DIFFUSION COEFFICIENT OF O IN MELILITE: IMPLICATION TO THE THERMAL HISTORY OF ALLENDE CAI. Hiroshi Nagasawa, Dept. of Chemistry, Gakushuin University, Mejiro, Toshima-ku, Tokyo 171, Japan.

An attempt was made to explain the heterogeneous distribution of O isotope anomalies in the Allende CAI by experimentally determining the self-diffusion coefficient of (D) of O in melilite.

The large O isotope anomalies in the CAI ($\delta^{18}O \simeq \delta^{17}O \simeq -40$ ‰) were found only in spinel and fassaite pyroxene and never found in coexisting melilite and anorthite. This unique distribution, if produced by a diffusion process, requires melilite and anorthite to have much higher diffusion rates compared with those in spinel and pyroxene. Since D in melilite is not known, it is a key factor to solve the problem of the isotopic heterogeneity in CAI.

EXPERIMENTAL METHOD. 1) A natural gehlenite sample was crushed to less than 300μm and sieved to 4 uniform grain size ranges and purified by electromagnetic separation. 2) The powdered samples were heated to 1190°C in an atmosphere of CO_2 doped with ^{18}O. 3) The diffusion annealed samples were fused with a 1:1-mixture of Na_2CO_3 and K_2CO_3 at about 800°C for 2 hours. 4) The fused cake ontained by the carbonate fusion was decomposed by 100% phosphoric acid. The released CO_2 was measured for $^{18}O/^{16}O$ ratio by a Varian-Mat 230 mass-spectrometer. 5) D was calculated by assuming the grains are spheres with a diameter equal to the average diameter of the grains, and by using the equation,

$$\alpha = \frac{\delta^{18}Of - \delta^{18}O \text{ in } CO_2}{\delta^{18}Oi - \delta^{18}O \text{ in } CO_2} = \frac{6}{\nu^2} \sum \frac{1}{\nu^2} \exp\left(-\frac{\nu^2 \pi^2 Dt}{r_o^2}\right) \qquad 1)$$

where $\delta^{18}Oi$ and $\delta^{18}Of$ are initial and final $\delta^{18}O$ value in melilite, respectively, t and r_o are time interval of diffusion heating and radius of the grain, respectively.

RESULTS AND DISCUSSION. D values of 1×10^{-13} cm^2/sec (r_o=20μm, t=2 hours, T=1190°C) and 5×10^{-13} (r_o= 40 μm, t=2 hours, T=1190°C)were obtained. The earlier, smaller value might have been due to incomplete reaction in carbonate fusion. Thus, the larger value, 5×10^{-13} cm^2/sec is tentatively taken as D in melilite at 1190°C. This value is approximately equal to that of diopside pyroxene (2), if ΔE of 90 kcal/mol is assumed for both melilite and diopside.

Since fassaite is unstable above 1250°C (3), 1200°C is approximately the highest possible T for diffusion process. At 1200°C D(diop) ≃D(mel) ≃ 6×10^{-13}cm^2/sec, and thus, no large O isotope heterogeneity between melilite and fassaite is expected, if the effect of change in chemical composition in the same crystal structure is small and the grain sizes of the minerals are similar.

If, alternatively, the effect of chemical composition and/or cleavage or defect in the crystals etc changes effective D (or D/a^2) in the minerals, then isotopic heterogeneity can be produced under certain thermal conditions. Let's assume over-all effect changes D by 1 order of magnitude, and for a favorable case, D(fass)is larger than D(mel) by 2 orders of magnitudes. The $\delta^{18}O$ value calculated for the case of D(fass) = 6×10^{-14}, D(mel) = 6×10^{-12} cm^2/sec, grain size r_o= 0.01 cm is shown in Fig. 1.

At 1200°C time interval of the thermal process must be within about 800 to 6000 hours. D change above 1000°C is approximately factor of 10/100°C, if ΔE of 90 kcal/mol (2) is assumed for melilite. Thus, at lower T, time interval necessary for complete change of O in melilite become longer corresponding to the change in D by the above ratio.

Nagasawa et al (4) proposed a model of partial melting of CAI, in which the rate of O isotope exchange between melt and the atmosphere is much larger than that between solid and atmosphere. However, melting of crystals is not always favorable for the faster exchange of O isotopes, since increase in

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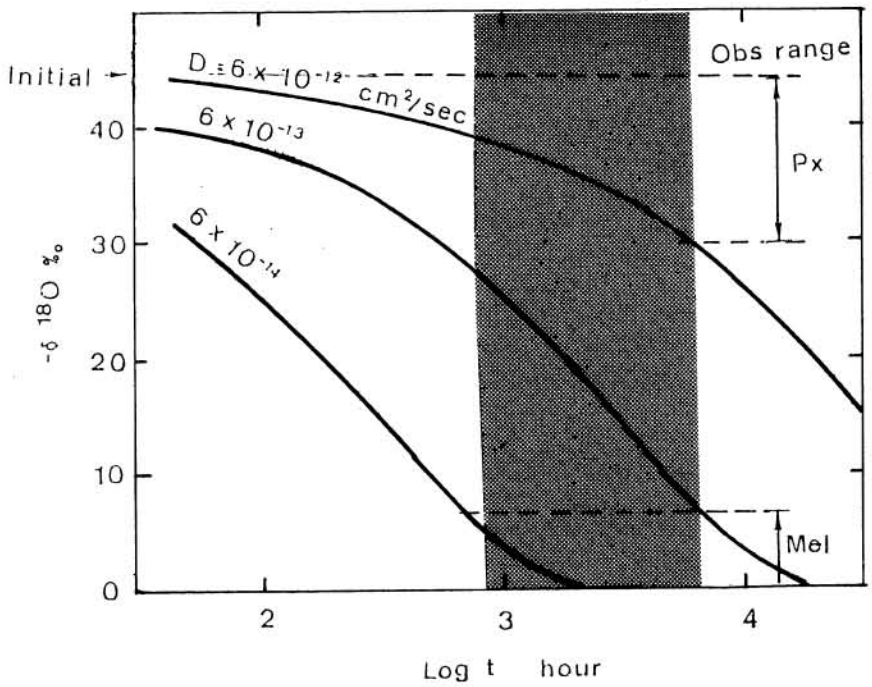

Fig. 1. $\delta^{18}O$ ‰ vs heating time interval (r_o = 0.01 cm). The observed range of O isotope ratios in fassaite and melilite in CAI are shown. Time interval of heating to 1200°C resulting $\delta^{18}O$ values observed in fassaite and melilite is limited to the shaded area. Since spinel has much lower D compared with melilite and pyroxene, no O isotope exchange is expected for spinel under a thermal process of this range.

diffusion length could compensate the increase in D or even could reduce D/a^2.

It is shown that diffusion coefficients of O in silicate phases involved in the formation of CAI are very important limiting factor for investigating the thermal history of CAI, since D constraints strongly temperature and time interval of heating. However, further study of diffusion coefficients is necessary to establish a model of thermal process that can explain the O isotope heterogeneity in the Allende CAI.

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