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Comparative Mössbauer studies of the Baszkówka ordinary chondrite and some other meteorites

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It was suggested that perhaps Mössbauer parameters of troilite could serve as a criterion for an interpretation of the origin of various meteorites. It was found that in three meteorites coming from the same parent body (Baszkówka, Mt. Tazerzait and Songyuan) the angle between the principal axis of the electric field gradient (EFG) and the direction of the magnetic field in troilite is around 60°. Further investigations showed that in some other meteorites, not belonging to this group, this angle is close to 90°.

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INTRODUCTION

Astronomical analysis as well as petrographic data indicate that some ordinary chondrites could come from the same parent body. For example, the majority of strongly shocked L chondrites are believed to originate from one parent body, whereas almost unshocked and highly porous L chondrites like Baszkówka, Mt. Tazerzait, and Tjerebon are supposed to be fragments of another parent body (Pilski and Walton, 1998). An attempt was made to see whether Mössbauer spectroscopy can find a common feature in spectra of meteorites originating from the same parent body, which can distinguish them from spectra obtained for meteorites from other parent bodies. Mössbauer studies of four samples taken from different parts of the Baszkówka meteorite showed a variability in mineral composition. This observation proves that the distribution of iron among different mineral phases cannot be a good criterion for the origin of various meteorites. It was suggested that the Mössbauer parameters of meteoritic troilite could serve as such a criterion (Gałązka-Friedman *et al.*, 1999).

MÖSSBAUER SPECTROSCOPY

Mössbauer spectroscopy is applicable to different isotopes, but is most sensitive for ^{57}Fe. The Mössbauer spectra reflect the properties of the environment of the iron atoms. Each iron-bearing compound gives a characteristic Mössbauer spectrum. The spectra show the counting rate after the absorption of the gamma rays (14.4 keV for ^{57}Fe emitted by a radioactive ^{57}Co source) in the investigated sample (the absorber), as function of the source velocity. Mössbauer spectra are characterised by the following Mössbauer parameters: the isomer shift (IS),

Table 1

Iron distribution (atomic %) among mineral phases in four samples of the Baszkówka meteorite

Subspectrum	Sample 1	Sample 2	Sample 3	Sample 4
Kamacite	23	14	13	29
Troilite	25	28	33	26
Olivine	33	35	28	26
Pyroxene	16	21	24	18
Fe^{3+}	3	2	2	1

the quadrupole splitting or quadrupole interaction (QS) and the value of the internal magnetic hyperfine field (H_{eff}). The isomer shift, quadrupole splitting and line widths are given in velocity units (mm/s). The relation between these units and energy units can be easily calculated from the equation:

$$\Delta E = E_\gamma (v/c)$$

where: E_γ — the energy of the emitted gamma rays (14.4 keV for ^{57}Fe); v — the velocity of the radioactive source, c — the velocity of light; for ^{57}Fe, a change in velocity of 1 mm/s is equivalent to a change in energy of $4.8\ 10^{-8}$ eV; the values of the magnetic hyperfine fields are usually given in kOe or Tesla.

In minerals there are usually two types of Mössbauer spectra for ^{57}Fe: doublets and sextets (consisting of two or six lines). Sextets are typical for iron compounds with magnetic ordering; in nonmagnetic compounds only doublets are observed. In doublets the distance between the two lines yields the quadrupole splitting and the centre between the two lines — the isomer shift. In magnetic spectra the positions of the lines depend on the value of the isomer shift (IS), the quadrupole interaction (QS), the hyperfine field (H_{eff}) and the angle between the principal axis of the electric field gradient (EFG) and the direction of the internal magnetic field.

Mössbauer studies of meteorites started shortly after the discovery of the Mössbauer effect in 1958. The first paper dealing with this subject was published in 1964 (Sprenkel-Segel and Hanna, 1964). Mössbauer measurements were used to identify different mineral phases and to determine their relative amounts.

A typical Mössbauer spectrum of non-weathered ordinary chondrite is a superposition of two sextets and two doublets. One sextet is connected with metallic iron within an iron-nickel alloy (kamacite), the other with Fe^{2+} in iron sulphide (troilite) of composition FeS. Doublets correspond to ferrous iron present in silicate phases such as olivine $(Mg, Fe)_2[SiO_4]$ and orthopyroxene $(Mg, Fe)[SiO_3]$.

Sometimes subspectra corresponding to ferric iron appear in meteoritic Mössbauer spectra. Many papers were devoted to the problem of the origin of this ferric iron. Some ferric compounds in meteorites could be the product of terrestrial weathering (Berry *et al*., 1994; Shinonaga *et al*., 1994) and others may be due to pre-terrestrial oxidation (Knudsen, 1989; Burns, 1994). Many papers were devoted also to kamacite (Scorzelli, 1991).

The first Mössbauer studies of the Baszkówka meteorite were presented in 1996 (Gałązka-Friedman *et al.*, 1996). The Baszkówka chondrite consists of chondrules and their fragments, rock fragments, silicate mineral crystals as well as of irregular grains of kamacite and troilite (Gałązka-Friedman *et al*., 1998; Stępniewski *et al*., 1998). The main silicate minerals of both the chondrules and the matrix are olivine and orthopyroxene. The composition of olivine determined by X-rays is about Fa_{20} (Siemiątkowski, 2001), while that determined by an electron microprobe ranges from Fa_{23} to Fa_{26}, giving an average value of $Fa_{25.3}$ (Stępniewski and Borucki, 2001). The microprobe analyses of orthopyroxenes give results ranging from Fs_{18} to Fs_{23} with an average value of $Fs_{20.4}$ (Stępniewski and Borucki, *op. cit.*).

Mössbauer studies performed on four different samples of the Baszkówka meteorite (Gałązka-Friedman *et al*., 1996, 1998) showed the presence of the four above-mentioned iron-bearing mineral phases: olivine, pyroxene, kamacite and troilite and a small amount of trivalent iron. The Mössbauer spectrum of this trivalent iron was a doublet with IS = 0.45 mm/s and QS = 0.75 mm/s. These parameters are somewhat different from those of the products of terrestrial weathering (Burns, 1994). As the Baszkówka meteorite is a comparatively young fall, we suggest that this trivalent iron was created during the cosmic life of the meteorite. This assumption is confirmed by Mössbauer measurements of isolated fractions of the Baszkówka meteorite (see below).

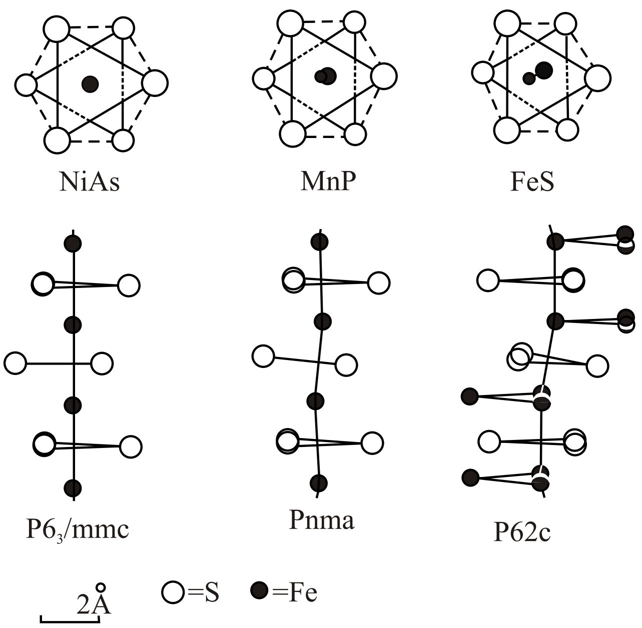

Fig. 1. Structures of FeS from left to right: NiAs-type, MnP-type and troilite structures; at the top, octahedral iron sites viewed from above are shown

Table 2

Mössbauer parameters of troilite from different meteorites

Meteorite	Reference	IS [mm/s]	QS [mm/s]	H [T]
Numakai	Shima *et al.* (1975)	0.76	–0.06	31.6
Toluca	Ousef *et al.* (1979)	0.76	0.26	31.5
Rittersgrun	Just *et al*. (1984)	0.67	–	31.3
Putinga	de Oliveira *et al.* (1988)	0.76	0.30	31.1
Torino	Ortalli and Pedrazzi (1990)	0.75	0.26	30.6
Alfinello	Bonazzi *et al.* (1992)	0.73	0.17	31.1
Alenak	Bonazzi *et al.* (1992)	0.79	0.18	31.1
Siena	Bonazzi *et al.* (1992)	0.75	0.17	31.1
Aigle	Bonazzi *et al.* (1992)	0.74	0.16	31.1
Pieve di Casa	Bonazzi *et al.* (1992)	0.77	0.19	31.6
Xinyang	Zhang *et al.* (1994)	0.61	0.31	30.2
Jilin	Zhang *et al.* (1994)	0.67	0.16	31.2
Al Kidirate	Gismelseed *et al.* (1994)	0.78	–0.15	31.2
RC 063	Berry *et al.* (1994)	0.76	–	31.5
RC 099	Berry *et al.* (1994)	0.68	–	30.6
RC 035	Berry *et al.* (1994)	0.43	–	28.7
Y74155	Shinonaga *et al.* (1994)	0.81	0.15	31.4
Y791428	Shinonaga *et al.* (1994)	0.82	0.15	31.4
ALH77299	Shinonaga *et al.* (1994)	0.81	0.15	31.4

The percentages of iron found in different mineral phases in four samples of the Baszkówka meteorite are listed in Table 1.

The variability of the mineral composition observed in the four samples of the Baszkówka meteorite indicates that the distribution of iron among different mineral phases cannot be a criterion for determining the parent body of various meteorites.

MÖSSBAUER STUDIES OF TROILITE PRESENT IN DIFFERENT METEORITES

THE CRYSTAL STRUCTURE OF TROILITE

Iron monosulphide phases are characterised by a multiplied unit cell with respect to the NiAs-type structure, and often possess a lower lattice symmetry (Hafner and Kalvius, 1966; Kruse and Ericsson, 1988; Kruse 1993; *cf*. Fig. 1). At room temperature FeS occurs as hexagonal troilite with P62c space group. In this structure, all Fe atoms form triangular three-atom clusters in planes perpendicular to the *c* axis. Each Fe atom is surrounded by a distorted sulphur octahedron. The Fe cluster breakup may be caused either by increased temperature, increased pressure, or by vacancies at Fe sites. On heating, the transition of troilite into the orthorhombic MnP-type structure (Pnma) can be completed at 140°C, but this transformation in meteoritic material is not fully reversible, as after slow cooling to 7°C, 20% of the Fe atoms may remain in the MnP structure. On heating, MnP-structured iron monosulphide transforms into the NiAs structure with space group $P6_3$/mmc at about 225°C.

HYPERFINE PARAMETERS OF TROILITE

Table 2 shows Mössbauer parameters of troilite obtained at room temperature from 19 different meteorites in 9 different

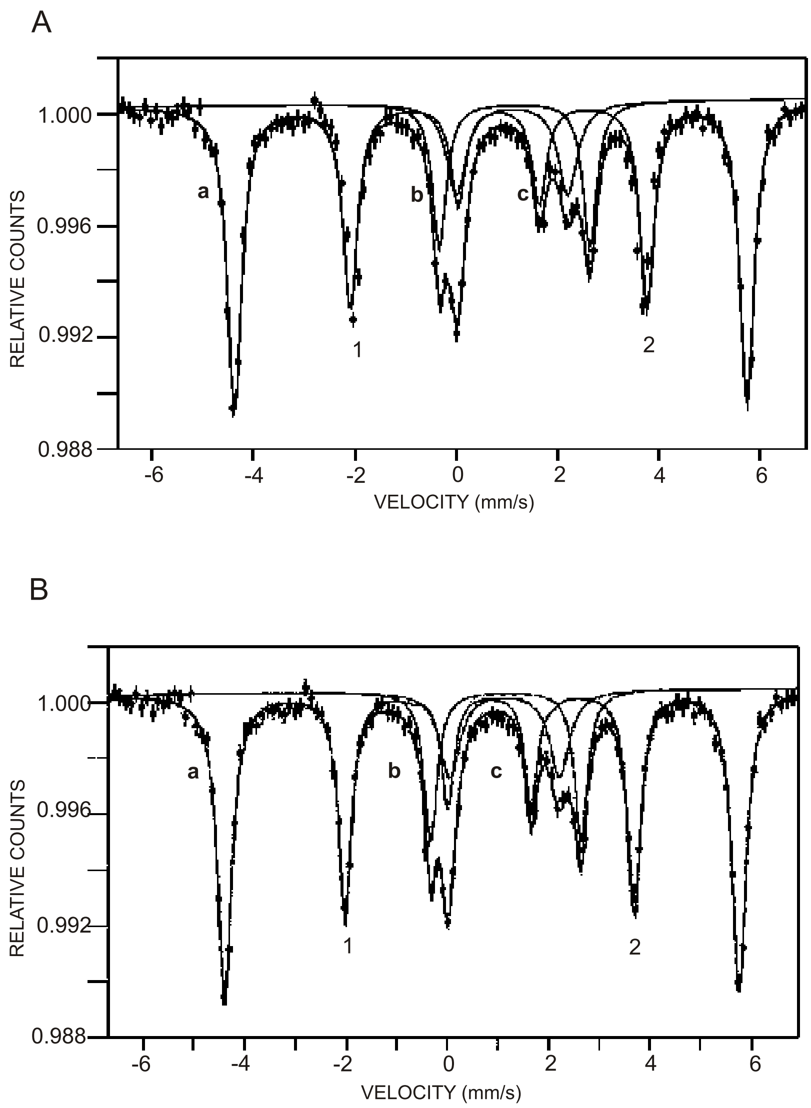

Fig. 2. Mössbauer spectrum of troilite isolated from the Baszkówka chondrite

The solid lines are the theoretical lines obtained from computer fits to the experimental data points and show the subspectra: sextet **a** for troilite, doublet **b** for olivine and doublet **c** for pyroxene; A — fit with Θ = 0; B — fit with Θ = 61.4°; observe the bad fit to lines 1 and 2 in A

laboratories. The data were published in 10 different papers between 1975 and 1994. In all these analyses of the troilite subspectrum it was assumed that the principal axis of the EFG is colinear with the direction of the magnetic hyperfine field.

In Table 2 isomer shifts are given relative to iron metal. Typical values given for the errors of isomer shifts (IS) are ±0.02 mm/s, of quadrupole splitting (QS) ±0.04 mm/s and of magnetic fields (H_{eff})±0.5 T. A large spread in the values obtained for these parameters can be easily noticed. For the isomer shift the values range between 0.67 and 0.82 mm/s and for QS between –0.15 and +0.31 mm/s. The reasons for such a spread could be different amounts of vacancies or impurities in different troilites (Ok *et al.*, 1982; Baek *et al.*, 1984; Kruse, 1990) or the neglecting of the angle between EFG and H_{eff} in these fits. As in all above measurements troilite was only a small fraction of iron-containing compounds in the samples, we decided to isolate troilite from Baszkówka and Mt. Tazerzait meteorites in order to obtain more accurate Mössbauer parameters.

NEW MÖSSBAUER STUDIES OF DIFFERENT IRON-CONTAINING COMPOUNDS ISOLATED FROM BASZKÓWKA AND MT. TAZERZAIT METEORITES

MATERIALS

In order to avoid iron oxidation, the samples of bulk meteorites were crushed, then ground into fine powder in an agate mortar under ethyl alcohol. After grinding, the samples were dried at room temperature and loaded, as soon as possible, into Mössbauer plastic holders. Troilite was isolated from the gradually crushed chondrites using combined magnetic and

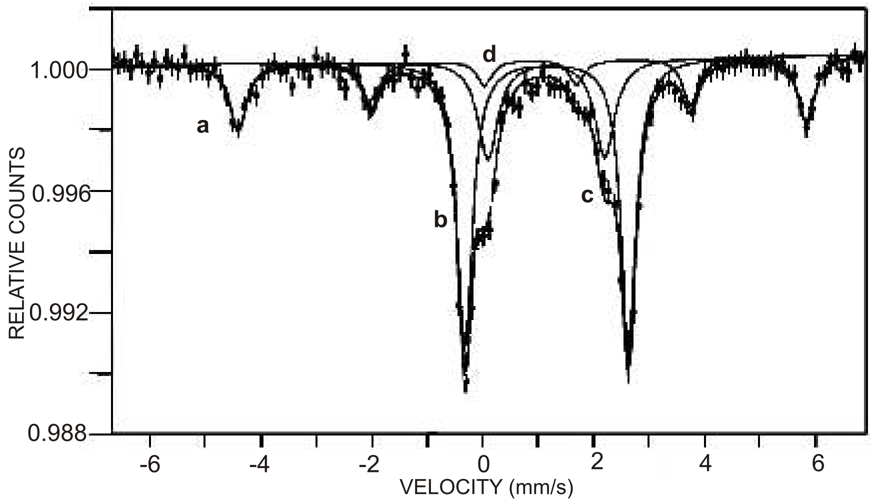

Fig. 3. Mössbauer spectrum of troilite separated from the Mt. Tazerzait chondrite

Subspectra **a**, **b** and **c** as in Fig. 2; doublet **d** corresponds to Fe^{3+}; in the theoretical spectrum Θ = 61.3°

hand-picking with a needle techniques. First, kamacite was removed from the crushed material using a permanent magnet, then troilite grains were isolated from the remaining minerals by hand-picking under a stereoscopic microscope. The last method was also applied for obtaining the concentrates of olivine. The fine-grained separates were loaded into the plastic holders. The separation into the different phases was not complete.

METHOD

Mössbauer spectra of ^{57}Fe were measured at room temperature with a conventional Mössbauer spectrometer. A 25 mCi ^{57}Co-in-Rhodium source, giving a narrow unsplit emission line, was used. The 14.4 keV gamma rays were detected using a proportional counter. The velocity scales were calibrated using an iron foil absorber at room temperature. All experimental Mössbauer spectra were fitted with Lorentzian lines, the line widths, quadrupole splittings (QS), isomer shifts (IS), the magnetic hyperfine fields (H_{eff}) and the relative intensities of various subspectra being free parameters in these fits. At first all spectra were analysed assuming that the angle between the principal axis of the EFG and H_{eff} is zero, but later it was realised that no good fits to the troilite subspectrum could be obtained with this assumption and that an additional parameter Θ = 0, representing this angle, has to be introduced.

RESULTS

In Figure 2 the Mössbauer spectrum of troilite isolated from Baszkówka is shown. Figure 2A shows the fit assuming Θ = 0, Figure 2B the fit with Θ as a free parameter, yielding Θ = 61.4°. The points with error bars are the experimental points and the lines represent the best theoretical fits to the experimental spectra. In the figure subspectrum **a** represents the sextet attributed to troilite, subspectrum **b** — the doublet attributed to olivine and subspectrum **c** — the doublet attributed to pyroxene. All subspectra are shown. Numbers 1 and 2 point to second and the fifth line of the troilite sextet. It is easy to notice, that in Figure 2A the theoretical lines do not follow the experimental points of these two peaks. In Figure 2B, when the angle Θ was introduced as a free parameter, the fit of the theoretical lines to the experimental points is perfect.

The spectrum obtained from troilite isolated from the Mt. Tazerzait meteorite is shown in Figure 3. This spectrum was also fitted with Θ as free parameter.

Mössbauer parameters obtained from the best fits to the subspectra of troilite isolated from Baszkówka and Mt. Tazerzait meteorites are given in Table 3. Within the limits of errors, these parameters are the same.

Table 3

Mössbauer parameters of troilite present in seven ordinary chondrites

Meteorite	IS [mm/s]	QS [mm/s]	H [T]	Θ [deg]
Baszkówka (isol)	0.76±0.01	1.03±0.02	30.9±0.1	61.4±0.1
Mt. Tazerzait (isol)	0.78±0.02	1.05±0.05	31.3±0.1	61.3±1.0
Songyuan	0.76±0.01	0.89±0.05	30.9±0.1	61±5.0
El Hammami	0.75±0.01	0.90±0.02	31.1±0.1	63±2.0
Grzempy	0.63±0.01	0.68±0.03	30.1±0.1	90±10
Mbale	0.68±0.01	0.68±0.02	30.7±0.1	90±10
Tsarev	0.66 ±0.01	0.88±0.06	30.3±0.1	90±15

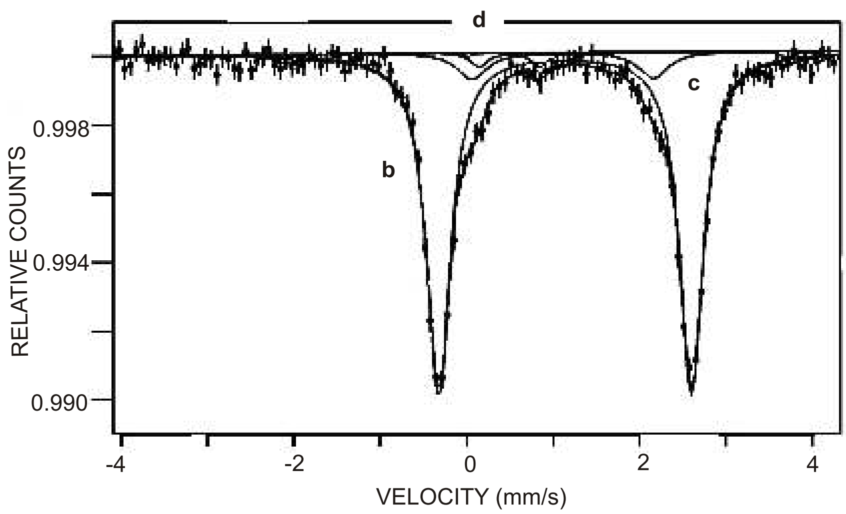

Fig. 4. Mössbauer spectrum of olivine (**b**) isolated from the Baszkówka chondrite

Small amounts of pyroxene (doublet **c**) and of trivalent iron (doublet **d**) are also present

Mössbauer spectra of the olivine and silicate (olivine and pyroxene) phases isolated from the Baszkówka meteorite were also obtained. In Figure 4 the Mössbauer spectrum obtained for olivine (**b**) is shown and a weak subspectrum corresponding to pyroxene (**c**) as well as a weak doublet with parameters characteristic for trivalent iron (**d**) are also observed.

Figure 5 shows the Mössbauer spectrum obtained for two silicate phases: olivine (**b**) and pyroxene (**c**). Again weak subspectra corresponding to other phases are also observed: (**d**) — the doublet corresponding to trivalent iron and (**a**) — the subspectrum due to troilite is seen here. The fact that the trivalent iron was separated with both silicate phases indicates that it is within the structure of these phases and is not a nano-phase product of terrestrial weathering.

PRELIMINARY MÖSSBAUER STUDIES OF SIX OTHER ORDINARY CHONDRITES

Mössbauer studies of six other ordinary chondrites: Songyuan (L6), Tsarev (L5), Mbale (L6), Grzempy (H5), El Hammami (H5) and Marlow (L5) were performed. The samples were prepared from bulk pieces of the meteorites, without isolation of the mineral phases. In five meteorites: Songyuan, Tsarev, Mbale, Grzempy and El Hammami the same minerals (kamacite, troilite, olivine and pyroxene) as in the Baszkówka meteorite were identified. In the sixth one (Marlow) there was no kamacite and only a very small amount of troilite. We were able to identify subspectra of oxides and hydroxides. It seems that the Marlow meteorite is strongly weathered and that the metallic and sulphide phases were oxidised in it. In the five non-weathered meteorites we were able to determine the angle Θ of troilite.

The value of Θ obtained for the Songyuan meteorite (61±5°) is very close to the value obtained for Baszkówka and Mt. Tazerzait. This result correlates well with other observations, which suggest that Songyuan belongs also to "the Baszkówka family" (Morgan, 1999, pers. comm.). The values of Θ obtained for other meteorites, not belonging to this group, are close to 90°. All obtained parameters are summarised in Table 3. There are differences also in the other Mössbauer parameters between the various meteorites.

Thus the isomer shift for the Grzempy, Mbale and Tsarev troilite is between 0.63 and 0.68 mm/s, whereas for others it is 0.77±0.02 mm/s. The QS of troilite from the 3 above mentioned meteorites is between 0.68 (Grzempy and Mbale) and 0.88 mm/s (Tsarev), whereas it is 1.02 for Baszkówka and Mt. Tazerzait troilite. For the Songyuan troilite it is 0.89±0.05 mm/s and for El Hammami 0.90±0.02 mm/s.

The Mössbauer parameters of the silicates are the same, within limits of errors, in all 7 meteorites and are: IS = 1.17±0.01 and QS = 3.00±0.03 mm/s for olivine and IS = 1.17±0.01 and QS = 2.13±0.03 mm/s for pyroxene.

As can be seen from Table 3, Θ was determined more accurately for the Baszkówka and Mt. Tazerzait meteorites than for the other meteorites. For the Baszkówka and Mt. Tazerzait meteorites the parameters were evaluated from isolated troilite, whereas for the other meteorites, bulk samples were used.

CONCLUSIONS

The Mössbauer studies of four bulk samples, taken from four different parts of the Baszkówka meteorite, showed the presence of four iron-bearing compounds: kamacite, troilite, olivine and pyroxene. This is the typical mineral composition

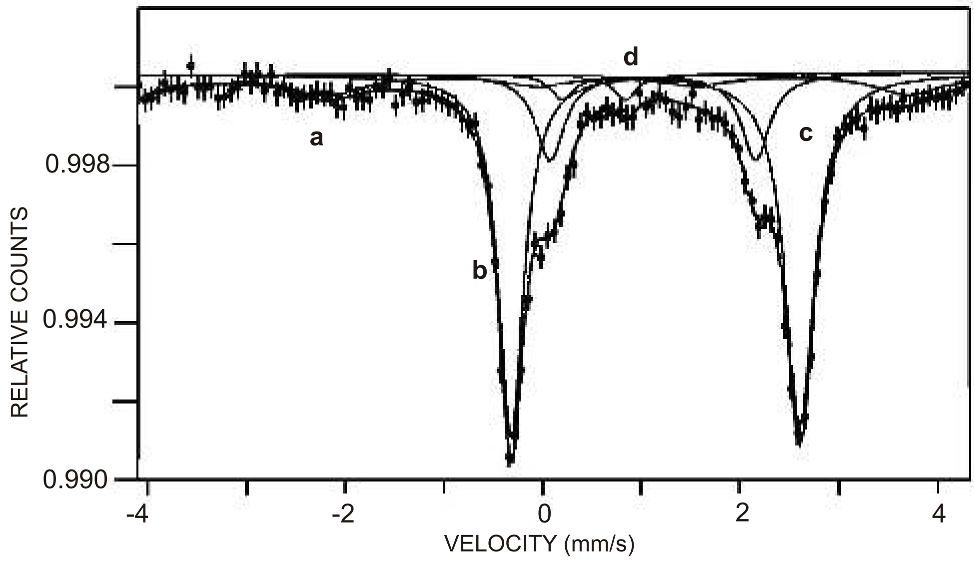

Fig. 5. Mössbauer spectrum of the silicate phases (olivine — **b** and pyroxene — **c**) isolated from the Baszkówka chondrite; subspectra corresponding to trivalent iron (**d**) and to troilite (**a**) are also observed

of ordinary non-weathered chondrites. The different distribution of the four mineral phases in various parts of the same meteorite shows that this distribution cannot serve as a good criterion for distinguishing between the parent bodies of various meteorites.

Mössbauer studies of troilite isolated from a large part of the Baszkówka meteorite point to the necessity of including the angle Θ (the angle between the main axis of the electric field gradient and the direction of the magnetic field) in the fitting of Mössbauer spectra of troilite. As astronomical analysis and petrographic features indicate that Baszkówka and Mt. Tazerzait meteorites could come from the same parent body, the equality of the angles Θ and the other Mössbauer parameters obtained for troilite isolated from Baszkówka and Mt. Tazerzait meteorites, brought about the hypothesis that these parameters could yield a good criterion for identifying the parent body of different meteorites. Preliminary Mössbauer studies of six other ordinary chondrites seem to support this hypothesis, however, in order to prove it, more spectra of troilite isolated from different meteorites have to be measured.

The presence of Fe^{3+} in the samples of isolated olivine and silicate phases (olivine and pyroxene) is in agreement with the suggestion that ferric iron found in the Baszkówka meteorite is a product of pre-terrestrial oxidation (Ostertag *et al.*,1984).

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