Dual-Modality Probe for Characterization of Heterogeneous Mixtures

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***Abstract*—This paper presents a laboratory feasibility study aiming at the development of a dual modality sensor for development within an oil and gas extraction plant to measure the composition of heterogeneous mixtures. The technology of manufacturing the sensor is briefly described and the preliminary measurements, conducted for mixtures of vegetable oil and saline water, using two modalities: ultrasound and electrical, are presented. The experimental results obtained from ultrasonic measurements are compared with theoretical predictions. Finally, conclusions and recommendations for future work are presented.**

***Index Terms*—Heterogeneous mixtures, speed of sound, thick-film, ultrasonic.**

I. Introduction

THE AIM OF THE presented study is to develop a reliable dual modality sensing technique to be deployed in the hostile environment of an oil and gas extraction plant, particularly within primary separation systems (both top-side and subsea) or, potentially, within the down-hole environment. Here, the heterogeneous mixtures consist of at least four components: crude oil, formation water, gas, and solid particulates. Typical operating conditions of such installations are characterized by high temperature reaching in excess of 150 °C pressure in excess of 150 bar, and the presence of highly aggressive chemical compounds. From the above, it is clear that any possible sensor design must meet at least two criteria: to survive the high operating pressure and temperatures and to ensure the chemical stability of the components exposed to process media.

Generally, heterogeneous mixtures of multiple phases or components are a common occurrence within many industrial processes; numerous examples can be given from areas as diverse as chemical, petrochemical, oil and gas extraction, pharmaceutical, food and drink and water treatment plants. The ability to monitor and control the multiphase phenomena is therefore one of the most fundamental issues.

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II. Probe Design and Manufacture

A dual modality probe using two measurements modalities: electrical and ultrasound, was designed, manufactured, and tested in laboratory conditions. The probe is built from two identical transducers. Each of them consists of two electrodes with a piezoelectric layer connected to the first and second electrode. The first electrode, second electrode, and piezoelectric layer of each transducer form a pressure wave transducer. Both transducers are displaced from one another such that the second electrodes from both transducers can be used for electrical measurements.

The fabrication of the transducers was undertaken at the University of Southampton within the School of Electronics and Computer Science. The devices were made using thick-film technology, with which the Southampton group have considerable experience, [1]. The sensors comprise three layers of thick-film material: a lower electrode layer, a piezoelectric layer, and an upper electrode layer. The substrate material was 96% alumina of thickness 635 μm. Each electrode layer was fabricated with a commercial gold paste and the piezoelectric layer was a special-purpose paste developed at the University. Lead zirconate titanate (PZT) powder type 5A, manufactured by Morgan Electronics, Ltd., was chosen as the active material for the piezoelectric paste. This has a Curie temperature exceeding 170 °C, allowing the sensors to operate at the desired specification of 150 °C, as fully discussed elsewhere.

The size of the lower gold electrode is 10 $\times$ 10 mm and this is screen-printed onto the alumina substrate, dried in an infrared drier at about 1500 °C and then fired in a belt furnace at a peak temperature of 8900 °C. The thick-film PZT layer was printed twice in order to obtain a thickness of around 50 μm and then subjected to a similar firing profile to the initial layer. The upper gold layer, of dimensions 25 $\times$ 25 mm, was then printed, dried, and fired in the same manner to the other layers. The resulting device is essentially a planar capacitor-type structure. After processing, the PZT layer has to be polarized in order to induce piezoelectric behavior. This is achieved by placing the substrate on a hot plate at a temperature of around 890 °C and applying a dc electric field of 4 MV/m across the sample for 10 min.

III. Experimental Setup

Pairs of transducers, as shown in Fig. 1, were attached to a fork-like probe, as shown in Fig. 2 in such a way that the measurement of the time of flight would conduct along a 25-mm

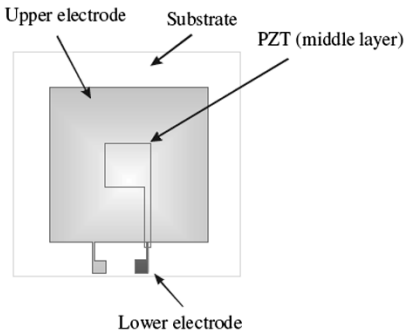

Fig. 1. Layout of the thick-film ultrasound sensor.

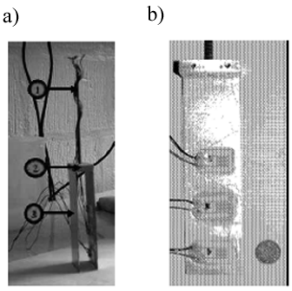

Fig. 2. (a) Fork-like probe used for supporting transducers. 1: support rod; 2: base plate; 3: aluminum "leaves" supporting the transducers. (b) An array of transducers on one of the aluminum "leaves."

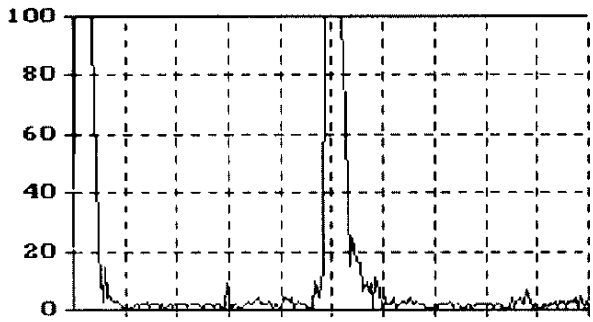

Fig. 3. Typical waveform of the sent and received signal obtained during experiments (arbitrary scale).

path in the horizontal direction between the transducers immersed in the mixture. The transducers were insulated with an epoxy layer with the thickness of 0.2 mm.

An ultrasonic flaw detector, EPOCH II, Model 2100, was used to excite one of the transducers while the other transducer acted as a receiver. The waveform of the excitation and the received signal were displayed on a computer screen connected to the flaw detector, which allowed the measurement of the time of flight of the ultrasonic wave between the two transducers.

Fig. 3 shows a typical waveform graph obtained in the experiments. From the waveform of the detector, the time between the excitation peak (the first peak in the waveform) and the receiving peak (the second peak in the waveform) can be read. With the distance between the two transducers (25 mm), and taking into account the time of flight within the layers of transducers insulation, the ultrasonic velocity can be determined. The trend of the speed of sound with the change of temperature was also investigated.

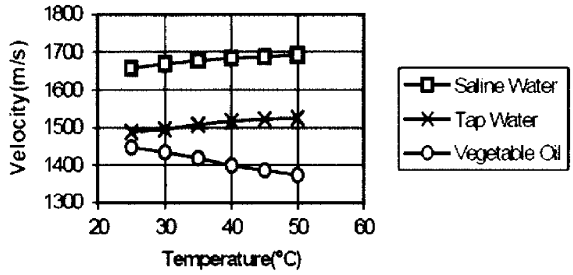

Fig. 4. Speed of sound in salty water and oil at different temperatures (data for tap water for reference).

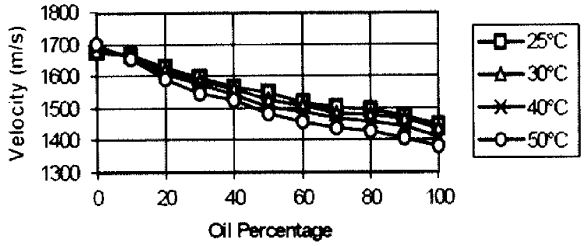

Fig. 5. Speed of sound in emulsions at different oil concentration and at different temperatures.

At the initial stage of the investigation, fluids present in the oil and gas extraction were simulated by the use of vegetable oil and saline water with a sodium chloride content of 20% by weight. They were used for both the safety aspects of the laboratory and the convenience of using relatively clean fluids. The high content of sodium chloride in saline water was chosen to mimic the density of the formation water present in oil extraction processes.

In order to obtain stable mixtures (emulsions), an aqueous surfactant solution was prepared by dissolving 1% by weight of Tween 20 (ICI Chemical & Plymers, Ltd.) in saline water. A series of vegetable oil-in saline water emulsions with volume concentration of oil varying between 10% and 50% in steps of 10% were prepared by blending the components in a glass beaker with a homogenizer (IKA-EUROTURRAX T20) at a rotational speed of 22 000 rpm. Similarly, a series of saline water in-oil emulsions with volume concentrations of saline water between 10% and 50% in steps of 10% were prepared using a similar procedure. In the latter case, to avoid confusion, instead if measuring water contents in oil, the mixtures are identified by volume fraction which varies between 59% and 100%. The Distribution Modal Size of droplets in the emulsions tested were in the range of 1.0–10.0 μm, as fully described elsewhere [2].

IV. Experimental Results

A. Ultrasonic Measurements

1) Speed of sound in pure liquids
Initial measurements were done for pure liquid and the speed of sound at different temperature was investigated. The results shown in Fig. 4 indicate that the speed of sound in saline water increases with the increase of temperature while the speed in vegetable oil decreases with the increase of temperature.

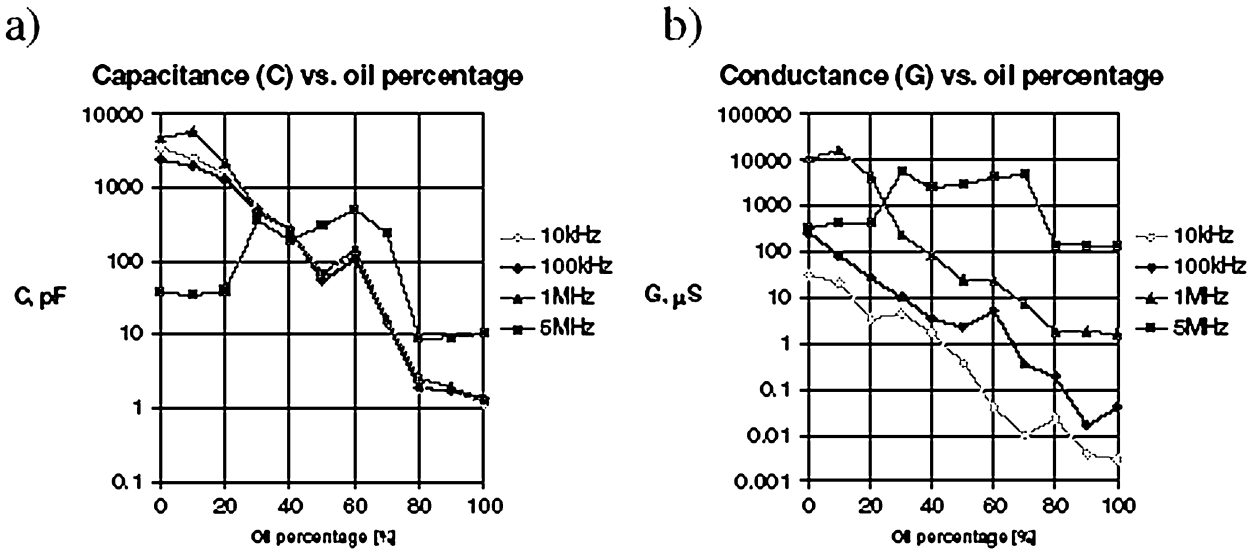

Fig. 6. Impedance measurements obtained using ac circuit: (a) capacitance and (b) conductance.

2) Speed of sound in heterogeneous mixtures
The second part of the investigation involved measurement of the sound velocity in vegetable oil and saline water emulsions. The measured velocity of sound versus oil volume fraction at different temperatures is shown in Fig. 5.

B. Electrical Measurements

The effect of varying frequency of electrical field on the measured capacitances and conductances is depicted in Fig. 6. The results of measuring the capacitance values using a charge-discharge circuit are presented elsewhere, [3]. The results shown in Fig. 6 were obtained using an impedance analyzer. Except a drastic change in the results for a frequency of 5 MHz, the measured capacitances are nearly independent of the applied frequencies, as illustrated in Fig. 6(a), whereas the conductance data shows a clear dependence on the applied frequency, as shown in Fig. 6(b).

C. Analysis of Ultrasound Data

A model describing the ultrasonic propagation in heterogeneous mixtures was γ developed by Yi *et al.* [4] and Tsouris and Tavlarides [5]. It predicts the time of flight of the ultrasonic signal as a function of the volume fraction of the dispersed phase by taking into account the change in the length of the sound path arising from reflection and refraction effects at the droplet interface.

Equation (1) summarizes the model obtained

$$\Phi = (t^* - t_c)/(g_d t_d - g_c t_c) \tag{1}$$

where Φ is the volume fraction, t^*—time of flight in dispersion, t_c—time of flight in the continuous phase; and t_d—time of flight in the dispersed phase.

The correction factors g_d, g_c for each phase account for the sound refraction phenomena and are given by the following relations:

$$g_d = 1/\gamma[1 - (1 - \gamma^2)^{3/2}] \tag{2}$$

$$g_c = 1 + 1\gamma^3/[1 - (1 - \gamma^2)^{3/2}] - 3/5[1 - (1 - \gamma^2)^{5/2}] - 2/5\gamma^2 \tag{3}$$

where γ is the ratio of the sound velocity in the dispersed phase to the sound velocity in the continuous phase. Equations (2) and (3) hold for $\gamma \leq 1$ which implies that the dispersed phase needs to have a lower sound speed than the continuous phase. This will be important for the results presented.

The relationship between speed of sound in the mixture and the volume fraction can be achieved by reorganizing (1)

$$1/v^* = \Phi(g_d/v_d - g_c/v_c) + 1/v_c \tag{4}$$

where v^* is the speed of sound in the dispersion, v_c is the speed of sound in the continuous phase, and v_d is the speed of sound in the dispersed phase. The sound velocity at a given temperature in a pure phase is constant, so there is a linear relationship between the volume fraction and the reciprocal of ultrasonic velocity in the mixed states.

The comparison of the experimental results with theoretical model is presented here for the temperature of 25 °C. Similar results can be obtained for other temperatures. From the measurements conducted in pure liquids, one can obtain the value of γ in (2) and (3). At 25 °C for the speed of sound in vegetable oil of 1446.8 ms^{-1} and that in saline water of 1700.6 ms^{-1}, the value of γ is 0.85. Equation (4) will then assume the following form:

$$1000/v^* = 0.0013\Phi + 0.5880. \tag{5}$$

It is clear from (4) and (5) that there is a linear relationship between the reciprocal of the speed of sound and the volume fraction. Therefore, it is more convenient to present data from

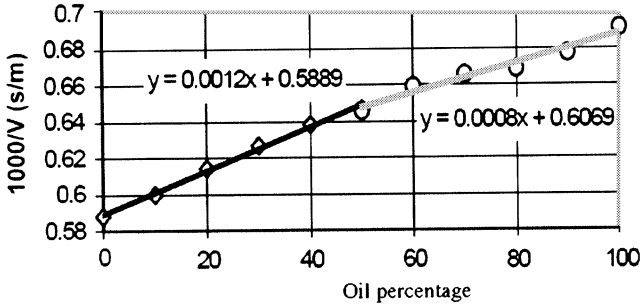

Fig. 7. Trend of 1000/V versus oil percentage at 25 °C.

Fig. 5 in the form shown in Fig. 7, where on the vertical axis the inverse of velocity multiplied by a factor of 1000 is plotted.

The data in Fig. 7 can be subjected to linear regression process. Given the condition that $\gamma \leq 1$, this can only be done for the first six points on the left of Fig. 7. The relationship obtained is given as (6)

$$1000/\mathrm{v}^* = 0.0012\Phi + 0. \tag{6}$$

It shows that the regression curve coincides almost identically with the theoretical prediction. Therefore, the theory explained with (1)–(5) can be applied to predict the fraction of the oil-in-saline water mixtures.

For oil volume fractions between 50% to 100%, there is no mathematical model $(\gamma > 1)$ available. However, it may be interesting to note that the regression analysis for this range yields the following relationship:

$$1000\mathrm{v}^* = 0.008\Phi + 0.6069. \tag{7}$$

Referring to Fig. 7, it can be seen that sound velocity in the emulsion investigated decrease with increase in the oil fraction in emulsions. From Fig. 4, the results show that the speed of sound in oil-saline water emulsions decreases with the increases of temperatures.

V. Conclusion

The concept of designing a dual-modality probe by using a thick-film technology, for metering the oil/water composition, was validated in laboratory conditions. The designed transducer consist of a layer sandwich structure. It is hoped that the materials used for the sensors will allow the sensor to survive high operating temperatures and pressures as well as contact aggressive media.

The probe has shown promising measurement characteristics in both ultrasonic and electrical modalities. Ultrasonic characteristics have been verified against a simple theoretical model.

Future work needs to concentrate on the following:

- long-term testing of the sensors in harsh conditions and evaluation of their performance;
- studies of the electrical modality in order to explain the character of the measurement obtained;
- application of dielectric coating on the sensor, most likely in the form of layer of glass sintered on the surface, and characterization of such a modified sensor design.

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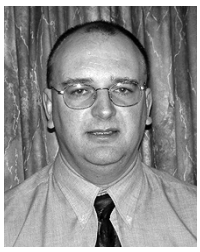

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He has many years of experience working in chemical plants. This very valuable industrial experience makes him very enthusiastic about the understanding of the process tomography methodology.