Optimal Vessel Materials for Indirect-contact Ultrasound Measurements

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Ultrasound can be used to detect small impurities or bubbles in opaque fluids, such as liquid metals, and measure fluid velocity based on the Doppler Effect. Ultrasound transducers, however, cannot always tolerate direct contact with the test fluid. When performing indirect-contact ultrasound measurements, a suitable vessel material is a prerequisite for good acoustic coupling and thus accurate flow measurement. Here, we present an experimental study focusing on finding the optimal vessel materials for applying indirect-contact ultrasound measurement in liquid gallium. We investigate the effects of the type, thickness, and wettability of vessel materials on ultrasound measurements. Our results suggest that the intensity transmission coefficient alone cannot predict the sound transmission behaviors accurately. Particularly in gallium, wetting plays an important role. Velocity measurements are less sensitive to the choice of vessel material than echo intensity measurements.

Keywords: Ultrasound Doppler velocimetry, Liquid metals, Acoustic coupling, Wetting, Indirect-contact ultrasound measurement

1. Introduction

Ultrasound is a powerful technique for studying the flow in a variety of fluids, especially in liquid metals [1-5]. In the past thirty years, ultrasound Doppler velocimetry (UDV) has been widely applied to industrially-motivated studies, such as measuring flow structures of metal melts in casting molds [6] and liquid metal batteries [7]. Besides measuring the flow velocity, ultrasound is also used to detect bubbles, impurities, and solid/liquid interfaces [8,9].

The working principles of ultrasound detection and ultrasound Doppler velocimetry have been introduced in detail in previous works [1,10]. As the UDV is not fully contactless, to transmit the sound waves from the ultrasound transducer into the test fluid, a continuous acoustic path is required [3]. Therefore, ultrasound transducers are usually inserted into the test fluid directly; we call this direct-contact measurement. However, the ultrasound transducer cannot always tolerate direct contact with the test fluid due to some restrictions of the experimental setup. Especially when the test fluid is metal melt, particular challenges are presented. First, the high temperature and corrosion of metal melts could damage transducers or destroy the piezoelectric materials. Second, as a foreign substance, the transducer itself might contaminate the metal melt. Third, since most of the commercially available UDV transducers are designed for water-based fluids, their acoustic coupling to liquid metals is often poor. In fact, most ultrasound studies of liquid metals were conducted using transducers designed for water. Acoustic waveguides provide one solution, but they reduce the signal quality significantly [3], and the restrictions of experimental setup persist. An alternative is to place the ultrasound transducer outside the vessel of the test fluid; we call this indirect-contact ultrasound measurement

Indirect-contact ultrasound measurement has been widely used for studying flows in many kinds of fluids, especially in liquid metals. Unlike with direct-contact measurement, the ultrasound waves must pass through an additional layer, the vessel wall, in the indirect-contact measurement. Obviously, the existence of the wall would affect the ultrasound measurements. In previous studies, a variety of materials have been used for the vessel. However, we are unaware of any prior publication that systematically studied the effect of different vessel wall materials on sound transmission and thus ultrasound measurement quality, either in water or in liquid metal.

In the indirect-contact measurement, an acoustic coupling medium should be used for achieving a continuous, lowloss acoustic path between the transducer and the vessel wall. The existence of vessel wall will induce two extra interfaces in the ultrasound path: the acoustic coupling medium/vessel wall interface and the vessel wall/test fluid interface. The acoustic coupling condition at each interface depends on the acoustic impedance $Z = c\rho$, where c is the sound speed and ρ is the density. Usually, good acoustic coupling occurs when the acoustic impedance mismatch is small. The effects of impedance mismatch on ultrasound transmission through two interfaces is expressed by [11]:

$$T_{I} = \frac{4}{2 + \left(\frac{Z_{3}}{Z_{1}} + \frac{Z_{1}}{Z_{3}}\right) \times \cos^{2}\frac{2\pi f L}{c_{2}} + \left(\frac{Z_{1}Z_{3}}{Z_{2}^{2}} + \frac{Z_{2}^{2}}{Z_{1}Z_{3}}\right) \times \sin^{2}\frac{2\pi f L}{c_{2}}}$$
(1)

where T_i is the intensity transmission coefficient, Z_i are the acoustic impedances (*i*=1: acoustic coupling medium, *i*=2: vessel material, *i*=3: test fluid), c_2 is the sound speed in the vessel material, and *L* is the thickness of vessel wall.

In Eq. (1), when *L* equals an integer multiple number of half-wavelengths, ($L = (n/2)\lambda$), the Z_2 term is eliminated, so that the vessel wall is predicted to be irrelevant. In this case, the transmission strength matches that of a sound wave transmitted into the test fluid directly. Interestingly, when *L* equals an odd number of quarter wavelengths ($L = (2n - 1)\lambda/4$) and $Z_2 = \sqrt{Z_1Z_3}$, then the T_I equals 100%. Thus, a material whose acoustic impedance equals the geometric mean of the impedances of the acoustic coupling medium and test fluid is predicted

to maximize the acoustic transmission. A wall made from such a material is called a matching layer [11].

Note that the Eq. (1) was originally developed for liquid phases. When an acoustic wave is normally incident on an interface, many solids obey the same equation [11], but other factors should be considered. For example, wetting becomes a key factor that determines the continuity of the acoustic path between the vessel and test fluid. This is especially true when the test fluid is a liquid metal, because metals have unusually high surface tension. The high surface tension tends to prevent liquid metal from wetting the vessel wall thoroughly, which would cause some air pockets left between the liquid metal and vessel wall. As the acoustic impedance mismatch between gas and liquid is huge, the air gaps will reflect ultrasound waves strongly. Therefore, if poor wetting forms air gaps, even small ones, ultrasound transmission is severely impeded.

We experimentally studied the effects of the vessel materials on ultrasound measurements in liquid gallium. To better understand the ultrasound transmission through a wall, the influences of wall thickness and wetting were investigated. Our results show that in practice, Eq. (1) is not accurate enough to predict which vessel material is optimal. Going beyond Eq. (1), we used contact angles to evaluate the wetting condition, which plays an important role in the ultrasound measurements.

2. Experimental methods

To investigate the effect of vessel material, we used test plates of various materials to perform indirect-contact ultrasound measurements. Figure 1 shows a schematic diagram of the experimental apparatus. A slot near the front wall of the container allowed test plates to be inserted. The apparatus also allowed measurements without any test plate in place, which are used for reference. Two ultrasound transducers with working frequency of 8 MHz (Signal Processing, Switzerland) were placed on the two opposite walls of the container and fixed by swage fittings. The transducer placed on the front wall was connected to a DOP3010 Velocimeter (Signal Processing, Switzerland) and operated in emit/receive mode for data acquisition. During experiments, this transducer was inserted into the acoustic coupling medium until gently touching the test plate. For each test, 1000 UDV profiles were recorded, and the averaged values were used as the final data for echo intensity and flow velocity, respectively. In all UDV measurements, a time gate compensation (TGC) with a uniform magnitude was applied to compensate the sound attenuation in fluid. The second transducer placed on the container's back wall was connected to an oscilloscope (Teledyne LeCroy, U.S.A.) and served as a hydrophone to measure the sound pressure at the backwall position. The pressure measurements were stored and displayed on the oscilloscope.

We used liquid gallium $(60^{\circ}C)$ as the test fluid and deionized water as the acoustic coupling medium. Flow was driven by a rotating magnetic field generated by a stir plate beneath the container. To produce the similar flow

for all tests, both the position of the container on the stir plate and the rotating speed were kept constant.



Figure 1: Experimental setup. The slot near the front wall of the container positions the test plate and rubber sealing layers. Transducer-1 was located on the front wall of the container and connected to the ultrasound velocimeter. Transducer-2 was located on the back wall of the container and connected to an oscilloscope.

The test plate materials we selected for liquid gallium are listed in table 1. Those materials were selected as they have been widely used as the vessel material in previous studies of gallium [5,6,9,12,13]. The thickness of each plate is listed in table 1. The thicknesses of selected test plate materials are near-integer multiples of half-wavelengths, except for nylon. The thickness of the nylon plate was chosen to be an odd number of quarter-wavelengths, since nylon has approximately the right acoustic impedance to be the matching layer.

Table 1: Test plate materials selected for liquid gallium

| Vessel Materials | Thickness/ half- wavelength | Intensity transmission coefficients | Contact angle |
|--------------------|-----------------------------------|---|-------------------------|
| Acrylic | 1.48 | 30.27% | $97^{o}\pm4.20$ |
| Nylon 6/6 | 2.23 | 95.81% | $130^{o}\pm2.92$ |
| Borosilicate glass | 4.71 | 45.23% | $109^{\text{o}}\pm8.06$ |
| Copper | 0.49 | 29.38% | $124^{o}\pm7.46$ |
| Steel | 0.52 | 27.85% | $121^{o}\pm4.71$ |

As mentioned above, the wetting condition becomes important when ultrasound waves pass through a solidliquid interface. Contact angles are usually used as the primary data in wettability studies, which indicate the degree of wetting between a liquid and solid [14]. In this study, to investigate the influence of wetting, the contact angles between each test plate material and gallium were measured by static sessile drop method [15] with a goniometer (AST Products Inc., U.S.A. For each test plate, the contact angle measurement was repeated five times; the average values are listed in table 1.

We also calculated the theoretical acoustic intensity transmission coefficients for each test material in gallium, by Eq. (1), and the results are listed in table 1 as well. Since the UDV transducer is designed for water, we assumed a 100% sound transmission rate from the transducer to the acoustic coupling water.

3. Results and discussion

Figure 2 shows the time-averaged echo intensity measured in liquid gallium with different test plate materials. The echo intensity of direct-contact measurement, without any test plate, is shown for reference. All selected test plate materials allowed detection of the loud echo from the back wall of the container, evident as a large peak at about 100 mm in the figure. In addition to detecting large interfaces, ultrasound has the potential to detect echoes reflected from small particles or bubbles suspended in the bulk of the fluid, which is also necessary for measuring fluid velocity. The acrylic test plate transmitted the strongest bulk echo signals, even stronger than the direct contact. Comparing with Table 1, though nylon is predicted to have the highest transmission coefficient, figure 2b shows that the nylon test plate did not transmit signals as strongly as the acrylic test plate. This discrepancy can be explained by wetting. The measured contact angle between the transducer surface material (Epotek epoxy) and liquid gallium is 137°, which means that wetting in direct-contact measurement is poor. The contact angle between nylon and gallium is similar (130°). So with nylon, wetting is similarly poor and the measured echoes have the similar strength as the direct contact, as shown in figure 2b. Acrylic, however, wets gallium much better, as indicated by its lower contact angle (97°). Thus, wetting, which determines the continuity of the acoustic path, seems to explain why indirect measurement through acrylic produces stronger echoes than the direct measurement.



Figure 2: Time-averaged echo intensity measured in gallium with different test plate materials. Results suggest that acrylic has a better performance in terms of sound transmission into liquid gallium than other materials, even better than the direct-contact measurement.

Figure 2 also shows that all test plate materials have induced strong artificial echoes near the transducer surface (0 mm). Those echoes are caused by the acoustic impedance mismatch among acoustic coupling medium (transducer surface), test plate, and test fluid. We refer to these strong echoes as front-wall noises. However, under the same TGC parameter, the front-wall noises are weak and mask bulk echo measurements for only about 5~10 mm when the test plate is plastic; when it is metal, frontwall noises are stronger and mask nearly 30 mm, as shown in figure 2 d-e. Those noises seriously affect the ultrasound measurement. As shown in figure 2e, the steel plate also produces many undesirable artificial peaks appearing in the bulk part of the curve. Those artificial peaks are explained by reverberation artifacts that occur when sound waves reflect repeatedly within the metal plates. Many reflections can occur because the metals have small acoustic damping coefficients, so sound waves are attenuated little as they traverse the metal.

Figure 3 shows the transient sound pressure measured by the transducer located at the back wall. Comparing with the reference (direct contact) pressure curve, more noises appeared in sound pressure curves (at times greater than 2 ms) of glass and metal test plates. In addition, figure 3e clearly shows that the sound pressure is weakened by the steel test plate. Interestingly, for the borosilicate glass test plate, although the echo profile shows strong artificial noises near the transducer surface (figure 2c), the measured bulk echo intensity and sound pressure (figure 3c) are better than that of direct contact. This may be explained by the high predicted transmission coefficient and relatively small contact angle between the borosilicate glass and liquid gallium.



Figure 3: Sound pressure measurements in gallium with different test plate materials. Each curve describes a transient pressure waveform in one pulse duration. More reverberation noises appeared within the metallic materials data.

Figure 4 shows the time-averaged flow velocity measured in liquid gallium with different test plate materials. All measured flow patterns are almost the same as for direct contact. However, the steel plate caused such strong reverberations that velocity could not be measured within 25 mm of the transducer. It seems like the velocity measurements are less sensitive to the choice of wall material than echo intensity measurements.

Considering the echo intensity and flow velocity results, as well as the machineability of the material itself, we would suggest using acrylic as the vessel material for ultrasound indirect-contact measurements in liquid gallium. Borosilicate glass could be used for hightemperature applications. If the purpose is only to detect large interfaces or measure a simple flow, stainless steel might also be a choice. However, copper is not recommended, since a slow reaction with gallium has been observed in our experiments. Our measurements suggest that wetting plays an important role in experiments with liquid gallium. During the experiment, we also observed that the gallium oxide layer formed at the gallium-vessel interface is likely to improve the wetting. However, this oxide layer would also intensify front noises and degrade ultrasound signals when it became thicker [4, 5].



Figure 4: Time-averaged flow velocity measured in gallium with different test plate materials. All selected materials could allow ultrasound velocity measurements in liquid gallium, and the measured mean flow structures are close to the reference data.

To further explore the effect of wetting on ultrasound transmission, two steel plates with different surface roughness were used for ultrasound measurements in gallium. As shown in figure 5, the contact angle between gallium and smooth steel is 121°, small enough that the gallium droplet spreads across the surface somewhat. With roughened steel, however, the contact angle is so large (164°) that the gallium hardly wets the surface at all. Thus, we would expect better acoustic coupling between gallium and smooth steel than roughened steel. That expectation is confirmed by figures 5(b) and (e), which show weaker reverberation noises and a much stronger back-wall echo with smooth steel plate. Little useful information is contained in the echo signals measured through roughened steel plate. Consequently, no real velocity is measured; only noise appears in figure 5(f).



Figure 5: Wetting experiments in gallium: (a) and (d) show the measured contact angle between a gallium droplet and steel plate; (b) and (e) show the time-averaged echo intensity; (c) and (f) show the time-averaged flow velocity. Gray curves are obtained from direct contact measurement and are used as reference. Roughened steel has a large contact angle with gallium and caused poor ultrasound measurements.

4. Summary

By carefully choosing the vessel material, indirect-contact ultrasound measurements can achieve the same or better measurements quality than direct contact. In this work, we experimentally studied the effect of vessel wall material on ultrasound transmission performance when using indirectcontact ultrasound measurements in liquid gallium. Through our study, we found that the calculated intensity transmission coefficients alone cannot predict the sound transmission precisely. In real measurements, especially when the test fluid is liquid metal, the wetting condition between vessel material and test fluid changes the acoustic coupling, as can be predicted from contact angles. For the same type of material, the surface roughness, which is one of the factors that determine its wetting properties, would affect acoustic coupling and thus ultrasound transmission. Therefore, proper surface treatments are desirable to achieve good ultrasound measurements.

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