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An asymptotic and numerical study of time-dependent homogeneous nucleation modelled by the Becker-Doering equations

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Among many other physical, chemical or biological processes, the precipitation of lipids in aqueous solutions to form micelles, the formation of crystals in undercooled liquids or crystallization of glasses may be examples of homogeneous nucleation [1]. In the limit of large critical sizes, we carry out an asymptotic and numerical study of the Becker-Doering kinetic equations which model transient homogeneous nucleation. These equations are a particular case of coagulation-fragmentation processes in which clusters can grow or decrease only by adding or losing a single monomer. The two kinetic constants of the model are related through the assumption of detailed balance with the usual free energy containing cluster volume and surface terms. This leaves only one free constant, a discrete diffusivity, which depends on the physical phenomena to be described. Starting from pure monomers, three eras of transient nucleation are characterized in the Becker-Doering equations with two different models of discrete diffusivity: the classic Turnbull-Fisher formula and an expression describing thermally driven growth of the nucleus. The latter diffusivity yields time lags for nucleation which are much closer to values measured in experiments with disilicate glasses. After an initial stage in which the number of monomers decreases, many clusters of small size are produced and a continuous size distribution is created. During the second era, nucleii are increasing steadily in size in such a way that their distribution appears as a wave front advancing towards the critical size for steady nucleation. The nucleation rate at critical size is negligible during this era. After the wave front reaches critical size, it ignites the creation of supercritical clusters at a rate that increases monotonically until its steady value is reached. Analytical formulas for the transient nucleation rate and the time lag are obtained that improve classical ones and compare very well with direct numerical solutions [1, 2, 3].

References

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