

# Cr diffusion in orthopyroxene: Experimental determination, $^{53}\text{Mn}$ – $^{53}\text{Cr}$ thermochronology, and planetary applications

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## Abstract

We have determined Cr diffusion coefficients ( $D$ ) in orthopyroxene parallel to the  $a$ -,  $b$ -, and  $c$ -axial directions as a function of temperature at  $f(\text{O}_2)$  corresponding to those of the wüstite–iron (WI) buffer. Diffusion is found to be significantly anisotropic with  $D(//c) > D(//b) > D(//a)$ , conforming to an earlier theoretical prediction. Increase of  $f(\text{O}_2)$  from WI buffer conditions to 4.5 log unit above the buffer at 950 and 1050 °C leads to decrease of  $D(\text{Cr})$  by a factor of two to three, possibly suggesting significant contribution from an interstitial diffusion mechanism. We have used the diffusion data to calculate the closure temperatures ( $T_c$ ) of the Mn–Cr decay system in orthopyroxene as a function of initial temperature ( $T_0$ ), grain size ( $a$ ) and cooling rate for spherical and plane sheet geometries. We also present graphical relations that permit retrieval of cooling rates from knowledge of the resetting of Mn–Cr ages in orthopyroxene during cooling,  $T_0$  and  $a$ . Application of these relations to the Mn–Cr age data of the cumulate eucrite Serra de Magé yields a  $T_c$  of 830–980 °C, and cooling rates of 2–27 °C/Myr at  $T_c$  and ~1–13 °C/Myr at 500 °C. It is shown that the cooling of Serra de Magé to the closure temperature of the Mn–Cr system took place at its original site in the parent body, and thus implies a thickness for the eucrite crust in the commonly accepted HED parent body, Vesta, of greater than 30 km. This thickness of the eucrite crust is compatible only with a model of relatively olivine-poor bulk mineralogy in which olivine constitutes 19.7% of the total asteroidal mass.

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## 1. INTRODUCTION

The Mn–Cr decay system, in which the short-lived nuclide  $^{53}\text{Mn}$  decays to  $^{53}\text{Cr}$  by electron capture, and has a half-life of 3.7 Myr, provides an important tool for dating early solar system objects that show radiogenic  $^{53}\text{Cr}$  ( $^{53}\text{Cr}^*$ ). Excess  $^{53}\text{Cr}$  relative to the terrestrial abundance, which is non-radiogenic, has been detected in the two most abundant meteoritic minerals, olivine and orthopyroxene, and in addition also in spinels and plagioclase from several meteorites (e.g., Hutcheon et al., 1998; Lugmair and Shuk-

olyukov, 1998; Nyquist et al., 2001; Sugiura, 2002). Although  $^{53}\text{Mn}$  is a short-lived nuclide, its half-life is relatively long in comparison to those of other short-lived nuclides such as  $^{26}\text{Al}$  (0.73 Myr),  $^{41}\text{Ca}$  (0.10 Myr),  $^{10}\text{Be}$  (1.5 Myr). As discussed by McKeegan and Davis (2004), this relatively long half-life of  $^{53}\text{Mn}$  (among the short-lived radionuclides), along with the fact that Mn and Cr are reasonably abundant elements, make the  $^{53}\text{Mn}$ – $^{53}\text{Cr}$  decay system an important chronological tool to explore the time period from nebular events to accretion and differentiation of early-formed planetesimals.

A mineral age determined by any decay system corresponds to the time period since the closure of the decay products within the mineral grains during cooling. The closure temperature,  $T_c$ , of a specific decay system in a mineral depends on a number of factors, namely, diffusion kinetic properties of the system, cooling rate of the host rock, and size and geometry of the mineral grains used for dating

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(Dodson, 1973). In addition, for slowly diffusing systems  $T_c$  also depends on the initial temperature,  $T_0$ , at the onset of cooling (Ganguly and Tirone, 1999, 2001). A proper interpretation of mineral age requires an unambiguous characterization of  $T_c$  as a function of the above variables. We have thus undertaken a systematic study of Cr diffusion in meteoritic minerals, especially in the two most abundant ones, olivine and orthopyroxene, to properly quantify the mineral closure temperatures. As shown elsewhere (Ganguly and Tirone, 1999, 2001; Ito and Ganguly, 2006), these data also enable us to develop thermochronological formulations that permit retrieval of cooling rates from  $T_0$ , grain size, and the extent of resetting of age or age loss between  $T_0$  and  $T_c$ .

The Cr diffusion data in olivine and the related closure temperature and thermochronological formulations have been recently presented by Ito and Ganguly (2006). These results are also used to retrieve the cooling rate of the pallasite (stony-iron meteorite) Omolon from the Mn–Cr age of olivine and thereby to constrain its burial depth, which corresponds to the core-mantle boundary, in the parent body. In this work, we present Cr diffusion data in orthopyroxene and the related closure temperature and thermochronological formulations. These results are applied to constrain the cooling rates and burial depths of a cumulate eucrite sample, and thus to provide constraints on the mineralogy and structure of the commonly accepted parent body, asteroid Vesta.

## 2. EXPERIMENTAL AND ANALYTICAL METHODS

### 2.1. Starting material and experimental method

The orthopyroxene crystals used in this work are gem quality natural crystals from Sri Lanka with ~96% MgSiO<sub>3</sub> (enstatite) content. The composition of a typical crystal, as determined by spot analyses in an electron microprobe, is given in Table 1. Since diffusion in orthopyroxene is expected to be anisotropic (Ganguly and Tazzoli, 1994; Schwandt et al., 1998), with the three principal diffusion axes being parallel to the crystallographic axes, the orthopyroxene crystals were oriented in a single crystal diffractometer and cut normal to the *a*-, *b*-, and *c*-crystallographic direc-

tions. The oriented cut faces were then polished stepwise to mirror finish, following the procedure described in Ito and Ganguly (2006).

The experiments were carried out in a vertical gas mixing furnace in which a desired  $f(\text{O}_2)$  condition was imposed by a computer controlled flowing mixture of CO and CO<sub>2</sub> and cross-checked by a zirconia sensor, as described by Ito and Ganguly (2006). Prior to the tracer diffusion experiments, the polished samples were thermally pre-annealed at  $T$ - $f(\text{O}_2)$  conditions at or close to those of the diffusion experiments in order to equilibrate the point defect concentrations of the crystals to the experimental condition or nearly so, and also to anneal the effects of strain caused by the polishing process. A thin layer of Cr was deposited on the polished surface of a pre-annealed sample by thermal evaporation under very high vacuum conditions. The sample was then inserted quickly from the top to the hot spot of the furnace that was already set to the desired  $T$ - $f(\text{O}_2)$  condition of an experiment. The furnace was completely sealed immediately after the insertion of the sample.

A new modification to the furnace involves forced recirculation of the gas mixture into the furnace and periodic venting of gas through a pressure release valve. This procedure greatly reduces the gas consumption and thus makes it possible to carry out diffusion experiments over a long period of time. Furthermore, we are now able to use much greater flow rate of CO<sub>2</sub> compared to what we were able to achieve earlier, and were thus able to study the effect of  $f(\text{O}_2)$  on the diffusion kinetics over significantly larger range of  $f(\text{O}_2)$  conditions compared to the earlier study of Ito and Ganguly (2006) on Cr diffusion in olivine.

### 2.2. Measurement of diffusion profiles

The experimentally induced diffusion profiles were measured by depth-profiling in an ion microprobe in one of two ion probe facilities, the Cameca ims-6f SIMS at the University of Tokyo and the Cameca ims-3f SIMS at the Arizona State University, with most of the measurements being carried out in the former facility. To minimize electrostatic charging of a sample by the primary ion beam irradiation, a thin film (~30 nm) of either Pd (Tokyo laboratory) or Au (ASU laboratory) was deposited on a sample surface. The Cr diffusion profiles were measured as the spatial variation of <sup>53</sup>Cr along with, occasionally, that of <sup>50</sup>Cr. In addition, spatial variation of the non-diffusing species <sup>56</sup>Fe, <sup>30</sup>Si, and <sup>104</sup>Pd or <sup>197</sup>Au were also measured simultaneously during the sputtering process. In several analyses, <sup>26</sup>Mg was measured instead of <sup>50</sup>Cr. The purpose of measuring the non-diffusing species was to help us locate the thin film and crystal surfaces and monitor the stability of the analyses. A typical set of analytical data is shown in Fig. 1.

Because of the mixing effect during depth profiling, the intensity of the Au or Pd layer gets smeared over certain distance. It is assumed that the surface of Cr thin film, over which the metal thin films were deposited, lies approximately at half-intensity for the counts for <sup>104</sup>Pd or <sup>197</sup>Au (Fig. 1a). The sample surface is assumed to be located in between this distance and that at which a non-diffusing species achieved plateau intensity. Since all non-diffusing species do

Table 1  
Electron probe data (average of six analyses) of enstatite used in the diffusion study

Oxide	wt% (1σ)		Atom proportion (1σ)
Na <sub>2</sub> O	0.0099 (0.0066)	Na	0.0006 (0.0004)
SiO <sub>2</sub>	57.4595 (0.1984)	Si	1.9407 (0.0023)
MgO	38.0118 (0.0616)	Mg	1.9139 (0.0037)
Al <sub>2</sub> O <sub>3</sub>	2.4380 (0.0460)	Al	0.0970 (0.0019)
CaO	0.1509 (1.052)	Ca	0.0055 (0.0002)
MnO	0.0185 (0.0141)	Mn	0.0005 (0.0004)
FeO	1.8049 (0.0567)	Fe	0.0510 (0.0016)
TiO <sub>2</sub>	0.0306 (0.0306)	Ti	0.0008 (0.0005)
Cr <sub>2</sub> O <sub>3</sub>	0.0049 (0.0022)	Cr	0.0001 (0.0001)
Total	99.9289	Cations	4.0102
		Oxygen	6.0000

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