Acoustic Chemical Sensors

Although pleased to see the potential of acoustical sensors reviewed by M. D. Ward and D. A. Buttry (*1*), we wish to address what appear to be misconceptions in their article concerning flexural Lamb wave acoustic sensors. The illustration of flexural motion shown in their figure 1 is inaccurate. The lowest order antisymmetric Lamb wave (*2*) propagates in the plane of the film rather like the ripples in a flag waving in the wind. The wavelength of the ripples is determined by the spatial period of the interdigital transducers that launch acoustic waves at one end of the plate and receive them at the other end, typically 50 wavelengths away. In our device, the Lamb waves travel in thin composite membranes of ZnO on silicon nitride (not Si) that are formed on an Si substrate. The Si is etched out underneath to leave a rectangular membrane in an Si frame.

We agree that operation at a relatively low frequency is advantageous for the reasons Ward and Buttry cite: less noise, simpler instrumentation, lower cost, and operation in viscous liquids at a frequency below the visocoelastic relaxation frequency, which permits viscous properties to be detected. The Lamb wave sensor has both the lowest operating frequency and the lowest minimum detectable mass change (MDM), as shown in their table 1. Most of the entries in that table appear to have come from a publication of our group (*3*), but the original sources do not appear to have been cited correctly by Ward and Buttry.

Equations 4 and 5 in the article by Ward and Buttry, which compare sensitivities of the shear-horizontal acoustic plate mode (SH-APM) and flexure-wave devices, lead one to conclude that the SH-APM device and the flexural-wave device can have comparable sensitivities. One should note, however, that the thickness, *b*, of the SH-APM (100 to 200 μm) is much larger than that obtainable with flexure-mode devices (3 μm down to less than 1 μm). This is because the SH-APM requires a crystalline substrate to generate the horizontal-shear motion, and so the device cannot be made extremely thin. Because of the different dimensions, the attainable sensitivity, $S_m = (\Delta f/f_0)(1/\Delta m)$, of flexure-mode devices can be much larger than that of the SH-APM sensor.

Because acoustic energy propagates throughout their full thickness, thin Lamb wave sensors respond equally to mass added to either side of the plate, as do quartz crystal microbalance and SH-APM sensors. Our sensors also have electrodes on only one face, and so have a design simplicity that Ward and Buttry attribute only to SH-APM sensors.

Finally, all the acoustic sensors considered are affected by the viscosity and density of a liquid overlay. For the Lamb wave sensor, these sensitivites have been well characterized (*4*), and the detection of interfacial mass changes in a liquid has been demonstrated electrochemically (*5*).

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Response: The representation of the flexural motion in our figure 1 (*1*) was limited by the difficulties inherent in illustrating complex motions. The figure accurately shows the direction of particle displacement during the wave propagation, but does not show it as "ripples in a flag waving in the wind." It shows one wavelength of the wave as it propagates. In this respect, it is similar to figure 2 of a previous paper from the Berkeley Sensor and Actuation Center (*2*), in which two and one-half wavelengths of this mode are graphically depicted. The description of their device as a "ZnO on Si" composite membrane derives from figure 1 in (*2*), in which the membrane is clearly shown as an "Si-based and ZnO-coated structure." In more recent papers [(*1*), references 11 and 12], the Berkeley Group appears to have changed the design to one in which the flexural waves travel in a composite membrane of ZnO on silicon nitride.

Unfortunately, the references in our table 1 were not correct. They should have been as follows: for surface acoustic wave (SAW), reference 33; for shear horizontal-acoustic plate mode (SH-APM), reference 13; for Lamb wave, references 11 and 12; and for quartz crystal microbalance (QCM), reference 14. Our use of the S_m formalism derives directly from a previous contribution by S. W. Wenzel and R. M. White, as indicated by our reference 12.

The fact that differences between S_m values for Lamb wave and SH-APM devices is related to their different thicknesses was not discussed explicitly in our article. However, our equations 4 and 5 indicate the influence of thickness, *b*, for both SH-APM and Lamb wave devices. The listings in table 1 also make clear that the S_m value for Lamb wave devices is much larger than that for SH-APM devices. However, the noise levels reported in the literature for Lamb wave devices [(*1*), reference 11] makes the value of minimum detectable mass change (MDM) found by B. J. Costello *et al.* not much better than that produced with an SH-APM. Since the MDM values can depend on the details of excitation and measurement equipment, it is possible that these values can be significantly improved.

Costello *et al.* are correct that Lamb wave devices respond equally well to mass changes on either side of the membrane (with or without an electrode). This is a design simplicity that we incorrectly attributed to SH-APM devices only, although to our knowledge this quality has not been previously discussed for Lamb wave devices.

While we did not explicitly state that Lamb wave devices are sensitive to the density and viscosity of the liquid overlayer, it was implicit in our discussion of these effects. We did state that the separation of mass and density-viscosity effects, in such a way as to allow for their separate determination, has not been conclusively demonstrated for Lamb wave devices. The literature in this area does not yet contain sufficient quantitative examples of this separation.

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