IN-SITU AC-HYSTERESIS MEASUREMENTS OF SPD-PROCESSED Cu_{20}(Fe_{15}Co_{85})_{80}

A PREPRINT

Martin Stückler¹, Stefan Wurster, Reinhard Pippan, Andrea Bachmaier
Erich Schmid Institute of Materials Science, Austrian Academy of Sciences
Jahnstraße 12, 8700 Leoben, Austria
¹martin.stueckler@oeaw.ac.at

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ABSTRACT

The changes of magnetic properties upon heat treatment of a metastable supersaturated solid solution processed by severe plastic deformation are investigated by in-situ AC-hysteresis measurements. Data are analyzed in the framework of dynamic loss theory, with correlative investigations of the microstructural properties. The evolution of hysteresis upon annealing points out that the single-phase supersaturated solid solution remains stable up to 400°C, then hindering of domain wall motion sets in at this temperature. At 600°C, a multi phase microstructure is present, causing a significant increase in coercivity.

1 Introduction

With high-pressure torsion (HPT), a technique of severe plastic deformation (SPD), it is possible to form bulk supersaturated solid solutions with grain sizes in the nanocrystalline regime [1, 2]. It has been shown, that with different ratios of Co to Cu, huge reductions in the coercivity can be achieved by means of SPD, resulting in soft magnetic materials [3, 4]. In the present study, Fe is added to lower the magnetocrystalline anisotropy to further reduce the coercivity [5]. To study the evolution of magnetic properties as a function of temperature, the hysteresis is recorded during in-situ annealing treatments with a concomitant investigation of the dynamic magnetic behavior for certain temperatures. The resulting data are discussed in the framework of dynamic loss theory and correlated to the evolving microstructure.

2 Experimental

Powders (Fe: MaTeck 99.9% -100+200 mesh; Co: GoodFellow 99.9% 50-150 μm; Cu: Alfa Aesar -170+400 mesh 99.9%) were mixed and hydrostatically consolidated in Ar-atmosphere. A coin-shaped specimen (diameter: 8 mm; thickness: 1 mm) was processed by two subsequent steps of HPT deformation (100 turns at 300°C; 50 turns at room temperature (RT)), as described elsewhere [4]. The sample was further processed into a ring shaped specimen and equipped with 68 primary windings and 61 secondary windings (Cu-wire; diameter: 0.315 mm and 0.200 mm, respectively). Electrical isolation between the sample and the windings was maintained with a high temperature adhesive (Minco FortaFix Autostic FC8). To apply the magnetic field, a sinusoidal current (4 A; 5-1000 Hz) was applied to the primary winding with a KEPCO BOP 100-4M power supply, according to [eq. 1a]. The voltage induced in the secondary windings [eq. 2] was measured with a National Instruments BNC-2110 terminal block. Data processing was carried out with LabView (version 14.0.1f3). The hysteresis measurement is described in more detail in Ref. [6]. For in-situ measurements, the sample was clamped between two Cu-blocks and heated by cartridge heaters (hotset hotrod HHP HT4030504). The temperature was measured by a K-type thermocouple, close to the samples’ position to ensure the hysteresis measurements take place at ±5°C of the target temperature. To maintain a homogeneously heated sample, measurements were started 15 min after stabilization of the target temperature. In-situ measurements were performed in a customized vacuum chamber to prevent oxidation, maintaining a pressure below 10⁻² mbar during the
Figure 1: Evolution of in-situ AC-hysteresis curves according to the temperature treatment in fig. 2. The as-deformed state is measured at RT (a). After in-situ temperature treatment at 150°C (b), 300°C (c), 400°C (d) and 600°C (e), the specimen is measured again after cooling down to room temperature (600°C-RT; f). The legend in (a) applies to all diagrams.

whole experiment.
Microstructural investigations were performed by X-ray diffraction using Co-Kα radiation (XRD; Bruker D2-Phaser) and scanning electron microscopy (SEM; Zeiss LEO1525) in backscattered electron (BSE) mode. The composition was determined by an energy dispersive X-ray spectroscopy (EDS; Bruker XFlash 6160) system.

3 Results and discussion

The magnetic field $H$ is controlled by the number of primary windings $N_p$, the applied current $I$ and the mean diameter $d_m$ [eq. 1(a)], resulting in the present case in a maximum magnetic field of 11.9 kAm$^{-1}$ ($d_{outer}=8.76$ mm; $d_{inner}=5.76$ mm).

$$H = \frac{N_p \cdot I}{\pi \cdot d_m} \quad \text{(1a)}$$

$$d_m = \frac{d_{outer} + d_{inner}}{2} \quad \text{(1b)}$$

Eq. 2 gives the magnetic induction $B$ as a function of the time dependent induced voltage $U_{ind}(t)$, the number of secondary windings $N_s$ and the cross-sectional area $A$ of the ring-core (here: $A=0.707$ mm$^2$).

$$B = \int \frac{U_{ind}(t)dt}{N_s \cdot A} \quad \text{(2)}$$

The specimen is measured in the as-deformed state at RT, shown in fig. 1(a). It can be seen, that saturation is achieved and the area of the hysteresis loop rises slightly with increasing frequency, indicating low eddy-current losses. The in-situ temperature treatment was performed according to the thermal profile shown in fig. 2. Measurements were started after settling the target temperature for 15 min, since similarly processed Co-Cu samples showed the majority of microstructural changes happening only during a short time period after reaching the target temperature. The time stamps of the measurements are represented by the black diamonds in fig. 2. Hysteresis are measured at 150°C (fig. 2(b)), 300°C (fig. 2(c)), 400°C (fig. 2(d)) and 600°C (fig. 2(e)). For the 150°C measurement, the temperature exceeded 160°C for a short period of time, but it was shown that the microstructure does not change in this temperature range since only relief of internal stresses takes place to a small extend [8]. The hysteresis exhibits similar shapes
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Figure 2: Temperature $T$ during the in-situ experiment as a function of time $t$. Black diamonds mark the periods, in which hysteresis measurements are performed.

up to 300°C, but at 400°C the frequency behavior changes significantly, which is clearly visible in the 1000 Hz measurement. The area of the 1000 Hz hysteresis increases again in the 600°C-state. The measurement of the in-situ treated specimen is repeated after cooling down to RT (fig. 1(f); 600°C-RT). For the 600°C-RT state saturation is not achieved in the 1000 Hz measurement. This means, the measured hysteresis represents a minor loop and the coercivity cannot be evaluated for this frequency. Lower frequencies reveal a larger area of the hysteresis loop with respect to the 600°C-state, arising most likely from the temperature dependence of the magnetocrystalline anisotropy according to Brukhatov-Kirensky [9].

In the following, a quantitative analysis on the evolution of the coercivity is carried out. Fig. 3 shows the measured coercivities as a function of frequency. To disentangle the (static) intrinsic magnetic properties from the dynamic losses, [eq. 3] is fitted to the data [10, 11, 12].

$H_C(f) = H_C(0) + b \cdot \sqrt{f} + c \cdot f$

In [eq. 3], the dynamic loss is separated into anomalous-loss $b$, caused by domain wall motion, and eddy current loss $c$, which is mainly controlled by the conductivity [13]. Since the conductivity of SPD-processed materials is significantly lowered with respect to coarse-grained materials [14], we assume the dynamic losses being mainly controlled by anomalous losses and therefore neglect the third term in [eq. 3]. In fig. 3 the measured coercivities are plotted versus the square-root of frequency, showing a linear scaling with $\sqrt{f}$, confirming the aforementioned statement. The results from linearly fitting the data are shown in fig. 4. Diminishing static coercivity, as well as anomalous loss, can be identified between the as-deformed, 150°C- and 300°C-annealed state. For SPD-processed Cu-Co and Cu-Fe-Co, a diminishing defect density was reported in this temperature window, but no apparent changes in the grain size have been determined [4][8]. A huge jump in $H_C(0)$ can be noticed at 600°C, indicating a large microstructural variation, such as grain growth. Large microstructural variations have been determined in similar materials at this temperature [4][8]. A further increase in coercivity is visible for the 600°C-RT state, which is again traced back to temperature dependence.

The anomalous loss parameter $b$ is closely related to the microstructure, and takes into account the energy needed for domain wall motion, which can be increased due to pinning at lattice defects or grain / phase boundaries. The increase in $b$ at 400°C therefore indicates the formation of pinning centers accelerating domain wall motion [16][17], which rushes ahead the demixing of the microstructure at 600°C. For the 600°C-state, the anomalous loss parameter stays rather constant.

The microstructure of the in-situ heat-treated sample is investigated by SEM and XRD and compared to the initial (as-deformed) state. For this purpose a second sample is fabricated, representing the as-deformed state. Fig. 5 shows SEM images of both samples. The as-deformed state (fig. 5(a)) shows a highly homogeneous, nanocrystalline microstructure. In the 600°C-RT state (fig. 5(b)), a significantly larger grain size in the ultra-fine grained regime is visible. Furthermore, phase contrast indicates a chemical inhomogeneity, showing demixing tendencies. Bright areas indicate high Z and therefore Cu-rich regions, whereas dark-grey or black areas point at the presence of low Z
In-situ AC-hysteresis measurements of SPD-processed Cu$_{20}$(Fe$_{15}$Co$_{85}$)$_{80}$ reveal a persisting soft magnetic behavior up to 400°C. The amount of eddy current losses is low by comparison, owing to the high resistivity of SPD-processed materials. At 400°C, pinning centers start to form, accelerating domain wall motion and causing an increase in dynamic loss. At 600°C, the microstructure has changed from the initial single-phase supersaturated solid solution into a multi-phase microstructure according to the thermodynamical equilibrium. The formation of pinning centers rushes ahead this phase change. The results demonstrate the capability of magnetic measurements capturing smallest microstructural changes before they become evident with other techniques.

4 Conclusion

In-situ AC-hysteresis measurements of SPD-processed Cu$_{20}$(Fe$_{15}$Co$_{85}$)$_{80}$ reveal a persisting soft magnetic behavior up to 400°C. The amount of eddy current losses is low by comparison, owing to the high resistivity of SPD-processed materials. At 400°C, pinning centers start to form, accelerating domain wall motion and causing an increase in dynamic loss. At 600°C, the microstructure has changed from the initial single-phase supersaturated solid solution into a multi-phase microstructure according to the thermodynamical equilibrium. The formation of pinning centers rushes ahead this phase change. The results demonstrate the capability of magnetic measurements capturing smallest microstructural changes before they become evident with other techniques.
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Data Availibility Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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