TEMPERATURE DEPENDENCE OF THE MÖSSBAUER PARAMETERS OF THE Fe—Ni PHASES IN THE SANTA CATHARINA METEORITE

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The temperature variation in the range 8-760K of the hyperfine parameters of the Fe-Ni phases in the Santa Catharina meteorite has been determined. It is suggested that the disordered 50-50 Fe-Ni phase actually consists of two distinct fractions, i.e. a completely disordered phase and one with intermediate long-range ordering parameter. The single-line subspectrum of the 28%-Ni phase was found to display magnetic ordering below approximately 25K.

A massive block of meteoritic material, weighing over 25 kg, was recovered from a dilapidated museum of the former "Rijkshandelshogeschool, Antwerpen". Subsequent mineralogical, chemical and physical analyses allowed to identify it as a fragment of the Santa Catharina meteorite (SCM). The specimen is to be seen as the fourth heaviest mass of SCM preserved in meteorite collections /1/.

^{57}Fe Mössbauer spectra, both transmission and conversion electron, of the SCM have been reported before /2-9/. The transmission spectrum at room temperature (RT) could be decomposed /9/ into a central unsplitted line, a narrow sextet S_1 with hyperfine field H_{hf} = 290 kOe, and a broader sextet S_2 with H_{hf} = 305 kOe. These components are attributed respectively to a paramagnetic Fe-Ni alloy with ~ 28 at.% Ni, a crystallographically ordered 50-50 Fe-Ni alloy and a non-ordered 50-50 Fe-Ni alloy. In mineralogical terms, nickel-rich Fe-Ni alloys are called *taenites*. The superstructure in the ordered alloy results in minute tetragonality and the phase has therefore been termed *tetrataenite*. The present contribution concerns a Mössbauer study at variable temperatures in the range 8 - 760 K in order to determine the temperature dependence of the hyperfine interactions of the various Fe-Ni phases present in the SCM.

Spectra of a representative, 50 μm thick slice at some selected temperatures are shown in Fig. 1. The asymmetry in depth and position of the four outer lines of the sextet component indicates the superposition of at least two Zeeman patterns with different hyperfine field and quadrupole shift. Following the interpretation of Scorzelli et al. /9/, the spectra were decomposed into two sextets (also plotted in Fig. 1) and one unsplitted line. The obtained hyperfine parameters at some relevant temperatures are given in Table 1, in which sextet #1 arises from the tetrataenite phase (dotted subspectrum in Fig. 1). The RT data are in fair agreement with those reported by Scorzelli et al. /9/. Note that the line width Γ for the ordered phase is relatively small: a 25 μm thick Fe foil gives $\Gamma = 0.31$ mm/s on the spectrometer used. A high degree of ordering may therefore be concluded.

The temperature variation of some hyperfine parameters are represented in Fig. 2. The full lines in Fig. 2a are the theoretical curves calculated using the Debye approximation of the lattice vibrations for the evaluation of the second-order Doppler shift /10/. The adjusted values for the characteristic Mössbauer temperature were 445 K and 570 K for the 50-50 Fe-Ni phases (both have equal δ within the experimental errors) and the 28%-Ni phase respec-

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Table 1
Mössbauer parameters at some selected temperatures for the Santha Catharina meteorite obtained from fitting two symmetric Lorentzian-shaped sextets and one unsplitted line.

	Sextet S1					Sextet S2					Unsplitted		
T	H_{hf}	δ^{Fe}	$2\varepsilon_Q$	Γ	RA	H_{hf}	δ^{Fe}	$2\varepsilon_Q$	Γ	RA	δ^{Fe}	Γ	RA
(K)	(kOe)	(mm/s)	(mm/s)	(mm/s)	(%)	(kOe)	(mm/s)	(mm/s)	(mm/s)	(%)	(mm/s)	(mm/s)	(%)
14	296.2	0.160	0.39	0.38	32	323	0.15	0.00	1.00	16	0.029	0.81	52
80	294.7	0.153	0.38	0.33	30	314	0.15	0.09	0.81	25	0.026	0.50	45
180	292.5	0.108	0.38	0.35	32	310	0.12	0.05	0.73	25	-0.015	0.43	43
300	286.6	0.036	0.39	0.31	30	298	0.05	0.11	0.52	24	-0.087	0.41	46
420	279.8	-0.042	0.38	0.34	33	290	-0.03	0.01	0.75	22	-0.153	0.41	45
520	271.5	-0.104	0.37	0.33	35	276	-0.10	0.07	0.89	21	-0.216	0.42	45
640	256.0	-0.206	0.38	0.30	27	257	-0.21	0.00	0.88	27	-0.298	0.40	46
720	236.4	-0.273	0.25	0.58	49	--	--	--	--	--	-0.343	0.38	51
764	--	--	--	--	--	--	--	--	--	--	-0.340	0.39	100
err.	(0.5)	(0.005)	(0.01)	(0.01)	(4)	(2.0)	(0.02)	(0.06)	(0.06)	(4)	(0.005)	(0.01)	(2)

Indicated errors are about three times the average computed standard deviations

tively. The reduced hyperfine fields both follow a $T^{3/2}$ law for T below ~340 K and indicate a stronger magnetic exchange interaction in the ordered alloy as compared to the non-ordered phase.

In contrast to the claims of some of the earlier cited authors, the spectrum of the 28%-Ni phase does change significantly at low temperatures. Below ~ 25K, the absorption lines of all three components drastically broaden. For the unsplitted line at 8K Γ is near 1mm/s. This can be explained by the existence of antiferromagnetic ordering at low temperatures, resulting in a small magnetic hyperfine field of ~15 kOe (8K) as estimated from the broadening. This suggestion is consistent with the results of Crowell and Walker /11/ who found for small particles of γ-phase Fe-Ni alloy with 28 at.% Ni, a transition temperature T_N of 30 K and a saturation field of 27(5) kOe. This T_N value is in good agreement with the T_N versus at%Ni curve reported by Rancourt et al. /12/ for Fe-Ni Invar alloys. The line broadening at low T for the two sextets is attributed to H_{hf} distributions, probably induced by varying magnetic interactions between the intimately intergrown domains of the taenite phases. This intimate intergrowth is also suggested to be the reason why the 28%-Ni phase does not transform martensitically to the α-phase /13/. Another explanation for the absence of this transformation could be that the domains have submicron dimensions, a feature which is known to suppress the martensitic transformation for synthetic iron-rich Fe-Ni alloys /14/.

The components S_1 and S_2 could no longer be resolved from the 720 K spectrum. Instead, one broad sextet, with quadrupole shift in between that expected for S_1 and S_2, is left. Simultaneously, the fraction of the 28%-Ni phase increases. These features indicate that the gradual break-down of the tetrataenite long-range order has started and that the magnetic ordering in the structurally unordered phase is collapsing. The latter suggestion is consistent with the data of Bansal et al. who reported a magnetic ordering temperature of 735 K for $Fe_{52}Ni_{48}$ /15/.

The considerable broadening for the second sextet throughout the entire temperature range is a consequence of structural variations of the neighbouring atomic shells of the probe iron nuclei in the unordered phase. This datum led us to the approach of analysing the spectra as a superposition of a single line, one sextet (tetrataenite) and a model-independent H_{hf} distribution with a linear correlation between H_{hf} and $2\varepsilon_Q$. At low T, up to ~400 K, the goodness-of-fit decreased by about 10%, but no improvement was obtained at higher T. The

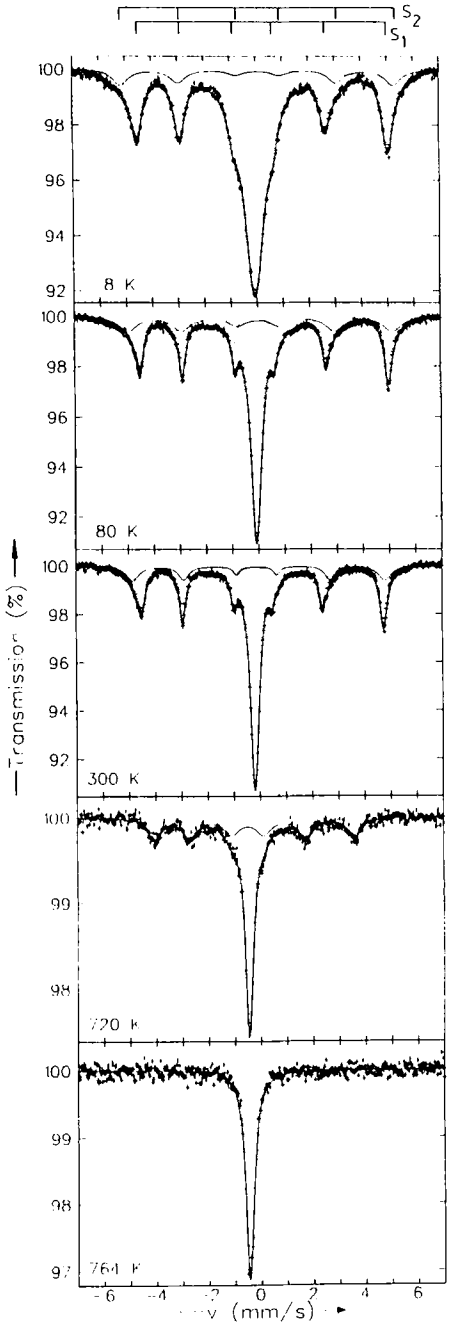

Fig. 1.
Mössbauer spectra at some selected temperatures of a thin slice of Santa Catharina meteorite.

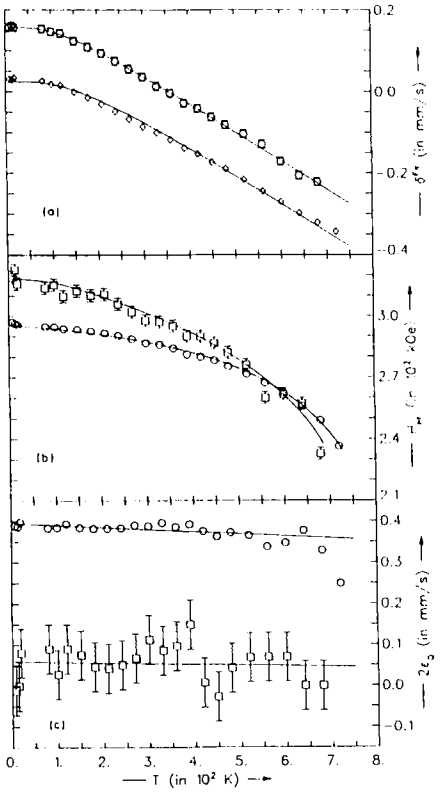

Fig. 2.
Temperature variation of (a) the isomer shifts, (b) the hyperfine fields and (c) the quadrupole shifts of the various Fe-Ni alloys of the Santa Catharina meteorite. (o) ordered 50-50 Fe-Ni, (□) disordered 50-50 Fe-Ni and (◇) 72-28 Fe-Ni. The full lines in (a) are calculated curves (see text); in (b) they serve as a guide for the eye.

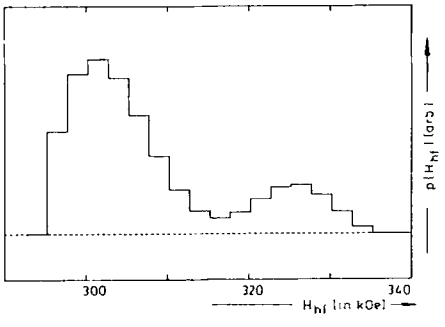

Fig. 3.
Distribution profile for the magnetic hyperfine field of the unordered 50-50 Fe-Ni phase at 80K.

parameters for the tetrataenite and the 28%-Ni phase were all in line with the values derived from the previous fitting approach. The coefficient of the $2\varepsilon_Q$-H_{hf} correlation was found to be scattered in the range 0.005-0.01 mm/(s.kOe). An example of an evaluated H_{hf} distribution, $p(H_{hf})$, is shown in Fig. 3 (T = 80 K). The bimodal structure was observed for all spectra and the relative positions and heights of the two maxima were found to change insignificantly with T. At RT, the maxima are at 293(1) kOe and at 311(1) kOe, with $2\varepsilon_Q$ of 0.17 mm/s and -0.02 mm/s respectively. The latter field value and the corresponding qaudrupole shift exactly coincide with those of Larsen's completely disordered sample of Cape York meteorite /4/. The first maximum would then correspond to a phase with an intermediate long-range order parameter.

In conclusion, the present study has shown that the Mössbauer spectra of the Santa Catharina meteorite at all temperatures up to ~700 K are consistently interpreted on the basis of the suggestion of Scorzelli et al. /9/. However, a better fit is in most cases obtained when the spectral component arising from the disordered 50-50 Fe-Ni phase is considered as a field distribution with correlated quadrupole shift. The bimodal structure of the evaluated probability profiles is a strong indication that this phase consists of a completely disordered phase and one with an intermediate long-range ordering parameter. The unsplitted spectrum from the 28%-Ni phase, previously claimed to remain unchanged (except for the isomer shift) down to 4.2K, was found to display magnetic ordering at temperatures below ~25 K.

ACKNOWLEDGEMENT

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