

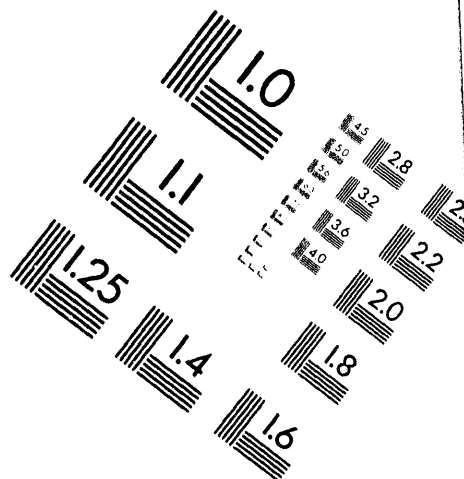
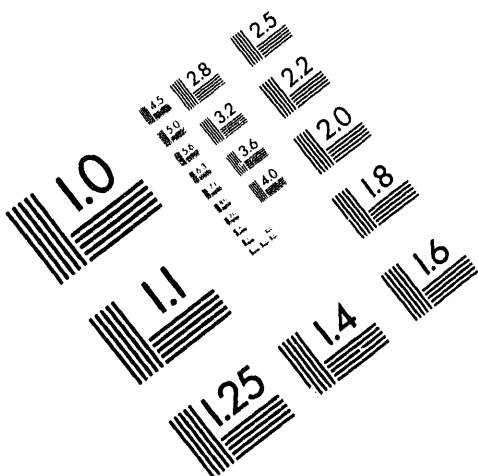


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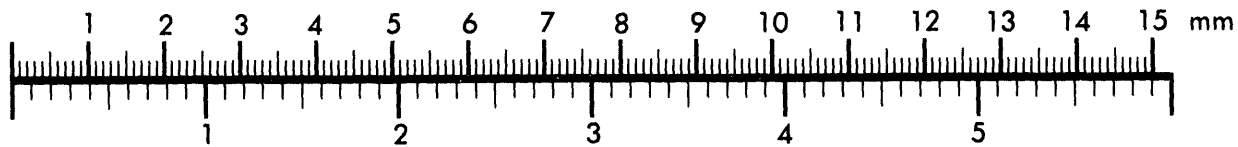
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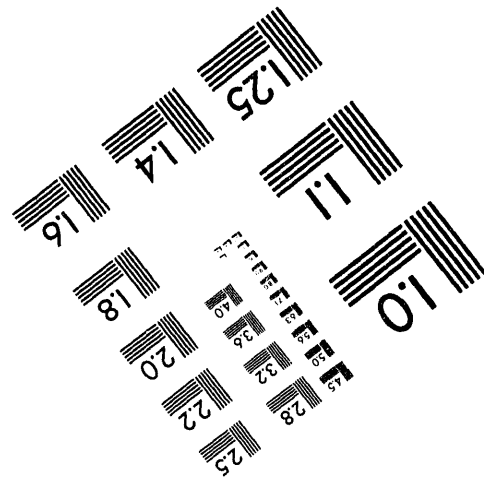
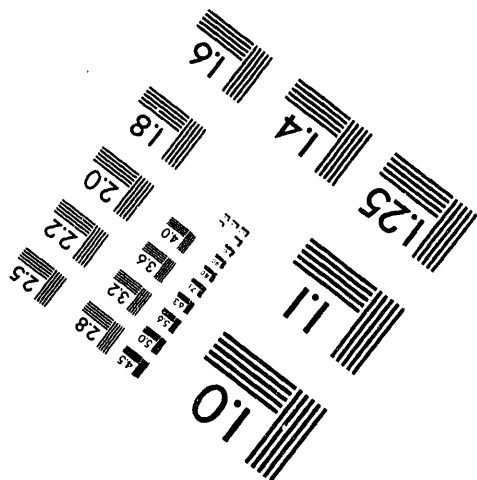
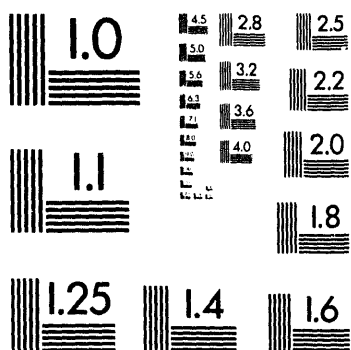
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**RESONATOR/OSCILLATOR RESPONSE TO LIQUID LOADING**

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A thickness-shear mode (TSM) resonator typically consists of a thin disk of AT-cut quartz with circular electrodes patterned on both sides. When connected to appropriate circuitry, the quartz crystal resonates at a frequency determined by the crystal thickness.

Originally used to measure metal deposition in vacuum, the device has recently been used for measurements in liquid. Since the mass sensitivity of the resonator is nearly the same in liquids as in air or vacuum, the device can be used as a sensitive solution-phase microbalance [1]. In addition, the sensitivity of the TSM resonator to contacting fluid properties enables it to function as a monitor for these properties.

Under liquid loading, the change in frequency of the resonator/oscillator combination differs from the change in resonant frequency of the device. Either of these changes can be determined from an appropriate application of an equivalent-circuit model that describes the electrical characteristics of the liquid-loaded resonator [2]. This circuit, shown in Fig. 1, consists of a capacitance  $C_o^*$  ( $C_o^* = C_o + C_p$ , where  $C_o$  is the "static" capacitance and  $C_p$  is the parasitic capacitance) in parallel with a "motional" branch ( $L_1$ ,  $C_1$ ,  $R_1$ ,  $L_2$ , and  $R_2$ ). The unperturbed (dry) device response is determined by the elements  $C_o^*$ ,  $L_1$ ,  $C_1$ , and  $R_1$ . Liquid coupling to the device surface increases the motional impedance, introducing the motional

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inductance ( $L_2$ ) and resistance ( $R_2$ ). These are related to the density  $\rho$  and viscosity  $\eta$  of the contacting fluid [2]:

$$L_2 = \frac{nL_1}{N\pi} \left( \frac{2\omega_s \rho \eta}{\mu_q \rho_q} \right)^{\frac{1}{2}} \quad (1a)$$

$$R_2 = \frac{n\omega_s L_1}{N\pi} \left( \frac{2\omega_s \rho \eta}{\mu_q \rho_q} \right)^{\frac{1}{2}} \quad (1b)$$

where  $n$  is the number of sides contacted by liquid,  $N$  is the resonator harmonic number,  $\omega_s$  is the angular series resonant frequency ( $\omega_s = 2\pi f_s$ ), and  $\rho_q$ ,  $\mu_q$  and  $K^2$  are the quartz density, shear stiffness, and electromechanical coupling factor, respectively.

If we define the series resonant frequency  $f_s$  as the frequency at which the motional inductance  $L_1$  and capacitance  $C_1$  resonate:

$$f_s = \frac{1}{2\pi \sqrt{(L_1 + L_2) C_1}} \quad (2)$$

then the equivalent-circuit model of Fig. 1, together with Eq. 1a, can be used to calculate the changes in  $f_s$  caused by liquid loading [2]:

$$\Delta f_s = -\frac{L_2 f_s}{2L_1} = -\frac{2f_s^2}{N\sqrt{\mu_q \rho_q}} \left( \frac{\rho \eta}{4\pi f_s} \right)^{\frac{1}{2}} \quad (3)$$

The change in motional resistance due to liquid loading is  $\Delta R_m = R_2$ , where  $R_2$  is given in Eq. 1b. As defined,  $\Delta f_s$  caused by liquid loading arises only from changes in the motional inductance  $L_2$  and not from changes in the motional resistance  $R_2$ . Eq. 3 agrees with the prediction of Kanazawa and Gordon [3], based on the definition of resonance given in Eq. 2. We will see below that the frequency of a resonator/oscillator does *not* follow Eq. 3 under liquid

loading conditions.

The changes in resonant frequency  $\Delta f_s$  and motional resistance  $\Delta R_m$  arising from liquid loading can be measured with a network analyzer. In many applications, however, it is preferable to use an oscillator circuit to track these changes [4,5]. Wessendorf has described an oscillator circuit capable of driving the TSM resonator in fairly damped liquid media [5]. This oscillator provides two outputs: an RF output indicating oscillation frequency, and a dc output proportional to the resonator motional resistance  $R_m = R_1 + R_2$ . This oscillator circuit will sustain oscillation for motional resistances up to approximately 4 k $\Omega$ . From Eq. 1, this enables resonator operation in liquids with viscosity values up to 130 cP (one-sided contact) and 34 cP (two-sided).

Oscillator circuits use a closed feedback loop that varies the oscillation frequency  $f_o$  to control the loop phase shift to zero degrees and unity gain. In the Wessendorf circuit, for the loop phase to be 0°, the resonator's impedance phase angle  $\phi$  will be close to 0°, independent of the resonator's impedance magnitude.

The dependence of the device impedance phase angle  $\phi$  on frequency and liquid properties can be found from the equivalent-circuit model of Fig. 1:

$$\phi = \tan^{-1}(A) - \cot^{-1}(B) \quad (4a)$$

where

$$A = \frac{(1 + \xi) - (\omega_{so}/\omega)^2}{1/Q_o + \xi} \quad (4b)$$

$$B = \frac{1}{\omega R_1 C_o^* (1 + Q_o \xi)} - A \quad (4c)$$

$$\xi = \frac{L_2}{L_1} = \frac{n}{N\pi} \left( \frac{2\omega_s \rho \eta}{\mu_q \rho_q} \right)^{\frac{1}{2}} \quad (4d)$$

and  $Q_o = \omega_s L_1 / R_1$ . The factor  $\xi$  that appears in  $A$  and  $B$  contains the influence of liquid loading in the density-viscosity product  $(\rho\eta)^{1/2}$ .

The oscillation frequency  $f_o$  is the solution to Eqs. 4 when  $\phi = \phi_o$ , where  $\phi_o$  is the impedance phase angle maintained by the oscillator. There are two solutions, one close to series resonance and one close to parallel resonance. Since the excess loop gain is much greater near  $f_s$ , the  $f_o$  solution near  $f_s$  is the frequency at which oscillation occurs. Changes in oscillation frequency  $\Delta f_o$  due to liquid loading arise from changes in *both*  $L_2$  and  $R_2$ .

Fig. 2 shows the calculated variations in  $f_s$  (Eq. 3) and  $f_o$  (Eqs. 4) vs. the liquid loading parameter  $(\rho\eta)^{1/2}$ . From Eq. 3,  $\Delta f_s$  varies linearly with  $(\rho\eta)^{1/2}$  (dashed line);  $f_o$  tracks  $f_s$  for small values of  $(\rho\eta)^{1/2}$ , but diverges at larger values. The point of departure depends on the impedance phase angle  $\phi_o$  maintained by the oscillator.

Fig. 3 shows the calculated variation in  $f_o$  vs.  $(\rho\eta)^{1/2}$  for several values of the parasitic capacitance  $C_p$ . We note that  $f_o$  tracks  $f_s$  over a wider range of  $(\rho\eta)^{1/2}$  values when  $C_p$  is small. This shows the importance of minimizing the parasitic capacitance.

Fig. 4 shows changes in  $f_s$  determined from network analyzer measurements made on a resonator contacted on one side by n-butanol. Viscosity, and to a lesser extent, density, were varied by changing the butanol temperature. Fig. 4 also shows the variation in  $f_o$  measured with

an oscillator driving the resonator. The oscillation frequency  $f_o$  tracks  $f_s$  at low values of liquid loading, but diverges at higher values, as predicted by Eqs. 3 and 4 and illustrated in Figs. 2 and 3.

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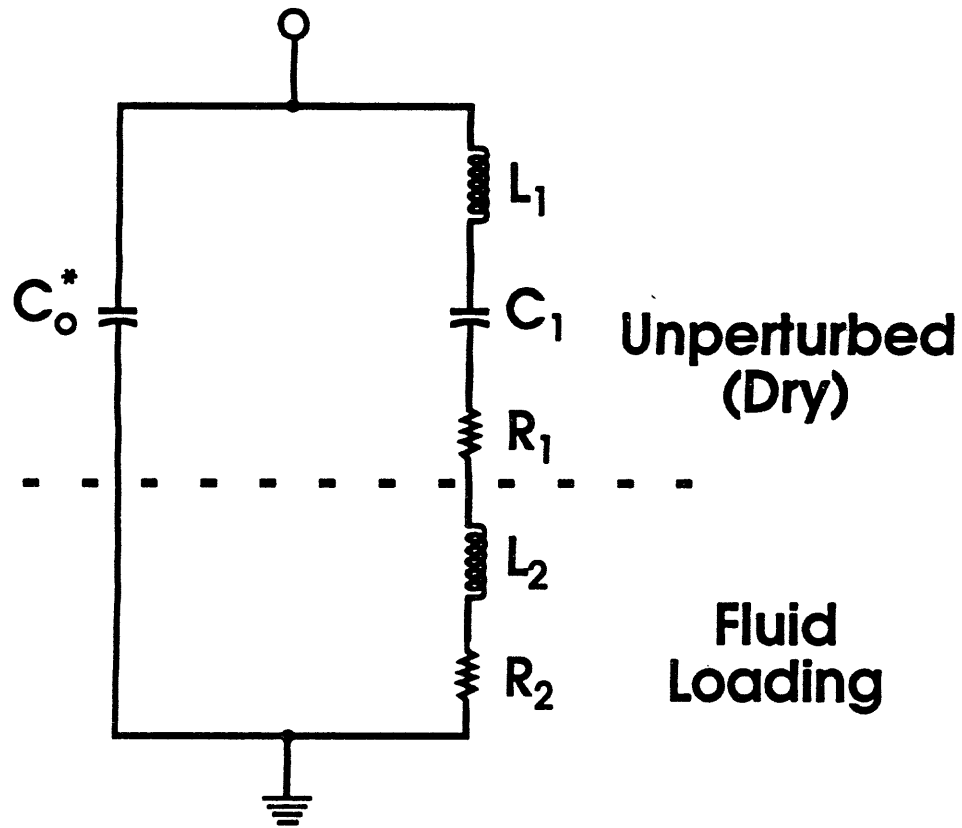
In conclusion, the changes in resonant frequency  $f_s$  and oscillation frequency  $f_o$  with liquid loading can be determined from an equivalent-circuit model that describes the electrical characteristics of the liquid-loaded resonator. The oscillation conditions pertaining to a particular circuit can be applied to the equivalent-circuit model to relate oscillation frequency to liquid properties. For the oscillator considered, a particular solution has been derived. The oscillator tracks  $f_s$  only at low values of liquid loading; at higher values,  $f_o$  falls below  $f_s$  due to the effect of liquid loading on the motional resistance  $R_2$ .

The authors wish to thank L. Casaus of Sandia National Laboratories for technical assistance. This work was performed at Sandia National Laboratories, supported by the U.S. Department of Energy under contract No. DE-AC04-94AL85000.

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*Fig. 1. Equivalent-circuit model to describe the electrical characteristics (for  $\omega$  near  $\omega_s$ ) of a TSM resonator with liquid loading.*

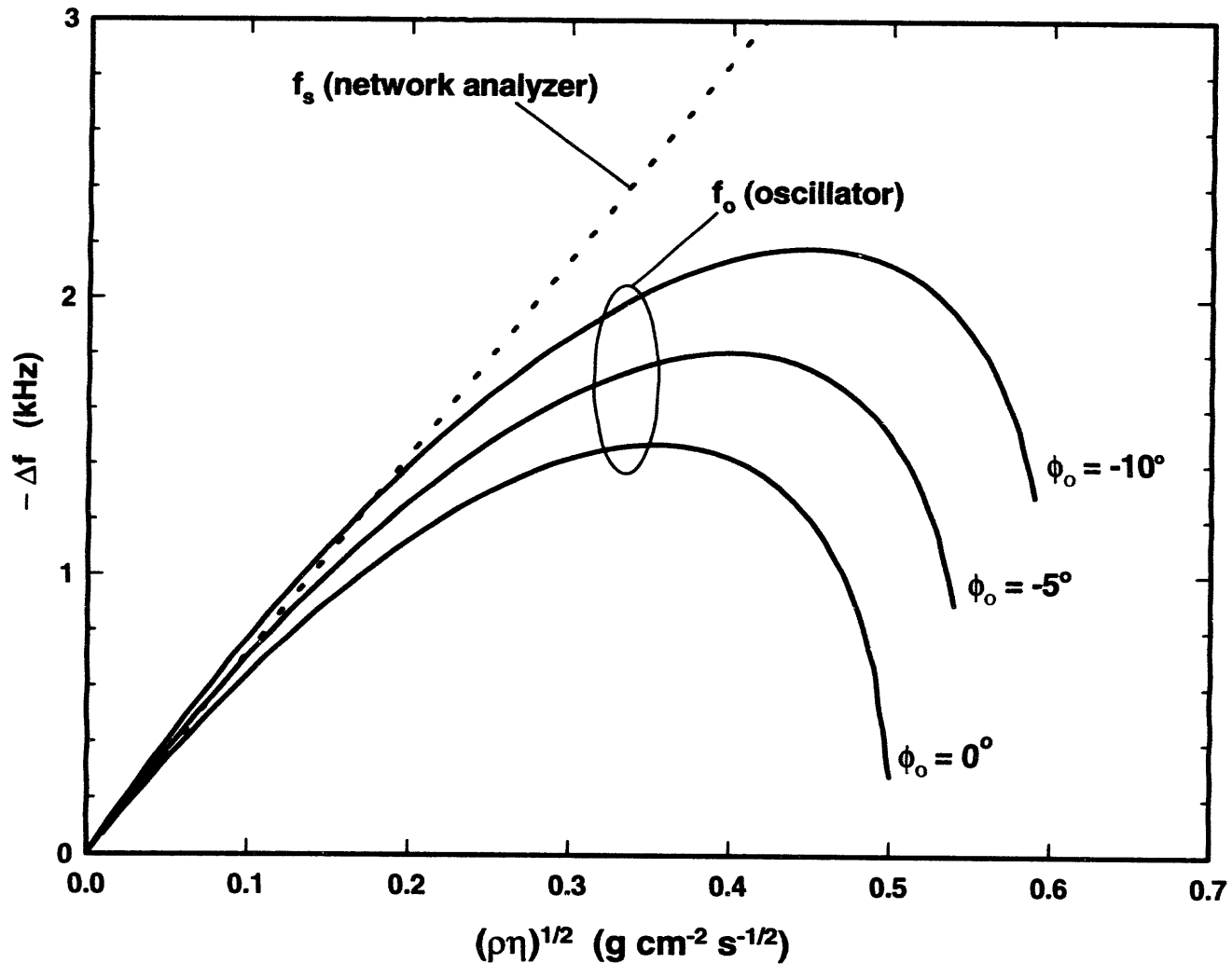


Fig. 2. Oscillator frequency shifts (solid lines) calculated vs.  $(\rho\eta)^{1/2}$ , for several values of the impedance phase angle ( $C_p = 5$  pF). The dashed line shows the variation in the series resonant frequency  $f_s$  measured with a network analyzer.

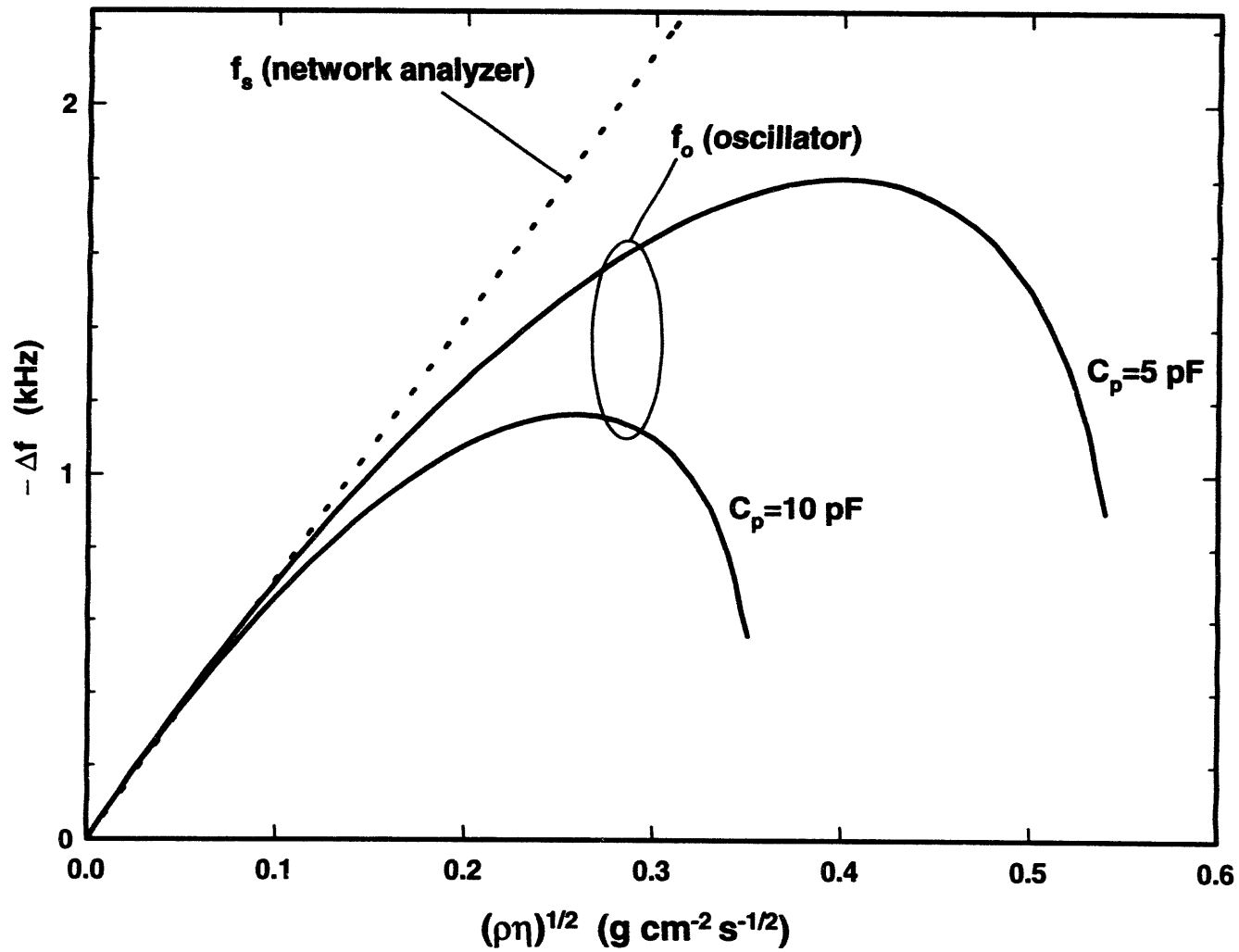


Fig. 3. Oscillator frequency shifts calculated vs.  $(\rho\eta)^{1/2}$ , for several values of parasitic capacitance  $C_p$  ( $\phi_o = -5^\circ$ ). The dashed line shows the variation in the series resonant frequency  $f_s$  measured with a network analyzer.

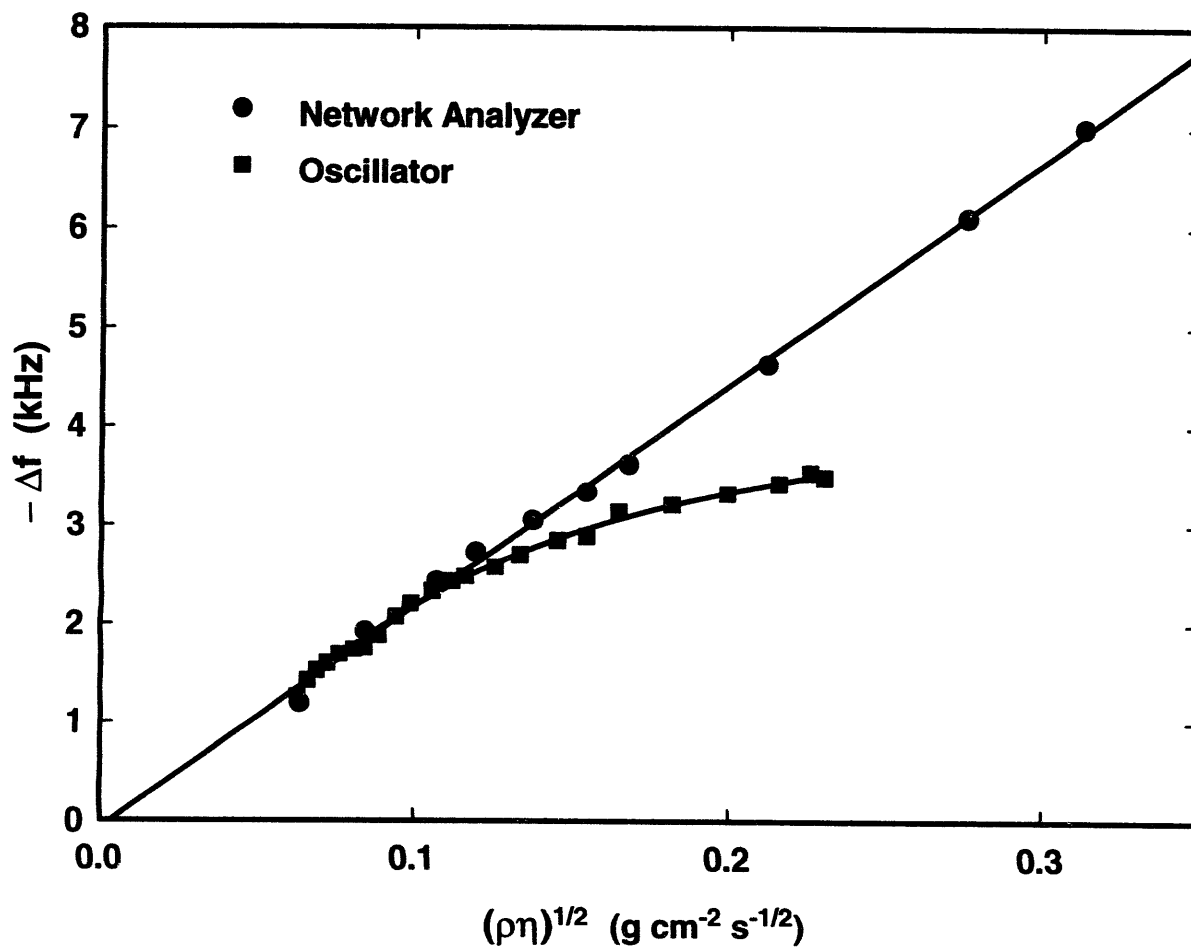


Fig. 4. Measured changes in series resonant frequency (network analyzer) and oscillation frequency (oscillator) vs  $(\rho\eta)^{1/2}$  of n-butanol as temperature was varied.

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## ABSTRACT FORM

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The change in resonant frequency of a thickness-shear mode resonator under liquid-loading conditions is compared with changes in oscillation frequency of a resonator/oscillator combination. For low values of liquid loading, oscillation frequency tracks crystal resonant frequency; as liquid loading increases, oscillation frequency deviates from the resonant frequency. The changes in resonant and oscillation frequency are determined from an equivalent-circuit model that describes the electrical characteristics of the liquid-loaded resonator.

This work was performed at Sandia National Laboratories, supported by the U.S. Department of Energy under contract No. DE-AC04-94AL85000.

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