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Development of Magnetic After-Effect Setup and Application in the Study of Relaxation Processes in Fe-C, Fe-Cu and Fe-Cr Alloys

Abstract. *A magnetic after-effect setup, based on an open magnetic circuit, is developed and presented. The magnetic after-effect experiment aims to determine the time and temperature dependence of the initial susceptibility, which is obtained by an AC measurement of the mutual inductance. After sample demagnetization, the change of the time dependent initial susceptibility is measured for particular ferromagnetic materials at a chosen temperature. The setup is applied for the investigation of relaxation processes of dislocations and carbon atoms in Fe-C, Fe-Cu and Fe-Cr alloys.*

Streszczenie. *Przedstawiono układ do badania lepkości magnetycznej w obwodach otwartych. Badano początkową susceptancję jako funkcję temperatury i czasu. Analizowano procesy relaksacyjne w stopach Fe-C, Fe-Cu i Fe-Cr. (**System do badania procesów relaksacyjnych w stopach Fe-C, Fe-Cu i Fe-Cr**)*

Keywords: magnetic after-effect, initial susceptibility, Fe-based alloys.
Słowa kluczowe: after –fekt – lepkość magnetyczna, początkowa susceptancja.

Introduction

Magnetic relaxation is the relaxation of a magnetic system to an equilibrium state or steady-state condition as the magnetic field is changed. The corresponding measurement technique, also known as Magnetic After-Effect (MAE) spectroscopy, has been constantly improved in the last few decades and applied to several pure ferromagnetic materials and a great number of alloys in order to obtain detailed information on intrinsic defects, impurity atoms and their interactions [1]. It is a non-destructive evaluation technique that belongs to the group of relaxation techniques which has the great advantage to be sensitive not only to the mere presence of the lattice defects - to what most other methods are confined - but, in addition, to detect defect movements such as local reorientations or diffusional migration [2].

The MAE is a general term for the following phenomenon: after demagnetization of the sample under consideration a time-dependent decrease of the initial reversible susceptibility, χ, (or alternatively, the increase of its reciprocal value, i.e. the reluctivity $r = 1/\chi$) is observed for particular ferromagnetic materials at specific temperatures. Such disaccommodation of the initial susceptibility is due to interactions between the magnetic domain walls and the thermally activated motion of microstructural defects towards energetically more favourable positions. These interactions result in a time-dependent process of magnetic domain wall stabilization [3], which leads to the reduction of domain wall mobility, and hence to the decrease of the initial susceptibility with increasing time.

Here, we developed a fully automated magnetic after-effect setup. It is used for the study of relaxation processes of carbon interstitials, dislocations as well as their interplay in Fe-C, Fe-Cr and thermally aged Fe-1%Cu alloys. This contributes to the development of a non-destructive evaluation technique for the analysis of irradiation induced embrittlement of nuclear reactor pressure vessel steels.

Experiment

The purpose of the MAE experiment is to determine the time and temperature dependence of the initial susceptibility, which is obtained by an AC measurement of the mutual inductance.

Magnetic after-effect setup:

Schematic representation of the magnetic relaxation setup is given in Fig. 1.

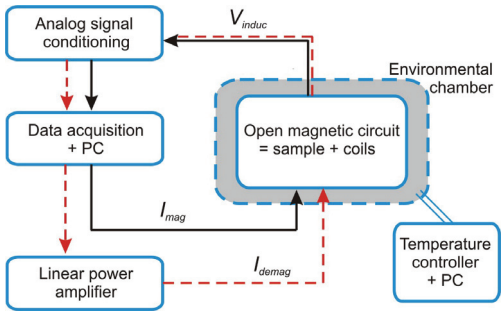

Fig. 1. Schematic representation of the magnetic after-effect setup. The core element of the setup is the ferromagnetic sample under test around which concentric coils are placed (open magnetic circuit). The open magnetic circuit is inserted in a temperature-controlled environmental chamber. The dashed line (- - -) represents the demagnetization stage and the full line (---) shows the magnetization stage to measure initial susceptibility.

As the core element of the setup, the sample under test is placed within three concentric cylindrical coils, as shown in Fig. 2.

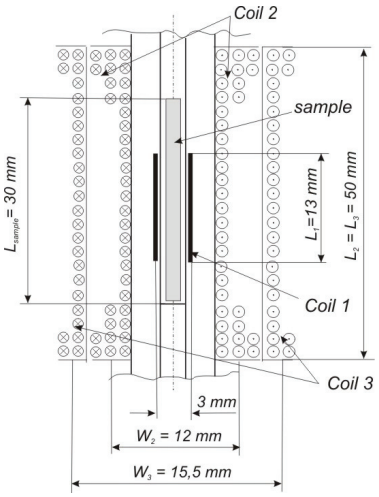

Fig. 2. The ferromagnetic sample placed as the core of three concentric cylindrical coils (open magnetic circuit). Coil 1 – to measure induced voltage; Coil 2 – to apply the excitation current during the magnetization stage; Coil 3 – to apply the excitation current during demagnetization stage.

The time dependent magnetic flux in the sample results in an induced voltage in coil 1, positioned closest to the sample. Coil 2 is used for applying the excitation current during the magnetization stage, while the outlying coil 3 is applied for the excitation current during the demagnetization stage. The MAE setup is designed for ferromagnetic rectangular samples with dimensions 1.3×1.3×30 mm^3. Since no additional means are present to close the flux path, this configuration can be considered as an open magnetic circuit. In order to determine the temperature dependence of the susceptibility this circuit is inserted in a temperature-controlled environmental chamber.

The experimental procedure consists of three steps:

1. The temperature of the environmental chamber is controlled to a predefined temperature set point by the combined action of resistive heating and liquid nitrogen cooling.

2. After the temperature stabilization at the set point, the demagnetization procedure begins, starting with an alternating magnetic field with initial amplitude of about 10 kA/m and decaying. Fig. 3 shows the electronic scheme of the MAE setup.

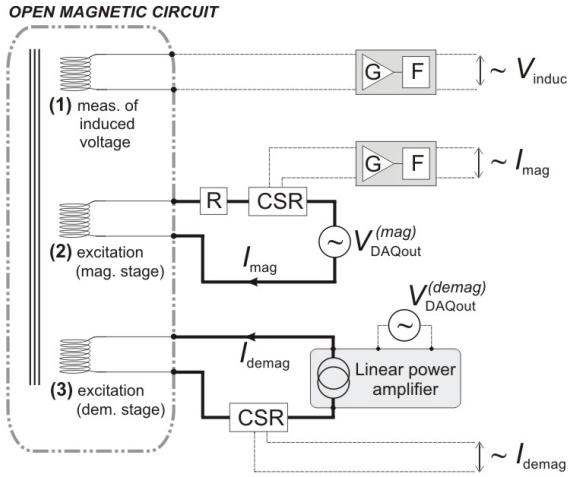

Fig. 3. Electronic scheme of the magnetic after-effect setup. The linear power amplifier is used as a current source. R – resistor of 1 kΩ, CSR – four-point current sense resistor with resistance value of 1 Ω, G –instrumentation amplifier with a gain of 1065, F – low pass filter with cut-off frequency f_c = 500 Hz.

The linear amplifier is used as source of the excitation current applied during the demagnetization phase. For the application of I_{demag}, coil 3 is placed around the sample under test.

3. Once the field metric demagnetization is finished, the demagnetizing electronic circuit opens. Then, a magnetic AC sinusoidal excitation is applied to the sample, with small constant effective amplitude, H_{eff}, of about 10 A/m, which is small enough to correspond with the reversible region of the magnetization curve (magnetic induction ≈ 1 mT). The design of coil 2 is done such that the magnetic field amplitude of 10 A/m corresponds with the excitation current amplitude of the order of mA. This order of magnitude for the electrical current enables us to apply the excitation directly from the DAQ, without additional amplifier (since the analogue output of the DAQ-card can deliver a maximum current of 5 mA), having the advantage that no DC - offset and no additional noise is introduced in excitation coil 2. An appropriate resistor (R) is added to the circuit in order to maximize the usage of the voltage range of the analogue output channel of the DAQ (10 V).

The measurement of the initial susceptibility:

During the sinusoidal field excitation, characterized by the amplitude H_{eff} *and* the excitation frequency, f_{excit}, the initial susceptibility of the sample under test at constant temperature set point T, is measured. The measurement is performed at specific time points t_i, starting from 1 s to 180 s, and with a time interval Δt_i. This interval is the interval between succeeding initial susceptibility measurements, and is set here to 1 s.

Moreover, in order to obtain the susceptibility value at a certain discrete time point, t_i, the waveforms of the excitation current $I_{mag}(t)$ (coil 2), and induced voltage $V_{induc}(t)$ in coil 1 are measured during a certain time scope δt around t_i, i.e. during $[t_i - \delta t/2,\ t_i + \delta t/2]$. The time interval δt and the frequency f_{excit} of the applied current $I_{mag}(T)$ are chosen in such a way that the following expression is fulfilled: $\delta t = j/f_{excit} < \Delta t_i$. The integer value j gives the number of periods of $I_{mag}(t)$ and $V_{induc}(t)$ that are measured during the time interval δt. The excitation frequency set point is an interplay between maximizing the signal-to-noise ratio and minimizing the skin effect. Moreover, with a higher j or δt, a higher accuracy of the susceptibility measurement is obtained. By using the parameters f_{excit} and δt which are set in the experiments to 275 Hz and 0.2 s, respectively, the number of measured periods of the waveforms $I_{mag}(t)$ and $V_{induc}(t)$ is j = 55.

The waveforms $I_{mag}(t)$ and $V_{induc}(t)$ which are obtained during the time interval δt, are served as input data for two Fourier transformations, resulting in $\hat{I}_{mag}(t)$ and $\hat{V}_{induc}(t)$ which are the amplitudes of the excitation current and the induced voltage respectively. Both amplitudes correspond to the set point of excitation frequency, f_{excit}. By using these amplitudes, we obtain the mutual inductance $\Lambda_m(t_i, T)$, between coil 1 and coil 2 of the open magnetic circuit around the sample:

$$\Lambda_m(t_i,T) = \frac{\hat{V}_{induc}(t_i,T)}{2\pi \cdot f_{excit} \cdot \hat{I}_{mag}(t_i,T)}. \tag{1}$$

where, the angular frequency ω is equal to $2\pi f_{excit}$. In order to obtain the initial value of the magnetic susceptibility, we have used the basic equations which describe the correlations between the relative permeability μ_r, and the magnetic susceptibility. Taking into account the relation between the magnetic inductance $B(t_i, T)$ and the effective magnetic field which acts on the sample, $H_{eff}(t_i, T)$, i.e. :

$$\hat{B}(t_i,T) = \mu \cdot \hat{H}_{eff}(t_i,T), \tag{2}$$

and considering the definition of the relative permeability $\mu_r = \mu/\mu_0$ as well as $\chi = \mu_r - 1$, we expressed the initial susceptibility in the following way:

$$\chi(t_i,T) = \frac{\hat{B}(t_i,T)}{\mu_0 \cdot \hat{H}_{eff}(t_i,T)} - 1. \tag{3}$$

The amplitude of the induced voltage in coil 1 is given as a function of the number of turns of coil n_1, cross section of the specimen S_{mat}, angular frequency ω, and the amplitude of the magnetic inductance:

$$\hat{V}_{induc}(t_i,T) = \beta_1 \cdot n_1 \cdot S_{mat} \cdot \omega \cdot \hat{B}(t_i,T). \tag{4}$$

An approximation related to the fact that the cutting frequency of 500 Hz might be too close to the excitation frequency of 275 Hz is taken into account by adding the correction factor β_1 (error for susceptibility $\beta_1 \leq 1$) in Eq. (4).

Using the following expression related to coil 2, where the amplitude of the effective magnetic field is proportional to the amplitude of excitation current [4]:

$$\hat{H}_{eff}(t_i,T)\cdot L_2 = \beta_2 \cdot n_2 \cdot \hat{I}_{mag}(t_i,T), \quad (5)$$

as well as Eqs. (1) and (3), the value of the initial magnetic susceptibility is obtained as follows:

$$\chi(t_i,T) = \frac{\Lambda_m(t_i,T)\cdot L_2}{\beta_1 \cdot \beta_2 \cdot n_1 \cdot n_2 \cdot S_{mat} \cdot \mu_0} - 1, \quad (6)$$

with n_2 - the numbers of turns of coil 2. However, due to the choice that the magnetic path length is the length of the coil 2 L_2, the correction factor β_2 appears in Eq. (5). For each discrete time point t_i, these measurements and processing are performed, resulting in one initial susceptibility value χ for each t_i.

Magnetic after-effect spectra:

One of the typical experimental results of these measurements is shown in Fig. 4 where the change of initial susceptibility as a function of time t_i is plotted.

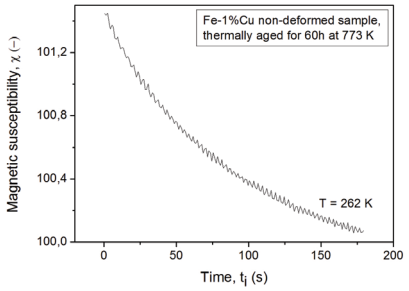

Fig. 4. The change of magnetic susceptibility as a function of time, of a non-deformed, for 60 h thermally aged at 773 K Fe-1%Cu sample. The measurement was performed at 262 K.

This measuring sequence - temperature control, demagnetization, and measurement of initial magnetic susceptibility, is then repeated for other temperature set points in a range of 200 K to 500 K and with a temperature step of 4 K. A few MAE curves, obtained from the tests performed at different temperatures, are presented in Fig. 5.

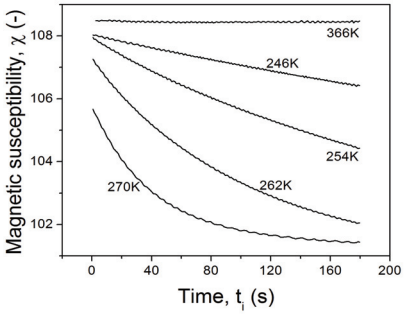

Fig. 5. MAE curves obtained from the measurements performed at different temperatures for non-deformed, thermally aged for 60 h at 773 K Fe-1%Cu- samples.

Once the measurements are performed for all temperatures T, in order to study the specific temperature dependence of the magnetic after-effect, the experimental isothermal data can be processed into isochronal curves. These curves or so-called MAE spectra show the relative change of initial susceptibility, S, as a function of temperature between two times t_{begin} and t_{end} in the following way:

$$S(T) = \frac{\Delta\chi}{\chi_{end}}(T) = \frac{\chi(t_i = t_{begin},T) - \chi(t_i = t_{end},T)}{\chi(t_i = t_{end},T)}, \quad (7)$$

Alternatively, it can be presented as the relative change of the reluctivity, r ($r = 1/\chi$) [5]:

$$\frac{\Delta r}{r_1}(T) = \frac{r(t_i = t_{end},T) - r(t_i = t_{begin},T)}{r(t_i = t_{begin},T)}, \quad (8)$$

with set parameters $t_{begin} = 1\ s$ and $t_i = 180\ s$, where $r_1 = r(t_{begin}, T)$. Notice that, when neglecting the second term in the RHS of Eqn. (6), the correction factors β_1 and β_2 disappear in (7) and (8).

If a certain thermally activated relaxation process (interaction between magnetic domain walls and the lattice defects) exists at a specific temperature, the decrease of the isothermal susceptibility with time is more pronounced, as shown in Fig. 5. Such interaction will be characterized by a peak occurring (local maximum) in the isochronal relaxation curve – the relative change of reluctivity as a function of temperature, see Fig. 6.

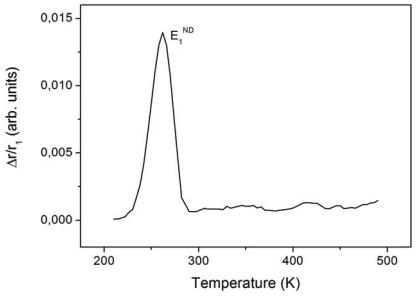

Fig. 6. Isochronal relaxation curve of non-deformed, thermally aged for 60h at 773 K Fe-1%Cu sample. The peak at about 262, denoted as E_1^{ND} is recognized as the Snoek relaxation process of carbon interstitial atoms in iron lattice.

Materials

The details of the Fe-C, Fe-1%Cu and Fe-2.5%Cr alloys production and the sample preparations are published elsewhere [5, 6, 7]. The chemical composition of the materials used in these studies is given in Table 1.

Table 1. The nominal composition of investigated alloys

Material	Nominal composition (wt.%)
Fe-C	(< 200 ppm impurities; ~ 30 ppm C)
Fe-1%Cu	1%Cu (~ 30 ppm C)
Fe-2.5%Cr	2.5%Cr; 0.01%C

The various stages of copper precipitation, which plays an important role in dislocation dynamics, in Fe-1%Cu alloys were achieved by a thermal aging process, consisting of time-dependent heat treatments at 773 K in argon atmosphere, and subsequent fast quenching into iced water. The duration of the heat treatment varied from 0.1 h to 480 h. The Fe-2.5%Cr sample was also thermally aged but on 823 K for 0.75 h followed by water quenching, thus avoiding creation of carbides. The Fe-C alloys were not thermally treated.

The second set of samples of Fe-Cu and Fe-Cr alloys was, prior to the MAE measurements, subjected to a torsional cyclic plastic deformation in a consistent way by an angle of $\pi/2$ over their length at room temperature. This corresponds to a local deformation of about 6% ($\alpha R/L$). Fe-C samples were deformed by an angle of $3\pi/2$ (18%), also at room temperature.

Results

The MAE spectra of non-deformed and cold-worked samples of Fe-C; Fe-1%Cu and Fe-2.5%Cr alloys are shown in Fig. 7.

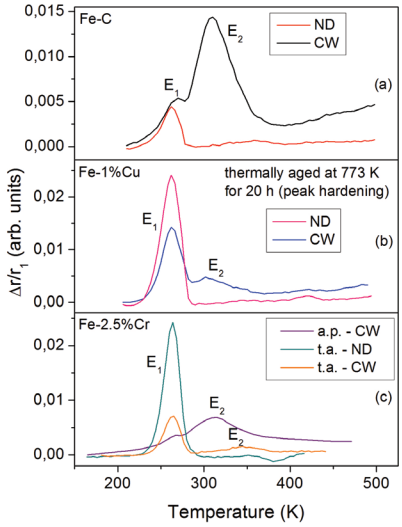

Fig. 7. The MAE spectra of non-deformed (ND) and cold-worked (CW) samples of Fe-C (a), Fe-1%Cu (b) and Fe-2.5Cr% (c) alloys. The Fe-1%Cu samples were thermally aged for 20h at 773 K – the aging time which corresponds to the peak hardening of this alloy. The as provided (a.p.) Fe-2.5%Cr sample, after plastic deformation and MAE measurement, was thermally aged (t.a.) for 0.75 h at 823 K and tested again.

In the case of non-deformed samples for all alloys, by increasing the temperature the relative change of reluctivity exhibits a peak at about 260 K, denoted as E_1. The full width at half maximum ($FWHM$), temperature position (T_C) and the integrated intensity are the main peak parameters which are used to analyze the relaxation processes. They are proportional to the distribution of relaxation times ($FWHM$), activation energy (T_C), and concentration of relaxed defects (integrated intensity) [8]. On the basis of previous investigations which show very good agreement with the peak parameters obtained in our measurements, the E_1-peak is assigned to the Snoek relaxation process of carbon interstitials [3].

As a consequence of plastic deformation we have a more complex structure, see Fig. 7. Beside the Snoek-peak, the MAE spectra of cold-worked samples in all alloys exhibit an additional structure, denoted as E_2, which temperature position is in the range between 320 K and 340 K. The previous investigations based on the MAE measurements have shown that the temperature of 330 K corresponds to the activation energy of 1.08 eV [9]. This value agrees well to the activation energy of the relaxation process related to dislocation motion obtained from the previous internal friction measurements [10]. Moreover, the $FWHM$ of the E_2-peak (≈ 60 K) which is much larger than in the case of E_1, indicates that this relaxation process is not governed by a single relaxation time, but a distribution of times. This gives an additional argument in favor of assignment of the E_2-peak to the relaxation process of line defects such as dislocation motion.

Contrary to the Snoek-peak in the spectra of non-deformed thermally aged Fe-1%Cu samples, the E_1 – peak observed in the spectra of plastically deformed samples shows a reduced intensity, see Fig. 7(b). Such behavior is expectable, since a certain amount of carbon atoms is segregated at dislocations during plastic deformation. In addition, the different ratio of the E_1 and E_2 peaks intensities in the spectrum of the CW Fe-C sample comparing the one in Fe-Cu and Fe-Cr samples is a consequence of the applied amount of plastic deformation.

Conclusion

To conclude, we developed a fully automated magnetic-after effect setup which is based on an open magnetic circuit consisting of three concentric cylindrical coils. The experimental procedure which enables the measurement of time dependent initial (reversible) magnetic susceptibility consists of three stages – temperature control, demagnetization and magnetization (which is the actual measurement of the time-dependent decay of the initial susceptibility at a fixed temperature). This measuring sequence is repeated for each temperature set point within the studied temperature range.

In order to observe relaxation processes by using the new developed magnetic after-effect setup, we performed measurements on Fe-C, Fe-1%Cu and Fe-2.5%Cr alloys. In the isochronal relaxation spectra of all alloys, two types of processes, observed at 260 K and around 330 K, are assigned to the Snoek relaxation of carbon interstitial atoms and thermal activation of dislocation motion (under plastic deformation), respectively.

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