Distilling determination of water content in hydraulic oil with a ZnO/glass surface acoustic wave device

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Detection of water content in hydraulic oil is critical to identify abnormal wear conditions for purpose of predicting possible machinery failure in hydraulic systems. The paper reports a feasibility study of measuring the water content in the hydraulic oil using a ZnO thin film surface acoustic wave (SAW) device combined with the standard distillation method. The shift of resonant frequency of the SAW device increases with the increase of water content in hydraulic oil, and reaches 919 kHz for 0.80 wt.% water content in oil samples. The results indicate that the ZnO SAW sensor can detect water content in hydraulic oil with high sensitivity.

Keywords: surface acoustic wave; frequency shift; ZnO sensor; detection of water content

1. Introduction

Water content in hydraulic oil is potentially harmful to the hydraulic systems and may even cause catastrophic damage to the machines. Water molecules can speed up oxidation and reduce life span of the systems by ways of rusting, corroding, etching and depositing. Therefore, measurement of the water content in hydraulic oil is extremely crucial. But quantitative water detection in oils is difficult because of perturbation of other impurity substances including dirt, soot, alkanes and synthetic compounds. So far, several qualitative and quantitative techniques such as electrostatic extraction [1], Karl Fischer titration test [2], distillation [3] and electric dehydration method [4] have been reported to determine water content in oils. Among them, the standard distillation method (Dean and Stark distillation method [5]) is regarded to be one of the most mature detecting methods, but is a fairly burdensome method and needs a substantial sample volume to guarantee the accuracy. The achievable detection limits of water contents in oils are between 0.05 and 25 wt.% [6].

Surface acoustic wave (SAW) devices are one of the key components for electronics, microsensors, and microsystems. SAW devices have been widely studied to use as a sensor to detect gas, humidity and moisture owing to their high sensitivity, small size, and ability to be interfaced with passive wireless systems, etc [7, 8]. For instance, Khan *et al.* studied SAW device based fluorine gas sensor integrated on ZnO nano thin film with palladium (Pd) on top as sensing layer [9]. Murakawa *et al.* integrated ionic liquid on a SAW resonator to detect hydrogen sulfide gas [10]. A SAW humidity sensor based on electrospun nanofibers sensitive layer was reported to have a sensitivity of 75 kHz/%RH from 20 to 90% RH [11]. SAW devices based on Rayleigh mode were also used for measuring a small variation of moisture in gas medium with high accuracy [12, 13].

Here, we firstly propose a novel way to measure water content in hydraulic oil by using SAW device as humidity sensor combined with the standard distillation method. It is shown that SAW devices can be used to detect water content down to 0.01 wt% in oils, overcoming the limitation of the distillation method, demonstrating its potential for the development of portable sensor system for monitoring the quality of hydraulic oil.

2. SAW sensor and measurement setup

The piezoelectric thin film SAW sensors were fabricated on a 2-inch Corning glass 2318 substrate with a thickness of 1.1 mm. A 3-μm-thick ZnO thin film was deposited on the glass as piezoelectric layer by direct current (dc) reactive magnetron sputtering method. The deposition conditions were optimized [8, 14]: substrate temperature of 100 °C, deposition pressure of 2 Pa, O_2/Ar mixture gas at a ratio of 50/100 sccm, sputtering power of 200 W, and bias voltage of −75 V. Fig. 1 shows a SEM picture of the ZnO thin film on the glass substrate. A clear columnar structure with large grain sizes indicates c-axis orientation of the ZnO film, which is crucial for the performance of SAW devices. Aluminum in thickness of 750 nm was used to fabricate the interdigitated transducers (IDTs), which was deposited by sputtering and patterned by lift-off process.

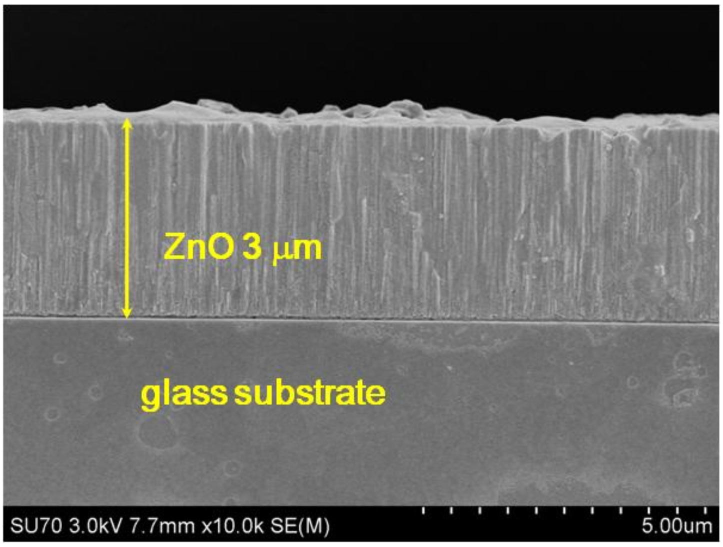

Fig. 1 Cross-section view of ZnO thin film on glass substrate

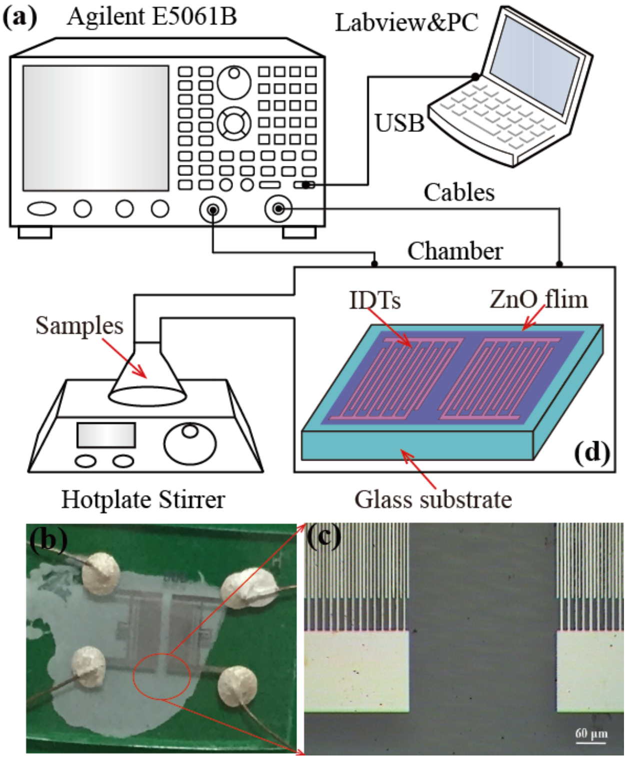

FIG. 2 (a) Schematic diagram of the experimental setup, (b) the fabricated SAW device, (c) an enlarged view of the IDT electrodes and (d) schematic view of the SAW device.

The distance between the two IDT electrodes was 20λ, with the wavelength λ determined by the IDT pitch. The reflector was in the IDTs to enhance the performance of the SAW devices. The IDT electrodes are composed of 60 pairs of fingers. For this research, only the

SAW devices with wavelength of λ=16 μm were used. Our previous work [16, 17] has demonstrated the fabricated SAW devices with a wavelength of λ=16 μm have the Rayleigh wave mode, performed well with resonant frequencies round ~171.7 MHz and have a high sensitivity to humidity. Fig. 2(b) and (c) are the photo images of the SAW device used for sensing experiments.

The measurement setup is shown in Fig. 2(a). Oil (petroleum-based hydraulic oil, L-HM46) containing various contents of water in a conical flask were heated on a hotplate. The conical flask was connected to a plastic cylinder chamber in a sealed way. The chamber was capped with thermal insulation foam to avoid thermal radiation from the hotplate which will influence the gas temperature in the chamber, hence the measurement accuracy. The SAW device was mounted in the chamber connected to the flask. The evaporated water moisture from the oil will be absorbed by the hydrophilic ZnO surface of the SAW device, and monitored by tracking the frequency shift based on the principle of SAW humidity sensor. Transmission properties of the SAW devices were measured by a network analyzer (Agilent E5061B) to analyze the frequency spectrum and frequency shift. A laptop with Labview program was used to conveniently control the measurement and collect data from the network analyzer. Since the water content ratios greater than 0.1 wt.% can cause severe harm to hydraulic systems, the experimental oil specimens in a volume of 50 ml were prepared by adopting the method of standard addition with water content ranging from less than 0.01 to 0.80 wt.%.

3. Results and discussion

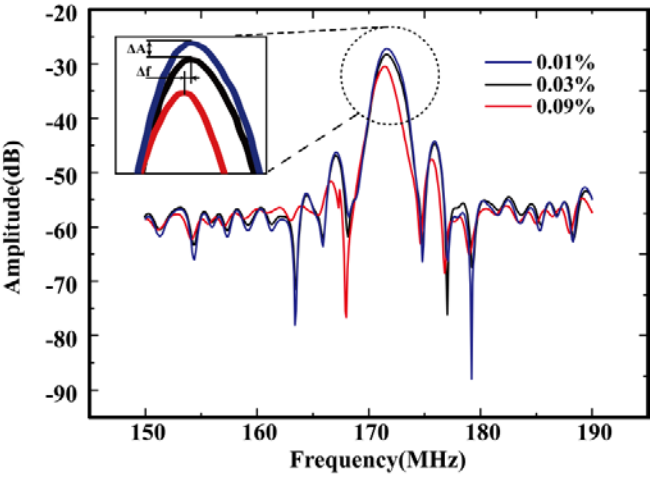

FIG. 3 Transmission spectra of the SAW device for oil samples with different water contents at the heating

time of 600 s.

When the SAW device was used to test oil samples with water contents of 0.01, 0.03 and 0.09 wt.%, it showed a similar characteristic with different resonant frequencies and amplitudes under the same heating time of 600 s (Fig. 3). Both of the amplitudes and frequency of the SAW device decreased quickly as the amounts of water in the samples increased. The characteristics agreed with the non-saturation behavior, which means that the SAW device can operate in this range of water contents. Attachment of additional substances such as water molecules on the surface of the SAW device will induce a shift of resonant frequency and attenuation of amplitude due to the mass loading effect. Since there was no absorption of inclusions (additives, soot and debris etc.) on the sensing film during the distillation, the mass accumulation is merely caused by moisture on the sensing film. The Sauerbery equation [18] gives the linear relationship between resonance frequency shift and additional mass attached on the resonator:

$$\Delta f = -Cf_0^2 \,\Delta m/A \tag{1}$$

where C is a constant related to the substrate, and f_0 is the resonant frequency, and $\Delta m/A$ is the mass change per unit area.

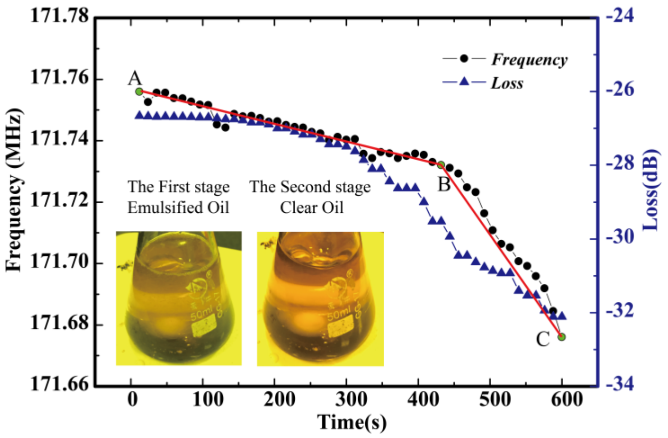

FIG. 4 Frequency responses and insertion loss of the SAW device with the heating time change.

Fig. 4 shows the change of resonant frequency and insertion loss of the SAW device treated with distilling the oil sample with 0.03 wt.% water. Apparently, the resonant frequency and insertion loss decreased continuously as the heating time increased from 0 to 600 s. More specifically, the maximum frequency shift approximated to 80 kHz and the insertion loss was clearly reduced by approximately 5.45 dB, reaching to around -32 dB.

More detailed observations revealed that the obtained dynamic responses of all the samples can be roughly divided into two regions with different slopes relative to humidity. As shown in Fig. 4, in the first region with time from 0 to 400 s (Point A to B), the maximum center frequency and insertion loss changed slowly with reduction only about 20 kHz and 1 dB, respectively. During the period of 0 to 400 s, the sample of hydraulic oil was severely emulsified according to the dissolved water content [19], as shown in the left insert of Fig. 4. These changes were primarily caused by the increase of temperature. Since little moisture was generated in the chamber, small change of mass caused small change of resonant frequency of the SAW device according to the equation (1). But in the second region from 400 to 600 s, the frequency and insertion loss of the sensor had a large change of about 60 kHz and 4dB, respectively, and the sample became more crystal-clear since water in oil was distilled, as shown in the right insert of Fig. 4. The characteristic changes of the sensing are mainly correlated to the absorption of moisture from the oil sample. At this stage with the steady high temperature, the frequency changed greatly because of the accumulation of more water molecules attached on the sensing surface of the SAW device.

More oil samples were tested with the water contents from less than 0.01 to 0.8 wt.%, as shown in Fig. 5. Similarly, the curves in Fig. 5 can be divided into two regions with different gradients. Time interval for the first region is different for different samples. In the first region, the change of frequency is due to the temperature variation, while in the second one, the frequency change is due to the water vapor loading on the SAW sensor. The pre-heating time (e.g. 400 s for 0.3 wt.%) for the first region decreased as the water content increased. The resonant frequencies of all the samples downshifted slightly with time in the first region, but decreased much quicker in the second stage. The larger the water content, the more the frequency changed. For the sample with a water content less than 0.01 wt.%, the resonant frequency shift was only 34 kHz. However, for the sample with water content of 0.09 wt.%, the frequency had a significant change of 389 kHz. Fig. 5(b) shows the frequency shift of the samples from 0.2 to 0.8 wt.% in the period of heating time from 450 to 600 s. The resonant frequencies of the samples (0.2~0.8 wt.%) fluctuated sharply, and even crossed with each other for the period from 500 to 550 s. This might be caused by the scattered formation of water vapor on the ZnO thin layer at the non-uniform generating speeds. Although hydraulic oil with such high level of water content is difficult to find in the market, it may exist in hydraulic systems in bad conditions.

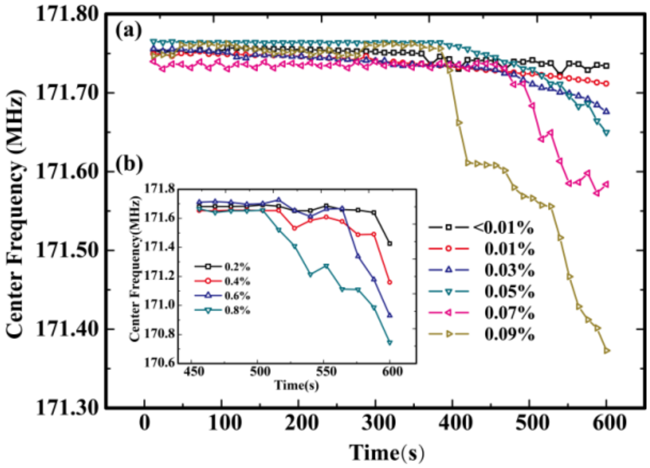

FIG. 5 Frequency shifts of the samples (with water content from 0.01% to 0.09 wt.%) and (b) the samples (with water content from 0.2% to 0.8 wt.%) as a function of heating time.

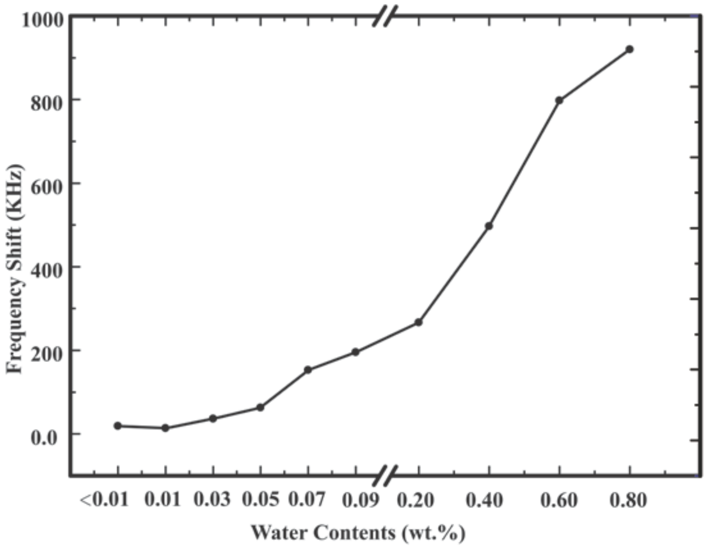

FIG. 6 Resonant frequency shift of all the samples at the heating time of 600s.

Fig. 6 shows the frequency shift and relative change ($\Delta f/f_0$) of all the samples as a function of water content at the heating time of 600 s. Both of the values of two indicators increased when water content increased. For the sample with water content less than 0.01 wt.%, the frequency shift and $\Delta f/f_0$ were only 34 kHz and 111 ppm, respectively. However, for the sample with 0.80 wt.% water content, the frequency shift and $\Delta f/f_0$ greatly increased to 919 kHz and 5300 ppm, respectively. As we can see, the proposed method can detect water content in hydraulic fluid with high sensitivity of frequency shift, and has advantages of simple operation and time saving (600 s). However, more work needs to be done to find the smallest detectable water content, and study the nonlinear relationship between water content and frequency shift.

4. Conclusions

In summary, a novel method based on the combination of the standard distillation and the Rayleigh mode SAW device was proposed to measure water content in hydraulic oils. The SAW sensor exhibits a good performance in response to the samples with various water contents. The water content in hydraulic oil can be detected from the frequency shift of the SAW sensor with high sensitivity. The proposed method provides a possibility to develop a portable device for measurement of water content in hydraulic oil in the future.

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References

[1] Tetsuji Hirato, Kazuya Koyama, Takahiro Tanaka, Yasuhiro Awakura, and Hiroshi Majima, Demulsification of water-in-oil emulsion by an electrostatic coalescence method, Materials Transactions, JIM 32 (3), 257 (1991).

[2] Steven K MacLeod, Moisture Determination Using Karl Fischer Titrations, Analytical Chemistry 63 (10), 557A (1991).

[3] Timothy E Knight and BM Hausen, Melaleuca oil (tea tree oil) dermatitis, Journal of the American Academy of Dermatology 30 (3), 423 (1994).

[4] Giles John Brereton and BA Bruno, Particle removal by focused ultrasound, Journal of sound and vibration 173 (5), 683 (1994).

[5] EW Dean and DD Stark, Industrial & Engineering Chemistry 12 (5), 486 (1920).

[6] Larry A Toms and Allison M Toms, Machinery Oil Analysis: Methods, Automation and Benefits: a Guide for Maintenance Managers, Supervisors & Technicians. (Society of Tribologists and Lubrication Engineers, 2008).

[7] Jian Zhou, Xingli He, Hao Jin, Wenbo Wang, Bin Feng, Shurong Dong, Demiao Wang, Guangyi Zou, and JK Luo, Crystalline structure effect on the performance of flexible ZnO/polyimide surface acoustic wave devices, Journal of Applied Physics 114 (4), 044502 (2013).

[8] Jian Zhou, Xing Li He, Wen Bo Wang, Qi Zhu, Wei Peng Xuan, Hao Jin, Shu Rong Dong, Miao De Wang, and Ji Kui Luo, Transparent surface acoustic wave devices on ZnO/glass using Al-doped ZnO as the electrode, Electron Device Letters, IEEE 34 (10), 1319 (2013).

[9] Saleem Khan, Sandeep Arya, Suresh Kumar, Parveen Lehana, Fabrication and characterization of highly sensitive ZnO/Si SAW device with Pd selective layer for F_2 gas sensing, Microsystem Technologies, volume 21, issue 9, pp 2011-2017, 2015.

[10] Yoshinobu Murakawa, Motoaki Hara, Hiroyuki Oguchi, Yuichiro Hamate, Hiroki Kuwano, Surface acoustic wave based sensors employing ionic liquid for hydrogen sulfide gas detection, Microsystem Technologies, volume 19, issue 8, pp 1255-1259, 2013 .

[11] Lin Q, Li Y and Yang M, Highly sensitive and ultrafast response surface acoustic wave humidity sensor based on electrospun polyaniline/poly(vinyl butyral) nanofibers. Anal. Chim. Acta 748, 73－80 (2012).

[12] Saurabh Kumar, PS Mukherjee, and NM Mishra, Online condition monitoring of engine oil, Industrial lubrication and tribology 57 (6), 260 (2005).

[13] Xiaohui Hu and Wuqiang Yang, Planar capacitive sensors - designs and applications, Sensor Review 30 (1), 24 (2010).

[14] X. L He, D. J Li, J. Zhou, W. B Wang, W. P Xuan, S. R Dong, H Jin, and J. K Luo, High sensitivity humidity sensors using flexible surface acoustic wave devices made on nanocrystalline ZnO/polyimide substrates，Journal of Materials Chemistry C, 1 (39), 6210 (2013).

[15] D. J Li, C Zhao, Y. Q Fu, and J. K Luo, Engineering silver nanostructures for surface acoustic wave humidity sensors sensitivity enhancement, Journal of The Electrochemical Society 161 (6), B151 (2014).

[16] W. P Xuan, L He, J.K Chen, W. B Wang, X. Z Wang, Y Xu, Z Xu, Y. Q Fu and J. K Luo, High sensitivity flexible Lamb-wave humidity sensors with a graphene oxide sensing layer, Nanoscale, 7, 7430 (2015).

[17] Weipeng Xuan, Mei He, Nan Meng, Xingli He, Wenbo Wang, Jinkai Chen, Tianjin Shi, Tawfique Hasan, Zhen Xu, Yang Xu, J. K. Luo, Fast response and high sensitivity ZnO/glass surface acoustic wave humidity sensors using graphene oxide sensing layer, Scientific Reports, 4,7206-7206 (2014)

[18] K. Bodenhöfer, A. Hierlemann, G. Noetzel, U. Weimar and W. Göpel, Selective detection of nitrogen and oxygen containing volatile organic compounds: use of metal-modified polysiloxanes as sensor coatings, Anal. Chem., 68, 2210 (1996).

[19] Bauer C, Day M. Water contamination in hydraulic and lube systems[J]. Practicing Oil Analysis Magazine, 2007, 9(9-10).