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3,291,736

GREASE COMPOSITIONS CONTAINING ALKYL SUCCINIC PARTIAL ESTERS

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9 Claims. (Cl. 252-49.6)

This invention relates to grease compositions and, in one of its aspects, the invention relates to grease compositions possessing non-staining rust inhibiting and improved structural properties. More particularly in this aspect, the invention relates to grease compositions possessing non-staining rust inhibiting and improved structural properties, employing certain surface-active additive compounds capable of imparting such characteristics to greases.

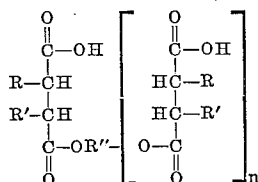
Surface-active compounds have been found to exert two types of effects on grease compositions. One type involves structure modification by changing the thickener surface or the interaction between the thickening agent and the lubricating vehicle. Such additives function as structure improvers, stringiness agents, and the like. The other type of effect is on the interaction between grease and metal surfaces. This grease-metal interaction is manifested in the form of corrosion inhibition, or as lubricity improvement, anti-wear, and other effects. Heretofore, various materials have been employed as rust inhibitors in grease compositions, or as structure improvers, making it possible to lower the concentration of the thickening agent. No attempt, however, has been made to incorporate into a grease formulation, a single additive material which is effective both as a rust inhibiting agent and as a structure improver. This is particularly applicable with respect to pigment thickened silicone grease compositions. Hence, prior to the present invention, no single additive material has been incorporated in grease formulations, which effectively imparts rust inhibiting and improved structural properties.

It is, therefore, an object of the present invention to provide new and improved grease compositions containing a single additive which imparts rust inhibiting and improved structural properties.

Another object of the invention is to provide new and improved pigment-thickened silicone grease compositions containing a single additive which imparts rust inhibiting and improved structural properties.

Other objects and advantages inherent in the invention will become apparent to those skilled in the art from the following detailed description.

In accordance with the present invention new and improved grease compositions are provided, exhibiting the aforementioned properties of rust inhibition and improved structure, which contain small amounts of a partial ester having the general formula:



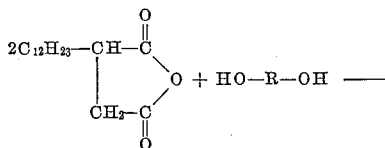
wherein of R and R' one is hydrogen, and the other is selected from the group consisting of alkyl and alkenyl

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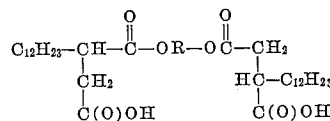
radicals containing from 8 to 18 carbon atoms and preferably from about 9 to about 15 carbon atoms, R'' is a residue of a polyhydric alcohol and preferably a residue of a polyhydric alcohol containing from 2 to 6 carbon atoms, and n is an integer from 1 to 4.

These corrosion inhibitors, as represented by the above-indicated general formula, are partial esters of an alkyl or alkenylsuccinic anhydride and, preferably, are the products obtained by the reaction of one molar equivalent of a polyhydric alcohol with two molar equivalents of the anhydride. The residue of the polyhydric alcohol, as represented by R'' may, generically, contain oxygen, sulfur or nitrogen in the chain as well as other substituents. The integer n represents a whole number from 1 to 4 to indicate that within the scope of the inhibitor compound are included the derivatives of diols, triols and other polyhydroxy compounds.

Illustrative of a typical procedure for the preparation of the inhibitor compounds employed in the novel grease formulations of the present invention, is the procedure in which the desired substituted succinic anhydride and alcohol charges are weighed into a suitable jacketed acid-resisting vessel equipped with agitation, temperature recording means and a reflux condenser. This charge is heated at a temperature from about 95° C. to about 100° C., while agitating, and held at this temperature, while the progress of the reaction is observed periodically by withdrawing a sample and determining the neutralization number thereof. Heating is continued until the desired neutralization number is obtained, or until it remains substantially constant. The resulting reaction is represented as follows:



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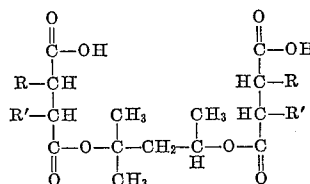
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A more detailed description of the preparation and properties of the above described partial esters comprising the corrosion inhibitors present in the novel grease formulations of the present invention, are described in Patent 3,117,091, issued January 7, 1964, and the disclosure thereof is incorporated in the present application, by reference. A particularly outstanding application of the use of the aforementioned partial esters, is their incorporation in silicone-containing greases, as corrosion inhibitors, in which small amounts of a partial ester having the following general formula, is incorporated in such greases:

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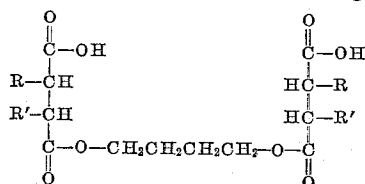
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wherein of R and R' one is hydrogen, and the other is a branched chain dodecyl group.

3

Another outstanding application of the use of the aforementioned partial esters, is their incorporation in silicone-containing greases, as corrosion inhibitors, in which small amounts of a partial ester having the following general formula, is incorporated in such greases:



wherein of R and R' one is hydrogen, and the other is a branched chain dodecenyl group.

The outstanding benefit realized by incorporating the above-described partial esters in grease formulations, resides in the unusual property that these partial esters not only impart corrosion inhibiting properties to the grease, making possible the lowering of the concentration of the thickening agent, and at the same time providing good storage stability. Coupled with these important properties is the additional characteristic of these partial esters to act as a reservoir for the oil vehicle, thereby enhancing the prolonged efficacy and life of the grease composition under the conditions of use.

In preparing the improved grease compositions of the present invention, the aforementioned partial ester inhibiting agents are combined with a grease-forming quantity of the thickening agent, and are present in the grease composition in an amount from about 0.01 to about 20 percent, by weight, and preferably in an amount from about 0.05 to about 5 percent, by weight, of the total composition. A wide variety of materials may be employed as the thickening or gelling agent. These may include any of the conventional metal salts or soaps, which are dispersed in the lubricating vehicle in grease-forming quantities, in such degree as to impart to the resulting grease composition, the desired consistency. Other thickening agents that may be employed in the grease formulations of the present invention in conjunction with the aforementioned partial esters, comprise the non-soap thickeners, such as surface-modified clays and silicas, aryl ureas, calcium complexes and similar materials. In general, grease thickeners may be employed which do not melt and dissolve when used at relatively high temperatures; however, in all other respects, any material which is normally employed for thickening hydrocarbon fluids for forming greases can be used in preparing the improved greases of this invention.

A particularly outstanding type of thickening agents are pigment thickeners comprising perylimid compounds. Of special importance are the p-methoxy perylimid compounds, and more specifically p-methoxyphenyl-perylimid, when employed as a pigment thickening agent in combination with the aforementioned rust inhibiting partial esters. The preparation of perylimid greases, having incorporated therein thickening agents of the aforementioned perylimid types, are described in detail in Patent 3,126,341 issued May 24, 1964, and the disclosure thereof is incorporated in the present application, by reference. Such perylimid greases, having incorporated therein the partial ester corrosion inhibitors of the present invention, have outstanding thermal stability for high temperature operations, and are capable of functioning at temperatures as high as about 600° F. It should be noted, however, that various other types of thickening agents may be employed, in forming the novel greases of the present invention, if so desired.

The lubricating vehicle employed in the novel greases of the present invention, may comprise any of the conventional hydrocarbon oils of lubricating viscosity, and may include mineral or synthetic lubricating oils, aliphatic phosphates, esters and diesters, silicates, siloxanes and oxalkyl ethers and esters. Mineral lubricating oils,

4

employed as the lubricating vehicle, may be of any suitable lubricating viscosity ranging from about 45 SSU at 100° F., to about 6000 SSU at 100° F., and preferably from about 50 to about 250 SSU at 210° F. These oils may have viscosity indexes, varying from below 0 to about 100 or higher. Viscosity indexes from about 70 to about 95 are preferred. The average molecular weights of these oils range from about 250 to about 800. The lubricating oil is employed in the grease composition in an amount sufficient to constitute the balance of the total composition.

As previously indicated, the oil vehicles employed in the novel greases of the present invention may comprise mineral or synthetic oils of lubricating viscosity. When high temperature stability is not a requirement of the finished grease, mineral oils having a viscosity of at least 40 SSU at 100° F., and particularly those within the range of about 60 SSU to about 6000 SSU at 100° F. may be employed. In instances, where synthetic vehicles are employed rather than mineral oils, or in combination therewith, as the lubricating vehicle, various compounds of this type may be successfully utilized. Typical synthetic vehicles include: polypropylene, polypropylene glycol, trimethylol propane esters, neopentyl and pentaerythritol esters, di-(2-ethyl hexyl) sebacate, di-(2-ethyl hexyl) adipate, di-butyl phthalate, polyethylene glycol di-(2-ethyl hexoate), fluorocarbons, silicate esters, silanes, esters of phosphorus-containing acids, liquid ureas, ferrocene derivatives, hydrogenated mineral oils, chain type polyphenyls, siloxanes and silicones (polysiloxanes), alkyl-substituted diphenyl ethers typified by a butyl-substituted bis-(p-phenoxy phenyl) ether, phenoxy phenyl ethers, etc. Particularly preferred, herein, are polysiloxanes, such as polymethylphenyl siloxanes, and m-bis-(m-phenoxyphenoxy) benzene.

The following examples will serve to illustrate the improved grease compositions of the present invention and their properties, as evidenced by respective bearing protection and 60 stroke worked penetration tests for the grease formulations therein described. The bearing protection against rust tests employed is identified as D1743-60T, in ASTM Standards on Petroleum Products, 1961. The penetration test employed is identified as D217-52, in ASTM Standards of Petroleum Products, 1961.

Example I

A lubricating grease composition was prepared by admixing 235 parts by weight of p-methoxyphenyl perylimid pigment, 60 parts by weight of denatured ethanol and 765 parts by weight of a polymethylphenyl siloxane. This polysiloxane had a molar ratio of methyl to phenyl groups of 20:1, molecular weight of approximately 4000 (as determined by vapor osmometry) and the following kinematic viscosity:

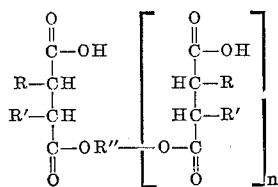
Temperature, ° F.:	Centistokes
@ 210 -----	30
@ 100 -----	80
@ -65 -----	2000

The components of the aforementioned mixture were paddled in a grease kettle, with the temperature being raised to 230° F. in three hours. The resulting grease was then homogenized in a colloid mill through a clearance of 0.002 inch. This resulting grease composition was found to have a soft appearance, and a 60 stroke worked penetration according to the aforementioned ASTM method D217-52, of 350. The grease was found to fail the bearing protection test against rusting, in accordance with the aforementioned ASTM D1743-60T.

Example II

A second lubricating grease composition was prepared in a manner similar to the procedure employed in Example I, except that 783.5 parts by weight of the polysiloxane of Example I were employed and 26.5 parts by

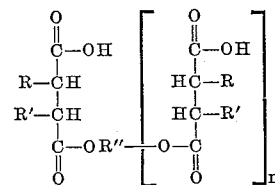
1. A grease composition containing a silicone as a lubricating vehicle; a thickening agent; and as a corrosion inhibitor, a small amount of a partial ester having the general formula:



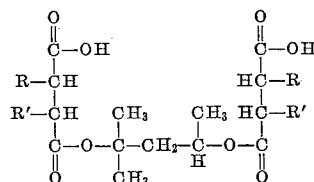
3. The grease composition of claim 1 wherein said partial ester is present in an amount, by weight, from about 0.05 to about 5 percent of the total composition. 75

$$\begin{array}{c} \text{O} \\ \parallel \\ \text{C}-\text{OH} \\ | \\ \text{R}-\text{CH} \\ | \\ \text{R}'-\text{CH} \\ | \\ \text{C}-\text{OR}'' \\ \parallel \\ \text{O} \end{array} \left[\begin{array}{c} \text{O} \\ \parallel \\ \text{C}-\text{OH} \\ | \\ \text{HC}-\text{R} \\ | \\ \text{HC}-\text{R}' \\ | \\ \text{C}-\text{O} \\ \parallel \\ \text{O} \end{array} \right]_n$$

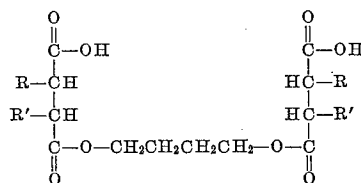
5. A grease composition containing a silicone as a lubricating vehicle; a thickening agent; and as a corrosion inhibitor, a small amount of a partial ester having the general formula:



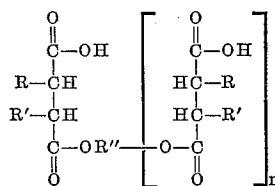
6. A grease composition containing a silicone as a lubricating vehicle; a thickening agent; and as a corrosion inhibitor, a small amount of a partial ester having the general formula:



7. A grease composition containing a silicone as a lubricating vehicle; a thickening agent; and as a corrosion inhibitor, a small amount of a partial ester having the general formula:



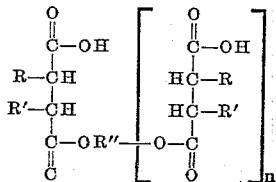
8. A grease composition containing a polysiloxane, as a lubricating vehicle; a perylimid, as a thickening agent; and, as a corrosion inhibitor, a small amount of a partial ester having the general formula:



7

wherein of R and R' one is hydrogen, and the other is selected from the group consisting of alkyl and alkenyl radicals containing from 8 to 18 carbon atoms, R'' is a residue of a polyhydric alcohol, and n is an integer from 1 to 4.

9. A grease composition containing polymethylphenylsiloxane, as a lubricating vehicle; p-methoxyphenyl perylimid, as a thickening agent; and, as a corrosion inhibitor, a small amount of a partial ester having the general formula:



8

wherein of R and R' one is hydrogen, and the other is selected from the group consisting of alkyl and alkenyl radicals containing from 8 to 18 carbon atoms, R'' is a residue of a polyhydric alcohol containing from 2 to 6 carbon atoms, and n is an integer from 1 to 4.

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DANIEL E. WYMAN, *Primary Examiner*.

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UNITED STATES PATENT OFFICE

CERTIFICATE OF CORRECTION

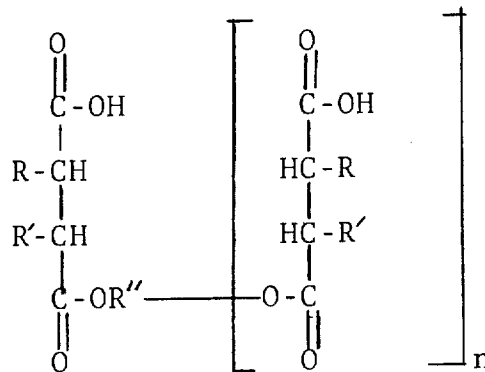
Patent No. 3,291,736

December 13, 1966

Fritz A. Buehler 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 7, lines 11 to 20, the formula should appear as shown below instead of as in the patent:



This certificate supersedes the certificate of correction issued September 19, 1967.

Signed and sealed this 22nd day of October 1968.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

EDWARD J. BRENNER
Commissioner of Patents