

AN EXPERIMENTAL STUDY OF CIRCUMSTELLAR SILICATE DUST CRYSTALLIZATION. A. Tsuchiyama¹ and K Murata¹, ¹Department of Earth and Space Science, Graduate School of Science, Osaka University, 1-1 Machikaneyama-cho, Toyonaka, 560-0043 JAPAN (akira@ess.sci.osaka-u.ac.jp).

Introduction: Infrared astronomical observation revealed that silicate dust is partially crystallized as olivine and pyroxene in circumstellar environments. The most abundant dust species around oxygen-rich evolved stars is amorphous silicate, and about 5% of the dust is composed mainly of olivine and pyroxene with Mg-rich compositions (Mg#>0.9) [1]. There is a possibility that the amorphous silicates condensed in stellar winds first, and then crystalline silicates crystallized from the amorphous silicates by annealing during cooling process [2]. It is widely accepted that interstellar silicate dust is almost amorphous [3]. Mg-rich olivine and pyroxene have been also observed around oxygen-rich young stars as well as abundant amorphous silicate (e.g., [4]). The crystalline phases are considered to be crystallized from the amorphous interstellar dust.

In order to understand the crystallization processes in the circumstellar environments, crystallization experiments from metastable amorphous silicates as dust analogues at relatively low temperatures below the solidus have been made (e.g., [5]). Lately, quantitative kinetic parameters for crystallization of olivine and pyroxene were determined by measuring the degree of crystallization as a function of temperature and time [6-8]. Metastable Mg-Fe partitioning between olivine and amorphous silicate was also determined to discuss the chemical composition (Mg#) of the crystalline phase [7]. In this paper, we briefly review these new data for the crystallization, and based on the data the chemical compositions of the amorphous silicates and crystallization behaviors around evolved and young stars are discussed.

Crystallization experiments: Crystallization experiments with three different CI-like compositions have been made; (A) S-free CI composition (SiO₂-Al₂O₃-MgO-CaO-FeO-NiO-Na₂O) [6], (B) S-free and Fe-depleted simple CI composition (SiO₂-MgO-FeO, where [Fe]=[Fe]_{CI}-[S]_{CI}) [7], and (C) S- and Fe-free simple CI composition (SiO₂-MgO) [8] (Mg#=0.543, 0.733 and 1, and (Mg+Fe)/Si=1.97, 1.46 and 1 for (A), (B) and (C), respectively). The starting materials of the amorphous silicates were synthesized by sol-gel method. Time evolution of the crystallization degree is expressed by Johnson-

Mehl-Avrami equation at a given temperature; $C=C_{\infty}[1-\exp\{-(t/\tau)^n\}]$, where C_{∞} is the final degree of crystallization, t time, τ time constant for the crystallization, and n kinetic parameter. The time constant is expressed as Arrhenius equation; $1/\tau=\nu_0(-E/kT)$, where ν_0 is frequency factor, E activation energy, and T temperature. Olivine crystallizes from Si-poor compositions (A) and (B) ((Mg+Fe)/Si~1.5-2) accompanied with heterogeneous nucleation on preexisting minor crystallites ($n=1.5$), while pyroxene (enstatite) from Si-rich composition (C) ((Mg+Fe)/Si~1) by nucleation and growth ($n=2.5$). The activation energy for pyroxene ($E/k=1.1\times 10^5$ K at 750-800°C for (C)) is larger than that of olivine ($E/k\sim 6\times 10^4$ K at 660-758°C for (A)). Mg is preferentially distributed into olivine with the effective partition coefficient of Mg/Fe between olivine and amorphous silicate, $K_d=(Mg/Fe)_{as}/(Mg/Fe)_{ol}$, of ~0.4 for (B).

Discussion: The chemical compositions of amorphous silicates around evolved and young stars can be constrained. If we assume that the Mg:Fe:Si ratio is in CI composition and Fe is present as FeO in silicates, metal and/or Fe sulfides, the Mg# and the (Mg+Fe)/Si ratio of the amorphous silicates are ~0.8-1 and 1.37-1.07, respectively, based on K_d .

TTT diagram can be made from the Arrhenius relations of τ for olivine and pyroxene. The large values of E (~ 1×10^5 K) show that crystallization occurs in narrow temperature ranges (approximately at 1000K) in a wide range of cooling or heating time scales (~ 10^5 - 10^{10} s). The small population of crystalline silicate dust around evolved stars can be explained by the first condensation of amorphous silicates followed by crystallization during cooling along the stellar winds. Pyroxene and olivine are expected to exist in inner and out regions of a protoplanetary disk, respectively.

References: [1] Tielens A. G. G. M. Et al. (1998) *Astrophys. Space Sci.*, 255, 415. [2] Seki J. and Hasegawa H. (1981) *Prog. Theor. Phys.*, 66, 903. [3] Kemper F. et al. (2005) *ApJ*, 633, 534. [4] Honda M. et al. (2006) *ApJ*, 646, 1024. [5] Hallenbeck S. L. et al. (1998) *Icarus*, 131, 198. [6] Murata K. et al. (2007) *ApJ*, 668, 285. [7] Murata K. et al. (2009) *ApJ*, 696, 1612. [8] Murata K. et al. (2009) *ApJ*, 697, 836.