Speed of sound measurements in helium at pressures up to 100 MPa compared with a Helmholtz energy EOS and an *ab initio* calculated virial EOS

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Helium is a noble gas with numerous applications in science and industry. For example, liquid helium is a crucial cryogen and due to its non-toxic nature and properties that differ from air, helium is a perfect gas for leak detection in high-pressure systems. Thus, sound and comprehensive knowledge of thermophysical properties with low uncertainties is desirable. However, common thermophysical property software such as REFPROP 10.0 considers an unpublished Helmholtz energy equation of state (EOS) by Ortiz-Vega et al. [1], which is the recommended EOS by NIST [2] although no detailed information on their results or uncertainty is available. A preliminary version of this EOS was published in the PhD thesis of Ortiz-Vega [3], but the results of the two EOS differ. For the fitting of the EOS from the PhD thesis, the considered speed of sound data in the gas phase was limited up to pressures of 15 MPa. In addition to empirical EOS, the EOS of helium is amenable to theoretical evaluation based on *ab initio* interatomic potential-energy functions. Gokul et al. [4] calculated acoustic virial coefficients for a virial EOS (VEOS) up to the order $n = 7$. The speed of sound calculations are in very good agreement with experimental data up to 15 MPa.

For both EOS, validation for higher pressure was lacking prior to this work. To extend the experimental database, we measured the speed of sound in helium using the dual-path pulse-echo technique along five isotherms in a temperature range from (273 to 373) K at pressures from (15 to 100) MPa with a relative expanded uncertainty ($k = 2$) from (0.02 to 0.04)%. In comparison to the Helmholtz energy EOS, deviations within the experimental uncertainty were found for all temperatures up to pressures of 60 MPa, followed by a consistent increase of negative deviations up to -0.26% at higher pressures. Remarkably, deviations from the VEOS agree within the experimental uncertainty at all investigated state points.

[1] Ortiz Vega DO, et al. unpublished 2015.
[2] Huber ML, et al. Ind Eng Chem Res 2022;61(42):15449-72.
[3] Ortiz Vega DO. PhD thesis. College Station, Texas, USA: Texas A&M University; 2013.
[4] Gokul N, et al. J Chem Eng Data 2021;66(8):3258-81.