RESEARCH PAPER | Engineering | Volume : 3 | Issue : 11 | Nov 2013 | ISSN - 2249-555X

A Study on The Preparation and Evaluation of Silicone Greases

KEYWORDS: silicone grease , silicone oil, fumed silica, polyethylene, Silicone rubber

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ABSTRACT *This study describes the formulation of silicone greases using two grades from silicone oil as fluid; Polyethylene, fumed silica, silicone rubber and styrene butadiene rubber using as thickener for prepared greases. In this respect, five formulations from silicone greases are prepared with different proportions of these compounds. The effects of the different ingredients ratios on efficiency of the formulated greases have been studied. Examination of the prepared greases including penetration, dropping point, and oil separation testes is carried out. It has to be mentioned that with respect to prepared greases on increasing silicone oxide and polyethylene by-product, oil separation decrease while consistency increase. The penetration also significantly decreases as fumed silica addition increases. The study reveal that the formulated greases have no dropping point, which proves that the prepared greases have stable structure at high temperature. This is due to the larger strength and small size of fumed silica. In addition, the electrical properties (tan delta, dielectric constant, and resistivity) of formulated greases are also investigated. It may be concluded that the prepared silicone greases, because of their improved mechanical, thermal and electrical properties, have high potential to be used in wide range of industries such as cable, vacuum seal and most of the industries that deals with heavy duty equipments.*

Introduction

Thermal greases typically offer better thermal performance and reduced manufacturing cycle times in comparison to other thermal interface material options. However, thermal greases are susceptible to a variety of failure mechanisms during their service life. Two main causes of increase in thermal resistance of a grease layer are grease pump-out and grease dry-out. The powering up or powering down of the device causes a relative motion between the die and the heat spreader, which tends to squeeze the thermal grease out of the interface gap. This phenomenon is referred to as "pump-out" and results in increased thermal resistance due to loss of grease material from the interface (1,5).

Many patents reported (6-8) the compositions of silicone greases. These patents relates to lubricating compositions and methods for their preparations. In general, silicone grease composition comprising organopolysilixane as thermal fluid and a thickening agent such as finally divided silica filler. The optimal preparation method for each application is dependent upon type of technology and its methodology. Within the area of lubricating greases, alternative sources of resins and additives were studied (9-11). In the present paper, it is desired to study of the effect of some fluids and thickening agents on silicone greases performance such as, mechanical, thermal and electrical properties.

Experimental

Materials

Silicone oil-100, silicone oil-1000 and paraffin oil were supplied by Nerol Chemical Company. Physicochemical properties of these oils were carried out as presented in Table 1. Two grade from polyethylene, film and blow molding grades, were provided by Sidi Kerir Petrochemicals Co; these grades were designated PE-1 and PE-2, respectively. Fumed silica (silicone oxide), octyl amine and silicone rubber, natural rubbers are used as commercial grade

Grease Formulations

Five formulations of silicone greases, designated Grease A, Grease B, Grease C, Grease D and Grease E were designed using different ratios from above mentioned materials as listed in Table 2. Certain amount of paraffin oil was heated at temperature of 150 ^{0}C at autoclave then Polyethylene EP-1, EP-2 and natural rubber were added. The mixture was continuously heated to ensure computability occurred and then cooled to 100 ^{0}C. After that, silicone dioxide, silicone oils, silicone rubber and octyl amine were added according to ratio as mentioned in Table 2. The stirring was continued at temperature of 100 ^{0}C for 2 hour then cooled to room temperature. The obtained greases were good homogenized and store prior to analysis.

Grease Characterization

Dropping point, evaporation loss and penetration tests of the investigated greases were performed according to ASTM methods, D-566, D-2595 and D-217, respectively. The mechanical properties for the prepared greases were carried out using ASTM Grease Worker. The prolonged worked penetration was measured after the greases have been worked from 1000 to 3000 double strokes. Also, Corrosion evaluation for formulated greases under study was performed according to ASTM D-4048 Method (Table 3).

Dielectric constant of the prepared greases was measured using Keithley Electrometer Model 6517B. The permittivity and resistivity were measured at 1KHz, 10KHz and 100 KHz for all investigated greases (Figure 1). Thermogravimetric analysis for selected Grease A, Grease D and Grease E was carried out by using Instrument: SDT Q600 V20.5 Build 15. Approximately 10 mg from grease sample was placed on a pt pan and heated from 30 to 600^{o}C at 10 ^{o}C per minute Figure1.

Result and Discussion

Table 1, shows the physicochemical properties of silicone oil-100, silicone oil -1000 and paraffin oil. There are some differences between these oils due to their chemical compositions. In this respect, Flash point, evaporation loss, pour point, total acid number and dynamic viscosity of both grades of silicon oils are better than in case paraffin oil concerning lubricating properties. This indicates that the silicone oils exhibit high thermal stability, oxidation resistance, very low pour point and high flash point in compared to paraffin oil.

Table 1: Physicochemical Properties of the Fluids

Test	Silicon oil -100	Silicone oil -1000	Paraffin oil
Flash point , °C	310	350	220
pour point °C	-3o	-30	+2

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Evaporation loss, 24hr at 150 °C	0.2	0.2	3.4
Evaporation loss, 48hr at 150 °C	0.22	0.21	3.9
Dynamic viscosity ,cP			
@ 40 °C	340	395	150
@100 °C	240	35o	75
Total acid number	0.01	0.01	0.04

Grease is the preferred form of lubrication in hard-to-reach places in a mechanically rubbing or dynamic systems. Grease acts as reservoir for lubricant based fluids and additive molecules. In this respect, many trials were done to reach the designed formulations of silicone greases with different material ratios as presented in Table 2. The prepared greases depend on the physical and chemical properties of their ingredients that are structurally related, which is obtained by the proper selection of ingredients and processing. Fume silica with excellent grease-forming properties as a result of its elongated or chain like structure and high oil absorbing properties, was used as a thickening agent (12). Therefore, fume silica percentage ranging from 4wt% to 9 wt% have good effect on evaporation loss, penetration and oil separation of formulated greases. Generally, with increasing of fume silica significantly increase the consistency and decrease oil separation. This may be due to the absorbing properties and small size of fume silica. The bonding of silicone oxide thickening agent in gel varied with the weight of the solids. Thus, the large strength of silicone oxide was thought to be due to the structure being knit together by means of primary valences bonds. The increases of silicone oxide concentration tend to increase the consistency of formulated greases.

Table 2: Formulated Silicone Greases and their Ingredients

Type of Grease / Ingredient	Grease -A	Grease -B	Grease -C	Grease -D	Grease -E
Silicone -100, Wt%	58	0.0	58	0.0	29
Silicone-1000, Wt%	0.0	58	0.0	58	29
Silicone rubber, Wt %	15-14	14-13	13-12	12-11	11-10
Silicone dioxide, Wt%	4-5	5-6	6-7	7-8	8-9
Paraffin oil, Wt%	20	20	20	20	20
Polyethylene-1, Wt%	3.0	0.0	3.0	0.0	1.5
Polyethylene-2, Wt%	0.0	3.0	0.0	3.0	1.5
Natural rubber	0.5	0.5	0.5	0.5	0.5
Octyl amine,Wt%	0.3	0.3	0.3	0.3	0.3

Table 3 ,show the variation of penetration with mechanical working at different strokes 1000 to 3000 as a function of binding forces between ingredients of prepared greases compared with data of unworked. It was noted that with increasing stroke number the consistency values of greases decreases in the order Grease-A> Grease-B > Grease-C>grease-D =Grease-E. This reveal that the greases C , D and E are resist deformation in consistency with increasing the stroke numbers. It could be speculate that the degree of texture deformation of prepared greases depend on many factors such as, type of bond, type of interaction and degree of the compatibility between grease compositions Also, Careful inspection in Table 3 shows improvement and reinforcement in all physicochemical properties for prepared greases. This is attributed to increasing thickener concentration lead to increasing the columbic interaction between silicone oxide, silicone rubber, natural rubber and silicone oil to form good three-dimensional network and good texture (sponge) that have good thermal, mechanical and oxidation stabilities.

Corrosion of metals in contact with lubricating greases may be caused either by a component formed during service or by some external compound. Thus, octyl amine was added as anticorrosion additives with o.3wt%. According to the ASTM Copper Strip Classification, the copper strip immersed in greases A, B, C, D and E which is a passing ASTM rating. It is evident that the octyl amine serves the purpose of an anticorrosive agent by taking up any corrosive compound. This means that the formulated greases under study may be used in new electrical installations and maintenance because of its excellent performance in keeping metals free from corrosion.

Table 3: Characterization of formulated greases

Test	Formulated Greases				
	Grease-A	Grease-B	Grease-C	Grease-D	Grease-E
Penetration Unworked	280	278	270	270	274
Worked 60 strokes	280	278	270	270	274
1000 strokes	281	279	271	270	274
2000 strokes	283	280	275	276	278
3000 strokes	288	283	280	283	284
NLGI consistency number	2	2	2	2	2
Dropping point	Non-melt	Non-melt	Non-melt	Non-melt	Non-melt
Evaporation loss	0.4	0.4	0.3	0.35	0.35
Oil separation	0.5	0.5	0.3	0.25	0.30
Oxidation Stability	1.4	1.3	1.1	1.4	1.5
Dynamic viscosity, cP.	22456 LB	22511 LB	22559 LB	22590 LB	22609 LB
Egyptian Standard Code					

Electrical properties

The experimental data in Table 4 show the permittivity values of formulated silicone greases under investigation. These greases have in general a high resistivity in order of TΩ m and have a very low tan delta less than 0.001. Therefore, dielectric constant of prepared greases is electrically insulating and does not break down when high voltage is applied. Despite strongly polarized Si-O bonds, silicone rubber is nonpolar, as the methyl side groups prevent Si-O dipoles from approaching each other too closely. It may be speculate that the intermolecular forces are weak and mainly composed of London-van der Waals interactions that decrease with the square of the distance between molecules.

Table 4: Dielectric properties of formulated silicone greases

Type of Grease / Resistivity	Grease A	Grease B	Grease C	Grease D	Grease E
E, at 1 kz	7.3	6.49	14.78	17.80	16.95
E, at 10 kz	7.3	6.61	14.83	17.75	17.18
E, at 100 kz	7.3	6.68	14.60	17.42	17.02
P, Ohm.m	3.37E+13	6.332E+12	3.230E+12	3.230E+12	6.399E+12

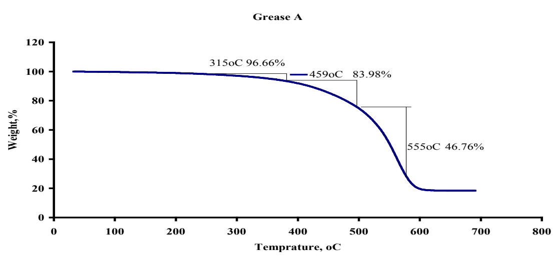
Volume : 3 | Issue : 11 | Nov 2013 | ISSN - 2249-555X

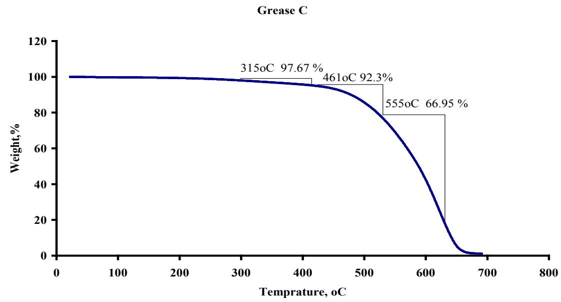
Thermal characterization

Thermal decomposition profile of the selected silicone greases, grease A, Grease C and Grease E, show similar behaviors as observed in Figure 1. According to weight loss data, it was observed that the thermal decomposition takes place in just three stages, which is identical for all the blends studied. The first step, obtained at temperature 315oC , is attributed to the departure of the paraffin oil and polyethylene from the sample. The second occurs between 315 to 461 oC is attributed to the departure of the silicone oil from the sample this is the most important step to determine the grease thermal stability order and third at temperature 555 oC , which corresponds to the grease degradation. Based on these data it may be concluded that the weight loss of selected greases is decreasing in the following order grease A< grease C < grease E. It has to be mentioned that with respect to prepared silicone greases on increasing silicone rubber thermal stability increase

Conclusion

Results indicate that the fume silica and polyethylene percents in formulated grease are directly proportional to the dynamic viscosity and consistency. The formulated greases produced are classified as NLGI 2 and suitable for high temperature up to 265°C. They are proven that polyethylene by product could be reuse as part of thickening agent in silicone grease formulation and reducing the cost of the grease produced. In addition, the results of corrosive substance test proved that the octyl amine suitable and proper anticorrosion for such greases. On other hand, the all formulated silicone greases consider as excellent dielectric materials and have high insulation.





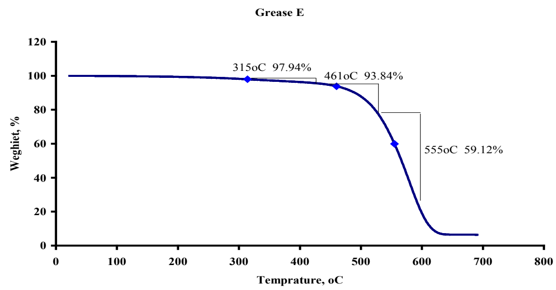


Figure 1.Thermal gravimetric analysis of selected greases, A , C and E.

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