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Degradation Factors of Semi-Synthetic Lubricant Oil for Gasoline-CNG Engine

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ABSTRACT

Compressed natural gas (CNG) as alternative energy has gained attention in public transport. CNG is clean energy, high octane, and less expensive than other fuels and can be operated in a dual-fuel engine system. This research studied various factors that affect the degradation of semi-synthetic engine lubricants for the gasoline-CNG engine. Samples were collected by distance lubricant oil analysis was conducted based on various ASTM methods. The kinematic viscosity (KV) analysis is an important property of Lubricant oil. It was measured at the structures of 40 and 100 °C. At 100 °C, the KV decreased from the heating phase, causing the intramolecular bonds to break, making the oil film thinner and leading to a catalytic oxidation reaction. The Fourier transform infrared (FTIR) spectroscopy showed that the lubricants undergo chemical changes in their properties due to increasing the carbonyl functional (C=O) concentration by oxidation product. The result indicated increased acid content and KV. It affects the corrosion of the internal engine parts. The total base number analysis indicates the cleaning agent performance of the lubricant. It was found that at a distance of over 15,000 km, the concentration of alkaline additive reduces half the amount of additive. This demonstrated that additives have been degraded from contaminations, such as dirt, water, metal, and soot.

KEYWORDS: Semi-synthetic lubricant oil, compressed natural gas (CNG), gasoline, lubricant oil degradation, additive depletion, free radical, contamination

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1. INTRODUCTION

In 2024, the compressed natural gas (CNG) market demand rising by approximately 65% in southeast Asia. It can continue to grow a further 80%. CNG is alternative energy popular for publication transport and passenger car in southeast Asia. Because it has been recognized as clean energy and reduces carbon dioxide emission from combustion. Disadvantage CNG room temperature combustion is used at around 800 °C. (Zheng *et al.*, 2022) It generates lubricant degradation more comfortably than other fuels and it changes the structure of engine parts. In this work interested in studying semi-synthetic lubricant oil products. Because the manufacturers provide end-of-time for semi-synthetic around 10,000 kilometers. This expert is commonly incorrect, specifically in varying engine conditions and users. (Karluk *et al.*, 2022) This paper analyses the lifetime of it can use between 15000 to 20000 kilometers. It can be used over the manufacturer recommendations. However, it can lead to a negative effect. Such as sluggish engines, engine wear, and engine failure. As we observe in appearance used lubricant oil. The first was changed to a darker color and the film oil was thicker of these physical changes in lubricant oil. Therefore, they can be monitored end of time in lubricant oil. (Maguire, 2010) The chemical reaction produces lubricant change structure. Initially, the oil structure is a long-chain hydrocarbon. While using operate under high temperatures and a high shear rate of the engine. It leads to a bond hydrocarbon break and leads to an oxidation reaction. The oxidation reaction is easy to react with free radicals and given by products are peroxide and hydroperoxide. It can produce various products in lubricant oil such as carboxylic acid, hydroxy ketones, and ketoaldehydes. (Barman, 2002) By the time that oxygen is in the environment and attaches chemical structures such as carbonyl, phenol, and alcohol can be measured from FTIR. It causes increased viscosity and acid number. (Guan *et al.*, 2011) The acid number measured the amount of acid generated in lubricant oil. It is an important indicator of efficacy in used lubricant oil. The acid could be corrosive to internal engine parts and wears metal contamination. The result demonstrates a defective engine during operation, leading to a road accident and contamination. The contamination can separate into two types in the engine. The first is internal contamination from wearing metal and soot combustion. The second is external combustion produced from water, dirt, and soil in the environment. These kinds lead to reduce efficiency and changes in the characteristics of the lubricant oil. It can form coagulation, vanish stains, and separate between oil and water. Contamination has a direct impact on the reduced concentration of alkaline additives. Calcium sulfonate is commonly used as a cleaning agent. Because it reduces rust performance, stability oxidation, and low cost. It will adhere to the surface of the calcium sulfonate additive. After a while, the concentration gradually degraded additive, and replace with dirt on the surface additive. The result show additive and lubricant oil degradation.

This article studied and analyzed the degradation factors of semi-synthetic lubricant oil in operated gasoline-CNG engines, following several ASTMs. They point out the degradation of semi-synthetic lubricant oil in the gasoline-CNG engine. Because the room temperature is higher than other fuel combustion.

2. MATERIALS AND METHODS

2.1 Samples

The lubricant oil was taken from the Toyota commuter 2018 model. The special engine has a VR38DETT V6, a capacity of 2,982 CC, and 132 horsepower is the engine's maximum power. The fuel is used in the combustion of CNG-gasoline engines. The initial distance to begin is approximately 120,000 km. It can inindicate the engine's real collect five points: 0, 5000, 7500, 10000, and 15000 km. The collected samples are used at very different distances because it is studied in real operation in the engine parts. To allow flexibility for the driver brings to collect the sample. Because vans are used in public transport systems, continuing work operates between 12 and 15 h and prevents the depletion of lubricant oil by more than 10%. The three samples produced for the CNG engine are lubricants A study was SAE 10W-40 lubricant oil. The lubricant B was SAE 15W-40 grade and lubricant C was SAE 15W-40 grade. Group B and C are different manufacturers. All samples were added to the engine to analyze factors degradation. The new lubricant oil properties are shown in table 1. The method was collecting lubricant oil, The first step set up the instrument, the tube was cut long enough to enable us to reach halfway into the oil sump. The tube was going to assemble the hose into the vacuum pump. The aluminum ring is going to tighten until it is snug. The new bottle includes a vacuum pump to ensure that the instrument is properly sealed. In the second step, clean around the valve to protect against contamination before opening the valve. The tube was inserted so it went halfway to the oil sump, and it was filled up with approximately 100 ml. We take the bottle and remove it. Finally, the bottle's top was sealed with a solid cap.

Table 1. The result shows the properties of new semi-synthetic lubricant oils.

Properties	Lubricant A	Lubricant B	Lubricant C
Grade	10W-40	15W-40	15W-40
KV40°C (mm^2/s)	98.01	112.7	115.3
KV 100°C (mm^2/s)	14.80	15.12	15.31
Viscosity index	158	140	140
Pour point (°C)	-45	-33	-33
Flashpoint (°C)	240	250	254

2.2 Fourier transforms infrared spectroscopy

FTIR is a promising instrument for identifying lubricant oil quality. It extracts quantitative chemical compounds from complex matrices and is completely used for hazardous substances. It is an undestroyed method and no required prepared sample. By measuring the amount of oxidation and nitration that occurs in the lubricant oil over a given distance, the results can be used to monitor the lubricant oil degradation. The Fourier transform Spectrometer (FT-IR) technique, Perkin Elmer model oil express series one. The sample cells used zinc selenide (ZnSe) transmission cells. It was used at an analytical absorbance wavelength of 4000-550 cm^{-1} and path the length was fixed at 0.1 mm. The FTIR instrument was kept in a humidity control box under 45% of the analysis at room temperature, according to ASTM E2412. The oxidation analysis range was done in the range of 1800 to 1670 cm^{-1} and the nitration was in the range of 1650 to 1600 cm^{-1}. (ASTM International Standards, 2019)

2.3 Total base number analysis

Determining the lubricant oil base number is an important oil analysis because it provides information on the alkaline concentration in lubricant oil. It helped neutralize acid by-products that formed issues of the degradation generated under operating of the engine part. Consequently, TBN protects the engine from being damaged by acid in the lubricant oil. Thus, when adding a new lubricant to the engine part, TBN concentration is maximum value but when it uses long-distance the concentration decreases. In this work, the total base number of the samples was measured, using automatic titration (905 Titrando, Metrohm) with LiCl 3M in an EtOH glass electrode. All the measurements followed ASTM D4739. (ASTM International Standards, 2018)

2.4 Total acid number analysis

TAN represents one of the most important factors that lead to lubricant oil degradation products. TAN is commonly used to indicate the quality of lubricant oil. The result of overheating and contamination in the combustion process. The lubricant oil acidity value can be used as an indicator for the engine wear contamination in the engine part. The automated titration (905 Titrando, Metrohm) with LiCl 3M in EtOH glass electrode was used in this analysis, following ASTM D664. This analysis method can trace the acidic content of the lubricant oil, but it cannot determine the type of acid present in the lubricant oil. The result shows the value of potassium hydroxide (KOH, in milligrams) to the titrate of one gram of sample. (ASTM International Standards, 2018)

2.5 Kinematic viscosity analysis

The kinematic viscosity was studied in physical properties and measured, following ASTM D445 at the test temperature of about 40 °C and 100 °C, using the Cannon instrument, model CAV 2200. A thermometer for the standard silicone fluids within the analytical apparatus had a maximum temperature tolerance of ± 0.1 °C. The kinematic viscosity is an indicator of three factors of problems in lubricant oil. As temperature, intermolecular interaction, and the motion state of the liquid molecules. (Al Sheikh Omar *et al.*, 2021), (ASTM International Standards, 2000) KV analysis is required to analyze at 40 and 100 °C. The lubricant oil is commonly analyzed at 40 °C following ISO 3448 likewise engine lubricant oil is typically measured at 100 °C following SAE J300. Additionally, 100 °C reduces the rise of measure interference for engine lubricant oil soot contamination. In other words, KV help considers change at low and high temperature of lubricant oil.

2.6 Additive contents and wear metal analysis

Metal analysis has acquired lubricant industry interest and analysis based on atomic spectrometric techniques. It has been improved to measure metal contaminants and organometallic compounds in lubricants. Inductively coupled plasma optical emission spectroscopy (ICP-OES) is a high-performance technique for the analysis of almost all metals. It can be measured for routine muti-element. The experiment argon (Ar) purity of 99.9996% and oxygen ($O2$) purity of 99%. The peristaltic pump of the ICP-OES instrument was used as a solution to the nebulizer and the sample was prepared by weight standard mix with white spirit solvent. The samples were performed with a PFA-100 nebulizer. It has given a high percentage recovery and accumulated the result of the metal analysis. The ICP-OES technique (Avio-500 model, Perkin Elmer) is used to determine the concentrations

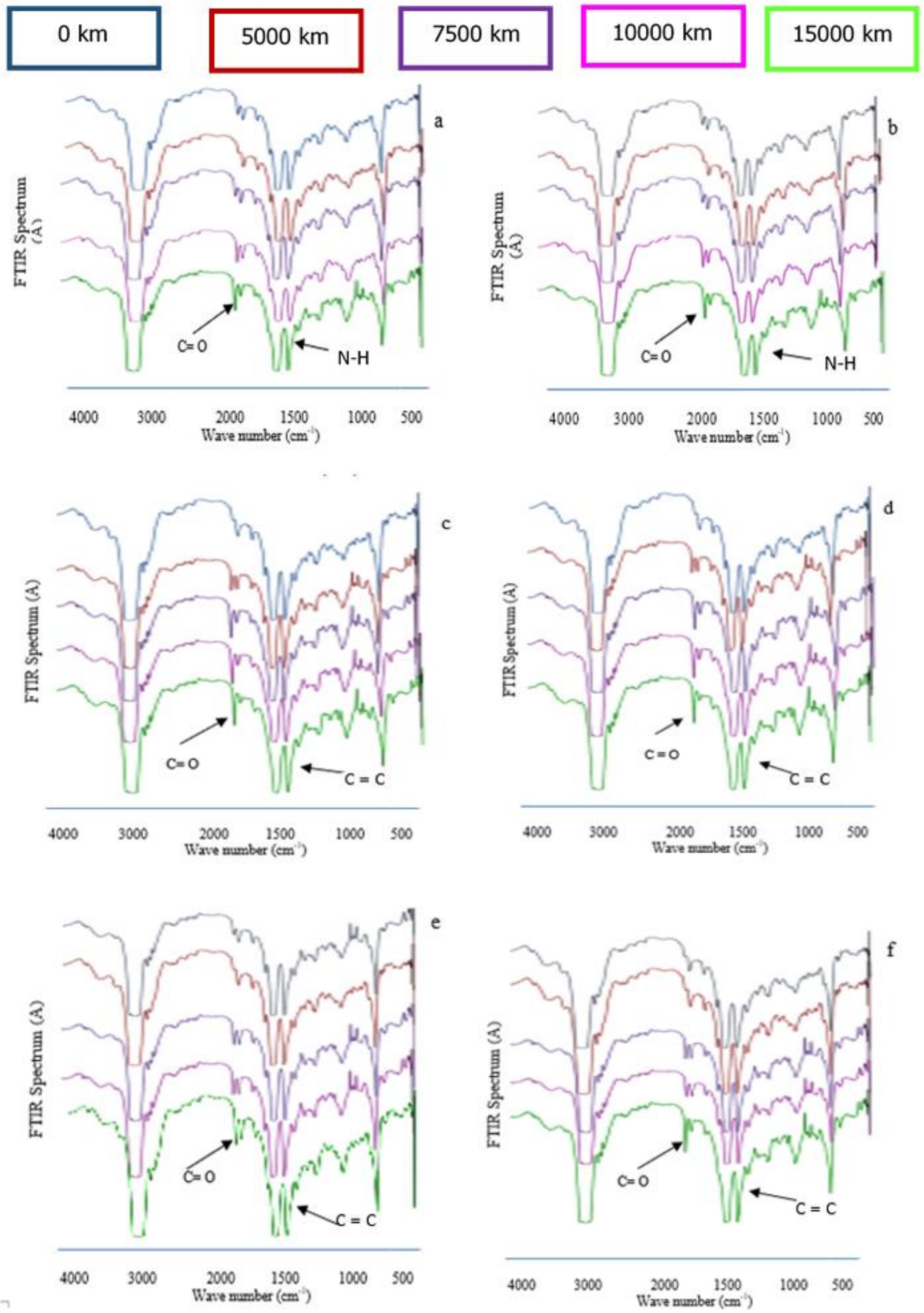


Figure 1 FTIR Spectrum of semi-synthetic lubricant oil with different kilometers travel a) Lubricant A van 1 b) Lubricant A van 2 c) Lubricant B van 1 d) Lubricant B van 2 e) Lubricant C van 1 f) Lubricant C van 2.

of iron (Fe) and aluminum (Al) in the used lubricant oils, following ASTM D5185. (ASTM International, 2019), (Lara *et al.*, 2015), (Goncalves *et al.*, 1998)

The analysis result of the metal-organic contents of additives can be used to determine the oil additives' lifetime and performance. This is due to additives that can help to improve the properties of the base lubricant oil. In this work, organic metals such as Zn, P, and Ca were investigated, as referred to in ASTM D4951. (R.Q. Aucélio *et al.*, 2007)

3. RESULTS AND DISCUSSION

3.1 FTIR analysis

FTIR has proved extremely successful in identifying chemical substances in the lubricant oil spectroscopy data and analyzing the effect of degradation in lubricant oil. However, when coupled with chemometric the average FTIR spectrum of used lubricant oil, the wavelength identified is 550-4000 cm^{-1}. Rang can be assigned to vibrational modes from hydrocarbons, carbonyl group, nitrite group, and water. Our interests vary in degrees of oxidation. As evidenced by the observation of the carbonyl group (C=O) absorption presented on the spectrum by an intense band around 1740 cm^{-1}. The two intended bands are presented around 1900-1750 $cm^{-1}$and 1250-1150 cm^{-1}. Oxidation is the main factor of degradation in lubricant oil. Another group of interesting function observation of the amine group (N-H) absorption was presented on the spectrum by an intense band around 1650-1580 cm^{-1} on the spectrum of their nitration. It is observed weak intensity at 0 km and high intensity at above 15000 km. It has caused the environment, such as rain acid, and erosion stone. (Macián *et al.*, 2021), (Sneha *et al.*, 2021) figure 1 shows the measured spectrum difference distance in used lubricant oil. At 0 km, the intensity of carbonyl and amine formed is as expected the radical initial chain reaction. Due to the carbonyl and amine low-intensity peak. The lubricant has been used for 5000-15000 km. It was found that the carbonyl and amine had increased as high-intensity peaks, corresponding to the operating distance. Because oxidation of the revolving oil is catalyzed by the metal and overheating temperature in the engine part. The oxidation leads to the destruction of new lubricant oil, additive depletion, and acid number. Molecules of base lubricant oil are nonpolar, but the effect of oxidation produces a polar in the compound of lubricant oil.

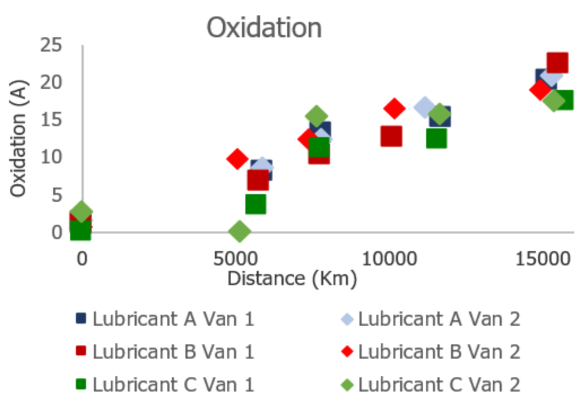


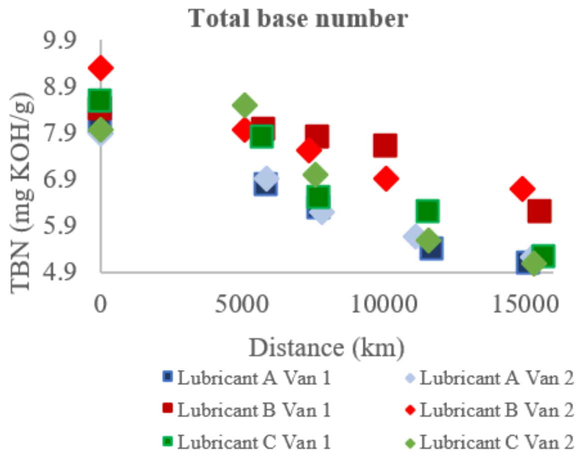
Figure 2 Evolution of amount oxidation analysis by FTIR

It leads to the production of ketone and alcohol in the compound. The accumulation of oxidation products could not be homogenized with the lubricant oil. It transforms the varnish adheres engine part.

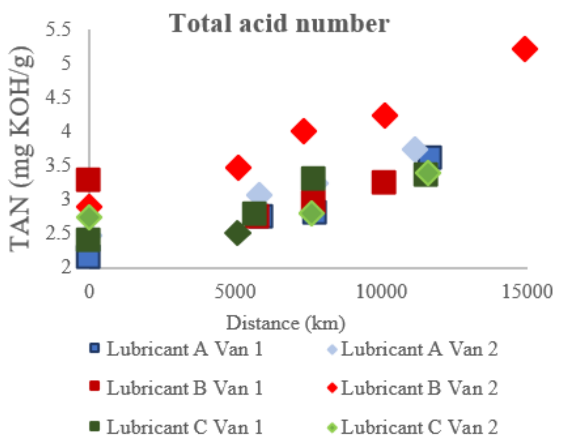
Figure 2 presents the amount of oxidation generated in the lubricant oil degradation. Lubricant A Van 1 found a concentration of oxidation increasing around 7.010 abs/ cm^{-1} at 5877 km and continuing to increase at 20.131 abs/ cm^{-1} at 15100 km. Similarly, concentration in van 2 increased around 6.931 abs/ cm^{-1} at 5864 km and continued to increase at 20.013 abs/ cm^{-1} at 15300 km. Lubricant B van1 found a volume of oxidation equal to 6.801 abs/ cm^{-1} at 5750 km and up the rise increased to 22.473 abs/ cm^{-1} at 15534 km. Likely, lubricant B brought quantity oxidation equal to 9.798 abs/ cm^{-1} at 5086 km, and up the rise increased to 18.912 abs/cm-1 at 14903 km. Lubricant C Van1 found a volume of oxidation equal to 3.561 abs/ cm^{-1} at 5698 km and up the rise increased to 17.598 abs/cm^{-1} at 15668 km. Equally, lubricant C van2 brings quantity oxidation equal to 0.875 abs/cm^{-1} at 5109 km, and up the rise increased to 17.591 abs/cm^{-1} at 15320 km. The result agrees with the FTIR spectrum.

3.2 Total base number analysis

The mechanism reacts with alkaline concentration. While the dirty build-up, alkaline additives are unable to work. The work of alkaline additive covers the surface of a metal engine part, and during operation dirty, adhere reduces the efficiency of alkaline additive. The result of TBN is presented in figure 3(a) presented the volume of TBN in the lubricant oil. Lubricant A



(a)



(b)

Figure 3 TBN (a) and TAN (b) values of relative difference distance travel by titration analysis.

Van 1 found the amount of TBN at 0 to 5877 km drop the rise estimated at 1.310 mg KOH/g and a distance of 15100 km occurred drop to 3.013 mg KOH/g. Similarly, lubricant A van2 occurs with a drop concentration estimate of 1.001 mgKOH/g at 5864 km and a lower concentration estimate of 2.752 mg KOH/g at 15300 km. Lubricant B found a TBN concentration drop rise of approximately 2.267 mg KOH/g at 15534 km in van 1 and 2.6 mg KoH/g in van 2 at 14903 km. Lubricant C TBN concentration decreased around 3.432 mg KOH/g at 15668 km and 2.915 mg KOH/g at 15320 km. The factors that affect TBN concentration drop the rise bring from contamination in the engine. The beginning contamination includes soot, condensed water, and acid generation. Another problem occurs poor seal of the joint. It can be contaminated in and generated new products in lubricant oil. (Macián *et al.*, 2021)

3.3 Total acid number analysis

The TAN results are presented in figure 3(b). Lubricant A found TAN concentration to rise approximately 1.510 mg KOH/g at 15100 km in van 1 and 1.473 mg KoH/g in van 2 at 15300 km. Lubricant B found a TAN concentration increase of approximately 1.533 mg KOH/g at 15534 km in van 1 and 2.321 mg KOH/g in van 2 at 14903 km. Lubricant C TBN concentration decreased around 1.353 mg KOH/g at 15668 and 1.151 mg KOH/g at 15320 km. Because the product from oxidation generated in molecule lubricant oil and detected the carboxylic acid functional group agrees with FT-IR analysis. (Macián *et al.*, 2021) The acid number can occur in the degradation of additives and catalytic oxidation. Oxidation products bring nitric acid occurs in gasoline and natural gas fuels. Sulfuric acid brings sulfur of antiwear and extreme pressure. All acids led to the corrosive and reduce performance engine part.

3.4 Kinematic viscosity analysis

The results of kinematic viscosity are presented in Figure 4(a). The kinematic viscosity at 40 °C can be slightly increased in all samples. The accepted degradation criteria are ± 15%. Lubricant A Van1, the KV increased at 5877 km up the rise at 15100 km. The end distance result show +1.152 % degradation. Similarly, lubricant A van 2 clearly increased at 5864 km and continued to increase at 15300 km. The result end distance demonstrates +2.743 % degradation. Lubricant B van 1 increased at 5750 km and continued to increase at 15534 km. The result end distance demonstrates +8.350 % degradation. Lubricant B Van2, the KV increased at 5086 km up the rise at 14903 km. The end distance result show +6.669 % degradation. Lubricant C van 1 clearly increased at 5698 km and continued to increase at 15668 km. The result end distance demonstrates +3.912 % degradation. Lubricant B Van2, the KV increased at 5109 km up the rise at 15320 km. The end distance result show +3.917 % degradation. The factor analysis affects the increase in viscosity properties. For instance, contamination and oxidation reaction are related to the size of molecules of lubricant oil. The initial oxidation

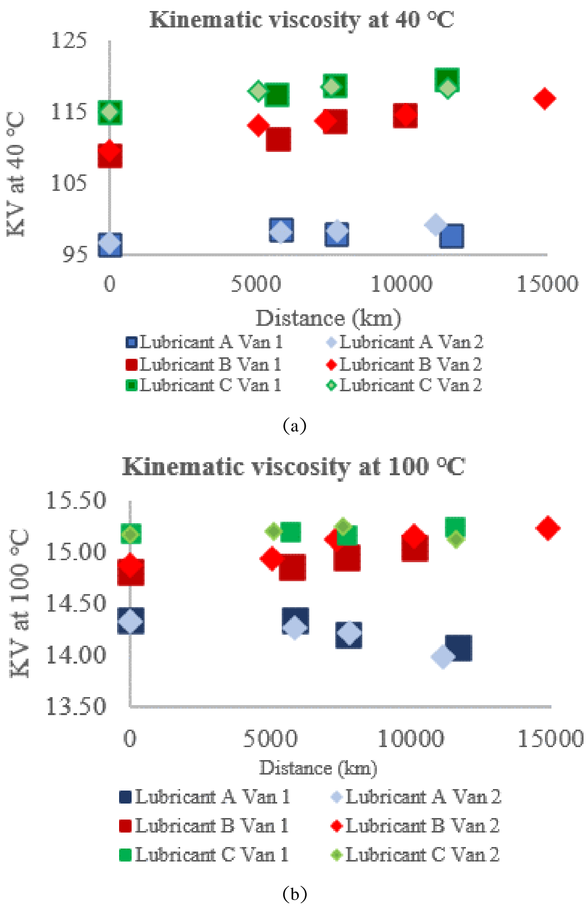


Figure 4 Variation of kinematic viscosity against three semi-synthetic lubricant oil and different distance: (a) 40°C, (b) 100 °C.

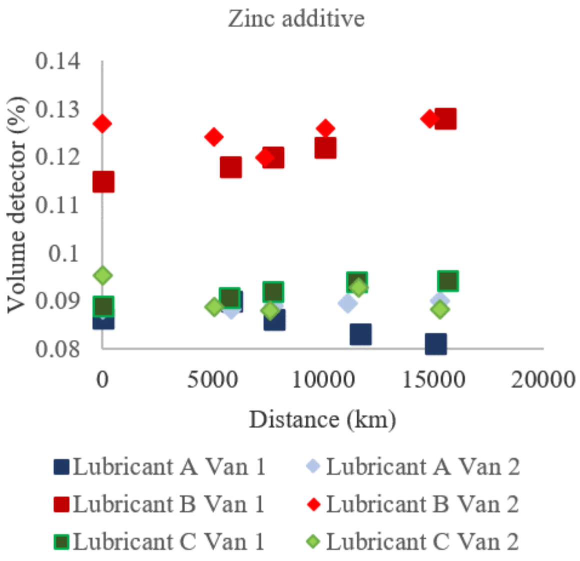
is the polymerization process that creates long-chain molecules of hydrocarbon. It increases the size of molecules and changes chemical properties in lubricant oil. This result agrees with the FT-IR analysis. Another related factor is contamination (soot, sludge, and dirty) in lubricant oil. It changes the appearance of lubricant oil such as contamination.

Figure 4b presents kinematic viscosity at 100 °C. The result shows that the A and C groups can be slightly decreased viscosity. The accepted degradation criteria are ± 15%. Lubricant A Van1, the KV begins decreased at 5877 km drop the rise at 15100 km. The end distance result show -2.442 % degradation. Similarly, lubricant A van 2 decreased at 5864 km and continued to reduce at 15300 km. The result end distance demonstrates -2.372% degradation. Lubricant B van 1 increased at 5750 km and continued to increase at 15534 km. The result end distance demonstrates +3.241% degradation. Lubricant B Van2, the KV increased at 5086 km up the rise at 14903 km. The end distance result shows +2.351 % degradation. Lubricant C van 1 clearly decreased at 5698 km and continued to decrease at 15668 km. The result end distance demonstrates -0.721 % degradation. Lubricant B Van2, the KV decreased at 5109 km dropping the rise at 15320 km. The end distance result show -1.192 % degradation. Because while lubricant oil work under high temperatures. It affects molecules of lubricant oil break and appearance lubricant oil film thinner. Other factors, contributes to the change property of lubricant oil such as fuel dilution and coolant leaks inside the engine. It could decrease the viscosity of the lubricant oil but the B group shows the resulting contract in another group. The effect relates to oxidation reaction. Because the spectrum of oxidation is higher than the other groups and agrees with the FT-IR analysis. (Heredia-Cancino *et al.*, 2018)

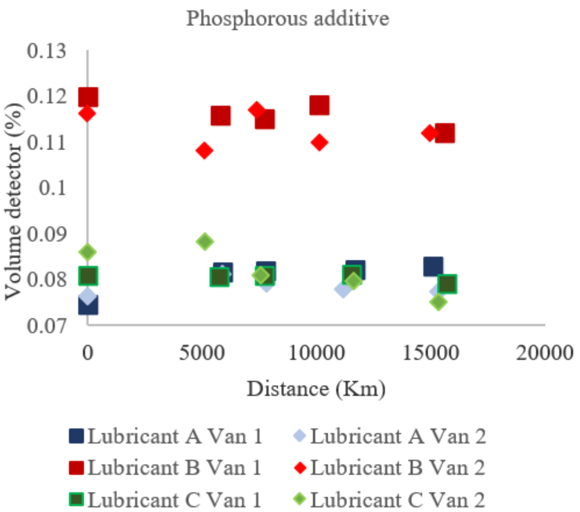
3.5 Additive contents and wear metal analysis

Five elements (Zn, P, Ca, Al, and Fe were measured by ICP-OES. The result is presented in figure (5). The concentrations of the elements in all sample groups were small quantity. Lubricant oil can protect the engine part. Figure 5(a) presented the concentration of zinc additive. It coated the steel elements to prevent rust and help anti-oxidation properties in lubricant oil. The result show concentration of zinc was increased concentration in the B and C groups. Because there were operated at long distances and Zn can be contaminated with lubricant oil. On the other hand, group A had more Zn content because of the end distance of A group less used operation in the engine. However, the concentration of Zn had little change and was acceptance critical.

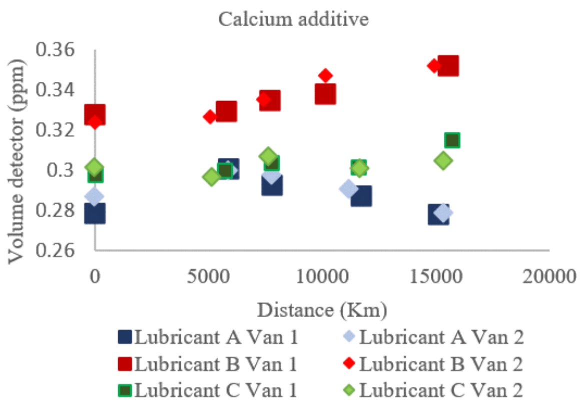
Phosphorous (P) additive works under extreme pressure. It helps to prevent damage to engine parts under operating high pressure such as gearbox starting and stopping of the engine. It was based on the P compound. The result is demonstrated in figure 5(b). B and C groups decreased concentration because long operations have been used which can lead to volatilization during engine operation. Group A has more P content because of the end distance of A group less used operation in the engine.



(a)



(b)



(c)

Figure 5 The Result of additive content by ICP-OES in used lubricant oil using a CNG-NGV engine. a) zinc, b) phosphorous, c) Calcium

Calcium (Ca) additive use detergent and dispersant properties in lubricant oil. It works clean deposits and metal contamination on surface engine parts. The result is shown in figure 5(c), the concentration increased quantitatively in all sample groups. It has multi-reason-produced high valves. The first cause is dust, It can abrasively dust particles from rock or silicon cement. The second cause hard water and the last cause are grease Ca contained cylinder help lubricant the engine.

Figure (6) presented the result of Wear metal being corrosive in the engine part iron (Fe) and aluminum (Al). The result of quantitative wear is corrosive in lubricant oil. Fe and Al were found in small quantities, and it did not affect the operation of the engine part. It demonstrates the lubricant's good performance. (Salem *et al.*, 2015), (Notay *et al.*, 2019), (Kumar *et al.*, 2005)

4. CONCLUSION

This work found the following 7 factors that produce lubricant oil degradation. The main factor is temperature related to KV. The KV increases at low temperatures and decreases the appearance of the film at high temperatures. Temperature led to the second and third-factor oxidation reaction and nitration. Its products such as varnish, sludge, and acid in the lubricant oil. It can be covering the engine part. Four factors occurred acid and corrosive engine parts that produce contamination in the system. Five-factor contamination can be coagulation as it accumulates at high volume. It affects to amount of TBN drop the rise and can be indicated point of lubricant oil degradation. This research expective users to realize the problems from lubricant degradation that led to a negative effect on engine.

All analytical methods in this work is important to indicate optimal performance for lubricant oil application. It was approved for lubricant oil quality inspection by ASTM and routine analysis performance lubricant oil. To conclude, it is suggested that the use of lubricant oil be carried out according to the manual book recommendation for protecting engine parts and reducing emissions to the environment.

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