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Review

Meteorites: Messengers from the Outer Space

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Milhares de meteoróides, corpos sólidos do espaço extra-terrestre, entram na atmosfera da Terra a cada ano. São pedaços de rocha, metal ou aglomerados de rocha e metal, variando em massa de frações de grama a centenas de quilogramas. Aqueles que sobrevivem à passagem através da atmosfera e caem na Terra, são chamados meteoritos. Os meteoritos são as rochas primitivas, tão antigas quanto o Sistema Solar. O estudo desses objetos enigmáticos intriga os cientistas desde tempos antigos, não apenas por seu local de origem, mas também pelas condições que prevaleceram na sua formação, dando origem a estruturas e composições química e mineralógica características. Essas rochas primitivas são mensageiras do espaço extraterrestre que carregam consigo preciosos segredos sobre a formação do Sistema Solar, depende de nós revelá-los. Nesse artigo apresentamos uma revisão do trabalho realizado no Centro Brasileiro de Pesquisas Físicas (CBPF/MCT), aplicando a espectroscopia Mössbauer ao estudo do sistema Fe-Ni meteorítico.

Tens of millions of meteoroids, solid bodies from the outer space, enter the Earth´s atmosphere each year. They are pieces of stone, iron or stony-iron conglomerates, ranging in mass from fractions of a gram to hundreds of kilograms. Those that survive the passage through the atmosphere and fall to Earth are called meteorites. Meteorites are the most ancient and primitive rocks as old as the Solar system. The study of these enigmatic objects intrigued scientists since ancient times, not only about their place of origin but also about the conditions that prevailed there and gave rise to their characteristic chemical and mineralogical composition and structures. These primitive rocks are messengers from the outer space that carry with them precious secrets about the formation of the Solar system, depends on us to reveal them. In this paper a review of the work done in the Centro Brasileiro de Pesquisas Físicas (CBPF/MCT), applying the Mössbauer spectroscopy to the study of the meteoritic Fe-Ni system will be reported.

Keywords: meteorites, Fe-Ni alloys, tetrataenite, antitaenite, Mössbauer spectroscopy

1. Introduction

Tens of millions of meteoroids, solid bodies from the outer space, enter the Earth´s atmosphere every year. They are pieces of stone, iron or stony-iron conglomerates, ranging in mass from fractions of a gram to hundreds of kilograms. Meteorites are meteoroids that survive passage through the atmosphere and fall to Earth.

Meteorites are the most ancient and primitive rocks as old as the Solar system. The study of these enigmatic objects intrigued scientists since ancient times, not only about their place of origin but also about the conditions that prevailed there and respond for their characteristic chemical and mineralogical composition and structures. These primitive rocks, are messengers from the outer space that carry with them precious secrets about the formation of the Solar system, it depends on us to identify, decipher and reveal them.

The study of meteorites effectively began, at the Brazilian Center for Physics Research, CBPF, by the late Professor Jacques Danon, in the 70´s, when the Danish scientist, Jens Martin Knudsen, called his attention to the importance of Mössbauer spectroscopy on this matter. Since then the Mössbauer effect has been largely applied in the CBPF, to deal with different problems in meteoritics.

In this paper a review of works done by our CBPF group focusing on those involving the application of Mössbauer spectroscopy on the study of the meteoritic Fe-Ni system will be reported.

2. Meteoritic Fe-Ni Alloys

The meteoritic metal does contain a unique complicated microstructure. It has been observed that in slowly cooled

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meteorites their metallic microstructures are dominated by a series of complex phase transformations occurring below 400 °C (Figure 1). These microstructures can be observed in the three main groups of meteorites: *i*) stony meteorites = chondrites (from the mantle of parent bodies); *ii*) stony-iron meteorites (from the mantle/core interface or from collision mixing); and *iii*) iron meteorites (from Fe-rich Fe-Ni cores of parent bodies).

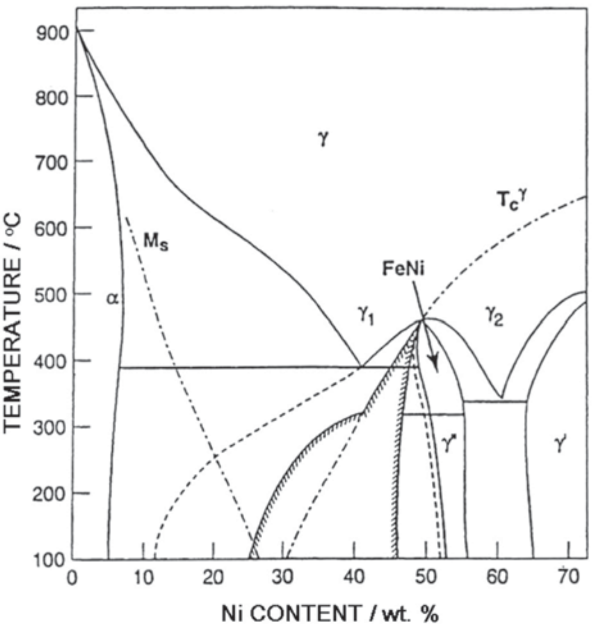

Figure 1. Low temperature portion of the Fe-Ni phase diagram adapted from ref. 20.

Being attracted by the new ideas brought by Prof. Danon after he came back from a visit to the Oersted Institute in Copenhagen, we started the study of meteorites at CBPF. J. M. Knudsen and collaborators discovered in the taenite lamellae extracted from the Cape York and Toluca meteorites (iron meteorites from the group of octahedrites), the presence of an ordered phase in the Fe-Ni system, $Fe_{50}Ni_{50}$, not found on Earth.[1]

Results obtained in a completely different domain allowed a better understanding of the nature of these alloys, detected in meteorites. Starting from Louis Néel results on the relations between ordering phenomena and magnetic properties of binary alloys, the group of the Centre d'Études Nucléaires de Grenoble (CENG), evidenced, in 1962, the presence of an ordered phase, superstructure L_{10}, in the neutron irradiated Fe-Ni 50-50 alloys.[2] This phase is distinguishable by the way it is formed and by their structural, themodinamic and magnetic properties.

As it was first shown in Grenoble, Mössbauer spectroscopy is a technique that allows a relatively easy identification of this ordered phase. The tetragonal distortion of the superstructure L_{10} has its origin in an electric field gradient created at the level of the iron atoms of the alloy. This gives rise to a Mössbauer spectra that is singular among all Fe-Ni alloys (Figure 2).

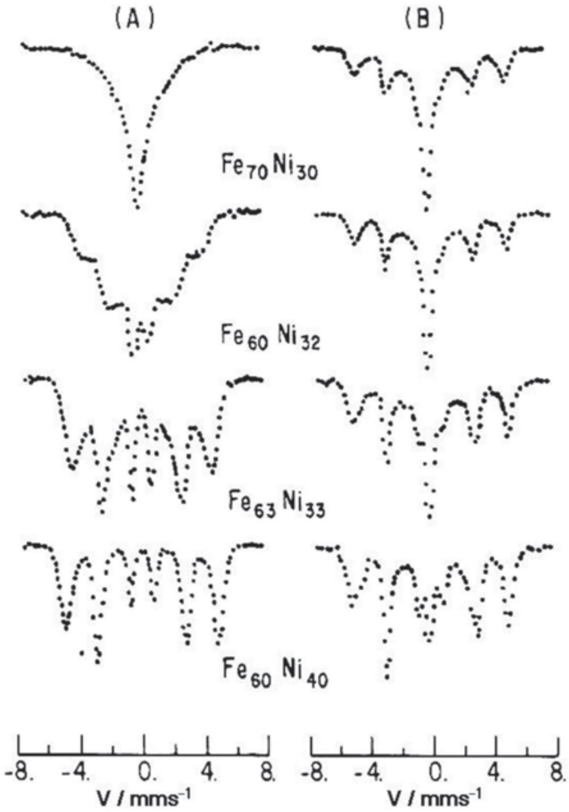

Figure 2. Mössbauer spectra at room temperature: (A) before irradiation and (B) after electron irradiation.

The order-disorder transition temperature of the L_{10} superstructure phase is 320 °C and this explains why it has not been possible to obtain this phase by annealing, since at this temperature the diffusion of metalic atoms is practically halts and a very long time is required for ordering. Cooling rates in the interior of the parent bodies of iron meteorites, as demonstrated by Wood,[3] are expected to be *ca.* 1 °C/10^6 years. It is then in meteorites that the ordered phase could occur in their Fe-Ni constituents. This was first confirmed, 15 years later.[1]

Based on these results we started studying the Santa Catharina meteorite (Figure 3), an iron meteorite of the *ataxite* group with a Ni content (*ca.* 35 wt%) that is one of the highest among the iron meteorites. Found in 1875 in the São Francisco do Sul island in Santa Catarina state, Brazil, this meteorite draw attention because of its high Ni content and its total mass, more than 25,000 kg. The site where it was found became a commercially exploited metalic mine, and its production was continuously exported to England until its complete exhaustion. The scientific interest of its discovery was only recognized by a French Mission, in 1876. But it was only in 1882, after the analysis of samples sent to Paris that it was recognized as a meteorite.

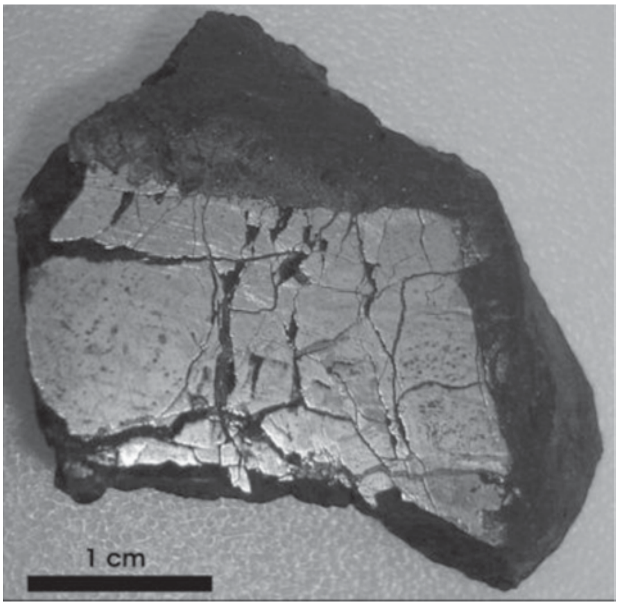

Figure 3. The Santa Catharina ataxite.

We developed a detailed study of the Santa Catharina meteorite, showing the occurrence of a massive amount of the ordered phase $Fe_{50}Ni_{50}$ along with a Ni-poor Fe-Ni phase (Figure 4). These two phases were later recognized as new minerals in meteorites and were named *tetrataenite*[4] and *antitaenite,*[5] repectively. These first results have been presented by Prof. Louis Néel in the Académie de Sciences de Paris, in October 1978.

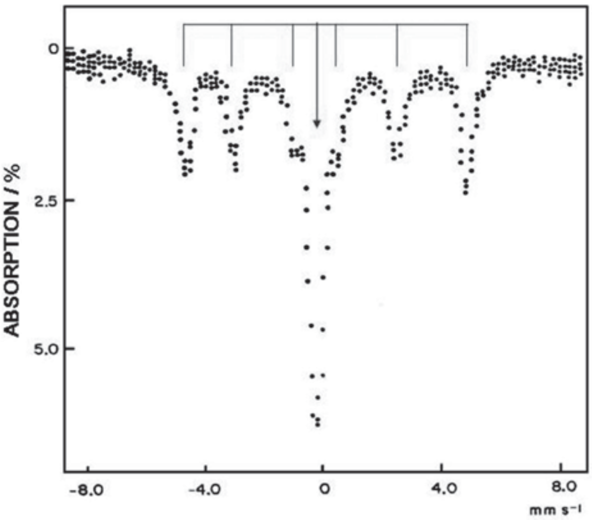

Figure 4. Mössbauer spectra of the Santa Catharina ataxite; the sextet corresponds to the ordered $Fe_{50}Ni_{50}$, tetrataenite, and the arrow corresponds to antitaenite.

We proceed studying the order-disorder processes in the Fe-Ni system of other ataxites with Ni composition in which the Fe-Ni alloys exhibit invar anomalies,[6] similarly as for other groups of meteorites: octahedrites, hexahedrites, metal particles of chondrites and mesosiderites. [7-9]

In the case of the metal particles of chondrites it was possible through the Mössbauer hyperfine parameters to learn about the different thermic and shock history of the 3 different types of chondrites: LL (low low Fe), L (low Fe) and H (high Fe). The behaviour of the ordered phase as a function of shock and temperature showed that the high relation Ni/Fe present in the LL chondrites favours the formation of the ordered phase, also common in L chondrites in minor proportion, and in certain cases can also be found in H chondrites.

The comparison between the meteoritic Fe-Ni alloys and electron irradiated Fe-Ni invar alloys showed that despite the similar composition of the meteorite phases and the irradiated alloys, the order degree is much higher in meteorites. The thermic variation of the lattice parameters indicates that the meteoritic Fe-Ni phases exhibit the same properties as for the irradiated Fe-Ni invar alloys. Basically, radiation enhances the phase segregation kinetics and the ordering of the alloy. The same process occurs in meteorites, but in a much different time scale, due to the exceptionally slow cooling rate of meteorites in their parent bodies.

Since shock effects have an important role in a meteorite history, we studied the solid state transformations induced by shock in ordered Fe-Ni alloys from meteorites. It is estimated that 65% of iron meteorites have been submitted to shock pressures above 130 kbar in pre-terrestrial impacts.[10] At that time the transformations introduced by shock effects in ordered Fe-Ni alloys was not well understood. The only interesting results reported were related to the Cu_3Au superstructure.[11] In order to study shock effects in the Fe-Ni system, we apply the flying plates technique[12] to simulate the effects of shock-waves up to 200 kbar on the disordering processes of meteoritic Fe-Ni ordered alloys, with Mössbauer spectroscopy, optical and electron microscopy, electron microprobe and X-ray diffraction techniques.

Results of the induced shock pressures showed that, in the Santa Catharina iron meteorite, the degree of long range order is reduced, similarly to what has been reported for the Cu_3Au alloy. In the metallic particles of chondrites, there is a tendency to decrease the degree of the long range order, but the alteration by shock is less evident in the hyperfine parameters, probably because the degree of order in the metal particles of chondrites is smaller than in the Santa Catharina ataxite. Consequently it would be necessary to apply higher shock pressures in chondrites, to have disordered effects in the same order of magnitude.

Since the experiments done in Grenoble there was an unsolved problem in the understanding of the irradiated and meteoritic alloys. It occurs that the ordered phase $Fe_{50}Ni_{50}$ superstructure L_{10}, in irradiated alloys as well as in meteorites, is always associated with another Ni-poor phase, which appears in the Mössbauer spectrum at room temperature as a paramagnetic singlet. This phase was initially referred to as "paramagnetic γ-phase".[13] This paramagnetic contribution is always observed in coexistence with a magnetic sextet corresponding to the ordered phase, *tetrataenite,* and was previously interpreted as being due to an atomic disordered γ-phase (*taenite*). The Ni content of this phase is low enough for its magnetic ordering temperature to be well below the room temperature. Another interpretation would be to consider this phase as an ordered Fe_3Ni.[14] However it is difficult to conciliate these two interpretations as their characteristics do not fit to any of the suggested phases.

Several experiments have been performed aiming to analyse the behaviour of this phase as a function of temperature and of applied magnetic fields. Variable temperature measurements[15] of the "paramagnetic phase" in the Santa Catharina ataxite have shown line broadening that sets in at an ordering temperature of *ca.* 25 K as the temperature lowered. At the lowest temperatures, the broadening is such that it must correspond to a very small saturation value of the hyperfine field. Under applied fields up to 80 kG[16,17] the hyperfine splitting remains small, showing that the small value is due to the electronic structure rather than dynamic effects such as superparamagnetism. Also, whenever the paramagnetic contribution is observed, it always coexists with a hyperfine sextet pattern that is unambiguously attributed to *tetrataenite* (atomically ordered or partially ordered FeNi phase, 50 at% Ni). [5,18,19]

The above mentioned difficulties in the interpretation of the Ni-poor phase, as seen by Mössbauer spectroscopy, can be resolved with a new interpretation in which we assign the meteoritic paramagnetic phase to a γ-low spin (γ_{LS}) Fe-Ni phase. Its magnetic ordering is the antiferromagnetic transition of the γ_{LS}-phase and T_N can be used to estimate its composition. Its low saturation hyperfine field value is a consequence of its low-spin electronic structure. The γ_{LS}-phase (with estimated composition *ca.* 25-30 at% Ni) has the same ambient conditions lattice parameter as *tetrataenite* (having various degrees of atomic order) in that the X-ray diffraction lines of the two phases have not been resolved in any meteorite sample by classical diffraction methods. Both phases have smaller lattice parameters than their respective same-composition γ-high spin (γ_{HS})-phase counterparts. In our interpretation the γ_{LS}-phase occurs in an epitaxial association with *tetrataenit*e. Both phases have practically indistinguishable lattice parameters and form a very fine intergrowth (< 0.1 μm), IG (Γ_{LS}/tetrataenite). This intergrowth is an indicative of an equilibrium state at low temperature, in this composition range. This interpretation suggest a specific name for this potential new mineral, γ_{LS}-phase Fe-Ni having *ca.* 25-30 at% Ni and the same ambient condition lattice parameter as *tetrataenite*. Since it is a taenite and its main fingerprint characteristic making it distinguishable from the γ_{HS}-phase is its antiferromagnetism, we proposed that it be called *antitaenite.* This also stresses its differences with ordinary (high-spin) taenite. The phase was then recognized as a new mineral and referred in the literature with the proposed name. However the great difficulty in detecting this phase by diffraction methods remained until recently when Synchrotron X-ray diffraction investigation of iron meteorites carried out at the *Laboratório Nacional de Luz Sincrotron* (Campinas, Brazil), confirmed for the first time, by a diffraction method, the presence of a disordered face centered cubic phase (FCC), space group Fm-3m, with stoichiometry $Fe_{0.6665}Ni_{0.3335}$ and lattice parameter of 0.3588 nm (Figure 5). This phase agrees with the previously proposed *antitaenite*.[5] The other Fe-Ni phase detected is the ordered phase and iron-deficient *tetrataenite* ($Fe_{0.4356}Ni_{0.5644}$).

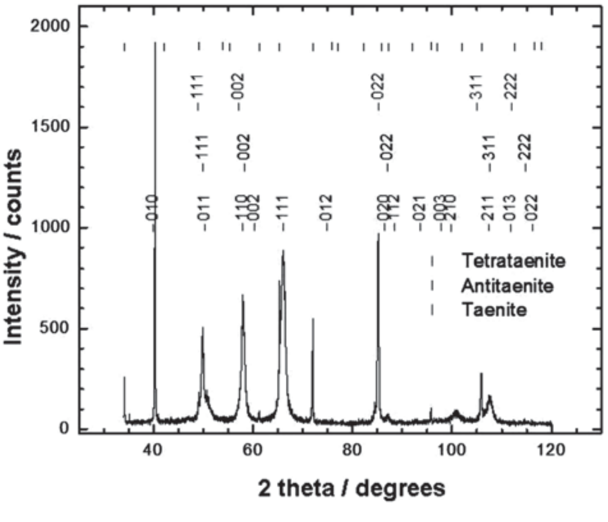

Figure 5. SR-XRD diffractogram of the Santa Catharina meteorite; tetratenite = 34 wt%, antitaenite = 63 wt%, disordered taenite = 3 wt%.

3. Conclusion

The study of meteoritic metal is an attempt to determine the Fe-Ni phase diagram experimentally. Meteoritic metal is basically a Fe-Ni alloy containing from 5 to 60 at% Ni with small amounts (<1 wt%) of Co, P, S and C. Because meteorites have cooled slowly over millions of years (1 to 1000 million years) in their asteroidal bodies, meteoritic metal contains a characteristic structure which cannot be completely duplicated in the laboratory due to the slow diffusion process at low temperatures. Therefore, meteorites are useful as indicators of the low temperature phase transformation which occurs in Fe-Ni alloys. The microstructure of the low temperature phase transformation products in meteoritic metal is similar in stony, stony-iron and iron meteorites. Differences in the microstructure are most likely a function of cooling history at low temperature. The Mössbauer spectroscopy technique, introduced by Prof. J. Danon in CBPF, is continuously giving a great contribution to the study of the meteoritic alloys as well as to other problems involved in meteoritics research.

Glossary

antitaenite, γ_{LS} (gamma low spin phase) – face-centered cubic γ-FeNi with Ni ℑ 30 wt%.

ataxite – an iron meteorite with high nickel content, composed almost entirely of taenite and having no obvious structure.

chondrite – an abundant class of stony meteorites characterized by chemical compositions similar to that of the Sun and by the presence of chondrules.

hexahedrite - an iron meteorite with low nickel content, consisting almost exclusevily of kamacite.

invar – FeNi alloy with Ni ∀35%. This alloy is known for its unique properties of controlled coefficient of thermal expansion.

iron meteorite – a meteorite composed primarily of iron-nickel metal (see **ataxite**, **hexahedrite**, **octahedrite**).

kamacite – body-centered cubic, γ-FeNi with Ni < 7.5 wt%.

lamellae – plural of lamella (latin). A thin layer within a geological material.

mesosiderite – a class of stony-iron meteorites consisting of metal and fragments of igneous rocks.

octahedrite – an iron meteorite of intermediate nickel content, containing both kamacite and taenite in a Widmanstätten pattern.

parent body – an object of asteroidal size or larger from which meteorites were derived.

taenite – face-centered cubic γ-FeNi; in meteorites the name is used for compositions of about 25-48% Ni.

tetrataenite – tetragonal FeNi with about 48-54% Ni, atomically ordered.

Widmanstätten pattern – a regular geometric intergowth of kamacite plates within taenite that occurs in some iron meteorites (see **octahedrite**)

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Chemist, Universidade Federal do Rio de Janeiro (1963); MSc. (Geochemistry), Universidade Federal Fluminense (1976); PhD. (Docteur d'Etat és Sciences, Sciences Phisiques), Université Pierre et Marie Curie - Paris VI - Mention Très Honorable - (1982); Pos-Doc, MIT, Francis Bitter Magnet Laboratory (1983); Full Professor, Centro Brasileiro de Pesquisas Físicas (CBPF/MCT); Vice-Director CBPF (2001 - 2005); Chief Condensed Matter Department (2001-2005); Vice-Coordinator Experimental Low Energy Physics Coordination; Main Topics of Interest: Meteoritics, Minerals and Rocks, Archaeometry; The International Astronomical Union named Scorzelli a metallic asteroid discovered by S. J. Bus. The homage was received in the 2006 Meteoritical Society Meeting, in Zürich, in recognition of the Mössbauer work on metallic meteorites. Since then this asteroid is referred to as 7735 SCORZELLI (1980 UL1), http://neo.jpl.nasa.gov/orbits/.

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