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SCATTERING MÖSSBAUER STUDIES OF WEATHERED AND UNWEATHERED MEXICAN IRON METEORITES

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Abstract: Non-destructive Mössbauer spectra (MS) of a series of nine Mexican iron meteorites belonging to the collection of the Instituto de Geología of the Universidad Nacional Autónoma de México (IG-UNAM) were obtained by 90° γ-ray scattering. We divided the set of nine meteorites in two groups: The first group shows complex spectra of iron oxides mixed with a paramagnetic phase; their spectra and Mössbauer parameters have noticeable differences. The spectra of the second group are quite similar and characteristic of Fe-Ni alloys.

Keywords: Mexican iron meteorites, Scattering Mössbauer spectroscopy

INTRODUCTION

In 1794, Ernst Chladni put forward the extraterrestrial origin of what we today call meteorites. Since then, gaining insights into the initial stages of the solar system has stimulated great interest in the study of meteorites. Although iron meteorites have been recognized around the world, the Mexican territory has been favored in such a way that Fletcher (1890) states:

Mexico is remarkable beyond any other part of the earth's surface for the number and magnitude of the masses of meteoric iron found within its borders: it has been generally assumed that widespread showers were necessary to the explanation of their occurrence.

Notwithstanding the large number of iron meteorites that have fallen in Mexican territory (Sonnenschmidt, 1804; von Humboldt, 1810; Bartlett, 1854; del Castillo, 1889; Tarayre, 1867; Bárcena, 1876; Ward, 1902; Angermann, 1903), very few studies, other than morphological, have been performed by Mexican scientists. Moreover, there are more and larger collections and studies of Mexican meteorites in other countries than in México. This paper represents the first systematic study of Mexican iron meteorites, nine in total, using Mössbauer spectroscopy (MS).

Most of the studied samples are fragments of known meteorites that belong to the collection of the Instituto de Geología of the Universidad Nacional Autónoma de México (IG-UNAM), and no destructive analysis can be performed on them. All of the specimens were finds, and the oldest one (Morito) is from the XVII century. Unfortunately, for most of these specimens we do not know the conditions to which they were exposed prior to their being gathered by the IG-UNAM. Figure 1 shows a photograph of one of the samples.

According to the IG-UNAM and Meteoritical Bulletin, all the samples are iron octahedrites. In the following list, we indicate their names, weights, the Mexico state and the year when they found:

1. Bacubirito, 17.0 g, Sinaloa (1863).
2. Cacaria, 44.0 g, Durango (1867).
3. Casas Grandes, 157.3 g, Chihuahua (1867).
4. Charcas (Catorce), 39.6 g, San Luis Potosí (1804).
5. Charcas, 11.6 g, San Luis Potosí (1804).
6. Chupaderos, 18.4 g, Chihuahua (1852).
7. Morito, 64.8 g, Chihuahua (1600).
8. Santiago Papasquiaro, 546.0 g, Durango (1958).

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Fig. 1. A fragment of the Xiquipilco no. 2 meteorite

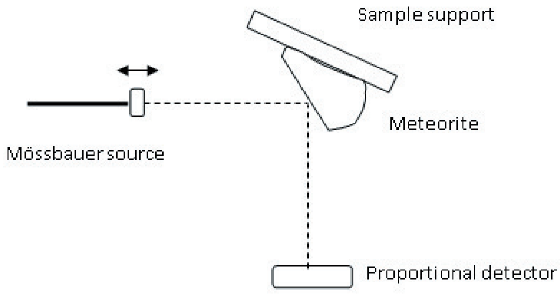

Fig. 2. Sketch of the experimental setting to record the Mössbauer spectra

9. Xiquipilco no. 2, 110.4 g, Estado de México (1949).

Casas Grandes, Charcas, Chupaderos, Morito and Cacaria are classified as IIIAB irons. Bacubirito and Santiago Papasquiaro are ungrouped iron meteorites. Xiquipilco no 2 is classified as an iron, meaning that it is an iron meteorite but not fully clasified as per the definition in the Meteoritical Bulletin.

EXPERIMENTAL

The Mössbauer spectra (MS) were recorded using the radiation scattered by the bulk sample to avoid damage on the meteorites. This technique is most sensitive to the surface than to the bulk; using gas or scintillation detectors the probing depth is between 20 and 100 µm. A proportional Kr-CO_2 (152 cmHg) detector was placed at 90° with respect to the direction of the collimated incident radiation (Fig. 2); the exposed area of the meteorite is about 5.0 cm^2. Aluminum foil was used to eliminate the 6.47 keV X-rays due to the internal conversion process, and14.4 keV was selected with a single channel analyzer. In order to get a meaningful signal to noise ratio, the accumulation times were quite long (about one-week continuous accumulation). The spectra were recorded with a constant acceleration spectrometer, using a 50 mCi (1.87 GBq) ^{57}Co Mössbauer source in Rh matrix, and the data were fitted using Recoil 1.05 software (Lagarec, 2002). All the isomer shifts (IS) are quoted with respect to metallic iron. To take into account possible electric field gradient and magnetic field distributions, the line widths were not restricted, so the reported magnetic field intensities (H) and quadrupole splitting (ΔQ) refer to average values of these distributions. After recording the MS, the meteorites were divided into two groups.

RESULTS

First Group

This group shows complex spectra of iron oxides mixed with a paramagnetic phase and comprises the Cacaria, Charcas (Catorce), and Xiquipilco no. 2 meteorites. Their Mössbauer spectra are quite complex and will be analyzed individually.

Cacaria meteorite

Figure 3a shows the experimental spectrum (dots) together with the least squares fit (continuous line). This spectrum consists of two sextets and a quadrupole doublet whose Mössbauer parameters (MP) are shown in table 1. For the sake of clarity and only in this case, we show the corresponding subspectra in figure 3b. The magnitudes of the hyperfine fields of the sextets correspond to those of magnetite $Fe^{3+}[Fe^{2+}Fe^{3+}]O_4$: the

Table 1. Mössbauer parameters of the Cacaria meteorite

	IS (mm/s)	ΔQ (mm/s)	H (T)	Γ (mm/s)
Sextet I	0.277(3)	0.012(3)	49.55(2)	0.32(5)
Sextet II	0.683(2)	0.017(2)	46.14(2)	0.36(3)
Doublet	0.258(7)	0.680(1)		0.32(6)

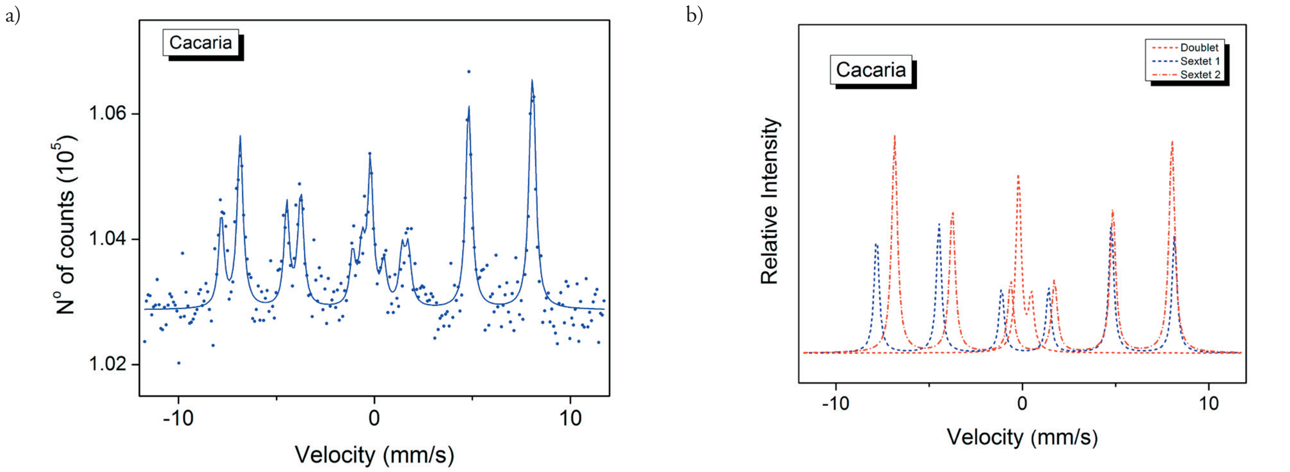

Fig. 3. Mössbauer spectrum of the Cacaria meteorite: a) Experimental spectrum (dots) of the Cacaria meteorite together with the least squares fit (continuous line); b) Subspectra of the fit analysis

49.55 T to Fe^{3+} in the A and B sites and the 46.14 T to (Fe^{2+} + Fe^{3+}) in the B sites of the AB_2O_4 spinel structure. The area relation between these sextets is 0.63. It is worth noting that the Cacaria meteorite spectrum is similar to the ones observed by Wdowiak and Agresti (1984) and Madsen et al. (1986) in the Orgueil CO carbonaceous chondrite. The first authors ascribe the doublet to superparamagnetic nanocrystallites of magnetite, but the last of these authors do not agree with this interpretation and suggest that it may be due to an amorphous iron (III) compound. It is important to note that an explanation for the presence of a superparamagnetic phase is not an easy task. It could be that, notwithstanding the very long cooling time elapsed since the creation of a meteorite to its arrival on Earth, once it penetrates the atmosphere its temperature rises in such a way as to favor the formation of nano-crystals on its surface. Nevertheless, comprehensive explanation for this doublet could be that, due to the atmospheric conditions to which the meteorite has been exposed (that we do not yet know), it is more likely the formation of paramagnetic γ–FeOOH at is surface (Buchwald, 1977).

The obtained Mössbauer parameters (Table 1) are in agreement with reported ones (Greenwood & Gibb, 1971; Johnson, 1969; Persoons, et al., 1986).

Charcas (Catorce) meteorite

One of the several fragments of an iron meteorite found near the town of Charcas, in the state of San Luis Potosí, was towed to another town call Catorce, for unknown reasons (Buchwald et al., 2005). Again, two magnetic sextets and a quadrupole doublet are present in the MS of this meteorite (Fig. 4). Their MP are shown in table 2. One of the sextets is associated with the α–FeNi (33.66 T) cubic phase (Oshtrakh et al., 2009; Böttger et al., 1994), and the other one may correspond to be spinel structure γ–Fe_2O_3. However, two important things are to be noted. First, the line widths values are high and, secondly, the magnetic field value of the γ–Fe_2O_3 phase is lower than the one reported in the literature (Greenwood & Gibb, 1971; Betteridge et al., 1995). The doublet parameters are equal to the ones reported by Madsen et al. (1986) and has been assigned to a paramagnetic phase of Fe^{3+}. These effects are most likely due to the weathering and mechanical processes that the Charcas meteorite fragment was subjected to. The area relation between the two sextets is 0.50.

Table 2. Mössbauer parameters of the Charcas meteorite

	IS (mm/s)	ΔQ (mm/s)	H (T)	Γ (mm/s)
Sextet I	0.474(9)	0.174(9)	47.24(5)	0.40*
Sextet II	0.006(5)	0.006(4)	33.66(3)	0.40(8)
Doublet	0.376(2)	0.735(4)		0.43(4)

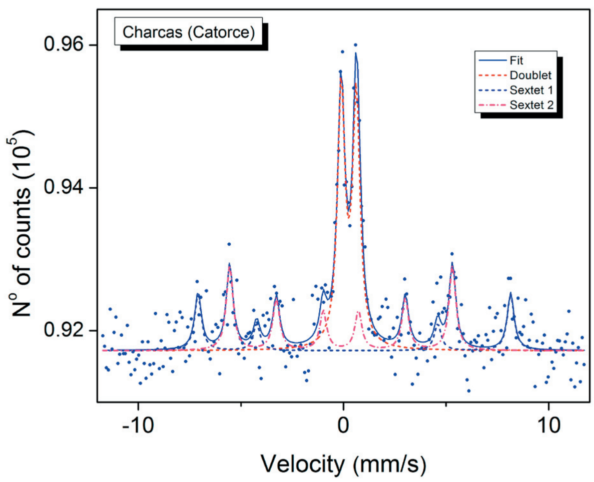

Fig. 4. Mössbauer spectrum of the Charcas (Catorce) meteorite

Xiquipilco no. 2

The MS of this meteorite (Fig. 5) shows a magnetic sextet and a quadrupole doublet whose parameters are listed in table 3. However, even at first sight the line-widths of the sextet are too large, so in the fitting process we left free this parameter to obtain an average hyperfine field that corresponds approximately to γ–Fe_2O_3 (49.24 T). The line-width (0.6 mm/s) of the sextet reveals a magnetic field distribution due to an amorphization probably caused by rapid cooling. The presence of the quadrupole doublet, whose line width is less than for the sextet, could be associated with a superparamagnetic effect due nanocrystallites formed in the cooling process.

Table 3. Mössbauer parameters of the Xiquipilco no. 2 meteorite

	IS (mm/s)	ΔQ (mm/s)	H (T)	Γ (mm/s)
Sextet	0.249(6)	0.015(5)	49.24(5)	0.60*
Doublet	0.333(1)	0.637(2)		0.40*

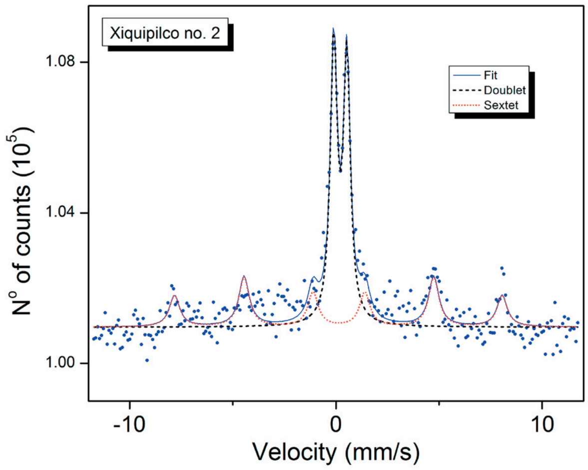

Fig. 5. Mössbauer spectrum of the Xiquipilco no. 2 meteorite

Second group

The Mössbauer spectra of the six meteorites comprised in the second group (Bacubirito, Casas Grandes, Chupaderos, Morito, Charcas and Santiago Papasquiaro), shown in figure 6, are nearly equal, notwithstanding that Buchwald mentions that the composition and morphology of first one and the last one are anomalous (Buchwald 1968a, b). Their line widths have reasonable values expected for iron meteorites subjected to fast cooling rates. All the spectra were fitted assuming a magnetic hyperfine field and an electric field gradient giving rise to an asymmetric magnetic sextet. The only noticeable differences among them are: a) minor variations of the relative areas of the different absorption peaks and b) even smaller variations in the isomer shift and quadrupole splitting values. The average hyperfine field value for all the samples of this group is 33.99 ± 0.03 T. These parameters, listed in table 4, are associated with a mixture of kamacite and taenite (Böttger et al., 1994; Scorzelli, 1997; Scorzelli et al., 1994; Bonazzi et al., 1994).

Table 4. Mössbauer parameters of Group II meteorites

Meteorite	IS (mm/s)	ΔQ (mm/s)	H (T)	Γ (mm/s)
Bacubirito	-0.022(5)	0.006(2)	33.89(8)	0.34(4)
Casas Grandes	0.018(6)	0.015(6)	33.88(4)	0.31(8)
Charcas	0.016(1)	0.002(1)	33.97(7)	0.42(2)
Chupaderos	0.019(9)	0.003(2)	33.88(6)	0.36(2)
Morito	-0.029(5)	0.004(5)	33.64(6)	0.36(8)
Santiago Papasquiaro	-0.024(3)	0.000(5)	34.07(6)	0.38(2)

CONCLUSIONS

There is general agreement that most of the meteorites that impact the Earth are fragments of the asteroid belt that were created in the initial stages of formation of the solar system. Their study supplies information about their composition and the initial conditions that prevailed in the solar nebulae around 4,600 million years ago.

From the nine studied meteorites, the specimens from group one, besides of the expected magnetic sextets, show a quadrupole doublet. The origin of the quadrupole doublet is hypothesized to have been due to the severe heating of the meteorite when it penetrates the Earth atmosphere followed by a very fast cooling that allows the formation of nanocrystals. Another possibility would be the formation of a crust of γ–FeOOH, due to atmospheric conditions. Unfortunately, no cleaning nor polishing process could be performed in the studied samples to discriminate between both possibilities. The complex spectra of the group one meteorites suggest that they were subjected to different heating and cooling rates as well as different environmental conditions.

Do to the fact that the MP of all the meteorites of the second group (Bacubirito, Casas Grandes, Chupaderos, Morito, Charcas and Santiago Papasquiaro) are essentially equal, we suggest that all of them may be classified as IIIAB. In the Meteoritical Bulletin Database only Bacubirito and Santiago Papasquiaro are clasified as Iron ungrouped; their confirmation as IIIAB must be done by standard chemical methods. All the rest are clasified as IIIAB.

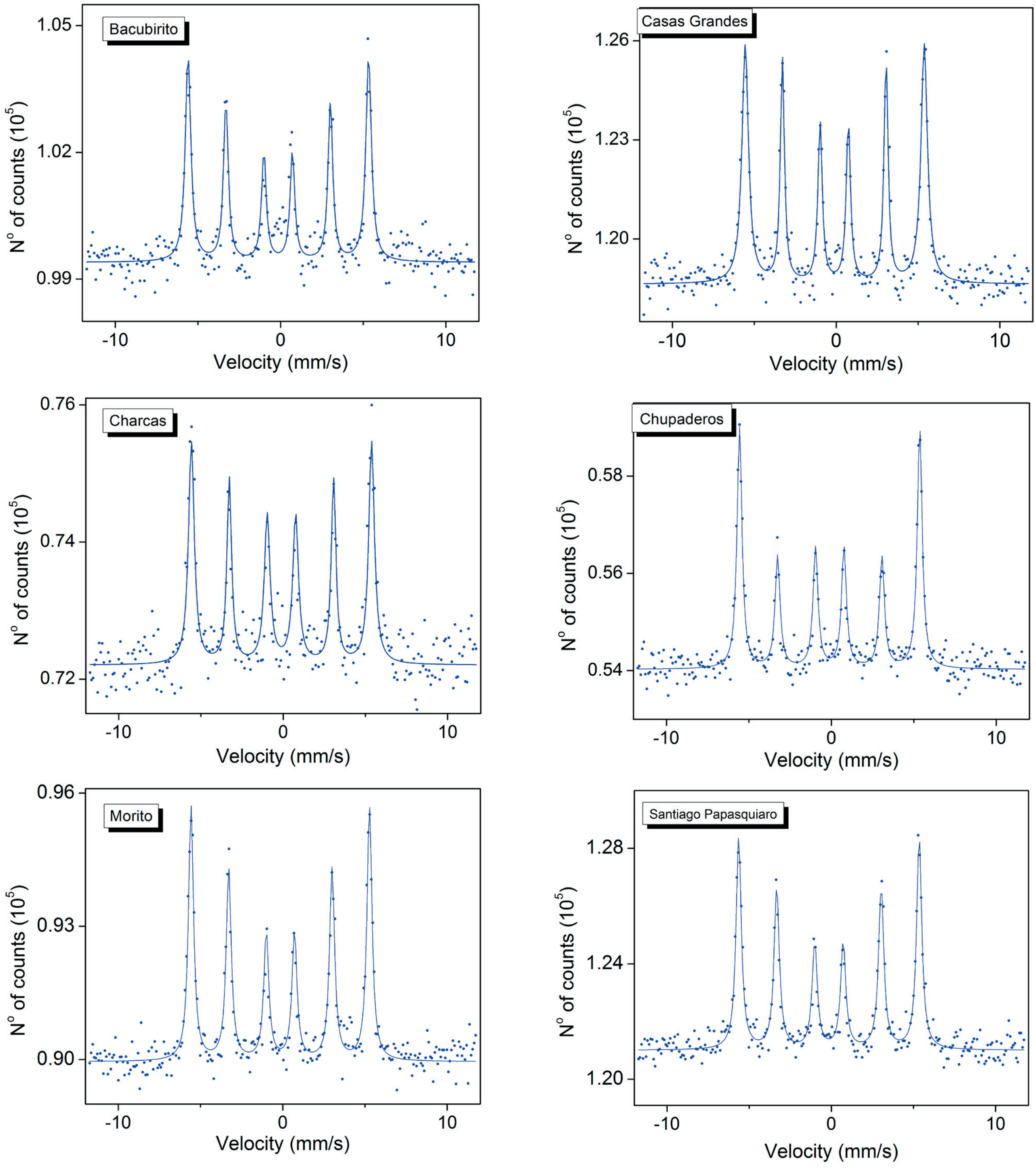

Fig. 6. Mössbauer spectra of the meteorites of the second group

REFERENCES

Angermann E., 1903 – El fierro meteórico de Bacubirito (Est. de Sinaloa): Inst. Geol. Mex., Parergones, V.1 (4),113-116.

Bárcena M., 1876 – On certain Mexican meteorites. Proceedings of the Academy of Natural Sciences. Philadelphia: 122.

Bartlett J.R., 1854 – Personal Narrative of Explorations and Incidents in Texas, New Mexico, California, Sonora, and Chihuahua: Connected with the United States and Mexican Boundary Commission, During the Years 1850, '51, '52, and '53 (Vol. I) and (Vol. II) New York: D. Appleton & Company.

Betteridge S., Dickson D.P.E., Pankhurst Q.A., 1995 – Using polarized Mössbauer spectroscopy to probe magnetic moment orientations in γ-Fe_20_3. *Journal of Magnetism and Magnetic Materials* 140-144, 2213-2214.

Bonazzi A., Jiang K., Ortalli I., Pedrazzi G., Zhang X., 1994 – Investigations on five iron meteorites. *Hyperfine Interactions*, 91, 541-545.

Böttger Ch, Campbell S.J., Wu E., Smith R.G., 1994 – Mössbauer and X-ray studies of an iron meteorite sample. *Hyperfine Interactions* 91, 563-569.

Buchwald V.F., 1968a – Reclassification of Mexican Meteorites. The Geological Society of America, Inc. Seven Meeting Abstracts for 1968, 40-41.

Buchwald V.F., 1968b – A unique iron meteorite from Santiago Papasquiaro. The Geological Society of America, Inc. Seven Meeting. Abstracts for 1968, 40-41.

Buchwald V.F., 1977 – The Mineralogy of Iron Meteorites. *Philosophical Transactions of the Royal Society of London* A. 286 (1336), 453-491.

Buchwald V.F., 2005 – Iron and steel in ancient times. Historisk filosofiske Skrifter 29: 17, The Royal Danish Academy of Science and Letters. Det Kongelige Danske Videnskabernes Selskab.

del Castillo A., 1889 – Catalogue descriptif des météorites (fers et pierres météoriques) du Mexique: avec l'indication des localités dans lesquelles ces météorites sont tombés ou ont été découverts: 15.

Chladni E.F.F., 1794 – Über den Ursprung der von Pallas gefundenen und anderer ihr ähnlicher Eisenmassen und über einige damit in Verbindung stehende Naturerscheinungen. Riga. J.F. Hartknoch.

Fletcher L., 1890 – On the mexican meteorites, with special regard to the supposed occurrence of wide-spread meteoritic showers". *Mineralogical Magazine and Journal of the Mineralogical Society* 9 (42), 91 and 129.

Greenwood N.N., Gibb T.C., 1971 – Mössbauer Spectroscopy. Chapman and Hall Ltd., London 1971.

von Humboldt A., 1810 – Vues des Cordillères et Monuments des Peuples Indigènes de l'Amérique. Paris: 416, Plate 45.

Johnson C.E., 1969 – Antiferromagnetism of γ-FeOOH: a Mossbauer effect study, *J. Phys. C: Solid State Phys.* 2, 1996-2002.

Lagarec K., 2002 – Recoil 1.05. Mössbauer Analysis Software for Windows.

Madsen M.B., Mørup S.M., Costa T.V.V., Knudsen J.M., Olsen M., 1986 – Superparamagnetic component in the Orgueil meteorite and Mössbauer spectroscopy studies in applied magnetic fields. *Nature* 321 (29), 501-503.

Oshtrakh M.I., Grokhovsky V.I., Abramova N.V., Semionkin V.A., Milder O.B., 2009 – Iron–nickel alloy from iron meteorite Chinga studied by Mössbauer spectroscopy with high velocity resolution. *Hyperfine Interactions* 190 (1–3), 317–324.

Persoons R.M., Chambaere D.G., Vandenberghe R.E., Bowen L.H., 1986 – Effect Study of Poorly Crystalline 7-FeOOH. *Phys. Chem. Minerals* 13, 61-67.

Scorzelli R.B., 1997 – A study of phase stability in invar Fe-Ni alloys obtained by nonconventional methods. *Hyperfine Interactions* 110, 143-150.

Scorzelli R.B., Souza Azevedo I., Oratlli I., Pedrazzi G., Bonazzi A., 1994 – Iron-nickel superstructure in metal particles of Alfianello meteorite. *Hyperfine Interactions* 83, 479-482.

Sonnenschmidt F.T., 1804 – Mineralogisch Beschreibung der vorzüglichsten Bergwerks-Reviere von Mexiko oder Neues Spanien, Schleiz: 288.

Tarayre E.G., 1867 – Rapport sur l'exploration minéralogique des regions mexicaines. *Arch. Comm. Sci. Mexique*, Paris, 3, 173, 343, 348, 402.

Ward H.A., 1902 – On Bacubirito or the great meteorite of Sinaloa, Mexico. *Rochester Science*, New Series, 16 (401), 395-398.

Wdowiak T.J., Agresti D.G., 1984. – Presence of a superparamagnetism in the Orgueil meteorite. *Nature* 311, 140-142.

Publishers: Wrocław University of Technology, Faculty of Geoengineering, Mining and Geology
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