CRYSTALLIZATIONS OF FORSTERITE FROM AMORPHOS Mg-BEARING SILICATE GRAINS. C. Kaito and Y. Kimura, Laboratory for Nano-Structure Science, Department of Physics, Ritsumeikan University, 1-1-1 Nojihigashi, Kusatsu-shi, Shiga, 525-8577, Japan. kaito@se.ritsumei.ac.jp, ykimura@se.ritsumei.ac.jp

Introduction: Magnesium-rich crystalline silicates have been discovered in some circumstellar shells [1], in disks around evolved stars [2], in protoplanetary accretion disks around young stars [3-5] and in comets [6] due to the observation of Infrared Space Observatory. The fact that part of the silicates produced around evolved stars are crystalline but that no crystalline silicates have been yet detected in the interstellar medium raises the question of the structural evolution of this crystalline dust during its life. Recently, abundant crystalline silicates in the disk of very low mass star have been reported [7]. In this paper, crystallization of Mg_2SiO_4 from amorphous Mg-bearing silicate grains due to the direct heating, electron beam irradiation and chemical reaction processes have been demonstrated by transmission electron microscope (TEM) based on the in-situ observation. Amorphous grain size and electron energy dependences on the crystallization have been presented as the one of the low-temperature crystallization.

Experimental setup: Amorphous Mg-bearing silicate grains with about 20-150 nm in diameter were produced by the coalescence between MgO and SiO smoke grains [8]. i) The direct observation of the crystallization process was carried out using transmission electron microscope equipped with a heating holder, which can be heated up to 800ºC. ii) The metamorphism of the carbon coated amorphous Mg-bearing silicate grains was directly observed using a Hitachi H-9000NAR TEM with a special holder, which can be heated up to 1500ºC. iii) Nonthermal crystallization was succeeded by preparing of the amorphous Mg-bearing silicate grains covered with a carbonaceous layer consisting of amorphous carbon, CH_4 and their derivatives. In order to observe the as-deposited structure of the sample before exposure to air, the sample was transformed to the TEM using special transfer holder and observed immediately. After that, it was exposed to air and then the same position was observed again. iv) By cooling amorphous Mg-bearing silicate grains in CH_4 gas at 80 Torr with liquid nitrogen, a lot of frosts of CH_4 were grown on the specimen surfaces. The frosts of CH_4 disappeared by taking out into air. The electron beam crystallization took place for the sample of the pre-treated CH_4 gas.

Results and Discussion: Typical crystallization samples by the four experiments are shown in Fig. 1. The direct heating method, the crystallization took place whole the particles, which can be clearly recognized Bragg reflection image. The size dependence was not observed. The crystallization temperature covered with carbon layer becomes 200ºC lower accompanying with the slight crystallization of an amorphous carbon layer.

Low temperature crystallization due to the exothermic chemical reaction [9] occurred concentrically beneath the carbon layer. The crystallization of amorphous carbon layer can be clearly observed. Therefore, chemical reaction energy with the oxygen gas in air introduced the crystallization of surface carbon layer and the reaction energy acted to the crystallization of the amorphous silicate. Electron beam crystallization by observing 90 minutes at the ordinary observation condition was observed as the crystallization of a few nanometers Mg_2SiO_4 crystal on the particle surface. The crystallization took place selectively for the 100 nm order amorphous silicate grains.

Direct heating crystallizations correspond to the thermal annealing model [10]. Last two experimental results correspond to the low-temperature crystallization. The chemical reaction based on Greenberg model and electron energy dependence on the crystallization were elucidated. The crystallization of amorphous dust started from the grain surface, the formation of a rim-like layer due to prenucleation was detected [11]. If the amorphous grain surfaces were covered with thin amorphous layer, the crystallization temperature decrement due to the graphitization of amorphous carbon layer becomes lower about 600ºC. The rim-like contrast was never observed. The crystallization process from some nucleuses at the surface layer was same for both experiments. As elucidated low-temperature crystallization of $MgSiO_3$ grains, the previous treatment by the irradiation of film by He+ ions indicated the crystallization at 300kV electron beam [12]. In the present grains, the electron beam crystallization hardly took place. If we adsorbed the CH_4 gas before the electron beam irradiation, the crystallization of 100 nm order amorphous Mg-bearing silicate grains predominantly took place at 100kV electron beam. The mean free path of the electron in the material was clearly observed. The crystallites of the order of 20 nm were distributed on the particle surface.

The nonthermal crystallization of the amorphous grains covered with carbon layer produced CH_4 gas atmosphere was directly observed accompanying with the alteration of carbonaceous layer to the graphitic structure due to oxidation at room temperature in air.

The differences of the crystallization processes for the four specimens were clarified by the present paper.

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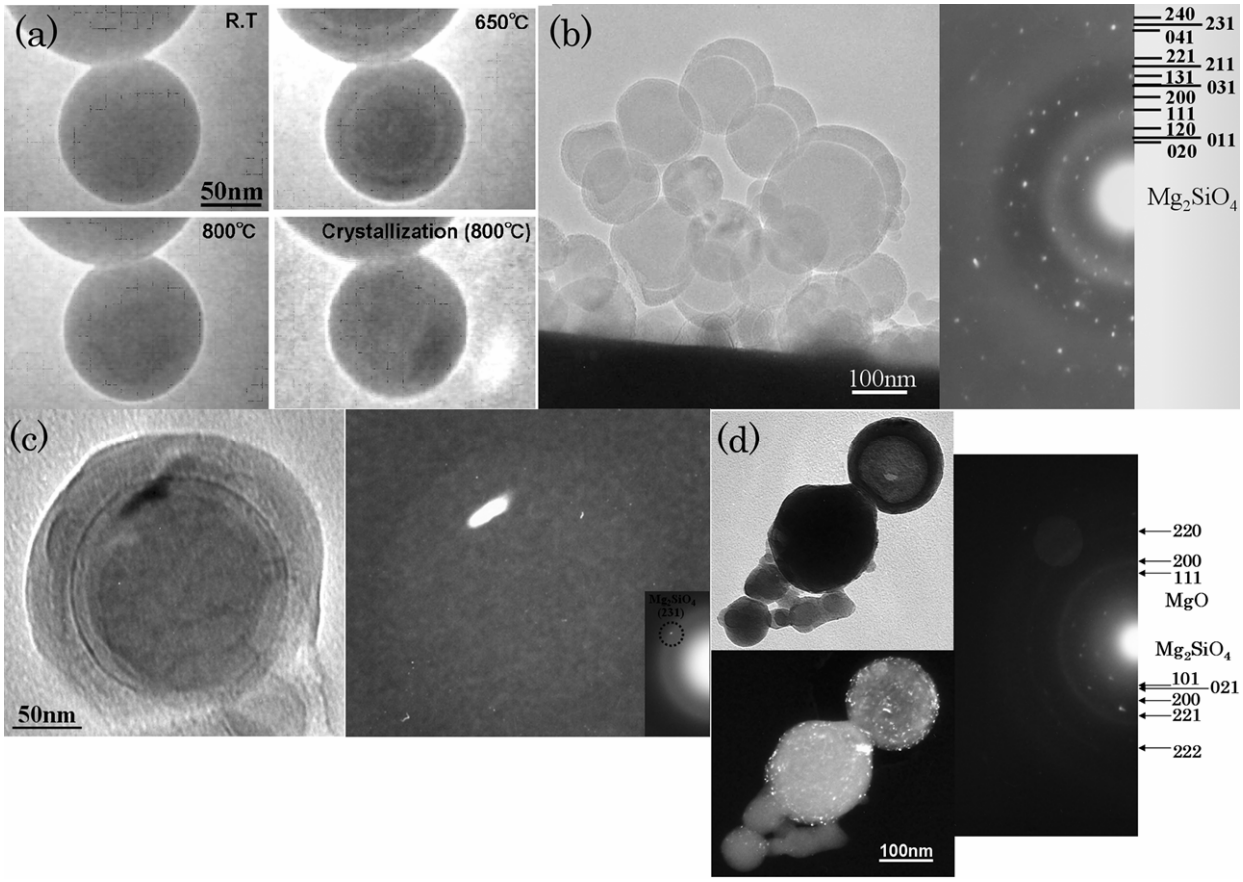

Fig. 1. (a) Alteration of an amorphous Mg-bearing silicate grain by heating in TEM. The appearance and disappearance of the white contrast ring are observed at 650 and 800°C. The Bragg reflection shows the crystallization of the grain. (b) Crystallization took place at 600°C. Electron diffraction pattern can be identified as the Mg_2SiO_4 crystal. Amorphous grains were crystallized lower temperature than (a). (c) Amorphous Mg-bearing silicate grains after adsorbed CH_4 gas. The grain was crystallized at room temperature from the surface. Large crystal formation on the part of Mg-bearing silicate grain can be seen. (d) Amorphous Mg-bearing silicate grains after adsorbed CH_4 gas. Electron beam irradiation at 100 kV induced the crystallization. In addition to Mg_2SiO_4 crystal, diffuse rings of MgO can be seen. The small crystallizations of MgO are also contained. The dark field image clearly shows the crystallization as light contrasts.