec-butyl-6-methyluracil)	80.0%
	montmorillonite clay	14.5%
	synthetic silica	2.0%
35	sodium alkyl naphthalene sulfonate	3.0%
	methylated cellulose	0.5%

The above ingredients are blended, micro-pulverized to particle size essentially below 50 microns and then reblended.

The formulation is suspended in water to provide a ratio of 14 pounds of active ingredient in 50 gallons of water which is applied on one acre of a grass rhizome barrier strip encircling citrus orchards. The chemical is thoroughly incorporated by disking or rototilling to a depth of 8 inches within 2 hours after application. Control of torpedoo grass (*Panicum repens*), Bermuda grass (*Cynodon dactylon*), Johnson grass (*Sorghum halepense*), and centipede grass (*Eremochola ophiuroides*) is obtained. There is no injury to the citrus plants provided barrier strips are no closer to trees than 50 feet.

The following compounds can be formulated and applied in like manner:

- (b) 1,1' - oxalyl bis (3 - isopropyl - 5 - bromo - 6-methyluracil)
- (c) 1,1' - oxalyl bis (3 - tert - butyl - 5-chloro - 6 - methyluracil)

(d) 1,1' - oxalyl bis [3 - cyclobexyl - 6,7-dihydro - 5 - cyclopenta (d)pyrimidine-2,4 - (1H,3H) - dione]

(e) 1,1' - malonyl bis (3 - sec - butyl - 5-bromo - 6 - methyluracil)

(f) 1,1' - terephthaloyl bis (3 - sec - butyl - 5-bromo - 6 - methyluracil)

(g) 1,1' - perhydroterephthaloyl bis (3 - sec-butyl - 5 - bromo - 6 - methyluracil)

(h) 1,1' - oxalyl bis (3 - tert - butyl - 5-bromo - 6 - methyluracil)

EXAMPLE 9

(a) 3 - tert - butyl - 5 - chloro - 1-(5 - bromo - 3 - tert - butyl-1,2,3,4 - tetrahydro - 6-methyl - 2,4 - dioxo - 1-pyrimidinyl carbonyl) - 6-methyluracil

kaolinite	25%
calcium sulfate dihydrate	69%
nonyl phenyl polyethylene glycol ether	2%
sodium lignin sulfonate	3%
	1%

The above ingredients are blended, micro-pulverized to a particle size essentially below 50 microns and then reblended.

A 25% wettable powder formulation is suspended in water and applied with a tractor mounted sprayer so that 2 to 4 lbs. of active ingredient is applied on one acre of asparagus in the spring before the spears emerge. Good control of lambs quarters (*Chenopodium album*), ragweed (*Ambrosia artemisiifolia*), mustard (*brassica nigra*), chickweed (*stellaria media*), crabgrass (*digitaria* spp.) and goosegrass (*Eleusine indica*) is obtained through the cutting season. No crop injury is experienced.

The following compounds can be formulated and applied in like manner:

(b) 1,1' - (1 oxoethylene) bis (5 - bromo - 3-sec-butyl - 6 - methyluracil)

(c) 1,1' - (1 - oxo - 2 - methylethylene) bis-(5 - chloro - 3 - tert butyl - 6 - methyluracil)

EXAMPLE 10

1,1' - oxalylbis(3 - tert - butyl-5 - chloro - 6 - methyluracil)	5.0%
calcium lignin sulfonate plus wood sugars	9.7%
dioctyl sodium sulfosuccinate	0.3%
15-30 mesh attapulgitic	85.0%

The active material, the calcium lignin sulfonate plus wood sugars and the wetting agent are micropulverized to a particle size substantially less than 100 microns. This product is suspended in water and sprayed on the granules of attapulgitic which are tumbled in a mixer. Following drying, the granules are ready for application.

Twenty pounds of the above granular formu-

lation (equivalent to 4 lb./acre of active ingredient) are spread on an acre of Louisiana sugar cane in the spring just after off baring. Good control of crabgrass (*Digitaria spp.*) barnyard grass (*Echinochloa crusgalli*), mustard (*Brassica spp.*), chickweed (*stellaria media*), lambsquarter (*Chenopodium album*), and Johnson grass (*Sorghum halepense*) seedlings is obtained with no significant injury to the crop.

10 EXAMPLE 11

	1,1' - phthaloylbis[3 - phenyl- 5,6,7,8 - tetrahydro - 2,4- (1H,3H) - quinazolinedione]	10%
15	diatomaceous silica	5%
	micaceous talc	85%

The above dust is prepared by blending and grinding the active material with the minor diluent, then blinding the resulting powder with the major diluent.

The above dust formulation is applied with a dust blower to general weedy vegetation around the swampy edge of a pond so that 20 lbs. of active ingredient are applied on one acre. Good control of general weed species is obtained permitting a freer flow of water, and thus reducing areas of impounded water that provide breeding grounds for mosquitos.

30 EXAMPLE 12

	1,1' - oxalylbis(5 - bromo - 3- tert - butyl - 6 - methyluracil)	10%
	sodium sulfate, anhydrous	10%
	calcium lignin sulfonate	5%
35	kaolinite	75%

The above components are ground to pass a 50 mesh screen, moistened with water and extruded and cut to form pellets approximately $\frac{1}{8} \times \frac{3}{32}$ in size. The pellets are dried and can be used as such or can be further subdivided into granules in the 15-60 mesh range.

The above pellets or granules are applied with an auger spreader on railroad rights of way so that 20 lbs. of active ingredient are applied on one acre of actually treated area. Good control of foxtail (*Setaria spp.*), broom-sedge (*Andropogon virginicus*), Bermuda grass (*Cynodon dactylon*), and maretail (*Erigeron canadensis*) is obtained.

EXAMPLE 13

	1,1' - sebacoylbis(3 - sec - butyl- 5 - bromo - 6 - methyluracil	15%
55	alkylaryl polyethylene glycol ether isophorone	5% 80%

The above emulsifiable oil is prepared by mixing the components with agitation until a homogeneous solution results. The oil may be

emulsified with water or extended with weed oil for application.

The formulation is extended with water or herbicidal oil in the proportion of 8 lbs. of active ingredient per 100 gallons of carrier and sprayed with a low pressure, large droplet forming sprayer on utility rights of ways. The general vegetation is sprayed thoroughly to wet both stems and foliage. Good control of maple (*Acer spp.*), Virginia pine (*pinus virginiana*), sweet gum (*liquidambar syraciflua*), tulip poplar (*Liriodendron tulipifera*), broomsedge (*Andropogon virginicus*), mountain laurel (*Kalmia latifolia*), and poison ivy (*Rhus radicans*) is obtained.

EXAMPLE 14

	1,1' - carbonylbis(5 - bromo - 3 - tert - butyl - 6 - methyluracil)	25%
	sodium lauryl sulfate	35%
	sodium lignin sulfonate	2%
	attapulgit clay	35%
	synthetic silica	3%

The above components are blended micro-pulverized to a particle size below 50 microns and reblended.

A mixture is made with water using the formulation mentioned above to provide a concentrate of 8 lbs. of active ingredient in 50 gal. of carrier and is sprayed on one acre of blackberries. The spray mixture is directed toward the base of the plant on to freshly cultivate soil just prior to the time new leaves emerge in the canes in the spring. A repeat application of 4 lbs. per acre one month later provides full season weed control. Good control of chickweed (*stellaria media*), henbit (*Lamium amplexicaule*), lambsquarter (*Chenopodium album*), field cress (*Rorippa austriaca*), wild mustard (*Brassica spp.*), crabgrass (*Digitaria spp.*) or goosegrass (*Eleusine indica*) is obtained.

EXAMPLE 15

	1,1' - carbonylbis(3 - sec - butyl- 5 - chloro - 6 - ethyluracil)	30%
	soya lecithin	2%
	mineral oil	63%
	sorbitan hexaoleate	5%

The above oil suspension is prepared by first grinding the active compound to pass a 40 mesh screen. All the components are then mixed and sand ground to a particle size substantially below 5 microns. The product is characterized by its extremely small particle size so that when diluted with weed oil or emulsified in water and sprayed, plant foliage is uniformly coated.

The above formulation is suspended in herbicidal oil to form a concentrated homogeneous suspension. The suspension containing 20 lbs. of active ingredient in 30 gallons of carrier is applied by high pressure sprayer

mounted on a helicopter and distributed over one acre of area for fire-break maintenance. Good control of crabgrass (*Digitaria spp.*), foxtail (*setaria spp.*), broomsedge (*Andropogon virginicus*), maple (*Acer spp.*) sprouts and mountain laurel (*Kalmia latifolia*) sprouts is obtained.

EXAMPLE 16

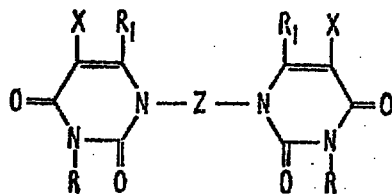
- 10 1,1' - carbonylbis - (5 - bromo - 3 - sec - butyl - 6 - methyluracil) 97%
alkylarly polyethylene glycol ether 3%

The above components are blended and ground to pass a 20 mesh screen. This composition is suitable for further formulation or under some circumstances can be used directly as a granular material.

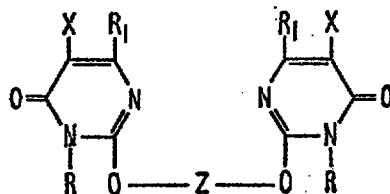
- 15 Without further alteration, this formulation is spread by hand or with a fertilizer spreader capable of distributing granules uniformly.
20 Two pounds of the partially granulated mixture is spread evenly over 1,000 square feet of crushed rock, sand, gravel or ballast before applying an asphalt road surface. Good control of Bermuda grass (*cynodon dactylon*), Johnson grass (*Sorghum halepense*) and torpedo grass (*Panicum repens*) is obtained.

WHAT WE CLAIM IS:—

1. A compound of the formula



30 or the corresponding isomeric formula

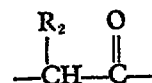
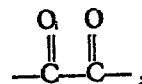


- 35 wherein each of the R radicals, which may be the same or different, represents an alkyl radical containing 3 to 8 carbon atoms, a phenyl radical, a benzyl radical, a cycloalkyl radical containing 5 to 8 carbon atoms or a substituted cycloalkyl radical containing 5 to 8 carbon atoms in which the or each substituent is a methyl or methoxy radical;
40 each of the R₁ radicals, which may be the same or different, represents an alkyl radical containing 1 to 3 carbon atoms;
45 each of the X radicals, which may be the same or different, represents a chlorine,

bromine or iodine atom or a methyl radical or, when the R radical on the same ring is a cycloalkyl or substituted cycloalkyl radical, a hydrogen atom; or an X radical and an R₁ radical on the same ring together form a trimethylene or tetramethylene radical; and Z represents a 1,2,3-tetrahydrophthaloyl radical of the formula

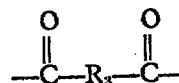


55



wherein R₂ represents a hydrogen atom or an alkyl radical containing 1 to 4 carbon atoms, or

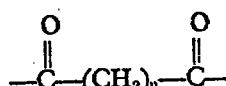
60



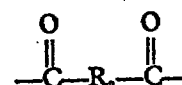
wherein R₃ represents an alkylene radical containing 1 to 12 carbon atoms, an alkenylene radical containing 1 to 12 carbon atoms, a cyclohexylene radical or a phenylene radical.

65

2. A compound according to Claim 1 wherein Z represents a radical of the formula



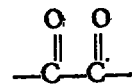
70



wherein n is 0 or an integer of 1 to 10, or wherein R₃ represents a cyclohexylene radical or a phenylene radical.

3. A compound according to Claim 1 wherein the two X radicals are the same, the two R₁ radicals are the same, the two R radicals are same and Z is the radical

75



4. 1,1' - Oxalylbis(5 - bromo - 3 - sec - butyl - 6 - methyluracil).
5. 1,1' - Oxalylbis(5 - bromo - 3 - tert - butyl - 6 - methyluracil).

80



mounted on a helicopter and distributed over one acre of area for fire-break maintenance. Good control of crabgrass (*Digitaria spp.*), foxtail (*setaria spp.*), broomsedge (*Andropogon virginicus*), maple (*Acer spp.*) sprouts and mountain laurel (*Kalmia latifolia*) sprouts is obtained.

EXAMPLE 16

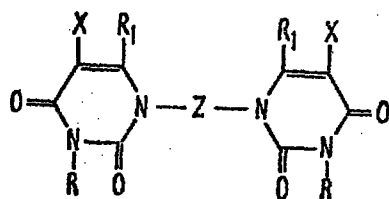
- 10 1,1' - carbonylbis - (5 - bromo - 3 -
sec - butyl - 6 - methyluracil) 97%
alkylarly polyethylene glycol ether 3%

The above components are blended and ground to pass a 20 mesh screen. This composition is suitable for further formulation or under some circumstances can be used directly as a granular material.

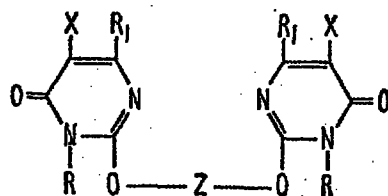
- 15 Without further alteration, this formulation is spread by hand or with a fertilizer spreader capable of distributing granules uniformly.
20 Two pounds of the partially granulated mixture is spread evenly over 1,000 square feet of crushed rock, sand, gravel or ballast before applying an asphalt road surface. Good control of Bermuda grass (*cynodon dactylon*), Johnson grass (*Sorghum halepense*) and torpedo grass (*Panicum repens*) is obtained.

WHAT WE CLAIM IS:—

1. A compound of the formula



- 30 or the corresponding isomeric formula

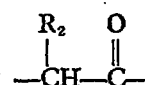
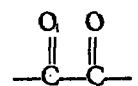


wherein each of the R radicals, which may be the same or different, represents an alkyl radical containing 3 to 8 carbon atoms, a phenyl radical, a benzyl radical, a cycloalkyl radical containing 5 to 8 carbon atoms or a substituted cycloalkyl radical containing 5 to 8 carbon atoms in which the or each substituent is a methyl or methoxy radical;

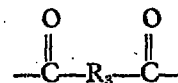
each of the R₁ radicals, which may be the same or different, represents an alkyl radical containing 1 to 3 carbon atoms;

each of the X radicals, which may be the same or different, represents a chlorine,

bromine or iodine atom or a methyl radical or, when the R radical on the same ring is a cycloalkyl or substituted cycloalkyl radical, a hydrogen atom; or an X radical and an R₁ radical on the same ring together form a trimethylene or tetramethylene radical; and
Z represents a 1,2,3-tetrahydrophthaloyl radical of the formula

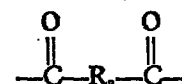
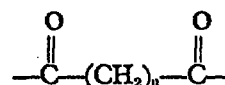


wherein R₂ represents a hydrogen atom or an alkyl radical containing 1 to 4 carbon atoms, or



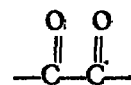
wherein R₃ represents an alkylene radical containing 1 to 12 carbon atoms, an alkenylene radical containing 1 to 12 carbon atoms, a cyclohexylene radical or a phenylene radical.

2. A compound according to Claim 1 wherein Z represents a radical of the formula



wherein n is 0 or an integer of 1 to 10, or wherein R₃ represents a cyclohexylene radical or a phenylene radical.

3. A compound according to Claim 1 wherein the two X radicals are the same, the two R₁ radicals are the same, the two R radicals are same and Z is the radical



4. 1,1' - Oxalylbis(5 - bromo - 3 - sec-butyl - 6 - methyluracil).

5. 1,1' - Oxalylbis(5 - bromo - 3 - tert-butyl - 6 - methyluracil).

6. 1,1' - Oxalylbis[5 - bromo - 3 - (1-ethylpropyl) - 6 - methyluracil].
7. 1,1' - Oxalylbis(3 - *sec* - butyl - 5-chloro - 6 - methyluracil).
- 5 8. 1,1' - Oxalylbis(3 - *tert* - butyl - 5-chloro - 6 - methyluracil).
9. 1,1' - Carbonylbis(5 - bromo - 3 - *sec*-butyl - 6 - methyluracil).
- 10 10. 1,1' - Carbonylbis(5 - bromo - 3 - *tert*-butyl - 6 - methyluracil).
11. 1,1' - Carbonylbis(5 - bromo - 3 - *sec*-butyl - 6 - methyluracil).
12. A compound according to Claim 1 specifically hereinbefore named.
- 15 13. A herbicidal composition comprising a compound as claimed in any one of the preceding claims and a carrier.
14. A composition according to claim 13 wherein the carrier is at least in part a solid.
- 20 15. A composition according to claim 13 or 14 wherein the carrier is at least in part a liquid immiscible with water.
16. A composition according to any one of claims 13 to 15 wherein the carrier is at least in part water.
- 25 17. A composition according to any one of claims 13 to 16 which contains a surface active agent.
18. A composition according to any one of claims 13 to 17 wherein the compound is as claimed in any one of claims 2 to 11.
- 30 19. A composition according to any one of claims 13 to 18 which contains in addition a substance known for use as a herbicide.
- 35 20. A composition according to claim 13 substantially as hereinbefore described.
21. A composition according to claim 13 substantially as described in any one of Examples 7 to 16.
22. A composition according to claim 13 substantially as described in any one of Examples 7, 8a, 9a and 10 to 16.
23. A method of inhibiting the growth of weeds at a locus which comprises applying to the locus a compound as claimed in any one of claims 1 to 12.
24. A method according to claim 23 wherein the compound is used to inhibit the growth of weeds growing amongst crops by applying the said compound at a rate which is sufficient to inhibit the growth of weeds but insufficient substantially to damage the crops.
25. A method according to claim 23 or 24 wherein the compound is applied in the form of a composition as claimed in any one of claims 13 to 22.
26. A method according to claim 25 wherein the composition is as claimed in any one of claims 13 to 17 and 21.
27. A method according to claim 25 wherein the composition is as claimed in claim 18 or 22.
28. A method according to claim 23 or 24 wherein the compound is as claimed in any one of claims 2 to 11.
29. A method according to claim 23 substantially as hereinbefore described.
30. A method according to claim 23 substantially as described in any one of Examples 7, 8a, 9a and 10 to 16.
31. A method according to claim 23 substantially as described in any one of Examples 7, 8a, 9a and 10 to 16.

J. A. KEMP & CO.
Chartered Patent Agents,
14, South Square,
Gray's Inn, London, W.C.1.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1969.
Published by the Patent Office, 25 Southampton Buildings, London, W.C.2, from which copies may be obtained.

ERRATUM

SPECIFICATION NO. 1,160,028

Page 1, For Index at Acceptance C2C only read:- (1E6K4, 1E6K6, 1E6K7, 1E7C2, 1E7E1, 1E7F1, 1E7H1, 1E7H2, 1E7N5, 3A8A4, 3A8B2, 3A8C7, 3A8G1, 3A12A4A, 3A12B5, 3A12C6, 3A14C3, 3A14C8A, 3A19A4, 3A19B2, 3A19B3, 3A19C2, 3A19C3, 3A19D3, 200, 215, 22Y, 221, 225, 247, 25Y, 250, 252, 28X, 31Y, 315, 351, 352, 355, 36Y, 364, 366, 368, 679, 79Y, 791, 172-195-285, 173-190-281, JA)

THE PATENT OFFICE,
29th January 1970

D 121710/45

