

## MAGNESIUM DIFFUSION IN MINERALS IN CAIs: NEW EXPERIMENTAL DATA FOR MELILITES AND IMPLICATIONS FOR THE Al-Mg CHRONOMETER AND THERMAL HISTORY OF CAIs.

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**Introduction:** Ca-Al-rich inclusions (CAIs) consist of high temperature minerals that represent the oldest known solid objects in the solar system. The CAIs are known to have contained the short-lived radionuclide <sup>26</sup>Al, which decays to <sup>26</sup>Mg, and has a half-life of 0.72 Myr. The <sup>26</sup>Al-<sup>26</sup>Mg decay system provides a high-resolution relative chronometer for the CAIs [e.g., 1]. However, the chronometer could have been partly disturbed by Mg diffusion in the minerals in CAIs during parent body and/or nebular processes [e.g., 1-3]. It has also been suggested that the non-zero initial  $\delta^{26}\text{Mg}^*$  of 0.7 ‰ in Efremovka (E44) melilite is a consequence of diffusive exchange of Mg isotopes between melilite and anorthite during a period of 10<sup>5</sup> years of residence of CAIs in the protoplanetary disk during which the (<sup>26</sup>Al/<sup>27</sup>Al)<sub>0</sub> changed from a supra-canonical (7x10<sup>-5</sup>) to a canonical (5x10<sup>-5</sup>) value [3]. Thus, studies of Mg diffusion kinetics in the minerals in CAIs are essential for proper interpretation of the Al-Mg chronological data, the extent of intercrystalline Mg isotope exchange and also the observed Mg isotopic heterogeneities within individual minerals in terms of the thermal histories of the CAIs [4,5].

Mg diffusion kinetics have been determined earlier in the CAI minerals anorthite [6] and spinel [7,8], and to a limited extent in melilite [4,9], which varies typically in composition from  $\text{Åk}_{20}$  to  $\text{Åk}_{60}$  in the CAIs. Since diffusion in melilite is known to be significantly anisotropic [10] and compositionally dependent in the åkermanite-gehlenite binary system [11], a systematic study of Mg diffusion in melilite as function of crystallographic orientation and composition is required for a proper understanding of its diffusion kinetic properties in the CAIs.

In this work, we report Mg diffusivity in the melilite end-members åkermanite and gehlenite as a function of temperature and crystallographic orientation. Limited data are also presented for the  $\text{Åk}_{70}\text{Geh}_{30}$  composition for Mg diffusion parallel to *a*-axial direction and combined with earlier data [9] to represent the Arrhenius relation. We apply these data and the available Mg diffusion data in anorthite and spinel to calculate their respective closure temperatures of Mg diffusion for appropriate range of grain sizes in the CAIs in order to evaluate (a) the relative robustness of these minerals for the purpose of Al-Mg chronometric studies, and (b) the possible resetting of Mg isotopic composition during the period of residence of the CAIs in the protoplanetary disk and by thermal processes in the parent body.

**Experimental:** Synthetic crystals of åkermanite and gehlenite were grown by the Czochralski pulling method [10], cut normal to the *a*- and *c*-axial directions, and polished on one side to mirror finish by a combination of mechanical and chemical polishing. The polished sections were thermally pre-annealed for 24 hours at 900°C and the diffusion source material was placed on the surfaces of the pre-annealed crystals by thermally evaporating <sup>25</sup>Mg-enriched MgO powder under a high vacuum condition. The diffusion experiments were run in air at temperatures of 900-1200°C and from 10 min to 1 month in duration, depending on the temperature, in a vertical tube furnace.

The experimental diffusion profiles of <sup>25</sup>Mg in the quenched gehlenite crystals were measured by depth profiling with an ion microprobe (Cameca, ims-3f SIMS at the Arizona State University) using a primary beam of mass filtered <sup>16</sup>O [10]. The crater depths were measured by a surface profilometer that was calibrated against known standards.

**Results:** Fig.1 shows an Arrhenian plot of the measured diffusion data (symbols) of Mg in åkermanite, gehlenite and  $\text{Åk}_{70}\text{Geh}_{30}$  composition, as determined in this work and in earlier studies [9]. Also shown for comparison are the available data for some other CAI minerals, namely anorthite [6] and spinel [7,8]. The parameters of the Arrhenian relation,  $D = D_0 \cdot \exp(-E/RT)$ , for Mg diffusion in melilite, as determined in this study (Table 1).

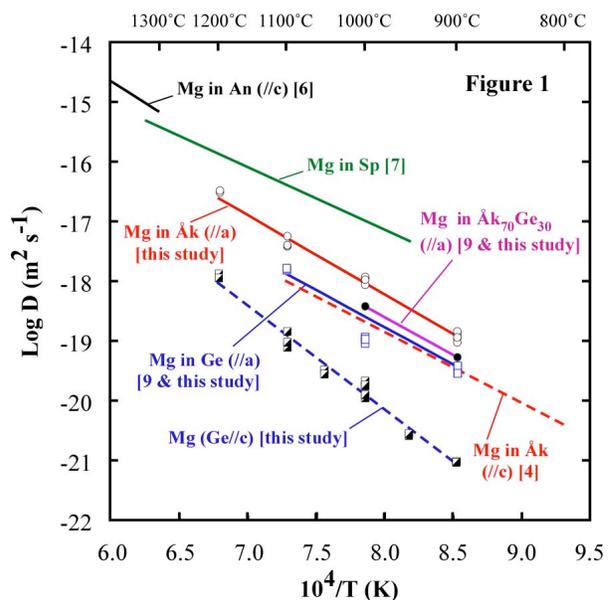


Table 1. Arrhenius Parameters for melilite

		Log Do (m <sup>2</sup> /s)	E (kJ/mol)	
Åkermanite	a	-7.59 ± 0.33	254 ± 8	[4]
	c	-9.30	228	
Gehlenite	a	-8.30 ± 0.89	251 ± 21	
	c	-6.11 ± 0.43	336 ± 11	
Åk <sub>70</sub> Ge <sub>30</sub>	a	-8.52	241	

**Closure Temperatures of Al-Mg System in CAI minerals:** Using the closure temperature formulation of Dodson, as modified by Ganguly and Tirone for application to systems with limited diffusion loss [12], we calculated closure temperatures ( $T_c$ ) of the Al-Mg decay system in anorthite, spinel, and melilite as function of appropriate grain size and cooling rate at the initial temperatures ( $T_0$ ) of 650 and 1200°C. On the basis of crystal morphology, we used a planesheet geometry for anorthite, spherical geometry for spinel and cylindrical geometry for melilite. The chosen range of melilite composition ( $\text{Åk}_{20}$ - $\text{Åk}_{60}$ ) and grain size for each mineral are dictated by the observational data in the CAIs [13]. Figure 3 shows  $T_c$  as a function of  $T_0$  for melilites ( $\text{Åk}_{20}$  and  $\text{Åk}_{60}$ ) for a cooling rate of 0.5°C/h. The suggested cooling rate for CAIs are in the range of 0.5 to 50°C/h [14,15]. Higher cooling rate and Åk content raise the  $T_c$  for a specific  $T_0$  and grain size values.

**Discussion:** From the results presented in Figs. 2a and 2b, we find melilite to be the most robust phase for <sup>26</sup>Al-<sup>26</sup>Mg chronometry of CAIs because of its relatively lower Mg diffusivity (Fig.1) and larger grain size in CAIs. Figures 2 and 3 show that original <sup>26</sup>Mg\* in melilite might survive through the thermal evolution of the parent body ( $T(\text{peak}) \sim 650^\circ\text{C}$ ) if the grains are 1 mm or more in dimension. For similar grain size, the Mg isotopic composition of melilites should not have been disturbed during cooling in the protoplanetary disk at a rate of at least 0.5°C/h. The Al-Mg system in spinel would be most disturbed among the CAI minerals.

**References:** [1] MacPherson G.J. et al. 1995. *Meteoritics* 30:365-377. [2] Thrane K. et al. (2006) *ApJ* 646:L159-L162. [3] Young et al., *Science* (2005) 308, 223 [4] LaTourrette T. and Hutcheon I.D. (1999) *LPS XXX*, 2003 [5] Ito M. and Messenger S. (2007) *Workshop on Chronology of Meteorites*, 4040. [6] LaTourrette T. and Wasserburg G.J. (1998) *EPSL* 158:91-108. [7] Liermann H-P. and Ganguly J. (2002) *GCA* 66:2903-2913. [8] Sheng Y.J. et al. (1992) *GCA* 56:2535-2546. [9] Ito M. et al. (2001) *LPS XXXII*, 1518. [10] Ito M. and Ganguly J. (2004) *MaPS* 39:1911-1919. [11] Nagasawa et al. (2001) *PCM* 28:706-710. [12] Ganguly J. and Tirone M. (1999) *EPSL* 170:131-140. [13] Grossmann L. (1980) *Ann. Rev. Earth Planet. Sci.* 8:559-608. [14] MacPherson G.J. et al. (1984) *J. Geology* 92:289-305. [15] Stolper E. and Paque J.M. (1986) *GCA* 50:1785-1806.

