Kinetics of air-hydrate nucleation in polar ice sheets

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Abstract

Nucleation of air clathrate hydrates in air bubbles and diffusive air-mass exchange between coexisting ensembles of bubbles and hydrate crystals are the major interrelated processes that determine the phase change in the air-ice system in polar ice. In continuation of Salamatin et al. where the post-nucleation conversion of single air bubbles to hydrates was considered, we present here a statistical description for transformation of air bubbles to air clathrate hydrates based on the general theory of evolution of these two ensembles, including the gas fractionation effects. The model is fit to data on ice cores from central Antarctica, and then compared to other ice-core data. The focus is on the rate of clathrate-hydrate nucleation, which is determined to be the product of the inverse relative bubble size raised to the power $\lambda \approx 5.8$ with the relative supersaturation to the power $\beta \approx 2$. The clathration-rate constant is $k0 \approx 3.2-4.5 \times 10-6$ yr-1 at 220 K. The N2- and O2-permeation coefficients in ice, at 220 K, are inferred to be DN(2) $0 \approx 1.8-2.5 \times 10-8$ mm2 yr-1 and DO(2) $0 \approx 5.4-7.5 \times 10-8$ mm2 yr-1, respectively. Comparison of observations to simulations of bubble-to-hydrate transformation in Greenland ice sheet gave estimates for activation energies of hydrate formation and air diffusion of QJ ≈ 70 kJ mol-1 and Qd ≈ 50 kJ mol-1, respectively.

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