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Report

Anomalous Mössbauer parameters in the second generation regolith Ghubara meteorite

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Abstract–We conducted Mössbauer spectroscopic studies on the Ghubara meteorite which had been described as at least two-generation regolith breccia on the macro scale. The isomer shift and quadrupole splitting of the Fe-Ni part are quite different from those obtained in ordinary chondrites, reflecting shock effects. We observed a large amount of magnetite that may have come from weathering of, primarily, the silicate fraction. We found very similar iron mineralogy in the Densmore meteorite.

INTRODUCTION

Iron-bearing minerals form an important component of all meteorites and carry a lot of information about their history (Dodd 1981; Rubin 1997; Bland et al. 2002). A number of techniques such as petrology, chemical analysis, XRD, INAA, etc., are commonly used to learn the elemental composition and other mineralogical aspects of meteorites. ^{57}Fe Mössbauer spectroscopy is particularly useful for the characterization of iron-bearing minerals because it gives first-hand information about the atomic environment of iron nuclei and, hence, leads to direct identification of the compounds and their chemical states. Recently, our group has conducted extensive studies to identify the correlations between the Mössbauer absorption areas and the classification of meteorites (Verma et al. 2002, 2003; Tripathi et al. 2000; Paliwal et al. 2000). It has been shown that different classes of meteorites exhibit characteristic Mössbauer parameters and shapes so that a "fingerprint" Mössbauer analysis can be quickly made to obtain a broad classification of a meteorite. Due to its sensitivity to Fe^{3+} and Fe^{2+} ions, Mössbauer spectroscopy has been particularly useful to study weathering patterns of meteorites (Bland et al. 1998, 2002).

Mössbauer spectra of ordinary chondrites exhibit iron in four major phases: olivine, pyroxene, troilite, and metallic alloy, with the largest absorption in olivine followed by pyroxene. The alloy phase occupies 10–20% of the absorption area in H chondrites and about 5% in L chondrites. Based on extensive studies carried out on the outer crust of a large number of weathered ordinary chondrites, Bland et al. (1998) have shown that a complete chain of weathering products like akagnite (β-FeOOH), goethite (α-FeOOH), lepidocrocite (γ-FeOOH), maghemite (γ-Fe_2O_3), magnetite (Fe_3O_4), etc., appears in addition to the usual four iron-based minerals in weathered meteorites.

Several large Ghubara specimens were found in the hot deserts of Oman beginning in 1954 and were described as internally fresh with "only slightly weathered crust" (Grady 2000). Ferko et al. (2002) have reported the irradiation history of the Ghubara meteorite that is an L5 ordinary chondrite. Ghubara is believed to have a complicated irradiation history. Based on its petrology, noble gas concentration, thermoluminescence, etc., it is argued that the Ghubara meteorite exhibits at least two generations of regolith exposure. This material was buried in the present host for 4–5 Ma and then was re-excavated. Uneven transient heating may have occurred due to an anisotropic shock-loading impact that sent the meteorite earthward. To the best of our knowledge, so far, Mössbauer studies have not been carried out on meteorites containing two-generation regolith breccia. Here, we report Mössbauer studies of the Ghubara meteorite. Our studies show that the Mössbauer parameters of the alloy phase are quite different than usual, indicating an unusual thermal and shock history of this meteorite. Apart from the usual four iron-based minerals, we also observed a large amount of magnetite. Conspicuously, other commonly occurring weathering products are not seen in significant proportion. A very similar iron mineralogy is found in the Densmore meteorite.

EXPERIMENTAL

Ghubara samples were procured from two different sources. One piece, a slice a few mm thick, was obtained from Schoolers Inc. The other piece was given to us by Professor N. Bhandari of the Physical Research Laboratory (PRL) in Ahmedabad. This piece was originally obtained from the Natural History Museum in London and is a sample taken several cm below the surface of the meteorite. The Densmore meteorite piece was obtained from Schoolers Inc. For Mössbauer analysis, the meteorite fragments were ground to a fine powder, and ~70 mg was sandwiched between transparent tapes to make one Mössbauer absorber 12 mm in diameter. Two samples (GS1 and GS2) were made from the Ghubara piece obtained from Schoolers Inc. and one sample (GB) was made from the PRL piece. While GS1 was made from the whole piece of meteorite, the surface portion (~0.2 mm) of the piece was removed before the piece was crushed to make GS2. The GB and Densmore samples were also prepared from whole pieces.

The Mössbauer spectra were recorded using a conventional constant acceleration Mössbauer spectrometer with ^{57}Co in Rh matrix as the gamma ray source. Spectra were computer-fitted using a least squares routine and assuming each spectrum to be a sum of Lorentzian functions. During the curve fitting, the width and the intensity of the two halves of a quadrupole doublet were constrained to be equal. In the case of a sextet, line widths of the outer, middle, and inner pairs were constrained to be in the ratio of 1.2:1.1:1.0. This ratio was chosen on the basis of pure iron spectra. The quality of fit was judged from the value of χ^2 which was close to 1.0 per degree of freedom. The isomer shift (IS) is reported with respect to α-iron. The reported values of IS and quadrupole splitting (QS) have an accuracy of ~0.02 mm/s, while the hyperfine magnetic field B has an accuracy of ~0.2 T. The areas are correct up to 3%. A closed cycle helium cryostat was used to collect spectra at 12 K.

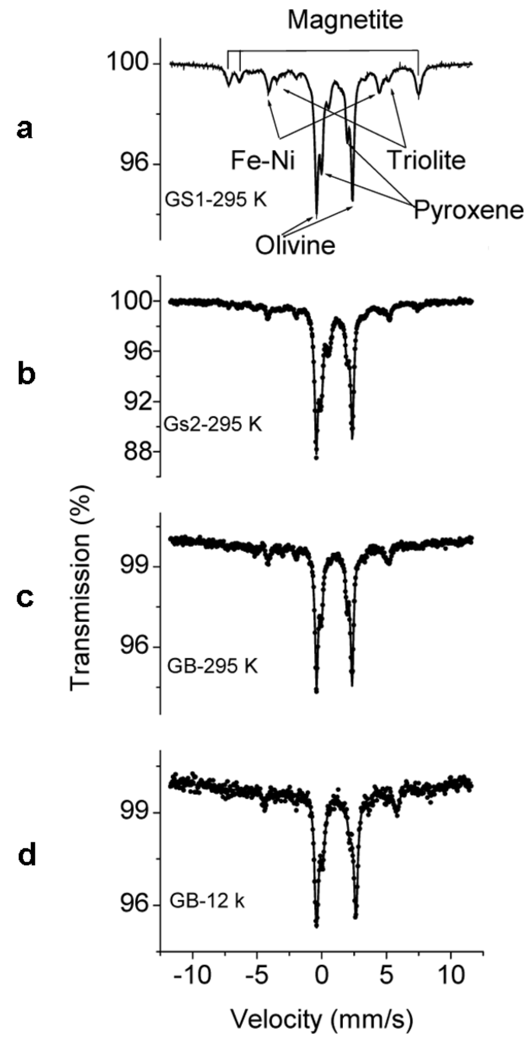

Fig. 1. The Mössbauer spectra of Ghubara meteorites. The samples' codes are explained in the text.

RESULTS AND DISCUSSION

The Mössbauer spectra of the three Ghubara samples are given in Fig. 1 and the Mössbauer parameters are given in Table 1. In all the spectra, one can see the four characteristic components of an ordinary chondrite: olivine, pyroxene, troilite, and the Fe-Ni alloy phase. While the paramagnetic minerals olivine and pyroxene give quadrupole doublets, troilite and Fe-Ni alloy give six-line patterns. The visible peaks are identified in Fig. 1a.

The Fe-Ni phase shows unusual Mössbauer parameters. Let us look at GS1. While the internal hyperfine magnetic field 33.5 T matches well the Fe-Ni alloy phases, the isomer shift of −0.12 mm/s and the quadrupole splitting of −0.37 mm/s are not seen in chondrites. In the ordinary chondrites, this phase has an almost zero isomer shift relative to pure iron and also a negligible quadrupole splitting. A large quadrupole splitting shows significant deviation from cubic symmetry and, hence, a large distortion in the bcc crystal structure. A large negative isomer shift also indicates distortion in the crystal structure and polarization of the electronic s-shells resulting in a larger overlap of electrons with the Fe nucleus. These results support the suggestion of Ferko et al. (2002) that the Ghubara meteorite must have been subjected to an unusual shock metamorphism. Both isomer shift and quadrupole splitting values are smaller (by magnitude) in GS2 and almost normal in GB, which shows that the shock-induced deformation changes in the following order: GS1 > GS2 > GB. This difference in shock level may be due to local heterogeneity.

Looking at the spectrum of GS1 and the corresponding Mössbauer parameters in Table 1, we find components other than the usual four components of ordinary chondrites. Two sextets with reasonably high intensity, one with HMF 45.8 T, and another with 49.1 T appear in the spectrum. The

Table 1. The Mössbauer parameters of the Ghubara and Densmore meteorites.

Sample	Temp. (K)	IS (mm/s)	QS (mm/s)	B(T)	Rel. area	Phase
GS1	295	–0.12	–0.37	33.5	5	Fe-Ni
		0.71	–0.16	30.9	9	Troilite
		0.25	0.02	49.1	15	Magnetite-1
		0.62	0.00	45.8	10	Magnetite-2
		1.12	2.98	–	35	Olivine
		1.12	2.11	–	15	Pyroxene
		0.32	0.66	–	11	Fe^{3+}
GS2	295	–0.05	–0.11	33.6	5	Fe-Ni
		0.73	–0.14	31.0	9	Troilite
		0.26	0.21	49.6	3	Magnetite-1
		0.61	–0.12	46.1	3	Magnetite-2
		1.10	2.99	–	43	Olivine
		1.09	2.14	–	16	Pyroxene
		0.32	0.70	–	20	Fe^{3+}
GB	295	–0.04	–0.02	33.5	8	Fe-Ni
		0.77	–0.15	31.3	11	Troilite
		0.31	–0.08	48.7	4	Magnetite-1
		0.74	–0.07	46.4	1	Magnetite-2
		1.15	3.01	–	48	Olivine
		1.12	2.17	–	19	Pyroxene
		0.43	0.60	–	8	Fe^{3+}
GB	12	–0.22	–0.16	34.0	10	Fe-Ni
		0.82	–0.06	31.6	15	Troilite
		1.01	–0.41	49.4	8	Magnetite-1
		1.22	3.03	–	43	Olivine
		1.12	2.02	–	18	Pyroxene
		–0.14	0.57	–	6	Fe^{3}

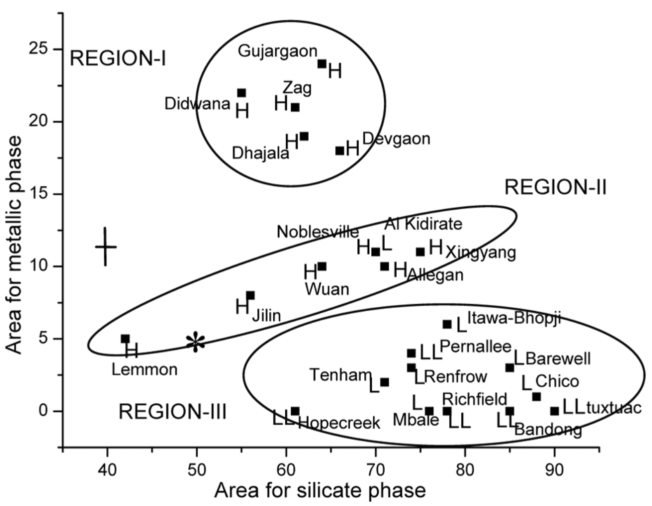

Fig. 2. Systematics of the Mössbauer absorption area of metal phase versus silicate phase in ordinary chondrites.

Mössbauer parameters of these sextets correspond to magnetite that has a spinel structure. The iron atoms occupy tetrahedral A sites and octahedral B sites. The two sextets correspond to iron in these two sites. The leftmost peaks of the two sextets are clearly visible in the spectrum, while the fifth and sixth are merged. The third and the fourth peaks of the two sextets are not clearly visible because they overlap with other components. The total absorption area corresponding to the magnetite phase in GS1 is ~25%. Apart from magnetite, we also have a doublet component with absorption area of ~11% that corresponds to an Fe^{3+} compound.

The GS2 sample, which was made from the portion obtained after removing the surface layer, shows much less magnetite. The GB sample from the National History Museum, which is taken from regions several cm below the surface, also shows a presence of magnetite that is more clearly visible in the spectrum at 12 K (Fig. 1).

Magnetite is known to occur in meteorites due to weathering during their terrestrial life. Assuming that the magnetite does, indeed, come from the weathering of the Ghubara meteorite, some interesting observations can be made. The sequence of weathering of ordinary chondrites from various regions has been extensively studied by Bland et al. (1998, 2002), and it has been found that, in most cases, the major weathering products are: α-FeOOH, β-FeOOH, and γ-Fe_2O_3. These components are not seen in the Mössbauer spectra of Ghubara even at 12 K, which indicates that the weathering mechanism is different from those that are commonly observed in ordinary chondrites that have fallen in deserts. However, such an odd combination of weathering minerals with dominating magnetite has, in some cases, been observed.

Several characteristics have established Ghubara as an L5 chondrite. Mössbauer studies of unweathered L chondrites have established that the alloy phase is small in these chondrites giving an absorption area of ~5% in the Mössbauer spectrum (Verma et al. 2003). Indeed, ~5% of the absorption area in the spectra is present in the alloy phase as expected for an L chondrite. This means that most of the weathering came from the silicate phase that produced magnetite as an end product. This conclusion is also supported by the relative Mössbauer areas of the alloy phase, olivine, and pyroxene. Verma et al. (2003) have shown that these areas show certain systematic behaviors, and plots of metal absorption area versus silicate absorption area separate out nicely for H and L chondrites (Fig. 2). The position of Ghubara is shown by the asterisk in Fig. 2. Clearly, it falls out of the L group due to the depletion of silicates. On the other hand, the olivine/pyroxene absorption area ratio (~2.3) is representative of L chondrites. This shows that both the silicate components are weathered at almost the same rate. This is a common behavior in the weathering of ordinary meteorites (Bland et al. 1998).

Some magnetite is present even in the GB sample. This means that the weathering has penetrated deep inside the meteorite because additional pores and fractures may have been produced in this meteorite during shock metamorphism.

One may also look into the possibility of formation of magnetite at the crust when Ghubara hit the Earth's atmosphere. Mössbauer studies carried out on a large number

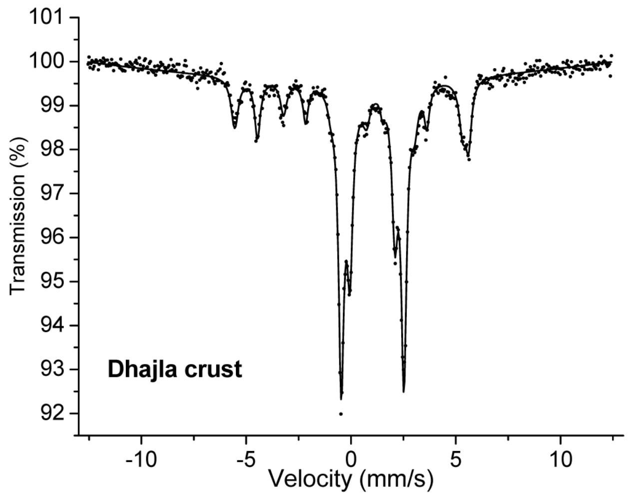

Fig. 3. The Mössbauer spectrum of the Dhajala meteorite crust.

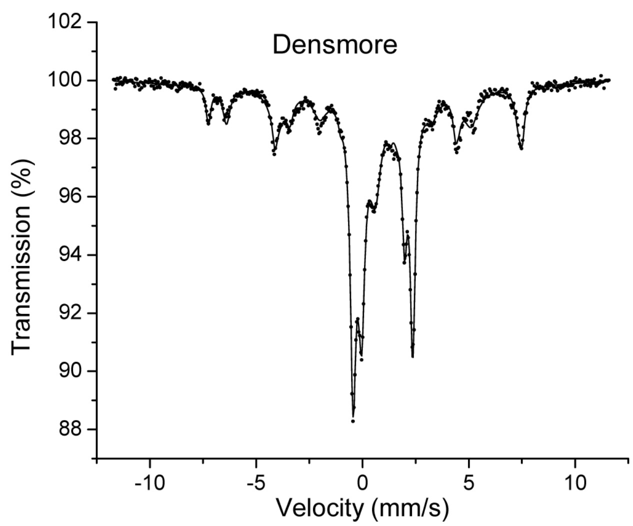

Fig. 4. The Mössbauer spectrum of the Densmore meteorite.

of ordinary chondrite samples made from the crust itself do not show the presence of magnetite in significant amounts. In Fig. 3, we give the Mössbauer spectrum of a sample made only from the crust layer of the Dhajla meteorite. There is no significant presence of magnetite in this spectrum.

Figure 4 shows the Mössbauer spectrum of the Desmore meteorite which is assumed to be an LL chondrite. A comparison of the spectra of this sample and GS1 shows that the two meteorites have an almost identical iron mineralogy, though the meteorites not only fell at different locations, but they also have different terrestrial ages. While the Ghubara chondrite has been extensively studied and its unusual preterrrestrial history has been established, not much work has been done on Densmore. In view of the almost identical iron mineralogy of the Densmore and Ghubara meteorites despite their different fall locations, we suggest that a systematic study of the irradiation history of Densmore be undertaken.

CONCLUSIONS

The Ghubara meteorite shows all the characteristic Mössbauer signatures of an L chondrite. However, the Fe-Ni phase shows large negative isomer shift and quadrupole splitting, showing a strong distortion in the bcc crystal structure. This is consistent with the suggestion that Ghubara is at least a two-generation meteorite and has had an unusual thermal and pressure history.

Magnetite appears to be the result of the weathering of the silicate phase. The abundance of magnetite decreases as one takes samples from deeper portions of the meteorite, but it is present even in a sample taken at the depth of several cm. Since the Ghubara meteorite has undergone a complicated history of shock metamorphism that makes it a two-generation regolith breccia, it is possible that the iron mineralogy is altered in a fashion that promotes formation of magnetite through weathering.

Another meteorite, Densmore, is found to have the same iron mineralogy as Ghubara. We suggest that a systematic investigation be made on Densmore to explore its preterrestrial history.

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