

CRYSTALLIZATION OF EVAPORATING FORSTERITE-RICH MELTS: TEXTURE AND MAGNESIUM AND SILICON ISOTOPIC COMPOSITIONS OF THE EVAPORATION RESIDUES.

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Introduction: Isotopic studies of Type B CAIs show correlated enrichments of heavy silicon, magnesium and oxygen isotopes by several per mil [1]. FUN CAIs extend this trend toward much larger values with $\delta^{26}\text{Mg}$ up to ~80‰ and $\delta^{30}\text{Si}$ up to ~30‰ [1-3]. These large isotopic fractionations, along with the coarse-grained igneous textures of Type B and most FUN CAIs, indicate that their precursors have experienced high-temperature melting and evaporation. In our isothermal evaporation experiments using Type B CAI-like melts [4, 5] and forsterite-rich melts [6, 7], we were able to produce residues with bulk chemical and isotopic compositions close to those of natural CAIs. However, no attempts have been made so far to experimentally reproduce the observed variations in isotopic composition between individual minerals of the CAIs, which most likely reflects crystallization of minerals from the still-evaporating melt. Here we present our first results on texture and isotopic compositions of the residues produced by cooling forsterite-rich melts in vacuum.

Experimental: FUN1 (forsterite plus 3 wt.% Al_2O_3 and 2.5% CaO) and FUN2 (33 wt.% MgO; 39% SiO_2 , 13% CaO, 15% Al_2O_3) compositions were used. The powders were loaded onto Ir-wire loops 2.5 mm in diameter, heated in a vacuum furnace to 1500° – 1850°C at total pressures of $<10^{-6}$ Torr and cooled at 30 to 120°C/hr. Mo-wire loops were used in 1 atm hydrogen experiments.

A JEOL JSM-5800LV scanning electron microscope with an Oxford/Link ISIS-300 x-ray microanalysis system was used to study the textures and chemical compositions of the run products. The magnesium and silicon isotopic compositions of the evaporation residues were measured using the UCLA Cameca IMS-1270 ion microprobe. San Carlos olivine and pyroxene, and melilite (pure gehlenite and åkermanite) standards were used to correct for instrumental mass fractionation and for matrix effects.

Results: When FUN1 melt was cooled from 1850°C (~30°C above liquidus) only forsterite (Fo) and pyroxene (Px) were crystallized regardless of the cooling rates used (30 to 120°C/hr) and temperatures at which samples were quenched. Typical textures of the evaporation residues are shown in Fig.1. Cooling to 975°C at 120°C/hr (Fig. 1a) resulted in formation of large, isotopically zoned Fo grains ($\delta^{26}\text{Mg}$ from 6‰ to

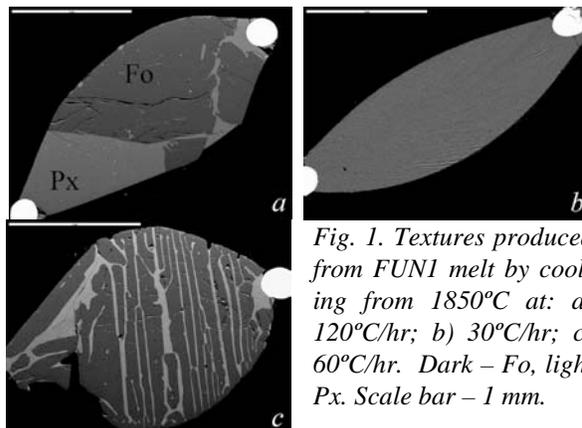


Fig. 1. Textures produced from FUN1 melt by cooling from 1850°C at: a) 120°C/hr; b) 30°C/hr; c) 60°C/hr. Dark – Fo, light Px. Scale bar – 1 mm.

11‰, and $\delta^{30}\text{Si}$ from 4‰ to 6‰). Px is isotopically heavier ($\delta^{26}\text{Mg}$ up to 17‰ and $\delta^{30}\text{Si}$ up to 12‰) suggesting that extra Mg and Si were evaporated after Fo was crystallized. When FUN1 melt was cooled at 30°C/hr only fine crystalline Fo and Px were observed (Fig. 1b). Cooling at 60°C/hr to 1700°C and then at 30°C/hr to 625°C resulted in the texture with parallel bars of forsterite (Fig. 1c) very similar to that of barred olivine chondrules. $\delta^{26}\text{Mg}$ of Fo bars varies from 15‰ to 20‰ and $\delta^{30}\text{Si}$ from 6‰ to 15‰. The barred texture was also observed in the sample cooled at 60°C/hr to 1750°C and then at 30°C/hr to 1700°C. Slower cooling at high temperatures (30°C/hr from 1750°C) is responsible for larger $\delta^{26}\text{Mg}$ (23‰ to 32‰) of the Fo bars in this sample. The fact that Px has the same $\delta^{26}\text{Mg}$ (26‰ to 33‰) suggests that Fo bars were formed right before the sample was quenched. If the sample was allowed to cool to lower temperatures, the molten Px would continue to evaporate, resulting in larger fractionations of Mg and Si isotopes in Px.

When FUN2 melt was cooled from 1850°C (~250° above liquidus) at 120°C/hr in vacuum, the elongated Fo crystals with large Sp and dendritic intergrowth of Px and melilite (Mel) were observed (Fig. 2a). $\delta^{30}\text{Si}$ varied from 6‰ to 10‰ in Fo and from 12‰ to 14‰ in Px and Mel. Cooling at 60°C/hr resulted in formation of very large Sp (up to 500 μm) and fine grained intergrowth of Px and Mel. Only fine grained Px and Mel were observed in samples cooled at 30°C/hr.

In experiments with FUN2 melt the coarse-grained igneous texture typical for Type B and most FUN CAIs was observed only when the melt was cooled from temperatures closer to the liquidus ($T \sim 1600^\circ\text{C}$).

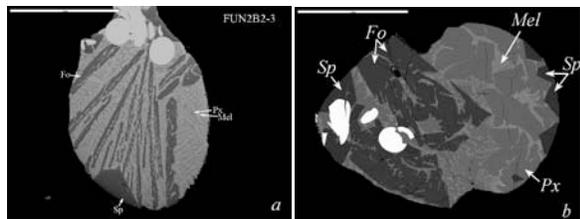
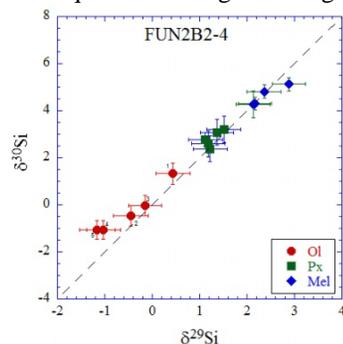


Fig. 2. Textures of evaporation residues of FUN2 melt: a) cooled from 1850°C at 120°C/hr; b) cooled from 1700°C at 60°C/hr.

Fig. 2b shows sample FUN2B2-4, cooled from 1700°C at 60°C/hr, consisting of large grains of Fo, Sp and Px, and interstitial Px, Mel and An. The Si isotopic composition is shown in Fig. 3. Fo grains are isotopically zoned with $\delta^{30}\text{Si} = -1.5\%$ in the center of the charge to $\delta^{30}\text{Si} = 1\%$ near the surface, suggesting that Fo was growing while melt was still evaporating. The much tighter ranges in $\delta^{30}\text{Si}$ of Px and Mel indicate that they crystallized within a narrow temperature range and/or when evaporation of Si slowed down significantly. If the same experiment was conducted in H_2 , a higher degree of evaporation of Mg and Si and much larger isotopic fractionations would be expected.

When FUN2 powder was heated to below liquidus temperatures, 10–50 μm Fo crystals enclosed within Px and Mel were formed. Fig. 4 shows a residue cooled from 1500° to 1100°C at 50°C/hr followed by reheating at 1300°C for 9 days in 1 atm of H_2 . It is the last stage that resulted in formation of forsterite-free outer parts first and crystallization of melilite mantle later. The texture of the experimental run product shown in Fig. 4 is very close to that of Vig1623-5 FUN CAI.

Discussion: Unlike crystallization in a closed system where textures are determined mostly by the cooling rate of the melt, it is the evaporation rate that determines the textures produced in our vacuum experiments. When forsterite-rich melt cools slowly, significant amounts of Mg and Si could evaporate from the melt, decreasing the liquidus temperature. As a result the droplet remains molten and continues to evaporate. As temperature decreases, the evaporation rates of Mg and Si also decrease and the system slowly approaches the liquidus. During this stage multiple nuclei of the



liquidus phase would start to form, resulting in formation of a fine-grained texture at lower temperatures.

Fig 3. Silicon isotopic compositions of FUN2B2-4.

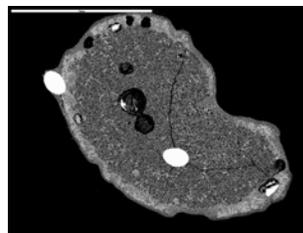


Fig. 4. Texture produced by cooling FUN2 powder from 1500°C to 1100°C at 50°C/hr followed by reheating at 1300°C in 1 atm of H_2 . Dark is Fo, dark gray is Px, light gray is Mel.

At high cooling rates the changes in melt composition due to evaporation are much smaller, the system approaches the liquidus much faster, and significantly fewer nucleation sites of liquidus mineral would form, resulting in a coarse-grained texture. In terms of the isotopes, slow cooling would produce larger fractionation compared to fast cooling. This agrees with the results that Fo from the sample cooled at 120°C/hr is characterized by $\delta^{26}\text{Mg}$ from 6‰ to 11‰ and $\delta^{30}\text{Si}$ from 4‰ to 6‰, while Fo bars of FUN1B2-4 formed by cooling at 60°C/hr are characterized by $\delta^{26}\text{Mg}$ from 15‰ to 20‰ and $\delta^{30}\text{Si}$ from 6‰ to 15‰. The degrees of evaporation and fractionations are expected to be much larger if such experiments are conducted at low total pressures in a hydrogen-rich gas.

Texture, mineralogy and bulk isotopic composition of Vig1623-5 ($\delta^{26}\text{Mg} = 60\%$ and $\delta^{30}\text{Si} = 20\%$) and close isotopic compositions of minerals composing the core of the inclusion [2, 8] can be explained by multiple heating/cooling events [9]: 1) evaporation of a very Fo-rich precursor at above liquidus temperatures that produces a residue with bulk chemical and isotopic compositions close to Vig1623-5, followed by very fast cooling to preserve the isotopic homogeneity within the droplet; 2) heating below liquidus ($\sim 1500^\circ - 1600^\circ\text{C}$) and cooling at $\sim 50^\circ\text{C/hr}$ to form 50–200 μm Fo crystals and fine Sp from multiple nuclei formed during quench in stage 1 without significant loss of Mg and Si; and 3) heating to $\sim 1300^\circ\text{C}$ and slow cooling to form a forsterite-free melilite mantle (Fig. 4).

As in the case of normal Type B CAIs, the coarse grained igneous texture of most FUN CAIs (like C1 or KT-1) and measured isotopic fractionations of Mg, Si and O could be reproduced by a single heating/cooling of a precursor with composition close to that of FUN2, used in our experiments.

References: [1] Clayton et al. (1988) *Phil. Trans. Roy. Soc. London*, A325, 483-501. [2] Davis et al. (1991) *GCA*, 55, 621-637. [3] Thrane et al. (2008) *Astrophys. J.*, 680, L141-144. [4] Richter et al. (2007) *GCA*, 71, 5544-5564. [5] Knight et al. (2009) *GCA*, 73, 6390-6401. [6] Mendybaev et al. (2008) *LPS XXXIX*, Abstract #2345. [7] Mendybaev et al. (2010) *LPS XLI*, Abstract #2725. [8] Marin-Carbonne et al. (2011) *MAPS*, 46, Suppl., A147. [9] Mendybaev R. A. and Richter F. M. (2011) *MAPS*, 46, Suppl., A157.