

Strong Localization and Rapid Time Scales of Superheating in Solid-State Nanopores

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Abstract

Extreme localized superheating and homogeneous vapor bubble nucleation have recently been demonstrated in a single nanopore in thin, solid state membranes [1]. Aqueous electrolytic solution present within the pore is superheated to well above its boiling point by Joule heating from ionic current driven through the pore. Continued heating of the metastable liquid can eventually lead to explosive nucleation of a vapor bubble in the pore.

Here we report on the consistency of theoretical predictions with experimental results concerning the thermal, spatial and temporal scales involved. The temperature and heating dynamics of the nanopore system were modeled using COMSOL Multiphysics® software. The calculation required solving the heat equation in the nanopore with inclusion of a Joule heating source term, appropriate boundary conditions and temperature dependent material properties of the electrolyte and membrane. The heat equation must be coupled with charge continuity and Ohm's law to determine the source term. This is done using the Joule Heating (jh) and Electric Currents (ec) physics interfaces in COMSOL. The intrinsic symmetry of the nanopore allowed for a 2D axisymmetric model to be used for the calculation. A primary challenge to modeling was the lack of experimental data for the electrolyte material properties at the extremely high temperature regime achieved in the nanopore heating experiments at atmospheric pressure. Specifically, the high temperature dependence of electrical conductivity, thermal conductivity, density, and heat capacity of the electrolyte were needed. Therefore, appropriate assumptions were made for these properties either by substitution of material data of pure water or by fitting to experimental data.

Our results show that the liquid at the center of the pore can be rapidly superheated to several hundred degrees kelvin above the boiling point within microseconds. The temperature within the pore is shown to increase by over 100K from the edge to the center of a pore 50nm in radius. This degree of localization strongly indicates that vapor nucleation is homogeneous due to the high temperature dependence of the kinetic nucleation rate. We show that these results agree with the experimentally observed phenomena of heating and vapor bubble nucleation in the nanopore heating experiments.

The nanopore heating experiments are a novel and elegantly simple platform by which superheating and homogeneous nucleation can be studied in a highly controllable manner. The

calculations indicate the extremity of the conditions induced within the nanopore subject to high applied voltage. This simple method for inducing such high levels of superheat and bubble nucleation will find application ranging from fundamental physics to chemistry in superheated solutions to microfluidic devices.

Reference

[1] Nagashima, G., E. V. Levine, D. P. Hoogerheide, M. M. Burns, J. A. Golovchenko, "Superheating and Homogeneous Vapor Bubble Nucleation in a Solid-State Nanopore," *Phys. Rev. Lett.*, Accepted: June 4, 2014.