Chapter

Grease Lubrication: Formulation Effects on Tribological Performance

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Abstract

Grease lubrication performance prediction is challenging. Only recently that empirical equations to predict grease film thickness for prevailing rolling conditions under fully flooded lubrication taking into account thickener properties and content for low, moderate, and high speeds were developed. At starved lubrication, although new insights about the supply and loss mechanisms that govern film formation have been published, contact replenishment and, consequently, film thickness predictions for long-term operation are still not available. Prediction of components efficiency requires film thickness values and properties, including film's molecular structure, which makes it even more challenging. When it comes to prevailing sliding conditions, the literature is scarce and most of the knowledge developed for prevailing rolling conditions is not applicable. During the sliding of the contacting bodies, boundary and mixed lubrication regimes are expected. In this situation, the tribological response is primarily defined by grease thickener and additives physicochemical interaction with the surface. This complexity leads many researchers to seek simpler relationships between grease formulation and properties with its performance. This review aims to present the state-of-art on grease lubrication and update some of these relationships.

Keywords: grease components, grease properties, grease performance, friction, wear, rolling contact fatigue

1. Introduction

The most common definition of lubricating grease is the one put forward by ASTM D 288: "A solid to semifluid product of a thickening agent in a liquid lubricant, other ingredients imparting special properties may be included" [1]. This means that grease is a thickened (not a thick) oil. It also states that lubricating grease is based on a multiphase system consisting of at least two well-defined components, namely a thickener and a lubricant fluid. The "other ingredients" refer to additives. The distribution of these components depends on the application but typically is 65–95 wt % base oil, from 5 to 35 wt % thickener, and from 0 to 15% additives. These separate components form a multiphase matrix, in which the thickener forms a structure that holds the base oil and the additives. This multiphase matrix endows the grease with a certain consistency and a yield pseudoplastic behavior that gives it many advantages over lubricant fluids, such as ease of use (it will not easily leak out due to its consistency), inherent sealing action, low friction, and protection against corrosion [2]. However, it also makes it quite difficult to characterize or model the individual influence of each of its components on the grease overall performance because their effectiveness depends on their interaction [3, 4], manufacturing process [5, 6], and system operating conditions, as observed with calcium sulfonate thickened greases that show excellent wear resistance in pure sliding tests [7, 8] but reduced rolling contact fatigue life in prevailing rolling conditions [9, 10].

The synergic effect among grease components (base oil, thickener, and additives) along with its properties dependency on the manufacturing process, pose significant challenges on modeling grease behavior in a tribological system. Consequently, there are very few scientific works modeling grease behavior as a multiphase fluid [11, 12], and most of the experimental work focuses on the comparison between grease performance with its composition or properties.

The available work modeling lubricating greases as a multiphase fluid is very promising and may bring significant new insights about grease performance, such as the grease lubricating phase (variation of volume fraction distribution of oil and thickener in the contact) as a function of time and position [11], and the rheological changes of lubricating greases as a function of thickener distribution and deformation [12]. Unfortunately, such models were not yet employed to compare different grease formulations with their flow and tribological responses. Analytical solutions to predict grease-lubricated rolling bearing life, such as ISO 281, still do not take into account the thickener type, although grease response depends on it.

The experimental work evaluating grease performance is abundant under operating conditions that resemble the ones from rolling bearings (low slide to roll ratio, SRR), which are the main grease application. Tests have been performed over the years by many researchers under such conditions with several lubricating greases. Their main outcomes—film thickness, friction, wear, and rolling contact fatigue—have been related to grease characteristics. By today, it is well known that under fully flooded conditions, grease does not always follow the film thickness predicted by the traditional elastohydrodynamic lubrication (EHL) theory [13] nor the coefficient of friction is depicted by the Stribeck curve [14]. This divergence is attributed to the presence of thickener in the contact [15] and depends on the operating conditions, in particular the lubrication regime [16]. In fact, it has been shown that thickener changes film thickness [13], coefficient of friction [14], crack propagation rate [15], and play a significant role in the lubricant supply to the contact under starved lubrication [17]. Although a good agreement between thickener content and morphology with film thickness measurements under fully flooded conditions was observed [13], its effect on friction, wear and crack propagation is still not well known [15].

Different from prevailing rolling contacts, when it comes to grease evaluation under pure sliding conditions, the literature is scarce and the effect of thickener and base oil on grease tribological response is unknown for most grease formulations. At typical concentrated sliding contact conditions, hydrodynamic lubrication is not prone to occur and the lubrication regime is the boundary or mixed [18]. In this situation, the thin film covering the surfaces is mostly composed of thickener and/or additives. Consequently, the thickener used to play a more significant role than the base oil viscosity on grease performance [19]. Most of the existent work under sliding conditions focuses on the development and evaluation of extreme pressure/anti-wear (EP/AW) additives and nanoparticles to control friction and wear [20–22]. Studying the effect of one specific additive or nanoparticle on grease performance is complicated by the presence of other additives and it is often most appropriate to consider the additive package since one additive can have a synergistic or an antagonistic effect on another one [20]. Furthermore, the additives performance also depends on the thickener type, which interferes with their mobility toward the surface through chemical interactions or attractive forces [21]. Consequently, it is difficult to observe general trends for the tribological response of a specific additive. Besides that, only general information about the additive package is provided by grease manufacturers.

In order to clarify the role of thickener type and base oil viscosity on grease film thickness, friction and wear, this review presents a compilation of the literature for prevailing rolling and prevailing sliding conditions through schematic graphics along with a ranking of the thickeners and base oils that contribute the most on the tribological response of the greases.

2. The role of thickener and base oil in prevailing rolling contacts

Lubricating grease is mostly used in prevailing rolling contacts, such as in rolling bearings and gears [23]. Therefore, most of the knowledge on grease lubrication is related to tests performed under the low slide-to-roll ratios. In these operating conditions, lubricating greases are prone to form elastohydrodynamic films that separate the contacting surfaces, and therefore, the study of film formation, friction, and rolling contact fatigue is of particular interest. The tests reported in the literature can be divided into many ways. Here, they are divided into two ways—fully flooded and starved contacts.

2.1 Fully flooded conditions

The large majority of grease film thickness and coefficient of friction measurements reported in the literature comes from the ball-on-disc experiments using a device that forces the grease back to the contact and ensures a fully flooded condition. A schematic view of this device is well depicted in [24]. In this situation, lubricating greases present different behavior than lubricating oils. **Figure 1** presents typical center film thickness measurements as a function of the entrainment speed for a lubricating grease and its base oil. The film thickness increases proportionally to entrainment speed ($h_{ff} \propto U^{0,67}$) for the oils, as predicted by EHL theory

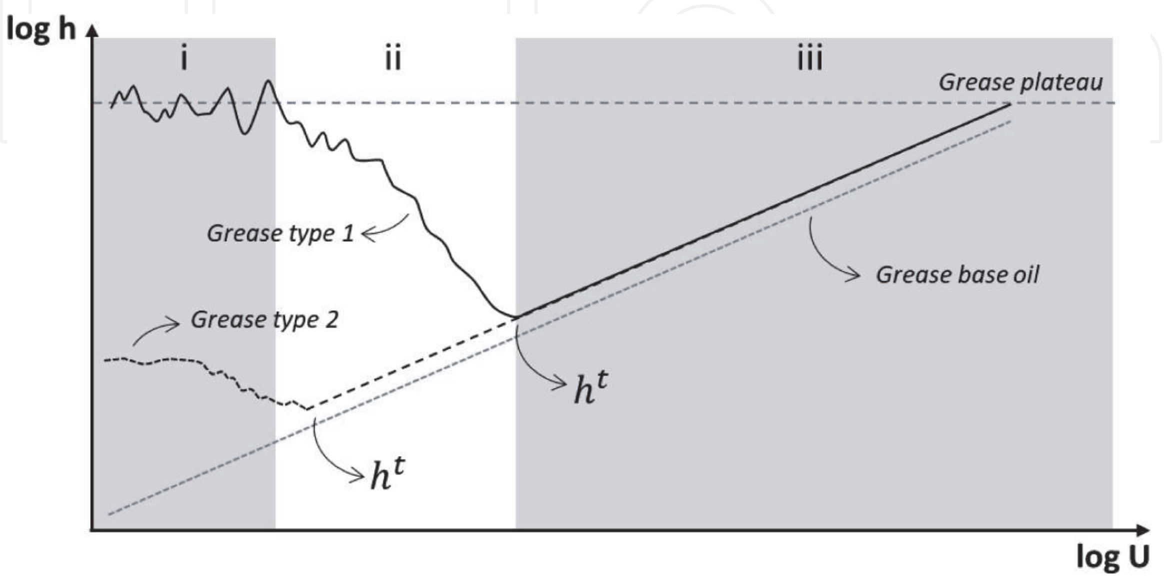

Figure 1.
Typical film thickness versus entrainment speed curves for grease and its base under fully flooded lubrication and low SRR.

for point contacts and fully flooded lubrication (Eq. (1)) [25], while lubricating greases present the well-known "V-shape" curve [15]. That difference is attributed to the thickener effect on film formation. At medium to high speeds grease presents the same behavior of its base oil (region iii) and used to be modeled with Eq. (1), although in general, it presents slightly higher film thickness. The difference was found to be proportional to the ratio of the thickener concentration (φ) to the average volume of the individual thickener particles (V), as shown by Eq. (2), which presented a good agreement with experimental results and better predicts lubricating grease film thickness [13]. The grease film thickness predicted with Eq. (2) was found to be 3–40% higher than the ones predicted with Eq. 1 [14], which shows the importance to take into account the thickener on film thickness prediction.

$$h_{ff} = 2.69 R_x \left(1 - 0.6 e^{-0.73k}\right) (E'\alpha)^{0.53} \left(\frac{\overline{u}\eta_0}{E' R_x}\right)^{0.67} \left(\frac{F}{E' R_x}\right)^{-0.067} \tag{1}$$

$$h_g = h_{ff} \left[\left(0.61 \left(\frac{\varphi}{V}\right) + 1.97 \right) + 1 \right] \tag{2}$$

As the entrainment speed is reduced, a transition occurs (h^t) and the grease does not follow the conventional EHL theory anymore (region ii). In this region, the film thickness is dominated by thickener material trapped on the contact interface and increases as the entrainment speed is further reduced. At very low speeds most lubricating greases reach a plateau with very high film thickness (region i). In these regions (i and ii), the measurements fluctuate significantly due to the thickener material crossing the contact [26, 27], and qualitative relationship among film thickness and thickener content, morphology, and properties have been presented [26–28]. The observed transition (h^t) occurs, for each type of grease, at a different specific film thickness [26]. Kanazawa et al. [16, 29] showed strong evidence that (h^t) is related to the ratio between film thickness and thickener particles size, and that colloidal nanoparticle dispersions present the same behavior. This should lead to new research lines in which grease film thickness must be modeled as a multiphase fluid and thickener characteristics should be taken into account. While such research lines are emerging, engineering solutions have been proposed. Morales et al. [30] modeled the trends observed in regions i and ii with good agreement with the experimental results. To do so, first film thickness measurements h_g are used to estimate the effective grease viscosity by solving Hamrock and Dowson equation for the dynamic viscosity η_0 at every speed $\overline{u}$ value (Eq. (1) [25]). The grease effective viscosity $\eta_{g.eff}$ is then approximated by a simple model described by Eq. (3), in which the constants A and f are obtained by applying a collocation method using one speed below and one at the transition (h^t). Finally, the grease effective viscosity ($\eta_{g.eff}$) is used instead of the grease base oil viscosity (η_0) in Eq. (1) to predict film thickness.

$$\eta_{g.eff} = \eta_{0.oil} [\coth(A\overline{u})]^f \tag{3}$$

These empirical equations are the state-of-art to predict grease film thickness under fully flooded conditions using analytical solutions for the three regions presented in **Figure 1**. At the oil-dominated region (iii), the film thickness can be reasonably predicted using only the base oil type and viscosity (Eq. (1)), which are provided by grease suppliers, although more accurate predictions can be done if thickener morphology and content are taken into account (Eq. (2)). At the thickener-dominated regions (i and ii) film thickness can only be predicted if prior film thickness measurements are carried out (Eq. (3)). Since thickener characterization and film thickness measurements are, in general, not available for end users, several researchers have presented qualitative relationships between grease type and film thickness that are quite useful [14, 26, 28, 30–35]. These observations show that—(i) mineral oils (M) present higher film thickness values than synthetic oils (S) with the same viscosity due to their greater pressure-viscosity coefficient [25]; (ii) film thickness increases with thickener content [36]; (iii) in general, thickeners that enhance grease film thickness the most in comparison to the corresponding base oil for lubricating greases formulated with mineral oils, above and below the transition (h^t), are as follows—calcium (C) > polyurea (Pu) > lithium (L); (iv) for lubricating greases formulated with synthetic oils—polypropylene (P) > lithium complex (Lc) > polyurea (Pu). Although lubricating greases formulated with synthetic oils present similar behavior, PAO-based greases present higher film thickness than ester-based greases. This response is attributed to the thickener structure, which is smaller for ester-based greases processed with the same thickener type [37].

From the aforementioned summary, in **Figure 1**, “grease type 1” refers to PS, LcS, and CM, while “grease type 2” refers to LM and PuM. However, it is important to stress that this general trend might not be always observed. Depending on the combination of thickener and base oil types, along with the manufacturing process, the thickener morphology changes [3, 6, 38], and consequently the film thickness response also changes [37].

The operating conditions also affect the trends presented in **Figure 1**. Temperature variation affects grease base oil viscosity, which in turn affects film thickness in a similar manner as the entrainment speed does. Therefore, the transition (h^t) occurs at higher speeds with an increase in temperature, but its value stays fairly constant [39]. Load and slip ratio variation were shown to have a minor effect on film thickness values, as predicted by standard EHL theory [39].

Figure 2 presents the typical coefficient of friction measurements as a function of the entrainment speed for lubricating grease and its base oil. The base oil coefficient of friction follows the Stribeck curve, which is characterized by high friction values at low speeds (low specific film thickness) due to asperities contact (region i), followed by a reduction of the coefficient of friction as the speed increases (region ii), up to a point in which the friction does not vary significantly (region iii).

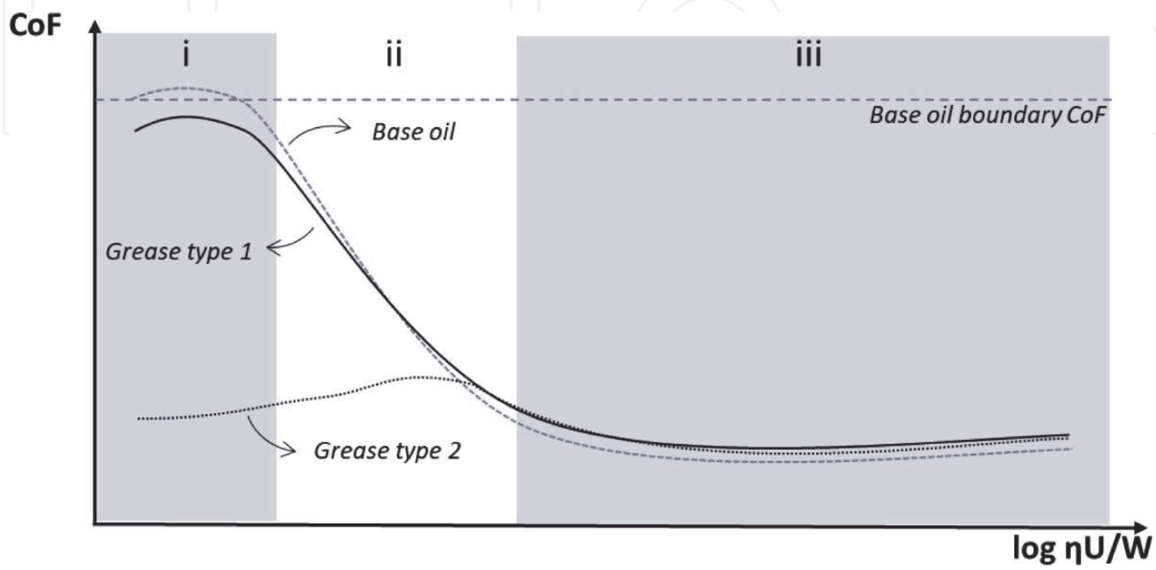

Figure 2.
Typical coefficient of friction versus entrainment speed curves for grease and its base oil under fully flooded lubrication and low SRR.

Different than their base oils, some lubricating greases do not follow Stribeck curve and present friction reduction as the speed is reduced from region iii to region i. That difference is also attributed to the thickener. At high speeds (region iii) greases and their base oils present the same behavior, although lubricating greases used to present friction values slightly different (lower) than their base oils. At low speeds (regions i and ii), at the thickener dominated region, two different trends are observed—(i) lubricating greases "type 1" follow the Stribeck curve and might present slightly higher friction values than their base oils as the speed is reduced; (ii) lubricating greases "type 2" present moderate friction increase followed by its stabilization or reduction as the speed is reduced. It is very important to mention the coefficient of friction does not necessarily reduce as film thickness increases. This is well depicted by calcium greases, which present very high film thickness, but also a high coefficient of friction, at the low-speed region [14, 15]. This occurs because the friction depends on the film properties and does not only on its thickness, and therefore, the typical coefficient of friction versus specific film thickness curves observed for base oils are not always observed for greases.

The grease coefficient of friction under EHL conditions depends on grease film thickness, thus, all the uncertainties around film thickness prediction are observed for the coefficient of friction prediction. Besides that, while film thickness formation is mostly governed by two "macro" properties above the transition (h^t)—ambient pressure viscosity η_0 and pressure viscosity coefficient α, friction has its origins in the lubricant molecular structure [40], including molecular shape and flexibility [41], which is obviously a much more complicated matter. Therefore, most of the published work focused on the qualitative relationship between grease type and friction [14, 16, 27, 28, 32, 35, 37, 42]. The summary of these works shows that—(i) mineral oils present a higher coefficient of friction than synthetic oils with the same viscosity due to their greater pressure-viscosity coefficient, which seems to be related to their limiting shear strength and viscosity index [14, 37]; (ii) greases formulated with PAO base oils seem to provide the lowest friction followed, in order, by ester, mineral paraffinic, and naphthenic base oils [43–46]; (iii) the higher the thickener content the smaller is the CoF for lubricating greases of "type 2" [35, 47] (see **Figure 2**) due to the increased probability of thickener crossing the contact and depositing on the surface; (iv) the low-pressure rheological properties (viscous and viscoelastic) are not the main influential factor in determining friction [48]; (v) in general, thickeners that reduce the coefficient of friction the most in comparison to the corresponding base oil for lubricating greases formulated with mineral oils are as follows—lithium (L) > polyurea (Pu) > calcium (C); (vi) for lubricating greases formulated with synthetic oils—polypropylene (P) > lithium complex (Lc) > polyurea (Pu).

Based on the aforementioned summary, in **Figure 2**, "grease type 1" refers to CM, PM + S, LM, while "grease type 2" refers to PS, LcS, and PuS. However, as mentioned before, the coefficient of friction depends on the properties of the separating film, which in turn depends on the combination of thickener, base oil, and the manufacturing process, hence caution should be taken while using these qualitative rankings.

The trends presented in **Figures 1** and **2** are of utmost importance for grease selection because rolling bearing life and efficiency are directly related to film thickness and friction. The current procedures for grease selection, based on ISO 281, and rolling bearing efficiency, based on tools provided by bearing manufactures [49], only take into account grease base oil viscosity. These current practices would lead to the selection of lubricating greases with very high base oil viscosity to ensure adequate lubrication (high specific film thickness—region iii of **Figures 1** and **2**) for rolling bearings operating at low speeds, which are usually observed in large-size bearings. However, high viscous greases used to present very low bleeding properties which can generate premature failure due to starvation [50], as discussed in the next section. An alternative is the selection of lubricating greases that generate high film thickness and low friction under low entrainment speed (low specific film thickness), such as lubricating greases formulated with PAO base oil and thickened with polypropylene or lithium complex (PS or LcS). Obviously, other parameters, out of the scope of this chapter, should be verified, such as environmental and operating factors.

Similar trends presented in **Figures 1** and **2** are also observed in full rolling bearing tests [27, 29, 30, 51–57] under fully flooded conditions or a short period of test, situations in which starvation and grease degradation do not take place. Under these operating conditions, if the rolling bearing is properly selected, the main source of failure is fatigue or grease degradation. In case of fatigue, a thickener type that has a high affinity with the bearing material and a morphology resembling small particles, such as calcium sulfonate, is more likely to enter the pre-formed cracks and propagate them faster, leading to severe wear [15].

2.2 Starved conditions

Although most of the published studies on grease lubrication are performed under fully flooded conditions, it is well known that most greased components, such as rolling bearings, operate under starved conditions. Therefore, it is crucial to understand the key aspects controlling the lubrication process when the lubricant is not available in abundance to the contact. Starved conditions are usually evaluated in ball-on-disc test rigs without the use of a device to channel the grease back to the contact or in long-duration rolling bearing tests. Generally, these tests are performed at a constant speed to evaluate grease film thickness variation over time, which is governed by competing for lubricant supply and loss mechanisms. This balance, which is not constant in time and can present a chaotic behavior [58], has not been modeled yet due to its dependence on a number of factors, such as—(i) centrifugal forces [59, 60]; (ii) surface tension [60, 61]; (iii) capillary forces [62]; (iv) contact forces between contacting bodies [63]; (v) transient operating conditions [63]; (vi) cage geometry and material [17]; (vii) bleed oil properties [17, 28]; (viii) grease rheology, in particular, its yield stress [50], (ix) grease bleeding [50] and (x) mechanical stability [50]. However, several of these individual factors have already been modeled and validated with experimental results. For instance, the grease-free flow due to centrifugal forces was shown to be dependent on the Herschel-Bulkley rheological parameters (τ_0, K, n) when oil separation is low (low temperatures), and also on the bleed oil properties when oil separation is high [64]. Grease bleeding due to centrifugal forces was numerically modeled for greases with fibrous structure, such as lithium thickened greases. This model considers grease base oil viscosity and density, thickener fiber diameter, and mass [65]. Static oil bleed, representing the oil bleeding from the grease settle at the outer ring shoulder and on the seal was recently described using a Washburn-like model based on Darcy's law with the great agreement with five lubricating greases [66]. Shear degradation at different temperatures was modeled for fibrous structured greases and the results were validated using grease collected from a grease worker and rolling bearings [67]. Due to these recent advances in the prediction of grease shear aging and oil bleeding, it is expected that soon all these models will be placed together and better predictions of grease film thickness under starved conditions will be available.

Figure 3 presents grease film thickness as a function of time for entrainment speeds higher than the transition thickness (h^t) in a log-log scale (see **Figure 1**).

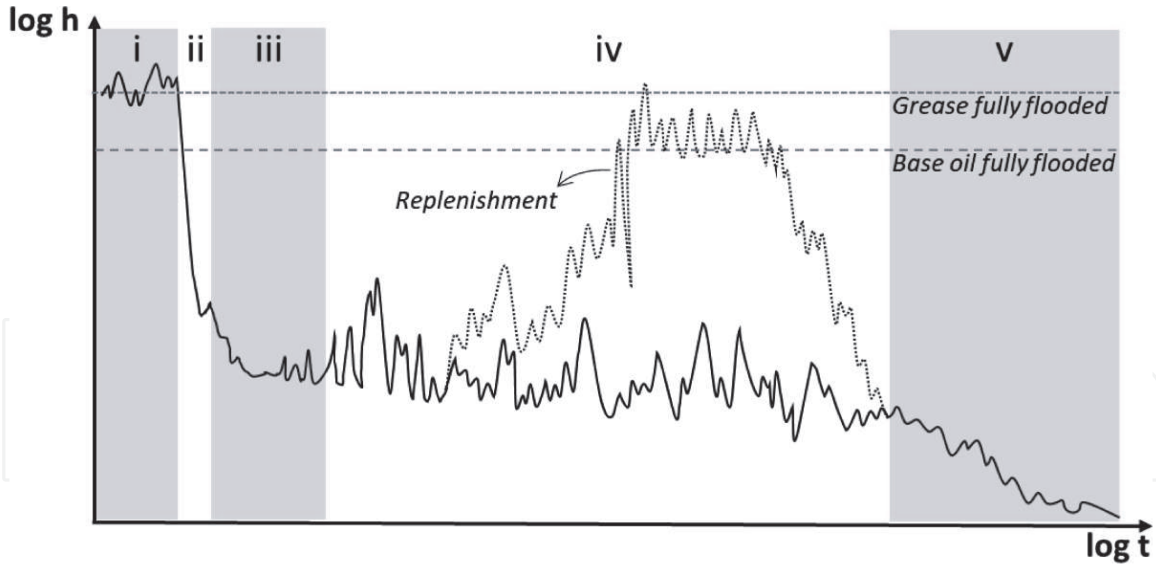

Figure 3.
Typical film thickness versus time curves for lubricating grease under starved lubrication and low SRR.

Two dashed lines representing grease and its base oil film thicknesses under fully flooded lubrication are also presented. Besides, the curve is divided into five stages—(i) at the beginning of the test the contact is fully supplied with grease and the same trends reported in Section 2.1 are observed, that is, most lubricating greases present higher film thickness than their base oils due to thickener contribution; (ii) as time progresses film thickness drops quickly; (iii) after that the film thickness decay rate slows considerably, (iv) up to the moment it reaches a stable value that might remain for a long period of time (hours). In this stage, the film thickness, which is formed by a thin layer of highly worked grease deposited on the track, changes little with time, and it may decrease or increase very slowly unless replenishment by grease or its bled-oil improves and increases film thickness considerably (dotted line). In this stage, the base oil viscosity is not directly related to film thickness values [50]; (v) at some point the lubrication availability to the contact becomes scarce and asperities contact takes place.

In every stage, the film thickness curve is characterized by significant scatter due to sudden local increases caused by thickener lumps crossing the contact. Such local increases occur at different rates and reach different thickness values depending on the thickener morphology [28, 68]. The duration, or even the existence, of each stage depends on the competing lubricant supply and loss mechanisms aforementioned. Therefore, some greases might not present the stabilization stage (iv) and quickly reach asperities contact (v), while others might present several replenishment cycles before reaching asperities contact. In terms of film thickness prediction, the first stage can be modeled using Eq. (2) since fully flooded lubrication occurs. Then, as starvation proceeds, which occurs in a few hours, the steady grease film thickness at the end of stage (iii) can be predicted by Eq. (4), which is related to the product of speed (U, m/s), dynamic viscosity (η, Pa s), and half-contact width (a, m).

$$h_{g\_starve} = h_{ff} \cdot b \cdot (U \cdot \eta \cdot a)^{-c} \tag{4}$$

This equation was validated in a full bearing test rig instrumented to measure film thickness [17]. The tests were performed with a deep-groove ball bearing 6309-2Z/C3 with a steel cage with 10 balls (full bearing) and 05 balls, and with a DGBB 6309-2Z/TN99 with a glass fiber reinforced polyamide 66 cage with 10 balls (full bearing) and 05 balls. Three lubricating greases with different thickener

(Li, LiC, and Pu) and base oil (M, E, and PAO) types were tested for several rotational speeds and loads. The results clearly showed that whatever is the effect of grease formulation, rheology, bleeding properties, number of balls in a bearing (overrolling time), cage geometry, and type of surface tension, all the experiments collapsed into a single curve represented by Eq. (4), which suggests that they are secondary at this stage. However, some spread between model and experiments was observed and attributed to these secondary effects, although it could also be related to the use of the fully flooded oil film thickness (h_{ff}, Eq. (1)) instead of the fully flooded grease film thickness (h_g, Eq. (2)) into Eq. (4).

It is also important to state that Eq. (4) is not a general film thickness equation for starved lubricated contacts since the values of the constants "b" and "c" were shown to be dependent on the tribological system (ball-on-disc, rolling bearing type) and might be affected by load direction (axial, radial, or combined) [17]. This equation clearly shows that starvation increases with speed, viscosity, and half-contact width. High speeds lead to faster starvation due to grease being pushed away from the contact and having less time for the film to recover [17, 62]. However, since the centrifugal force acting on the side reservoirs of grease increases with speed, bleeding increases, and the replenishment of grease and its bled oil also takes place more often. Such effect was not observed in the experiments due to their short duration (20 h). High base oil viscosity reduces replenishment and oil bleeding due to a reduction of oil mobility [62]. Increasing half-contact width requires more time for replenishments to the center of the contact [17].

As the time further progresses and stage (iv) is reached, grease properties and formulation effect on film formation become relevant. Long-term tests (100 h) were performed with full deep groove ball bearing 6209-2Z/C3 at constant axial load and rotational speed for six lubricating greases with different thickeners (Cs, L, Lc, Pu) and base oil (M, E, PAO, SS) types. Grease rheological parameters, shear stability, and bleeding properties were all different. The measured film thickness results clearly showed that lubricating greases with low shear stability and high oil bleeding were able to replenish the contact within the tested time, and therefore to present higher film thickness values, which were closer or higher to the oil fully flooded film thickness prediction. Grease shear aging caused an increase in oil separation and a decrease in the thickener particle size. This led to a reduction of the yield stress which enhanced the mobility of the grease. Furthermore, smaller thickener particles contribute the most to film thickness enhancement as shown by Eq. (2). Other researchers also observed improved replenishment for lubricating grease with a higher oil-bleed rate, lower viscosity, consistency, and mechanical stability [50, 55, 69]. From these results, it is verified that replenishment at stage (iv) is mainly driven by oil bleeding and grease shear aging, being the other aforementioned effects secondary [50]. Although 100 h may be long for a lab test, it is extremely short compared to the expected life of rolling bearings. Therefore, grease quality should not be associated with low mechanical stability and high oil bleeding rate, since it may cause severe starvation in the long run due to grease leakage [50]. Film thickness variations during stages (iv) and (v) have been only qualitatively presented and numerical models to predict grease contact replenishment are not available yet.

The schematic film thickness curve presented in **Figure 3** changes with the operating conditions and grease type. As temperature increases, the decay rate decreases leading to later starvation and better replenishment due to decreased consistency, reduced bled oil viscosity, and improved oil bleeding [50]. The scatter on the results increases significantly at higher temperatures, which indicates the thickener lumps are crossing the contact at a higher rate. Load reduction reduces the Hertzian half-contact width which slows down the onset of starvation [50]. Ball spin was also shown to improve replenishment by dragging the grease and bled-oil of the sidetrack into the contact inlet [55].

In terms of grease formulation, some thickener types produce higher film thickness values and enter the contact much more often than others at stage (iv), without considering replenishment effects. Here, the same trends observed for fully flooded conditions were detected. In general, thickeners that enhance grease film thickness the most are as follows—polypropylene (P) > calcium (C) > polyurea (Pu) ≈ lithium (L) [28, 33, 34, 39, 68, 70].

Friction measurements performed under starved conditions in a ball-on-disc test rig at entrainment speeds higher than the transition thickness (h^t) also present 05 stages, as shown in **Figure 4**—(i) at the very beginning of the test (first seconds) the contact is fully supplied with grease, and the same trends reported in Section 2.1 are observed, that is, lubricating greases present similar coefficient of friction than their base oils under fully flooded conditions; (ii) as time progresses coefficient of friction increases quickly (few minutes); (iii) after that the friction coefficient increase rate slows considerably, (iv) up to the moment, it reaches a stable value that remains for a long period of time (hours); (v) and finally the grease is gradually expelled from the contact and friction tends to asperities friction values (dry CoF).

The coefficient of friction curve presented in **Figure 4** changes with the operating conditions and grease type. In terms of operating conditions, by increasing the entrainment speed, the CoF grows quicker and its value in the stabilization zone is higher than that at lower entrainment speeds, which should be due to stronger starvation. The same effect is observed for tests performed at higher SRR values, but in this case, the value of the CoF in the stabilization zone is even higher. On the other hand, at higher operating temperatures lower CoF values are observed due to the decrease in the viscous friction and better replenishment due to the reduced grease consistency and improved oil bleeding [68]. Lewis et al. [71] showed the time required to reach stage (v) (dry coefficient of friction) depends more on samples' roughness than on grease properties. Samples with low roughness (Ra < 2 μm) reach stage (v) up to four times later than samples with high roughness (Ra > 3 μm).

In terms of grease formulation, it is known that lubricating greases formulated with mineral and high viscous oils produce higher friction than the ones formulated

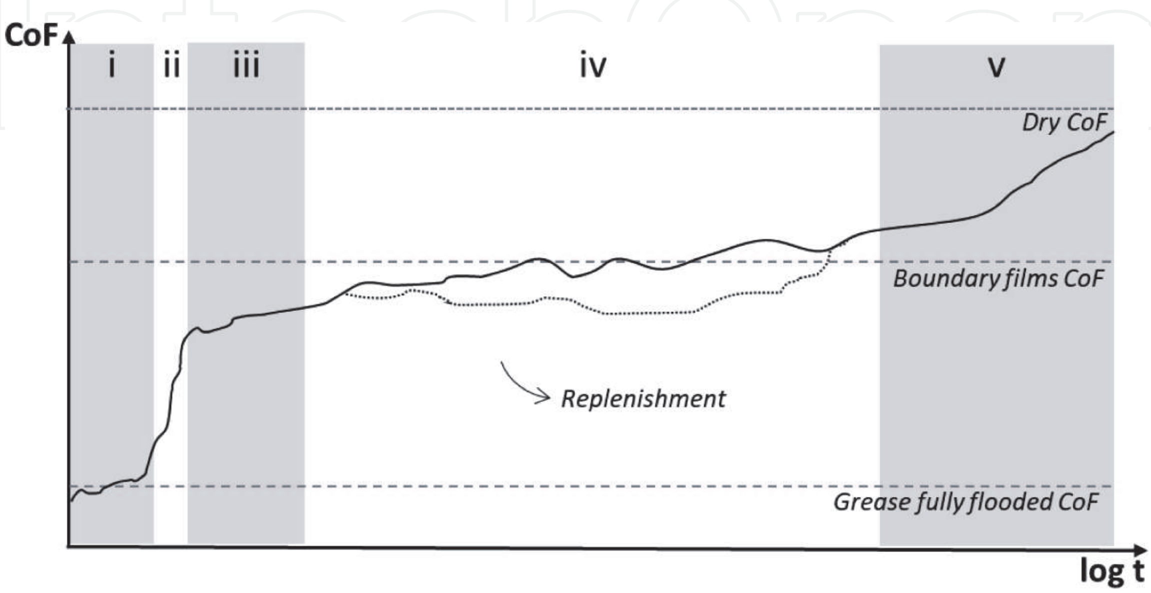

Figure 4.
Typical coefficient of friction versus time curves for lubricating grease under starved lubrication and low SRR.

with synthetic and low viscous oils [14, 72]. When it comes to the thickener, similar to fully flooded lubrication, lower friction values are observed for polypropylene (P) > polyurea (Pu) > lithium (L) > calcium (C). This occurs because the coefficient of friction is a response of the boundary films, which are mainly formed by sheared thickener. As discussed in Section 2.1, some thickener types present high friction properties, while others present low friction properties, such as calcium and polypropylene, respectively.

Analogously to the closure of Section 2.1, grease selection for applications that run under starved lubrication should not blindly follow the procedure defined by ISO 281, since it does not take into account the thickener type, nor critical grease properties such as oil bleeding. In fact, the starved film thickness, before any mechanism of grease/bleed-oil supply to the contact takes place, does not present any trend with the base oil viscosity, but with grease formulation (thickener and base oil type). Higher film thickness values are observed for synthetic oils thickened with polypropylene, lithium complex, or polyurea [50]. These formulations also present lower friction values and higher efficiency. The maintenance of this film depends on contact replenishment, which in turn depends on grease rheological properties, shear stability, and bleeding rate. All these properties can be changed by playing with the thickener content and grease manufacturing process. However, at the moment, there are no tools or models to provide end users with optimum values of these properties for specific applications. The general advice is to look for the unworked and worked grease consistency (related to its rheology and mechanical stability), the oil bleeding rate, thickener type, and operating temperature of lubricating greases that operate successfully in similar applications to establish reference values. Alternatively, bearing manufacturers could be contacted to assist user needs.

3. The role of grease components in sliding contacts

Grease is used in a wide range of applications. Its usage in components that operate under prevailing sliding conditions, such as railway lubrication, represents just a small fraction of grease usage. This explains the short amount of systematic research on grease lubricated sliding contacts. In these conditions, lubricating greases are not prone to form full films, thus boundary and mixed lubrication regimes are expected. Therefore, the study of wear and friction is of particular interest and reported in the following sections considering fully flooded and starved contacts.

3.1 Fully flooded conditions

Under sliding conditions, lubricating greases are mainly evaluated in four-ball machines or pin-on-disc test rigs under unidirectional or reciprocating motions. These types of tests are, in general, not instrumented to measure film thickness, although electric methods (capacitance, resistance) are sometimes employed as an indirect measure of film thickness. Alternatively, the lubrication regime is qualitatively evaluated based on friction and wear analysis.

Figure 5 shows the coefficient of friction as a function of time in a grease-lubricated pin-on-disc test under low contact pressures. The general trend is high friction at the very beginning of the test (region i) to overcome the static condition followed by a stable regime (ii) in which some scatter is observed due to thickener crossing the contact and eventual asperity contact since mixed lubrication regime is expected. At this stage, if the grease supply fails, a rapid increase in friction is

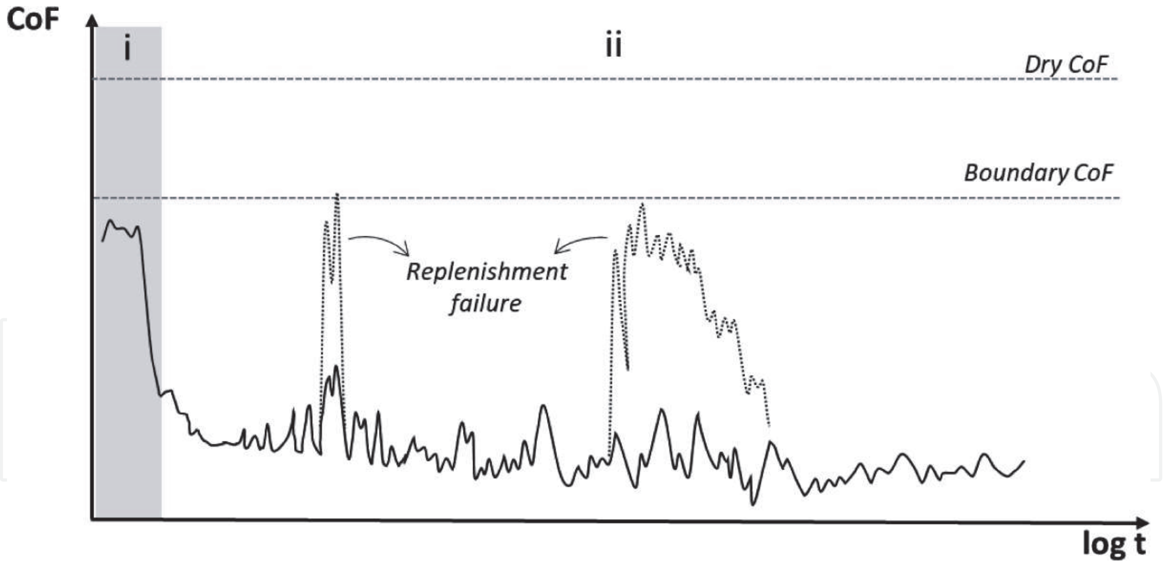

Figure 5.
Typical coefficient of friction versus time curves for lubricating grease under fully flooded lubrication and pure sliding condition.

observed. Here, the coefficient of friction results from shearing a thin layer of grease, mostly composed of thickener deposited onto the surfaces, and/or reactions films composed by additives (boundary CoF). These trends are observed for lubricating greases whatever is their formulation [18, 19]. Even with constant grease supply to the contact, sometimes replenishment failure occurs because the parallel contacting surfaces are not prone to hydrodynamic film formation.

Figure 6 shows a schematic representation of the coefficient of friction as a function of the Hersey parameter under pure sliding conditions for two grease types. These trends were obtained from the pin-on-disc evaluation of six in-house fully formulated NLGI 2 greases formulated with a commercial additive package for EP application and mineral base oil. The two most common thickener types, lithium and calcium, and three base oil viscosities, 50 mm^2/s, 200 mm^2/s e 500 mm^2/s, were used in the formulations. More details about these lubricating greases can be found in [15]. The pin-on-disc tests were performed under a constant sliding speed of

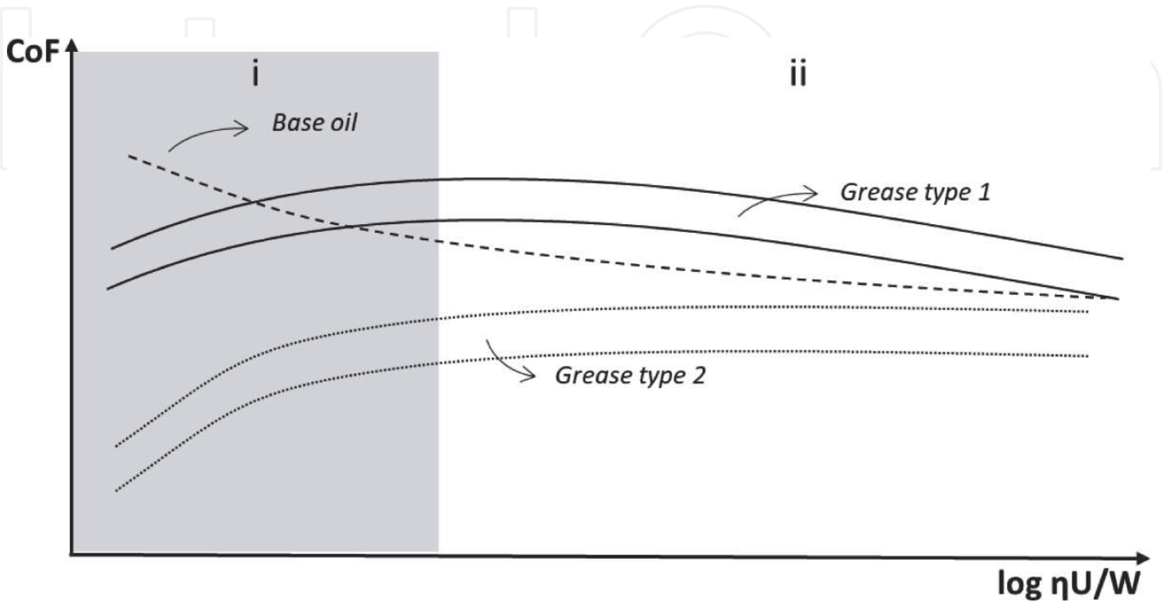

Figure 6.
Typical coefficient of friction versus Hersey number curves for lubricating grease under fully flooded lubrication and pure sliding condition.

0.5 m/s and contact pressure of 2.5 MPa for 60 min. The flat-tip pins and discs were manufactured from pearlitic steel with a hardness of 360 HV and composite roughness of S_q = 1.2 μm [19]. Different from the observations from prevailing rolling conditions the coefficient of friction does not vary significantly with the product of speed and viscosity [19, 45]. This occurs because pin-on-disc tests performed under pure sliding conditions are not prone to build thick films due to their parallel surfaces. The surfaces are either parallel from the beginning of the test when flat-tip pins are used, or they quickly turn flat and parallel due to wear when round-tip pins are used [18]. Therefore, the usual lubrication regimes are boundary (stage i) and early mixed lubrication (stage ii) for usual engineering surface finishing and contact pressure. In the case of very smooth surfaces, low loads, high speeds, and fully flooded conditions, the coefficient of the friction curve may resemble the one from **Figure 2**. Although, in general, the effect of speed, viscosity, and load on friction coefficient is rather small [19, 22, 73].

Under boundary lubrication conditions (stage i), the film is composed mainly of thickeners under moderate loads, and therefore, the coefficient of friction depends on its properties. In this situation, lubricating greases thickened with calcium (grease type 1) are used to present higher friction values than greases thickened with lithium (grease type 2) [7, 8, 19]. This is also observed under prevailing rolling conditions and fully flooded lubrication [74]. As the base oil viscosity or speed increases (region ii) the effect of thickener on the coefficient of friction becomes slightly less relevant, as in this region the film is also composed of base oil. The friction variation from regions i to ii is small and depends pretty much on the grease formulation. Lithium thickened greases in general present lower shear resistance than their corresponding base oil and, therefore, the coefficient of friction is lower when the film is mostly formed by thickener (low Hersey number) [7, 8, 19]. Calcium thickened greases, on the other hand, in general present similar shear resistance to its base oil, and therefore, the coefficient of friction varies little as a function of the film composition (%thickener + %oil) [7, 8, 19].

Figure 7 shows the sum of the mass loss of the tested pins and discs as a function of the Hersey parameter under fully flooded lubrication and pure sliding conditions. Analogously to **Figure 6** the zones i and ii correspond to the boundary and early mixed lubrication regime, respectively. The dominant wear mechanism under these operating conditions is two-body abrasion since the adhesive forces between

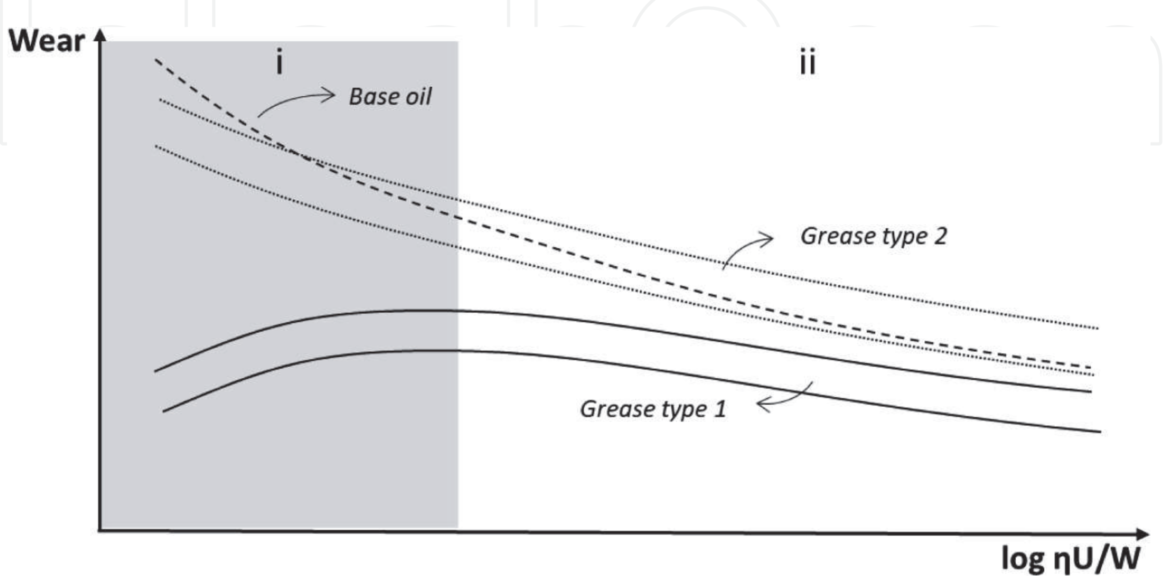

Figure 7.
Typical mass loss versus Hersey number curves for lubricating grease under fully flooded lubrication and pure sliding condition.

contacting asperities are reduced substantially by the introduction of grease [18]. In these conditions calcium greases (grease type 1) present lower wear than lithium greases (grease type 2), as also observed by [7]. As the base oil viscosity increases (region ii), the effect of thickener on wear becomes less relevant and similar wear values are observed for both greases, as in this region the film is also composed of base oil. The wear variation from regions i to ii depends on grease formulation. Lithium thickened greases present wear reduction due to a slight increase in film thickness, which reduces the contact severity. At region i, wear is quite high because of the poor anti-wear properties of lithium thickener [15, 75]. Calcium-thickened greases present similar wear values for the whole range of the Hersey number. This occurs because as viscosity decreases (transition from regime ii to regime i) the film composition becomes dominated by calcium thickener that minimizes wear under boundary lubrication due to its well-known anti-wear properties [8, 76].

3.2 Starved conditions

Figure 8 shows the coefficient of friction as a function of time under pure sliding conditions and starved lubrication. Three-dashed lines representing the coefficient of friction measured without lubrication (dry), the coefficient of friction resulting from tribofilms derived from thickener and/or additives (boundary CoF), and the coefficient of friction measured when grease supply to the contact is abundant (grease fully flooded lubrication) are also presented. Besides, the curve is divided into five stages—(i) at the beginning of the test, static friction is overcome and the lubricant between the contacting bodies accommodates, thus high friction values are observed. This period lasts a few seconds and it is followed by (ii) very low fiction values, which are equivalent to the fully flooded lubrication presented in **Figure 5** since at this stage, there is lubricant enough to supply the contact. This stage also used to last a few seconds [77]; (iii) as time progresses starvation proceeds and the coefficient of friction increases up to its stabilization at stage (iv). In this stage, that last several hours [78], the coefficient of friction, which is governed by a thin layer of highly worked grease deposited in the track and/or tribofilms derived from chemical reactions between additives and the surface, changes little with time, and it may decrease or increase very slowly unless replenishment occurs and the

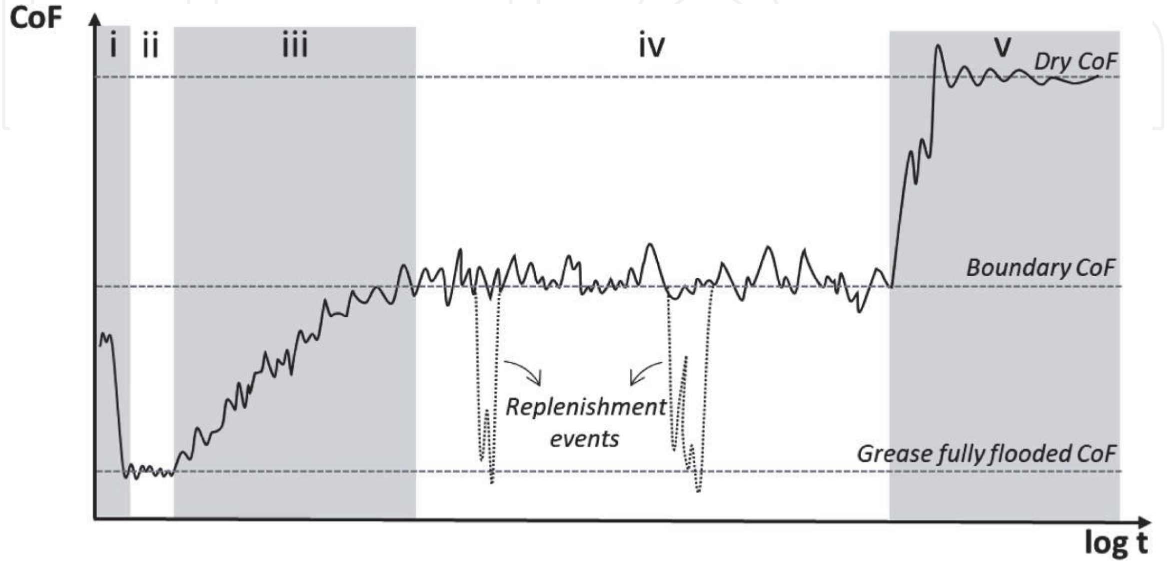

Figure 8.
Typical coefficient of friction versus time curves for lubricating grease under starved lubrication and pure sliding condition.

coefficient of friction reduces considerably (dotted lines); (v) at some point the tribofilm derived from thickener and/or additives wears off and the coefficient of friction quickly reaches dry friction values, that phenomena are usually reported as film breakdown [78]. The duration, or even the existence, of each stage presented in **Figure 8** depends on the competing lubricant supply and loss mechanisms, as well as the formation of reaction films. Therefore, some greases might not present the boundary stage (iv) and quickly reach asperities contact (v), while others might present several replenishment cycles before reaching asperities contact. The transition from stages (ii) to (iv) and from stages (iv) to (v) are defined as lubricant film breakdown [77, 78], although the related phenomena are different. The transition from stages (ii) to (iv), defined from here on as lube film breakdown, describes the starvation process, that is, the progressive absence of lubricant supply to the contact. The transition from stages (iv) to (v), defined from here on as boundary film breakdown, described the wear process of the boundary film formed from physical and or chemical interaction of thickener and additives with the contacting surfaces.

The schematic film thickness curve presented in **Figure 8** changes with operating conditions and grease type. In terms of operating conditions, a low coefficient of friction is observed for a longer period of time (stage ii) for the greater initial amount of lubricating grease, lower contact pressure, and higher sliding speed [77]. According to Hersey number and Stribeck curve, higher load (thus higher contact pressure) and lower speed will generate nominal thinner films (**Figures 1** and **2**), leading to premature lube film breakdown and a high coefficient of friction. This, however, only occurs at very low speeds. At high speeds, a significant decrease in load carrying capacity occurs due to the effect of frictional heating in the contact zone, causing an appreciable decrease in effective lubricant viscosity [79], and due to low replenishment as a result of grease being pushed away from the contact and having less time for the film to recover [62]. Lundberg [80] showed that low roughness increases the period in stage (ii), which is in agreement with the observations under prevailing rolling conditions [71]. In this case, low roughness indicates thicker film, and therefore, a larger period of time to occur lube film breakdown. It is important to point out the replenishment events presented in **Figure 8** occur less often under pure sliding than under rolling conditions, mainly because grease or bled-oil that might flow into the contact is more likely to be ejected than to find its way back in between the contact bodies due to the flat contact geometry. Besides that, the influence of operating conditions on the replenishment events follows the ones described in Section 2.2.

In terms of grease formulation, base oil nature, thickener type, and additive package play a major role in the starved coefficient of friction. For lubricating greases formulated with the same additive package, calcium greases present a higher coefficient of friction and reach stage (iv) faster than lithium greases, just like observed in **Figures 2** and **6**, and also for the same reasons. At stage (iv), calcium greases continue to present higher friction values than lithium greases, however, the tribofilms derived from calcium thickened greases last much longer. Considering the goal of lubricating greases is to prevent or minimize the chances to reach stage (v), associate with severe wear [78], and the time between stages (i) and (iii) used to be just a fraction of the time spent in stage (iv), one can conclude that calcium greases, in particular calcium sulfonate greases, present higher performance than lithium greases under concentrated sliding contacts. Therefore, for starved sliding contacts, lubricating greases that are highly reactive to the surface are used to present better performance [7, 8].

Mass loss of pins and discs under pure sliding conditions and starved lubrication depends on the five stages presented in **Figure 8**. The mass loss might be quite high at stage (i), but it drops rapidly and remains low at stage (ii). At these stages, the surfaces are partially separated and the dimensional wear coefficient used to be less than $k \approx 10^{-9}$ mm^3 (N m)$^{-1}$. During this period the separation might increase due to surface smoothening, which keeps the wear under low levels. At stages (iii) and (iv), the mass loss is higher but still acceptable for many engineering applications—$k \approx 10^{-6}$ to 10^{-8} m^3 (N m)$^{-1}$. At these stages, there is a thin film of lubricant material (thickener and/or additives) adsorbed to the surface that controls wear. Since these films are sparse and do not cover the whole contacting area, asperities contact is expected. In general, stages (i)–(iv) are oxidative in character, although some adhesive mechanisms related to unprotected asperity contacts might occur. As time progresses, these films are worn off up to the moment the metallic contact takes place, generating high wear rates, with dimensional wear coefficient typically greater than $k \approx 10^{-}5$ mm^3 (N m)$^{-1}$. High loads and high speeds increase the severity of the contact leading to premature scuffing (stage v). Wear resistance in pure sliding conditions depends on the likelihood of thickener and additives to get onto the contacting surfaces and form tribofilms. This is achieved using lubricating greases with high chemical affinity with the contact bodies and small size thickener particles, such as greases thickened with calcium sulfonate, formulated with ester base oil, and additives with high load-carrying capacity based on sulfur and phosphorus.

4. Conclusions

Lubricating greases play a significant role in the component's life and efficiency. In order to maximize this role, they must meet specific performance requirements. However, defining such specifications is challenging because they are system-dependent and their simulation in the laboratory might require elaborate equipment and tests with a long duration (>100 h).

Under prevailing rolling conditions and fully flooded lubrication, the current practice, which works reasonably well, is to predict film thickness using traditional EHL theory for oil lubrication (Eq. (1)) when the operating conditions lead to film thickness higher than the transition film thickness (h^t). Although an improved film thickness equation for grease-lubricated contacts that account for the thickener effect (Eq. (2)) was developed and more accurate results can be achieved. These equations are reliable to predict film thickness in situations in which fully flooded conditions are expected, such as in rolling bearings lubricated by centralized systems or during the first days of operation of sealed for life rolling bearings. Below the transition film thickness (h^t), typically observed in large size bearings running under low rotational speeds, grease and oil behavior differs the most. In this condition, the film thickness can only be predicted based on previous film thickness measurements (Eq. (3)). Due to the coasts related to large-size bearings maintenance, further research on film formation below the transition speed is of utmost importance.

When it comes to starved lubrication, which is the operating condition of rolling bearings sealed for life during most of their lifetime, several phenomena occur simultaneously and grease film thickness prediction is challenging. Initially (first few minutes), grease film thickness is larger than the fully flooded base oil, indicating fully flooded conditions with thickener contribution. At this stage, Eq. (2) can be used to predict grease film thickness. Then film thickness decreases due to starvation (first hour) and remains stable for a few more hours. At this stage, the grease film thickness can be predicted with Eq. (4). At a certain moment, grease film thickness increases again to values similar or higher than its fully flooded base oil, mostly, due to oil bleeding, and grease shear aging. This stage strongly depends on grease type, particularly on its bleed properties and mechanical stability (rheological behavior). The higher is the bleed rate and the lowest in the mechanical stability the faster grease film recovery is observed. However, it does not mean that a high bleed rate and low mechanical stability properties are desired for lubricating greases since their long-term effects (after 100 h) are not known. This last stage has not been modeled yet because it requires more accurate bleeding and shear aging models to be implemented along with film thickness predictions.

The coefficient of friction prediction depends on grease film thickness and properties (molecular structure). Consequently, all uncertainties around film thickness prediction impact friction. Besides, the need to include the film properties at the high-pressure zone makes it a much more complicated matter. In the case of mixed and boundary lubrication, the role of surface topography and properties on the physicochemical reactions with additives and thickener must be also taken into account. Therefore, the existent numerical models, to predict coefficient of friction, are far less developed than the ones to predict film thickness and still do not take into consideration the role of thickener.

All these challenges related to film thickness and coefficient of friction prediction lead several researchers to evaluate formulations and properties of lubricating greases that promote high film thickness and low coefficient of friction over a wide range of operating conditions. The results point out for lubricating greases formulated with low viscosity synthetic oils, in particular PAO, thickened with polypropylene, polyurea, or lithium. These combinations, in particular PAO with polypropylene or polyurea, also lead to long grease life at moderate temperatures [81]. Obviously, the grease properties such as oil bleeding and mechanical stability, which can be adjusted during the manufacturing process, must match the application. Those general trends do not qualify PPAO, PuPAO, or LPAO as a multiuse grease. In fact, for specific situations, that is, very low speeds, high loads, high temperature, and humid environment, over based calcium sulfonate complex greases, are the best option due to their high dropping point, excellent EP/AW characteristics, and water resistance [81].

Under prevailing sliding conditions, the lubrication regime for usual engineering surfaces is mainly boundary and mixed lubrication, whatever the contact is fully flooded or starved since the contact conditions are less prone to film formation. Thus, the research lies mostly on the effect of grease type and properties on wear and friction, instead of film thickness. Comparatively to prevailing rolling conditions, the effect of base oil viscosity, speed, and load on the coefficient of friction is rather small since lubrication regime transitions are less likely to occur. In this situation, in which nominal specific film thickness is low, the film that partially separates the surfaces is mainly composed of grease thickener and/or additives, which primarily govern the friction and wear response of the system. Thickener types used to be qualified by their thermomechanical stability, water resistance, load carrying capacity, lubricity (friction), additive response, and compatibility with other greases by grease manufacture. As discussed earlier, as grease properties depend on their constituents, interaction, and the manufacturing process, the thickener properties do not necessarily translate the grease properties. Still, in the large majority of the evaluations reported in the literature, calcium greases present higher load carrying capacity (wear resistance) but slightly lower lubricity (friction) than lithium greases in tribological tests, which is in agreement with the general classification provided by grease suppliers. The durability of the films formed by thickeners and/or additives on the surface is still an open topic that requires further investigation.

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