PERMEABILITY DISACCOMMODATION IN α Fe-C-N IN THE TEMPERATURE RANGE FROM $-40°$ TO $+180°C$

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The disaccommodation of permeability in several samples of pure and very pure iron, containing carbon and nitrogen in the form of a solid solution, was studied in the range of temperatures from $-40°$ to $+180°C$. Apart from time decrease of permeability associated with the carbon and nitrogen Snoek relaxation and with C—C pairs, a new disaccommodation effect was found to appear at temperatures over 0°C. The effect of quenching, ageing and plastic working on this phenomenon was investigated.

1. Introduction

Hydrogen, carbon and nitrogen, and presumably boron and oxygen as well, form interstitial solutions when dissolved in alpha-iron. The atoms of these elements occupy the octahedral sites of the bodycentered cubic lattice, and they can easily jump between adjacent positions of equilibrium. The anisotropic distribution between the three possible types of gaps, associated with the existence of external stress or magnetization, leads to the appearance of the so-called diffusive mechanical and magnetic after effects.

The magnetic after effect is observed in the form of several separate phenomena of which the most important are permeability disaccommodation, the ΔH effect, and the additional losses in measurements with alternating current which arise besides those due to hysteresis and eddy currents. These effects, although associated with the same elementary processes, have different relaxation times. The relaxation times change with temperature T according to Arrhenius' law

$$\theta = \theta_0 \exp \left[\frac{Q}{RT} \right]$$

where Q is the activation energy, θ_0 a pre-exponential factor and R is the gas constant. Relaxation times of 0.5 to 100 minutes correspond to the disaccommodation effect. The ΔH effect, which appears as a lag of the magnetic induction behind field strength, is commonly observed for θ within the interval from 10^{-2} to 10^{3} seconds. The maximum of relaxation losses, at an angular frequency of magnetizing current ω, comes at $\omega\,\theta = 1$. In practice,

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the frequencies used are from 10 to 1000 cps, thus the relaxation times corresponding to the loss peaks are contained in the range from 10^{-2} to 10^{-4} sec.

The pronounced differences in the relaxation times cause the individual effects of the magnetic aftereffect to appear within different temperature ranges. Starting out from the low temperatures, the first to be observed is disaccommodation. The region of this effect partially overlaps the region of the ΔH effect. On the other hand, the peaks of relaxation losses appear at temperatures higher than the upper limit of the ΔH region.

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2. *Magnetis aftereffects in αFe in the temperature range from —40° to +180°C*

Apart from the aftereffects associated with the presence of single interstitial atoms, known as Snoek relaxation, there appears in this temperature range a number of other processes which are due to more complex relaxation centers. Disaccommodation from single carbon and nitrogen atoms is observed below 0°C [1]–[5]. Only Richter [6, 7] studied the ΔH effect of Snoek relaxation; it shows up in the range of temperatures from —11° to +97°C. The maxima of the magnetic relaxation losses of this phenomenon, at frequencies between 33 and 5000 cps, are observed at temperatures from 85° to 160°C [8]–[10].

Disaccommodation due to pairs of carbon atoms had been observed at 0°C during the research described in Ref. [11].

In iron samples, submitted to cold working of the order of 0.1 per cent, additional peaks of losses were observed in the range of temperatures above the Snoek maximum ($f = 33$ cps and $T = 73$°C) [12]; they are presumably associated with the relaxation in αFe—C as revealed in the internal friction data by Rawlings and Robinson [31]. Considerable plastic working (of the order of 20 per cent) leads to the appearance of disaccommodation at approximately 180°C [14]. This effect is linked with the so-called Köster relaxation [15].

3. *Research on disaccommodation in αFe—C—N in the —40° to +180°C range*

As is seen in the review given in Sec. 2, the time decrease of permeability had not been examined hitherto in the range of temperatures from 0° to 180°C. In materials with impurities forming substitutional solutions some disaccommodation processes may appear here due to jumps of individual interstitial atoms, neighbouring with one or more substituting atoms. On the other hand, any time-changes of permeability should not be found in non-deformed samples of pure iron containing only carbon and nitrogen. Nonetheless, losses in αFe—C—N have been detected recently, and they are presumably of relaxation origin, to which there would correspond disaccommodation in the temperature range from 0° to 100°C [16].

3.1. Some remarks on the experiments

The time-changes of permeability were measured on two alternating current bridges: a Maxwell–Wien bridge, described in detail in Ref. [12], and a Wilde bridge, described in Ref. [17]. For obtaining temperatures lower than room temperature a thermostat was used, in which the cooling agent was dry ice in methyl alcohol. Temperatures higher than that of the surroundings were stabilized in a thermostat with air circulation. The temperature was determined by means of a copper-constantan thermocouple. The thermoelectric force was measured with a KM-76 compensator.

The samples were demagnetized by means of Kinel's electronic demagnetization device [18].

A number of specimens produced in two different series were examined. The first series (GR) gave samples produces from carbonyl iron by powder metallurgy; they contained less than 0.1 per cent of impurities forming the substitutional solution, approximately 0.02 per cent of carbon and 0.001 per cent of nitrogen. The samples of the second series (P) were produced from electrolytic iron and contained only 0.01 per cent of substitutional impurities. The carbon content was from 0.01 to 0.02 per cent, and the nitrogen concentration was higher than in the GR series. The samples were in the form of toroids made by looping strip 0.1 mm thick and 10 to 15 mm wide. The examined material did not show any texture.

Measurement were performed on stres-relieved and quenched samples. In the first case, the specimens were relieved of stress under a dynamical vacuum (10^{-4} mm Hg) for several hours, and then cooled slowly with the furnace (in the 700° to 300°C range the cooling rate averaged 1°C/min). In the second case, the samples were sealed in evacuated quartz tubes which, after relief annealing, were thrown into a water and ice bath and shattered. Four hundred windings of 0.5 mm coil wire were wound around each sample.

After the temperature became fixed the specimens were demagnetized by a 50-cps alternating field, whose initial amplitude was several oersted. The demangetization time was five seconds. Next, the bridge was brought to equilibrium in a continuous manner and after 0.5 and 15 minutes from initiation of demagnetization its indications were read. The results of the measurements are shown in the form of the dependence $\left(\frac{\Delta\mu}{\mu_p}\right) \cdot 100\% = f(T)$, where $\Delta\mu$ is the difference between the permeability after 0.5 and 15 minutes from the instant of demagnetization, and μ_p is the permeability after 0.5 minute. When the curves of the temperature-dependence of the magnetic losses were determined, the procedure used was like that given in Ref. [10].

3.2. Results of measurements

3.2.1. Stress-relieved samples

Figure 1 presents an example of $\frac{\Delta\mu}{\mu_p}$ as a function of temperature obtained for the GRII-2 sample (0.02% C and 0.001% N) after the following stress-relieving treatment: annealing at 900°C for 18 hours and at 700°C for one hour, and then cooling with the furnace. It is seen that apart from the large disaccommodation of the order of 35 per cent appearing below 0°C (the region of carbon and nitrogen Snoek relaxation) there is a weak effect above room temperature. Measurements repeated many times have shown that the $\frac{\Delta\mu}{\mu_p} = f(T)$ curve is not monotonic, but has distinct local extrema. Figure 2 gives the results from the region between 60° and 140°C for the same sample, after ageing at room temperature.

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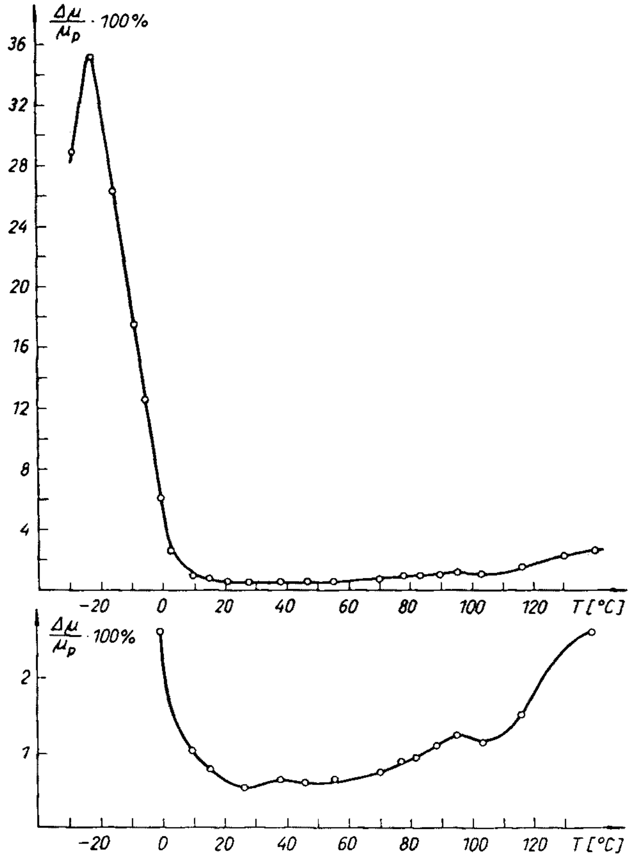

Fig. 1. $\frac{\Delta\mu}{\mu_p}$ *versus* temperature for GRII-2 sample, cooled slowly

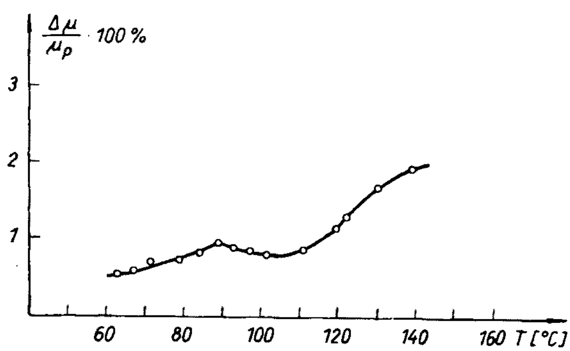

Fig. 2. Disaccommodation in GRII-2 sample after ageing at room temperature; the region from + 60° to +140°C

In Fig. 3 we see the disaccommodation in the range from —30° to 110°C obtained for the P-1 sample, also cooled slowly. Measurements of the tangent of the angle of losses as a function of temperature have shown that this sample contains only nitrogen in solid solution [10].

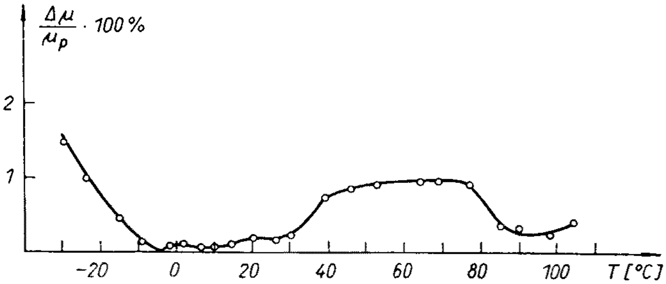

Fig. 3. Disaccommodation in P-1 sample, cooled slowly

The effect of ageing at 25°C and higher temperatures on disaccommodation in the range from —40° to 110°C was examined. Figures 4 and 5 present the results obtained for the GRII-2 sample. Additional ageing at 200°C for 24 hours brought about a drop by one half of the disaccommodation in the Snoek region, and a slight change of it above room

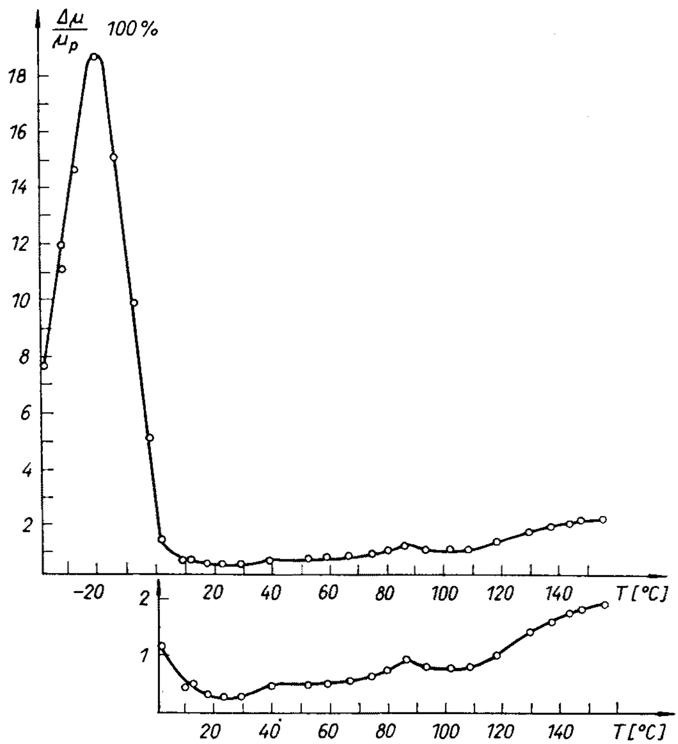

Fig. 4. Disaccommodation in GRII-2 sample after ageing at room temperature

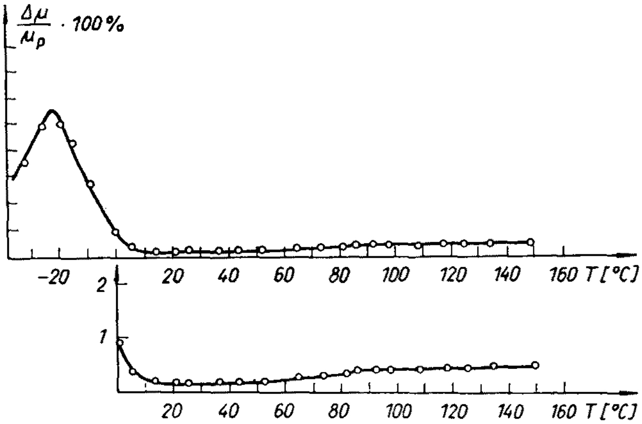

Fig. 5. The dependence $\frac{\Delta\mu}{\mu_p}=f(T)$ in GRII-2 sample after ageing at 200°C

temperature. It was also found that ageing at 50°C does not affect $\frac{\Delta\mu}{\mu_p}$ in the entire examined range of temperatures, but ageing at 100°C decreases it in the Snoek range (from 28 to 18 per cent) and only slightly affects the region above room temperature.

3.2.2. Quenched samples

Figure 6 depicts the curve $\frac{\Delta\mu}{\mu_p}=f(T)$ obtained for a sample of the GR series, quenched from 700°C. As is seen, this curve has a minimum near 50°C and a maximum at 85°C. The

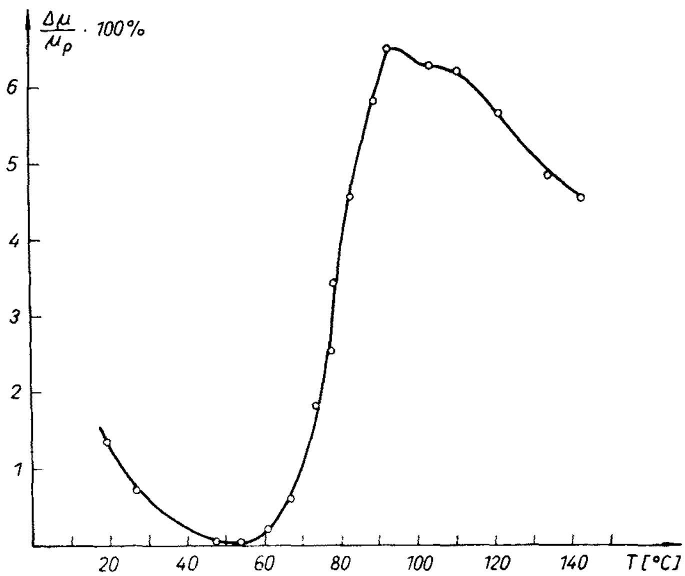

Fig. 6. $\frac{\Delta\mu}{\mu_p}$ *versus* temperature in GRII-2 sample after quenching from 700°C

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ageing which supervenes during performance of the measurement heightened the value in the first region and lowered it in the second.

In the case of a P series sample (P2) after quenching there was no disaccommodation in the 30° to 60°C region (Fig. 7). In this range time decrease of permeability appeared in the second measurement.

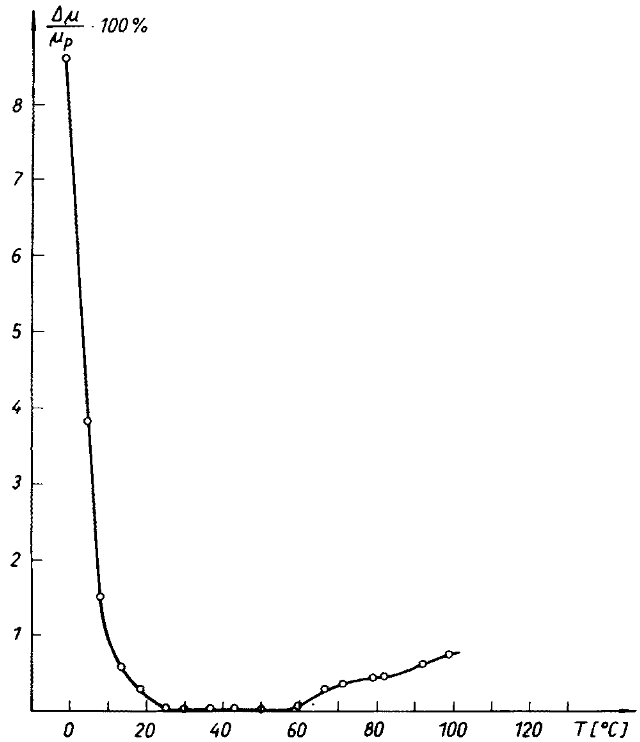

Fig. 7. The dependence $\frac{\Delta\mu}{\mu_p} = f(T)$ in P-2 sample quenched from 700°C

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3.2.3. The influence of plastic working

The stress-relieved GR series sample was submitted to plastic working by cold rolling of the order of 25 per cent. The obtained dependence $\frac{\Delta\mu}{\mu_p} = f(T)$ is shown in Fig. 8 (curve 1). During the second measurement there proved to be a considerable drop in disaccommodation in the 30° to 120°C range (curve 2).

3.3. Discussion of results

It is seen distinctly in Figs 1 and 3 that apart from disaccommodation associated with carbon and nitrogen Snoek relaxation which appears below 0°C there is in the stress-relieved αFe—C—N samples a weak, but well reproducible, time-change of permeability above

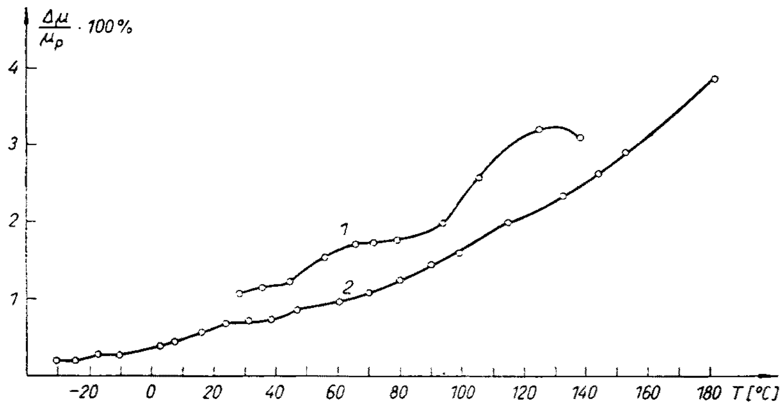

Fig. 8. Disaccommodation for GRII-2 sample after cold working of the order of 25%. Curve *1* — measurement immediately after working, curve *2* — second measurement

room temperature. In the case of the P-1 sample cooled slowly and containing only nitrogen, this effect appears already below 0°C.

Research on the effect of ageing showed that ageing temperatures up to 50°C bear little effect on the shape of the $\frac{\Delta\mu}{\mu_p} = f(T)$ curve in the entire range of temperatures. This influence increases with higher ageing temperature, and is particularly large for 200°C.

In quenched samples, the $\frac{\Delta\mu}{\mu_p} = f(T)$ curves have a different shape (Figs 6 and 7). In a certain region of temperatures disaccommodation has a very low value or is even entirely absent. Below and above this region the changes of permeability with time are much larger than in specimens which had been relieved of stress and cooled slowly.

The increase of $\frac{\Delta\mu}{\mu_p}$ in the region below the temperature of the minimum is due to disaccommodation processes from small clusters of interstitial atoms (pairs, triplets, *etc.*), which appear there [11, 19, 20]. After quenching the concentration of the interstitial impurities is much higher than when cooling is slowly executed. On the other hand, the concentration of the pairs is proportional to the square, and that of triplets to the cube, of the concentration of the single atoms. Therefore, the disaccommodation due to these clusters increases very rapidly after quenching. This reasoning is corroborated by the results obtained for the P-2 sample (Fig. 7). The measurements of tan $\delta_r = f(T)$, made for this sample after quenching, gave a Snoek peak of height corresponding to a 0.01 per cent carbon concentration in the solution (in fine agreement with the result of chemical analysis), hence, twice as low as in the GRII-2 sample. Because of this the temperature region from 30° to 45°C has been revealed in Fig. 7.

The examined permeability disaccommodation in αFe—C—N, appearing above —10°C (Fig. 3), is not due to interstitial C and N atoms jumping in the neighbourhood of a foreign substitutional atom. Simple calculations show that in the P series samples, containing only 0.01 per cent of impurities substituting iron in the solution, the intensity of this type of

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disaccommodation should be lower by an order of magnitude than the effect observed. The results obtained for the quenched samples (Fig. 6 and 7), *viz.*, the decrease or disappearance of the time-changes of permeability in a certain region of temperatures, also confirm this hypothesis. For it is well known that the quenching process does not affect disaccommodation due to substitutional impurities very much.

The quenching experiments show also that when the samples are heated to 700°C the relaxation centers of the investigated effects decay, and then the quenching processes freeze them in. During ageing or measurement these centers reappear, leading to the formation or increase of disaccommodation. The results concerning the effect of ageing on disaccommodation in samples cooled slowly suggest that the studied phenomenon is closely related to the extraction of carbon and nitrogen from the solid solution in α Fe.

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