HIGH TEMPERATURE HEATING OF THE ALLENDE METEORITE II.

A synthesized silicate glass simulated to the Allende CAI (Ca, Al-rich inclusion in the Allende meteorite) doped with reagent REE (rare earth element) oxides was heated with a vacuum furnace for the purpose of studying the cosmochemical fractionation of REE in the early history of the solar system.

The powdered, homogenized glass sample was pressed to 3-mm-diameter pellets and heated to 1600-2000°C on a heating element of spiral Mo wire of 1mm in diameter under vacuum or in controlled atmosphere of H₂-H₂O mixture with total pressure of 2.6 x 10⁻⁴ to 0.64 Torr and H₂O/H₂ ratio of ≤ 10⁻⁴ - 6.9 x 10⁻⁷ using the heating system shown in Figure 1. The residues remained on the heating element after heating were analyzed by INAA (instrumental neutron activation analysis) for REE.

Figure 1. Furnace and gas pressure controlling system for high temperature heating. W: water saturation column, T₁: spiral cold trap, M: H₂O-manometer. L: gas leak. T₂: cold trap, F: vacuum furnace, VG: Pirani and ionization gauge, I: infrared thermometer. Desired H₂O/H₂ ratios were obtained by reducing partial pressure of H₂O in water-saturated H₂ gas using T₁ which was cooled by chloroform and/or dichloroethane as cooling agent. Total pressure was controlled by flow rate at L and speed of evacuation. Temperatures measured by I were not accurate because of the temperature gradient within the heating zone and unsteady emissibility of the infrared source consisting of Mo metal wire and silicate melt which have quite different emissibility.

The results of REE analyses are plotted as enrichment factor (ratio of concentration of a REE in residue to that in the unheated sample) versus REE atomic number in Figure 2. Because of the poor accuracy of the temperature values measured by the IR thermometer, the values of residual fraction (%), R, are expected to be better indicator for the degree of heating and evaporation. The results for the experiment under vacuum and 10⁻⁴ total pressure show large negative Ce anomalies which dominate those of Eu and Yb. The observed large Ce anomalies are probably due to higher oxygen partial pressure for the experiments compared with the experiments under higher H₂ pressures. The residual gas on evacuation is believed to be mainly of water vapor and

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1) Vacuum ($1 \times 10^{-6}$ Torr)

2) Total pressure $2.6 - 5.2 \times 10^{-4}$ Torr

3) Total pressure 0.64 Torr

4) Total pressure 0.64 Torr

Figure 2. Enrichment factor of REE in the residues. $R$ is residual fraction (%) defined by the ratio of weight recovered to that of the original sample. Chemical compositions of the original sample are: SiO$_2$, 29.8; Al$_2$O$_3$, 31.6; MgO, 10.1; CaO, 26.8; TiO$_2$, 1.0 (%); La, 110; Ce, 1300; Pr, 82; Nd, 1080; Sm, 7.1; Eu, 6.4; Gd, 210; Tb, 63; Dy, 61; Ho, 65; Er, 198; Tm, 126; Yb, 58; Lu, 64 (ppm). Accuracy of INAA are better than 10 % usually. However, some Pr, Gd, Ho and Tm values are subjected to larger errors due to poor counting statistics and/or unsteady baseline.
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other O-containing molecules adsorbed on the surface inside the furnace, which maintain oxygen partial pressure high enough to evaporate Ce as CeO$_2$ as predicted by Boynton$^1$. Negative anomalies of Eu are seen in all samples except most of those with the largest $R$ values among each group of the experiments. Negative anomaly of Yb is seen for the sample with $R = 27$ under high H$_2$/H$_2$O atmosphere, while it is slightly seen in the late stage residue in vacuum ($R = 5$). Relative enrichment of heavy REE over light REE is seen for $R = 5$ under vacuum. The above characteristics of REE fractionation are consistent with those calculated for condensation fractionation of REE by Boynton$^2$.

Although the starting material used in the experiment has chemical composition similar to CAI, the results can readily be applicable to the evaporation of the material with the average solar composition, because evaporation of the latter material proceeds through a stage in which residue has the chemical composition of CAI$^3$. Since evaporation and condensation are reverse process the results can also be applicable to the condensation fractionation of REE in the solar nebula, if the observed fractionation of REE are close enough to those in the equilibrium process. A direct implication of the results is that the flat REE patterns with negative Ce anomalies found in the Cl Allende CAI$^1$ and the cosmic spherule from oceanic sediment$^4$ are confirmed to have been produced by condensation or partial evaporation in high O/H environment (CI possibly was produced as supernova condensate and the spherule possibly by heating of a meteoritic material in the earth's atmosphere). Negative anomalies of Ce in the fractionated REE pattern reported for the bulk sample and mineral fractions of Melrose-b howardite$^5$ could also be produced by heating in high O/H atmosphere.

In conclusion, (1) the observed REE fractionation in the high temperature heating experiment is consistent with the results of thermodynamic calculation$^1,2$, and (2) it is confirmed experimentally that relative sizes of Ce, Eu and Yb anomalies reflect the O/H ratio in the atmosphere in which the fractionation took place. Further examination of heating condition is under way.