Measurement of Sound Speeds in Fluids at High Pressure from Near Freezing to Super Critical Temperatures

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Sound speeds measured as a function of pressure and temperature provide a robust path to extend ambient pressure thermodynamic properties to high pressures. Our group developed novel methods for ultrasonic measurements to 700 MPa between 250 K and 400 K and for diamond anvil measurements of sound speeds to 10 GPa and temperatures greater than 600 K. With each experiment, the challenges lie in accurate calibration of pressures and temperatures, as well as in creating reliable methods for the determination of sound speeds. In a conventional hydraulic system, two commercially available pressure transducers, continuously monitored to track temporal drift and environmental temperature variations, were calibrated against a deadweight absolute pressure system. The entire pressure vessel is heated or cooled; accurate temperature determinations require use of an RTD and several thermocouples. The sound speed measurements are based on a well characterized path length and digital signal processing of a frequency modulated source to obtain reproducible time-of-flight determinations. Average standard errors for pressure range from 0.02 MPa to 0.1 MPa at the highest pressures, 20 mK for temperature and 0.01% for sound speed determinations. In diamond anvil measurements, pressure determinations near room temperature are accurate to about 20 MPa. Pressure determinations at high temperatures are dependent on calibrations that are not fully consistent between research groups. A pressure uncertainty of about 50 MPa is commonly asserted for measurements above 400 K. Spectroscopically determined sound speeds (both Brillouin and Impulsive Stimulated Scattering) are reproducible to better than 0.2%. Associated mostly with pressure uncertainties, sound speeds for water to 9 GPa and 725 K reported by three separate groups can be correlated using a Gibbs energy representation with a root-mean-square misfit near 1%. Noteworthy is that pressure transducers are subject to temporal drift and show hysteresis with increasing and decreasing pressure cycles. Furthermore, absolute pressure calibration in the 1000 MPa range involves massive, not readily transportable, infrastructure. We suggest that sound speeds in water could serve as a drift- and hysteresis-free secondary standard for pressure.