57Fe Mössbauer spectroscopy studies of chondritic meteorites from the Atacama Desert, Chile: Implications for weathering processes

P. Munayco · J. Munayco · M. Valenzuela · P. Rochette · J. Gattacceca · R. B. Scorzelli

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Abstract Some terrestrial areas have climatic and geomorphologic features that favor the preservation, and therefore, accumulation of meteorites. The Atacama Desert in Chile is among the most important of such areas, known as dense collection areas. This desert is the driest on Earth, one of the most arid, uninhabitable locals with semi-arid, arid and hyper-arid conditions. The meteorites studied here were collected from within the dense collection area of San Juan at the Central Depression and Coastal Range of Atacama Desert. 57Fe Mössbauer spectroscopy was used for quantitative analysis of the degree of weathering of the meteorites, through the determination of the proportions of the various Fe-bearing phases and in particular the amount of oxidized iron in the terrestrial alteration products. The abundance of ferric ions in weathered chondrites can be related to specific precursor compositions and to the level of terrestrial weathering. The aim of the study was the identification, quantification and differentiation of the weathering products in the ordinary chondrites found in the San Juan area of Atacama Desert.

Keywords Weathering · Meteorite · Atacama desert
1 Introduction

The study of meteorites depends largely on finding samples that may have been under the influence of the environment for over hundred years. An interdisciplinary study is under way with meteorite samples from the Atacama Desert, one of the oldest and driest desert of the world [1], in an attempt to understand the weathering processes acting on these primitive materials and the conditions of the accumulation surfaces that have preserved them. Those samples may have undergone weathering processes that may result in large changes of their original phases.

Oxidation is certainly the most important reaction to consider and their products are usually crystalline oxides associated to the iron matrix. The degree of weathering in ordinary chondrites (OC) can be quantitatively measured using Mössbauer spectroscopy to determine the proportions of the Fe-bearing phases occurring mainly as Fe$^0$ in the Fe-Ni metal, Fe$^{2+}$ in the ferromagnesian silicates and troilite, and Fe$^{3+}$ in the terrestrial alteration products. In newly fallen equilibrated OC the amounts of Fe$^0$ (kamacite and taenite) and Fe$^{2+}$ (olivine, pyroxene, and troilite) are known within narrow limits. Thus, the abundance of oxidized iron in weathered chondrites can be related to specific starting compositions and to the level of terrestrial weathering.

The aim of this study is the identification and quantification of the weathering products of OC found in San Juan areas of the Atacama Desert (AD) [2].

2 Materials and methods

$^{57}$Fe Mössbauer spectroscopy in transmission geometry was performed at room temperature (RT) and 4.2 K in a 512 channels Halder spectrometer. The drive velocity was calibrated using a $^{57}$Co in Rh matrix source and an iron foil both at RT. All measurements were performed at high velocity (12 mm/s), average recording time of 24 h per sample. Mössbauer absorbers were prepared with 90 mg/cm$^2$ of the bulk meteorite sample. NORMOS code [3] was used for the spectrum analysis based on least-square fitting routine, assuming each spectrum to be a sum of Lorentzian lines grouped into quadrupole doublets and magnetic sextets. The isomer shifts was reported in relation to $\alpha$-Fe.

The Mössbauer measurements have been performed on 9 meteorites collected in the San Juan area. The meteorites are weathered ordinary chondrites (OC) and include the chemical groups H, L and LL.

3 Results and discussion

The Mössbauer spectra at 300 K show an overlapping of paramagnetic and magnetic phases as observed in Fig. 1 for two selected samples from San Juan area, showing a low (San Juan 013) and high (San Juan 025) weathering degree. In San Juan 012 magnetic components associated with iron oxides are not observed indicating a low weathering degree. On the other hand, magnetic components associated to iron oxides indicate a high weathering degree in San Juan 025 sample. The spectra were fitted with three quadrupole doublets, two quadrupole doublets attributed to
Fe²⁺: one associated with olivine (IS = 1.19 mm/s; QS = B3.0 mm/s) and the other to pyroxene (IS = 1.21 mm/s; QS = 2.14 mm/s). The third doublet is due to Fe³⁺, whose Mössbauer parameters (IS = 0.36, QS = 0.77) are typical of a number of oxides and oxyhydroxides in a superparamagnetic state (e.g., ferrihydrite, small-particle goethite, lepidocrocite, akaganéite), and additionally, four magnetic sextets: two of them associated with the Fe-Ni primary phases (kamacite and taenite) and Troilite (FeS), the two remaining associated to goethite (Bhf = 38 T) and maghemite (Bhf = 50 T) [4].

Mössbauer measurements at low temperature (4.2 K) exhibit a too complex spectra exhibiting relaxation effects but apparently no magnetic ordering due to olivine and pyroxene and magnetic components associated to iron oxides/oxyhydroxides (Fig. 1b).

Mössbauer spectra at low temperature were fitted with the same components as in RT fitting: olivine, pyroxene and four magnetic components (troilite, goethite, maghemite, kamacite/taenite). In addition, it was observed one magnetic component (not observed at RT) with an hyperfine field of 48–49 T with nearly the same relative area of the Fe³⁺ doublet observed at RT. This magnetic component identified by their hyperfine parameters as akaganéite [5] can be associated to the magnetic splitting of the Fe³⁺ doublet at 4.2 K. In order to take into account the relaxation effects due olivine and pyroxene, it was necessary to use the spherical relaxation model of the fitting program Normos [3].
Table 1  Relative area for primary phases and weathering products obtained from the fitting of the Mössbauer spectra of meteorites recovered in the San Juan area at room and low temperature

| Meteorite  | Type | Primary mineralogy | Weathering products | Total   |
|-----------|------|--------------------|---------------------|---------|
|           |      | Olivine (%)        | Pyroxene (%)        | FeNi (%)| Troilite (%)| Akaganéite (%)| Goethite (%)| Maghemite (%)| Fe$^{3+}$| oxide (%)|
|           |      |                    |                     |         |             |                |             |                 |         |         |
| $T = 300$ K |      |                    |                     |         |             |                |             |                 |         |         |
| San Juan 004 L | 40.4  | 27.5              | 4.2                | 13.4    | 0.0          | 0.0            | 0.0          | 14.6            | 14.6    |
| San Juan 008 LL | 56.7  | 21.9              | 1.0                | 1.0     | 0.0          | 0.0            | 0.0          | 19.4            | 19.4    |
| San Juan 006 H | 26.0  | 16.4              | 5.1                | 8.7     | 0.0          | 16.5           | 4.8          | 22.6            | 43.8    |
| San Juan 007 H | 25.6  | 17.0              | 2.9                | 7.6     | 0.0          | 17.7           | 13.4         | 15.7            | 46.9    |
| San Juan 012 H | 24.2  | 15.7              | 4.3                | 7.7     | 0.0          | 17.9           | 13.0         | 17.2            | 48.1    |
| San Juan 011 H | 23.6  | 17.7              | 3.2                | 4.8     | 0.0          | 22.1           | 10.5         | 18.1            | 50.7    |
| San Juan 010 H | 21.6  | 16.6              | 2.3                | 3.6     | 0.0          | 27.7           | 4.9          | 23.4            | 55.9    |
| San Juan 003 H | 17.2  | 10.6              | 2.7                | 12.7    | 0.0          | 31.3           | 8.2          | 17.5            | 56.9    |
| San Juan 005 H | 18.3  | 11.0              | 2.6                | 10.3    | 0.0          | 20.0           | 13.5         | 24.3            | 57.8    |
| $T = 4.2$ K   |      |                    |                     |         |             |                |             |                 |         |         |
| San Juan 004 L | 41.6  | 25.5              | 6.5                | 13.4    | 7.4          | 5.5            | 0.0          | 0.0             | 13.0    |
| San Juan 008 LL | 55.0  | 22.8              | 1.0                | 1.0     | 10.6         | 9.6            | 0.0          | 0.0             | 20.2    |
| San Juan 006 H | 27.7  | 16.8              | 5.3                | 9.4     | 14.5         | 20.9           | 5.5          | 0.0             | 40.8    |
| San Juan 007 H | 26.7  | 16.2              | 2.9                | 9.7     | 9.0          | 21.7           | 13.7         | 0.0             | 44.4    |
| San Juan 012 H | 25.7  | 17.3              | 4.3                | 7.8     | 7.7          | 25.0           | 12.2         | 0.0             | 44.9    |
| San Juan 011 H | 23.8  | 14.6              | 3.6                | 5.7     | 20.7         | 21.7           | 9.9          | 0.0             | 52.3    |
| San Juan 010 H | 21.7  | 15.8              | 2.4                | 3.7     | 14.4         | 35.1           | 6.9          | 0.0             | 56.4    |
| San Juan 003 H | 18.9  | 11.0              | 2.2                | 10.2    | 8.9          | 40.2           | 8.5          | 0.0             | 57.6    |
| San Juan 005 H | 20.2  | 11.2              | 1.2                | 11.6    | 17.1         | 26.7           | 12.0         | 0.0             | 55.8    |
Fig. 2 Spectral area of total Fe$^{2+}$ (%) and opaques against total ferric oxidation derived from Mössbauer spectra recorded from weathered samples of San Juan area, showing that both are weathered almost at a constant rate.

The phase quantification was done using the relative areas taken from fitted Mössbauer spectrum (see Table 1) assuming that all of the phases have the same recoil-free fraction.

The samples show a wide range in total oxidation between 10 % and 58 %, given by the presence of Fe-oxides appearing as magnetically ordered Fe$^{3+}$ (at low temperature) or as paramagnetic Fe$^{3+}$ (at room temperature).

In order to study the weathering effect on the primary mineralogy of the samples we compare spectral areas of Fe$^{2+}$ total (olivine and pyroxenes) and opaque (Fe-Ni and troilite) with the total amount of oxidation. Thus it was possible to determine which phases are the most susceptible to weathering (Fig. 2). The decrease in primary phases with increasing oxidation suggest that all iron-containing minerals within the meteorite are affected by weathering to some degree, as previously reported for OCs from other hot deserts [6].

4 Conclusions

From $^{57}$Fe Mössbauer spectroscopy results we are able to conclude that the weathering degrees of San Juan OC samples vary in the range of $\sim$10 to 57 %. The observed iron oxides are present as magnetically ordered Fe$^{3+}$ (magnetite, maghemite, goethite) and paramagnetic Fe$^{3+}$. The last one, identified as akaganéite, the first product of Fe-Ni oxidation found in all samples in the range of 7 to 22 %. The decrease in primary phases with increasing oxidation suggests that all iron-containing minerals within the meteorite are affected by weathering to some degree.
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