

## Uses of biodegradable lubricants, a challenge of today's societies

Andreea Mirela Teleasa<sup>1</sup>, Sorin Cananau<sup>1</sup> and Alexandru Valentin Radulescu<sup>1</sup>

<sup>1</sup>Machine Elements and Tribology Department, University POLITEHNICA of Bucharest, Romania

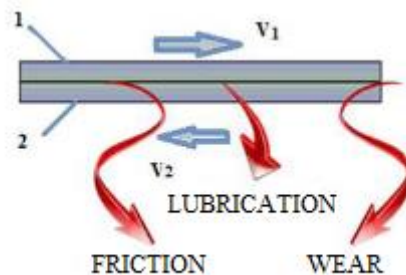
E-mail: teleasa.andreea@yahoo.ro

**Abstract.** In this paper, aspects regarding lubricants, their classification, their influence on the environment and their areas of use are presented. Lubrication represents an important process for various equipment and machines with moving parts. The correct choice of lubricant can lead to increased energy efficiency and at the same time decrease the rate of wear of various moving elements. We must take into account the fact that the choice of the lubricant requires special attention because at the moment a large part of the lubricants used are those based on mineral oils, which leads to damage to the environment, human health and high costs. According to the above, we can turn our interest to biodegradable lubricants (based on vegetable oils and animal fats), because they represent renewable and easily biodegradable resources.

**Keywords:** *biodegradable, lubricants, properties, viscosity.*

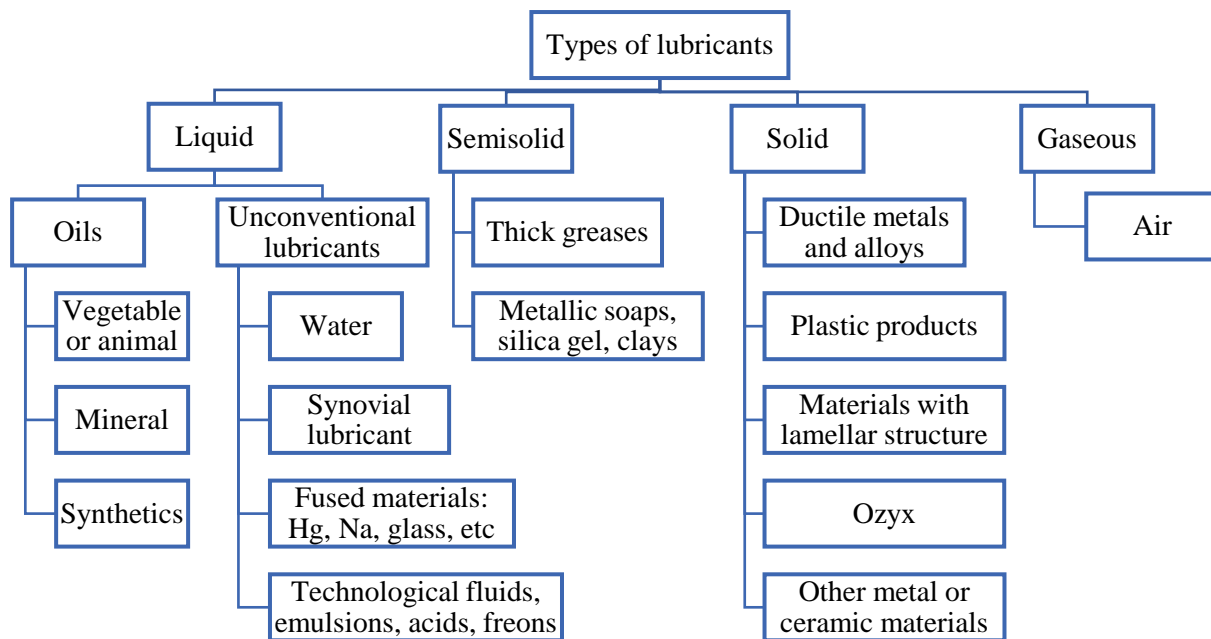
### 1. Introduction

According to the Standard ASTM Designation G40-95 [1], the lubricant is represented by any substance interposed between two surfaces in order to reduce friction and/or wear between them. Some authors [2], [3] state that the lubricant represents the third body interposed between the surfaces of the friction coupling. This may be naturally present or may be intentionally introduced to reduce friction and wear and/or to dissipate heat generated by friction. Although the term lubricant often suggests oils or thick greases, these being the most used lubrication materials, the notion of lubricant is much broader, being represented by a wide variety of substances in all states of aggregation [2]. Figure 1 shows the schematic representation of the interaction between two surfaces in relative motion.



**Figure 1.** Schematic representation of the interaction between two surfaces in relative motion with the highlighting of the three basic phenomena specific to tribology [3].

There is a growing interest in research studies on the use of oils from renewable resources as substitutes for mineral oils, but these are not on a large economic scale and the processing technology to the lubricant is not yet very cost-effective. Hence the interest of researchers to study their rheological and tribological behavior. Human activity since the beginning of society has required technologies and materials to reduce friction to transport materials in construction, agriculture, but also in other fields of activity [2]. Figure 2 shows the classification of lubricants.



**Figure 2.** Classification of lubricants [2].

## 2. Biodegradability of lubricants

Biodegradable lubricants have a long history. Specifically, natural oils and fats, being made up of unsaturated triglycerides, such as castor oil, palm oil, soybean oil, tallow, lard are historically well documented for their lubricating properties. However, their range of use is generally limited by lower stability against thermal and hydrolytic oxidative stress and partially inferior cold flow properties. These limits can be improved in several ways, for example by chemical or genetic modification or by additive [4].

The often-used prefix "bio" refers to an association with a renewable resource of biological origin, such resources generally excluding fossil fuels. Biodegradation is the decomposition or chemical transformation of a substance caused by organisms or their enzymes.

Biodegradability is not just a "property" or characteristic of a substance, but is also a system concept, i.e., a system with conditions that can determine whether a substance in it is biodegradable.

For the most part, when the term "biodegradable" is used to describe a lubricant product, it means that the product is more biodegradable than petroleum-based stocks or formulations [4].

The key to recipes for biodegradable lubricants is closely related to two aspects: the selection of the base oil and the selection of the additive package, which establishes an acceptable compromise in terms of biodegradability, performance and price. The main groups of base fluids that would meet the above requirements, which also have practical applicability, are synthetic esters, natural esters and polyglycols (these are also part of the classification of biodegradable lubricants) [2], followed by fatty oils, mineral base fluids, synthetic base fluids and complex esters:

- natural esters (vegetable oils) - such as those obtained from rapeseed, sunflower or soybean seeds);

- synthetic esters (esters of dicarboxylic acids or polyolesters) from mineral or vegetable base oils;
- polyglycols (especially polyethylene glycols) from mineral base oils.

### 2.1 Properties of biodegradable lubricants

The evaluation characteristics of lubricants are included in two main groups: physico-chemical characteristics and performance characteristics. The physico-chemical characteristics can be divided into two categories: recognition (commercial) and rheological. When designing, approving, manufacturing, choosing and recommending lubricants, these characteristics must be examined as a whole. From the product catalogs, norms and quality standards, information is obtained especially regarding the (commercial) recognition characteristics, this of course being subjected to quality guarantees. From the point of view of the manufacturer and the user, it is important to know more about the respective lubricants, namely, other characteristics related to their operating behavior [2].

Table 1 shows physico-chemical properties of vegetable oils.

**Table 1.** Physico-chemical properties of vegetable oils [5]–[9].

Oil type	Property								
	Density [kg/m <sup>3</sup> ]	Viscosity index	Kinematic viscosity at 40°C [mm <sup>2</sup> /s]	Kinematic viscosity at 100°C [mm <sup>2</sup> /s]	Saponification index [mg KOH/g]	Acid value [mg KOH/g]	Pour point [°C]	Flash point [°C]	Iodine value [mg I <sub>2</sub> g <sup>-1</sup> ]
Rubber tree seed oil	0.922	182	32.80	-	206	13	-9	295	135
Coconut oil	0.925	159	29.00	-	265	0.4	22	320	9
Sunflower oil	0.934	176	40.05	8.65	186	4	-18	332	127
Soybean oil	-	219	32.93	8.08	189	0.16	-9	240	144
Rapeseed oil	-	216	45.60	10.07	180	1.40	-12	240	104
Jojoba oil	-	233	24.90	6.43	94.69	1.10	9	-	98

Vegetable or natural oils are triglycerides of natural fatty acids, for example palmitic acid, stearic acid, oleic acid, vegetable oils, linolenic acid, etc. Due to the relatively high content of unsaturated fatty acids, natural oils tend to have low oxidation stability. A high unsaturated acid content is characterized by a lower iodine number which in turn tends to result in higher solidification temperatures. This means poorer cold flow behavior. Rapeseed oils present a fairly good compromise between low temperature behavior and oxidation stability [10]. Low temperature flow properties and oxidative stability and especially the relationship between the two are related to the fatty acid profile of the oil [11].

### 3. Impact of lubricants on the environment

Biodegradable lubricant formulation is a reversal from the current mineral oil dominated lubricant market. It is accepted that over 90% of all lubricants could be rapidly biodegradable. Currently, some countries in Western Europe and Scandinavia are the largest consumers of biodegradable lubricants. In 2005, in Germany, 5% of lubricating base oils were rapidly biodegradable esters (natural and synthetic). Over the years, the focus on lubricants has shifted from biodegradability to regenerability.

This influences the design of such products. Descriptors such as green or compatible lubricants suggest that there is no interaction with the environment. Since any oily substance stains the soil, it has been suggested that the term "environmentally friendly lubricants" expresses minimal damage and danger to nature [12], [13].

### *3.1 Driving forces for reducing environmental impact*

Ways and measures to reduce environmental damage that mean overcoming or at least reducing the problems caused by the contact of lubricants with the environment in general are initiated or intensified by the following driving forces [11]:

- Environmental facts;
- Public awareness;
- Government directives and regulations;
- Globalization of markets;
- Economic incentives.

Bartz [11] pointed out that no lubricant can be truly environmentally friendly (i.e. not harming the environment); at best, a lubricant can remain environmentally neutral (i.e. harmless). The best that can be hoped for is that the lubricant affects the environment to an acceptable degree (minimum impact).

Consideration of the environmental aspects of lubricants should cover the entire life cycle from production to disposal. In particular, the production and use of lubricants should not cause resource depletion, disposal problems, waste formation or emissions, should have low energy consumption, should not present health hazards, should be ecotoxicologically acceptable and rapidly biodegradable.

The terminology used in relation to "environmental compatibility" is both subjective (unmeasurable) and objective (measurable). The subjective criteria are ecological and compatible with the environment. The objective criteria for "bio lubricants" are biodegradability of at least 60% according to OECD 301 (aerobic biodegradation test) or 80% according to CEC L-33-A-93 (test method for biodegradability) [4].

## **4. Fields of use of biodegradable lubricants**

Lubricants are used in various industrial applications. The most important categories in which lubricants are used are [14]:

- Oils used in industrial applications such as machine oils, compressor oils, metalworking fluids and hydraulic oils, etc.;
- Automotive oils used in the automotive and transportation industry, such as engine oils, gears, transmission fluids, gear oils, brake and hydraulic fluids, etc.;
- Special oils such as process oils, white oils, tool oils, etc.

Biodegradable lubricants offer significant advantages as an alternative lubricant for industrial and maintenance applications due to their inherent superior qualities. Due to the environmental benefits of biodegradable lubricants, they allow their use in sensitive environments and ensure pollution prevention. Biodegradable lubricants have the ability to be used in various industrial and maintenance applications [15].

### *4.1 Experimental studies of biodegradable lubricants for different applications*

Ibrahim et al. [16] carried out a study in which they experimentally analyze the performance of sunflower oil and soybean oil as lubricants for helical gears. The machine used for the experimental testing was a gear test rig. Oil input data for the tests were taken periodically from the gear test facility over 80 consecutive hours. Oil performance properties such as kinematic viscosity were measured immediately after oil sampling using Brookfield LVDV-II with PRO digital programmable viscometer. The kinematic viscosity parameter was measured in cSt units at temperatures of 40°C and 100°C using the ASTM D445-06 method [17]. All measured data were subsequently compared with the SAE 75W synthetic gear lubricant brand Renolin B10 [16]. Following experimental results show that sunflower oil, soybean oil and Renolin B10 have similar viscosity properties at high temperatures, but soybean oil has poor properties at low temperatures compared to sunflower oil. At low temperatures, soybean oil is less viscous, which can affect the contact surface between the two gears. The oil will allow the surface to contact each other and cause friction to increase and damage to the gear surface will occur. This can lead to reduced gear life in low temperature applications [18].

According to the above, we can say that sunflower oil is suitable as a lubricant for bevel gears compared to soybean oil which is in the SAE 75W classification. By comparison, sunflower and soybean oils have poor low temperature properties and have a wide range of viscosity over a temperature range that indicates the oil will not perform well throughout. However, both sunflower and soybean oils can perform well at higher temperatures [16].

In 2011, Majdan et al. [19] carried out an experimental study in which they present a comparison of the quality of two fluids. The first was the UTTO type mineral oil that is commonly used in the transmission and hydraulic systems of agricultural tractors. The second fluid tested was a biodegradable hydraulic fluid of the ERTTO type that could replace toxic mineral oil. Both fluids were tested under the same test conditions using a special test device according to STN 11 9287 (1983). Selected hydrostatic pump parameters were evaluated. The tests were evaluated according to the parameters that describe the technical condition of the hydrostatic pump as follows: the flow efficiency, the decrease in the flow efficiency and the level of cleanliness of the tested fluid. Based on the results obtained, we can state that the biodegradable hydraulic fluid does not exert any harmful influence on the technical condition of the hydrostatic pump. Therefore, the tested biodegradable fluid can be used for agricultural tractors. The selected parameters have been shown to be suitable for evaluating the hydraulic fluid during its working performance.

Permsuwan et al. [20] conducted tests to determine the effect of vegetable oils on piston deposits and cylinder bore deposit formation, engine oil consumption and viscosity stability when used in a gasoline engine on a period, representing the normal oil change period (i.e., 7 hours with a low tank refill compared to 250 hours of full tank operation). The method used to evaluate the degree of varnish deposition was Demerit Rating (DR), which is a standard method of the Petroleum Institute [21].

Several vegetable oils have been tested with this technique and satisfactory results have been obtained. The vegetable oils selected for testing were: R30 castor oil (which is a high-performance castor base oil), crude castor oil (which is unprocessed and toxic), and pure coconut oil. The tests were carried out on a Villiers C-30 industrial gasoline engine. The motor load was supplied by a coupled generator having an adjustable electrical resistance. The duration of the test for each oil was 7 hours [20]. The viscosity of the tested vegetable oils was raised using this type of test, namely, R30 castor oil (which is a commercial high-performance race car oil), its viscosity increased by 37% during the 7-hour test period and coconut and castor oil could be used as engine lubricants in the test engine. During testing, the engine ran with these oils without any problems or breakdowns. The results show that all vegetable oils protect the piston jacket from lacquer deposition. Crude castor oil gave the best result as there was no deposit on any area of the piston except the crown. Coconut oil also gave a good result with little deposit in the first ring groove. Coconut oil crown deposition was the lowest [20].

Lovell et al. [22] conducted an experimental study in which they introduced and analysed a new environmentally friendly lubricant to determine its potential use in sheet metal stamping processes. This lubricant is based on a combination of boric acid and rapeseed oil, both of which are natural, environmentally friendly and have independently demonstrated good lubrication potential. To evaluate the frictional characteristics of rapeseed oil and boric acid lubricant in a forming operation, a strip stretch friction simulator was used. Using the test apparatus, the coefficient of friction between the mold and a steel sheet was measured for four different lubrication conditions: unlubricated, canola oil, transmission fluid, and a combination of boric acid and canola oil. The author demonstrated that this apparatus effectively models the deformation behavior encountered in a deep drawing process and accurately reproduces the frictional effects experienced during sheet bending. Based on the experimental results, the boric acid and canola oil lubricant significantly outperformed the other lubrication conditions in terms of measured friction coefficient, final sheet surface properties, and overall sheet formability. These results indicate that boric acid and canola oil lubricant has substantial potential to provide the manufacturing community with a commercially reliable and environmentally friendly alternative that will enable the forming of complex parts.

In their paper, A.V. Radulescu et al. [23] determined the main rheological properties of pork fat, which is one of the most important sources of animal fat waste in the food industry. From this point of

view, two kinds of pork fat were tested: pork fat from the back of the neck and from the breast. The measuring equipment for the tests was a Brookfield Cap 2000+ rotary cone and plate viscometer. The viscometer uses CAPCALC32 software for complete control and data analysis. The viscometer is suitable for digital data acquisition and provides the ability to determine the variation of viscosity as a function of temperature. All measurements can normally be made in a temperature range between 5 and 75°C [24], [25]. Three types of tests were performed for these greases: shear tests, time influence on viscosity and temperature performance.

Following the experimental results, the conclusions are as follows [23]:

- The rheological properties of pork fat depend on the portion of the pork body from which they were collected;
- Pork fat has an important thixotropic behavior, due to the breaking of the structure when shearing at a given rate;
- The transition area, between semi-plastic and fluid behavior, is accentuated for a range of temperatures between 25°C and 35°C;
- The appearance of pork fat, in a fluid state, depends on the portion of the pork body from which it was collected.

The paper of I. Radulescu [26] is focused on the rheological study of biodegradable fats based on rapeseed oil and beeswax, added with graphene or graphite nanoparticles. For this study, 4 types of grease samples were prepared, namely: pure grease, additive grease, with antioxidant (1%) and antiwear additives (2%), graphene grease, with antioxidant (1%), antiwear (2 %) and graphene powder (0.5%), graphite grease, with antioxidant (1%), antiwear (2%) and thermally expanded graphite (0.5%). Each of them was thermally analyzed in the temperature range of 20°C...50°C. Rheological tests were performed using a Brookfield CAP 2000+ viscometer with cone and plate geometry. The experimental test consists of a loading from 10 s<sup>-1</sup> to 2000 s<sup>-1</sup> shear rate gradient, followed by an unloading to highlight the thixotropy of the lubricant - "shear memory". The experimental results show that the addition of nanoparticles increases the homogeneity of the grease and increases the range of use of biodegradable lubricants up to the melting point of beeswax. The highest values of the yield point and viscosity are obtained for the grease added with graphite nanoparticles, compared to the one added with graphene nanoparticles. From the adhesion point of view, graphite nanoparticle grease is more suitable than graphene nanoparticle grease.

## 5. Viscosity index

### 5.1 Variation of viscosity with temperature for vegetable oils

The viscosity index (VI) is an arbitrary measure of the change in viscosity of a fluid in relation to a change in temperature. It is also defined as the dimensionless number that indicates how changing temperature can affect the viscosity of an oil (engine oil and automatic transmission oils and power steering fluids). The higher the VI, the smaller the change in fluid viscosity for a given change in temperature, and vice versa. Thus, a fluid with a low viscosity index will experience a relatively large change in viscosity as the temperature changes. Liquids with high VI, on the other hand, are less affected by temperature changes [27].

The ASTM D2270 standard calculates the viscosity index by measuring the kinetic viscosity of oils at 40°C and 100°C [27] using the relationship below:

$$VI = \frac{L - U}{L - H} \times 100, \quad (1)$$

where [28]:

- VI = viscosity index;
- L = kinematic viscosity at 40°C of an oil of 0 viscosity index, having the same kinematic viscosity at 100 °C as the oil whose viscosity index is to be calculated, mm<sup>2</sup>/s;

- H = kinematic viscosity at 40°C of an oil of 100 viscosity index, having the same kinematic viscosity at 100 °C as the oil whose viscosity index is to be calculated, mm<sup>2</sup>/s;
- U = kinematic viscosity at 40°C of the oil whose viscosity index is to be calculated, mm<sup>2</sup>/s.

Another indication of the change in kinematics viscosity with temperature, which is less arbitrary than the viscosity index, is the viscosity-temperature coefficient (VTC) defined by the relationship [29]:

$$VTC = \frac{A - B}{A}, \tag{2}$$

where:

- A = viscosity (mm<sup>2</sup>/s) at 40°C;
- B = viscosity (mm<sup>2</sup>/s) at 100°C.

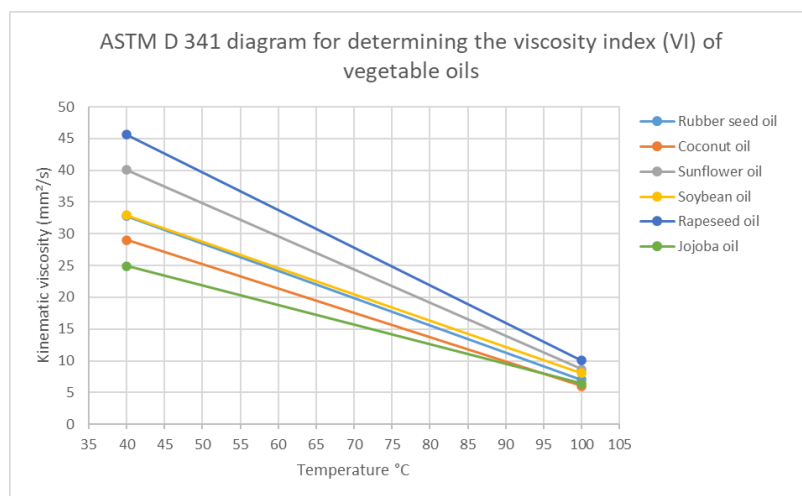
**Table 2.** Experimental results of biodegradable oils regarding viscosity index and temperature-viscosity coefficient

Oil	Kinematic viscosity at 40 °C (mm <sup>2</sup> /s)	Kinematic viscosity at 100 °C (mm <sup>2</sup> /s)	Viscosity index (VI)	Viscosity temperature coefficient (VTC)
Rubber seed oil	32.80	7.02	184	0.7860
Coconut oil	29.00	6.02	161	0.7924
Sunflower oil	40.05	8.65	203	0.7840
Soybean oil	32.93	8.08	234	0.7546
Rapeseed oil	45.60	10.07	216	0.7792
Jojoba oil	24.90	6.43	231	0.7418

Table 2 shows experimental results regarding the characteristics of biodegradable oils (rubber seed oil, coconut oil, sunflower oil, soybean oil, rapeseed oil, jojoba oil) together with the kinematic viscosity at temperatures of 40°C and 100°C corresponding to each oil, with which the viscosity index could be determined using relation (1) and the temperature-viscosity coefficient using relation (3).

From Table 2 it can be seen that the values for VTC are much lower than those of VI, so we can see that the highest value for VTC is represented by coconut oil, and the lowest by jojoba oil. From the point of view of VI, coconut oil has the lowest VI, and the highest is represented by soybean oil.

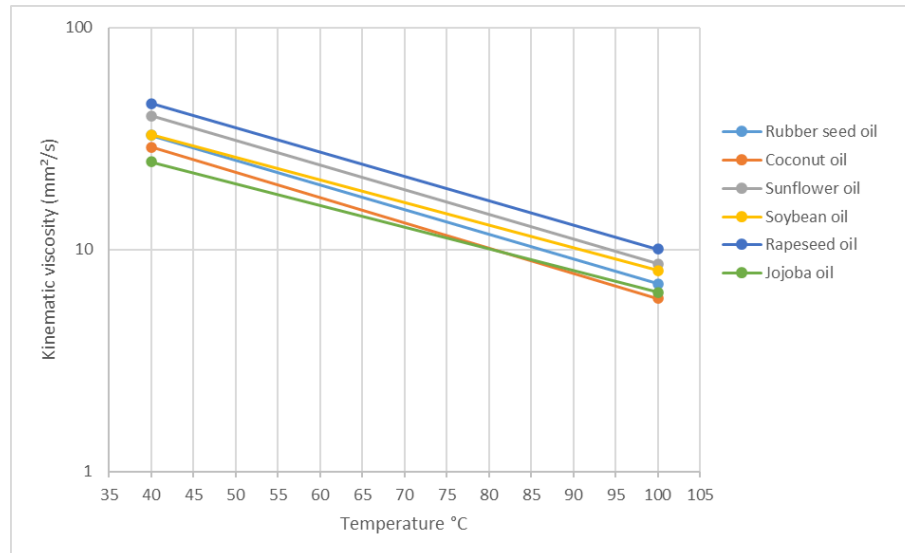
The viscosity index could also be determined using the ASTM D 134 chart, as can be seen in Figure .



**Figure 3.** ASTM D 341 diagram for determining the VI viscosity index of vegetable oils

Kinematic viscosities for temperatures of 40°C and 100°C were plotted in Figure 4 for the six oils. These points were joined by lines for each component separately. The lower the viscosity index, the less dependent the viscosity is on temperature variation.

From Figure 4 it can be seen that jojoba oil has the smallest slope, which means that the viscosity variation with temperature is smaller.



**Figure 4.** ASTM D341 chart for determining the viscosity index (VI) of vegetable oils with logarithmic kinematic viscosity

## 6. Conclusions

Following the theoretical research carried out in this first report, it was found that biodegradable lubricants are of great interest both from the point of view of the environment and for their use in agricultural, food, hydraulic machinery, etc., but also their testing to identify properties and influencing factors. For these biodegradable lubricants, vegetable oils, polyglycols and synthetic ester oils can be used as base oils. Replacing petroleum-based lubricants (which are an exhaustible resource and at the same time polluting the environment) with vegetable oils will represent a renewable resource over time.

The viscosity of biodegradable oils varies with their temperature, as the viscosity decreases, the temperature increases because the molecules vibrate more and interact less [29].

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