Tribological Performance of Bio-based Greases with Polymer Thickener Systems

Tribologische Leistungsfähigkeit von biobasierten Schmierfetten mit polymeren Verdickersystemen

Von der Fakultät für Maschinenwesen der Rheinisch-Westfälischen Technischen Hochschule Aachen zur Erlangung des akademischen Grades eines Doktors der Ingenieurwissenschaften genehmigte Dissertation

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Abstract

Currently, most commercially available lubricating greases are produced from petrochemical materials. Petrochemical lubricants used in machines and systems in various industries get leaked or disposed of into the environment, which imposes significant threats to the ecosystems. Toxic petrochemical materials pollute water, air, and soil, which leads to several diseases and health problems for humans and animals. A promising solution to these threats lies in the transition to bio-based, non-toxic lubricants.

A growing interest in scientific research towards synthesizing bio-based lubricants, including lubricating greases, has been observed in recent decades. However, such developed bio-based alternative greases were mostly plant-based and could not compete with their polymeric petrochemical counterparts regarding their performance in rolling element bearings; thus, they were often not integrated into the industrial machines. To produce competitive lubricating greases for application in rolling element bearings, it is essential to consider both development and evaluation process, while considering their correlations.

This thesis employs a methodological development of bio-based lubricating greases with different polymer thickener systems and a comprehensive assessment of their tribological performance at the tribometer and component level. It also includes the tribological performance of the bio-based polymeric greases compared to the petrochemical polymeric counterpart. Understanding the correlation between the grease's physicochemical characteristics and tribological performance on different scales elucidates the potential for the design of competitive bio-based greases. Therefore, the correlation of tribological performance with physicochemical parameters in different scales is also covered in this work. The findings of this work facilitate the future development of lubricating greases, specifically bio-based greases, and contribute to the transition toward the integration of sustainable lubricating greases in rolling element bearings across various industry sectors.

Zusammenfassung

Derzeit werden die meisten marktüblichen Schmierfette aus petrochemischen Stoffen gewonnen. Die Entsorgung und das Leckage von petrochemischen Schmierstoffen, die in Maschinen und Anlagen in verschiedenen industriellen und landschaftlichen Sektoren verwendet werden, stellt ein Umweltrisiko dar. Toxische Stoffe können Wasser und Boden verunreinigen, was zu schwerwiegenden Gesundheitsproblemen für Mensch und Tier führt. Eine vielversprechende Lösung für diese Gesundheitsbedrohungen liegt in der Umstellung auf biobasierte, ungiftige Schmierstoffe, die diese Risiken möglicherweise verringern können.

Das Interesse an der wissenschaftlichen Forschung zur Synthese von biobasierten Schmierstoffen, einschließlich Schmierfetten, hat in den letzten Jahrzehnten zugenommen. Allerdings waren die entwickelten alternativen Schmierfette meist pflanzenbasiert und konnten hinsichtlich ihrer Leistung in Wälzlagern nicht mit ihren polymeren petrochemischen Gegenstücken konkurrieren; daher wurden sie oft nicht in die Industriemaschinen integriert. Um konkurrenzfähige Schmierfette für den Einsatz in Wälzlagern herzustellen, ist es wichtig, sowohl den Entwicklungs- als auch den Bewertungsprozess zu berücksichtigen und dabei die Korrelationen zu beachten.

Diese Arbeit umfasst eine methodische Entwicklung von biobasierten Schmierfetten mit verschiedenen Polymerverdickersystemen und eine umfassende Bewertung ihrer tribologischen Leistung auf Tribometer- und Komponentenebene. Sie umfasst auch die tribologische Leistung der biobasierten polymeren Schmierfette im Vergleich zu ihrem petrochemischen polymeren Gegenstück. Das Verständnis der Korrelation zwischen den physikochemischen Eigenschaften des Schmierfetts und der tribologischen Leistung in verschiedenen Ebenen gibt einen Einblick in das Potenzial für die Entwicklung konkurrenzfähiger biobasierter Schmierfette. Daher wird in dieser Arbeit auch die Korrelation zwischen der tribologischen Leistung und den physikochemischen Parametern in verschiedenen Ebenen behandelt. Die Ergebnisse dieser Arbeit erleichtern die künftige Entwicklung von Schmierfetten, insbesondere von biobasierten Schmierfetten, und tragen zum Übergang zur Integration nachhaltiger Schmierfette in Wälzlagern in verschiedenen Industriesektoren bei.

Preface

This dissertation was written during my doctoral studies at the Institute for Machine Elements and Systems Engineering (MSE) at Rhenish-Westphalian Technical University, RWTH Aachen.

I would like to express my gratitude to my supervisor, Univ.-Prof. Dr.-Ing. Georg Jacobs, for his valuable guidance throughout my academic journey. His insightful critical comments and technical support were a constant source of motivation during my doctoral research. My sincere thanks go to Univ.-Prof. Dr.-Ing. Schmitz for reviewing this dissertation and serving as the second reviewer and examiner in my doctoral examination. I am also grateful to Univ.-Prof. Dr.-Ing. Schmitt for his role as the chairman of my doctoral examination.

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Finally, I would like to express my special thanks to my wife for the understanding and patience during my academic journey and dedicate this dissertation to her and our expected little child.

Aachen, August 2025

Content

Nomenclature

Symbol	Description	Unit
C_F	Damping constant of the fluid	[N.s/m]
C_s	Damping constant of the structure	[N.s/m]
E'	Reduced elasticity module	[Pa]
F	Normal load	[N]
f	Frequency	[Hz]
G	Dimensionless material parameter defined as	[-]
	$G = \alpha_p . E'$	
G'	Storage module	Pa
G''	Loss module	Pa
h_0	Film thickness	[m]
H_{min}	Dimensionless minimum film thickness	[-]
h_{min}	Minimum film thickness	[m]
k	Ellipticity parameter	[-]
	$k = 1.03. (\frac{R_y}{R_x})^{0.64}$	
K_F	Stiffness of the fluid	[N/m]
K_s	Stiffness of the structure	[N/m]
P_n	Polymerisation degree	[-]
R_q	Root-mean-square roughness	[m]
R_a	Arithmetic mean roughness	[m]
R_x	Radius in x-direction	[m]
R_y	Radius in y-direction	[m]

R_z	average roughness depth	[m]
U	Dimensionless velocity parameter	[-]
	$U = \frac{\eta_0 . u_m}{E' . R_x}$	
u_m	Hydrodynamic effective velocity	[m/s]
W	Dimensionless load parameter	[-]
	$W = \frac{F}{E' . {R_x}^2 \prime}$	
α_p	Pressure-viscosity coefficient	[-]
δ	Mutual approach	[m]
ζ	Damping ratio	[-]
η_0	Base viscosity	[Pa.s]
λ	Specific film thickness	[-]

Abbreviations

Abbreviation	**Description**
ASTM	American Society for Testing and Materials
AW	Anti-Wear
BD	1,4-butanediol
BS	Succinic acid
DDS	Dodecanedioic acid
EHD	Elastohydrodynamic
EP	Extreme Pressure
FE8	Axial bearing test rig
FFT	Fast Fourier Transformation
FNR	Agency for Renewable Resources (Fachagentur Nachwachsende Rohstoffe e. V.)
FTIR	Fourier Transform Infrared
H-NMR	Proton nuclear magnetic resonance
LPC	Liquid Crystal Polymers
MDI	Methylenediphenyl diisocyanate
NLGI	National Lubricating Grease Institute
NMR	Nuclear Magnetic Resonance
PAG	Polyalkylene glycols
PAO	Polyalphaolefine
PDI	Pentamethylene diisocyanate
PrD	1,3-propanediol
PTFE	Polytetrafluororthylene
RLP	Radial bearing test rig

RMS	Root mean square
SEM	Scanning Electron Microscope
SRR	Slide to Roll Ratio
TDI	Toluene diisocyanate
TU Dortmund	Technical University of Dortmund

1. Introduction

The negative effects of using fossil-based materials on the environment have been considered a growing concern and have been widely acknowledged by scientists, policymakers, and the general public. The extraction of oil from natural resources to produce petrochemical materials in recent decades has caused severe damage and pollution of air, water, and soil. Furthermore, the manufacturing process of petrochemical materials, on the one hand, and the disposal, on the other hand, has threatened the ecosystems and wildlife by increasing greenhouse gases and polluting the environment. Additionally, the usage of petrochemical materials by humans has caused severe health effects, respiratory problems, and cancer. Recently, a trend has been observed in various industries to decrease the harmful impact of petrochemical materials. One of the ways to decrease such effects is to develop alternative biodegradable and bio-based materials. [1].

The transition to bio-based materials as the first step toward producing biodegradable materials is crucial to reducing the harmful effects of petrochemical materials on the environment. This transition will additionally address the urge to protect the current oil and gas resources and pursue alternative clean ones according to the conservation of resources (COR) theory. The intention to apply bio-based materials in the lubricant industry to substitute petrochemical materials to reduce the environmental impact and serve resource conservation has been observed in recent decades. [1].

One important application of lubricants in rotating machinery is to lubricate rolling/sliding contacts such as in rolling element bearings. According to SKF, more than 80% of the rolling element bearings are lubricated with lubricating greases [2]. According to the latest official data, around 40000 t of lubricating grease were produced in 2011 [3], from which the share of bio-based greases — i.e. the greases produced from a renewable raw material — was only 1500 t or less than 4% [4]. Another recent report [5] also indicates that more than 90% of lubricating greases are manufactured based on petrochemical oils. To be able to substitute the petrochemical greases with bio-based, non-toxic alternatives, there is a clear need for further development of the applicable bio-based greases in the lubrication market.

1.1. Problem statement

While there have been previous attempts to substitute petrochemical lubricating greases with sustainable, environmentally friendly and biodegradable greases, there are still main challenges to overcome. The first challenge is the development of greases from sustainable materials, which are easily available. The second challenge is the performance of the lubricating greases in applications, which can vary based on the quality and manufacturing process and chemical structure of the grease components, i.e., base oil, thickener, and additives. To be able to substitute petrochemical greases with competitive eco-friendly alternatives, further research toward the development of alternative greases which are comparable to petrochemical counterparts in terms of tribological performance are needed.

For the development of fully bio-based greases, both the thickener and the base oil must be derived from renewable materials. The current fully bio-based greases mainly consist of vegetable oils and vegetable-based thickeners, which exhibit inferior tribological performance to conventional petrochemical greases, such as those with polymeric thickener systems. To produce competitive bio-based greases, polymeric bio-based thickener systems must be developed. Furthermore, the tribological performance of the targeted fully bio-based greases with polymeric thickener systems, such as polyurea, polyester, and polyamide, must be evaluated and compared to the petrochemical polymeric counterpart.

The development process of greases for rolling element bearings requires multiple steps. It begins with a fundamental understanding of the chemical structure and synthesis of greases, physicochemical characterizations, and tribological behavior of the grease during screening tests. This is followed by fine-tuning of the chemical structure during the early-stage development and ends up with the upscaling and overall evaluation of the performance at the component level. The influencing parameters of the chemical synthesis, physicochemical evaluations, and tribological assessments must also be identified and correlated. Such a procedure facilitates development of bio-based lubricating greases in this work and in the future. Hence, the aim of this work is to reduce the negative effects of petrochemical materials on the environment by a methodological development and tribological evaluation of new bio-based lubricating greases.

In the following, the state of research on the lubricating greases, their components, the evaluation methods, and the bio-based alternatives are covered. The research need is then identified, and the research hypotheses and the objectives of the work are derived at the end.

2. State of the research

This chapter provides a comprehensive analysis of the current situation of the lubricating greases. The following subchapters cover essential aspects of lubricating grease composition, current grease types and existing bio-based alternatives and their limitations. Additionally, an overview of the existing tribological performance evaluation methods, such as lubricating film formation, friction and wear is provided. Moreover, based on the identified needs in the current state of the art, the research topic for this dissertation is defined.

2.1. Lubricating greases

According to the definition from National Lubricating Grease Institute (NLGI), a lubricating grease is "'a solid to semi-fluid product or dispersion of a thickening agent in a liquid lubricant. Other ingredients imparting special properties may also be included" [6] .Lubricating greases play a crucial role in industrial machines to decrease wear and friction in machine elements, thus preventing the premature failure due to insufficient lubrication. Other important roles of the lubricating grease are to cool down the machine elements by increasing the heat exchange rate to decrease the temperature caused by friction as well as better sealing against contamination and corrosion. The lubricating greases are used in machine elements in various industrial sections such as oil and gas, food industry, aerospace [6], electrical vehicles (EVs) [7]. This includes machine elements such as rolling element bearings and gears. One of the most important machine elements, in which the lubricating greases is used is rolling element bearing. In rolling element bearings, the lubricating grease separates the rolling elements and the raceways to avoid asperity contact and wear of the materials.

The tribological performance of rolling element bearings, as a widely used machine element in rotating machines, plays a crucial role in the flawless operation of mechanical systems. To analyse the lubricating greases in rolling element bearings in terms of tribological performance, first, the common standard definitions such as *Tribology*, *Friction,* and *Wear* must be defined. *Tribology* is the "the science and technology of mutually interacting surfaces in relative motions" [8]. These contacts are commonly associated with three key parameters: friction, wear, and lubrication. Understanding these key parameters is critical for the design of machine elements such as rolling element bearings.

Friction is, by definition, the resistance to motion [9]. In the rolling element bearings, the contact areas between rolling elements and inner/outer raceways or between the rolling elements and the cage are the main sources of friction and wear. *Wear* is the loss of material due to two surfaces sliding [9]. Common types of wear between two moving surfaces are adhesive, where some amount of material sticks to other surfaces and is transported by another surface, and abrasive, in which the harder surface removes some amount of the material from the softer surface. In machine elements such as rolling element bearings, the wear sources are usually the single contact points where two surfaces may have contact due to insufficient lubrication between them. In the context of grease performance, the greases that cause a lower amount of wear and friction in the rolling element bearings are associated with better tribological performance.

The performance of the greases in machine elements including the rolling element bearings is directly correlated with the characteristics of the composition elements of the grease. The greases are normally composed of three different materials: base oil (80-90 wt. %), thickener (5-20 wt. %) and the additives (0-5 wt. %) as shown in Figure 2-1. In following, the common types of materials in production of each grease components are covered.

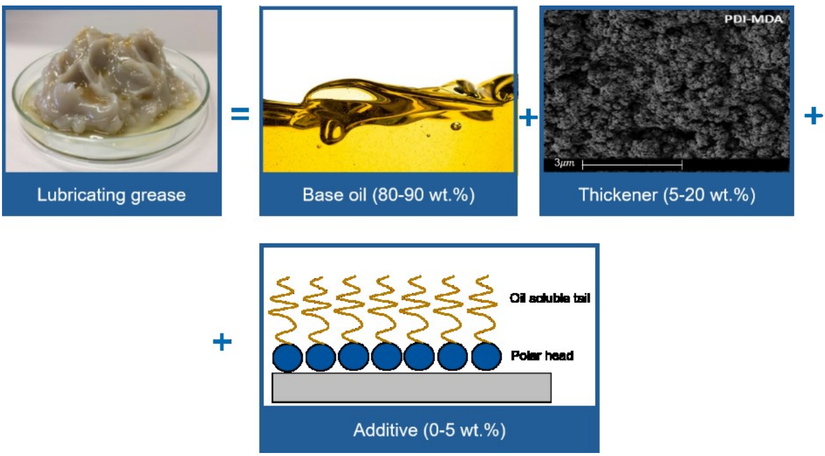


Figure 2-1: Components of the conventional lubricating greases according to [5]

2.2. Existing petrochemical greases

Base oil is the primary component of lubricating greases with major share of around 80-90 wt.%. The base oil plays a crucial role in the lubrication mechanism within the contact zones of rolling element bearings. Hence, the choice of base oil from a specific type and its viscosity significantly influence the performance of lubricating greases [10,11].

Commonly used base oils include mineral oils, synthetic oils, and vegetable oils. Mineral oils such as paraffinic, naphtenic and aromatic oils provide good lubricity and thermal stability but may have limited low-temperature performance. Synthetic oils offer a wide range of viscosities, excellent high-temperature properties, and improved oxidation resistance. The use of ester oils in lubricating greases is well known and they are already used in commercially available products [12]. Popular synthetic oils are polyalphaolefine (PAO), polyalkylene glycols (PAG) as well as ester-based oils. The most common base oil used in greases applied in rolling bearings are the mixture of the PAO and mineral oils [10], which has good thermal stability and low-temperature fluidity of the PAO and inherits the cost efficiency of the mineral oils.

Thickeners of lubricating greases provide the structure and consistency to lubricating greases. The thickener system performs as a reservoir to provide the rolling element bearing contacts with enough oil in the contact area under load and shear effects from rotation of rolling element. Thickener of the greases usually form a three-dimensional structure that keep the base oil by Van-der-Waals or capillary forces inside the pores [10] as in Figure 2-2.

Thickeners are classified into soap, non-soap and complex thickeners. Soap thickeners are synthetized by reacting of a fatty acid and a base. The product of the reaction is a soap and water as a by-product. To produce metal soap thickeners, the fatty acids usually made from natural fats, are neutralized by metal hydroxide or metal oxides as the base. The most common metallic soaps thickeners are lithium, calcium, sodium and aluminum complexes. Lithium greases exhibit excellent mechanical stability and water resistance [10]. Calcium greases offer acceptable anti-wear properties and corrosion resistance [13]. Aluminum greases are sensitive to shear and temperatures and are not suitable for rolling element bearings applications [10].

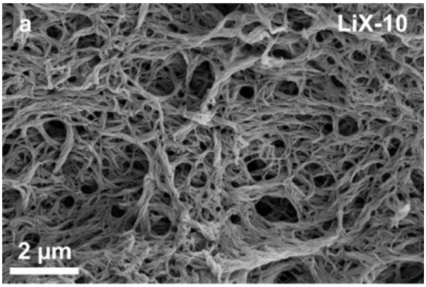


Figure 2-2: Three-dimensional structure of a lithium soap thickener under scanning electron microscope (SEM) from [14]

Non-soap thickeners include other types of thickeners which are not produced based on soaps. They may root in inorganic materials such as clay and silica [15] or organic materials [16] such as polymers, polytetrafluorethylene (PTFE) or a combination of them. Polymeric thickener systems are shown to provide enhanced mechanical and thermal stability and compatibility with other grease components [17]. Advancements include the development of hybrid thickener systems that combine the benefits of different thickener types, leading to improved performance under various operating conditions [18].

Conventional lubricating greases can optionally include different amounts of additives (0-5 wt.%). Additives play a crucial role in enhancing the performance and functionality of lubricating greases. They can be added to improve anti-wear (AW), extreme pressure (EP), corrosion resistance, oxidation stability and noise reduction characteristics of the greases [19,20]. Common additives include anti-wear agents, friction modifiers, corrosion inhibitors, antioxidants, and viscosity index improvers [10]. Some additives even have an impact on modifying the grease microstructure [21].

2.3. Alternative bio-based greases

As of 2019, more than 90% of the lubricating greases were manufactured based on petrochemical materials [5], which can be replaced by sustainable alternatives bio-based materials. To form a fully bio-based grease as an alternative to petrochemical greases, both base oil and thickeners musts be produced from bio-based materials.

As the base oil for alternative greases, vegetable oils are gaining attention due to their biodegradability and renewable nature [22,23]. A recent study compared the friction behaviour of a bio-based lubricant, mineral oil, and synthetic oil in a journal bearing. Results indicated that the bio lubricant exhibited lower friction than the mineral lubricant, though the synthetic oil displayed the lowest friction [24]. Specifically, attention has turned to vegetable oils such as palm oil and sunflower oil to identify suitable bio-based lubricants. Compared to mineral oils, vegetable oils are renewable, readily available, environmentally friendly, and thus sustainable [25]. Additionally, there have been suggestions for improving bio-based lubricants based on soybean oil with epoxy compounds, as demonstrated in [26]. Bio-based lubricating oils, like jatropha vegetable oil [27], castor oil [28], rice bran oil [29] [30,31], soybean oil [32], and sesame oil [33] are already available on the market, offering an alternative to mineral oils as the base oil in greases.

In recent years, there has been considerable research into the development of fully bio-based greases, in which both thickener and base oil is primarily produced from renewable materials. Studies, such as those detailed in [34], have explored the use of cellulose pulp from the Eucalyptus Globulus plant as a raw material for producing a thickener system together with castor oil for bio-based greases. Another study [35] optimized cellulose pulp as a potential material for bio-based thickener production. Furthermore, research presented in [36] demonstrated the development of bio-based greases using castor oil and high-oleic sunflower oil, with cellulose fibres serving as the thickening agent. In other studies, sorbitan monostearate glyceryl monostearate [37], methyl [38] and ethyl cellulose [39,40,41], chitin and chitosan [42] are utilized to synthetize different bio-based thickener systems. Among developed bio-based greases only a few greases with thickeners from eucalypt and pine woods have shown similar or lower friction coefficients compare to the lithium soap grease samples as in [43]. In other studies [44,45], lignocellulosic materials have been modified and used as thickener agents in castor oil to form a fully bio-based grease. Also, beeswax have been used as bio-based thickener together with castor oil [46]. The grease samples were able to show comparable friction to the commercial soap grease. Beeswax have also been used as bio-based thickener together with rapeseed oil to form bio-based grease [47]. There have been other developments with bio-based thickeners from cellulose pulps such as pine-kraft and eucalyptus-kraft [48,49]. Also, epoxide-functionalized alkali lignins have been dispensed in castor oil to form bio-based greases [50]. There have also been studies [51,52], which produced bio-based thickener agent from silica together with vegetable oils or esters to form bio-based grease. In another study, beef tallow and graphene are also been combined to form bio-based greases [53]. Moreover, research [54] has indicated that wear in bearings lubricated with some of ester-based bio-based greases can be lower than that observed with petrochemical soap greases.

The polymeric petrochemical greases have been shown to exhibit superior tribological performance [55]. Despite the promising results of bio-based greases in terms of their tribological performance compared to soap-based greases, they still cannot compete with conventional petrochemical polymeric greases.

Research Need #1: Based on the review, it can be concluded that despite recent advancements in bio-based grease technology, most of the developed thickener systems for bio-based greases are vegetable-based thickeners. Although they show promising results compared to petrochemical soap greases, they still cannot compete with polymeric petrochemical greases regarding the tribological performance such as friction, wear, and lubrication film formation. There is a need for the development of polymeric thickeners from renewable materials for bio-based greases to improve the competitiveness in terms of tribological performance compared with conventional petrochemical greases.

The developed bio-based greases must be evaluated regarding tribological performance and compared to petrochemical greases. In the next section, the standard tribological evaluation methods for the greases are discussed.

2.4. Tribological evaluation of greases and lubrication mechanism

Several tribological evaluation methods have been developed to characterize film formation, friction, and wear in the oil and grease-lubricated rolling element bearings. The tribological performance of the greases can be evaluated on the isolated single rolling contact of the bearings on the lab using tribometer set-ups and/or done on the component level, i.e., rolling element bearing test rigs; each method is entitled to specific limitations. The state-of-the-art evaluation methods are covered in two categories in the following section: (a) on single contact level on the tribometer and (b) the component tests.

2.4.1. Grease performance on single rolling contact level (tribometer)

The production process of lubricating greases is associated with high costs during chemical design, troubleshooting, final synthesis, scale-up in higher amounts of greases and performance evaluation of the greases on component tests. Therefore, repeating this costly chemical optimization process over and over is not feasible for early-stage development. To evaluate the suitability of developed grease samples with developed thickeners in smaller amounts in early-stage development, screening methods at single rolling contact on tribometers can be proposed.

For the study of film formation, friction, and wear mechanism, which is strongly affected by thickener chemical structure [56], various tribometer types are used to simulate the situation of an isolated single contacts in the laboratory, as the usual methods are shown in Figure 2-3.These methods are covered shortly in the following.

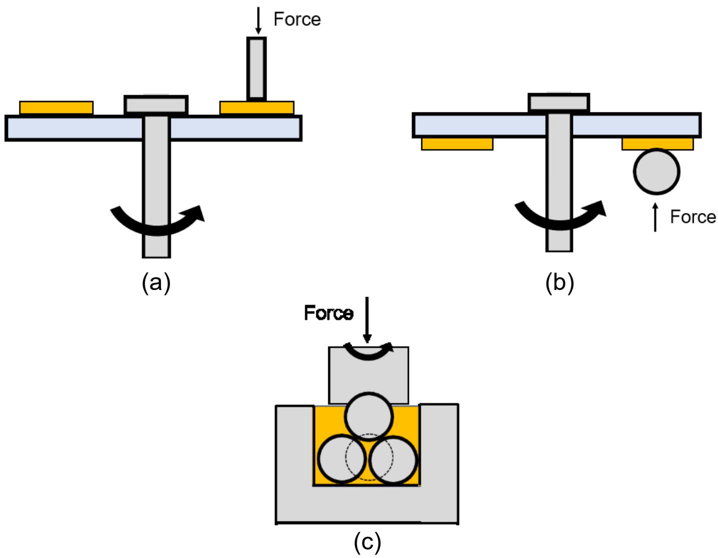


Figure 2-3: Schematic demonstration of pin-on-disc (a) and ball-on-disc (b) and four ball (c) tribometers

Pin-on-disc [57] tribometer: This setup consists of a stationary test pin, which is pressed against a rotating disc with the specified load. Oil or grease might also be used as the lubricant on the contact area and the frictional force is measured as the disc rotates. Different pin materials are used to investigate the friction coefficients of various materials under various loads and speeds. The limitation of this method is that it only represents sliding contacts and cannot simulate the rolling contacts between rolling elements and raceways with point or line contacts.

Ball-on-disc [58] tribometer: In this setup, a steel ball is pressed against a steel/glass disc under defined force, and the ball and the disc will separately rotate to simulate the motion between the rolling element and the raceways in the rolling element bearings. Oil or grease might be used as the lubricant on the contact area, and the torque sensors on the ball or the disc measure the friction forces. Film thickness can be captured by the camera, and the friction coefficients for different speeds, loads, or materials are then determined based on the measured frictional forces.

Four-ball tribometers: The next method of evaluation of lubricating greases is to assess the extreme pressure performance and anti-wear properties of greases by subjecting three stationary balls to a rotating fourth ball [59]. The test measures the scar diameter or wear volume at the contact level. Modifications to the test, such as incorporating a variable load and temperature, provide possibility of predicting real-world performance. There have been studies that evaluated the anti-wear and additive evaluations of the developed bio-based greases using four-ball tribometers and showed promising results of lubricating greases with bio-based additives [60,61,62].

The advantage of the ball-on-disc tribometer over other mentioned tribometers is that it represents the rolling contacts and delivers more information about the formation of the lubricating film between the rolling element and raceways in the rolling element bearings. This includes information about the thickness of the lubrication film and the friction induced in the contact area, which can be directly correlated. Therefore, this evaluation method on tribometer will be focused on in the scope of this work.

Remark: The published approaches for methodological development of greases in the literature consider the standard chemical synthesis and standard chemical evaluations including measurements of viscosity using rheometer [48], thermal stability [63], and employing Fourier-transform infrared spectroscopy FTIR [32]. However, when it comes to evaluation of the tribological performance, if at all, they are mainly limited to four-ball screening tests [31] or Translatory Oscillation Tribometer (SRV) tests [64]. Such methods do not provide explicit information about the lubrication mechanism, e.g. film formation of the greases. Measurement of film formation under various conditions provides fundamental understanding of lubrication mechanism and can lead to better understanding and estimation of wear [65]. The analysis of lubrication mechanism also provides potentials for the fine-tuning of grease formulations to achieve desired performance characteristics in early-stage development. Additionally, the existing development methods usually do not integrate component tests to evaluate the final performance of the greases and are only limited to tribological screening tests. The current thesis employs the comprehensive methodological development of the grease with the focus on the lubrication mechanism and film formation as well as component evaluation tests to address the existing gap in the literature. As a test case for such approach, development of bio-based greases is focused in this work.

In the following section, the film formation in the contact area and the effect of thickener on it at the tribometer level will be discussed.

2.4.1.1. Film formation and effect of thickener

During the operation of grease-lubricated bearings, the base oil is released from the thickener systems due to the composition of applied load, temperature, and shear. The motion of the contact partners helps convey the base oil into the contact area, establishing a hydrodynamic lubricating film that ideally separates the moving surfaces. These contacts, experiencing deformation under high pressures, are known as elastohydrodynamic (EHD) contacts (Figure 2-4) and are essential to ensure the longevity of bearings. The film formation in single contact of oil-lubricated rolling element bearings has been understood widely in the literature by several researchers and summarized by [66] for different geometries and conditions. The most popular estimation formulas for dimensionless minimum film thickness in elliptical contacts have already been proposed by Hamrock and Dowson [67]. The effect of thickener system on the film formation of the grease is neglected in this model.

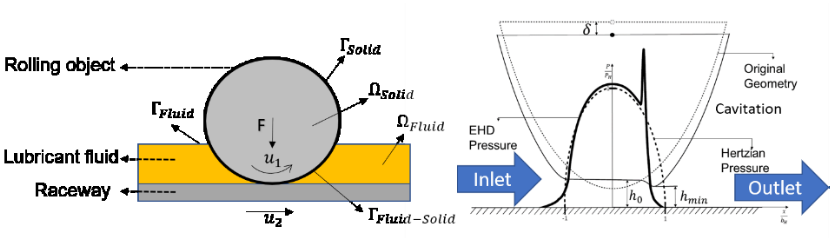


Figure 2-4: Single roller of a rolling element bearing and film formation in elastohydrodynamic (EHD) contact between roller and a raceway

Research Need #2: In elucidation of the effect of thickener on lubrication mechanism, evaluation of film formation on ball-on-disc tribometer has been used in various studies [68,69,70,71]. However, there is a research need to investigate the potential of the ball-on-disc tribometer in selecting bio-based thickener candidates (related to research need #1) as the screening method in early-stage development and to determine the effects of bio-based thickeners properties, such as thickener type and chemical structure, on the film formation.

2.4.2. Grease performance on component level (rolling element bearing)

The results of the tribometer level evaluation methods in terms of film formation and friction cannot solely be used as the single performance evaluation method of greases due to limited parameter range and not considering complex dynamics of bearings such as rolling element forces and their interactions with cage or centrifugal forces as well as manufacturing parameters or imperfection geometries of the bearing's components. Therefore, the performance of the greases has to be further evaluated on the component level to check the competitiveness of the developed bio-based greases. In the following, the standard methods for tribological evaluation of the greases on rolling element bearing level will be discussed.

Component test setups are needed to cover more complex effects of bearings, such as centrifugal forces, or more operational conditions range, such as rotational speeds and forces. This is necessary to evaluate the final performance of the developed greases. Among others, measurement of wear protection characteristics on FE8 with gravimetric analysis or surface profilometry [72], lifetime of grease-lubricated rolling element bearings on FE9, corrosion on false brinelling and micro pitting and EMCOR, vibration on GRW noise test are usually used at rolling element bearings level. The standard tribological methods both at tribometer and component levels and the limitations are also summarized in Figure 2-4.

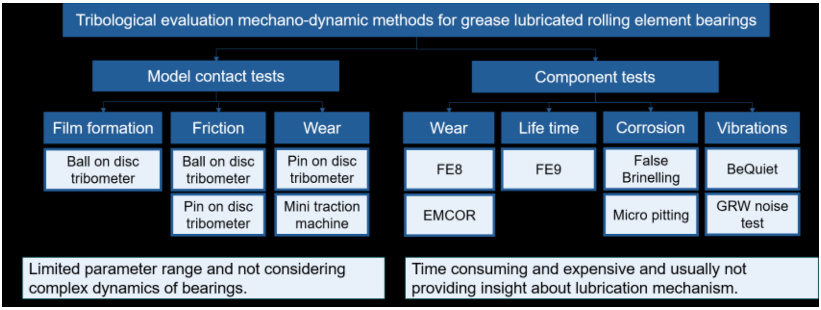


Figure 2-4: Tribological evaluation methods for the lubricating greases in rolling element bearing applications divided into model contact level and component tests

Previous studies have shown that greases with different thickener types have different friction coefficients and wear protection characteristics such as in [73] and [74]. As an example, in [74], it has been shown that different thickener types, e.g., lithium, polyurea, and calcium sulfonate complex greases, have resulted in different wear protection results. Additionally, the thickeners play an important role in the lifetime of rolling element bearings [75,76]. It has been shown that different thickener types, e.g. lithium soap and diurea, have resulted in different lifetimes of the bearings by advancing the lubrication in the contact areas and thus supporting the longevity of the bearings [75]. As the two main aspects for the evaluation of grease performance, only the wear and lifetime assessment methods will be focused in this work.

The performance evaluation with component tests is usually associated with high costs and is time-consuming; thus, integrating them in the early-stage development phase is usually not economically feasible. There are only limited studies that assessed bio-based lubricants (mostly the bio-based oils) and compared them to petrochemical lubricants in component tests [77]. Furthermore, solely the screening methods are not adequate for evaluation of the overall performance of the greases, as the suitability of the greases are highly dependent on the operational conditions which cannot be covered fully by limited parameter ranges of screening methods. Therefore, utilization of screening and component tests are not interchangeable [78].

Research Need #3: Previous studies have shown that greases with different thickener types have resulted in different wear protection and lifetime of the rolling element bearings. The performance of the developed bio-based thickener for bio-based greases (research need #1) has to be investigated regarding wear protection and the lifetime of the rolling element bearing. Additionally, there is a need to verify the convertibility of the results of the screening method on the tribometer (research need #2) on the final performance of the greases on the rolling element bearing tests.

One of the aspects of lubricating oil and greases in rolling element bearings is to dampen the dynamic forces in bearings. These dynamic loads originate from various sources, such as external dynamic loads or imperfections in geometries, which may degrade the overall performance [79]. Therefore, besides the previously mentioned methods, evaluation of vibration and noise reduction characteristics of the lubricant in rolling element bearings can be used. Vibration analysis of oil-lubricated contacts has already been investigated at the tribometer [80] as well as component level [81].

For the evaluation of damping characteristics of the different lubricating greases on the rolling element bearings, the standard method is the vibration tests according to the *BeQuiet* method from SKF [82]. This method observes the vibration levels of the radial ball bearing lubricated with different greases under different axial loads. This method has been previously used as an evaluation method to compare different thickeners and base oil combinations [83,84]. However, previous studies about the effect of thickener on the film formation on the tribometer have revealed that film formation in the contact area, as well as the thickness of the thickener deposition layer [85] is dependent on the radial force. In industry-relevant radial loads (in the GPa range) and different temperatures, the effect of thickener on noise reduction of the greases is unknown.

The vibration analysis of the rolling element bearings as an important tool has not only been used to evaluate the damping characteristics of the lubricating oil and greases, but also it provided the researchers the opportunity to build condition monitoring models of the rolling element bearings to evaluate the overall tribological performance of the bearing such as the wear [86]. The vibration analysis is also used to identify the failures in rolling element bearings, such as pitting and cracks on raceways, which usually occur due to insufficient lubrication [87]. For accurate condition monitoring and fault detection and preventing premature failure in rolling element bearings, several experimental vibration analyses such as [88] have been conducted, and different simulative vibration models for oil-lubricated contacts have been proposed [89,80]. However, due to the complexity of the thickener system, such models for grease-lubricated contacts are missing. Furthermore, it is still not known if the developed bio-based greases will demonstrate undesired noise characteristics comparable with petrochemical greases.

Research Need #4: Vibration analysis has been used as a tribological performance evaluation method to characterize grease thickeners [83,84]. However, the role of bio-based thickeners in damping characteristics of the bio-based grease has not been identified. Additionally, the previous studies on noise reduction of greases are limited in explaining the role of thickeners in the overall noise reduction of grease due to the limitation of standard tests. A fundamental research is needed to identify the role of thickener in vibration characteristics of the grease by conducting separate vibration tests on complete greases including the thickener as well as only the base oils.

Based on the findings of the literature review, it can be concluded that the author could not find any public holistic methodological development for bio-based grease for rolling element bearings applications that thoroughly evaluates and tests the bio-based greases against petrochemical grease in terms of film formation at tribometer level and wear and lifetime, and noise performance at component level and covers the correlation of the performance both at tribometer and component levels.

2.5. Overview on the research needs

As emphasized in the preceding chapters, the primary function of lubricants is to minimize friction and wear. Oils and greases are the common lubricating substances employed in various machine applications. These commonly utilized lubricants are typically derived from petrochemical sources. However, it is widely recognized that petrochemical materials have negative impacts on the environment [1], necessitating a shift towards more environmentally friendly alternatives. The demand for eco-friendly lubricants has become widely acknowledged due to their potential to decrease the negative impacts on the environment. Summarizing the previous identified research needs, there are two visible aspects of the bio-based lubricating greases that should be investigated:

Development of the bio-based thickeners for greases: Considering the development goals, both the base oil and thickener system in lubricating greases must be derived from bio-based sources in order to create a fully bio-based lubricating grease. Bio-based lubricating oils on the basis of sunflower oil, soybean oil, and castor oil have already been developed and evaluated previously. Previous studies have also explored the development of bio-based non-polymeric thickeners. However, these thickeners exhibit inferior performance compared to conventional polymeric petrochemical greases, especially in rolling element bearing applications where higher shears, loads, and temperatures are present. Hence, there is a need for the development of polymeric thickeners that are not only produced from renewable resources but also perform well regarding the tribological behaviour compared to the petrochemical greases in rolling element bearing applications (**Research Need #1**).

Thoroughly evaluation of the tribological performance of the developed lubricating greases in both contact and component level: When it comes to the evaluation of developed greases for rolling element applications, it is essential to identify the influencing parameters of bio-based thickener such as thickener type, and chemical structure on the film formation at early-stage development of the greases (**Research Need #2**). This approach is adopted to minimize the expenses and lengthy duration associated with conducting component tests on rolling element bearings. Tribometer tests are limited in parameter range and do not consider complex situations of bearings, such as interactions with cage or centrifugal forces; therefore, only considering the tribometer evaluation for greases does not reflect the overall performance of the greases. Therefore, there is a need to verify suitability of this screening method for thickener development and convertibility of the results on the final performance of the greases on the rolling element bearing test rigs (**Research Need #3**). In addition to tribological performance evaluation methods in terms of lifetime and wear protection, vibration analysis has been used in the literature to characterize grease thickeners. However, the role of bio-based thickeners in damping characteristics of the bio-based grease has not been identified (**Research Need #4**). To ensure a comprehensive evaluation of the ultimate performance of developed bio-based lubricating greases, it is necessary to evaluate them at both the tribometer and component levels.

3. Research concept

A structured research concept is outlined in this chapter by defining an objective and several hypotheses based on the derived research needs and defined research topic. These will specify an approach to investigate the research topic and explain the methods. An overview of the most important points is presented at the end of the chapter.

3.1. Objective

Previous studies indicate that existing bio-based greases show inferior performance compared to their petrochemical counterparts. The general research need behind this work is to reduce the negative environmental effects of petrochemical lubricants by introducing alternative novel bio-based polymeric lubricating greases, without compromising on their tribological performance.

The specific objective of this work is **to determine the tribological properties (such as film formation, friction, and wear) of the developed polymeric bio-based greases both at tribometer and bearing level and comparison to the petrochemical counterpart**. Additionally, the correlation of tribological performance and physicochemical characterization results of newly developed greases in development phase will be clarified.

A holistic approach with the focus on the lubrication mechanism is employed to support early-stage development and evaluate the final performance of the greases on the component tests.

3.2. Hypotheses

Based on the state of the research and identified research needs, the following hypotheses are defined:

1. The film formation of bio-based greases on tribometer level is significantly influenced by the thickener type and can be correlated with chemical parameters of thickener, such as structure, and flow limit.
2. The performance of developed greases at the component level on rolling element bearing test rigs can be correlated to the results of

tribological screening tests and confirms the suitability of the screening method.

Additionally, it is assumed that the vibration analysis can be used as a further tool to assess the noise reduction characteristics of the developed thickeners. To ensure that the developed greases with bio-based polymeric systems do not show adverse effect on the noise characteristics compared to the petrochemical reference grease, the following hypothesis is formulated:

3. The vibration characteristics of rolling element bearings will not critically deviate between those lubricated with developed bio-based greases and those lubricated with the petrochemical reference grease.

3.3. Structure of the methods

In order to investigate the hypotheses outlined in chapter 3.2, this chapter elaborates on the methods employed to test them. An overview on the structure of the thesis is demonstrated in Figure 3-1.

Initially, the work begins by explanation of the chemical synthesis and development of bio-based greases with three different polymeric systems, in this work, polyurea, polyester, and polyamide, as well as the pre-screening physicochemical characterization methods for the created greases in development stage.

To test the first hypothesis, it is essential to assess the performance of the developed greases regarding film formation at the tribometer level. Therefore, this work starts with evaluation of the developed bio-based grease samples on single contact on ball-on-disc tribometer in Chapter 5. By comparing the performance of the developed grease candidates with polyurea, polyester and polyamide thickener groups and different physicochemical characteristics, the validity of the hypothesis 1 will be tested. Additionally, a comparison will be made with an existing petrochemical grease available on the market to determine the performance of developed bio-based greases compared to the petrochemical grease and filter the best candidates.

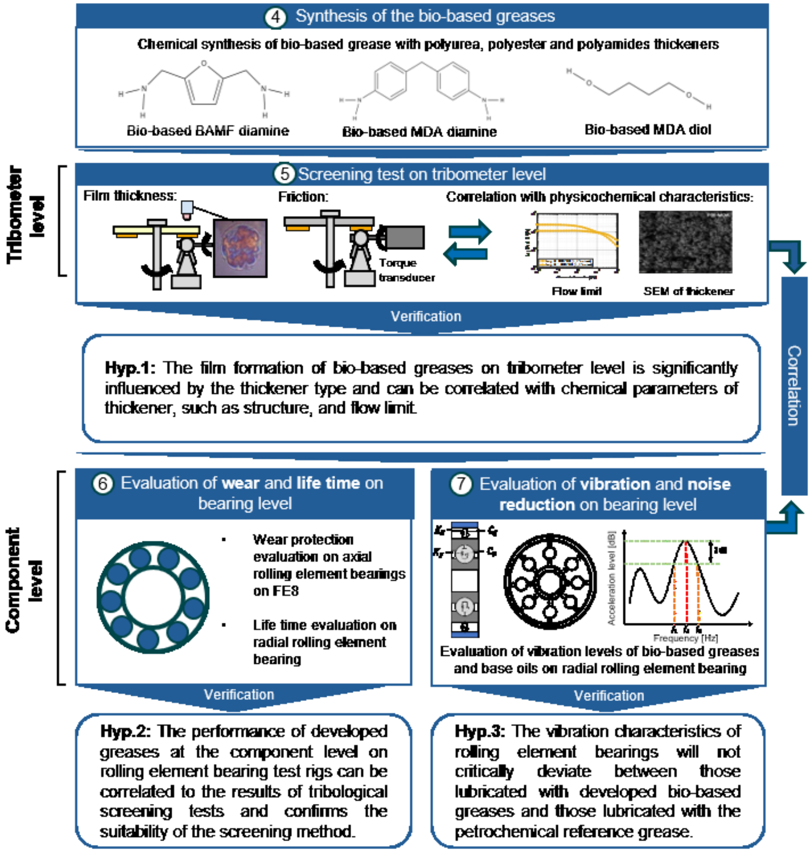


Figure 3-1: Structure of the presented work

To verify the second hypothesis in Chapter 6, component tests will be conducted on the axial and radial rolling element bearing test rigs on the best candidates to assess the wear protection and lifetime characteristics of the greases. The friction, wear, and lifetime in the final performance assessments at the component level will be correlated with the film thickness and friction coefficients observed at the single contact level to confirming the suitability of the screening method.

To test hypothesis 3, vibration analysis on radial rolling element bearings will be conducted in Chapter 7. The base oils and the complete grease samples will be applied separately on the test bearings and the vibration signals will be collected by vibration sensors in different rotational speeds and loads. The results of the vibration analysis will be compared between the base oils, bio-based grease samples, and petrochemical grease to evaluate their respective noise reduction characteristics. This final segment aims to provide insights into the vibration characteristics and tendencies exhibited by the greases, thus wrapping up the chapter on testing methods.

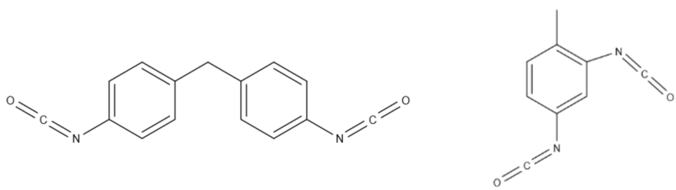
By employing these comprehensive, established testing approaches, the main aim will be achieved: determination of tribological performance and behaviour of the developed bio-based lubricating greases with polymeric thickeners in rolling bearing applications. Through these methods, we expect to thoroughly understand how these greases perform under various conditions and how comparable are they to traditional petrochemical lubricants. This analysis will provide crucial information for further optimization of the performance of these bio-based lubricants and ensuring their suitability for real-world applications.

4. Synthesis and development of the bio-based greases

Polymeric thickeners have shown excellent high temperature and antioxidation performance over non-polymeric thickeners [10]. Therefore, in the scope of a mutual project with TU Dortmund University, bio-based greases with three polymeric thickener systems of polyurea, polyester and polyamide have been synthesised [90,91]. Synthesis of the produced bio-based greases and the methods of petrochemical characterizations are briefly covered in this chapter. Parts of this chapter have already been initially reported in the author's previous publications [92,93]. For more details about the synthesis of the greases, the reader can refer to the thesis of Max Jopen [91] and the project final report [90]. The results of the characterization methods which are relevant to the scope of this work will be covered in the next chapters and correlated with tribological performance.

4.1. Polyurea

The greases with polyurea thickener systems have been used since 1980s [10]. They have good physicochemical and tribological characteristics such as mechanical stability, oxidation and high temperature performance [94]. The majority of polyurea greases are produced from reaction between a diisocyanate such as methylenediphenyl diisocyanate (MDI) or toluene diisocyanate (TDI) with diamines or fatty amines [95,96]. The chemical structures of the MDI and TDI molecules are shown in Figure 4-1.



(a) (b)

Figure 4-1: Chemical structure of the usual diisocyanates in production of conventional polyurea thickeners: (a) MDI and (b) TDI

To produce bio-based alternative polyurea thickeners, an available bio-based diisocyanate was utilized. To synthesize the bio-based polyurea thickener systems, an in-situ polymerization process of the monomers in castor oil was conducted. The reaction followed a polyaddition mechanism. Three polyureas were synthesized using pentamethylene diisocyanate (PDI) reacted with one of the diamines (a–c) (Figure 4-2). The reaction was carried out in an ULTRA TURRAX® Tube Drive with an ST-20 mixing vessel as the reaction vessel. The monomers were heated to melt and then mixed in the castor oil at 100 °C. PDI was added to the reaction mixture as the final component under stirring. After the addition of all components was completed, stirring continued for another 2 minutes. Consequently, a gel-like structure formed within 10 seconds [92].

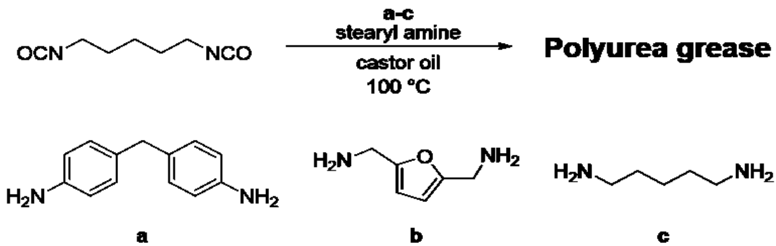


Figure 4-2: Reaction of bio-based diisocyanate (PDI) with three candidate diamines to create bio-based polyurea thickener systems from [92]

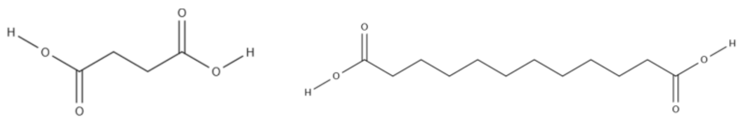
The MDA diamine is produced from Aniline; thus PDI-MDA is half bio-based (Figure 4-2 (a)). However, PDA is derived from corn starch, while BAMF is derived from sugar, making them entirely bio-based (Figure 4-2 (b-c)). Polymerisation degree determines the thickening abilities of the thickener system as known from petrochemical polyurea greases. Based on the previous knowledge, the theoretical polymerisation degree has been set to $P_n = 9$ and the share of thickener system in all the polyurea samples is 15 wt. %. The bio-based polyurea grease samples with the corresponding share of bio-based carbon atoms are listed in Table 4-1.

Table 4-1: The bio-based polyurea greases

Grease Nr.	Base oil	Thickener	Share of bio C [%]
1	Castor oil	(a) PDI-BAMF	98
2	Castor oil	(b) PDI-MDA	91
3	Castor oil	(c) PDI-PDA	98

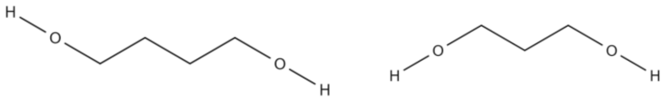
4.2. Polyester

As the next polymeric system, polyesters have been chosen to produce bio-based thickener systems for greases. This decision was initiated from good availability of bio-based polyester raw materials and their biodegradability. Polyesters are usually produced from reaction of dicarboxylic acids with diols. To produce the bio-based polyesters thickener systems for bio-based greases, various combinations of bio-based dicarboxylic acids including succinic acid (BS), dodecanedioic acid (DDS), and bio-based diols such as 1,3-propanediol (PrD), 1,4-butanediol (BD) were reacted (Figure 4-3 and Figure 4-4). The polyester thickener systems were prepared through quasi-in-situ polymerization, assuming a theoretical degree of polymerization of 9. The monomer ratio was set at dicarboxylic acid: diol: alcohol 1: 0.75: 0.25. The theoretical chain length was calculated using the Carothers equation [97]. Share of thickener system in all the polyester samples was 30 wt. %. More details for production method can be found in [93].



(a) (b)

Figure 4-3: Chemical structure of bio-based dicarboxylic acids in production of bio-based polyester thickeners: (a) BS and (b) DDS



(a) (b)

Figure 4-4 Chemical structure of the bio-based diols in production of bio-based polyester thickeners: (a) BD and (b) PrD

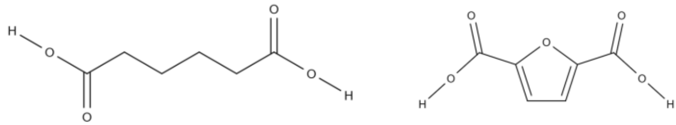
From the combination of the reactions between mentioned materials, three samples have shown thickening effects in reaction with castor oil. The bio-based polyester grease samples with the corresponding share of bio-based carbon atoms are listed in Table 4-2.

Table 4-2: The bio-based polyester greases

Grease Nr.	Base oil	Thickener	Share of bio C [%]
1	Castor oil	(a) DDS-BD	100
2	Castor oil	(b) DDS-PrD	100
3	Castor oil	(c) BS-BD	100

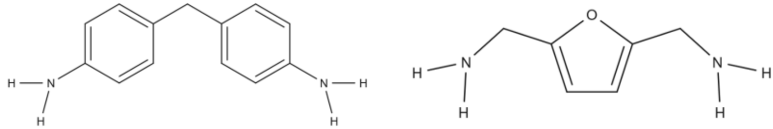
4.3. Polyamide

The third polymeric group selected to produce bio-based thickener systems, was polyamide. The application of the polyamide greases is already known since 1960s. Polyamides are produced as a reaction between dicarboxylic acid and diamines. To produce the bio-based polyamides, bio-based dicarboxylic acids (Figure 4-5) are reacted with bio-based diamines (Figure 4-6). The dimer salt was formed at 160 °C, followed by polymerization in the melt at 240 °C. The castor oil was then added at 160 °C, and the thickener was melted into the base oil. Thus, the production followed a quasi in-situ polymerization approach similar to the polyesters. Analogous to the polyesters, a thickener content of 30 wt. %. was selected.



(a) (b)

Figure 4-5: Chemical structure of bio-based dicarboxylic acids (a) AS and (b) DCF in production of bio-based polyamide thickeners



(a) (b)

Figure 4-6 Chemical structure of the bio-based diamines in production of bio-based polyamide thickeners: (a) MDA and (b) BAMF

From the combination of the reactions between mentioned materials, two samples have shown thickening effects in reaction with castor oil. The bio-based polyamide grease samples with the corresponding share of bio-based carbon atoms are listed in Table 4-3.

Table 4-3: The bio-based polyamide greases

Grease Nr.	Base oil	Thickener	Share of bio C [%]
1	Castor oil	(a) AS-MDA	91
2	Castor oil	(b) DCF-BAMF	100

4.4. Physicochemical characterizations

Physicochemical characterization methods are used to ensure the correct production process of the greases and are utilized as pre-assessment tools for their functionality. In the following, some of the standard methods that have been used to produce the greases will be discussed.

Nuclear Magnetic Resonance (NMR) spectroscopy [98] as well as Fourier Transform Infrared (FTIR) spectroscopy [99] are usually used to ensure that the chemical formulation of the produced grease matches the designed formulation. FTIR spectroscopy identifies the present functional groups such as hydroxyl (-OH), carbonyl (C=O), and amine (NH) in the grease formulation which can be compared to the theoretical designed formulation. Beside identification of the components, NMR spectroscopy also provides information about mobility of the components within the grease matrix (Figure 4-7). Molar mass and the number of repeat units for the developed greases were determined using ^{1}H-NMR spectroscopy through end-group analysis, as described in [92]. A DPX-400 Bruker spectrometer (Berlin, Germany) was employed for this purpose. The results of this characterization methods is not considered in this work and can be found in [91,92,93].

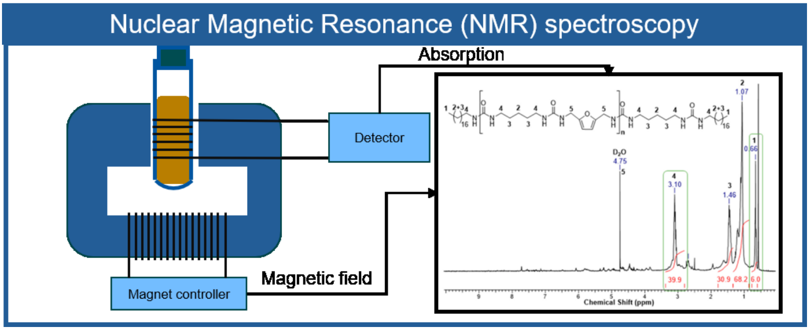


Figure 4-7: Usual physicochemical characterization methods of the developed grease in early phase

The resistance of grease to deformation under loads, known as its consistency, is typically measured using the American Society for Testing and Materials (ASTM) work cone penetration method [100], as outlined in ASTM D 217. This method quantifies the relative hardness of the grease. Greases are classified for application based on their NLGI class, which can range from 1 to 3 in rolling element bearing applications. The NLGI class for the developed bio-based greases was determined by unworked penetration according to DIN ISO 2137, utilizing a PNR 10 Penetrometer from Anton Paar ProveTec (Dahlewitz, Germany). The penetration of a cone into the grease at room temperature was measured and assigned to the respective NLGI class based on the immersion depth.

Greases exhibit viscoelastic behaviour [101], crucial for retaining oil within and releasing it as needed in contact area. Rheometers [102] are commonly employed to examine this viscoelastic behaviour (Figure 4-8). In this setup, grease is sandwiched between two discs that rotate at varying frequencies. The flow limit or yield point is determined by identifying the intersection of the viscous and elastic components of the results. The flow limit of the developed bio-based greases was investigated using an MCR 302 Anton Paar rheometer (Ostfildern, Germany). Oscillation measurements were conducted according to DIN 51810-2 at 25 °C, employing a plate-plate system with a geometry diameter of 25 mm. The flow limit was determined by analysing the intersection of the logarithmically plotted curves from the storage (G´) and loss modulus (G´´). The relevant results of measurements of the developed greases on the rheometer will be discussed in Chapter 5.

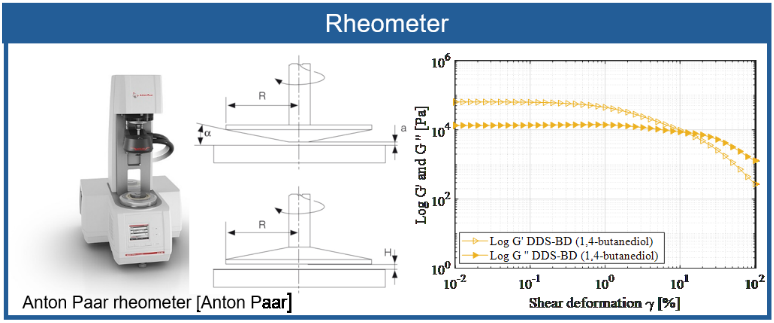


Figure 4-8: Usual physicochemical characterization methods of the developed grease in early phase

As the temperature rises in the application, the grease undergoes a phase change. Each type of grease has a specific temperature at which it transitions from a semi-fluid to a liquid state. This temperature, known as the drop point temperature, signifies the maximum temperature at which the grease can effectively function. Thermal analysis is usually conducted to assess how the greases behave across varying temperatures [103]. Dropping point analysis on the developed bio-based greases was performed using a DP70 instrument from Mettler Toledo (Gießen, Germany) following DIN ISO 2176 standards. The prepared greases were equilibrated at 180 °C and then heated to 300 °C at a rate of 5 °C/min. The dropping point was determined by observing the first drop that crosses an optical square of a camera at an appropriate temperature. The results of this characterization methods is not considered in this work and can be found in [91,92,93].

Tribological evaluations as stated in the state of the research in Chapter 2 are essential to identify mechanical performance of the lubricating greases in machines. In the following, tribological evaluation of develop bio-based greases will be covered.

5. Screening tests on tribometer level

The objective of this chapter is to experimentally validate the proposed first research hypothesis, which is to verify if the film formation of the developed greases is influenced by developed thickeners and to identify any correlation between the greases' tribological performance on tribometer screening results, i.e., film thickness and friction, and the chemical parameters, such as structure, manufacturing method and flow limit. To achieve this, the tribological performance of the developed bio-based greases will be demonstrated through screening tests on a ball-on-disc tribometer. Additionally, the performance of the bio-based greases will be compared to a reference petrochemical grease available on the market. In the end, the results of tribological measurements on the three grease thickeners, polyurea, polyester and polyamides will be summarized, greases will be ranked, and correlated with the chemical structure of the thickener systems.

The description of methods, parts of the results description, and parts of results interpretations in this chapter have already been published in the author's previous publications [92], [93] as well as reported by the author in the final report of the project “Entwicklung biobasierter Verdickersysteme zur Herstellung von Schmierfetten” [90].

5.1. EHD Film thickness measurements

The first important role of the lubricating greases in rolling element bearings is to separate contact partners by forming a thin lubrication film with thickness ranging from nano- to micrometres by releasing the base oil into the single contacts between the rolling elements and the inner and outer raceway. Formation of this thin fluid film plays a crucial role in life time of the rolling element bearing. To evaluate the ability of the greases in terms of film formation in EHD contact, the thickness of the thin film in the contact area for different operational conditions can be measured using available screening methods such as tribometers as a commonly used method as discussed in Chapter 2. Tribometers simulate the situation of single contact of the rolling element bearings in the lab. Among other types ball-on-disc tribometers provide the possibility to evaluate the lubrication characteristics of the developed greases in early phase of grease developments as in the literature [104,105,106,107]. In this work the ball-on-disc EHL-2 tribometer (Figure 5-1) of the PCS instruments (London, United Kingdom) is used to investigate two important aspects of the developed greases: film thickness and friction coefficients in EHD contact.

The film thickness in EHD contact can be directly measured on a tribometer test rig using a glass disc and camera, which allows visualization of the EHD contact area.

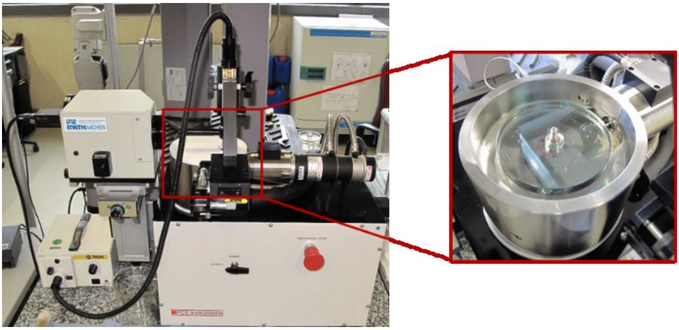


Figure 5-1: EHL-2 ball-on-disc tribometer from PCS Instruments from [92]

To determine the film thickness values, the tribometer test rig benefits from white-light interferometry effects described in [108]. This method was also used in literature [109,105]. Before starting the film thickness measurement, the grease is distributed evenly on the surface of the glass disc with a grease distributer as shown in Figure 5-2. This enables a defined and reproducible grease layer with height of approx. 0.1 mm on disc surface. In the next step, the disc with the grease on the surface was heated up to 40 °C in the heater from Binder GmbH (Tuttlingen, Germany). The tribometer pot was also heated up to 40 °C for 5 min, hence allowing the disc and grease to reach steady temperature.

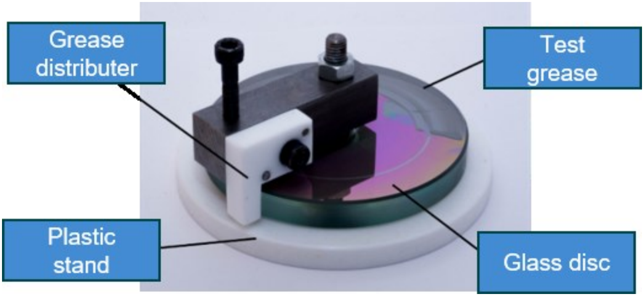


Figure 5-2: Grease distributer for film thickness measurements setup [93]

For the film thickness measurements, normal force on the ball was set to 47 N, which is equivalent to Hertzian pressure of 700 MPa. Slide to Roll Ratio (SRR) is defined as ratio of rolling speed to relative speed of disc and ball, which varies from zero to two and can be demonstrated in percentage. During the film thickness measurements SRR was set to be zero, i.e., pure rolling conditions. As run-in-phase, steel ball was rolled on the disc surface covered with grease layer for 15 min with 30 mm/s, thus distributing uniform grease on the ball path. Rolling speed was increased from 4 to 1000 mm/s in approximately 45 min. The correlation of film thickness and the rolling speed for base oil is known to be linear in double logarithmic diagrams [10]. Therefore, to compare the film thickness of greases over rolling speed with their base oil, film thickness was measured 6 times at 61 rolling speeds with logarithmically equal intervals. Film thickness on each rolling velocity was averaged to avoid any inaccuracy of the measuring device. For each grease, three independent measurements were conducted to ensure reproducibility of the measurement results.

To ensure that the results of measurements are not affected by mixed lubrication conditions, specific film thickness is calculated for the measurements setting as defined in [110,111]:

$$\lambda = \frac{h_0}{\sqrt{\left(R_{q,1}^2 + R_{q,2}^2\right)}} \qquad \text{Equation 5-1}$$

where h_0 is the film thickness and R_q is the root-mean-square roughness of contact bodies. Operational conditions leading to lambda values of higher than 3 are known to ensure separating film thickness in contact, known as fluid regime, as in [110,111]. For given surface roughness values of disc (0.8 nm) and the steel ball (6.1 nm) [112], the minimum film thickness for fluid regime can be calculated. Corresponding film thickness of lambda value of 3 was calculated equal to 18 nm for steel ball and glass disc. For the steel ball on steel disc setting in friction measurements, the minimum film thickness to ensure fluid regime was calculated equal to 23 nm. Therefore, it was assumed that measurements were not affected by the mixed lubrication effects.

Developed greases with bio-based polyurea, polyester and polyamide thickener systems have been considered in EHD film thickness measurements and the results are discussed as following.

5.1.1. Polyurea

In the first set of tribological characterization, the film thickness of three developed greases with polyurea thickener systems was measured varying the rolling speed up to 1000 mm/s. The results of film thickness measurements are shown in Figure 5-3 to Figure 5-6.

As a reference, film thickness measurement of castor base oil with a measured kinematic viscosity of 254 mm²/s at 40 °C is also shown. As stated before, three measurements were averaged to represent the trend of film thickness dependent on rolling speed. The aim of the development of bio-based greases was to identify the bio-based thickener and base oil combinations, which can be used in industry instead of harmful petrochemical based greases and thus reduce harmful environmental influences. Therefore, a petrochemical reference grease available on the market (Berutox FH 28 EPK 2) was proposed by industrial partner (Carl Bechem company). Film thickness measurements were also carried out on the reference grease for comparison to the bio-based greases.

Figure 5-3 shows the results of the lubricant film thickness measurement of polyurea PDI-BAMF at 40 °C and approx. 700 MPa. The lubricating film formation of the castor oil (base oil of the bio-based greases) was also measured on the tribometer and is shown in the diagram. Up to a rolling speed of 50 mm/s, the lubricating film thicknesses of the polyurea grease PDI-BAMF and reference grease Berutox FH 28 EPK 2 are approximately the same. The lubricating gap height of the polyurea grease PDI-BAMF is higher from approx. 50 mm/s compared to the reference grease. From a rolling speed of 100 mm/s, the lubricating film thickness increases with increasing rolling speed, similar to the measured base oil curve. Thus, the contact shows a similar hydrodynamic behaviour as with pure oil lubrication. This can be explained by the so-called bleeding behaviour of the lubricating grease [10].

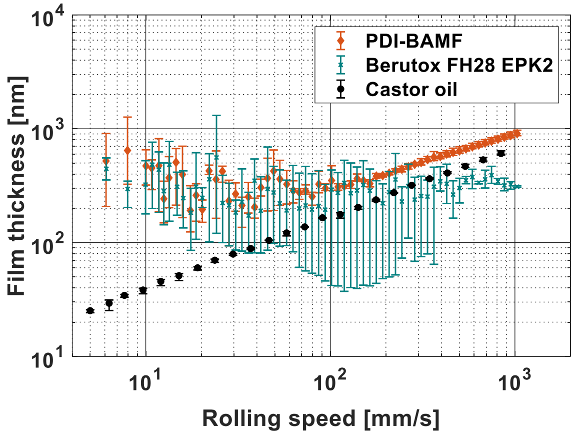


Figure 5-3: Film thickness of bio-based polyurea grease PDI-BAMF over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [92]

This effect describes the releasing of base oil from the thickener matrix during operation. The released base oil leads to approximately the same lubricant film formation as in a test with pure base oil. Therefore, it can be stated that the PDI-BAMF grease shows a good oil bleeding behaviour on the screening test. This should be separately confirmed by specific oil bleeding measurement methods which is out of the scope of this work. The offset between the two data sets can be explained by the thickener layer that additionally forms on the surfaces of the disk and ball [113].

The measurement results of the second polyurea grease PDI-MDA compared to the reference grease, Berutox FH 28 EPK 2, are depicted in Figure 5-4. The film thickness is approximately 400-700 nm lower at lower rolling speeds, up to about 20 mm/s, compared to the reference grease Berutox FH 28 EPK 2. Lower film thickness of the grease compared to the base oil in this region indicates minimal or almost absence thickener effects [56] of the polyurea grease PDI-MDA in lower speeds. Beyond approximately 20 mm/s, the film thickness increases with the rising rolling speed, similar to the test with pure base oil. This can be explained by the film formation of the base oil contained in the grease due to hydrodynamic film formation [114]. From around 150 mm/s onward, the film thickness decreases with further increase in rolling

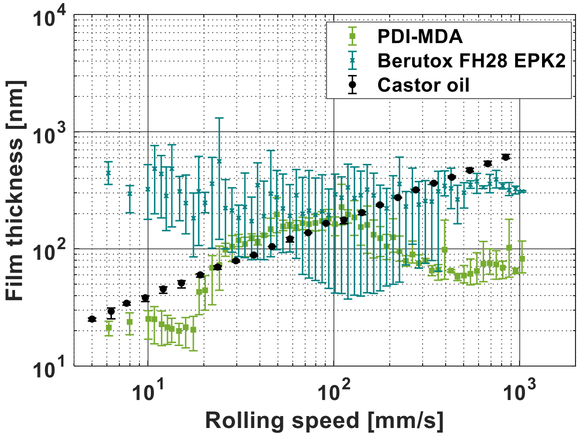


Figure 5-4: Film thickness of bio-based polyurea grease PDI-MDA over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [92]

speed. This effect, known in the literature [115] as starvation, occurs at a characteristic rolling speed when the contact does not receive sufficient lubricant. Insufficient oil quantity increases the risk of solid contact and, consequently, premature bearing failure. From around 70 to 150 mm/s film thickness of the PDI-MDA is comparable to the petrochemical grease. Except for that speed region, the film thickness of this grease is lower than the petrochemical grease.

The third bio-based grease with a polyurea thickener PDI-PDA was also tested against the reference grease Berutox FH 28 EPK 2, and the results are shown in Figure 5-5. The hydrodynamic behavior of polyurea PDI-PDA starts at a speed of 20 mm/s, leading to effective film formation with minimal variability compared to the reference grease Berutox FH 28 EPK 2. This grease does not demonstrate the starvation point below 1000 mm/s. Therefore, it can be stated that the PDI-BAMF grease shows even better oil bleeding behaviour on the screening test compared to the petrochemical reference grease. This should be separately confirmed by further oil bleeding measurements, which is out of the scope of this work.

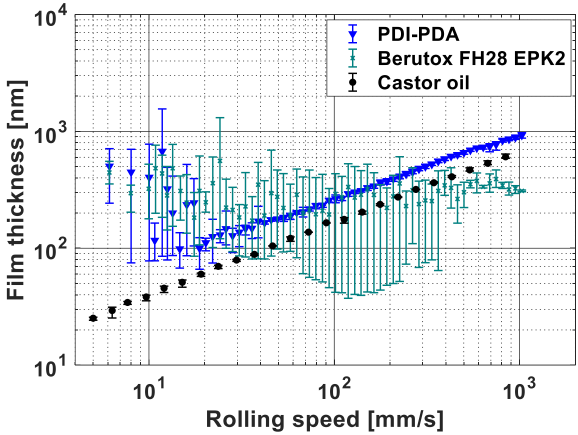


Figure 5-5: Film thickness of bio-based polyurea grease PDI-PDA over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [92]

In Figure 5-6, all three bio-based greases with polyurea thickeners are compared. The results indicate that PDI-BAMF and PDI-MDA exhibit similar behavior in terms of film formation. PDI-BAMF, up to a speed of 40 mm/s, shows a higher film thickness (approximately 100 to 300 nm) compared to PDI-PDA. Due to the comparable results in film formation, both PDI-BAMF and PDI-PDA can be considered suitable thickener candidates for further evaluation of the alternative bio-based lubricating greases with polyurea thickeners.

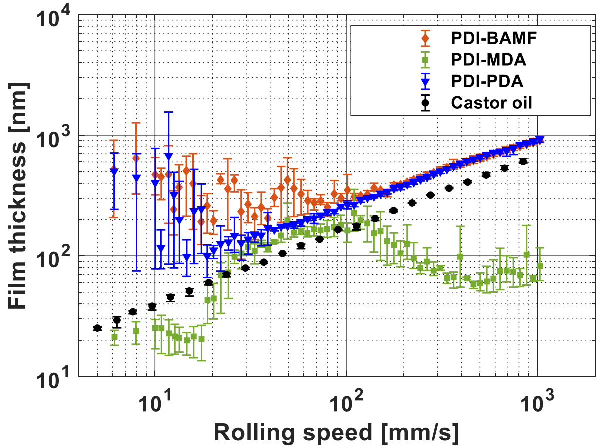


Figure 5-6: Film thickness of bio-based polyurea greases over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [92]

5.1.2. Polyester

Next, the grease samples with bio-based polyester thickeners are evaluated and shown in Figure 5-7 to Figure 5-10. The film thickness at low rolling speeds up to 40 mm/s for the first bio-based polyester grease DDS-BD is around 100 nm lower than film thickness of reference grease as in Figure 5-7. However, the difference in film thickness between the two greases increases by increasing the rolling speed. In higher rolling speeds up to 1000 mm/s, the difference in film thickness between DDS-BD and reference grease reaches the maximum value of around 350 nm. At lower rolling speeds up to 50 mm/s, bio-based grease DDS-BD shows up to around 150 nm higher film thickness than the pure castor oil. This is known as thickener effect due to formation of the deposited-thickener layer on the roller and raceway [56]. From rolling speed of 50 mm/s, the grease DDS-BD shows a lower film thickness compared to the castor oil due to starvation effect starvation and replenishment definition [115,116,117,118]. The starved lubricated contact will then be provided with oil in higher rolling speeds which is called replenishment effects [119,120]. The film thickness of DDS-BD decreases

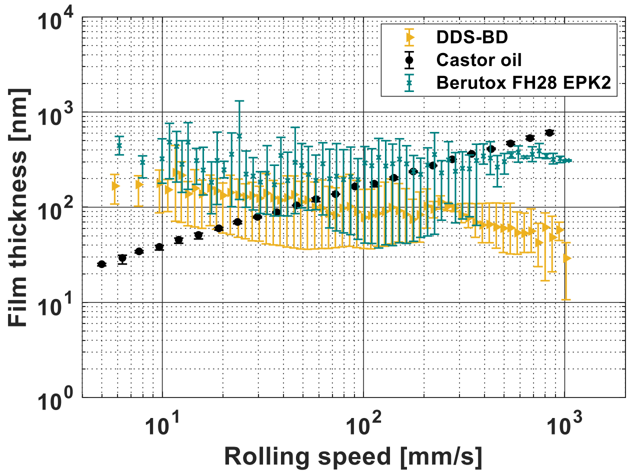


Figure 5-7: Film thickness of bio-based polyester grease DDS-BD over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [93]

down to around 30 nm when the speed is increased from 50 up to 1000 mm/s due to lack of lubricant replenishment in the contact.

Figure 5-8 shows the results of the lubricating film thickness measurement of second bio-based polyester grease with DDS-PrD thickener system. It can be seen that for DDS-PrD, the lubricant film thickness increases continuously with increasing rolling speed. Thus, DDS-PrD shows a similar hydrodynamic behavior as with pure oil lubrication over the rolling speed. It can be seen that at lower rolling speeds up to 200 mm/s, the DDS-PrD has a lower film thickness than the petrochemical reference grease. The effect of the thickener at lower rolling speeds cannot be seen in this grease type. At higher rolling speeds from 200 to 500 mm/s, comparable film thickness of DDS-PrD with reference grease film thickness can be observed. From about 500 to 1000 mm/s, the film thickness of DDS-PrD is around 200 nm higher compared to the reference grease. The starvation point for this polyester grease could also not be observed below 1000 mm/s similar to polyurea greases PDI-PDA and PDI-BAMF.

For the third polyester grease BS-BD, the lubricant film thickness is approximately the same at lower rolling speeds up to about 30 mm/s

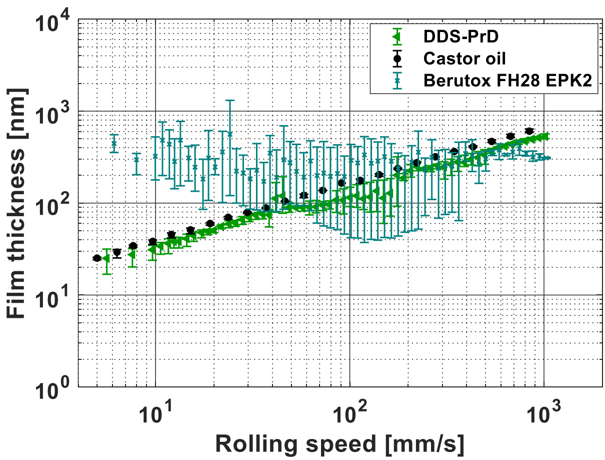


Figure 5-8: Film thickness of bio-based polyester grease DDS-PrD over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [93]

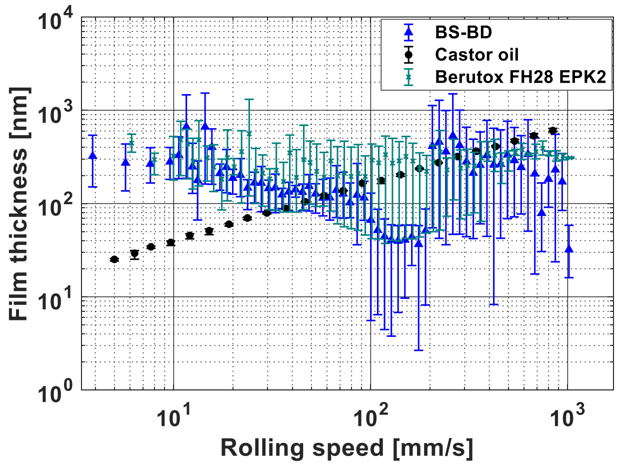


Figure 5-9: Film thickness of bio-based polyester grease BS-BD over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [93]

compared to the reference grease as shown in Figure 5-9. From 30 to 60 mm/s, the lubricant film thickness increases with further increase in rolling speed similar to the castor oil. This effect can be explained by domination of film formation by base oil, thus leading to hydrodynamic behaviour of the grease. From 60 mm/s, starvation occurs and the lubricant film thickness decreases with increasing rolling speed. At higher rolling speeds from 200 mm/s, the results show greater scatter, which can be explained by transition from starved contact to replenishment effects and can thus lead to higher film thickness [119,120]. However, in order to avoid overestimation of the film formation and thus the load carrying capacity of the lubricating film, the lower error bars of the diagram should be considered.

The comparison among all the greases shows that at lower rolling speeds up to about 60 mm/s, the film thickness of DDS-BD and BS-BD is higher than DDS-PrD as in Figure 5-10. This is due to the better thickening effect of DDS-BD and BS-BD at lower speeds. At higher speeds from 60 mm/s, starvation occurred with DDS-BD and BS-BD. Also, it could be observed that the film thickness under starvation is comparable or higher for BS-BD than reference grease. However, for DDS-PrD, the trend is the same as that observed for

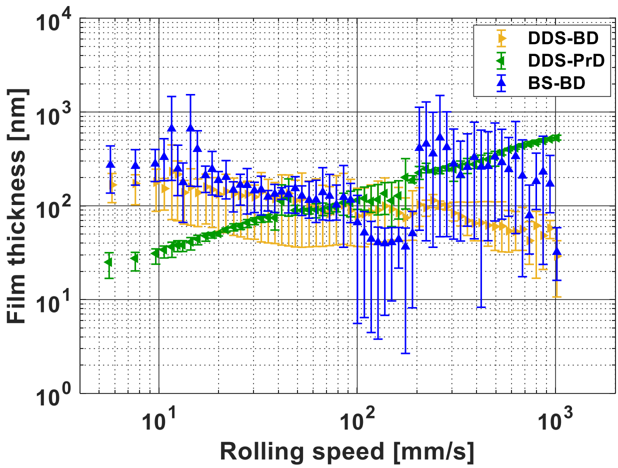


Figure 5-10: Film thickness of bio-based polyester greases over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer from [93]

pure base oil. It can therefore be concluded that DDS-PrD thickener does not perform as expected in other greases.

5.1.3. Polyamide

Next, the grease samples with bio-based polyamide thickeners are measured and the results are demonstrated in Figure 5-11 to Figure 5-13. The results of the lubricating film thickness measurements for the first developed bio-based polyamide AS-MDA and the petrochemical reference grease, Berutox FH 28 EPK 2, are presented in Figure 5-11. The lubricating film thickness of polyamide AS-MDA in is within the same range as that of the reference grease up to 10 mm/s. Beyond 30 mm/s, the lubricating film thickness of polyamide AS-MDA becomes less than that of the base oil, indicating the onset of a starvation effect. Beyond 50 mm/s, the lubricating film thickness of polyamide AS-MDA decreases due to severe boundary lubrication and passage of thickener particles into the contact area.

As depicted in Figure 5-12, the lubricating film thickness of Polyamide DCF-BAMF is higher than that of the base oil and the reference grease,

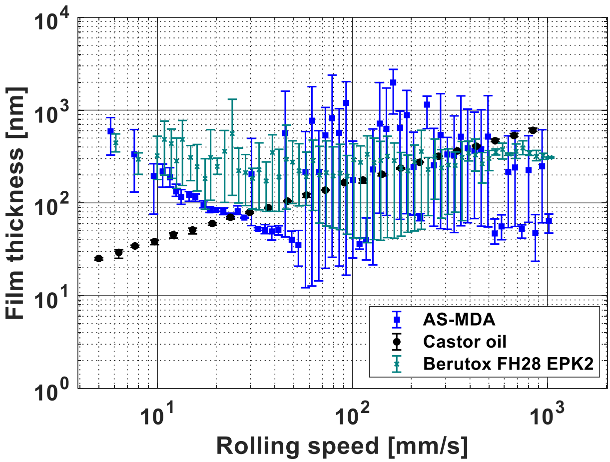


Figure 5-11: Film thickness of bio-based polyamide grease AS-MDA over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer

Berutox FH 28 EPK 2, up to approximately 160 mm/s. Beyond this speed, starvation begins, as evidenced by the increased scattering in lubricating film thickness at higher rolling speeds.

Figure 5-13 illustrates the trends for both polyamide greases. DCF-BAMF exhibits a higher lubricating film thickness compared to AS-MDA at lower rolling speeds up to 160 mm/s, which is advantageous from a tribological perspective.

It is also to mention that both bio-based polyamide greases show high scattering in film thickness results. This is due to the overserved crystal formations in the greases, which was observed during chemical characterizations of the greases as well. This will be discussed more in details in Chapter 5.3.

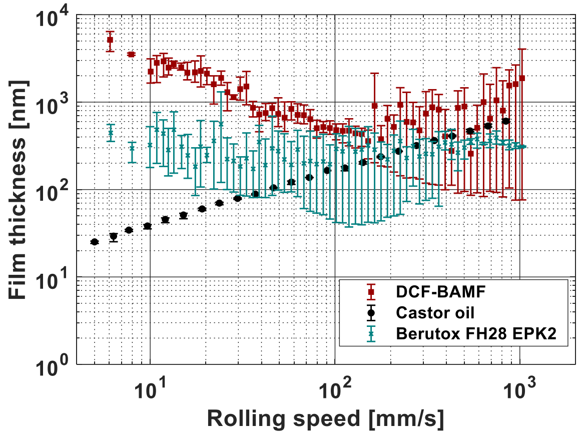


Figure 5-12: Film thickness of bio-based polyamide grease DCF-BAMF over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer

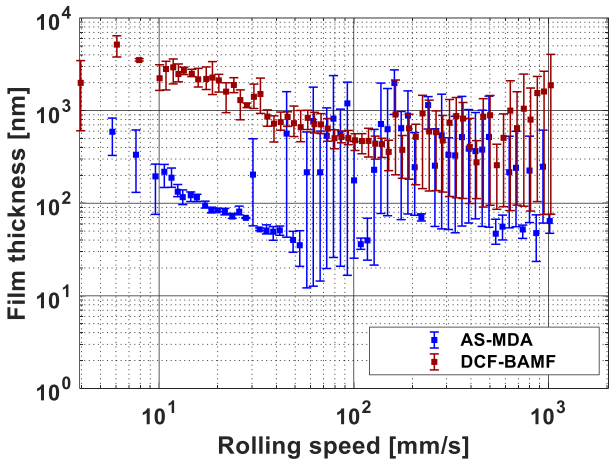


Figure 5-13: Film thickness of bio-based polyamide greases over rolling speed compared to reference grease and castor oil at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 0 on steel ball on glass disc tribometer

5.2. EHD friction measurements

The lubricants play a crucial role in reducing friction forces between contacting surfaces, thereby enhancing the lifespan of machine components. In rolling element bearings, both overall friction loss and longevity are influenced by the type of lubricating oil or grease used. The capacity to decrease friction in contact areas is thus a critical tribological characteristic of lubricating greases. When evaluating tribological performance, the friction coefficient must be considered. This has been already utilized as a standard evaluation of greases in the literature [121].

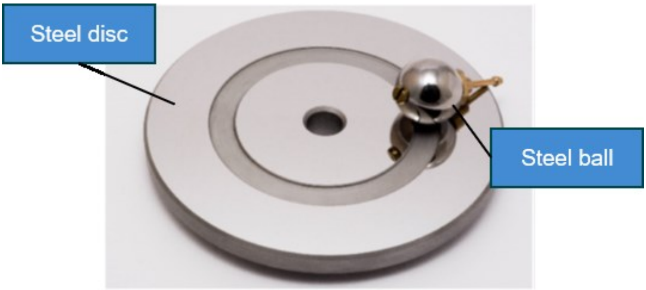


Figure 5-14: Steel ball and steel disc for friction measurement setup of the EHL-2 tribometer of PCS instruments from [93]

To evaluate the bio-based greases in terms of tribological performance in this work, the friction coefficients on EHD contact on steel ball on steel disc of the EHL-2 tribometer (Figure 5-15) were measured by varying the rolling speed. Friction measurements were also conducted on the petrochemical reference grease to have a comparison with the bio-based greases.

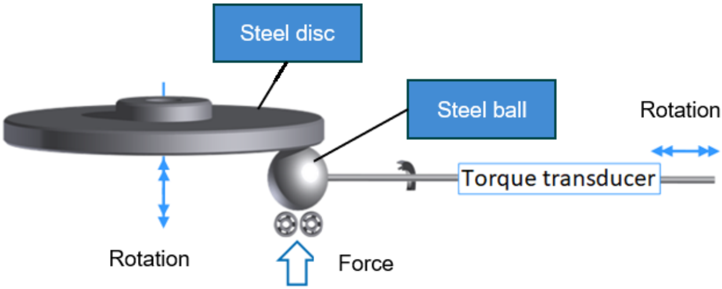


Figure 5-15: Schematic demonstration of the friction measurement setup of EHL-2 tribometer of PCS instruments from [92]

In friction measurements, rolling speed was increased from 9 to 1000 mm/s in 44 equally distributed speed points for constant Slide-to-Roll-Ratio (SRR) for approximately 15 min. For each grease, both positive and negative SRR values were measured, thus considering both scenarios of faster ball rotation than the disc and vice versa. For each SRR value, three independent measurements were conducted (Figure 5-15).

In following friction measurements results on the developed bio-based greases are discussed separately for polyurea, polyester and polyamide greases.

5.2.1. Polyurea

The results of friction measurements of polyurea greases are shown in Figure 5-16. It can be seen that friction coefficient in higher speeds, up to 800 mm/s, was same between bio-based PDI-BAMF and the reference grease. The higher scatter in results was also observed in lower speed due to small grease particles passing through the contact, as was also reported in [109].

The bio-based grease PDI-MDA shows similar friction coefficients to the reference grease up to 30 mm/s in Figure 5-18. In average speeds, from 30 mm/s to 400 mm/s, friction coefficient of the PDI-MDA is lower than the reference grease, and by higher speeds, from 30 mm/s, show both greases again the same friction coefficients.

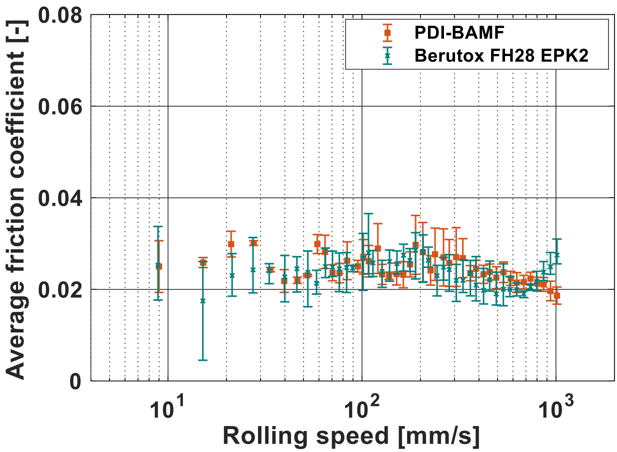


Figure 5-16: Average friction coefficient of bio-based polyurea grease PDI-BAMF over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [92]

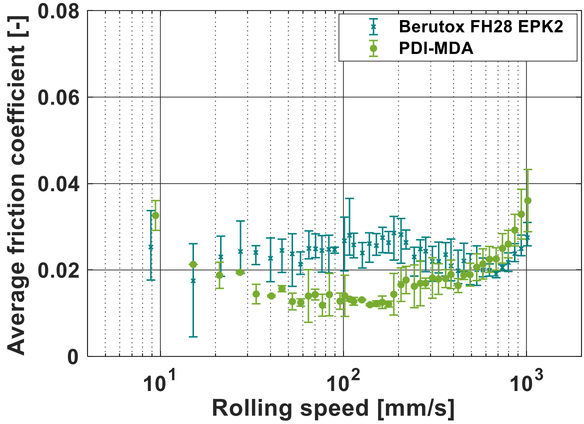


Figure 5-18: Average friction coefficient of bio-based polyurea grease PDI-MDA over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [92]

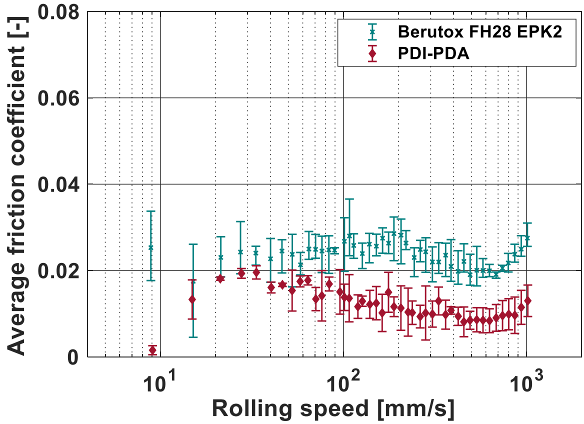


Figure 5-17: Average friction coefficient of bio-based polyurea grease PDI-PDA over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [92]

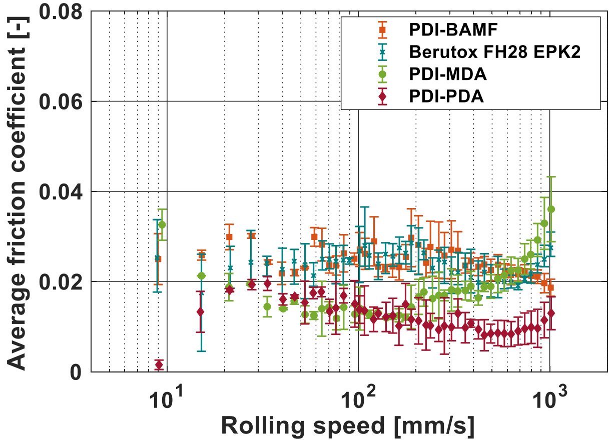


Figure 5-19: Average friction coefficient of bio-based polyurea greases over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [92]

The bio-based greases are compared among each other in Figure 5-19. It can be seen that the friction coefficient of bio-based grease 3 is lower than the reference grease in all speed ranges up to 1000 mm/s. The friction coefficient of PDI-PDA decreases even further with increasing rolling speed. It can be generalized that PDI-MDA results in the smallest friction coefficient. Up to 200 mm/s, PDI-MDA and PDI-PDA lead to similar values.

5.2.2. Polyester

In Figure 5-21, friction coefficient of bio-based grease DDS-BD over rolling speed is compared with reference petrochemical grease. In lower to medium speed range up to about 400 mm/s shows the bio-based grease DDS-BD lower friction coefficient than the petrochemical reference grease. However, in higher rolling speeds from 400 mm/s, friction coefficients of DDS-BD increase sharper than friction coefficients of reference grease, resulting in higher friction coefficients of DDS-BD. This can be correlated with occurrence of starvation of the DDS-BD.

DDS-PrD is compared with reference grease in Figure 5-20. It can be seen that DDS-PrD has relatively lower friction coefficients compared to reference grease in rolling speeds up to around 300 mm/s. From 300 mm/s to 600 mm/s

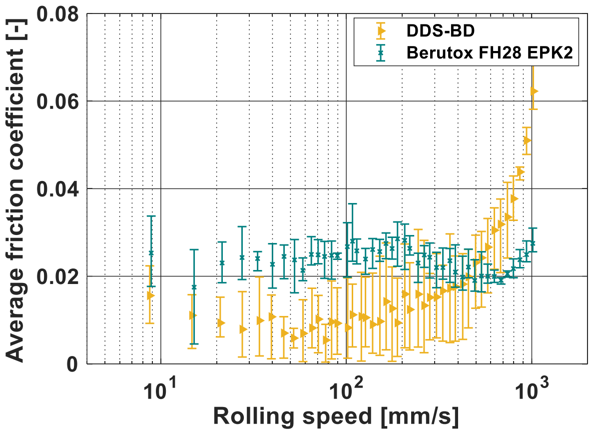


Figure 5-21: Average friction coefficient of bio-based polyester grease DDS-BD over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [93]

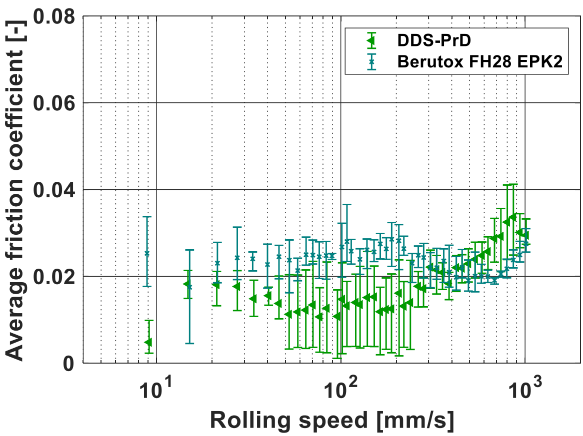


Figure 5-20: Average friction coefficient of bio-based polyester grease DDS-PrD over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [93]

show both greases relatively similar friction coefficients. From 600 mm/s onwards, DDS-PrD grease has around 0.01 higher friction coefficients than reference grease.

Figure 5-22 compares friction coefficients of BS-BD and references grease. In low rolling speeds up to 30 mm/s show both greases similar friction coefficients. However, BD-BD has lower friction coefficients than reference grease from 30 mm/s up to around 300 mm/s. In higher speed region from 300 mm/s, trend reverses and friction coefficient of BS-BD is higher than reference grease.

The order of friction coefficient is similar among the greases as compared in in Figure 5-23; however, DDS-BD shows relatively lower friction coefficient in lower speeds to 50 mm/s. Friction coefficients of DDS-PrD are lowest among the other bio-based polyester greases in higher speeds from about 650 mm/s.

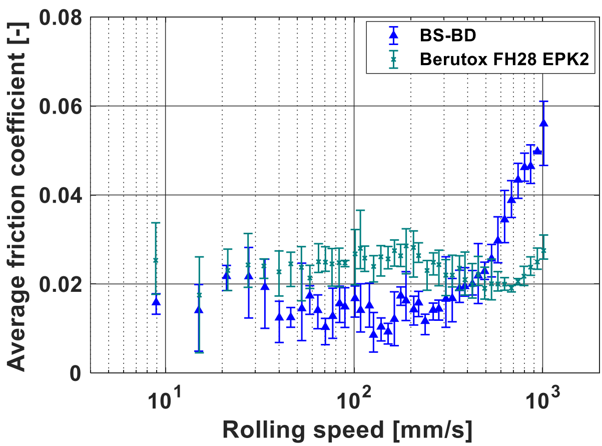


Figure 5-22: Average friction coefficient of bio-based polyester grease BS-BD over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [93]

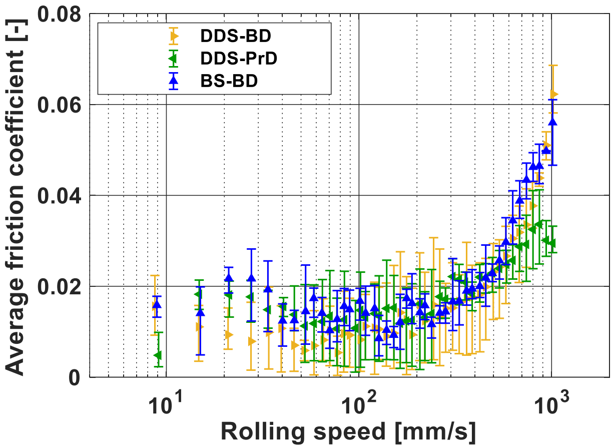


Figure 5-23: Average friction coefficient of bio-based polyester greases over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer from [93]

5.2.3. Polyamide

Figure 5-24 to Figure 5-26 present the results of friction measurements using bio-based polyamide greases. In Figure 5-25, the friction coefficient of bio-based polyamide AS-MDA is compared to the petrochemical reference grease, Berutox FH 28 EPK 2. At low rolling speeds, up to 20 mm/s, the friction coefficients between the reference grease and the bio-based grease are comparable. From 30 mm/s to approximately 200 mm/s, the friction coefficient of polyamide AS-MDA is higher than that of the reference grease. Between 200 mm/s and around 500 mm/s, polyamide AS-MDA and the reference grease exhibit similar friction coefficients. However, at higher rolling speeds, from 500 mm/s to 1000 mm/s, the friction coefficient of polyamide AS-MDA is once again higher than that of the reference grease.

Polyamide DCF-BAMF demonstrates comparable friction coefficients to the reference grease (Figure 5-24). Polyamide AS-MDA exhibits relatively higher friction coefficients at lower rolling speeds, up to around 200 mm/s, and at higher speeds above 500 mm/s when compared to polyamide DCF-BAMF (Figure 5-26).

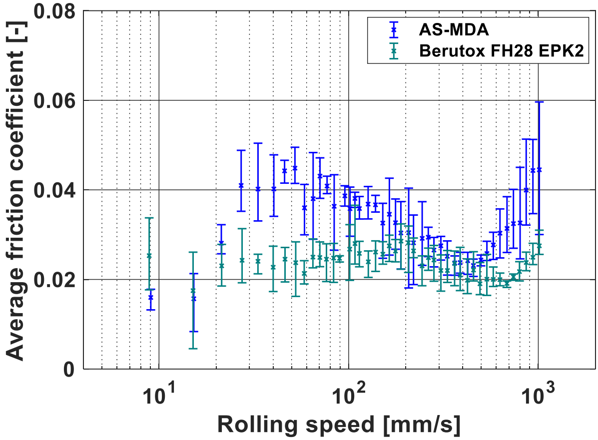


Figure 5-25: Average friction coefficient of bio-based polyamide grease AS-MDA over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer

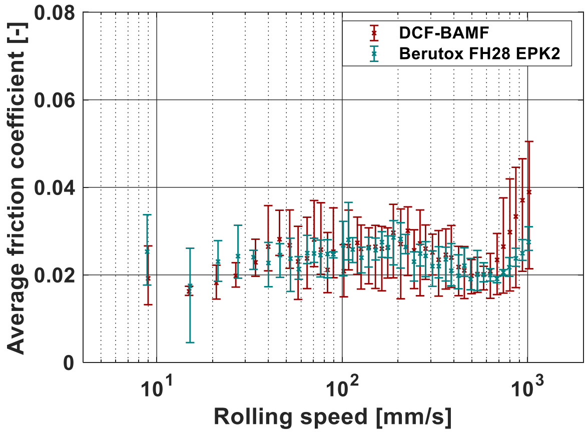


Figure 5-24: Average friction coefficient of bio-based polyamide grease DCF-BAMF over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer

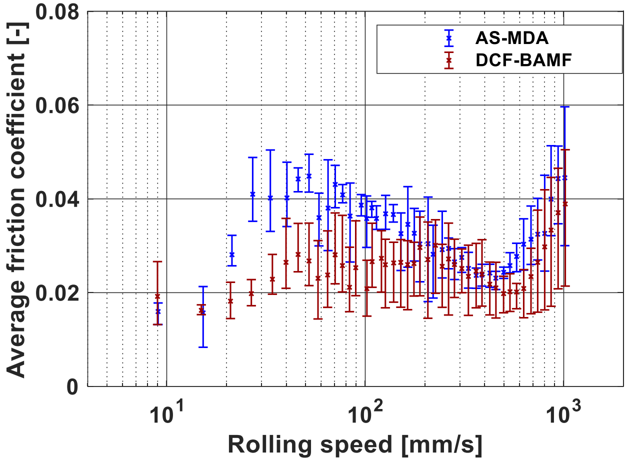


Figure 5-26: Average friction coefficient of bio-based polyamide greases over rolling speed compared to reference grease at Hertzian contact pressure of 700 MPa at 40 °C with SRR = 15% on steel ball on steel disc tribometer

5.3. Correlation of the physicochemical characterization with tribological performance of the bio-based greases

In previous sections, it was shown that the two important tribological parameters on tribometer level namely film thickness and friction coefficient of the developed greases are significantly influenced by the thickener type. In following the physicochemical characterization results of the greases will be correlated with the performance of the greases.

5.3.1. Polyurea

A general conclusion regarding the tribological performance of the developed greases over the entire measured speed range cannot be drawn, as the performance is dependent on operational conditions such as speed. Therefore, the film formation and friction results of the polyurea greases are summarized in Table 5-1.

Table 5-1: Summarized comparison of film thickness (h_0) and friction coefficient (μ) of the polyurea greases measured on ball-on-disc tribometer at 40 °C and 700 MPa

	Rolling speed u_r [mm/s]		
	4 to 200	**200 to 700**	**700 to 1000**
h_0	PDA≈BAMF≻MDA	PDA≈BAMF≻MDA	PDA≈BAMF≻MDA
μ	PDA≈MDA≺BAMF	PDA≺MDA≺BAMF	PDA≺BAMF≺MDA

Higher film thickness and lower friction coefficients are two important parameters to rank the tribological performance of the greases. The film thickness of the PDI-PDA and PDI-BAMF greases is similar to each other and higher than that of the PDI-MDA grease across the entire speed range, indicating superior tribological performance compared to PDI-MDA. (Meaning PDA≈BAMF≻MDA).

To cave the reason behind demonstrated tribological performance of the greases, the results of physicochemical characterizations of the greases in development phase as described in Chapter 4 will be reviewed. The results of physicochemical characterizations have been already published in author's previous publications [92,93] and dissertation of Max Jopen [91].

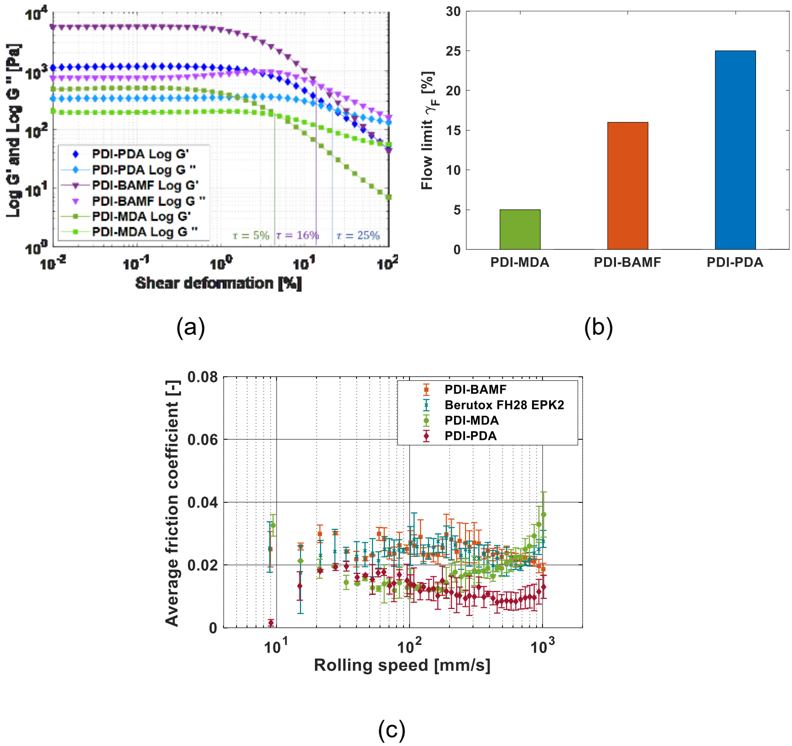


Figure 5-27: Physicochemical characterization of developed polyurea greases on rheometer. (a) results of G' storage module and G'' loss module over shear deformation from [92] (b) summarized flow limit γ_F for the greases (c) the summary of friction measurements on polyurea greases from Chapter 4

Among other methods, the rheological results of measured greases provide understanding about the correlation of chemical structure and lubricating characteristics of the greases. The results of the measurements on rheometer with the polyurea greases are demonstrated in Figure 5-27. The results show the storage module G' which indicates storage of elastic energy, and the loss module G'' that stands for energy dissipation, over shear deformation. Up to a flow limit, lubricating greases exhibit a decisive proportion of elastic deformation ($G''/G' \ll 1$). Above the flow limit, plastic deformation is dominant ($G''/G' \gg 1$). In industrial applications in the bearings, however, greases are always subjected to loads far above the flow limit. Since available plate-plate rheometers are not able to consider higher pressures and shear stresses in lubricating greases to simulate industrial operational conditions in the bearings, the rheological results cannot be directly associated with the film thickness or friction values from measurements on disc-on-ball tribometer. However, the qualitative comparison can be discussed as in following.

The flow limit is the point at which the elastic deformation capacity of the system is reached. Beyond this point, the fibres structure and the bonds to the base oil molecules are broken, causing the grease to begin flowing. This phenomenon is known as thixotropic behaviour of the greases. The rheometer results indicate that the PDI-PDA system has the highest flow limit, followed by PDI-BAMF, and PDI-MDA greases demonstrating the lowest flow limit (PDA≻BAMF≻MDA). It means the PDI-MDA needs the minimum starting shear deformation to break the molecular chains and flow. Since the flow limit has shown to have reverse correlation to the chain mobility [91], it can be stated that the chain mobility of the PDI-MDA is higher. This can be tracked back to so called odd-even effect. Previous studies reported that the tribological behaviour of the greases such as wear and friction is influence also by the odd or even number of the carbon atoms in the chain [122,123], due to deviation of chain mobility of the system [124,125]. The PDI-MDA except for other systems of PDI-BAMF and PDI-PDA, has odd number of carbon atoms and tend to have different flexibility of the molecules showing exceptional behaviours, as have been seen previously for other materials with odd number of carbons [124]. The lower flow limit of the PDI-MDA and PDI-BAMF can also be tracked back into their chemical structure, as PDI-BAMF and PDI-MDA have both aromatic structures which make their structure comparable to Liquid Crystal Polymers (LPCs) [91]. It is known about the LPCs that the higher aromatic compounds lead to better orientation of the system towards flow direction [126], thus higher chain mobility and lower flow limit of the greases.

Looking into morphological parameters of the thickener systems as shown under SEM in Figure 5-28, the polymer fibres in PDI-PDA and PDI-BAMF are more uniformly distributed compared to the PDI-MDA which shows agglomerated particles. It is also to observe that the structure of the thickener systems shows polymer fibres with higher densities in PDI-PDA and PDI-BAMF compared to the PDI-MDA. Comparing with the previously mentioned results in Figure 5-27 (a), it can be seen that he measured storage module G' is the highest for PDI-BAMF and PDI-PDA and lowest for PDI-MDA. In the literature it has been observed that higher storage module G' of the tested greases was correlated with higher density of the thickener network [102]. The higher density of the thickener structure leads to higher porosity of the system. The evaluated performance of the greases (PDA≻BAMF≻MDA) can therefore be correlated with the density and porosity of the polyurea thickener systems (PDA≻BAMF≻MDA). It can be stated that better porosity of the thickener may cause better ability to act as the reservoir of the oil, thus better bleeding behaviour and better tribological performance of the greases.

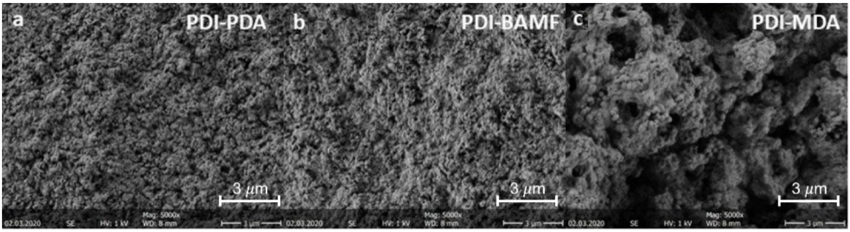


Figure 5-28: Microstructure of the bio-based polyurea greases under scanning electron microscope (SEM): (a)PDI-PDA, (b) PDI-BAMF and (c) PDI-MDA from [91]

5.3.2. Polyester

To correlate the tribological performances with chemical boundary conditions of the polyesters, the results of the tribometer measurements are first summarized for different speed ranges in Table 5-2. It can be seen that except for starvation region (100-200 mm/s), the BS-BD shows comparable film thickness to DDS-BD and higher than DDS-PrD. The friction coefficients of three greases are very close in the middle speed range and only slightly different in low and high-speed regions. Overall, it can be stated that due to superior film formation of the BS-BD in a big speed range, the overall tribological performance of the polyester greases can be ranked as BS-BD≈DDS-BD≻DDS-PrD.

Table 5-2: Summarized comparison of film thickness (h_0) and friction coefficient (μ) of the polyester greases measured on ball-on-disc tribometer at 40 °C and 700 MPa

	Rolling speed u_r [mm/s]		
	4 to 100	**100 to 200**	**200 to 1000**
h_0	BS-BD≈DDS-BD≻DDS-PrD	DDS-PrD ≻DDS-BD≻BS-BD	BS-BD≈DDS-BD≻DDS-PrD
	4 to 60	**60 to 650**	**650 to 1000**
μ	DDS-BD≈DDS-PrD≺BS-BD	DDS-BD≈DDS-PrD≈BS-BD	DDS-BD≈BS-BD≺DDS-PrD

To provide insights about the performance of the bio-based polyesters, analog to polyurea greases the physicochemical characteristics are focused. The results of the measurements on rheometer are demonstrated in Figure 5-29. As it can be seen the highest flow limit is shown by BS-BD and then DDS-BD system. DDS-PrD systems shows the lowest flow limit among other systems (BS-BD≻DDS-BD≻DDS-PrD).

To correlate the results, DDS-BD and BS-systems are focused first. Although film thickness values of BS-BD over rolling speeds show similar results to DDS-BD film thickness, minimal differences are captured. In comparison to DDS-BD, BS-BD shows relatively higher film thickness. The lower film thickness and minimal decrease in performance of DDS-BD over BS-BD can

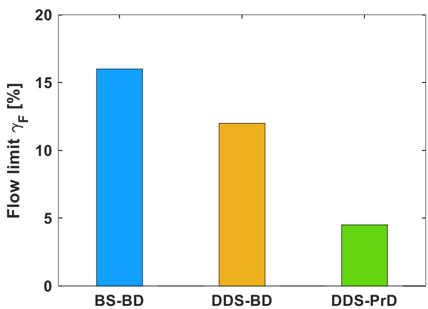


Figure 5-29: Summarized flow limit γ_F of physicochemical characterization on rheometer with developed polyester greases from [92]

be contributed to the higher number of CH2 groups in DDS-BD with the same diol, thus higher crystallization potential and reducing the chain mobility. By higher crystallization potentials, the amount of bleeding oil from thickener released to the contact can be decreased. Consequently, the film thickness in contact area will decrease, thus increasing the risk of solid contact and decreasing in tribological performance [93]. It can also be seen that the flow limit decreases with higher number of CH2 group (DDS-BD > BS-BD).

In comparison to DDS-BD, DDS-PrD shows different behaviour in film thickness over the rolling speed diagram; even number of CH2 groups [127] in the dicarboxylic acid and the odd number of CH2 groups in the diol DDS-PrD can be responsible for the different behaviour. However, to clarify special behaviour of DDS-PrD, further physicochemical investigations are needed.

It can be observed that analog to the discussion for polyurea greases, the tribological performance of the polyester greases are in qualitative correlation with flow limits from rheometer measurements.

In terms of morphology of the greases, SEM images of the polyester thickeners are shown in comparison to polyurea thickener in Figure 5-30. In polyester greases, almost no self-assembled microstructure of the polyesters

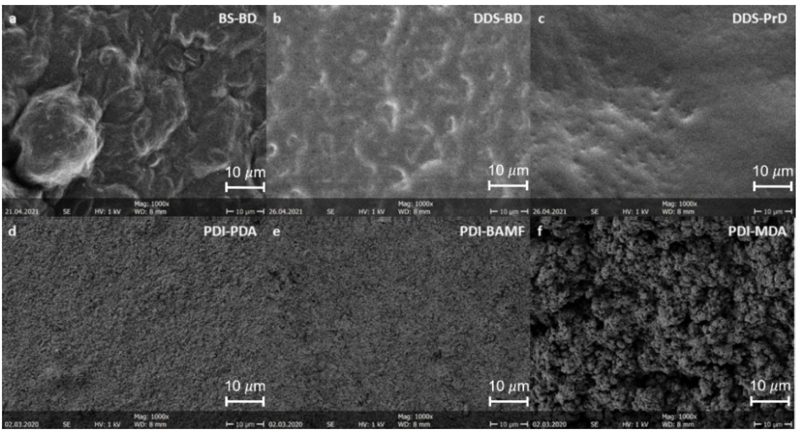


Figure 5-30: Microstructure of the bio-based polyester greases (a-c) compared to polyurea grease (d-f) under scanning electron microscope (SEM) from [91]: (a)BS-BD, (b) DDS-BD, (c) DDS-PrD, (d) PDI-PDA, (e) PDI-BAMF and (f) PDI-MDA

compared to polyurea thickeners could be seen. BS-BD exhibits more structure than DDS-BD, while DDS-PrD shows almost no structure and a flat surface. This can be correlated with previously discussed flow limits in rheometer (Figure 5-29) and oil bleeding abilities. The reason for lower foam-like structure of the polyester thickeners is the lack of chemical bonds between thickener molecules to form the structure. In polyurea thickeners the hydrogen bonds play an important role in thickener chains however, in polyesters there are not free NH and OH groups to form such bonds. Therefore, physical interactions between thickener fibers and base oil are more plausible. Since the castor oil is a polar oil, the interaction of the base oil with the thickener polymer fibers is responsible for formation of the gel-like structure of the polyester greases and not the foam like structure of the thickener [91,93].

5.3.3. Polyamide

To correlate the tribological performances with chemical boundary conditions of the polyamides, the results of the tribometer measurements are first summarized for different speed ranges in Table 5-3. To emphasize on the trend of tribological performance of the greases, the few exceptional points

Table 5-3: Summarized comparison of film thickness ($\boldsymbol{h_0}$) and friction coefficient ($\boldsymbol{\mu}$) of the polyamide greases measured on ball-on-disc tribometer at 40 °C and 700 MPa

	Rolling speed u_r [mm/s]	
	4 to 150	**150 to 1000**
$\boldsymbol{h_0}$	DCF-BAMF≻AS-MDA	DCF-BAMF≈AS-MDA
$\boldsymbol{\mu}$	DCF-BAMF≺AS-MDA	DCF-BAMF≈AS-MDA

which show different film and frictions are not considered in the Table 5-3. The tribological performance of the DFC-BAMF is shown to be comparable or superior to the AS-MDA (DCF-BAMF ≻AS-MDA).

Flow limit of the polyamides are also shown in the Figure 5-31. It can be seen that the flow limit of the DCF-BAMF is higher than AS-MDA. It was observed that the tribological performance hence show the same trend as the flow limit from rheometer measurements.

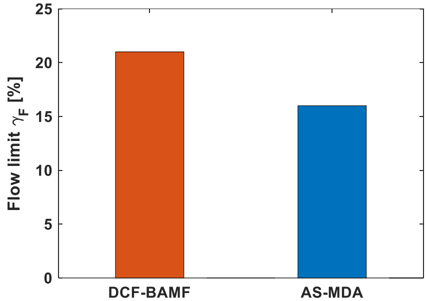


Figure 5-31: Summarized flow limit γ_F of physicochemical characterization on rheometer with developed polyamide greases from [92]

SEM images reveal the morphology of thickener particles in the polyester grease in comparison to the polyurea in Figure 5-32. It can be seen that AS-MDA and DCF-BAMF thickeners demonstrate structure which are comparable from the general shape to the polyurea thickeners. This is due to the ability of the chemical structure of the polyamide thickeners to build hydrogen bonds between thickener particles [91], thus forming microstructures analog to

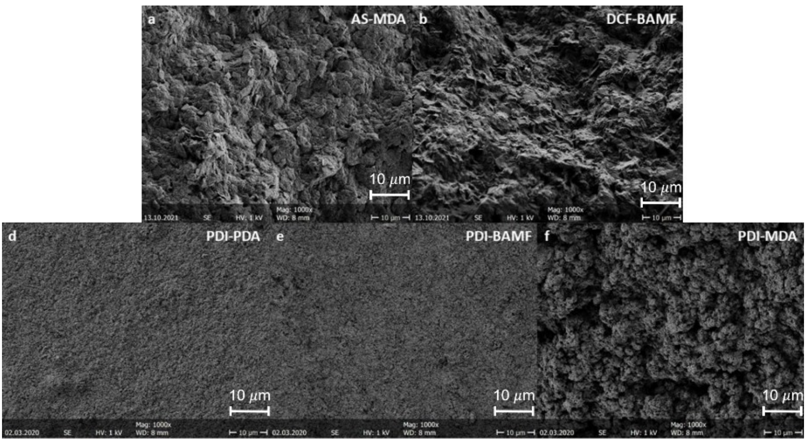


Figure 5-32: Microstructure of the bio-based polyamide greases (a-c) compared to polyurea grease (d-f) under scanning electron microscope (SEM) from [91]: (a)AS-MDA, (b) DCF-BAMF, (c) PDI-PDA, (d) PDI-BAMF and (e) PDI-MDA

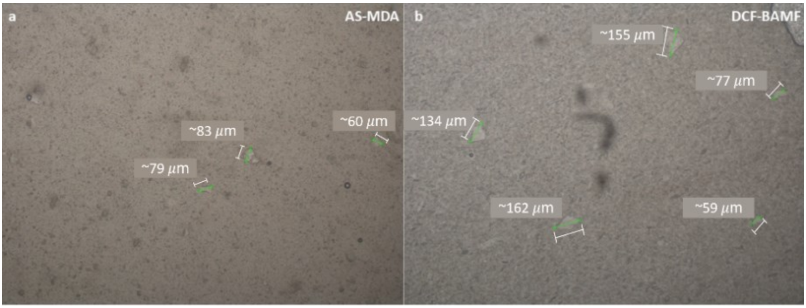


Figure 5-33: Light microscopy of the bio-based polyamide greases and crystal formation from [91]: (a) AS-MDA, (b) DCF-BAMF

polyurea formation. However, polyamide thickeners show significantly bigger polymeric fibers. Furthermore, compared to the polyurea thickeners, the polyamides do not show pores in the structure.

In the polyamide greases after a few weeks at room temperature, formation of macroscopic crystals was observed. Agglomerated grease particles (Figure 5-3) were formed which were observed by [91]. In grease lubrication, these agglomerated particles pass through the contact area causing random film thickness depending on the size of the particles. This phenomenon was already observed in film thickness and friction of both polyamide greases with high scattering of the results. This disturbs the film formation of the greases significantly which may cause sometimes very low film thickness and lead to higher chance of asperity contacts in the bearings. It is mentioned by [91] that the crystal formation can be accelerated with higher temperatures. Since the temperature in the EHD contact is raised in normal applications that means this effect will disturb the lubrication more significantly making the developed bio-based greases as not suitable for rolling element bearing applications.

While the base oil is the same between developed bio-based polyurea, polyester and polyamide greases in this work the manufacturing and processing methods vary, as do the physical interactions and the superstructure. Polyureas are known to form self-assembling superstructures [128]. This takes place mainly via hydrogen bonds between the urea units. The oil is incorporated in the thickener network. For the polyesters, this type of superstructure formation is rather unlikely, since they cannot form self-assembly via hydrogen bonds due to their chemical structure. These systems must therefore be based on a different mechanism. Hydrogen bonds to the polar castor oil (free OH group) would be conceivable here. The network would therefore be influenced by oil-thickener interactions rather than by thickener-thickener interactions, as is the case of polyurea. The thickener would thus have an overall higher contribution to the total lubrication of the polyesters. Therefore, the thickener particles can be detached from the structure and carried into the contact area with the base oil by higher shear forces, thus resulting in higher scatters in higher speeds in film-thickness results. In polyamides the superstructures are also formed, due to the ability to form hydrogen bonds. The primary interaction forces between the thickener and oil in polyamide greases are also hydrogen bonds. Overall show polyurea greases superior tribological performance on the tribometer to other thickener types which could be successfully tracked back into chemical structure and agrees with physicochemical findings in development phase.

5.4. Chapter conclusion

The primary objective of chapter 5 was to experimentally validate the first proposed research hypothesis:

1. The film formation of bio-based greases on tribometer level is significantly influenced by the thickener type and can be correlated with chemical parameters of thickener, such as structure, and flow limit.

Therefore, tribological performance of the developed bio-based greases were evaluated regarding film thickness and friction on screening tests on a ball-on-disc tribometer.

The results of tribological evaluation in terms of the film thickness and friction showed significant deviations between greases with polyurea, polyester and polyamide thickeners. The results showed superior performance of polyurea and polyesters over polyamide thickeners. While polyamides exhibit partially acceptable performance in tribological tests at lower speeds, they are considered unsuitable for rolling element bearing applications due to the formation of crystals inside the grease and disrupted film formation at middle to higher speeds. The polyurea system is shown to be superior to other systems with highest film thickness and lowest friction coefficients. By correlation of the tribological results with the chemical structure and physicochemical characterization results, it could be concluded that the formation of a foam-like thickener system is essential for suitable performance of the greases. A significant effect of chemical parameters such as flow limit of grease and microstructure of the thickeners as well as chemical bounds in thickener structure on the tribological performance of the lubricating greases were observed. **These observations proved the first proposed hypothesis of this work** and confirmed that the thickener type and chemical structure of the thickener can be correlated with the tribological performance of the greases at the tribometer level regarding film formation and friction. By evaluating the film thickness and friction, the thickener system with the best tribological performance, i.e., polyurea thickener, was identified.

Next objective of Chapter 5 was to evaluate the comparability of the developed bio-based greases with a petrochemical grease available on the market on tribometer screening tests. Therefore, film formation and friction coefficients of the developed bio-based greases were compared to a reference petrochemical grease. Among bio-based polyurea samples, PDI-PDA and PDI-BAMF systems showed comparable tribological performance to the petrochemical reference grease. From bio-based polyester systems, BS-BD was shown to be comparable with the reference grease in terms of film formation and friction. Bio-based polyamide thickeners were not able to pass the tests and therefore not comparable to the petrochemical reference grease.

Bio-based polyurea thickener showed the best performance on the tribometer evaluations among the other systems as well as comparable to petrochemical reference grease. Therefore, it was selected for further evaluations. In the next chapter, the tribological performance of the selected candidates of bio-based polyurea will be further investigated at the component level.

6. Evaluation on rolling element bearing component level

The objective of this chapter is to experimentally validate the second proposed research hypothesis, that is, to determine whether the results of tribological screening tests on film thickness and friction at the tribometer level can be correlated and confirmed by the results of rolling element bearing component tests. To achieve this, the tribological performance of two selected greases (PDI-BAMF and PDI-MDA) with the bio-based polyurea thickener system, as the best identified bio-based thickener system, will be demonstrated regarding wear protection on the axial bearing test rig (FE8) and lifetime on the radial bearing test rig component tests. Ultimately, the results of component measurements will be correlated with the tribometer results at the contact level. Additionally, the performance of the bio-based greases will be compared to the reference petrochemical grease to rank the greases.

The description of methods, parts of the results descriptions and images, and interpretations in this chapter have already been reported by the author in the final report of the project “Entwicklung biobasierter Verdickersysteme zur Herstellung von Schmierfetten” [90], by German FNR agency.

6.1. Evaluation of the performance on short time measurements on axial rolling element bearings

Lubricating greases are applied in rolling element bearings for various reasons, including minimizing asperity contacts and protecting the contact bodies against wear. Previous results on film formation and friction on the tribometer are insufficient for evaluating the overall performance of the developed greases, as they only pertain to single-contact performance. To address the third hypothesis, the overall performance of the developed bio-based greases must be evaluated at the component level. Evaluation of the greases regarding wear protection has been already suggested [129] and conducted [130,131] on FE8 testing system.

Following the promising outcome of bio-based polyurea greases from Chapter 5, two candidates (PDI-BAMF and PDI-MDA) were chosen for the scale-up process, whereas PDI-PDA was not upscaled. The scaled-up greases included additives similar to those of the petrochemical reference grease. Carl Bechem Company conducted the scale-up of the selected samples as part of the mutual project [90]. Due to technical limitations a new method had to be developed to produce polyesters. However, the method for production of polyurea greases could be integrated based on the existing reactors. For more information the reader is referred to the project report. In following, the wear protection characteristics of scaled-up polyurea samples will be demonstrated.

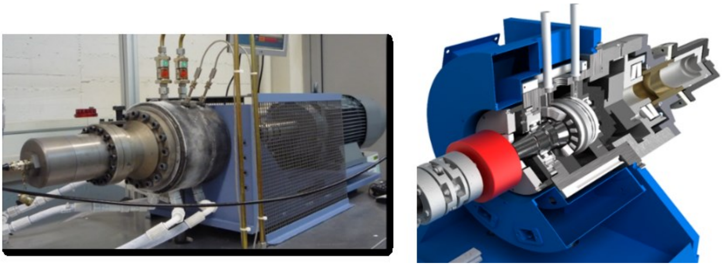


Figure 6-1: Axial bearing test rig FE8

The wear tests were carried out on the FE8 axial bearing test rig Figure 6-1 according to DIN 51819-3 (standard operating conditions for wear measurements at 7.5 min^{-1} and 80 KN and 80 °C for 80 hours) greases in order to evaluate the manufactured bio-based greases with regard to their wear protection properties. In the test rig, two thrust bearings of type 81212 (Figure 6-2) were used as test bearings, which were relubricated with 50 mm^3 of grease every 10 minutes during the measurements using a grease pump.

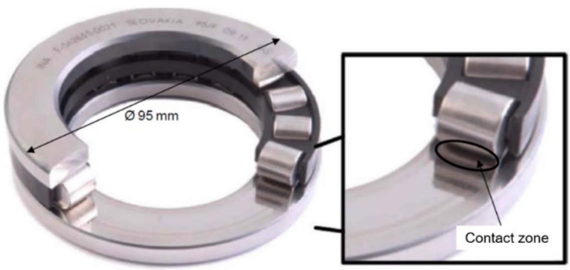


Figure 6-2: Axial bearings 81212 with polyamide cage as test bearings for FE8 measurements

Before and after the tests, the axial bearings were cleaned and the surface roughness measurements were carried out on the surface of the raceways and the rolling elements at three marked points. From the roughness measurements, the characteristic values (the arithmetic mean roughness value (R_a), the square mean roughness value (R_q) and the average roughness depth (R_z)) were determined in accordance with DIN EN ISO 4287. In addition, the bearings were weighed before and after the tests to calculate the wear mass as the parameter to evaluate wear protection characteristics of the greases. During the tests, the temperature and torque were continuously measured on the test rig. Wear tests on the FE8 rolling bearing test was carried out for PDI-BAMF and PDI-MDA, as well as the petrochemical reference grease, Berutox FH 28 EPK 2. The measurement setup was heated to 80 °C up to steady state and then the measurements were conducted.

6.1.1. Results and discussions

After 80 hours of testing, the samples were removed from the test stand, cleaned, and subjected to surface and weight measurements. Figure 6-3 to Figure 6-5 present the results of surface roughness measurements on the shaft side (WS), the coupling side (KS), and on the rolling elements (WK) before and after the FE8 test. The average deviations in the roughness values of the roller after the test are quite minimal for reference grease, on the order of 0.002 µm for R_a, indicating very slight alterations on the surfaces.

	Before	Ra	Rq	Rz
Bearing 1	WS point 1	0.060	0.070	0.420
	WS point 2	0.060	0.070	0.370
	WS point 3	0.060	0.080	0.420
	Average	0.060	0.073	0.403
	GS point 1	0.070	0.090	0.440
	GS point 2	0.060	0.080	0.410
	GS point 3	0.060	0.080	0.430
	Average	0.063	0.083	0.427
	WK 1	0.050	0.070	0.380
	WK 2	0.060	0.070	0.390
	WK 3	0.060	0.070	0.400
	Average	0.057	0.070	0.390

	After	Ra	Rq	Rz
Bearing 1	WS point 1	0.050	0.060	0.310
	WS point 2	0.040	0.060	0.320
	WS point 3	0.040	0.050	0.290
	Average	0.043	0.057	0.307
	GS point 1	0.060	0.070	0.370
	GS point 2	0.060	0.080	0.390
	GS point 3	0.060	0.070	0.390
	Average	0.060	0.073	0.383
	WK 1	0.050	0.070	0.350
	WK 2	0.050	0.060	0.420
	WK 3	0.050	0.070	0.380
	Average	0.050	0.067	0.383

Difference bearing 1

Difference	Ra	Rq	Rz
WS	-0.017	-0.017	-0.096
GS	0.003	-0.010	-0.043
WK	-0.007	-0.003	-0.007

	Before	Ra	Rq	Rz
Bearing 2	WS point 1	0.060	0.080	0.420
	WS point 2	0.070	0.080	0.410
	WS point 3	0.050	0.070	0.360
	Average	0.060	0.077	0.397
	GS point 1	0.080	0.090	0.460
	GS point 2	0.060	0.070	0.380
	GS point 3	0.060	0.070	0.400
	Average	0.067	0.077	0.413
	WK 1	0.040	0.050	0.360
	WK 2	0.040	0.050	0.370
	WK 3	0.030	0.040	0.280
	Average	0.037	0.047	0.337

	After	Ra	Rq	Rz
Bearing 2	WS point 1	0.040	0.050	0.260
	WS point 2	0.040	0.050	0.310
	WS point 3	0.050	0.060	0.340
	Average	0.043	0.053	0.303
	GS point 1	0.050	0.06	0.340
	GS point 2	0.040	0.05	0.290
	GS point 3	0.040	0.05	0.320
	Average	0.043	0.053	0.317
	WK 1	0.040	0.050	0.300
	WK 2	0.050	0.060	0.330
	WK 3	0.050	0.060	0.430
	Average	0.047	0.057	0.353

Difference bearing 2

Difference	Ra	Rq	Rz
WS	-0.017	-0.023	-0.093
GS	-0.023	-0.023	-0.097
WK	0.010	0.010	0.017

Average	Ra	Rq	Rz
Bearing 1 & 2 WK	0.002	0.003	0.005

Figure 6-3: Surface roughness (R_a,R_q and R_z) before and after FE8 wear tests on the test axial bearing 81212 with reference grease Berutox FH 28 EPK 2 on the shaft side (WS), the coupling side (GS), and on the rolling elements (WK)

Before		Ra	Rq	Rz
Bearing 1	WS point 1	0.060	0.070	0.380
	WS point 2	0.050	0.060	0.330
	WS point 3	0.060	0.070	0.350
	Average	0.057	0.067	0.353
	GS point 1	0.050	0.070	0.430
	GS point 2	0.050	0.070	0.360
	GS point 3	0.060	0.070	0.370
	Average	0.053	0.070	0.387
	WK 1	0.040	0.050	0.360
	WK 2	0.050	0.070	0.380
	WK 3	0.050	0.060	0.380
	Average	0.047	0.06	0.373

After		Ra	Rq	Rz
Bearing 1	WS point 1	0.050	0.070	0.370
	WS point 2	0.060	0.080	0.410
	WS point 3	0.050	0.060	0.370
	Average	0.053	0.070	0.383
	GS point 1	0.050	0.070	0.390
	GS point 2	0.060	0.070	0.430
	GS point 3	0.060	0.070	0.390
	Average	0.057	0.070	0.403
	WK 1	0.040	0.060	0.320
	WK 2	0.040	0.050	0.280
	WK 3	0.040	0.050	0.310
	Average	0.040	0.053	0.303

Difference bearing 1

Difference	Ra	Rq	Rz
WS	-0.003	0.003	0.030
GS	0.003	0	0.017
WK	**-0.007**	**-0.007**	**-0.070**

Before		Ra	Rq	Rz
Bearing 2	WS point 1	0.050	0.070	0.330
	WS point 2	0.050	0.070	0.330
	WS point 3	0.060	0.070	0.350
	Average	0.053	0.070	0.337
	GS point 1	0.080	0.100	0.550
	GS point 2	0.060	0.080	0.470
	GS point 3	0.070	0.090	0.460
	Average	0.070	0.090	0.493
	WK 1	0.060	0.070	0.430
	WK 2	0.070	0.080	0.490
	WK 3	0.050	0.070	0.340
	Average	0.060	0.073	0.420

After		Ra	Rq	Rz
Bearing 2	WS point 1	0.040	0.050	0.260
	WS point 2	0.040	0.050	0.260
	WS point 3	0.050	0.070	0.450
	Average	0.043	0.057	0.323
	GS point 1	0.050	0.070	0.370
	GS point 2	0.050	0.070	0.360
	GS point 3	0.050	0.070	0.400
	Average	0.050	0.070	0.377
	WK 1	0.060	0.070	0.370
	WK 2	0.050	0.060	0.350
	WK 3	0.050	0.060	0.350
	Average	0.053	0.063	0.357

Difference bearing 2

Difference	Ra	Rq	Rz
WS	-0.010	-0.013	-0.013
GS	-0.020	-0.020	-0.117
WK	**-0.007**	**-0.010**	**-0.063**

Average	Ra	Rq	Rz
Bearing 1 & 2 WK	**-0.007**	**-0.008**	**-0.067**

Figure 6-5: Surface roughness (R_a,R_q and R_z) before and after FE8 wear tests on the test axial bearing 81212 with bio-based grease PDI-BAMF on the shaft side (WS), the coupling side (GS), and on the rolling elements (WK)

Before		Ra	Rq	Rz
Bearing 1	WS point 1	0.040	0.050	0.250
	WS point 2	0.040	0.050	0.300
	WS point 3	0.040	0.050	0.270
	Average	0.040	0.050	0.273
	GS point 1	0.050	0.060	0.310
	GS point 2	0.050	0.060	0.330
	GS point 3	0.050	0.060	0.330
	Average	0.050	0.060	0.323
	WK 1	0.030	0.040	0.190
	WK 2	0.030	0.040	0.250
	WK 3	0.030	0.050	0.270
	Average	0.030	0.043	0.237

After		Ra	Rq	Rz
Bearing 1	WS point 1	0.040	0.050	0.310
	WS point 2	0.050	0.060	0.320
	WS point 3	0.040	0.050	0.290
	Average	0.043	0.053	0.307
	GS point 1	0.040	0.050	0.290
	GS point 2	0.050	0.060	0.340
	GS point 3	0.050	0.060	0.320
	Average	0.047	0.057	0.317
	WK 1	0.060	0.080	0.370
	WK 2	0.060	0.060	0.280
	WK 3	0.060	0.070	0.370
	Average	0.06	0.070	0.340

Difference bearing 1

Difference	Ra	Rq	Rz
WS	0.003	0.003	0.033
GS	-0.003	-0.003	-0.007
WK	**0.030**	**0.027**	**0.103**

Before		Ra	Rq	Rz
Bearing 2	WS point 1	0.030	0.040	0.220
	WS point 2	0.030	0.040	0.230
	WS point 3	0.030	0.040	0.200
	Average	0.030	0.040	0.217
	GS point 1	0.030	0.040	0.170
	GS point 2	0.040	0.050	0.280
	GS point 3	0.040	0.050	0.290
	Average	0.037	0.047	0.247
	WK 1	0.030	0.040	0.250
	WK 2	0.030	0.040	0.240
	WK 3	0.030	0.040	0.280
	Average	0.030	0.040	0.257

After		Ra	Rq	Rz
Bearing 2	WS point 1	0.050	0.0600	0.330
	WS point 2	0.040	0.0600	0.360
	WS point 3	0.040	0.0600	0.310
	Average	0.043	0.060	0.333
	GS point 1	0.050	0.060	0.360
	GS point 2	0.040	0.050	0.290
	GS point 3	0.040	0.060	0.310
	Average	0.043	0.057	0.322
	WK 1	0.050	0.070	0.430
	WK 2	0.050	0.060	0.330
	WK 3	0.040	0.060	0.340
	Average	0.047	0.063	0.367

Difference bearing 2

Difference	Ra	Rq	Rz
WS	0.013	0.020	0.117
GS	0.007	0.010	0.073
WK	**0.017**	**0.023**	**0.110**

Average	Ra	Rq	Rz
Bearing 1 & 2 WK	**0.023**	**0.025**	**0.107**

Figure 6-4: Surface roughness (R_a,R_q and R_z) before and after FE8 wear tests on the test axial bearing 81212 with bio-based grease PDI-MDA on the shaft side (WS), the coupling side (GS), and on the rolling elements (WK)

For the bearings lubricated with PDI-BAMF grease the average deviation of the surface roughness R_a, on the rolling element is also quite small equal to 0.007 µm. For PDI-MDA however, the averaged R_a is 0.023 µm which is slightly higher than PDI-BAMF and the reference grease.

Additionally, the surfaces of the raceways and rolling elements were examined under a light microscope after the measurements. The results of the optical examination under the light microscope are presented in Figure 6-6 to Figure 6-8. On the surfaces of the bearings lubricated with PDI-MDA, clear signs of wear in the form of scratches are visible, while the raceways and rolling elements lubricated with PDI-BAMF exhibit small scratches on their surfaces. It can be observed that there are minimal signs of wear on the raceways and rolling elements lubricated with the reference grease, Berutox FH 28 EPK 2. The optical surface analysis and roughness measurements lead to the following ranking: the best wear protection is achieved when using the reference grease, followed by PDI-BAMF and PDI-MDA grease.

Figure 6-6: Light microscopy of surface of the coupling side (KS) after 80 hours of wear testing in the FE8 test rig at 80°C with 1900 MPa: PDI-MDA (left), PDI-BAMF (middle), and Berutox FH 28 EPK 2 reference grease (right)

Figure 6-7: Light microscopy of surface of the rolling elements (WK) after 80 hours of wear testing in the FE8 test rig at 80°C with 1900 MPa: PDI-MDA (left), PDI-BAMF (middle), and Berutox FH 28 EPK 2 reference grease (right)

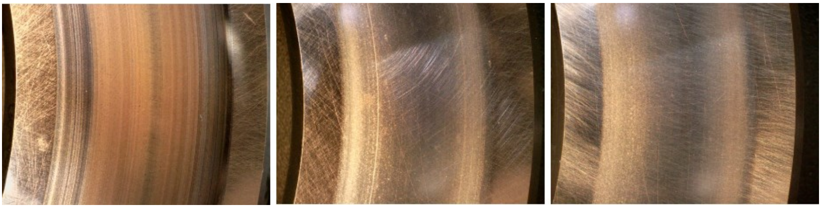


Figure 6-8: Light microscopy of surface of the shaft side (WS) after 80 hours of wear testing in the FE8 test rig at 80°C with 1900 MPa: PDI-MDA (left), PDI-BAMF (middle), and Berutox FH 28 EPK 2 reference grease (right)

In the final step, the mass of the bearings was measured, and the difference from the initial measurement was calculated. Following the DGMK Project 774 [132] guidelines, the difference in the mass of the 15 rolling elements was used to assess the wear protection of a grease. This assessment categorizes the wear protection into four classes, as shown in Table 6-1.

The results of wear mass and surface roughness changes were summarized and compared between the bio-based greases PDI-BAMF, PDI-MDA, and the reference grease Berutox FH 28 EPK 2 (see Figure 6-9). It is evident that the reference grease, followed by PDI-BAMF, exhibits the least surface roughness changes on the rolling elements and the lowest wear mass (0.6 mg for the reference grease and 4.8 mg for the PDI-BAMF grease). Therefore, both are rated as very good wear protection according to Table 6-1. While

Table 6-1: Classification of wear masses of the rolling elements and assessment of wear protection effectiveness using the FE8 axial bearing test rig according to DGMK 774 [132]

Wear mass of the rolling elements (WK) after 80 hours	**Assessment of wear protection effectiveness**
< 10 mg	Very good wear protection
10 – 30 mg	Good wear protection
30 – 100 mg	Moderate wear protection
> 100 mg	Excessive wear

PDI-MDA

Difference	Ra	Rq	Rz
WS	0.008	0.012	0.075
GS	0.002	0.003	0.033
WK	**0.023**	**0.025**	**0.107**

Average	Mass [mg]
Bearing 1 and 2 WK	**10.3**

Good wear protection

PDI-BAMF

Difference	Ra	Rq	Rz
WS	-0.007	-0.005	0.008
GS	-0.008	-0.010	-0.050
WK	**-0.007**	**-0.008**	**-0.067**

Average	Mass [mg]
Bearing 1 and 2 WK	**4.8**

Very good wear protection

Berutox FH 28 KN EPK2

Difference	Ra	Rq	Rz
WS	-0.017	-0.020	-0.095
GS	-0.013	-0.017	-0.070
WK	**0.002**	**0.003**	**0.005**

Average	Mass [mg]
Bearing 1 and 2 WK	**0.6**

Very good wear protection

Figure 6-9: Surface roughness deviation before and after wear tests and average wear mass on the FE8 test rig for bio-based polyurea PDI-BAMF, PDI-MA, and reference petrochemical grease Berutox FH 28 EPK 2 at 80°C and 1900 MPa after 80 hours

PDI-MDA displays higher wear masses (10.3 mg) and roughness deviations, it can still be considered to offer good wear protection according to Table 6-1.

It is observed that the results of the wear measurements on rolling element bearing component test align well with the findings from film formation and friction measurements in the contact level on the EHD2 tribometer (PDI-BAMF≻PDI-MDA). It will be discussed in more details in Chapter 6.2.

6.1.2. Lifetime of radial rolling element bearings lubricated with the developed bio-based greases

The applicability of the greases on radial bearings and fill-for-life suitability of best bio-based grease samples was investigated using a radial bearing test rig (RLP). For this purpose, two sealed deep groove ball bearings of size 6305 were used as test bearings as shown in the design in Figure 6-10.

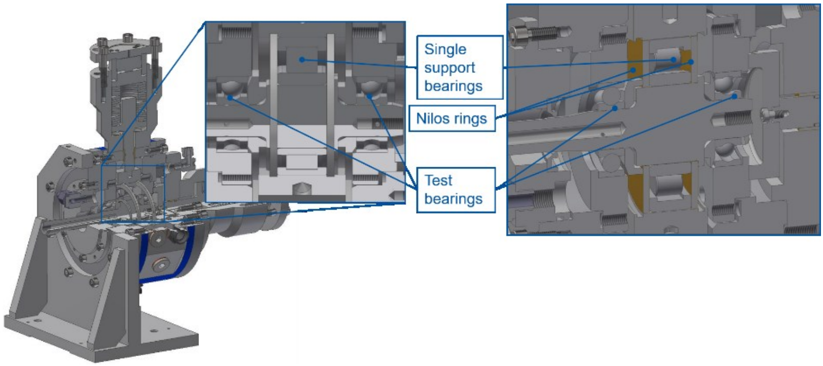


Figure 6-10: Radial bearing test rig with two test ball bearings of 6305 for evaluation of bio-based greases in rolling bearing applications

The radial loads were applied by disc spring assemblies. The radial load acts on the support cylindrical roller bearing NU310, which in turn results in a load on the housing-supported test bearings. The use of sealed, grease-lubricated bearings required an adaptation design and modification of the initial test rig. The design with sealed cylindrical roller bearing NU310 by so called Nilos rings is shown in Figure 6-10.

In the RLP, the force sensor measured the radial forces acting on the disc springs. The force is then transmitted to the support bearing, shaft, and the two test bearings. Two PT 100 temperature sensors additionally measured the temperatures at the test bearings as shown in Figure 6-11. A vibration sensor was used to define failure of the greases. Before commencing the measurements, the covers of the test bearings were removed to replace the original grease with the test greases. According to the catalog specifications of the bearing manufacturer, 90% of the free bearing space, i.e., 12 ml, was filled with grease. Subsequently, the cover was replaced and checked to ensure that the bearings rotate smoothly (Figure 6-12).

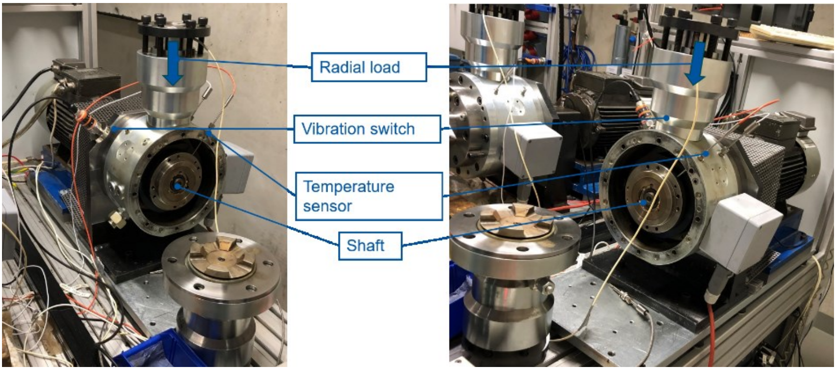


Figure 6-11: Radial bearing test rig set up and measuring sensors

The tests were conducted at a radial load of 10 kN, a shaft speed of 2500 min^{-1}, and a temperature of 50 °C. Given the stochastic nature of bearing lifetime, directly comparing the lifetime results for greases can be challenging. To create a standardized basis for visual comparison of failure in the bearings, a time limit of 300 hours was set for each grease test. That means each grease test was performed until the failure or measurement time reached 300 hours for the first time. In the scope of this work, more comparable results were attempted to be generated with the help of a 300-hour threshold; however, more repetition of the measurements is essential for better comparisons and ensuring the reliability of the results.

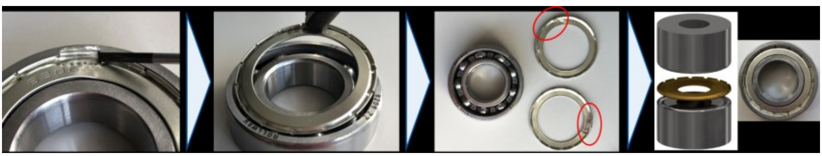
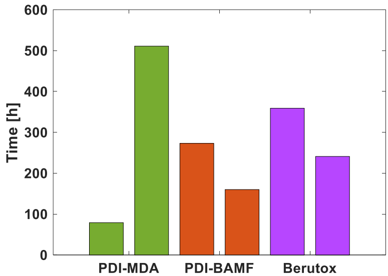
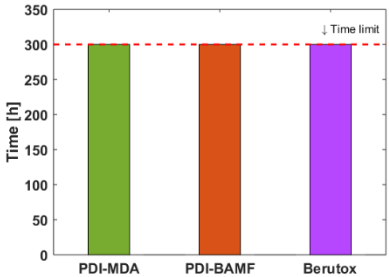


Figure 6-12: Process of replacing the original grease of the ball bearings with the test greases

The results of the measurements were divided into two groups: one with a maximum run time of 300 hours and the other without a time limit. The results of the life-time tests for the bio-based greases with polyurea thickener systems, PDI-BAMF and PDI-PDA, and the petrochemical reference grease, Berutox FH 28 EPK 2, are presented in Figure 6-13. The results of measurements without time limit shows high variation of the life time for PDI-MDA grease. For bio-based PDI-BAMF and petrochemical grease, comparable life times can be observed.



(a)



(b)

Figure 6-13: Results of the life-time tests on the radial bearing test rig for the bio-based greases with polyurea PDI-BAMF and PDI-MDA, as well as the petrochemical reference grease Berutox FH 28 EPK 2, at 10 kN, 2500 min^{-1}, and 50 °C: (a) limited to 300 hours, (b) without time limit until failure

The samples were removed from the test rig and cleaned after the measurement. The outer ring of the test bearing was cut open after to conduct optical inspections on the surface of the inner and outer rings, the rolling elements, and the cage for identification of the damage.

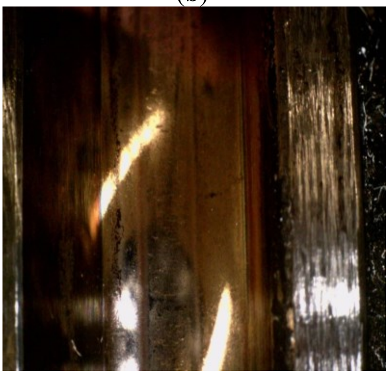
For the initial comparison, the samples with bio-based greases were evaluated for the quality of the inner bearing surfaces following their first tests without a time limit. The bearing surfaces were examined visually and under a light microscope after failure. The results are depicted in Figure 6-14. As shown in Figure 6-14 (a) and Figure 6-14 (b), the inner ring of the bearing lubricated with PDI-MDA appears darker compared to PDI-BAMF, indicating presence of higher temperatures and eventually mixed friction condition during operation. This aligns with the temperatures measured at failure point on the test rig for PDI-MDA (higher than 180 °C) and PDI-BAMF (around 120 °C).

Noticeable wear marks are also present on the inner ring, as confirmed by the microscope images in Figure 6-14 (c) and Figure 6-14 (d). The cage of a bearing lubricated with PDI-MDA fractured after 79 hours, and under the microscope, the pittings on the rolling elements are clearly visible, as shown in Figure 6-14 (e) and Figure 6-14 (g). The damage pattern on the test bearing lubricated with PDI-BAMF is only minimally observed after 273 hours of testing.


(**a**)

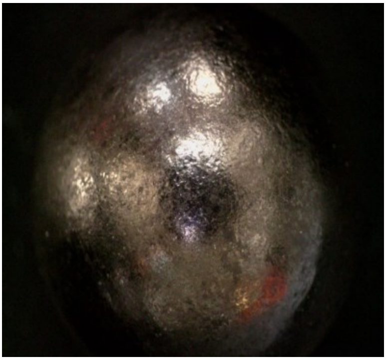

(**b**)


(**c**)


(**d**)


(**e**)


(**f**)



(g)

(h)

Figure 6-14: Optical evaluation of the test bearings for PDI-MDA after 79 hours (a-g) and PDI-BAMF after 273 hours (b-h): (a) Inner ring of the bearing with PDI-MDA, (b) Inner ring of the bearing with PDI-BAMF, (c) Inner ring of the bearing under the light microscope with PDI-MDA, (d) Inner ring of the bearing under the light microscope with PDI-BAMF, (e) Cage and balls of the bearing with PDI-MDA, (f) Cage and balls of the bearing with PDI-BAMF, (g) Cage and balls of the bearing under the light microscope with PDI-MDA, (h) Cage and balls of the bearing under the light microscope with PDI-BAMF

Figure 6-13 shows significant scattering in the results of life time measurements without time limits. Additionally, when comparing the condition of PDI-MDA after 79 hours and PDI-BAMF after 273 hours, different types of failures are evident, including cage damage, pitting, and wear grooves. Therefore, another method is beneficial to support the life time results. It is assumed that, after a specific measurement time, the greases experienced the same conditions, and therefore, the surface protection characteristics of the greases eventually became more comparable. In the measurements with a time limit, as shown in Figure 6-13 (a), even after 300 hours, no signs of peaks in temperature and vibration signals were detected for the greases. Therefore, it was assumed that the bearings were operating under normal conditions. However, minor deviations in surface quality were expected after 300 hours. Therefore, comparing the surface qualities under similar conditions after 300 hours will provide insights into the lubricating performance of the greases (Figure 6-15). The inner ring of the bearing lubricated with PDI-MDA exhibits wear patterns relatively similar to those of PDI-BAMF. Minor pittings can be observed on the inner ring's surfaces in the case of PDI-MDA. However, the surfaces of the bearings from the tests with PDI-BAMF show no surface alterations due to mixed lubrication and demonstrate no damage, as shown in Figure 6-15 (d). The wear marks on the ball surfaces of the bearing lubricated with PDI-MDA indicate a slight disadvantage compared to PDI-BAMF.

(a)

(b)

(c)

(d)

(e)

(f)

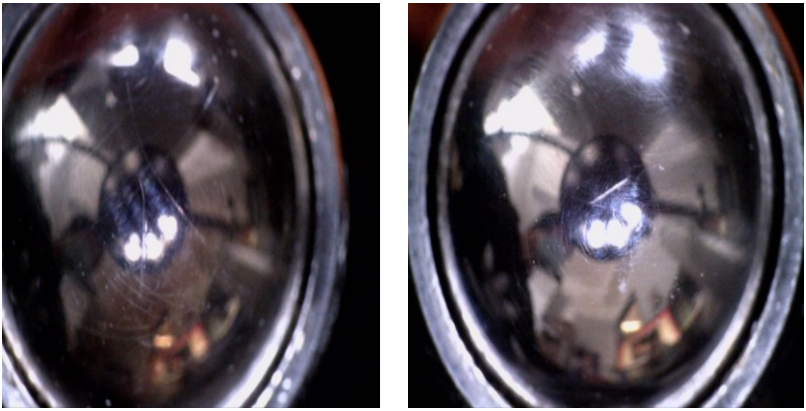


Figure 6-15: Optical evaluation of the test bearings for PDI-MDA after 300 hours (a-g) and PDI-BAMF after 300 hours (b-h): (a) Inner ring of the bearing with PDI-MDA, (b) Inner ring of the bearing with PDI-BAMF, (c) Inner ring of the bearing under the light microscope with PDI-MDA, (d) Inner ring of the bearing under the light microscope with PDI-BAMF, (e) Cage and balls of the bearing with PDI-MDA, (f) Cage and balls of the bearing with PDI-BAMF, (g) Cage and balls of the bearing under the light microscope with PDI-MDA, (h) Cage and balls of the bearing under the light microscope with PDI-BAMF

The comparison of the results from the tests with bio-based greases to the test with the reference grease, Berutox FH 28 EPK 2, is presented in Figure 6-16 and Figure 6-17. The scratches on the surfaces of the rolling elements of the bearing lubricated with the reference grease are less pronounced compared to those in the bio-based greases (Figure 6-16). Overall, the results indicate that the bio-based greases exhibit wear protection performance comparable to petrochemical greases, considering that the additive packages for the bio-based greases have not yet been optimized.

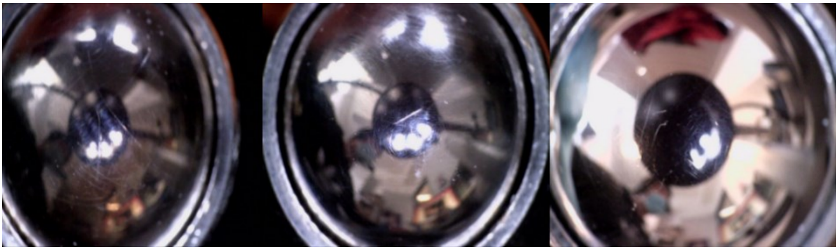


Figure 6-16: Optical evaluation of the surfaces of rolling elements of bearings with PDI-MDA (left), PDI-BAMF (middle), reference grease Berutox FH 28 EPK 2 (right) after 300 hours on the radial bearing test stand at 50 °C with 2180 MPa

The surface qualities of the inner rings in the systems with bio-based greases and the reference grease, Berutox FH 28 EPK 2, are compared in Figure 6-17. In certain areas, small pittings can be observed on the surface of the inner ring of the bearing lubricated with PDI-MDA, indicating the starting time of formation of pitting in the bearing with PDI-MDA. This is an indication of progressive degradation, which could lead to the eventual failure of the bearing. However, PDI-BAMF and the reference grease show no surface alterations, suggesting better lubrication properties.

Based on the results of the life time measurements on radial bearing test as mentioned in this section, the performance of the greases can be ranked as following: (Reference>PDI-BAMF>PDI-MDA). These results agree with previously discussed results of tribometer and FE8 tests in Chapter 4 and 5.

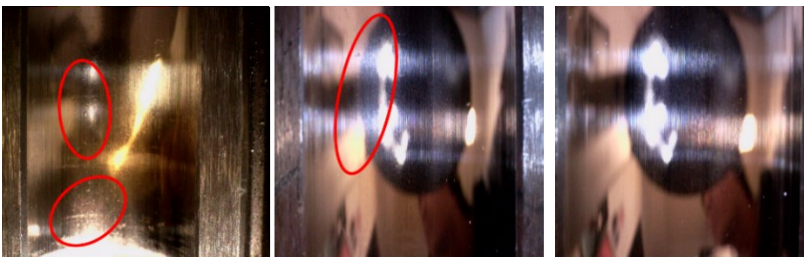


Figure 6-17: Optical assessment of the surfaces of the inner rings of bearings with PDI-MDA (left), PDI-BAMF (middle), and reference grease Berutox FH 28 EPK 2 (right) after 300 hours on the radial bearing test rig at 50 °C with 2180 MPa

6.2. Correlation of the tribological performance on screening method with the results of component tests

To compare the tribological results on component tests with tribometer scale, first the FE8 wear test results of PDI-MDA and PDI-BAMF with the film formation and friction results on tribometer level will be compared. Figure 6-18 (a,b) shows the measured temperature and torque during the wear tests on FE8. The results show that the measured torque of the greases reaches a steady state after a few hours. At the steady state point, the bearing with the petrochemical grease shows relatively higher torque values. The bearing with PDI-BAMF exhibits lower torque compared to the reference grease, while the

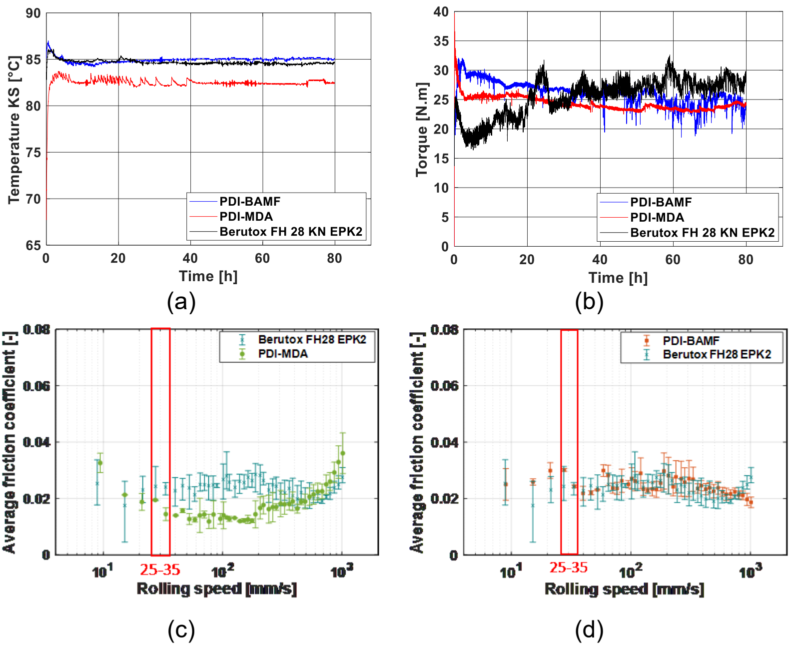


Figure 6-18: Correlation of results from component test on FE8 and tribometer level (a) temperature evolution of the FE8 test (b) measured torque from FE8 measurements (c) friction coefficient of PDI-MDA compared to reference grease (d) friction coefficient of PDI-BAMF compared to reference grease on tribometer

PDI-MDA grease shows the lowest torque. This trend is also evident in the temperature results shown in Figure 6-18 (b). The trend can be correlated with the findings of friction measurements on tribometer shown in Figure 6-18 (c). It is due to the fact that higher friction of petrochemical grease and PDI-BAMF in this rolling speed range cause higher torque, thus higher temperatures of reference grease and PDI-BAMF compared to PDI-MDA. It is to notice that although the results can be correlated, the values of friction coefficients from tribometer do not represent the exact friction coefficient and film thickness of the FE8 measurements due to different temperature, pressure, load, and type of contact as mentioned compared in Table 6-2.

PDI-BAMF and Berutox FH 28 EPK 2 showed higher film thickness in the measurement speed range compared to the PDI-MDA grease. This indicated better film formation abilities of PDI-BAMF and PDI-MDA. The life time measurements on RLP also showed comparable performance of the PDI-BAMF and Berutox FH 28 EPK 2 and better lubricity compared to PDI-MDA. Although it seems that the the results from life time tests on RLP agree with

Table 6-2: Different contact situations with different test methods of tribometer level and bearing component level

	Test methods		
	Tribometer	**FE8**	**Radial bearing**
Pressure [MPa]	700	1900	2020
Temperature [°C]	40	80	50
Rolling speed [mm/s]	4 to 1000	≈ 25 to 35	≈ 3370
Type of contact	Point contact	Line contact	Point contact
Roller diameter [mm]	19.5	11	10
Raceway	Disc	Disc	Groove
Lubrication	Grease layer on the surface of the disc	Relubrication every 10 min with 50 mm^3	12 ml initial lubrication

the tribometer film formation, the differences must be considered carefully. The tribometer measurement were performed in hydrodynamic lubrication regime ($\lambda > 3$), however, the results from axial bearing test rig are in boundary or mixed lubrication regime ($\lambda < 3$) and therefore cannot directly be correlated. It is also to notice, despite comparable results of the methods the film thickness and friction coefficient of the single contact of the RLP test rig during the measurement cannot exactly be predicted by tribometer results due to limited rolling speed range, pressures of tribometer measurements (Table 6-2.), and different lubrication regimes.

Although overall performance of the greases from different evaluation methods of tribometer and component tests in this work show consistent ranking of the greases, not all the effects of the grease lubrication can be captured by tribometer tests such as centrifugal forces in higher rolling speeds as in rolling element bearing test [133]. At higher velocities the differences between tribometer results and the bearing results are expected to be more pronounced.

6.3. Chapter conclusion

The primary objective of chapter 6 was to experimentally validate the second proposed research hypothesis:

2 The performance of developed greases at the component level on rolling element bearing test rigs can be correlated to the results of tribological screening tests and confirms the suitability of the screening method.

Therefore, tribological performance of the developed bio-based greases were evaluated at component level on FE8 and radial bearing test rig in terms of wear protection and lifetime. The resulting ranking of the greases (Berutox FH 28 EPK 2≈BAMF≻MDA) were in agreement to the previous tribometer film formation and friction results. **The second hypothesis is, therefore, proved by the results.**

FE8 results showed comparable wear protection characteristics of the bio-based greases with the petrochemical grease. The radial bearing test rig also demonstrated the suitability of using bio-based grease samples in rolling element bearing applications. Thus far, the component tests have shown comparable performance of bio-based greases compared to their petrochemical counterparts. In the next chapter, the comparability of the noise reduction performance of the bio-based greases with the petrochemical grease will be further investigated.

7. Vibration analysis of the greases

This chapter primarily aims to validate the third research hypothesis of this study concerning the noise characteristics of the developed bio-based greases: “The vibration characteristics of rolling element bearings will not critically deviate between those lubricated with developed bio-based greases and those lubricated with the petrochemical reference grease.” This chapter, therefore, incorporates comparisons of the noise reduction performance of the bio-based greases with the petrochemical reference greases. These comparisons will provide the complementary information to address comparability of the tribological performance of the developed bio-based greases with that of petrochemical grease. Consequently, vibration measurements are conducted on the PDI-PDA and PDI-BAMF as the most promising bio-based polyurea candidates from previous evaluations in this work, and compared accordingly.

To elucidate the effect of thickeners on noise reduction performance of the grease, vibration measurements on rolling element bearings have been conducted initially with only the base oils and subsequently with complete greases. Furthermore, the reproducibility of the vibration results has been verified. Given that the vibration behaviour of the lubricated rolling element bearings is dependent on the operational conditions [134], this chapter also includes exploratory investigations into the vibration behaviour of the greases under different load and temperature conditions along with discussions.

7.1. Measurement set-up

The tests were performed on the same radial bearing test rig as in Chapter 6.1.2 which was additionally equipped with two integrated piezoelectric accelerometers of PCB 356A01 with measurement range of ±9810 m/s², transverse sensitivity ≤ 5 %, resonance frequency ≥ 50 kHz. The accelerometers were mounted at two different positions on the outer metal housing of the test rig with recommended glue from the sensor manufacturer. The sensors were installed with 90- and 30-degree radial angle as shown in Figure 7-1 to ensure measuring vibrations of the test rig in different vibration modes.

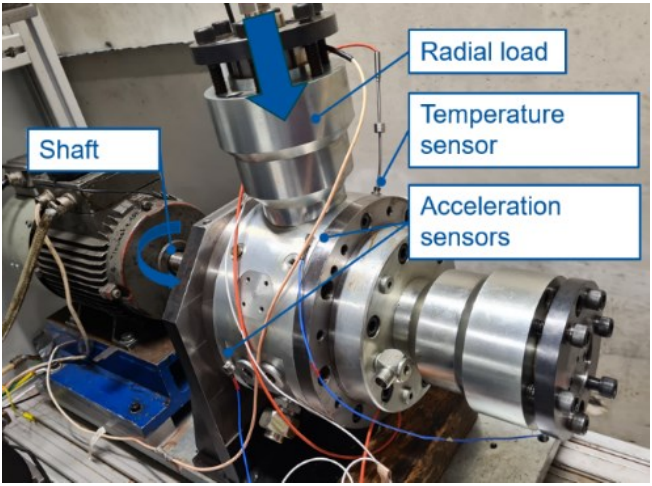


Figure 7-1: Vibration measurement set up on radial bearing test rig

Time-domain signals from the accelerometers were attained through the data acquisition hardware of Müller BBM PAK MK2, MF03 with the ICP 42 modules with sampling rate of 32768 Hz (Figure 7-2).

Lubricating of the test bearings was performed with the same method as explained in Chapter 6.1.2. The test rig was heated by heating elements to reach steady state for each case and the temperatures were measured during the measurements using two PT 100 temperature sensors. Run-in phase for

Figure 7-2: Data acquisition system of Müller BBM PAK MK2 for analysis of vibration signals

the bearings was set to 5 min with 100 min^{-1} to assure uniform distribution of the lubricating greases inside the rolling element bearings. The measured signal duration of 95 s was recorded and processed for each measurement.

To investigate the effect of operational conditions on the vibrations of the rolling element bearings with the greases, the measurements were conducted with various loads of 1, 5, and 10 kN, corresponding to Hertzian contact pressures of 1.33, 1.75, and 2.02 GPa, respectively. The measurements were conducted on bio-based polyurea greases PDI-MDA and PDI-BAMF, with castor oil as the base oil, which showed superior performance compared to other developed samples as in Chapter 5 and Chapter 6, as well as on the petrochemical reference grease and PAO base oil. The tested lubricants and the operational conditions of the measurements are summarized in Table 7-1. During the measurements the rotational speed was increased from still stand to 3000 min^{-1} and decreased back to zero.

The measured accelerations in each direction of the sensors were measured in time domain and processed using the data acquisition system. The Root Mean Square (RMS) amplitude of the vibrations have been calculated by the software.

Table 7-1: Operational conditions for vibration measurements on bio-based greases and the castor oil as well as petrochemical grease and PAO

		Radial load [kN] (Hertzian pressure [GPa])	**Temperature [°C]**	**Rotational speed [min^{-1}]**
Bio-based	Castor oil	1,5,10 (1.33, 1.75, 2.02)	40	Up to 3000
	PDI-BAMF	1,5,10 (1.33, 1.75, 2.02)	25, 40	Up to 3000
	PDI-BAMF (2)	1 (1.33)	40	Up to 3000
	PDI-MDA	1,5,10 (1.33, 1.75, 2.02)	40	Up to 3000
Petro.	PAO	1,5,10 (1.33, 1.75, 2.02)	40	Up to 3000
	Berutox	1,5,10 (1.33, 1.75, 2.02)	40	Up to 3000

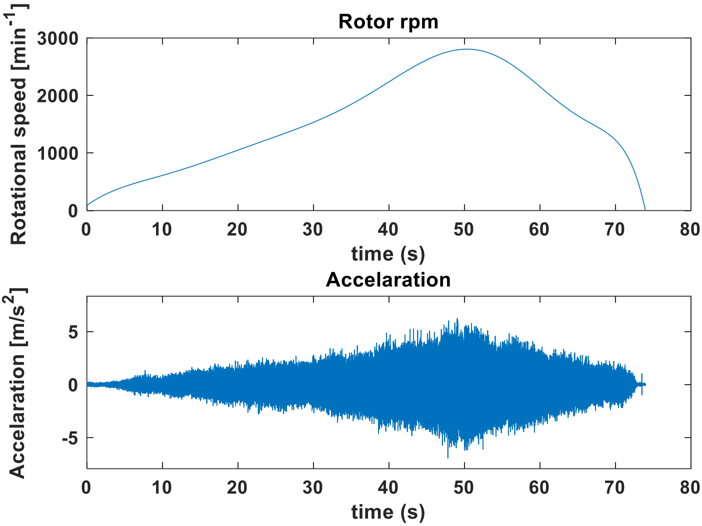


Figure 7-4: Acceleration signals of vibration measurements in time domain

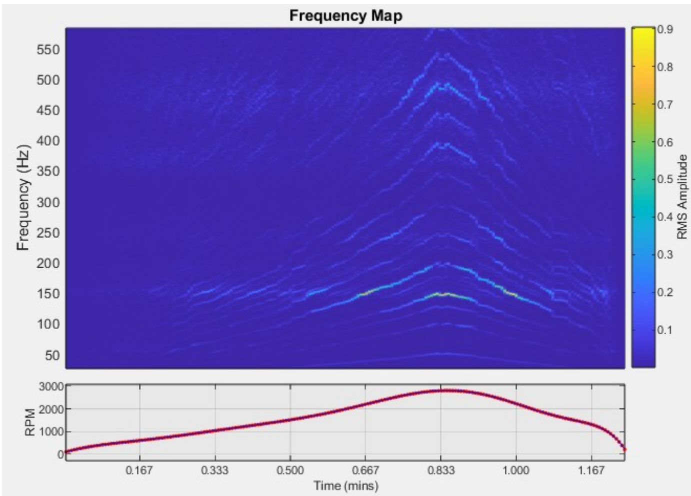


Figure 7-3: Converted acceleration signals of the vibration measurements to frequency domain in waterfall diagram using FFT

The time domain results (Figure 7-4) are converted to frequency domain results using Fast Fourier Transformation (FFT) and plotted in the format of waterfall diagrams (Figure 7-3). This assures capturing the vibration resonances in different rotational speeds of the shaft.

The noise reduction performance of the greases and oils was compared by comparison of the resulting vibration of the bearings with different lubricants in terms of RMS amplitude at the resonance point of the system. Since the excitations rise with higher loads, higher vibration amplitudes are not directly linked to different characteristics of the greases; therefore, additionally, the damping ratio at the resonance points was considered to evaluate the greases. The damping ratio was calculated according to half power method as cited in [135] as following:

$$\zeta = \frac{f_2 - f_1}{2f_0} \qquad \text{Equation 7-1}$$

where ζ is the damping ratio and f_1 and f_2 are the corresponding frequency of the resonance point with frequency of f_0 as shown in Figure 7-5.

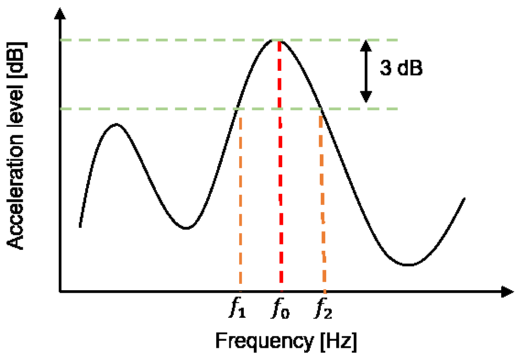


Figure 7-5: Schematic demonstration of resonance peak and calculation of the damping ratio based on half power method

7.2. Vibration analysis of oil lubricated rolling element bearings

The initial series of vibration measurements were conducted on the base oils of both bio-based and petrochemical lubricating greases, namely castor oil and PAO, at 40 °C under 1, 5 and 10 kN. To provide insight into the dominant components of the vibration signals, the time dependent acceleration results of the vibration tests were converted to the frequency domain using FFT and are demonstrated in the following spectrogram or waterfall diagrams as in Figure 7-6 and Figure 7-7. The waterfall diagrams show the frequency content of the acceleration signal over rotational speeds and are widely used in vibration analysis as in [136,137]. The RMS amplitudes of vibration signals for each test are converted to dB for better demonstration. The diagrams depict the acceleration level in dB, represented by contour colours corresponding to the frequency, along with the fitted rotational speeds. The RGB colour legend method indicate acceleration levels and resonances of the system.

The brighter lines indicate the frequency-rpm relations and are referred to as harmonics or orders in vibration analysis which are synchronous with the rotational speed of the test rig shaft. The peaks in the diagrams show the highest vibration amplitudes or the system resonances. For comparison of the vibration signals on the most dominant orders, order cut as a signal processing technique is used to isolate the vibration signals on the harmonics. Therefore, extra diagrams are plotted which show the acceleration level of the signals over the rotational speed. The lubricants in rolling element bearings are the main source of high damping [138]. Therefore, as an extra tool to evaluate the vibration results of different lubricants, the damping ratio ζ values at the resonances are calculated using half power method as described in Chapter 7.1.

In the results of vibration analysis of castor oil, it can be seen that the two dominant orders, namely third and fourth harmonics can be considered as the most dominant orders among others as in the in Figure 7-6. However, the results of the PAO oil shows only one dominant harmonics of fourth order in Figure 7-7. The dominant resonance point with the highest acceleration is located on the fourth order for both PAO and castor oil. Therefore, only the vibration signals on the fourth order for both oils have been extracted and demonstrated. From comparison of the waterfall diagrams between petrochemical PAO and bio-based castor oil, it can be observed that lubricating with PAO can eliminate vibrations related to 3rd harmonics eventually due to better lubrication. However, to compare the results quantitatively, the acceleration amplitudes at the most dominant resonances on fourth order for both oils will be compared in following.

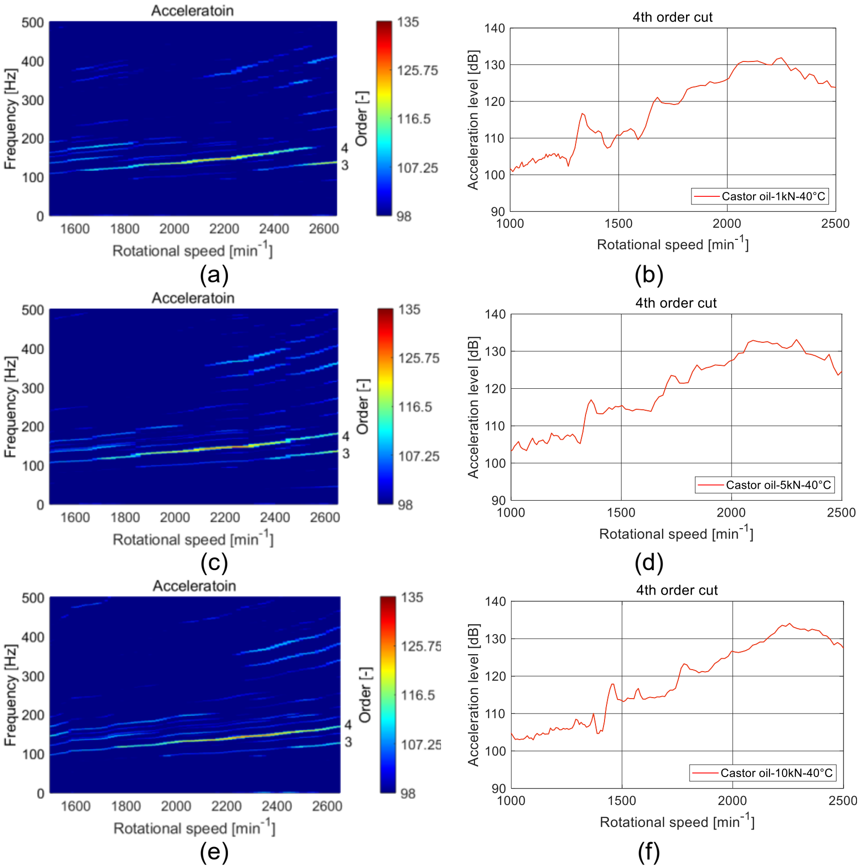


Figure 7-6: Waterfall (a-c) and order cut (b-f) diagrams of vibration measurements on radial rolling element bearing test rig lubricated with castor oil at 40°C with rpm sweep up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

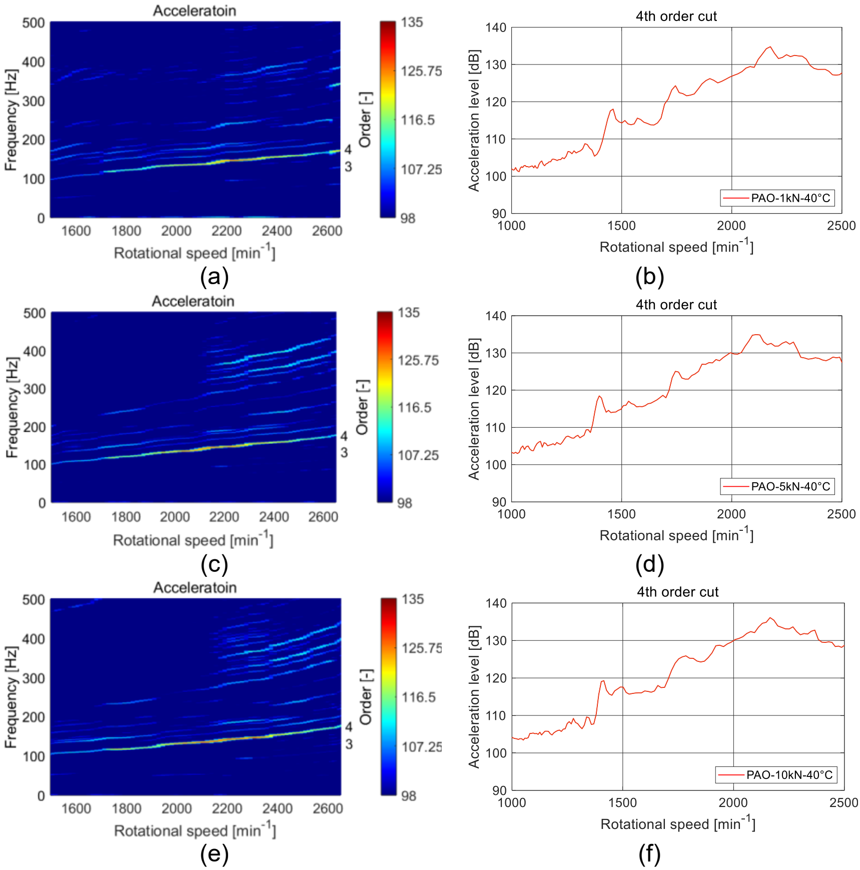
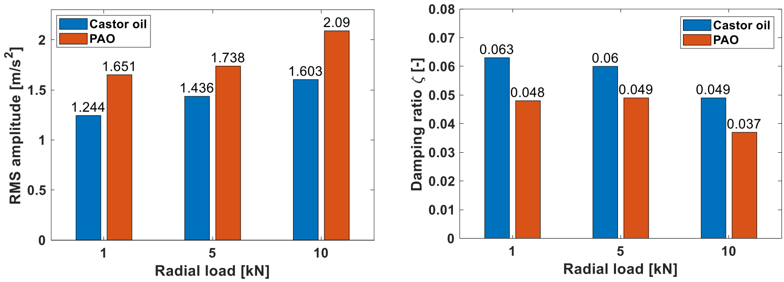


Figure 7-7: Waterfall (a-c) and order cut (b-f) diagrams of vibration measurements on radial rolling element bearing test rig lubricated with PAO at 40 °C with rpm sweep from up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

To discuss the vibration parameters for the oils quantitatively, information from waterfall diagrams at the operating points with the highest vibration amplitudes are summarized into the Table 7-2 and Figure 7-8. The frequency and rotational speed of the peaks shows minimal deviations. This occurred because the measurement of the rotational speed on the test rig was not automatically synchronized with the acceleration measurements and had to be set manually. Therefore, the deviation in these two parameters will not play an important role in the interpretation of the results.

Table 7-2: Vibration peaks of vibration measurements on the rolling element bearings lubricated with castor oil as the base oil of developed bio-based greases and PAO as the base oil of petrochemical reference grease at 40°C for different applied radial loads

	Radial load [kN]	**Frequency [Hz]**	**Rotational Speed [rpm]**	**RMS Amplitude [m/s^2]**	**Damping Ratio ζ [-]**
Castor oil	1	149.96	2249.5	1.244	0.063
	5	152.95	2294.3	1.436	0.060
	10	150.44	2256.7	1.603	0.049
PAO	1	145.01	2175.3	1.651	0.048
	5	140.69	2110.4	1.738	0.049
	10	144.31	2164.7	2.090	0.037



(a) (b)

Figure 7-8: Comparison of the (a) RMS amplitude and (b) damping ratio of the vibration analysis of the rolling element bearings lubricated with castor oil and PAO as the base oil of developed bio-based and petrochemical greases at 40°C

It can also be observed that lubricating with castor oil, RMS amplitudes are increased by 15% and 11% by increasing the radial applied load from 1 to 5 kN and from 5 to10 kN. Overall, the RMS values increase around 28% from 1 to 10 kN. The vibration peaks of the bearings with PAO show an increase of the RMS amplitudes of 5% and 20% (overall 26%) by increasing the radial applied load from 1 to 5 kN and from 5 to10 kN. It is to notice that for both castor oil and PAO oil, the RMS amplitudes of the resonance peaks increase significantly with an increase in load. It has been observed in the time-dependent raw data of the measurement, as well as in the literature, that for constant loads, the RMS vibration amplitudes of the rolling bearings increase with increasing rotational speed [139]. Assuming to have the vibration peaks at the same rotational speeds for different loads, increasing the radial load will increase the stiffness between the joints of the rolling bearing and the housing parts of the test rig (assumed to be K_s in Figure 7-9) and therefore increase the vibration energy as also explained in the literature [140]. It can also be observed that the PAO shows around 20 to 30% higher RMS amplitudes compared to the bio-based castor oil. This can be tracked back to the viscosities of the based oils. PAO kinematic viscosity is measured 220 mm^2/s and castor oil has 254 mm^2/s at 40°C. It has been reported in the literature that the higher viscosities will cause lower vibration RMS amplitudes [140,141,142].

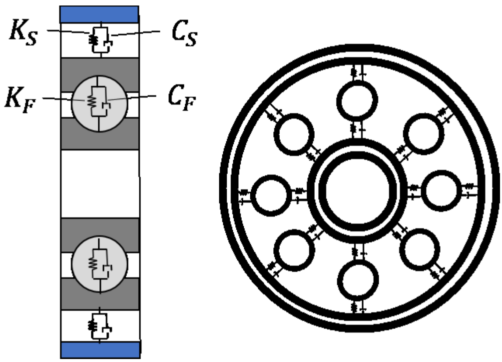


Figure 7-9: Equivalent stiffness and damping of a rolling element bearing with indices of "S" for structure and "F" for the lubricant fluid

By increasing the applied radial load on the rolling element bearings lubricated with castor oil the damping ratio ζ shows decrease of 5% and 22% (overall 28%) from 1 to 5 kN and from 5 to 10 kN. For the PAO oil, the damping ratio ζ stays almost constant from 1 to 5 kN and shows a decrease of 29% from 5 to 10 kN radial load increase. It can be seen that for the bearings with both castor oil as well as PAO as lubricants, the damping ratio decreases with the increase in radial load. This can be addressed to the decrease of the damping constant of the oils. The stiffness and damping of the single contact of the bearing is demonstrated in Figure 7-10 (a). The elastohydrodynamic film in the contact area is responsible mostly for the damping behaviour of the rolling element bearings. That means C_F dominates C_S. By increasing the load for constant rotational speed, the film thickness will decrease minimally and at the same time the contact area as well as the fluid film viscosity will be increased. Consequently, the ability of the lubricant film to damp the vibrations will also decrease as reported in the literature. The decrease of the damping of the lubricant oil by increasing the load has been previously reported by several researchers as in [143,81]. See Figure 7-11 (a).

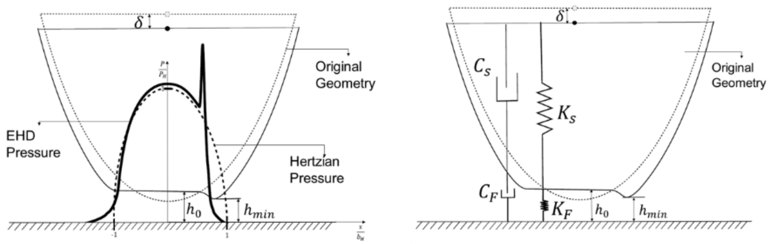
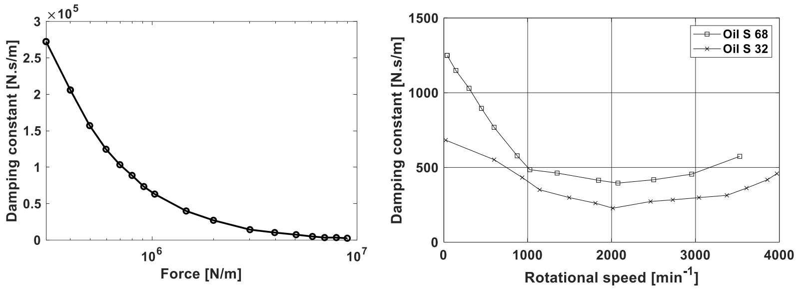


Figure 7-10: Formation of EHD contact and the equivalent stiffness and damping of a single contact of a rolling element bearing with indices of “F” for fluid and “S” for structure

It can also be seen that the damping ratio values of the castor oil is higher for all the load cases. This can be addressed to relatively higher viscosity of the castor oil compared to PAO. The effect of viscosity on the damping of the fluid film in single contact of the rolling element bearings is seen in the literature as shown in Figure 7-11 (b), which shows an increase of damping constant by application of higher viscosity oil as the lubricant [134].



(a) (b)

Figure 7-11: (a) effect of load on the damping constant of oil lubricated single contact of a rolling element bearing from [143] and (b) effect of viscosity of the oil on the damping of the rolling element bearing from [134]

Comparing castor oil with PAO, the higher damping ratios and lower RMS amplitudes show comparable to better performance of the bio-based base oil of the developed greases compared to the petrochemical base oil on fourth harmonic.

7.3. Vibration analysis of grease lubricated rolling element bearings

To verify the research hypothesis regarding differentiation of the vibration behaviour of the grease and base oil, the vibration measurements have been also conducted on the developed bio-based as well as petrochemical grease.

7.3.1. Verification of reproducibility of the vibration measurements on the greases

To verify the reproducibility of the measurement on greases, two independent vibration measurements on the grease PDI-BAMF have been conducted at 1 kN at 40 °C. The waterfall and order cut diagrams are demonstrated in Figure 7-12. The vibration measurement results are summarized in Table 7-3. The RMS amplitudes results show small deviations of around 0.019 m/s^2. The damping ratio results show also small deviations of around 0.006. Comparing this deviation with deviations due to change in grease later on, confirms reproducibility of the results, thus the errors will be considered for interpretation of the results in next sections.

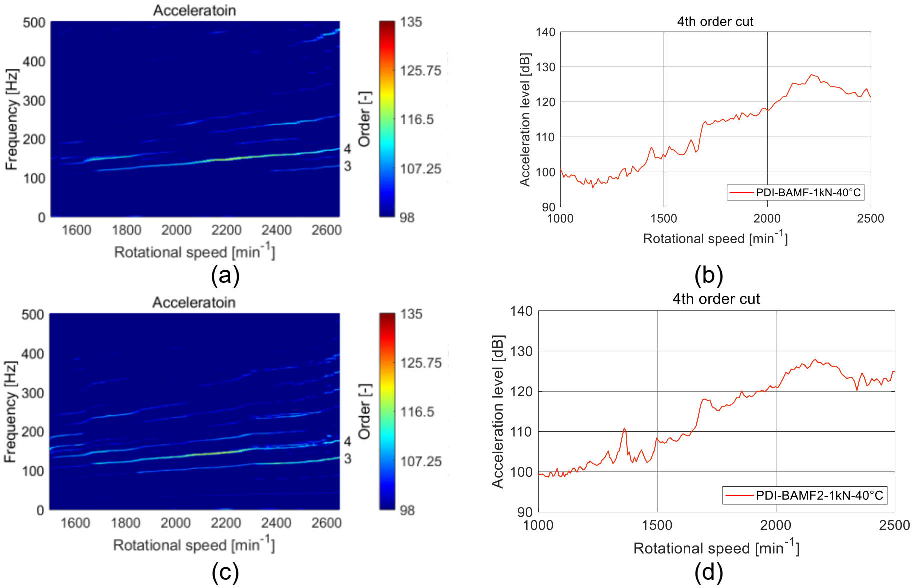


Figure 7-12: Waterfall and order cut diagrams of vibration measurements on radial rolling element bearing test rig lubricated with PDI-BAMF at 1 kN equal to 1.33 GPa and 40°C (a-b) first and (c-d) second measurement with rpm sweep from up to 3000 min^{-1}

Table 7-3: Difference of vibration peaks of vibration measurements on the rolling element bearings lubricated with developed bio-based greases PDI-BAMF for first and second measurement at 40°C

	Frequency [Hz]	**Rotational Speed [rpm]**	**RMS Amplitude [m/s^2]**	**Damping Ratio ζ [-]**
1st	147.45	2211.8	0.772	0.041
2nd	144.30	2164.6	0.791	0.047
Diff	3.14	47.2	0.019	0.006

7.3.1.1. Vibration measurements on the bio-based greases and comparison to petrochemical grease

The vibration measurements are conducted on the most promising bio-based polyurea greases PDI-BAMF, PDI-MDA as well as Berutox FH 28 EPK 2 as the reference petrochemical grease under varied radial loads of 1, 5 and 10 kN. The waterfall and colormap diagrams as well as order cuts are demonstrated in Figure 7-13, Figure 7-14 and Figure 7-15 which show only minimal difference between bio-based greases and the petrochemical grease.

Comparing the waterfall diagrams, it can be seen that Berutox FH 28 EPK 2 shows only one dominant harmonic at fourth order. This observation is similar to what was noted in previous section for PAO as its base oil. The bio-based greases show two dominant harmonics of third and fourth order. This observation is also consistent with what was noted for castor oil. The vibration parameters of the resonance peaks for grease measurements are extracted on the fourth order from the diagrams and summarized into the Table 7-4.

Table 7-4: Vibration peaks of vibration measurements on the rolling element bearings lubricated with petrochemical Berutox FH 28 EPK 2 and developed bio-based greases PDI-BAMF and PDI-MDA at 40°C for different applied radial loads

	Radial load [kN]	**Frequency [Hz]**	**Rotational Speed [rpm]**	**RMS Amplitude [m/s²]**	**Damping Ratio ζ [-]**
Berutox	1	148.36	2225.4	0.795	0.058
	5	140.79	2111.9	0.822	0.060
	10	145.92	2188.9	0.901	0.066
BAMF	1	147.45	2211.8	0.772	0.041
	5	145.58	2183.8	0.944	0.042
	10	144.87	2173.1	1.164	0.049
MDA	1	143.63	2154.6	0.951	0.015
	5	142.52	2137.8	0.907	0.034
	10	143.47	2152.1	1.162	0.050

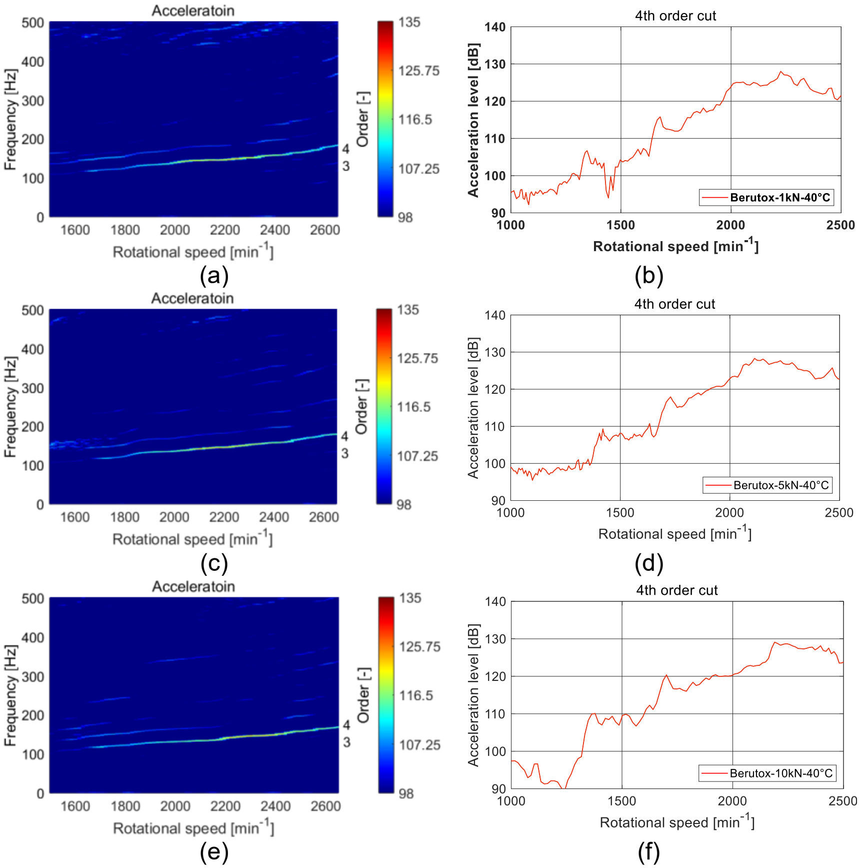


Figure 7-13: Waterfall (a-c) and order cut (b-f) diagrams of vibration measurements on radial rolling element bearing test rig lubricated with Berutox FH 28 EPK 2 at 40°C with rpm sweep from up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

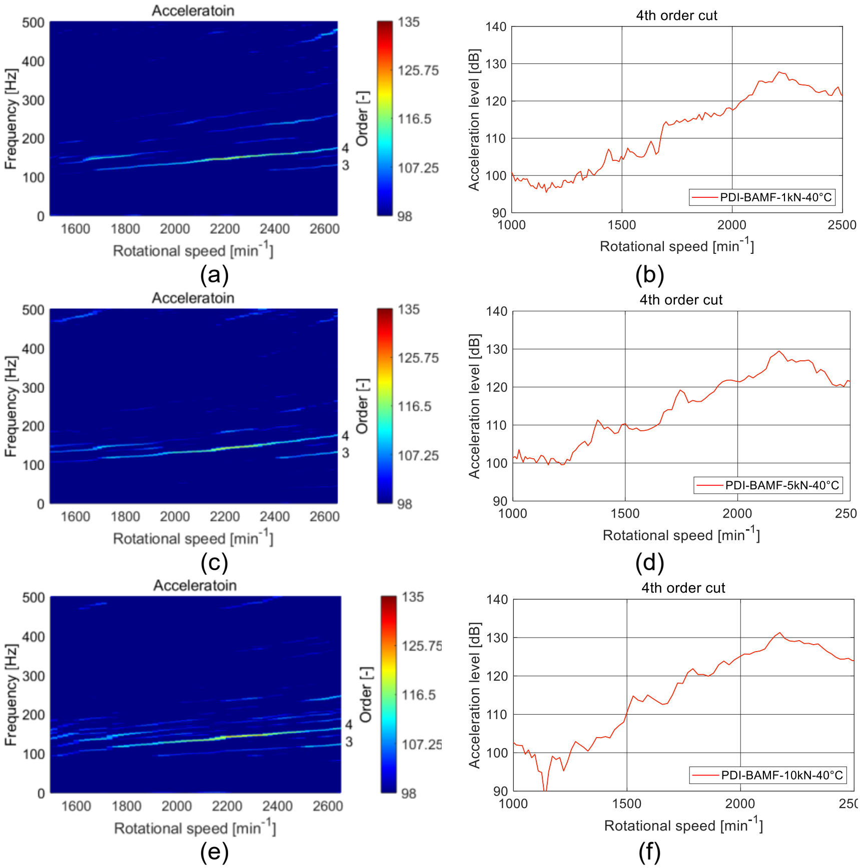


Figure 7-14: Waterfall (a-c) and order cut (b-f) diagrams of vibration measurements on radial rolling element bearing test rig lubricated with PDI-BAMF at 40°C with rpm sweep from up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

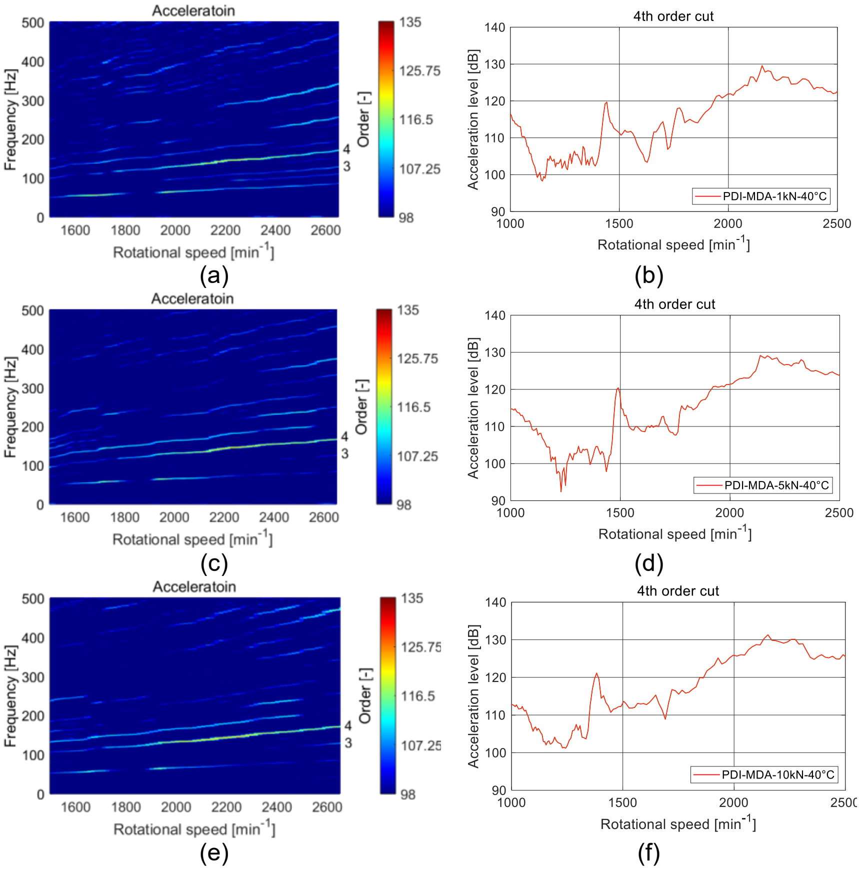


Figure 7-15: Waterfall (a-c) and order cut (b-f) diagrams of vibration measurements on radial rolling element bearing test rig lubricated with PDI-MDA at 40°C with rpm sweep from up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

The RMS and damping results of the petrochemical grease is compared to its base oil and demonstrated in Figure 7-16. It can be observed that RMS amplitude of the Berutox FH 28 EPK 2 shows 4% and 10% (overall RMS difference of 0.11 around 14%) by increasing the radial load from 1 to 5 kN and from 5 to 10 kN. PAO oil however, shows a clear overall increase of 26% (overall RMS difference of 0.45) in RMS amplitude over increasing radial load from 1 to 10 kN. The increase in RMS amplitudes in grease lubrication with increasing the load have been already observed in the literature [144]. The results from Figure 7-16 show that by adding suitable thickener to the oil and using full grease formulation, the vibration amplitudes is decreased drastically thus increasing the noise reduction performance of the rolling element bearing.

At 1, 5 and 10 kN radial loads, Berutox FH 28 EPK 2 shows respectively 50%, 53% and 57% and lower RMS amplitude compared to PAO. In particular in higher loads shows Berutox FH 28 EPK 2 grease more advantages in decreasing vibration amplitudes over PAO base oil.

The damping ratio of Berutox FH 28 EPK 2 show an increase around 4% from 1 to 5 kN and 10% by increasing the radial load from 5 to 10 kN. The overall increase in damping ratio of Berutox FH 28 EPK 2 was 14% (damping ratio difference of 0.008). As seen previously the accuracy of the damping calculation was 0.006. To avoid misinterpretation due to this error, it can be concluded that at least the trend of decrease in damping ratio with increase in load which was observed in PAO does not occur in case of using complete grease.

At 1 kN radial load, Berutox FH 28 EPK 2 shows 20% higher damping ratio compared to PAO. By increasing the load to 5 kN, Berutox FH 28 EPK 2 results in 22% lower damping ratio compared to the base oil. At 10 kN, Berutox FH 28 EPK 2 shows 78% higher damping ratio compared to PAO. The damping of the grease shows higher deviation to the damping of the PAO over load. This shows the advantage of using the grease specially in higher loads to increase the noise reduction of the rolling element bearings.

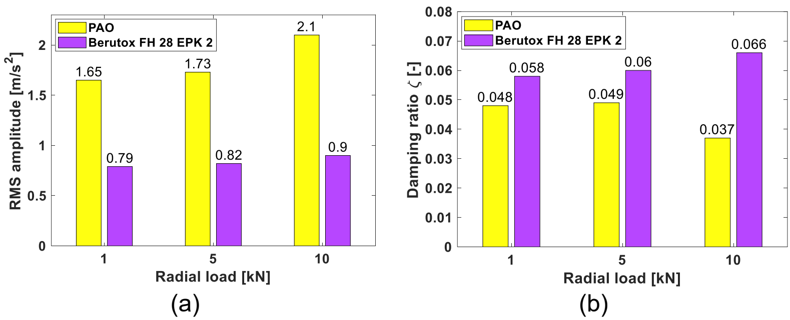
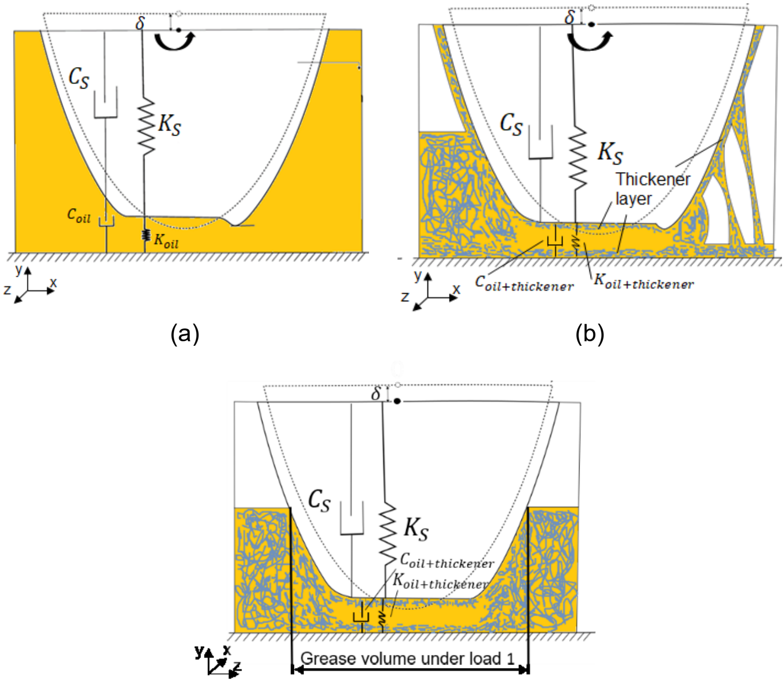


Figure 7-16: Comparison of the (a) RMS amplitude and (b) damping ratio of the vibration analysis of the rolling element bearings lubricated with petrochemical grease Berutox FH 28 EPK 2 and PAO as the base oil at 40°C

To discuss the effect of thickener of the grease on its vibration behaviours, the vibration models of the EHD contact must be compared between grease and oil lubrication. In case of fully flooded oil lubrication, the stiffness and damping of the contact are composed of both the structural components and the fluid film stiffness and damping, as mentioned by several researchers [134,143]. This is schematically shown in Figure 7-17 (a). Since damping of the structure is shown to be smaller than the lubricant ($C_S \ll C_{oil}$), the equivalent damping of the single contact is dominated by EHD damping.

In the case of grease lubrication, an adhesive thickener layer forms on the roller and the raceways as reported in previous researches [145,146] as shown in Figure 7-17 (b). Therefore, the stiffness and damping of the EHD contact will be determined by both the stiffness and damping of the oil and the thickener together. Greases show also a sticky behaviour which is called tackiness of the grease [10], which may also have an influence on the dynamic behaviour of the contact. Under starved conditions in grease lubrication, the amount of the released oil in the contact area is limited, thus lower film thickness compared to fully flooded oil lubrication with the same operational conditions [147]. Looking into the contact area from front view, it can be seen that the grease usually forms a track line on the raceway (Figure 7-17 (c)). This grease layer gets pushed to sides by passing the roller at the beginning of the rotation in so called churning phase [10]. In this case only a fraction of the grease is under the load and shearing of the roller and the rest only provides base oil through capillary effect into the contact area as a reservoir [148].



(c)

Figure 7-17: Schematically demonstration of stiffness and damping of the single contact of a rolling element bearing in (a) fully flooded oil lubrication from side view (b) grease lubrication side view (c) grease lubrication front view

Only a fraction of the grease volume is under the dynamic squeezing between the roller and the raceway. Therefore, only the volume of the grease which is under the squeezing and shearing of the roller has to be considered in dynamic stiffness and damping model of the contact. It is hard to measure how much of the stiffness and damping of the contact is dominated by the oil volume and grease thickener separately due to measurements limitation. In vibration measurements in this work it could be seen that by increasing the load the damping of the grease increases despite the decrease of the damping of the fully flooded oil. Under higher loads the contact bodies are deformed more, thus increasing the contact area (Figure 7-18 (a,b)). By increasing the contact area, there is more grease volume between the roller and the raceway which is applied to squeezing and shearing effects of the contact bodies. Larger volume of the damping material in the contact area may cause better damping abilities of the grease by increasing load compared to the fully flooded oil lubricated contacts, where amount of oil is constant. Furthermore, by increasing the load for constant rotational speeds the film thickness in fully flooded lubrication decreases. However, by increase in the load, the thickness of deposited-thickener layer stays almost constant [85]. Therefore, the film thickness stays more or less constant due to domination of deposited thickener layer.

Looking closer to contact area, under harsh conditions of higher loads the film thickness decreases in oil lubrication to nanometre scales. In this case the roughness of the contact bodies also plays and important role. In case of grease lubrication, the thickener layer may fill the valleys Figure 7-18 (c,d). In mixed lubrication conditions, the grease layer can damp the vibrations caused by collision of the roughness local peaks of the roller and the raceway as well as the roller and the cage, thus increasing the damping of vibrations compared to the oil lubrication.

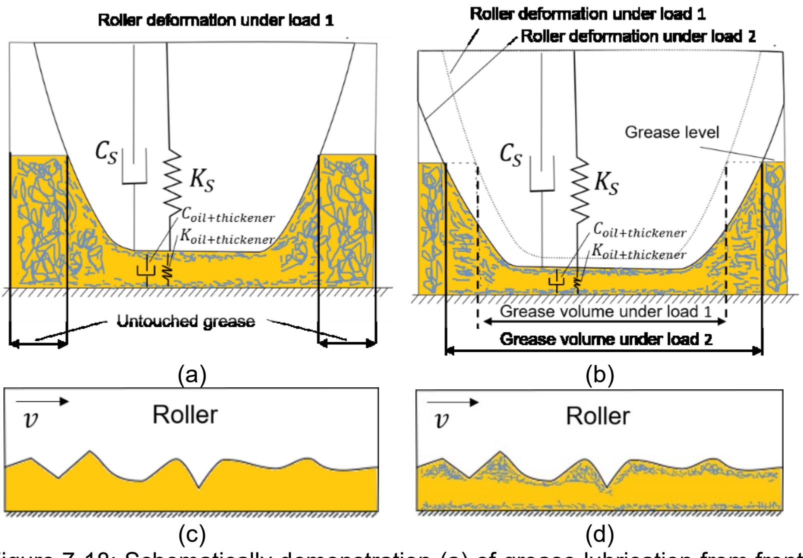


Figure 7-18: Schematically demonstration (a) of grease lubrication from front view under with smaller load, (b) higher loads, (c) oil lubrication contact area with roughness and (d) grease lubrication contact area with roughness

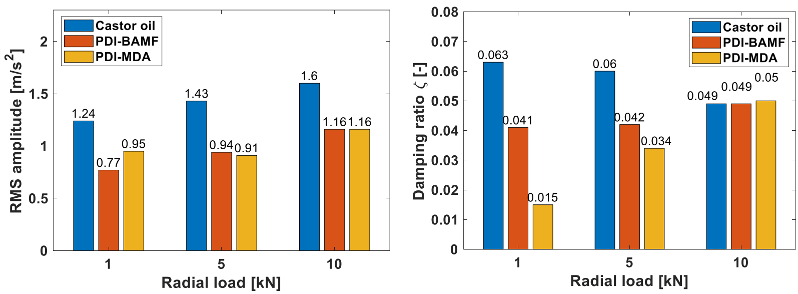
Next, the comparison of the results for bio-based polyurea greases PDI-BAMF and PDI-MDA with their base oil will be discussed. To do so, the RMS and the damping ratios are visualized and compared in Figure 7-19.

It can be observed, that the RMS amplitude of the PDI-BAMF shows an increase of 22% and 23% (overall RMS difference of 0.39 around 50%) by increasing the radial load from 1 to 5 kN and from 5 to 10 kN. The PDI-MDa shows around 4% (RMS difference of 0.04) decrease from 1 to 5 kN which may be due to measurements error (RMS difference of 0.019) or inhomogeneity of the grease formulation. From 5 to 10 kN shows PDI-MAD 22% increase in RMS amplitude. Overall shows PDI-MDA 22% increase in RMS values over the whole load range. This shows higher sensitivity of the PDI-MDA RMS amplitudes to the load compared to PDI-BAMF grease. For both greases the trend of increasing the RMS values with increasing the load is observed in the diagrams. This effect has been already observed in the literature [144].

At 1, 5 and 10 kN radial loads, PDI-MDA shows around 30%, 57% and 37% lower RMS amplitude compared to castor oil as the base oil. The RMS amplitudes of PDI-BAMF are 61%, 52% and 37% lower at 1, 5 and 10 kN radial loads compared to the castor oil. The data shows a decrease in ability of the PDI-BAMF thickener to decrease the RMS values of the pure base oil by increasing the load. However, the results of PDI-MDA grease seem to scatter more and a clear trend could not be concluded.

The damping ratio of the PDI-BAMF shows minimal increase by increasing the radial load from 1 to 5 kN. By further increase of the load from 5 to10 kN, the damping ratio increases more significantly around 16%. Overall, PDI-BAMF damping ratio increase around 19% over the load range. The PDI-MDA shows however more significant increase of 126% (damping ratio difference of 0.019) by increasing the radial load from 1 to 5 kN and 47% increase by further increase of the load from 5 to 10 kN. Overall, shows PDI-MDA significant increase of 223% in damping ratio over the whole load range. Both greases show the trend of increasing the damping ratio by increasing the load.

At 1 kN radial load, PDI-MDA shows 76% lower damping ratio compared to castor oil and PDI-BAMF shows 34% lower damping ratios compared the base oil results. By increasing the load to 5 kN, PDI-MDA results in 43% lower damping ratio compared to castor oil and PDI-BAMF shows 29% lower damping ratio compared to the castor oil. At 10 kN, PDI-MDA and PDI-BAMF show comparable damping ratio to the castor oil. It can clearly be seen that the deviation of the damping between the base oil and both greases decrease with increasing the load. That means damping ability of the rolling element bearings lubricated with the developed bio-based greases will be enhanced and at some point, comparable with base oil lubrication by increasing the load. The unexpectedly lower damping of the bio-based greases compared to the base oil under lower loads may be due to the different gel-like formation of the thickeners compared to the reference grease. This phenomenon needs to be investigated in more detail, which is beyond the scope of this work.



(a) (b)

Figure 7-19: Comparison of the (a) RMS amplitude and (b) damping ratio of the vibration analysis of the rolling element bearings lubricated with developed bio-based greases PDI-BAMF and PDI-MDA and castor oil as the base oil at 40°C

In lower loads the bio-based polyurea thickener PDI-BAMF shows advantages over PDI-MDA (lower RMS and higher damping values). This may be tracked back to their thickener structures as shown in Figure 7-20 from [91]. The PDI-BAMF thickener show smaller pores and the distribution of the fibres is smoother thus acting better as reservoir for the base oil. This causes also better bleeding abilities of the grease. In PDI-MDA however, the thickener fibres are longer and form bigger thickener particles which are poor distributed and agglomerated. The pores in the structure are sometimes around 0.9 μm It was also seen that more hydrogen bonds were captured in IR spectroscopy which may be correlated to bigger particles as reported by [91]. By increasing the force, the thickener system of PDI-MDA may be deformed and sheared creating more uniformly distributed thickener around the contact area which causes the oil to bleed easier to the contact area thus better lubrication of the contact higher damping of the vibration signals.

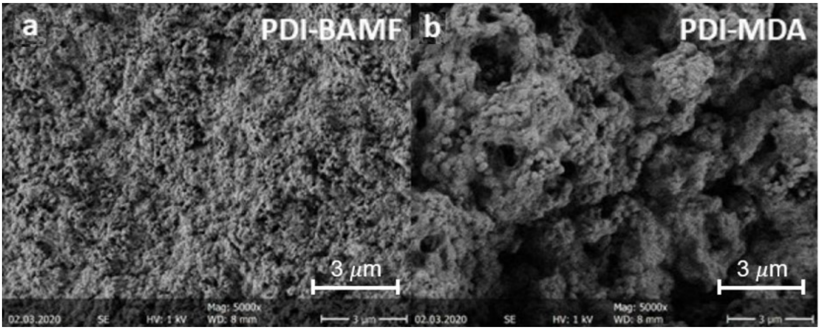
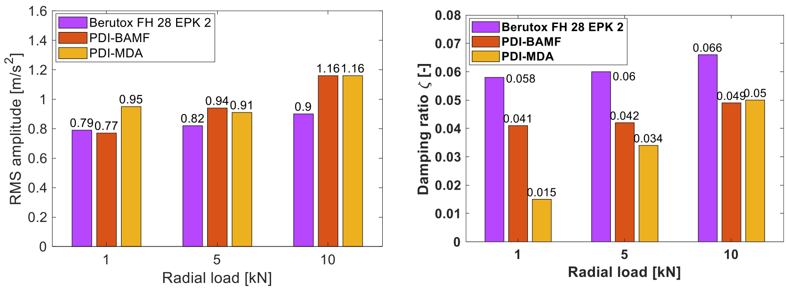


Figure 7-20: Thickener structure of bio-based PDI-BAMF and PDI-MDA under SEM as reported by [91]

To evaluate the noise reduction abilities of the developed bio-based polyurea greases with the petrochemical grease, the vibration results of PDI-BAMF, PDI-MDA and Berutox FH 28 EPK 2 are shown in Figure 7-21. At 1 kN, petrochemical grease Berutox FH 28 EPK 2 shows 20% lower RMS amplitudes compared to PDI-MDA and 5% higher than PDI-BAMF. By increasing the load to 5 kN, the petrochemical grease shows 10 and 14% lower RMS amplitude compared to the bio-based greases. At 10 kN, the petrochemical grease shows 28% lower RMS amplitude compared to the bio-based greases. In low load ranges are the RMS reduction ability of the bio-based PDI-BAMF is comparable to better than petrochemical grease.

The damping ratio of the Berutox FH 28 EPK 2 at 1 kN radial load is higher than PDI-MDA by 280% and 41%higher than PDI-BAMF. By increasing the load to 5 kN, the petrochemical grease shows 76 and 42% higher damping ratio compared to PDI-MDA and PDI-BAMF. At the highest radial load 10 kN, shows Berutox FH 28 EPK 2 around 34% higher than both bio-based greases. Damping ratios of all three greases increase with the increasing radial load.

Other advantage of the petrochemical grease is that in the waterfall diagrams only 4th harmonics was identified as the dominant order. However, in both bio-based greases 3rd order vibrations were also significant. Overall it can be observed that the petrochemical grease Berutox FH 28 EPK 2 shows the best results regarding decrease of vibration compared to its base oil as well as enhancement of the damping ratios over the load range. The noise reduction ability PDI-BAMF is comparable to the petrochemical grease. At higher loads, the noise characteristics of the PDI-MDA shows less deviation to the petrochemical grease. The slightly better noise performance of the petrochemical grease may be tracked back to the optimized thickener system and compatible additive packages compared to the bio-based and need to be further investigated in the future works.



(a) (b)

Figure 7-21: Comparison of the (a) RMS amplitude and (b) damping ratio of the vibration analysis of the rolling element bearings lubricated with petrochemical grease Berutox FH 28 EPK 2 and developed bio-based greases PDI-BAMF and PDI-MDA at 40°C

7.3.2. Effect of temperature on vibration behaviour of grease lubricated rolling element bearings

To investigate the effect of temperature on the vibration behaviours of the rolling element bearings lubricated with greases, measurements have been conducted on PDI-BAMF at room temperature at 25 °C and compared to the previous shown results at 40 °C. The results of the measurements are summarized into Table 7-5.

Comparing RMS amplitudes at 1 kN, PDI-BAMF results at 25 °C and 40 °C show very close results. However, by increasing the radial load to 5 kN the RMS amplitude of the results of the measurements at 25 °C show 76% higher amplitudes. By increasing the radial load further to 10 kN the RMS amplitudes of the measurements at 25 °C still show 20% higher values than the measurement at 40 °C.

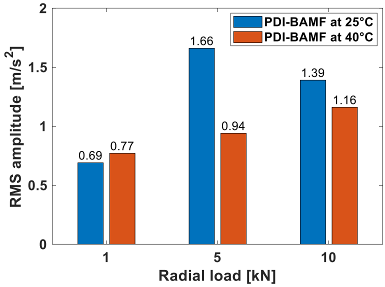
The damping ratio of the PDI-BAMF lubricated rolling element bearing at 1 kN stays almost contestant by increasing the temperature to 40 °C. At 5 kN the increase of damping ratio by increasing the temperature is measured to be more significant about 250%. At 10 kN the damping ratio is increased by 88% with increasing the temperature from 25 to 40 °C.

Table 7-5: Vibration peaks of vibration measurements on the rolling element bearings lubricated with developed bio-based greases PDI-BAMF at 25°C and at 40°C for different applied radial loads

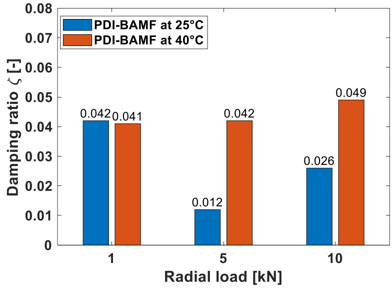
	Radial load [kN]	Frequency [Hz]	Rotational Speed [rpm]	RMS Amplitude [m/s^2]	Damping Ratio ζ [-]
25 °C	1	152.47	2287.1	0.694	0.042
	5	145.73	2186.1	1.665	0.012
	10	144.59	2168.9	1.393	0.026
40 °C	1	147.45	2211.8	0.772	0.041
	5	145.58	2183.8	0.944	0.042
	10	144.87	2173.1	1.164	0.049

For better comparison of the RMS and damping ratios, the results are visualized into Figure 7-22.

Results show an increase in RMS amplitude for both temperatures by increasing the radial load. However, the measurements at 40 °C for medium to higher radial loads from 5 kN show significantly lower RMS values. Also, the damping ratios for medium to higher radial loads from 5 kN are higher than the measurements at 25 °C and thus, better vibration behaviour. It has been shown by physicochemical characterisations in [91] that the polyurea grease samples showed almost solid behaviour at 25 °C. That means the oil bleeding in the contact area at 25 °C under lower loads is not enough to release the oil into contact area and therefore only grease particles lubricate the contact. By increasing the temperature to the 40 °C, the oil inside the thickener has lower viscosity and more oil volume can reach the contact area through the thickener due to better permeability of porous thickener system [148].



(a)



(b)

Figure 7-22: Comparison of the (a) RMS amplitude and (b) damping ratio of the vibration analysis of the rolling element bearings lubricated with bio-based grease PDI-BAMF at 25 and 40°C

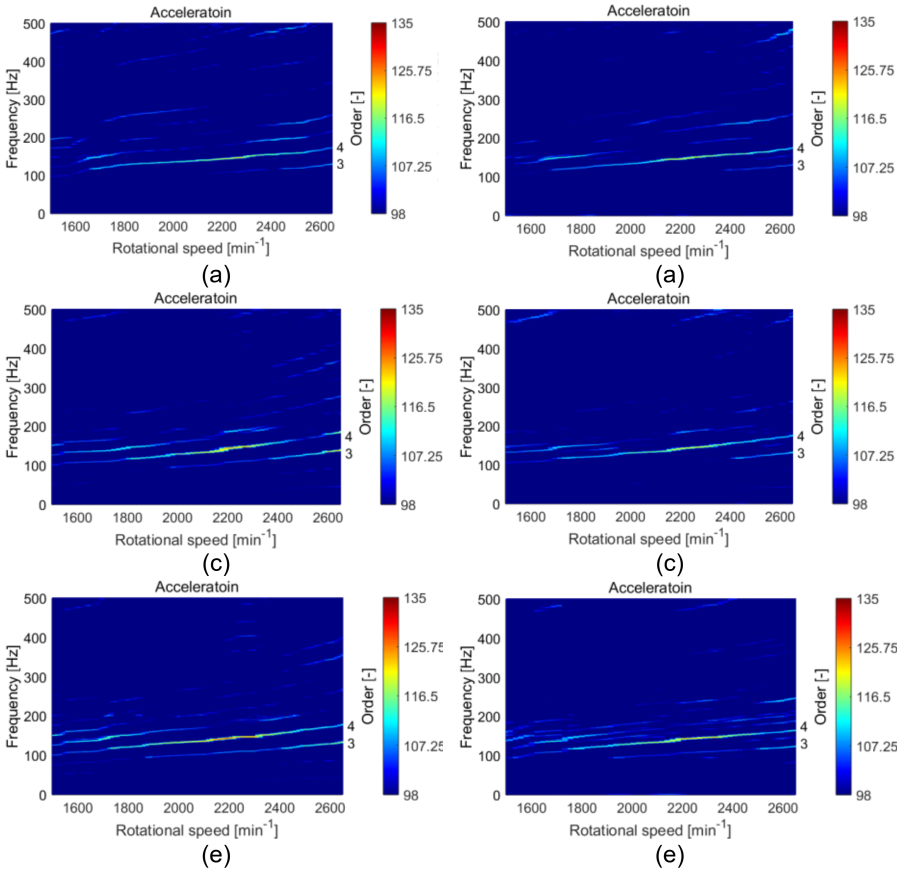


Figure 7-23: Waterfall diagrams of vibration measurements on radial rolling element bearing test rig lubricated with PDI-BAMF at (a-e) 25°C and (b-f) 40°C with rpm sweep from up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

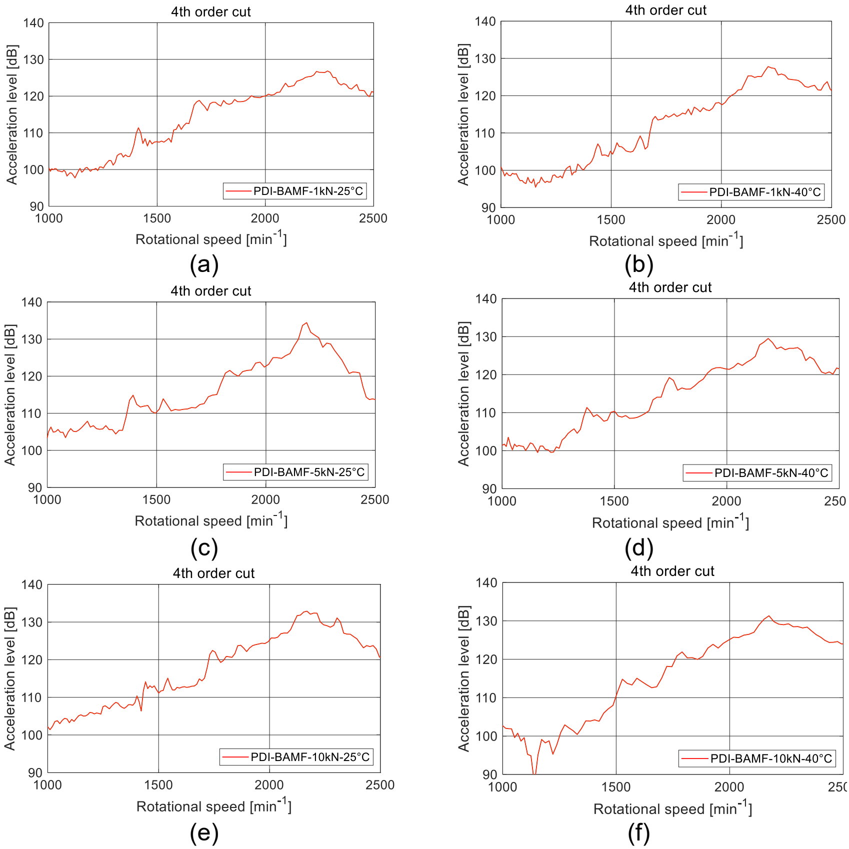


Figure 7-24: Colormap and order cut diagrams of vibration measurements on radial rolling element bearing test rig lubricated with PDI-BAMF at (a-e) 25°C and (b-f) 40°C with rpm sweep from up to 3000 min^{-1}. (a,b) under 1 kN radial load equal to 1.33 GPa (c,d) under 5 kN radial load equal to 1.75 GPa (e,f) under 10 kN radial load equal to 2.02 GPa

7.4. Chapter conclusion

The primary objective of chapter 7 was to experimentally validate the third proposed research hypothesis:

3 The vibration characteristics of rolling element bearings will not critically deviate between those lubricated with developed bio-based greases and those lubricated with the petrochemical reference grease. .

To this end, vibration characteristics of the developed bio-based and petrochemical greases were compared to their base oils and to one another using a radial bearing test rig equipped with acceleration sensors. The results indicated that the RMS amplitude of the grease-lubricated system is significantly lower compared to oil lubrication mainly due to the presence of thickener in the grease. Further investigations over varying loads revealed that damping of the oil-lubricated system showed a descending trend upon increasing load. However, in grease lubricated scenarios in this work, the damping ratio was observed to be either constant or increasing by increasing the load. These observed differences in vibration characteristics of the oils and greases elucidates the role of thickener system. The resulting ranking of the greases (Berutox FH 28 EPK 2≈BAMF≻MDA) in terms of noise reduction characteristics, as determined from the vibration tests, could be directly correlated with the previous tribometer results, as well as the results of wear and lifetime tests conducted on the FE8 and radial bearing test rig. It was observed that the damping ratio of bio-based polyurea grease PDI-BAMF was comparable to Berutox FH 28 EPK 2 as the petrochemical grease. However, for PDI-MDA there is a need to enhance the thickener system to increase the noise reduction abilities. **Except for lower load ranges in the measurements, the noise characteristics of the bio-based greases did not show a significant deviation to the petrochemical reference grease and therefore the third hypothesis of this work is validated.**

8. Summary and outlook

In the field of machine elements, lubricants primarily consist of petrochemical substances. Over 80% of rolling element bearings are lubricated with greases derived mainly from petrochemical sources. However, there has been a notable shift within the lubrication industry towards adopting more sustainable resources for lubricant production. To create fully sustainable bio-based lubricating greases as an alternative to petrochemicals, it is necessary to develop both the base oil and thickener systems using bio-based materials.

Despite recent advancements in bio-based grease technology, most of the developed thickener systems for bio-based greases in the literature are vegetable-based and show inferior tribological performance, such as friction, wear, and lubrication film formation, compared to conventional petrochemical lubricating greases, specifically with the polymeric thickeners. In a mutual project between TU Dortmund, Carl Bechem and MSE, almost fully bio-based polymeric greases based on bio-based polyurea, polyester, and polyamide thickeners have been introduced [90].

The specific objective of this work was **to determine the tribological properties (such as film formation, friction, and wear) of the developed polymeric bio-based greases both at tribometer and bearing level and to compare it to the petrochemical counterpart**.

To support the objective, three hypotheses have been proposed and investigated during the work. The hypotheses are as following:

1. The film formation of bio-based greases on tribometer level is significantly influenced by the thickener type and can be correlated with chemical parameters of thickener, such as structure, and flow limit.

To evaluate the first hypothesis, tribological tests regarding film formation and friction on a ball-on-disc tribometer were conducted on the greases developed with bio-based thickener systems, i.e., polyurea, polyester and polyamide thickeners (Chapter 5). The ball-on-disc tribometer has been used as the screening method to identify grease candidates with promising tribological performance and support early-stage development. It was shown that the grease candidates with polyurea (PDI-BAMF and PDI-PDA) and polyester (DDS-PrD at higher speeds) thickeners resulted in comparable film thickness and friction coefficients in comparison to the reference petrochemical grease Berutox FH 28 EPK 2. Although the results depended on the rolling speeds, it was generally shown that the polyurea greases have superior film formation and friction performance compared to other polymeric thickeners. Therefore, only this thickener system has been selected for further component evaluation tests. The tribological performance of the grease samples at the tribometer level was also correlated with the chemical parameters of the thickener, such as structure, flow limit, and chemical bonds within the chemical structure of the thickeners. This correlation provided essential information for estimating the tribological behaviour of the polymeric greases in an early-stage development phase, e.g., the formation of a foam-like thickener system is essential for the suitable performance of the greases. The observations from the tribometer tests confirmed the validity of the first hypothesis.

2. The performance of developed greases at the component level on rolling element bearing test rigs can be correlated to the results of tribological screening tests and confirms the suitability of the screening method.

The bio-based polyurea and polyester grease samples have shown promising results and comparable performance to the petrochemical grease at the tribometer level. In particular, the grease samples with the bio-based polyurea thickener systems have shown comparable to superior performance regarding the film formation and friction coefficients compared to the petrochemical grease. Therefore, bio-based polyurea greases have been further evaluated regarding wear protection characteristics on FE8 and lifetime on radial bearing test rigs at component level (Chapter 6). The results of ranking of the greases from the component tests regarding wear protection characteristics and lifetime were in agreement with the results at the tribometer level regarding film formation and friction (Berutox FH 28 EPK 2≈BAMF≻MDA). The results at the component level and the comparability with the results at the tribometer level confirmed suitability of screening tests and the validity of the second hypothesis.

Additionally, the vibration analysis was used as a further tool to assess the noise reduction characteristics of the developed thickeners. To ensure that the developed greases with bio-based polymeric systems do not show adverse effect on the noise characteristics compared to the petrochemical reference grease, the following hypothesis was formulated:

3. The vibration characteristics of rolling element bearings will not critically deviate between those lubricated with developed bio-based greases and those lubricated with the petrochemical reference grease.

The vibration properties of the developed bio-based and petrochemical greases were analysed, comparing them to their respective base oils and one another. The measurements were conducted using a radial bearing test rig with acceleration sensors (Chapter 7). The results indicated that the vibrations of the grease-lubricated system are significantly lower compared to oil lubrication, primarily due to the presence of thickener in the grease. Further investigations over varying loads revealed that damping of the oil-lubricated system showed a descending trend upon increasing load. As the novel finding of this measurement, it was shown that in grease-lubricated scenarios in this work, the damping ratio was observed to be either constant or increasing by increasing the load. By increasing the contact area, there is more grease volume between the roller and the raceway which is applied to squeezing and shearing effects of the contact bodies. Larger volume of the damping material in the contact area may cause better damping abilities of the grease by increasing load compared to the fully flooded oil lubricated contacts, where amount of oil is constant.

The observation from vibration measurements showed that the vibrations of the grease-lubricated contacts are different from those of the oil-lubricated contacts. Except for the lower load ranges in the measurements, the noise characteristics of the bio-based greases did not show a significant deviation from the petrochemical reference grease, and therefore, the third hypothesis of this work has been validated. When comparing the noise characteristics of the bio-based polyurea greases with the petrochemical reference grease, comparable performance was observed, which is in agreement with the performance results from film formation and friction tests at the tribometer level (Chapter 5), as well as the wear protection and lifetime results at the component level (Chapter 6).

By testing the research hypotheses of this work, a holistic approach for the evaluation of lubricating greases for rolling element bearings at both tribometer and component levels was employed that provides a fundamental understanding of the lubrication mechanism and supports early-stage development. The correlation of the results from different methods at different levels could confirm the competitive tribological performance of the developed bio-based polyurea greases and the petrochemical grease. “The tribological properties (such as film formation, friction, and wear) of the developed polymeric bio-based greases both at tribometer and bearing level and comparison to the petrochemical counterpart” were determined, and this work's objective was achieved.

The bio-based polyurea greases in this work have been produced on a kilogram scale (scaled up) using the same reactor used for production of petrochemical polyurea greases. The scale-up and further evaluation have been done by the industrial project partner, Carl Bechem company, and the bio-based polyurea greases are planned to be optimized and released to the market. Furthermore, the development methodology and the knowledge gained from the correlation of physicochemical and tribological results supported Carl Bechem in optimizing existing products and producing further lubricating greases.

The results of this study provide the basis for estimating the impact of chemical parameters on tribological performance and supporting the synthesis process during the future early-stage development of the greases. Additionally, the tribological evaluation results across different levels provide insights into the grease lubrication mechanism and can serve as a workbench framework for future assessments of bio-based greases for rolling element bearing applications. Additionally, the findings from vibration tests on greases contribute to understanding the role of thickeners in vibration models of grease-lubricated contacts. This serves as a basis for future vibration analyses of grease-lubricated rolling element bearings.

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