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A Brownian model for crystal nucleation

M.A. Durán Olivencia; F. Otálora Muñoz

Abstract-

In this work a phenomenological stochastic differential equation is proposed for modelling the time evolution of the radius of a pre-critical molecular cluster during nucleation (the classical order parameter). Such a stochastic differential equation constitutes the basis for the calculation of the (nucleation) induction time under Kramers' theory of thermally activated escape processes. Considering the nucleation stage as a Poisson rare-event, analytical expressions for the induction time statistics are deduced for both steady and unsteady conditions, the latter assuming the semiadiabatic limit. These expressions can be used to identify the underlying mechanism of molecular cluster formation (distinguishing between homogeneous and heterogeneous nucleation from the nucleation statistics is possible) as well as to predict induction times and induction time distributions. The predictions of this model are in good agreement with experimentally measured induction times at constant temperature, unlike the values obtained from the classical equation, but agreement is not so good for induction time statistics. Stochastic simulations truncated to the maximum waiting time of the experiments confirm that this fact is due to the time constraints imposed by experiments. Correcting for this effect, the experimental and predicted curves fit remarkably well. Thus, the proposed model seems to be a versatile tool to predict cluster size distributions, nucleation rates, (nucleation) induction time and induction time statistics for a wide range of conditions (e.g. time-dependent temperature, supersaturation, pH, etc.) where classical nucleation theory is of limited applicability.

Index Terms- A1. NucleationA1. Induction timeA1. Stochastic process

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