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ORIGINAL PAPER

Assessment of Water Contamination on Grease Using the Contact Angle Approach

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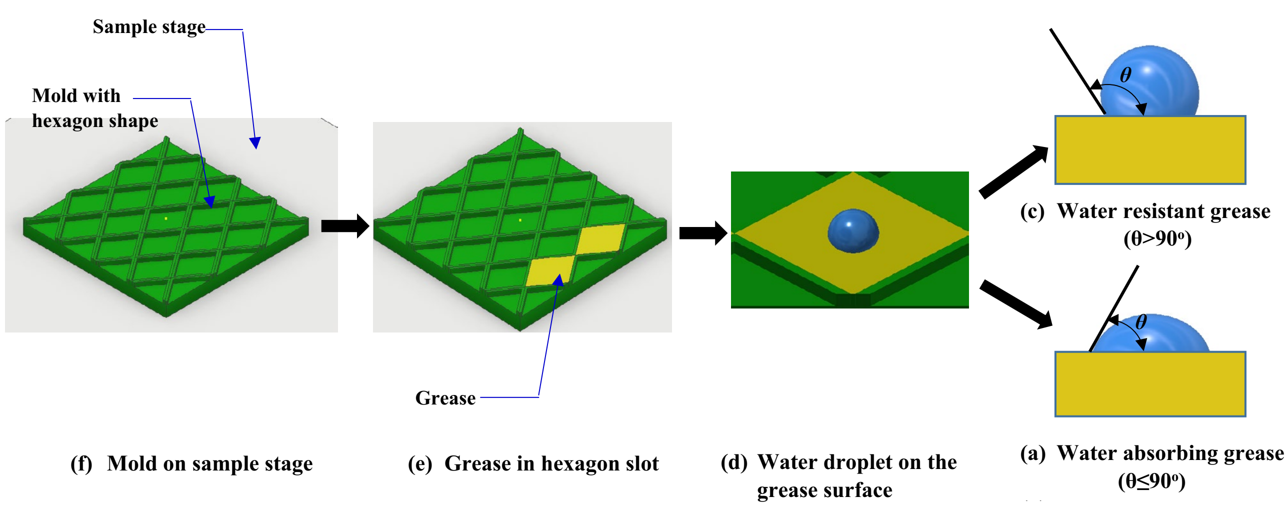
Abstract
Many grease-lubricated machinery components operate in wet environments, making them susceptible to water contamination. It has been reported that the presence of a mere 1% water contamination in grease reduces the life of a bearing by 90%. Yet there are only a few standards available to characterize grease performance in the presence of water, most of which are primarily qualitative, providing inconclusive information, often relative to standard samples, at a significant cost in time and materials. In this paper, a unique approach is proposed for evaluating grease capability for repelling water. The method utilizes the contact angle of a water droplet on a grease surface to quantify grease water resistance. It is hypothesized that the higher the hydrophobic nature of the grease, the greater the resistance to water and vice versa. The validation of the hypothesis is established by performing extensive contact angle measurements on seven types of commercially available greases. The efficacy of contact angle results is established by measuring the change in the yield stress values obtained using a rheometer. From the obtained results, it is shown that the proposed approach is reliable and capable of distinguishing greases as water repellent/attractive.

Graphic Abstract



Keywords Grease performance · Contact angle · Rheological properties · Water contamination · Water resistance

1 Introduction

Grease is a highly complex lubricant composed of thickener, additives, and base oils [1]. Compared to lubricating oil, grease provides good sealing properties, requires less

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maintenance, and can even operate in a solid-contaminated environment [1]. Further, the availability of greases in a range of viscosities offers application flexibility that satisfies the requirement of different industry sectors ranging from automotive, chemical processing, metalworking, power generating, and food and beverage processing factories [2].

Research shows that the working life of grease is profoundly affected by contaminants [3], such as water, which tends to modify their chemical structure and degrade their rheological properties, which, in turn, negatively affects the performance of the machinery components [4]. The significance of water contamination can be comprehended from the findings provided in many reports; some of the findings are summarized below:

- Duncanson [5] reported deprivation of the grease additives and the formation of acid occurs 10 times faster in the presence of water.
- The presence of water in a bearing resulted in the reduction of film thickness, causing flash vaporization, erosive wear and hydrogen embrittlement due to hydrolysis [6].
- Mere 1% of water reduced the life of a journal bearing by 90% [7].
- Water content as low as 0.02% (200 ppm) negatively affects the service life of roller bearings [8].
- An increase in water content from 13 to 38% resulted in a sizable increase in the four-ball wear scar for calcium sulfonate complex grease from 0.38 and 0.48 mm [9].
- The emulsification of grease due to water led to a significant reduction in adhesive and cohesive properties [10, 11]. This behavior deteriorate the performance in steel rolling mills [12].
- Some greases become firmer with water, leading to a reduced ability to replenish the contacts [12] and resulting in excessive power consumption [9].
- Free water in the water repellent greases entered through the seal, causing corrosion and resulting in bearing failure [11, 13].
- A very recent review article [14] states that many commercially available greases are marketed as water-resistant without providing a quantitative justification and that the overall criteria for defining the water resistance properties of the grease need to be improved.

Clearly, water contamination triggers several structural and chemical mechanisms leading to premature failure of grease. Thus, appropriate grease selection can prolong the safe working operation of vital components/machinery. For example, it has been reported that by applying a water-resistant grease in a pipe mill bearing, the failure rate of bearings due to water washout was reduced from a frequency of 4 per month to 1 per month, saving $5500 in bearing costs and $20,000 in production costs [2].

Numerous standardized tests have been developed over the years to characterize the behavior of grease in the presence of water. The most pertinent ones are DIN 51807-1, ISO 11009:2000/ASTM D1264, ASTM D7342, ASTM D4049, ASTM D1743/D6138/D5969/D7038/D4048, and a few non-technical testing procedures that rely on visual inspection, static and dynamic water absorption test, crackle test, etc. Despite best attempts, practitioners and researchers contend that these standard tests provide only qualitative and inconclusive information on the performance of lubricating greases with water contamination [10]. Further, it is even harder to find any literature relating to the studies that examine real applications [10].

Researchers [4, 15–18] have reported using the rheological properties of grease, such as yield stress, zero shear viscosity to quantitatively evaluate the change in grease performance in the presence of water, often by employing a rheometer. This approach does provide satisfactory results; however, the evaluation of grease performance is determined relative to a standard value. To this end, it is important to mention that the behavior of the grease chemistries to water contamination is not the same. Specifically, some calcium/lithium-based soaps thicken in response to water contamination, while other greases tend to become thin. This trend makes it difficult for the practitioner to select the best water repellant candidate between two greases having the same magnitude of change where one thickens while other thins. Therefore, this approach fails to assess the water-repellant properties in a single platform. Further, the quantity of sample required for testing makes this approach difficult to use in the assessment of used grease samples collected from process operations.

The present paper reports the development of a unique measuring technique for correlating the behavior of the grease in the presence of water. As reported by Leckner [10], the behavior of grease in the presence of water is strongly dependent on the availability of polar components and the arrangement of surface-active thickener and additives in the grease. As reported by Cyriac [4], the water-repelling or absorption capability of grease depends on the hydrophobic/hydrophilic nature of the surface-active components and the type of base oil in the grease. This technique leverages the chemistry behind the interaction with surface-active polar components within the grease structure, to assess the behavior of grease in the presence of water.

A simple and effective methodology available for determining the water-repelling nature of grease is to measure the contact angle of a water droplet on a grease surface. A grease having good water-repellent properties will have poor wettability, appear hydrophobic in nature, and exhibit a large contact angle. By the same token, a grease attracted to water tends to have good wettability properties, appear hydrophilic in nature, and exhibits a smaller contact angle.

This methodology provides a quick tool for researchers looking to optimize hydrophobic or hydrophilic characteristics while screening different combinations of additive chemistries and thickeners.

The usefulness of measuring the contact angle values been realized in numerous applications such as oil industries [19], engines [20], super-hydrophobic coatings [21], printing [22, 23], self-cleaning [24, 25], nano-fluids [26, 27], electowetting [28], friction and wear [29], wettability of graphene on plasmonic nano-hole arrays [30], and for examination of the effect of piezoelectric direct discharge plasma in modifying the surface of orthopedic materials [31]. The present work extends the evaluation of the response of grease in the presence of water. It is a direct approach to assess the water resistance property of grease and enables the practitioners to formulate the water resistance property of grease during its development stage to achieve the desired properties.

To validate the hypothesis, as a first step, the resistance of grease is determined by measuring the rheological property (yield stress) using a rheometer. In this approach, the change in rheological properties between the pristine and water contaminated greases is determined to estimate the water resistance property. In the present work, the samples are contaminated with water by the static absorption method, i.e., grease is immersed in water for a definite duration, and the samples are tested after draining the water. Lastly, the contact angle values of the water droplets are measured on the pristine grease surface using a drop shape analyzer. Obtained values of rheological properties and contact angle values are then compared to establish the efficacy of the proposed approach.

The outline of this paper is as follows. Section 2 covers the details of the instruments used for measuring the rheological properties, and the contact angle measurement of greases are provided. Results are presented in Sect. 3, followed by a detailed discussion in Sect. 4. Summary and concluding remarks are presented in Sect. 5.

2 Materials and Methods

Experiments are performed on seven different types of commercially available NLGI grade 2 greases. The details of the greases used are provided in Table 1.

Table 1 Grease designation with thickener and base oil types

Grease label	Thickener types	Base oil type
LiC-m	Lithium complex with molybdenum disulfide	Mineral oil
LiC-P	Lithium complex with molybdenum disulfide and graphite	Poly-alpha-olefin oil
CaS	Calcium sulfonate	Mineral oil
PU	Poly-urea	Mineral oil
Si	Silicone with PTFE	Synthetic oil
AlC	Aluminum complex	Mineral oil
WLi	White lithium	Mineral oil

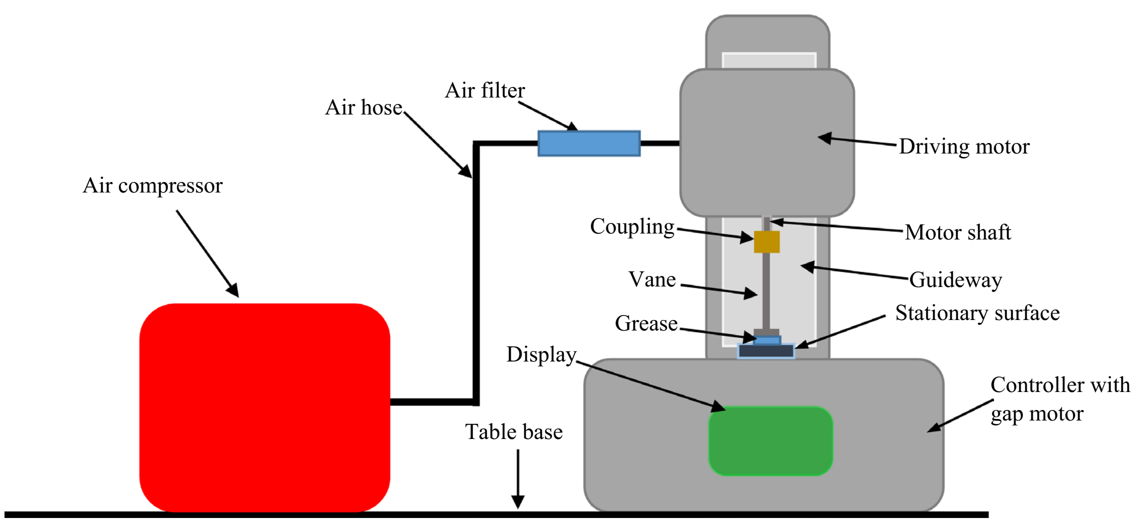


Fig. 1 Schematic representation of rheometer

It is known that mixing grease with water through mechanical shearing action leads to the degradation of grease. Therefore, the grease samples are contaminated in a static-type condition using the following procedure. First, ~ 5 g of a grease sample is immersed in 50 g of water at room temperature (25 °C) for 24 h. After 24 h, water is drained out and the weight of the grease is measured. From the weight of the pristine W_{pr} and water contaminated W_{con} greases, the percentage of water absorption W by the grease is determined using Eq. (1).

$$W = \left(\frac{W_{pr} - W_{con}}{W_{pr}} \right) \times 100 \tag{1}$$

The water-contaminated grease samples are tested in a rheometer for determining the change in the rheological properties, as described in Sect. 2.1. Further, the measurement of the contact angle of water droplets using the droplet size analyzer is given in Sect. 2.2.

2.1 Rheometeric Tests

In the present work, the rheological properties of the grease are studied using the Anton Paar MCR 301 rheometer (see Fig. 1), and the details of this rheometer is provided elsewhere [32]. It is well known that the change the rheological behavior with the presence of water does not adequately represent the water absorption/repletion capacity of grease. In fact, existing approaches are incapable of providing such details [14]. Nevertheless, among the existing test/standards, rheological measurements are more accurate and reliable. Hence, to gain better insight into the performance of greases in the presence of water, they are applied in this study.

To measure the rheological properties, a sample (2 mm thick covering the plates' 15 mm-diameter) is placed on to the stationary surface. When the grease sample is pressed between the plate and stationary surface, a deformation history is observed. To eliminate the deformation history, the gap between the plate and sample is closed by controlling the force until the required sample thickness of 1 mm is reached. The excess sample protruding outside the plate is then trimmed off and sufficient rest time is provided for the applied force to relax. There are several rheological properties through which the grease performance can be evaluated; however, in the present work the yield stress values are considered for evaluating the performance of the grease.

According to Cyriac et al. [16], the yield stress of the grease is defined as the point on the stress–strain curve where the linear behavior of the grease deviates from the experimental data by a prescribed amount (see Fig. 2), or it is taken at the stress level at which the Maxwell model is no longer linear. For a lubricating grease, the linear region is attributed to the solid characteristics of the thickener structure and it is observed to be occurring at the lower level of strain. For a grease, there exists no defined elastic–plastic behavior and the linearity gradually reduces with the increase in strain values (see Fig. 2). Cyriac et al. [16] defined the yield stress as the stress at which the linear behavior deviates by 0.5%, akin to that of steel. In other words, the yield stress is the point where the coefficient of determination (R^2) between the third-order polynomial fit of the experimental values and the linear fit is greater than 99.5%.

The stress–strain plot is obtained by adopting the procedure proposed by Cyriac et al. [16], where the plate of the rheometer is oscillated at a fixed frequency of 1 Hz while maintaining the grease sample thickness of 1 mm and sweeping the amplitude of the strain from 0.01 to 1 [16, 33, 34]. Cyriac et al. [16] concluded that the oscillatory strain sweep approach is more robust, reliable and repeatable compared to other existing methods due to the negligible slip of the

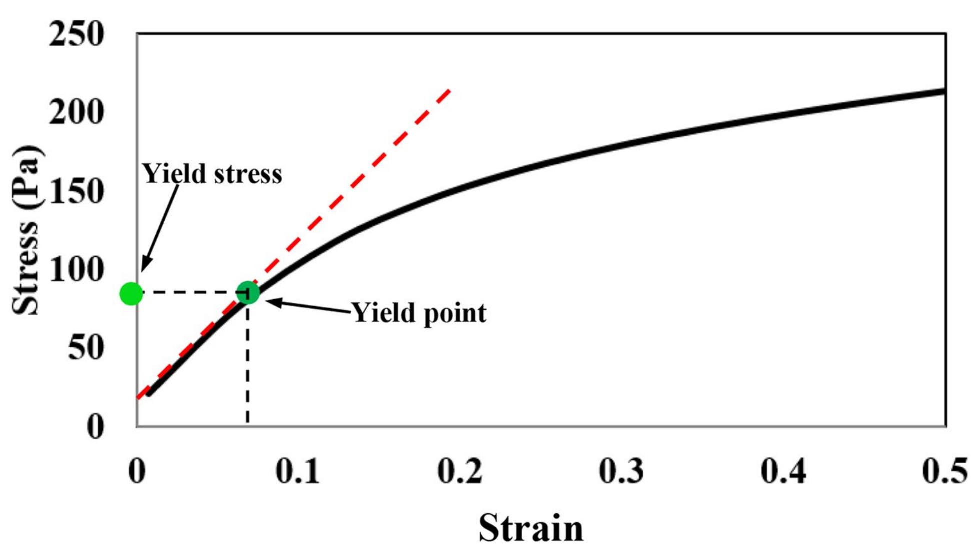


Fig. 2 Stress–strain rate plot with yield point and stress marked

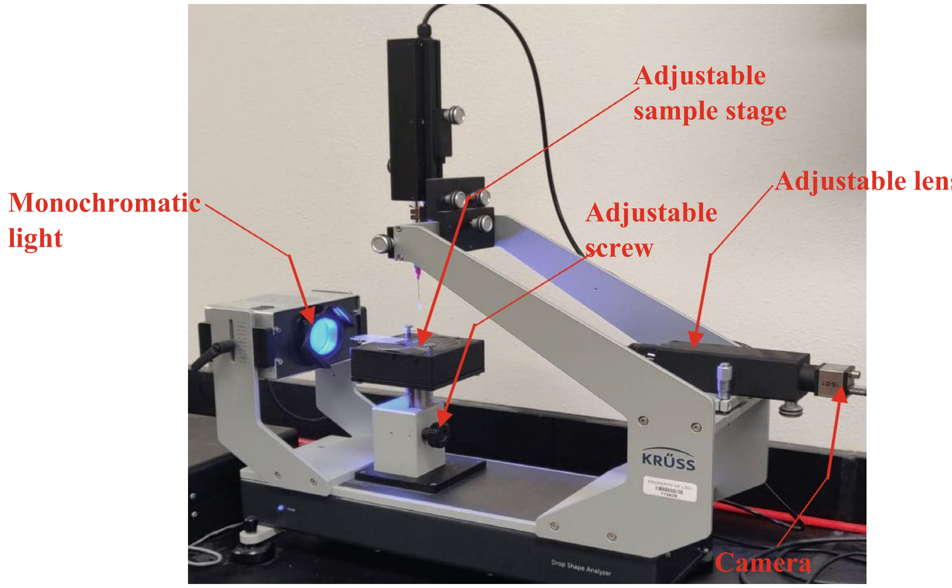


Fig. 3 Drop shape analyzer

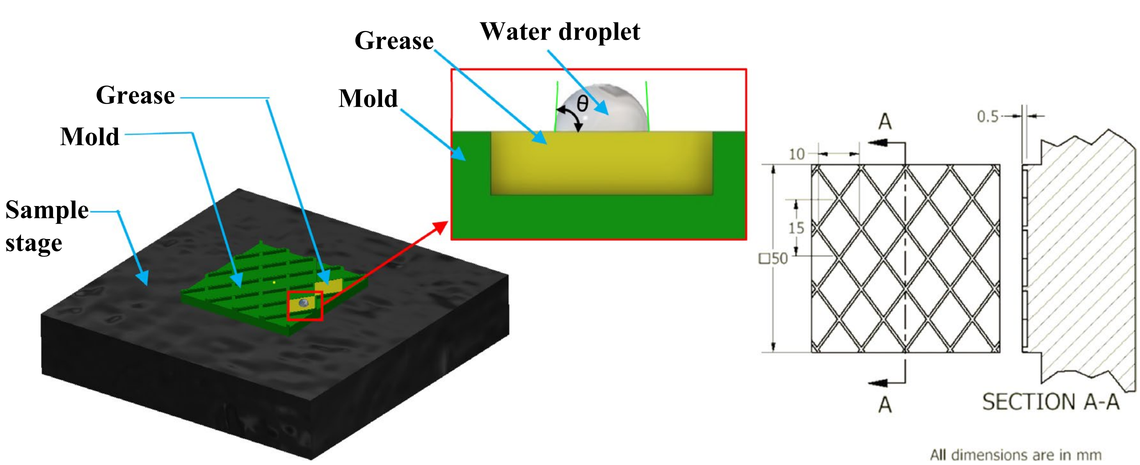
plate in linear viscoelastic (LVE) range. The measurement obtained during oscillatory strain sweep is insensitive to by the plates' geometry, surface roughness, the gap between the plates, and the frequency of shearing.

The stress–strain plot is obtained by oscillating the vane of the rheometer at a fixed frequency of 1 Hz while maintaining the grease sample thickness of 1 mm and sweeping the amplitude of the strain from 0.01 to 1. To determine the point at which a deviation occurs in the stress–strain plot, the procedure proposed by Cyriac et al. [16] is adopted. According to this procedure, the yield stress is the point where the coefficient of determination (R^2) between the third-order polynomial fit of the experimental values and linear fit, is greater than 99.5%.

The level of degradation of the grease with water contamination is determined from the percentage change in the yield stress properties $\tau_{diff}\%$ between the pristine and the water contaminated grease. The value of $\tau_{diff}\%$ is determined using Eq. (2).

$$\tau_{diff}\% = \frac{(\tau_{pr} - \tau_{con})}{\tau_{pr}} \times 100 \tag{2}$$

where τ_{pr} and τ_{con} are the yield stresses obtained for the pristine and water contaminated greases, respectively.



(a) 3D view of the mold **(b)** Dimension of the mold

Fig. 4 Schematic representation of 3D view, contact angle measurement and dimension of the mold

2.2 Water Droplet Analyzer

To measure the contact angle of a water droplet on the grease surface, a drop shape analyzer shown in Fig. 3 is employed. In this setup, grease samples to be tested are placed on the adjustable sample stage and the height of the stage is set to the level of the lens using an adjustable screw. A water droplet is placed on the grease surface using a 10 µl syringe, and the focus and zoom of the droplet are adjusted using the adjustable lens (Thorlabs AC254-075-A-ML Lens). The image of the droplet is captured using the camera (IDS UI-5480CP-M-GL GigE camera) attached to it. The monochromatic blue light provides a clear black image of the droplet. From the captured image, the contact angle θ is calculated by the built-in software.

To achieve repetitive results, it is necessary to have an unvarying thickness of the grease and uniform surface during every test. For this purpose, a special mold made of polymer, consisting of several rhombus-shaped slots, is employed (see Fig. 4), and a systematic procedure is followed in all measurements.

The schematic representation of the mold in 3D view with grease in the slots and water droplet on the grease surface is shown in Fig. 4a. The dimensions of the rhombus slots are as follows: length, 15 mm, breadth, 10 mm, and thickness, 0.5 mm (see Fig. 4b). From the dimensions of the polymer mold, it can be inferred that this mold requires a small quantity of grease for testing. This allows the practitioner to perform several tests on a degraded grease sample extracted, for example, from a bearing in the field. Further, the multiple slots in the mold reduce the sample preparation as well as testing time, providing a constant grease height for all tests. The constant height of the samples allows easy identification of the base of the water droplet and for easy detection of the droplet edges for accurate measurement of contact angle.

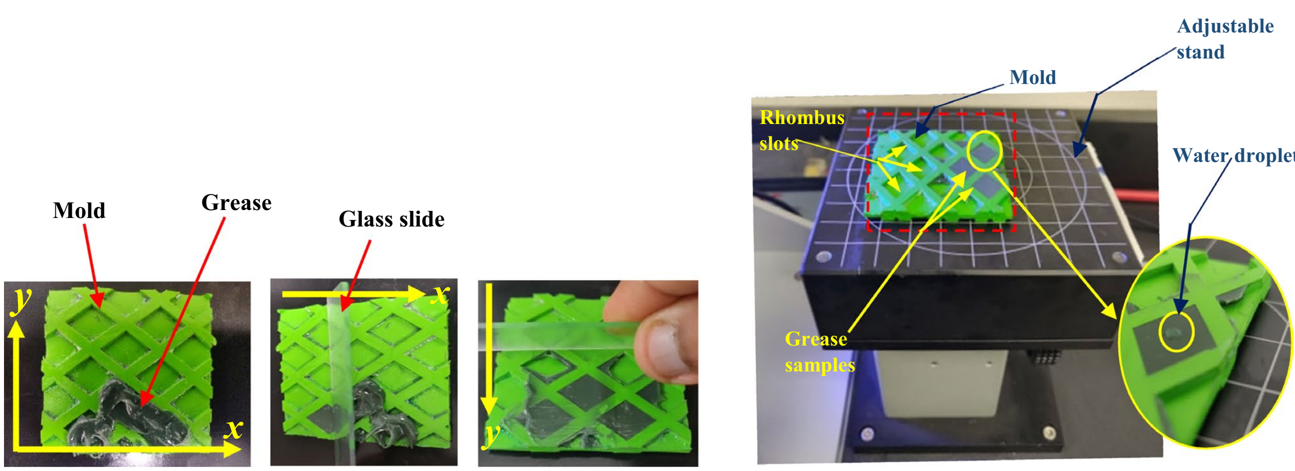
To achieve a grease surface of the same height of the mold surface as well as for attaining a uniform surface finish of the grease samples, three steps are followed (see Fig. 5a–c). In Step 1: the grease sample is collected from the grease cup using a spatula and applied on top of the slots of the mold. Step 2: using a glass slide, the excess grease is wiped off in the *x*-direction. Step 3: Step 2 is repeated in the *y*-direction. The slot containing the grease to be tested is kept in front of the camera and a 5 µl water drop is placed on the grease surface. The droplet is captured using the camera (see Fig. 5d) and analyzed using the software.

3 Results

The measured weight difference in percentage for all the greases due to the water contamination is provided in Fig. 6.

3.1 Rheometeric Measurements

The yield stress values for the pristine and water-contaminated LiC-m, LiC-P, CaS, PU, Si, AlC, and WLi greases are determined from the shear stress–strain values. The log-plots of shear stress and strain for LiC-m, CaS, PU, and WLi greases and shown in Fig. 7a–d, respectively. To avoid redundancy, the values for other types of greases are not plotted. Figure 7a and b reveal that the values of shear stress for pristine LiC-m and CaS greases are lower than water contaminated greases, while Fig. 7c and d, suggest an opposite trend for PU and WLi greases.



(a) Step 1 (b) Step 2 (c) Step 3 (d) Mold with grease and water droplet

Fig. 5 Procedure followed for applying the samples in the rhombus slots and the mold with water droplet on the grease surface

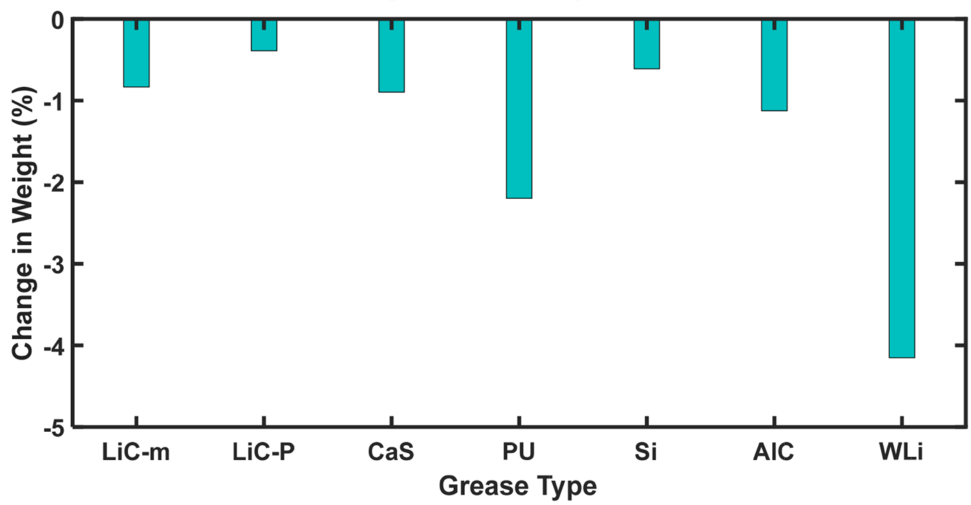


Fig. 6 Percentage change in the weight of the greases with the presence of water

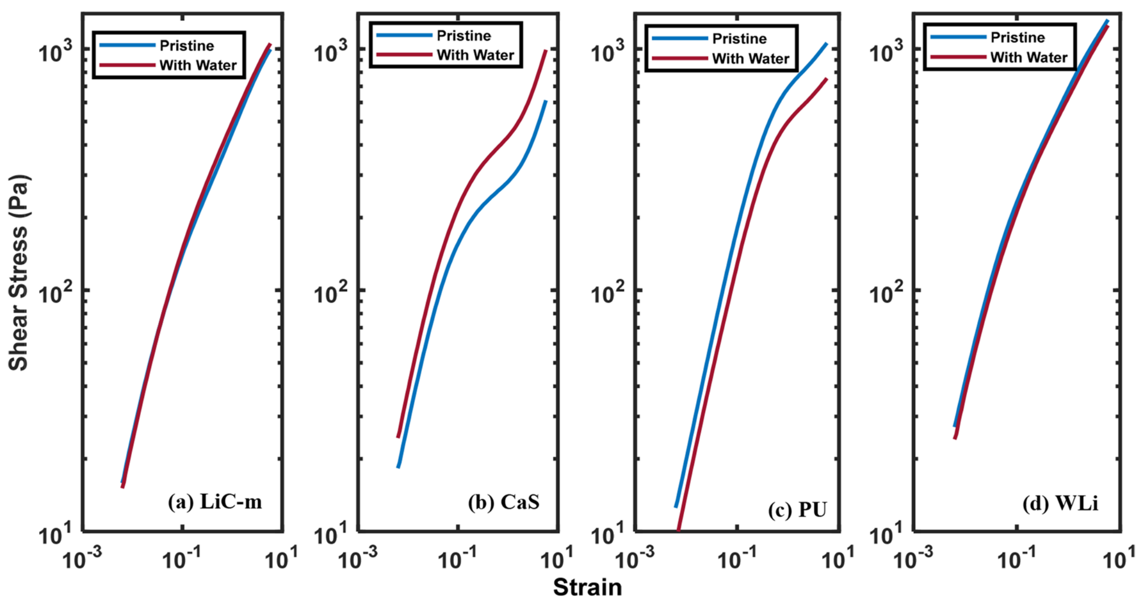


Fig. 7 Log–log plot of shear stress versus strain for LiC, CaS, PU, and WLi greases

From the plotted shear stress vs. strain values, and using the linear fit function in Matlab, the yield stress values are determined for $R^2 > 99.5\%$. Experimentally determined shear stress–strain values and their linear fits for both the pristine and water contaminated CaS grease are shown in Fig. 8a. The percentage change in the yield stress values determined using Eq. (2) for all the greases are provided in Fig. 8b. From this figure it can be observed that the variation in the yield stress values for all the greases follows the same trend. For LiC-m and CaS greases, the change in yield stress values is negative, indicating that these greases thickened with water contamination. For other types of greases, the yield stress values decreased with water contamination.

3.2 Water droplet analyzer

Grease is a semi-solid substance with a complex structure. When a water droplet is placed on a grease surface, it tends to spread and/or gets absorbed by the grease. Therefore, the size of the droplet and the contact angle values change with time. To address these issues, a video of the droplet is recorded at 3 frames/s and the images at different intervals are analyzed for the contact angle. The contact angle values for the droplet are determined using the length/breadth approach, an option provided in the instrument's software. In this approach, the length and breadth of a rectangle are manually adjusted to the edges and peak point of the droplet, respectively (see Fig. 9).

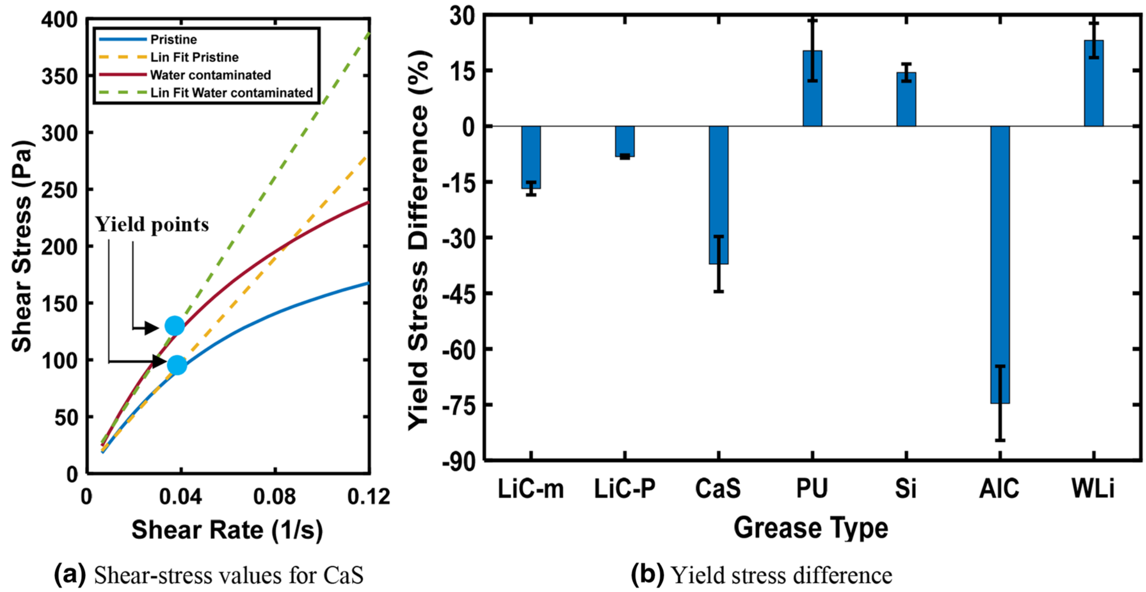


Fig. 8 Determination of yield point from experimental and linear fit data for CaS grease, determined shear stress difference for all types of greases

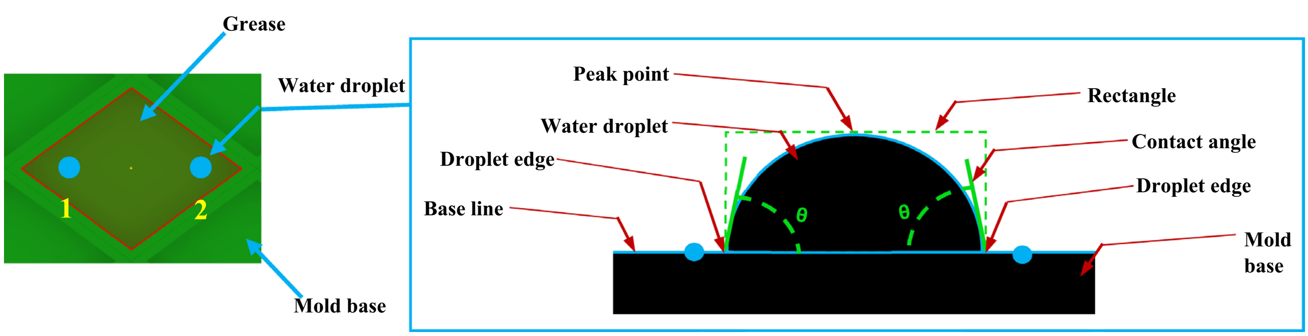


Fig. 9 Rhombus slots with water droplets and schematic representation of contact angle

Further, this establishes the baseline for calculating the contact angle. The schematic representation of the methodology followed to determine the contact angle is provided in Fig. 9. To gain more confidence on the obtained results, the contact angle values are determined for two droplets on the edges of the rhombus slots (see Fig. 5). For each type of grease, a minimum of nine sets of readings is taken.

Three sets of measurements from nine series of experiments on a pristine LiC-m grease are shown in Fig. 10. This figure also provides the captured water droplet images at 5, 30, 60, 150 and 300 s. Referring to this figure it can be observed that the contact angle varies with time, and that the values decrease non-linearly for the first 150 s, after which the rate of change becomes small and linear. It is interesting to note that the variation in the contact angle values among the samples are somewhat scattered during the first 45 s of the process, but become invariant and "stabilize" thereafter.

Similarly, the contact angle values and captured droplet images for PU, CaS, and WLi greases are shown in Fig. 11a–c, respectively. The progression of contact angles is similar in all cases, exhibiting a nonlinear transient behavior that stabilizes after 150 s.

Now, referring to the values of contact angle plotted in Figs. 10 and 11, the following general observations can be made. (i) contact angle values reduce with time, (ii) the variation in the contact angle values among different samples tend to be somewhat scattered until 60 s, but become invariant thereafter (iii) the values decreased non-linearly until 150 s and, subsequently, the values stabilize and drop linearly at a relatively low rate of change. Evaluation of contact after 60 s can be considered to provide adequate information for practical purposes. The average values of contact angle for all the nine samples of the greases at 60 s are determined and plotted in Fig. 12.

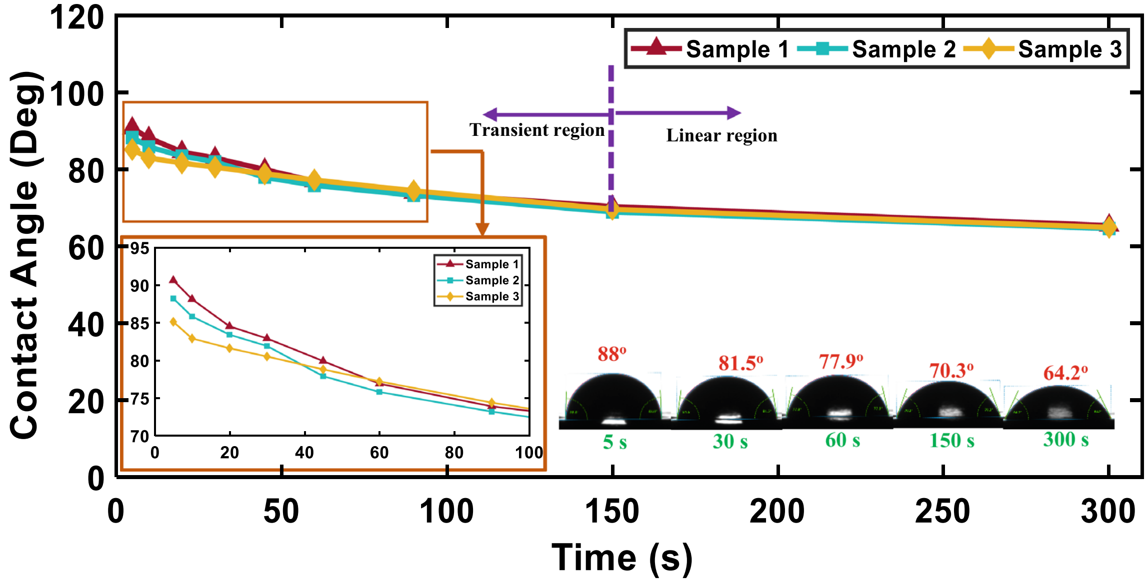


Fig. 10 Contact angle values and water droplet image of LiC-m grease

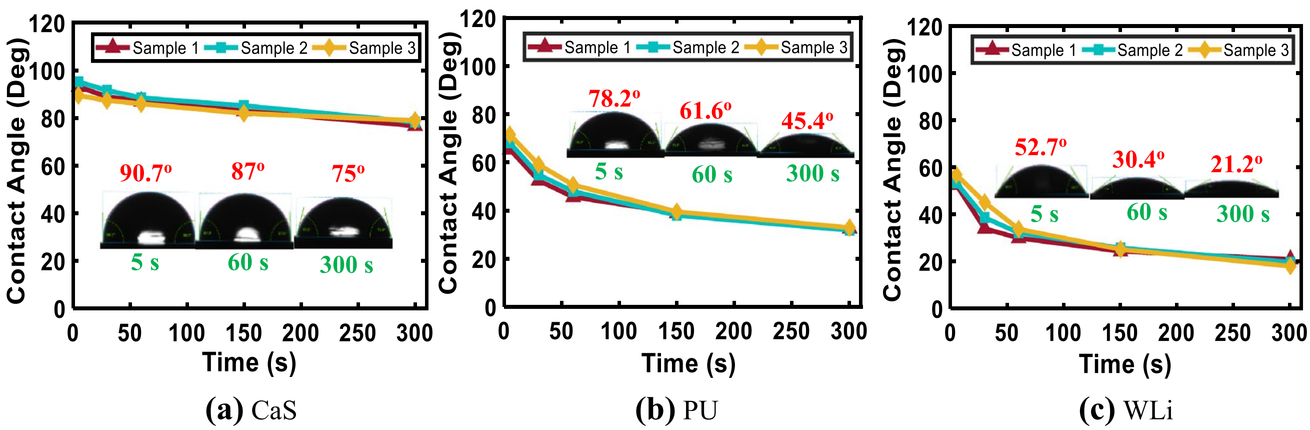
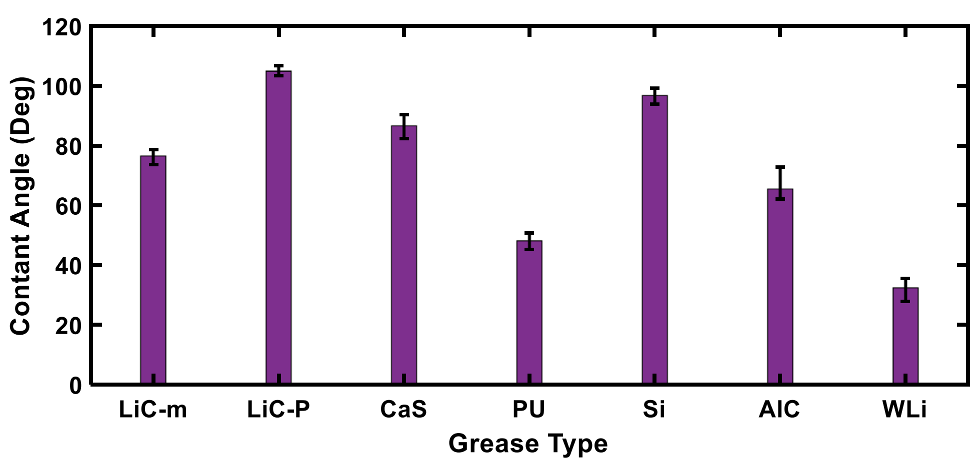


Fig. 11 Contact angle values and water droplet images for CaS, PU, and WLi greases

Fig. 12 Average contact angle (60 s)



4 Discussion

From Fig. 6 it can be observed that, LiC-P grease yielded lower while WLi grease yielded higher change in the weight percentage with water contamination. In other words, WLi grease has the highest capacity for absorbing water, while LiC-P grease exhibits the best water repelling properties. The order of percentage change in the weight with the contamination of water is:

LiC-P > Si > LiC-m > CaS > AlC > PU > WLi

However, measuring the change in grease with the presence of water can mislead the practitioner as it is very difficult to remove all of the free water from the grease samples. Further, some water may react with the grease and that it is possible that a portion of the grease components may be discarded in the process of draining. The change in the yield stress values with water contamination are provided in Fig. 7b. From these figures it can be observed that grease property is affected, both negatively and positively, with water contamination. Note that the change in the yield stress values for LiC-m, LiC-P, CaS and AlC greases are negative, while for PU, Si and WLi they are positive. These negative and positive values of the change in yield stress indicate that the grease tends to either thicken or dilute in the presence of water. Similar observations for LiC-m, CaS, and PU greases—i.e., increase in the consistency with the addition of water for LiC-m and CaS greases and decrease in the consistency for PU grease—have been reported by others [16]. The drastic positive and negative values in the rheological properties make the interpretation of grease resistance to water difficult for the practitioners. Specifically, among the PU, Si and WLi greases, it is clear that Si grease will exhibit better water-resistant characteristics, while the WLi grease will not have the same water-resistant characteristics in the presence of water. However, it is not clear whether the test performance of the Si grease is better than LiC-m, LiC-P, CaS, and AlC greases. Therefore, an alternative approach to assess the grease's performance in water contamination is necessary.

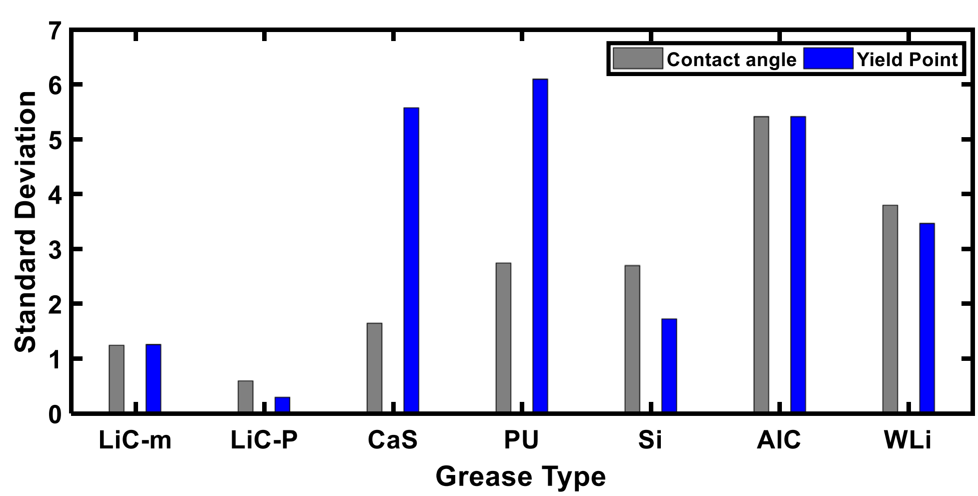
Table 2 Comparison of change in yield point difference percentage and contact angle

Grease types	Yield point difference (%)	Contact angle (°)
Si	14.42	96.8
PU	20.3	48.23
WLi	23.06	32.5
LiC-P	−8.2	104.95
LiC-m	−16.81	76.58
CaS	−37.13	86.7
AlC	−74.63	65.58

In the present research, an attempt is made to assess the nature of the grease surface-active components by measuring of contact angle of a water droplet. The primary advantage of the proposed approach is that only a minute quantity of grease is needed for testing. For the circular cross-section plate of 25 mm diameter and for 1 mm grease thickness, the volume of grease used in the rheometer is ~ 490 mm^3. On the other hand, the volume of grease used in the rhombus slot is determined to be 75 mm^3, roughly sixfold less sample vs. the rheometer.

The average values of the contact angle for all the considered samples of the greases (measured at 60 s) are plotted in Fig. 12. This figure suggests that the contact angle values for LiC-P and Si greases are greater than 90°, and for CaS grease it is approximately equal to 90°. This indicates that the water droplet remains on the grease surface as free water for a relatively long time. In the case of LiC-m grease the value of $\theta < 90°$, and for PU, AlC and WLi greases $\theta \ll 90°$, indicating the grease either spreads or gets absorbed faster into the grease. The contact angle results indicate the order of water repellant properties are:

Fig. 13 Standard deviation values determined for change in yield point difference and contact angle



LiC-P > Si > CaS > LiC-m > AlC > PU > WLi

Now, comparing the predicted order of performance from the percentage change in weight and contact angle values, it is concluded that except for CaS and LiC-m greases, the predicted order of performance follow the same trend. Simillarly, comparison of the values of yield point and contact angles are carried out using their values provided in Table 2. From this table it is observed that the first three greases illustrated positive change, while the last four values showed a negative change in the yield points. Further, both approaches predicted the same order of performance in the presence of water for having a positive change of yield point, i.e. Si > PU > WLi. However, in the case of a negative change of yield point, the order predicted for LiC-m and CaS greases are opposite. Leckner [10] and Nagarkoti [12] reported that calcium sulfonate provides better washout properties than the lithium complex followed by lithium-based greases. Therefore, it can be concluded that the order predicted by the contact angle approach is correct compared to the rheological and weight measuring approaches.

To establish the precision of the proposed approach, the standard deviation in the values of percentage difference of yield point and contact angle values are determined for different greases and presented in Fig. 13. From this figure it can be observed that the values of standard deviation determined using both approaches are almost the same expect for CaS and PU greases. In the case of CaS and PU greases, the values of the standard deviation determined for the rheological method are higher than the contact angle approach. Therefore, it can be concluded that the proposed contact angle approach provides equivalent or better precision than the rheological approach.

From the above discussion it can be concluded that the contact angle approach can quantitatively determine the water resistance properties of the grease.

5 Conclusions

An attempt is made to characterize the water repulsion properties of seven commercially available greases (LiC-m, LiC-P, CaS, PU, Si, AlC and WLi), by measuring their rheological property (yield stress), and comparing these data with a new method that measures the contact angle of a water droplet on the greases' surface. For determining the water-resistance properties using the rheological approach, shear stress–strain plot values are obtained for pristine and water contaminated greases. From the obtained results it was observed that for LiC-m, LiC-P, CaS and AlC greases, the consistency of the grease increased with the inclusion of water, while for the other three greases: Pu, Si, and WLi the consistency was reduced. Therefore, it was concluded that the yield point analysis approach is a relative approach and not capable of providing clear differentiation in water repulsion performance in a single test. In contrast, it was established that the contact angle approach, which requires six-fold less volume of grease compared to the rheological approach, will allow the practitioner to test multiple numbers of times. Further, the proposed approach is able to provide a comparative order of water-repelling properties of all the greases in a single platform. The values of contact angle θ observed for different greases are: LiC-P, and Si > 90°, CaS $\theta \sim 90°$, LiC-m < 90°, and PU, AlC and WLi $\theta \ll 90°$. For this limited study, the greases with $\theta \geq 90°$ can retain water on the grease surface as free water for a relatively long time while for $\theta \ll 90°$, the water either spreads or gets absorbed faster into the grease. The order of the grease having water repellant properties are: LiC-P > Si > CaS > LiC-m > AlC > PU > WLi. The order of water-repelling properties predicted using the contact angle approach is more accurate than the rheological approach and, additionally, the standard deviation values predicted using the proposed contact angle approach either provided equivalent or better precision than the rheological approach. Future studies that correlate the contact angle values for grease experiencing water contamination with mechanical shearing are highly desirable. Work is in progress toward this objective.

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