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(54) **Preparation of emulsifiable pesticidal solid compositions**

(57) An emulsifiable pesticidal solid composition is prepared by heating and melting a pesticide having a melting point of not higher than 70°C and at least one water soluble polymer selected from polyethylene glycol, polyoxyethylene polyoxypropylene glycol, polyoxyethylene polyoxybutylene glycol and polyoxyethylene polyoxypropylene polyoxybutylene glycol, which is in a solid form at room temperature, optionally in the presence of a surfactant, solvent and/or water soluble carrier, and solidifying the resulting mixture. The resultant emulsifiable solid concentrate is easy to handle and can be readily emulsified.

**GB 2 230 954 A**

EMULSIFIABLE PESTICIDAL SOLID COMPOSITIONS

1           The present invention relates to emulsifiable  
pesticidal solid compositions which can be readily  
emulsified, when diluted with water upon use.

5           Emulsifiable concentrates of pesticides are  
uniform solutions obtained by dissolving pesticides and  
surfactants in organic solvents. The emulsifiable concen-  
trates are general formulations which are easy in handling  
and can stably exhibit their effects.

10           However, since the emulsifiable concentrates  
contain organic solvents in large quantities, problems  
of environmental pollution, malodor, inflammability, etc.,  
due to vaporization of the organic solvent, are involved.  
In addition, these formulations have sometimes such  
problems as phytotoxicity to crops due the solvents.

15           To solve these problems, various investigations  
have been hitherto made to make emulsifiable concentrates  
into a powdery form. For example, there is reported a  
method which comprises absorbing       emulsifiable concen-  
trates onto carriers such as starch, cellulose powders,  
20   urea, cork powders, inorganic silicates, type II anhydrous  
gypsum, etc. In conventional techniques, however, these

1 encounter problems that an absorbability of emulsifiable  
concentrate onto the carriers is too small to prepare  
emulsifiable pesticidal solid composition which has  
good flowability, or emulsifiability is insufficient  
5 when diluted with water.

In order to obtain excellent emulsifiable  
pesticidal solid compositions, the present inventors have  
made various investigations and as a result, they have  
found a process for preparing emulsifiable pesticidal  
10 solid compositions having good flowability which can  
be readily emulsified, and have accomplished the present  
invention.

That is, the present invention provides emulsi-  
fiable pesticidal solid compositions obtained by heating  
15 and melting pesticides together with at least one water  
soluble polymer, which is in a solid form at ambient or  
room temperature (ca. 25°C) and is selected from poly-  
ethylene glycol, polyoxyethylene polyoxypropylene glycol,  
polyoxyethylene polyoxybutylene glycol and polyoxyethylene  
20 polyoxypropylene polyoxybutylene glycol, in the presence  
of or absence of surfactants, solvents and/or water  
soluble carriers to mix them and solidifying the resulting  
mixture.

In general, it is difficult to solidify pesticides  
25 in a liquid form at room temperature. It is also difficult

1 to solidify even pesticides which have melting points  
around room temperature. In addition, these pesticides  
involve a problem in storage stability even if they are  
prepared into a solid form. According to the present  
5 invention, however, excellent emulsifiable pesticidal  
solid compositions can be obtained even if the pesticides  
have a melting point of 70°C or below.

The pesticide as used herein not only refers  
to a single compound but also include a mixture of two  
10 or more compounds. In the case of mixture, the present  
invention is directed to a mixture showing a melting  
point of not higher than 70°C.

As polyethylene glycol used in the present  
invention, there is generally used polyethylene glycol having an average  
15 molecular weight of 1,000 or more; it is particularly  
preferred to use polyethylene glycol having an average  
molecular weight of 4,000 to 20,000 from the viewpoint  
of water solubility, etc.

As polyoxyethylene polyoxypropylene glycol,  
20 there is generally used polyoxyethylene polyoxypropylene glycol  
having an ethylene oxide weight of 80% or more in the  
molecule thereof and having an average molecular weight  
of 1,000 or more in the propylene oxide moiety.

As polyoxyethylene polyoxybutylene glycol and  
25 polyoxyethylene polyoxypropylene polyoxybutylene glycol,  
there are generally used those having an ethylene oxide,  
weight of 80% or more in the molecule thereof and having  
an average molecular weight of 1,000 or more in the

1 butylene oxide moiety or in the propyleneoxide moiety.

What is specifically meant by the solid form at room temperature as used herein refers to the state in which the melting point is approximately 30°C or

5 higher.

Specific examples of the water soluble polymer such as polyethylene glycol include polyethylene glycol having an average molecular weight of 1,000, 4,000, 6,000, 10,000 and 20,000 (hereinafter referred to PEG-1000, 10 PEG-4000, PEG-6000, PEG-10000 and PEG-20000, respectively); NEWPOL<sup>®</sup> PE-68 (manufactured by Sanyo Chemical Industries Co., Ltd.; polyoxyethylene polyoxypropylene glycol having an ethylene oxide weight of 80% in the molecule thereof and having an average molecular weight of 1,750 in the 15 propylene oxide moiety), NEWPOL<sup>®</sup> PE-78 (manufactured by Sanyo Chemical Industries Co., Ltd.; polyoxyethylene polyoxypropylene glycol having an ethylene oxide weight of 80% in the molecule thereof and having an average molecular weight of 2,050 in the propylene oxide moiety), 20 NEWPOL<sup>®</sup> PE-88 (manufactured by Sanyo Chemical Industries Co., Ltd.; polyoxyethylene polyoxypropylene glycol having an ethylene oxide weight of 80% in the molecule thereof and having an average molecular weight of 2,250 in the propylene oxide moiety), NEWPOL<sup>®</sup> PE-108 (manufactured by 25 Sanyo Chemical Industries Co., Ltd.; polyoxyethylene polyoxypropylene glycol having an ethylene oxide weight of 80% in the molecule thereof and having an average molecular weight of 3,250 in the propylene oxide moiety),

1 and the like. These water soluble polymers may be used  
singly or as appropriate admixture thereof. The water  
soluble polymer may be used in an amount sufficient to  
take a solid form in the final preparation form. That  
5 is, the amount is generally in a range of 20 to 99 wt%,  
preferably in a range of 50 to 90 wt%, based on the  
total weight of the composition.

In the present invention, surfactants may also  
be used upon fusion of pesticides together with the  
10 water soluble polymers. Examples of such surfactants which  
can be used include glycerine fatty acid esters, sucrose  
fatty acid esters, sorbitan fatty acid esters, fatty acid  
salts, alkyl sulfates, alkylbenzene sulfonic acid salts,  
alkylammonium salts, quaternary ammonium salts, alkyl  
15 aryl ethers and polyoxyethylenated products thereof,  
ethylene oxide addition products of higher alcohol;  
polyoxyethylene polyoxypropylene glycol in a liquid or  
paste form at room temperature such as NEWPOL<sup>®</sup> PE-64  
(manufactured by Sanyo Chemical Industries Co., Ltd.;  
20 polyoxyethylene polyoxypropylene glycol having an  
ethylene oxide weight of 40% in the molecule thereof  
and having an average molecular weight of 1,750 in the  
propylene oxide moiety), etc. These surfactants may be  
used singly or in a suitable combination. An amount of  
25 the surfactants added is generally in a range of 0.1 to  
20 wt%, preferably in a range of 1 to 10 wt%, based on  
the total weight of the composition. Surfactants  
which become liquid by heating and fusion are preferred

1 but it is not always necessary to use such surfactants.  
It is sufficient that surfactants be dissolved in water  
when the preparation is diluted with water.

Where a melting point of pesticide is in a  
5 range of 0 to 70°C or where surfactants are in a paste  
or solid form at room temperature, a small quantity  
of solvent may also be added to the composition, if  
necessary and desired, for purposes of reducing a viscosity  
upon preparation and preventing crystallization of the  
10 pesticides in the composition when stored at a low temper-  
ature. As the solvent, non-volatile solvents or low  
volatile solvents are used. Examples of such solvents  
used to regulate the viscosity of the composition and  
prevent crystallization of pesticides in the composition  
15 include vegetable oil, mineral oil, liquid paraffin,  
aromatic hydrocarbons, ketones, polyethylene glycol  
which have an average molecular weight of 200 to 600 and  
are liquid at room temperature, polypropylene glycol and  
glycol ethers, etc. An amount of the solvents added is  
20 generally in a range of 10 to 1000 wt%, preferably in  
a range of 30 to 200 wt%, based on the pesticide.

Upon fusion of pesticides and the water soluble  
polymer in the present invention, water soluble carriers  
may also be added to the composition. Examples of the  
25 water soluble carrier which can be used include water  
soluble polymer such as hydroxypropyl cellulose, sodium  
CMC, etc.; urea, lactose, ammonium sulfate, sucrose,  
sodium chloride, Glauber's salt, etc. These water soluble

1 carriers may be appropriately added in such an amount  
that a concentration of the carriers in a spray mix  
prepared upon sprinkling is less than the solubility of  
these carriers in water.

5           In addition to pesticides, surfactants, solvents  
and water soluble carriers, the emulsifiable pesticidal  
solid compositions in accordance with the present invention  
may also appropriately contain stabilizers, synergists,  
coloring agents, etc.

10           However, mineral carriers should not be added,  
in view of the nature that the preparations are emulsi-  
fiable pesticidal solid compositions.

          The emulsifiable pesticidal solid compositions  
in accordance with the present invention are used by  
15 diluting with water to a suitable dilution magnification.

          Specific examples of the pesticides which can  
be used in the present invention are given below but  
the present invention is not deemed to be limited only  
to these examples.

20	Compound No.	Compound
	(1)	$\alpha$ -Cyano-3-phenoxybenzyl 2-(4-chlorophenyl)-3-methylbutyrate
	(2)	(S)- $\alpha$ -Cyano-3-phenoxybenzyl (S)-2-(4-chlorophenyl)-3-methylbutyrate
25	(3)	$\alpha$ -Cyano-3-phenoxybenzyl 2,2,3,3-tetra- methylcyclopropanecarboxylate



- 1 (4) 3-Phenoxybenzyl 3-(2,2-dichlorovinyl)-  
2,2-dimethylcyclopropanecarboxylate
- (5) 3-Phenoxybenzyl chrysanthemate
- (6)  $\alpha$ -Cyano-3-phenoxybenzyl
- 5 3-(2,2-dichlorovinyl)-2,2-dimethyl-  
cyclopropanecarboxylate
- (7)  $\alpha$ -Cyano-3-(4-bromophenoxy)benzyl  
3-(2,2-dichlorovinyl)-2,2-dimethyl-  
cyclopropanecarboxylate
- 10 (8)  $\alpha$ -Cyano-3-(4-fluorophenoxy)benzyl  
3-(2,2-dichlorovinyl)-2,2-dimethyl  
cyclopropanecarboxylate
- (9)  $\alpha$ -Cyano-3-(3-bromophenoxy)benzyl  
3-(2,2-dichlorovinyl)-2,2-dimethyl-  
cyclopropanecarboxylate
- 15 (10)  $\alpha$ -Cyano-3-(4-chlorophenoxy)benzyl  
3-(2,2-dichlorovinyl)-2,2-dimethyl-  
cyclopropanecarboxylate
- (11)  $\alpha$ -Cyano-3-phenoxybenzyl chrysanthemate
- 20 (12)  $\alpha$ -Cyano-3-(4-bromophenoxy)benzyl  
2-(4-chlorophenyl)-3-methylbutyrate
- (13)  $\alpha$ -Cyano-3-(3-bromophenoxy)benzyl  
2-(4-chlorophenyl)-3-methylbutyrate
- (14)  $\alpha$ -Cyano-3-(4-chlorophenoxy)benzyl  
2-(4-chlorophenyl)-3-methylbutyrate
- 25 (15)  $\alpha$ -Cyano-3-(4-fluorophenoxy)benzyl  
2-(4-chlorophenyl)-3-methylbutyrate

- 1 (16)  $\alpha$ -Cyano-3-phenoxybenzyl  
2-(4-bromophenyl)-3-methylbutyrate
- (17)  $\alpha$ -Cyano-3-phenoxybenzyl 2-(4-tert-  
butylphenyl)-3-methylbutyrate
- 5 (18)  $\alpha$ -Cyano-3-phenoxybenzyl 2-(3,4-  
methylenedioxyphenyl)-3-methylbutyrate
- (19)  $\alpha$ -Cyano-(4-fluoro-3-phenoxy)benzyl  
3-(2,2-dichlorovinyl)-2,2-dimethyl-  
cyclopropanecarboxylate
- 10 (20)  $\alpha$ -Cyano-3-phenoxybenzyl 2-chloro-  
4-(trifluoromethyl)anilino-3-methyl-  
butyrate
- (21)  $\alpha$ -Cyano-3-phenoxybenzyl 2-(4-difluoro-  
methoxyphenyl)-3-methylbutyrate
- 15 (22) Cyano-(5-phenoxy-2-pyridyl)methyl  
3-(2,2-dichlorovinyl)-2,2-dimethyl-  
cyclopropanecarboxylate
- (23)  $\alpha$ -Cyano-3-phenoxybenzyl 2,2-dimethyl-  
3-(1,2,2,2-tetrabromoethyl)cyclopropane  
carboxylate
- 20 (24)  $\alpha$ -Cyano-3-phenoxybenzyl 2,2-dimethyl-  
3-(1,2-dichloro-2,2-dibromoethyl)cyclo-  
propanecarboxylate
- (25)  $\alpha$ -Cyano-3-phenoxybenzyl 1-(4-ethoxy-  
phenyl)-2,2-dichlorocyclopropane-  
carboxylate
- 25

- 1 (26)  $\alpha$ -Cyano-3-phenoxybenzyl 2,2-dimethyl-  
3-(2-chloro-3-trifluoromethylvinyl)-  
cyclopropanecarboxylate
- (27) 2-(4-Ethoxyphenyl)-2-methylpropyl  
5 3-phenoxybenzyl ether
- (28) 3-Phenoxybenzyl 2-(4-ethoxyphenyl)-  
3,3,3-trifluoropropyl ether
- (29) O,O-Dimethyl-O-(3-methyl-4-nitro-  
phenyl)phosphorothioate
- 10 (30) O,O-Dimethyl-S-[1,2-di(ethoxycarbonyl)-  
ethyl]phosphorothioate
- (31) O,O-Dimethyl-O-(4-cyanophenyl)-  
phosphorothioate
- (32) O,O-Dimethyl-S-( $\alpha$ -ethoxycarbonyl-  
15 benzyl)phosphorodithioate
- (33) O,O-Diethyl-O-(2-isopropyl-4-methyl-  
6-pyrimidinyl)phosphorothioate
- (34) O,O-Dimethyl-O-[3-methyl-4-(methyl-  
thio)phenyl]phosphorothioate
- 20 (35) O-Ethyl-O-(2,4-dichlorophenyl)-S-  
n-propylphosphorodithioate
- (36) O-(4-Bromo-2,5-dichlorophenyl)-O,O-  
diethylphosphorothioate
- (37) 2-Methoxy-4H-1,3,2-benzodioxo-  
25 phospholine-2-sulfide
- (38) O,O-Diethyl-O-(2,3-dihydro-3-oxo-  
2-phenyl-6-pyridazyl)phosphorothioate

- 1 (39) O,O-Dimethyl-O-(2,4,5-trichloro-phenyl)phosphorothioate
- (40) O,O-Diethyl-O-(3,5,6-trichloro-2-pyridyl)phosphorothioate
- 5 (41) O,O-Dimethyl-O-(3,5,6-trichloro-2-pyridyl)phosphorothioate
- (42) O-(4-Bromo-2,5-dichlorophenyl)-O,O-dimethylphosphorothioate
- (43) O-(4-Cyanophenyl)-O-ethyl-O-phenyl-phosphorothioate
- 10 (44) O,O-Dimethyl-S-(N-methylcarbamoyl-methyl)phosphorodithioate
- (45) 2-sec-Butylphenyl N-methylcarbamate
- (46) 3-Methylphenyl N-methylcarbamate
- 15 (47) 3,4-Dimethylphenyl N-methylcarbamate
- (48) 2-Isopropoxyphenyl N-methylcarbamate
- (49) 5-Ethoxy-3-trichloromethyl-1,2,4-thiadiazole
- (50) O,O-Diisopropyl-S-benzyl phosphorothiolate
- 20 (51) O-Ethyl-S,S-diphenyl dithiophosphate
- (52) Polyoxin
- (53) Blastocidin S
- (54) 3,4-Dichloropropionanilide
- (55) Isopropyl N-(3-chlorophenyl)carbamate
- 25 (56) Ethyl-di-n-propyl thiocarbamate
- (57) 3-methoxycarbonylamino-phenyl  
N-(3-methylphenyl)carbamate

- 1 (58) 2-Chloro-(2,6-diethyl-N-methoxy-  
methyl)acetanilide
- (59)  $\alpha,\alpha,\alpha$ -Trifluoro-2,6-dinitro-N,N-  
dipropyl-p-toluidine
- 5 (60) S-(4-Chlorophenyl)methyl-N,N-  
diethylthiol carbamate
- (61) S-Ethylhexahydryl-1H-azepine-1-  
carbothioate
- (62) N-Butoxymethyl-2-chloro-(2,6-diethyl-  
10 acetanilide
- (63) O-Ethyl-O-(5-methyl-2-nitrophenyl)-  
sec-butylphosphoramidothioate
- (64) N-(Chloroacetyl)-N-(2,6-diethyl-  
phenyl)glycine ethyl ester

15 These pesticides are contained generally in a  
range of 1 to 80 wt%, preferably in a range of 10 to 40  
wt%, based on the total weight of the composition.

The emulsifiable pesticidal solid compositions  
in accordance with the present invention can be prepared,  
20 for example, as follows.

The emulsifiable solid compositions can be  
prepared by heating the water soluble polymer in a solid  
form at room temperature, e.g., polyethylene glycol or  
polyethylene polyoxypropylene glycol, in a container  
25 at a temperature of 50°C or higher, generally at 80 to  
95°C; adding pesticides and if necessary and desired,  
surfactants, solvents and/or water soluble carriers

1 to the water soluble polymer while stirring to uniformly  
mix them; spreading the thus obtained melt mixture onto a  
vat, a glass plate, or the like and cooling to solidify;  
pulverizing and then sieving. In a larger scale of  
5 preparation, the emulsifiable solid compositions can be  
obtained by spraying and solidifying the melt mixture  
described above in a chamber equipped with a cooling  
apparatus.

[Examples]

10 Hereafter the present invention is described  
in more detail by referring to the preparation examples  
and test examples but is not deemed to be limited only  
thereto.

In the following preparation examples, parts  
15 are all by weight, unless otherwise indicated.

Preparation Example 1

Ninety parts of PEG-6000 were added to 10 parts  
of Compound Nos. (1), (2), (3), (4), (5), (6), (31),  
(33), (46), (47), (48), (49) and (66), respectively.  
20 Each mixture was heated to 80°C to fuse and thoroughly  
mix them with each other. The melt mixture was spread  
onto a glass plate and cooled and solidified. Then,  
the solid was ground and sieved to 1000 to 297  $\mu\text{m}$  to give  
an emulsifiable pesticidal solid composition in a granular  
25 form.

1 Preparation Example 2

Ninety parts of NEWPOL<sup>®</sup> PE-68 (supra) were added to 10 parts of Compound Nos. (2), (3), (4), (5), (6) and (31), respectively. Each mixture was heated to 80°C to fuse and thoroughly mix them with each other. The melt mixture was spread onto a glass plate and cooled and solidified. Then, the solid was ground and sieved to 1000 to 297 μm to give an emulsifiable pesticidal solid composition in a granular form.

10 Preparation Example 3

Forty parts of NEWPOL<sup>®</sup> PE-68 (supra) and 50 parts of PEG-6000 were added to 10 parts of Compound Nos. (2), (3) and (31), respectively. Each mixture was heated to 80°C to fuse and thoroughly mix them with each other. The melt mixture was spread onto a glass plate and cooled and solidified. Then, the solid was ground and sieved to 1000 to 297 μm to give an emulsifiable pesticidal solid composition in a granular form.

Preparation Example 4

20 Five parts of SORPOL<sup>®</sup> 355LSA (surfactants, manufactured by Toho Chemical Co., Ltd.) and PEG-6000 were added to 10, 20, 30 and 40 parts of Compound No. (31) to make up 100 parts, respectively. Each mixture was heated to 80°C to fuse and thoroughly mix them with each other. The melt mixture was spread onto a glass plate and cooled and solidified. Then, the solid was

25

1 ground and sieved to 1000 to 297  $\mu$ m to give an emulsifiable  
pesticidal solid composition in a granular form.

Preparation Example 5

5 Twenty parts of HISOL SAS-296 (solvent, manu-  
factured by Nippon Petrochemicals Co., Ltd.), 10 parts of  
SORPOL<sup>®</sup> 3598 (surfactant, manufactured by Toho Chemical  
Co., Ltd.) and 60 parts of PEG-6000 were added to 10 parts  
of Compound Nos. (2), (3), (4), (5), (6) and (29),  
respectively. Each mixture was heated to 80°C to fuse and  
10 thoroughly mix them with each other. The melt mixture  
was spread onto a glass plate and cooled and solidified.  
Then, the solid was ground and sieved to 1000 to 297  $\mu$ m  
to give an emulsifiable pesticidal solid composition in  
a granular form.

15 Preparation Example 6

Sixty parts of PEG-1000, PEG-4000, PEG-20000,  
NEWPOL<sup>®</sup> PE-68 (supra), NEWPOL<sup>®</sup> PE-78 (supra), NEWPOL<sup>®</sup>  
PE-88 (supra) or NEWPOL<sup>®</sup> PE-108 (supra) were added to  
10 parts of Compound No. (3), 20 parts of HISOL SAS-296  
20 (supra) and 10 parts of SORPOL<sup>®</sup> 3598 (supra). Each  
mixture was heated to 80°C to fuse and thoroughly mix  
them with each other. The melt mixture was spread onto a  
glass plate and cooled and solidified. Then, the solid  
was ground and sieved to 1000 to 297  $\mu$ m to give an  
25 emulsifiable pesticidal solid composition in a granular  
form.



1 Preparation Example 7

Ninety parts of PEG-20000 were added to 10 parts of Compound No. (3). The mixture was heated to 80°C to fuse and thoroughly mix them with each other.

5 After the melt mixture was cooled to about 50°C, the mixture was sprayed in a chamber cooled to -5°C and solidified to give an emulsifiable pesticidal solid composition in a granular form.

Preparation Example 8

10 Eighty parts of PEG-6000 were added to 20 parts of Compound No. (1) and the mixture was heated to 80°C to fuse and thoroughly mix them with each other. The melt mixture was spread onto a glass plate and cooled and solidified. Then, the solid was ground and sieved to 1000

15 to 297  $\mu\text{m}$  to give an emulsifiable pesticidal solid composition in a granular form.

Preparation Example 9

Sixty parts of PEG-6000 were added to 10 parts of Compound No. (3), 10 parts of HISOL SAS-296 (supra) and

20 5 parts of SORPOL<sup>®</sup> 3598 (supra) and the mixture was heated to 80°C to fuse and thoroughly mix them with each other. After 15 parts of lactose, urea or Glauber's salt were added to the melt mixture to disperse, the dispersion was spread onto a glass plate and cooled and solidified.

25 Then, the solid was ground and sieved to 1000 to 297  $\mu\text{m}$  to give an emulsifiable pesticidal solid composition in

1 a granular form.

Comparative Example 1

Ninety parts of PEG-6000 powders were added  
to 10 parts of Compound Nos. (31), (33) and (34),  
5 respectively. Each mixture was attempted to thoroughly  
mix in a mortar with a pestle. However, the resulting  
mixture was extremely sticky so that any fluidizable  
product was not obtained.

Comparative Example 2

10 Sixty parts of PEG-20000 powders were added  
to 10 parts of Compound No. (3), 20 parts of HISOL SAS-296  
(supra) and 10 parts of SORPOL<sup>®</sup> 3598 (supra). The mixture  
was attempted to thoroughly mix in a mortar with a  
pestle. However, the resulting mixture was extremely  
15 sticky so that any fluidizable product was not obtained.

Comparative Example 3

After 15 parts of lactose, urea or Glauber's  
salt were added to 10 parts of Compound No. (3), 10  
parts of HISOL SAS-296 (supra), 5 parts of SORPOL<sup>®</sup> 3598  
20 (supra) and 60 parts of PEG-6000 powders, the mixture  
was attempted to thoroughly mix in a mortar with a  
pestle. However, the resulting mixture was extremely  
sticky so that any fluidizable product was not obtained.

1 Test Example 1

Hundred milligrams of each of the emulsifiable  
pesticidal solid compositions in a granular form produced  
in Preparation Examples 1, 2 and 3 were charged in  
5 250 ml glass stoppered cylinders, each of which contained  
100 ml of 3° hard water (53.4 ppm hard water as CaO).  
Inversion of the cylinder was repeated 30 times at a  
rate of once per 2 seconds to emulsify. The granules  
were fully dissolved in all of the compositions. Then,  
10 each cylinder was kept for 15 minutes in a thermostat at  
20°C to examine stability of the emulsion. In any case,  
isolated oil cream was hardly observed.

Test Example 2

One gram of each of the emulsifiable pesticidal  
15 solid compositions in a granular form produced in  
Preparation Examples 4, 5 and 6 was charged in 250 ml  
glass stoppered cylinders, each of which contained 100 ml  
of 3° hard water. Inversion of the cylinder was repeated  
30 times at a rate of once per 2 seconds to emulsify.  
20 The granules were fully dissolved in all of the composi-  
tions. Then, each cylinder was kept for 2 hours in a  
thermostat at 20°C to examine stability of the emulsion.  
In any case, isolated oil cream was hardly observed.

Test Example 3

25 One gram of each of the emulsifiable pesticidal  
solid composition in a granular form produced in

1 Preparation Example 9 was charged in 250 ml glass  
stoppered cylinders, each of which contained 100 ml of  
3° hard water. Inversion of the cylinder was repeated  
30 times at a rate of once per 2 seconds to emulsify.  
5 The granules were fully dissolved in all of the composi-  
tions. Then, each cylinder was kept for 2 hours in a  
thermostat at 20°C to examine stability of the emulsion.  
In any case, isolated oil cream was hardly observed.

Reference Example

10 Acute toxicity test was performed to determine  
LD<sub>50</sub> value by orally administering the emulsifiable  
pesticidal solid composition in accordance with the  
present invention, produced in Preparation Example 8,  
to ICR strain male and female mice of 6 week old.

15 With respect to an emulsifiable concentrate  
having a conventional formulation [The emulsifiable  
concentrate was prepared by mixing 20 parts of Compound  
No. (1), 10 parts of SORPOL<sup>®</sup> 3005X (surfactants, manu-  
factured by Toho Chemical Co., Ltd.) and 70 parts of  
20 xylene], LD<sub>50</sub> value was also determined in a similar  
manner.

The results are shown in the table below.

	LD <sub>50</sub> value (mg/kg)	
	<u>Male Mouse</u>	<u>Female Mouse</u>
Preparation Example 8	1330	944
Conventional emulsifiable concentrate	514	510

1           The emulsifiable pesticidal solid compositions  
in accordance with the present invention are excellent  
preparations showing good flowability and having  
properties enabling to easy handling. In addition, the  
5 emulsifiable solid compositions can readily be emulsified  
when diluted with water.

CLAIMS:

1. A process for preparing an emulsifiable pesticidal solid composition which comprises heating and melting
  - (a) a pesticide having a melting point of not higher than 70°C, and
  - (b) at least one water soluble polymer which is in a solid form at room temperature and is selected from the group consisting of polyethylene glycol, polyoxyethylene polyoxypropylene glycol, polyoxyethylene polyoxybutylene glycol and polyoxyethylene polyoxypropylene polyoxybutylene glycol, in the presence of or absence of
  - (c) a surfactant, a solvent and/or a water soluble carrier, and solidifying the resulting mixture.
2. A process according to claim 1, wherein the said water soluble polymer is present in an amount within a range of from 50 to 90 wt% inclusive based on the total weight of the composition.
3. A process according to claim 1 or claim 2, which process is substantially as herein described and exemplified.
4. An emulsifiable pesticidal solid composition whenever produced by a process according to any preceding claim.