

Room temperature ^{57}Fe Mössbauer spectroscopy of ordinary chondrites from the Atacama Desert (Chile): constraining the weathering processes on desert meteorites

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Abstract We report the results of a study on the weathering products of 21 meteorites found in the Atacama Desert (Chile) using room temperature ^{57}Fe Mössbauer spectroscopy (MS). The meteorites are weathered ordinary chondrites (OCs) with unknown terrestrial ages and include the three chemical groups (H, L, and LL). We obtained the percentage of all the Fe-bearing phases for the primary minerals: olivine, pyroxene, troilite and Fe–Ni metal, and for the ferric alteration products (composed of the paramagnetic Fe^{3+} component and the magnetically ordered Fe^{3+} components) which gives the percentage of oxidation of the samples. From the Mössbauer absorption areas of these oxides, the terrestrial oxidation of the Atacama OC was found in the range from ~5% to ~60%. The amount of silicates as well as the opaques decreases at a constant rate with increasing oxidation level.

Keywords Weathered ordinary chondrites · Iron oxo-hydroxides · Hot desert environments · Atacama Desert

1 Introduction

Ordinary chondrites (OCs), one of the most abundant type of known meteorites (~80%), are excellent standard geological samples to track the effects of terrestrial weathering because: (1) their initial composition before weathering is very well known from the analysis of modern falls [1–4], (2) all the iron in an equilibrated OCs is considered to be present as Fe^{2+} or Fe^0 (in silicates and sulphides in the first case, and Fe–Ni metal in the second), thus any

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Fe^{3+} present in an ordinary chondrite find (as opposed to an observed fall) may be interpreted as the product of terrestrial alteration that can be quantified by ^{57}Fe Mössbauer spectroscopy, (3) their terrestrial ages (i.e., the time they have spent on Earth since falling) can be constrained through ^{14}C [5], ^{81}Kr , ^{36}Cl [6] or ^{26}Al analyses to provide a chronology of events, and (4) because these meteorites fall more or less uniformly over the Earth's surface, they provide a standard sample to study weathering processes in very different environments.

The aim of this study is the identification and quantification of the weathering products of 21 weathered OCs found in two main areas of the Atacama Desert, northern Chile, one of the oldest and driest deserts in the world [7, 8], in order to understand the weathering processes operating in the Atacama Desert (AD), and to constrain the effect of the terrestrial environment on the primary signatures of this extraterrestrial material.

This study is being complemented with other techniques such as optical and scanning electron microscopy (SEM), X ray diffraction (XRD), physical and magnetic properties characterization, ^{14}C dating, and bulk and mineral grain chemistry of major and trace elements.

2 Background

Almost 90% of the normative mineralogy of ordinary chondrites is made up of olivine, pyroxene, troilite and Fe–Ni alloys, all of which contain Fe. The alteration products that have been recognized in desert OCs are mainly iron oxo/hydroxides (e.g., akaganéite, goethite, maghemite and magnetite) formed by the oxidation of the primary phases. As Mössbauer spectroscopy is extremely sensitive to changes in the Fe valence state, it can provide an overview of the effect of weathering on the whole sample and quantify the relative proportion of ferrous, ferric and metallic components, allowing the quantification of the oxidation of the samples.

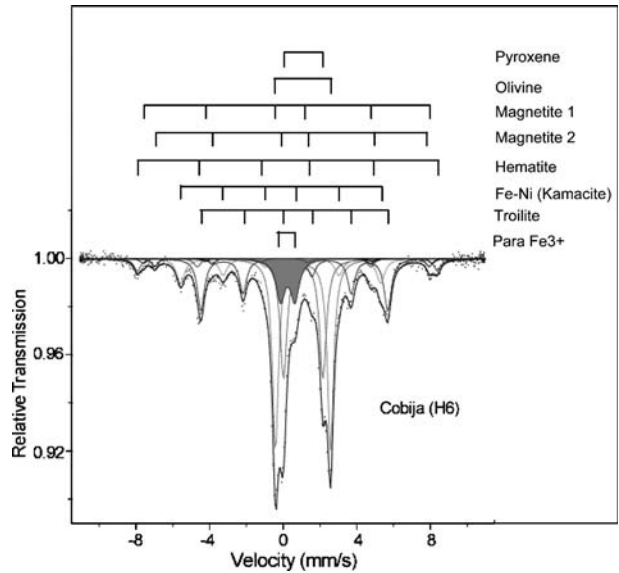
3 Experimental

About 1–2 g of sample taken from the outer part of the meteorites (but excluding any surviving fusion crust) were crushed to obtain powders for ^{57}Fe Mössbauer spectroscopy and XRD.

Mössbauer spectra were recorded at room temperature (RT), in high velocity, in transmission geometry using a $^{57}\text{Co}(\text{Rh})$ source, in a Halder spectrometer with 512 channels. The velocity of the drive shaft tracks the input waveform to the drive control unit. With a sinusoidal wave input, the shaft velocity changes uniformly in time, that is, the acceleration is constant and a velocity reference signal positive. The drive velocity was calibrated with the same source and a metallic iron foil at RT. Average recording time was 48 h per sample. Mössbauer absorbers containing ~250 mg of the bulk meteorite powdered sample were used. Isomer shifts were measured relative to $\alpha\text{-Fe}$. NORMOS code [9] was used for the spectrum analysis.

The phase quantification was done using the relative areas taken from fitted Mössbauer spectrum, since they are proportional to the number of resonant nucleus that represents the Mössbauer effect occurrence probability. It should be mentioned that, for a given temperature, this probability is not equal for all the phases. However, in this paper we

Fig. 1 RT Mössbauer spectrum of the Cobija (H6) meteorite



are considering as a good approximation, that all the phases have the same probability according to reported low temperature results in similar samples.

4 Results and discussion

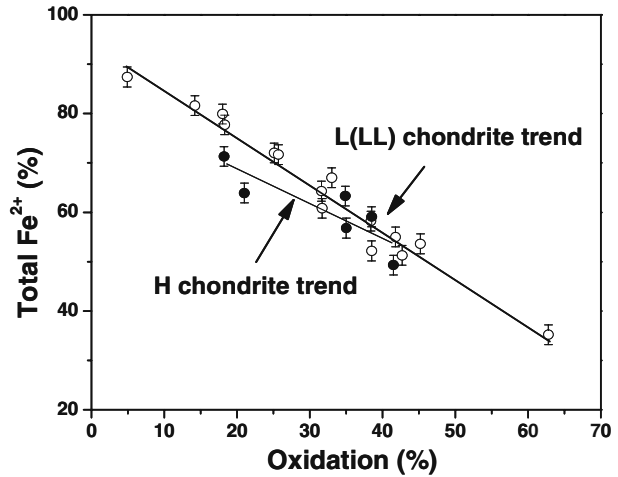
The first petrographic studies with traditional microscopy and SEM showed a wide range of weathering grades for the set of samples given by different patterns of alteration, mainly of the opaque phases, and also in shock veins [10]. From XRD, the alteration products detected were akaganéite, goethite, maghemite, hematite and magnetite [10, 11].

The relative proportions of the major Fe-bearing phases of the samples (as pyroxene, olivine, Fe–Ni and troilite), as well as the alteration products, were recognized and quantified by Mössbauer spectroscopy. As can be seen in Fig. 1, the Mössbauer spectrum shows an overlapping of paramagnetic and magnetic phases. The spectra were fitted with three quadrupole doublets, two of them attributed to Fe^{2+} , one associated to olivine ($IS=1.19$ mm/s; $QS=3.0$ mm/s) and the other to pyroxene ($IS=1.21$ mm/s; $QS=2.14$ mm/s). The third is due to Fe^{3+} ($IS=0.36$ mm/s; $QS=0.77$ mm/s) which can be associated to superparamagnetic oxides and/or iron hydroxides (small particles of goethite, akaganéite, lepidocrocite), that will be identified by low temperature measurements, under way. Finally, the magnetic components have been associated to large-particle goethite ($B_{\text{hf}}=38.2$ T), Fe–Ni ($B_{\text{hf}}=33.5$ T), troilite ($B_{\text{hf}}=32.5$ T), hematite ($B_{\text{hf}}=51.8$ T) and magnetite (two sextets $B_{\text{hf}}=49.1$ T and $B_{\text{hf}}=46.2$ T).

The samples show a wide range in total oxidation (between 4.9% and 62.8%), given by the presence of Fe-oxides appearing as magnetically ordered Fe^{3+} or as paramagnetic Fe^{3+} .

To study the weathering effect on the primary mineralogy of the samples we compare the spectral areas of ferromagnesian silicates (olivine and pyroxenes) and opaque (Fe–Ni and

Fig. 2 Spectral area of total Fe^{2+} (%) against total ferric oxidation derived from Mössbauer spectra recorded from weathered Atacama OCs, showing that ferromagnesian silicates are weathered almost at a constant rate



troilite) with the total amount of oxidation to determine which phases are most susceptible to weathering (Figs. 2 and 3). We can observe a decrease in primary phases with increasing oxidation, almost at a constant rate for the H and L–LL groups, suggesting that all iron-containing minerals within the meteorite are affected by weathering to some degree.

From the Mössbauer data we plot the percentage oxidation of the samples as a frequency histogram. Comparing this oxidation frequency distribution of the Atacama Desert (AD) OCs (Fig. 4c) with the oxidation frequency distribution of other hot desert OCs [12], from the Nullarbor Region (NR) and Roosevelt County (RC; Fig. 4a) and from the Sahara Desert (Fig. 4b) we can observe that AD OCs show a peak around 35%, similar to Sahara Desert samples, in contrast with the peak value between 40–45% of NR and RC samples.

The authors of Ref. [13] related this peak to the level of weathering required before meteorites begin to be eroded and lost from a population. As this property is related to the physical weathering of OCs, in principle it should be the same for all collection areas. Therefore, the difference we observe can be related to the stability and age of the accumulation surfaces. AD OCs display a distribution pattern similar to SD OCs that corresponds to a younger accumulation area compared with NR and RC [12].

5 Conclusions

The decrease in primary phases with increasing oxidation suggests that all iron-containing minerals within the meteorite are affected by weathering to some degree, as previously reported for OCs from other hot deserts [12].

Although we have not yet obtained the terrestrial ages of these meteorites we can suppose from the oxidation frequency distribution pattern that the main accumulation area, in which most of the samples have been found, can not be as old as the time required to reach the peak value of about 40–45%, found for other hot desert meteorites [12, 13].

Fig. 3 Spectral area of opaques (Fe–Ni and troilite) against total ferric oxidation, showing that both groups of Atacama OCs are affected at almost constant rate by weathering

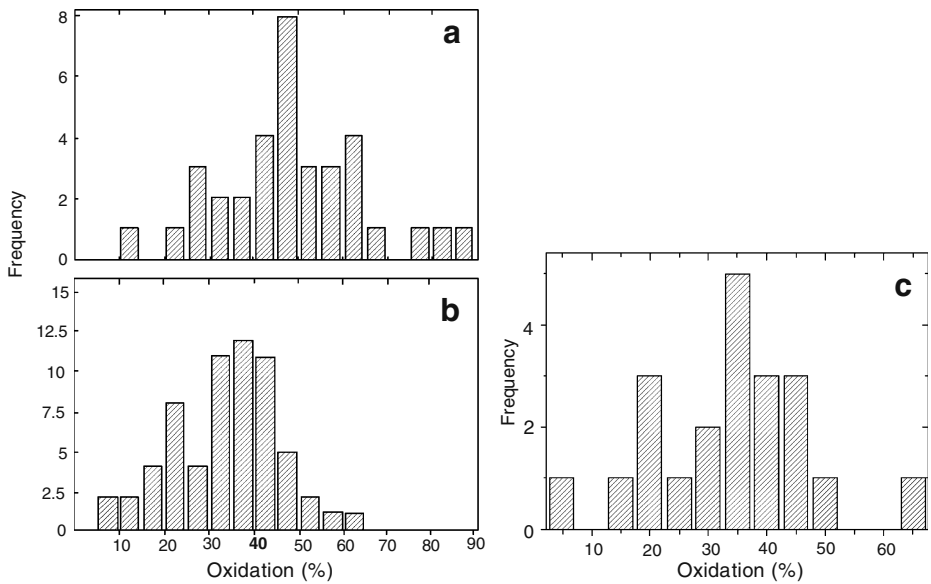
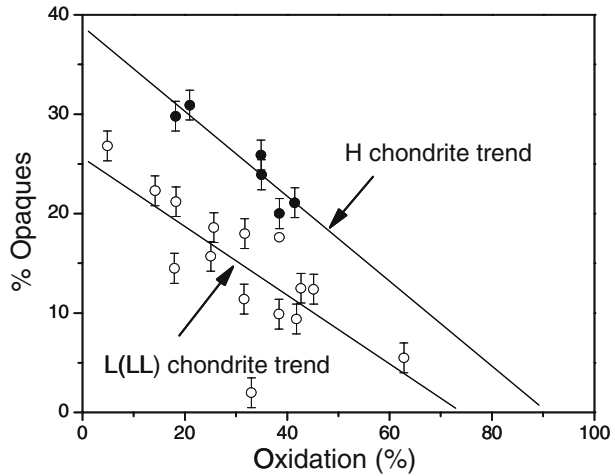


Fig. 4 Percentage oxidation (in 5% bins) against frequency for all the hot desert OCs studied by [12], split between (a) non-Sahara Desert (i.e., Roosevelt County and Nullarbor Region) samples and (b) Sahara Desert samples. **c** Shows the oxidation-frequency distribution for Atacama Desert (AD) samples obtained in the present study

As pointed out by [12], at least two factors are significant in determining the level of oxidation: terrestrial age and initial chemistry. H OCs should show, for a given terrestrial age, higher oxidation levels than L(LL) OCs, because of their high Fe–Ni content, so that the next step of terrestrial age dating will be crucial in understanding the weathering processes in the Atacama Desert.

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