

Experiments on reaction between forsterite and Si-rich gas;
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Mg and Si are the most abundant refractory elements in the solar system. The Mg/Si ratios of chondrites are different among different chemical groups. This difference should be formed by fractionation in the primordial solar nebula. If Mg-silicates were formed by condensation in the cooling nebula, forsterite can react with Si-rich gas to form enstatite based on the equilibrium condensation model (e.g. [1]). Because the Mg/Si ratio of the solar abundances of the elements is similar to that of enstatite, only a small degree of fractionation was expected in the case of complete reaction, while a large amount of fractionation in the case of incomplete reaction. Thus, the reaction is essentially important for the Mg/Si fractionation in the nebula. In the present study, experiments on reaction between forsterite and Si-rich gas were carried out to investigate kinetics of the reaction, such as reaction rates and textures.

A "double capsule method" was newly designed for the present experiments (Fig.1). In this method, a single crystal of forsterite (5x1.25x1mm along a, b, and c axis, respectively) and cristobalite powder as a source for Si-rich gas are put into a Knudsen cell. The forsterite crystal is placed in an inner capsule to avoid contact with the cristobalite powder. When this double capsule is heated in vacuum, Si-rich gas in equilibrium with cristobalite (mainly SiO and O₂ based on thermochemical calculation) will be formed in the Knudsen cell, and enstatite will be formed by reaction with the forsterite crystal.

The experiments were carried out at temperatures ranging from 1345 to 1510°C for durations ranging from 5 to 236 hrs in vacuum (2×10^{-6} – 6×10^{-5} Torr). In the experiments, a reaction layer of enstatite surrounding the forsterite crystal was formed (Fig.2) except for some runs at low temperatures. The width of the enstatite is almost constant in a single run irrespective of the crystallographic orientation of the forsterite substrate. Polysynthetic twins observed in the enstatite indicates that the enstatite was formed as protoenstatite during the run and transformed to clinoenstatite by cooling after the run. The enstatite crystals usually have their c-axes normal to the forsterite surface.

The width of the enstatite layer increases with time and temperature. At constant temperatures the width, x , is nearly proportional to the square root of time, t (Fig.3) ($x = (kt)^{1/2}$, where k is the parabolic rate constant). This x - t relation indicates that the reaction is controlled by diffusion of elements in the enstatite layer. The activation energy and pre-exponent term of k were obtained by the method of least squares (approximately 330 KJ/mol and 4×10^{-6} m²/sec, respectively) although the data are scattered to some extent probably due to uncertainty of the temperature measurement and effect of stress by volume change during the reaction.

We can roughly estimate reaction rates in the primordial

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solar nebula from k obtained in the present experiments. This experimental value of k gives the maximum reaction rate because the partial pressure of SiO in the nebula was smaller than that in the experiments, where the gas is in equilibrium with the silica mineral. When we adopt the maximum duration for the reaction as the ^{129}I - ^{129}Xe formation interval (approximately 10^7 yr [2]), the maximum width of enstatite formed by the reaction is about 0.1-1mm at 1000-1200K. If the size of forsterite condensates is similar to that of matrix minerals (about $1\text{ }\mu\text{m}$), it takes more than about 10^3 - 10^0 yr at 1000-1200K to complete the reaction. The present results indicate that the reaction was completed when the size of forsterite was sufficiently smaller than $1\text{ }\mu\text{m}$, while maximum fractionation took place when the size was sufficiently larger than $1\text{ }\mu\text{m}$.

References: [1] Grossman, L. and Larimer, J. W. (1974) *Rev. Geophys. Space Phys.*, 12, 71-101. [2] Podosek, F. (1970) *Geochim. Cosmochim. Acta*, 34, 341.

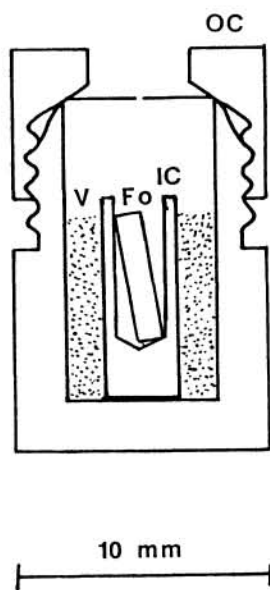


Fig.1

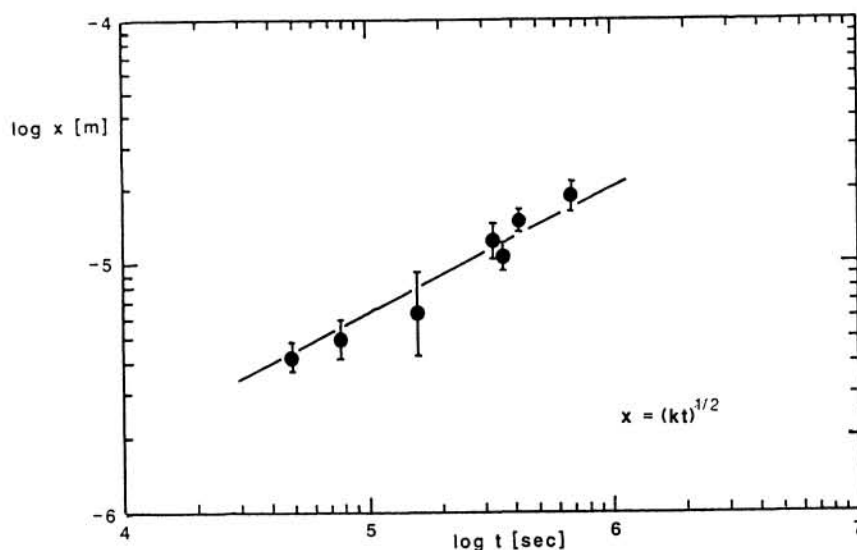


Fig.3

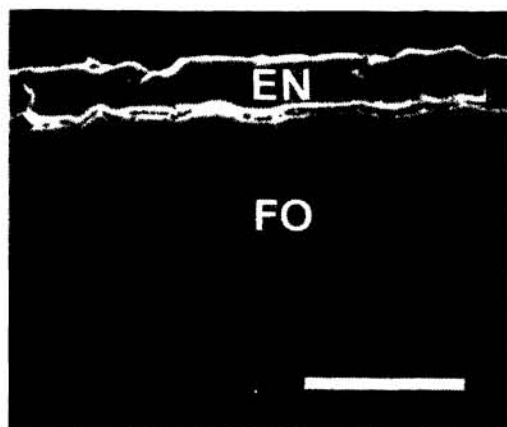


Fig.2

Figure 1. A schematic illustration for a "double capsule method". OC: outer capsule of Mo used as a Knudsen cell, IC: inner capsule of Mo, Fo: a single crystal of forsterite, V: cristobalite powder as vaporization source.

Figure 2. A SEM image of a run product (1435°C for 117.4 hrs). Scale bar $50\text{ }\mu\text{m}$. (FO: forsterite, EN: enstatite)

Figure 3. A relation between the width of the reaction layer of enstatite, x , and time, t , at temperatures of 1533 - 1560°C .