

Nonthermal crystallization of amorphous silicates in comets

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Recent observations show the ubiquity of crystalline silicate in various objects, among which comets provide the best opportunity to study possible processing of pristine matter during their formation and evolution. While thermal processing of bare amorphous silicates in the hot inner solar nebula has been invoked, its drawback is a difficulty in explaining the interstellar composition of cometary ices. Here we apply a model of core-mantle interstellar grains to propose nonthermal crystallization of the amorphous silicate core due to the energy released by chemical reactions in the organic refractory mantle when moderately heated by solar radiation. By formulating the nonthermal crystallization, we find that the degree of crystallinity is determined by a single parameter that is proportional to the amount of the released energy. We show that the present mechanism of crystallization is capable of reproducing the strengths of crystalline silicate features observed in comets. Our model can reconcile crystalline silicate in the comae of comets with the interstellar composition of ices in their nuclei.

Key words: Astrochemistry, radiation mechanisms: non-thermal, comets: general, solar system: formation, planetary systems: protoplanetary disks, dust, extinction.

1. Introduction

The presence of crystalline silicates in comets was first identified by infrared spectra of dust in the coma of comet 1P/Halley (Bregman *et al.*, 1987). Since then, infrared features of crystalline silicate have been observed in a number of comae, but the origin of crystalline silicates is a matter of debate. Experimental studies on thermal annealing of amorphous magnesium silicate smokes show that a dust temperature in a coma of a typical comet is too low for amorphous silicate^{*1} to crystallize in situ (Hallenbeck *et al.*, 1998, 2000). Therefore, thermal annealing of amorphous silicates in the hot inner solar nebula and incorporation of crystalline silicate into comet nuclei are suggested (see a review by Hanner, 1999). In contrast, the interstellar composition of gas in the comae indicates the preservation of interstellar volatiles in the cold outer nebula (Biermann *et al.*, 1982; Mumma, 1996). To reconcile hot and cold materials, crystalline silicates are considered as being transported from the inner solar nebula to the outer comet-formation region (Bockelée-Morvan *et al.*, 2002). Nuth *et al.* (2000) claim that the fractional abundance of crystalline silicate in the solar nebula increases with time and thus can be used to date a comet's formation age. Harker and Desch (2002) considered in-situ annealing of silicate at elevated temperatures by nebular shocks that could proceed with crystallization up to 20 AU, depending on the model parameters (Nakamoto and Miura, 2005). Without the outward transport, however, the in-situ annealing mechanism predicts no crystalline silicate in the Kuiper Belt comets, contrary to

observations (Watanabe, 2004). As noticed by Huebner (2002), none of the thermal mechanisms allow comets to retain the interstellar composition of ices in their nuclei.

We notice that a detection of crystalline silicates in a cometary coma does not necessarily mean the presence of the crystallites in the nucleus. Comet nuclei may consist of interstellar ices along with interstellar dust that is almost free from crystalline silicates. If low-temperature crystallization of amorphous silicate, as pointed out by Molster *et al.* (1999), takes place in a comet coma, then one could automatically reconcile observable signatures of both interstellar ices and crystalline silicates in the coma. Recently, Carrez *et al.* (2002) have found that MgSiO₃ glasses irradiated by 300 keV electrons crystallize at room temperature. We propose yet another nonthermal mechanism to preserve interstellar composition of volatiles in comets.

2. Nonthermal Crystallization Model

It is considered that a comet is an agglomeration of interstellar dust consisting of an amorphous silicate core, an organic refractory (OR) inner mantle, and an icy outer mantle (e.g., Greenberg, 1982; Jessberger *et al.*, 1988). Because of the fluffiness of cometary dust, crystallization does not make essential difference between the aggregates and their constituent interstellar grains. The radius of the amorphous silicate core of the interstellar grain is $a = 0.07 \mu\text{m}$ and the thickness of the OR mantle is $h_r = 0.03 \mu\text{m}$ (Li and Greenberg, 1997). We assume Mg-bearing glass as the main component of the amorphous silicate core (see Jessberger *et al.*, 1988; Kimura *et al.*, 2003). Figure 1 depicts our nonthermal mechanism of crystallization at moderate temperatures

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*1 We use the term “amorphous silicate” meaning noncrystalline materials with a stoichiometric silicate composition following the astronomical literatures, although it may not be a correct mineralogical term.

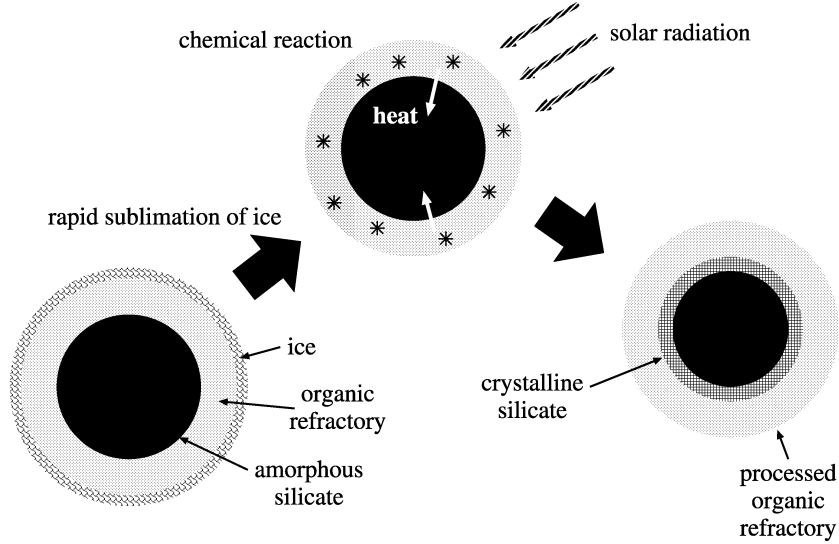


Fig. 1. Illustration of our nonthermal crystallization mechanism.

(Yamamoto and Chigai, 2005). A typical temperature of cometary dust is several hundred K at heliocentric distances around 1 AU from the sun. This is insufficient for the amorphous silicate core to crystallize but is sufficiently high for the icy mantle to sublime and produce the observed interstellar composition of gas in a coma. Such moderate heating also increases the diffusion rate of reactive molecules and triggers reactions among them in the OR mantle. In consequence, the energy released by the reactions increases the temperature and subsequently expedites further reactions. This is a positive feedback process resulting in chain reactions, a sudden rise in the temperature of the OR mantle, and heat flow into the amorphous silicate core. The heat flow brings about crystallization from the core surface towards the interior.

The degree of crystallinity $f_c(r, t)$ at a distance r from the center of the core at a time t is given (Haruyama *et al.*, 1993) by

$$1 - f_c(r, t) = \exp\left(-\int_0^t \frac{dt'}{Ae^{E_c/kT(r,t')}}\right), \quad (1)$$

where k is Boltzmann's constant and E_c is the effective activation energy of crystallization. In Eq. (1), $Ae^{E_c/kT}$ indicates the timescale for amorphous-to-crystalline transition (Brucato *et al.*, 1999; Fabian *et al.*, 2000). Here, the vibration period A of diffusing atoms in silicate is 5×10^{-14} s (Duschl *et al.*, 1996). Equation (1) would overestimate the crystallization degree because it allows crystallization above the melting point (see Section 4). The temperature $T(r, t)$ is determined by the equation of heat conduction:

$$\frac{\partial T}{\partial t} - \frac{\chi}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right) = 0, \quad (2)$$

where χ is the thermal diffusivity. Heating due to deposition of latent heat of crystallization and cooling due to thermal emission and sublimation of organic molecules are negligible because the time scales of these processes are much longer than that of heat conduction.

The duration of heating by reactions in the OR mantle is almost instantaneous compared to the timescale of heat conduction into the silicate core. We model such heating by putting an instantaneous spherical surface heat source on the silicate core surface and take the temperature distribution of the silicate core at $t = 0$ to be $T(r, 0) = \text{const} \cdot \delta(r - a)$. Here the proportional constant (i.e., the strength of the source) is determined from the total energy released by the reactions given by

$$\frac{4\pi c_p}{\Omega} \int_0^a r^2 dr T(r, 0) = 4\pi g a^2 n_r h_r E_r, \quad (3)$$

where c_p is the specific heat per unit cell, $g = 1 + h_r/a + (1/3)(h_r/a)^2$ is a geometrical factor, n_r is the number density of reactive molecules in the OR mantle, E_r is the amount of energy released per reaction, and Ω is a volume of the unit cell of silicate. The initial temperature distribution in the core is thus given by

$$T(r, 0) = \frac{g\Omega n_r h_r E_r}{c_p} \delta(r - a). \quad (4)$$

We ignore the temperature prior to heating by the reactions because no crystallization takes place in practice at temperatures before the reactions. The solution to Eq. (2) for the initial condition (4) is given (Carslaw and Jaeger, 1959, p. 259) by

$$T(z, t) = \frac{g\Omega n_r h_r E_r}{2c_p (\pi \chi t)^{1/2}} e^{-z^2/4\chi t} \quad (5)$$

for $z \ll a$, $z = a - r$ being the depth from the core surface. In most cases of interest, the crystallized region is confined to the silicate core surface as shown below. Inserting Eq. (5) into Eq. (1), one obtains the degree of crystallinity at depth $\zeta = z/(\chi A)^{1/2}$ for $t \rightarrow \infty$ to be

$$f_c(\zeta, \infty) = 1 - \exp\left[-\theta^2 \int_0^\infty dx x \exp\left\{-x \exp\left(\frac{\zeta^2}{2\theta^2 x^2}\right)\right\}\right], \quad (6)$$

where

$$\theta = \frac{gk\Omega n_r h_r E_r}{E_c c_p (2\pi\chi A)^{1/2}}, \quad x = \frac{2E_c c_p (\pi\chi t)^{1/2}}{gk\Omega n_r h_r E_r}. \quad (7)$$

Note that the material dependence of $f_c(\zeta, \infty)$ is put together into the dimensionless parameter θ . The degree of crystallinity on the surface is given from Eq. (6) by

$$f_c(0, \infty) = 1 - e^{-\theta^2}. \quad (8)$$

This indicates that $\theta > 1$ is the condition of substantial crystallization near the surface.

3. Degree of Crystallinity

The parameter θ is the key quantity that determines the degree of crystallinity. For forsterite whose density is $\rho = 3.2 \text{ g cm}^{-3}$ and molecular weight is $\mu = 140$, we find $\Omega = \mu m_H / \rho = 7.3 \times 10^{-23} \text{ cm}^3$ where m_H is the hydrogen mass. The specific heat per unit cell is expressed as $c_p = fk$ at temperatures higher than the Debye temperature, where f is freedom of motion of atoms (plus contributions from electrons) composing a unit cell. The specific heat measured for forsterite is $40\text{--}50 \text{ cal mol}^{-1} \text{ K}^{-1}$ at $T = 900\text{--}2000 \text{ K}$ (Chase *et al.*, 1985) which corresponds to $f = 21\text{--}26$. Following Hofmeister (2001), we estimate the thermal diffusivity of amorphous silicate with the sound velocity $c_s \simeq 3 \times 10^5 \text{ cm s}^{-1}$ and phonon mean free path $\lambda \approx (3\Omega/4\pi)^{1/3} \simeq 2.6 \times 10^{-8} \text{ cm}$ to be $\chi = c_s \lambda / 3 \sim 2.6 \times 10^{-3} \text{ cm}^2 \text{ s}^{-1}$ and the length scale $(\chi A)^{1/2}$ to be $1 \times 10^{-8} \text{ cm}$. We take $n_r = 10^{21}$ to 10^{22} cm^{-3} , corresponding to concentrations of reactive molecules of roughly 1 to 10% in the OR mantle (Greenberg, 1976). The energy released per reaction, E_r , is a few to 10 eV (Schutte and Greenberg, 1991; Sorrell, 2001). Using these values, θ is expressed as

$$\theta = 2.7 \left(\frac{n_r}{3 \times 10^{21} \text{ cm}^{-3}} \right) \left(\frac{h_r}{300 \text{ \AA}} \right) \left(\frac{E_r}{5 \text{ eV}} \right) \cdot \left(\frac{10^{-16} \text{ cm}^2}{\chi A} \right)^{1/2} \left(\frac{\mu}{140} \right) \left(\frac{21}{c_p/k} \right) \left(\frac{3.5 \text{ eV}}{E_c} \right). \quad (9)$$

The θ value being on the order of unity implies that amorphous silicates in comets do crystallize to a certain degree by the present mechanism. Figure 2 shows the degree of crystallinity $f_c(r, \infty)$ calculated from Eq. (6) for $\theta = 0.9$ to 9 ($n_r = 10^{21}\text{--}10^{22} \text{ cm}^{-3}$). Also plotted is the case for $\theta = 18$ (e.g., $n_r = 10^{22} \text{ cm}^{-3}$, $E_r = 10 \text{ eV}$) as a possible upper limit. The crystallized region is localized near the core surface and hence the assumption posed in deriving Eq. (6) is not violated.

We examine whether the range of crystallinity derived from the present model provides the strengths of crystalline silicate peaks as observed in the $10 \mu\text{m}$ band. The strength of a crystalline silicate peak is evaluated by the absorption cross section at a wavelength λ of the peak relative to that at $\lambda = 8 \mu\text{m}$, where the spectra show a continuum baseline arising from emission by the OR mantle. The absorption cross section is calculated for three-layered spherical grains consisting of an amorphous silicate core coated by crystalline olivine together with an outermost OR mantle (see

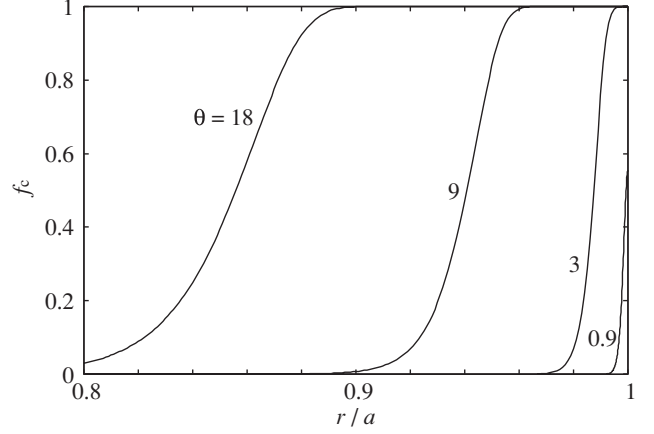


Fig. 2. The degree of crystallinity $f_c(r, \infty)$ for $\theta = 1$ to 18 as a function of distance r from the center of the silicate core having the radius a .

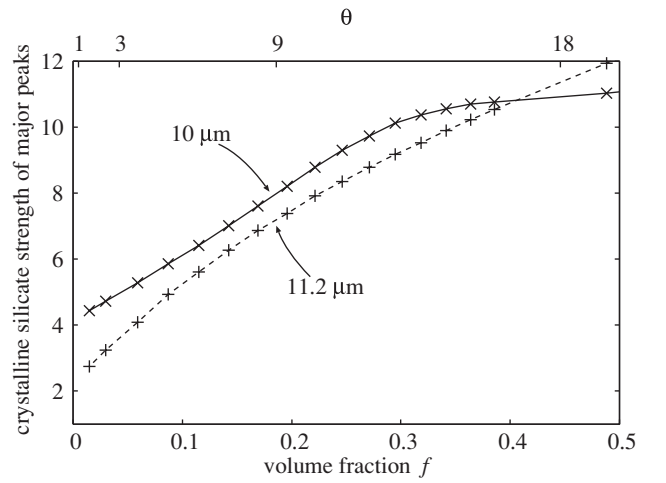


Fig. 3. Crystalline silicate strengths expected for observed major features as a function of crystallinity f_c .

Friedman, 1998). We use the complex refractive indices for organic refractory given by Li and Greenberg (1997), amorphous silicate by Scott and Duley (1996), and forsterite by Sogawa *et al.* (2006). The fraction \bar{f}_c of crystalline silicate in the volume V of the silicate core is estimated as

$$\bar{f}_c = \frac{1}{V} \int dV f_c(\zeta, \infty), \quad (10)$$

which gives $\bar{f}_c = 0.4\text{--}20\%$ for $n_r = 10^{21}\text{--}10^{22} \text{ cm}^{-3}$ ($0.9 \lesssim \theta \lesssim 9$) from Fig. 2. We focus for simplicity on the strengths of the major peaks at 10.0 and $11.2 \mu\text{m}$, which are attributed to forsterite. Figure 3 shows that the silicate peak strengths increase with crystallinity f_c . The strengths at 10.0 and $11.2 \mu\text{m}$ are comparable and, for $\theta = 0.9\text{--}9$, the strengths of both peaks could reach up to 2–3 times the strengths due to amorphous silicate ($f_c = 0$). These characteristics of the peaks are in harmony with infrared observations of cometary comae.

4. Discussion

We have proposed a nonthermal crystallization model of cometary silicate grains. The degree of crystallinity depends only on θ , which measures the amount of energy de-

posited in the OR mantle. The volume fraction of crystallites as much as 20% is realized depending on the θ -values. As shown in Fig. 3, the crystallinity obtained in the present model may reproduce the observed strengths of crystalline silicate features in the 8–13 μm spectra. See Kimura *et al.* (2008) for discussion of the detailed spectra. In spite of the success in reproducing the observed infrared features, the present mechanism does not require mixing of amorphous and crystalline silicates. A natural consequence of the present model is that volatiles of interstellar composition may be preserved composition in comets.

The present mechanism would also work for grains in other objects such as in protoplanetary disks if (1) the condition $\theta > 1$ to induce crystallization is fulfilled and (2) moderate heating is available to trigger exothermal reactions in OR mantles. Crystalline silicates found in debris disks might be brought in part from comet-like objects in the disks. It is also possible to make crystalline silicate by the present mechanism in the vicinity of protostars. Recent observations showed the existence of crystalline silicates around protostars at an early stage of 10^5 years (Ciardi *et al.*, 2005). This mechanism may also be triggered by shock waves and work at high temperature regions near O or B stars, although radiation pressure by these stars prevent any dust from staying in the vicinity of the stars. An upper limit of the degree of silicate crystallinity in the interstellar medium is estimated to be about 5% based on a comparison between a model and infrared observations (Li *et al.*, 2007). To assess the degree of silicate crystallinity in the interstellar medium, it would be necessary to investigate the balance between the nonthermal crystallization and amorphization by cosmic rays. More observations of crystalline silicate features in low-temperature environments will provide a clue to specify the temperature that triggers the nonthermal crystallization.

Our results show a possibility of efficient crystallization by nonthermal process. As stated in Section 2, our model permits amorphous silicate to crystallize above its melting temperature. If we exclude crystallization at higher temperatures than the melting temperature, we would have a lower volume fraction of crystalline silicate. On the other hand, our model neglects the finiteness of the particle size for the determination of the temperature. Actually the heat transmitted to the center of the particle cannot run away easily. As a result, the mean temperature of the particle would not decrease as we have modeled. We expect that this particle-size effect increases the volume fraction of crystal. Therefore, it is worth examining this nonthermal process more in detail by taking into account a more realistic crystallization model and the particle-size effect.

Crystallization is a process for atoms to overcome an activation energy of crystallization at an appreciable probability. In general, low-temperature crystallization is possible if (1) a significant amount of input energy is stored and (2) there is a trigger to release the stored energy to induce crystallization. d'Hendecourt *et al.* (1982) observed that the energy stored in their OR is released at ~ 30 K. Recently, Kaito *et al.* (2006) prepared *particles* of amorphous Mg silicate coated with amorphous carbon. They found that their crystallization temperatures are lowered substantially due

to the energy released by graphitization of amorphous carbon. Inclusion of CH_4 in the amorphous carbon mantle further reduces the crystallization temperature to room temperature (Kaito *et al.*, 2007). Although their experiments does not necessarily simulate crystallization of cometary silicate, they prove that a silicate core proceeds with forsterite crystallization upon releasing energy in its outer layer. The temperature to trigger the nonthermal crystallization could be further lowered, if the stored energy is released at much lower temperatures. Experiments on a search for mechanisms of low-temperature crystallization are encouraged to better understand the ubiquity of crystalline silicate in various objects.

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