This invention relates to improved lubricating greases thickened with finely divided solids, and more particularly to such greases containing small amounts of formamide, and to methods of preparing such greases.

We have found, in the preparation of solids thickened greases, that a small amount of formamide present in the grease mixture during the homogenization process assists in the development of the gel structure and very materially improves the yield and stability of the product. This effect obtained by the use of formamide is unexpectedly very much greater than that obtained with various other liquids such as water, which are commonly employed in solids thickened greases in small amounts to assist in the gel formation.

The solids thickened greases which are benefited by the presence of small amounts of formamide in accordance with this invention are those wherein the solid thickening agent is a finely divided polar material, such as, for example, silica and certain naturally occurring siliceous materials such as bentonite, organo-clays obtained by treating bentonite with amine of various types, organic pigments of various types such as phthalocyanines, indigo, etc., having a measurable surface polarity, and metal oxides of various types such as zinc oxide, magnesium oxide, aluminum oxide, etc., having the property of thickening lubricating oils when they are dispersed therein in finely divided form in suitable proportions. Such materials are suitably employed in the form of particles below about 5 microns in diameter, and usually having a diameter below about 2 microns in diameter, although the most suitable particle size and form will vary somewhat with the type of material employed.

In general, it is found that the effectiveness of formamide as a yield improving agent increases with increasing polarity of the solid thickening agent. However, special advantages are obtained in certain cases with thickening agents of only relatively low polarity. For example, in the preparation of organo-clay thickened greases employing the so-called "in situ" process, wherein the organo-clay is formed by treating bentonite with an amine or an amine salt in the presence of lubricating oil comprising all or a portion of the oleaginous liquid component of the grease, it has been found that an unexpected yield advantage is obtained when the formamide is present during the organo-clay reaction. Accordingly, the preparation of such greases wherein an organo-clay is formed in situ in the presence of the formamide, is regarded as comprising a preferred embodiment of this invention.

The greases of this invention comprise an oleaginous liquid as the chief component, containing about 0.05 to about 5.0 percent of formamide, based on the weight of the grease, and a finely divided solid of the character described above in a sufficient amount to thicken the lubricating oil to a grease consistency with the aid of the formamide which is present. Ordinarily the amount of solid thickening agent is present in amounts from about 5 to about 45 percent by weight, based on the weight of the grease, and preferably in amounts from about 7 to about 25 percent by weight, according to the thickening action of the solid and the grease consistency desired. The formamide is preferably employed in an amount between about 0.1 and 3.0 percent of the weight of the grease composition.

The lubricating oils employed in these greases may be any suitable oils of lubricating characteristics ordinarily employed in lubricating greases, including the conventional mineral lubricating oils, synthetic oils obtained by various refining processes, such as cracking and polymerization, and other synthetic oleaginous compounds, such as high molecular weight ethers, esters, silicones, etc. Suitable mineral oils include paraffinic and naphthenic oils having viscosities in the range from about 80 seconds Saybolt Universal at 100° F. to about 225 seconds Saybolt Universal at 210° F., and are preferably those having viscosities in the range from about 100 to about 600 seconds Saybolt Universal at 100° F.

Additives of the usual types employed in solids thickened greases, including oxidation inhibitors, rust and corrosion inhibitors, extreme pressure agents, etc., may be employed in these greases where no antagonistic effect results between such additive and the formamide. Such materials are employed in minor amounts, such as from about 0.05 to about 10.0 percent by weight, and preferably in amounts from about 0.5 to about 5.0 percent by weight of the grease. A very suitable class of oxidation inhibitors are the aromatic amines, such as, for example, diphenylamine, alpha- and beta-naphthylamines and para-phenylenediamine. Very advantageously a compound of the class of higher aliphatic amines and amides, wherein an aliphatic hydrocarbon group contains about 10-25 carbon atoms, is employed in the composition in order to improve the water resistance and other properties. We have found that formamide offsets or overcomes the yield depressing effect commonly resulting from the use of compounds of this type in solids thickened greases to a sufficient extent that solids thickened greases having good water resistant properties are obtained in satisfactory yields by employing both higher aliphatic amines or amides and a small amount of formamide in the composition.

As an example of a preferred embodiment of this invention, greases were prepared containing a surface treated bentonite as the thickening agent and containing a small amount of formamide, which was present during the bentonite-amine reaction. The preparation of the solid thickening agent was carried out by treating a micron sized bentonite with a commercial quaternary amine salt mixture, comprising about 75 percent by weight of dioctadecyl- and dihexadecyl-dimethyl ammonium chlorides in a weight ratio of 70:30, respectively, in isopropyl alcohol. The reaction was carried out in the presence of the mineral lubricating oil employed in the grease, employing stoichiometric amounts of the amine salts and bentonite on the basis of the measured base exchange capacity of the bentonite. A typical grease preparation was carried out in the following manner: 113 grams of the commercial amine salt mixture was mixed with 2,112 grams of a refined paraffinic residual oil having a gravity, API of 26.0, a Saybolt Universal viscosity at 100° F. of 977 seconds and a viscosity index of about 80, the mixture heated to 250° F. and circulated in a Manton-Gaulin homogenizer at 5000 pounds pressure dif-
ferential while 200 grams of bentonite and then 4.8 grams of formamide were added. The circulation was continued for 45 minutes. The following table shows the yield and stability of the product thus obtained as compared with a grease prepared in the identical way except that 4.8 grams of water was added instead of the formamide.

### Table I

<table>
<thead>
<tr>
<th>Composition, Wt. Percent, Calculated:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Organin-85°</td>
</tr>
<tr>
<td>Sodium chloride</td>
</tr>
<tr>
<td>Water</td>
</tr>
<tr>
<td>Formamide</td>
</tr>
<tr>
<td>Mineral lubricating oil</td>
</tr>
<tr>
<td>Penetration ASTM, at 77° F./Unworked</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>Worked—60 strokes</td>
</tr>
<tr>
<td></td>
</tr>
</tbody>
</table>

As shown by the above table, a stable grease was obtained in satisfactory yield by employing 0.2 weight percent of formamide in conjunction with the organo treated bentonite as the thickening agent. On the other hand, the same amount of water was ineffective to produce a stable grease structure in this composition. By the use of larger amounts of water, up to about 1 percent, substantial yield improvements were obtained in greases of this type, but these were still considerably lower than those obtained with equivalent or lower amounts of formamide.

Table II below shows the effect of formamide in improving the yield in a different solids thickened grease, and also shows its effect in offsetting the yield depressing effect of higher aliphatic amines and amides which were employed to improve the water resistant properties of the grease. The greases comprised 15.0 percent by weight of finely divided silica, having an average particle size of 0.022 micron and a surface area of 160–175 square meters per gram, in a refined naphthenic base oil having a Saybolt Universal viscosity at 100° F. of about 310 seconds. The preparation was carried out by mixing together the solid thickener and additives with the lubricating oil at room temperature and finishing by milling in a Premier colloid mill with two passes at 0.002 inch clearance.

### Table II

<table>
<thead>
<tr>
<th>Additive</th>
<th>Penetration ASTM, at 77° F.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unworked</td>
<td>240</td>
</tr>
<tr>
<td>Worked—60 strokes</td>
<td>150</td>
</tr>
<tr>
<td>Worked—100,000 strokes</td>
<td>300</td>
</tr>
</tbody>
</table>


As shown by the data given in the tables, very materially improved yields are obtained by employing small amounts of formamide in greases thickened with polar solids of various types, including such greases containing higher aliphatic amines and amides having a yield depressing effect. In addition, the greases containing formamide have greatly improved working stability, as shown by the smaller amount of change which occurred in the greases of the examples upon working 10,000 or 100,000 strokes in the ASTM worker tester.

Obviously many modifications and variations of the invention, as hereinbefore set forth, may be made without departing from the spirit and scope thereof and only such limitations should be imposed as are indicated in the appended claims.

We claim:

1. A lubricating grease consisting essentially of a major amount of lubricating oil, about 35–45 percent by weight of a finely divided polar solid having the property of forming gels with lubricating oils, selected from the class consisting of bentonites which have been surface treated with higher aliphatic amines, silica and oxides of zinc, magnesium and aluminum, and about 0.50–5.0 percent by weight of formamide, said finely divided polar solid being present in an amount sufficient to thicken the composition to a grease consistency.

2. A lubricating grease according to claim 1 wherein the formamide is present in an amount of 0.1–3.0 percent by weight.

3. A lubricating grease according to claim 1 wherein the said finely divided polar solid is bentonite which has been treated with a compound chosen from the class consisting of higher aliphatic amines and salts thereof.

4. A lubricating grease according to claim 1 wherein the said finely divided polar solid is silica.

5. A lubricating grease according to claim 1 wherein the said polar solid is zinc oxide.

6. A lubricating grease according to claim 1 wherein the said grease contains about 0.05–10.0 percent by weight, sufficient to improve its water resistant properties, of a compound selected from the class consisting of higher aliphatic amines and amides.

7. A lubricating grease according to claim 1 wherein the said amides are alkylamides of higher fatty acids.

8. A lubricating grease consisting essentially of a major amount of mineral lubricating oil, about 7–25 percent by weight, sufficient to thicken the composition to a grease consistency, of finely divided silica, about 0.1–3.0 percent by weight of formamide and about 0.5–5.0 percent by weight of a compound chosen from the group consisting of higher aliphatic amines and amides.

9. A lubricating grease according to claim 8 containing 0.5–5.0 percent by weight of N,N-di-(betahydroxyethyl)-12-hydroxy stearamide.

10. A lubricating grease according to claim 8 containing 0.5–5.0 percent by weight of a compound having the formula RNHC6H4CH2, where R is an aliphatic hydrocarbon group containing 10–25 carbon atoms.

11. The method of grease preparation which comprises providing a mixture consisting essentially of a lubricating oil, about 5–45% by weight based on the weight of the mixture of a finely divided insoluble high melting solid having a measurable surface polarity and having the prop-

### Table III

<table>
<thead>
<tr>
<th>Additives</th>
<th>Penetration at 77° F.</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5% Formamide</td>
<td>200 strokes</td>
</tr>
<tr>
<td>Liquid</td>
<td>341</td>
</tr>
</tbody>
</table>

- 30% Penetration at 80 strokes
- Too soft for penetration

Table III Penetration at 77° F.

<table>
<thead>
<tr>
<th>Additives</th>
<th>Liquid</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5% Formamide</td>
<td>341</td>
</tr>
</tbody>
</table>

[note: Table III is repeated in the text for emphasis]
2,986,517

5

ert of forming gels in lubricating oils, and about 0.05–5% by weight of formamide, and subjecting the said mixture to shearing until a grease consistency is obtained.

12. The method of grease preparation which comprises heating together a mixture of lubricating oil, a minor amount of a compound chosen from the class consisting of higher aliphatic amines and salts thereof, about 5–45% by weight of bentonite and about 0.05–5.0% by weight of formamide and shearing the said mixture until a grease consistency is obtained.

References Cited in the file of this patent

UNITED STATES PATENTS


FOREIGN PATENTS

735,711 Great Britain __________ Aug. 24, 1955
UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 2,986,517
May 30, 1961

Norman R. Odell et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 4, line 35, for "0.50-5.0" read -- 0.05-5.0 --.

Signed and sealed this 17th day of October 1961.

(SEAL)
Attest:

ERNEST W. SWIDER
Attesting Officer

DAVID L. LADD
Commissioner of Patents
USCOMM-DC