KFKI-1981-03

KHALID AL ANI
I. DÉZSI
ZS. KAJCSOS
A. BALOGH
B. MOLNÁR
G. BRAUER

# POSITRON ANNIHILATION STUDY OF $Fe_{1-x}$-$B_x$ CRYSTALLINE BINARY SYSTEM

*Hungarian Academy of Sciences*

**CENTRAL RESEARCH INSTITUTE FOR PHYSICS**

**BUDAPEST**

KFKI-1981-03

# POSITRON ANNIHILATION STUDY OF $Fe_{1-x}$-$B_x$ CRYSTALLINE BINARY SYSTEM

Khalid Al Ani, I. Dézsi, Zs. Kajcsos, A. Balogh, B. Molnár

Central Research Institute for Physics
H-1525 Budapest 114, P.O.B. 49, Hungary

G. Brauer

Zentralinstitut für Kernforschung Rossendorf
DDR-8051, Dresden, Pf. 19, GDR

HU ISSN 0368 5330
*ISBN 963 371 779 5*

## ABSTRACT

The $Fe_{1-x}-B_x$ system with X=0, 0.025, 0.099, 0.150, 0.185, 3.307, 0.333, 0.500 and 1.00 was studied by measuring positron angular correlation and Doppler broadening of annihilation γ-line energy. The dependence of the h parameter indicates a tendency for a preferred annihilation of positron at a B atomic site in the concentration range $x \leq 0.333$.

The S parameter obtained from the energy distribution of the annihilation line gives the same conclusion as does the dependence of the h parameter.



## АННОТАЦИЯ

Изучалась система $Fe_{1-x}-B$ при x = 0; 0,025; 0,099; 0,150; 0,185; 0,307; 0,333; 0,500 и 1,00 с помощью методов угловой корреляции 2γ и измерения расширения Допплера γ-линии аннигиляции. Изменение параметра h в области концентрации $x \leq 0{,}333$ указывает на повышенную склонность позитронов к аннигиляции на местах атомов B.

Параметр S, полученный из распределения энергии аннигиляционных линий, позволяет сделать аналогичный вывод, что и изменение параметра h.



## KIVONAT

A $Fe_{1-x}-B_x$ rendszert X=0, 0.025, 0.099, 0.150, 0.185, 0.307, 0.333, 0.500 és 1.00 esetén vizsgáltuk 2 γ-szögkorreláció és az annihilációs γ-vonalak Doppler-kiszélesedése mérésének segitségével. A h-paraméter változása az $x \leq 0.333$ koncentráció-tartományban a pozitronoknak a B-atomhelyeken történő annihilációra való fokozott hajlamát mutatja.

Az annihilációs vonalak energiaeloszlásából nyert S-paraméter ugyanarra a következtetésre vezet, mint a h-paraméter változása.

## 1. Introduction

A positron in binary systems will generally not annihilate with electrons of each sort of atom in proportion to the concentration [1,2].

The positron spatial distribution is affected to a greater or lesser extent by the potential difference between the two kinds of atoms and hence the positron may be attracted more to one kind of atom and then annihilate preferentially there. Lock and West [3] developed a simple theory using the h parameter of the angular distribution curve to study the positron relative affinity in disordered binary alloys.

Since the S parameter obtained from the Doppler broadening of the annihilation $\gamma$-line gives similar information to the h parameter, it seemed resonable to perform such parallel studies.

The Fe-B system has already been studied in the amorphous form using positron annihilation but no results for crystalline samples were given there [4]. Polycrystalline samples were chosen for the present studies to avoid the anisotropic annihilation effects in single crystal samples.

## 2. Experimental

Three experimental methods were used to obtain the presented data: the angular correlation of the 2 $\gamma$-annihilation photon, that of the positron lifetime, and the investigation of the Doppler-broadening of the annihilation $\gamma$-energy.

The experimental data of the 2$\gamma$-angular correlation of the annihilation radiation were obtained using longslit geometry with 140 mm long NaI(Tl) detectors. The detector-sample distance was 220 cm. Additional collimators /2 mm width/ were used in front of the detectors to define the angular resolution of the instrument. The positron radioactive source was 16 mCi $^{22}$Na. The coincidence resolution time was 200 nsec, resulting in a peak-to-background ratio of 0.4-0.5 %. The angular correlation curve scanning and the data collection were performed automatically by using a special program in a CAMAC system. All measurements were done in a vacuum of $10^{-5}$ torr to prevent the annihilation of positrons in air.

For lifetime measurements a fast-slow coincidence unit was used with Philips XP-1023 photomultipliers and NE 111 fast plastic scintillators. The lifetime spectra were stored in a 4096 channel ICA-70 analyser. A few $\mu$Ci of $^{22}$Na was used as a positron source. Such a source was prepared by evaporating a neutralized aqueous solution of carrier-free $^{22}$NaCl onto a thin /0.5-0.9 mg.cm$^{-2}$/ foil of Al. The deposited active spot was covered with the same foil. The time resolution of the instrument measured for $^{60}$Co source was 300 psec. The measured spectra were analysed by least-squares fitting using the POSITRONFIT EXTENDED computer program [5].

In the Doppler broadening method, the energy distribution measurements were performed by using a high resolution ORTEC 1000 hyperpure germanium low energy photon spectrometer detector + preamplifier connected with a vertical cryostat for cooling. The active volume was 1 cm$^3$. The detector was followed by a main amplifier and a biased amplifier /ORTEC 716 A and 444 Models, respectively/. The spectra were recorded by a 4096 Multichannel Analyser. With this system a resolution of 1.064 $\pm$ 0.004 keV was achieved for the 514 keV $\gamma$-line of $^{85}$Sr.

Also the 569.69 keV $\gamma$-line of $^{207}$Bi, was used for energy calibration; the calibration factor was 54.3 eV/channel. For the positron source, about 3 $\mu$Ci of $^{22}$Na evaporated on 6 $\mu$m Hostaphan foil was used.

To avoid electronic drift it was necessary to collect the data in such a way that every sample was measured 12 times in 80 minutes /i.e. 400 sec one time/ then the data were summed up by computer program. /1-1.6/ x $10^6$ events were collected for every sample with a 0.2 peak-to-background ratio.

The samples were prepared as follows. Alfa Research amorphous boron of high purity was mixed with high purity iron in powder form. The mixture was pressed by of 10 ton /$cm^2$ and was then melted and homogenized in a pure $Al_2O_3$ crucible under vacuum in a high-frequency induction oven. The contentration of the components was determined by using atomic absorption spectrometry. The samples were cut to proper sizes then polished and annealed again in vacuum. Annealing was done at 200 $^oC$ below the melting temperature for 1/2 - 1 day then samples were cooled slowly to room temperature.

3. Results

3.1 Measurements of the angular correlation

The angular distribution curves are shown in Fig. 1. The measured curves showed an asymmetry less than 1 % between positive and negative positions of the moving detector. The measured spectra were corrected for background then normalized to the same area and fitted assuming a low momentum component /resulting in a parabolic shape/ and a high momentum one /resulting in a Gaussian distribution/; for iron the data analysis agrees well with the literature [6]. The pure boron sample was in the form of pressed powder. The best fit was obtained by assuming one parabolic and one Gaussian. The first parabola has an intensity of /4.5 % ± 0.8/, its cut off at 2.7 ± 0.35 mrad. The width of this component suggests the existence of positronium in the sample and it was subtracted from the angular distribution curve. The second parabolic may correspond to annihilations on valence electrons.

The h parameter /the peak per total area/ increased with increasing concentration of boron and is shown in Fig. 2a. The low momentum component which can be related to the relative probability of the annihilation rate on the conduction electrons is shown in Fig. 2b., while Fig. 2c. shows the Fermi cut-off.

### 3.2 Lifetime measurements

The lifetime parameters necessary for the calculation of the h and s parameters are given in the following sections.

The observed lifetime results for the Fe 15 % B and the pure boron are given in Table 1.

Table 1. Positron lifetime values

| Sample | $\tau_1$ psec | $\tau_2$ psec | $I_2$ % |
|---|---|---|---|
| Fe-15 % B | 122 $\pm$ 3 | 445 $\pm$ 14 | 7.43 |
| B | 229 $\pm$ 5 | 907 $\pm$ 13 | 13.49 |

The iron lifetime value /110 psec/ was obtained from the literature [6]. For the pure boron the measured lifetime value is in very good agreement with the value of 229 psec [7].

The second component $\tau_2$ is attributed to annihilation of positron on the sample surfaces and gain boundaries and positronium annihilation.

### 3.3 Doppler broadening measurements

The energy distribution of the annihilation $\gamma$-line was measured for various samples. In addition, a solid polycrystalline boron sample was also measured. One of the measured energy distribution annihilation $\gamma$-lines is shown Fig. 3.

As can be seen in this figure, the measured line-shapes show an asymmetry around the peak position favouring the low-energy tail. This effect is the consequence of photoelectron escape. A line-shape correction regarding the asymmetry was made according to the literature [8] by taking a straight-line background between estimated "end points" of the peak then from each channel a fraction of the value equal to the ratio between the peak intensity at that channel and the total peak area was subracted.

On the basis of the energy distribution the contribution of the conduction electron /central part/ and the core electrons /wing parts/ was chosen as shown in Fig. 3.

The S parameter, i.e. the sum of counts in a central part of the peak relative to the total area of the peak, may reflect the fractional contribution of different electronic states in the annihilation process. Therefore, after background and line-shape correction this parameter was calculated for all samples /see Fig. 4a/. In Fig. 4b and 4c the core contribution and the ratio between the contribution of conduction electrons to the core ones are shown respectively.

As seen in these figures, the difference in the S parameter between the pressed powder and the solid boron samples supports the conclusion of the angular correlation results.

## 4. Discussion

Lock and West suggest a theoretical model taking into account the angular distribution h parameter in long slit geometry [9] for a polycristalline sample and assuming that the thermalized positron annihilates from a ground state Bloch wave function. They described the h parameter of the positron annihilation in binary alloys by the equation

$$h_{AB}=h_A\lambda_A+c_B\eta_B(h_B\lambda_B-h_A\lambda_A)/\lambda_A+c_B\eta_B(\lambda_B-\lambda_A)$$

where $h_i$ , $\lambda_i$ and $c_i$ represent the h parameter, the positron annihilation rate and the atomic concentration of the pure components, respectively; $\eta$ is a factor describing the positron relative affinity between the two types of cells in the alloys.

Regarding the theoretical model, the $\eta$ factors are undetermined parameters of the order of unity, but one has to consider their effect on the measurable results by comparing the measured $h_{AB}$ as a function of the concentration with the calculated values according to the above equation.

As seen in Fig. 2a, the tendency of the h parameter may be attributed to different positron relative affinity for the two kinds of atom. In the iron-rich region, the behaviour of the h parameter may be explained by a mixture of phases such as $\alpha$-Fe and $Fe_2B$ present in the samples after the heat treatment. If every h is replaced by S with respect to the experimental results the same tendency is seen as indicated by the h parameter /see Fig. 4a/. In view of this, it is clearly a complementary method for the study of the relative positron affinity in binary alloys.

The tendency of the h and the S parameters to exhibit stepwise behaviour is attributed to the annihilation of positron in different crystal sturctures of intermetallic compounds existing in the Fe-B alloys such as $Fe_2B$ and FeB.

The transition metals are characterized by the presence of "holes" in their d-bands, though upon alloying the boron atom may donate electrons to the d-bands [10-12] thus, the position of the Fermi level may rise. This effect can be seen clearly in Fig. 2c where the Fermi cut-off increases as a function of boron content.

In the transition metal-rich regions, the boron atoms are isolated from each other whereas in monoborides they are arranged along one dimensional zig-zag chains. As a conclusion, the tendency of the h and S parameters may suggest, in addition, that the structure influences the positron's relative affinity in the Fe-B crystalline alloy.

References

[1] P. Hantojärvi: Positrons in solids, Springer, Berlin /1979/

[2] M.J. Stott, A.T. Stewart and P. Kubica: App . Phys. 4, 213 /1974/

[3] D.G. Lock and R.N. West: J. Phys. F: Metal. Phys. 4, 2179 /1974/

[4] Proceeding of the Fifth International Conference on Positron Annihilation, April 8-11, 1979; Lake Yamanaka, Japan

[5] P. Kirkegaard and M. Eldrup: Compt. Phys. Commun. 3, 240 /1972/; Compt. Phys. Commun. 7, 401 /1974/

[6] D.O. Welch and K.G. Lynn: Phys. Stat. Sol /b/ 77, 277 /1976/

[7] R.M. Singru, K.B. Lai and S.J. Tao: Positron annihilation data tables - Atomic data and nuclear data tables 17, 272 /1976/

[8] K. Fransson, A. Nilsson, J.D. Raedt and K.B. Rensfelt: Nucl. Instr. Meth. 138, 479 /1970/

[9] D.C. Connors, V.H.C. Crisp and R.N. West: J. Phys. F: Metal Phys. 1, 355 /1971/

[10] B.D. Hanson, M. Mahnig and Louis E. Tóth: Z. Naturforsch. 26a, 739 /1971/

[11] Less-common metals /proceedings of the 6th International Symposium on Boron and Borides/, vol:67, N$^{o}$ 1 /1979/

[12] V.I. Matkovich: Boron and refractory borides, Springer, Berlin /1977/

Figure 1. Angular distribution curves. The solid line shows the result of the fit.

Figure 2. a/ The h parameter as a function of boron content. The solid line is the calculated one.

b/ Low momentum components as a function of boron content.

c/ Fermi cut off values.

Figure 3. Energy distribution of annihilation $\gamma$-line in pure Fe. c, the central part, represents the conduction electron contribution in the annihilation process; A + B represents the core contribution.

Figure 4. a/ The S parameter obtained from Doppler broadening measurements as a function of boron content. The solid line is the one calculated by substituting h with S.

b/ Core contribution

c/ Ratio of conduction to core electrons

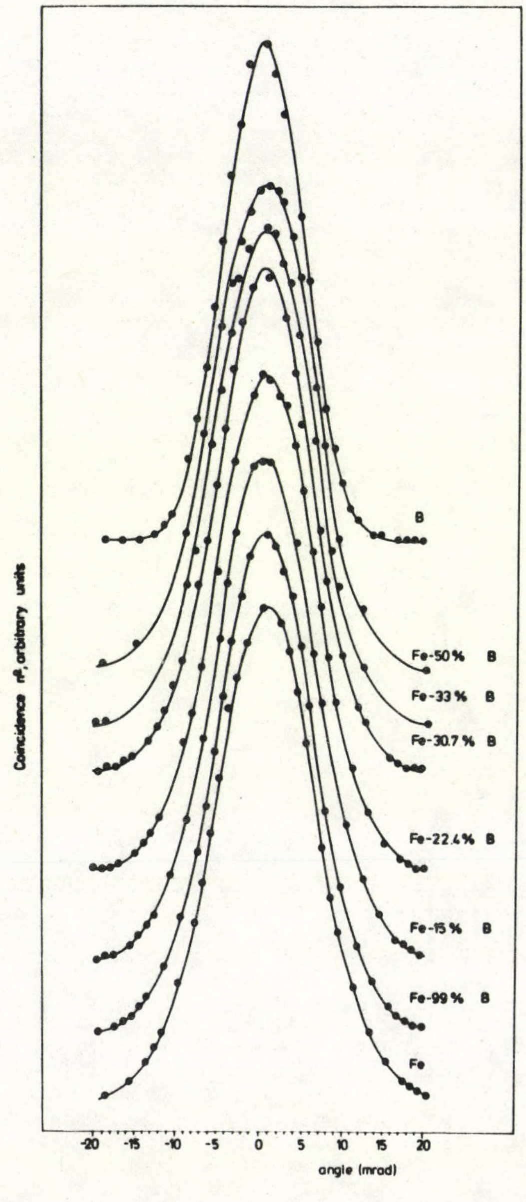

B
Fe-50% B
Fe-33% B
Fe-30.7% B
Fe-22.4% B
Fe-15% B
Fe-99% B
Fe
Coincidence nº, arbitrary units
-20 -15 -10 -5 0 5 10 15 20
angle (mrad)

Fig. 1

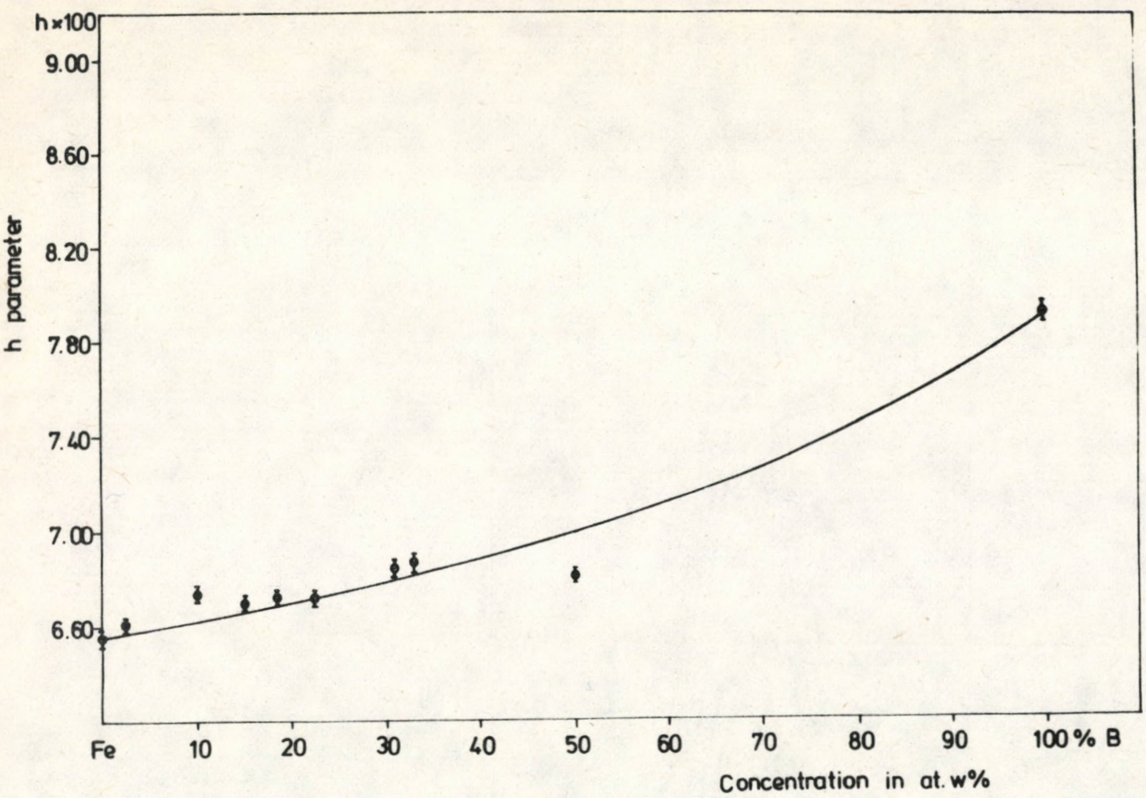

h×100
9.00
8.60
8.20
7.80
7.40
7.00
6.60
h parameter
Fe
10
20
30
40
50
60
70
80
90
100% B
Concentration in at. w%

Fig. 2a

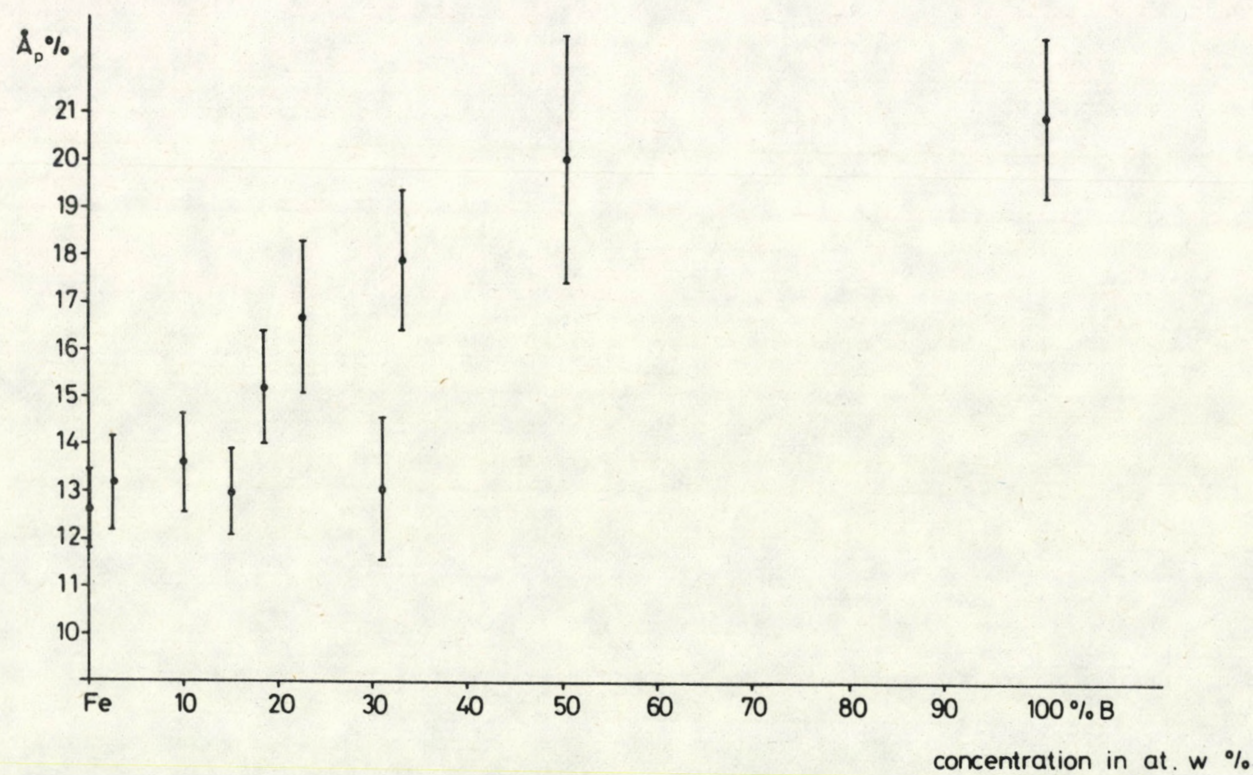
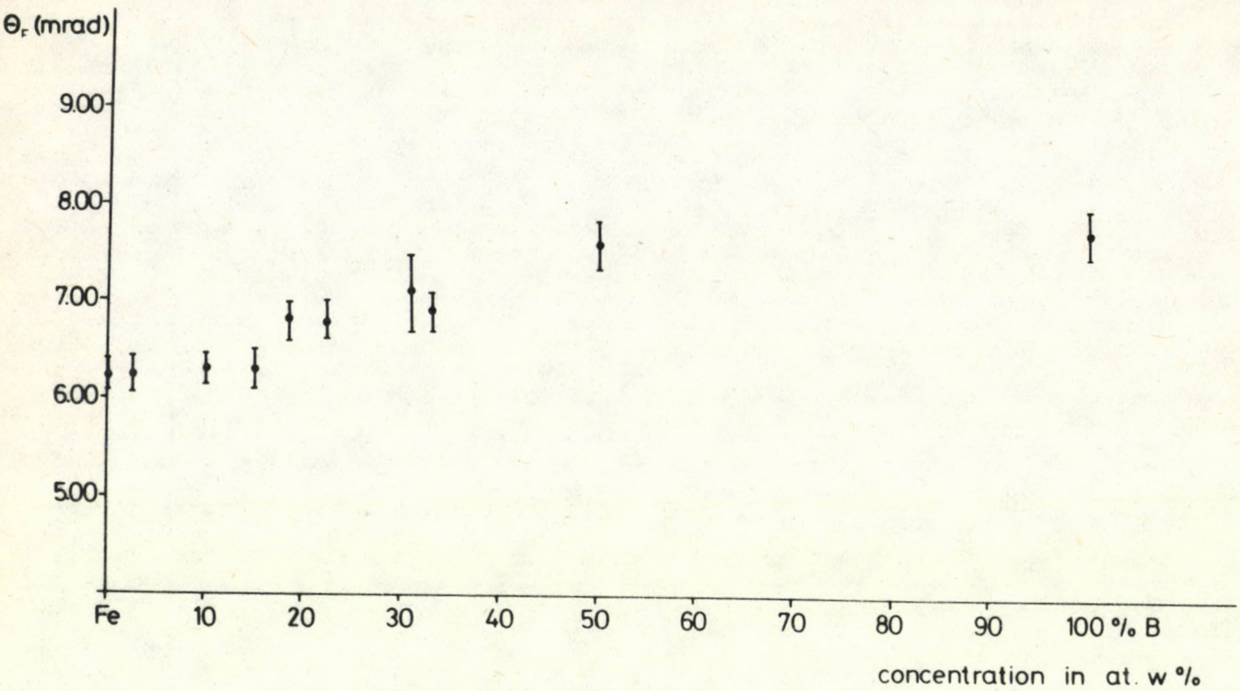
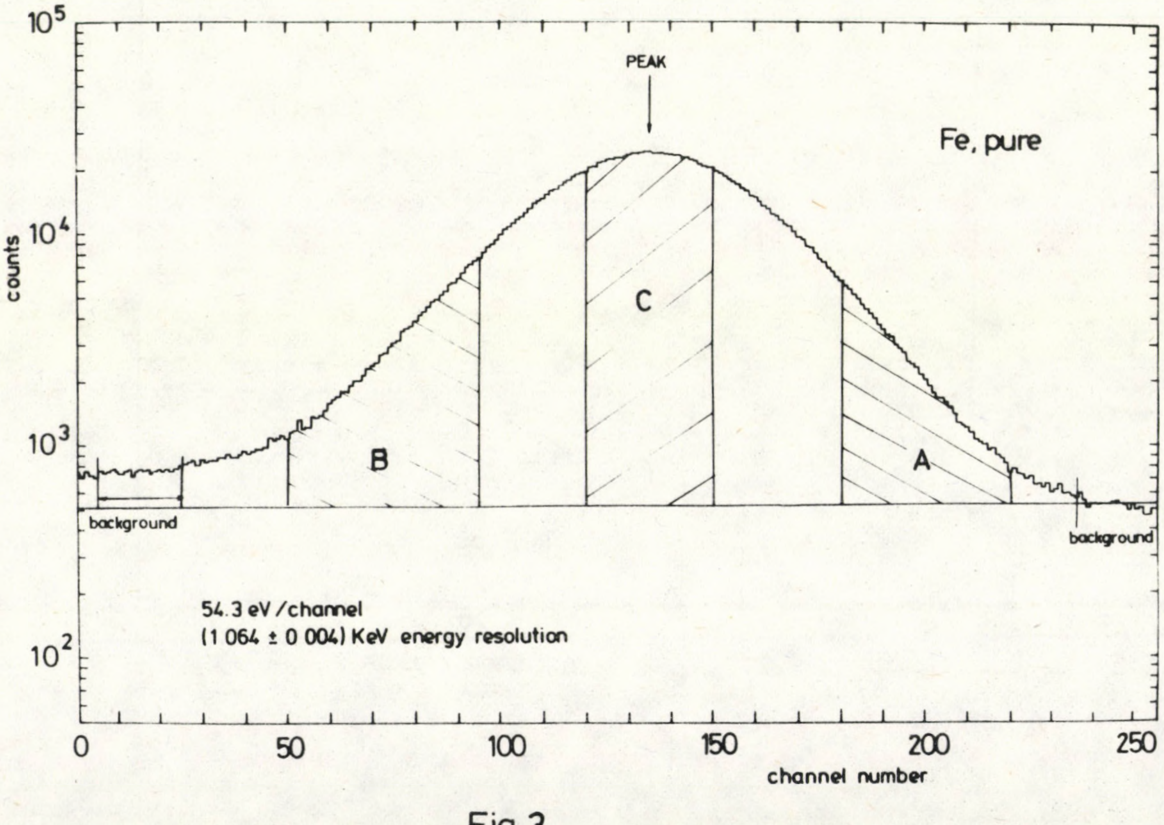

Å_p %
21
20
19
18
17
16
15
14
13
12
11
10
Fe
10
20
30
40
50
60
70
80
90
100 % B
concentration in at. w %

Fig. 2b



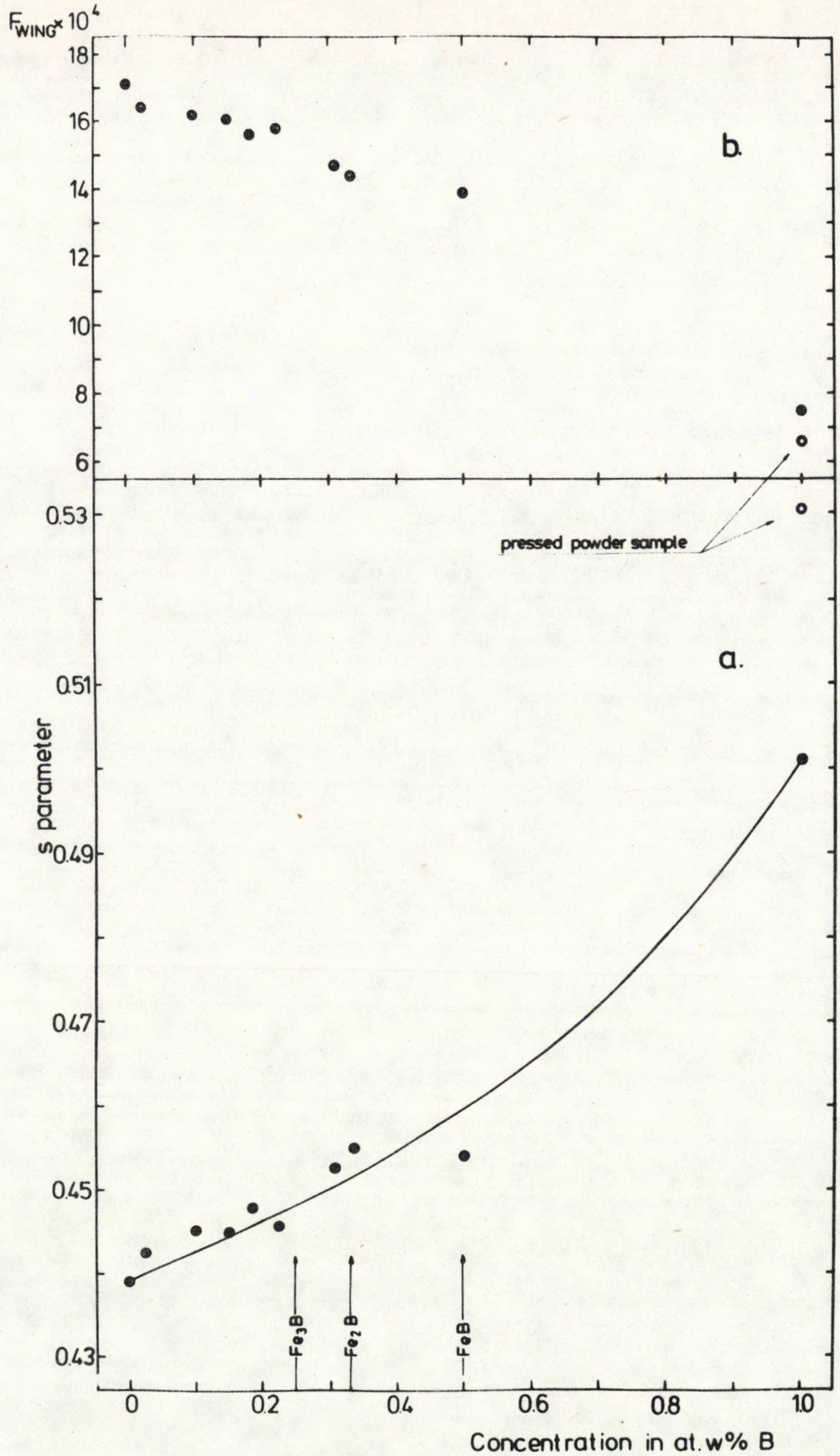
Θ_F (mrad)
9.00
8.00
7.00
6.00
5.00
Fe
10
20
30
40
50
60
70
80
90
100 % B
concentration in at. w %


Fig. 2c



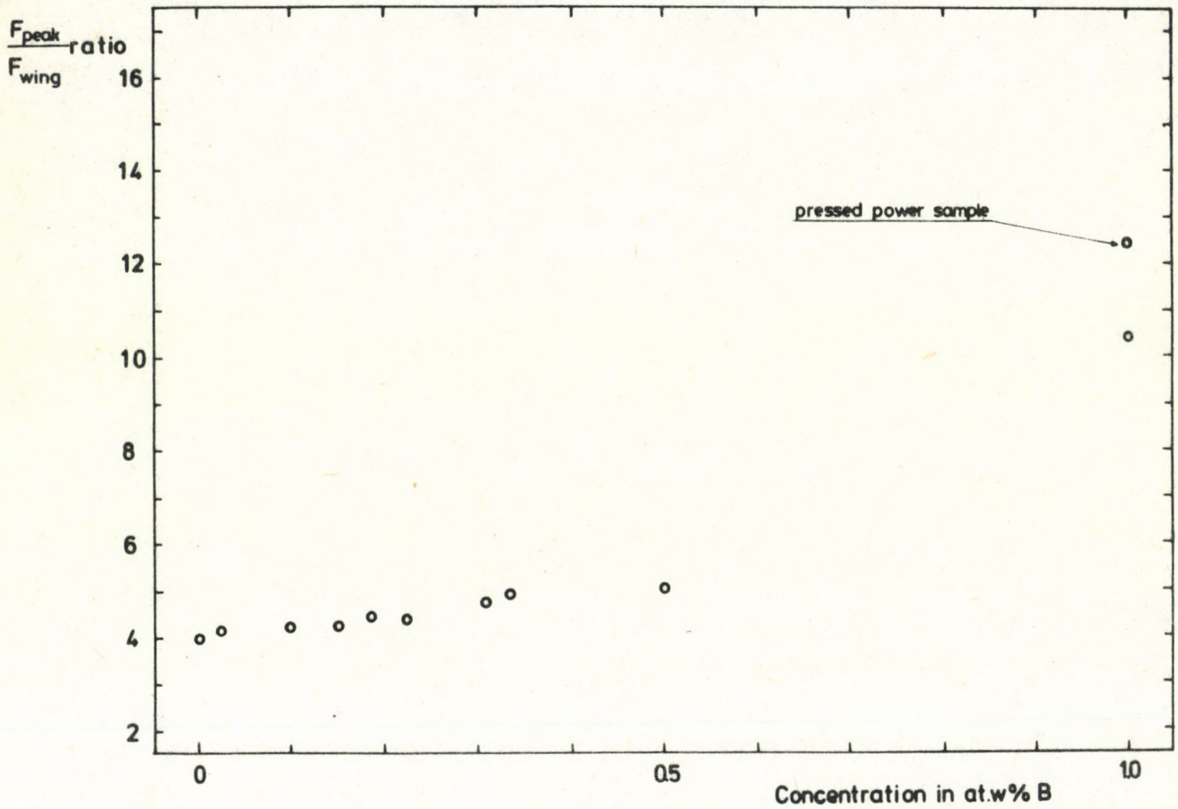
PEAK
Fe, pure
counts
C
B
A
background
background
54.3 eV/channel
(1.064 ± 0.004) KeV energy resolution
channel number
0
50
100
150
200
250


Fig. 3



F WING × 10^4
18
16
14
12
10
8
6
b.
0.53
0.51
0.49
0.47
0.45
0.43
s parameter
pressed powder sample
a.
Fe3B
Fe2B
FeB
0
0.2
0.4
0.6
0.8
1.0
Concentration in at.w% B


Fig. 4



F peak / F wing ratio
16
14
12
10
8
6
4
2
pressed power sample
0
0.5
1.0
Concentration in at.w% B


Fig. 4c

Kiadja a Központi Fizikai Kutató Intézet
Felelős kiadó: Szegő Károly
Szakmai lektor: Nagy Dénes Lajos
Nyelvi lektor: Harvey Shenker
Példányszám: 750 Törzsszám: 81-13
Készült a KFKI sokszorosító üzemében
Felelős vezető: Nagy Károly
Budapest, 1981. január hó