Prediction of Transport Coefficients in Dense, Cryogenic Gases using Revised Enskog Theory

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New technological solutions are currently being developed to phase hydrogen into the society as a clean energy carrier, or as a reduction agent in zero emission metal production. For this purpose, accurate prediction of transport properties such as thermal conductivities, diffusion coefficients, viscosities and thermal diffusion coefficients is needed for hydrogen-rich mixtures.

The Enskog solutions to the Boltzmann equations have long been known to provide accurate predictions of transport properties of dilute gases. However, at elevated densities predictions have been confined to mixtures of hard spheres.

In recent developments, Barker-Henderson perturbation theory has successfully been applied to develop the Revised Enskog Theory for Mie fluids (RET-Mie), giving accurate predictions for the transport properties of real, dense gas mixtures [1].

While the RET-Mie has been shown to give accurate predictions for a range of fluids and conditions, it fails in systems with sufficiently large thermal de Broglie wavelengths, for example mixtures containing hydrogen, deuterium, helium or neon at low temperatures. For such mixtures, Feynman-Hibbs quantum corrected Mie potentials have been shown capable of accurately reproducing equilibrium properties, through the SAFT-VRQ Mie equation of state, at temperatures above 20 K.

We have developed a Revised Enskog Theory for quantum-corrected Mie fluids, using thermodynamic perturbation theory to obtain accurate values for the radial distribution function at contact. This allows us to predict the transport properties of mixtures in which quantum effects are significant, even at elevated densities.

A major advantage of the theory is that it gives a possibility of using potential parameters fitted to equilibrium properties, such as vapour-liquid equilibria, vaporisation enthalpies, etc. for prediction of transport properties.

The theory is tested by comparing theoretical predictions to simulation results obtained using a recent implementation of the Feynman-Hibbs corrected Mie potential in LAMMPS, as well as by comparing to experimental results.

References

1. Vegard G. Jervell, Øivind Wilhelmsen; Revised Enskog theory for Mie fluids: Prediction of diffusion coefficients, thermal diffusion coefficients, viscosities, and thermal conductivities. *J. Chem. Phys.* 14 June 2023; 158 (22): 224101. https://doi.org/10.1063/5.0149865