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Piezoelectric-excited membrane for liquids viscosity and mass density measurement

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ABSTRACT

This paper presents a piezoelectric-excited membrane device for rapid measurement of liquids viscosity and density. The working principle of the device is based on membrane's resonant frequency and Q factor responses to the damping effects of a surrounding liquid. The dependences of the resonant frequency and Q factor on liquids viscosity and mass density were theoretically investigated using a sphere-end oscillator model in viscous liquids and compared to experimental results. The theory and experimental results show that the piezoelectric-excited membrane can be used to measure liquids viscosity in a range from 19.88 cP to 1733 cP and mass density in a range from 0.829 g/cm^3 to 0.886 g/cm^3. Hence, the piezoelectric-excited membrane device is a promising candidate for rapid measurement of liquids viscosity and mass density, especially for wide-range viscosity measurement.

1. Introduction

Rapid measurement and real-time monitoring of liquids viscosity and mass density are of substantial importance and essentially affect the designs of production and transportation in various industries of the modern world [1–4]. In the petroleum industry, viscosity and density measurement is conducted to monitor the oil quality and to determine the design of pipeline elements [5,6]. In the food industry, the texture of liquid foods is maintained by controlling the viscosity and density during production, which provides high production efficiency and cost effectiveness [7–9]. In the healthcare industry, treatment of certain vascular diseases requires the control of blood fluidity, which is achieved by monitoring blood viscosity [10–12]. Hence, there is an increasing demand for low-cost devices that can measure liquids viscosity and density rapidly and can be easily integrated into target industrial systems.

Conventional viscometers, such as capillary tube viscometers and rotating viscometers, require a relatively large amount of samples and a long measurement time, which means those devices are not suitable for rapid testing. For in-field monitoring applications, various resonant viscosity sensors have been reported since they are small, responsive, inexpensive, and can be installed as immersive units. The working principle is based on changes in their vibrational behavior that are induced by damping forces form the surrounding liquid medium. Typically, changes in the resonant frequency and quality factor (Q factor) are considered as signals.

For example, thickness-shear mode resonators, such as quartz crystal microbalance (QCM) and flexural vibration oscillators, such as cantilever sensors, have been extensively investigated to realize in-field monitoring of liquids viscosity and density, especially for small-volume liquid samples [13–19]. QCM is the simplest and most commonly used acoustic viscosity sensor, which is usually made of a thin quartz disk sandwiched between two electrodes and operates in a shear mode. When a liquid is in contact with the disk, a thin layer of the liquid is viscously entrained with the shear movement, leading to changes in the resonance frequency and Q factor of the oscillating quartz crystal. However, typical lateral displacement of QCM operating in the thickness-shear mode is about 1–2 nm and its high frequency shear waves quickly dissipate in a 0.25 μm penetration depth in liquids [20]. Especially for nonlinear liquids containing polymers or higher molecular weight surfactants, QCM is out of operation because the lateral vibration displacement is much smaller than the chain length of the polymers or surfactants [21,22]. Cantilever sensors working in bending modes normally have lower resonant frequencies and larger displacements than thickness-shear mode devices and are considered more suitable for measuring viscosity of nonlinear liquids. Cantilever sensors have been well studied in viscosity and density measurements [23–32]. For instance, viscosity measurements of engine oils, silicone oils and glycerol solutions with cantilevers have been reported. In

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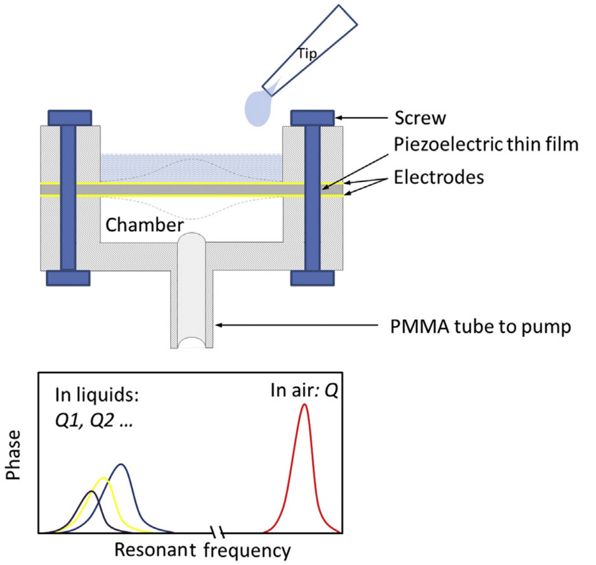

Fig. 1. Schematic representation of piezoelectric-excited membrane used for liquids viscosity and density measurement.

addition, many research studies have been demonstrated to explain the relationship between resonant responses of cantilevers and liquid properties. However, viscous damping forces in liquids cause energy losses of vibrating cantilevers, which significantly reduces the measurement resolution of cantilevers. Furthermore, in high viscosity liquids, the energy loss deteriorates the flexural vibration displacement, and no resonant frequency and associated Q factor can be measured [33]. As a result, the usability of cantilever sensors is limited to low viscosity and mass density ranges.

In this paper, a piezoelectric-excited membrane with a fully clamped boundary was presented for liquids viscosity and density measurement. Similar to the aforementioned resonant sensors, the resonant frequency and Q factor of the membrane are affected by the viscosity and mass density of liquid environments. Membrane devices in different sizes were made from Polyvinylidene Fluoride (PVDF) piezoelectric thin film and tested in liquids with various viscosities and densities. The piezoelectric-excited membrane was found to be operational in a wide viscosity range from 19.88 cP to 1733 cP and a mass density range from 0.829 g/cm^3 to 0.886 g/cm^3.

2. Theory

The membrane used in this work is composed of three layers: a top electrode, a piezoelectric thin film and a bottom electrode. And the boundary of the membrane is fully clamped. Underneath the membrane is a small chamber that is connected to a vacuum system to induce a stress in the membrane. The top surface of the membrane is in contact with a tested liquid. The geometry and operational principle of the membrane device are given in Fig. 1.

As discussed in our previous work, the stress induced by the pressure difference between the top and bottom sides of the thin film structure makes it easier to actuate the vibration of the membrane and improves the signal-to-noise ratio as well as the Q factor[34]. Due to the fully clamped boundary condition, application of an alternating electrical signal induces contraction and expansion of the piezoelectric layer, thereby making the membrane vibrate in a flexural mode. The resonant frequency and Q factor of the membrane can be expressed as [35]:

$$f_{ij} = \frac{\alpha_{ij} 2a}{2}\sqrt{\frac{T}{\rho h}} = \frac{\alpha_{ij} 2}{2}\sqrt{\frac{T}{M}} \tag{1}$$

$$Q = \frac{f_{ij}}{FWHM} \tag{2}$$

where α_{ij} is the dimensionless frequency parameter of the membrane, ρis the density of the membrane, T is the stress in the membrane, a and h are the size and thickness of the membrane, M is the mass of the membrane, $FWHM$ is the band width of resonant phase peak, respectively.

To well describe the resonant behavior of the membrane in liquids, a sphere-end (radius R) oscillator model in viscous liquids was used to correlate the resonant frequency and Q factor of the membrane to the viscosity and mass density of liquid environments. In this theoretical model, the resonant function of a membrane can be expressed as [36]:

$$\frac{d^2\tau}{dt^2} + \mu\frac{d\tau}{dt} + 2\pi f_{ij} 2\,\tau = \frac{F_0 e^{i2\pi f_{ij} t}}{m'} \tag{3}$$

where τ is a function of time t, μ is the damping factor, m’ is the generalized mass of the membrane, $F_0 e^{i2\pi f_{ij} t}$ is the driving force from the applied electric field and reversed piezoelectric effect.

The drag force in liquids can be approximated by considering that a fixed determinable volume of liquids will be carried with the membrane during its vibration. The drag force is given by [37]:

$$P_l = \frac{2}{3}\pi\rho_l R^3 \frac{d^2\tau}{dt^2} + 3\pi R^2\sqrt{2\eta_l\rho_l 2\pi f_{ij}}\frac{d\tau}{dt} \tag{4}$$

where the η_l and ρ_l are the viscosity and density of the liquid environment, respectively.

When the membrane is operated in a liquid environment, the total drag force could be expressed as:

$$P = F_0 e^{i2\pi f_{ij} t} - \left(\frac{2}{3}\pi\rho_l R^3 \frac{d^2\tau}{dt^2} + 3\pi R^2\sqrt{2\eta_l\rho_l 2\pi f_{ij}}\frac{d\tau}{dt}\right) \tag{5}$$

Substituting Eq. (5) into Eq. (3) and making the function in the same format:

$$\frac{d^2\tau}{dt^2} + \mu'\frac{d\tau}{dt} + 2\pi f_{ij}{}' 2\,\tau = \frac{F_0 e^{i2\pi f_{ij} t}}{M'} \tag{6}$$

where $M' = m' + \frac{2}{3}\pi\rho_l R^3$, $\mu' = \frac{\left(m'\mu + 3\pi R^2\sqrt{2\pi\eta_l\rho_l f_{ij}}\right)}{M'}$ and $f'_{ij} = f_{ij}\sqrt{\frac{m'}{M'}}$ are considered as the effective mass, effective damping factor and membrane’s resonant frequency in the liquid, respectively.

It is easy to calculate the resonance frequency from the above discussions as:

$$f'_{ij} = \frac{\alpha_{ij}}{2}\sqrt{\frac{T}{M + \frac{2\pi\rho_l R^3 M}{3m'}}} \tag{7}$$

Written in the same format as Eqs. (1) and (7) can be expressed as:

$$f'_{ij} = \frac{\alpha_{ij}}{2}\sqrt{\frac{T}{M + M^*}} \tag{8}$$

where $M + M^*$ is the symbolic mass of the membrane in the liquid and $M^* = \frac{2\pi\rho_l R^3 M}{3m'}$ is induced by the liquid mass loading.

From Eq. (7) and Eq. (8), the resonant frequency is a function of the liquid density and can be expressed in a simplified form as:

$$\frac{1}{f_{ij}{}'^2} = a + b\rho_l \tag{9}$$

where a and b are constants.

For a mechanical resonator, the Q factor is given by [35]:

$$Q_{liquid} = \frac{2\pi M' f'_{ij}}{\mu'} \tag{10}$$

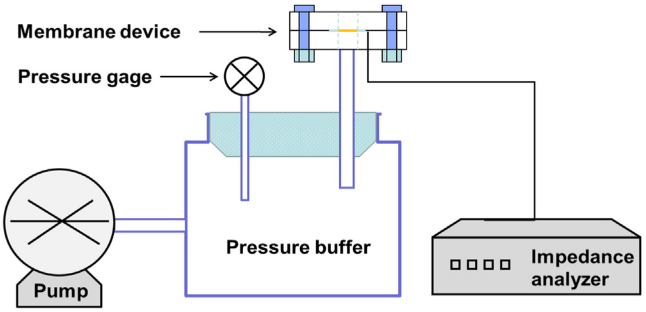

Fig. 2. System for liquids viscosity and density measurement using piezoelectric-excited membrane.

The Q factor is a function of both the density and viscosity of the liquid and can be expressed in a simplified form as:

$$\frac{1}{Q} = c + d(\eta_l\rho_l)^{1/2} \tag{11}$$

where c and d are constants.From Eq. (9), it can be seen that the inverse of the squared resonance frequency ($1/{f_{ij}'}^2$) is proportional to the mass density (ρ_l) of the liquid. In addition, from Eq. (11), the inverse of the Q factor ($1/Q$) is proportional to the square root of the viscosity and density product ($\sqrt{\eta_l\rho_l}$) of the liquid. Hence, the viscosity and density can be obtained by measuring responses of the resonant frequency and Q factor of the vibrating membrane in the liquid of interest.

3. Experiment

Membranes with various sizes (2 mm, 3 mm and 4 mm) were fabricated as the follow process, starting with 30-μm thick piezoelectric PVDF thin film. The PVDF film was covered with a hard steel mask with patterned holes through which 100 nm gold was deposited by sputtering to form the top and bottom electrodes. Then the membranes were firmly clamped using a PMMA frame with epoxy and screws. The schematic representation of the measurement system is given in Fig. 2.

The experimental setup consists of a vacuum system (including a pump and pressure buffer chamber) and an impedance analyzer (Agilent, HP 4294A). The pump and the pressure buffer chamber are used to induce a stable pressure difference (650 Torr) between the top and bottom sides of the membranes, which induces a stress in the membrane acting as a restore force of vibration. Liquids with different densities and viscosities were added to the top surface of the membrane by using a micro pipette. Phase and impedance signals of the membrane were recorded using an impedance analyzer HP4294A. The resonance frequency, f_{ij}, of the membrane was determined as the phase peak frequency, and the Q factor was calculated by $f_{ij}/FWHM$.

Reference oils from the SavanTech were used in the experiments. The viscosity and density of those oils are shown in Table 1.

4. Results and discussion

Typical phase spectra of a 3-mm membrane in air and in liquid (R-100 oil) are given in Fig. 3.

Table 1
Viscosity and density of the oils used in the experiments.

Liquids	R-100	R-300	R-2350	R-2450	R-600
Viscosity (cP)	19.88	61.94	152.51	372.66	1733
Density (g/cm^3)	0.829	0.853	0.861	0.870	0.886

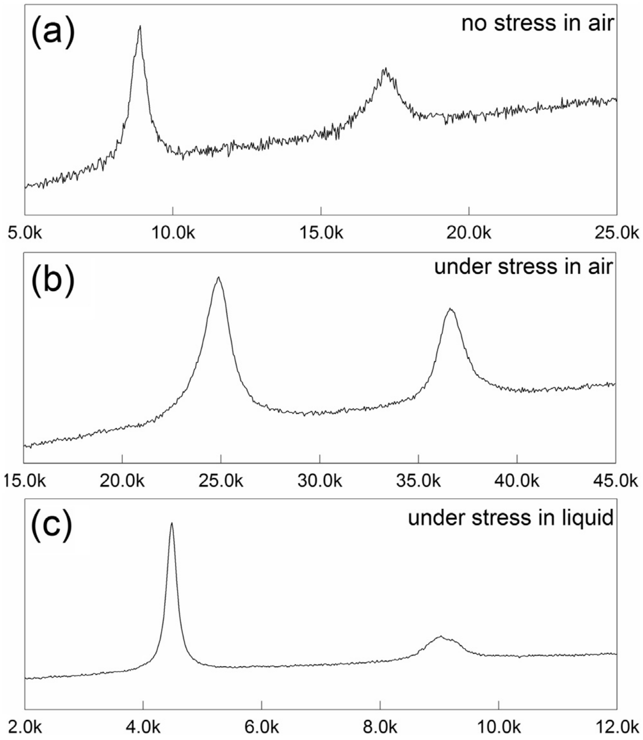

Fig. 3. Resonance behaviors of a 3 mm membrane (a) no stress in air, (b) under stress in air and (c) under stress in liquid (R-100).

From Fig. 3, it is clear that the resonances of the membrane occur in the range of kilohertz. The first and second resonant peaks can be found in these spectra. Frequencies and Q factors of each peak are given in Table 2.

From Fig. 3(a), it is clear that when the membrane is under zero stress, the phase spectrum looks noisy, and the resonance is quite weak resulting in a low signal-to-noise ratio. The frequencies of the first and second peaks are 8794 Hz and 17,198 Hz, and the Q factors are 10.2 and 13.8, respectively. From Fig. 3(b), when there is a stress induced by the pressure difference, the resonant frequencies shift to a higher range, and the signal-to-noise ratio improves. The frequencies of the first and second peaks are 24,888 Hz and 36,622 Hz, and the Q factors are 13.4 and 20.5, respectively. As discussed in our previous work, the stress induced by the pressure difference actuates the flexural mode vibration more easily, and thus, the signal-to-noise ratio can be improved. It also makes the device work in a higher frequency range and improves the Q factor. From Fig. 3(c), when the membrane is immersed in a liquid, the resonant frequencies are 4473 Hz and 9070 Hz, which are much smaller than those measured in air. This can be attributed to the mass load of the liquid as expressed in Eq. (8). It can also be found that the Q factor of the first peak in liquid (21.8) is much higher than that in air (13.4). From Eq. (10), the Q factor of the membrane in liquid is proportional to M'/μ'. Since the effective mass is much more significant than that on effective damping factor, the Q factor in liquid increased. This is an advantage of the membrane device

Table 2
Resonant frequencies and Q factors of 3 mm membrane under different conditions.

Peak	No stress in air		Under stress in air		Under stress in liquid (R-100)	
	Frequency	Q	Frequency	Q	Frequency	Q
1	8794	10.2	24,888	13.4	4473	21.8
2	17,198	13.8	36,622	20.5	9070	15.9

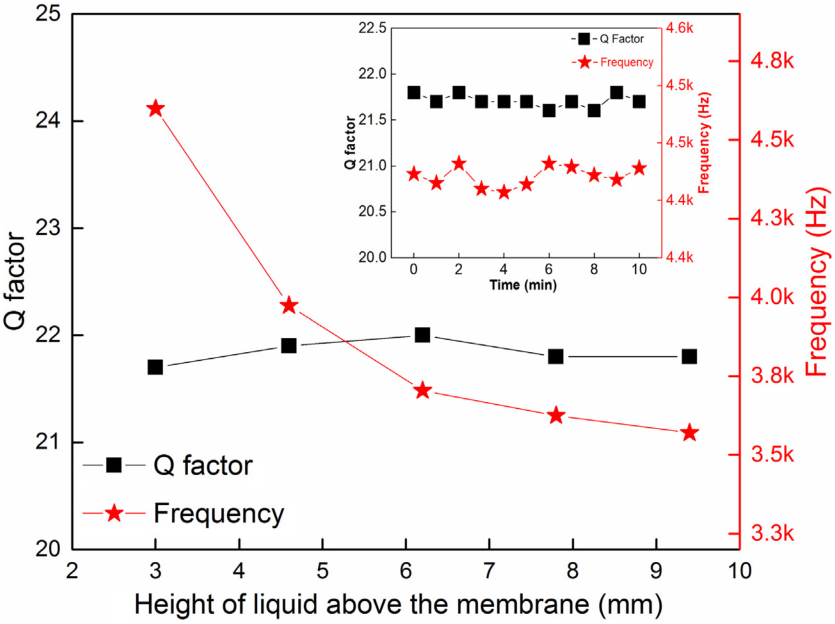

Fig. 4. The resonant frequencies and Q factors of a 3 mm membrane in R-100 of different heights above and the stability testing in 3 mm R-100 (the insert figure).

over conventional cantilever sensors for liquids viscosity and density measurement. The Q factor of the second peak in liquid (15.9) is smaller than that in air (20.5), which can be explained by the asymmetrical vibration of the membrane [38]. In cantilever-based sensing, only a thin layer of liquid is entrained on the cantilever surface and its Q factor is mainly affected by the effective damping factor, which often results in unmeasurable signals in liquid as mentioned earlier.

In consideration of the significant influence of the mass load on the resonance frequency, the amount of liquids on the membrane should be precisely adjusted in the experiments. The stability of the resonance frequency and Q factor signals is quite important for the real applications. To settle down the discussions above, the resonant frequency and Q factor of a 3 mm membrane were tested in R-100 oil with 3 mm in height for 10 min. The data were recorded every minute. After that, the height was changed by adding additional R-100 oil on top of the membrane using a micro pipette and data were recorded subsequently. The resonant frequencies and Q factors with different heights of R-100 oil on top of the membrane are shown in Fig. 4.

It can be seen that when the height of R-100 oil increases from 3 mm to 10 mm, the Q factor stays in the range of 21.7–22.0, which indicates that the Q factor is not very sensitive to the liquid height (hence, liquid weight) in the testing range and mainly influenced by the viscosity. By contrast, the resonant frequency of the membrane clearly decreases from 4475 Hz to 3570 Hz due to the increased

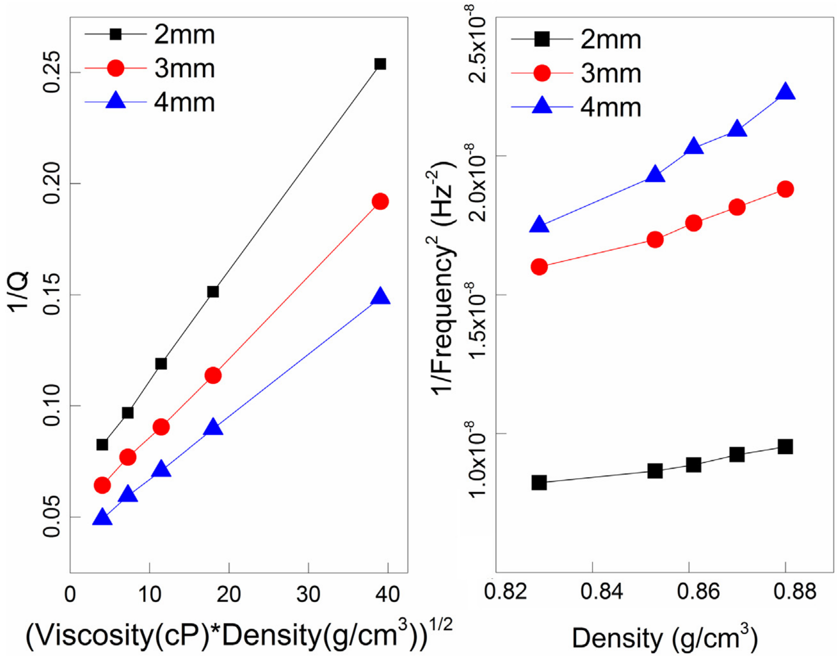

Fig. 5. Resonant frequencies and Q factors of membranes with various sizes (2 mm, 3 mm and 4 mm) tested in liquids of different viscosities and densities.

mass load of the liquid as discussed in Eq. (8). As shown in the inset of Fig. 4, the resonant frequency stays between 4457 Hz to 4482 Hz, and the Q factor stays between 21.6 and 21.8, which means that time-dependent stability of the membrane device has been demonstrated. The Q factor and resonant frequency data at 0 min in the inset of Fig. 4 also indicate that the response is instantaneous after the addition of a liquid to the membrane. This can be attributed to the fact that damping of the membrane in liquid environment occurs instantaneously. Hence, the membrane sensor can be used for rapid measurement and real-time monitoring of liquids viscosity and mass density.

Membranes with various sizes (2 mm, 3 mm and 4 mm) were tested in different liquids with 3 mm in height. The volumes of liquid samples needed in the testing were only about 0.012 ml, 0.027 ml and 0.048 ml for the 2 mm, 3 mm and 4 mm membranes, respectively, which indicates that only very small volumes of liquid samples are needed for viscosity and density measurement using our membrane device.

The resonant frequencies and Q factors of membranes with different sizes (2 mm, 3 mm and 4 mm) tested in liquids of different viscosities and densities are shown in Fig. 5.

From Fig. 5, it is clear that the inverse of the Q factor ($1/Q$) increases linearly with the square root of liquids viscosity and density product ($\sqrt{\eta_l \rho_l}$), which is in good agreement with the theoretical analyses discussed in Eq. (11). It indicates that the membrane device can measure the liquid viscosity in a range from 19.88 cP to 1733 cP. In addition, the nearly linear dependency between the inverse of the squared resonant frequency ($1/f_{ij}{'}^2$) and liquid density (ρ_l) in Fig. 5 agrees with the theory too. It also indicates that the membrane device can measure the liquid mass density even in a narrow range from 0.829 g/cm^3 to 0.886 g/cm^3.

5. Conclusions

In this paper, a piezoelectric-excited membrane liquids viscosity and density sensor is designed, theoretically investigated and the feasibility is demonstrated by experiments. It was found that the inverse of the Q factor ($1/Q$) of the membrane is linearly proportional to $(\eta_l \rho_l)^{1/2}$ and the inverse of the squared resonant frequency ($1/f_{ij}{'}^2$) is linearly proportional to ρ_l in a viscosity range from 19.88 cP to 1733 cP and in a density range from 0.829 g/cm^3 to 0.886 g/cm^3. In conclusions of the theoretical analyses and experimental results, the piezoelectric-excited membrane can be used for liquids viscosity and mass density measurement, especially for wide-range viscosity measurement.

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