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FOR NONINVASIVE CHEMICAL DIAGNOSTICS

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SWEPT-FREQUENCY ACOUSTIC INTERFEROMETRY TECHNIQUE FOR NONINVASIVE CHEMICAL DIAGNOSTICS

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ABSTRACT

Swept-Frequency Acoustic Interferometry (SFAI) is a noninvasive fluid characterization technique currently being developed for chemical weapons treaty verification. The SFAI technique determines sound speed and sound attenuation in a fluid over a wide frequency range from outside a container (e.g., reactor vessel, tank, pipe, industrial containers etc.,). From the frequency dependence of sound attenuation, fluid density can also be determined. These physical parameters, when combined together, can be used to identify a range of chemicals. This technique can be adapted for chemical diagnostic applications, particularly in process control where monitoring of acoustic properties of chemicals (liquids, mixtures, emulsions, suspensions, etc.,) may provide appropriate feedback information. The SFAI theory is discussed and experimental techniques are presented. Examples of several novel applications of the SFAI technique are also presented.

INTRODUCTION

To meet the requirements of the Chemical Weapons Convention, the treaty that calls for eventual destruction of all chemical weapons [1], it is essential to have techniques that can be used to monitor compliance and destruction of existing stockpiles in a verifiable way. In particular, the requirements are for nondestructive and noninvasive techniques that are fast and reliable. The Swept-Frequency Acoustic Interferometry (SFAI) technique, among several others, is being developed by the US Defense Special Weapons Agency to meet such requirements.

The SFAI technique is a general-purpose, noninvasive fluid physical property characterization technique that has many applications in industry as a diagnostic and monitoring tool. In this paper, we describe the underlying principles, capabilities, and several novel applications of the SFAI technique.

The SFAI technique is a novel adaptation of the acoustic resonator interferometer technique developed several decades ago [2,3] for determining sound velocity and

absorption in liquids and gases. In the original technique, and also in more recent modifications of the technique [4,5], the transducers (sensors) needed to be in direct contact with the fluid being tested. This requirement effectively restricted the use of this technique to highly specialized laboratory characterization of fluids. In contrast, the SFAI technique extends the capabilities of the ultrasonic interferometry technique significantly and allows the determination of velocity and attenuation of sound in a fluid (liquid, gas, mixtures, emulsions, etc.,) inside sealed containers (e.g., pipes, tanks, chemical reactors, etc.,) over a wide frequency range, completely noninvasively. In addition, if the container material properties (density and sound velocity) are known, the liquid density can also be determined using the SFAI technique. Our measurements show that it is possible to uniquely identify various chemical warfare compounds and their most significant precursors based on these physical parameters (sound velocity, attenuation, frequency dependence of sound attenuation, and density).

For liquid mixtures, both sound speed and attenuation are related to mixture composition. Both mixture composition and solution concentration can be monitored with great resolution (<0.01 %) using SFAI. In the case of emulsions, the frequency-dependent sound attenuation information from this technique can be used to determine particle size distribution. The technique can also be adapted for measurements on a single drop of a liquid. Our objective in this paper is to point out some interesting applications of this general purpose technique.

PRINCIPLE OF SFAI TECHNIQUE

The underlying principle is simply the setting up of standing waves in a resonator cavity using external excitation and its simultaneous detection. This is described in Fig. 1 where a swept sine-wave voltage excitation, a sine-wave gradually increasing in frequency with time (see top frame), is applied to a disk-shaped piezoelectric transducer (transmitter) that forms one wall of an enclosure. A second identical and parallel transducer (receiver) on the opposite side of the enclosure detects the standing wave signal. The space between the two transducers is filled with a fluid. The transmitter transducer converts the sine-wave voltage signal to sound waves that propagate through the liquid and are detected by the receiver transducer as an electric signal. Resonance occurs as a result of sound waves bouncing back and forth between source and receiver. When a reflected wave incident on (returning to) the source is in synch, that is -- in phase, with the new wave being generated, the two waves interfere constructively to produce a large amplitude resonant standing wave pattern. This in-phase condition can only occur at those frequencies for which an integral number of wavelengths fit exactly in the round trip distance from the source to receiver and back. Thus a series of equally spaced (in frequency) pronounced resonance peaks, also known as interference peaks, result from a frequency sweep measurement (see bottom frame, Fig. 1). The spacing between any two consecutive interference peaks $\Delta f_n = f_n - f_{n-1}$ is related to the path length d (separation between the two

opposing transducers) and the sound velocity c_n in the liquid at frequency f_n as $c_n = 2 \cdot d \cdot \Delta f_n$. Here, n simply indicates the n -th peak.

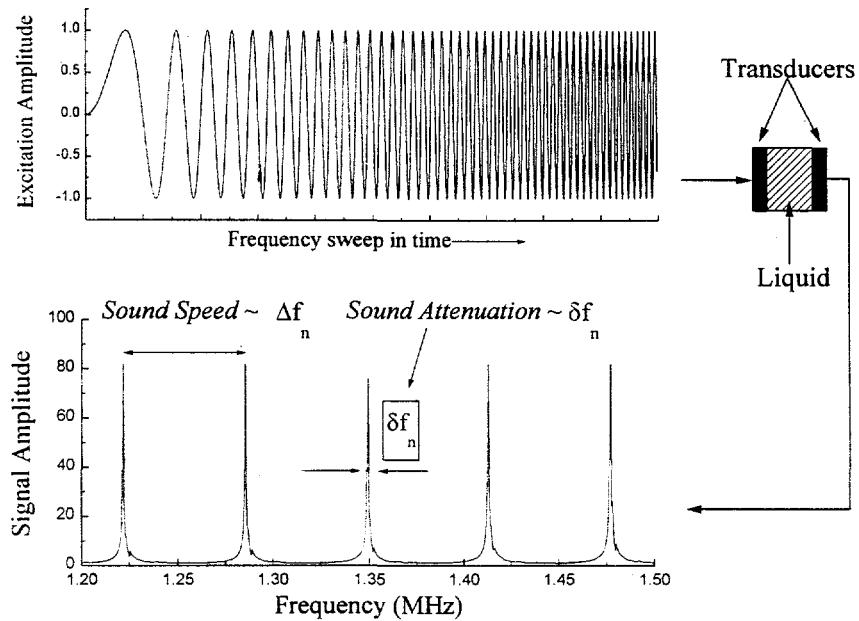


Figure 1. Principle of the swept-frequency interferometry technique. As the frequency applied to one transducer is swept from low to a high value (see top frame), the detected signal from the second transducer shows the characteristic interference pattern (see bottom frame) that develops in the liquid in the resonator cavity

The situation is slightly modified when measurements are made noninvasively with transducers attached to the outside wall of a container (e.g., pipe, tank, reaction chamber, etc.,) that contains the liquid. The spectrum in this case is a superimposition of the interference peaks in the liquid and in the wall thickness. The wall peaks are typically well separated in frequency and the liquid peaks are easily discerned in the regions between the wall peaks.

The width of the peaks δf_n contain valuable information about total sound attenuation (loss) α_n in the resonator system. In a noninvasive measurement, one needs to include the reflection loss of the container wall, that is related to the acoustic impedance (density \times sound speed) mismatch between the wall and the liquid, in addition to the absorption in the liquid. Thus, for any observed peak, contributions from both these loss mechanisms are combined together and individual contributions cannot be easily separated. However, such a separation is possible if peak-width measurements are made over a wide frequency range. The loss due to sound absorption in the liquid is frequency dependent and increases as the square of the frequency. In contrast, the wall reflection loss is frequency independent. Figure 2 shows the frequency-dependent behavior of the peak width. If the

peak-width measurements are extrapolated to zero frequency, the width δf_0 is entirely due to impedance mismatch. Because the liquid sound speed is independently determined from the liquid peak spacing, the liquid density can now be determined if the wall sound speed is known. Incidentally, the sound speed of the container wall can also be determined independently from the interference spectrum by observing the spacing between the wall peaks. Thus, a single sweep measurement can be used to derive multiple physical properties of a liquid that include: sound speed, sound absorption, frequency dependence of sound absorption, and liquid density. A combination of several of these properties can be used to uniquely identify all the chemical warfare compounds and their most significant precursors.

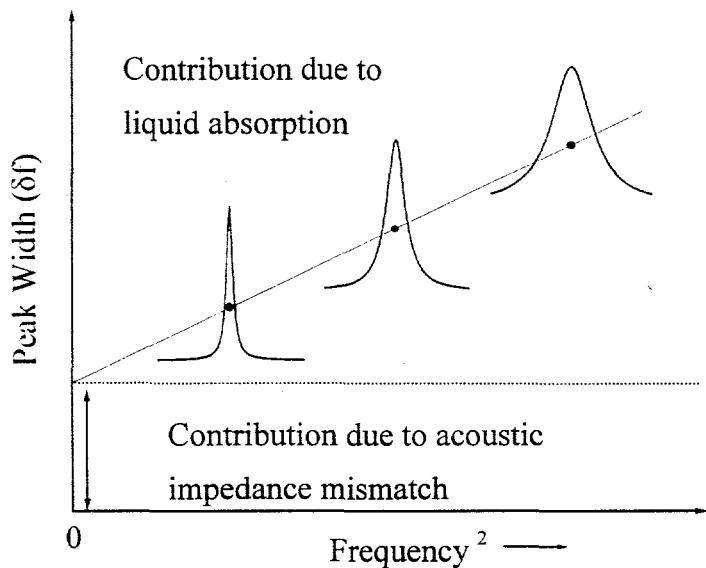


Figure 2. Determination of liquid density from frequency-dependent peak width. The intercept δf_0 at zero frequency is due to acoustic impedance mismatch. The difference $\delta f - \delta f_0$ provides a measure of the actual sound absorption in the liquid.

The frequency-dependent information is obtained in real-time and one does not need to manipulate the measured data through any kind of Fourier Transform as would be necessary for the conventional pulse-echo technique. Because this technique relies on resonance characteristics of the cavity, very little power is required for any measurement. The cavity provides a mechanical gain for the resonance signal and this gain depends on the cavity Q (quality factor). The SFAI technique also provides very high (~ 90 dB) signal-to-noise ratio in the data. For example, the data shown in Fig. 1 (bottom frame) is the raw data from a single sweep measurement without any averaging or data smoothing. This technique provides excellent quality frequency-dependent acoustic measurements that are difficult to obtain using pulse-echo techniques. The frequency resolution can be as high as 1 Hz at 10 MHz (1 part in 10 million) and, thus, the sound speed measurement can have correspondingly high resolution. It is difficult to match the accuracy and resolution of the measurement capabilities of the SFAI technique by any other acoustic

methods (e.g., pulse-echo). Note that for the SFAI technique, it is not necessary to have the transmitter and receiver transducers opposite each other as described above. We have found that custom-built dual-element transducers, that have two transducer elements side by side but are electrically and acoustically isolated from each other, can also be used for those applications requiring access from one side only. The liquid cavity can also be cylindrical, such as pipes, drums, and reaction chambers.

APPLICATION EXAMPLES

We now describe several novel applications of the SFAI technique. In the first example, we show how it is possible to monitor concentration of a solution with very high resolution. Here we show data for salt (NaCl) solution in water measured noninvasively from outside a glass container. Both sound speed and attenuation are dependent on salt concentration. The sound speed change is reflected in the shift in the peak positions, whereas the attenuation variation can be seen in both peak width and peak amplitude. Note that for sound speed it is the peak spacing that is observed and not the position of any individual peak. The inset shows the data on sound speed variation with

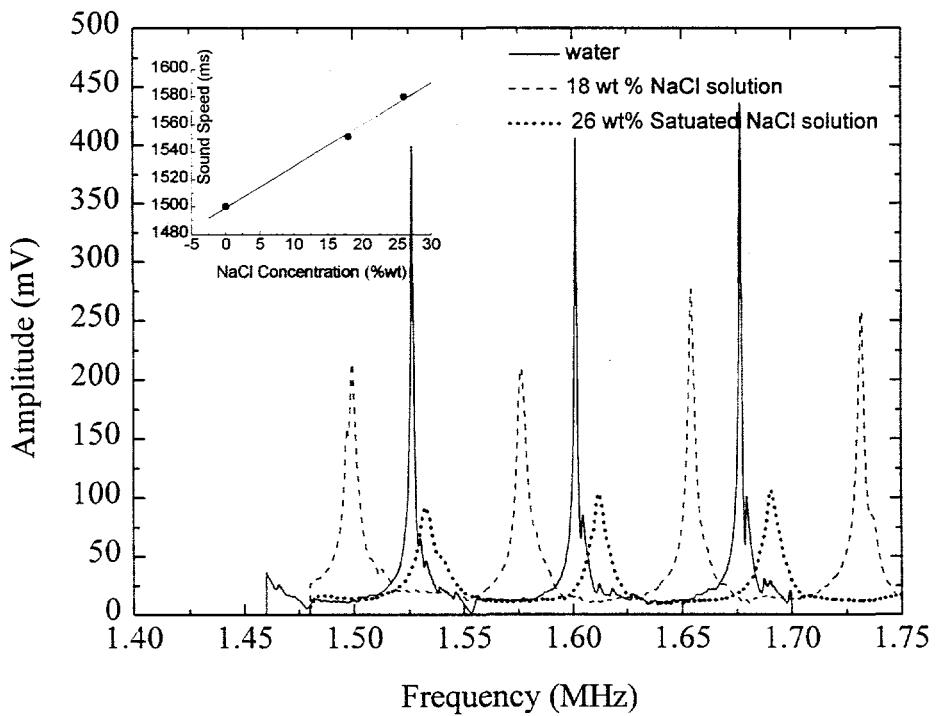


Figure 3. Sound speed and attenuation variation as a function of salt concentration. The inset shows peak-spacing (sound speed) as a function of concentration.

concentration. The slope of the least-squares fitted line is 3.03 m/s for each percent in concentration. The sound speed resolution can be better than 0.01 m/s with this technique. This represents better than 0.01 percent resolution in concentration change.

In certain situations, only a very small sample may be available for testing. The SFAI technique can be easily adapted for testing liquid samples smaller than a single drop. This becomes possible by using highly miniaturized transducers that are less than 1 mm in diameter. Various implementations are possible. One simple mechanical arrangement of the system involves aligning two such transducers, fixed inside thin stainless steel tubes, along a straight line with a small gap (~ 1 mm) between the two opposing transducer faces as shown in the inset of Fig. 4. A drop of the liquid sample to be tested is placed in the gap. Surface tension of the liquid automatically pulls in the drop within the transducer gap. This obviates the need for having a container for the liquid, and cleaning is very straightforward. Figure 4 shows some typical data taken with such a system for three different liquids. As expected, each liquid shows different peak spacing (i.e., sound speed) and peak width (sound attenuation).

Many novel studies using only minute quantities of liquid are possible with this arrangement. One example is that for photo-active chemicals, where one can focus a light beam of appropriate wavelength on the drop and observe the effect of light on the ultrasound characteristics. For sensitive measurements, the light beam is chopped and the

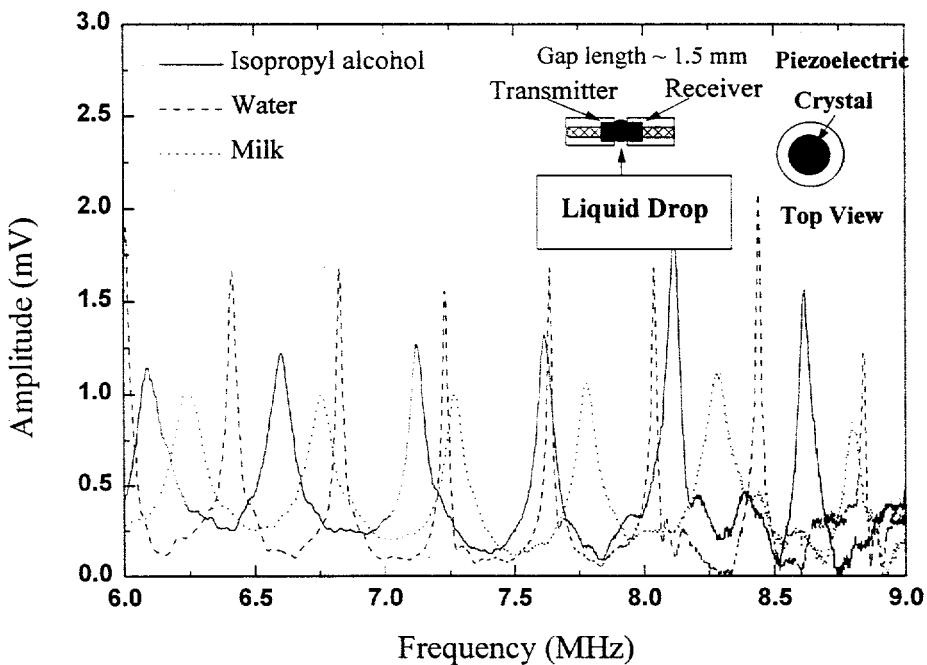


Figure 4. Ultrasonic interference measurements for single drop of liquids. The inset shows the transducer arrangement.

modulation of a single interference peak is followed using a lock-in amplifier. For observing single drops of liquids that have magnetic properties, an external magnetic field can be applied and the effect on ultrasonic characteristics observed. As in the case of the optical modulation, the magnetic field can be modulated by applying a sine-wave current signal through a coil to generate the magnetic field. Many variations of this technique are possible. This technique is not restricted to studying liquids but can be used for gels, tissues (soft materials), and even solids.

Among other application examples, we have successfully used the SFAI technique for noninvasive characterization of various types of suspensions (e.g., TiO_2) useful in paint, paper and pulp industries. We have also shown how this technique can be used to characterize petroleum products and also determine spoiled milk in sealed milk containers (e.g., paper cartons, plastic bottles, and Tetra Pak pouches).

In summary, the SFAI technique is highly adaptable and versatile. It has excellent resolution in detecting minute changes in liquid characteristics. Although developed for chemical weapons verification, the technique has applications in many areas of industry.

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