Low Temperature Crystallization of Dust

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Crystalline silicate in various kinds of objects

- Observed in
  - evolved stars
    - C-rich giant stars, post AGB stars
  - Herbig Ae/Be stars
  - T Tauri stars
  - young MS stars
  - Comets
  - ZL dust
  - IDPs
  - other galaxies (ULIRG)

- Not observed in
  - ISM & molecular clouds
  - Protostars

(Molster et al. 1999)
Crystallization: Transition from amorphous to crystalline state

- L: latent heat of crystallization
- Must overcome energy barrier $E_c$ for crystallization

Internal energy: $E_{\text{crystal}} < E_{\text{amorphous}}$
Energy source to overcome Ec

Annealing induced by fluctuation of thermal energy
(Hallenbeck et al. 1998, Fabian et al. 2000, Kamitsuji et al. 2005, and many others)

But other energy sources:

• irradiation of electrons or high energy particles
  (Carrez et al. 2002, Y. Kimura et al. 2008)

• heat of chemical reactions
  (Yamamoto & Chigai 2005, Yamamoto+2010)
Basic idea of crystallization by heat of reactions

0) Suppose a silicate grain coated with a mantle including chemically reactive molecules (radicals).
1) Moderate heating induces chemical reactions in the mantle
2) heat of reactions leads to crystallization

(Yamamoto & Chigai 2005, Yamamoto+2010)
Series of experiments by Kaito et al. (Kamitsuji et al. 2005; Kaito et al. 2007a,b)

Exp.1
- Amorphous silicate
- Heating in vacuum
- $T \sim 1000\text{K}$
- D$_{\sim 100\text{nm}}$

Exp.2
- Amorphous silicate
- Amorphous carbon
- Heating in vacuum
- $T \sim 870\text{K}$
- D$_{\sim 10\text{nm}}$

Exp.3
- Amorphous silicate
- Amorphous carbon
- CH$_4$
- Exposed to the air
- $T \sim 300\text{K}$
- D$_{\sim 10\text{nm}}$
HREM image of the sample and its magnification of the interface region

crystallization of Mg silicate

Kaito et al. 2007b
Picture of crystallization in Exp. 3 by Kaito et al. (2007b)

1. Release of heat of chemical reactions (oxidation of CH₄) in C mantle

2. Induce graphitization in the C mantle

3. Latent heat deposited by graphitization leads further temperature elevation

4. Induce crystallization of silicate from the interface of C mantle and silicate core

5. Cessation of crystallization due to cooling
Modelling of Exp. 3

Time variation of temperature of the particle

\[ \frac{4}{3} \pi a^3 \rho c \frac{dT}{dt} = \frac{4}{3} \pi [(a + h)^3 - a^3] \dot{\varepsilon} - \Lambda_{\text{air}} - \Lambda_{\text{rad}} + H_{\text{si}} + H_{\text{c}} \]

Rate of generation of heat of reactions in mantle

\[ \dot{\varepsilon} = -\frac{dn_{\text{rad}}}{dt} E_r \quad \frac{dn_{\text{rad}}}{dt} = -\nu_r e^{-E_{\text{ar}}/kT} n_{\text{rad}} \]

Crystal growth

\[ \frac{d\alpha_{\text{sil}}}{dt} = a_0 \nu e^{-E_{\text{sil}}/kT} (1 - e^{-q_{\text{sil}}(T_m - T)/kT}) \]
Key physical quantities

- Stored energy density: $Q = n_{\text{rad}}(0) E_r$
- Time scale of the reactions: $\tau$
- Ambient gas pressure
Feature of crystallization

( $n_{\text{rad}}(0) \, E_r = 10^{27} \text{ K cm}^{-3}; \, t_r = 10^{-8} \text{ s, gas density} = 10^{-3} \text{ g cm}^{-3} = 1 \text{ atm} \)
Feature of crystallization (2)
small deposition of heat of reaction

\( n_{\text{rad}} \) \( E_r = 0.9 \times 10^{27} \text{ K cm}^{-3} \); \( t_r = 10^{-8} \text{ s} \), gas density = \( 10^{-3} \text{ g cm}^{-3} = 1 \text{ atm} \)
(a) $Q=1.1 \times 10^{27}$ Kcm$^{-3}$

(b) $Q=1.0 \times 10^{27}$ Kcm$^{-3}$

(c) $Q=0.9 \times 10^{27}$ Kcm$^{-3}$

\[ \text{(P = 1 atm)} \]
Low gas density, large energy deposition, fast reactions easy crystallization

Radical concentration >1 - 10% leads to substantial crystallization
Crystallization degree in steady accretion disks

\( Q = n_{rad}(0) \, E_r = 10^{27} \, \text{K cm}^{-3} \)

Crystallization degree

- Complete crystallization
- Partial crystallization
- No crystallization

Crystalline region is much extended than by annealing.

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Concluding remarks (1/3)

- Crystallization triggered by chemical reactions may explain ubiquity of crystalline silicate in various objects

  Similar phenomenon

  - Wigner energy release known in nuclear reactor engineering
  - Sudden release of energy stored in graphite moderator irradiated by neutrons upon moderate heating (Spontaneous energy release)
Concluding remarks (2/3)

- The present mechanism does not depend on the details of the chemistry but depends only on the amount of reactive molecules times heat of reactions, $Q = n_{\text{rad}}(0) E_r$, and reaction timescale $\tau$. 
Concluding remarks (3/3)

• Similarity of ice compositions in molecular clouds and comets:
  - Present mechanism can explain the coexistence of crystalline silicate and ice of IS composition in comets without mixing.

• Search for crystalline silicate at low temperature environments is encouraged by future high resolution observations of disks.