United States Patent

[11] 3,609,079

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[21] Appl. No. 783,537
[22] Filed Dec. 13, 1968
[45] Patented Sept. 28, 1971

[54] **SILICONE LUBRICANTS**
6 Claims, 6 Drawing Figs.

[52] U.S. Cl. **252/46.3,** 252/49.6, 252/32.7
[51] **Int. Cl.** **C10m 1/52,** C10m 1/50, C10m 1/38
[50] **Field of Search** .... 252/46.3, 45, 47, 47.5; 260/302 SD

[56] **References Cited**

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OTHER REFERENCES

Devine et al. " Molybdenum Disulfide Diester Lubricating Greases," NLGI Spokesman, 320- 326, Jan. 1964

Primary Examiner—Daniel E. Wyman
Assistant Examiner—W. Cannon
Attorneys—E. J. Brower and A. W. Collins

ABSTRACT: Lubricant compositions having increased lubricity are obtained by forming a mixture of a major amount of a silicone lubricant and a minor amount of a sulfur containing compound dissolved in the silicone and including at least two sulfur atoms per molecule. According to a preferred form of the invention, from 1 to 10 parts by weight of di-tertiary-nonyl polysulfide are admixed with from 99—90 parts by weight of a silicone oil to provide a new composition which functions properly as a lubricant under loads as high as 240 kilograms when employing the Shell 4-Ball EP machine. Without the polysulfide additive, the silicone oil is not useful as a lubricant under loads exceeding about 40 kilograms when employing the Shell 4-Ball EP machine.

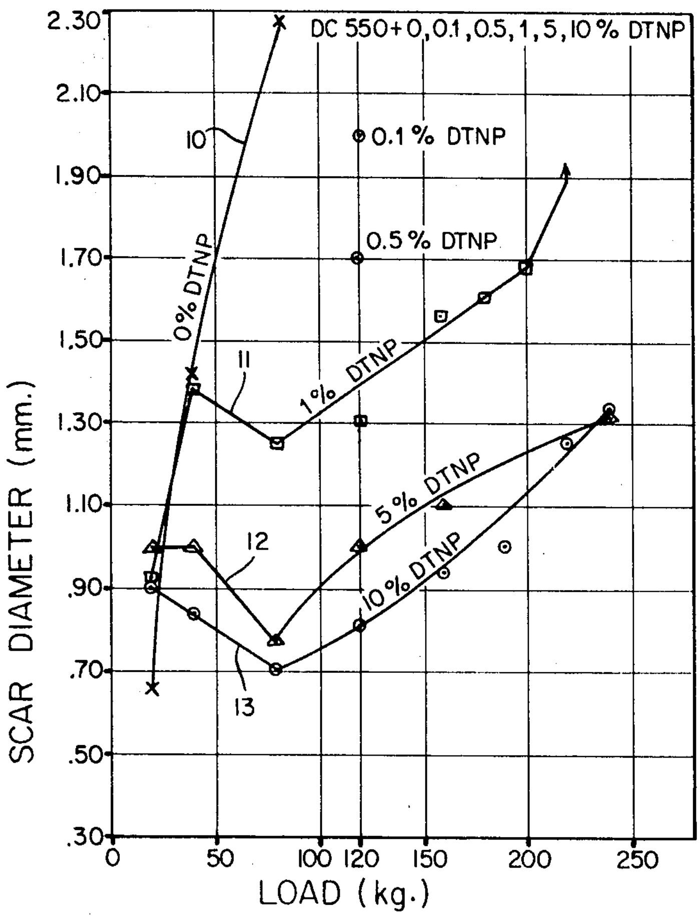

DC 550+0,0.1,0.5,1,5,10% DTNP
0.1% DTNP
0.5% DTNP
0% DTNP
1% DTNP
5% DTNP
10% DTNP
10
11
12
13
SCAR DIAMETER (mm.)
LOAD (kg.)

Fig. 1

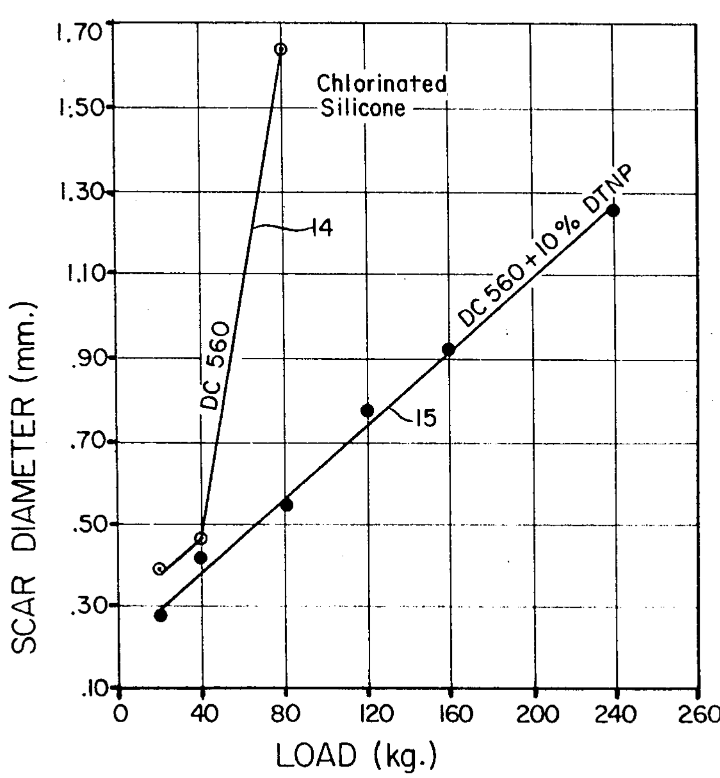

Chlorinated Silicone
DC 560
DC 560+10% DTNP
14
15
SCAR DIAMETER (mm.)
LOAD (kg.)

Fig. 2

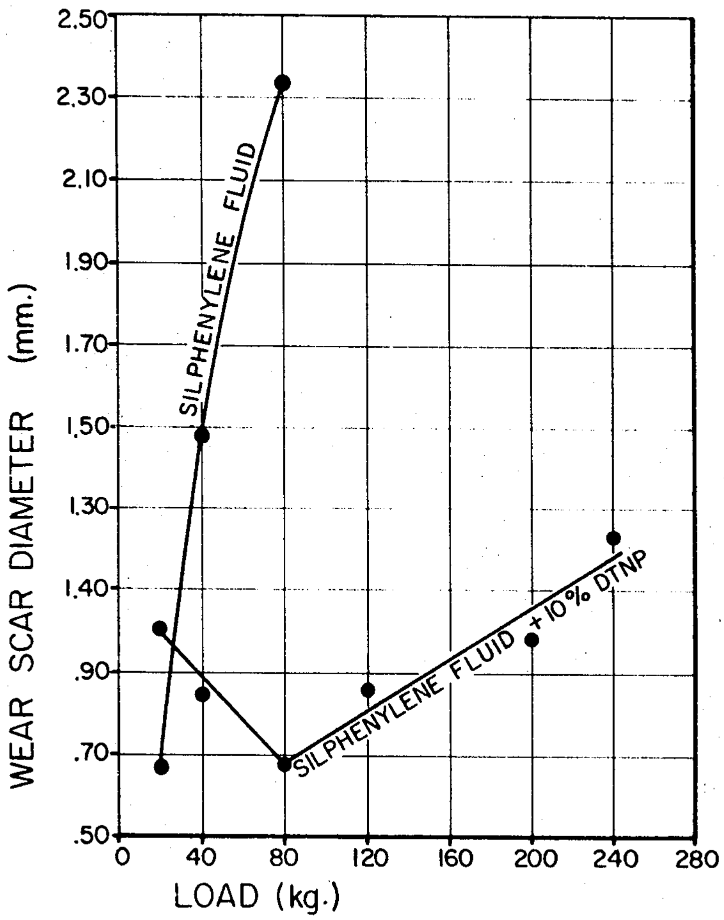

2.50
2.30
2.10
1.90
1.70
1.50
1.30
1.40
.90
.70
.50
WEAR SCAR DIAMETER (mm.)
0
40
80
120
160
200
240
280
LOAD (kg.)
SILPHENYLENE FLUID
SILPHENYLENE FLUID + 10% DTNP

Fig. 3

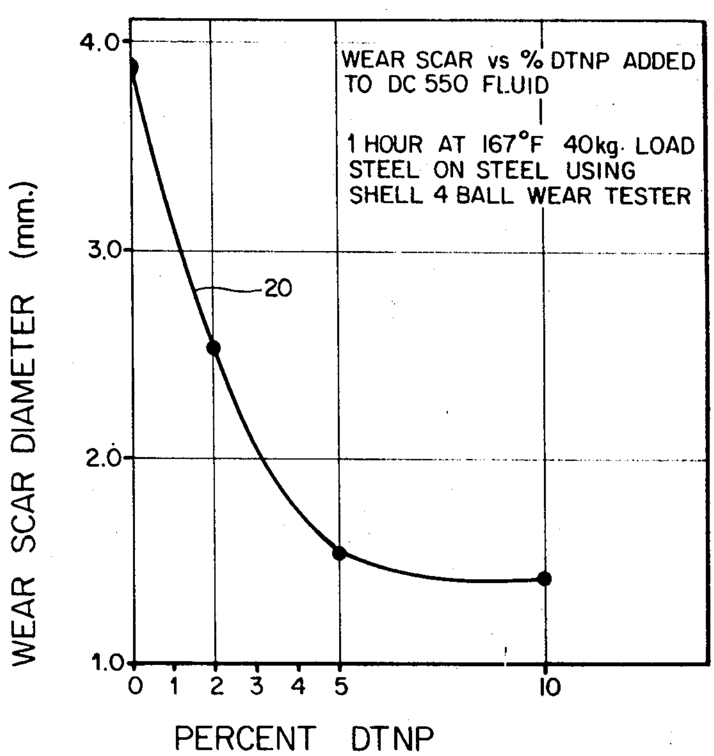

4.0
3.0
2.0
1.0
WEAR SCAR DIAMETER (mm.)
0
1
2
3
4
5
10
PERCENT DTNP
WEAR SCAR vs % DTNP ADDED TO DC 550 FLUID
1 HOUR AT 167°F 40kg. LOAD STEEL ON STEEL USING SHELL 4 BALL WEAR TESTER
20

Fig. 4

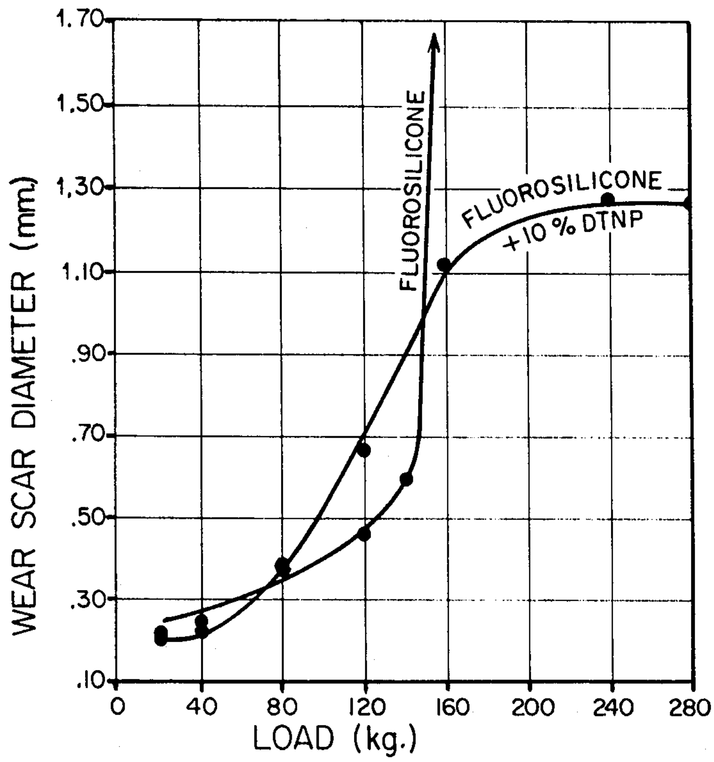

1.70
1.50
1.30
1.10
.90
.70
.50
.30
.10
WEAR SCAR DIAMETER (mm.)
FLUOROSILICONE
FLUOROSILICONE
+10 % DTNP
0
40
80
120
160
200
240
280
LOAD (kg.)

Fig. 5

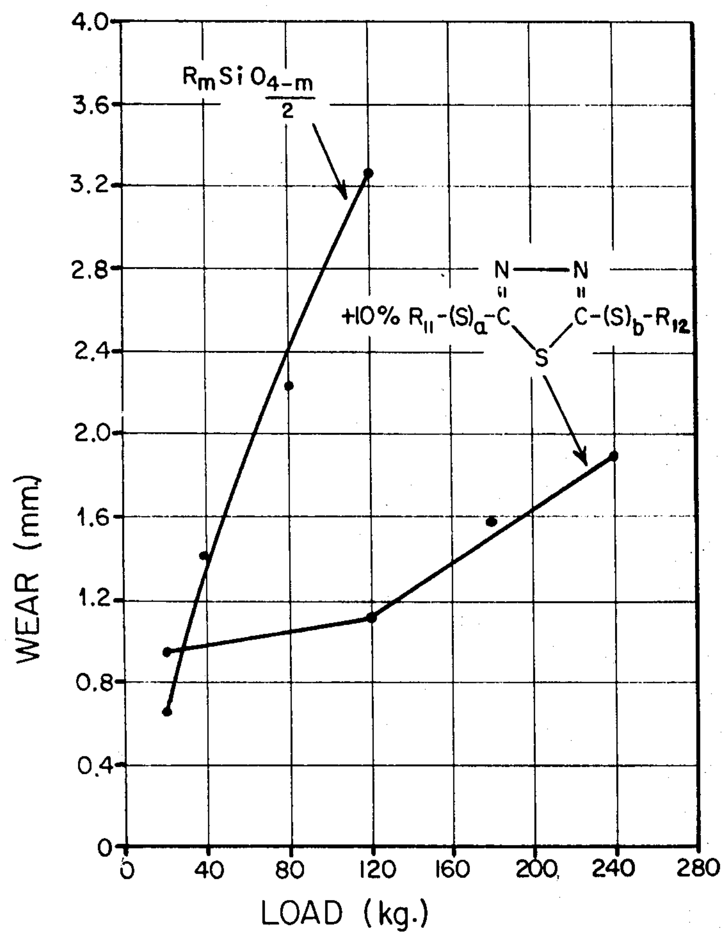

4.0
3.6
3.2
2.8
2.4
2.0
1.6
1.2
0.8
0.4
0
WEAR (mm.)
RmSiO4-m/2
+10% R11-(S)a-C N N C-(S)b-R12 S
0
40
80
120
160
200
240
280
LOAD (kg.)

Fig. 6

SILICONE LUBRICANTS

BACKGROUND OF THE INVENTION

Silicone oils and lubricants compounded from them are well-known, commercially available materials which have presented numerous advantages over other types of lubricant compositions. Generally speaking, for equivalent viscosities, silicone lubricants have (1) lower evaporation rates, (2) smaller changes in viscosity with unit change in temperature, (3) lower freezing or solidification points, and (4) better oxidation and thermal degradation resistance than other types of lubricants. However, a major disadvantage of such silicone lubricants is their poor load-carrying capacities. In this respect, the silicone lubricants are considerably less useful than most other classes of lubricants, including the commonly used petroleum and diester lubricants. In fact, the ability of the silicone lubricants to lubricate steel vs. steel combinations in sliding motion under boundary conditions is practically nil. The well-known antiwear and EP additives which are readily soluble in the petroleum and diester lubricants cannot be employed with silicones because of their poor solubility therein.

SUMMARY OF THE INVENTION

This invention relates to silicone lubricant compositions and has for an object greatly increasing the wear-prevention properties of a silicone lubricant, especially under conditions of sliding motion, by adding thereto a sulfur containing compound soluble in the silicone and including at least two sulfur atoms per molecule.

Unexpectedly, we have discovered that the addition, to a silicone lubricant, of a minor amount of a sulfur-containing compound dissolved in that lubricant and including two or more sulfur atoms per molecule significantly enhances the lubricity properties of the silicones, thereby, greatly extending the applications to which such silicone lubricants can be directed.

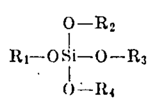
In general, any of the well-known silicone fluids and greases may be used to provide the lubricant compositions of the invention. Such silicone lubricants generally are liquids, or solids liquifiable on warming. As used herein, the term "silicone lubricant" includes the well-known organopolysiloxanes and organosilanes, silicate esters, silarylene-type compounds, such as silphenylene, as well as alkyl and aryl disilazane such as hexamethyl disilazane, for example.

Among the organopolysiloxanes which may be used to provide the lubricant compositions of the invention are those having the formula

$$\text{I} \quad R_mSiO_{\frac{4-m}{2}}$$

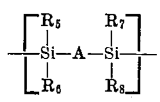
where R represents a member selected from the class consisting of alkyl radicals (e.g., methyl, ethyl, propyl, isopropyl, butyl, octyl, etc. radicals); cycloalkyl radicals (e.g., cyclohexyl, cycloheptyl, etc., radicals); aryl radicals (e.g., phenyl, diphenyl, naphthyl, etc., radicals); alkaryl radicals (e.g., tolyl, xylyl, ethylphenyl, etc., radicals); aralkyl radicals (e.g., benzyl, phenylethyl, etc., radicals); haloaryl radicals (e.g., monochlorophenyl, dibromophenyl, tetrachlorophenyl, monofluorophenyl, etc., radicals); cyanoalkyl radicals (e.g., cyanomethyl, β-cyanomethyl, γ-cyanopropyl, etc., radicals). Preferably at least 50 mole percent of the R groups are lower alkyl groups having from one to two carbon atoms, preferably the methyl radical, and m has a value from 1.98 to 3. Examples of such organopolysiloxanes are given in Patnode U.S. Pat. Nos. 2,469,888 and 2,469,890, and in Pfeifer U.S. Pat. No. 2,704,748, wherein are disclosed linear as well as branch-chained organopolysiloxanes coming within the above formula as well as cyclic organopolysiloxanes, e.g., hexamethylcyclotetrasiloxane, octamethylcyclotetrasiloxane, tetraphenyltetramethylcyclotetrasiloxane, octaethylcyclotetrasiloxane, etc.

The silicate ester lubricants which can be used to provide the compositions of the invention will generally correspond to the formula



where R_1, R_2, R_3, and R_4 may be the same or different and are selected from the same class of radicals as designated for R in formula I above.

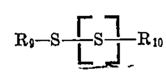
The silarylene-type materials useful to provide the lubricant compositions of the invention will generally comprise polymeric materials characterized by recurring structural units of the formula



where R_5, R_6, R_7 and R_8 are the same or different and are selected from the same class of radicals as R above, and A is an arylene radical (e.g., phenylene, diphenylene, naphthylene, etc. radicals); alkarylene radicals (e.g., tolylene, xylylene, ethylphenylene, etc. radicals); haloarylene radicals e.g., monocholorophenylene, dibromophenylene, tetrachlorophenylene, monofluorophenylene, etc., radicals).

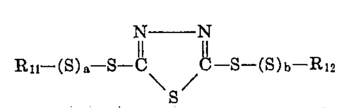
Other examples of silicone fluids which can be used in the practice of the invention may be found in *Synthetic Lubricants*, R. C. Gunderson and A. W. Hart, Reinhold Publishing Corp. 1962.

In general, any sulfur-containing compound, soluble in the silicone lubricant with which it is to be admixed, and including at least two atoms of sulfur per molecule, may be employed to provide the improved silicone lubricants of the invention. Among the sulfur containing compounds which may be used are those having the formula



where R_9 and R_{10} are hydrocarbon radicals which can be the same or different, and which are selected from the same class of radicals as R above, and x is a number from 1 to about 20.

One particular group of sulfur compounds which can advantageously be used are those corresponding to the general formula



where R_{11} and R_{12} are the same or different hydrocarbon radicals selected from the same class of radicals as R above, a and b are whole numbers from 0 to about 8, the sum of a and b being at least 1, and preferably 2 to about 16. Specific examples of polysulfides falling within the scope of the above formula, as well as methods of their preparation, can be found in U.S. Pat. No. 2,719,125—Roberts.

Specific examples of sulfur compounds which can be employed in the practice of the present invention are, for instance, tetraethyl thiuram disulfide, antimony diamyl dithiocarbamate, antimony diamyl phosphordithioate, di-tertiary butyl polysulfide, di-tertiary amyl polysulfide, di-tertiary octyl polysulfide, di-tertiary-nonyl polysulfide, di-tertiary dodecyl polysulfide and the disulfide derivative of 2,5-di-mercapto-1,3,4-thiadiazole, and polysulfide polymers such, for example, as those corresponding to the formula

VI

$$H—S—(C_2H_4—O—CH_2—O—C_2H_4—S_2)_x—C_2H_4—0—CH_2—0—C_2H_4—S—H$$

where x is 6 or 23.

Other examples of suitable polysulfides which are soluble in the silicone lubricant with which they are to be admixed and which include at least two sulfur atoms per molecule will readily occur to those skilled in the art.

The amount of the sulfur-containing compound used in the practice of the present invention can be varied within wide limits. Satisfactory results can be obtained in improving the wear-resistant properties of the silicone lubricant by adding to the silicone from as low as about one-tenth of 1 percent to as high as 25 percent or more, based upon the total weight of the silicone lubricant plus sulfur compound employed. Preferably, the amount of the sulfur compound advantageously employed is the minimum required to impart the desired load-carrying characteristics to the silicone lubricant that is used. It is essential in this connection that the sulfur compound used be dissolved in the silicone in the amount employed so as to form a homogeneous solution therewith. Preferably, therefore sulfur compounds which are soluble in the silicone should be employed. In instances where the sulfur-containing compound is not soluble in the silicone, adequate solubility may be obtained by employing a small amount of a common solvent for the silicone and the sulfur compound so as to facilitate the production of a homogeneous solution of the sulfur compound in the silicone. Such common solvents will readily occur to those skilled in the art.

DESCRIPTION OF DRAWINGS AND PREFERRED EMBODIMENTS

Referring now to the drawings, FIGS. 1–6 respectively present the results of test of a plurality of silicone lubricants with and without soluble sulfur additives which include two or more sulfur atoms per molecule.

A Shell 4-Ball Extreme Pressure Tester was used for the comparison presented in the graphs of FIGS. 1–6, inclusive. The pressure tester used to compile the data shown in FIGS. 1–3, 5 and 6 is an accepted testing device comprising three balls which are maintained stationary and located 120° apart. A fourth ball is supported by the three stationary balls, all of them being of steel. In the test, the steel balls were of the composition of specification AISI-C52100.

Referring now to FIG. 1, the ball supported by the other three was rotated with an initial load of about 20 kilograms. The four-ball tester includes a cup within which the balls are located, and which is filled with the lubricant whose lubricity is to be tested. Curve **10** was drawn from data acquired when the cup was filled with a poly(methyl-phenyl)siloxane containing no additive. After rotation of the supported steel ball at 1,800 revolutions per minute for 10 seconds, the test was terminated and the wear scar diameter measured. The four balls, or test specimens, are one-half inch in diameter and the reference to wear scar diameter is the width of the brightened and worn circular area produced on the stationary ball. With initial loading (20kg.), this wear scar diameter had a width of 0.75 millimeters. The four balls were then replaced and the load was increased to 40 kilograms. The wear scar diameter had increased to about 1.42 millimeters. At 75 kilograms load, the wear scar diameter had further increased to 2.15.

We have found that the addition of an organic compound having at least two or more sulfur atoms per molecule dissolved in the silicone liquid greatly decreases the wear. The addition of but one-tenth of a percent of the sulfur-containing additive results in a discernible improvement. If only 1 percent be added, then a load of 40 kilograms up to 110 kilograms was found to result in a wear scar diameter not exceeding about 1.375 millimeters. As a matter of fact, with a load of 75 kilograms, the curve **11** indicates a decrease in wear scar diameter to 1.25. As the load is further increased to 220 kilograms, it will be seen that the scar diameter increased more or less linearly until the value of about 200 kilograms was reached, and then increased at a much more rapid rate, seizure having taken place at the load of 220 kilograms.

By adding 5 percent of the sulfur-containing compound, in this case the di-tertiary-nonyl polysulfide in 5 percent of the weight, the initial scar diameter at 40 kilograms load had not increased beyond its magnitude at 20 kilograms (see curve **12**). There was then the unexpected decrease in scar diameter to 75 kilograms load with a gradual rise in scar diameter until a loading of 240 kilograms was reached. At this heavy loading, the scar diameter was only 1.35 in contrast with the scar diameter for the silicone liquid without additive of 2.27 at 80 kilograms of load, and as compared with a scar diameter of about 1.9 for the 1 percent of additive at a load of 220 kilograms.

Curve **13** shows the results obtained by the addition of 10 percent by weight of di-tertiary-nonyl polysulfide to the silicone employed. The scar diameter was depressed throughout the loading with the final scar diameter at 240 kilograms about the same as that for the 5 percent addition.

The results as presented by the curves of FIG. **1** demonstrate that silicone lubricants have been provided with greatly increased wear-resistant characteristics which now make them suitable for a wide variety of applications for which the silicones alone were wholly unsuited.

While we prefer to describe lubrication where the oil film is maintained between the bearing surfaces as hydrodynamic, we recognize that others refer to such characteristics as complete or viscous lubrication, meaning that the friction developed arises due solely to the internal fluid friction in the film. This simply means that in hydrodynamic lubrication, the surfaces are separated. In respect of boundary lubrication, the lubricating film becomes extremely thin and we consider that this encompasses surface-to-surface contact.

Referring now to FIG. **2**, curve **14** illustrates the relationship between load and wear scar diameter for a chlorinated silicone available on the market as Dow Corning 560. Here it will be seen that between the loading of 20 to 80 kilograms, the scar diameter rapidly increased to 1.65, at which time seizure occurred. The addition of 10 percent of di-tertiary-nonyl polysulfide completely changed the wear characteristics. The initial scar diameter was less, and at a loading of 240 kilograms, it has increased to 1.25, the scar diameter without the additive having been attained as shown by curve **14**, at about 55 kilograms load.

Table I below sets forth the wear scar diameters (in millimeters) obtained from testing, at various load levels, poly(methyl-phenyl)siloxane alone, and in admixture with 5 percent by weight of various alkyl polysulfides corresponding to the formula

$$R_{13}-\left(S\right)_4-R_{13} \qquad \text{(VII)}$$

in which R_{13} was an alkyl radical including from four to 12 carbon atoms.

TABLE I

Carbon chain length	C_4	C_5	C_8	C_9	C_{12}	Base fluid
Load (kg.):						
80	0.59	0.81	1.00	0.78	0.93	2.2
120	0.90	0.95	1.13	1.00	0.99	3.3
240	1.34	1.39	1.52	1.33	1.36	Weld

The results set forth in the foregoing table also make clear the unexpectedly decreased wear to bearings operating under heavy load when lubricated with the compositions of the present invention.

FIG. **3** shows a comparison of wear scar diameters attained at various loads in the test apparatus using silphenylene lubricant alone, and silphenylene lubricant including 10 percent by weight of di-tertiary-nonyl polysulfide. FIG. **3** thus demonstrates that the silphenylene fluid affords little wear resistance since the wear scar diameter rapidly increases from a value of just below 0.7 millimeters at 20 kilograms load to over 2.2 millimeters at 80 kilograms load. On the other hand, the addition of 10 weight percent of di-tertiary-nonyl polysulfide extended the permissible load to 240 kilograms with a scar diameter of only 1.2 millimeters.

FIG. **4** illustrates the effect on lubricity of increased addition to the silicone lubricant of a sulfur-containing compound in accordance with the invention. In this instance, the Shell 4-

Ball Wear Test was conducted. FIG. 1 demonstrates that the addition of only one-half of 1 percent of the sulfur additive (di-tertiary-nonyl polysulfide) materially decreases the scar diameter. The chart shown in FIG. 4 also demonstrates that the optimum amount of sulfur compound addition is at about 8 percent, based on the weight of the silicone liquid used.

FIG. 5 shows the results of tests made on a fluorosilicone lubricant alone, compared to the fluorosilicone lubricant containing 10 percent by weight of the di-tertiary-nonyl polysulfide. It will be observed that the fluorosilicone without the additive is useful as a lubricant up to about 130 kilograms load, at which point, any additional loading greatly increases the scar diameter. In contrast, the composition of the invention comprising the fluorosilicone including 10 weight percent of di-tertiary-nonyl polysulfide extends the usefulness of the lubricant to a loading in excess of 280 kilograms and with a scar diameter of only 1.25 millimeters.

FIG. 6 illustrates the unexpected results achieved by adding to the silicone lubricant 10 weight percent of a polysulfide comprising an alkyl derivative of 2,5-di-mercapto-1,3,4-thiadiazole.

The following table II sets forth the data obtained from an oscillator motion test conducted with an organic dye thickened poly(methyl-phenyl)siloxane with, and without, a sulfur compound additive (alkyl derivative of 2, 5-di-mercapto-b 1, 3, 4Thiadiazole of the type disclosed in U.S. Pat. No. 2,719,125).

TABLE II

	Cycles to Failure
Siloxane alone	577
Siloxane plus 5% by weight of sulfur additive	2,385
Test Conditions	
AISI 4620 steel ring oscillation against a stationary AISI 4130 steel block.	
Grease Properties	
Penetration (worked 60 strokes) =	297
Dropping point =	>450° F.
Oscillations/Min.	87.5
Load	90 lbs.
Temperature	77° F.

The following table III sets forth the wear scar diameters (in mm.) obtained at various load levels using a poly(methyl-phenyl)silicone modified with 15 percent di-tertiary-nonyl polysulfide and with 25 percent of said polysulfide. The Shell 4-Ball EP Tester was used.

TABLE III

Load (kg.)	40	80	120	240	300
85% silicone +15% polysulfide	0.57	0.97	0.88	1.44	1.58
75% silicone +25% polysulfide	0.40	0.68	0.87	—	1.59

Table IV below sets forth the wear scar diameters (in mm.) obtained at various load levels using (a) poly(methyl-phenyl)siloxane plus 10 percent by weight di-tertiary-nonyl polysulfide, (b) poly(methyl-phenyl)siloxane siloxane plus 10 percent by weight of an alkyl derivative of 2,5-di-mercapto-1,3,4-thiadiazole; and (c) poly(methyl-phenyl)siloxane plus 5 percent by weight of di-tertiary-nonyl polysulfide and 5 percent by weight of an alkyl derivative of 2,5-di-mercapto-1,3,4-thiadiazole. The Shell 4-Ball EP Tester was used.

TABLE IV

SCAR DIAMETERS (mm.) AT VARIOUS LOADS

	at 40kg.	at 80kg.	at 120kg.	at 180kg.	at 240kg.
(a)	0.84	0.71	0.81	1.03	1.33
(b)	1.01	0.90	1.15	1.57	1.89
(c)	0.95	0.83	1.15	1.45	1.60

Formulation (c) combines the antioxidant and corrosion inhibitor qualities of the alkyl derivative of 2,5-di-mercapto-1,3,4-thiadiazole with the antiwear properties of the di-tertiary-nonyl polysulfide, and would be preferred to provide rust preventive properties as well as antiwear and extreme pressure characteristics.

The following table V provides a comparison of metal response (AISI–52100 and AISI 440C) at various load levels using (a) poly(methyl-phenyl)siloxane lubricant alone and (b) poly(methyl-phenyl)siloxane plus 5 weight percent of di-tertiary-nonyl polysulfide. The Shell 4-Ball EP Tester was employed.

TABLE V

WEAR SCAR DIAMETER (mm.) AT VARIOUS LOADS (kg.)

	at 40	at 60	at 120	at 160	at 200
AISI–52100					
with (a)	1.4	1.9	3.3	—	—
with (b)	1.0	0.88	1.0	1.11	1.23
AISI–440C					
with (a)	2.6	3.2	—	—	—
with (b)	1.37	1.51	1.94	2.38	2.87

The following table VI sets forth the values of wear scar diameters (in mm.) at various loads using, as a lubricant, (a) poly(methyl-phenyl)siloxane; (b) poly(methyl-phenyl)siloxane plus 10 percent by weight of di-2-ethyl hexyl sebacate (a well-known and widely used diester fluid); (c) poly(methyl-phenyl)siloxane with 9.5 weight percent di-2-ethyl sebacate and 5 percent by weight of di-tertiary-nonyl polysulfide; and (d) 85.05 weight percent poly(methyl-phenyl)siloxane, 9.45 weight percent di-2-ethyl hexyl sebacate, 5 percent di-tertiary-nonyl polysulfide and 0.5 percent phenyl alphanaphthylamine (a well-known antioxidant). The Shell 4-Ball EP Tester was used.

TABLE VI

WEAR SCAR DIAMETERS (mm.) AT VARIOUS LOADS (kg.)

	at 80kg.	at 120kg.	at 240kg.
(a)	2.2	3.3	weld
(b)	2.7	weld	weld
(c)	0.8	1.02	1.45
(d)	0.8	1.01	1.40

It should be understood that while the present invention has been described in considerable detail with respect to certain specific embodiments thereof, it is not to be considered limited to those embodiments, but may be used in other ways without departure from the spirit of the invention or the scope of the appended claims.

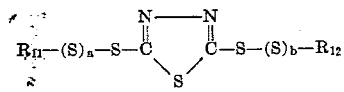
What is claimed is:

1. A lubricant composition consisting essentially of a major proportion of an organic silicon-containing lubricant selected from the class consisting of organopolysiloxanes, organosilanes, silicate esters, silarylenes, and alkyl and aryl disilazanes, having dissolved therein a minor proportion, sufficient to improve the load-carrying properties of said organic silicon-containing lubricant, of a sulfur-containing compound selected from the group consisting of:

a. a di-tertiary alkyl polysulfide wherein the alkyl group includes from four to 12 atoms;

b. a compound having the general formula $H—S—(C_2H_4—O—CH_2—O—C_2H_4—S_2)_x—C_2H_4—O—CH_2—O—C_2H_4—S—H$ wherein x is 6 or 23; and

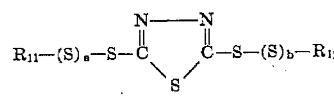
c. a hydrocarbon polysulfide derivative of 2,5dimercapto-1,3,4,-thiadiazole having the general formula



wherein R_{11} and R_{12} are the same or different hydrocarbon radicals selected from the class consisting of alkyl, cycloalkyl, aryl, alkaryl, aralkyl, haloaryl, and cyanoalkyl radicals; and *a* and *b* are whole numbers from 0 to about 8, the sum of *a* and *b* are whole numbers from 0 to about 8, the sum of *a* and *b* being at least 1.

2. The lubricant composition of claim 1 in which the sulfur containing compound ranges from about 0.1 percent to about 25 percent by weight of the total weight of said silicone and said sulfur-containing compound.

3. The lubricant composition of claim 1 in which the sulfur-containing compound is a hydrocarbon polysulfide derivative of 2,5-dimercapto-1,3,4,-thiadiazole having the general formula



wherein R_{11} and R_{12} are the same or different hydrocarbon radicals selected from the class consisting of alkyl, cycloalkyl, aryl, alkaryl, aralkyl, haloaryl, and cyanoalkyl radicals; and *a* and *b* are whole numbers from 0 to about 8, the sum of *a* and *b* being at least 1.

4. The lubricant composition of claim 1 wherein the sulfur-containing compound comprises a compound having the general formula $H—S—(C_2H_4—O—CH_2—O—C_2H_4—S_2)_x—C_2H_40—CH_2—O—C_2H_4—S—H$ wherein x is 6 or 23.

5. The lubricant composition of claim 1 wherein the sulfur-containing compound comprises a di-tertiary alkyl polysulfide wherein the alkyl group includes from four to 12 carbon atoms.

6. The lubricant composition of claim 5 wherein the sulfur-containing compound is di-tertiary-nonyl polysulfide.

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CERTIFICATE OF CORRECTION

Patent No. 3,609,079 Dated: September 28, 1971

Martin J. Devine and
Edward R. Lamson

It is certified that errors appear in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 7, lines 29 and 30, delete the phrase "the sum of a and b are whole numbers from 0 to about 8,".

Signed and sealed this 21st day of November 1972.

(SEAL)
Attest:

EDWARD M.FLETCHER,JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents