Evaluation of Graphene Oxide Dispersed in Spindle Oil as a Potential Lubricating Grease.

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ABSTRACT

With the advancement of nanomaterial research, scientists around the world have focused their efforts on exploring the use of these materials in a wide range of fields, including microelectronics, medicine, and tribology. It is widely recognized that carbon-based nanomaterials, when added to greases, have significant potential to reduce friction in moving parts of machinery, thus improving efficiency and durability.
In this study, graphene oxide (GO) was used with an initial concentration of 13.6 mg/mL mixed with Spindle oil to assess the dispersion, homogeneity, tribological performance, and effectiveness of this material as an additive in greases for automotive applications.
After preparing the dispersion, a high viscosity of the solution was visually observed. Tests were conducted to evaluate the viscosity and also tribological tests to ensure the quality of the material and its feasibility as a potential grease.

1 - LITERATURE REVIEW

1.1 – TRIBOLOGY

Tribology can be defined as a multidisciplinary field that encompasses all information related to lubrication, friction, and wear between contacting surfaces. It is the science that aims to study the performance and improvements of mechanisms such as bearings, rolling elements, gears, etc., through the optimization of aspects like kinematics, geometries, clearances, lubricants, materials, and surface treatments [1].

The selection of lubricants and the understanding of their behavior under different operating conditions are fundamental to ensuring system performance and durability. However, challenges remain, as the ideal combination of bulk and surface properties is rarely achieved by a single material [2,3].

Among the strategies to mitigate wear under specific operating conditions, solid lubricants have distinguished themselves as an effective solution, particularly in automotive applications [2].

Unlike liquid lubricants, materials such as molybdenum disulfide (MoS_2), graphite, and carbon-based compounds are capable of maintaining their effectiveness even in environments with high temperatures, extreme pressures, or in the absence of conventional lubrication. These materials form a film on the contact surfaces, reducing friction and preventing direct wear between components. In the automotive sector, solid lubricants are widely used in engines, braking systems, drive chains, and other components that require non-dripping lubrication and are exposed to harsh conditions. They are less sensitive to gravity, pressure, and centrifugal forces, contributing to increased durability, reduced maintenance needs, and optimized vehicle performance [2].

1.2 – GREASES

1.2.1 - GREASE COMPONENTS

Greases are lubricating substances with a consistency between semi-solid and solid, due to the addition of thickening agents. This consistency is their main characteristic, as it helps prevent the lubricant from dripping or being wasted from the application site. Most greases are produced by thickening a petroleum-derived mineral oil with a type of metallic soap [4].

Lubricating greases are complex compounds, mostly composed of a thickener, a base oil, and often additives that provide special Properties [4,5].

Although greases do not provide the same cooling effect as lubricating oils, they perform several essential functions in equipment lubrication. Due to their semi-solid texture, greases are able to remain longer on surfaces, even

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under adverse conditions, protecting against wear and corrosion. Additionally, they help minimize friction, prevent the ingress of contaminants such as dust and water, and maintain their structural stability throughout use, even at extreme temperatures. Their consistency also facilitates application, contributing to reliable performance during storage and operation. Another important aspect is that greases are compatible with seals and other components, and can tolerate a certain level of contamination without losing effectiveness [4,5].

Lubricating greases are essentially composed of three main elements: a base oil, a thickener, and additives. Among these components, thickeners play a fundamental role in forming the semi-solid structure of the grease, being responsible for providing the consistency that prevents the lubricant from flowing under normal operating conditions. Thickeners can be classified into three categories: metallic soaps, inorganic thickeners, and organic thickeners [4, 5].

Metallic soaps are the most common thickeners and are formed through the reaction between animal or vegetable fats and alkalis, such as metal hydroxides. This process is known as saponification. The type of metal used in soap formation - such as sodium, lithium, calcium, or barium - directly influences the final grease properties, including water resistance, operating temperature, and mechanical stability. For example, calcium soap-based greases exhibit higher water resistance, while those formulated with sodium soap are water-soluble [4,5].

In addition to simple base soaps, it is also common to use mixed base metallic soaps, in which two or more metals are combined to provide a balance between desirable properties such as water resistance and high-temperature performance. These mixed base greases are widely used when greater versatility in application and performance under severe conditions is required [5, 6].

The choice of thickener type is based on the specific application requirements, considering factors such as operating environment, exposure to water, thermal variations, and type of motion involved. Proper selection ensures that the grease performs its role efficiently, protecting mechanical components and extending equipment service life [7].

When discussing metallic thickeners, calcium, lithium, and sodium soaps stand out, as well as complex soaps - formed by combining simple soaps with aliphatic acid salts - which offer higher thermal resistance, enabling applications over wider temperature ranges [8].

When referring to inorganic thickeners, bentonite clay, silica gel, and carbon black are commonly encountered. These materials are employed due to their capability to operate at high temperatures. However, some, such as silica, may cause abrasive wear due to agglomeration. Organic thickeners, such as polyureas and synthetic fibers, stand out for their chemical stability and resistance to hydrocarbons, making them suitable for severe working conditions and elevated temperatures [9].

The base oil, the predominant component of grease (70 to 95% of the composition), can be of mineral or synthetic origin. Petroleum-derived mineral oils are widely used due to their good balance between performance and cost, but their selection depends on the application: high-viscosity oils are recommended for high loads, while low-viscosity oils are suitable for high speeds. Synthetic oils, such as silicones, polyesters, and phosphate esters, are preferred under extreme conditions due to their thermal resistance, oxidation stability, and ability to operate at very low temperatures. The correct combination of thickener type and base oil is essential to ensure component protection, extend system service life, and guarantee lubricant efficiency [10].

Regarding grease composition, synthetic fluids such as silicones, phosphate and silicate esters, polyalkylene glycols, among others, can be used when the objective is to operate under critical conditions. These fluids provide the grease with aging resistance, excellent low-temperature fluidity, and a high flash point [7,8].

Finally, to confer special properties applicable in various situations, whether extreme or not, additives are incorporated into grease formulations, such as graphite, molybdenum disulfide, among others. These additives can act as rust inhibitors, oxidation inhibitors, extreme pressure agents, anti-wear agents, dyes, and more [7].

Table 1 – NLGI Grade.

Fonte: ASTM International, 2021 [23].

NLGI GRADE	**Worked Penetration (25 ºC)**
000	445-475
00	400-430
0	355-385
1	310-340
2	265-295
3	200-250
4	175-205
5	130-160
6	85-115

Greases are classified according to their NLGI grade (National Lubricating Grease Institute), as shown in Table 1, through the worked penetration test. The method that specifies the procedures for measuring grease consistency using a penetrometer is ASTM D-217. The higher the NLGI number, the thicker the grease [11].

1.2.2 - ADVANTAGES OF USING GREASES

Several factors influence the properties of greases, such as pumpability, structural and mechanical stability, consistency and texture, dropping point, load capacity, protection against rust and corrosion, among others. Each criterion is linked to the grease composition. Greases provide better sealing compared to oils, effectively assisting in sealing by preventing the ingress of abrasive dust and water [4,5,7].

1.3 – GRAPHENE

1.3.1 -STRUCTURE AND PROPERTIES OF GRAPHENE AND ITS DERIVATIVES.

Graphene and its derivatives, such as graphene oxide (GO) and reduced graphene oxide (rGO), have been extensively studied as lubricant additives due to their exceptional tribological properties. Their two-dimensional structure enables the formation of ultrathin protective films on contact surfaces, reducing friction and wear. Although they all share similar lubrication mechanisms, they differ in solubility, electrical conductivity, mechanical strength due to variations in their chemical structures, and even lubrication methods. GO, for example, contains oxygenated functional groups that increase its solubility in polar solvents, whereas rGO exhibits better electrical conductivity and mechanical strength while retaining some of GO's enhanced solubility [12, 13].

1.3.2 – LUBRICATION MECHANISMS OF GRAPHENE.

The effectiveness of graphene as a lubricant additive is related to its ability to form tribofilms on contacting metal surfaces. The weak interlamellar forces between graphene layers allow easy sliding, resulting in low shear resistance. Additionally, graphene can fill microscopic imperfections on surfaces, providing smoother movement and reducing wear. These lubrication mechanisms are effective under various load, pressure, and temperature conditions, making graphene a versatile additive for grease applications [14, 15].

1.3.3 – IDEAL GRAPHENE CONCENTRATION IN GREASES.

Studies indicate that the concentration of graphene in grease significantly influences its tribological performance. Research has shown that the addition of 0.1 wt.% graphene, GO, or rGO in lithium greases may not be sufficient to form an effective tribofilm. Unlike what is observed in lubricating oils, higher concentrations in greases, such as 0.5 wt.% or 1 wt.%, are more effective. These studies demonstrated significant improvements in friction and wear reduction due to the formation of more robust protective films on contact surfaces [16, 17, 18].

1.3.4 – GRAPHENE DISPERSION METHODS IN GREASES.

Uniform dispersion of graphene in grease is crucial to maximize its tribological benefits. Due to the high viscosity of greases, methods such as direct ultrasonication may be ineffective. A common approach is pre-mixing graphene with the base oil, followed by incorporating this mixture into the grease, facilitating a more homogeneous distribution. Mechanical stirring, especially when combined with heating, is also used to improve graphene dispersion in high-viscosity greases [19, 20].

1.3.5 - INFLUENCE OF GRAPHENE PARTICLE SIZE.

The size of graphene particles directly affects tribofilm formation and, consequently, the lubricating performance of the grease. Research has shown that smaller graphene flakes tend to form more uniform and effective films, resulting in lower coefficients of friction and wear. Therefore, controlling the particle size of graphene is essential to optimize the tribological properties of lithium greases enriched with graphene [21].

1.3.6– IMPACT OF GRAPHENE ON THE GREASE MICROSTRUCTURE.

The incorporation of graphene in greases not only improves their tribological properties but also alters their microstructure. Studies have shown that the addition of graphene can increase the viscosity and elasticity of the grease due to the thickening of the grease structure, which also contributes to better adhesion to contact surfaces. These structural changes help prolong the lubricating action and highlight the need to study graphene's effect not only as an additive but also within the grease matrix [22].

2 - METHODOLOGY

The dispersed rGO used in the tests was produced and supplied by IME (Military Institute of Rio de Janeiro) at an initial concentration of 13,6 mg/ml. The base oil used in the tests was Spindle 60, a low-viscosity oil (40 ºC – Celsius = 10.33 cSt - Centistokes) provided by Vibra Energia S.A.

For use in the tests, it was necessary to reduce the rGO concentration from 13,6 mg/ml to a final concentration of 10 mg/ml (milligrams per milliliter). Thus, aiming for a total volume of 200 ml (milliliter) of the final formulation, the concentration calculation was performed, resulting in volumes of 147,050 ml of rGO and 52,941 ml of Spindle 60.

The homogenization of rGO in Spindle 60 was initially carried out by magnetic stirring for 15 minutes at room temperature. This initial mixture can be seen in Figure 1.

Figure 1 – Homogenization of the rGO/Spindle 60 dispersion after 15 minutes of magnetic stirring.

Source: Author.

Subsequently, washing by centrifugation was performed. The dispersion was centrifuged for 20 minutes at a temperature of 23 ºC, with 10,000 RPM, an acceleration time of 3 minutes, and a deceleration time of 15 minutes to ensure the integrity of the reduced graphene oxide sheets. The centrifugation washing process was repeated six times, discarding and replacing the oil in each wash. The purpose of centrifugation was to remove all water present in the dispersion originating from the rGO manufacturing process and to ensure better homogenization of the rGO sample with the base oil.

After the washing steps, it was verified that the rGO at a final concentration of 10 mg/ml was effectively homogenized in the base oil, imparting the sample with a viscosity typical of lubricating greases, as can be seen in Figures 2 and 3.

Figure 2 – rGO/Spindle 60 sample immediately after completion of centrifugation.

Source: Author.

Figure 3 – Visual appearance of the rGO/Spindle sample after completion of the centrifugation washing process.

Source: Author.

Tribological performance tests were conducted on the final dispersion, including the Four Ball Wear test and the drop point test. To determine the NLGI grade of the rGO/Spindle 60 grease, the worked penetration test (60 strokes) was performed.

3 - RESULTS AND DISCUSSION

1.3.1 – VISUAL ASPECT.

It was observed that, likely due to the solubility limit of the rGO dispersion in the base oil, not all solute was adsorbed by the solvent. Thus, it was verified that after the completion of the six washing cycles performed via centrifugation and subsequent oil removal, a residual amount of base oil remained in the dispersion.

This residue, as can be seen in Figure 2 where the phase separation due to the centrifugation process is evident, was discarded, and only the precipitate at the bottom of the vial was used for the tests.

This rGO/Spindle 60 sample was subjected to a visual stability test and remained stable, maintaining its initial form for a period of 10 months. This demonstrates that the grease production process using an rGO dispersion through the centrifugation washing method was visually effective.

1.3.2 – WORKED PENETRATION 60X – NLGI GRADE.

For the characterization of the consistency of the developed grease, the worked penetration test was performed according to the ASTM D217-21 standard, with the range of values shown in Table 1 [23]. The test was conducted at the Lubrax laboratories in Rio de Janeiro.

The test consisted of applying 60 worked cycles to the grease using a standard cone penetrometer, aiming to simulate the mechanical kneading that occurs during use. The obtained penetration value fell within the range corresponding to NLGI grade 2, indicating a grease with intermediate consistency, commonly used in a wide range of automotive and industrial applications. This result confirms that the formulation has a stable structure and rheological behavior compatible with typical market requirements, which correspond to the mechanical stresses the grease is subjected to that affect its physical properties [23, 24].

1.3.3 – DROPPING POINT.

The dropping point test is an assay that aims to determine the maximum temperature at which the grease being tested transitions from a semi-solid to a liquid state. In other words, this test indicates the highest temperature at which the grease remains stable, maintaining its structure before beginning to flow [25]. This test was conducted at the Senai Lençóis Paulista School in São Paulo.

Through the ASTM D566 method, it is possible to perform quality control between production batches, compare the thermal stability of different formulations, and also determine the classification of greases according to the type of thickener and their possible practical application [25, 26].

The method applied for the dropping point test on the rGO/Spindle 60 sample was ABNT NBR 6564:2009 – Lubricating Grease – Determination of Dropping Point, which is technically equivalent to ASTM D566 – Standard Test Method for Dropping Point of Lubricating Grease [26, 27].

The result obtained for the rGO/Spindle 60 sample according to the ABNT NBR 6564:2009 method was above 288 ºC, indicating excellent thermal resistance and making the grease suitable for applications in high-temperature environments. The incorporation of additives such as graphene, graphene oxide, and reduced graphene oxide has been associated with improvements in the thermal and tribological properties of greases. Recent studies have demonstrated that the addition of graphene to greases can significantly increase the dropping point, as well as enhance wear resistance and reduce the coefficient of friction when effectively dispersed [27, 28, 29].

1.3.4 – FOUR BALL WEAR.

The ASTM D2266 method is used to measure a lubricant's ability to prevent wear between sliding steel surfaces. The test consists of applying a standard load of 392 N (Newtons) to a rotating ball against three stationary balls, all immersed in grease. The assembly is rotated for 60 minutes at a speed of 120 RPM and a constant temperature of 75 °C. After the test, the average diameter of the wear scars on the stationary balls is measured, with the result expressed in millimeters. The smaller the wear diameter, the better the lubricant's performance [30].

Thus, the test indicates the effectiveness or ineffectiveness of the grease in protecting against wear, the integrity of the lubricant film, and the homogeneity and dispersion of additives incorporated into the lubricant formulation [30].

The wear scar diameter obtained in the Four Ball Wear test, conducted according to ASTM D2266, was 1.130 mm. This value is considered high, as typical values for high-quality greases range between 0.4 mm and 0.6 mm. Therefore, the test performed on the rGO/Spindle 60 sample indicates unsatisfactory tribological performance in wear prevention [30, 31].

This performance, which was lower than expected, can be attributed to the absence of anti-wear and extreme pressure additives in the grease formulation. Another factor to consider is the possible poor dispersion of rGO, which may lead to nanoparticle agglomeration, compromising the formation of an effective lubricating film. These factors are critical for wear protection, especially in applications involving high loads and high speeds [31].

4 - CONCLUSION

It is believed that, since the sample developed in this work did not contain extreme pressure and anti-wear additives in its formulation, this could explain the high value obtained in the Four Ball Wear test. Another relevant point is the sample homogenization, which may have contributed to the elevated result due to the agglomeration of rGO sheets [31].

Regarding the applicability of this grease, by maintaining the original composition and manufacturing process of the rGO/Spindle 60 sample, it may be suitable for use in applications that do not require high load capacity [31].

However, it is important to emphasize that this grease represents a preliminary technical feasibility test, and further complementary tests will be necessary to continue the research and to confirm the potential applicability of this product.

5 - FUTURE RESEARCH

In future studies to further investigate and confirm the feasibility of this grease, it will be necessary to reformulate the grease by adding the pre-prepared rGO dispersion in the base oil, employing ultrasonication to ensure better homogenization of the nanoparticles.

Another important aspect will be the execution of additional performance tests complementary to those conducted in this study, in order to ensure the quality and applicability of the product.

Finally, conducting tests with additive-enhanced formulations, such as the incorporation of extreme pressure additives and others, is necessary to ensure a broader range of applicability for this product and to obtain more conclusive results regarding its performance.

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