# Comment on "The nucleation behavior of supercooled water vapor in helium"

## COMMENTS

# Comment on "The nucleation behavior of supercooled water vapor in helium" [J. Chem. Phys. 117, 5647 (2002)]

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In a recent paper[1] by Peeters *et al.* new experimental data on nucleation rates of water in helium in the temperature range of 200–235 K were presented. The original data are shown in Fig. 1. Nucleation rates have been reduced with classical nucleation theory predictions as described in the original paper.[1] In this temperature range a drastic change in the nucleation process was observed at 207 K, which according to the author's suggestion was due to a transition from vapor/liquid to vapor/solid nucleation. To investigate this phenomenon more carefully some additional experiments were made in the same temperature range and using the same experimental setup as well. The new measurements did not show any sign of such transition in the indicated temperature range. To explain the discrepancy, the raw data obtained by Peeters *et al.* were closely investigated and an error was found in the calculation of the vapor fraction of water in the test gas mixture.

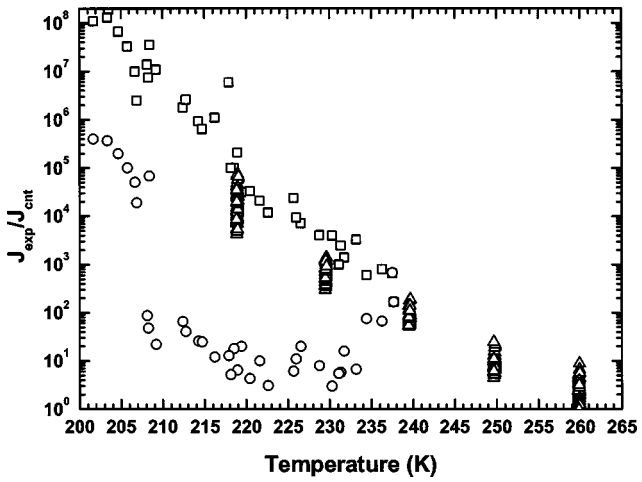


FIG. 1. Experimental nucleation rates as a function of temperature. The circles represent the original data by Peeters *et al.*, the squares mark these data after correction, and the triangles mark data obtained by Wölk and Strey. The nucleation rates are scaled with the classical nucleation theory (for vapor/liquid nucleation).

The test gas mixture originates from a combination of two different streams. One is a "wet" helium gas stream. This gas stream is saturated with water vapor by bubbling it through two containers, half-filled with water at a constant pressure and temperature. In that way, the wet gas stream has a constant vapor fraction. It can be diluted by the second gas stream, which consists of dry helium only. The gas streams are controlled by mass flow controllers (MFCs). Setting a different ratio of the flows through the MFCs alters the composition of the gas–vapor mixture.

The experimental setup allows measuring nucleation rates in a "window" between $10^{14}$ and $10^{17}$ $m^{-3}\,s^{-1}$. For different temperature regions, different initial saturations have to be chosen. This is achieved by changing the degree of dilution of the wet gas stream. The error made by Peeters *et al.* was that they used a wrong calibration curve for the MFC controlling the dry gas (helium) stream. As a result, the real vapor fraction in the test gas was lower than the calculated one. Peeters *et al.* observed a sudden change in nucleation rates (Fig. 1) at 207 K. It has become clear now that the nucleation rate jump was caused by a discontinuous change in the dilution procedure at 207 K. The raw data of Peeters *et al.* have been reinterpreted using the correct calibration data and they are shown in Fig. 1. The recalculated data in tabulated form can be obtained directly from the authors by e-mail or from EPAPS.[2] The corrected data agree quite well with the data by Wölk and Strey[3] (see Fig. 1). Wölk and Strey obtained their data using a completely different type of experimental setup.

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[1] P. Peeters, J. J. H. Gielis, and M. E. H. van Dongen, J. Chem. Phys. **117**, 5647 (2002).

[2] See EPAPS Document No. E-JCPSA6-120-004410 for the recalculated nucleation data in tabular form. A direct link to this document may be found in the online article's HTML reference section. The document may also be reached via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html) or from ftp.aip.org in the directory /epaps/. See the EPAPS homepage for more information.

[3] J. Wölk and R. Strey, J. Phys. Chem. B **105**, 11683 (2001).

 