ARRHENIUS LAW PARAMETERS FOR CARBON SNOEK RELAXATION IN ALPHA IRON

By L. Kozłowski

Institute of Metallurgy, Academy of Mining and Metallurgy, Cracow,

J. W. Moroń, J. Przybyła and J. Rasek

Institute of Physics, Silesian University, Katowice*

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By utilizing the measurement of permeability disaccommodation curves the parameters of Arrhenius' law for the carbon Snoek relaxation in alpha iron were determined in the case of two series of samples: 1. samples made from pure and very pure iron, and 2. samples made from very pure iron containing an addition of 0.116% Al.

1. *Introduction*

For many years the activation energy and pre-exponential factor in Arrhenius' law for the carbon Snoek relaxation in αFe—C have been assumed to be equal to the values obtained by Wert, namely, $Q = 20100$ cal/mol and $D_0 = 0.02$ cm^2/sec [1]. Apart from data received from measurements of anelastic phenomena and other low-temperature data, Wert made use in his calculations of diffusion coefficients determined at high temperatures. As time went by, however, it became obvious that results obtained by after-effect techniques and results of diffusion measurements cannot be described by a mutual Arrhenius law; lower values of Q are always obtained at low temperatures [2]. Homan [3] and McLellan and collaborators [4] have made an attempt to explain this phenomenon.

In papers [4] and [5] the results of many authors, obtained at low temperatures, primarily be mechanical techniques, were utilized to give a new determination of Q and D_0. The obtained values, arranged in table, are much lower than all previously published ones, Q always being nearly equal to or greater than 20000 cal/mol and $D_0 \geqslant 0.01$ cm^2/sec. Also in a recent study [6] the value $Q = 19400$ cal/mol was assumed.

In the research mentioned above the method of magnetic after-effects did not play any greater role. In contradistinction to anelastic measurements, the analysis of magnetic

* Address: Instytut Fizyki, Uniwersytet Śląski, Katowice, Bankowa 12, Poland.

measurements required the introduction of a continuous spectrum of relaxation times [7–9], and in the most extensive studies an enormous scatter of activation energies corresponding to different parts of the spectrum was found (for the shortest relaxation time 11300 cal/mol, for the longest — 24300 cal/mol, and for intermediate time — 17200 cal/mol [9]).

The studies [10] and [11] dealt with very pure iron containing carbon and nitrogen in the form of a solid solution. It was shown that in the case of weak magnetizing fields the permeability disaccommodation and temperature dependence of the tangent of the magnetic loss angle for this material can be described by means of formulae derived on the basis of Néel's theory for a single relaxation time, identical with the relaxation time of mechanical after-effects. Employing these results, the phenomenon of permeability disaccommodation was examined in several samples of pure iron and iron containing about 0.1% Al, this phenomenon arising due to the carbon Snoek relaxation.

2. *Samples*

Investigations involved several samples of pure and very pure iron (99.9 or 99.99% Fe without considering the impurities forming interstitial solutions), described in detail in [10], one sample manufactured from Johnson and Matthey iron (99.999% Fe) and two samples of iron containing 0.116% Al in solid solution form. The last two samples were prepared at the Department of Physical Chemistry of the Institute of Iron Metallurgy in Gliwice from electrolytic iron of their own manufacture (99.99% Fe) and an iron-aluminum alloy. The samples of pure iron contained up to 0.02% of carbon and less than 0.01% of nitrogen, whereas the Fe-Al (0.1%) samples had 0.02% C, 0.013% N and traces of Al_2O_3. Some of the samples were saturated with carbon in an atmosphere produced by the decomposition of isohexane.

The samples were in the shape of toroids formed by rolling up tape from 0.10 to 0.15 mm thick and about 10 mm wide; the inner diameter was 30 mm, the outer 40 mm.

The samples of pure iron were stress relieved by annealing in vacuum at approx. 700°C for several hours, after which they were cooled with the furnace (in the range from 700°C to 300°C the mean cooling rate was about 1°C/min).

The Fe-Al (0.1%–C samples were annealed in a vacuum furnace at 950°C for three hours and then cooled with the furnace to 700°C and kept at this temperature for the hours, after which they were cooled with the furnace to room temperature. As is known from the literature [12, 13], this kind of heat treatment causes the nitrogen in the iron to become chemically bounded with aluminum, giving AlN, and thereby this addition does not participate in the after-effects.

About 400 windings of 0.5 mm diameter coil wire were wound on the samples.

3. *Experimental procedure*

The measurements of permeability disaccommodation were made with a Maxwell type bridge of a design like that described in Ref. [14]. After the temperature became settled, the sample was demagnetized within several seconds by a 50 Hz alternating field of maximum strength of several oersteds with the use of the device described in Ref. [15]. The bridge was balanced in a continuous manner, readings being taken at fixed intervals. Each measurement was repeated several times, and the results were averaged.

The obtained disaccommodation curves, in agreement with the results of the paper [11], were broken up into two or three elementary processes which comply with single time constants by the method given in Ref. [16].

4. *Results*

In the case of the pure iron samples a relaxation was obtained apart from the carbon Snoek relaxation which had a much lower intensity and a time constant Θ five times longer. At the highest measuring temperature there was observed a very slow decrease

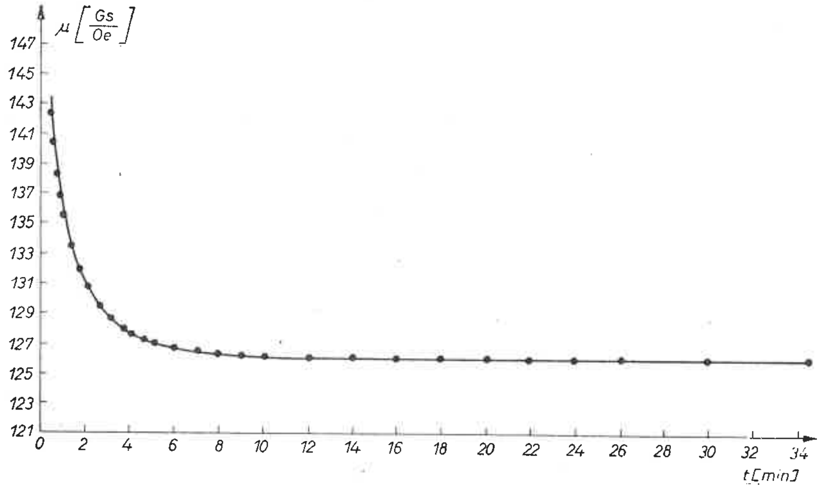

Fig. 1. Changes of magnetic permeability with time in sample of pure iron; temperature $-12.5°C$

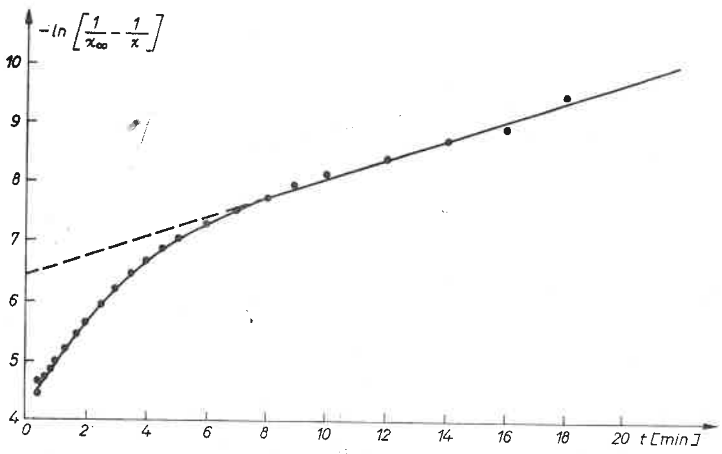

Fig. 2. First analysis of disaccommodation curve of Fig. 1

of permeability with time; this effect had a relaxation time 250 times longer than that of the Snoek effect. The presence of disaccommodation associated with nitrogen Snoek relaxation was noticeable only at the lowest temperatures.

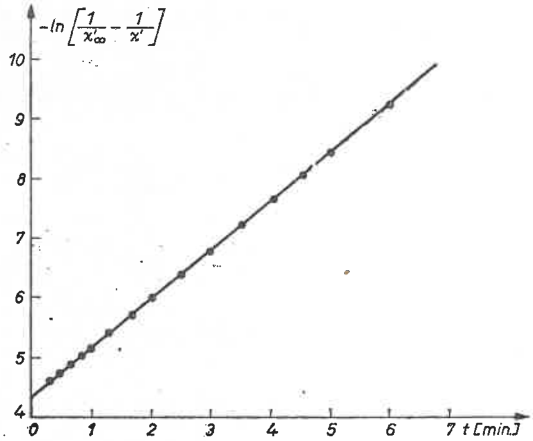

Fig. 3. Second analysis of disaccommodation curve of Fig. 1

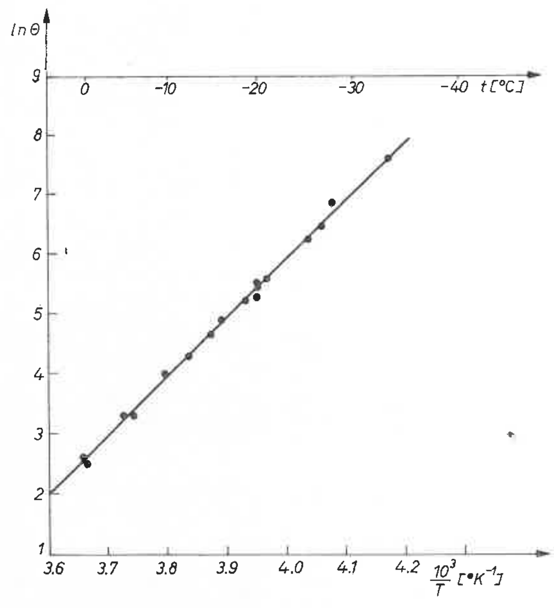

Fig. 4. Graph of $\ln\Theta$ *vs* $1/T$ found for pure iron sample

In the case of Fe-Al/0.1%) —C only the Snoek process was looked into, denoted II in Ref. [13]; in that paper this material was used for a detailed investigation of the disaccommodation effect. The intensity of the drop in permeability with time, linked with this relaxation, was at least five times higher than the intensity of the disaccommodation originating due to other relaxation phenomena, whereas the time constants of the various components differed by five to ten times.

As an example, Fig. 1 presents the disaccommodation curve found for a pure iron sample at $-12.5°C$. Figure 2 and 3 show two subsequent analyses of this curve.

The graphical representation in Figs 4 and 5, in the form of a dependence of $\ln \Theta$ on reciprocal absolute temperature, shows the results obtained for Snoek relaxation in Fe-C

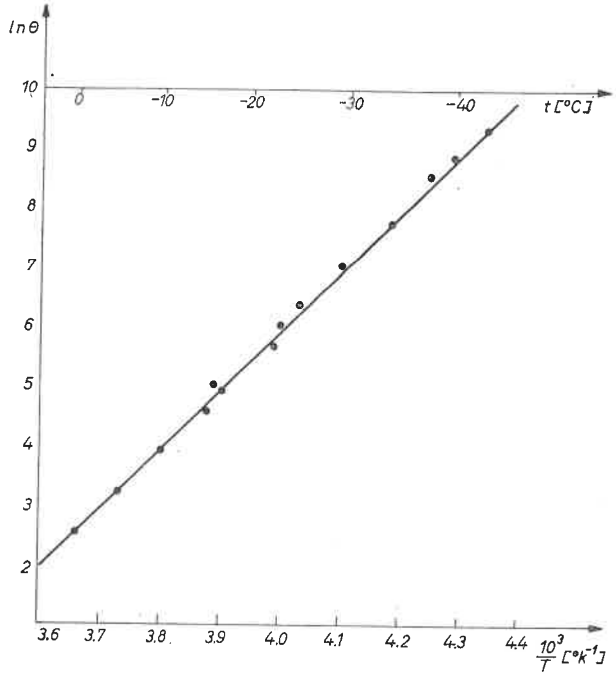

Fig. 5. Graph of $\ln \Theta$ *vs* $1/T$ for carbon Snoek relaxation in Fe-Al-C samples

and in Fe-Al/0.1%/$-$C. It is seen that rectilinear runs are obtained, in accord with Arrhenius' law. Results corresponding to other relaxation effects are described in detail in Ref. [17]. By the method of least squares the following parameters of Arrhenius' law were found:

1) for the iron samples

$$Q = (19500 \pm 220) \text{ cal/mol}$$

$$\Theta_0 = \begin{pmatrix} 3.2 & \begin{matrix} +1.7 \\ +1.1 \end{matrix} \end{pmatrix} \times 10^{-15} \text{ sec}$$

2) for the samples of iron containing 0.1 Al

$$Q = (19510 \pm 270) \text{ cal/mol}$$

$$\Theta_0 = \begin{pmatrix} 3.5 & \begin{matrix} +2.3 \\ -1.5 \end{matrix} \end{pmatrix} \times 10^{-15} \text{ sec.}$$

5. *Discussion of results of measurements*

The Table compares the results of the present work with those of Lord and Beshers [5] and Mc Lellan *et al.* [4].

It is clear that the measurements of permeability disaccommodation give for Q and D_0 values which are close to the results obtained by compilation from the work of many authors, usually employing internal friction technique. It seems, however, that in the paper [5] there is too low a value of activation energy of carbon Snoek relaxation.

TABLE

Comparison of Arrhenius law parameters for carbon Snoek relaxation found recently by various authors

Author	Q [cal/mol]	D_0 [cm²/sek]	Comments
Mc Lellan *et. al.* [4]	19300	0.0033	Compilatory calculations on the basis of many authors' data obtained primarily by internal friction
Lord and Beshers [5]	19160	0.00394	
this work	19500±220	$0.0070^{+0.0025}_{-0.0040}$	Fe samples
	19510±270	$0.0065^{+0.0030}_{-0.0045}$	Fe-Al (0.1%) samples from permeability disaccommodation measurements

Employing the parameters found for Fe-C, the temperature of the peak for the vibrational frequency of 1 Hz was obtained. It is found to be 38.3°C, instead of the 39°C usually quoted in the literature.

The results of the present work are thus a confirmation that the parameters of Arrhenius' law for the carbon Snoek relaxation differ considerably from the values found by Wert now commonly accepted as true [1]. They also imply that the same results are obtained on the basis of the disaccommodation phenomenon as by the method of anelastic effects. It is also proved that an addition of 0.1% of aluminum bears no influence at all on the carbon Snoek relaxation in alpha iron.

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