



Mössbauer spectroscopic study on the composition of Fe-containing minerals in ordinary chondrites, Miller Range 07710 and Yamato 790272

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Abstract ^{57}Fe Mössbauer spectroscopy was applied to compositional studies of Fe-containing minerals in two different ordinary chondrites, MIL 07710 (L4) and Y-790272 (H4), collected in Antarctica. For both samples, spectral analysis revealed that there are five quadrupole doublets and two magnetic sextets in the room-temperature spectra, suggesting the presence of olivine (M1 and M2), pyroxene (M1 and M2), a high-spin Fe^{3+} containing phase, antiferromagnetic FeS (troilite), and ferromagnetic Fe-Ni alloy. The large relative area of the oxidized Fe^{3+} phases and the small intensity of the metal phases signify a possible effect of terrestrial weathering especially for Y-790272 (H4).

Keywords Chondrite · Meteorite · Mössbauer spectroscopy · MIL 07710 · Y-790272 · Terrestrial weathering

1 Introduction

Chondrites are the most primary source of planets and asteroids formed in the early solar system. Because they preserve primitive compositions of early parent bodies, compositional studies on minerals containing iron, a main constituent element in chondrites, are of great

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importance for understanding of the early environment and formation process of our solar system. As the first stage of investigation, it is essential to examine electronic states of constituent Fe atoms to provide insight into chemical compositions of Fe containing minerals in chondrites.

For that purpose, in the present work, we employed ^{57}Fe Mössbauer spectroscopy among many analytical techniques because it can provide information on oxidation states, local structures, and magnetism of Fe atoms involved in various minerals and compounds through hyperfine interactions with high sensitivity. Another notable feature of this spectroscopy is the analytical capability of the chemical states of the probe element in nondestructive manner; therefore, invaluable samples can be offered for other researches after thorough Mössbauer investigation. Taking these advantages, this spectroscopy has long been applied to the study of meteorites to provide valuable information on chemical compositions through discussion based on hyperfine interaction parameters [1–5].

In the present study, we investigated chemical compositions of Fe-containing minerals in two different types of ordinary chondrites, MIL 07710 and Y-790272, collected at Miller Range in 2007 and at Yamato Mountains in 1979, Antarctica, respectively, by means of ^{57}Fe Mössbauer spectroscopy. Ordinary chondrites are divided into three groups on the basis of total iron content and metallic iron content: H (high iron content), L (low iron content), and LL (low iron and low metal content). MIL 07710 and Y-790272 are classified as L4 and H4 ordinary chondrites, respectively [6–8], where the number “4” means a petrologic type. In this work, the constituent iron-bearing minerals were identified through their hyperfine interaction parameter values. The effect of terrestrial weathering is also discussed based on the proceeded oxidation of the metals and sulfides.

2 Experiments

After removal of the surface portion, which has long been directly exposed to terrestrial atmosphere in the ice sheet of Antarctica, fragments of the samples (MIL 07710 and Y-790272) were ground into powder in a clean agate mortar, and about 150 mg aliquots of the powders were separately mounted on thin aluminum plates to make the sample area of about 5 cm^2 (about $6\text{--}8.4\text{ mg Fe/cm}^2$), and sealed with polyimide films. Each of the mounted powder samples was fixed onto the sample holder, and ^{57}Fe Mössbauer spectra were obtained in 1024 channels (before folding) at room temperature on a triangular velocity reference signal mode using a WissEl Mössbauer Spectrometer equipped with an MVT-1000 velocity transducer and an MRG-500 drive unit. The instrumental error for the present measurements estimated by a proposed method was $\pm 0.046\text{ mm/s}$ [9]. The radioactivity of $^{57}\text{Co/Rh}$ source was approximately 75 MBq at the time of the measurement. The isomer shift (I. S.) and velocity were calibrated with those of an $\alpha\text{-Fe}$ foil with $25\text{ }\mu\text{m}$ thickness measured at room temperature. The Mössbauer spectra were analyzed with Lorentzian curves using an analytical program MossA [10].

3 Results and discussion

3.1 MIL 07710 (L4)

The ^{57}Fe Mössbauer spectrum for MIL 07710 obtained at room temperature is shown in Fig. 1. As observed for other L ordinary chondrites [3], the spectrum is comprised of several

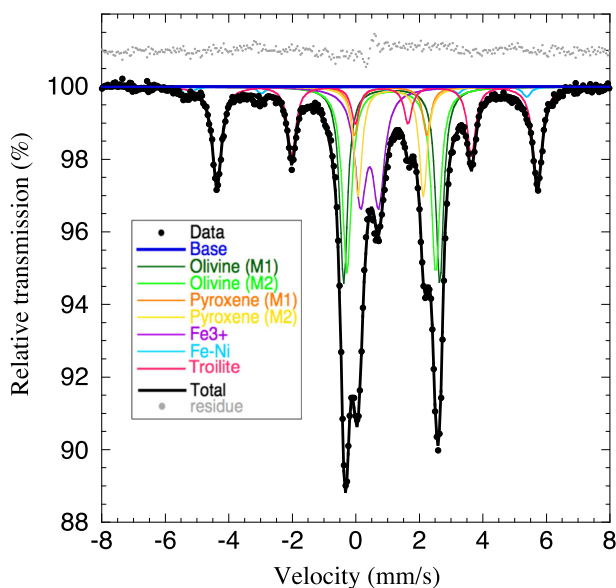


Fig. 1 ^{57}Fe Mössbauer spectrum for MIL 07710 (L4) measured at room temperature. Subspectra shown with different colors are assigned based on their Mössbauer parameters as indicated in the legend

components. The set of two prominent peaks at the velocities of around 2.6 and -0.4 mm/s is a well-known quadrupole doublet assignable to olivine. Because it is known that there are two different sites occupied by divalent metal ions in olivine, spectral fitting was performed with initial values for the Mössbauer parameters in reference to literatures [3, 11]. The same procedures were also taken for the two giant peaks adjacent to those for olivine assuming the presence of two components in pyroxene, a major mineral in chondrites. Thus, we first assumed the presence of 4 main doublets for two olivine components, M1 and M2, and two pyroxene components, M1 and M2. In order to reproduce a considerably clear shoulder peak at the velocity of around 1 mm/s, another doublet was incorporated into the analysis; the isomer shift value suggests that the electronic state for the component is a high-spin Fe^{3+} . There can be seen distinct sextet peaks; the component is assigned to troilite (antiferromagnetic FeS at room temperature) from the I. S. and magnetic hyperfine field of $B_{\text{hf}} = 31.1$ T, which were obtained by the analysis with three symmetric doublets [11]. In addition to the intense sextet, small satellite peaks in the spectrum were reproduced assuming another sextet of ferromagnetic Fe-Ni alloy (kamacite and/or taenite). Thus, the spectrum was finally analyzed with the five quadrupole doublets and two magnetic sextets. The Mössbauer parameter values obtained by the fit are listed in Table 1.

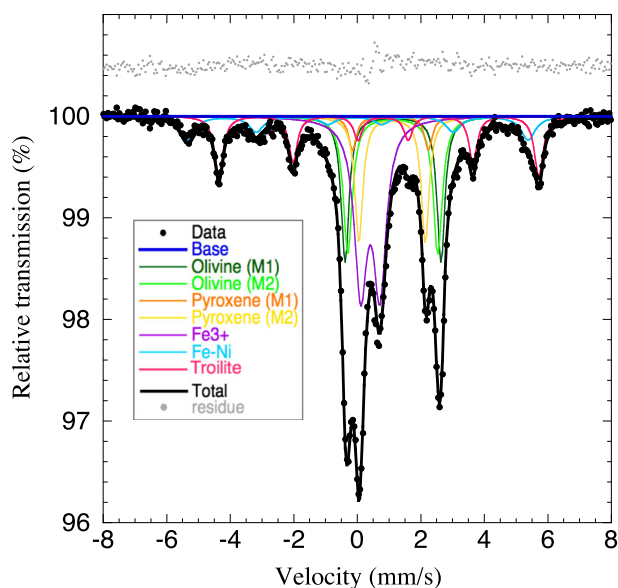
Because MIL 07710 is a “find” meteorite, it is inferred that the chondrite has been contiguous to the ice and atmospheric air for a considerable period of time. The large relative area of oxidized Fe^{3+} component of 16.4% is hence attributed to the result of oxidation by terrestrial weathering. (Note that the relative area is the ratio of only Fe atoms in the sample, and minerals without Fe atoms are excluded from the present consideration.) This inference is consistent with experimental results that a “fall” chondrite Saratov, classified into the same L4 group as MIL 07710, contains only 6 to 7% of Fe^{3+} component [3]. Taking into consideration the report that metal phases are the most susceptible to terrestrial

Table 1 Mössbauer parameter values obtained by the analysis of the room temperature spectrum for MIL 07710 (L4)

I. S. (mm/s)	Q. S. (mm/s)	Relative Area (%)	B_{hf} (T)	FWHM (mm/s)	Phase
1.14(1)	3.02(1)	20.3(7)	—	0.30 ^a	Olivine (M1)
1.13(1)	2.80(1)	21.8(8)	—	0.34(1)	Olivine (M2)
1.10(1)	2.29(3)	5.0(7)	—	0.30 ^a	Pyroxene (M1)
1.11(1)	2.06(1)	11.3(7)	—	0.30 ^a	Pyroxene (M2)
0.44(1)	0.57(1)	16.4(3)	—	0.44(1)	Fe ³⁺
0.75(1)	-0.14(2)	23.0(6)	31.1(1)	0.33(5)	Troilite
0.09(8)	-0.07(2)	2.1(2)	33.1(1)	0.30 ^a	Fe-Ni

a: Fixed values

Fitting errors are shown

**Fig. 2** ^{57}Fe Mössbauer spectrum for Y-790272 (H4) measured at room temperature. Subspectra shown with different colors are assigned based on their Mössbauer parameters as indicated in the legend

weathering [12], the present low concentration of the Fe-Ni alloy (2.1%) can be explained by its oxidation to the Fe^{3+} phase having proceeded in a long period of time.

3.2 Y-790272 (H4)

The room-temperature ^{57}Fe Mössbauer spectrum for Y-790272 is shown in Fig. 2. We analyzed the complex spectrum assuming the presence of seven components of minerals in the same way as MIL 07710. The Mössbauer parameters for those components are tabulated in Table 2, and each mineral listed there was assigned based on the parameter values. Because Y-790272 is grouped into H4, one can expect that minerals formed in reducing atmosphere such as Fe-Ni alloy and Fe sulfide (troilite) are relatively rich and those formed in oxidizing atmosphere are deficient compared with L4 chondrites. Contrary to the expectation, however, little difference, or rather opposite compositions are seen in the sum of the relative

Table 2 Mössbauer parameter values obtained by the analysis of the room temperature spectrum for Y-790272 (H4)

I. S. (mm/s)	Q. S. (mm/s)	Relative Area (%)	B_{hf} (T)	FWHM (mm/s)	Phase
1.14 (1)	3.02(4)	15(8)	—	0.30(3)	Olivine (M1)
1.13(1)	2.84(6)	16(8)	—	0.35(3)	Olivine (M2)
1.05(1)	2.39(2)	3.6(6)	—	0.30 ^a	Pyroxene (M1)
1.10(1)	2.09(6)	13.1(16)	—	0.30 ^a	Pyroxene (M2)
0.42(1)	0.61(1)	28(4)	—	0.48(1)	Fe ³⁺
0.76(2)	-0.13(4)	14.9(7)	31.1(2)	0.32(5)	Troilite
-0.02(1)	0.10(2)	8.9(12)	33.2(1)	0.54(3)	Fe-Ni

a: Fixed values

Fitting errors are shown

Table 3 Room-temperature Mössbauer parameter values for β -FeOOH and γ -FeOOH reported in literature

Sample	I. S. (mm/s)	Q. S. (mm/s)	Relative Area (%)	Literature
β -FeOOH	0.39	0.95	39	[13]
	0.38	0.55	61	
γ -FeOOH	0.35	0.55	100	[14]

areas of the reduced minerals (Fe-Ni + troilite), and the area of the oxidized Fe³⁺ component is actually greater than that of MIL 07710 (L4). The present opposite result for the sum of the compositions of reduced minerals therefore implies a possibility that terrestrial weathering effect has been more accumulated for Y-790272 compared with MIL 07710 due to, for example, a longer terrestrial age. (It seems natural that the relative area for Fe-Ni alloy in Y-790272 (H4), 8.9%, is larger than that in MIL 07710 (L4), 2.1%. However, taking into consideration the fact that the contents of metallic iron in H and L ordinary chondrites are 15–19% and 4–10%, respectively, it can be said that oxidation has proceeded further in the H4 chondrite than in the L4 [5].) For the discussion of sample oxidation, it may also be important to take into account the difference in the elapsed time after their finding and in the state of sample preservation in the laboratory.

3.3 Oxidized Fe³⁺ phase

For both of the present samples, identification of the oxidized Fe³⁺ phase has not been attained yet at the present stage. However, we can still extend discussion for this phase. The presence of ferric oxyhydroxides such as ferrihydrite, goethite (α -FeOOH), akaganeite (β -FeOOH) and lepidocrocite (γ -FeOOH) has been discussed in compositional studies on meteorites. Considering the present Mössbauer parameter values of I. S. and quadrupole splittings (Q. S.), however, possible candidates for the compounds are β -FeOOH and/or γ -FeOOH, both of which are known to be components of iron rust occurring in nature. Table 3 displays their Mössbauer parameter values reported in literature [13, 14]. β -FeOOH has two different components with their Q. S. of 0.55 and 0.95 mm/s; however, a single-component analysis can also be carried out with a wide line width assumed, giving the Q. S. value of 0.67 mm/s [13]. This value is fairly close to those for the unknown Fe³⁺ components in MIL 07710 (0.57 mm/s) and Y-790272 (0.61 mm/s). Because the line widths (FWHM) of their peaks are relatively wide, the Fe³⁺ subspectra for both chondrites would also consist of more than one component, which allows us to attribute the oxidized mineral to β -FeOOH [15, 16]. For the identification of the oxidized Fe³⁺ phases, low temperature measurements

should be valid due to the difference of their Néel temperatures to antiferromagnetic phase, $T_N = 293$ K and 50 K for β -FeOOH and γ -FeOOH, respectively.

4 Summary

In the present work, compositions of Fe-containing minerals in MIL 07710 (L4) and Y-790272 (H4) were first investigated by means of ^{57}Fe Mössbauer spectroscopy. The Mössbauer spectra consist of five doublets, corresponding to olivine (M1 and M2), pyroxene (M1 and M2), and an oxidized Fe^{3+} phase; and of two sextets, troilite and Fe-Ni alloy, for both samples as observed in other ordinary chondrites of similar chemical and petrological types. Because both samples are find meteorites, they have long been exposed to terrestrial environment, which should have caused chemical change by terrestrial weathering. The large relative areas of the oxidized Fe^{3+} phases are possible evidence of the weathering, which is more prominent in Y-790272 (H4).

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