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Kinetic effects of multi-component nucleation

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Abstract

The process of multicomponent condensation is considered. The global evolution of a metastable state originally created in the system has been investigated. Kinetic description of the ordinary one channel nucleation has been corrected. The theory taking into account several channels of nucleation is constructed. The analytical approximate description of the whole condensation process including the change of channels is given. The specific phenomena of the secondary nucleation is outlined and described analytically. The possibility of the temporary partial reverse condensation and the methods to describe it have been discussed.

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1. Introduction

Multicomponent nucleation is widely spread in nature and the task to describe it theoretically is rather actual. Creation of the classical theory of nucleation [1] allows to investigate the case of multicomponent nucleation. The stationary rate of multicomponent nucleation was found by Stauffer [2]. The stationary rate of nucleation depends on kinetic coefficients for embryos near the critical one which was clearly shown by Wilemsky and Wyslouzil [3]. Since the cited publications the

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stationary rate of nucleation can be considered as a known result confirmed by numerical simulations.

Contributions by Greer et al. [4] determined the stationary rate of nucleation for the systems of different nature (for the crystal nucleation in glasses the definition of kinetic coefficients is a special complex problem). The relation of nucleation phenomena to spinodal decomposition was considered by Binder et al. [5]. Here one has to stress that the theory considered here describes the nucleation started not far from the binodal curve and far from critical point. These restrictions are necessary to take the standard expression for the free energy of a critical embryo.

These results gave the opportunity to describe kinetics of the global evolution during the whole phase transition. The final period of evolution is known—it is the period of the recondensation (coalescence) and it is described by the Lifshic–Slezov theory [6]. But coalescence is mainly the redistribution of surplus substance (despite a small part of the surplus substance is gradually going from the mother phase to the new phase) and the real transition of practically all surplus substance occurs earlier. Before the coalescence, in fact, a phase transition takes place. To describe the phase transition it is necessary to determine the total number of droplets appeared in the process of nucleation. This quantity is the quasi-integral of evolution after the end of nucleation and before the coalescence. The characteristic times of coalescence ordinary strongly exceeds the characteristic times of experimental observations and the characteristic time of a practically all surplus substance consumption exceeds the time of nucleation (i.e., the duration of the intensive appearance of droplets). Then the problem to determine the number of droplets is the central one.

The problem to describe the global evolution until coalescence for multicomponent case was formulated by Stauffer [2] who considered the growth of supercritical droplets. The approximate analytical approach [7] determined the number of droplets after the end of nucleation (i.e., the end of the active appearance of droplets). Recent publications by Djikaiev et al. [8,9] repeat (with some errors) the consideration made earlier [7]. All mentioned publications gave the consideration of the most spread case of multicomponent condensation when the droplets of the only composition will appear in the system. Then the description of the nucleation can be reduced by renormalizations to the one component case. Therefore it was sufficient to give the corresponding references to the one component case [14]. Nevertheless Djikaiev et al. [8,9] reproduced the method of solution of one component situation and unfortunately it was done with errors. This requires to give here detailed analysis of this problem.

Besides the consideration of nucleation with the only composition of components in droplets the case of multiple compositions will be considered here. To see that the consideration based exclusively on the only composition is not complete we can imagine the following simple example. Let us suppose that the system contains two immiscible substances and both substances in a vapor phase are supersaturated. Then it is evident that the two separate processes of condensation will take place. As the result there will be droplets of the first substance and of the second substance.

When the combination of vapors in the system allows several equilibrium concentrations of different components it means that there are several paths to