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In-situ lubricating oil condition sensoring method based on two-channel and differential dielectric spectroscopy combined with supervised hierarchical clustering analysis

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ABSTRACT

In this article, the two-channel and differential dielectric spectroscopy (TD-DES) has been proposed and employed to obtain the dielectric submicroscopic phase (DSP) and heterogeneous characteristics of the lubricating oil system compared with the Fourier transform infrared spectroscopy (FT-IR) data. The hierarchical clustering analysis (HCA) results based on TD-DES data were well consistent with those based on the FT-IR data. The two phases including the DSP and heterogeneous characteristics of the lubricating oil system featured by TD-DES were the special with regard to the function-group characteristics expressed by FT-IR data, which proved that the TD-DES method was more effective and sensitive than FT-IR to monitor the lubricant systems condition during the early oxidation degradation. A new and qualitative oil condition monitoring technique based on TD-DES and supervised hierarchical clustering analysis (S-HCA) has been proposed, by means of which the in-service lubricating oil samples can be classified well-separately into six clusters: (1) the fresh stage (cluster F), (2) the initial oxidation stage (cluster A), (3) the moderate oxidation stage (cluster B), (4) the accelerated oxidation stage (cluster C), (5) the strong oxidation stage (cluster D), (6) the heterogeneous system stage (cluster E). The signal for an oil change need should be sent in the stage (5) and the lubricating oils in stage (6) could damage the engine. By the leave-one-out cross-validation (LOO CV), the 33 in-service lubricant samples were all clustered correctly, which proved that the TD-DES analysis method combined with S-HCA can provide significant oil change advice.

1. Introduction

Lubricating oil plays a key role in industrial and automotive equipment with the multiple enhancing functions such as sealing, lubrication, anti-friction, cooling, cleaning and anti-corrosion. It is imperative that the lubricating oil should be monitored on a regular basis, to identify oil condition arising from the engine and to evaluate the equipment working status. In fact, the lubricating oil monitoring and analysis include two types: one is the detection and analysis of wear metal debris, the other is the analysis of the oil itself condition, whose goal is to reduce the likelihood for costly repairs due to overdue maintenance and equally eliminate unnecessary maintenance. The analysis of the metal debris resulted from the moving parts are usually conducted by the atomic emission spectrometry (AES), atomic absorption spectrometry (AAS), ferrography and inductively coupled plasma (ICP) methods and so on. All the wear measurement techniques' purpose is to determine the elemental content of each debris particle and the debris size [1], which can tell us which parts may be in bad working status. Much attention has also been paid to the analysis of the lubricating oil matrix during its life. The goal of the oil analysis is to figure out the substantial physical and chemical changes of lubricating oil due to the oxidative high temperature degradation and contamination by water, ethylene glycol, fuel, soot, which can detect the lubricating oil degradation stages and signal the need for an oil change when the oil's condition warrants it.

Traditionally oil condition monitoring is performed using a range of chemical and physical tests such as viscosity, total acid number (TAN), total base number (TBN), insolubles content (IC), fuel and water dilution, glycol contamination, etc. However these testing are always tedious, time-consuming and expensive to carry out. So a lot of new and efficient sensors and spectroscopic strategies have been proposed for lubricating oil analysis [2]. Actually, the exact lifetime or useful life remaining of the lubricating oil cannot be predicted because it depends on various parameters such as different oil formulations and equipment working circumstances. But it is imperative to determine which stage lubricating oil stay and signal the

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need for an oil change. Currently, most of the sensor and spectroscopic strategies for lubricating oil analysis are based on the standard chemical and physical testing results including TAN, TBN, viscosity, water and fuel contents, glycol contaminant, soot, etc. The most widely applied sensor and spectroscopic strategies were Fourier transform infrared (FT-IR) and dielectric spectroscopy (DES sometimes called impedance spectroscopy, and also known as electrochemical impedance spectroscopy (EIS)) [2,3].

FT-IR monitoring parameters including oxidation, nitration, sulfate by-products, soot, glycol contamination, diesel fuel dilution, anti-wear depletion and water contamination have broadly been applied in rapid laboratory analysis of lubricating oil quality [4–9]. Especially, FT-IR oil condition monitoring by direct trending [9] is a semi-quantitative procedure, in that no calibration against a primary reference method is performed. Instead, results are reported in terms of peak areas or heights (or baseline tilt in the case of soot) and interpreted in relation to empirically established criteria [7]. A series of ASTM standard practices such as E2412 [9], D7412 [10], D7414 [11], D7415 [12], D7624 [13], D7844 [14], etc. have been published and applied widely [15]. The main disadvantage of the FT-IR technology is its poor environmental adaptability, which makes FT-IR not suitable for on-line or in-situ lubricating oil monitoring.

DES as a non-invasive and rapid analysis method, has been proved to be a useful tool for analysis of industrial lubricants and can offer the opportunity to characterize, evaluate and provide insights into mechanisms of lubricant performance and degradation. Combined with simple, well fabricated and utilized sensors, dielectric and electrochemical properties have been shown practical for automotive oil condition monitoring [16–20]. Lvovich, et al. has analyzed the EIS characterization of industrial lubricants in the frequency range from 10 MHz to 1mHz and established a relationship between the lubricant chemical composition and EIS data [21]. Electrochemical sensors based on EIS and cyclic voltammetry have been utilized to detect water leaks and continuously monitor the time dependent dynamics of water/oil interactions following the injection of water into industrial lubricant reported by Smiechowski et al. [22]. The EIS study of carbon black based model systems in the frequency range from 10 MHz to 10 Hz has demonstrated that the complex mechanism of interaction between charged carbon black particles and surfactants (primarily dispersants) was responsible for maintaining dispersions in engine oils by Smiechowski et al. [23].

Spectroscopic analysis technologies including FT-IR and DES can provide enough and useful information about the chemical and physical properties of the lubricating oil during its degradation. And FT-IR and DES results correlate well with physical and chemical recognition sensors combined with chemometrics multivariate data analysis [2–6,21–26].

According to the results currently reported, normal lubricating oil analysis technologies based on FT-IR and DES spectroscopic strategies usually include two steps. Firstly, the relationships between normal lubricant properties such as TAN, TBN, viscosity, water and fuel contents, glycol contaminant, soot, etc., and FT-IR or DES (EIS) data should be established. Secondly, the analysis results of normal lubricant properties could be used to determine the oil condition and signal the need for an oil change. Actually, it could be more efficient and effective to directly determine the oil condition by the lubricant physical and chemical composition information from FT-IR or DES data. As for lubricating oil monitoring, simple and efficient analysis method is indispensable to qualitatively determine the degradation degree of lubricating oil and signal the need for and oil change. So the qualitative analysis models which can determine the lubricant degradation degree and give the direct advice for oil change are enough for oil condition monitoring.

In this article, two-channel and differential dielectric spectroscopy (TD-DES) analysis method has been applied to obtain the TD-DES data of 33 in-service lubricating oils collected from the in-service engines. Chemometrics statistical methods including principal component analysis (PCA) and modified hierarchical cluster analysis (HCA), called supervised hierarchical cluster analysis (S-HCA), have been put forward and used to establish the qualitative analysis models based on the TD-DES data and FT-IR data of the in-service lubricants. This article aimed to develop an effective technique for lubricating oil condition analysis based on the combination TD-DES and S-HCA, which can determine the oil condition and signal a need for oil change in time efficiently.

2. Material and methods

2.1. Samples

Thirty-three in-service engine lubricant samples including 32 used oil and their corresponding fresh oil XingPu CD10W-40, all the used oil were collected from 32 vehicles of the same type with different mileages. The samples were labeled and listed in Table 1.

2.2. FT-IR tests

Perkin Elmer 400 FT-IR spectrum was used to collect the FT-IR spectra of the samples. A pair of well-polished KBr windows with 0.1 mm spacer was used to acquire the sample spectral data at room temperature about 23 ± 1 °C in the range of 4000 cm^{-1} to 400 cm^{-1} at a 2 cm^{-1} resolution.

2.3. TD-DES tests

A TD-DES Analyzer made by Logistical Engineering University (LEU) was employed to obtain the DES data. The principle of TD-DES detection is simultaneously applied excitation signal to the sample sensor and the reference sensor, followed by the differential operation to eliminate the influence of substrate (such as the substrate material of planar interdigital capacitor sensor) and enhance the sensitivity significantly [27]. A pair of planar interdigital capacitor sensors were adopted for convenient sample attachment. The sensor with gold plated copper electrode combs and substrate material of Rogers RO4003C has a round shape with 33 mm diameter and the spacing between the comb fingers and their width are both 450 μm.

The TD-DES spectra were obtained at room temperature 20 ± 1 °C under the sine wave excitation voltage of 18 V (peak to peak) with the frequency range from 1 kHz to 95 kHz at an interval of 500 Hz.

2.4. Theory of the supervised hierarchical clustering analysis (S-HCA)

Identifying natural patterns in data is one of the most important goals of chemometrics. Specifically, clustering techniques are almost indispensable as a tool for data mining, which can be often divided into unsupervised and supervised methodologies [28]. As one of unsupervised clustering techniques, HCA can be applied to multidimensional data sets, in order to study similarities (or dissimilarities) of objects in the variables space, or similarities of variables in the objects space [29–33].

In this article, TD-DES data have been preprocessed by PCA. And the HCA has been modified into a kind of supervised clustering technique, called S-HCA. The calibration model based on S-HCA can be called S-HCA calibration model. The samples data set which will be used to build the S-HCA calibration model could be called calibration set. The unknown samples to be analyzed by