

INTER-DIFFUSION IN MELILITE: DETERMINATION OF AL+AL VS. MG+SI INTER-DIFFUSIVITIES.

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Introduction: Inter-diffusion of Al+Al vs. Mg+Si in the gehlenite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$)-åkermanite ($\text{Ca}_2\text{MgSi}_2\text{O}_7$) solid solution system of melilite along the a-axis has been studied in the temperature range of 1200~1350°C.

Melilite is one of the early condensing refractory minerals in the early solar nebula and commonly found in the Ca, Al-rich inclusions (CAIs) in the carbonaceous chondrites. Melilite in CAIs is generally gehlenitic in composition and often shows compositional zoning. Compositional zoning in melilite in the CAIs was essentially produced in the process of crystallization of melilite, but can be affected by a later thermal event. In this respect variation and distribution of gehlenite-åkermanite compositions in melilite crystals in CAIs may provide information on the thermal history of CAIs.

Experimental: We have determined inter-diffusion coefficients for Al+Al vs. Mg+Si pairs by coupled annealing of synthetic gehlenite and åkermanite crystals followed by determination of Al+Al vs. Mg+Si diffusion profile near the boundary of the two end-member crystals by using a scanning electron microprobe equipped with an energy-dispersive X-ray analyzer.

Results and Discussion. A typical example of diffusion profile is shown in Fig 1. The diffusion profile shows a characteristic pattern with steep inclinations at the both ends and a long plateau at the center, which is similar to the tentative results of Al+Al vs. Mg+Si inter-diffusion in the melilite system along the c-crystallographic axis reported in Morioka et al(1995)[1]. The observed diffusion profile indicates that the inter-diffusion coefficient is strongly dependent on composition, i.e., diffusion coefficient is high in the intermediate composition and decreases rapidly towards end-member compositions. The values of composition-dependent diffusion coefficients are calculated by Boltzmann-matano equation [2] (Fig. 2). These values are subjected to fairly large uncertainty, especially in the high D range. The position of maximum is at about Ak₈₀, near the composition with lowest melting point in the phase diagram (Flynn's rule [1,3]). However, since the calculated D values diverge so widely, about two orders of magnitude, it is unlikely that the observed relation between D and composition can be explained only by the Flynn's rule. Difficulty in charge balancing by paired ions under diluted condition may be another explanation for the steep decrease of D observed at the both end of the diffusion profile. Mg²⁺ ion diffusing through gehlenite lattice must find extra Si⁴⁺ in the neighboring T site. The low probability for Mg²⁺ ion to find pairing Si⁴⁺

under a diluted condition, i.e., at the diffusing front would prevent rapid diffusion of Mg²⁺ in the gehlenite lattice.

Temperature dependence of D for the composition range of Ak₅-Ak₂₅, which corresponds to the low D range in Fig 2, is shown in Fig 3. Activation energy of diffusion calculated from the values in the figure is approximately 400-500 kJ/mol, which is relatively large compared with those for divalent cations and oxygen measured in melilite previously [1,4]

The observed values of D for gehlenite-åkermanite inter-diffusion for the composition range of Ak₅-Ak₂₅ are in the same order of magnitude as those of oxygen in gehlenitic melilite, but slightly lower than those oxygen.

References: [1] Morioka M and Nagasawa H. (1991) GCA, **55**, 751-759. [2] e.g., Crank J. (1975) The Mathematics of Diffusion, Oxford. [3] Flynn, C.P. (1972) Point Defects and Diffusion, Clarendon Press. [4] Yurimoto H. et al (1989) GCA, **53**, 2387-2394.

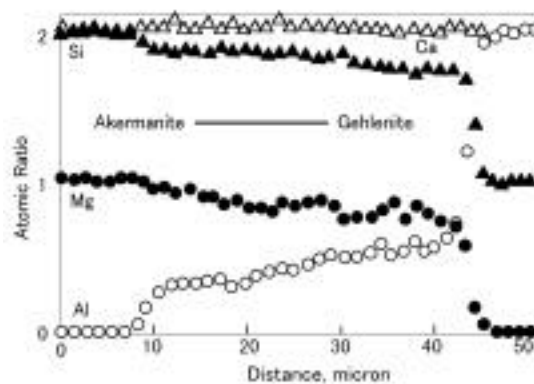


Figure 1. Diffusion profile of the gehlenite-åkermanite system annealed at 1350°C for 5 days.

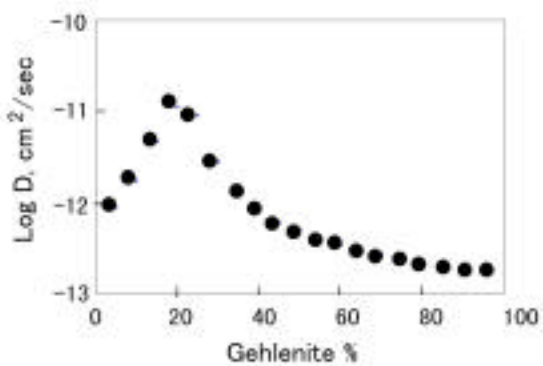


Figure 2. Log D (cm²/sec) vs. composition.

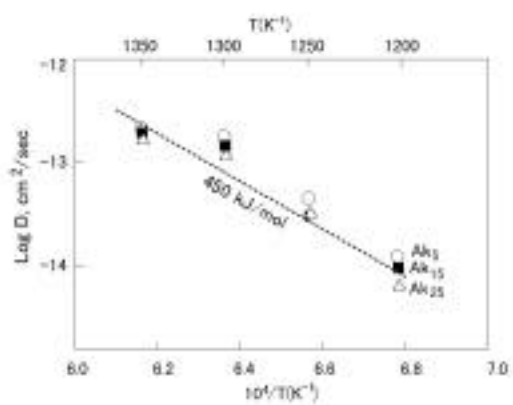


Figure 3. Log D (cm²/sec) vs. 1/T. Dotted line indicates activation energy for diffusion.