INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

Inventors:


Title: SPONTANEOUSLY WATER DISPERSIBLE CARRIER FOR PESTICIDES

Abstract
This invention relates to natural diatomaceous earth granule compositions which can be broadcast spread on the soil to deliver a pesticide. Granules retain their physical integrity when spread, and have the unique property to spontaneously disperse when irrigation water is applied or rainfall hits the particle. Upon wetting, the particle disperses (blooms) to cover the soil surface. This bloom can cover an area many times the external surface of the particle. The granules have high loadings of natural diatomaceous earth, i.e., from about 35 to about 95 weight percent and contain from about 5 to about 40 weight percent of a surfactant system which exhibits excellent dispersing; rewetting and binding properties. Bioactive compounds can be loaded at up to 60 weight percent of the granule. Bioactive compounds may be formulated products or technical grades and may be homogeneously distributed throughout the granule or spray impregnated onto the granule.
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SPONTANEOUSLY WATER DISPERSIBLE CARRIER FOR PESTICIDES

Background of the Invention

Field of the Invention

This invention relates to highly dispersible compositions; their use as carriers of bioactive materials; and methods for preparing and using said compositions. More specifically, the invention relates to surfactant-diatomaceous earth compositions for agricultural use in the form of dry spreadable granules and methods of preparing same, and even more specifically for their use in the delivery of pesticides to the soil.

Description of Prior Art and Problems

An organic pesticide is a bioactive material which destroys or inhibits the action of plant or animal pests. The general term pesticide includes insecticides, herbicides, rodenticides, and miticides.

Organic pesticides are widely used in soil and turf by both consumers and commercial operators. Pesticide compounds may be used alone; however, usually they are formulated into conventional forms
such as dust, granule, microgranule, wettable powder, flowable powder, emulsion, microcapsule, oil, aerosol, etc., using techniques well known in the art. To improve or stabilize the effects of the pesticide, the pesticide is blended with suitable adjuvants and then used as such or after dilution if necessary. Examples of adjuvants include carriers, diluents, spreaders, emulsifying agents, wetting agents, dispersion agents, or fixing agents.

The present invention is directed towards dry spreadable diatomaceous earth granules, and towards methods of preparing such granules, which can be applied with a dry spreader to a target area and, when exposed to water via, for example, rain or irrigation, will not only readily disintegrate, but actively disperse so as to achieve disperse area to granule diameter ratios larger than that heretofore realized. These granules are primarily used as carriers for bioactive materials such as pesticides. These dry spreadable, water dispersible granules are easy to formulate, ship, store, and apply.

**Summary of the Invention**

This invention provides highly dispersible compositions; their use as carriers of bioactive...
materials; and methods for preparing and using said compositions. Specifically, the invention relates to diatomaceous earth compositions in the form of dry spreadable granules and methods of preparing same, and more specifically to their use in the delivery of pesticides to the soil.

The granules utilize, in addition to the diatomaceous earth, a surfactant composition designed to provide binding, rewetting and dispersion properties to the granules.

**Detailed Description of the Invention**

The instant invention relates to the discovery of a dry spreadable, water dispersible diatomaceous earth granule which, when exposed to water, has a high dispersion area to granule diameter ratio. As a result, when the granules of this invention are dry spread and water treated, the area affected by the granule formulation is much enhanced over that realizable with the dry spreadable granules of the prior art.

By diatomaceous earth is meant a silica material characterized by a large surface area per unit volume. This is the result of an enormous number of fine pores within the structure. Diatomaceous
earth is a naturally occurring material and consists mainly of accumulated shells or frustules of intricately structured amorphous hydrous silica secreted by diatoms. Diatoms are single-celled golden brown algae of the Bacillariophyceae class. The enhanced absorption/adsorption properties of the diatomaceous earth result from the very large internal surface area, hence its valuable capacity to disperse readily upon contact with water.

The effective diatomaceous earth of this invention has a surface area in the range of from greater than about 5 square meters per gram to less than about 90 square meters per gram, preferably from greater than about 10 to less than about 60 square meters per gram and a pore volume in the range of from greater than about 2 cubic centimeters (cc) per gram to less than about 5 cc per gram, preferably from greater than about 3 cc per gram to less than about 4 cc per gram. Natural diatomaceous earth is suitable for this invention.

Diatomaceous earth is present in the granule composition at from about 35 to about 95 weight percent, preferably from about 50 to about 95 weight percent of the diatomaceous earth, surfactant, and pesticide granule composition.
The surfactant composition of the present invention is present at from about 5 to about 40 weight percent, preferably from about 4.995 to about 20 weight percent of the diatomaceous earth/surfactant/pesticide granule composition. The surfactant composition provides three essential properties to the dry spreadable, water dispersible granules.

First, the surfactant composition must act as a binder for the particles in the granule so that after the processing and agglomerating water used in the initial preparation of the granule has been driven off, residual surfactant will act as a binder to form a hard, easily handleable, i.e., dry spreadable granule.

Second, the surfactant composition must possess a good rewetting ability such that when the granule is applied to the site to be treated and exposed to water, the surfactant composition will enable the water to easily penetrate the interstices of the individual diatomaceous earth particles quickly, i.e., "wet" the internal surface areas of the particles. It is theorized that this rapid rewetting of the internal surfaces results in a displacement of the air initially entrapped within the particles which
then provides an additional dispersion mechanism for the granules of this invention.

Third, the surfactant composition must be able to aid in the breaking up and rapid dispersion of the particles initially in the granule.

In summary, the surfactant composition should possess the characteristic properties of being a good binder, rewetting agent, and dispersing aid. These properties could, of course, be found to a limited extent in a single component surfactant composition, but preferably at least a two-component composition is utilized. As will be noted below, polymeric or bulky dispersants often can function as acceptable binders. A three-component surfactant composition in which each surfactant is optimized for one of the aforementioned, desired properties of the composition is most preferred.

The dispersants of the instant invention are usually present at from about 3 to about 15 weight percent; preferably from about 6 to about 10 weight percent based on the weight of the diatomaceous earth, surfactant, and pesticide granule composition. Certain nonionics can function as dispersants, typically high molecular weight materials, i.e., the polymeric.

However, the preferred dispersant surfactants of this
invention are the solid (at room temperature, i.e., 24°C), high molecular weight anionic surfactants.

The rewetting agents of the instant invention are usually present at from about 2 to about 5 weight percent, preferably from about 2 to about 3 weight percent based on the weight of the diatomaceous earth, surfactant, and pesticide granule composition. Certain nonionics can function as rewetting agents, typically the low molecular weight materials, i.e., non-polymeric. However, the preferred rewetting surfactants of this invention are the solid (at room temperature), lower molecular weight anionic surfactants. These surfactants are selected for their ability to move more quickly in aqueous solutions than those anionics used as dispersants.

The binders for use in the granules of this invention, are usually present from about 0 to about 10 weight percent, preferably from about 2 to about 6 weight percent based on the weight of the diatomaceous earth, surfactant, and pesticide granule composition. These binders can be traditional binders well known in the art such as the starches, the sugars, etc., but preferably the granules of this invention use residuals or the secondary characteristics of the
dispersing agents and/or rewetting agents described herein as the binding agents.

The anionics useful in the surfactant compositions of this invention include the alkyl and alkyl ether sulfates. These materials have the respective formulae $ROSO_3M$ and $RO(C_2H_4O)_xSO_3M$ wherein R is an alkyl, alkenyl or alkylauryl group of about 8 to about 22 carbon atoms, $x$ is 1 to 10, preferably 1 to 4, and M is a water-soluble cation such as ammonium, sodium, potassium, magnesium, triethanolamine (TEA), etc. The alkyl ether sulfates useful in the present invention are condensation products of ethylene oxide and monohydric alcohols having about 8 to about 22 carbon atoms. Specific examples of the above sulfates include ammonium lauryl sulfate, magnesium lauryl sulfate, sodium 2-ethyl-hexyl sulfate, sodium acetyl sulfate, sodium oleyl sulfate, sodium tridecyl sulfate, triethanolamine lauryl sulfate, ammonium linear alcohol, ether sulfate ammonium nonylphenol ether sulfate, and ammonium monoxyxynol-4-sulfate.

Another suitable class of anionic surfactants are the water-soluble salts of the general formula:

$$R_1SO_3M$$

wherein $R_1$ is selected from the group consisting of:
i) a straight or branched chain, saturated aliphatic hydrocarbon radical having from 8 to 24, preferably 12 to 18 carbon atoms;

ii) a mono-, di-, or tri- C₁₋₆ alkyl substituted aryl wherein the aryl is preferably a phenyl or naphthyl group;

iii) alpha-olefins having 12 to 24 carbon atoms, preferably 14 to 16 straight chain carbon atoms, most preferably 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosene, and 1-tetracosene; and

iv) naphthalene-formaldehyde condensation products.

Specific examples include Supragil MNS/90, a trademark of Rhône-Poulenc Inc. for an alkynaphthalenesulfonate-formaldehyde condensate and Supragil WP, a trademark of Rhône-Poulenc Inc. for an alkynaphthalenesulfonate.

Additional examples of anionic synthetic surfactants which come within the terms of the present invention are: i) the isethionates, i.e., the reaction products of fatty acids esterified with isethionic acid and neutralized with sodium hydroxide
where, for example, the fatty acids are derived from coconut oil; and ii) the n-methyl taurates, i.e., the sodium or potassium salts of fatty acid amides of methyl tauride in which the fatty acids, for example, are derived from coconut oil. Other anionic synthetic surfactants of this variety are set forth in U.S. Pat. Nos. 2,486,921; 2,486,922; and 2,396,278, which are incorporated herein by reference.

Still other anionic synthetic surfactants include the classes designated as the sulfosuccinates and sulfo succinamates. These are of the general formulae:

\[
\begin{align*}
\text{(i) and} & \quad R_2-O-C-CH_2-CH-C=O-\text{Na} \\
& \quad \text{SO}_4\text{Na}
\end{align*}
\]

\[
\begin{align*}
\text{(ii) respectively,} & \quad \text{Na-O-S-CH-COONa} \\
& \quad R_2/CH_2\text{-CON} / (R_2 \text{ or H})
\end{align*}
\]

wherein \( R_2 \) is a \( C_2-C_{20} \) alkyl or alkylamido. These classes include such surface active agents as disodium \( N \)-octadecylsulfo-succinamate; tetrasodium \( N-(1,2- \)

dicarboxyethyl)-N-octadecylsulfo-succinamate; diamyl ester of sodium sulfosuccinic acid; dihexyl ester of sodium sulfosuccinic acid; and dioctyl esters of sodium sulfosuccinic acid.

Another class of anionic organic surfactants are the B-alkyloxy alkane sulfonates. These compounds have the following formula:

\[
\text{OR}_4 \text{H} \\
\mid \mid \\
\text{R}_3 \text{-} \text{C} \quad \text{C-} \text{SO}_3 \text{M} \\
\mid \mid \\
\text{H} \quad \text{H}
\]

where \( R_3 \) is a straight chain alkyl group having from 6 to 20 carbon atoms, \( R_4 \) is a lower alkyl group having from 1 to 3 carbon atoms, and \( M \) is a water-soluble cation as hereinbefore described.

Specific examples of B-alkyloxy-alkane-1-sulfonates, or alternatively 2-alkyloxy-alkane-1-sulfonates include: potassium-B-methoxydecanesulfonate, sodium 2-methoxytridecanesulfonate, potassium 2-ethoxytetradecylsulfonate, sodium 2-isopropanoylhexadecylsulfonate, lithium 2-t-butoxytetradecylsulfonate, sodium B-methoxyoctadecylsulfonate, and ammonium B-n-propoxydodecylsulfonate.
Also to be included in the anionic class of surfactants are the disulfonates of the general formula:

\[ (R_5 \text{ or } H) - \begin{array}{c}
\text{O} \\
\text{SO}_3M' \\
\end{array} - \begin{array}{c}
\text{O} \\
\text{SO}_3M' \\
\end{array} \]

wherein \( R_5 \) is a \( C_8- C_{20} \) alkyl group and \( M \) is a watersoluble cation as hereinabove described. The preferred anionics of the disulfonate class are disodium dodecyl diphenyloxide disulfonate and ethoxylated nonylphenyl ammonium disulfonate. All of the above-described anionic surfactants and mixtures thereof may or may not be ethoxylated with from about 1 to about 10 ethylene oxide units per "R" unit.

Also useful as a dispersant surfactant for the granules of the instant invention are the alkali and alkali earth metal salts of the lignosulfonates.

Illustrative of the nonionics which are useful in the surfactant compositions of this invention include the following:

A) Amides such as:

i) Alkanolamides of the formula -
-13-

\[
\begin{array}{c}
\text{O} \\
\text{R'} \\
\text{R - C - N} \\
\text{R''}
\end{array}
\]

wherein \( R' \) and \( R'' \) each can be \(-H, \ -\text{CH}_2\text{CH}_2\text{OH}, \) or \(-\text{CH}_2 - \text{CH} - \text{OH};\)

\( \text{CH}_3 \)

ii) ethoxylated alkanolamides of the formula -

\[
\begin{array}{c}
\text{O} \\
\text{CH}_2\text{-CH}_2\text{-O)}_x\text{H} \\
\text{R - C - N} \\
\text{(CH}_2\text{-CH}_2\text{-O)}_y\text{H}; \text{ and}
\end{array}
\]

iii) ethylene bisamides of the formula -

\[
\begin{array}{c}
\text{O} \\
\text{R - C} \\
\text{N-CH}_2\text{-CH}_2\text{-N} \\
\text{H} \\
\text{C-R} \\
\text{O}
\end{array}
\]

B) Esters such as:
i) fatty acid esters of the formula -

\[ \text{R - C - O - R}_1; \]

ii) glycerol esters of the formula -

\[ \text{R - C - O - CH}_2 - \text{CH - CH}_2 - \text{O - R}_1 \]

\[ \text{OH}; \]

iii) ethoxylated fatty acid esters of the formula -

\[ \text{R - C - O (CH}_2\text{CH}_2\text{O})_x\text{-R}_1; \]

iv) sorbitan esters of the formula -

\[ \text{CH - CH}_2 - \text{O - C - R}; \text{ and} \]

\[ \text{OH} \]

v) ethoxylated sorbitan esters of the formula -
C) Ethoxylates such as:

i) alkylphenol ethoxylates of the formula -
\[ R - \text{C}_6\text{H}_4 - \text{(OCH}_2\text{CH}_2)_n\text{OH}; \]

ii) alcohol ethoxylates of the formula -
\[ R - \text{O} - \text{(CH}_2\text{CH}_2)_n\text{H}; \]

iii) tristyrylphenol ethoxylates of the formula -
iv) mercaptan ethoxylates of the formula -
\[ R - S - (CH_2CH_2O)_yH \]

D) End-capped and EO/PO block copolymers such as:

i) alcohol alkoxylates of the formula-
\[ \text{CH}_3 \]
\[ R - (OCH_2CH_2)_x \cdot (O - \text{CH} - \text{CH}_2)_m - \text{OH}; \]

ii) ethylene oxide/propylene oxide block copolymers of the formula -
\[ \text{CH}_3 \]
\[ \text{HO} - (CH_2CH_2O)_x \cdot (CH_2-CH-O)_m \cdot (CH_2CH_2O)_y - \text{H}; \]
iii) copolymers of the formula -

\[
\begin{align*}
\text{H} & \text{(OCH}_2\text{CH}_2\text{)}_x \ - \ (\text{OCHCH}_2)_m \ - \ (\text{CH}_2\text{CHO})_n \ - \ (\text{CH}_2\text{CH}_2\text{O})_y \ - \ \text{H} \\
\text{or} & \\
\text{H} & \text{(OCH}_2\text{CH}_2\text{)}_x \ - \ (\text{OCHCH}_2)_m \ - \ (\text{CH}_2\text{CHO})_n \ - \ (\text{CH}_2\text{CH}_2\text{O})_y \ - \ \text{H}
\end{align*}
\]

iv) chlorine capped ethoxylates of the formula -

\[
\begin{align*}
\text{R} & \ - \ (\text{OCH}_2\text{CH}_2\text{)}_x \text{Cl}; \ and \\
v) \text{tetra-functional block copolymers of the formula -}
\end{align*}
\]

\[
\begin{align*}
\text{CH}_3 & \text{CH}_3 \\
\text{H} & \text{(OCH}_2\text{CH}_2\text{)}_x \ - \ (\text{OCHCH}_2)_m \ - \ (\text{CH}_2\text{CHO})_n \ - \ (\text{CH}_2\text{CH}_2\text{O})_y \ - \ \text{H} \\
\text{or} & \\
\text{H} & \text{(OCH}_2\text{CH}_2\text{)}_x \ - \ (\text{OCHCH}_2)_m \ - \ (\text{CH}_2\text{CHO})_n \ - \ (\text{CH}_2\text{CH}_2\text{O})_y \ - \ \text{H}
\end{align*}
\]

wherein...
R is a fatty alkyl group, preferably a C₆-
C₂₂ fatty alkyl group, most preferably a C₆-
C₁₈ fatty alkyl group;

R₁ is -H or a fatty alkyl group,
preferably -H or a C₆-
C₂₂ fatty alkyl group, most
preferably -H or a C₆-
C₁₈ fatty alkyl group;

x, x', y, y' and n are each independently
moles of ethylene oxide preferably 1 to 300; most
preferably 1 to 150; and

m, m', l and l' are each independently
moles of propylene oxide, preferably 1 to 300; most
preferably 1 to 150.

Among the preferred dispersing agents
are: the alkylnaphthalenesulfonate-formaldehyde
condensates; sodium lignosulfonate, diphenyloxide,
ethoxylated tristyrylphenol ethoxylates, ethoxylated
tristyrylphenol phosphates, the ethylene
oxide/propylene oxide block copolymers, and acid,
salts and copolymers of the polyacrylates.

Among the preferred rewetting agents are:
the alkylnaphthalenesulfonates, sodium methyloleoyl
taurate, the sulfosuccinates, the carboxylates, the
alkylarylsulfonates, the ethoxylated alkyl phenols and
the ethoxylated alcohols.
Although electrostatic dispersants such as the anionics are preferred, steric dispersants such as the polyvinyl alcohols can also be used.

Although the dry spreadable, water dispersible granules of this invention can be prepared by a standard pan granulation process, it is preferred that the granules, with or without the presence of a pesticide, are prepared via a homogeneous extrusion process. Of note, granules that are prepared in the absence of a pesticide by the homogeneous extrusion process can subsequently be sprayed with pesticide to adhere same to the granules.

The extrusion granulation process is as follows: First, each component is weighed, combined, and blended. Water is then added with continuous mixing until from about 10 to about 50 weight percent, preferably about 25 to about 45 weight percent moisture content is realized. The mixture is then fed to a low pressure basket or radial extruder. The wet mass is extruded through a die having openings with diameters of from about 0.8 mm to about 2.0 mm, preferably about 1 mm. The extrudate is then fed to a vibratory fluidized bed dryer which reduces the moisture content to below about 5; preferably to below about 4 and most preferably below about 2.5 weight
percent; the weight percents being based on the total weight of the composition at that respective step in the process.

In the spray formulation process, diatomaceous earth/surfactant granules are prepared by the homogeneous extrusion process described above, and the pesticide is dissolved in an appropriate solvent, e.g., iso-propanol or xylene in an amount sufficient to make a suitably viscous sprayable solution, typically from about 10 to above about 50 percent by weight pesticide and sprayed onto the granules. It is also recognized that certain actives can be melt sprayed onto the granules. The spray is continued onto the granules with continuous mixing. The sprayed granules are then dried, preferably in a fluid-bed dryer to final moisture content as indicated above. The product, however made, is then sieved in a particle classifier to remove oversized and undersized granules. It is preferred that the final product is that which passes through a 10 mesh screen but will not pass through a 60 mesh screen.

A characteristic of the dry spreadable, water dispersible diatomaceous earth granules of this invention is the speed with which the granules disperse in water. When the granules of this
invention with size of 1x5 (diameter x length in millimeters) are gently placed on the surface of cold tap water (about 20°C) in, for example, a beaker, the granules, with no mechanical energy input, will completely disintegrate within two (2) to ten (10) seconds. Most conventional water dispersible granules require from five (5) to thirty (30) or more inversions of the beaker to effect a similar result in a similar time period.

As indicated above, a further preferred embodiment of the present invention is the use of the granules described above as a carrier for the delivery of pesticides, e.g., insecticides, herbicides, fungicides and plant growth regulators. For the purpose of expediency, the term "pesticide" herein is also defined to include fertilizers such as urea. Specifically, the present invention is directed towards a granule as described above, methods of preparing said granules, use of said granules as a carrier to deliver pesticides, and the pesticidal granule per se.

Although the instant invention is universally applicable to all pesticides, the preferred classes of pesticides include the pyridines, carbamates and dithiocarbamates, chlorinated
hydrocarbons, organophosphorous compounds, dinitroanilines, diphenyl ethers and synthetic pyrethroid compounds.

Other well known specific pesticides include oxadiazon, iprodione, aluminum salt of ethyl hydrogen phosphonate, (fosetyl Al), and those described in U.S. Patent 5,317,834 which is incorporated herein by reference.

In this embodiment, the pesticide is present in the granule described above, in an effective amount that will not interfere with the desired properties of the granules, i.e., dry spreadability and ability to disperse readily. For example, the pesticide must not be present in an amount that would result in "sticky" granules which cannot be spread, or in an amount such that the granules will not disperse readily upon contact with water.

Although the pesticide may be present up to about 60 weight percent, the pesticide is preferably present at from about 0.005-30 weight percent, most preferably 0.5-20 weight percent, based upon the total weight of the diatomaceous earth, surfactant, and pesticide in the granule.
The granules, containing the pesticide can be applied with a dry spreader to a target area and, when exposed to water via, for example, rain or irrigation, readily decompose and actively disperse. The high dispersibility allows the pesticide to be delivered over a larger surface area than that immediately covered by the granules resulting in a much more effective delivery of the pesticides to the targeted area.

The following examples are intended to illustrate the claimed invention and are not in any way designed to limit its scope. Numerous additional embodiments within the scope and spirit of the claimed invention will become apparent to those skilled in the art.

**Materials**

The following materials are used in the Examples. Each of the carrier compounds was obtained from Celite Corp., Lompac, CA.
### Diatomaceous Earth

<table>
<thead>
<tr>
<th>Compound</th>
<th>How Manufactured</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 209</td>
<td>Natural diatomaceous earth</td>
</tr>
<tr>
<td>Celite 392</td>
<td>Natural diatomaceous earth</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Natural diatomaceous earth</td>
</tr>
<tr>
<td>Celite 503</td>
<td>Flux Calcinated diatomaceous earth</td>
</tr>
<tr>
<td>Celite 560</td>
<td>Flux Calcinated diatomaceous earth</td>
</tr>
</tbody>
</table>

### Synthetic Compound

<table>
<thead>
<tr>
<th>Compound</th>
<th>How Manufactured</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microcel E</td>
<td>Calcium silicate</td>
</tr>
<tr>
<td>Microcel T-49</td>
<td>Calcium silicate</td>
</tr>
<tr>
<td>Microcel A</td>
<td>Calcium silicate</td>
</tr>
<tr>
<td>Microcel B</td>
<td>Calcium silicate</td>
</tr>
<tr>
<td>Celite R-685</td>
<td>Magnesium silicate</td>
</tr>
</tbody>
</table>

---

**Surfactants:**

- **Supragil WP**: a trademark of Rhône-Poulenc Inc. for an alkylnaphthalenesulfonate.
- **Supragil MNS/90**: a trademark of Rhône-Poulenc Inc. for an alkylnaphthalenesulfonate-formaldehyde condensate.
- **Soprophor S/40-P**: a trademark of Rhône-Poulenc Inc. for a tristyrylphenol ethoxylate.
Tests

The Speed of Dispersion Test is a determination of the degree of breakup of the granules in a petri dish after contact with droplets of water.

To determine the Speed of Dispersion, a test is conducted as follows:

Approximately 0.1-0.2 grams of granules are placed into a petri dish. The granules are placed approximately 5 mm apart. A single droplet of water is squirted onto each granule. After 5-10 seconds, the granule is observed and rated as discussed below. Application of single drops of water is continued until total disintegration occurs.

Speed of Dispersion is measured on a scale of 0 to 9, with 9 being the highest and 0 being the lowest. A rating of 9 indicates that the granule disperses immediately on contact with water, spreading out evenly without additional water needed. A rating of 5 indicates that the granule loses integrity after about 2 seconds on contact with water. Almost no material spreads from the original body of the granule. On contact with the second water droplet, material disperses into very small fragments. The fragments disperse totally on contact with a third
water droplet. A rating of 0 indicates that water has no effect on the granule.

Susceptibility Test: Susceptibility is defined as the amount of ingredients suspended after a given time in a column of liquid of a stated height. Susceptibility is expressed as a percentage of the amount of the ingredient in the original suspension. Susceptibility was measured using CIPAC Method MT168. The water used was CIPAC Standard Water D (342 ppm hardness).

Breakup Test: Breakup measures the breakup of the granules in a column of water and is evaluated on a scale of 0 to 9 with 9 being the highest. A rating of 9 indicates that the granules immediately disperse into a cloud of material which very slowly spreads downward, eventually filling the entire column. A rating of 5 indicates that small amounts of the granules disperse into a cloud of material. The remaining material migrates downward in the form of fragments, with approximately 1/2-1/4 of the original mass of granules reaching the bottom of the cylinder as fragments. A rating of 0 indicates that the granules sink to the bottom of the cylinder unaffected by the water.
To determine Suspensibility and Breakup, a graduated cylinder test is conducted as follows:

Approximately 0.2 grams of granule formulation are placed into separate graduated cylinders containing 100 mls. of water. The results are observed, and for Breakup, a numerical value from 0-9 is assigned.

Rating: An overall rating is given each granule formulation based on the Speed of Dispersion and Breakup results as follows: if both are individually 6 or better, the formulation is given a "Good" rating; if both are individually 4 or better, the formulation is given a "Fair" rating; and if below the "Fair" standard, the formulation is given a "Poor" rating.

Example 1

Two different diatomaceous earth surfactant granule formulations are prepared as follows: 85 grams of the diatomaceous earth; 8 grams of Supragil MNS/90; 4 grams of Soprophor S/40-P; and 3 grams of Supragil WP are weighed on a balance; added to a food processor; and mixed for 2 minutes at the highest rate. Then about 90 grams of water is added and the material mixed for an additional 2 minutes.
Care is taken to ensure good uniformity of the material. The mass is then transferred to a benchtop basket extruder and extruded through die holes as indicated below. The wet granules are dried in a fluid-bed dryer at a temperature of 50 - 60°C for about 10 minutes. The moisture content of the granules after drying is from about 2.2 to 2.5 weight percent.

Speed of Dispersion and Breakup tests are conducted on two granule sizes made from Celite 500 and one granule size made from Celite 209. The results are as indicated in Table I below.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Granule Size</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 500</td>
<td>1 mm</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>2 mm</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 209</td>
<td>1 mm</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
</tbody>
</table>

**Example 2**

A series of dry spreadable, water dispersible granules is prepared as described above using several different diatomaceous earth samples. The granules are then dried in two sets; one at 50°C
for 20 minutes and the other at 100°C for 10 minutes. The final moisture content for all granules is less than 4%.

For comparison, water dispersible granules containing synthetic calcium and magnesium silicates are also prepared.

The formulation for all granules is as follows:

| Weight Percent (%) | Diatomaceous earth or synthetic silicate compound | 85
| Supragil WP | 3 |
| Supragil MNS/90 | 8 |
| Soproponor S/40P | 4 |

The performance results are as indicated in Table II below.

**Table II**

| Sample | Suspensibility
| 50°C | 100°C | Breakup
| 50°C | 100°C |
|-------|-------|-------|-------|
| Celite 209 | 43.5 | 38.8 | 8 | 8 |
| Celite 392 | 34.7 | 32.8 | 7 | 7 |
| Celite 500 | 42.5 | 36.4 | 8 | 8 |
| Celite 503 | 32.5 | 33.2 | 2 | 2 |
| Celite 560 | 26.1 | 25.3 | 2 | 3 |
-30-

<table>
<thead>
<tr>
<th>Sample</th>
<th>Suspensibility 50°C</th>
<th>Suspensibility 100°C</th>
<th>Breakup 50°C</th>
<th>Breakup 100°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microcel E</td>
<td>21.0</td>
<td>15.4</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel T-49</td>
<td>10.5</td>
<td>13.6</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel A</td>
<td>18.3</td>
<td>18.1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel B</td>
<td>30.9</td>
<td>31.4</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Celite R-685</td>
<td>26.2</td>
<td>24.6</td>
<td>2</td>
<td>2</td>
</tr>
</tbody>
</table>

From the results shown in Table II above, the performance of the natural diatomaceous earth samples is superior to that of the synthetically made materials. The natural diatomaceous earth samples show overall better Suspensibility, as well as better Breakup in water.

For Celite 209, 392 and 500 Suspensibility is 32% or higher with Breakup values of 7-8. From Table II, one also observes that drying temperature has little effect on Breakup. However, drying temperature appears to influence Suspensibility; low drying temperatures improving the Suspensibility.

**Example 3**

The series of dry spreadable, water dispersible carrier granules of Example 2 are tested to determine the Speed of Dispersion of the granules. The results of the Speed of Dispersion tests are as shown below in Table III.
### Table III

<table>
<thead>
<tr>
<th>Sample</th>
<th>Speed of</th>
<th>Dispersion</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>50°C</td>
<td>100°C</td>
</tr>
<tr>
<td>Celite 209</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Celite 392</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>Celite 500</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Celite 503</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Celite 560</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>Microcel E</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel T-49</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel A</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel B</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Celite R-685</td>
<td>2</td>
<td>2</td>
</tr>
</tbody>
</table>

The petri dish dispersion performance of the natural diatomaceous earth samples are much superior to that of the synthetically-made materials.

**Example 4**

The physical properties of the dry spreadable, water dispersible granules of Examples 2 and 3 are determined and set forth in Table IV below.
### Table IV

<table>
<thead>
<tr>
<th>Sample</th>
<th>WA'</th>
<th>Mpas'</th>
<th>SA'</th>
<th>MPo's</th>
<th>PV'</th>
<th>D'</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 209</td>
<td>185</td>
<td>5</td>
<td>10-20</td>
<td>0.88</td>
<td>3</td>
<td>8</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Celite 392</td>
<td>215</td>
<td>25</td>
<td>10-20</td>
<td>2.6</td>
<td>3.2</td>
<td>8</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>Celite 500</td>
<td>230</td>
<td>22</td>
<td>19</td>
<td>2.5</td>
<td>1.4</td>
<td>7</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>Celite 503</td>
<td>220</td>
<td>31</td>
<td>1.0</td>
<td>9.5</td>
<td>2.6</td>
<td>12</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Celite 560</td>
<td>240</td>
<td>52</td>
<td>0.7</td>
<td>26.6</td>
<td>2.6</td>
<td>16</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Celite R685</td>
<td>*</td>
<td>10</td>
<td>175</td>
<td>2</td>
<td>5.5</td>
<td>17</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Microcel A</td>
<td>410</td>
<td>17</td>
<td>100</td>
<td>1.6</td>
<td>5.2</td>
<td>7.2</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel B</td>
<td>300</td>
<td>14</td>
<td>91</td>
<td>2.6</td>
<td>3.8</td>
<td>14.6</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel E</td>
<td>500</td>
<td>23</td>
<td>120</td>
<td>1.8</td>
<td>5.5</td>
<td>5.8</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Microcel T-49</td>
<td>115</td>
<td>7</td>
<td>105</td>
<td>2.2</td>
<td>1.7</td>
<td>11.6</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

'Water Absorption, % by Weight
'Median Particle Size - microns
'Surface Area - (meters$^2$/gram)
'Median Pore Size - microns
'Pore Volume - cc/g
'Bulk Density - lb/cubic foot

Thus, as can be seen by the results in Table IV above, unexpectedly superior Breakup and Speed of Dispersion performance results can be
realized with the granules of this invention when natural diatomaceous earth having a surface area in
the range of from greater than about 5 to less than
about 90 square meters per gram; preferably, from
greater than about 10 to less than about 60 meters per
gram and a pore volume in the range of from greater
than about 2 cubic centimeters (cc) per gram to less
than about 5 cc per gram, preferably, from greater
than about 3 cc per gram to less than about 4 cc per
gram, is utilized.

Example 5

The following is illustrative of the preparation of a homogeneous spreadable, water
dispersible granule (SWDG) containing 2 weight percent
(active ingredient (a.i.)) chlorpyrifos.

The homogeneous formulation is composed of:

2.0 wt.% Chlorpyrifos
3.0 wt.% Supragil WP
4.0 wt.% Soprophor S/40-P
8.0 wt.% Supragil MNS/90
83.0 wt.% Diatomaceous Earth (Celite 500)

The test material is prepared by using a
benchtop basket extruder. The batch size is 50 grams.
First, chlorpyrifos technical (99%, low odor), is dissolved in iso-propanol to make a 20 weight percent chlorpyrifos stock solution. To make a batch of 2 weight percent a.i. SWDG homogeneous formulation, the following materials are weighed on a balance and mixed for 2 minutes at the highest rate in a compact food processor: 41.5 grams Celite 500, 4.0 grams Supragil MNS/90, 2.0 grams Soprophor S/40-P, and 1.5 grams Supragil WP. Then, 5 grams of the 20 weight percent chlorpyrifos (in iso-propanol) solution is added using a pipet. The mixture is mixed for one more minute. Then 44.5 - 45.5 grams of water is added and the material mixed for another 2 minutes. Care is taken to ensure a good uniformity of the material. The mass is transferred to a benchtop basket extruder and extruded. The dye has 1 mm holes. The wet granules are dried in a fluid-bed dryer at a temperature of 50°C (set-point) for 10 minutes. The actual drying temperature is about 60°C. The moisture content of the granules after the dryer is 2.2 - 2.5 weight percent. The dry granules have a good visual flowability. The tap density of the granules is 0.35 grams/cc. The Speed of Dispersion rating from the petri dish test is 8. From the graduated cylinder test, the Breakup rating is 7-8.
The moisture content of raw material Celite 500 is 3.7 weight percent. So, the weight of raw material Celite 500 is close to the dry weight of Celite 500. According to the product specifications of Supragil MNS/90, Soprophor S/40-P, and Supragil WP, the moisture contents of these components are no higher than 2 weight percent. Also, samples of chlorpyrifos technical were tested for volatile materials in a ventilating oven. The weight loss after 2 hours at 141°F (≈60.5°C) was only 0.6 weight percent. Hence, the granules should not lose significant amount of chlorpyrifos at the dryer. Therefore, the SWDG product as manufactured above contains approximately 2.0 weight percent chlorpyrifos.

Example 6

The following is illustrative of the preparation of a 2 weight percent a.i. chlorpyrifos SWDG whereby the chlorpyrifos is sprayed onto the diatomaceous earth-surfactant substrate to make the SWDG pesticidal-carrier granules.

The activity and final formulation of the 2 weight percent a.i. chlorpyrifos SWDG spray-on granule is almost identical to that of the 2 weight percent a.i. chlorpyrifos SWDG homogeneous granule. However,
the spray-on granule is prepared using a different process from that of the homogeneous formulation. An SWDG inert granule is prepared first, then chlorpyrifos solution (in iso-propanol) is sprayed on the inert granules to make 2 weight percent a.i. chlorpyrifos SWDG spray-on material. The SWDG inert formulation is composed of the following ingredients:

- 3.0 weight percent Supragil WP
- 4.0 weight percent Soprophor S/40-P
- 8.0 weight percent Supragil MNS/90
- 85.0 weight percent Diatomaceous Earth (Celite 500)

The inert material is made in a process similar to that used to prepare the 2 weight percent a.i. chlorpyrifos SWDG homogeneous granule. To make a 50 gram batch, the following ingredients are mixed in a food processor: 42.5 grams Celite 500, 4.0 grams Supragil MNS/90, 2.0 grams Soprophor S/40-P, and 1.5 grams Supragil WP. Then, 46.5 - 47.0 grams of water is added and the material is well-mixed to become an extrudable mass. The mass is transferred to the benchtop basket extruder and extruded. The benchtop extruder has a 1 mm dye. The wet granules are dried in the fluid-bed dryer at the temperature of 50°C (set-point) for 8 minutes. The moisture content of the inert granules after drying is 2.1 - 4.6 weight
percent. The dry inert granules have a good visual flowability. The tap density of the inert granules is 0.34 grams/cc. The Speed of Dispersion rating from the petri dish test and the Breakup rating from the graduated cylinder test are 8 and 8 respectively.

The next step is to spray the pesticide active ingredient onto the SWDG inert material. Chlorpyrifos (99%, low odor), is dissolved in iso-propanol to make a 20% weight percent solution. A benchtop tumbling mixer is used. 98 grams of the above SWDG inert granules are added to the mixer and the mixer turned on. Then 10.0 grams of 20% chlorpyrifos (in iso-propanol) solution are sprayed onto the inert granules using a hand sprayer in a time period of 30 seconds. The material is mixed for another 5 minutes. The spray-on granules are dried in the fluid bed dryer at a temperature of 50°C (set-point) for 8 minutes. The moisture content of the granules after the dryer is 2.1 - 2.2 weight percent. The dry granules have a good visual flowability. The tap density of the granules is 0.35 grams/cc. The Speed of Dispersion rating from the petri dish test and the Breakup rating from the graduated cylinder test are 8 and 8 respectively.
Example 7

Additional dry spreadable, water dispersible granules are made by the pesticidal spray-on process described in Example VI using different final concentrations of chlorpyrifos on the granules. The performance results are as indicated in Table V below.

Table V

<table>
<thead>
<tr>
<th>Sample</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.4% a.i. Chlorpyrifos</td>
<td>8</td>
<td>-</td>
</tr>
<tr>
<td>2.0% a.i. Chlorpyrifos</td>
<td>8</td>
<td>8</td>
</tr>
<tr>
<td>2.1 % a.i. Chlorpyrifos</td>
<td>8</td>
<td>-</td>
</tr>
</tbody>
</table>

Example 8

A series of dry spreadable, water dispersible granules is prepared as described above using Celite 500 and several different pesticides.

The following pesticides are used:

- Oxadiazon: Chipco® Ronstar® 50WP Brand Herbicide, a trademark of Rhone-Poulenc for a formulated wettable powder containing 50% oxadiazon, a herbicide.
Fosetyl Al: Chipco® Aliette® WDG Fungicide, a trademark of Rhone-Poulenc for a water dispersible granule containing 80% aluminum salt of ethyl hydrogen phosphonate, (fosetyl-Al), a fungicide.

Iprodione: Chipco® 26109, a trademark of Rhone-Poulenc for a formulated wettable powder containing 50% iprodione, a fungicide.

Chlorpyrifos: Chlorpyrifos Technical (99% a.i.) obtained from Micro Flo Company, an insecticide.

Mancozeb (DG): a coordination product of Zn²⁺ and manganese ethylene bis(dithiocarbamate), (75% a.i.) a fungicide, obtained from Lesco Inc.

Mancozeb (Dithane): a coordination product of Zn²⁺ and manganese ethylene bis(dithiocarbamate) (80-85% a.i.), a fungicide, obtained from Rohm & Haas Company.

Chlorothalonil: Bravo® W-75, a trademark of ISK Biotech for terachloroisophthalonitril (75% a.i.), a fungicide.

Prodiamine: Barricade 65 WG Brand herbicide, a trademark of Sandoz Crop Protection Corp. for a formulated water dispersible granule containing Prodiamine (65% a.i.).

The formulation for all granules which are prepared by the aforesdescribed homogeneous process
(except where indicated) and except those containing chlorpyrifos, is as follows:

| Celite 500 and pesticide (Total of Two) | 85 |
| Supragil WP                       | 3  |
| Supragil MNS 90                   | 8  |
| Soprohor S/40P                    | 4  |

Although the amount of the pesticide may be varied, see Tables VI and VII, "% Active", the total amount of pesticide and Celite 500 remains constant at 85% of the total weight.

The granules containing chlorpyrifos are prepared as described in Example 5.

Table VI represents performance results realized using formulated pesticidal products as the pesticidal starting material in the granules of this invention whereas Table VII represents performance results realized using "technical" grade pesticides as the pesticidal starting material.

Breakup and Speed of Dispersion of each of the formulations utilized are determined and are listed below in Tables VI and VII. In addition, a
determination of the overall rating of each sample is shown in the last column.

The first series of granules in Table VI are formulated using Ronstar 50WP, a wettable powder containing 50% oxadiazon as the active herbicide ingredient. The granules in this series contain 0.5, 1.0, 2, 5, 10, 15, 20 and 30 weight percent oxadiazon based on the total weight of a granule of 1.0 mm size.

As can be seen in Table VI, good results are obtained for up to 15 weight percent oxadiazon; above 15 weight percent dispersibility is reduced. Therefore it appears that 15% is the preferred upper limit of the range defining peak performance.

The next series of granules in Table VI are formulated using Aliette, a fungicide containing ethyl hydrogen phosphonate (fosetyl Al) as the active pesticide ingredient. This series contains 0.8, 1.6, 4, 8, and 16 weight percent active ingredient based on the total weight of a granule of 1.0 mm size. The results shown in Table VI indicate that with up to 4 weight percent Aliette, good results are obtained.

The third series of granules in Table VI are formulated using Chipco 26019, a fungicide containing 50% iprodione as the active ingredient. This series contains 0.5, 1.0, 2.5, 5, 10, 15, 20 and
30 weight percent active ingredient based upon the total weight of a granule of size 1.0 mm. As can be seen in Table VI, good results are obtained for up to 15 weight percent pesticide; it appears that 15% is the preferred upper limit for peak performance with respect to dispersibility.

A fourth set of samples, containing chlorpyrifos, an insecticide, is prepared as described in Example 5. This series contains 2.0, 5.0, 10.0 and 15.0 weight percent active ingredient based upon the total weight of a granule of 1.0 mm size. As can be seen in Table VI, granules containing 2.0%, 5.0%, and 10.0% chlorpyrifos perform well.

Test results from various other pesticides are as indicated in Table VI. The overall results from these different active ingredients indicate that formulated pesticides can be loaded onto water dispersible granules to a certain degree. The approximate peak values when using the formulated pesticides in the studies are:
Oxadiazon 15%
Ethyl Hydrogen Phosphonate 4%
Iprodione 15%
Chlorpyrifos 10%

5
Mancozeb (DG) 10%
Mancozeb (Dithane) 15%
Chlorothalanil > 5%

(Above percents are weight percents based upon the total granule weight.) Beyond the peak values there is reduced dispersibility indicating that at a point, the presence of the formulated pesticide interferes with the ability of the granule to disperse rapidly in water. However, in the instant invention, these peak values are equal to or superior to concentrations found in existing granular spreadable products as will be seen in Table VIII below.
### Table VI

<table>
<thead>
<tr>
<th>Carrier</th>
<th>Pesticide</th>
<th>% Active</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>0.5</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>1</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>2</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>5</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>10</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>15</td>
<td>7</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>20</td>
<td>5</td>
<td>5</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiazon</td>
<td>30</td>
<td>2</td>
<td>2</td>
<td>Poor</td>
</tr>
<tr>
<td>Celite 500</td>
<td>fosetyl Al</td>
<td>0.8</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>fosetyl Al</td>
<td>1.6</td>
<td>8</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>fosetyl Al</td>
<td>4</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>fosetyl Al</td>
<td>8</td>
<td>6</td>
<td>3</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>fosetyl Al</td>
<td>16</td>
<td>2</td>
<td>0</td>
<td>Poor</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>0.5</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>1</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>2.5</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>5</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>10</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>15</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>20</td>
<td>3</td>
<td>2</td>
<td>Poor</td>
</tr>
<tr>
<td>Celite 500</td>
<td>iprodione</td>
<td>30</td>
<td>1</td>
<td>0</td>
<td>Poor</td>
</tr>
<tr>
<td>Celite 500</td>
<td>chlorpyrifos</td>
<td>2</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>chlorpyrifos</td>
<td>5</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>chlorpyrifos</td>
<td>10</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>chlorpyrifos</td>
<td>15</td>
<td>6</td>
<td>5</td>
<td>Fair</td>
</tr>
<tr>
<td>Carrier</td>
<td>Pesticide</td>
<td>% Active</td>
<td>Speed of Dispersion</td>
<td>Breakup</td>
<td>Rating</td>
</tr>
<tr>
<td>------------</td>
<td>-----------------</td>
<td>----------</td>
<td>---------------------</td>
<td>---------</td>
<td>--------</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorpyrifos Spray*</td>
<td>1.4</td>
<td>8</td>
<td>-</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorpyrifos Spray*</td>
<td>2.1</td>
<td>8</td>
<td>-</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (DG)</td>
<td>1</td>
<td>8</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (DG)</td>
<td>2</td>
<td>8</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (DG)</td>
<td>5</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (DG)</td>
<td>10</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (DG)</td>
<td>20</td>
<td>6</td>
<td>5</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (DG)</td>
<td>30</td>
<td>5</td>
<td>3</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (Dithane)</td>
<td>15</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancoset (Dithane)</td>
<td>30</td>
<td>6</td>
<td>5</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil</td>
<td>2</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil</td>
<td>5</td>
<td>7</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Prodimine</td>
<td>6.5</td>
<td>8</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 501</td>
<td>Prodimine</td>
<td>13</td>
<td>6</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 502</td>
<td>Prodimine</td>
<td>19.5</td>
<td>3</td>
<td>4</td>
<td>Poor</td>
</tr>
</tbody>
</table>

*Prepared by a spray process as follows:

1. Inert SWDG is prepared as in Example 5 1.
2. Micro Flo Chlorpyrifos Pro 6mup (obtained from Micro Flo Company) is diluted in xylene at a weight ratio of 1 to 9.
3. 5 grams of SWDG inert granule is placed in a petri dish with a diameter of 10 cm. The granules are spread out as a thin layer.
4. The chlorpyrifos/xylene solution is sprayed on the granules using a hand sprayer.

5. The wet granules are dried in a fluid-bed dryer at a temperature of 80°C for 20 minutes.

**Table VII**

As previously indicated, the following Table VII presents performance results realized using technical grade pesticides as the pesticidal starting material in the granule homogeneous formulation process of this invention.

<table>
<thead>
<tr>
<th>Carrier</th>
<th>Pesticide</th>
<th>% Active</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 500</td>
<td>Oxadiason, Tech</td>
<td>20</td>
<td>6</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiason, Tech</td>
<td>30</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiason, Tech</td>
<td>40</td>
<td>6</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Oxadiason, Tech</td>
<td>50</td>
<td>5</td>
<td>5</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Iprodione, Tech</td>
<td>20</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Iprodione, Tech</td>
<td>30</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Iprodione, Tech</td>
<td>40</td>
<td>4</td>
<td>5</td>
<td>Fair</td>
</tr>
</tbody>
</table>

15 The above Table VII illustrates the significantly higher pesticidal loadings that can be realized by using technical grade pesticide as the starting
material in the SWD granules of this invention as compared to the already superior loadings achievable with formulated pesticide usage that was shown in Table VI. It is believed that the "technical" provides better performance than use of the formulated starting material because of the absence of carrier and formulation aids that are contained in the formulated products.

For comparative purposes, and to vividly underscore the unexpectedly high pesticidal loadings realizable with the SWD granules of this invention, the following Table VIII contains a listing of typical, commercially available dry spreadable, granular pesticidal products and their respective pesticidal (% active) loadings.
Table VIII

<table>
<thead>
<tr>
<th>Product</th>
<th>% a.i.</th>
<th>Generic Name</th>
<th>Manufacturer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dursban</td>
<td>2.32</td>
<td>Chlorpyrifos</td>
<td>The Andersons Lawn Products Division</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Maumee, OH</td>
</tr>
<tr>
<td>Insecticide III</td>
<td>1.34</td>
<td>Chlorpyrifos</td>
<td>The O.M. Scott and Sons Company</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Marysville, OH</td>
</tr>
<tr>
<td>Ronstar G</td>
<td>2</td>
<td>Oxadiazon</td>
<td>Rhone Poulenc Ag Company Research Triangle Park, NC</td>
</tr>
<tr>
<td>Goosegrass/</td>
<td>1.31</td>
<td>Oxadiazon</td>
<td>The O.M. Scott and Sons Company</td>
</tr>
<tr>
<td>Crabgrass Control</td>
<td>5.25</td>
<td>Bensulide</td>
<td>Marysville, OH</td>
</tr>
<tr>
<td>5% Daconil</td>
<td>5</td>
<td>Chlorothalonil</td>
<td>The Andersons Lawn Products Division</td>
</tr>
<tr>
<td>Fungicide</td>
<td>1.3</td>
<td>Iprodione</td>
<td>Maumee, OH</td>
</tr>
<tr>
<td>Fungicide X</td>
<td>1.3</td>
<td>Iprodione</td>
<td>The O.M. Scott and Sons Company</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Marysville, OH</td>
</tr>
</tbody>
</table>

Example 9

Granules of this invention are prepared by the homogeneous process as set forth in Example 5 with the chlorpyrifos being replaced by 17.5% urea fertilizer. The performance results are given in Table IX below.

Table IX

<table>
<thead>
<tr>
<th>Sample</th>
<th>% Active</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 500</td>
<td>Urea</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>(17.5%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Example 10

For purposes of comparison, various other materials well known in the pesticidal carrier art, are substituted for the diatomaceous earth and examined in the granule surfactant systems of this invention. At weight loadings of 85%, the materials are substituted for the diatomaceous earth in the process as set forth in Example 1. The performance results are given in Table X below.

Table X

<table>
<thead>
<tr>
<th>Sample</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kaolinite</td>
<td>0</td>
<td>0</td>
<td>Poor</td>
</tr>
<tr>
<td>Lattice NT</td>
<td>1</td>
<td>0</td>
<td>Poor</td>
</tr>
<tr>
<td>Attacote</td>
<td>3</td>
<td>1</td>
<td>Poor</td>
</tr>
<tr>
<td>AC FD 181</td>
<td>0</td>
<td>0</td>
<td>Poor</td>
</tr>
<tr>
<td>Sulfur</td>
<td>1</td>
<td>1</td>
<td>Poor</td>
</tr>
<tr>
<td>Vermiculite Dust</td>
<td>3</td>
<td>1</td>
<td>Poor</td>
</tr>
</tbody>
</table>

Example 11

For additional comparative purposes, a synergistic blend of an anionic surfactant and a lignosulfonate (Ag RHO SP-33D, a trademark of Rhone-
Poulenc) is substituted for the Supragil MNS/90 surfactant of this invention in the process of Example 5 and homogeneous process granules are prepared with three pesticides as set forth in Table XI below together with performance results.

**Table XI**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Pesticide</th>
<th>% Active</th>
<th>Speed of Dispersion</th>
<th>Break-up</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 500</td>
<td>Mancozeb (DG)</td>
<td>10</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancozeb (DG)</td>
<td>20</td>
<td>6</td>
<td>4</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancozeb (DG)</td>
<td>30</td>
<td>5</td>
<td>3</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancozeb (Dithane)</td>
<td>15</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Mancozeb (Dithane)</td>
<td>30</td>
<td>6</td>
<td>5</td>
<td>Fair</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil</td>
<td>2</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
</tbody>
</table>

**Example 12**

It is also an embodiment of this invention to incorporate multiple pesticidal products into the SWD granules. This can be accomplished in several ways, for example: 1) blending two or more pesticides in the starting material of the homogeneous process; 2)
spraying two or more pesticides upon the diatomaceous earth-surfactant granules as in the spray-on process described above; or 3) a combination of these processes, that is, incorporating one or more pesticides in a homogeneous process followed by the spraying of one or more pesticides upon the homogeneously prepared granules.

SWD granules containing two pesticides are prepared in accordance with the homogeneous process of Example 5. The weight percentages of the two pesticides; their identities and the performance results of the final granulated products are set forth in Table XII below.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Pesticides</th>
<th>Ingredient Percentages</th>
<th>Speed of Dispersion</th>
<th>Breakup</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil, Iprodione</td>
<td>3% / 97%</td>
<td>8</td>
<td>8</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil, Iprodione</td>
<td>44 / 16</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil, Iprodione</td>
<td>44 / 24</td>
<td>7</td>
<td>7</td>
<td>Good</td>
</tr>
<tr>
<td>Celite 500</td>
<td>Chlorothalonil, Iprodione</td>
<td>12% / 38%</td>
<td>7</td>
<td>6</td>
<td>Good</td>
</tr>
</tbody>
</table>
Example 13

A series of homogeneously processed granules are produced as set forth in Example 5 using a 1.0 mm or a 2.0 mm die on the extruder to determine the effect of particle size on Speed of Dispersion performance. The resulting data are indicated in Table XIII below.

Table XIII

<table>
<thead>
<tr>
<th>Sample</th>
<th>% Active</th>
<th>Speed of Dispersion (1.0 mm)</th>
<th>Speed of Dispersion (2.0 mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oxadiazon</td>
<td>2</td>
<td>8</td>
<td>6</td>
</tr>
<tr>
<td>Fosetyl Al</td>
<td>1.6</td>
<td>8</td>
<td>7</td>
</tr>
<tr>
<td>Iprodione</td>
<td>2.5</td>
<td>8</td>
<td>8</td>
</tr>
</tbody>
</table>
What is claimed is:

1. A dry spreadable, water dispersible granular composition, comprising:
   a) from 35 to 95 weight percent of diatomaceous earth having an internal surface area in the range of from greater than about 5 to less than about 90 square meters per gram and a pore volume in the range of from greater than about 2 to less than about 5 cubic centimeters per gram; and
   b) from 5 to 40 weight percent of a surfactant composition comprising a surfactant selected from the group consisting of nonionics, anionics, and combinations thereof, wherein the weight percents are based on the total weight of the granular composition.

2. The dry spreadable, water dispersible granular composition of claim 1 wherein the surfactant composition comprises:
   i) from 3 to 15 weight percent of a high molecular weight nonionic or anionic surfactant dispersant.
ii) from 2 to 15 weight percent of a low molecular weight nonionic or anionic surfactant rewetting agent; and

iii) from 0 to 10 weight percent of a binder.

3. The dry spreadable, water dispersible granular composition of claim 2 wherein the binder comprises:

i) a high molecular weight nonionic or anionic surfactant dispersant, or

ii) a low molecular weight nonionic or anionic surfactant rewetting agent.

4. The dry spreadable, water dispersible granular composition of claim 2 wherein:

i) the surfactant dispersant is a high molecular weight anionic; and

ii) the surfactant rewetting agent is a low molecular weight anionic.

5. The dry spreadable, water dispersible granular composition of claim 2 wherein the anionic surfactant is selected from the group consisting of:

i) alkyl and alkyl ether sulfates;
ii) water soluble salts of the formula:
   \[ R_1 - S\text{O}_3^- - M \]
wherein \( R_1 \) is selected from the group consisting of
   a) a straight or branched chain,
   b) a mono-, di-, or tri- \( C_1 - C_6 \) alkyl substituted aryl;
   c) alpha-olefins having 12 to 24 carbon atoms; and
   d) naphthalene-formaldehyde condensation products;
   iii) isethionates;
   iv) N-methyl taurates;
   v) sulfo succinates;
   vi) sulfo succinamates;
   vii) B-alkoxy alkane sulfonates;
   viii) disulfonates;
   ix) alkal or alkali earth metal salts of lignosulfonates;
   x) ethoxylated derivatives of the above-identified anionic surfactants; and
   xi) mixtures thereof.
6. The dry spreadable, water dispersible granular composition of claim 2 wherein the nonionic surfactant is selected from the group consisting of amides, esters, ethoxylates, end-capped block copolymers, EO/PO block copolymers, and mixtures thereof.

7. The dry spreadable, water dispersible granular composition of claim 2 wherein the nonionic surfactant is selected from the group consisting of

   i) Alkanolamides of the formula -

      \[ \begin{array}{c}
         O \\
         / \\
        \end{array} \quad (CH_2-CH_2-O)_xH \\
        \begin{array}{c}
         / \\
        \end{array} \\
         R - C - N \\
        \begin{array}{c}
         \backslash \\
        \end{array} \\
         \begin{array}{c}
         \backslash \\
        \end{array} \\
         R'' \\
      \end{array} \]

   wherein R' and R'' each can be -H,
   -CH_2CH_2OH, or -CH_2 - CH - OH;

   ii) ethoxylated alkanolamides of the formula -

      \[ \begin{array}{c}
         O \\
         / \\
        \end{array} \quad (CH_2-CH_2-O)_yH \\
        \begin{array}{c}
         / \\
        \end{array} \\
         \begin{array}{c}
         \backslash \\
        \end{array} \\
         \begin{array}{c}
         \backslash \\
        \end{array} \\
         R - C - N \\
        \begin{array}{c}
         \backslash \\
        \end{array} \\
         (CH_2-CH_2-O)_yH; \]
iii) ethylene bisamides of the formula -

\[ \text{O} \]
\[
\begin{array}{cc}
R \quad & C \\
\downarrow & \quad \downarrow \\
N-CH_2-CH_2-N & \\
\downarrow & \downarrow \\
H & C-R \\
\end{array}
\]

iv) fatty acid esters of the formula -

\[ \text{O} \]
\[
R \quad C \quad O \quad \quad R_1;
\]

v) glycerol esters of the formula -

\[ \text{O} \]
\[
R \quad C \quad O \quad CH_2 \quad CH \quad CH_2 \quad O \quad \quad R_1
\]
\[ OH; \]

vi) ethoxylated fatty acid esters of the formula -

\[ \text{O} \]
\[
R \quad C \quad O \quad \{CH_2CH_2O\}_x - R_1;
\]

vii) sorbitan esters of the formula -
viii) ethoxylated sorbitan esters of the formula -

\[ \text{H} - (\text{OCH}_2\text{CH}_2)_n\text{O} - (\text{CH}_2\text{CH}_2\text{O})_x\text{H} \]

ix) alkylphenol ethoxylates of the formula -

\[ R - \quad \text{O} - (\text{OCH}_2\text{CH}_2)_n\text{OH}; \]

x) alcohol ethoxylates of the formula -

\[ R - \quad \text{O} - (\text{CH}_2\text{CH}_2\text{O})_n\text{H}; \]
xi) tristyrylphenol ethoxylates of the formula -

\[
\begin{align*}
\text{CH}, & \quad \text{CH} \\
\text{CH}_3 & \quad \text{CH} \\
\end{align*}
\]

xii) mercaptan ethoxylates of the formula -

\[
R - S - (\text{CH}_2\text{CH}_2\text{O})_n\text{H}
\]

xiii) alcohol alkoxylates of the formula -

\[
\begin{align*}
\text{CH}, & \quad \text{CH} \\
R - (\text{OCH}_2\text{CH}_2)_x - (\text{O} - \text{CH} - \text{CH}_2)_m - \text{OH} \\
\end{align*}
\]

xiv) ethylene oxide/propylene oxide block copolymers of the formula -

\[
\begin{align*}
\text{CH}, & \quad \text{CH} \\
\text{HO} - (\text{CH}_2\text{CH}_2\text{O})_x (\text{CH}_2\text{CH}_2\text{O})_m - (\text{CH}_2\text{CH}_2\text{O})_y - \text{H} \\
\end{align*}
\]
xv) copolymers of the formula -

\[
\begin{align*}
\text{CH}_3 & \quad \text{CH}_3 \\
& \quad \text{HO(\text{CH-CH}_2\text{O})_m - (\text{CH}_2\text{CH}_2\text{O})_x - (\text{CH}_2\text{-CH}_2\text{O})_1 \text{H}}
\end{align*}
\]

xvi) chlorine capped ethoxylates of the formula -

\[
\begin{align*}
R & = (\text{OCH}_2\text{CH}_2)_x\text{Cl}
\end{align*}
\]

xvii) tetra-functional block copolymers of the formula -

\[
\begin{align*}
\text{CH}_3 & \quad \text{CH}_3 \\
& \quad \text{H(OCH}_2\text{CH}_2)_x \quad - \quad (\text{OCHCH}_2)_m \quad - \quad (\text{CH}_2\text{CHO})_1 \quad - \quad (\text{CH}_2\text{CH}_2\text{O})_y \text{H}
\end{align*}
\]

or

\[
\begin{align*}
\text{CH}_3 & \quad \text{CH}_3 \\
& \quad \text{H(OCH}_2\text{CH}_2)_m \quad - \quad (\text{OCH}_2\text{CH}_2)_x \quad - \quad (\text{CH}_2\text{CHO})_1 \quad - \quad (\text{CH}_2\text{CH}_2\text{O})_y \text{H}
\end{align*}
\]

and

\[
\begin{align*}
\text{CH}_3 & \quad \text{CH}_3 \\
& \quad \text{H(OCH}_2\text{CH}_2)_1 \quad - \quad (\text{OCH}_2\text{CH}_2)_y \quad - \quad (\text{CH}_2\text{CHO})_1 \quad - \quad (\text{CH}_2\text{CH}_2\text{O})_1 \text{H}
\end{align*}
\]
xviii) Mixtures thereof;

wherein

R is a fatty alkyl group;
R₁ is -H or a fatty alkyl group;
x, x', y, y' and n are each independently moles of
ethylene oxide preferably 1 to 300; and
m, m', 1 and 1' are each independently moles of
propylene oxide, preferably 1 to 300.

8. The dry spreadable, water dispersible
granular composition of claim 1 wherein the
diatomaceous earth has an internal surface area in the
range of from greater than about 10 to less than about
60 square meters per gram and a volume in the range of
from greater than about 3 to less than about 4 cubic
centimeters per gram.

9. A dry spreadable, water dispersible
granular composition comprising:

a) from about 50 to 95 weight percent
diatomaceous earth having an internal surface area in
the range of from greater than about 5 to less than
about 90 square meters per gram and a pore volume in
the range of from greater than about 2 to less than about 5 cubic centimeters per gram;

b) from 3 to 10 weight percent alkynaphthalenesulfonate-formaldehyde condensate;

c) from 1 to 5 weight percent alkynaphthalenesulfonate; and

d) from 1 to 5 weight percent tristyrylphenol ethoxylate, all of the weight percents being based on the total weight percent of the granular composition.

10. The dry spreadable, water dispersible granular composition of claim 9 additionally comprising from about 1 to 6 weight percent of a nonionic, ethylene oxide/propylene oxide block copolymer.

11. A dry spreadable, water dispersible granular composition comprising:

a) 35-95 weight percent of diatomaceous earth having an internal surface area in the range of from greater than about 5 to less than about 90 square meters per gram and a pore volume in the range of from
greater than about 2 to less than about 5 cubic centimeters per gram;

b) from about 0 to 60 weight percent of a bioactive compound; and

c) from 5 to 40 weight percent of a surfactant composition comprising a surfactant selected from the group consisting of nonionics, anionics, and combinations thereof, wherein the weight percents are based on the total weight of the granular composition.

12. The dry spreadable, water dispersible granular composition according to claim 11 wherein the bioactive compound is a pesticide.

13. The dry spreadable, water dispersible granular composition according to claim 11 wherein the bioactive compound is a fertilizer.

14. The dry spreadable, water dispersible granular composition of claim 12 wherein the surfactant composition comprises:

i) from 3 to 15 weight percent of a high molecular weight nonionic or anionic surfactant dispersant;
ii) from 2 to 15 weight percent of a low molecular weight nonionic or anionic surfactant rewetting agent; and

iii) from 0 to 10 weight percent of a binder.

15. The dry spreadable, water dispersible granular composition of claim 14 wherein the binder comprises:

i) a high molecular weight nonionic or anionic surfactant dispersant, or

ii) a low molecular weight nonionic or anionic surfactant rewetting agent.

16. The dry spreadable, water dispersible granular composition of claim 14 wherein:

i) the surfactant dispersant is a high molecular weight anionic; and

ii) the surfactant rewetting agent is a low molecular weight anionic.

17. The dry spreadable, water dispersible granular composition of claim 14 wherein the anionic surfactant is selected from the group consisting of:

i) alkyl and alkyl ether sulfates;
ii) water soluble salts of the formula:
   \[ R_1^{-} \cdot SO_3^{-} \cdot M \]
   wherein \( R_1 \) is selected from the group consisting of
   a) a straight or branched chain,
   5  saturated aliphatic hydrocarbon radical having from 8 to 24 carbon atoms;
   b) a mono-, di-, or tri- \( C_1 \) - \( C_6 \) alkyl substituted aryl;
   c) alpha-olefins having 12 to 24 carbon atoms; and
   d) naphthalene-formaldehyde condensation products;
   iii) isethionates;
   iv) N-methyl taurates;
   15  v) sulfosuccinates;
   vi) sulfosuccinamates;
   vii) B-alkoxy alkane sulfonates;
   viii) disulfonates;
   ix) alkali or alkali earth metal salts of lignosulfonates;
   20  x) ethoxylated derivatives of the above-identified anionic surfactants; and
   xi) mixtures thereof.
18. The dry spreadable, water dispersible granular composition of claim 14 wherein the nonionic surfactant is selected from the group consisting of amides, esters, ethoxylates, end-capped block copolymers, EO/PO block copolymers, and mixtures thereof.

19. The dry spreadable, water dispersible granular composition of claim 14 wherein the nonionic surfactant is selected from the group consisting of

i) Alkanolamides of the formula -

\[
\begin{array}{c}
O \\
\parallel / \\
R - C - N \\
\downarrow \\
R' \\
\end{array}
\]

wherein R' and R" each can be -H, -CH₂CH₃OH, or -CH₂ - CH - OH;

ii) ethoxylated alkanolamides of the formula -

\[
\begin{array}{c}
O \\
\parallel / \\
R - C - N \\
\downarrow \\
(\text{CH}_2 - \text{CH}_2 - \text{O})_x \text{H} \\
\end{array}
\]
iii) ethylene bisamides of the formula -
\[
\text{O} \\
R - C - H \\
\text{N-CH}_2\text{-CH}_2\text{-N} \\
\text{O} \\
\]
iv) fatty acid esters of the formula -
\[
\text{O} \\
R - C - O - R_1; \\
\]
v) glycerol esters of the formula -
\[
\text{O} \\
R - C - O - \text{CH}_2 - \text{CH} - \text{CH}_2 - O - R_1 \\
\text{OH}; \\
\]
vi) ethoxylated fatty acid esters of the formula -

\[
R - \overset{\text{O}}{\text{C}} - O - (\text{CH}_2\text{CH}_2\text{O})_x - R_1;
\]

vii) sorbitan esters of the formula -

\[
\begin{array}{c}
\text{HO} \\
\text{O} \\
\text{CH} - \text{CH}_2 - O - \overset{\text{O}}{\text{C}} - R; \\
\text{OH}
\end{array}
\]

viii) ethoxylated sorbitan esters of the formula -

\[
M - (\text{OCH}_2\text{CH}_2)_n - O - (\text{CH}_2\text{CH}_2\text{O})_x - H
\]

\[
\begin{array}{c}
\text{O} \\
\text{CH}_2 - \overset{\text{O}}{\text{C}} - R \\
\text{O} - (\text{CH}_2\text{CH}_2\text{O})_y - H;
\end{array}
\]
ix) alkylphenol ethoxylates of the formula -

\[
R - \overset{\text{R}}{\text{O}} - (\text{OCH}_2\text{CH}_2)_n\text{OH};
\]

x) alcohol ethoxylates of the formula -

\[
R - \overset{\text{O}}{\text{O}} - (\text{CH}_2\text{CH}_2\text{O})_n\text{H};
\]

xi) tristyrylphenol ethoxylates of the formula -

\[
\overset{\text{CH}_3}{\text{CH}} - \overset{(\text{O CH}_2\text{CH}_2)_n\text{OH}}{\text{CH}} - \overset{\text{CH}_3}{\text{CH}} - \overset{\text{CH}_3}{\text{CH}};
\]

xii) mercaptan ethoxylates of the formula -

\[
R - \overset{\text{S}}{\text{S}} - (\text{CH}_2\text{CH}_2\text{O})_n\text{H};
\]
xiii) alcohol alkoxylates of the formula:

\[
\text{CH}_3 \quad \text{R-} \text{(OCH}_2\text{CH}_2\text{)}_x \cdot \text{(O - CH - CH}_2\text{)}_m \cdot \text{OH};
\]

xiv) ethylene oxide/propylene oxide block copolymers of the formula:

\[
\text{CH}_3 \quad \text{HO-} \text{(CH}_2\text{CH}_2\text{O)}_x \cdot \text{(CH}_2\text{-CH-O)}_m \cdot \text{(CH}_2\text{CH}_2\text{O)}_y \cdot \text{H};
\]

xv) copolymers of the formula:

\[
\text{CH}_3 \quad \text{CH}_3 \quad \text{HO(} \text{CH-CH-O)}_m \cdot \text{(CH}_2\text{CH}_2\text{O)}_x \cdot \text{(CH}_2\text{-CH-O)}_1 \cdot \text{H};
\]

xvi) chlorine capped ethoxylates of the formula:

\[
\text{R-} \text{(OCH}_2\text{CH}_2\text{)}_x \text{Cl}; \text{ and}
\]

xvii) tetra-functional block copolymers of the formula:

\[
\text{CH}_3 \quad \text{CH}_3 \quad \text{H(} \text{OCH}_2\text{CH}_2\text{)}_x \cdot \text{(OCHCH}_2\text{)}_m \cdot \text{(CH}_2\text{CH-O)}_m \cdot \text{(CH}_2\text{CH}_2\text{O)}_x \cdot \text{H}
\]

\[
\text{NCH}_2\text{CH}_3\text{N}
\]

\[
\text{CH}_3 \quad \text{CH}_3 \quad \text{H(} \text{OCH}_2\text{CH}_2\text{)}_y \cdot \text{(OCHCH}_2\text{)}_1 \cdot \text{(CH}_2\text{CHO)}_1 \cdot \text{(CH}_2\text{CH}_2\text{O)}_y \cdot \text{H}
\]

\[
\text{CH}_3 \quad \text{CH}_1
\]
or

\[
\begin{align*}
5 & \quad \text{CH}_3 \\
& \quad (\text{OCH}_2 \text{CH}_2)_m \quad - \quad (\text{OCH}_2 \text{CH}_2)_x \\
& \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quan
21. A dry spreadable, water dispersible granular composition comprising:
   a) from about 50 to 95 weight percent diatomaceous earth having an internal surface area in
   the range of from greater than about 5 to less than about 90 square meters per gram and a pore volume in
   the range of from greater than about 2 to less than about 5 cubic centimeters per gram;
   b) from 0.05 to 30 weight percent of a pesticide;
   c) from 3 to 15 weight percent alkynaphthalenesulfonate-formaldehyde condensate;
   d) from 3 to 15 weight percent alkynaphthalenesulfonate; and
   e) from 2 to 10 weight percent tristyrylphenol ethoxylate, all of the weight percents being based on the total weight percent of the composition.

22. The dry spreadable, water dispersible granular composition of claim 21 additionally comprising from 1 to 6 weight percent of a nonionic, ethylene oxide/propylene oxide block copolymer.
INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 A01N25/14 A01N25/30

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic database consulted during the international search (name of database and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
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<th>Relevant to claim No.</th>
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<td>DATABASE WPI&lt;br&gt;Week 9309&lt;br&gt;Derwent Publications Ltd., London, GB; AN 93-070986&lt;br&gt;XP002051078&lt;br&gt;&amp; JP 05 017 305 A (SDS BIOTECH. CORP.), 26 January 1993</td>
<td>1-5, 8, 11-17, 20</td>
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<td>EP 0 689 866 A (RHONE-POULENC CHIMIE) 3&lt;br&gt;January 1996&lt;br&gt;see the whole document&lt;br&gt;---</td>
<td>6, 7, 9, 10, 18, 19, 21, 22</td>
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Date of the actual completion of the international search: 23 December 1997

Date of mailing of the international search report: 14/01/1998

Name and mailing address of the ISA:
European Patent Office, P.B. 5818 Patentlaan 2<br>NL-2280 HV Rijswijk<br>Tel. (+31-70) 340-2040, Tx. 31 651 apnl, Fax: (+31-70) 340-3016

Authorized officer: Muellners, W
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