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Mössbauer spectroscopy of pyroxene in the light-dark structure of the Kapoeta meteorite: implications for thermal history of the Kapoeta parent body

Y A Abdu¹ and F C Hawthorne²

¹Department of Applied Physics and Astronomy, University of Sharjah, P.O. Box 27272, Sharjah, United Arab Emirates.

² Department of Geological Sciences, University of Manitoba, Winnipeg, MB R3T 2N2, Canada.

E-mail: yabdu@sharjah.ac.ae

Abstract. We report an experimental study of the intracrystalline distribution of Fe²⁺ and Mg between the non-equivalent octahedral sites, M1 and M2, in orthopyroxene from the Kapoeta meteorite by single-crystal X-ray diffraction and Mössbauer spectroscopy. The Fe²⁺-Mg ordering closure temperatures obtained by X-ray structure refinement for two orthopyroxene crystals from Kapoeta (357±30 °C and 411±18 °C) are very similar to those reported for diogenites, and indicate a slow cooling rate. The powder Mössbauer spectra of the light and dark regions in Kapoeta are identical, and their hyperfine parameters are characteristic of orthopyroxene. The closure temperature obtained by Mössbauer spectroscopy for the Kapoeta orthopyroxene (744 °C) is rather high, and indicates a fast cooling rate. The results are discussed in connection with the thermal history of the Kapoeta parent body.

1. Introduction

Pyroxene is one of the most common minerals in stony meteorites and on the surface of terrestrial planets and the moon. The intracrystalline Fe²⁺-Mg exchange reaction between the M1 and M2 octahedral sites in pyroxenes records the latest thermal event of the host rock. The unshocked pyroxene slowly cools to ~ 500 °C and below. Fe²⁺ occurs at M2 site while Mg predominately occurs at M1 site. In crystals that have been rapidly cooled, a more disordered Fe²⁺-Mg distribution is observed over the M1 and M2 sites. Kapoeta is an achondrite meteorite that belongs to the howardite group. Howardites, eucrites, and diogenites (HED) are believed to originate from igneous processes on the asteroid 4 Vesta. Kapoeta is a polymict regolith breccia with a light-dark structure, and the rock consists almost entirely of pyroxene [1].

In this paper, we report the distribution of Fe²⁺ between the M1 and M2 sites of pyroxene in the light and dark regions of the Kapoeta meteorite by Mössbauer spectroscopy and single-crystal X-ray diffraction (XRD). We compare our results with previous Mössbauer work on the whole-rock powdered sample [2] and single-crystal XRD data [3], in an attempt to better understand the thermal history of the Kapoeta parent body.

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2. Experimental setup

The selection of single crystals of orthopyroxene suitable for X-ray diffraction was not easy, as the Kapoeta material is brecciated. Two homogeneous and inclusion-free orthopyroxene crystals, hereafter referred to as X11 and X12, were selected for X-ray diffraction. X-ray diffraction data were collected with a BRUKER P4 four-circle diffractometer equipped with monochromatic Mo K α radiation and a 4K CCD area detector. X-ray diffraction intensities were collected up to 60° and unit-cell parameters were refined by using least squares. Mössbauer-spectroscopy measurements were done at room temperature (RT) using a ⁵⁷Co(Rh) point source. For preparing the powdered Mössbauer absorber, the sample was mixed with sugar and finely ground under acetone to avoid oxidation. The mixture was then loaded into a Pb ring (2 mm inner diameter) and covered by tape on both sides. The spectra were analyzed using a Voigt-based quadrupole-splitting-distribution (QSD) method.

3. Results, Discussion and Conclusion

The degree of Fe^{2+} -Mg ordering for each orthopyroxene crystal was determined from the intracrystalline distribution coefficient, K_D , given by:

$$K_{\rm D} = \left[X_{\rm Fe}^{2+}{}_{(\rm M1)}.X_{\rm Mg(M2)}\right] / \left[X_{\rm Fe}^{2+}{}_{(\rm M2)}.X_{\rm Mg(M1)}\right]$$
(1)

where $(X_{Fe}^{2+}(M1), X_{Mg(M1)})$ and $(X_{Fe}^{2+}(M2), X_{Mg(M2)})$ are the M1 and M2 site occupancies, respectively. The closure temperature, T_C, of the Fe²⁺-Mg ordering reaction was calculated from the relation [4]:

$$\ln K_{\rm D} = -2557(\pm 49)/T(K) + 0.547(\pm 0.048)$$
(2)

The M1 and M2 site occupancies determined by X-ray structure refinement, along with K_D and T_C values, for the Kapoeta orthopyroxene crystals X11 and X12 are given in Table 1. The distribution of Fe²⁺ and Mg over the M1 and M2 sites gives T_C values of 411±18 °C and 357±30 °C for X11 and X12, respectively, in accord with those previously reported for orthopyroxene crystals from Kapoeta having similar compositions to our orthopyroxenes [3]. The low T_C values of the Kapoeta orthopyroxene crystals indicate a slow cooling rate. The composition of these pyroxenes and their T_C values are very similar to those reported for diogenites [5], and may suggest a diogenitic origin of these orthopyroxenes deep within the parent body of the HED meteorites. The studies of orthopyroxene single crystals are not representative of the bulk Kapoeta meteorite, as it is a polymict breccia, and hence the low Tc values obtained are not representative of bulk Kapoeta breccia. To obtain a closure temperature for the bulk Kapoeta orthopyroxene, we use Mössbauer spectroscopy, as discussed below.

Table 1. M1 and M2 site occupancies, intracrystalline distribution coefficient (K_D) and closure temperature (T_C) for the Kapagta arthomyzogna (opy)

for the Kapoeta orthopyroxene (opx).						
	Opx-Xl1	Opx-Xl2	Opx-bulk			
	XRD	XRD	Mössbauer			
$X_{Fe}^{2+}(M1)$	0.047(2)	0.018(3)	0.12(3)			
$X_{Mg(M1)}$	0.953(2)	0.982(3)	0.88(3)			
X_{Fe}^{2+} (M2)	0.548(2)	0.379(3)	0.50(5)			
X _{Mg(M2)}	0.452(2)	0.621(3)	0.50(5)			
$Mg/(Mg+Fe^{2+})$	0.70	0.80	0.69			
K _D	0.041(2)	0.030(5)	0.14(4)			
$T_{C}(^{\circ}C)$	411 ± 18	357 ± 30	744 ± 119			

The RT Mössbauer spectrum of the light region in the Kapoeta meteorite is shown in Fig. 1. It is fitted to a QSD model having one generalized QSD site for Fe^{2+} with two Gaussian components. The QSD profile for Fe^{2+} (Fig. 1) displays two well-resolved Gaussian components centered at QS ~ 2.6 and 2.1 mm/s. Such a QSD profile can be used to obtain Fe site occupancies [6], and we assign the two components to Fe^{2+} at the M1 (QS ~ 2.6 mm/s) and M2 (QS ~ 2.1 mm/s) sites in orthopyroxene. The spectrum of the dark region is almost identical to that of the light region, and both spectra have the same Mössbauer relative areas (within experimental error) for Fe^{2+} at the M1 and M2 sites (table 2).



Figure 1. The room-temperature Mössbauer spectrum of the light region in the Kapoeta meteorite is shown along with the QSD profile for Fe²⁺.

Table	2.	Mössbauer	hyperfine	parameters	for	orthopyroxene	(Opx)	in	the
light a	nd	dark regions	in the Kap	ooeta meteor	rite.				

Sample	Fe-site	CS	QS	Γ	A
		(mm/s)	(mm/s)	(mm/s)	(%)
Opx (Light region)	$Fe^{2+}(M1)$ $Fe^{2+}(M2)$	1.18 1.15	2.56 2.06	0.29	20 80
Opx (Dark region)	$Fe^{2+}(M1)$ $Fe^{2+}(M2)$	1.18 1.15	2.50 2.05	0.28	21 79

CS = center shift relative to α -Fe (±0.02 mm/s); QS = quadrupole splitting (±0.05 mm/s); Γ = linewidth of the elemental doublets of the QSD (±0.02 mm/s); A = relative area (±4%).

Using the Mössbauer spectroscopy the relative areas of $Fe^{2+}(M1)$ and $Fe^{2+}(M2)$ for orthopyroxene in the light region are observed and shown in Table 2. The site occupancies $X_{Fe}^{2+}(M1)$ and $X_{Fe}^{2+}(M2)$ are also observed and listed in Table 1 of 0.12 and 0.50, respectively. $X_{Mg(M1)}$ and $X_{Mg(M2)}$ are then calculated from $1 - X_{Fe}^{2+}(M1)$ and $1 - X_{Fe}^{2+}(M2)$, respectively, and the results are given in Table 1, along with K_D and T_C . A value of K_D equal to 0.14(4) for orthopyroxene (bulk sample) is obtained. This value is significantly smaller than that obtained from the Mössbauer spectrum of the whole-rock powdered sample (0.57) [2]. The difference between the two Mössbauer results could be attributed to the presence of carbonaceous-chondrite inclusions in the Kapoeta whole-rock sample and the different fitting procedures.

In contrast to the low T_C values determined by X-ray structure refinement, the T_C value obtained by Mössbauer spectroscopy for the Kapoeta orthopyroxene is high (744 °C, see Table 1), indicating that the

rate of cooling is fast. This could be due to shock and reheating of the regolith material on or near the surface of the meteorite parent body. The interpretation of the single-crystal XRD and Mössbauer results is consistent with the nature of the Kapoeta rock (and howardites in general) being a polymict, eucrite+diogenite, breccia [7].

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