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EXPERIMENTAL RESEARCHES ON RHEOLOGICAL PROPERTIES OF BIOGREASES

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Abstract: *Currently, more and more manufacturers are producing biodegradable, high-performance soybean oil-based greases, available for various industrial and automotive applications. These products offer environmental benefits, such as non-toxicity and rapid biodegradability, as a sustainable alternative to conventional petroleum-based greases. This paper is focused on the rheological study of biodegradable greases based on soybean oil and aqueous sodium stearate, additivated with graphene or graphite nanoparticles in different concentrations. For this purpose, different biogrease samples have been prepared and each of them were thermal analysed in the range of 20⁰C … 75⁰C on a Brookfield CAP 2000+ viscometer, using Bingham rheological model. Finally, the thermal variation of the rheological parameters (yield stress and viscosity) was obtained, taking into account the presence of nano-additives.*

Keywords: *Rheology, biogreases, thermal, nano-additives*

1. Introduction

Biodegradable greases are semi-solid lubricants specifically formulated to be braked down by biological microorganisms in the environment, thereby reducing negative environmental impact [1]. They are used in applications where there is a risk of leakage or contamination of soil and water. They are minimally toxic to aquatic organisms, plants, and animals and do not bioaccumulate in the food chain. In case of spills or leaks (common in "total-loss" applications like chainsaws, food industry or rail curve grease), the environmental damage and associated clean-up costs are significantly lower than for petroleum-based products [2].

Currently, more and more manufacturers are producing biodegradable, high-performance soybean oil-based greases, available for various industrial and automotive applications [3].

Soybean oil-based greases are available in a variety of formulations, often incorporating different thickeners and additives to achieve performance comparable to traditional mineral oil products [4].

This paper proposes to study biodegradable greases based on soybean oil and aqueous sodium stearate as the thickener. Sodium stearate acts as a soap-based thickener, which forms a fibrous network within the soybean oil to create the grease structure [5], [6]. The use of an aqueous sodium stearate component facilitates the initial mixing and reaction. Water is typically heated and stirred with the sodium compound and stearic acid to dissolve the components and allow for the saponification reaction. The concentration of the sodium soap thickener in the base oil significantly impacts the final consistency of the grease. Ratios of oil-to-thickener are adjusted to achieve desired properties, typically within the 5-30% thickener range [7], [8].

The main advantage of using aqueous sodium stearate as the thickener is a high operating temperature range and a high level of biodegradability and non-toxicity.

Nanoparticles are highly effective as grease additives, significantly enhancing the grease's performance by reducing friction and wear, increasing load bearing capacity, and improving thermal stability and conductivity. They achieve this through several mechanisms at the microscopic level, such as: formation of the protective tribofilms at the surface level, rolling effect in the contact due to spherical nanoparticles etc. [9], [10].

In this paper, the nanoparticles used as additives for biodegradable greases are based on two allotropes of carbon with a hexagonal lattice structure: graphite and graphene. Graphite is a mineral that naturally occurs in metamorphic rock. It is formed as a result of the reduction of sedimentary carbon compounds during metamorphism. Contrary to common belief, the chemical bonds in graphite are actually stronger. So, graphene is fundamentally one single layer of graphite; a layer of bonded carbon atoms arranged in a honeycomb (hexagonal) lattice. However, graphene offers some impressive properties that exceed those of graphite as it is isolated from its “mother material” [11], [12].

2. Experimental methodology

Biodegradable greases used for experiments were prepared using soybean base oil with aqueous sodium stearate thickener in concentration of 8% (wt). Four different types of samples have been tested, with the following composition:

- Pure grease (Fig. 1);
- Additivated grease, with antioxidant (1%) and antiwear (2%) additives (Fig. 2);
- Graphene grease, with antioxidant (1%), antiwear (2%) and graphene powder (0.3%) (Fig. 3);
- Graphite grease, with antioxidant (1%), antiwear (2%) and thermal expanded graphite (0.3%) (Fig. 4).

Fig. 1. Pure grease

Fig. 2. Additivated grease

Fig. 3. Graphene grease

Fig. 4. Graphite grease

The rheological measurements were performed on a Brookfield viscometer CAP2000+ equipped with four cone-and-plate geometry and using a Peltier system for controlling the temperature. The CAP 2000+ Series Viscometers are medium to high shear rate instruments with Cone Plate geometry and integrated temperature control of the test sample material, [13]. A typical view of the viscometer is presented in Fig. 5, with all the four geometries cone and plate.

Fig. 5. Geometry of Brookfield viscometer

Concerning the technical parameters of the viscometer, rotational speed selection ranges from 5 to 1000 RPM. Viscosity measurement ranges depend upon the cone spindle and the rotational speed (shear rate). Viscosity is selectively displayed in units of centipoise (cP), poise (P), or Pascal seconds (Pa•s). Temperature control of sample is possible between either 5°C (or 15°C below ambient, whichever is higher) and 75°C or 50°C and 235°C depending on viscometer model. The viscometer uses a CAPCALC32 software for complete control and data analysis. The geometry of testing cones and the viscosity range are presented in Table 1.

Table 1: Geometry and viscosity range of testing cones

Cone number	Cone radius, mm	Cone angle, degree	Viscosity range, Pa.s
3	9.53	0.45	0.083 ... 1.87
5	9.53	1.8	0.333 ... 7.50
6	7.02	1.8	0.833 ... 18.7
8	15.11	3	0.312 ... 3.12

A “velocity imposed gradient” test was used, with the temperature range between 20^{0}C and 75^{0}C and cone geometry number 8. The experimental results were numerically treated assuming the validity of Bingham rheological model:

$$\tau = \tau_0 + \eta \frac{\partial u}{\partial y} \quad (1),$$

where: τ – shear stress
τ_0 – yield stress
η – dynamic viscosity
$\frac{\partial u}{\partial y}$ – shear rate

3. Results

The experimental test consists of a load from the 10 s^{-1} to 2000 s^{-1} shear rate gradient, followed by an unload in order to highlight the thixotropy of the lubricant - "shear memory”. The test is repeated three times for each temperature (20^{0}C, 26^{0}C, 32^{0}C, 38^{0}C, 44^{0}C, 50^{0}C, 66^{0}C and 75^{0}C) and the duration of homogenization (soaking time) of the sample at a certain shear rate was 300 seconds. The rheograms are obtained by plotting shear stress as a function of the shear rate, as an average of 30 points, using the software Capcalc 32 specific for the viscometer. Fig. 6 shows a typical rheogram at 20^{0}C, for all four types of greases: pure grease, additivated grease, graphene grease and graphite grease.

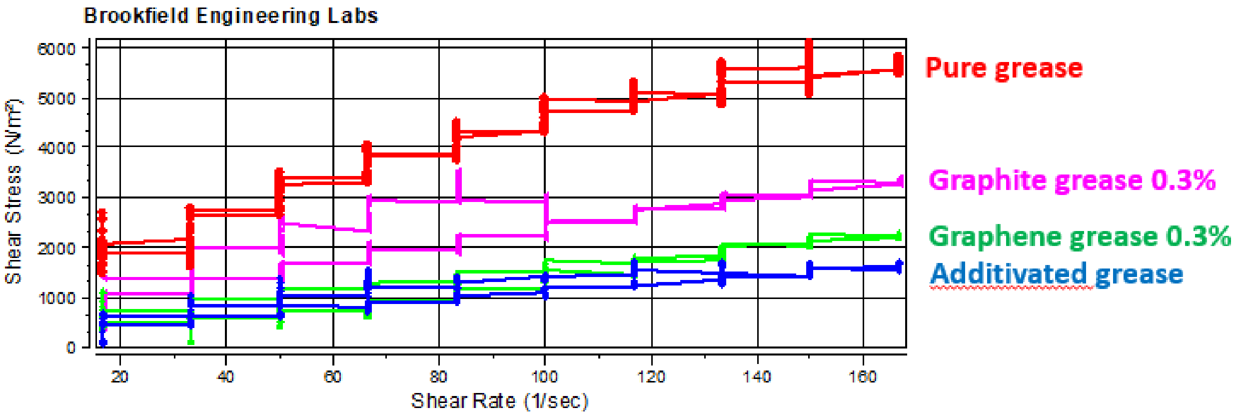

Fig. 6. Lubricant rheograms for the four greases, at 20^{0}C

Analysing the rheograms from Fig. 6 the following observations can be made:

- The grease in pure state has a homogenous structure, with a reduce thixotropy;
- The addition of antioxidant and antiwear additives to the grease decreases the values of the corresponding rheological parameters (yield stress and viscosity) by comparison with the pure grease;
- The supplementary addition of graphene nano-additive increases the values of rheological parameters by comparison with the additivated grease;
- The supplementary addition of graphite nano-additive increases the values of rheological parameters by comparison with the additivated grease with graphene, but it reduces the homogeneity of the grease structure.

The results regarding the variation of the rheological parameters (yield stress and viscosity) with the temperature are presented centralized in Tables 2, 3, 4 and 5.

Table 2: Rheological parameters for the pure grease

Temperature, ^{0}C	Yield stress (τ_o), Pa	Viscosity (η), Pa·s
20	2179	3.51
28	1857	1.98
37	1205	1.41
47	745	1.12
57	419	0.96
66	275	0.72
75	162	0.52

Table 3: Rheological parameters for the additivated grease

Temperature, ^{0}C	Yield stress (τ_o), Pa	Viscosity (η), Pa·s
20	532	1.83
28	337	1.24
37	197	1.10
47	125	0.93
57	112	0.66
66	60	0.57
75	41	0.31

Table 4: Rheological parameters for the graphene grease

Temperature, ^0C	Yield stress (τ_o), Pa	Viscosity (η), Pa·s
20	881	1.92
28	482	1.59
37	314	1.42
47	244	1.16
57	120	0.88
66	79	0.69
75	52	0.40

Table 5: Rheological parameters for the graphite grease

Temperature, ^0C	Yield stress (τ_o), Pa	Viscosity (η), Pa·s
20	1652	2.98
28	1483	1.46
37	1009	1.25
47	772	1.02
57	497	0.87
66	397	0.57
75	153	0.50

4. Discussions

Analyzing the experimental results concerning the variation of the rheological parameters with the temperature (Tables 2, 3, 4 and 5), it can be observed the influence of the additives and nanoparticles on the grease behavior.
The yield stress of the pure grease decreases by the temperature, considering the entire range of temperatures values: from 20^{0}C until 75^{0}C (Fig. 7). For the additivated grease, the yield stress at 20^{0}C is much smaller than the same value for the pure grease; over 20^{0}C, the yield stress is continuous decreasing, on the whole temperatures interval. The addition of the graphene nanoparticles increases the values for yield stress by comparison with the additivated grease, but they still remain smaller than for the pure grease. The greatest values for the yield stress are obtained for grease additivated with graphite particles.

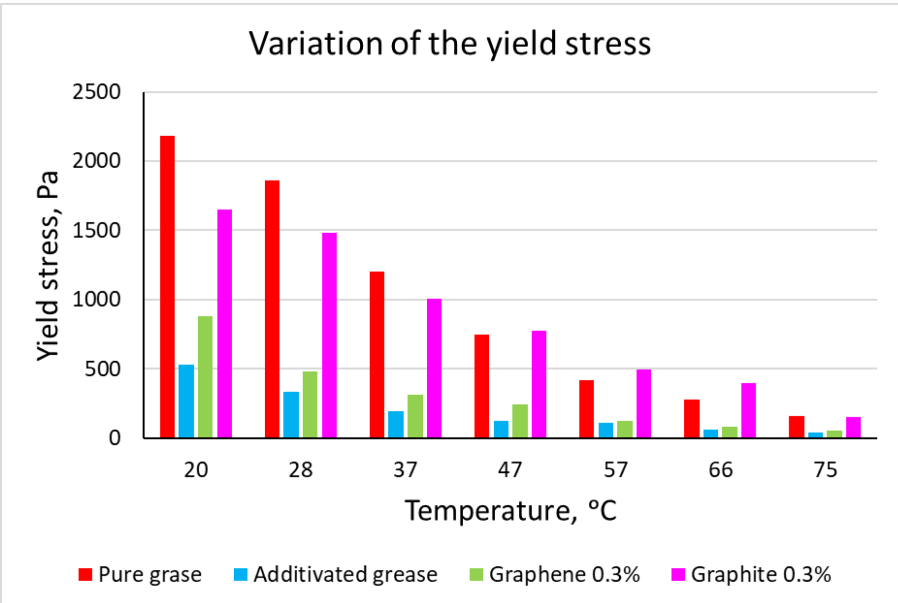

Fig. 7. Variation of the yield stress with temperature

The viscosity of the pure grease decreases by the temperature on whole interval of studied temperatures: from 20⁰C until 75⁰C (Fig. 8). The viscosity of the additivated grease decreases by the temperature and particularly - at 20⁰C - this viscosity is half value of the pure grease viscosity.
Once with the increasing of temperatures, the difference between both grease viscosities is diminishing. The addition of the graphene nanoparticles increases a little bit the viscosity values - by comparison with the additivated grease, but the differences remain quite small.
The greatest values for the viscosity are obtained for grease additivated with graphite particles, even if they are always smaller than the corresponding pure grease. At the end of temperature interval (around 66⁰C ... 75⁰C) the differences between viscosities are almost insignificant.

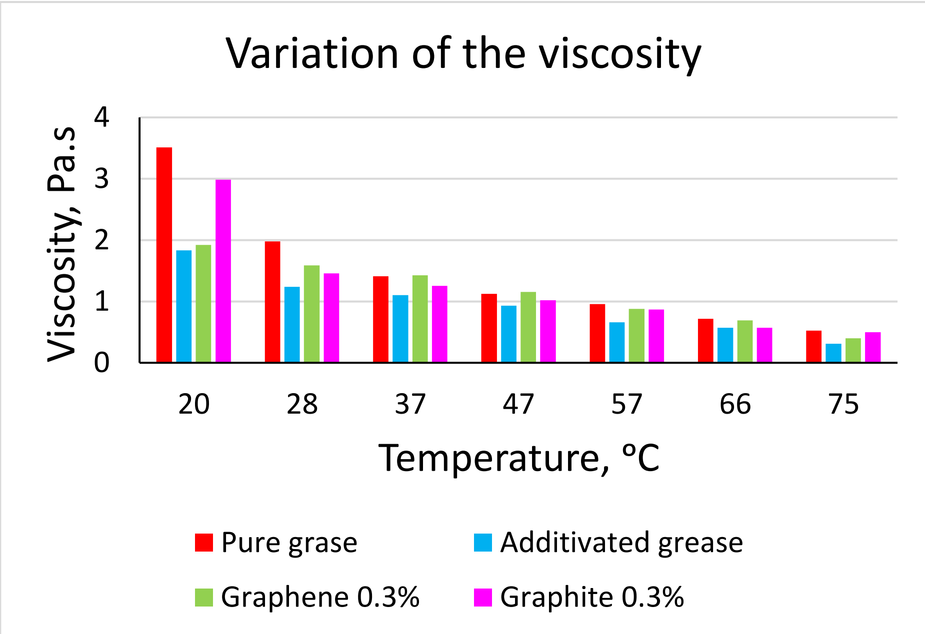

Fig. 8. Variation of the viscosity with temperature

4. Conclusions

1. The paper presents the study of the rheological properties (yield stress and viscosity) of biodegradable greases based on soybean oil with aqueous sodium stearate thickener, in concentration of 8%, additivated with 0.3% graphene or graphite nanoparticles.
2. The rheological properties were investigated in the range of temperature of 20⁰C … 75⁰C and the results were processed according to Bingham model.
3. It can be observed that the addition of the nanoparticles decreases the values of the rheological parameters (yield stress and viscosity), by comparison of the same values for pure grease.
4. The greatest values for the yield stress and for the viscosity are obtained for the grease additivated with graphite nanoparticles, by comparison with the grease additivated with graphene nanoparticles, but they still remain smaller, by considering the pure grease.
5. Supplementary investigations must be performed in order to study the tribological properties of these additivated grease with nanoparticles.

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