

# 非晶質シリケートの結晶化実験による 星周塵化学組成の制限

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## MENU

### ・ 村田D論の紹介

Murata et al. (2007) *ApJ* 668, 285

Murata et al. (2009) *ApJ* 696, 1612

Murata et al. (2009) *ApJ* 697, 836

(Murata et al. (2009) *ApJ* 698, 1903)

### ・ 星周塵化学組成の議論



# Circumstellar and interstellar dust

- Evolved stars (e.g., Waters et al., 1996)

  - Amorphous silicates (AS) (~95 %)

  - Crystalline silicates (~5 %)

    - olivine (OL):**  $(\text{Mg,Fe})_2\text{SiO}_4$  Mg#(=Mg/(Mg+Fe)) > 0.9

    - pyroxene (PX):**  $(\text{Mg,Fe})\text{SiO}_3$

    - Amorphous silicates condensed from gas → Crystallization**  
(Seki & Hasegawa, 1981; Gail, 1999; Rietmeijer et al., 1999)

- Interstellar medium (Kemper et al., 2004)

  - Amorphous silicates (AS)

- Young stars (e.g., Waelkins et al., 1996)

  - Amorphous silicates (AS) (80-90%)

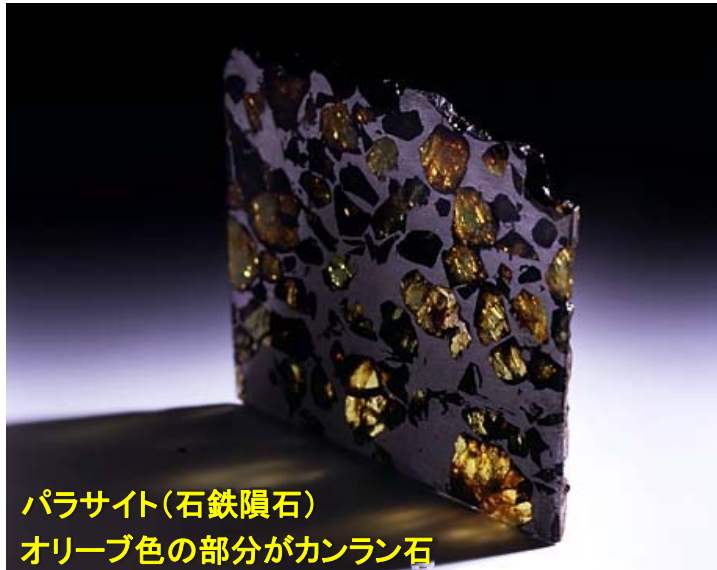
  - Crystalline silicates (10-20%)

    - olivine (OL):**  $(\text{Mg,Fe})_2\text{SiO}_4$  Mg# > 0.9(?)

    - pyroxene (PX):**  $(\text{Mg,Fe})\text{SiO}_3$

    - Crystallization of interstellar amorphous silicates**  
(Bockelee-Morvan et al., 2002; Hallenbeck et al., 1998)

# 宇宙における鉱物 (crystalline silicates) の主役たち



カンラン石 (olivine)  
 $(\text{Mg,Fe})_2\text{SiO}_4$



Caに乏しい輝石 (Ca-poor pyroxene)  
 $(\text{Mg,Fe})\text{SiO}_3$

# Crystallization experiments of amorphous silicates (AS)

Table 1 Chemical compositions of starting materials and CI and GEM compositions.

Composition	A	B'	C	C'	Fo-n	En-n	En80	En80-g	CI	GEMS	GEMS <sup>%</sup>
	gel <sup>#</sup>	gel <sup>#</sup>	gel <sup>#</sup>	gel <sup>#</sup>	TP <sup>\$</sup>	TP <sup>\$</sup>	gel <sup>#</sup>	glass <sup>&amp;</sup>			w/o metal
Mg	1.07	1.07	1.07	1.07	2	1	0.8	0.8	1.07	0.663	0.663
Si	1	1	1	1	1	1	1	1	1	1	1
Fe	0.9	0.39					0.2	0.2	0.9	0.461	0.06
Al	0.085		0.085						0.085	0.084	0.084
Ca	0.061		0.061						0.061	0.025	0.025
Na	0.057		0.057						0.057	n.d	n.d.
Ni	0.049		0.049						0.049	0.033	
S									0.52	0.165	0.165
O	4.24	3.46	3.34	3.07	4	3	3	3	4.24	3.31	2.87
Mg#=Mg/(Mg+Fe)	0.54	0.73	1	1	1	1	0.8	0.8	0.54	0.59	0.92
NBO*	-0.47	1.08	1.33	1.86	0	2	2	2	-0.47	1.38	2.25
Major phase**	ol	ol	ol (fo)	px (en)	ol (fo)	px (en)	px	ol			
Reference	[1]	[2]	[3]	[4]	[5]	[6]	[7]	[7]	[8]	[9]	

\* Number of bridging oxygen

\*\* ol: olivine, px: pyroxene, fo: forsterite, en; enstatite

#: Synthesized by sol-gel method

\$: Glass nanoparticles (~100 nm) produce by condensation from R-F induction thermal plasmas

&: Glass quenched from melt

?: (Fe<sub>0.9</sub>Ni<sub>0.1</sub>) metal is removed.

[1] Murata K. et al. (2007) ApJ, 668, 285.

[2] Murata K. et al. (2009) ApJ. 696, 1612.

[3] Noguchi R. et al. (2009) Japan Geoscience Union Meeting 2009, abstract.

[4] Murata K. et al. (2009) ApJ, 697, 836.

[5] Imai Y. et al. (2009) Japan Geoscience Union Meeting 2009, abstract.

[6] Imai Y. (2009) private communication.

[7] Seidler S. (2009) private communication.

[8] Anders and Grevesse (1989)

[9] Bradely (1988, 1994ab) and Bradely and Ireland (1996)



# Crystallization experiments of amorphous silicates (AS)

Solar abundance  
 Si:Mg:Fe~1:1:1  
 S~0.5  
 Al, Ca, Ni, Na<0.1  
 O>C

## Primitive amorphous silicates

- (1) CI (=solar abundance)
- (2) GEMS (Glass embedded with metal and sulfides)

## Amorphous silicate SM

- (1) CI and derivatives
  - A: CI
  - B: CI-FeS ( $\text{FeO} = \sum \text{Fe} - \text{FeS}$ )
  - C: CI- $\sum \text{Fe}(\text{Fe} + \text{FeS}; \text{FeO} = 0)$
- (2) Olivine composition
  - Fo:  $\text{Mg}_2\text{SiO}_4$
- (3) Pyroxene composition
  - En:  $\text{MgSiO}_3$
  - En80:  $(\text{Mg}_{0.8}\text{Fe}_{0.2})\text{SiO}_3$

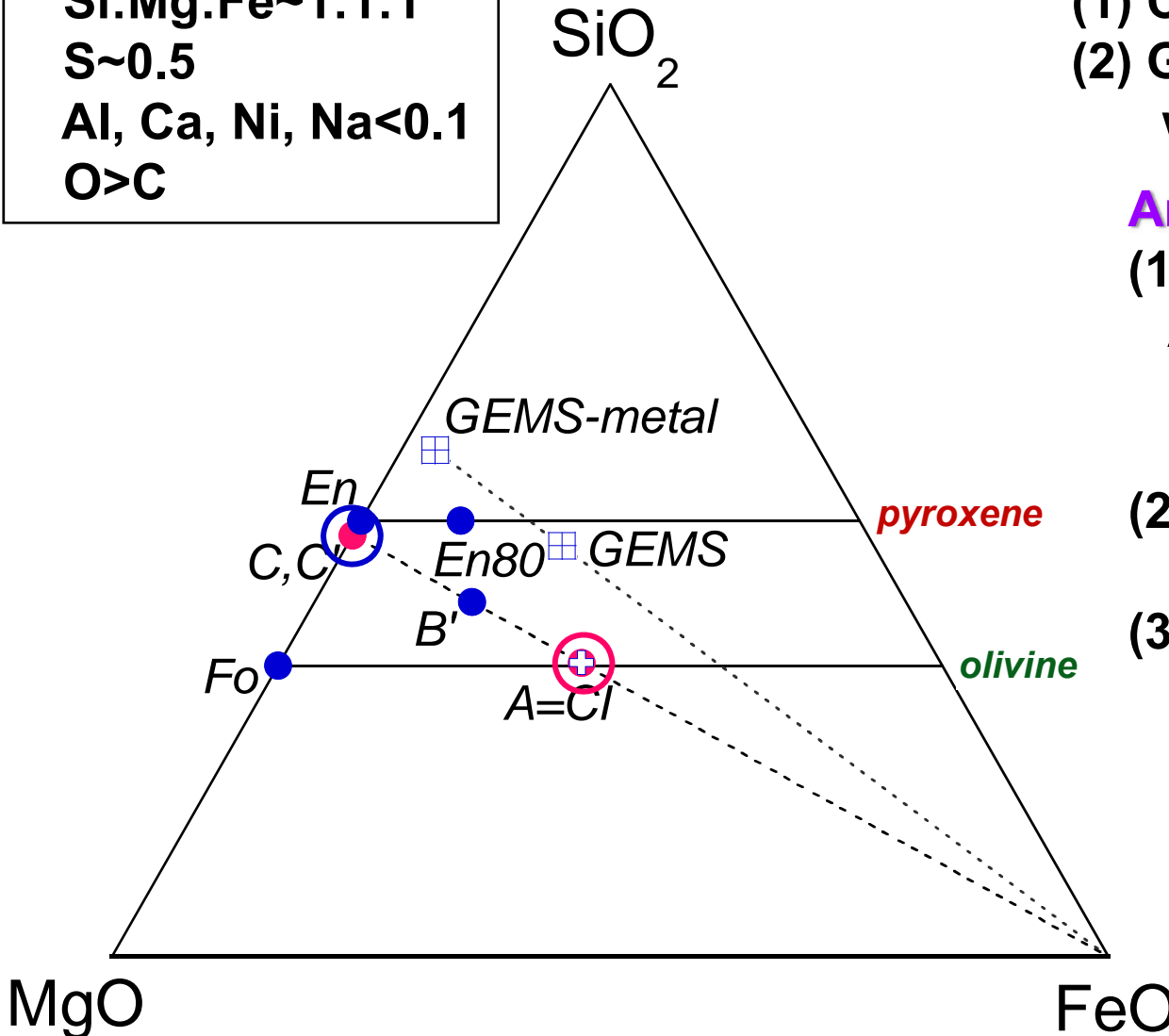
## SiO<sub>2</sub>-MgO-FeO-X system

(X: Al<sub>2</sub>O<sub>3</sub>-CaO-NiO-Na<sub>2</sub>O)

A, C

## SiO<sub>2</sub>-MgO-FeO system

B', C'



# Crystallization time scale and activation energy

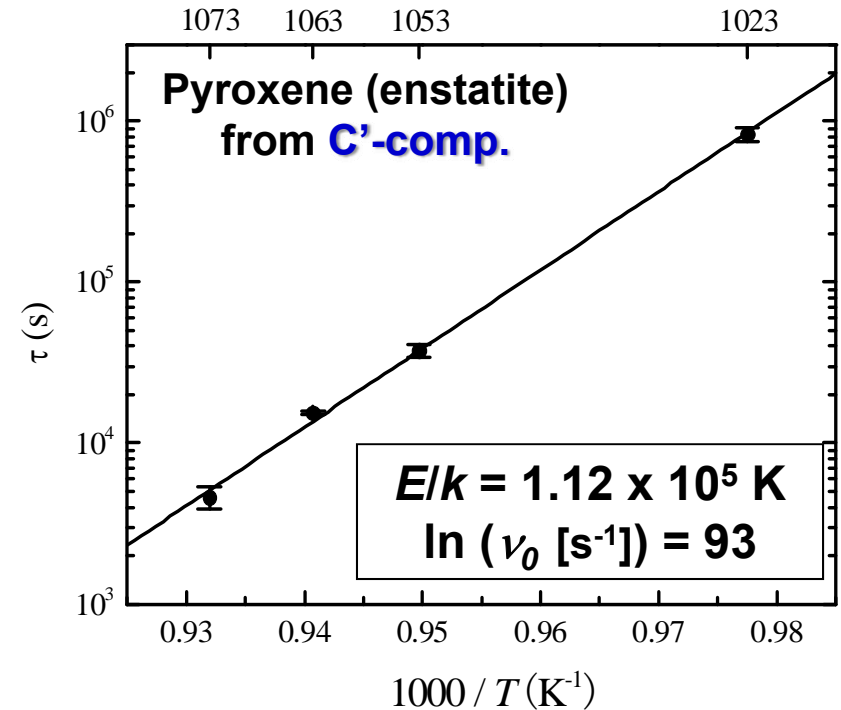
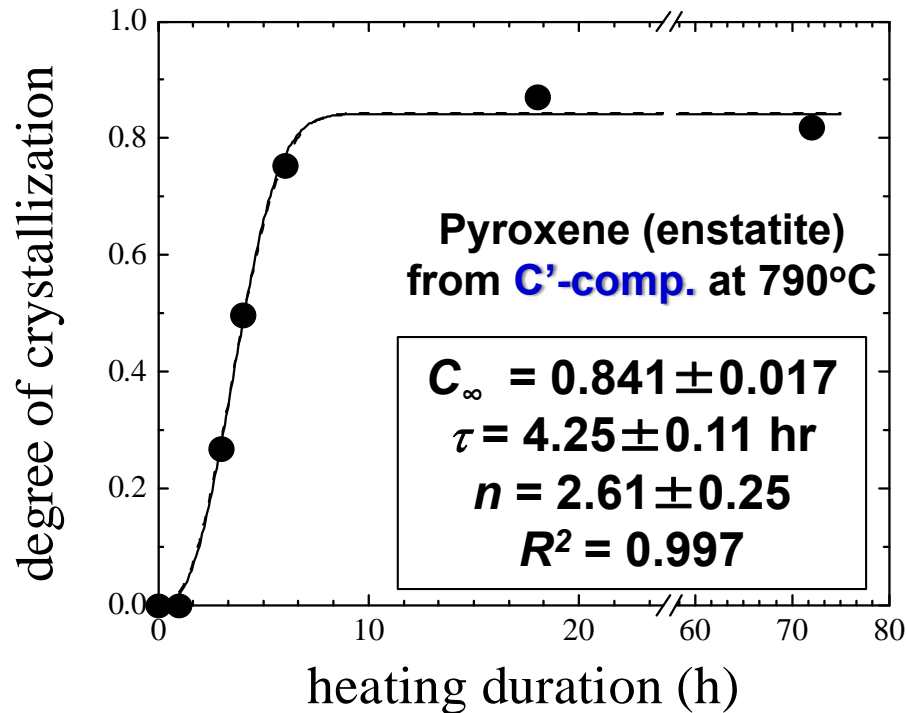
OL crystallization (**A-comp.**: Murata et al., 2007, *ApJ* 668, 285)

PX crystallization (**C'-comp.**: Murata et al, 2009b, *ApJ* 697, 836)

Degree of crystallization,  $C$ ,  $\Leftarrow$  IR spectrum

Crystallization time scale,  $\tau$ ,  $\Leftarrow$  Fitting of time evolution,  $C(t)$ , by JMA eq.

Activation energy,  $E$ , and pre-exponent term,  $\nu_0 \Leftarrow$  Arrhenius plot of  $\tau$   
 $T(K)$



Experiments	NBO	$E/k$ (K)	$\ln \nu_0$ ( $\text{s}^{-1}$ )	mode of crystallization
OL from <b>A</b>	-0.47	$\sim 6 \times 10^4$	$\sim 52$	growth from pre-exist. xst.
PX from <b>C'</b>	1.86	$1.12(3) \times 10^5$	93(2)	nucleation and growth

# Crystallization around evolved and young stars

**TTT (Time-Temperature-Transformation) diagram** for ol & px crystallization

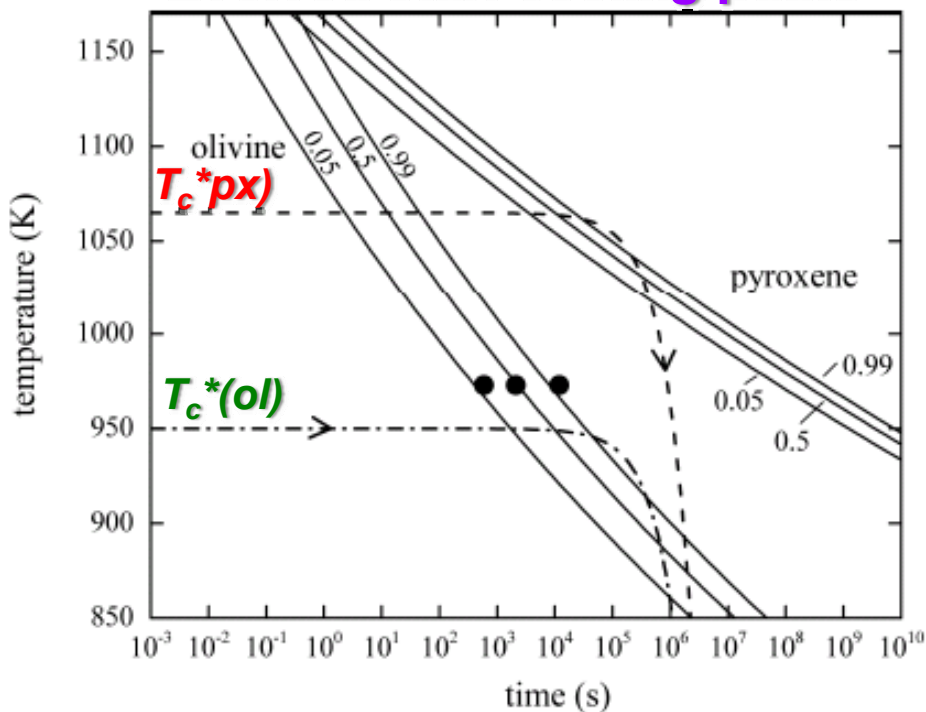
Degree of crystallization,  $C$ ,  $\Leftarrow$  IR spectrum

Crystallization time scale,  $\tau$ ,  $\Leftarrow$  Fitting of time evolution,  $C(t)$ , by JMA eq.

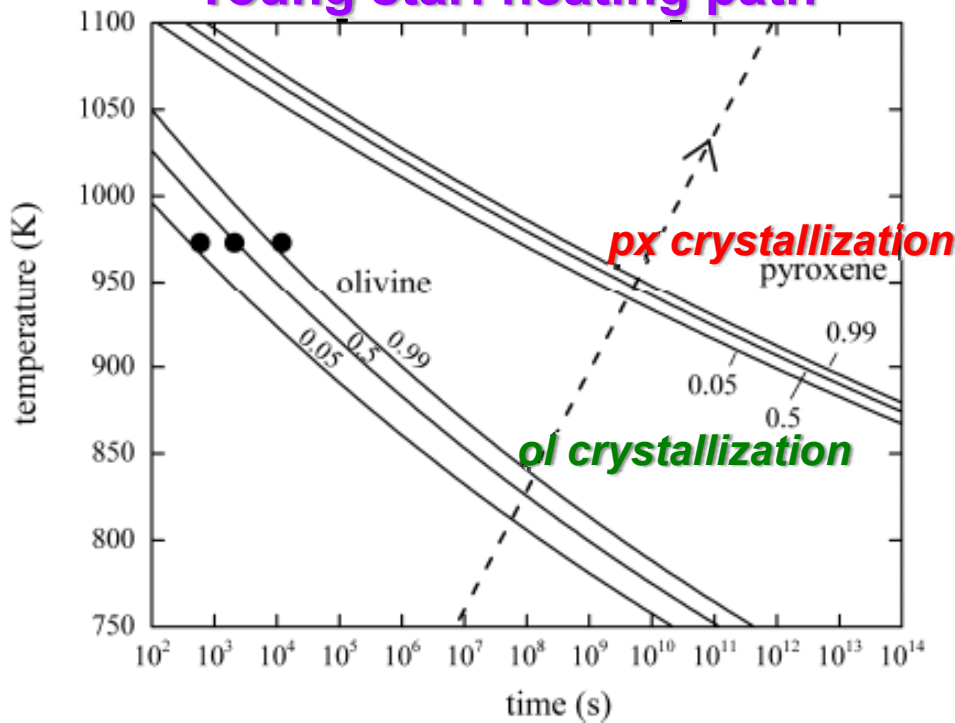
Activation energy,  $E$ , and pre-exponent term,  $\nu_0$   $\Leftarrow$  Arrhenius plot of  $\tau$

Experiments	NBO	$E/k$ (K)	$\ln \nu_0$ ( $s^{-1}$ )	mode of crystallization
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**Evolved star: cooling paths**

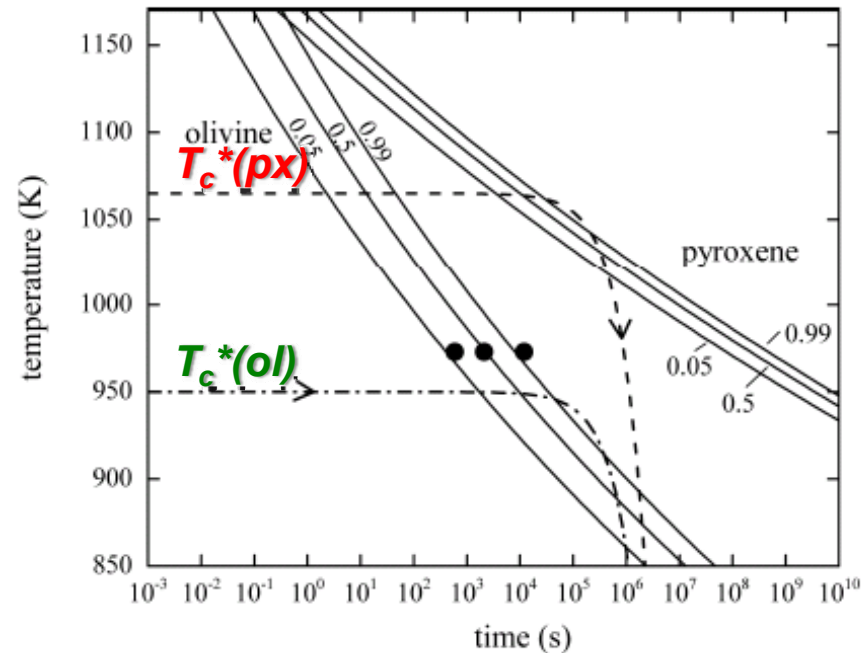
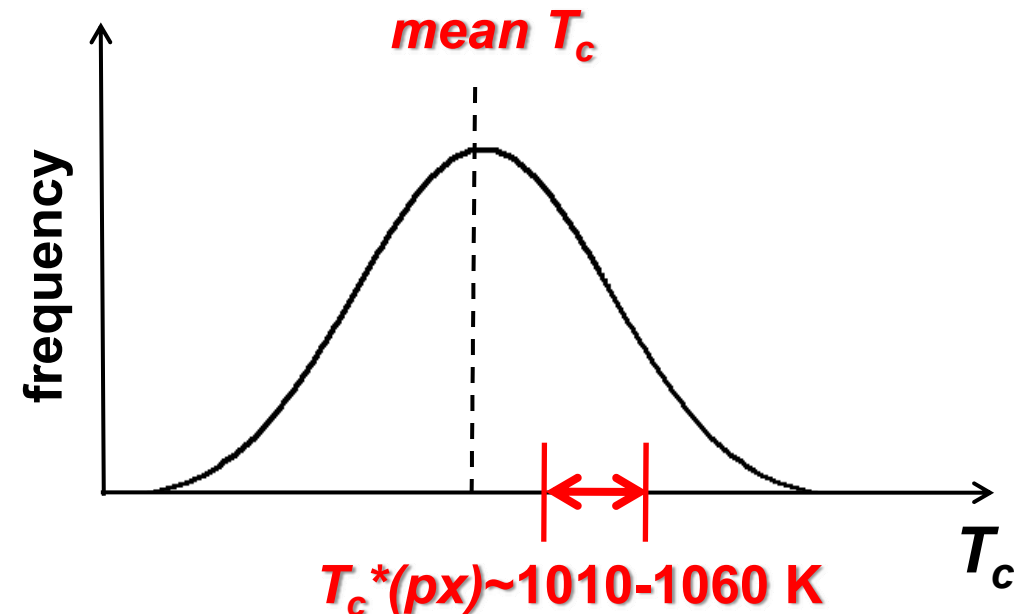
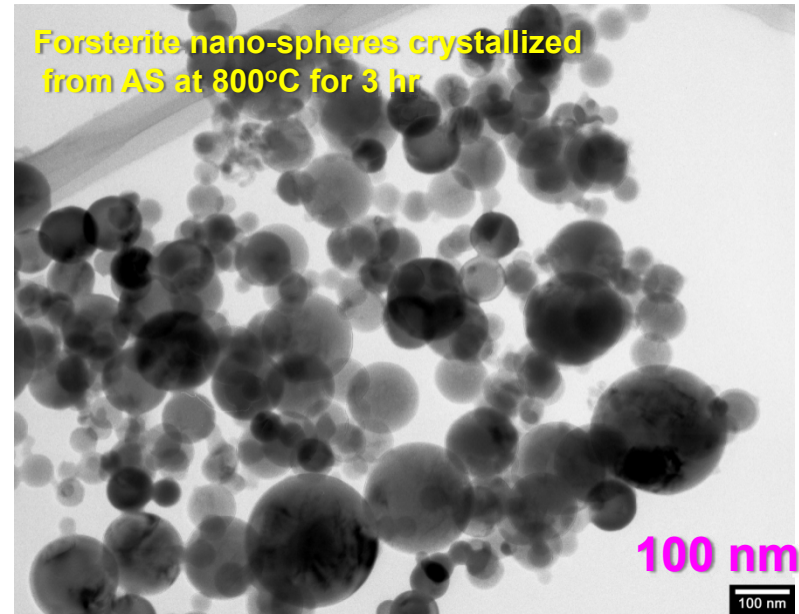


**Young star: heating path**



# Crystalline dust around evolved stars

- Condensation of **AS as spheres** followed by
- **Partial crystallization** of the spheres can explain following observations:
  - Population of crystalline dust (~5 %)
  - Unidentified 33  $\mu\text{m}$  feature in the IR spectra for olivine



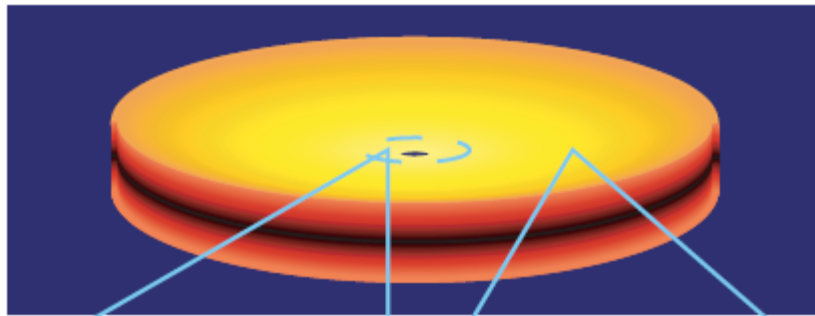
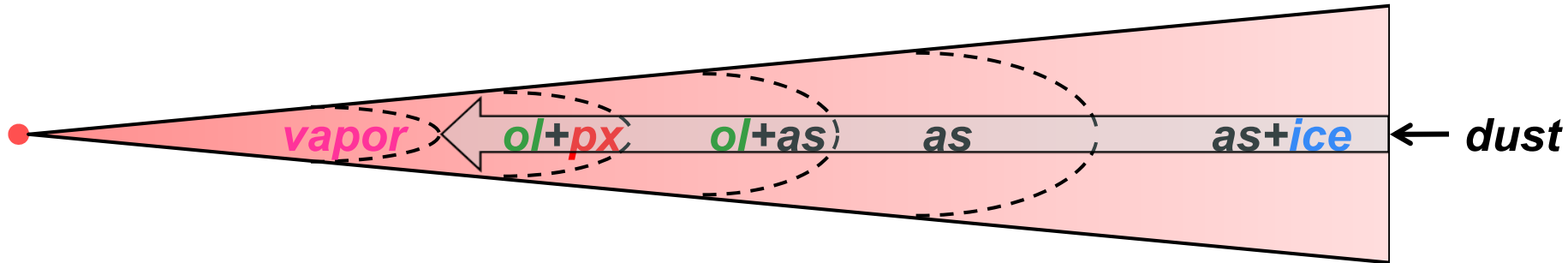


# Crystalline dust around and young stars

- Narrow temperature interval for crystallization for AS dust falling towards the central star

OL:~30°C, PX:~10°C

⇒ Zonal distribution of AS, OL and PX along the heliocentric distance



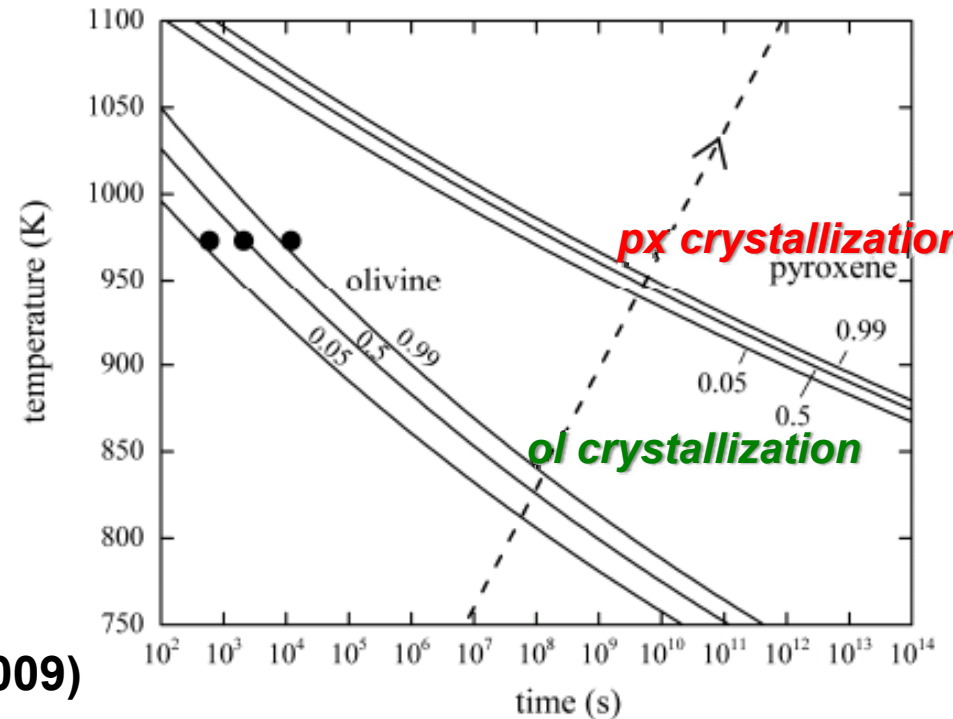
Inner disk

Outer disk

pyroxene-rich



olivine-rich



Watson et al. (2009), Sergent et al. (2009)

# 非晶質宇宙塵の化学組成への制約

## (1) Mg-richな星周塵結晶質ケイ酸塩の化学組成

Evolved stars: Mg#>0.9

Young stars: mostly Mg#>0.9

⇒ ASとOL間のFe-Mg元素分配実験  
(Murata et al, 2009a, *ApJ* 696, 1612)

## (2) 星周塵におけるOLとPXの共存

Evolved stars: ?

Young stars: Mixture of SiO<sub>2</sub>-poor and rich ASs

⇒ 結晶化実験: 出発物質(AS)の化学組成依存性

OL/PX crystallization vs. **NBO**

**NBO**: Number of bridging oxygen (架橋酸素数)

# ASとOL間のFe-Mg元素分配

OL crystallization (B'-comp.: Murata et al., 2009a, *ApJ* 696, 1612)

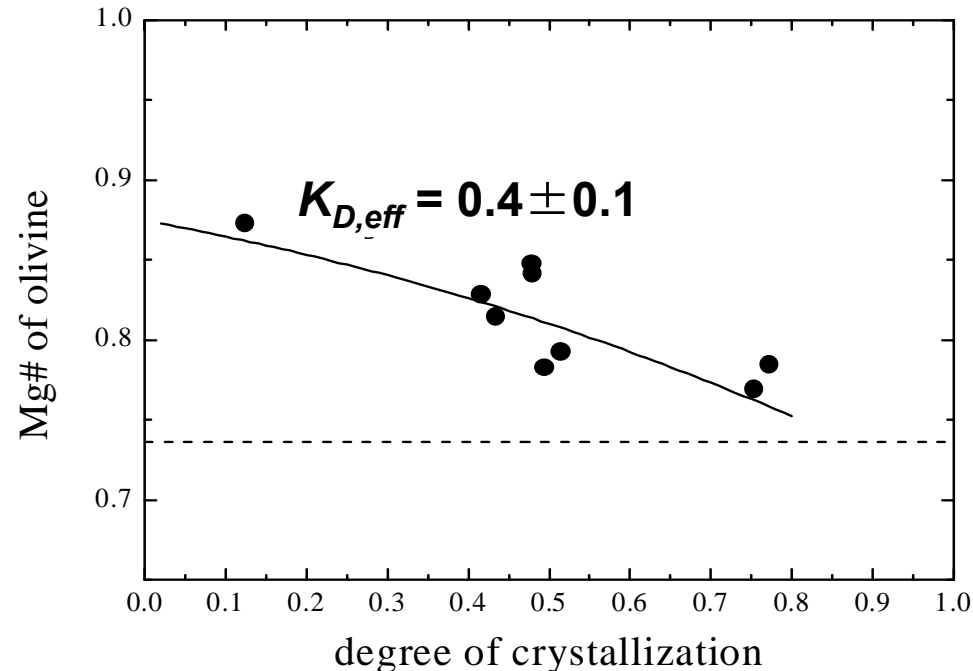
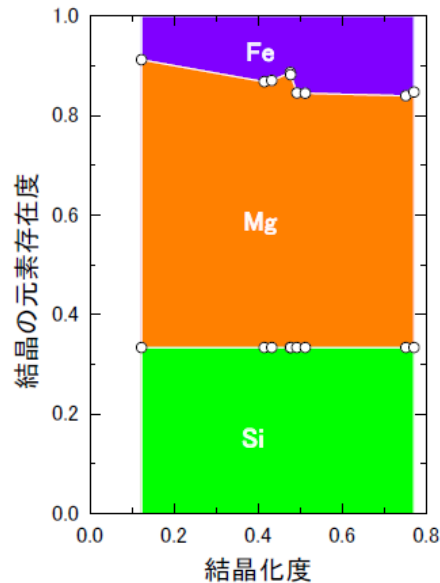
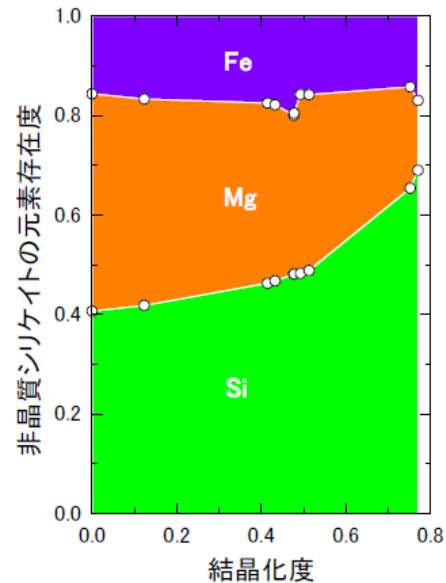
Degree of crystallization,  $C$ ,  $\Leftarrow$  IR spectrum

Mg# of olivine as a function of  $C$ ,  $\Leftarrow$  XRD

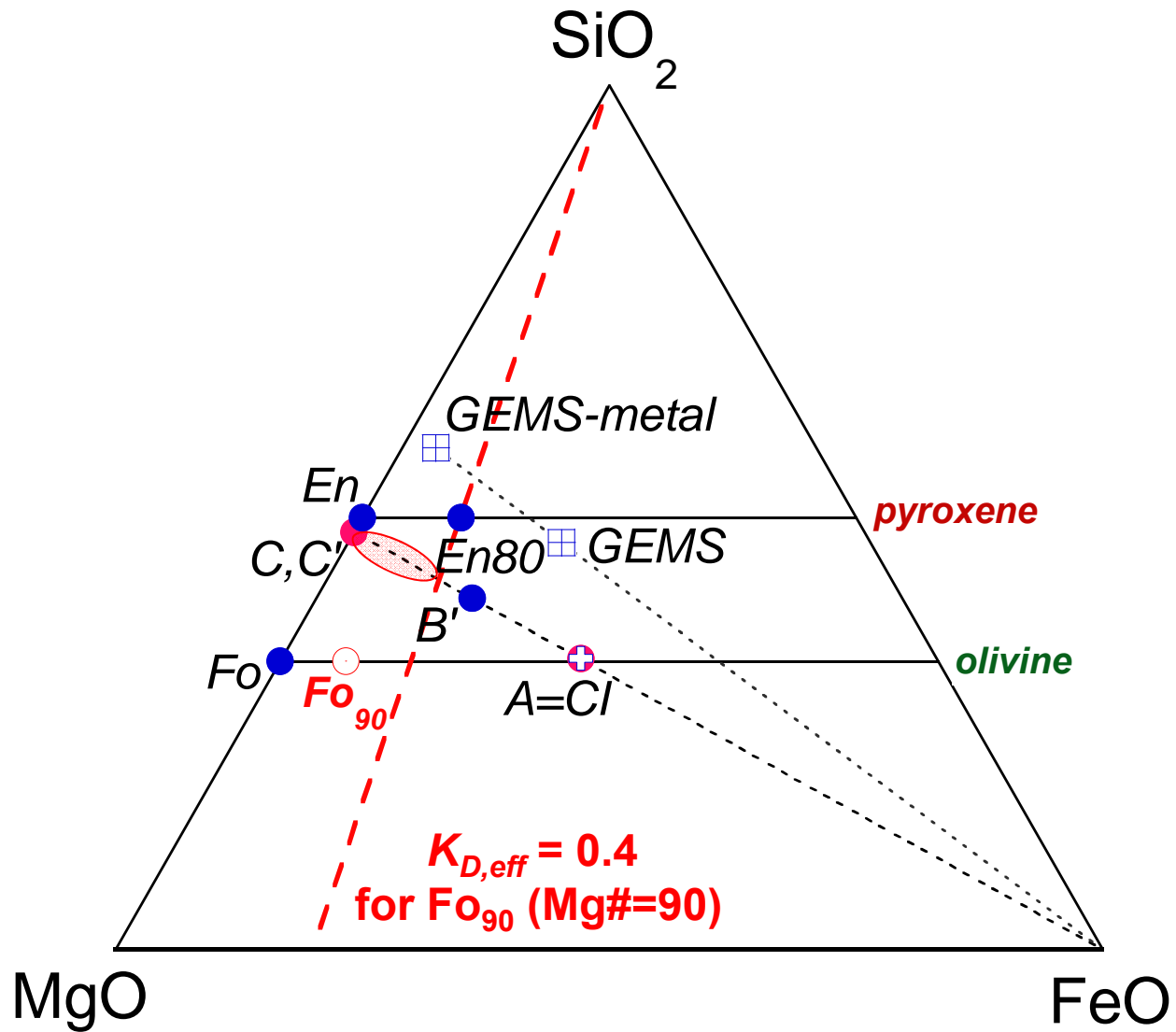
Chemical composition of residual AS (Mg#) as a function of  $C$

$\Rightarrow$  **Effective distribution coefficient** of Mg-Fe between OL and AS,  $K_{D,eff}$

$$K_{D,eff} = \frac{(Mg/Fe)_{AS}}{(Mg/Fe)_{OL}} = 0.4 \pm 0.1$$



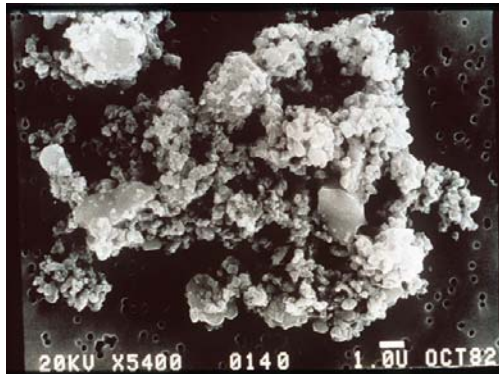
# Mg-richな星周塵結晶質ケイ酸塩の化学組成



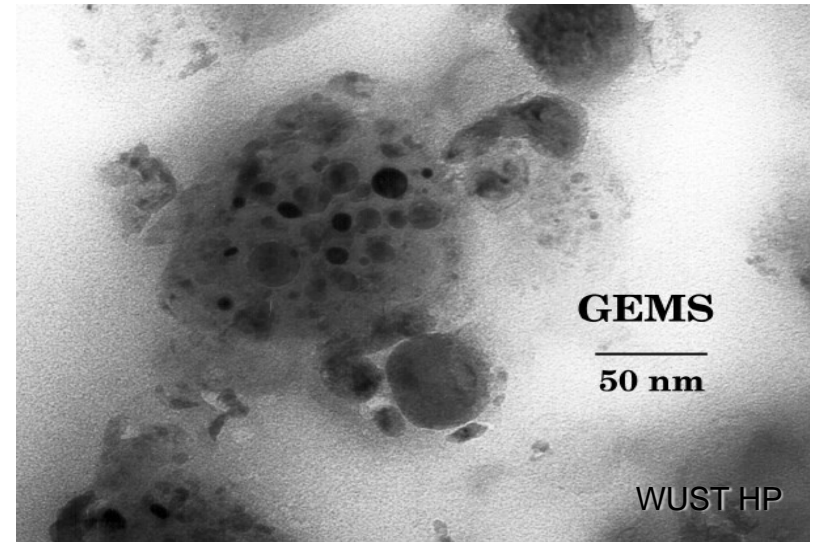
# GEMS

**GEMS** (glass with embedded metal and sulfides) (Bradley, 1995)  
彗星起源の宇宙塵(無水惑星間塵)を特徴づける始原的な非晶質ケイ酸塩

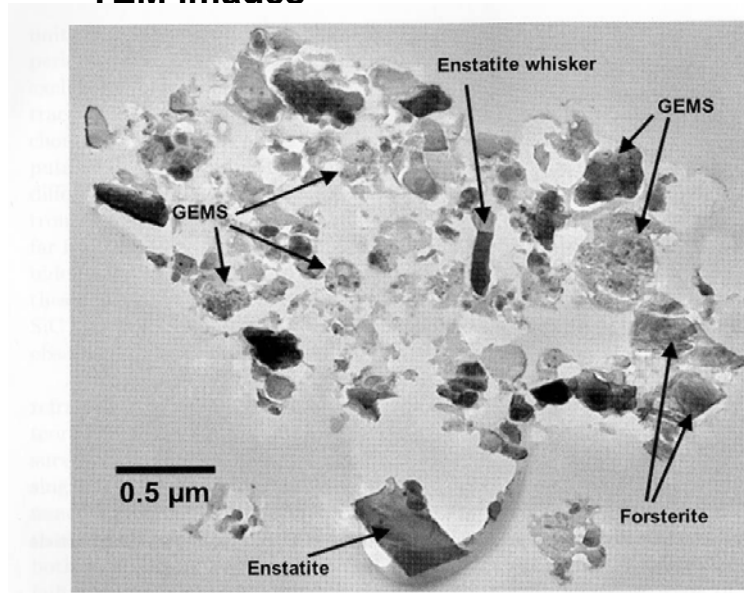
SEM images



TEM images



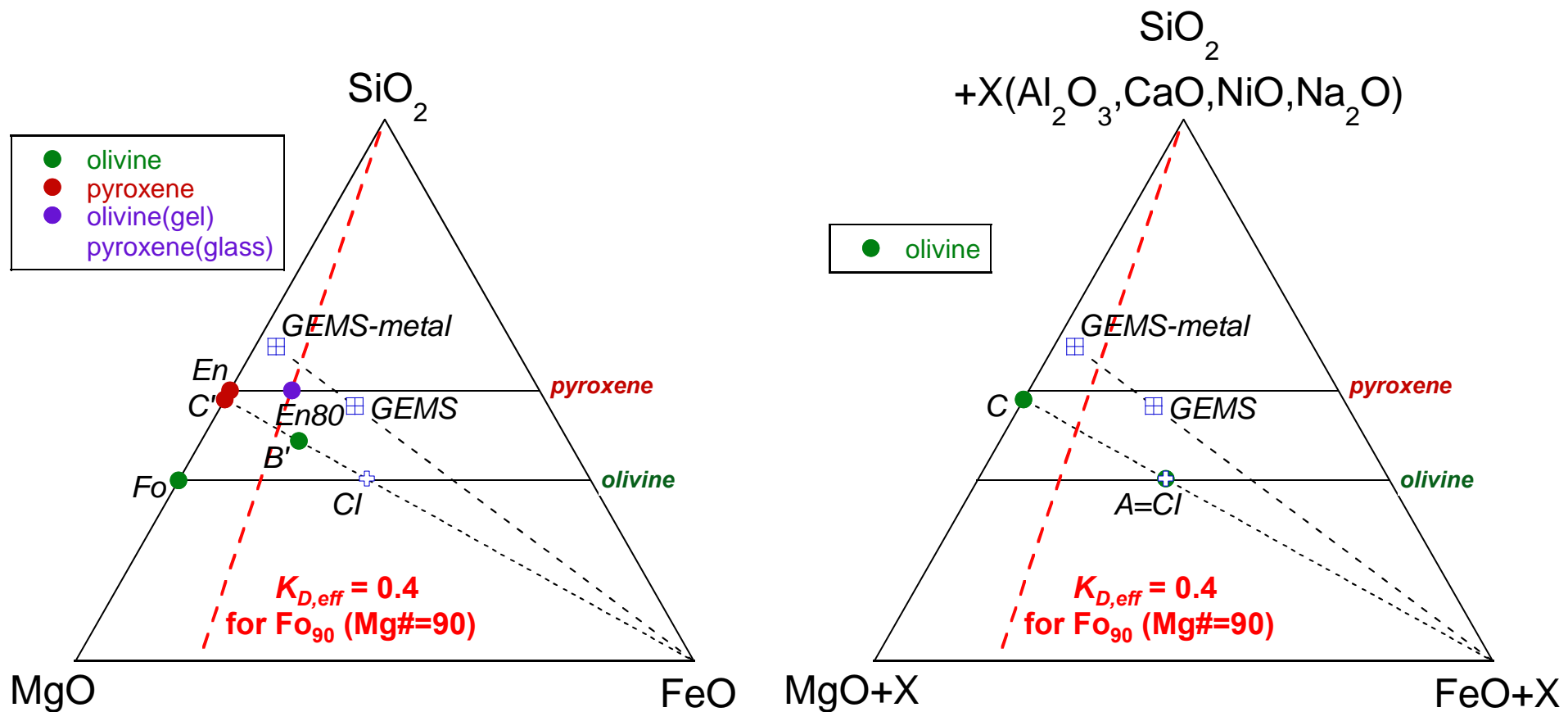
TEM images



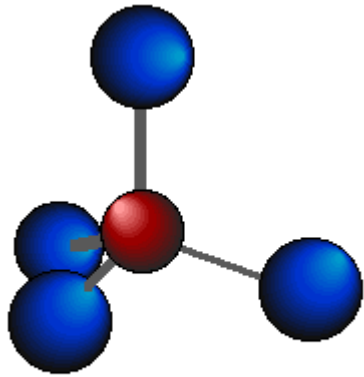
Bradley (2003)



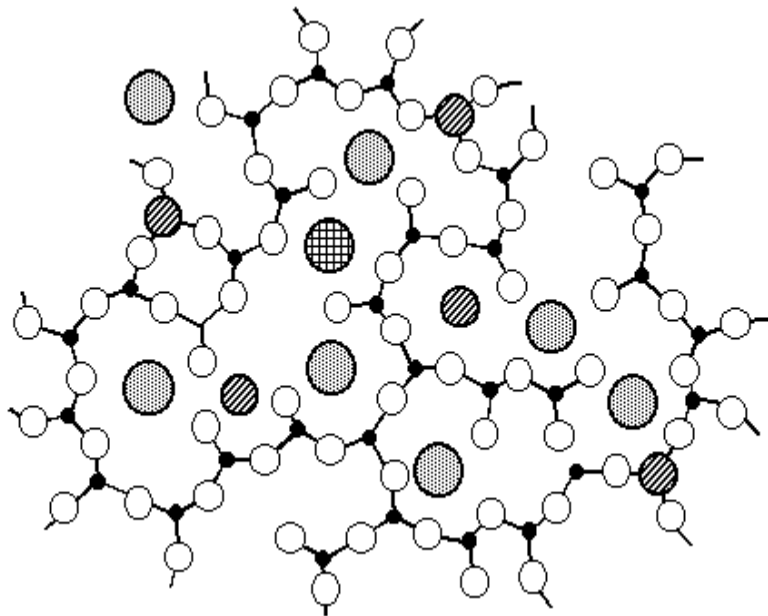
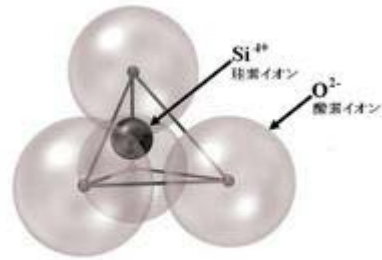
# 出発物質(AS)の化学組成依存性: OL/PX?



# 非晶質/結晶質シリケートの構造



$[\text{SiO}_4]^{4-}$  配位4面体



非晶質シリケートの模式的な構造

- 珪素
- 酸素
- 修飾陽イオン  
例: Na
- 中間陽イオン  
例: Ca

陽イオン:  
 $\text{Mg}^{2+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Na}^+$ ...



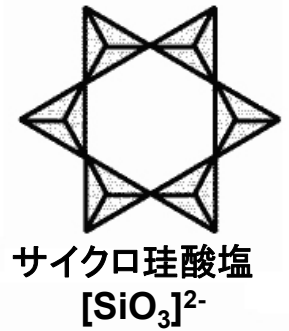
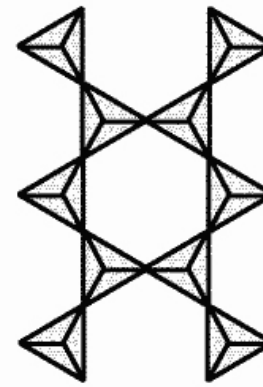
ネソ珪酸塩  
 $[\text{SiO}_4]^{4-}$



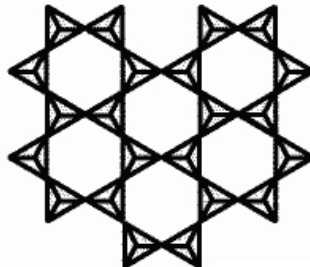
ソロ珪酸塩  
 $[\text{SiO}_{3.5}]^{3-}$



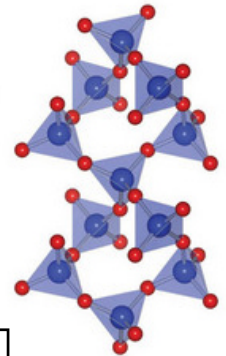
イノ珪酸塩  
 $[\text{SiO}_3]^{2-}$ ,  $[\text{SiO}_{2.5}]^-$



サイクロ珪酸塩  
 $[\text{SiO}_3]^{2-}$



フィロ珪酸塩  $[\text{SiO}_{2.5}]^-$



テクト珪酸塩  
 $[\text{SiO}_2]$

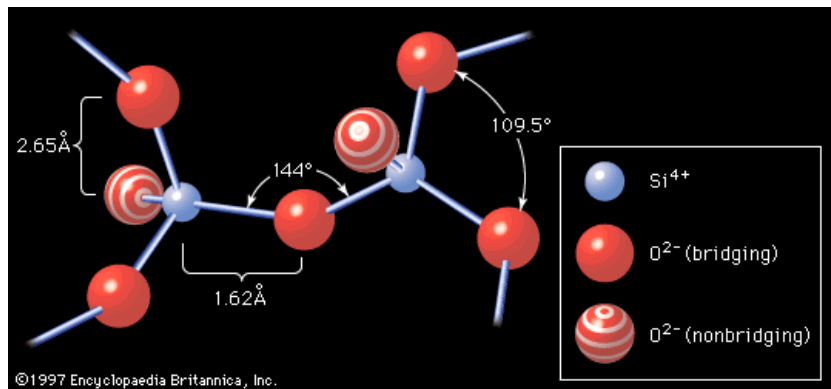
珪酸塩鉱物(結晶)の分類

# NBO(Number of Bridging Oxygen) 架橋酸素数

架橋酸素(bridging oxygen): 2つの $\text{SiO}_4$ 四面体を共有する酸素(Si-O-Si結合)

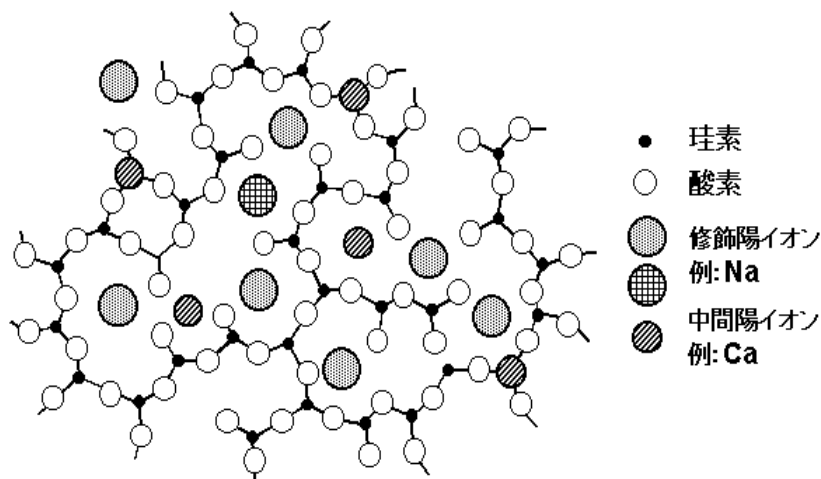
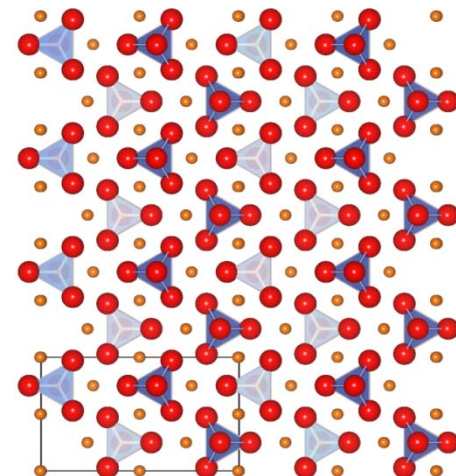
非架橋酸素(non-bridging oxygen):  $\text{SiO}_4$ 四面体を共有しない酸素

架橋酸素数(NBO): Si原子1個あたりの架橋酸素の平均数



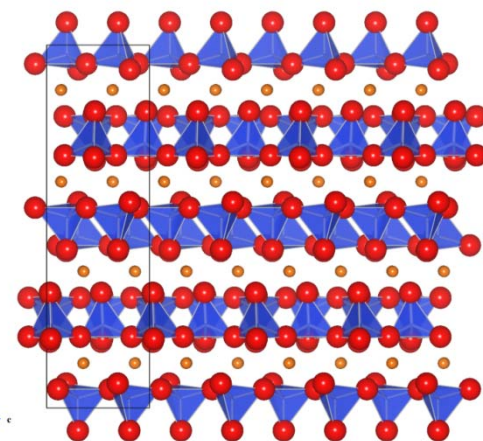
olivine

NBO=0  
(ネソケイ酸塩)

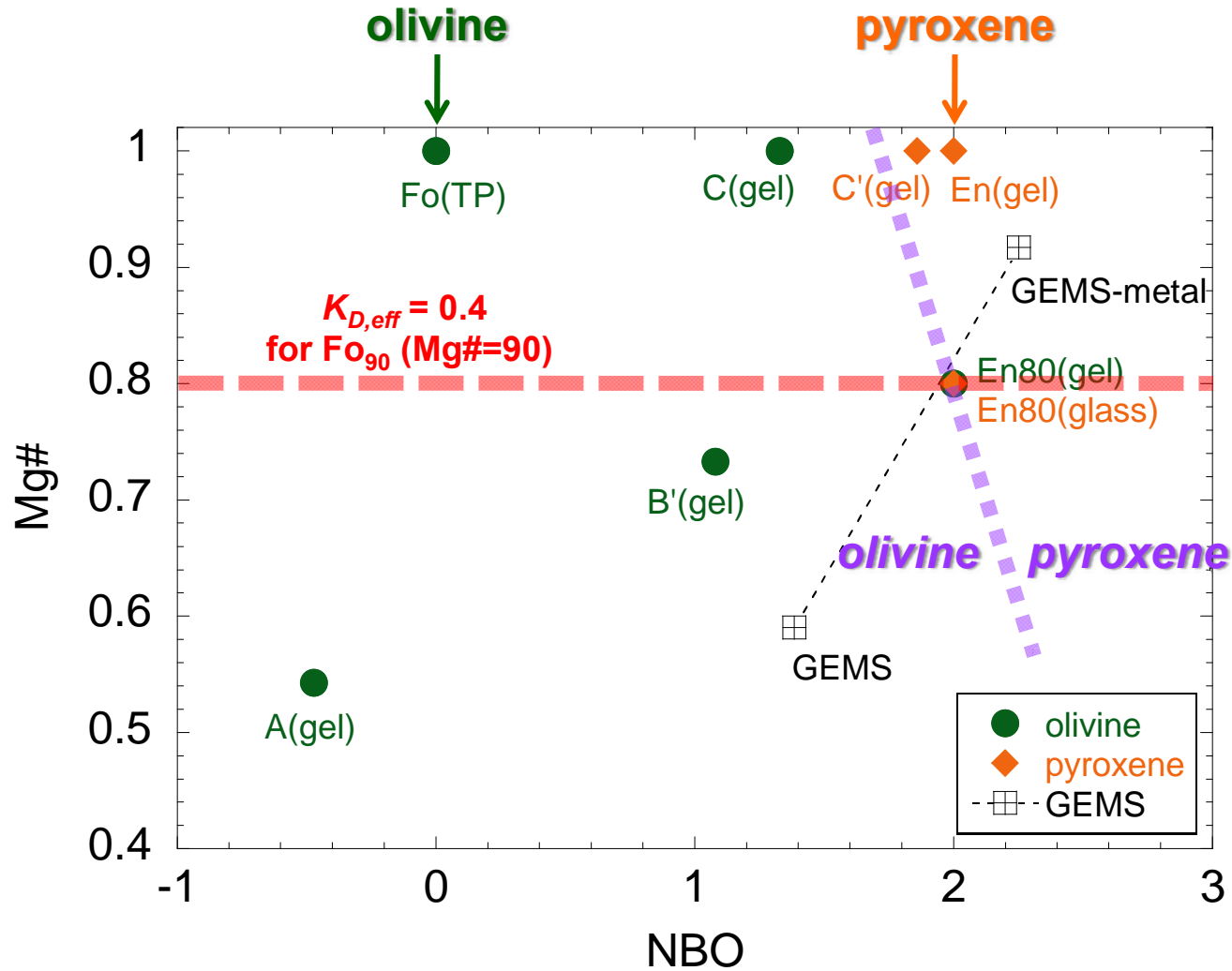


pyroxene

NBO=2  
(イノ珪酸塩, chain silicate)



# Olivine/pyroxene crystallization vs. NBO



# 晩期星星周塵でのOL/PXの共存

- 非晶質シリケートの化学組成:  
ほぼ一定の場合

NBO~1.5-2

AS の構造 (凝縮温度などに依存) で OL/PX

Cl組成 (-Fe) よりもやや SiO<sub>2</sub>-rich ?

(GEMS的) ?

- 非晶質シリケートの化学組成:  
不均一の場合

NBO < 1.5 ~ > 2 まで変化

組成変化の原因:

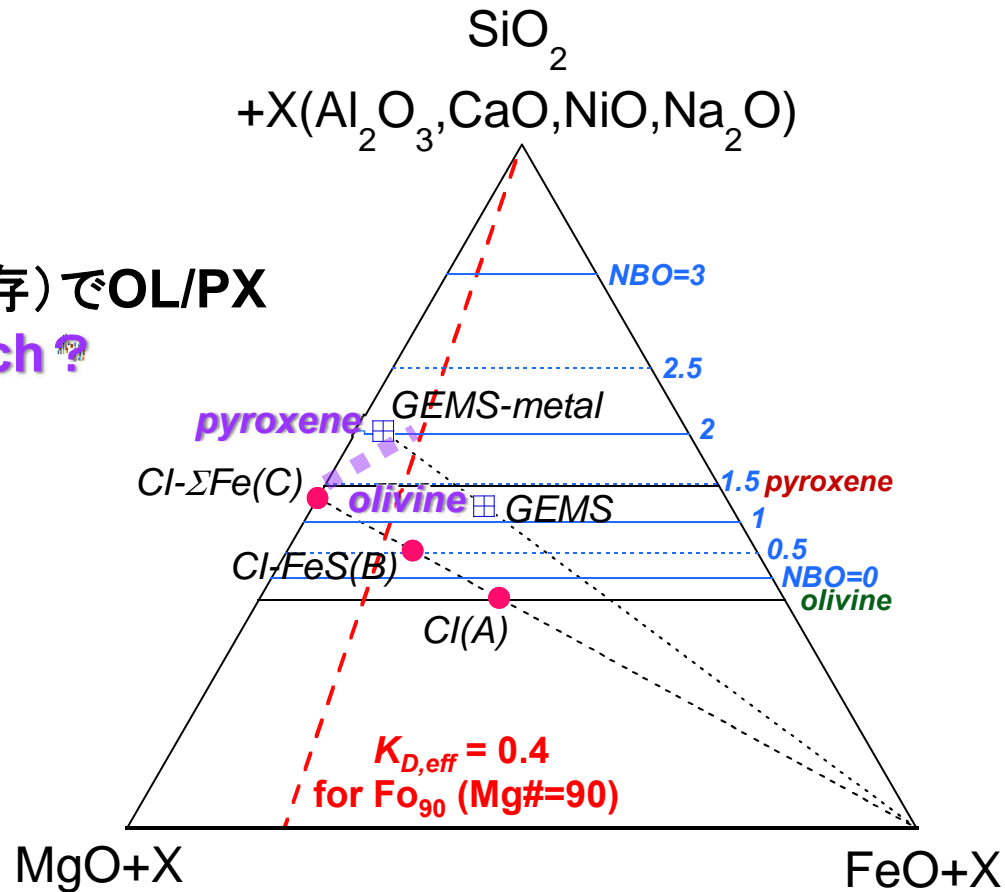
摂動 or 時間変化 ?

- いずれにせよ

Fe の少なくとも一部は FeS, metal として存在 (low FeO)

Mg-rich OL/PX だけでなく

PX の結晶作用も説明できない





# まとめ

- 星周塵(晩期星/若い星)に観測される結晶質シリケート(OL/PX)は非晶質シリケートの加熱による結晶作用によって生成されたという**作業仮説**の基で、非晶質シリケートの結晶化実験から**星周塵の化学組成**を議論した。
- Murata (2009, D-thesis) のレビューをおこなった。**作業仮説**により、晩期星星周塵における結晶質シリケートの割合が説明できる。  
(Murata et al., 2009b)。  
若い星で観測されたOL/PX分布(Watson et al., 2009; Sergent et al., 2009)が説明できる。
- ASのMg#は、OL-AS間のFe-Mg分配実験(Murata et al., 2009a)より、制限を受ける(**Mg#>0.8**)。
- OL/PXのどちらが主要な結晶相であるかを、ASの**NBO**(架橋酸素数)によりおおよそ説明できる。
- 以上より、星周塵の平均的な化学組成は**ClよりSiO<sub>2</sub>-rich**である可能性があること、Feの多くの部分はFeSやmetalとして存在している(**low-FeO組成**)ことが示唆される。