DIFFUSION COEFFICIENTS OF CATIONS AND OXYGEN IN SYNTHESIZED SINGLE CRYSTAL MELILITES AND THEIR IMPLICATIONS TO THE THERMAL HISTORY OF ALLENDE CAI

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Interdiffusion of major cations in gehlenite(Ge)-âkermanite(Ak)-Co-âkermanite(CoAk) system, tracer diffusion (Ni, Co, Fe, Mn, Ca, Sr, and Ba) in Ak and self-diffusion of O in end-member melilite have been measured.

SAMPLES AND EXPERIMENTAL METHODS. Melilite single crystals were synthesized from stoichiometrical mixtures of oxide and carbonate reagents by Czocharalski method(1). Each sample was cut into cubes of about 3 mm in size along the crystallographic axes and C-plane was polished and used for diffusion anneals. Interdiffusion. Two end-member samples were heated in contact with each other by the polished C-plane. Diffusion profiles were determined by EDXA. Tracer diffusion. A drop of aqueous solution of radioactive tracer(65Ni, 60Co, 54Mn, 43Ca, 88Sr or 133Ba) was dried up on the polished C-plane and heated for diffusion annealing. Penetration of the radioactive tracers was determined by step-wise grinding (about 5 µm in each step) of the C-plane, followed by gamma-ray measurement and/or beta-counting of the ground sample. O-self-diffusion. Each sample was heated with focused infrared lamps in a 0-18-enriched CO2 atmosphere. Penetration of 0-18 along the C-axis was measured by a Cameca 3f ion microprobe analyzer.

RESULTS AND DISCUSSION. Diffusion profiles are shown in Figs.1 and 2 and the calculated diffusion coefficients(D) are plotted in Fig.3.

D of Co in Ak determined by the tracer diffusion experiment fall approximately in the range of those observed for Co-Mg interdiffusion in Ak-CoAk system. This indicates that the mechanism of tracer diffusion of Co in Ak is essentially the same as in Co-Mg interdiffusion.

D of divalent ions in Ak increase with ionic radius(Fig.4). This appears to indicate that smaller Co and Fe substitute only the smaller Mg site in Ak, while larger Ca, Sr and Ba substitute the larger Ca site that would make the larger cations easier to diffuse through the Ak crystal lattice.

Diffusion profile observed for Ge-Ak interdiffusion experiment(Fig.1) shows very low D for Mg-Si-Al-Al interdiffusion, and possibly a complex change in diffusion rate over the whole range of Ge-Ak solid solution.

D of O self-diffusion in Ge determined in this study, though the value is tentative and subjected to a large uncertainty in temperature, is considerably lower than those observed by Hayashi and Muehlenbacks(2). This discrepancy may be due to the difference in defect density of the melilite samples used in both experiments.

IMPLICATIONS. The very low interdiffusion rate observed for Mg-Si-Al-Al pair in Ge-Ak system implies that the zoning in melilite crystals in CAI would have been unchanged under a most extensive heating event. On the other hand, the much higher D for divalent cations in Ak suggest that these divalent cations can easily enter into the melilite crystals, if Ak components in the melilite crystals are high enough to transport the divalent cations through Ak crystals.

D for O self-diffusion observed in this study, though tentative, is apparently not high enough to explain the heterogeneous distribution of O isotopes in CAI by exchange of O in melilite by those in a gaseous phase in a heating event(3). However, higher rate of exchange might have occurred in melilite crystals with higher defect densities.
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Fig. 1. Profile of interdiffusion in Ge-Ak(A) and Ak-CoAk(B) system along C-axis.

Fig. 2. Penetration of $^{95}$Sr in Ak crystal along C-axis.

Fig. 3. D vs. temperature.

Fig. 4. D vs. ionic radius of diffusing ion.

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