AN EXPERIMENTAL STUDY INTO
THE INFLUENCE OF GREASE COMPOSITION ON FRICITION
IN EHL CONTACTS

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Abstract

Rolling bearings are among the most widely used machine elements. As a result, reducing bearing frictional losses can have a significant impact on the overall energy efficiency of the vast majority of engineering systems. Given that rolling bearings are typically lubricated with grease, one of the most effective ways to improve bearing efficiency is through a suitable grease formulation. However, at present there is a lack of understanding of grease frictional behaviour in typical non-conformal, highly stressed bearing contacts. This limits the development of low-friction bearing greases, and thus further improvements in bearing efficiency. The current research work attempts to address this topic through an experimental investigation.

Friction and film thickness tests were conducted on a wide selection of greases in ball-on-disc tribometers, under conditions broadly representative of rolling bearing contacts. Initial work was carried out with a series of commercially available greases, selected to cover a wide spectrum of thickener types, base oil types and viscosities, and practical bearing applications. These tests served to identify the most influential aspects of grease formulation on friction, and provide a reference line in terms of the current state of the art in low-friction bearing greases. Subsequently, the work was focussed on the study of a series of custom lithium-thickened greases. These were designed and manufactured with the purpose of isolating and investigating the effects of particular aspects of interest in grease formulation, including base oil type, base oil viscosity and the presence of friction modifiers. In addition, the performance of the corresponding bled and base oils was also studied to help shed some light on the prevailing mechanisms.

The results show that two distinct operating regions, at high and low speeds, can be identified in the tribological performance of greases. At high speed greases behave approximately as their base oils. In this region, oil type was found to be the major parameter of influence on friction, with synthetic oils (particularly PAO) providing the lowest friction coefficients. In the same high speed region, the influence of base oil viscosity is that expected from a classical Stribeck relationship for oils, i.e. higher base oil viscosities in the grease formulation tend to produce higher friction as a result of thicker films. In contrast, at low speed grease behaviour is much more complex and is strongly influenced by both base oil and thickener properties. In this region the combination of lithium and PAO was shown to provide both thick films and low friction. No benefit in friction reduction was observed when a friction modifier, namely oleic acid, was added to the formulation of the greases. The influence of mechanical degradation of greases on their tribological performance was also evaluated. It was established that mechanical degradation
has the effect of diminishing the friction and film thickness benefits provided by some of the greases at low speeds. A remarkable exception was found in the tribological behaviour of greases formulated with lithium and PAO oil, which was overall practically unaffected by mechanical degradation. Finally, based on the observed results, the combination of a low viscosity PAO base oil and lithium thickener was suggested as the most suitable formulation for a low-friction rolling bearing grease.
Declaration of Originality

I, Nicola De Laurentis, declare that this thesis is my own original work. The use of information coming from other sources has been properly referenced.

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\( \alpha \) – Pressure-viscosity coefficient \([\text{Pa}^{-1}]\)

\( \dot{\gamma} \) – Shear rate \([\text{s}^{-1}]\)

\( \eta \) – Dynamic viscosity \([\text{Pa s}]\)

\( \Lambda \) – Lambda ratio \[
\Lambda = \frac{b_i}{\sqrt{R_{q1}^2 + R_{q2}^2}}
\]

\( \mu \) – Friction coefficient

\( \nu \) – Kinematic viscosity \([\text{m}^2 \text{ s}^{-1}]\)

\( \nu_1, \nu_2 \) – Poisson’s ratio

\( \omega \) – Rotational speed \([\text{rpm}]\)

\( \tau \) – Shear stress \([\text{Pa}]\)

\( E_1, E_2 \) – Young’s modulus \([\text{Pa}]\)

\( \bar{G} \) – Dimensionless material parameter

\( h_i \) – Central film thickness \([\text{m}]\)

\( h_o \) – Minimum film thickness \([\text{m}]\)

\( Q \) – Power losses \([\text{W}]\)

\( R_{q1}, R_{q2} \) – RMS surface roughness \([\text{m}]\)

\( R_{x1}, R_{x2} \) – Principal radius of contact in x-direction \([\text{m}]\)

\( R_{y1}, R_{y2} \) – Principal radius of contact in y-direction \([\text{m}]\)

\( R_x', R_y' \) – Reduced radius \([\text{m}]\)

\( SRR \) – Slide/roll ratio \[
SRR = \left| \frac{U_1 - U_2}{U} \right| \times 100
\]

\( T \) – Friction torque \([\text{N m}]\)

\( U \) – Entrainment speed \([\text{m s}^{-1}]\) \[
U = \frac{U_1 + U_2}{2}
\]

\( \bar{U} \) – Dimensionless speed parameter

\( U_1, U_2 \) – Tangential speed \([\text{m s}^{-1}]\)

\( VI \) – Viscosity index

\( W \) – Normal load \([\text{N}]\)

\( \bar{W} \) – Dimensionless load parameter

‘1’ refers to body 1

‘2’ refers to body 2

\( x = \) parallel to rolling direction

\( y = \) transversal to rolling direction
Chapter 1. Introduction

1.1 Background

Grease is a multi-phase lubricant consisting of two main components: a fluid, which is typically a lubricating oil, and a solid ‘thickener’, usually a metal soap. This composition provides grease with some inherent benefits over lubricating oils including a certain rigidity, which allows grease to stay in place without leaking, as well as sealing abilities. These properties make grease particularly suitable for application in rolling bearings. Indeed, this is by far the most important application of lubricating greases, with more than 90% of rolling bearings employing grease as the lubricant\(^1\). Rolling bearings are ubiquitous machine elements: they are used in almost all engineering systems, and across many different sectors of industry and transportation. It is reported that over 50 billion bearings are operating in the world at any time\(^1\). As most machines utilise multiple bearings, the cumulative impact of bearing frictional losses on the energy efficiency and sustainability of mechanical systems is undeniable. Although several strategies can be followed to reduce friction losses in a bearing, it is recognised that the employment of a suitable ‘low-friction’ grease can have a large impact in this respect. Furthermore, increasing energy efficiency through improved lubrication performance is usually more cost-effective and versatile than other potential approaches, such as optimisation of bearing geometry and materials, application of coatings or surface texturing. As a consequence, better understanding and optimisation of grease frictional performance can offer one of the most pervasive ways of improving the overall efficiency of mechanical systems, with associated savings in energy and reduction in carbon emissions. Traditionally, the selection of grease for a specific bearing application has been governed by a number of other non-related factors, such as cost, thermal stability, water washout et cetera. With the current quest for energy efficiency, driven by regulations and customer demand, low-friction grease formulations have gained in importance.

The primary role of grease in a rolling bearing is to lubricate the highly-stressed, non-conformal contacts at the interface between the rolling elements and the raceways of the bearing, where a large contribution to the overall friction is generated. Such non-conformal contacts operate in what is termed as elastohydrodynamic lubrication (EHL). The mechanisms of lubricant film formation, and to some extent friction, in these EHL contacts, are relatively well known for oils. However, the equivalent mechanisms and theories for grease are more complex and still rather unclear. Indeed, given the multiphasic chemistry of greases, their tribological behaviour depends on a large number of different properties and individual components in the overall formulation.
The degree to which these aspects affect the chemistry and physics of the lubricating film, on which friction depends, is still poorly understood. Despite the significant effort that has been put into attempting to understand the fundamental mechanisms of grease lubrication, a comprehensive, universally accepted theory does not exist yet. This hinders the development of a scientific framework for the choice or formulation of low-friction greases, and consequently makes it difficult to afford further improvements in rolling bearing efficiency.

1.2 Primary research objectives

This study explores the mechanisms of friction in rolling-sliding concentrated contacts lubricated with grease. As the focus of the research is grease composition rather than bearing factors, standard laboratory tests were used to measure friction coefficient and film thickness under contact conditions pertinent to rolling bearing applications. A wide selection of commercially available and custom-made greases were employed as samples for the experimental investigation. The commercial greases were selected and studied to provide basic guidelines on the relationship between grease formulation and friction, as well as a reference line in terms of the current state of the art in low-friction greases. Subsequently, the custom greases were formulated and manufactured with closely controlled composition to enable studies of the influence of specific aspects of the grease formulation on friction. The overall objectives of this research are:

- To develop new knowledge of grease tribological behaviour (friction and film thickness) in concentrated contacts;
- To gain a deeper understanding of the influence of grease composition on friction in concentrated contacts;
- To provide guidelines for the formulation of low-friction greases for potential use in rolling bearings.

1.3 Thesis outline

In this section the structure of the thesis as divided into its chapters is presented. Every chapter starts with an introduction to briefly outline its contents, and ends with a conclusion to stress the main highlights.
Chapter 2 provides a wide ranging literature survey on the behaviour of lubricating greases in EHL contacts. The definition of lubricating greases and a brief introduction on their history are given at the beginning of the chapter, with specific focus on bearing greases. Their general formulation and rheological properties are introduced and discussed. The basic concepts of EHL lubrication theory, which represents the typical mode of lubrication in rolling bearings, are presented. A comprehensive review on film formation and friction in greased EHL contacts is then given.

Chapter 3 describes the range of test lubricants chosen to perform the experimental work. It explains how the commercial greases were selected, and the custom greases designed and manufactured. This chapter also describes the methods used to characterise the greases, and their corresponding results.

Chapter 4 introduces the experimental methods employed to investigate friction and film forming behaviour in grease-lubricated non-conformal contacts. The test rigs are described, and the choice of test conditions are explained and justified.

Chapter 5 presents the experimental results obtained with the commercial greases. Friction and film thickness results are compared under different conditions of speed, temperature and contact surface roughness. Results obtained with mechanically degraded greases are also presented.

Chapter 6 presents the experimental results obtained with the custom greases. Friction and film thickness results are compared under different conditions of speed, temperature and lubricant supply. Results obtained with mechanically degraded greases are also presented here.

Chapter 7 provides a discussion on all findings obtained in this study. The observed trends in grease friction and film formation are interpreted in relation to grease composition and contact condition. An attempt is made to discuss the possible mechanisms at play. The chapter also discusses potential low-friction grease formulations based on the evidence obtained in the experimental results.

Chapter 8 summarises the main conclusions from this study, and recommends possible future work.
Chapter 2. Literature survey

This chapter provides an overview of the current state of knowledge on the use and tribological behaviour of lubricating greases in concentrated contacts. Firstly, the significance of low friction in greased rolling bearings is discussed, the definition of lubricating grease is given, and the basic chemical and rheological properties summarised. Given that this research concerns the study of grease frictional behaviour in rolling bearings, the theory that applies to the more general family of concentrated contacts is briefly introduced. The survey then reviews the literature on the existing experimental studies on film formation and friction in grease-lubricated concentrated contacts. Current theories behind the possible mechanisms of grease lubrication in rolling bearings are also presented. At the end of the chapter, gaps in the current state of knowledge are identified, and the present study is thus contextualised.

2.1 Introduction

Rolling bearings are an example of machine elements operating under the elastohydrodynamic lubrication regime (EHL), characterised by extremely high shear rates and pressures, and thus producing a very severe environment for the lubricating film. These conditions impose a dramatic change to the rheological properties of the lubricant, so that its frictional response to shear cannot be easily correlated to its properties at low shear rate, or at ambient pressure. Due to its complex nature, the general theory of EHL friction is still being widely investigated and disputed. A further complication to the understanding of friction in EHL contacts arises from the use of grease, for which the mechanism of film formation itself is still not entirely understood. The grease base oil properties are commonly used as a guideline to estimate grease film thickness and general tribological behaviour. Although the thickener type and concentration are also known to be important in this respect, their influence is generally only considered in light of some grease chemical and physical properties that are not directly related to film formation. Nevertheless, the literature reports many examples of studies in which it is shown that the thickener actively takes part in the lubrication process, with the consequence that the lubrication rules for oils are not strictly applicable. Currently, broad guidelines for the selection of bearing greases are used, based on extensive laboratory analysis and observations from the field, suggesting generalized selection rules to fulfil the specific application requirements. However, there is a lack of detailed understanding of the relationship between grease composition and friction in rolling bearings, which is partly due to the absence of a comprehensive investigation of the topic. In fact, very few studies exist, in general, on the
relationship between grease formulation and friction in concentrated contacts. With the current lack of hard data, in addition to the poor understanding of the mechanisms of grease film formation and frictional behaviour, reducing bearing power losses through optimised grease lubrication is a hard challenge.

2.2 Friction in rolling bearings and the role of grease

Rolling bearings are complex mechanical systems comprised of several tribological contacts. These include concentrated, as well as conformal contacts, at the interfaces between rolling elements and raceways, rolling elements and cage, seal and rings. These contacts give rise to several mechanisms of energy loss occurring during lubrication, including rolling, sliding and churning friction. Rolling bearings are intrinsically low-friction machine elements, as they are designed to effectively replace sliding friction with the (much lower) rolling friction originating at the rolling elements/raceways contacts. The latter is generated by the repeated loading/unloading hysteresis cycle of the rolling surfaces. In reality, sliding friction is not excluded. The very high pressures involved cause an elastic deformation of these elements, generating losses due to micro-sliding. Unequal curvatures in several contact areas also generate sliding friction. Macro-sliding also occurs with some types of bearings, when the rolling elements are able to spin. Sliding friction even tends to be larger than rolling friction when low viscosity lubricants are used. Finally, the seal and cage purely slide against the rings and rolling elements respectively, producing relatively high friction. These contributions often represent a considerable proportion of the overall friction losses, depending on the operating conditions. Churning losses are due to several lubricants effects: shearing the lubricant in between rolling elements, cage and raceway surfaces, as well as mechanically working the lubricant dispersed inside the bearing cavity, leads to energy waste. These losses are dependent on the bulk characteristics of grease, particularly its base oil viscosity and consistency. Experience suggests that the churning phase lasts for about one day of operation when grease is employed. Albeit this might not seem significant in determining the overall efficiency, due to its limited time of contribution, it does exert a crucial effect on the running-in, and therefore affects the energy efficiency of the bearing also in the long term. Indeed high churning losses might produce very high temperatures, which degrade the grease thermally, and in turn mechanically, thus compromising the life of the grease and its bearing. Grease degradation is commonly a major cause of premature bearing failure. In the formulation of energy efficient greases, the thickener type and concentration, and the viscosity of the base oil, need to be carefully controlled to avoid such inconveniences. However, when the correct grease is used in the right quantity (30 - 50 %
of the free volume in sealed/shielded bearings\textsuperscript{3}, churning losses can be considered negligible\textsuperscript{10}. This is one of the main reasons why grease is preferable to oil in applications where low friction is required. Grease is reported to be capable of reducing friction by a factor of threefold compared to its corresponding base oil\textsuperscript{11}. The choice of the right composition for the grease to achieve low friction will depend on the different ways in which it simultaneously affects these friction components, at the operating conditions of the bearing.

The primary role of grease in rolling bearings is to provide lubrication to the rolling elements/raceways concentrated contacts. Therefore, this study is focussed on the friction contribution given by these grease-lubricated EHL contacts. Here, friction is primarily determined by the lubricant film properties, including:

- Film thickness and the extent of surface separation;
- Film chemical composition and physical properties;
- Film formation dynamics.

Reducing friction is particularly important in high speed conditions, as the power losses $Q$ are proportional to the product of friction and speed. Indeed:

$$Q = T \times \omega = \mu \times W' \times \omega$$

where $T$ is the friction torque, $\omega$ the rotational speed, $\mu$ the friction coefficient and $W'$ the normal load. Therefore, since $U$ and $W'$ are fixed parameters, dictated by the operating conditions of the specific application, $\mu$ is the only manipulable parameter which can be potentially changed by employing different lubricants.

At low speeds, high friction is often associated to asperity interaction, due to the formation of lubricating films that are too thin to entirely separate the surfaces. Although this high friction is not particularly critical for power losses, because of low speeds and thus low $Q$ values, it does suggest the presence of inadequate lubrication conditions, which can be detrimental for the life of the bearing. High friction generates heat, which can lead to evaporation losses and oxidative thickening of the grease components, hence reducing its mobility. These circumstances could potentially affect the lubrication conditions also at high speed, with associated power losses and damage to the bearing. In fact, also in full film conditions, high friction is not only inconvenient for the power losses, but also harmful, due to the associated high temperatures.
Energy efficient greases should be suitably designed so as to reduce the overall friction whilst ensuring a long bearing life. The higher demand of energy efficiency required in modern bearings is associated with the minimisation of the lubricant film thickness\textsuperscript{12}, in order to reduce energy losses due to lubricant shearing and rolling resistance, and in turn lower the service temperatures. Ideally, the lubricant employed should have the minimum effective viscosity required at the expected operating temperature. However, this would imply the imminent risk of having a film too thin to separate the contact surfaces at low speeds, or when the relative motion stops intermittently. Hence, the selection of the right grease for friction reduction needs to be driven by a very meticulous, comprehensive reasoning. The grease components should be specifically selected using a holistic approach, as they mutually interact to determine film formation and its friction properties.

2.3 Lubricating greases

Greases are special lubricants with a complex rheology, resulting from a multiphase formulation which includes the presence of fluids and solids. They are employed in a large number of machines, and are the most preferred lubricant type in rolling bearings. With growing performance requirements, greases have acquired an important role in the design of rolling bearings.

2.3.1 Grease composition

The word ‘grease’ comes from the Latin crassus, which means ‘fat’. This is because in ancient times it was common to use animal fat (tallow), which is nowadays referred to as ‘cooking grease’\textsuperscript{11}, as a lubricant. In many languages the literal translation of grease has remained ‘fat’. In other languages, grease is called a ‘consistent mass’, since for centuries the term ‘grease’ has been used to loosely and generically indicate a certain category of thick, consistent lubricants. Today the definition of lubricating grease still remains ambiguous, and this is also due to the increased availability of many different components which can be used in grease formulation, thus imparting very different chemical and physical properties. A standard, concise but comprehensive definition of grease does not exist, as this would be too long to fully describe both its chemical composition and flow characteristics. However, some relatively simple definitions of grease can be found in the literature. For example, the American Society for Testing and Materials (ASTM) describes grease as ‘a solid to semi-solid product of dispersion of a thickening agent in a liquid lubricant’, often including other ingredients imparting ‘special properties’\textsuperscript{13}. The liquid lubricant, which is the dispersion medium, is in most cases a base oil.
The thickening agent, simply called ‘thickener’, consists of small solid particles. The remaining ingredients are additives. These components coexist representatively in weight percentages of approximately 85, 10 and 5 respectively\textsuperscript{12}, although these can vary much from grease to grease. Greases are often classified by the type of thickener used to give the structured matrix and consistency. The thickeners employed in greases are typically subdivided into three broad categories: soaps, complex soaps, and non-soap thickeners. The most widespread group of thickeners are soaps, produced by combining a fatty acid with an active metal. The thickener structure develops during the manufacturing process, and forms as a skeleton which penetrates the base oil throughout its volume. Figure 2.1 displays pictures of bulk lithium greases taken with a cryo-TEM (from literature\textsuperscript{14}, Figure 2.1 (a)), and with variable pressure SEM (on one of the samples analysed in this study, Figure 2.1 (b)). The three-dimensional matrix of lithium thickener fibres, appearing as stable particles dispersed throughout the base oil, is clearly shown. Thickener molecules interact with each other to keep the framework together by intermolecular forces including Van der Waals, hydrogen bonding and ionic forces\textsuperscript{1}.

![Figure 2.1. Examples of lithium-thickened grease microstructure, pictures taken with (a) cryo-TEM\textsuperscript{14} and (b) SEM](image)

The oil is thought to be bound to the three-dimensional thickener framework due to a series of dynamics, including capillary forces and mechanical occlusion\textsuperscript{15}. The classic analogy for grease is that of a sponge holding water\textsuperscript{12}; in the same way the thickener holds the base oil within its pore-like spaces, and this is released when the grease is sheared in service\textsuperscript{16}. Although the thickener is the component that differentiates grease from oil, and the grease ‘type’ is often identified by the nature of thickener used, many of the grease properties depend on the base oil. Moreover, the base oil characteristics, including solvency/polarity, chemical type, and
boiling range, substantially affect the structure formation and the thickening effect of the dispersed phase\textsuperscript{17}. As an example, Figure 2.2 shows SEM pictures of lithium thickener structures obtained in greases fabricated with the same amount and type of lithium thickener, but with base oils of different viscosities\textsuperscript{18}. It is clear that the characteristics of the base oil strongly affect the development in shape and dimension of the lithium thickener fibres.

![Figure 2.2 Lithium thickener structure obtained in greases formulated with a mineral oil of viscosity (a) 11 cSt, (b) 334 cSt and (c) 657 cSt at 40 °C\textsuperscript{18}](image)

The effectiveness of the forces that hold the framework ‘together’ depends on a number of factors: thickener type, quantity and morphology (shape, size and distribution), chemical structure and viscosity of the base oil, and ultimately the manufacturing process. As an example, Figure 2.3 shows the micrographs of two lithium greases having the same formulation, but manufactured with different processes to obtain distinct dimensional characteristics of the soap structure\textsuperscript{19}.

![Figure 2.3 Lithium thickener structures obtained by changing the manufacturing procedure: (a) fine and (b) coarse\textsuperscript{19}](image)

Additives can be included in the grease at any stage of the manufacturing process, depending on the type and usage. Typical multipurpose greases can contain friction modifiers, anti-wear,
extreme pressure, antioxidants, corrosion inhibitors, polymer and solid lubricants. To be able to perform well, the majority of additive systems are made with an optimised solubility in base oils, which can actually be a disadvantage when used in greases. Indeed most thickener structures are highly polar in character, and can cover the surfaces of the lubricated contact, thus preventing access to the additives. For this reason insoluble additives are preferred, for them to work correctly. These are often attached to the soap itself, so that they can take advantage of its strong polarity.

2.3.2 Grease rheology

Despite being made of mostly oil, grease shows remarkably different and complex rheological features that are dependent on the mutual interaction of its components. When a stress is applied to grease, both the thickener fibres and the oil resist deformation, opposing elastic and viscous forces respectively. Moreover, the grease structure, being fragile and easily deformable, is subjected to continual changes during handling, use, and with time. Obviously, depending on the extent to which grease is sheared and stressed, and for how long it is subjected to these conditions, its flow characteristics can be strongly affected. Grease can be defined, on the basis of its rheological characteristics, as a ‘lubricant which under the action of small loads at ordinary temperatures exhibits properties of a solid, and when the load has reached a critical value, starts deforming as a plastic and flowing like a liquid; when the load has been removed, it acquires again the properties of a solid’. This definition embraces some of the main rheological characteristics of grease, a material that is:

- Solid below a critical stress;
- Viscoelastic, showing characteristics of both a liquid and an elastic solid, above this critical stress;
- Thixotropic, i.e. the change in rheological properties with speed of deformation and time are, to a certain extent, reversible.

At very low stresses, no fluid behaviour can be observed in greases within a reasonable time. Under these conditions the fibre contacts are able to restrict the grease flow by mechanical obstruction. It is said that greases display apparent yield behaviour, i.e. they only experience irreversible flow after a minimum stress or strain has been applied, called ‘yield stress’. The yield stress is important for a number of practical reasons, since, for example, it prevents grease
leakage and determines its sealing capability in a bearing. Above the yield stress, grease flow behaviour is non-Newtonian, i.e. the relationship between shear stress and shear rate is not linear. In this respect, greases are classified as shear-thinning materials; they display a decrease in viscosity (often by several orders of magnitude) with an increase in shear rate or stress. Therefore in the case of grease it is more correct to use the term ‘apparent shear viscosity’, rather than viscosity, to merely express the ratio between the shear stress $\tau$ and the shear rate $\dot{\gamma}$. With increasing shear rates, eventually, a Newtonian value of viscosity is also reached in greases. This is generally different from their base oil viscosity, and is primarily dependent on the viscosity of the base oil, and the type and amount of thickener. The differences in the rheological behaviour of grease and its base oil are illustrated in Figure 2.4, where $\tan(\delta)$ is the viscosity of the base oil, and $\tan(\varepsilon)$ is the ‘apparent Newtonian viscosity’ of the grease, which is only reached at sufficiently high shear rates.

Figure 2.4 Schematic of the rheological behaviour of base oil (Newtonian) and grease (shear-thinning with yield stress)

There is no common formula that accurately describes the full rheological regime in the entire range of shear rates, but many rheological models are presented in the literature. Understanding the rheological behaviour of greases in the context of rolling bearings is of extreme importance, since both their static and dynamic properties highly affect their tribological behaviour. The range of typical shear rates to which grease is subjected in a bearing is very wide. Grease located on the bearing shoulders or on the seals only creeps and experiences shear rates as little as $10^{-6}$ s$^{-1}$. Conversely, grease lubricating the contacts at the interface between the rolling elements and the raceways is sheared at extreme values of the order of $10^{6}$ s$^{-1}$. At these shear rates the thickener microstructure degrades over time, and forms a dispersion of broken particles suspended in the base oil. Figure 2.5 shows the effect of prolonged shearing on the structure of a
lithium-thickened grease, in two pictures taken with an atomic force microscope. The thickener agglomerates appear greatly reduced in size, and more dispersed, after shearing.

![Figure 2.5 AFM pictures of (a) fresh and (b) shear-degraded lithium thickener microstructure.](image)

However, unless heavily shear-degraded, this process is macroscopically reversible. Cessation of shearing leads to re-solidification of the grease, and to a gradual recovery of its structural viscosity with time. Greases are thus classified as ‘thixotropic’ materials. Hence, the structural changes during flow are generally the result of the conflict between the incessant breakdown and reconstruction activity of thickener bonds. This process does not only depend on the level or duration of shear stress, but obviously also the mode in which the shear stress is applied, which is a function of the specific application. Due to the complexity of the degradation process, no universally accepted explanation for its mechanisms exists yet.

### 2.3.3 Use of grease in rolling bearings

Albeit an essential and very common product, lubricating greases only cover about 5% of the global consumption of lubricants, 1% of all petroleum products, and their market constitutes just 3% of the total amount of lubricants processed in the world. The inconsistency between their great versatility and common use on one side, and their relatively low manufactured and marketed volume on the other, is perhaps due to the relatively little amount of grease needed to lubricate a certain machine element.

Grease is extensively employed across different sectors, including many industrial and transport applications. Due to their solid-like consistency, greases are preferred to oils in applications where a spontaneous leakage of lubricant under the force of gravity is undesired. For example,
greases are most commonly used in mechanisms which can only be re-lubricated infrequently, or in areas where a continuous supply of oil cannot be retained. The same characteristic consistency provides grease with an inherent sealing capability, shock absorption action, and protection against corrosion. Rolling bearings account for a large proportion of grease use due to these lubrication requirements. In many applications, rolling bearings are only greased once, i.e. at the beginning of their life cycle. These sealed-for-life bearings are particularly employed in areas that are difficult or hazardous to reach for maintenance. Grease has a limited life, which depends on its composition, application and lubrication conditions. If a bearing is not re-lubricated, its life is ultimately determined by the grease life. Clearly, grease is an extremely important component of the bearing. Indeed greases are increasingly being regarded as a customised product, and part of the integral design of a rolling bearing, rather than a commodity at its service. In order to comply with the current high-standard specifications, the trend for the composition of modern bearing greases is moving towards the use of synthetic oils, novel thickeners and additives, and the employment of high-tech manufacturing processes. Likewise, thanks to the current improved understanding of grease behaviour in rolling bearings, grease lubrication is gradually being considered more as an interdisciplinary science, a complex mixture of chemistry, rheology, tribology, physics and engineering. Despite being neglected by the scientific community for many years, in recent times there has been a revival of interest in grease lubrication, possibly driven by the required higher efficiency and reliability standards for lubricated machine elements performance. With growing interests in environmental issues and energy efficiency, one of the most challenging demands is low friction.

2.4 Classical EHD lubrication

In order to discuss the possible mechanisms by which the formulation of grease can affect friction in bearings, it is important to first consider the general lubrication theory that applies to concentrated contacts. The type of contact existing between the rolling elements and the raceways in a rolling bearing, generating a considerable contribution to the overall bearing friction, belongs to this family. The classical lubrication theory for these types of contact is generally only valid when the lubricant is a simple, Newtonian fluid. Although this work is mostly concerned with friction in grease lubrication, the theory still offers an important background, setting the basis to evaluate and interpret the complex mechanisms of lubrication in greased bearings.
2.4.1 Introduction

The lubrication theory of concentrated contacts, where the load is applied over a tiny area of contact and generates pressures of the order of a few GPa, is relatively recent and only goes back to the 1940s\textsuperscript{31}. By this time, the Reynolds equation, describing the pressure distribution and load-carrying capacity of lubricating films in conformal contacts of journal bearings, was already 60 years old\textsuperscript{32}. The concepts behind the lubrication of conformal contacts had already been well established and verified for a variety of applications. However, lubrication in concentrated contacts, such as those found in rolling bearings, gears and cams, was poorly understood. For a long time it was impossible to explain how these machine elements could be effectively lubricated. Scientists soon realised that pure hydrodynamic theory was not applicable in these conditions. The calculated molecular-scale values of hydrodynamic film thicknesses could not be correct, as the observed low wear rates could only be justified by the existence of a film sufficiently thick to separate the contact surfaces. Therefore the existence of a somewhat different lubrication theory for concentrated contacts was suspected long before it could be proved or described using specific scientific concepts\textsuperscript{33}, but the first realistic model providing an approximate solution for film thickness was officially published only in 1949\textsuperscript{34}. The generation of two phenomena, induced by the typical extreme pressures of these contacts and crucial to the effectiveness of lubrication, needed to be considered jointly with the hydrodynamic theory:

- The increase in the lubricant viscosity under high pressure, incentivising the formation of a thicker film;
- The elastic deformation of the contact surfaces, forming a locally conforming area.

Because of these two effects, this type of lubrication is often referred to as ‘piezoviscous-elastic’\textsuperscript{35}. The lubricated contacts in which these effects take place are said to be operating in the elastohydrodynamic (EHD) regime, meaning that the contacting surfaces deform elastically under the great pressure generated in the lubricant film. Consequently, this type of lubrication is referred to as EHD lubrication, or simply EHL.
Figure 2.6 (a) Schematic representation of film formation in an EHL contact (not to scale) and (b) optical interferometry image of a ball-on-disc rolling contact (semi contact width ≈ 0.14 mm)

Three distinct regions can be distinguished in an EHL contact: the inlet, the Hertzian region, and the outlet. These are schematically shown in Figure 2.6 (a) for a representative point contact between a ball and a flat surface. The figure is purposely not to scale for clarity, and height is exaggerated. In reality, the ratio between the film thickness $h_i$ and the contact width is typically of the order of $10^{-3}$, thus the film is much thinner (in relation to the ball size) than as represented. The lubricant, which forms the film separating the surfaces in the Hertzian region, is entrained in the inlet and discharged in the outlet. Figure 2.6 (b) displays a typical optical interferometric image of a lubricated point contact. The following sections briefly review the mechanisms that rule film formation in the inlet, and the generation of sliding friction in the Hertzian zone of the contact.

### 2.4.2 Film formation

A thorough understanding of the film forming process must precede friction examination, since the film thickness establishes the shear rate in the Hertzian region of the contact, where EHL friction is generated. The formation of the EHL film starts to take place in the converging inlet region of the contact, as shown in Figure 2.6 (a). The available lubricant is entrained into this zone due to viscous drag forces exercised by the motion of the bodies. Consequently, the EHL film starts to form by generation of hydrodynamic pressure and relative motion of the surfaces, with tangential speeds $U_1$ and $U_2$. The pressure of the film gradually increases in the inlet region, and in turn facilitates its entrainment due to the significant increase in viscosity and therefore resistance to flow. This prevents the contact from squeezing the lubricant out. Within the Hertzian region the maximum pressure is of the order of a few GPa. At these high pressures the contact surfaces are elastically flattened and parallel, and separated by a film of essentially constant thickness $h_c$, which reaches the outlet of the contact almost undisturbed, with negligible
side leakage. Hence, the EHL film thickness is already established in the inlet, and will depend solely upon the rheological properties of the lubricant at the inlet pressure, which is of the order of tens of MPa$^{37}$. Under these conditions, the shear stress is still sufficiently low for most base fluids to behave in a Newtonian fashion$^{31}$. This makes it possible to predict EHL film thickness on the basis of the low shear rate properties of a Newtonian fluid.

Several equations exist to predict EHL film thickness in Newtonian fluids, the majority of which are regression equations fitted to numerical solutions. One of the most common equation for point contacts, extensively used in the current work, is Hamrock and Dowson’s$^{38}$, reported below:

$$
\frac{h_c}{R_x'} = 2.69 \bar{U}^{0.67} \bar{G}^{0.33} \bar{W}^{0.067} (1-0.61 e^{-0.75(\frac{R_y'}{R_x'})^{0.64}})
$$

where $\bar{U}$, $\bar{G}$ and $\bar{W}$ are the dimensionless groups defined as:

- Speed parameter, $\bar{U} = \frac{U \eta_0}{E R_x'}$
- Material parameter, $\bar{G} = \alpha E'$
- Load parameter, $\bar{W} = \frac{W}{E R_x'^2}$

with:

- $U$ the entrainment speed, i.e. the average speed of contact bodies 1 and 2, $U_1$ and $U_2$;
- $R_x'$ and $R_y'$ the reduced radii of the contact geometry in x and y direction respectively.
  \[
  \frac{1}{R_x'} = \frac{1}{R_{x1}} + \frac{1}{R_{x2}}, \quad \frac{1}{R_y'} = \frac{1}{R_{y1}} + \frac{1}{R_{y2}},
  \]
  $R_{x1}$, $R_{x2}$, $R_{y1}$ and $R_{y2}$ are the principal radii of the contact bodies;
- $E'$ the reduced Young’s modulus, linked to the material characteristics of the contact bodies by the relationship $\frac{2}{E'} = \frac{(1-\nu_1^2)}{E_1} + \frac{(1-\nu_2^2)}{E_2}$. $E_1$ and $E_2$ are the Young’s moduli, and $\nu_1$ and $\nu_2$ the Poisson’s ratios of contacting bodies 1 and 2 respectively;
- $W$ the total normal load;
- $\eta_0$ the lubricant dynamic viscosity at atmospheric pressure, and shear rate and temperature of the inlet;
• $\alpha$ the lubricant pressure-viscosity coefficient.

Given the geometry ($R_x', R_y'$), materials ($E'$) and application conditions ($U, W$), the central film thickness is governed by the values of lubricant dynamic viscosity $\eta_0$ and pressure-viscosity coefficient $\alpha$ at the operating temperature of the inlet.

In respect to the operating conditions, the effect of entrainment speed $U$ is very important, while load $W$ is not very influential. Indeed an increase in load contributes more to a further compression of the surfaces, rather than the lubricating film, without a significant contribution to the deformation of the inlet shape, and thus to the overall film formation process.

Obviously, in the case of grease, the film formation mechanism will generally be different, since this cannot be ascribed to a single, Newtonian component as for simple base oils. Grease is a far more complex rheological and chemical system. The nature of the entrained film (thickness, composition, microstructural characteristics, rheological properties), generally unknown, will determine EHL friction. This uncertainty represents a preliminary hindrance to the understanding of EHL friction behaviour of greases.

### 2.4.3 Friction

EHL friction, which is the focus of this work, is produced by the sliding motion of the contact surfaces, and it is therefore associated to the shear stresses and rates inside the Hertzian zone of the contact. As a consequence, it is clearly dependent on the thickness of the lubricating film. This determines both the extent of surface separation, and the shear rate operating on the lubricating film, on which friction depends.

The surfaces roughness plays a major role in determining the nature of sliding friction, as it establishes the conditions of film thickness in which asperity contact occurs. Below a minimum value of film thickness, which depends on surface roughness, friction is not entirely ‘fluid’, but it is also affected by the direct interaction of the asperities of the solids in contact. This condition is not only associated to the generation of high friction losses, but it also entails surface damage, and thus represents a potential risk for the reliability of the lubricated system.

Above this value of minimum film thickness, friction is entirely determined by the shear resistance of the lubricant film, which is often unknown. In fact, while the interpretation of the mechanism behind the formation of EHL films is generally understood, and the film thickness solutions (at least in full film conditions) represent an accepted approximation, EHL friction is still an unresolved topic. EHL film thickness and friction, due to the remarkably different pressure conditions in the contact regions where they are determined (the inlet for film
thickness, and the Hertzian area for friction), are in fact decoupled, and thus depend on different lubricant properties. In Section 2.4.2 it has been shown that, for a given set of operating conditions and contact geometry, film thickness formation is mostly governed by two ‘macro’ properties: ambient pressure viscosity $\eta_0$ (in agreement with hydrodynamic theory), and pressure-viscosity coefficient $\alpha$. Instead, friction has its origins in the lubricant molecular structure\(^3\), including molecular shape and flexibility\(^4\), which is obviously a much more complicated matter. At the typical severe pressures and shear rates that are found in the Hertzian region of EHL contacts, the lubricant film ceases to behave in a Newtonian fashion. In reality the lubricant is not able to reach the value of viscosity associated with the Hertzian contact pressure (piezoviscosity). As a consequence, the shear stress falls to much lower values than as predicted with Newton’s law, which establishes a proportional relationship between shear stress and shear rate. Despite the progress made, the physical explanation of this phenomenon has not reached consensus yet, and is still at the centre of a fierce debate\(^5\). Two main schools of thought exist to explain lubricant friction theory in EHL contacts. Corrections to these theories are implemented continuously to improve their ability to fit, or explain the experimental results. One of them is based on the observed frictional behaviour of lubricants in tests carried out on laboratory tribometers. The lubricating film is seen to display a non-Newtonian, shear-thinning behaviour above a certain value of shear stress, called ‘Eyring stress’\(^6\). The effect of shear heating, taking place with increasing shear rates, and associated with a reduction of viscosity, is also observed\(^7\). However, even when incorporating this effect, a condition of maximum, insurmountable shear stress is observed to occur at certain combinations of pressure, temperature and shear rate. This condition is believed to be due to a phase transition of the lubricant, from a viscous into a solid glassy state\(^8\), with obvious implications for the generated friction. Indeed under such conditions the lubricant cedes like a pseudo-solid rather than a liquid. The lubricant exhibits a rheological behaviour which is largely independent of viscosity, due to the fact that a solid can only withstand a finite shear stress. This, in turn, translates into a limited maximum value of friction. This value of stress, referred to as ‘limiting shear stress’, was first observed in experiments conducted using high-pressure rheometry\(^9\). The second school of thought bases its observations on experimental results obtained using these high-stress viscometers. According to this theory, the rheological response of lubricants under EHL conditions can be interpreted solely as a function of the limiting shear stress. Therefore, the lubricant involved in the EHL film formation can only acquire Newtonian or ‘limiting shear stress’ behaviour. It has been shown that both approaches can fit EHL friction data fairly well, especially in conditions of relatively low shear stresses\(^2\). Nevertheless, since the friction properties of EHL films are almost independent of the
atmospheric rheological properties of the lubricant, friction in EHL lubrication is still difficult to predict, and is usually measured using laboratory equipment, either tribometers or rheometers. In the end, it is generally recognised that a large part of the EHL theory is either confirmed or inspired by experimental observations\textsuperscript{45}, and this is particularly true in the case of EHL friction. A comprehensive understanding of friction in EHL contacts is further limited by a number of uncertainties related to the flow characteristics of the lubricant. These are not only associated to the lubricant rheology, but also to the interfacial forces which depend on the whole tribological system\textsuperscript{46}.

Despite the great attention given to energy efficiency of lubricated systems, the literature is currently lacking the availability of hard data on the frictional behaviour of lubricants in EHL contacts. This consideration applies even to simple base oils\textsuperscript{47}. Indicatively, there are strong suggestions of a positive correlation between pressure-viscosity coefficient and limiting shear stress\textsuperscript{48,39}. This relation implies that, while high values of pressure-viscosity are a desirable property in lubricants, as they yield thicker films (thus limiting or preventing solid-solid contacts), these might also be a characteristic of lubricants with a high value of limiting shear stress, giving high friction. Therefore, as it will be also stressed in the next section, the choice of the right lubricant is often the result of a compromise between two incompatible requirements: high film thickness and low friction.

2.4.4 The Striebeck curve

As mentioned in the previous section, when the lubricant film is not thick enough for the surfaces to be fully separated, friction becomes the result of two contributions: one hydrodynamic component, attributable to the shearing resistance of the lubricant, and one boundary component, which is dependent on the interaction between the surface asperities and the lubricant. The extent of surface separation, and the relative contributions to friction generated in a contact, determine different regimes of lubrication. The relationship between friction and film thickness can be visually represented in a so-called Striebeck curve\textsuperscript{49}. A typical Striebeck curve displays the way in which friction varies as a function of the classical Striebeck parameter (SP), defined as:

\[ SP = \frac{\eta_0 U}{W} \]
SP is purely a ‘service parameter’ to roughly estimate the film thickness, for friction trend representation\(^5\). The original work of Stribeck was on journal bearings, characterised by conformal contacts and therefore low pressures. However, in EHL contacts film thickness is influenced by the piezoviscosity of the lubricant in the inlet region, and friction depends on its shear-thinning properties at the high pressure of the Hertzian region. Both these properties are not contemplated in SP. Other service parameters can be used to express the relationship between friction and film thickness in an EHL contact. A commonly employed alternative parameter is the lambda ratio \(\Lambda\)^\(^{51,52}\), also known as ‘specific film thickness’. This is the ratio of lubricant film thickness to composite surface roughness of the two surfaces in contact:

\[
\Lambda = \frac{h}{\sqrt{R_{q1}^2 + R_{q2}^2}}
\]

where \(h\) is a representative value of lubricant film thickness (central or minimum), and \(R_{q1}\) and \(R_{q2}\) are the root mean square (RMS) surface roughness of the contacting surfaces of body 1 and body 2. Depending on the value of \(\Lambda\), the contact will work in different lubrication regimes. A generic representation of a Stribeck curve is given in Figure 2.7. The corresponding lubrication regimes, essentially defined by the extent to which the surfaces are separated by the lubricating film, are also indicated. These are defined based on two specific values of \(\Lambda\), i.e. \(\Lambda_f\) and \(\Lambda_s\), with \(\Lambda_f < \Lambda_s\):

- \(\Lambda \leq \Lambda_f\) - Boundary lubrication. At very low \(\Lambda\), the maximum value of the friction coefficient is typically found. The definition of SP suggests that either the lubricant has a low viscosity, the speed is too low, or the load is too high to form a separating hydrodynamically-generated film. The lubricant film is so thin that the load is almost entirely supported by the asperity contacts. This regime of lubrication is known as ‘boundary’. Under these conditions the role of adsorbed or reacted molecular films of additives is crucial in protecting against wear and reducing friction. Friction is governed by the boundary lubrication properties of the lubricant, which largely depend on the chemistry of the lubricant-additive-surface system. This provides the surfaces with lubricity, reducing typical dry-contact friction coefficients from 0.5 - 0.8 to much lower values of 0.1 - 0.15\(^4\);

- \(\Lambda_f < \Lambda < \Lambda_s\) - Mixed lubrication. As \(\Lambda\) is increased, friction starts decreasing due to the formation of gradually thicker films, partially separating the surfaces. In these conditions
the load is partly supported by asperity contacts, and partly by the lubricant, and therefore the contact is known to be operating in ‘mixed’ lubrication conditions;

- $\Lambda \geq \Lambda_2$ - Full film lubrication. The lowest friction coefficient is reached in correspondence to the minimum film thickness which guarantees a full separation of the surfaces ($\Lambda = \Lambda_2$). Above this value, the lubricant fully supports the load. Hence, here friction is of a fluid nature, and the contact is said to work in ‘full film’ lubrication conditions. From this point onwards, friction is determined entirely by the lubricant in-contact shear resistance, which generally depends on the operating conditions. The slope of the Striebeck curve in the full film region is positive, as shown in the figure, if shear heating is limited, i.e. at relatively low sliding, not too low temperatures, not too high viscosity and speeds.

![Schematic of Striebeck curve and corresponding lubrication regimes](image)

**Figure 2.7 Schematic of Striebeck curve and corresponding lubrication regimes**

The values of $\Lambda_1$ and $\Lambda_2$ are often not known, and are usually estimated experimentally. The lambda ratio is used more like a tool to approximately evaluate the extent of surface separation, rather than an absolute quantity on which to rely in the calculation of specific film thickness and therefore friction values. Indeed, references in literature report different values. Rough estimates for $\Lambda_1$ and $\Lambda_2$ are 1 and 3 respectively\(^{35}\). However, it is known that many engineering components operate successfully, without failure, with a calculated minimum film thickness of the same order as the surface roughness. Conformity and asymmetry in the surface topography often allow for a much lower ‘effective’ roughness than the measured, and therefore for higher ‘effective’ $\Lambda$ values. Typical values of nominal $\Lambda$ in rolling bearings are thought to be comprised between 0.8 and 2\(^{35}\). Due to this poorly understood process, the lubrication regimes are
sometimes defined on the basis of the measured friction coefficient value, rather than film thickness. Therefore, a value of friction coefficient $\mu > 0.1$ usually denotes that the contribution to friction given by asperity interaction dominates, and in these conditions the lubrication regime can be defined as boundary. Similarly, the shape of an experimental Stribeck curve can suggest the borders between the three lubrication regimes.

2.4.5 Starvation

An important assumption of classical EHL lubrication theory is that the quantity of available lubricant in the contact inlet is always sufficient for the lubricant film pressure build-up to start enough far upstream, and induce its full development\textsuperscript{45}. In this case the lubricant supply conditions are known as ‘fully flooded’. However, this is an assumption that, in reality, is not always satisfied. If the inlet is not fully filled, the film pressure build-up is shifted closer to the Hertzian loaded region\textsuperscript{54}, such that the actual formed film is thinner than as calculated by the EHL theory. When this condition, known as starvation\textsuperscript{55}, occurs, the EHL theory collapses. Some of its rules actually turn upside down, as for example the film thickness is shown to decrease with lubricant viscosity\textsuperscript{55} and speed\textsuperscript{56}. With increasing speed, eventually the central film thickness becomes comparable to the thickness of the film layer adsorbed on the contact surfaces\textsuperscript{55}, which in turn is proportional to the inverse of speed\textsuperscript{57}. Concurrently, the higher is the oil viscosity, the slower is the oil speed of flow. In turn, this means that the reflow time needed for the lubricant to replenish the contact inlet from the contact sides is longer\textsuperscript{58}. For example, in the operation of rolling bearings, the lubricant is pushed to the sides after each rolling element passage, and needs enough time to flow back onto the track to supply the contact inlet of the following rolling element. If the operating speed is too high, a viscous oil might not have sufficient time for replenishment, leading to a starved contact inlet.

However, starvation does not necessarily have a negative impact. For a given surface roughness of the contact, the lubricant can still be thick enough to provide full separation of the surfaces. The limited availability of lubricant in the inlet, leading to lower values of film thickness than in regular fully flooded conditions, also produces lower lubricant shearing losses. In fact, this condition can improve the overall efficiency of machine elements. With suitable design (geometry and surface roughness), and choice of lubricant, ‘partial starvation’ may actually be favourable in view of friction reduction.
2.5 Grease EHD lubrication

Despite the absence of an established theory on grease lubrication in concentrated contacts, the scientific literature reports a considerable number of research works on grease EHL lubrication, in both fully flooded and starved conditions. Grease film thickness in particular has been the subject of a significant amount of work, while grease friction is a somewhat less investigated topic. These studies have contributed to the understanding of the fundamental rules that govern EHL grease lubrication, often by comparison with the simpler and more conventional EHL theory for oil. Some of the more relevant studies are reviewed in this section.

2.5.1 Film formation in grease-lubricated contacts

Understanding film-forming behaviour in grease lubrication is a first important step towards improved grease formulations for energy efficiency and reliability purposes. As the grease main component is base oil, grease film thickness rules are often compared to the corresponding base oil rules, or interpreted as a function of them. Therefore the corresponding base oil film thickness behaviour, and the relative EHL theory, can still be used as a reference. However, while in oil lubrication the EHL rules can be applied with a certain degree of confidence, predicting film thickness in grease is more complex. The EHL theory might still be applied by replacing the base oil Newtonian viscosity with the grease non-Newtonian rheology at the contact inlet, provided that an adequate rheological model is employed. Accordingly, grease is modelled as a continuum, single-phase material with a shear-thinning behaviour. On the other side of the spectrum, part of the literature suggests that at the typical strong shear field of an EHL contact the thickener particles break down, forming a suspension in the oil medium. The result is that grease becomes ‘similar’ to an oil, and therefore is simply modelled as a Newtonian oil with a different (often higher) viscosity value. As the experimental work in the literature broadly suggests, both these interpretations oversimplify the true film formation mechanism. Furthermore, they disregard other important aspects, such as the effect of chemistry in the interaction between the lubricant and the contact surfaces. In reality, film thickness behaviour in grease lubrication is peculiar to the specific grease composition, and strongly dependent on the operating conditions. A universal theory on the topic does not exist yet. However, the experimental studies conducted so far can provide some common guidelines.

The majority of studies reviewed here were conducted using optical interferometry, which is one of the most popular techniques used to carry out research on EHL film thickness. Wedeven et al. were among the first to apply this method to grease, more specifically lithium greases, which were tested on a ball-on-disc device. The base oils were subjected to the same investigation for
comparison. The analysis revealed that grease film thickness behaviour as a function of speed is similar to that of a base oil. Nevertheless, grease formed thicker films than the corresponding base oil across the entire range of speeds used in the test. Moreover, thicker films were observed in greases containing a higher thickener content. These trends are shown in Figure 2.8. Dyson and Wilson found similar results in a twin disc machine using contact capacitance measurements to provide an indication of film thickness. In this study a trend was recognised also in relation to base oil viscosity, with greases formulated with a more viscous base oil displaying a higher increase in effective viscosity compared to the corresponding base oils.

Figure 2.8 Lithium greases and base oil film thickness results obtained by Wedeven

Wedeven et al. also observed a different shape of the EHL film as a function of the operating conditions. While at moderate speed this was horseshoe-like and thus similar to oil, at low speeds (and in static conditions, as shown in Figure 2.9 (a)) the film was very thick and heterogeneous when fresh grease was provided. However, after several overrolling cycles this film showed to become thinner and more uniform (see Figure 2.9 (b)).
Palacios et al.\textsuperscript{63}, also through optical interferometry, found equivalent results in terms of film enhancement by grease within the range of conditions covered in their tests. Based on their results, they also suggested that grease behaves as a Newtonian fluid in an EHL contact. Not only did they compare the film thickness behaviour of lithium greases (of several thickener contents) with their base oils, but also with suspensions of lithium thickener particles (with the same corresponding thickener content) in the same base oils. Only the greases with low thickener contents gave results similar to their suspensions. However, generally the greases formed thicker films than the suspensions, which in turn gave thicker films than the base oils. The conclusion was that, in general, grease films composition can be reduced to neither a base oil, nor a mere suspension of thickener in base oil. The influence of different thickener types on the grease effective viscosity was studied. It was found that, depending on thickener type and content, the enhancement in effective viscosity could range from negligible to very high. In a 30\% w/w lithium grease the effective viscosity was four times its base oil value.

Aström\textsuperscript{64} used interferometry to observe film formation at low speeds, for a selection of lithium and sodium greases manufactured using the same base oil. Sudden variations of film thickness were observed in the contact, locally disturbing the EHL horseshoe shape. These variations were more evident in greases containing more thickener. Moreover, different results were found according to thickener type: sodium thickener grease produced a temporary lift, while lithium grease gave very large film thickness variations within the contact. This was explained as a result of the different size of thickener particles in the two greases, the sodium thickener consisting of larger and longer fibres than the lithium thickener. The sketches of the corresponding contact interferograms, reproduced from the experimental results, are shown in Figure 2.10.
Figure 2.10 Sketches of EHL film shape observed by Aström\textsuperscript{64} at low speeds: (a) momentary lift with sodium grease and (b) sudden local variations with lithium grease

Cann et al.\textsuperscript{59} also used interferometry to observe film thickness formation in lithium and calcium greases at low speeds in a ball-on-disc tribometer. When compared to the base oils, a film increase comprised between 5 and 25 % for lithium greases, and between 35 and 81 % for calcium greases, was observed depending on thickener content. A reduction of film thickness was measured during the test. As suggested by the use of microspectroscopy, this time effect was attributed to a decreasing quantity of thickener being entrained with increasing time and speed, which in turn was explained as a result of both thickener degradation, and loss to the sides of the contact. The interpretation of the mechanism of film formation was described with a simple sketch, shown in Figure 2.11. According to this model, the grease structure starts to break down into smaller clumps of entangled fibres, separated by base oil, at a relatively long distance from the Hertzian contact. These particles are progressively sheared as they approach the inlet region, until they reduce to smaller, individual particles, which are partly expelled to the sides of the contact. It was suggested that the contribution of the thickener to the formation of the film depends on how the thickener affects the shear stability of the grease. The more the grease is shear stable, the more the thickener fibres are able to survive the shear rates and stresses in the inlet region, and thus the greater is the film thickness enhancement. Hence, the thicker film observed with the calcium-thickened grease compared to the film obtained with lithium-thickened grease was explained as the result of a higher shear stability.
However, the shear stability is also a function of shear rate. The same greases were tested by Williamson\textsuperscript{65} at medium speeds. As opposed to what was found by Cann et al.\textsuperscript{59} at low speeds, the lithium grease gave a thicker film than the calcium grease. Moreover, this study suggested that the thickener type not only influences the extent to which this participates to increase the effective viscosity of the base oil, but also the way in which it affects the rheology of grease in the contact inlet. Indeed, in the test conditions used in this study, it was found that the greases′ behaviour was either Newtonian (in lithium and calcium greases) or shear-thinning (in a polyurea grease). Hence, as previously indicated in the literature\textsuperscript{66}, these results led the authors to believe that not all the grease in the contact inlet is degraded. Instead, part of it can form a ‘core’ of bulk grease which remains undisturbed. Accordingly, entire grease structures would be allowed to survive and be entrained in the contact. Hence, the film formation mechanism proposed in Figure 2.11 was slightly changed according to Figure 2.12. It was suggested that the thickness of this core is related to the grease yield stress, controlling the resistance of the grease to deform plastically.

Figure 2.11 Grease film formation mechanism according to Cann et al.\textsuperscript{59}
Kaneta et al.\textsuperscript{67} investigated the influence of thickener structure (aromatic, alicyclic and aliphatic) of diurea greases on film formation, at low speeds. Although highly non-uniform within the contact due to the passage of thickener lumps, the shape of the grease films displayed a clear trend. The peaks and valleys of these films were found to be determined, respectively, by the thickener type and the viscosity of the base oil. Despite this decoupling, it was suggested that the base oil viscosity does influence the interaction of the thickener to the formation of such films, by controlling the entrainment of thickener fibres. Large fibres (as in the aliphatic grease studied in this work) do not easily cross the contact when in combination with a base oil of low viscosity (at the test temperature), and thus contribute less to film formation. In another study Kaneta et al.\textsuperscript{68} confirmed the relationship found between film thickness and thickener microstructure, and suggested that the thicker film obtained with the aromatic and alicyclic greases could be due to both thinner fibres and their better ability to adhere to the contact surfaces. Examples of the interferograms obtained in the film thickness measurements with the three greases are shown in Figure 2.13\textsuperscript{68}. However, the film thickness fluctuations were not observed anymore when mechanically worked greases were tested in place of fresh samples.
An Experimental Study into the Influence of Grease Composition on Friction in EHL Contacts

Figure 2.13 EHL film interferograms with (a) aromatic, (b) alyclic and (c) aliphatic diurea greases, as found by Kaneta et al.\textsuperscript{65}; semi contact width $\approx 0.18$ mm

Couronné et al.\textsuperscript{69}, from a comparison of film thickness, rheological and chemical measurements conducted on a series of model greases, also found that the contribution of thickener to film formation depends on thickener type; aluminium complex was found to traverse the contact, as opposed to lithium. In contrast, viscosity improver additives traversed the contact in every grease tested in this work.

Cousseau et al.\textsuperscript{70} compared both viscosity and film formation behaviour of a series of greases, and their base and bled oils. Base and bled oil viscosities were found to be generally different, due to the presence of thickener particles in the bled oil. As a consequence, this difference in viscosity was highly dependent on thickener type. The bled oil and grease film thickness measurements gave very similar results within the test speed range, suggesting that the bled oil properties, including viscosity and pressure-viscosity coefficient, could be used to predict grease film thickness under certain operating conditions. Depending on thickener type, the differences in viscosity and pressure-viscosity coefficients were more or less apparent. For example, a large increase in viscosity and reduction in pressure-viscosity coefficient was observed in a polypropylene-thickened grease.

Cen et al.\textsuperscript{71} conducted an investigation on the film thickness behaviour of a series of commercial greases. As had previously been observed by Hurley and Cann\textsuperscript{72} amongst others, the authors discerned between two regimes of lubrication, separated by a speed of transition. Above this speed, the grease film thickness behaviour was similar to that of a Newtonian base oil. Below this speed, the film thickness increased with decreasing speed, suggesting the entrainment or deposition of a thickener-rich film, so that in a log-log scale the film thickness versus entrainment speed curve appeared as ‘v-shaped’. An increase in temperature produced an increase of transition speed, but did not affect the minimum film thickness. The film thickness at low speeds, and the speed at which the transition between the two lubrication regimes occurred, were found to be highly dependent on thickener type. The results for a lithium and a calcium sulphonate complex greases are shown in Figure 2.14 (a) and Figure 2.14 (b) respectively. The
same authors carried out film thickness measurements on mechanically aged greases, obtained by degrading the fresh greases in a roll stability tester\textsuperscript{73}. Thinner films were formed by the aged greases at low speeds, although still thicker than in the corresponding base oils. However, fresh and aged greases approached the same film thickness values at higher speeds. In both works it was suggested that, at even higher speeds, the contribution of the thickener to film formation becomes negligible, and the formed grease film becomes equivalent to the base oil film, in direct contradiction to some earlier studies cited above\textsuperscript{55,63,70}.

![Figure 2.14](image)

Figure 2.14 Film thickness measurements for (a) lithium and (b) calcium sulphonate complex greases by Cen et al.\textsuperscript{71}; (c) film thickness measurements for polypropylene grease by Gonçalves et al.\textsuperscript{74}

Similar results were produced by Gonçalves et al.\textsuperscript{74}, who performed film thickness measurements with polypropylene-thickened greases, containing different thickener contents. At low speeds these greases formed thick, speed and temperature-independent films, thus appearing as a plateau in a film thickness versus speed curve. The results obtained from this study with one of the greases at different temperatures are shown in Figure 2.14 (c). When reaching a certain speed, the film thickness was found to decrease abruptly. A ‘transition speed’ was also identified
in this case, above which the grease film thickness started increasing in an EHL fashion. The transition speed, as well as the film thickness at values of speed above it, were found to increase with higher thickener content. However, the film thickness in correspondence to the low-speed plateau, and the film thickness at the transition speed, was the same for all greases, regardless of thickener content. These results suggested that at certain conditions of speed, the film thickness is entirely controlled by some properties of the grease thickener, and independent of its content or test conditions (speed and temperature).

Cyriac et al.\textsuperscript{75} identified a specific dimensional feature of the thickener that directly affects grease film formation. The existence of an intermediate regime of lubrication between low and high speeds was suggested. Here, the degree of thickening of the thickener on the base oil (resulting in an effective viscosity increase) was proportional to the ratio of thickener volume fraction to the size of the thickener particles. The participation of the thickener to film formation was found to be very low in lithium greases, medium in polyurea and calcium sulphonate complex greases, and high in lithium complex greases (with thickener particles size decreasing in this order). The result was confirmed by testing the lithium grease after a period of mechanical degradation. The decrease in particles size due to the degradation process resulted in an increased film thickness, i.e. more thickener was found to participate to the formation of the film.

Cousseau et al.\textsuperscript{76} studied the effect of both mechanical and thermal aging on greases, using a roll stability tester and an oven to simulate the two. It was shown that aged greases usually display higher film thickness than in fresh conditions. This was explained as a result of oil evaporation, (increasing the effective viscosity), and consumption of additives (allowing more thickener to be deposited on the contact surfaces).

Despite the numerous studies on grease film thickness, at present no universal model is available to correctly explain it. In oil lubrication it is possible to predict EHL film thickness from basic properties of the oil (viscosity and pressure-viscosity coefficient) with sufficient accuracy. However, generally the classical EHL theory cannot be used to describe film formation in grease lubrication. The ‘nature’ of the grease film, including its composition and process of formation (governed by hydrodynamic and/or adsorption/deposition rules), is usually unknown and seems to be strongly dependent on the operating conditions and grease properties. To complicate even more the understanding of the topic, it is observed that the occurrence of starvation is very frequent in grease lubrication.
2.5.2 Starvation in grease-lubricated contacts

The previous section has shown that, in the majority of studies on film thickness conducted in fully flooded conditions, grease is reported to form thicker films than its base oil at low entrainment speeds, and in some studies even at higher speeds. To avoid starvation, these studies were carried out at relatively low speeds, or with some external means used to forcibly fill the contact inlet with grease. In fact, experimental research indicates that grease is highly susceptible to starvation, as this inevitably occurs in correspondence to certain operating conditions. In oil lubrication, a bath or a jet lubrication system are often employed to provide lubricant to the contact. These are usually sufficient to deliver fully flooded conditions, unless the lubricated system operates at very high speeds or low temperature. On the contrary, grease is typically just applied to the lubricated surfaces before the running-in. Subsequently the supply conditions rely on the grease natural ability to feed the contact, which is also a function of the bearing geometry and operating conditions. As bearings lack the presence of a forced, controlled bulk supply mechanism, the availability of lubricant in the inlet is determined by a balance of lubricant lost from and supplied to the track. The lubricant is easily lost due to displacement, and cross or squeeze flow from the contact. Grease is removed from the inlet zone of the contact due to the rolling action of the contact surfaces and centrifugal forces, and does not readily flow back in bulk form to re-supply the contact due to its rheological properties, particularly the yield stress. However, because of the same rheological features, grease is able to naturally form a reservoir around the contact, which subsequently feeds it by a number of possible mechanisms. In starved conditions, grease film thickness can reach lower, or higher levels than what could be achieved using the corresponding base oil, depending on the operating conditions and the grease composition. The understanding of grease starvation mechanisms is particularly important in the lubrication of rolling bearings, as they usually operate in full film conditions only during the initial churning phase, just after the bearing has been filled with grease. Following this phase, as grease is pushed aside by the passage of rolling elements, thinner films will form, and the bearing will usually run under ‘partial’ starvation conditions. This means that, despite the fact that the inlet is usually not fully flooded, the film is still reasonably thick to suitably lubricate the surfaces and avoid failure.

Poon monitored grease film thickness decay with rolling time using magnetic reluctance in a twin disc machine, testing lithium, sodium and clay thickened greases. In all the tests with grease the film thickness reduced, with time, to values that were lower than in the corresponding base oils. The result obtained on a sodium grease is shown in Figure 2.15. A contraction of the inlet meniscus, which was related to a decrease of lubricant supply, was observed. However, these
findings were mostly explained as a result of grease degradation. It was suggested that the reduction of grease supply induced higher shear rates in the contact inlet, contributing to a further reduction of film thickness due to a faster process of mechanical degradation. Similar results were found by Wilson in rolling bearings lubricated with a lithium grease. However, in this case the results were attributed entirely to starvation, rather than to a time effect.

Figure 2.15 Grease film thickness decay with time and comparison with base oil as observed by Poon

In regard to the effect of grease properties on starvation behaviour, in Poon’s work it was reported that the preservation of thick films is favoured by a combination of coarse thickener structure and low viscosity base oil. Accordingly, it was suggested that the choice for the optimal grease base oil viscosity is the result of a compromise: sufficiently high viscosity is required to produce thick films, but the viscosity must also be sufficiently low to retain these films inside the contact.

The correct operating conditions of concentrated contacts in starved conditions not only rely on the grease capability to be retained within the rolling track, but also on its ability to replenish the contact. Aström focussed on this mechanism by observing the grease reservoir at the side of a ball-on-disc contact lubricated with grease. A sketch of the observed grease distribution around the starved lubricated contact is reproduced in Figure 2.16. The figure shows that the grease present in the primary reservoir (i.e. in the area close to the contact) reaches out the rolling track in correspondence to the ball position, hence providing replenishment. This was explained as the result of combined extensive shear, softening the grease and making it behave in an oil-like fashion, and surface tension forces. The same forces would help holding the reservoir in place.
Cann\textsuperscript{20} used interferometry to study the starved film formation behaviour of a lithium grease in a ball-on-disc rig. The results suggested that replenishment is dominated by two mechanisms. One is bulk grease flow, associated with the motion of the ball, squeezing the grease adjacent to it and dragging it back to the track. The second mechanism is also initiated by the overrolling action of the ball, releasing bled oil from the grease reservoir at the side of the rolling track. Then, this bled oil feeds the contact by reflow, driven by surface and capillary forces, acting closer to the track and forming a local supply around the contact. This concept is illustrated in Figure 2.17, taken from another work by Cann\textsuperscript{54}. Interferometry images of a starved, greased ball-on-disc contact, from a test performed at constant speed, are shown in the figure. At the beginning of the test (Figure 2.17 (a)), the film is relatively thick, despite the contact being heavily starved. Figure 2.17 (b) shows that after many revolutions, most of the lubricant gets removed and the separating film becomes very thin. Here, the film is actually thicker at the sides of the contact, where the ball squeezes and overrolls the adjacent grease. This incessant action degrades the grease, which consequently becomes more mobile and replenishes the contact in a more efficient way, as is evident in Figure 2.17 (c) where the film becomes thicker. Longer rolling times\textsuperscript{81} (further degrading the grease at the sides of the contact) and higher temperatures\textsuperscript{20} (reducing the viscosity of the bled oil) are thought to improve the supply conditions by increasing the quantity of available bled oil, and the speed at which it flows back to the contact respectively. As for oil lubrication, the common rules that govern grease film formation in fully flooded conditions as discussed in Section 2.5.1 are inverted when the contact inlet starves. In particular, starvation increases with higher rolling speed, base oil viscosity and thickener content\textsuperscript{82}. These factors
reduce, respectively, the available time for base oil reflow, the bled oil reflow speed, and the grease oil bleeding capability.

Figure 2.17 Interferograms of a starved ball-on-disc contact lubricated with grease, showing the evolution of film thickness with time; semi contact width ≈ 0.14 mm. Reproduced from Cann54

The base oil viscosity is also considered to affect starvation behaviour in relation to the mechanisms that govern thickener deposition. Vengudusamy et al.83, based on observations from a study carried out on lithium greases with different base oil viscosities, proposed the mechanism of grease starvation behaviour sketched in Figure 2.18. According to this model, at low speeds there is sufficient grease to provide fully flooded conditions. Therefore, as described in Section 2.5.1, the film thickness versus speed curve appears as v-shaped in a log-log plot (‘Region I’ and ‘Classical EHL region’). In both regions the film is said to be composed of a hydrodynamic component (made of bled oil, consisting of thickener particles discretely dispersed in base oil), and a static component (mostly composed of thickener particles, deposited on the surfaces at very low speeds). When Region II is approached, the onset of starvation is reached. This condition was found to occur in correspondence to the same value of the ‘Stribeck parameter’, defined in Section 2.4.4. Region II corresponds to an initial phase of starvation, where the hydrodynamic contribution to film thickness gradually decreases due to increasing speed. Finally, Region III is reached, where the film thickness is stabilised to a constant, speed-independent value. Here, the measured film thickness had approximately the same value as the static films measured at zero speed. It was suggested that, in these conditions, film formation is mostly dominated by the thickener and additive system of the grease, and not the base oil viscosity. The latter is only indirectly important, as it affects the mechanisms that control thickener entrainment and deposition. Indeed, thicker static and stabilisation films were formed at high temperature, or with low viscosity base oil greases. This result was interpreted as a consequence of the reduced availability of oil in the contact inlet, favouring the entrainment and deposition of thickener.
Another aspect to consider in starved grease contacts is surface chemistry, as this severely affects both film adhesion and oil reflow. Cann\textsuperscript{84} observed the appearance of grease films deposited in the rolling track of a glass disc, at the end of a starved ball-on-disc test. Depending on the grease formulation, the layer of lubricant appeared as a thick, viscous layer (Figure 2.19 (a)), or as a dispersion of isolated oil droplets (Figure 2.19 (b)). Figure 2.19 (b) corresponds to a test where the grease, based on an ester oil, failed to suitably replenish the contact. It was suggested that the presence of a non-wetting surface chemistry deleteriously inhibits the deposition of grease, and its back flow towards the rolling track. Huang et al.\textsuperscript{84} also found that greases formulated with ester oils, generating a finer thickener microstructure, are less prone to bleed, and thus less able to replenish the contact. In the same work Huang et al. suggested that the thickener itself can also give a relevant contribution to replenishment. If the thickener particles are able to resist shearing forces without breaking (as was found with the SiO\textsubscript{2} thickener), this can further lift the contact surfaces by flowing together with the base oil.
Grease reflow is seen to be highly affected by thermal aging. As found by Hurley et al.\textsuperscript{85} in a study on lithium greases, thermal aging causes evaporation of low molecular weight base oil species and base oil oxidation, resulting in a viscosity increase which slows down the reflow of bled oil. However, with moderate thermal aging, oil release is improved, hence counterbalancing this effect. The opposite was observed with long aging times. With the combined reduction of oil bleeding and increase of viscosity, grease fails to correctly replenish the contact, and severe starvation occurs.

Low pressure rheology also plays a major role in starved conditions, as it affects grease behaviour at the sides of the contact, which is responsible for replenishment. Greases with lower shear stability, therefore with a structure that is more prone to lose oil, are likely to form thicker films in starved conditions, as found by Cann and Lubrecht\textsuperscript{86}. Couronné et al.\textsuperscript{87} studied the starved film thickness behaviour of a series of greases containing lithium and aluminium complex thickeners, mineral and ester oils, with very different mechanical properties. They used pure rolling conditions and a relatively low contact pressure of 300 MPa. No direct effect of the parameters that are usually taken as indicators of the grease capability to produce thick films in starved conditions, such as oil bleeding and mechanical stability, was detected. However, it was noticed that certain combinations of oils and thickeners can lead to the development of strongly interconnected structure, resulting in a higher elasticity, making them more susceptible to starvation. For example, this was noticed to occur in lithium-thickened ester base oil greases, due to the high solvency of ester base oil towards the lithium soap. It was suggested that the interaction between the grease single components is more important than the single components on their own.

In summary, these studies have shown that, as in oil lubrication, generally the rules that govern grease film formation under starvation conditions are opposite to the ones that govern its
behaviour in fully flooded conditions. The understanding of grease starvation is particularly important in view of the fact that, in practical applications, starvation occurs in grease lubrication much more frequently than with oil. However, as in oil lubrication, generally several degrees of grease starvation exist, and partial starvation can be advantageous if accompanied by a reduction in viscous friction while still providing sufficiently thick films for surface protection. An emblematic anecdote is the accidental result obtained during an experiment conducted on a large journal bearing by Vermeulen. A grease supply pump was used to provide fully flooded conditions during the test, while friction was continuously monitored. The grease supply pump failed to work for eight hours, during which the machine was left unattended. Unexpectedly, the measured friction had gradually decreased during the test, until reaching half the value measured when the grease pump was on (see Figure 2.20). Correspondingly, the grease layer separating the bearing surfaces, estimated to be about 5 micron thick, was slowly, but not entirely, removed.

Therefore, as in the case of oil, in grease-lubricated applications there is an optimum value of film thickness which minimizes friction whilst protecting the surfaces from asperity contact. Continual flow of bulk grease is undesirable also because, by producing high torques and excessive temperatures, it leads to a faster deterioration of the grease properties through extensive milling, thus reducing its life.
2.5.3 Friction in grease-lubricated contacts

More often than not, the properties of the oil, particularly its viscosity, are used to model grease lubrication in practical applications. However, there is considerable evidence that the thickener does participate in the lubrication process\(^9\), and thus can influence friction:

- Directly, by affecting the shear strength of the film\(^9\);
- Indirectly, by determining the film thickness, and therefore the lubrication regime\(^9\).

Most of the studies on grease lubrication have focussed on film formation rather than friction\(^9\). Additionally, the available published work on grease friction is rather discontinuous: the few existing research works have been conducted with very dissimilar test geometries, operating conditions, and grease formulations. These studies can be divided into two main categories, depending on the test methods:

- Tests with laboratory devices, with carefully controlled and known lubrication (inlet supply and lubrication regime) and operating (temperature, load, slide/roll ratio) conditions;
- Tests with full bearings, more representative of real applications. However, as shown in Section 2.2, they carry the disadvantage of encompassing numerous, uncontrolled variables. The resulting, inevitable uncertainties make the interpretation of the results difficult, and often tackled with a series of assumptions.

The literature on grease friction is presented here according to this classification order. One of the first studies on grease friction was conducted by Godfrey\(^9\), who used a pin-on-disc tribometer to test the effect of temperature on the boundary lubrication properties of a series of greases. A temperature increase produced a decrease of friction with certain soap-thickened greases. As the opposite was observed in the corresponding base oils, this effect was attributed to the decreased shear strength of the soaps with the increased temperature. The corresponding dry soaps were also subjected to the same friction tests, confirming the results. The same trends were observed in tests carried out in a real bearing: grease friction was found to decrease with heating, and increase with cooling. Base oils gave a contrasting result as expected at the employed conditions, due to the reduction of film thickness and consequent greater contribution to friction given by asperity interaction under the mixed/boundary lubrication regime. This study
provided evidence of the participation of the soap, and its effect on friction, in grease lubrication of concentrated contacts.

The adsorption properties of lithium thickener were studied by Rong-Hua \(^{34}\) in a 4-ball machine. It was found that these properties depend on the carbon number of the fatty acid, and whether this is odd or even. Better adsorption, and consequently lower friction, were observed in greases made from low and odd (with the methyl and carboxyl groups on the same side of the molecule) carbon number fatty acids.

Figure 2.21 Traction curves displaying friction against slide/roll ratio under fully flooded conditions for a 5 % and a 15 % w/w lithium greases and their base oil, at 60 °C and 0.5 m/s \(^{32}\).

Cann \(^{32}\) investigated the effect of thickener content on friction, in two lithium greases made with 5% and 15% w/w thickener, in a ball-on-disc machine and under fully flooded conditions. Both greases gave lower friction than their base oil, particularly the one with a higher thickener content. The results obtained in this study are shown in Figure 2.21, plotted against slide/roll ratio as ‘traction curves’. The difference in friction between the greases and the base oil was attributed to the physical deposition of thickener (favoured in the grease containing a higher thickener content), and the consequent formation of a low-shear strength boundary layer. Elsewhere the formation of this low-friction boundary layer was found to be incentivised with longer shearing time \(^{95}\). Other researchers have explained this reduction of friction with time as a result of grease mechanical degradation with continuous shear, equivalent to a decrease of effective viscosity with time as usually observed in a rheometer \(^{36}\).
Cann\textsuperscript{4} also investigated the friction properties of a series of model greases, containing different thickener types and the same mineral base oil, in a ball-on-disc rig. Substantial differences in friction were observed at all speeds, and particularly at high temperatures. The results obtained in this study with the lithium, lithium complex and tetraurea greases at 100 °C are shown in Figure 2.22. Friction was very dependent on thickener type: compared to the base oil, it was lower in the lithium complex, similar in the lithium, and higher in the tetraurea grease. At low speeds and high temperatures, the friction coefficient versus speed curve shows the inverse shape of a ‘classical’ Stribeck curve. Therefore this result was referred to by Cann as ‘inverse-Stribeck’ behaviour. Indeed, friction decreases with lower speeds, instead of increasing as may be expected in presence of a thinning film and growing contribution given by surface asperity interaction. Again, this behaviour was ascribed to the formation of a low-friction ‘boundary’ film. However, not all the thickeners behaved in this way: the calcium sulphonate complex grease gave high friction at low speeds, and scratches were found on the disc used for the test.

![Figure 2.22 Friction coefficient with lithium (LiOHM), lithium complex (LiXM) and tetraurea model greases, formulated with the same mineral base oil, as measured by Cann\textsuperscript{4}](image)

Gonçalves et al\textsuperscript{97} studied the friction behaviour of a series of polymer greases containing different thickener content in a similar ball-on-disc rig. It was shown that, even in the case of a non-polar thickener grease, friction is strongly reduced when it is compared to friction in the base oil, especially at low speeds. Some of the results obtained in this study are shown in Figure 2.23 and Figure 2.24, where the coefficient of friction is plotted against a modified, dimensionless Strubeck parameter $Sp$\textsuperscript{98}, accounting for the effect of speed, load, viscosity and
A trend was recognised (Figure 2.23): with higher thickener content, friction at low \( Sp \) values and the maximum measured friction are lower, and the \( Sp \) value up to which the presence of more thickener gives lower friction is higher. This result was attributed to a higher frequency of thickener lumps passing through the contact, or a higher consistency value, with greases containing more thickener. The Stribeck curves of one of these greases and its base oil are compared in Figure 2.24 at different temperatures. The grease always displays lower friction than its base oil, and this effect becomes more evident at high temperatures.

![SRR 5% chart](image)

**Figure 2.23** Effect of thickener concentration of polymer greases on friction as measured by Gonçalves et al.\(^7\): thickener content increases in the order M1, M2 and M3

![Friction behaviour chart](image)

**Figure 2.24** Polymer grease and base oil friction behaviour at different temperatures as measured by Gonçalves et al.\(^7\)

The effect of temperature on friction is highly dependent upon the combination of thickener and base oil in the grease formulation. By integrating the area under the Stribeck curves obtained with a ball-on-disc rig, Lorimor et al.\(^9\) calculated the power loss (per unit load) given by different
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greases over the whole range of test speeds. The grease formulated with polyurea and mineral oil, and lithium complex and PAO oil, generated higher friction with higher temperature. In the polymer and PAO oil grease, friction decreased with temperature at all speeds. However, the comparison was made between greases with very different compositions. Moreover, the observed trends were not discussed in relationship to the active lubrication regimes, i.e. the study was not complemented with film thickness measurements.

As for film thickness behaviour, aging of greases also has an important effect on frictional performance. Recently Gonçalves et al. investigated the friction behaviour of polymer and lithium greases as fresh and after thermal aging. The aged greases showed a very different frictional and film thickness behaviour depending on thickener type. Generally the effective viscosity and pressure-viscosity of these greases changed after thermal degradation, and this was taken into account in the evaluation of the friction results. While thermal degradation did not significantly change the tribological behaviour of lithium greases, the polymer greases behaviour was strongly affected. The trends in friction for the polymer greases changed according to some mechanism that could not be explained purely on the basis of film thickness variation.

The effect of grease rheology on friction has been investigated in a limited number of works. Delgado et al. managed to separate the effect of rheology from the composition, by fabricating greases with the same formulation, but different manufacturing procedures giving different rheological properties. The isolated effect of grease rheology on friction was evaluated in ball-on-disc tests. Several linear trends between some rheological (viscous and viscoelastic) parameters and the friction coefficient were found. However, despite the remarkably different rheological properties, the differences in friction were shown to be relatively small, indicating that the low-shear, low-pressure rheological characteristics of greases are not the main influential factor in determining friction, at least in full film conditions. Nevertheless, since grease rheology is one of the main factors determining lubricant supply to the contact in starved conditions (see Section 2.5.2), this is expected to also influence friction when the lubricant inlet is not full. Heyer and Läuger investigated the influence of grease consistency on friction using a modified rheometer based on a ball-on-plates geometry. Greases with different NLGI numbers (conveying the ‘stiffness’ of grease, see Section 3.3.4.1) were used as samples. Greases with high NLGI numbers gave lower static friction, probably because a high yield also entails the formation of a thicker film at the start of the test. However, the same greases produced high friction at high speeds, due to a lack of back flow which led to starvation.

Rolling bearings have been employed in a number of research studies on grease friction. Wikström and Höglund stressed the importance of grease base oil type on the start and steady
torque of a cylindrical roller bearing, at low temperatures. PAO base oil greases were found to give lower starting torques than naphthenic base oil greases. No differences were detected between base oils and the corresponding greases. The same authors investigated the relationship between this starting torque and the rheological/tribological characteristics of the grease\textsuperscript{102}, focussing on several parameters of interest. The base oil viscosity was recognised as the only characteristic to have a direct effect on friction, with lower viscosity base oils giving lower friction under the test conditions employed in this study.

Muennich and Gloeckner\textsuperscript{103} used a thrust cylindrical roller bearing to study the effect of thickener type on film thickness and friction, in fully flooded and starved conditions. The results they found are shown in Figure 2.25. The greases were prepared with the same base oil, but different thickeners. In fully flooded conditions, friction was very dependent on thickener type, and generally higher in the greases that formed thicker films. It was observed that the dependence of viscosity on temperature is different in greases and base oils, and general conclusions on the thickener ability to control film thickness and friction behaviour could not be made, as the observed trends varied as a function of temperature. The thickener type was crucial also in influencing the onset of starvation and the final film thickness and friction, both becoming almost speed-independent. Depending on the degree of starvation, friction was either higher or lower than with the base oil. In general, however, it was lower than in the grease fully flooded case, indicating a reduction of viscous friction. It was also noticed that an increased thickener content produces a higher film thickness in fully flooded conditions, but anticipates the speed at which starvation occurs. It was concluded that the thickener can contribute to friction through its capability of increasing the Newtonian viscosity of the oil in the inlet of the contact, or its impact on starved film thickness and friction behaviour.
Wilson\textsuperscript{79} carried out film thickness measurements on a double-row spherical roller bearing and a single-row cylindrical roller bearing, lubricated with a lithium hydroxystearate grease and its base oil. A relative comparison between the friction values given by these lubricants was also made based on temperature measurements. It was found that, under the same operating conditions, the bearings starved when lubricated with grease. However, because of a decrease in viscous friction due to starvation, the self-induced temperature remained relatively low. As a consequence, the formed films were thicker or almost as thick (depending on the bearing type) as the ones produced by the base oil in fully flooded conditions. Indeed, due to the higher temperature generated in full film conditions with the base oils (producing higher viscous friction), the film thickness decreased due to a reduced base oil viscosity. Therefore, film thickness diminished in both cases, with grease because of starvation, and with oil because of shear heating. Despite the generation of similar film thickness in both cases, the corresponding relative decrease in friction with grease, deductible by a comparison between the measured temperatures, was much more evident.

Cousseau et al. did extensive work on a modified 4-ball machine\textsuperscript{104} to study friction in rolling bearings. A comprehensive analysis into the influence of base oil type and viscosity, thickener type, and the effect of their interaction on the rheological and tribological properties of greases of different compositions was advanced. Greases made of different combinations of base oils (mineral, PAO, ester and vegetable oil) and thickeners (lithium and PTFE, calcium, polyurea, polypropylene) were tested\textsuperscript{91}. It was stressed that the presence of thickener can highly impact the

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**Figure 2.25** Friction and film thickness results obtained by Muennich and Gloeckner\textsuperscript{103} in tests with cylindrical roller bearings in (a) fully flooded and (b) starved conditions with bentonite (A), lithium (B), sodium (C), calcium complex (D) and barium (E) greases;
overall rheological properties of grease\textsuperscript{105,106}, which in turn affect film formation and lubrication regime, by influencing:

- The viscosity of the bled oil, believed to be the ‘active’ lubricant participating in the lubrication process. At low speeds, the higher is the viscosity of the bled oil, the thicker is the film. This mechanism can reverse at high speeds, where a thicker bled oil, by replenishing the contact more slowly, can form thinner films due to starvation;
- The ‘flow index’, which is an indication of the shear-thinning behaviour and thus the film-forming capability. The lower is its value, the more the grease shear-thins, and the higher is the grease capability to replenish the contact.

It was suggested that, depending on the lubrication regime in which the bearing is running, starvation can either increase or decrease the overall friction. Limited lubricant supply is only desired if the conditions are such that a full separation of the surfaces is maintained. Below this minimum film thickness, further starvation increases the contribution of asperity contact, thus increasing friction and producing surface damage. Consequently, it was suggested that there exists an ideal combination of bled oil viscosity and flow index value which minimise friction. In order to discern between distinct contributions to the friction generated in the bearing, the SKF rolling bearing friction torque model was applied to better interpret the findings\textsuperscript{10}. Since the bearing used had no seal, and the drag losses are negligible when grease is used as the lubricant, the overall torque was separated into the rolling and sliding components only. Figure 2.26 displays some of these results\textsuperscript{107}, where the total friction torque (Figure 2.26 (a)) is divided into its rolling (Figure 2.26 (b)) and sliding (Figure 2.26 (c)) components. Based on the friction results, the greases were classified in three groups: mineral base oil, ester base oil and polymeric (containing polymer, i.e. polypropylene or PTFE) greases, giving lower bearing torques in this order. It was suggested that greases based on ester oils, with high values of flow indices, tend to starve and operate in mixed/boundary lubrication conditions, generating relatively high sliding torques, possibly higher than as predicted on the basis of the SKF model and shown in Figure 2.26 (c)\textsuperscript{106}. The most efficient grease (PPAO) was formulated with a PAO base oil of low viscosity, thickened with polypropylene. The low friction was explained as the result of the equilibrium of convenient properties that, overall, reduced both the rolling and the sliding friction, especially at low and high speeds. Supposedly, rolling friction was low because of the formation of thinner films at high speeds, due to poor replenishment. This was due to the high viscosity of the bled oil (which was almost three orders of magnitude higher than in the base oil),
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given the thickening effect of the specific polypropylene thickener used in this grease. Moreover, a series of reasons were suggested as responsible for low sliding friction:

- At low speeds, because of the low flow index value of the grease, able to easily release the high viscosity bled oil, forming thick films and thus limiting asperity contact;
- At high speeds, because of the ‘nature’ (i.e. type or molecular structure) of the base oil, in this case a PAO. A correlation was proposed between friction and the ‘lubrication parameter’, defined as the product of dynamic viscosity $\eta_0$ and pressure-viscosity coefficient $\alpha$ of the base oil: the lower the lubrication parameter, the lower the friction$^{105}$.

The above review has attempted to include the most relevant studies for the current research work, although there is very little published work on friction mechanisms in grease-lubricated contacts. A selection of studies have focussed on the effect of operating conditions, such as degradation time$^{108}$, high$^{93}$ or low$^{101,102}$ temperature. Others have attempted to explain grease friction through rheometry$^{106,99}$. Only a few have explored the effect of grease chemical composition on its friction behaviour, on a thrust ball bearing$^{105,91}$, four-ball machine$^{94}$ and ball-

Figure 2.26 (a) Total, (b) rolling and (c) sliding torque, with greases LiMi (lithium + mineral oil), LiMi2 (lithium + mineral oil), LiE (lithium + ester oil), LiPuE (lithium/calcium and polyurea + ester oil), PPAO (polypropylene + PAO oil), PE+V (polypropylene + ester and vegetable oils) as measured by Cousseau et al.$^{107}$
on-disc machine. Some of these studies have shown the importance of the base oil in grease formulation, with low viscosity and low pressure-viscosity oils suggested as a preferred choice for low friction in EHL conditions. Furthermore, thickener type, composition and texture have also been shown to affect friction, with some thickeners able to reduce friction, particularly under boundary and high temperature conditions. However, it is clear that grease frictional behaviour cannot be entirely ascribed to one single component of grease, but rather it is determined by the interaction between all grease elements, which in turn strongly depends on the specific operating conditions.

2.6 Models of grease lubrication in rolling bearings

A unanimously accepted theory of grease lubrication mechanisms in rolling bearings does not exist yet. In fact, the main reason for design limitation of bearing greases is the lack of understanding of their tribological behaviour, which leads to unpredictable and sometimes inexplicable failures. A number of different interpretations exist, explaining how grease lubricates the bearing surfaces. Some of their aspects collide, some come to an agreement. One of the main uncertainties, and a topic of wide discussion, regards the nature of the lubricating film. The existing theories are here reviewed to contextualise this research work.

The ‘bulk’ model states that the lubricating film is made of none other than grease. In this case the lubricating film would have the same chemical composition and structure of the bulk grease. However, reported evidence supporting this theory is inexistent. Furthermore, it is thought that the flow of the entire grease through the contact would generate excessively thick lubricant films, with consequent disproportionate friction losses and heating.

A widely accepted theory is that grease is a reservoir of oil, which is supplied by bleeding. Base oil would be liberated from the grease close to the track, as it is progressively worked by the rolling action of the elements. Therefore this oil would feed the contacts of a rolling bearing by a replenishment mechanism. This is the ‘sponge’ model, where grease is the sponge that holds oil in place and releases it when needed. In this case the ability of the grease to bleed oil, and the base oil viscosity, are taken as an indication of lubricating performance. In this respect, the use of a low viscosity base oil, low thickener content, and poor shear stability grease would facilitate oil bleeding. In this model the thickener does not play an active role in the lubrication process. Accordingly, grease friction is entirely attributable to the properties of the base oil. Full bearing tests performed with greases and their base oils, with results that show no difference in friction torque values, and in grease life, are in support of this model, especially at high speed conditions where friction losses become very important in terms of energy efficiency. Oil
bleeding is recognised, regardless of the validity of this theory, as one of the most important feed mechanisms in the lubrication of the rolling elements.

While the ‘sponge’ model implies a spontaneous leak of oil, the ‘bled oil’ model\(^7\) suggests that oil bleeding is more likely to be due to a breakdown of the thickener structure in the grease present at the sides of the contacts. In fact, experience suggests that the separation of oil and thickener would be difficult without degrading the grease structure\(^6\), and would only occur at very high temperatures. Moreover, due to strong indications suggesting that the thickener does participate in the lubrication process\(^1\), this bled oil is likely to carry thickener particles while it flows to replenish the contact\(^7\). Therefore part of the grease which actively participates to the lubrication of the bearing would be transformed into a suspension of thickener particles in oil. This bled oil would feed the contact in a classic hydrodynamic fashion, but generally with a different viscosity from the original base oil. Hence, the presence of thickener would affect film formation and the lubrication regime in which the bearing is operating. In view of that, grease friction could be predicted from the bled oil properties.

Another theory, the ‘viscous layer’ model, suggests that lubrication is provided by a thin layer of highly degraded grease, deposited on the tracks after the first minutes of operation\(^1\). Being enriched with broken thickener particles, this lubricant layer is described as an oily, viscous material, with a residual yield stress which prevents it from leaking. The remaining grease in the bearing would only act as a sealant, without participating in feeding the rolling track by oil bleeding. According to this model, which strongly competes against the ‘sponge’ model, the thickener has a primary role. Firstly, the thickener would be a determinant factor for the grease tackiness (i.e. cohesiveness and adhesiveness) and mechanical stability. The former controls the retention of a thick, viscous layer, and the latter the preservation of this layer with time. Secondly, the thickener would directly affect friction, being part of the active lubricant. At the same time, the base oil would still give a large contribution, as the viscous layer would contain a large proportion of it.

The ‘hybrid’ model proposes the joint existence of the ‘bled oil’ and ‘viscous layer’ models\(^2\). The idea behind this theory is that that the lubricating films are formed by the co-operation of various contributions, corresponding to different dynamics. Part of the total film thickness would be generated by a hydrodynamic component, the bled oil, formed by base oil thickened with thickener particles\(^5\). The second contribution would be of a boundary type, given by a static, speed-independent, physically deposited thickener-rich layer\(^4\). The existence of this component has been confirmed by SEM inspection\(^1\) and FTIR analysis\(^1\) of bearing surfaces after use. The grease lubrication mechanism, and the trend in film thickness versus time
according to this model, are illustrated in Figure 2.27 (a) and Figure 2.27 (b) respectively. Initial overrolling of grease would entrain large thickener structures, part of which are physically degraded and deposited on the rolling track, forming a high viscous layer, as observed at the outlet of the contact. Part of this grease is displaced to the side of the contact (Figure 2.27 (b), time 1). This bulk inlet flow would eventually end, and the deposited thickener would be further degraded, until forming a solid, residual layer. The physical degradation of this layer would release oily, degraded grease, which is relocated to the sides of the contact, and that subsequently feeds the contact intermittently by continuous squeeze flow and capillary action, determining a flow balance (Figure 2.27 (b), time 2). The overrolling action at the contact sides would further destroy the thickener structure, diminishing the grease viscosity and hence improving its mobility towards the inlet of the contact. Consequently, the film thickness would increase with time (Figure 2.27 (b), time 3). Therefore, depending on the operating conditions, whether they are starved or fully flooded, and on the rheological state of the grease, one contribution (the static layer or the bled oil) would be higher than the other. Fully flooded conditions usually exist during the initial churning phase of the bearing. Afterwards, the deposited layer would be partly removed with time due to continuous rubbing. However, bulk flow is known to be stimulated by high temperatures and low speed, side slip, vibrations and the action of the cage, which are all factors that contribute to regenerate the formation of the layer.

Figure 2.27 (a) Grease lubrication mechanism according to ‘hybrid model’ and (b) evolution of film thickness with overrolling time

Clearly, the understanding of the mechanisms of grease lubrication in rolling bearings is still in a muddled state. Cann et al. used FTIR to analyse the composition of lithium grease after an extended time of operation in a bearing. It was found that the primary lubricant is ‘free’ oil released from the grease by the action of the cage. However, discrete amounts of grease containing an increased thickener content (compared to the amount present in the bulk grease)
were also found, suggesting an alternative mechanism of lubricant supply. In another study, Cann et al.\textsuperscript{90} found evidence of the participation of lithium thickener throughout the whole life of operation in a bearing, i.e. up to its failure. The composition of the lubricant found on the inner raceway was observed and analysed at different times of operation during a simulation test. In the lithium grease containing an additive package, both the thickener and base oil were found in the rolling track, deposited as a thin, continuous layer. This was mechanically and thermally degraded with time, forming a mobile, viscous layer. Both the thickener and the base oil were found oxidised at failure, and a higher thickener concentration was detected, indicating a loss of base oil. However, the presence of thickener was reduced when the same test was performed at high speeds, suggesting that most of the thickener was ejected from the rolling tracks, or rapidly shear degraded. The existence of different lubrication mechanisms depending on bearing type was indicated in both studies\textsuperscript{90,116}. These results suggest that the theories of grease lubrication mechanism presented above might all be correct. However, each case should be considered individually by taking into account the influence of grease composition, operating conditions and bearing design. Given the application (bearing type and operating conditions), the composition and properties of the lubricating grease film determining EHL friction are likely to depend on the bulk grease formulation: the base oil is certainly an important component, but the verified presence of thickener in the track suggests that this should be taken into account in the design of a low friction bearing grease.

2.7 Discussion and conclusions

This chapter provided the background to the current study. Lubricating greases, their composition and rheological properties were described and discussed in terms of application in rolling bearings. It was shown that the lubrication theory that applies to the type of concentrated contacts existing between the rolling elements and the raceways of rolling bearings, known as EHL, is relatively well established for oil lubrication. In this case the mechanisms of film formation are better understood, and film thickness can be predicted with sufficient accuracy, although the explanation and prediction for the friction force arising from shearing this film still remains the subject of an unresolved debate. However, in oil lubrication the relationship between friction and film thickness, relative to the surface characteristics of roughness, is well known and described by the characteristic ‘Stribeck curve’. On the other hand, the review on grease film formation has shown that, in general, many adjustments to the EHL theory need to be made when grease is used as the lubricant. As compared to oils, grease shows a complex behaviour that depends on a much more extended number of variables. Furthermore, the degree to which
these properties contribute to film formation is strongly affected by the operating conditions, particularly temperature and speed\textsuperscript{65,71}. Depending on these, the thickener interferes with film formation according to rules that escape classical EHL theory for oils. As a result, different regimes of lubrication should be identified\textsuperscript{72}. These might be ruled by the base oil\textsuperscript{71}, thickener\textsuperscript{68}, or most commonly their combined effect\textsuperscript{69,70}. This picture is further complicated by the effect of thermal\textsuperscript{74} and mechanical\textsuperscript{67} degradation of grease, commonly occurring in the majority of applications and dramatically affecting its structure and rheological behaviour. It is suggested that the extent to which the single grease components participate to the formation of the EHL film is governed by:

- Hydrodynamic rules, when the active lubricant is either oil\textsuperscript{71,73}, or a Newtonian suspension of thickener particles in oil\textsuperscript{55,63,62}. Several studies report that the thickener particles have a thickening effect on the base oil, and therefore the higher is the thickener concentration, the higher is the film thickness. However, the participation of the thickener to the overall increase in base oil viscosity is highly different depending on thickener type\textsuperscript{63,64,65,69,70}, dimensional features\textsuperscript{75} and base oil viscosity\textsuperscript{62};
- The non-Newtonian rheology of grease, when shear-thinning and yield stress characteristics are considered to be relevant to the formation of the film. As the film thickness is determined in the contact inlet, it is suggested that the higher is the grease shear stability\textsuperscript{59} and yield stress\textsuperscript{65}, the higher is the capability of the grease to resist inlet shear and form a thicker film;
- Structural and chemical features of the thickener\textsuperscript{67}, particularly at low rolling speeds when the lubrication conditions are fully flooded\textsuperscript{74}. In these conditions the film formation has been attributed to various mechanisms, including adsorption of boundary layers\textsuperscript{20}, or entrainment by hydrodynamic action\textsuperscript{81}.

Starvation is seen to occur far more often with grease than with oil, which contributes to complicate the problem. Several studies comparing the oil and grease film forming behaviour without a forced lubricant supply have shown that grease films are often thinner than those with oil\textsuperscript{19,79}, as opposed to what is often observed in fully flooded conditions. The mechanisms that rule film formation in starved conditions depend on additional variables, and are often competing. For example, it was shown that the base oil viscosity must be adequately high to generate thick films, but also low enough to preserve them\textsuperscript{19}. Another discrepancy is found in regard to the effect of thickener type and structure, which also greatly affect lubricant supply. On
one hand, coarse thickener structures seem to support the formation and retention of thick films in starved conditions\textsuperscript{19}. In agreement, the reduction of thickener particles size due to mechanical degradation diminishes the film thickness. On the other hand, increased oil bleeding as a consequence of partial destruction of thickener improves the grease ability to re-supply the contact and form thicker films\textsuperscript{20,34,86}. An additional conflicting aspect is thermal aging. Depending on its extent, this can improve film thickness due to the increased effective viscosity of the bled oil\textsuperscript{76,85}, or restrict its flow back to the contact inlet, thus reducing film thickness\textsuperscript{85}. Concerning the low-shear rheology of greases, lower elasticity and lower shear stability promote the formation of thicker films due to better replenishment properties, which is the exact opposite of what is suggested for fully flooded conditions. A lower elasticity of grease may facilitate its deformation and the release of free lubricant, increasing the grease capability to form thick films\textsuperscript{87}. Similarly, in greases with lower shear stability the yield stress would reduce faster with mechanical degradation\textsuperscript{86}. This is desirable, as a strong yield behaviour may restrict the back flow of grease, reducing the film thickness and leading to severe starvation and increased friction\textsuperscript{100}, and ultimately failure. However, at the same time the yield stress should also remain sufficiently high, as a rapid loss of consistency makes the grease more prone to leak, reducing the availability of lubricant for potential replenishment.

The relationship between friction and film thickness in grease lubrication is even more unclear. As the scientific literature reports, a major difficulty in interpreting grease frictional behaviour is represented by the lack of understanding of grease film formation. A Stribeck curve for oil describes well the link between friction and film thickness. With moderate sliding, the point of minimum friction can be directly related to a condition of ‘minimal’ lubrication. In correspondence to this value, the film thickness is just sufficient to ensure full separation of the contact surfaces, without causing unnecessary extra hydrodynamic friction generated by an excessive amount of lubricant. Below and above this value, friction is theoretically higher, due to increased asperity contact and viscous shear respectively. In grease lubrication the relationship between friction and film thickness is more intricate for a number of reasons. A significant complication is due to the fact that, as previously reviewed, grease film thickness is generally not known. In addition, nominal lambda ratio is not, on its own, a suitable parameter to describe the relationship between film thickness and friction in the shape of a Stribeck curve. Indeed, the lubricating film composition is often not exactly known, and, as such, neither are its friction properties. While a few studies have concluded that the only relevant grease component in determining friction is base oil\textsuperscript{101,102}, the majority have claimed the importance of both base oil and thickener. As EHL friction is strongly dependant on the molecular structure of the
lubricating film, and the main grease component is the base oil, the base oil type may intuitively be expected to exert a high influence. Greases formulated with PAO base oils seem to provide the lowest friction\textsuperscript{101,102,104,118}, followed, in order, by ester\textsuperscript{104}, mineral paraffinic\textsuperscript{118} and naphtenic\textsuperscript{103,102,118} base oils. However, the thickener has been shown to affect friction in a number of ways. Because of the adsorption properties of certain types of polar thickeners\textsuperscript{93,94}, these are able to adhere and form a thin residual ‘boundary’ layer\textsuperscript{20} on the contacting surfaces, particularly at low speeds and high temperatures\textsuperscript{93}. As a lubricant itself, the thickener is characterised by its own shear strength, resulting in lower friction when some types of thickeners are used in the grease formulation\textsuperscript{119}. Moreover, it has been found that the viscosity and pressure-viscosity properties of the active fluid that lubricates the contact, whichever it might be, are different in base oils and greases\textsuperscript{70}. In this sense, the thickener has an implicit influence on friction, by affecting film formation. The thickener type\textsuperscript{4,103} and concentration\textsuperscript{92,97} affect both friction and film formation in EHL contacts. In practical applications, thickener type and concentration also influence the onset of starvation\textsuperscript{103}. In starved conditions the low-pressure rheology of greases, particularly yield stress\textsuperscript{100} and shear-thinning behaviour\textsuperscript{105,107}, is of primary importance in determining lubricant supply and friction. These properties depend on both base oil, thickener type and concentration, and also the manufacturing protocol\textsuperscript{120}, all contributing to grease efficiency. The strong time-dependent behaviour of greases further contributes to the complexity of the topic, generating more chaos. In particular, the reduction of friction which is often observed with increasing time has been explained either as the consequence of a decrease in film thickness and thus viscous friction due to starvation\textsuperscript{105}, the changing rheology during use\textsuperscript{96}, or the physical deposition of a thickener-rich film\textsuperscript{92,121,4} forming a boundary layer.

Despite the significant amount of research work that has been dedicated to study grease lubrication, there seem to be no universal agreement on the theory. Instead, the complexity of the topic has led to many ‘dark areas’ in the understanding of the mechanisms themselves. The apparent inconsistencies between the bearing lubrication theories presented in Section 2.6, which have been inspired by the studies revised in this chapter, can possibly be explained as the result of two main reasons:

- The studies have been carried out on very different test rigs, and in very different operating conditions, so that it is difficult to compare across them. Moreover, much of the research work has covered a relatively restricted range of conditions. In view of the fact that the contact conditions strongly affect the lubrication regime in which grease is
operating, this limits the generality of the findings and therefore the validity of each theory;

- The great variety of grease formulations used in these experimental studies further complicates the comparison and interpretation of the results. Due to the fact that the lubrication mechanisms depend on a large number of variables in the grease composition, which in turn affect the grease rheological properties, the comparison of results obtained from such different greases can be misleading. The studies have rarely been methodical in terms of carefully isolating each variable so that its influence on the grease tribological behaviour could be individually studied.

Evidently, the literature is missing a systematic work aimed at clarifying the influence of the grease single components, and their mutual interaction, on its tribological behaviour. This is especially true in view of the effect of grease composition on friction, since this specific topic has only been barely explored. The main objective of the current research work is to fill this gap by:

- Exploring the EHL behaviour of commercially available greases, used to create a ‘map’ for orienting the choice of grease formulation in function of their friction response;
- Investigating specific ‘directions’ inside this map, i.e. the effect of oil viscosity, oil type and the presence of friction modifiers. The formulation and manufacturing of custom greases was necessary for this specific purpose.

The research approach is conceptually summarised in the diagram shown in Figure 2.28. A series of commercial greases were first chosen and subjected to several tribological measurements, mostly focussed on friction. The aim was to evaluate the overall frictional performance of different grease formulations, and possibly identify potential combinations of thickener and base oils giving low and high friction. This investigation also served to get a reference line in terms of current compositions for low-friction greases. Moreover, it provided guidelines for designing and manufacturing custom-made greases. These were also subjected to a tribological study, closing the diagram loop presented in Figure 2.28. The purpose of using this procedure was to improve the knowledge on greases frictional behaviour to an increasingly detailed level, by iterating the research work. The final aim is to be able to answer the question: “what makes grease a ‘low friction’ grease?” As the answer is likely to depend on the specific application in which grease is employed, the tribological tests were designed to provide a range of different operating conditions.
Figure 2.28 Schematisation of ‘research approach’
Chapter 3. Test greases

As the subject of this thesis is grease formulation and its influence on EHL friction, this chapter is entirely dedicated to the description and reasoning behind the choice of the test greases. In the first section, the range of chosen commercial grease samples is introduced. In later sections, the selection of basic materials and their use in the manufacturing process of the custom greases is presented. The techniques used to characterise the greases, and manipulate them to obtain further lubricants for testing (e.g. oil bleeding, mechanical degradation) are also described.

3.1 Introduction

In order to reach the ultimate objective of enhancing the current understanding of frictional behaviour of greases, the current study considered two sets of lubricants. The first group is a series of commercial bearing greases, which were studied to develop a knowledge base for the influence of grease formulation on friction. This was necessary to develop a preliminary general understanding on how EHL friction is affected by different formulations of thickener and base oil, and provide a reference line in terms of the performance of current low-friction greases. The guidelines obtained in this manner, based on the results later presented in Chapter 5, were then used to formulate and manufacture a set of custom greases. With the tightly controlled composition and manufacturing process of these custom greases, a specific investigation into particular aspects of interest could be carried out. These include the effect of base oil type, base oil viscosity, and presence of additives, which could thus be analysed in the subsequent tribological study presented in Chapter 6.

3.2 Commercial greases

The selection of commercially available bearing greases tested in this work is introduced in this section. An in-depth description of the greases, including their composition and common applications, is given. Further information on the greases was obtained from the available data, or by employing a series of experimental techniques. The original greases were converted into simpler components, i.e. bled oils and thickener, which were then tested separately as additional lubricant samples. Finally, other test samples were obtained by mechanically degrading a selection of the original greases.
3.2.1 Reasoning for the selection of commercial greases

A number of commercially available greases were chosen for this experimental study, on the basis of three main requirements:

- To cover a broad range of bearing applications and operating conditions;
- To include a wide range of base oil/thickener combinations;
- To include some current state-of-the-art low friction bearing greases.

<table>
<thead>
<tr>
<th>Grease Name</th>
<th>Oil</th>
<th>Thickener</th>
<th>Oil Viscosity @ 40°C [cSt]</th>
<th>Oil Viscosity @ 100°C [cSt]</th>
<th>NLGI</th>
<th>Application</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiMi</td>
<td>Mineral</td>
<td>Lithium</td>
<td>74</td>
<td>8</td>
<td>3</td>
<td>Reference Grease</td>
</tr>
<tr>
<td>LiMi2</td>
<td>Mineral</td>
<td>Lithium</td>
<td>200</td>
<td>16</td>
<td>2</td>
<td>High load extreme pressure</td>
</tr>
<tr>
<td>LiEs</td>
<td>Ester</td>
<td>Lithium</td>
<td>25</td>
<td>5</td>
<td>2</td>
<td>Low friction</td>
</tr>
<tr>
<td>LiPAO</td>
<td>PAO</td>
<td>Lithium complex</td>
<td>191</td>
<td>22</td>
<td>2-3</td>
<td>Wide temp range</td>
</tr>
<tr>
<td>LiCPAO</td>
<td>PAO</td>
<td>Lithium complex</td>
<td>191</td>
<td>22</td>
<td>2-3</td>
<td>Wide temp range</td>
</tr>
<tr>
<td>PoEs</td>
<td>Ester</td>
<td>Polyurea</td>
<td>70</td>
<td>9</td>
<td>2-3</td>
<td>High temp</td>
</tr>
<tr>
<td>CaSMi</td>
<td>Mineral</td>
<td>Calcium sulphonate complex</td>
<td>420</td>
<td>26</td>
<td>2</td>
<td>Heavy duty high temp</td>
</tr>
<tr>
<td>CaSMi</td>
<td>Mixed mineral/synthetic</td>
<td>Calcium sulphonate complex</td>
<td>80</td>
<td>8.6</td>
<td>1-2</td>
<td>Heavy duty low temp</td>
</tr>
</tbody>
</table>

Table 3.1 lists the commercial greases properties and typical applications. These data were mostly obtained from public sources. Some additional information, particularly on application, was obtained from the industrial sponsor, a bearing company and hence the end user of these greases. Instead of using their commercial names, the greases are identified using a naming nomenclature, shown in the first column of this table. The nomenclature reflects the grease
composition; for example, ‘LiMi’ indicates a lithium-thickened grease based on a mineral oil. Base oils range from mineral to PAO, ester and mixed mineral/synthetic oils. They are characterised by very different viscosities, ranging from a minimum of 18 cSt to a maximum of 420 cSt at 40 °C. The thickeners include lithium, lithium complex, polyurea and calcium sulphonate complex. Four greases are based on lithium 12-hydroxystearate, and one is a lithium complex thickened grease. This is broadly representative of the global market of lubricating greases, 70% of which are currently based on lithium soaps\textsuperscript{122}. Two of the selected greases, LiEs and LiPAO, are categorised as ‘low friction’ greases.

3.2.2 Description of base oils, thickeners and practical uses of the selected greases

Little is known about the selected commercial greases; the only information available is shown in Table 3.1. A brief, general description of their thickeners and oils, together with an attempt to interpret the grease formulations as a function of their application, are given in this section. Further data, obtained from simple calculations or measurements, are also provided.

3.2.2.1 Thickeners

The majority of the test greases (LiMi, LiMi2, LiEs, LiPAO) are thickened with lithium soaps, according to the patent originally issued by Clarence E. Earle\textsuperscript{123}. This is obtained from the saponification reaction of a fatty acid, usually 12- hydroxystearic acid\textsuperscript{12}, using lithium hydroxide as the base. Lithium greases are labelled as ‘multipurpose’, due to their good general performance and cost effectiveness\textsuperscript{17}, making them versatile for use in a large number of applications. Their properties include excellent mechanical stability, good water resistance and reasonably good high temperature performance, up to 120 °C\textsuperscript{124}. LiCPAO is thickened with lithium complex. Complex soaps are generally thickeners obtained from the saponification reaction between a single metal with a combination of different types of acids. The most commonly used complexing agents in lithium complex greases are dimer acids such as azelaic, adipic or sebacic acids. The main advantage of complex greases over simple soap greases is their ability to withstand high operating temperatures, of up to 240 °C. Lithium complex grease is regarded as the new generation, multipurpose grease with a wide temperature range\textsuperscript{12}.

PoEs is made with polyurea, which is normally a reaction product between different types of isocyanates and amines. Polyurea greases exhibit extremely good high-temperature performance, oxidation and water resistance, making them suitable for filled-for-life applications.
CaSMi and CaSMix are calcium sulphonate complex greases. The main components from which this thickener is obtained are overbased calcium sulphonate, and calcium 12-hydroxystearate as the complexing agent. The leading characteristics of these greases are their inherent extreme-pressure properties, excellent mechanical stability and water resistance. They also have high dropping points and can thus be used at high temperatures.

3.2.2.2 Base oils

Oil manufacturers are often reluctant to disclose more precise information on the origin and composition of the oils they sell. Mineral oils are generally made of mixtures of different hydrocarbons, containing thousands of distinct chemical compounds. As a result the precise base oil chemistry is often unknown. Furthermore, it varies from batch to batch. The grease suppliers do not specify what kind of esters are used in the formulation of LiEs and PoEs. PAO oils are probably the best known oil type among the base oils present in the selection of commercial greases. Despite these uncertainties, some further information can be extracted from knowing the base oil type and viscosity values at 40 °C and 100 °C. This data was used to estimate the oils pressure-viscosity-temperature behaviour, by calculating their pressure-viscosity coefficient $\alpha$ and viscosity index $VI$. The $\alpha$ values were calculated by following a procedure described by Khonsari and Booser\(^5\). In this calculation the density was assumed to be 850 kg m\(^{-3}\) for all base oils, since no exact figures were available. The $VI$ values were calculated using the ASTM D2270 standard. These estimated values are all shown in Table 3.2.

The pressure-viscosity values were later used to approximate the base oils EHL film thickness by employing the Hamrock-Dowson EHL film thickness equation\(^38\).

<table>
<thead>
<tr>
<th>Grease name</th>
<th>LiMi</th>
<th>LiMi2</th>
<th>LiEs</th>
<th>LiPAO</th>
<th>LiCPAO</th>
<th>PoEs</th>
<th>CaSMi</th>
<th>CaSMix</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$ [GPa(^{-1})]</td>
<td>26</td>
<td>22</td>
<td>16</td>
<td>13</td>
<td>17</td>
<td>20</td>
<td>25</td>
<td>24</td>
</tr>
<tr>
<td>$VI$</td>
<td>81</td>
<td>79</td>
<td>121</td>
<td>175</td>
<td>139</td>
<td>114</td>
<td>85</td>
<td>72</td>
</tr>
</tbody>
</table>

Table 3.2 Estimated pressure-viscosity coefficients $\alpha$ and viscosity index $VI$ for the base oils of the commercial greases

LiMi, LiMi2, and CaSMi are made with mineral oils. CaSMix base oil is a mixed mineral/synthetic oil, which usually implies the presence of a small fraction of synthetic oil in a mineral oil, although the exact proportions present here are not known. The American
Petroleum Institute (API) categorises mineral oils into 3 groups according to the proportion of saturated and unsaturated compounds, the presence of sulphur and the viscosity-index value$^{12}$:

- **Group I** as the base stocks containing $< 90\%$ saturates and/or $> 0.03\%$ sulphur, and with a viscosity index $VI: 80 \leq VI < 120$;
- **Group II** as the base stocks containing $\geq 90\%$ saturates and $\leq 0.03\%$ sulphur, and $80 \leq VI < 120$;
- **Group III** as the base stocks containing $\geq 90\%$ saturates and $\leq 0.03\%$ sulphur, and $VI \geq 120$.

Since all the mineral base oils in the present greases have estimated $VI$s that are apparently lower than 120 (an anomalous value of 72 is calculated in CaSMix), these are likely to be either group I or group II oils, corresponding to the most common mineral groups in the market.

LiEs and PoEs are made of ester oils. These are synthetic oils obtained from the reaction between an acid and an alcohol, and are classified as group V according to API. Ester oils can be tailored to provide physical or chemical properties that cannot be achieved using conventional mineral oil products. Therefore they are often custom-designed for the specific application. For this reason, thousands of different ester oils exist, but unfortunately no extra information is available on the specific types used for LiEs and PoEs.

LiPAO and LiCPAO base oils are polyalphaolefins (PAO), classified as API group IV. These are hydrogenated oligomers of a $\alpha$-olefin, usually 1-decene$^{12}$. They are more popular than ester oils, not least because of their lower costs. Their molecular structure is very similar to petroleum isoparaffins. PAOs usually have better viscosity-temperature features than mineral oils, as is evident in the higher estimated VI values for these oils reported in Table 3.2.

3.2.2.3 Practical applications of selected commercial greases

All the chosen greases are typically employed in the lubrication of rolling bearings. In Table 3.1, LiMi is branded as the ‘reference grease’, due to a composition which is suitable for a wide range of applications. Indeed, LiMi is made of a cost-effective mineral oil of medium viscosity, and a multipurpose lithium thickener.

LiMi2 is employed in high load, extreme pressure applications. The manufacturers mention the presence of extreme pressure additives.
LiEs and LiPAO are the two ‘low friction’ greases, and are both made of low viscosity oils, as expected in bearing applications where energy efficiency is required\textsuperscript{26}. LiCPAO is employed in wide temperature range applications. This could explain the use of lithium complex, able to withstand high operating temperatures, and a high viscosity, high VI base oil.

PoEs is made with polyurea, which is a common thickener type in greases employed for high temperature applications.

CaSMi and CaSMix are used in heavy-duty applications. Calcium sulphonate complex thickeners are known to be characterised by inherent extreme-pressure characteristics. CaSMi is used at high temperatures, and therefore is made of a very high viscosity base oil. CaSMix base oil has a much lower viscosity, as it is more suitable for low temperature applications.

### 3.2.3 Determination of thickener content

One of the bits of missing information for the commercial greases is their thickener content. As the literature review in Chapter 2 suggests, the amount of thickener affects both the grease film thickness and friction behaviour.

In order to address this uncertainty, a common method to determine the thickener/oil ratio was applied. For each grease, a small sample was dissolved in petroleum ether in an ultrasonic bath. The obtained suspension was then pushed through a filter with a mesh size of 0.45 μm at the end of a syringe. The mesh of the filter is supposed to be small enough to allow for the solution of oil and solvent to pass through the filter, while retaining most of the thickener. The filter was dried in an oven at 40 °C for two days to let the solvent evaporate, while the solution of oil and solvent was distilled in a rotary evaporator to separate the solvent from the oil. The thickener weight was obtained by measuring the weight increase of the filter. The oil weight after solvent distillation was also measured. The measured absolute difference between the sum of thickener and oil weights and the initial weight of the grease sample was lower than 10 %. The missing information on thickener concentration was achieved by comparing the thickener weight with the initial grease sample weight. The obtained results are shown in Table 3.3. These measurements are affected by large uncertainties, not least due to the presence of additives in the commercial greases. These could have passed with the oil through the mesh\textsuperscript{76,75}, as well as filtered together with the thickener. In theory, based on the assumption that the thickener was entirely filtered during the process, which might not be true depending on thickener type\textsuperscript{70}, these measurements are susceptible to errors of the order of at least ±5 % (taking 5 % as an indicative
concentration of additives in the formulation of grease). Therefore these values are only approximations.

<table>
<thead>
<tr>
<th>Grease name</th>
<th>Thickener w/w %</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiMi</td>
<td>14</td>
</tr>
<tr>
<td>LiMi2</td>
<td>13</td>
</tr>
<tr>
<td>LiEs</td>
<td>11</td>
</tr>
<tr>
<td>LiPAO</td>
<td>13</td>
</tr>
<tr>
<td>LiCPAO</td>
<td>22</td>
</tr>
<tr>
<td>PoEs</td>
<td>11</td>
</tr>
<tr>
<td>CaSMi</td>
<td>23</td>
</tr>
<tr>
<td>CaSMix</td>
<td>24</td>
</tr>
</tbody>
</table>

### Table 3.3 Measured thickener concentration in commercial greases

3.2.4 Bled oils

A top-down approach was used for the commercial grease samples, meaning that the initial greases were disassembled into their simpler components, i.e. bled oil and thickener. In this way it was possible to test the single grease components separately, so as to disentangle their individual influence on friction.

A simple centrifuge was used to extract the bled oil from a filter cylinder (≈ 100 mm in diameter) filled with grease. This is shown in Figure 3.1.

![Figure 3.1](image-url) (a) Picture of centrifuge used to extract bled oil from commercial greases and (b) a schematic of bled oil extraction process

The temperature and rotational speed of the cylinder were adjusted, for each grease, to collect enough oil to carry out the tribology tests. The composition of these greases was determinant in
their ability to bleed oil, and the flow rate of the bled oil through the centrifuge filter was generally dependent on thickener type, base oil type and viscosity. An indicative ranking of the oil bleeding capability of each grease is given in Table 3.4, where:

- ++ stands for high bled oil volume: the bled oil of LiMi, LiMi2 and LiPAO was easily extracted, and a substantial amount of it was collected from each grease sample;
- + means that the extraction of oil was achieved with more difficulty; higher temperature and speeds were used for LiEs, LiCPAO and PoEs to bleed enough oil;
- 0 means that the extraction of bled oil through the centrifuge filter was impossible. CaSMi and CaSMix bled oils did separate from the thickener structure, but remained inside the centrifuge, as if a barrier of thickener was produced on the centrifuge filter, preventing the escape of oil. In order to obtain their bled oils, the original grease was dissolved with petroleum ether, and filtered with a fine mesh sieve to separate the solution of oil and solvent from the thickener. Finally, the solvent was distilled off through a rotary evaporator.

The bled oil kinematic viscosity was measured at 40 °C in an Anton Paar Stabinger SVM 3001 viscometer. The obtained values are shown in Table 3.4, together with the base oil viscosity for comparison. The majority of bled oils show viscosity values that are slightly higher than in the base oils. The maximum positive deviation was observed in LiCPAO bled oil, with a value of viscosity ≈ 24 % higher than its base oil. A negative deviation was observed in LiMi2 bled oil, with a viscosity ≈ 5 % lower than its base oil. These differences might be attributed to the presence of thickener or additives in the bled oils, or to an altered balance (in the bled oil) between the different chemical species originally present in the base oil.
Table 3.4 Comparison between base and bled oil kinematic viscosity in commercial greases

<table>
<thead>
<tr>
<th>Grease name</th>
<th>Oil bleeding ranking</th>
<th>Base oil viscosity @ 40 °C [cSt]</th>
<th>Bled oil viscosity @ 40 °C [cSt]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiMi</td>
<td>++</td>
<td>74</td>
<td>74</td>
</tr>
<tr>
<td>LiMi2</td>
<td>++</td>
<td>200</td>
<td>189</td>
</tr>
<tr>
<td>LiEs</td>
<td>+</td>
<td>25</td>
<td>28</td>
</tr>
<tr>
<td>LiPAO</td>
<td>++</td>
<td>18</td>
<td>18</td>
</tr>
<tr>
<td>LiCPAO</td>
<td>+</td>
<td>191</td>
<td>236</td>
</tr>
<tr>
<td>PoEs</td>
<td>+</td>
<td>70</td>
<td>76</td>
</tr>
<tr>
<td>CaSMi</td>
<td>0</td>
<td>420</td>
<td>434</td>
</tr>
<tr>
<td>CaSMix</td>
<td>0</td>
<td>80</td>
<td>87</td>
</tr>
</tbody>
</table>

The remaining LiEs and LiPAO grease left after the bled oil extraction process was collected from the centrifuge pot and used as a lubricant for some of the friction tests. From a comparison between the initial grease weight and the weight of the collected bled oil, the thickener content of these ‘de-oiled’ greases corresponds to circa 24 % for LiEs and 27 % for LiPAO.

### 3.2.5 Mechanically degraded greases

During extended time of operation in a bearing, grease is subjected to a severe mechanical degradation process, which is likely to change its film forming and friction properties\(^{16}\). These changes cannot be accounted for by testing fresh greases in laboratory devices like the ones employed in this study, which can only produce a mild, local form of degradation. Therefore, in order to simulate the grease degradation process occurring in a bearing, some of the greases were subjected to an intense process of mechanical shear in a different apparatus. The mechanically degraded greases were eventually used as additional samples for the friction analysis.

The Shell roll stability tester (ASTM D1831), shown in Figure 3.2, was used for this purpose. The test consists in rotating, for a definite time and at a specific temperature, a steel cylinder containing a 5 kg steel roller and a certain amount of grease. The roller is 60.2 mm in diameter and 176.2 mm in height. The grease, interposed between the roller and the cylinder during rotation, is thus subjected to a long, heavy mechanical degradation cycle. The standard aim of the test is to measure the grease variation in consistency (the grease ‘stiffness’, see Section 3.3.3) before and after the test. This gives an indication of the grease mechanical stability\(^1\), i.e. its
capacity to retain its consistency after an extended period of mechanical stress. However, in this study the test was used to prepare mechanically degraded samples for the following friction testing analysis. Only a selection of the greases, namely LiMi, LiEs, LiPAO and CaSMix, were studied. A sample of 50 grams for each grease was mechanically worked for 50 hours, at a temperature of 80 °C, by rotating the cylinder at a speed of 200 rpm.

Figure 3.2 Pictures of (a) Shell roll stability tester and (b) detail of roller and cylinder

3.3 Custom greases

As presented in Section 3.2, many variables differentiate the composition of the commercial greases. It will be shown in Chapter 5 that the experimental results obtained with these greases provided some basic guidelines as to the influence of grease composition on the frictional behaviour of greases. However, given the much variability in these ready-made commercial grease compositions, it was difficult to focus the study on the influence of any specific aspects of grease formulation on friction. To achieve this, the second group of lubricants used for the tribological investigation consists of a series of custom greases with controlled composition, which were formulated so as to 'isolate' specific properties in the greases formulation. These were manufactured using PAO, mineral and ester oils, and were all thickened with 12 % w/w lithium 12-hydroxystearate. Some of them were also additised with oleic acid after processing. The individual components of these greases, the manufacturing process and their consequent characterisation are here presented.
3.3.1 Base oils

The particular focus of the second part of the experimental work was the effect of base oil viscosity and type on friction. These two variables were isolated by testing model greases made with the same type of oil but different viscosities, or with different oil types of similar viscosity. By choosing the same type and amount of thickener to fabricate the greases, the number of variables of influence in the greases formulation was reduced to the minimum, i.e. the only ones of interest.

Due to the nature of EHL friction, finding trends in the effect of base oil viscosity on friction can only be attempted if greases made of similar base oil type are compared\textsuperscript{120}. PAO oil was chosen as the most suitable oil type for this investigation, based on the performance of PAO-based commercial greases, as shown later in Chapter 5. In addition, PAO oils have a definite, homogeneous and consistent composition, with consequent improved repeatability of the results and validity in their comparisons. Indeed different viscosity PAO oils have very similar molecular compositions, which mostly vary in chain length from one another. The higher is the viscosity of the PAO oil, the longer are its molecules. This means that, by choosing PAO oils for this investigation, it was possible to restrict the study to oils with the ‘same’ chemical composition.

PAO-based oils with five different viscosities were prepared to fabricate lithium-PAO base oil greases, identified as the ‘LP’ greases. Their oils were chosen and prepared so that their viscosities (ν) could be spaced in the following way:

\[ \nu_{LP5} \approx 2 \nu_{LP4}; \nu_{LP4} \approx 2 \nu_{LP3}; \nu_{LP3} \approx 2 \nu_{LP2}; \nu_{LP2} \approx 2 \nu_{LP1} \]

LP1, LP2 and LP5 are made from pure base oils. The base oils of LP3 and LP4 were obtained by mixing appropriate amounts (estimated with the Refutas equation\textsuperscript{127}) of LP2 and LP5 base oils (64 % and 36 %, and 28 % and 72 % respectively).

The investigation on the effect of oil type on friction was carried out by fabricating three more model greases. LM1 and LM2 are mineral oil-based greases, and LPE is a mixed PAO/ester oil-based grease. LM1 and LM2 base oils are group II mineral oils with different viscosities. LPE base oil is made of 20 w/w % dibasic ester and 80 w/w % PAO (the same PAO as used for LP1). Therefore, this investigation was conducted by comparing the behaviour of LP1 and LPE greases with that of LM1, and the behaviour of LP3 grease with LM2, as these comparison groups were formulated with base oils of different types, but similar viscosity. An additional investigation on the effect of base oil viscosity on friction within the mineral oil greases, as opposed to the PAO based greases, could be carried out by comparing LM1 and LM2 friction.
behaviour. However, in this case, in contrast with the PAO oils, the two mineral base oils might have very different molecular structures.

The possible comparative investigations with these greases, as a function of these base oil characteristics, are shown in Figure 3.3.

![Figure 3.3 Subgroups of greases as a function of the type of investigation (base oil viscosity and type)](image)

The greases base oils are listed in Table 3.5 with their main properties. The base oils viscosity in the table were measured using Anton Paar Stabinger SVM 3001 viscometer. The values obtained with the pure base oils are comparable to the ones stated by the manufacturers. The remaining properties are listed as reported on the base oils Technical Data Sheet provided by the manufacturers.

As the friction tests were carried out at 40 °C, 60 °C and 80 °C, the kinematic viscosity was also measured at these temperatures. These data are very close to the values of viscosity estimated using the ASTM D341-722 standard formula, also known as Walther’s equation:

$$\log_{10} (\nu_T + 0.7) = b - c \times \log (T)$$

where $\log$ is $\log_{10}$, $\nu_T$ is the kinematic viscosity at the temperature $T$ (in Kelvin), while $b$ and $c$ are two constants that can be found by knowing the viscosity at 40 °C and 100 °C. Therefore this equation was used to predict the base oil viscosity at temperatures other than 40 °C, 60 °C, 80 °C or 100 °C, where direct measurements were relied upon.
Table 3.5 List of base oils used for the fabrication of custom greases and relative properties

<table>
<thead>
<tr>
<th>Base oil</th>
<th>Type</th>
<th>Oil viscosity @ 40°C [cSt]</th>
<th>Oil viscosity @ 100°C [cSt]</th>
<th>Viscosity index</th>
<th>Density @ 15.6 °C [kg dm³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>LP1 BO</td>
<td>PAO</td>
<td>26</td>
<td>5</td>
<td>145</td>
<td>0.825</td>
</tr>
<tr>
<td>LP2 BO</td>
<td>PAO</td>
<td>47</td>
<td>8</td>
<td>136</td>
<td>0.831</td>
</tr>
<tr>
<td>LP3 BO</td>
<td>PAO</td>
<td>98</td>
<td>14</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>LP4 BO</td>
<td>PAO</td>
<td>211</td>
<td>28</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>LP5 BO</td>
<td>PAO</td>
<td>413</td>
<td>50</td>
<td>186</td>
<td>0.846</td>
</tr>
<tr>
<td>LPE BO</td>
<td>Mixed PAO/ester</td>
<td>26</td>
<td>5</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>LM1 BO</td>
<td>Mineral</td>
<td>35</td>
<td>5</td>
<td>102</td>
<td>0.872</td>
</tr>
<tr>
<td>LM2 BO</td>
<td>Mineral</td>
<td>102</td>
<td>11</td>
<td>96</td>
<td>0.886</td>
</tr>
</tbody>
</table>

3.3.2 Thickener

All the greases were fabricated using the same amount of nominally 12 % w/w lithium 12-hydroxystearate \((C_{18}H_{35}LiO_3)\) thickener. This was produced through the saponification reaction between 12-hydroxystearic acid \((C_{18}H_{36}O_3\), abbreviated to HSA) and a dispersion of micron-sized anhydrous lithium hydroxide \((LiOH)\) particles in mineral oil, conducted in the specific base oil chosen for the given grease batch.

3.3.3 Grease manufacturing

A grease kettle for small batches production, consisting of a reactor connected to a heating and cooling system, was used to fabricate the custom greases. This is shown in Figure 3.4 (a). Figure 3.4 (b) displays the mixing paddle used to stir the various ingredients used to manufacture the
greases. The approximate size of the reactor cylinder is 250 mm in diameter and 450 mm in height.

![Picture of grease kettle and mixing paddle](image)

Figure 3.4 Pictures of (a) grease kettle (with heating/cooling system and reactor) and (b) detail of mixing paddle

The same general recipe, including the actual quantity of ingredients, was used for all the greases. The aim was to keep constant manufacturing conditions, so as to maintain consistent results. About 4 kg of grease were obtained for each batch. Much effort was also put into controlling time, amount and insertion order of every single compound.

A common, well-known manufacturing process for simple lithium greases consisting of 4 stages (saponification, soap dissolution, re-crystallization and homogenization) was employed. A referenced manufacturing cycle\textsuperscript{129}, with some modifications coming from improvements following experimentation, was adopted for the fabrication of all greases. The first 3 stages of this process can be followed on the temperature cycle represented in Figure 3.5.
At first, the vessel was filled with half of the total oil used to produce 1 batch of grease (initial oil load). The vessel was locked, and the impeller switched on, so as to continually stir the mixture throughout the length of the process. When the oil reached a stable temperature of 80 °C, slightly above the HSA melting point, this was added to the oil. After 25 minutes, sufficient for melting all the HSA, LiOH was slowly poured inside the vessel through a threaded hole at the top of the vessel, initiating the saponification reaction. A spontaneous temperature and torque increase was observed here. When all the LiOH was poured in the vessel, the stirring speed was increased to facilitate a good distribution of the forming thickener phase, and the hole was closed to build pressure in the vessel and promote the saponification reaction. The temperature was increased following the gradient shown in Figure 3.5. The hole was then re-opened to allow for evaporation of water as the by-product of the saponification reaction. The temperature was further increased until the melting point of lithium 12-hydroxystearate was reached, just above 200 °C. The temperature was kept at this high value for 10 minutes to allow for all the thickener to melt. Subsequently the temperature was again reduced to start the re-solidification and crystallisation of the thickener. Specific temperature gradients were followed, as indicated in the figure. The remaining half of the oil was slowly poured inside the vessel, and its flow speed adjusted to match the nominal temperature ramp-down curve. During this stage the torque gradually increased, thus the rotational speed of the impeller was adjusted accordingly to keep it within a maximum, safety value. When the temperature reached values lower than 70 °C, the grease was moved to the roller mill for the last homogenization phase. Homogenization is a mechanical treatment, aimed at improving the uniformity of the grease composition, and at refining the thickener structure, thus enhancing its thickening effect. This was conducted in a
roller mill, depicted in Figure 3.6 (a). The approximate diameter of each roller is 150 mm. The grease was introduced in the position between rollers 1 and 2 (Figure 3.6 (b)), and forced to pass through gap A, and successively through gap B between rollers 2 and 3. The grease path is indicated by the black arrows in Figure 3.6 (b).

![Figure 3.6](image)

Figure 3.6 (a) Picture of roller mill used for homogenisation of custom greases and (b) schematisation of homogenisation procedure;

The homogenization phase consisted of three steps. The gap between the rollers was gradually reduced step after step, in order to further decrease the size of the thickener fibres and improve homogenisation. In step 1, the grease batch was worked once between the rollers, with nominal roller gaps $A = 150 \, \mu m$ and $B = 95 \, \mu m$. In step 2, the grease homogenized in step 1 was worked twice between the rollers, with nominal roller gaps $A = 150 \, \mu m$ and $B = 77 \, \mu m$. Finally, in step 3, the grease homogenized in step 2 was worked once between the rollers, with nominal roller gaps $A = 23 \, \mu m$ and $B = 5 \, \mu m$. The gap values for each homogenisation step are listed in Table 3.6.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>After manufacturing</td>
<td>Step 1</td>
<td>150</td>
<td>95</td>
</tr>
<tr>
<td>After step 1</td>
<td>Step 2 (x 2)</td>
<td>150</td>
<td>77</td>
</tr>
<tr>
<td>After step 2</td>
<td>Step 3</td>
<td>23</td>
<td>5</td>
</tr>
</tbody>
</table>

A preliminary analysis in the friction investigation presented in Chapter 5 revealed that the differences in friction between samples collected at the end of each homogenisation stage were negligible. Therefore, the greases chosen for the friction analysis are all samples collected after homogenization step 3, so that the samples were all post-processed using the same
homogenisation procedure. Nevertheless, some of the samples collected after homogenisation steps 1 and 2 were also characterised for completeness.

3.3.4 Grease characterisation

In order to gain basic information on the properties of the finished greases, some standard laboratory techniques were used to characterise them. Cone penetration, oil bleeding and SEM results are presented in this section.

3.3.4.1 Cone penetration test

A sample of grease was saved after every homogenisation step, and a penetration test (ISO 2137/ASTM D217) was performed on all the collected samples. The penetration test apparatus, and the schematic of its principles, are shown in Figure 3.7.

The preparation to the test consists, at first, in shearing a fixed amount of grease (equivalent to the volume of the standard cup size) with 60 double-strokes using a grease worker. The cup used for these measurements is a ‘half scale’ cup, with dimensions of 37.9 mm in diameter and 31.7 mm in height. The worked grease is then levelled with the top of the cup by means of a spatula. A standardized cone is placed vertically so that its tip barely contacts the top of the levelled grease, in correspondence to the central position of the cup. This cone is then allowed to fall into the grease, for 5 seconds and at 25 °C.

![Figure 3.7](image)

*Figure 3.7 (a) Cone penetrometer, (b) grease worker and cup, (c) schematic of penetration test;*

The penetration depth, indicating the stiffness of the grease, is measured in $10^1$ mm. The higher is the penetration depth, the softer is the grease. Greases are classified according to their consistency number, which is linked to the penetration depth though the NLGI scale as indicated in Table 3.7.
Table 3.7 NLGI classification for grease consistency

<table>
<thead>
<tr>
<th>NLGI</th>
<th>Penetration ([10^1 \text{ mm}])</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>445–475</td>
</tr>
<tr>
<td>00</td>
<td>400–430</td>
</tr>
<tr>
<td>000</td>
<td>355–385</td>
</tr>
<tr>
<td>1</td>
<td>310–340</td>
</tr>
<tr>
<td>2</td>
<td>265–295</td>
</tr>
<tr>
<td>3</td>
<td>220–250</td>
</tr>
<tr>
<td>4</td>
<td>175–205</td>
</tr>
<tr>
<td>5</td>
<td>130–160</td>
</tr>
<tr>
<td>6</td>
<td>85–115</td>
</tr>
</tbody>
</table>

The NLGI consistency number is a very popular type of specification for the designation of greases, and one of the most important in determining the grease suitability to a specific application. The majority of greases employed in the lubrication of rolling bearings have an NLGI number in between the range of 2 and 3. This value usually represents a good compromise between contrasting requirements, a grease that flows easily, but without leaking spontaneously, i.e. both fluid and stiff respectively.

Averaged penetration depths (3 measurements) are displayed, for each grease and homogenization step, in Figure 3.8. The same values were converted into NLGI consistency numbers as presented in Table 3.7. In the majority of the greases the penetration depth decreases, i.e. the consistency increases, after every homogenisation step. This is the result of decreasing the distance between the rollers in the homogenisation phase. The thickener fibres dimensions are progressively reduced, and their distribution in the base oil improved, thus refining and homogenising the microstructure. One exception is LPE, with a penetration depth that remains virtually constant. This indicates the presence of a ‘finished’ grease microstructure, i.e. a grease where the distribution and size of thickener fibres is already set and cannot be further modified.

Except for LP1, the PAO base oil greases display a clear trend of consistency decrease with higher base oil viscosity.

The use of mineral oil in greases LM1 and LM2 leads to considerably higher values of consistency. The literature reports that the affinity of mineral oils for lithium 12-hydroxystearate is higher than in PAO oils. Indeed a smaller quantity of thickener is usually required to obtain an equivalent value of consistency\(^1\).
An oil bleeding test (DIN 51817 IP 121) was carried out on a selection of the greases, namely LP1, LP2 and LP3, on samples taken after every homogenization steps.

The test consists in filling a wire screen cone (placed on top of a cup) with fresh grease, and lightly loading the grease with a weight. The cup diameter is 55 mm. The apparatus, schematically shown in Figure 3.9, is subsequently kept in an oven at a temperature of 40 °C for 168 hours. The amount of bled oil is weighed and expressed as a w/w percentage of the initial grease weight.

![Figure 3.9 Oil bleeding test (a) apparatus with sieve, cup and weight, and (b) schematic of test;](image)

This experiment was conducted as it is one of the major, standardised tests to evaluate the quality of manufactured greases. A minimum quantity of oil separation is required to guarantee a good lubricity during operation. However, it is important that this quantity does not exceed a
maximum value, which often indicates an incomplete saponification reaction, and consequently a poor development of the thickener microstructure.

The outcome of the test is shown in Figure 3.10 (a). One evident result is that each homogenization step, contributing to the increase of grease stiffness, also inhibits the separation of base oil.

The greases do not show any correlation in relation to base oil viscosity. However, the bled oil quantity seems to compare well with the penetration depth values found for each sample. This is shown in Figure 3.10 (b). The results obtained with the penetration test can thus be used as guidelines as to the ability of the greases to retain the base oil. The higher the NLGI consistency number, the less the grease bleeds oil.

Typical values of oil separation required for commercial greases are in between the range of 0.5 and 4 %\(^1\). Except for LP1 after homogenisation step 1, the remaining custom greases have values within these limits. The decrease in bled oil percentage with decreasing penetration depth is approximately linear.

![Figure 3.10](image)

**Figure 3.10** (a) Measured bled oil percentage values of custom greases LP1, LP2 and LP3 after homogenisation steps 1, 2, and 3; (b) comparison with penetration depth values

### 3.3.4.3 Thickener microstructure - SEM analysis

The grease microstructure was investigated by scanning electron microscopy (SEM), using a Hitachi S-3400 N microscope. The samples were prepared by first spreading a thin layer of each grease on a cylinder mount. These grease layers were repeatedly flushed with toluene to dissolve and separate the base oil. Finally only the residual thickener was left on the mounts. An Agar sputter coater was used to cover the thickener samples with gold, and thus improve conductivity. Pictures of the thickener samples were taken under high vacuum, at a voltage of 5 kV and various magnifications. Caution should be exercised in the interpretation of these pictures in
terms of their relation to the actual thickener structure as it would appear in the original grease. The leakage of base oil out of the spaces between the thickener particles causes them to collapse on each other, due to the absence of the supporting oil. Furthermore, the action of flushing toluene physically moves the thickener particles. Therefore the preparation of the samples itself will induce some changes in the original grease microstructure. The use of such a technique is indeed debated\(^1\), as the obtained pictures could be somewhat misleading\(^2\).

Pictures of the grease microstructures at 5000 X and 1000 X magnification are shown in Figure 3.11 and Figure 3.12. All the greases display the typical microstructure of lithium greases, described in the literature as a complex network of highly entangled fibres\(^3\). However, the pictures show differences in fibres length, size and interaction, as a function of the base oil type and viscosity.

It is difficult to recognise a trend in these features as a function of the characteristics of the base oil. For example, the influence of base oil viscosity, which may be expected to be apparent when comparing the LP greases in Figure 3.11\(^4\), is unclear. Among these, the thickener fibres in LP2 are the thinnest. In the remaining LP greases the degree of agglomeration between the fibres, and the hollow spaces between them, is bigger, especially in LP4 and LP5. All the single fibres have an average diameter size in the range 60 – 90 nm.

LPE grease is made of much thinner (30 – 50 nm in diameter) and shorter fibres. Indeed, a considerable difference in microstructures is noticed between this grease and the LP1 grease, despite the relatively similar base oil viscosities. Other research works also report the presence of thinner lithium fibres in ester-based greases\(^5\).

The mineral base oil greases LM1 and LM2 have long, but generally thinner (especially LM1) fibres than the LP greases. This can probably explain why these greases have higher consistencies as observed in Section 3.3.4.1. Indeed there are indication in the literature\(^6\) that the higher the ratio between the fibre length and diameter, the higher is the grease consistency.
Figure 3.11 SEM micrographs of LP greases lithium fibre networks
3.3.5 Grease additivation

The study on the effect of additives on the grease friction behaviour was limited to one type of organic friction modifier, oleic acid \((C_{18}H_{34}O_2)\). This choice was made because of the relatively well understood mechanism of friction behaviour of this type of additive\(^{132}\). Oleic acid is reported to react with metal surfaces to form dimers and higher oligomers as a result of thermal polymerization. Their polar head is anchored to the contact surfaces, while their outer layer can be easily sheared off, thus allowing for a low friction coefficient\(^{133}\).

LP1, LPE and LM1 were mixed with 5 % w/w oleic acid using a small roller mill, pictured in Figure 3.13. The approximate diameter of each roller is 50 mm. The corresponding base oils were also additised to enable comparative tests. Such a concentration for the oleic acid is relatively high, compared to the values in which it is normally used as a friction modifier in oils.

Figure 3.12 SEM micrographs of LPE, LM1 and LM2 greases lithium fibre networks
The purpose was to maximise its effect, and consequently emphasize the differences in friction behaviour between additised and non-additised greases and base oils.

![Figure 3.13 Picture of roller mill used to mix additives in custom greases](image)

### 3.3.6 List of custom greases

A summary of all the custom greases and their main properties are presented in Table 3.8.

<table>
<thead>
<tr>
<th>Grease Name</th>
<th>Base oil</th>
<th>Oil viscosity @ 40°C [cSt]</th>
<th>Oil viscosity @ 100°C [cSt]</th>
<th>Additive</th>
<th>NLGI number</th>
</tr>
</thead>
<tbody>
<tr>
<td>LP1</td>
<td>PAO</td>
<td>26</td>
<td>5</td>
<td>None</td>
<td>2</td>
</tr>
<tr>
<td>LP1a</td>
<td>PAO</td>
<td>26</td>
<td>5</td>
<td>Oleic acid</td>
<td>NA</td>
</tr>
<tr>
<td>LP2</td>
<td>PAO</td>
<td>47</td>
<td>8</td>
<td>None</td>
<td>3</td>
</tr>
<tr>
<td>LP3</td>
<td>PAO</td>
<td>98</td>
<td>14</td>
<td>None</td>
<td>3</td>
</tr>
<tr>
<td>LP4</td>
<td>PAO</td>
<td>211</td>
<td>28</td>
<td>None</td>
<td>2-3</td>
</tr>
<tr>
<td>LP5</td>
<td>PAO</td>
<td>413</td>
<td>50</td>
<td>None</td>
<td>2</td>
</tr>
<tr>
<td>LPE</td>
<td>Mixed PAO/ester</td>
<td>26</td>
<td>5</td>
<td>None</td>
<td>2-3</td>
</tr>
<tr>
<td>LPEa</td>
<td>Mixed PAO/ester</td>
<td>26</td>
<td>5</td>
<td>Oleic acid</td>
<td>NA</td>
</tr>
<tr>
<td>LM1</td>
<td>Mineral</td>
<td>35</td>
<td>5</td>
<td>None</td>
<td>3</td>
</tr>
<tr>
<td>LM1a</td>
<td>Mineral</td>
<td>35</td>
<td>5</td>
<td>Oleic acid</td>
<td>NA</td>
</tr>
<tr>
<td>LM2</td>
<td>Mineral</td>
<td>102</td>
<td>11</td>
<td>None</td>
<td>3-4</td>
</tr>
</tbody>
</table>
3.3.7 Mechanically degraded custom grease

In order to simulate and evaluate the tribological performance of the custom greases after an extended period of use in a bearing, the Shell roll stability tester (see Section 3.2.5) was used to mechanically degrade one of the custom greases. LP1 was subjected to the same degradation process described in Section 3.2.5. LP1 was chosen for this analysis due to its very similar formulation to the commercial grease LiPAO (see Section 3.2.1), so that their tribological behaviour could be compared also after mechanical degradation. However, the outcome of the test was very poor in this case. The mechanically degraded LP1 grease contained a considerable amount of wear particles, originating from the asperity interaction of the roller and cylinder surfaces during the degradation process. Since the mechanically degraded commercial greases did not show such problem, it was proposed that this could be due to the absence of additives in LP1, particularly antiwear additives. In order to overcome this issue, LP1 was additised with 3 % w/w Vanderbilts Chemicals Vanlube 73 before performing the test. This additive is described as a dispersion of antimony tris(dialkyldithiocarbamate) in oil (containing 6.5 – 7.2 % antimony, and 10 – 12.5 % sulphur), with anti-wear, extreme pressure and antioxidant properties. The roll stability test carried out on this grease sample was successful, clearly showing a lower amount of wear debris.

3.4 Conclusions

In this chapter the lubricants used for the present tribological investigations, the results of which are presented in Chapter 5 and Chapter 6, were thoroughly described. The reasoning behind the choice of these particular grease formulations was explained in relation to the research objectives of this study. The techniques used to characterise these lubricants and the results of these characterisations were presented in detail. In addition, the process of mechanical degradation used to alter the initial samples to better simulate their use in a bearing was also described. The first set of lubricants consists of commercial greases, their bled oils and de-oiled greases collected at the end of the bleeding process, as well as their mechanically degraded counterparts. The second set of lubricants consists of greases that were custom-made for this study (with and without additives), their base oils and a mechanically degraded sample.
Chapter 4. Experimental methods

This chapter presents the experimental methods employed in this study to investigate the friction and film thickness behaviour of greases. The tribology test devices, specimens, operating procedures and test conditions used for the investigation are described in detail.

4.1 Introduction

Despite the availability of standard or custom bearing test rigs, an important contribution to research in the lubrication of rolling bearings is still given by experimental work carried out on single-contact laboratory devices, such as twin-disc and ball-on-disc machines. The reason behind the choice of such test devices, rather than full bearings, lies in their simplicity and versatility. Indeed in these machines the number of possible variables influencing the outcome of each test (e.g. load, temperature, rolling and sliding speeds) can be independently controlled, thus allowing for the influence of any one condition of interest to be isolated and consequently investigated. These characteristics are particularly important in the study of greases, as the understanding of their tribological behaviour is still quite limited. In contrast, the use of full bearing test rigs bring numerous complications, such as uncontrolled grease supply to the contact, unknown slide/roll ratio and temperature, to name a few. With the exception of a few studies, the simultaneous friction and film thickness investigation in full bearings is not possible. In some cases film thickness has been effectively measured, and friction trends have been only deduced indirectly from monitoring temperature variations. Vice-versa, in other cases friction has been measured, and film thickness estimated based on temperature observations.

With single contact test rigs, no such issues are encountered, and the interpretation of the test results is far less ambiguous owing to full control of the relevant variables. This allows for more fundamental studies to be conducted, providing deeper understanding of grease behaviour. Full bearing tests could of course complement the single-contact experiments if the aim is to assess the behaviour of any new formulations under realistic application conditions. Therefore, with the aim of providing fundamental understanding of grease behaviour, ball-on-disc tribometers for friction and film thickness measurements, with a contact geometry suitable for simulating a typical concentrated contact pertinent to rolling bearings, were chosen for the current experimental work. In order to study the influence of grease composition itself on friction, it is necessary to exclude extra complications introduced by starvation. Single-contact test methods carry the additional advantage of easily maintaining a full inlet supply. This allows
An Experimental Study into the Influence of Grease Composition on Friction in EHL Contacts

for unequivocal interpretation of results since lubricant supply is tightly controlled. Hence, almost all of the tests in this study were conducted under fully flooded conditions, and effects of starvation were only briefly examined. Friction behaviour of greases was investigated over a range of temperature and speed conditions, such to identify the main parameters of influence and investigate their effects on friction. Film thickness was also measured in order to support the friction investigations and provide an insight into the prevailing mechanisms at play.

4.2 Friction experiments

EHL friction can be measured using a number of laboratory devices, including twin-disc, ball-on-disc, pin-on-disc, 4-ball machines, but also various rheometers and bearings. The ball-on-disc machine was preferred as the most appropriate rig to conduct the current study, seeking to investigate grease frictional behaviour in single bearing-like contacts. Understandably, the general lubrication conditions in a real rolling bearing are still far from that simulated in a ball-on-disc machine. However, the measurements obtained with this test rig have shown to correlate well with full bearing test results, validating the ball-on-disc test as a suitable method to screen the behaviour of greases in real bearings135,136. Compared to a test in a real bearing, a ball-on-disc laboratory rig allows for carefully and precisely controlled test conditions, which are an essential requirement for the scope of the current study as discussed above.

4.2.1 Description of the MTM friction test rig

The PCS Instruments MTM rig was used to test grease friction behaviour. The rig is schematically illustrated in Figure 4.1 (a)137, in its standard ball-on-disc configuration (Figure 4.1 (b)). The ball and disc specimens are mounted on shafts which are independently driven by DC servo motors to allow for a wide range of rolling/sliding conditions. The shaft carrying the ball is supported by a gimbal arrangement. This makes it possible to apply the ball load on the disc at an angle, minimising the contribution to friction given by spin while permitting the rotation of the ball. The frictional force between the ball and the disc is measured by a force transducer. Application of the load and restraint of the traction force is made through high stiffness force transducers, appropriately mounted in the gimbal arrangement to minimise the overall support system deflections. The load is applied via a stepper motor. The rig is fully controlled via the PC. The ball and disc specimens are contained in a small pot made of stainless steel, and covered with a stainless steel lid. In turn, the covered pot is contained in a PTFE thermal insulating jacket. Heat is applied through cartridge heaters, mounted inside the pot, and the temperature is measured and controlled through two resistance thermometers (RTDs). One RTD is positioned

99
inside the pot reservoir (lube probe), and the other inside the stainless steel pot itself (pot probe). The test rig has been previously shown to be able to successfully measure friction in grease lubrication\textsuperscript{108,4,119}.

![Figure 4.1](image_url)  
(a) Schematic representation of the MTM friction test rig\textsuperscript{137}; (b) close-up picture of the ball-on-disc specimens arrangement

Friction measurements on the MTM rig under mixed rolling/sliding conditions are made by taking two successive measurements, one with the disc rotating faster than the ball (generating a tension force on the load cell), and the other with the ball rotating faster than the disc (generating a compression force). Friction is then measured by halving the absolute difference between the two measurements. The primary reason for doing this is to remove any potential offset in the load cell attached to the ball drive shaft. However, this method of calculation also cancels any rolling friction force, as this does not change sign depending on the relative speed of the surfaces\textsuperscript{138}. Moreover, the contribution to friction coming from churning is negligible, due to the minimal amount of lubricant which is used for the test.

### 4.2.2 Test specimens

Both ball and disc specimens are made of AISI 52100 standard bearing steel, so that their material properties and surface chemistry is similar to that found in rolling bearings. A standard 19.05 mm diameter superfinished ball ($R_q \approx 15 \text{ nm}$) and 46 mm polished disc ($R_q \approx 10 \text{ nm}$), provided by PCS Instruments, were used for most tests. Some tests were also carried out with rougher discs, in order to test the lubricants over a wider range of nominal $\Lambda$ ratio value. In particular, this made it possible to accomplish much lower values of $\Lambda$, as the values of speed needed to achieve the same with smooth discs would be too low and beyond the means of control and precision of the MTM motors.
‘Rough’ discs with an average surface roughness of $R_q \approx 270$ nm were provided by PCS Instruments. With the purpose of obtaining medium values of roughness (and $A$ ratios), some smooth discs were also roughened using an ISO/FEPA P800 grit silicon carbide abrasive paper on a metallurgical polisher. These discs are labelled as ‘roughened’, and have $R_q$ values ranging from 100 to 113 nm, with an average $R_q$ value across all roughened discs of 106 nm. The smooth, roughened and rough discs all have isotropic roughness. The average values of the discs $R_q$ roughness, measured on a Veeco Wyko NT9100 optical profiler, are given in Table 4.1.

<table>
<thead>
<tr>
<th>Disc type</th>
<th>Roughness $R_q$ [nm]</th>
<th>Composite disc + ball roughness $R_q$ [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Smooth</td>
<td>10</td>
<td>18</td>
</tr>
<tr>
<td>Roughened</td>
<td>106</td>
<td>107</td>
</tr>
<tr>
<td>Rough</td>
<td>270</td>
<td>270</td>
</tr>
</tbody>
</table>

### 4.2.3 Friction test procedure and conditions

The test profiles used for the friction analysis are here introduced. Each test profile aims at investigating different characteristics in the greases friction behaviour. The definition of two variables used in this experimental work, i.e. entrainment speed and slide/roll ratio, are given below.

The entrainment speed $U$ is the average speed of the contact surfaces of the ball and the disc:

$$U = \frac{U_1 + U_2}{2}$$

where $U_1$ and $U_2$ are the tangential speeds of the ball and disc surfaces.

The slide/roll ratio (SRR), expressed as a percentage, is the ratio between the absolute sliding speed and the entrainment speed:

$$SRR = \left(\frac{|U_1 - U_2|}{U}\right) \times 100$$

### 4.2.3.1 Fully flooded friction tests

The majority of the results presented in this work look at the ‘Stribek’ behaviour of the test lubricants, i.e. at how friction changes with entrainment speed, while keeping the remaining variables (SRR, normal load, temperature) constant.
These friction tests consist of 4 steps. In the first, preliminary step, a small ball load of 1 N was applied on the disc, at a constant entrainment speed of 5 mm s\(^{-1}\), for 5 minutes and in pure rolling conditions. This preliminary step was introduced with the purpose of pre-shearing the grease, equivalently to common protocols used to examine greases flow and viscoelasticity in a rheometer\(^{23}\), and ensuring the same initial conditions for each test. The friction data were then obtained in 3 consecutive speed-sweep steps, where the speed was increased from 10 mm s\(^{-1}\) to 1000 mm s\(^{-1}\) at a constant value of maximum Hertzian pressure of 0.96 GPa, and SRR of 10 %. With this procedure, the step repeatability within the same test, and the analysis of the effect of time dependency on the friction coefficient evolution after each step could be monitored. Friction tests were carried out at 40 °C, 60 °C and 80 °C. Such operating conditions were chosen as they were considered both representative of typical contact conditions in a rolling bearing, and compatible with the capability of the rig. Typical SRR values estimated for rolling bearings range between 3 and 5 %\(^9\). A slightly higher nominal value of 10 % SRR was chosen, as this can be achieved with higher precision with the MTM disc and ball motor controllers even at the lowest speeds employed. For the same reason, the minimum speed was limited to 10 mm s\(^{-1}\). Below this value the combined speed control of disc and ball motors becomes poor, and the actual value of SRR cannot be guaranteed to be equal to the desired one. The maximum speed was restricted by the limited functionality of the test setup in providing inlet fully flooded conditions at high speeds. The test conditions used for every step in the fully flooded friction test are summarised in Table 4.2.

<table>
<thead>
<tr>
<th>Table 4.2 Conditions for fully flooded friction tests</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Hertzian pressure [GPa]</td>
</tr>
<tr>
<td>Temperature [°C]</td>
</tr>
<tr>
<td>Slide/roll ratio [%]</td>
</tr>
<tr>
<td>Entrainment speed range [mm s(^{-1})]</td>
</tr>
</tbody>
</table>

In a preliminary study it was observed that, within reason, friction in these tests does not depend on the amount of grease used, meaning that with the current test setup the conditions were always fully flooded. Nevertheless, the same quantity of grease of one gram was weighed each time on a precision balance, and distributed by means of a spatula all over the disc track. In order to maintain the fully flooded lubricant conditions supply at the contact inlet over the entire speed range, a small scoop was used to collect the grease displaced to the sides of the disc by the rolling action of the ball, and push it back into the rolling track. The scoop, schematically shown in
Figure 4.2, consists of a PTFE block with a channelling groove. This is positioned between the ball and the inlet in the rolling direction, and pushed down onto the disc with a spring. When testing oils the scoop was not used, as in this case the fully flooded conditions are simply achieved by filling the pot, so oil just covers the surface of the disc. The same test profile was used to study the behaviour of the bled and base oils.

Prior to each test, ball and disc were individually placed in toluene, and cleaned in an ultrasonic bath for 15 minutes. The same procedure was repeated with isopropanol, after which the specimens were dried with a heat gun. Repeatability of results was generally excellent and was confirmed by conducting every friction test for each grease and test condition at least twice, with 3 Strubeck-like steps in each test as described above. Nevertheless, all reported friction coefficient values are an average of two last steps in each test and its repeat, i.e. an average of two values.

Figure 4.2 Schematic representation of grease scoop supply mechanism

4.2.3.2 Starvation friction test

In order to test the greases ability in feeding the contact inlet without a forced supply, as it would occur in a real bearing, a few additional tests were conducted under nominally starved conditions, i.e. with no forced grease supply to the contact inlet. The friction tests in which this particular aspect was investigated are referred to as ‘starvation’ tests. Friction measurements in these conditions have already been successfully employed in the past to study the starvation behaviour of oils on the same test rig. A procedure was followed to achieve a consistent initial distribution of the grease, since these tests are normally highly susceptible to poor repeatability. One gram of grease was initially distributed over the disc track. Similarly to the fully flooded test profile, the grease scoop was
employed in a preliminary step where the grease was ‘worked’ at a constant speed of 5 mm s$^{-1}$, for 5 minutes and in pure rolling conditions, with 1 N ball load. At the end of this step, all the excess grease present on the sides of the rolling tracks of the disc and the ball, and on the grease scoop, was removed with a spatula and weighed. Only a thin layer of grease was left on the disc and ball rolling tracks, corresponding to circa 30 mg of grease (in the 27-33 mg range). The grease scoop was then removed, so that from this point forward there was no forced grease supply to the contact inlet. Therefore the grease supplied to the contact was determined by just the grease properties and contact conditions. A constant speed of 50 mm s$^{-1}$ was first applied for 20 min, at 0.96 GPa maximum Hertzian pressure and SRR of 10%, i.e. the same load and SRR employed in the fully flooded tests. At the end of this step the motion was stopped and the load removed. The test was then immediately restarted again at the above conditions. This procedure was repeated 4 more times. This stop-and-go sequence was used to incentivise lubricant reflow around the contact, allowing the friction coefficient to be monitored over a longer time. Eventually, the same tests were carried out at speeds of 100, 200 and 500 mm s$^{-1}$, or until the friction coefficient reached high values indicating severe starvation conditions. The test conditions employed in the ‘starvation’ tests are listed in Table 4.3.

The inlet supply conditions could be deduced from the interpretation of the friction curves. The level of inlet starvation was, as expected, dependent on the type of grease and the test conditions.

<table>
<thead>
<tr>
<th>Table 4.3 Conditions for starvation friction tests</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Hertzian pressure [GPa]</td>
<td>0.96</td>
</tr>
<tr>
<td>Temperature [˚C]</td>
<td>40</td>
</tr>
<tr>
<td>Slide/roll ratio [%]</td>
<td>10</td>
</tr>
<tr>
<td>Entrainment speed [mm s$^{-1}$]</td>
<td>50, 100, 200, 500 (5 X each)</td>
</tr>
<tr>
<td>Total time at each speed [min]</td>
<td>100</td>
</tr>
</tbody>
</table>

### 4.3 Film formation experiments

*Friction in grease-lubricated contacts should be evaluated in light of the sources that generate it. Firstly, it is essential to understand if friction is entirely due to the shearing resistance of the lubricant, or if asperity contacts are also contributing. Secondly, it is important to understand how the shearing resistance of this lubricant film correlates to its thickness. Finally, due to the multiphase composition of greases, the film nature (i.e. its composition and rheological behaviour) determining friction is, in principle, unknown, and is likely to vary*
An Experimental Study into the Influence of Grease Composition on Friction in EHL Contacts

depending on the operating conditions. For these reasons the mechanism of grease film formation was investigated in order to complement the friction results and help establish the friction mechanisms.

4.3.1 Description of the film thickness test rig

Optical interferometry is extensively applied to the study of lubricated EHL contacts, and its validity and versatility in this respect have been fully verified. An extensive review of the principles of this technique as applied to film thickness measurements can be found in a paper by Spikes and Cann. All the film thickness measurements carried out on the commercial greases were taken using the PCS Instruments EHD rig, illustrated schematically in Figure 4.3. This machine also employs a ball-on-disc geometry. The ball and disc specimens are end mounted on shafts which are independently driven by electric motors to allow for any sliding/rolling conditions. The load is applied by a stepper motor moving the ball upwards, against the disc. The ball is a standard 19.05 mm diameter AISI 52100 steel ball, with roughness \( R_q \approx 15 \) nm, i.e. the same ball specimen as used in the MTM friction tests. The disc is made of glass, with a coating of approximately 20 nm of chromium (CrSiO\(_3\)), overlaid by 500 nm of silica (SiO\(_2\)) on its lower surface. White light is shone vertically through the disc, focussed into the centre of the contact. Part of this light is reflected by the chromium layer, the remaining portion by the steel ball. These two beams recombine at wavelength values that depend on the difference in their path length, which in turn is determined by the thickness of the film separating the ball from the disc. This interference light is then passed into a spectrometer slit where it is dispersed to produce a set of interference fringes. The resulting spectral image is then detected by a black-and-white CCD camera and captured by a frame grabber. The wavelength corresponding to the maximum constructive interference in the central region of the contact is then processed by the software to calculate the central film thickness.

One of the limitations of this technique, commonly known as ‘spacer layer optical interferometry’\(^{141}\), is that it can only provide one film thickness measurement over a specific area of the contact. In oil lubrication the film formation mechanism and shape are generally known, and the film in the central area of contact is relatively flat. However, in the case of grease lubrication, depending on the operating conditions the typical EHL horseshoe shape can appear highly disturbed\(^{142}\), and the description of film formation in terms of a single absolute film thickness value provides very limited information. In order to improve this aspect, a different rig was used in the second experimental part of this research work. All the measurements carried out on the custom greases were taken with the Wedeven Associates Inc. WAM5 rig, with nearly
identical settings to the EHD rig. The main difference between the standard setups present in the two rigs consists in the method used to process the interference image of the contact. Instead of using a spectrometer to determine the wavelength of maximum interference, a high resolution CCD colour camera is employed on the WAM5 to grab an image of the whole contact. This technique is known as ‘spacer layer imaging method’ (SLIM). With this setup the whole film thickness map across the contact can be dynamically observed, and the formation of films with different shapes than classical EHL films can be identified. In this way the film thickness measurements, also taken with the SLIM technique in the tests performed with the custom greases, could also be accompanied by a dynamic visual image of the film shape inside the contact. In this case the film thickness was calculated as an average of the predominant colours in the central area of the contact, for each picture taken at the same speed.

4.3.2 Film thickness test procedure and conditions

All the tests were performed in fully flooded conditions, hence a grease scoop similar to the one described in Section 4.2.3.1 was used. Pure rolling conditions were chosen to limit scratching of the glass disc. The film thickness testing procedure consisted of 3 steps. The first step is equivalent to the preliminary step used in the fully flooded friction test. A low load was applied to ‘work’ the grease and ensure equal grease distribution and pre-shearing conditions before commencing each test. The film thickness measurements were taken during the following two speed-sweep steps, where the speed was first increased from 5 mm s\(^{-1}\) to 1000 mm s\(^{-1}\), and then decreased back to 5 mm s\(^{-1}\). Five measurements were taken during each step at the same speed. The reported film thickness results are an average of the speed decrease steps in each test and its
repeat, i.e. an average of ten values. The tests were run at 40 °C and ambient temperature (22 ± 1 °C), depending on the lubricant samples. The test conditions are summarised in Table 4.4. Prior to each test, ball and disc specimens were cleaned using the same procedure employed for the friction test specimens.

<table>
<thead>
<tr>
<th>Table 4.4 Conditions for the film thickness measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Hertzian pressure [GPa]</td>
</tr>
<tr>
<td>Temperatures [°C]</td>
</tr>
<tr>
<td>Slide/roll ratio [%]</td>
</tr>
<tr>
<td>Entrainment speed range [mm s⁻¹]</td>
</tr>
</tbody>
</table>

4.4 Conclusions

In this chapter the experimental methods used for the tribological investigation of the test greases have been described. Two ball-on-disc tribometers have been employed to study the grease friction and film thickness behaviour, due to their suitability in simulating rolling bearings-like concentrated contacts, whilst allowing for carefully controlled test conditions. Their principle of operation has been briefly explained. The material and surface characteristics of the specimens have been described. The test profiles and test conditions have been specified. The next two chapters report the experimental results obtained with these techniques.
Chapter 5. Experimental results with commercial greases

This chapter contains the experimental results on friction and film thickness obtained with the commercial greases and their bled oils. Properties of the commercial greases tested here are presented in Table 3.1 in Chapter 3. The investigation is focused on recognizing trends between grease composition and friction and film thickness, at different operating conditions (speed and temperature), surface roughness and rheological state of grease (fresh or mechanically degraded), as well as establishing a reference for the performance of currently available low-friction greases.

5.1 Introduction

As evident from Chapter 2, the current knowledge about the frictional behaviour of lubricating greases in typical bearing rolling/sliding contacts is very limited. This is partly due to an equivalent lack of understanding of their film forming behaviour, on which friction depends, which in turn is a function of the grease formulation and operating conditions. In the attempt to identify general trends of interest regarding the frictional response of very different grease formulations (base oil and thickener types, and base oil viscosities), readily available, commercial bearing greases were studied first in the experimental investigation. The relative influence of the basic components was investigated, and film thickness measurements were presented to support friction results. The influence on friction of different contact surface roughness, and of mechanical degradation of grease, was also investigated.

5.2 Grease friction

The friction results obtained with the commercial greases are shown in Figure 5.1, Figure 5.2 and Figure 5.3 at 40°C, 60°C, and 80°C respectively. The measurements are displayed using a log-speed plot so as to highlight the friction values obtained at low speeds, where many differences in the results were observed in terms of both magnitude and trends. The most striking feature in these figures is the very different frictional behaviour of the greases. In particular, at low speeds the friction values cover a range of almost one order of magnitude, from below 0.01 to almost 0.09. It is interesting to compare the behaviour of greases currently marketed as ‘low-friction’, namely LiEs and LiPAO. LiPAO clearly displays the lowest friction coefficient throughout the whole speed range, while LiEs only reaches a relatively low friction coefficient at high speeds. At lower speeds LiEs actually gives higher friction than some standard greases.
Figure 5.1 Friction coefficient of commercial greases at 40°C plotted against entrainment speed

Figure 5.2 Friction coefficient of commercial greases at 60°C plotted against entrainment speed
Figure 5.3 Friction coefficient of commercial greases at 80°C plotted against entrainment speed

The trends in the relationship between friction and speed are very different depending on the specific grease. Better comparisons can be made by observing Figure 5.4, where the results obtained for each grease at the three test temperatures were re-plotted individually. In all lithium greases LiMi, LiMi2, LiEs, LiPAO and LiCPAO and the polyurea PoEs grease, the friction curves obtained at low speeds are generally different from the shape of a typical Stribeck\textsuperscript{49} curve of an oil. With increasing speed, LiMi2 shows constantly increasing friction at all temperatures, while for LiMi, LiEs, LiPAO, LiCPAO and PoEs the friction coefficient rises with speed, up to a local maximum (appearing as a ‘bump’), after which it starts descending. This behaviour only appears at 60 °C in LiEs, LiCPAO and PoEs. The ‘bump’ generally moves to higher speed with increasing temperatures. On the contrary, in the case of calcium sulphonate complex greases CaSMi and CaSMix, the maximum friction coefficient value is found at the lowest speed at all temperatures for the given test conditions, and monotonically decreases as the speed is increased. Therefore the curves described by CaSMi and CaSMix appear more similar to Stribeck-like curves typically obtained with lubricating oils. At sufficiently high speeds the friction coefficient curves for each grease become flatter, and the speed at which this condition is reached for each grease increases with temperature.
Figure 5.4 Friction coefficient of commercial greases at 40 °C (blue markers), 60 °C (red markers) and 80 °C (green markers) plotted against entrainment speed.
To better describe the influence of temperature on grease friction, Figure 5.5 shows a bar chart of the measured friction coefficient at the three temperatures, and at a fixed speed of 1000 mm s\(^{-1}\). This speed is sufficiently high for all the greases to develop a full EHL film (i.e. \(\Lambda > 3\), where \(\Lambda\) is the specific film thickness estimated as the ratio of calculated central film thickness with the base oil to composite \(R_q\) roughness). Clearly, an increase in temperature leads to a decrease in friction for each individual grease at this speed. However, the effect of temperature on friction is much less obvious at lower speeds. Figure 5.6 shows the equivalent bar chart for friction measured at 50 mm s\(^{-1}\). It is apparent that at this lower speed the effect of temperature on friction is different depending on the specific grease. Friction either decreases with temperature, as for LiMi or LiMi2, is unaffected as in the case of CaSMi, or decreases with temperature as for CaSMix. Interestingly, an opposite behaviour can be seen in the two low-friction greases: in LiPAO friction decreases with temperature, while the opposite is observed in LiEs.
In an attempt to investigate a possible correlation between base oil viscosity and measured friction across all greases, Figure 5.7 plots the friction coefficient measured at 1000 mm s\(^{-1}\) and 40 °C (i.e. \(A > 3\)) against the base oil kinematic viscosity. Although no clear correlation is found, the results clearly show that the lowest friction coefficient values are achieved with the synthetic base oil greases, LiEs, LiPAO, PoEs and LiCPAO as indicated in the figure.

![Figure 5.7 Friction coefficient of commercial greases at 1000 mm s\(^{-1}\) and 40 °C plotted against kinematic viscosity of base oils](image)

### 5.3 Bled oil friction versus grease friction

The bled oil friction behaviour was investigated with the purpose of establishing the relative influence of thickener and base oil on friction. It should be noted that the bled oils, obtained according to the procedure described in Chapter 3, may well be different in composition, and hence in performance, from the base oils, as they are likely to contain additives\(^{76}\) and thickener particles\(^{91}\). However, the comparison between the friction behaviour of LiPAO bled and base oil (the only base oil from these commercial greases available for this study) showed minor differences, only evident within the mixed regime where the bled oil displays slightly lower friction than the base oil. This comparison is shown in Figure 5.8 at 80 °C, the temperature at which these differences are most apparent.
Figure 5.8. Friction coefficient of LiPAO base and bled oil at 80 °C plotted against entrainment speed

For clarity, only selected results are shown in this section, chosen to illustrate the general trends observed when comparing the performance of commercial greases and their respective bled oils. Further results are shown in Section 5.6 for other purposes. The friction coefficient of LiPAO and its bled oil is plotted in Figure 5.9 at the three test temperatures. The result obtained with the ‘thickener’, i.e. the de-oiled grease after the centrifugation of grease, is also shown for comparison. It is immediately apparent that LiPAO friction is much lower than that of its bled oil in the low speed region, thus giving an ‘inverse-Stribeck’ behaviour. However, as the speed increases, a point is reached where the friction curves become almost identical. The speed value where this occurs corresponds to conditions of mixed lubrication with the bled oil, where friction is still decreasing with speed. This ‘transition speed’ value also increases with temperature, i.e. the ‘inverse-Stribeck’ behaviour of LiPAO at low speed is extended up to higher speed when temperature is increased. The same behaviour was observed in all other lithium-thickened greases, as well as the polyurea grease. It is interesting to notice that the ‘thickener’ behaves as the grease at low speeds, while as soon as the grease and bled oil curves join, ‘thickener’ friction starts to rapidly increase, most likely due to the occurrence of starvation at these higher speeds. The same result was found in tests with LiEs ‘thickener’.

Both calcium sulphonate complex greases showed contrasting behaviour to that of lithium and polyurea greases described above. To illustrate this, the equivalent CaSMix and its bled oil friction results are plotted in Figure 5.10. It is evident that this grease gives a much higher friction than its bled oil in the low speed region, with the relative difference in friction becoming larger at higher temperatures. The transition speed after which grease and oil exhibit the same friction is again identified. As was the case for lithium greases, this speed increases with temperature. In the case of calcium sulphonate greases this effect appears even more
pronounced, so that at the highest temperature of 80 °C the grease and bled oil friction curves do not even start merging within the shown range of test speeds (< 1000 mm s⁻¹).

Figure 5.9 Friction coefficient of LiPAO, its bled oil and thickener at (a) 40°C, (b) 60°C and (c) 80°C plotted against entrainment speed
Figure 5.10 Friction coefficient of CaSMix and its bled oil at (a) 40°C, (b) 60°C and (c) 80°C plotted against entrainment speed
5.4 Grease film thickness

The film thickness results for LiPAO, LiEs and CaSMix greases are displayed in Figure 5.11, Figure 5.12 and Figure 5.13 respectively, as a representative selection of all film measurements made. For comparison, the theoretical film thickness values for all corresponding base oils, obtained using the Hamrock-Dowson EHL film thickness equation\textsuperscript{38}, are also shown. A thicker film at low speed, when compared to the base oils, was observed for all greases, but differences in behaviour are evident. LiPAO (Figure 5.11) shows the highest reached film thickness at the lowest speed of 10 mm s\textsuperscript{-1}, where it is more than 5000 % larger than that of the corresponding base oil. As the speed increases, the film thickness reduces and its minimum (\(\approx 90\) nm) is reached at a speed of about 300 mm s\textsuperscript{-1}, after which the film thickness starts to increase with speed as expected from the EHL theory. However, LiPAO film thickness remains higher than that of its base oil over the whole speed range tested here. LiEs (Figure 5.12) film thickness is much lower than that of the LiPAO grease at all speeds, despite LiEs and LiPAO being made of base oils of similar viscosity and the same thickener type. Nevertheless, the general trends are the same, with LiEs grease film thickness being much greater than that of its base oil at low speeds, and gradually reducing as the speed is increased. However, in LiEs the speed of transition into the EHL-like behaviour that may be expected from its base oil alone is already reached at around 10 mm s\textsuperscript{-1}, where the film thickness is \(\approx 10\) nm. This v-shaped film thickness versus speed curve has been observed in earlier grease studies\textsuperscript{95,71}. In contrast to the lithium based greases, CaSMix (Figure 5.13) film thickness does not exhibit the ‘v-shape’ curve. Instead, its film thickness constantly increases with speed within the test speed range. Similar behaviour was observed with the other calcium sulphonate complex grease, CaSMi, tested here. However, once again the grease film thickness is higher than that expected from the corresponding base oil in the low speed range, below about 200 mm s\textsuperscript{-1}. 
Figure 5.11 LiPAO film thickness and base oil film thickness prediction (solid line) at 40 °C plotted against entrainment speed

Figure 5.12 LiEs film thickness and base oil film thickness prediction (solid line) at 40 °C plotted against entrainment speed

Figure 5.13 CaSMix film thickness and base oil film thickness prediction (solid line) at 40 °C plotted against entrainment speed
5.5 Effect of roughness on grease friction

The friction results obtained at 40 °C using smooth ($R_q \approx 10$ nm), roughened ($R_q \approx 106$ nm) and rough ($R_q \approx 270$ nm) discs with the lithium and polyurea greases are shown in Figure 5.14. By testing the greases using rougher discs, generally higher friction is found. However, the trends are not entirely clear, especially in LiPAO where the friction coefficient at the lowest speeds reaches similar (exceptionally low) values regardless of roughness configuration.

![Friction coefficient graphs for different greases and roughness configurations.](image-url)

Figure 5.14 Friction coefficient of commercial greases with smooth ($R_q \approx 10$ nm, in blue markers), roughened ($R_q \approx 106$ nm, in red markers) and rough ($R_q \approx 270$ nm, in green markers) discs at 40 °C plotted against entrainment speed.
5.6 Effect of mechanical degradation on grease friction and film thickness

The friction curves of mechanically worked LiMi, LiEs, LiPAO and CaSMix at 40 °C are shown in Figure 5.15. The curves obtained with the corresponding fresh greases and bled oils are also displayed for comparison. The result of mechanically degrading LiMi, LiEs and CaSMix is to alter their original friction behaviour observed with the fresh greases at low speeds, which now becomes almost identical to their bled oil. Since the friction obtained with the fresh greases at high speeds is also equivalent to the bled oil friction, these mechanically degraded greases behave overall as their bled oils in these test conditions. Therefore the mechanical degradation of LiMi and LiEs produces a friction increase at low speeds, while the opposite is found in CaSMix. In contrast, the mechanically worked LiPAO grease behaves the same as in its fresh state, i.e. mechanical degradation had negligible effects in this case.

**Figure 5.15** Friction coefficient of mechanically worked LiMi, LiEs, LiPAO and CaSMix at 40 °C plotted against entrainment speed; comparison with results obtained with fresh greases and bled oils
Film thickness measurements at 40 °C were taken with the mechanically degraded LiEs and LiPAO greases. These are compared to the original results obtained with the fresh greases in Figure 5.16.

In LiEs a reduction in film thickness is observed at low speeds. The original increase in film thickness with decreasing speed, producing the v-shaped curve, is not displayed by the worked grease. On the contrary, as in the corresponding friction measurements, no significant effect is shown on the film thickness behaviour of LiPAO after its mechanical degradation.

![Graph](image_url)

**Figure 5.16** Film thickness of fresh and mechanically worked LiEs and LiPAO at 40 °C plotted against entrainment speed

### 5.7 Summary

The friction and film thickness results obtained with the commercial greases was presented in this chapter. The effect of grease composition, operating conditions (speed, temperature and surface roughness) and mechanical aging of grease on friction was investigated. Markedly different friction behaviour was observed between the tested greases. An attempt to assess the
relative influence of the individual grease components on friction was made by studying the behaviour of the corresponding bled oils under the same conditions. As a general trend, it was observed that the tribological behaviour of grease can differ significantly from that of its bled oil below a certain transition speed. This speed increases with temperature and appears to depend on grease composition. In terms of influence of grease composition on friction, it was observed that:

- Above the ‘transition speed’, friction is lower in the synthetic oil-based than in the mineral oil-based greases, while no obvious correlation between grease friction and base oil viscosity was observed amongst the greases;
- Below the ‘transition speed’, all tested greases are able to form a thicker film than their base oils. The corresponding friction behaviour seems to be strongly dependent on thickener type. Compared to their bled oils, lithium and polyurea greases exhibit lower friction, while the opposite is observed in the calcium sulphonate complex greases. In the majority of the greases this thick film forming behaviour is diminished by mechanical aging, so that the aged greases behave more similarly to their bled oils. The notable exception was the LiPAO grease, whose film thickness and friction behaviour remained unaffected by mechanical degradation. Moreover, at sufficiently low speeds this grease is able to provide very low friction even with very rough surfaces.

The findings presented in this chapter provided general guidelines for the selection of the basic ingredients, as well as the ones to avoid, in the formulation of low-friction greases. Moreover, the results shed light onto some of the possible mechanisms responsible for the tribological behaviour of greases. This preliminary work successfully laid the foundations for a more detailed study employing custom greases, the formulations of which were driven also by the observations made here. The results obtained with this set of custom greases are presented in the next chapter.
Chapter 6. Experimental results with custom greases

This chapter continues the investigation on the influence of grease composition on friction commenced in Chapter 5, by using a set of custom greases, designed to enable a study of the effects of more specific aspects of grease formulation, namely:

- Base oil viscosity;
- Base oil type;
- Presence of additives.

Properties of the custom greases employed here are presented in Table 3.8 in Chapter 3. In addition to studying fresh greases in a fully flooded contact, the analysis presented here incorporates the influence of grease mechanical degradation on friction, and a short investigation into the frictional behaviour of greases in starved lubrication conditions.

6.1 Introduction

Once some general trends were identified in the preliminary investigation conducted with the commercial greases and presented in Chapter 5, this second part of the study considered custom-made greases. Although very useful, the investigation with the commercial grease samples presented above inevitably suffers from uncertainties regarding the exact composition of the commercial greases. Only basic information was provided by the manufacturers, resulting in limited knowledge of the base oil type, thickener percentage, presence of additives and manufacturing cycle. The known composition and controlled manufacturing process employed for the custom greases tested here allowed for such uncertainties to be minimised. Moreover, the formulations of the commercial greases differ from each other in more than one component and/or property, which makes it difficult to accurately evaluate the specific influence of any single grease component on the general grease frictional behaviour. These constraints are removed with the custom greases by implementing a logical reasoning for the design of their composition, so that only one variable is varied at a time, as described in Chapter 3. Therefore the present study with the custom greases complements the knowledge obtained with the commercial greases by focussing on the effects of very specific aspects of grease formulation,
including the influence of base oil type, base oil viscosity and the presence of friction modifiers, on friction.

6.2 Tribological behaviour of base oils

The film thickness and friction results obtained with the base oils used in the preparation of the custom greases are presented first. Properties of these base oils are listed in Table 3.5 in Chapter 3. These results are used as a baseline for the evaluation of the tribological behaviour of the custom greases.

6.2.1 Base oil film thickness

The film thickness results obtained with the base oils (BO) at ambient temperature (22 ± 1 °C) are plotted against the entrainment speed $U$ in Figure 6.1.

![Figure 6.1. Film thickness of base oils at ambient temperature (22 ± 1 °C) plotted against entrainment speed](image)

The base oil film thickness measurements were used to calculate their pressure-viscosity coefficients $\alpha$, by back-calculation using the Hamrock-Dowson EHL film thickness equation$^{38}$. The results are reported in Table 6.1. The PAO oils have comparable values of $\alpha$, while the corresponding values for the mineral oils LM1 BO and LM2 BO are significantly higher. The
obtained values are compatible with experimental data found elsewhere for these types of oils\textsuperscript{144}. Although the pressure-viscosity coefficients are reported to depend on a series of operating conditions, in this study they were assumed to be constant. These were later used to estimate film thickness in the friction tests.

<table>
<thead>
<tr>
<th>Base oil</th>
<th>LP1</th>
<th>LP2</th>
<th>LP3</th>
<th>LP4</th>
<th>LP5</th>
<th>LPE</th>
<th>LM1</th>
<th>LM2</th>
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<tbody>
<tr>
<td></td>
<td>BO</td>
<td>BO</td>
<td>BO</td>
<td>BO</td>
<td>BO</td>
<td>BO</td>
<td>BO</td>
<td>BO</td>
</tr>
<tr>
<td>$\alpha$ [GPa\textsuperscript{-1}]</td>
<td>15</td>
<td>17</td>
<td>16</td>
<td>16</td>
<td>16</td>
<td>15</td>
<td>21</td>
<td>26</td>
</tr>
</tbody>
</table>

### 6.2.2 Base oil friction

The friction curves obtained with the base oils are shown in Figure 6.2, Figure 6.3 and Figure 6.4 at 40°C, 60°C, and 80°C respectively. The very different behaviour of the oils at low speeds is apparent. This is mostly due to the wide viscosity range, which in turn produces different lubrication regimes (boundary, mixed and full film) in the current test conditions, and therefore different extents of asperity interaction. At higher speeds all the PAO and mixed PAO/ester oils display very similar values of friction, and their curves are almost indistinguishable. The mineral oils of LM1 and LM2 give evidently higher friction.
Figure 6.2 Friction coefficient of base oils at 40 °C plotted against entrainment speed

Figure 6.3 Friction coefficient of base oils at 60 °C plotted against entrainment speed
Figure 6.4. Friction coefficient of base oils at 80 °C plotted against entrainment speed

6.3 Tribological behaviour of custom greases

Film thickness and friction results obtained with the custom greases are presented in this section.

6.3.1 Grease film thickness

Figure 6.5 shows examples of interferograms taken at different speeds for all the greases on the ball-on-disc film thickness rig described in Chapter 4. Under conditions of low speeds, the surfaces of the disc and ball appear to be separated by a ‘lumpy’ film. The smooth classical EHL horseshoe shape is eventually reached at a sufficiently high speed. Focussing on the lithium-PAO (LP) greases, a trend can be recognised, where the lower is the base oil viscosity, the higher is the speed of transition at which the horseshoe shape is reached.
The corresponding film thickness measurements for these greases are plotted against the entrainment speed in Figure 6.6. Starting from the lowest speed of 5 mm s$^{-1}$, it can be seen that the film thickness in all LP, and LM1 and LM2 greases falls from a value of $\approx 150$ nm to a minimum of $\approx 70$ nm with increasing speed. From here on, their film thickness starts increasing with speed with a slope which is close to 0.7 in the log-log plot, as predicted by the EHL theory for oils. LPE film thickness behaviour clearly diverges from this common trend. At 5 mm s$^{-1}$ LPE forms a film of $\approx 120$ nm. However, this decays very rapidly with increasing speed, reaching a minimum film thickness of 24 nm at a speed of only 25 mm s$^{-1}$, where the transition to the oil-like film thickness behaviour occurs. The corresponding transition in LP1, with the same base oil viscosity, occurs at a speed above 100 mm s$^{-1}$. 

Figure 6.5 Colour film thickness images of custom greases at different speeds
Grease friction in fully flooded conditions

The friction curves obtained with the custom greases are shown in Figure 6.7, Figure 6.8 and Figure 6.9 at 40°C, 60°C, and 80°C respectively. As found in the commercial lithium greases in Chapter 5, the greases generally display the previously mentioned ‘inverse-Stribeck’ behaviour. The curves ‘bump’, corresponding to the maximum value of friction reached, is particularly evident in all LP greases and LPE at 80°C in Figure 6.9. All the LP greases give very low friction at all speeds and temperatures. As found with their base oils, the friction coefficient values of these PAO-based greases at high speeds are very similar. Any trends in the relationship between base oil viscosity and grease friction are not immediately apparent, but these will be discussed in detail in Chapter 7. The mineral oil greases LM1 and LM2 give overall higher friction coefficients, especially at high speeds. As compared to the LP greases, LPE generally provides higher friction. While this result is confined to relatively low speeds at 40°C and 60°C, at 80°C this is evident across a broader range of speeds.
Figure 6.7 Friction coefficient of custom greases at 40 °C plotted against entrainment speed

Figure 6.8 Friction coefficient of custom greases at 60 °C plotted against entrainment speed
6.3.3 Inspection of grease deposited film

The friction tests carried out with LP1 and LPE at 40 °C were stopped in sequence at speeds of 20 mm s⁻¹, 50 mm s⁻¹, 100 mm s⁻¹, 200 mm s⁻¹, 500 mm s⁻¹ and 1000 mm s⁻¹. Pictures of the MTM discs were taken with an optical microscope in correspondence to the rolling track, to examine the morphology of the lubricant film deposited in the wake of the contact, on the half portion of the disc where the grease was just overrolled by the action of the ball, as shown in Figure 6.10. These pictures are overlaid at the appropriate speed points in Figure 6.11 and Figure 6.12, where the friction curves obtained with LP1 and LPE are compared with the corresponding curves of their base oils.
Plots and images in Figure 6.11 and Figure 6.12 suggest that the greases transit to their base oil-like behaviour at the same speed where the ‘bump’ in the grease friction curve occurs. It is from here on that, with increasing speed, grease friction starts to follow the same trend of its base oil. This speed of transition increases with temperature. These results are in line with the trends found for the commercial greases and their bled oils in Chapter 5. Moreover, an additional trend is evident in the LP greases: as for their film thickness results, a consistently lower transition speed in regards to friction was found with greases of higher base oil viscosity. The same observations were made in the comparison between the mineral oil-based greases LM1 and LM2. Eventually, when the oil viscosity (at the test temperature) is too high, this speed becomes too low to be identified within the speed range employed here.

With LP1 in Figure 6.11 (a), a thick uniform layer of lubricant is left in the rolling track at 20 mm s⁻¹. This is gradually removed as the speed is increased, until what is left in track at the maximum speed of 1000 mm s⁻¹ appears as a dispersion of oil drops, probably separated by thickener particles.
Figure 6.11 Friction coefficient of LP1 grease and base oil at (a) 40 °C, (b) 60 °C and (c) 80 °C plotted against entrainment speed; corresponding MTM disc micrographs (rolling track width ≈ 0.24 mm) at 40 °C are shown in (a).

The corresponding plots and pictures for LPE are shown in Figure 6.12. The speed of transition of LPE into its base oil-like behaviour is lower than in LP1, at all temperatures, despite the same value of viscosity of their base oils. As in LP1, the morphology of the deposited film changes as the speed is increased, from a grainy structure to a gradually fluid-like appearance, until only oil drops are found in the track at the maximum speed of 1000 mm s⁻¹. However, the deposited film at low speeds looks patchier and thinner than in LP1.
6.3.4 Effect of additives on grease and base oil friction

The influence of oleic acid friction modifier on the behaviour of custom greases was investigated by comparing the friction results of additised and non-additised base oils and greases at 80 °C, the test temperature at which the effect of oleic acid was the most apparent. These are plotted in Figure 6.13. The additised and non-additised oils are displayed in the left column (Figure 6.13 (a), (c), (e)). The corresponding greases are shown in the right column (Figure 6.13 (b), (d), (f)). The benefits of oleic acid are evident in the base oils at low speeds (in mixed lubrication conditions), where friction is reduced in the base oils of LP1, LPE and LM1. In contrast, the friction behaviour of additised and non-additised greases appears to be practically the same. The significance of this observation will be discussed in Chapter 7.
Figure 6.13 Friction coefficient of (a) LP1 base oil, (b) LP1 grease, (c) LPE base oil, (d) LPE grease, (e) LM1 base oil, (f) LM1 grease, with and without oleic acid, at 80 °C plotted against entrainment speed

6.3.5 Effect of mechanical degradation on grease friction

The friction results obtained with the mechanically worked and fresh LP1 grease are compared at all test temperatures in Figure 6.14. It should be noted that for practical reasons, the worked LP1 was prepared with an anti-wear additive (see Section 3.3.7). As found for the commercial grease based on lithium thickener and PAO oil, LiPAO grease in Section 5.6, no significant differences are apparent in the friction curves of custom-made LP1 before and after mechanical aging.
An Experimental Study into the Influence of Grease Composition on Friction in EHL Contacts

Figure 6.14 Friction coefficient of fresh and mechanically worked LP1 at (a) 40 °C, (b) 60 °C and (c) 80 °C plotted against entrainment speed

6.3.6 Grease friction in starved conditions

The results obtained with the LP greases under nominally starved conditions, i.e. without forced grease re-supply using the scoop, are presented in Figure 6.15. These tests were conducted at
fixed speeds of 50, 100, 200 and 500 mm \(s^{-1}\), for 100 minutes at each speed. However, they were stopped and immediately re-started every 20 minutes. The procedure is fully explained in Section 4.2.3.2. All the greases were found to severely starve after a certain time and/or speed. In the grease with the highest base oil viscosity, LP5, the friction coefficient gradually increases with time throughout the test at the minimum speed employed of 50 mm \(s^{-1}\). The halt of the ball and disc motion, and the load/unload sequence of the ball after 20 minutes from the start of the test do not help the recovery of the grease supply conditions, and the contact relentlessly starves until it runs dry. This condition corresponds to values of friction coefficient of 0.4 – 0.6, not shown in the figure for clarity. With greases of lower viscosity base oils, the speed at which severe starvation occurs increases to 100 mm \(s^{-1}\) for LP4 and LP3, 200 mm \(s^{-1}\) for LP2, and 500 mm \(s^{-1}\) for LP1: a sharp increase in friction can be noticed as soon as the tests are started at these speeds. The slope of the friction curve here is the same for all greases. However, it is possible that starvation may occur before these maximum speeds are reached, since these tests were not run at intermediate speeds.

Different lubrication phases can be recognised before severe starvation is reached. In LP4 it is possible to notice a gradual increase in friction (‘controlled starvation phase’) as the test is started. This is followed by a decrease in friction (‘recovery phase’) after about 10 minutes. The controlled phase of increasing friction starts again after 30 minutes, and lasts for the entire duration of the portion of the test conducted at the lowest speed of 50 mm \(s^{-1}\). In LP3 the recovery phase is also identified after 10 minutes, followed by a stabilisation (‘semi-starved steady phase’) of the friction coefficient. A phase of controlled increase in friction, followed by its stabilisation, is observed in LP2, both occurring at a higher speed of 100 mm \(s^{-1}\). Finally, in LP1 the friction coefficient remains relatively stable and low (‘low-friction steady phase’) at all speeds lower than 500 mm \(s^{-1}\), although a relatively large scatter in the friction results is observed at the lowest speed of 50 mm \(s^{-1}\). This stable phase terminates as soon as the maximum speed of 500 mm \(s^{-1}\) is applied, when friction is observed to abruptly increase (‘severe starvation phase’), as found with all other greases at lower speeds.
A starvation test was also carried out with LPE. The resulting curve is shown in Figure 6.16, together with the equivalent result for LP1. LPE gives higher friction than LP1 at 50 mm s$^{-1}$ and 100 mm s$^{-1}$. However, the severe starvation phase is reached at the same speed of 500 mm s$^{-1}$. In
the same figure, the result obtained with the additised LP1a grease is also shown. Almost identical friction behaviour was observed between LP1a and to its non-additised equivalent LP1, at 50 mm s\(^{-1}\) and 100 mm s\(^{-1}\). However, LP1a shows signs of controlled starvation already at 200 mm s\(^{-1}\), while the onset of severe starvation is observed at the same maximum speed of 500 mm s\(^{-1}\).

### 6.4 Summary

In this chapter the results of the experimental investigation on the tribological behaviour of custom-made greases were presented. This second part of the work was restricted to the study of lithium greases only, which were purposely manufactured to isolate single composition variables, specifically base oil viscosity, base oil type and presence of additives. In this way the influence of these particular variables on grease frictional behaviour could be investigated. The effect of mechanical aging of greases was also examined. The study also included a brief investigation into grease frictional behaviour under staved conditions. Most of the general trends which were briefly presented at the end of Chapter 5 with the commercial greases were confirmed in the investigation carried out with the custom greases. In the next chapter, the findings obtained with the two sets of greases are related in an attempt to identify and discuss some of the evident mechanisms of grease lubrication in general, and grease friction in particular.
Chapter 7. Discussion

The main trends and their significance in terms of mechanisms of grease lubrication and influence of grease composition on friction, as observed in the results chapters, will be discussed here. The first part of this study focussed on the frictional performance of fully-formulated commercial greases. This helped to identify the trends of interest. Subsequently, the second part of the study employed custom greases with controlled composition, designed to isolate the influence of particular aspects of grease composition, namely base oil type and viscosity, and presence of additives. The results obtained from these two sets of greases were complementary. This section discusses the implication of the observed trends and the physical mechanisms that may be responsible for them. Finally, potential formulations for a low-friction grease are discussed.

7.1 General trends

The results for both the commercial and custom greases have shown that the rules of grease friction behaviour in concentrated contacts differ according to grease composition, and strongly depend on the operating conditions. Despite this, the results point to the presence of some consistent general trends in grease behaviour.

7.1.1 Grease versus oil

The comparison between the experimental results obtained with the test greases and their corresponding bled or base oils provides evidence of the existence of two distinct operating regions in the tribological behaviour of greases in EHL contacts under fully flooded conditions, namely:

- ‘High speed’ region, where grease and its base oil follow the same trends in terms of film thickness and friction behaviour. In this region the standard EHL theories can be used to predict grease film formation and friction rules;
- ‘Low speed’ region, where grease gives a markedly different response than its base oil. In particular, greases generally form much thicker films than oils in this region. The thickness and friction behaviour of these films does not follow the classical EHL theory for oils, and is determined by the combined effect of thickener and oil type.

The same type of general behaviour has been observed before in other studies on grease lubrication\textsuperscript{72,74,97}. The speed at which the transition between these two regions occurs, defined as ‘transition speed’\textsuperscript{71}, has shown to be highly dependent on grease composition and contact
conditions, including temperature. In particular, higher temperatures have the effect of moving the speed of transition to higher values, and thus extending the range of speeds over which grease behaves differently from its base oil in the ‘low speed’ region. It is obvious that operating conditions other than just speed are relevant in this respect. Nevertheless, in discussing the experimental results the demarcation between these two operating regions is based on speed for convenience.

### 7.1.2 Grease film formation behaviour

The results obtained in Chapter 5 and Chapter 6 have revealed that the understanding of grease film formation mechanisms are of major importance in the interpretation of grease frictional behaviour in EHL contacts. It has been shown that, at sufficiently high speed, the log-log film thickness versus speed plots in both commercial greases (Figure 5.11, Figure 5.12 and Figure 5.13) and custom greases (Figure 6.6) follow a straight line, with a slope of approximately 0.7, in accordance with EHL theory. However, below the speed of transition mentioned above, greases film thickness behaviour completely diverges from that of base oils. In this low speed region grease generally forms thicker films, which also follow complex rules of formation that differ depending on grease composition. It was observed that with certain types of thickener, such as lithium (the thickener type on which this work was mostly focussed), film thickness increases with decreasing speed within this low-speed region, which is the exact opposite of what is predicted by the EHL theory for oils. This is clearly shown, for example, in Figure 6.6 for the custom greases, which were all fabricated using lithium. All these curves display, in fact, a v-shaped curve in the log-log plot, as already found in earlier studies on grease film thickness. A very unstable and non-uniform film is observed at these speeds, suggesting the passage of thick lumps of thickener/base oil, rather than the presence of a classical hydrodynamic film. This behaviour may be associated to either a residual, speed independent film deposited within the rolling track, a dynamic accumulation of thickener material entrained in the contact at low speed, or a combination of both.

Grease film thickness behaviour could be illustrated from a rheological perspective. For explanatory purposes, the Hamrock-Dowson EHL film thickness equation was applied to back-calculate the apparent viscosity of the grease at the inlet of the contact, from the film thickness measurements carried out with the custom greases. The values of pressure-viscosity coefficients of the base oils were used in these calculations, assuming they are the same for the greases. In Figure 7.1 the apparent viscosity is plotted against the maximum shear rate in the inlet of the contact, estimated as $3U/2h_c$ with $U$ the entrainment speed and $h_c$ the central film
thickness. In this way the film thickness measurements were turned into a rheological flow test, where the apparent viscosity is calculated at increasing values of estimated shear rate. The greases shear-thinning behaviour with increasing shear rates (at sufficiently low shear rates) is evident. The shear-thinning portion of the curve for each grease corresponds to the ‘low speed’ region described above. Here, all lithium-PAO (LP) greases have approximately the same apparent viscosity for a given shear rate, and thus grease apparent viscosity here appears to be independent of base oil viscosity. Therefore, by modelling grease with EHL rules in which a shear-thinning apparent viscosity is used in place of a Newtonian value, it is seen that for a given base oil type, it is the thickener that controls film formation. The finding is in line with published results of rheological measurements carried out on greases formulated with base oils of different viscosity, but the same amount of lithium thickener. Eventually, a Newtonian plateau is reached, where the apparent viscosity becomes approximatively constant, and where its value is now dependent on base oil viscosity. This is where grease starts operating in the ‘high speed’ region, since here its film thickness behaviour becomes analogous to that of a Newtonian base oil.

Figure 7.1 Apparent viscosity (calculated from measured film thickness values) of custom greases at ambient temperature (22 ± 1 °C) plotted against estimated inlet shear rate
7.1.3 Grease frictional behaviour

The majority of test greases displayed a very complex frictional behaviour. The base or bled oil friction results, as well as the grease film thickness results, are of major importance in supporting the interpretation of grease friction. With the final aim of understanding if grease film forming behaviour could be used to partly explain friction in EHL contacts, an attempt was made to directly correlate the friction and film thickness results. These measurements were taken on separate instruments with different slide/roll ratios, materials and loads, and therefore are not strictly comparable. However, the presence of moderate slide/roll ratio is known to have negligible influence on grease film thickness in EHL contacts under fully flooded conditions. Moreover, in a series of preliminary tests, it was observed that pressure does not seem to greatly affect the observed general trends. It is therefore believed that the influences of differing load and material elastic properties on film thickness can be quantified using the EHL theory. The differences are minimal, but have nevertheless been accounted for by using the Hamrock-Dowson EHL film thickness equation, leading to the following relationship:

$$\frac{h_c_{MTM}}{h_c_{EHD}} = \left( \frac{E'_{EHD}}{E'_{MTM}} \right)^{0.073} \left( \frac{W_{EHD}}{W_{MTM}} \right)^{0.067} \approx 0.92$$

where $h_c$ is the central film thickness, $E'$ the reduced Young’s modulus, $W$ the load, and the suffixes MTM and EHD refer to the friction and film thickness tests on the MTM and EHD rig respectively. This factor was used to project the film thickness measurements from the EHD rig onto the MTM rig, making it possible to create single plots which directly relate the film thickness and friction trends.

As an example, the measured values of film thickness and friction for the commercial CaSMix grease are both plotted against the entrainment speed in Figure 7.2, together with the friction result obtained with its bled oil and the prediction of its base oil film thickness. Presumably, base and bled oil have equivalent behaviour, and therefore the bled oil friction curve is simply referred to as ‘base oil friction’ in the figure. The two dashed horizontal lines mark the approximate boundaries between the three nominal lubrication regimes of the base oil: full film ($\Lambda \geq 3$), mixed ($1 < \Lambda < 3$) and boundary lubrication ($\Lambda \leq 1$). Clear parallels between friction and film thickness behaviour are shown in the figure. Both friction and film thickness curves for grease and base oil start overlapping at the same entrainment speed of about 200 mm s$^{-1}$. CaSMix grease forms a film that is thick enough for the surfaces to be fully separated even at the lowest speeds,
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unlike its base oil. However, the region where grease produces this relatively thick film coincides with the region where grease gives significantly higher friction than the base oil.

A similar plot for the commercial LiPAO grease is shown in Figure 7.3. LiPAO film thickness is again high enough, even at low speeds and despite the very low viscosity of its base oil, to keep the contact in the full film regime throughout the whole speed range. However, unlike CaSMix grease, the abnormally thick grease film at low speeds results in a friction coefficient that is much lower than that of the bled oil (again, referred to as ‘base oil friction’ in the figure) within the same low speed range. These results point to the fact that factors other than simple $\Lambda$ ratio govern grease friction at low speeds, and that the specific grease composition is very important in this respect.

**Figure 7.2 Correlation of friction and film thickness for CaSMix and its base oil at 40 °C**
All the studied lithium greases display the same ‘inverse-Stribeck’ behaviour of LiPAO in Figure 7.3. Indeed, below a certain speed of transition, as the grease film thickness starts to increase with decreasing speed (resulting in a v-shaped curve in the log-log plot), friction correspondingly decreases. This is the exact inverse of what was observed with the corresponding bled or base oil, where friction increases with decreasing speed, due to a drop in film thickness and the resulting greater number of asperity contacts in the mixed and boundary lubrication regime. In this work the speed of transition in grease friction has usually been observed to appear where the corresponding bled or base oil operates under conditions of mixed lubrication, i.e. where oil friction is still decreasing with increasing speed. As a result, the lithium greases friction curves display a ‘bump’ at approximately this speed. Indeed, starting from the lowest speed and moving to higher values, friction in these greases is seen to gradually increase and, after reaching a maximum value (i.e. the curve ‘bump’), it begins to decrease.

It is clear that these characteristic low-speed thick films are responsible for a likewise peculiar behaviour in friction, which in turn is strongly dependent on grease formulation. Because of the existence of these clearly distinct tribological operating regions of grease, at high and low speed, the corresponding effects of grease composition on friction are discussed in detail separately, in Section 7.2 and 7.3 respectively.
7.2 Effect of grease composition in the high speed region

In the high speed region, it is the base oil characteristics that were found to be the dominant influencing factor on grease friction, while no obvious influence of thickener type was observed under these conditions. Therefore only the effect of base oil characteristics on friction is discussed in this section.

7.2.1 Influence of base oil viscosity

Base oil viscosity on its own is not considered to be a good indicator for the estimation of grease friction in EHL lubrication. As Figure 5.7 suggests, no correlation exists between grease EHL friction and the kinematic viscosity of the base oil, when greases containing very different base oil types (as in this case for the commercial greases tested in this work) are compared. The experimental study conducted with the custom LP greases, presented in Chapter 6, allowed to better assess the effect of base oil viscosity on grease friction, by enabling a comparison between greases made with the same base oil type. In this way the additional effect of base oil type, which is instead an important variable of influence, could be effectively excluded. The LP greases friction curves obtained at the three test temperatures, and shown in Figure 6.7 - Figure 6.9, do not provide evidence of clear trends in the relationship between friction and base oil viscosity. However, since the base oil viscosity is known to highly affect film thickness in the first place, an attempt was made to capture the effect of film thickness in these measurements. The same curves are plotted against the nominal $\Lambda$ ratio (calculated as the ratio of central film thickness of the base oil to composite $R_q$ roughness) in Figure 7.4, Figure 7.5 and Figure 7.6 at the three test temperatures. For simplicity purposes, the central film thickness was estimated using the Hamrock-Dowson EHL film thickness equation based on the viscosity and pressure-viscosity $\alpha$ of the base oils, with $\alpha$ taken from Table 6.1 and the base oil viscosity as measured at the temperature of each test. Figure 7.4 - Figure 7.6 are able to describe the tribological behaviour of the greases relatively well. By adopting this change of abscissa, it is possible to notice that all LP greases friction values are similar at a given value of $\Lambda$, and that the curves form a common ‘master’ inverse-Stribeck-like curve. The value of $\Lambda$ in correspondence to the ‘bump’ of these curves is approximately the same in all LP greases and at any temperature. Most of the differences in friction between the LP greases are observed at values of $\Lambda$ close to the position of the ‘bump’, especially at high temperature (Figure 7.6). Here, friction might be generated by the complex interaction occurring between the grease components (base oil and thickener), and the surface asperities of the contact. However, these differences are marginal.
In contrast, with the mineral oil-based greases, i.e. LM1 and LM2, the values of friction still appear very different even when compared at the same nominal $\Lambda$. However, unlike the PAO base oils, the mineral oils of LM1 and LM2 might have very different molecular structures as already pointed out in Chapter 3.

Figure 7.4 Friction coefficient of custom greases at 40 °C plotted against lambda ratio $\Lambda$
It should be noted that the actual values of $\Lambda$ are likely to be higher than the nominal ones shown in the plots presented above, at both low and high speed. At low speed (and therefore in correspondence to values of friction to the left of the ‘bump’ in the LP greases curves) the actual
trend for $\Lambda$ should be the inverse of what is displayed. Indeed, according to the film thickness measurements shown in Figure 6.6 for these greases, here $\Lambda$ should increase with decreasing speed, as opposed to the predicted and plotted base oil $\Lambda$.

However, focussing on the ‘high speed’ region (at values of friction to the right of the curves ‘bump’), a simple calculation was carried out for these greases to estimate the relative increase in effective lambda compared to the nominal $\Lambda$ shown in the plot. As shown in Figure 7.1, at high values of estimated inlet shear rate, the apparent viscosity curve for these greases appears relatively flat. These values correspond to the high speed operating region, and indicate that at these speeds the behaviour of the grease in the contact inlet is approximately Newtonian. Some studies\textsuperscript{63} suggest that under these conditions the lubricating film reduces to a suspension of thickener particles in base oil, which thicken the base oil and lead to an increased effective Newtonian viscosity. A rough approximation for this apparent Newtonian viscosity for each grease can be calculated as an average of all apparent viscosity values within the Newtonian plateau shown in Figure 7.1. The differences between the calculated Newtonian apparent viscosity of the greases and the viscosity of their corresponding base oils (at the test temperature) is plotted in Figure 7.7. All the greases display higher values of Newtonian viscosity when compared to their base oils. The absolute difference in viscosity between the LP greases and their base oils increases with higher base oil viscosity. The percentage increase in viscosity goes from a minimum of 23.5 % for LP3 to a maximum of 56.3 % for LP5. Consequently, the greases friction curves within the high-speed region of Figure 7.4 - Figure 7.6 should probably be shifted to the right by the respective factors (at the test conditions), due to an enhanced film thickness through an increase in apparent viscosity.
Figure 7.7 Greases versus base oils absolute and percentage apparent kinematic viscosity increase

It is interesting to compare these estimated increases in apparent viscosity with some equivalent predictions using formulas available in the literature. A modified version\textsuperscript{62} of the original Einstein’s formula for suspensions\textsuperscript{147}, already suggested to solve the film thickness problem in grease lubrication\textsuperscript{148}, predicts that:

$$
\nu_G = \nu_{BO} \times \left(1 - \frac{5}{2} \varphi\right)^{-1}
$$

where $\nu_G$ is the kinematic viscosity of the suspension (i.e. the apparent kinematic viscosity of the grease), $\nu_{BO}$ is the kinematic viscosity of the base oil, and $\varphi$ is the volume fraction of the suspended thickener particles. Given that $\varphi$ is the same in all custom greases (as they were all made with the same w/w % of thickener), this formula predicts a constant percentage increase in kinematic viscosity of grease in relation to the viscosity of the base oil. This is in contrast with the estimated percentage increase in grease viscosity shown in Figure 7.7, which varies depending on the viscosity of the base oil. However, assuming (within an error of 10 %\textsuperscript{59}) that the thickener volume fraction equals its weight fraction, the formula predicts a percentage increase in viscosity (at the temperature of the film thickness tests) of 42.8 % for all LP greases. This prediction is very close to the overall average value of viscosity increase of 42.5 % in the LP greases, calculated using the approximated values shown in Figure 7.7.

In summary, within the high speed region a correlation between base oil viscosity and grease friction was found for greases made with the same base oil type, i.e. PAO in this study. Grease
friction seems to be reasonably well correlated with the estimated lambda ratio, with the friction curves for all LP greases falling on the same master curve at a given test temperature, in line with a classical Strubeck relationship. Accordingly, this result would suggest that the higher is the viscosity of the base oil, the thicker is the lubricating film at a given speed, and thus the higher is friction in this high speed region.

However, it should be noted that friction in this high speed EHL-like region cannot be entirely explained through lambda ratio only. The effect of operating conditions should also be considered, in particular the influence of temperature. Figure 7.8 displays the same friction curves plotted against nominal lambda ratio, where the results obtained at the three test temperatures are compared for each grease. It is observed that high-speed friction is always lower at higher test temperatures for a given lambda ratio. The same observations were made for all the lubricant samples tested in this study, including the bled and base oils. This may intuitively be expected to arise from the thermal effects on the EHL traction coefficient, given that at these high speeds the greases, like their base oils, appear to follow the standard EHL theory. Accordingly, film thickness is mainly determined by the value of viscosity at the inlet, and EHL friction by the physical properties of the lubricant within the contact, which are strongly affected by the in-contact temperature.
Figure 7.8 Friction coefficient of custom greases at 40 °C (blue markers), 60 °C (red markers) and 80 °C (green markers) plotted against lambda ratio Λ.
7.2.2 Influence of base oil type

In regards to the absolute value of friction in EHL contacts, the overall results have shown that, in the high speed region, the type of base oil employed in the grease formulation is one of the most influential aspects. As shown in Figure 5.7 with the commercial greases, much lower friction coefficients were found with greases formulated with synthetic base oils, i.e. PAO and ester in the current study. This was confirmed by the investigation on the custom greases, which compared results obtained with greases made with base oils of similar viscosity, but different type, so as to isolate the effect of base oil type. Comparing LP1 with LM1 (PAO and mineral oil-based greases respectively with similar base oil viscosities), or equivalently LP3 with LM2 (ditto), the mineral base oil greases systematically display much higher friction, even at the same nominal lambda ratio (Figure 7.4 - Figure 7.6). This result is compatible with earlier work on EHL friction in oil-lubricated contacts\textsuperscript{47,39}. In particular, the literature reports that low friction is often achieved with synthetic oils of low pressure-viscosity\textsuperscript{39} and high viscosity index\textsuperscript{149,150}. Therefore, an attempt was made to correlate the estimated pressure-viscosity and viscosity-temperature properties of the commercial greases base oils (estimated according to the procedure described in Section 3.2.2.2) with grease friction, within this high speed region where grease appears to follow EHL theory. Considering the wide differences between the base oils of these greases, a relatively strong relationship was found between grease friction and these base oil properties, as shown in Figure 7.9. The relationship between friction and pressure-viscosity is nearly linear, with only LiMi and CaSMix diverging with higher values of friction. In addition, an approximate inverse proportionality was found between these friction results and the calculated viscosity index $V/I$ values. The explanation for this finding can possibly lie in the molecular origins (particularly molecular rigidity and shape\textsuperscript{39}) of EHL friction, and is beyond the scope of this study.
An Experimental Study into the Influence of Grease Composition on Friction in EHL Contacts

Figure 7.9 Friction coefficient of commercial greases at 1000 mm s⁻¹ and 40 °C plotted against (a) estimated pressure-viscosity coefficient and (b) estimated viscosity index of base oils

It should be noted that although a ‘low friction’ oil type may not ensure low friction under all operating conditions, and low speeds in particular as discussed below, reduced friction at high speeds can provide the most significant contribution to the reduction of overall machine power losses. Indeed, the two low-friction commercial greases tested here, LiEs and LiPAO, are made with very low viscosity synthetic base oils.
7.3 Effect of grease composition in the low speed region

In the low speed region, thickener and base oil type were recognised as the main variables of influence on grease friction. These variables not only affect friction behaviour, but also the range of speeds in which the grease operates in this low-speed region. In this respect the base oil viscosity is also of major importance.

7.3.1 Influence of thickener type

As described above, all the test greases were able to form thick films in the low speed region. However, the results suggest that in addition to any influence of \( A \) ratio, friction is governed by the actual nature of these thick films. The friction behaviour at these speeds is shown to strongly depend on thickener type. Based on this, the tested range of greases can be broadly divided into two groups: lithium and polyurea greases, and calcium sulphonate complex greases. The differences between the two groups of greases in terms of friction and film build-up mechanism are apparent (see, for example, Figure 7.2 and Figure 7.3). All the lithium and polyurea greases tested in this work displayed a v-shaped film thickness curve: below the transition speed their film thickness increases with decreasing speed. These thick films also produce lower friction than in the corresponding base oils, resulting in ‘inverse-Stribeck’ behaviour. In contrast, in the calcium sulphonate complex greases, thinning films were observed with a reduction of entrainment speed, although this decrease of film thickness occurs at a lower rate than in base oils. However, the most striking difference is that the calcium sulphonate complex greases provide higher friction than their bled oils in this region. This was very apparent in CaSMix at 80 °C, probably as a result of its high thickener content (Table 3.3). Therefore, even if this grease is able to move the contact from mixed to full film lubrication regimes as compared to its base oil (see Figure 7.2), friction may actually increase. The nature of these films is not yet fully understood, but some of the basic properties of significance might include roughness, morphology, viscosity and mechanical and chemical properties. However, since it is known that the morphological microstructure of lithium and calcium sulphonate complex greases is very different, it is perhaps not entirely surprising that these two sets of greases give dissimilar friction results. Lithium thickener is described as typically consisting of entangled fibres\(^{130,131}\), while calcium sulphonate complex thickener is a fine dispersion of spherical and agglomerated particles\(^{131}\). If thickener material, in the form of particles, is entrained or deposited on the contact surfaces, the friction coefficient at low speed, when little oil is available inside the contact, may be expected to in some way depend on thickener morphology. Evidence of the presence of hard
thicker particles in a calcium sulphonate grease, damaging the lubricated contact surfaces, was found in an earlier study\textsuperscript{4}.

The fact that the presence of lithium or polyurea thickeners provide protection for the surfaces at low speeds, accompanied by a reduction in friction, was further confirmed by friction measurements carried out with the commercial greases using discs of higher roughness. The friction results presented in Figure 5.14 are re-plotted in Figure 7.10, here as a function of predicted lambda ratio $\Lambda$, which was again calculated based on the properties of the base oils. Once more, as shown for the custom greases in Figure 7.4 - Figure 7.6, with this change of abscissa the friction curves overlap for a limited range of nominal lambda ratio values. In particular, Figure 7.10 suggests the existence of a common curve, which is representatively drawn for every grease as a black dotted line, and has the appearance of a classical Stribeck curve for an oil. All the measured values of friction coefficient that fall below this curve correspond to real $\Lambda$ values that are higher than the nominal ones, hence giving lower friction in all the lithium and polyurea greases. The very low value of friction found in LiPAO with the rough disc at the lowest speeds suggests that this grease is capable of forming extremely thick film, considering that the composite roughness $R_q$ is 270 nm with this disc. Above these values of speed, friction is seen to rapidly increase, most probably due to the progressive removal of this film, and thus to an increasing contribution of the asperity contacts on friction.
7.3.2 Influence of base oil viscosity

Base oil viscosity is seen to have negligible influence on the greases friction coefficient within the low speed region, when the results are compared at equivalent lambda ratio values (Figure 7.4 - Figure 7.6). However, the base oil viscosity is observed to highly influence the range of speeds over which grease operates in the ‘low speed’ region. In particular, the lower is the base oil...
viscosity at the test temperature, the higher is the speed of transition between the low and high speed regions. This is particularly evident in the friction plots that compare the commercial greases with their bled oils (Figure 5.9 and Figure 5.10) and the custom greases with their base oils (Figure 6.11 and Figure 6.12) at different temperatures, and is further confirmed by the film thickness results of the LP greases (Figure 6.6), which were manufactured with base oils of different viscosity. The possible reasons for this behaviour are presented in Section 7.5, which discusses in more detail the general trends observed in the relationship between grease composition and the ‘transition speed’.

7.3.3 Influence of base oil type

The base oil type has been seen to play a significant role in grease friction even at low speeds. Looking at Figure 7.4 - Figure 7.6, it is seen that also in the low speed region (to the left of the greases ‘bump’) the LP greases friction measurements collapse into a single curve at the same nominal Λ. However, the corresponding friction values measured with the ester-based LPE grease, and also with the mineral oil-based LM1 and LM2 greases, are higher. Alternative explanations can be given for the results obtained with LPE, and with LM1 and LM2. As shown in Figure 6.6, LPE forms much thinner films at low speeds than LP1, despite their very similar composition in all respects other than the presence of a small percentage of ester oil in the formulation of LPE. In the comparison between Figure 5.11 and Figure 5.12, a similar result can also be observed in the study on the commercial LiPAO and LiEs greases. LiPAO and LiEs are both lithium greases, but made of PAO and ester oils (of very similar viscosity) respectively. Much higher friction was also observed in LiEs at low speeds (Figure 5.1-Figure 5.3 and Figure 5.14). This means that the same type of result, that of thinner films in greases containing ester oil, and consequently higher friction, was consistently observed. The SEM analysis of the thickener microstructure of these two sets of commercial and custom greases provides some further insight into the reason for this different behaviour observed in greases formulated with ester oils. The pictures of the thickener (prepared as outlined in Chapter 3) of LP1, LPE, LiPAO and LiEs at a magnification of 10k X are compared in Figure 7.11. It is evident that the lithium fibres displayed by the PAO-based greases (LP1 and LiPAO) are much thicker and longer than the ones observed in the ester-based LPE and LiEs. Smaller lithium particles have already been observed in earlier papers30,107 in ester-based greases. Published literature suggests that shorter lithium thickener fibres produce thinner films at low speeds and, as a consequence, higher bearing torques154. These results suggest that the presence of ester oil in greases might have compromised the development of long lithium thickener fibres during the
manufacturing process. In turn, this thinner microstructure is responsible for the diminished film enhancement at low speeds, and consequent higher friction due to asperity contact, observed in these greases. This result is further supported by the differences observed in the comparison between the deposited grease film in LP1 (Figure 6.11) and LPE (Figure 6.12) during the friction tests.

As opposed to LPE, the mineral oil-based LM1 and LM2 greases film thickness behaviour at low speed is equivalent to the one found in the LP greases (Figure 6.6). The higher friction observed in these greases could be simply due to the fact that at low speeds the thick lubricating film is still made, to a large extent, of base oil. It could be expected, therefore, that in this case the observed trends in the relationship between base oil type and grease friction are equivalent to the ones presented in Section 7.2.2 at high speeds: friction remains lower in the LP greases, followed by LM1 and finally LM2. These findings suggest that the ‘low-friction’ feature of the lithium thickener, at least in these conditions, is simply due to its capacity of producing thick films at low speeds, thus limiting or excluding metal-metal contact, and without generating significant friction itself (as found, in contrast, with the calcium sulphonate thickener).
7.4 Influence of additives

An interesting consequence of the thick low-speed grease films is its significance in terms of action of additives in the grease formulation. Under the employed test conditions, oleic acid did not show any influence on the greases friction behaviour. In contrast, oleic acid did show a reduction in friction at low speeds when added to the base oils, in the mixed lubrication regime where metal-to-metal contact is expected to occur (Figure 6.13). The most plausible reason for the ineffectiveness of oleic acid in the grease formulation is that at these low speeds, where oleic acid would normally be expected to act, the thick film formed by grease already provides low friction due to much higher $\Lambda$ values than with oils, rendering the oleic acid redundant. To the best of author’s knowledge, this study shows for the first time that some friction modifiers, in combination with certain grease formulations, might be irrelevant for friction reduction. Obviously, these results do not exclude the influence of oleic acid on friction in different contact conditions, or of other friction modifier additives. However, they do suggest that under certain conditions, additive-free grease formulations may be just as acceptable as their fully-formulated, and more expensive, equivalents.

7.5 Transition between low speed and high speed regions

The ‘transition speed’, i.e. the speed which marks the two regions of lubrication described above, has been mentioned several times in the course of this work. The friction results obtained with the de-oiled LiPAO grease at the three test temperatures (‘thickener’ in Figure 5.9) provides some evidence for its significance. This lubricant sample appeared to follow the same behaviour of the original grease at low speeds, but only up to the ‘transition speed’. As soon as this speed was exceeded, friction was observed to abruptly increase, most likely due to the onset of starvation. The result indicates that, in correspondence to this speed, the thickener is removed from the contact, or not entrained anymore. This is the point where, as observed with all test greases studied in this work, oil takes over. However, due to little oil available in the contact inlet when this ‘thickener’ lubricant sample was used, the contact consequently starves. All the results show that, for a given grease, this transition speed increases with temperature and with a reduction in base oil viscosity. In terms of the potential temperature effects, the literature reports different explanations, including that higher temperatures would produce:

- An increased growth of thickener material layers onto the surfaces, due to favourable thermal conditions \(^{73}\);
• A loss in consistency, claimed to improve the feeding mechanism of grease\textsuperscript{74};
• A reduction of shear stress in the contact inlet (due to a reduction in viscosity with temperature), allowing the grease thickener lumps to survive and be entrained up to higher speeds\textsuperscript{82}.

However, Figure 7.4 - Figure 7.6, where the friction results for the custom greases have been plotted against the corresponding base oils lambda ratio $\Lambda$ (therefore accounting for the effect of both speed and temperature on film thickness), clearly suggest that this speed is not strictly related to temperature, but to its effect on viscosity and, in turn, film thickness. This result indicates that the focus on the sole influence of temperature on the transition speed is somewhat futile. Indeed the transition speed of the LP greases, approximately corresponding to the curves ‘bump’, is approximately found at the same value of $\Lambda$, at all temperatures. Furthermore, Figure 6.6 clearly shows that all LP greases transit between the two operating regions of low and high speed at the same value of minimum film thickness, and that therefore it is the film thickness that determines the value of transition speed, rather than speed itself. These observations indicate that, for a given thickener and base oil type, and surface characteristics (e.g. roughness), the transition occurs at the same value of (minimum) film thickness. This ‘transition film thickness’ is a more universal parameter than the ‘transition speed’, as it encompasses the effect of speed, viscosity and temperature. This characteristic film thickness value might be expected to be related to the size of lithium thickener particles. Indeed much lower ‘transition film thickness’ values (and consequently ‘transition speed’ values) were found in the commercial and custom ester-based greases, made of much smaller lithium thickener particles as discussed above. Moreover, approximately the same ‘transition film thickness’ value was displayed by the LP greases and the mineral oil-based LM1 and LM2 greases, which have a very similar thickener microstructure as shown in Figure 3.11 and Figure 3.12.

In light of these observations, Figure 7.12 attempts to summarise the overall findings made on the influence of base oil type and viscosity in the custom greases. Here the observed trends are illustrated through descriptive plots, in which film thickness and friction are mutually related as a function of speed. The dashed lines are base oil film thickness and friction curves. The corresponding behaviour for the greases is represented by the continuous lines. In greases made with the same thickener (lithium) and base oil type, but with base oil of different viscosities (Figure 7.12 (a)), the transition to the classical EHL region occurs at the same value of ‘transition film thickness’ (b). The differences in friction at all speeds can be accounted for through film thickness, as the results would merge into a common master curve if plotted against $\Lambda$. On the
other hand, the differences in friction between lithium greases formulated with different base oil types, but base oils of the same viscosity (Figure 7.12 (b)) can be a result of either:

- Different film formation behaviour at low speeds (as in the comparison between grease A and grease C), with a more significant friction contribution given by asperity interaction with grease A; or
- Different friction response of the base oils, at both high and low speeds (as in the comparison between grease B and grease C).

![Figure 7.12 Schematic of film thickness and friction behaviour of grease as a function of (a) base oil viscosity and (b) base oil type](image)

### 7.6 Influence of mechanical degradation of greases

As the tribological behaviour of greases might change after many working hours of operation in a bearing, the result of mechanical degradation on friction was investigated on a selection of test greases. The results obtained with the commercial greases (Figure 5.15, Figure 5.16) suggest that the effect of prolonged mechanical working of grease is to generally annul (totally or partially) the enhancing effect of thickener to film formation, and therefore its influence on
friction, at low speeds. With this loss of thickener effects, the mechanically worked greases behave more similarly to their base oils, which results into an increase in friction in the lithium greases, and a decrease in the calcium sulphonate complex greases, as compared to their original behaviour in their fresh state. However, the notable exception to this are the lithium-thickened PAO greases (both the commercial LiPAO and custom LP1 greases), whose film formation and friction behaviour do not seem to be much affected by mechanical degradation. The reason for this finding is not clear, but it could be due to the non-polar nature of the PAO oil, allowing an abundant entrainment or deposition of thickener fibres even after they have been mechanically degraded. In any case, these results show that by choosing an appropriate formulation for the grease, the original tribological characteristics of fresh grease, including its capability to provide low friction and surface protection, can be preserved over long terms of operation in a bearing.

7.7 Effect of grease composition in starved conditions

The results obtained in the starvation tests suggest that the natural supply conditions in grease lubrication are affected by a complex set of interdependent variables, including lubricant distribution, time, speed, and the dynamic nature of the lubricating film. Therefore it is very difficult to assess the way in which they individually influence the lubricant supply to the contact inlet. Grease lubrication under starvation conditions is a complex topic, and its proper treatment is beyond the scope of this study. These results were used to assess the performance of the custom greases in presence of different conditions of lubricant supply than fully flooded, and verify whether some of the trends observed in fully flooded conditions could be confirmed also in starved conditions. The aim was to simulate more closely the behaviour of the greases in a real bearing, where no mechanism of forced grease supply exists. However, it should be noted that the real conditions of operation of grease in a bearing are still rather different from this ball-on-disc starvation test, due to the presence of many factors that change the grease distribution and its supply to the rolling contacts\textsuperscript{152}. Nevertheless, a basic attempt can be made to interpret the observed results. Several friction stages were identified under the nominally starved conditions (Figure 6.15 and Figure 6.16):

- in the ‘severe starvation phase’, speed is too high to either retain the lubricant layer in the rolling track, or allow enough time for the bled oil to reflow back to the contact and feed it\textsuperscript{58}. This condition leads to a rapid loss mechanism that soon makes the contact relatively dry. In accordance with literature data\textsuperscript{82-20}, this occurs at higher speeds with greases of
lower base oil viscosity. The maximum speed at which this phase was found is 500 mm s\(^{-1}\) for the LP1 grease. This might seem a low value in comparison to typical real bearing speeds. However, it is important to consider that, despite the general absence of an external mechanism that forcibly pushes the grease into the rolling contacts of a bearing (i.e. the lack of an equivalent for the ‘grease scoop’ used in the fully flooded tests), the supply conditions, nevertheless, could be much less severe than in the ball-on-disc test employed in this study. The availability of lubricant in bearing contacts can be enhanced by a number of mechanisms, induced by the presence of a generally different contact geometry, the cage and the occurrence of vibration\(^{77}\);

- The ‘controlled starvation phase’ suggests a monotonic thinning of the lubricant film. Since this occurs at constant speed, it is likely to be due to the displacement and removal of the deposited lubricant film by the overrolling action of the ball;
- In the ‘recovery phase’ a decrease in friction can be associated with replenishment. This is likely to be in the form of either bled oil released from the grease, made available from the mechanical degradation of the grease which creates more mobile lubricant\(^{108}\), or bulk grease resupply;
- In the ‘semi-starved steady phase’ a stable friction value is obtained. However, this value of friction is evidently higher than in fully flooded conditions, suggesting mixed lubrication conditions. This phase is likely to be governed by the presence of a constant film thickness, which could be due to the formation of a static layer of lubricant deposited on the rolling track, a stabilised rate of reflow of bled oil from the sides of the contact, or their combined effect\(^3\);
- The ‘low-friction steady phase’ indicates the presence of a film which is sufficiently thick to completely separate the surfaces, and thus to provide friction values comparable to the ones obtained in fully flooded conditions. Therefore in this case the supply conditions are likely to be equivalent to the fully flooded case.

As previously found in earlier studies\(^88\), it is seen that a wide range of supply conditions exists between the extremes of fully flooded and severe starvation, probably encompassing replenishment mechanisms through bulk reflow and oil bleeding.

A starvation test was carried out also with LPE, since a remarkable difference in the fully flooded friction and film thickness behaviour was observed between this grease and the LP greases. In line with the results seen in the fully flooded tests, LPE gives higher friction than LP1 at the lowest speeds employed in the starvation test, i.e. 50 mm s\(^{-1}\) and 100 mm s\(^{-1}\). It was observed
that, in fully flooded conditions, the friction behaviour of the base oils contained in LP1 and LPE is very similar. This suggests that, in starved conditions, the lubricant supply and consequent friction behaviour can be either driven by oil reflow, or by the low-speed thick films. In particular, oil reflow may be the main mechanism in LPE at 50 mm s\(^{-1}\) and 100 mm s\(^{-1}\), as this grease has already transited to the ‘high speed’ region at these speeds. Conversely, the lower friction observed in LP1 indicates that this transition has not occurred yet. The low-speed film formed in LP1 was retained in the rolling track for the relatively long duration of the test, and consequently provided lower friction. Oil reflow may be the leading mechanism responsible for the supply conditions and friction behaviour of both greases at 200 mm s\(^{-1}\) and 500 mm s\(^{-1}\), where both have transited to the ‘high speed’ operating region. Indeed at these speeds the behaviour of LP1 and LPE is practically the same.

While in fully flooded conditions the greases friction coefficient did not appear to be affected by oleic acid, under starved conditions oleic acid is seen to reduce the speed at which starvation occurs. With limited lubricant supply the oleic acid might encounter more favourable conditions for interacting with the metal surfaces, with consequent formation of a low-shear strength boundary layer at the contact interfaces. It has been reported that additives can be particularly reactive in starved conditions, and that they might limit the deposition of thickener in the track.\(^{108,76}\) This might possibly explain the earlier starvation observed in the additised grease.

### 7.8 Low-friction grease formulation

Given the wide range of greases tested here, current results indicate what compositions might be suitable for a low-friction grease. Based on present findings, low friction in the ‘high speed’ region can be achieved by using synthetic base oils with low EHL traction coefficient. For most machines, this ‘high speed’ region is the one that largely determines the overall power losses, so ensuring low friction here is of primary concern. The ‘low speed’ region may be less important in terms of the overall energy losses, but the grease here must ensure that lubricant films are thick enough to protect the surfaces from coming into direct contact. This is particularly important when using low-viscosity base oils which, without correct choice of other grease components, may quickly result in mixed/boundary lubrication as speed is reduced. Many greases are able to form such thick films, but lithium and polyurea greases seem to offer an additional benefit of low friction even at these low speeds.

In particular, a potential good formulation for an energy efficient grease was found in greases formulated with lithium and PAO. This composition provides low friction:
• In the ‘high speed’ region, due to a low-viscosity synthetic-type (PAO) base oil;
• In the ‘low speed’ region, due to the combined effect of PAO base oil and lithium thickener, which provides both thick and low-friction films.

Moreover, this specific formulation was found to be the only one, within the tested grease samples, to maintain the low friction characteristics even after severe mechanical degradation. This feature could potentially indicate a long grease life in certain bearing applications as an important additional advantage to low friction.

The friction curves of the custom LP1 grease and the commercially available LiPAO grease (marketed as a low-friction grease), both lithium-thickened low-viscosity PAO greases, are plotted together for comparison in Figure 7.13. The curves are very similar, with LP1 giving slightly higher friction, but only at low speeds and high temperatures. This result shows that, although the commercial LiPAO grease is very likely to contain other components in its formulation, its low friction performance is primarily determined by the basic formulation (base oil and thickener) used for the fabrication of LP1.
Figure 7.13. Friction coefficient of custom-made LP1 and commercially available LiPAO greases (both lithium-thickened low viscosity PAO greases) at (a) 40 °C, (b) 60 °C and (c) 80 °C plotted against entrainment speed.
Chapter 8. General conclusions

This chapter summarises the most important findings of this study. Some suggestions on potential future work, which could provide further understanding of the topic, are also given at the end of the chapter.

8.1 Summary

This research investigated the frictional behaviour of lubricating greases in rolling-sliding concentrated contacts pertinent to rolling bearings. Specific objectives of this research were:

- To improve the understanding of grease tribological behaviour in EHL contacts;
- To correlate the observed frictional response with grease chemical formulation;
- To identify potential formulations for low-friction bearing greases.

The significance of this research lies in the fact that almost all engineering systems employ multiple rolling bearings, the vast majority of which are lubricated with grease. It is recognised that optimised bearing lubrication through the choice of a suitable grease can significantly reduce friction, so that a provision of low-friction bearing grease is one of the most pervasive ways of improving the overall efficiency of mechanical systems. The primary function of grease in bearings is to lubricate the concentrated contacts between the rolling elements and the raceways, where an important contribution to the overall bearing friction is generated. Therefore, the current investigation on the relationship between grease composition and friction behaviour was conducted under conditions pertinent to typical rolling bearing contacts, where the lubrication regime is known as EHL.

First, a comprehensive review of the existing literature on grease lubrication, with specific focus on friction, was conducted in Chapter 2. It was shown that, due to their multiphasic composition and complex rheology, greases behave in an unconventional way, and generally do not obey the classical EHL theory for oils. Therefore, in contrast to oil lubrication, it is difficult to identify bulk characteristics of grease which can be used to predict the thickness and composition of the grease lubricating films. Instead, film formation mechanisms in grease lubrication depend on many variables, which change as a function of the operating conditions, grease formulation and microstructure, and its mechanical and thermal history. It has been shown that the complexity of the topic is at the core of a debate between different theories of grease lubrication in rolling bearings, which are contrasting and incompatible on many aspects. It was stressed that this could
be the result of the fact that the scientific literature is lacking the presence of comprehensive studies aimed at investigating the impact of grease formulation on film formation in a systematic manner. Moreover, EHL friction in grease lubrication was shown to remain a relatively unexplored area in the scientific literature, and consequently even less understood than film thickness. Without a better understanding of the mechanisms of grease lubrication in EHL contacts, and in particular the corresponding impact of grease formulation, it is difficult to create a background of rules for the choice or development of low-friction grease formulations. Therefore, a research programme was designed to address this lack of understanding and provide insight into the influence of grease composition on friction, with the ultimate aim of establishing basic guidelines for potential formulations of low-friction greases.

Chapter 3 described the greases used for the experimental investigation:

- The first set was a selection of commercially available greases, strategically chosen to encompass a wide variety of formulations, comprising different thickener types, base oil types and viscosities. The selection included currently available low-friction greases, so that their performance served to establish a reference line for further investigations. In addition, these greases were also centrifuged so as to obtain their bled oils, which were used as additional samples. This helped to establish the relative importance of thickener and base oil on the general grease frictional behaviour in EHL contacts. Some of the original grease samples were also subjected to a process of mechanical degradation, so that its effect on grease performance could be evaluated;

- The second set was a series of custom-made lithium greases, which were all fabricated in a systematic manner so that only one composition variable was changed at a time. This allowed for specific aspects of grease composition, namely the effects of base oil type and viscosity, and presence of friction modifiers (more specifically, oleic acid), to be effectively isolated. In this way the influence of these specific aspects of grease composition on friction could be investigated, and concrete conclusions could be drawn by direct comparison between grease samples differing in composition by only one variable.

These two sets of greases were subjected to a tribological investigation by employing the experimental techniques and procedures described in Chapter 4. The tests were carried out on ball-on-disc tribometers, mainly under fully flooded conditions for both friction and film thickness measurements. The experimental programme was focussed on the effect of speed and
temperature on grease tribological behaviour. In addition, specimens of different roughness were also employed for some of the experiments. Although the work was mostly conducted in fully flooded conditions, some of the tests were performed with the purpose of investigating friction behaviour in presence of a starved contact inlet.

Chapter 5 and Chapter 6 presented the results obtained with the commercial and custom greases respectively. Subsequently, Chapter 7 presented a discussion which brought all the observations together, and analysed the apparent trends and the physical mechanisms that may be responsible for them. The main findings are summarised below.

8.2 Main findings

The investigation revealed the existence of two operating regions, at low and high speeds, referred to as ‘low speed’ and ‘high speed’ regions, where the tribological behaviour of greases (under fully flooded conditions) is governed by different rules:

- In the ‘high speed’ region the grease friction and film thickness trends are similar to those of their base oils. Consequently, grease behaves according to the EHL theory, and its tribological performance is largely determined by the base oil properties;
- In the ‘low speed’ region greases are able to form a much thicker film than their corresponding base oils. Moreover, the rules that govern the relationship between film thickness and speed do not follow the EHL theory. The corresponding frictional behaviour at these speeds strongly depends on grease formulation.

The results provide evidence that grease film thickness and friction behaviour, although determined by the specific composition of the grease, are related in these two regions. The influence of grease formulation on friction generally depends on the lubrication region in which the grease operates, and it is therefore convenient to describe the trends observed in the high and low speed regions separately.

In the ‘high speed’ region, it was found that:

- Base oil viscosity and grease friction are not strictly related if greases made of different base oil types are compared. However, within greases made of the same base oil type, the influence of base oil viscosity is that expected from a classical Strubeck relationship, i.e.
the observed differences in friction can be accounted for by comparing the results at the same film thickness;

- Base oil type is one of the most influential factors on friction in this regime. Friction was systematically lower in greases formulated with synthetic base oil types (i.e. PAO and ester in this study) when compared to mineral oil-based greases;
- Although thicker films were generally observed with greases than with their equivalent base oils even in this high speed region, no direct evidence of thickener influence on friction was clearly observed.

In the ‘low speed’ region, it was found that:

- Thickener type significantly affects friction. Although some differences were observed in the film-forming behaviour of greases depending on thickener type, the most apparent difference is in their frictional behaviour. In particular, the low-speed thick films obtained with lithium-thickened and polyurea-thickened greases provided low friction, while high friction was given by the calcium sulphonate complex greases;
- Base oil viscosity does not affect friction in this region for a given oil type. However, it does affect the range of speeds over which greases operate in the ‘low speed’ region. In particular, the lower is the base oil viscosity, the higher is the maximum speed up to which greases operate in this region. The effect of temperature is equivalent, so that higher temperatures (reducing the base oil viscosity) also increase the value of speed of transition to the ‘high speed’ region;
- Base oil type is shown to affect low-speed friction in two ways. Firstly, the presence of ester base oil in the grease formulation influences the development of lithium thickener fibres during the manufacturing process, making them shorter. The study provides direct evidence that the size of lithium thickener particles affects the film thickness in the low-speed region: thicker films were formed with lithium-PAO greases, which have relatively large fibres, than in the equivalent lithium-ester greases which have evidently shorter and thinner fibres. Consequently, lithium-ester greases produce higher friction due to an increased level of asperity interaction within this low-speed region. Secondly, being the film thickness the same, base oil type can affect friction at low speeds in the same way as it does at high speeds: friction was shown to be lower in the PAO-based greases than in the equivalent mineral-based greases;
Oleic acid, used as a friction modifier in the lithium custom greases, proved to be ineffective in further reducing friction. While its beneficial effect on friction was obvious in the tests with base oils, no apparent differences were observed in the friction behaviour of additised and non-additised greases. It was proposed that the thick boundary films that greases form at these speeds already provide very low friction when ‘low-friction thickeners’, such as lithium in this case, are used in the grease formulation, and thus no additional benefit is derived from oleic acid. In contrast, since under the same conditions the base oils operate in mixed or boundary lubrication, i.e. with much thinner separating films, in this case oleic acid is enabled to interact with the steel surface and reduce friction;

Mechanical aging of greases generally diminishes their capability to form thick films. As a consequence, film thickness and friction performance of aged greases become much closer to those of their base oils. Therefore the effect of mechanical aging is to move the ‘transition speed’ to lower values, and generally to reduce grease film forming ability. A notable exception was found in greases formulated with lithium and PAO base oils, the tribological behaviour of which was unaffected by mechanical degradation.

The results provide direct evidence that the transition between the low and high speed regions for a given thickener structure occurs at a characteristic ‘transition film thickness’, rather than the more commonly employed ‘transition speed’. This ‘transition film thickness’ appears to be related to the size of thickener particles. Larger particles seem to generate higher values of ‘transition film thickness’. The same ‘transition film thickness’ was found in all custom greases formulated with PAO and mineral oils, which indeed have similar thickener microstructure, while a lower value was observed in the equivalent ester-based greases, containing much smaller thickener particles as already mentioned above.

Although very limited, the investigation on the starvation behaviour of greases did show some trends in the relationship between grease composition and friction. In line with earlier studies, severe starvation occurs at higher speeds with greases made with lower base oil viscosity. Furthermore, the results provide some evidence that the presence of polar additives in the grease formulation (oleic acid in this case) may promote the onset of starvation.

Based on the results of this study, and within the range of grease formulations and test conditions used here, the combination of low viscosity PAO base oil and lithium thickener is
suggested as the best formulation for an energy efficient, enduring bearing grease. Indeed this formulation was shown to give low friction under all operating conditions, and after extensive mechanical degradation. As an additional advantage, this composition also provides thick films at low speeds, affording enhanced protection to the contacting surfaces.

8.3 Suggestions for future work

In order to build on the understanding gained in this study, suggested potential further research may include:

1) Additional testing employing improved analysis techniques, to help better explain the prevailing mechanisms of grease film build-up and friction:

   a) Chemical/physical analysis of lubricating films in-situ during the film thickness and/or friction tests - Fourier Transform Infrared micro-spectroscopy could be used to attempt to characterise the composition of the lubricating film within the rubbing contact during the film thickness and/or friction tests. Ideally this set-up would allow to distinguish between thickener and base oil components inside the contact, hence establishing their relative contributions under different operating conditions. Fluorescence could also be used in adjunct to observe thickener and oil flow inside and around the contact;

   b) Chemical/physical analysis of lubricating films post-test - The combination of FTIR, SEM and AFM techniques could be employed to inspect and assess the chemical and physical properties of the lubricating film left in the rolling track of the MTM discs, and raceways of bearings in case of full bearing tests. This would provide valuable information on the composition of any residual grease films in relation to the formulation of the employed grease, and would therefore help establishing which grease components contribute to the development of any residual layers;

   c) Traction curves at low speeds - Friction measurements carried out at constant low speed and varying slide/roll ratio could offer an improved interpretation of the mechanisms of formation of these low-speed films. Tests with different inlet and in-contact shear rate conditions could indicate whether these films are built by adsorption or entrainment, and to what extent these mechanisms concur to build the lubricating film;
2) Full bearing tests should be carried out before suggested grease formulations are employed to manufacture and market ‘low-friction’ greases, in order to investigate:

a) Friction performance - Full bearing friction tests to complement the study on friction in single contacts, and confirm that the low-friction grease formulations suggested in this study perform equally well in real bearing application conditions;

b) Grease life performance - The results obtained with the mechanically degraded lithium-thickened PAO-based greases studied in this work are promising. It has been shown that, with this specific formulation, film thickness and friction behaviour remains unaffected by severe mechanical degradation. In view of the importance of grease life in bearing applications, this particular aspect should be verified in bearing endurance tests;

3) Further research on the influence of grease formulation on friction should be conducted with custom-made model greases, as this is the only way to systemically study the influence of any one aspect of grease formulation in controlled experiments. As done in the current work, the model greases should be formulated in a systematic manner so as to methodically concentrate the study upon the influence of a single aspect of grease formulation on friction at a time. The investigation could then be focussed on:

a) The influence of alternative thickener types and concentration - Given that lithium shortages and price increases have recently been reported, primarily due to its high demand for the production of batteries\textsuperscript{154}, it would be timely to investigate the performance of non-lithium thickeners in more depth;

b) The influence of additives - The fact that oleic acid was shown not to have any significant effects in the formulation of the tested greases does not exclude the potential influence of other additive types. In particular, the suitability of other friction modifiers in preserving low friction, under conditions where the low-speed thick films are absent or mostly removed, should be assessed. It would also be interesting to understand whether the mechanical properties of these low-speed layers could be optimised by using certain additives, such as tackifiers, for maintaining their functionality over a wider range of operating conditions;
c) The influence of grease microstructure - The results obtained in the current work have shown that, although the same manufacturing cycle, amount and type of lithium thickener were used for producing the custom greases, the structure obtained in the ester oil-based grease was much thinner. This, in turn, was shown to affect film thickness and friction behaviour. It would be interesting to understand the way in which the combination of thickener and oil affects the development of the grease microstructure itself, and how the manufacturing process could be optimised accordingly to produce the desired grease microstructure and consequently its tribological properties;

4) A model for the prediction of grease film thickness in EHL contacts should be developed. In view of the fact that many friction results in the current work have been explained by means of film thickness measurements, and that film thickness results have shown a distinctive trend in greases formulated with a given base oil and thickener type, it would be practicable and useful to develop a formula for film thickness prediction in grease-lubricated concentrated contacts as a function of grease composition;

5) Investigation into alternative ‘types’ of lubricant design, that would retain the basic beneficial properties of grease, i.e. its ability to keep the lubricant in place and seal the bearing, but may introduce additional benefits in terms of friction and surface protection. The basic task of a grease is simply to supply a lubricating media at the right place and at the right time throughout the lifetime of the component. Grease is able to do this primarily due to its multiphase structure. It is quite feasible, albeit challenging, that the same basic properties can be attained with alternative lubricant designs or principles, utilising new materials with intrinsic favourable lubricating properties.
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