



An investigation of factors influencing the response of screen printed thick film electrochemical sensors in the monitoring of lubrication oil ageing

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ABSTRACT

Based on condition based maintenance (CBM), oil can be replaced according to its measured status or in-situ with online monitoring systems. The acidity of oil, e.g. the total acid number (TAN), is an important parameter to evaluate lubricant ageing. Previous studies have shown the feasibility of thick film (TF) sensors based on ion selective electrodes (ISE) for the detection of engine oil acidity changes due to oxidation. Further study has shown the TF sensors generally have very long response times due to the extremely low conductivity of oils. This work focuses on the effect on the sensor response time and accuracy caused by introducing metal particles in oil, and the influence of water content in the oil. Two types of TF working electrodes (WE) have been tested and TAN was measured for each oil sample. The results show glass-based TF sensors can work under high temperature up to 120 °C and the metal particles in the oil can help to reduce the sensor response time significantly. It was also found that the presence of water in the oil has a minor influence on the voltage output of the TF sensor but can be helpful in the ionization and detection mechanism of H⁺.

1. Introduction

Modern oil lubricants for machinery containing high percentages of base oil degrade over the time of their usage producing acidic by-products such as carboxylic acid via a number of ageing processes of the base oil [1]. Hence total acid number (TAN) of oils has been used as an important indicator of oil degradation. So far, this is normally conducted by analyzing oil samples in laboratory under the guidance of industrial standard testing methods, for example, the titration methods ASTM D974 and ASTM D664. This offline oil condition monitoring method has been proven to be robust, however it has problems such as non-real time and the potential of contamination due to sampling etc. Online monitoring of lubricants has always been a desire for lubricant users. A number of attempts have been made to develop online sensors for oil chemical and physical properties, such as oil viscosity [1], density [2], dielectric constant [3], particle concentration distribution [4], water content in oil [5], and total ferrous debris [6]. Some of these have been applied to lubricant online monitoring of marine diesel engines on ships [7], wind turbines [8], turbine oil [9], and hydraulic machinery [10].

Thick film (TF) technology has been existent for approximately the last 60 years. TF sensors are compact and utilize low-cost mass

production for the application of this technology. TF pH sensors have been proven to be robust and accurate for aqueous solution acidity measurement [11] and soil condition monitoring [12,13]. Previous studies have also demonstrated the feasibility of using TF ruthenium oxide sensors in detecting acid content in artificially aged base and fully formulated engine oils [14,15]. It was found that TF sensors, including a ruthenium oxide working electrode and three types of reference electrodes (RE), were able to respond to the acidity change (oil ageing) under two temperatures (50 °C and 80 °C) and a linear correlation between the oil TAN and sensor outputs at 50 °C was found.

One of the major issues in the further development of TF sensors for oil acidity monitoring has been the relatively long sensor response time and influences of lubricant working conditions, e.g. water, wear debris etc. on the sensor responses under real service conditions. As the electro potential output of an electrochemical sensor reflects the H⁺ ion concentration in oil, in measurement using ion-selective ruthenium oxide electrodes the H⁺ concentration indicates an electrical property of the oil. Hence water content can change oil conductivity and/or dielectric constant of oil. This paper presents the results from a study on factors that influence the responses of TF sensors in detecting oil ageing including relative positions of the two electrodes, metal particle and water content at three different oil temperatures.

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2. Experiment setup

2.1. Oil samples

Fresh and oxidized oil samples were prepared at the Shell Houston laboratories using a proprietary in-house blown NO_x (BNO_x) and heating oxidization method. During the oxidization process, 350 ml samples of Star6 R13–3622 base oil, a group of base oils containing no additives (hydro treated base oil composed of hydrocarbons), were heated to 155 °C whilst an air/NO₂ mixture (NO₂ concentration of 3000 ppm) was blown into the oil at 200 cc/min [14]. Oil samples were taken at six oxidation time intervals: 2, 4, 8, 16, 20 and 24 h. Since the oil samples do not contain additives, the oil was extensively oxidized after 24 h, whereupon the oil viscosity had increased significantly, making its use for TF sensor testing beyond that time interval impractical.

The acidities (TAN) of the oil samples were measured at room temperature before the TF sensor experiments by using a Kittiwake test kit (FG-K1–110-KW) according to ASTM D974–12 standard (with color-indicator titration). The water content in the oil samples was measured by using Kittiwake EasySHIP DIGI Water in Oil test kit (FG-K1–102-KW), with a manufacturer's stated accuracy of 100 ppm. Table 1 shows the measurement details of TAN and water content in seven different oil samples.

2.2. Thick film acidity sensors

Thick-film screen printing technology consists of the printing and firing of ink paste onto a substrate. The main steps of the fabrication process are printing, drying (at about 150 °C depending on the paste used) and firing at the specific firing temperature of the ink paste used. The detail information has been described in [14]. Inks were printed layer by layer on 50.8 × 50.8 mm aluminum oxide substrate (Corstech) where each paste serves a specific purpose.

The working electrode used platinum gold (ESL 5837) as an underlying first layer for interconnection of conducting wires, while the reference electrode used silver (ESL 9912) as the underlying conductive layer (see Fig. 1). A second layer of dielectric insulator (ESL 4905) was deposited to leave an exposed window of the underlying conductor layer. The WE has a ruthenium oxide layer printed over the conductor layer through the exposed window. The RE does not employ any further layers, simply consisting of the bare silver layer exposed through the bottom square window. As mentioned before, polymer ruthenium oxide paste may be destroyed at high temperature [14]. So a glass based ruthenium oxide paste (Cermet Resistor System, 3980 Series, Electro-Science Laboratories, Inc.) was used to fabricate a new WE that can be used at higher temperature. For the ink with polymer binder the firing process (firing in a six zone belt furnace (BTUVQ41) with a maximum firing temperature 850 °C) was not employed for the final ruthenium oxide layer as the polymer material would be burned at such a high temperature.

Thick-film ion selective electrodes (ISE) and bare silver reference electrodes work together as a pH sensor producing an electro potential

Table 1

Details of the oil samples, their TAN and water content.

Sample No.	Oxidation (h)	Total Acid Number (mg KOH/g)	Water content (ppm)
1	0 (Fresh Oil)	0	0
2	2	1.3	0
3	4	5.2	1400
4	8	5.6	2100
5	16	9	3400
6	20	12.5	5100
7	24	13	100

voltage output in order to measure the acidity of oil. The electrodes of the TF sensor are shown in Fig. 1(a) while Fig. 1b–d shows the dimensions and layer structure of the finished electrodes. The reference electrodes should have a stable potential in the test solution while the ion selective electrode (i.e. the working electrode) responds to the change of the H⁺ concentration in oil. RuO₂ working electrodes have been shown to have the highest relative sensitivity to pH variation compared to other metal oxide materials tested in pH buffer solutions [11,16]. The bare silver conductor based electrode was utilised as a reference electrode because of its previously observed better performance than some other reference electrodes[15].

The following two different types of TF working electrodes were fabricated:

- Platinum gold conductor with a layer of polymer-based ruthenium oxide (RuO₂).
- Platinum gold conductor with a layer of glass-based ruthenium oxide (RuO₂).

The two types of working electrodes were tested in order to compare their performance in oil. Polymer-based electrodes has been tested in a previous study at 50 °C and 80 °C [14]. However, the polymer-based electrodes can only be tested up to a maximum temperature of approx. 100 °C but glass-based electrodes can be tested at much higher temperatures. Most of the time, engine lubricants work in very high temperature environments in combustion cylinders, and the oil will be heated to close to 400 °C–600 °C [17]. Even in an oil sump, the oil temperature will be higher than 150 °C. Consequently the glass-based working electrode should be better suited for online monitoring of engine lubricant if its performance is similar to or better than the polymer-based working electrode.

2.3. Experimental set up

Lubricant oil generally contains wear debris particles and water, and it also usually works at high temperature, especially in combustion engines. All of above define the working condition of the lubricant. Hence the influence of metal particle concentration, water content and temperature on TF sensors should be considered in the design of the experimental conditions. To investigate the influence of electrode relative positions, water and metal particles in oil on the TF sensor responses, the oil samples have been tested under a variety of conditions. Firstly, differing amounts of 316 L stainless steel powder were added to oil samples to simulate different particle concentration environments. The particle diameter distribution in the stainless steel powder was between 20 and 40 μm, which are within the size range of used oil from an engine with mild wear [18]. Additionally a slight amount of water was detected in oxidized oil samples, with the water content increasing with oil ageing (oxidization hours). The water contents of the oil sample are shown in Table 1. Furthermore, the oxidized oil samples were heated to 50 °C, 80 °C and 120 °C to simulate the possible working temperatures in engine oil sumps.

The oil acidity measurement experimental set up is shown in Fig. 2. The oil sample was put in a beaker and heated by a heater located beneath the beaker. A magnetic stirrer was used to keep the oil at a uniform temperature. The voltage output of the TF sensor was connected to a high input impedance voltmeter in order to record the voltage output during the experiment. The output voltage was typically in the millivolt range.

3. Results and discussion

3.1. Calibration of thick-film sensors

After a one and half hour hydration process in pH buffer solution, a stable output voltage was obtained from the TF sensors [15]. The WE and RE were placed in a fixed support structure, as they would be if manufactured as a sensor. In order to account for any positional

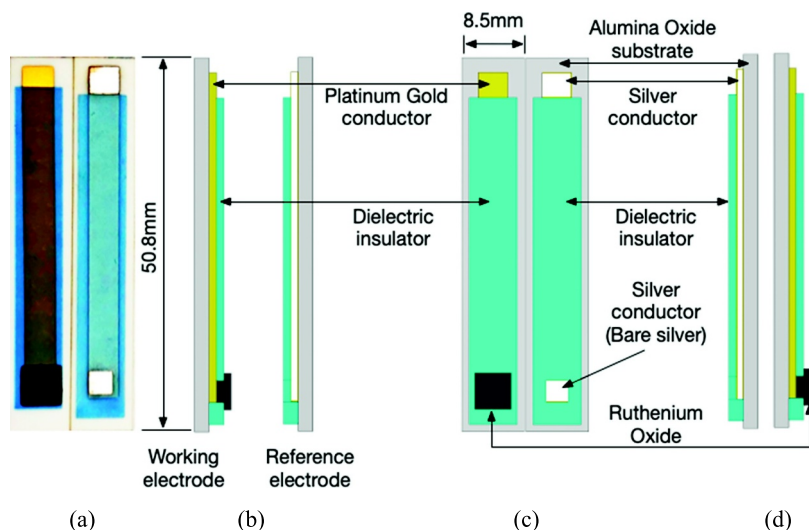


Fig. 1. Thick-film sensor dimension and placements of working and reference electrodes. (a) WE and RE of TF sensor, (b) face to face, (c) side by side, (d) back to back.

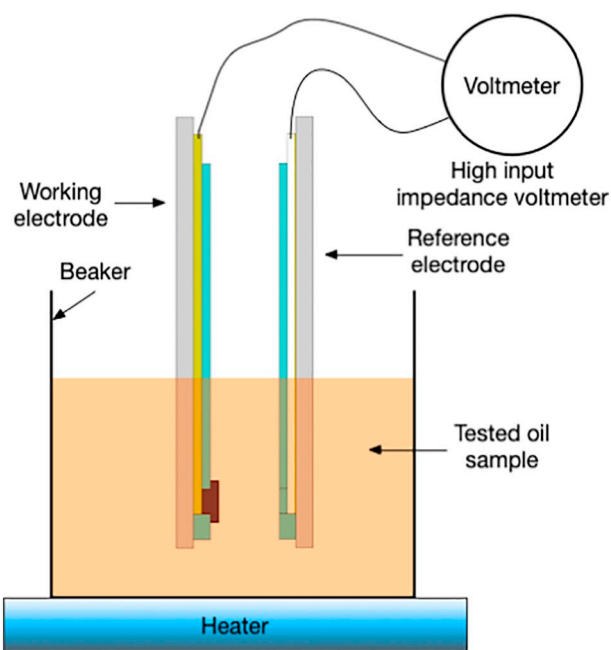


Fig. 2. Oil acidity measurement setup using a thick film sensor.

differences of the WE and RE affecting the measurement, the potential outputs were measured during the calibration process while the two electrodes were placed in three different configurations, as shown in Fig. 1b–d. The voltage outputs show that the arrangement of the electrodes placement has little influence on the voltage output. For both polymer-based WE and glass-based WE, the face to face structure gave the highest slope of potential versus pH, so this was chosen as the preferred experimental structure. Fig. 3 and Fig. 4 show the voltage output of the TF sensor versus pH value of buffer solution when using polymer based and glass based WEs. The slopes of the glass based WE are larger than the ones of the polymer based WE. This means that the glass based working electrode has a higher sensitivity than the polymer based one. The calibration results show that the TF sensors outputs have a very good linear response to pH value of buffer solutions. Therefore, they were considered to be good candidates for use as electrochemical sensors for testing the oil acidities in later experiments.

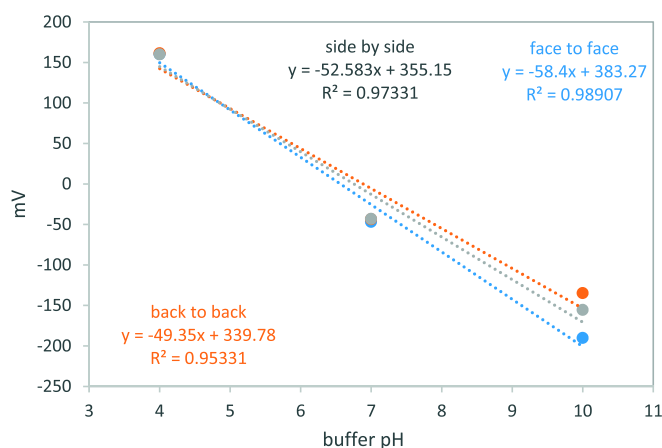


Fig. 3. Voltage output using polymer based electrode.

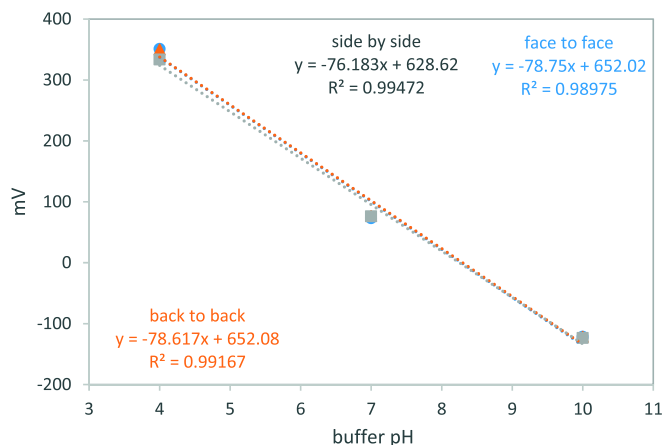


Fig. 4. Voltage output using glass based electrode.

3.2. Metal particle concentration influence on sensor response time

While using the TF sensor to measure the acidity of pure oil, the output voltage demonstrates fluctuation before gradually settling to a stable value. It takes approx. 30 min to 1 h to obtain a stable voltage output from the electrodes. This is a long response time for online oil

condition monitoring. In practical working conditions with used lubricant, there will almost certainly be metal particles mixed in with the lubricant. The metal particles mainly come from the wear debris of friction pairs and the working environment of the machine. Consequently the influence of the metal particles was considered in the TF sensor experiment. 316 L stainless steel powder with particle diameter distribution between 20 and 40 μm was added into the oil samples to see if this could decrease the sensor responding time. Stainless steel powder was selected because it does not react with acid in oil, so the consumption of acid by the added metal materials should be negligible. Four different amounts (3 mg, 5.2 mg, 7 mg, 10 mg) of 316 L powder were measured using an electronic balance equipment for adding into the 25 ml oil samples in order to test the relationship between metal particle concentration and reaction time of the TF sensor.

To account for the maximum possible metal particle amount in lubricant of approx. 350 ppm (about 7.17 mg) [19], the max amount of 316 L powder added in 25 ml oil sample was limited to no more than 10 mg (representing almost 488 ppm). The reaction speed of the TF sensor was measured under different metal particle concentrations in oil. The results show that with increasing metal particle concentration the reaction time is reduced, as shown in Fig. 5. This means the sensor will give a stable output in a shorter time. In 10 mg metal particle concentration, the stabilisation time was approx. 3 min. Therefore, the metal particle or wear debris in the lubricant was found to be helpful in improving the measurement speed of the TF sensor, hence improving real-time response.

Additionally, the voltage outputs under different amounts of oil metal particle content in a stable acidity level oil were measured, as shown in Fig. 6. There was only approx. 20 mV differential fluctuation in voltage output and the characteristic is almost linear. Hence the influence of metal particle concentration variation on the mV output of the TF sensor can be considered as a stable function of particle concentration. Thus it can be eliminated by data processing, and the system error can be overcome.

Furthermore, the conductivity of each oil sample with different amounts of 316 L particles was measured respectively using a liquid conductivity meter (Model L40, Wolfson Electronics). The relationship between conductivity and TF sensor stabilisation time is plotted in Fig. 7. Because the metal particle concentration is the only parameter changed in the oil, the result shows that the conductivity increase in oil is beneficial towards improving the reaction speed of the TF sensor. It was evident from the experiments that the reaction speed is influenced by metal particle concentration.

3.3. Influence of temperature on TF sensor oil acidity measurement

Fig. 8 illustrates the responses of the polymer based WE versus bare silver RE in seven oil samples at room temperature, 50 °C and 80 °C. The

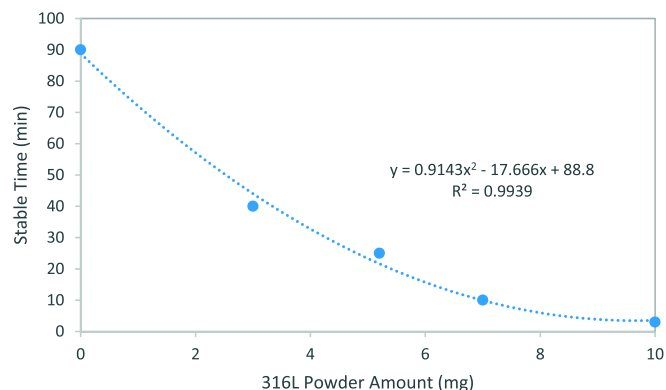


Fig. 5. TF sensor output stabilisation time versus amount of 316 L powder in oil at room temperature.

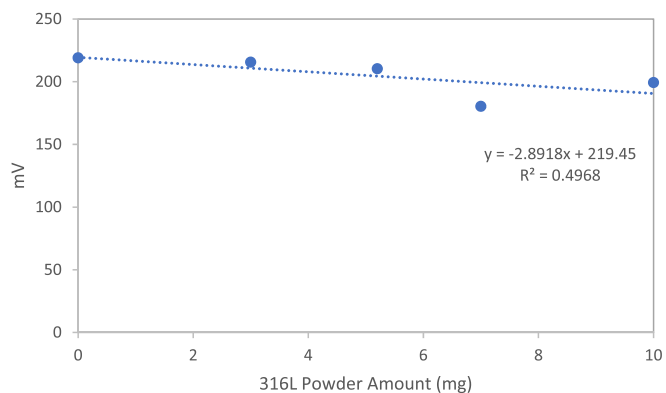


Fig. 6. TF sensor voltage versus amount of 316 L powder in oil at room temperature.

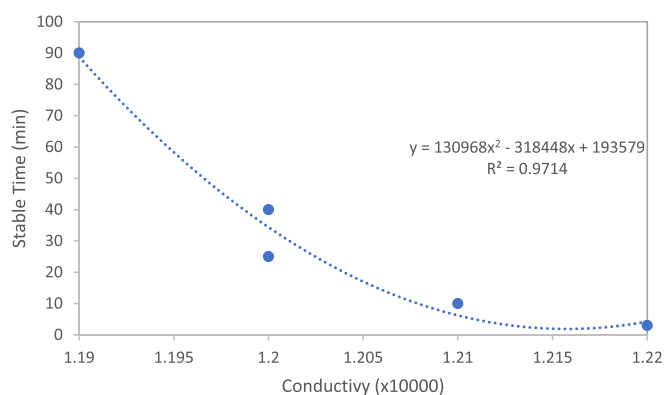


Fig. 7. TF output stabilisation time versus conductivity of oil at room temperature.

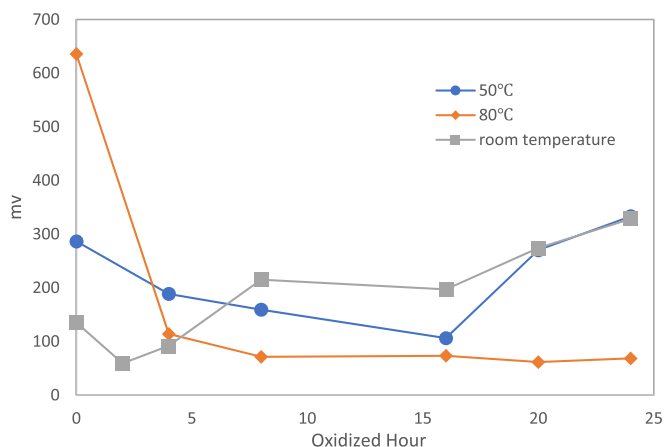


Fig. 8. TF output versus oxidized hours (Polymer based WE) at room temperature (≈ 28 °C), 50 °C, 80 °C.

polymer based electrode cannot be used at higher temperature because the physical properties of the ruthenium oxide and polymer composite layer will change in that environment. Most of the time engine oil will operate at a temperature less than 400 °C with the oil temperature in the oil tank (sump) being typically slightly more than 100 °C, therefore, the glass based electrode was tested at temperatures up to 120 °C in addition to 50 °C and 80 °C, as shown in Fig. 9. High temperature testing is helpful to understand the performance of the TF sensors when used in a practical engine working environment.

The total acid number (TAN) of all oxidized oil samples was tested by using Kittiwake EasyShip TAN test kit. The TAN of oil samples and

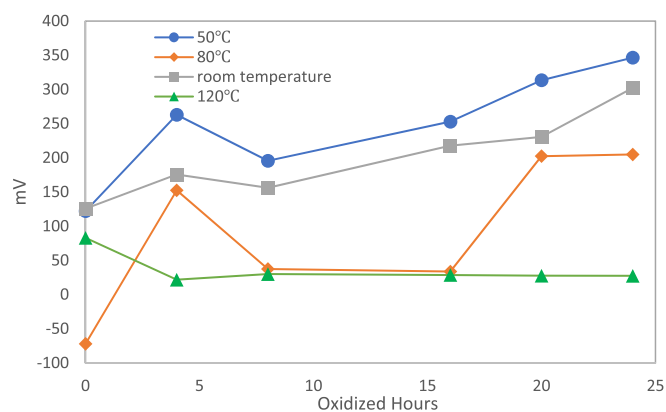


Fig. 9. TF output versus oxidized hours (Glass based WE) at room temperature ($\approx 28^\circ\text{C}$), 50°C , 80°C , 120°C .

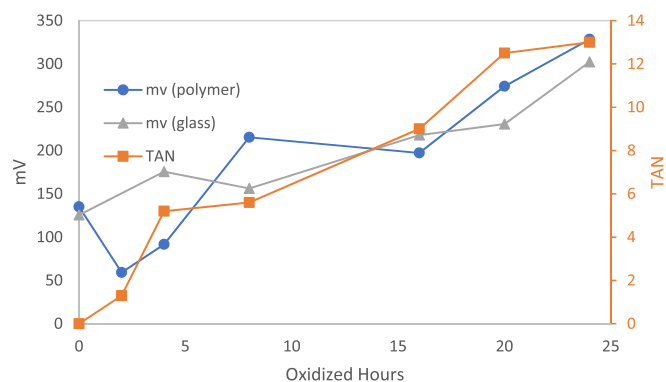


Fig. 10. TF outputs of both WEs and oil TAN versus Oxidized hours at room temperature.

voltage outputs of the two TF sensors are plotted in Fig. 10. TAN and mV outputs have the same trend with increasing oxidized hours. Through respective comparison of TAN, mV output of polymer based WE and mV output of glass based WE in Fig. 11(a) and (b), the mV outputs of both types of WE were found to be approx. Linear with TAN. The glass based WE has a relatively higher correlation than polymer based WE according to the R^2 value.

3.4. Influence of water content on TF sensor output

Fig. 12 illustrates the variation of water content in oil samples and mV outputs of TF sensor with oxidized hours. For the water content measurement, 5 ml of oil was measured with a Kitiwake water content

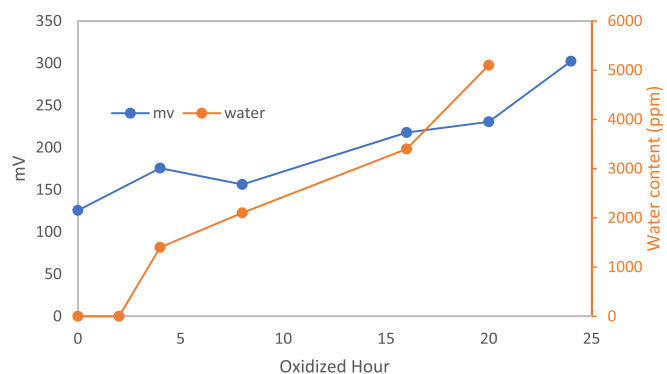


Fig. 12. Water content and voltage output of glass-based TF sensor increasing with oxidized hours at room temperature.

test kit. The water content of the oil sample after 24 h of oxidization of the oil sample was about 100 ppm and the oil was very sticky when it was tested. It is thought that given the increase in oil viscosity, water may have separated from it so that the water no longer stays in the oil. With the exception of that one isolated sample, the water content in oxidized oil samples tends to follow a linear relationship with oxidized hours, as shown in Fig. 13.

The TAN of the oil samples at room temperature increased with oxidation time as shown in Fig. 10. The oxidation process produces significant amounts of organic acid, such as carboxylic acid. Base oil is easily oxidized without the anti-oxidant additives that are designed to prevent the acidity increase and this oxidation process can be observed through the time series of TAN in oil samples.

However after the oil samples were tested at 120°C , the water contents of all oil samples was again tested and the results show that there was no measurable water in any of the oil samples (instrument reading of 0 ppm). Through comparison of the grey line (room temperature) and the green line (120°C) in Fig. 9 and the two plots in Fig. 12, it was observed that the voltage output of the TF sensor increased with increasing water content in oil. The voltage output of the TF sensor versus water content in oil is shown in Fig. 13 and shows a highly linear correlation. In contrast, the plot of 120°C in Fig. 9 shows that the voltage output only fluctuated over a small range and is almost flat, coincident with no water existing in the oil at this temperature. The water will have evaporated while heating the oil sample up to 120°C , although the carboxylic acids have a slightly higher boiling point, for example acetic acid has a boiling point of approx. 118°C . Consequently with the water content decreasing and very little change in the acid content in oil, there was little variation in sensor outputs in the differently aged oil samples. This can help explain the influence of water content on the voltage output of the TF sensor, since the acid in the oil has to dissolve in

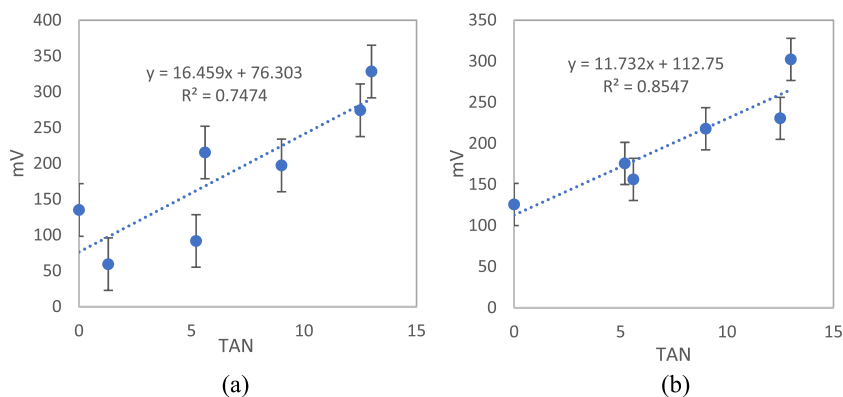


Fig. 11. (a) Polymer based TF output versus TAN of oil (b) Glass based TF output versus TAN of oil.

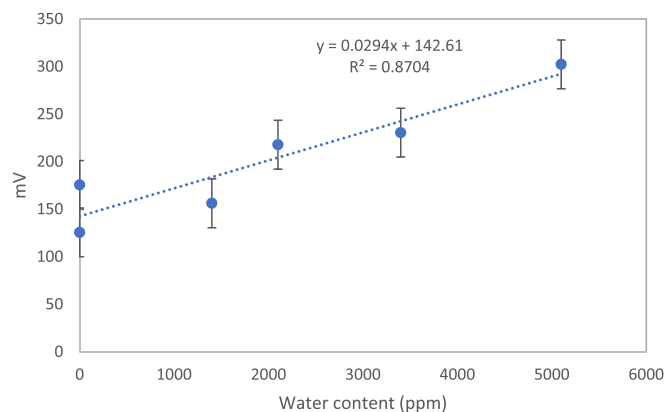
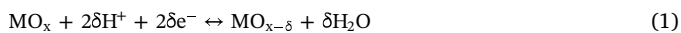


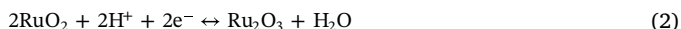
Fig. 13. Relationship between water content in oil sample and voltage output of TF sensor at room temperature.

water to produce H^+ . The TF sensor produces a voltage output due to the H^+ ion selectivity of the WE and reference potential output voltages combined together. Since it is the H^+ concentration in oil that is the reason for the voltage output of the TF sensor, a certain amount of water in oil might be necessary to allow acidity measurement with the TF sensor.

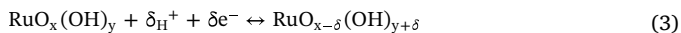
The response of ruthenium oxide to pH as proposed by Fog & Buck [16] involves a potentiometric pH response caused by either an equilibrium between two solid phases of the oxide or the intercalation of ionic species into the oxide structure. The former mechanism can be represented as a typical equilibrium for metal oxides as follows:



where MO_x is a higher metal oxide and $MO_{x-\delta}$ is a lower metal oxide. In the case of ruthenium oxide this can be written as:



Hence the electrode potential will be determined by the H^+ proton activity in the liquid, typically aqueous, phase. This model tends to lend weight to the above argument that the increasing water level in the aged oils affects the proton activity resulting from the concomitant increase in carboxylic acid levels and their dissolution in water. The above model does not however adequately explain the response of the TF ruthenium oxide WE in very low water content oils. In addition to solid phase and intercalation equilibria, Fog and Buck also suggest that metal oxide pH response could be due to ion exchange in a surface layer containing OH^- groups. The presence of OH^- groups on the oxide surface is thought to result in surface groups behaving as weak acids or bases that can exchange protons and hence where the electrode potential is subsequently determined by the local H^+ proton activity:



Consequently this proposed surface charge mechanism would appear to represent a better model for the WE response in low water content oils.

4. Conclusions

The oxidized base oil samples were produced under controlled environments and working conditions with different numbers of oxidized hours. They were used to evaluate the performance of two different acidity sensors that consist of thick-film electrodes. According to the experimental result and analysis, the main conclusions are:

- (1) The buffer solution test and calibration of the acidity sensors demonstrated that the relative position of the two electrodes has only a slight influence on the output voltage of the sensor. Also the outputs of both sensors exhibit a linear response to different values of pH buffer solution, with the glass based WE sensor displaying a higher sensitivity to pH variation.
- (2) The stabilisation time for testing of oil samples while adding 316 L powder showed that the reaction speed of the acidity sensor can be influenced by metal particle content of the oil and the concentration of metal particle gives a positive correlation with the reaction speed of the sensor. Consequently the sensor will have a faster reaction speed in practical monitoring of used engine oil in which a high level of metal wear debris exists.
- (3) The comparison of voltage outputs from polymer-based and glass-based WE sensors demonstrated that the glass-based electrodes can reflect the variation of acidity better than polymer-based electrodes and can work at higher temperatures.
- (4) Water content measurement supported the previously assumed mechanism of acidity measurement using TF sensors. The existence of water has a positive influence on measured acidity of oil samples when using TF sensors. Consequently the presence of water in the oil is helpful towards H^+ dissolution in oil and is a worthwhile subject for further research.

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