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Air-hydrate crystal growth in polar ice

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Abstract

Based on the theory of precipitation from supersaturated solutions proposed by Lifshitz and Slyozov (J. Phys. Chem. Solids 19 (1/2) (1961) 35), we develop a mathematical description of post-formation growth (ripening) of mixed air clathrate-hydrate crystalline inclusions in polar ice sheets. The growth is controlled by oxygen and nitrogen diffusion through the ice matrix. Hydrate populations in general go through three sequential stages: (1) a short transient characterized by the rapid composition relaxation and dissolution of the smallest hydrates, (2) a slow transformation of the resulting size distributions towards a steady-state pattern that is an attribute of (3) the asymptotic stage of ripening. A regularization procedure is used to numerically solve the initial value problem. Computer simulations of the hydrate size distributions are compared to the data from a 3300-m ice core from Vostok Station, East Antarctica. The asymptotic stage is likely unattainable in natural conditions. Data from the GRIP ice core (central Greenland) suggest that the activation energy of hydrate growth increases at the elevated temperature near the ice-sheet bottom. The theory predicts extinction of the climatically induced fluctuations in the hydrate number-concentration and mean-radius profiles in ice sheets with depth.

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1. Introduction

Growth of the larger particles of a dispersed phase in a second continuous saturated phase at the expense of the smaller ones is known as Ostwald ripening. The enhanced curvature-induced solubility of the small particles from the Thomson–Freundlich–Gibbs relation (see e.g., Refs. [1,2]) explains this process qualitatively, however, quantitative prediction for specific multi-component systems is difficult. The theory of the evolution of dispersed precipitates in the two-phase mixtures has been developed by Greenwood [3], Lifshitz and Slyozov [4], and Glicksman [2], among others, and can be used as a basis to consider more complicated natural systems.

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