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Improved magnetic performance of Co-based ribbons by nano-crystallization through magnetic annealing

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Abstract

Different techniques of experimental analysis have been combined to extract complementary information about the phase transformations, DC and AC magnetic properties through the stages of nano-crystallization of Co-based amorphous ribbons, via transverse magnetic annealing. The nano-crystallization starts by nucleation and growth of soft magnetic meta-stable Co_{23}B_6 phase with less nucleation activation energy compared to other, thermodynamically favored, stable phases. Then, Co_2B and Co_3B , as a semi-hard magnetic phase, are identified in the samples, magnetic annealed at 525 and 550°C, respectively. Field-induced anisotropy dominates over the residual contributions of magneto-crystalline anisotropy by the different components, after field annealing at 510, 515 and 525°C. The anomalous loss is found to account for more than 90 % of the total loss. The same is significantly reduced by annealing in a transverse magnetic field, which results in an easy axis perpendicular to the length and a hard axis along the length of the ribbons, leading to a change of the dominant magnetization mechanism. In addition, magnetic annealing also causes a measurable decrease in the domain width, which, in turn, promotes pinning and inhibits domain wall motion, thus further favoring coherent domain rotation as the main mechanism of magnetization. This combination accounts for a 70 % decrement in the anomalous loss in the so-processed ribbons at 525°C and renders them attractive for applications in mid- and high-frequency power supplies and inverters.

Keywords: Amorphous alloys; Nano-crystallization; Magnetic properties; Magnetization process; Magnetostructural transformations

1. Introduction

Co-based soft ferromagnetic nanocomposite materials are obtained by crystallizing amorphous ribbons of compositions which typically include both large e.g., Zr or Nb and small e.g., B and Si atoms relative to the ferromagnetic transition-metal elements. In these complex alloy systems, the

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large and small atoms provide for a good glass forming ability and they enable one to obtain a composite microstructure, which consists of nanocrystals of a transition-metal-rich phase embedded within an intergranular amorphous phase, enriched in the glass formers, after the first ‘primary’ crystallization step [1,2].

At higher annealing temperatures, resulting in ‘secondary crystallization’, the glass-former enriched intergranular amorphous phase crystallizes as well, resulting in the formation of intermetallic compounds. A number of different intermetallic compounds are noted in the literature depending on the exact alloy composition including $(\text{Fe,Co,Ni})_2\text{B}$, $(\text{Fe,Co,Ni})_2\text{Zr}$, and $(\text{Fe,Co,Ni})_3\text{B}$. More complex phases exist, exhibiting the Cr_{23}C_6 prototype structure, such as $(\text{Fe,Co,Ni})_{23}\text{B}_6$ which has a large unit cell, with a size of more than 1 nm, containing 92 medium-sized ferromagnetic transition-metal elements and 24 small metalloid atoms [3]. This phase is a metastable one, which can easily decompose into $(\text{Fe,Co,Ni})_3\text{B}$ and $(\text{Fe,Co,Ni})_2\text{B}$. However, when the main transition metal site is partially substituted for by another transition metal atom, such as Nb, Co, or Ni, the phase is stabilized and more easily retained [4].

The precipitation of M_{23}B_6 phase throughout the amorphous matrix results in a significant improvement in the soft magnetic properties and low losses at high frequencies [5]. In addition, the network-like structure of Co_{23}B_6 , which requires long-range atomic rearrangements of constituent elements and a large negative entropy, leads to the high stability of the supercooled liquid against crystallization. The details of such transformations during the annealing process, inherently involving a definite sequence of compositional as well as structural ordering processes, are however still rather unclear. Furthermore, the 23:6 phases are not as well understood as other crystallization products, because they exhibit complex face-centered-cubic structures with 116 atoms per conventional unit cell and they are often metastable [6]. It is therefore important to study the underlying structure and microstructure of these phases in order to understand the underpinning of all important properties for the technical application of these materials [4].

Power loss during electrical conversion accounts for a large percentage of the total dissipation (primarily heat) during power transmission and distribution [7]. This is felt both close to the end user, resulting in a decreased efficiency of the ever more ubiquitous consumer-grade power electronic components; but also in the growing share of high-power applications such as power inverters used within photovoltaic installations [8].

The overall power loss can be attributed broadly to three different mechanisms, namely, hysteresis loss, eddy current loss and anomalous loss. It is well understood that as the operating frequency f increases, the eddy and anomalous losses become dominant at $f > 100$ kHz, for metallic, high polarisation materials. On the other hand, recent advances in power conversion circuits have pushed their switching frequencies into the 100s of kHz and even into the megahertz range (in on-chip applications) [9,10]. The utilization of these benefits is more readily attainable using ferrite-based low-moment materials, which are insulating. However, the low flux density typical to ferrites (0.3-0.4 T) requires considerable cross-sectional area (and volume), which is a penalty in many deployments [11]. Thus, to put conducting and insulating core materials on a level playing field, a specific key metric has to be used, say for transformer applications, which is a direct indication of the overall efficiency of the end device. One such choice is the material power loss performance. Presently, ferrites [12], iron powder and tape wound cores [13] are primarily used for relatively low-power applications, where only grams of material are required per installation. An affordable, yet efficient, high-moment magnetic material, allowing for operation beyond 100 kHz, would enable mass application in higher power export-to-grid invertors and power-regulation circuits (in the kW range), which are expected to become prolific in the near future as many countries are adopting new regulations with respect to renewable energy sources.

Anomalous loss dominates the total dynamic loss in most soft magnets with a uniaxial anisotropy oriented longitudinally to the applied magnetic field. It originates from localized eddy currents around moving domain walls under the driving action of an external magnetic field. The anomalous loss can be decreased through changing the mechanism of magnetisation from predominantly domain wall motion and depinning to coherent rotation of domains, which can be done using transverse magnetic annealing. The mechanisms of inducing magnetic anisotropy by magnetic annealing include spatial atomic pair ordering resulting in directional order in the sample, structural relaxation and the rearrangement of free volume in amorphous materials.

The objective of the present paper is to give a more detailed characterization and a deeper understanding of the phase transitions, DC magnetic properties and anomalous loss contribution in transverse field annealed nanocrystalline ribbons. For this purpose, we have investigated the structure and magnetization process in amorphous and nanocrystalline ribbons with different strengths of transverse field-induced anisotropy at frequencies of up to 1 MHz. It is shown that

Co_{23}B_6 crystalizes as the primary phase at 510°C and then Co_2B and Co_3B form by increasing the magnetic annealing temperature to 525°C and 550°C , respectively.

2. Experimental

Melt-spun ribbons of $20\text{ }\mu\text{m}$ thickness and 1 mm width are obtained using fabrication conditions explained in reference [14]. The ribbons are annealed at 5 T magnetic field at different temperatures; 510 , 515 , 525 and $550\text{ }^\circ\text{C}$. The equilibrium phase diagram of the composition is predicted using a thermodynamic modelling software called PandatTM and its 2019 version of Cobalt-based database. X-ray powder diffraction (XRD) data of the melt-spun ribbons are collected using a Phillips PANalytical X'pert Pro diffractometer, with an operational wavelength of $1.5405\text{ }\text{\AA}$ ($\text{Cu K}\alpha$ anode). Rietvel refinement analysis is carried out on the diffraction patterns. For transmission electron microscopy (TEM) analysis, each selected ribbon has been processed using a focused ion beam microscope (FEI Helios Nanolab 600i). A protective metallic (Pt-based) layer has been then deposited on the surface before milling is carried out to create an ultrathin lamella, followed by in-situ micromanipulator transfer to a TEM sample holder. TEM imaging of the ultrathin lamella is performed using a JEOL-2100 TEM operating at 200 kV . In addition, the very-small coercivity and anisotropy field of the ribbons, along its length, is accurately measured by a B-H loop tracer running at 10 Hz with applied in the magnetic field up to 0.2 T . In addition, Evico Magnetics Kerr Microscope & Magnetometer is used for the visualization of magnetic domains. Thermomagnetic measurements are done in a Magneto-Thermo-Gravimetric instrument [15].

The power loss characterization of the ribbons is performed using a custom design air-core solenoid inductor. 150 turns of 28 gauge Cu wire are wound on a 5 cm long hollow cylinder of 5 mm outer radius with near-zero spacing between the turns to create a uniform excitation field inside the solenoid. The impedance of the air core solenoid, Z_a , is measured using an LCR meter (HP4284A) at various excitation current levels. The ribbon samples ($2\text{ cm} \times 1\text{ mm} \times 20\text{ }\mu\text{m}$) are fixed on a sample holder and inserted inside the solenoid, along its axis, where the uniformity of the field is best (Fig. 1). Without moving the probes from the test fixture, the ribbons are centered inside the solenoid, to be at the same distance from both open ends, where the field lines are no longer parallel to the axis. In this way, the magnetic core material is exposed to a uniform

excitation field along its length. The Cu turns are at sufficient distance from the solenoid axis, so that the demagnetisation field does not alter the field lines around the turns maintaining the same AC winding resistance as in the case of air core solenoid measurement. Therefore, the difference between the impedance measurement with and without magnetic core material, Z_c and Z_a respectively, at different frequencies, represents the contribution of the magnetic core to the inductance and power loss at each frequency. As the cross-sectional area of the ribbons is significantly smaller than that of the solenoid, the effect of the insertion of the magnetic core on the real part of the inductance is negligible. Thus, the magnetic material power loss is calculated as:

$$P_{loss} = \text{Re}(Z_c - Z_a)I^2, \quad (1)$$

where $\text{Re}(Z_c - Z_a)$ is the real part of the complex impedance difference and I is the excitation current measured using the LCR meter. The induction field inside the ribbon samples is calculated by

$$B = \frac{\text{Im}(Z_c - Z_a)I}{(2\pi fS)} \pi r^2, \quad (2)$$

where f and S are the measurement frequency and the total cross-sectional area of the ribbons, and r is the radius of the coils. The power loss measurement method has been tested with commercial samples and the results are in good agreement with the reported power loss in standard datasheets.

3. Results and discussions

3.1 Structural characterization

Figure 2 shows the XRD patterns of the ribbons as a function of magnetic annealing temperature up to 550°C. The XRD pattern of the as-quenched ribbons exhibits a broad diffuse peak at an angle of 44.40° which represents the amorphous structure of the ribbons before any heat treatment. Magnetic annealing the amorphous ribbons at 510°C results in the formation of a broad peak, but with a more noticeable tip at 44.78°. TEM studies shows that this observation is due to nucleation of nanocrystals of metastable Co_{23}B_6 phase with cubic crystal structure with the average size of 5 nm in the glassy matrix (Fig. 3a and 4a). Annealing the samples at 515°C shifts the broad peak to 44.79°. By increasing the temperature to 525°C several additional peaks start appearing in the XRD pattern belonging to Co_{23}B_6 and Co_2B , an orthorhombic equilibrium phase. Annealing the samples at 550°C results in an increase in the intensity of peaks corresponding to Co_{23}B_6 and Co_2B

phases. Simultaneously, the appearance of a new orthorhombic equilibrium phase, Co_3B , is suspected as hinted by the presence of overlapped peaks at 41.79° , 44.67° and 48.68° in addition to a low intensity peak at 73.9° . The formation of the as-mentioned phase is confirmed by Rietveld refinement method and the percentage of the phase is estimated as 12% in the sample annealed at 550°C .

To study the crystalization process in more details, TEM micrographs of ribbons annealed at 510°C and 525°C are shown in Figs 3 and 4. The distance between lattice planes in three different directions is equal ($6.057 \text{ \AA}$) in Fig. 4a and corresponds to the $\{111\}$ planes. This lattice spacing is in good agreement with the calculated distance based on the lattice parameter of Co_{23}B_6 phase, estimated from rietveld refinement analysis of XRD patterns. In addition, the refined lattice parameter, $10.54 \text{ \AA}$, is in good agreement with the reported lattice parameter of this metastable phase [5]. In addition to the Co_{23}B_6 phase in the samples annealed at 510°C (Fig. 4a) and 525°C (Fig. 4b), $\{100\}$ planes and $\{110\}$ planes of Co_2B phase are identified in the sample annealed at 525°C (Fig. 4c and 4d). As can be seen in Fig. 4a and b, the size of nanocrystals increases with increasing annealing temperature and nanocrystals with different atomic orientations (Fig. 4 b-d) form at the expense of the amorphous matrix (Fig. 4a). Furthermore, the size of crystallites in both samples; magnetically annealed at 525°C and 550°C , calculated by Rietveld refinement method, is 33.53 and 33.54 nm , respectively, which is in good agreement with the apparent TEM images of sample annealed at 525°C (Fig. 3b and Fig. 4b-d).

Additionally, some thermodynamic and kinetic aspects of these phase transitions are studied further as following. First, the phase transitions are studied by evaluating the equilibrium phase diagram of the system using PandatTM software. Thermodynamic calculation shows that three main phases including Co_2B , Co (hcp) and Co_3B are stable from room temperature to 780°C in this alloy system. However, based on microstructure analysis of magnetically annealed samples (Fig. 2-4), the signs of presence of the metastable Co_{23}B_6 phase can be seen after magnetic annealing at 510°C , then Co_2B is identified in the sample annealed at 525°C and at the end Co_3B forms in the sample annealed at 550°C . Thus, this discrepancy between experiments and thermodynamic modelling should be addressed accordingly.

The nucleation of a metastable phase could be likely kinetic in origin and is kinetically favored compared to that of the stable phase [5]. According to classical nucleation theory [16], the

activation threshold for formation of a critical nucleus is ΔG^* , which is the free energy required to form the critical nucleus;

$$\Delta G^* = \frac{16\pi\sigma^3}{3\Delta G_v^2} f(\Theta), \quad (3)$$

Where $f(\Theta)$ is the catalytic potency factor for heterogeneous nucleation, which depends on the wetting angle Θ . ΔG_v is the Gibbs free energy difference between the liquid and the solid:

$$\Delta G_v = \Delta H_f \frac{T_m - T_{ma}}{T_m}, \quad (4)$$

where ΔH_f , T_m , and T_{ma} denote the enthalpy of fusion, melting temperature, and magnetic annealing temperature, respectively. In addition, the interface energy σ can be estimated by the model developed by Spaepen and Meyer [17]:

$$\sigma = \alpha \frac{\Delta S_f T_{ma}}{(N_L V_m^2)^{1/3}}, \quad (5)$$

where α is a factor dependent on the structure of the solid nucleus and ΔS_f , N_L ; and V_m are the entropy of fusion, Avogadro's number, and molar volume of the phase, respectively. Without the knowledge of the contact angle Θ of the crystal nucleus to the substrate, the nucleation is assumed to be homogenous. Based on this theory, the activation threshold of the stable and metastable possible phases in this system is estimated and shown in Table 1. As can be seen, the necessary activation energy for the formation of Co_{23}B_6 at 510°C is much less than other stable phases. Therefore, from kinetics point of view, the nucleation of this phase is more favorable.

Table 1. Gibbs free energy and interface energy difference between the liquid and solid; ΔG_v and σ , the activation threshold for formation of a critical nucleus ΔG^* at 510°C.

Phase	ΔG_v (kJ/mol)	σ (J/m ²) $\times 10^5$	ΔG^* (J) $\times 10^{23}$	ΔG^* (meV)
Co₂₃B₆	155	3.71	3.55	0.355
Co₂B	7.95	1.62	114	11.4
Co	9.03	1.12	29.2	2.92
Co₃B	29.4	5.4	304.8	30.48

3.2. DC magnetic properties

Transverse field annealing changes the magnetisation easy axis by inducing transverse anisotropy. This effect can be noticed by comparing the preferred orientation of domain in MOKE images of as-quenched and magnetic annealed samples at 510 and 525°C (Fig. 5). It can be seen that in as-quenched ribbons the easy axis is along the length of the ribbon because of shape anisotropy (Fig. 5a). However, transverse magnetic annealing at 510°C shifts the easy axis to the width of the ribbons (Fig. 5b) and also cause limited crystalization of Co₂₃B₆ in the amorphous matrix (Fig. 4a). The type and the number of nucleated nanocrystals increase after magnetic annealing at 525 °C (Fig. 2-4), resulting in a more complex microstructure and magnetic structure (Fig. 5c). Simultaneously, the domain structure changes from wide domains (Fig 5a and 5b) to a pattern of small, irregular domains (Fig. 5c).

The evolution of hysteresis loops measured along the length of the ribbons as a function of magnetic annealing temperature is shown in Fig. 6. The shape of hysteresis loops by increasing magnetic annealing temperature is becoming more flat which is due to the effect of transverse field-induced anisotropy on different magnetic parameters which will be addressed accordingly. First, the as-mentioned effect can be measured quantitatively by the induced anisotropy energy (K_u) (Fig. 7) which is calculated based on [18]:

$$K_u = \frac{H_k \mu_0 M_s}{2} \quad (6)$$

where μ_0 is the free space permeability and M_s is magnetization saturation. The anisotropy enhancement is related to directional atomic ordering along the direction of the local magnetization in order to minimize the spin orbit coupling energy [12]. Since the directional ordering of solute atoms is induced by the local spontaneous magnetization, during heating of the sample, K_u could only be induced when the field annealing temperature is lower than T_C . Thus, a crucial factor influencing the field-induced K_u in the multiple-phase microstructure of nanocrystalline soft magnetic alloys is the Curie temperature (T_C) of the phases. Based on this theory, thermomagnetic measurements are carried out on different samples as shown in Fig. 7. As can be seen, Curie temperature of the amorphous phase is approximately 310°C, while the Curie temperature of Co_{23}B_6 is more than 610°C. It is worth mentioning that the optimum annealing temperature, for realizing the ‘magnetically softest’ microstructure, in nanocrystalline soft magnetic alloys often lies in a range between the Curie points of the two constituent phases [19], so the Co-based ribbons in this study are magnetically annealed at temperatures between these two Curie points.

Additionally, the effect of the microstructural evolution of samples during magnetic annealing can be observed as in magnetization change continuous temperature scans (Fig. 7). First, due to single-phase amorphous structure of as-quenched ribbon, just one phase with a Curie temperature of 310°C can be observed. However, in magnetically annealed samples at 510, 515 and 525°C, in addition to the amorphous matrix phase, another phase with the Curie point of 610°C, which is possibly related to Co_{23}B_6 , can be distinguished. In addition, the decreasing portion of amorphous phase by enhancing the magnetic annealing temperature (Fig. 2-4) can be ascribed to the area under the M-T curve in the amorphous region. In other words, at point * in Fig. 7, M-T curves of magnetic annealed samples at 525, 515 and 510°C diverge, respectively, indicating that the portion of amorphous phase is lower in the samples annealed at higher temperatures.

The metastable equilibrium between Co_{23}B_6 as the primary bcc phase, and the residual amorphous phases is maintained in the nanocrystalline soft magnetic alloy. An obvious consequence of this two-phase microstructure is that the bulk field-induced K_u of the sample reflects the volume-weighted average of the two local K_u contributions from the two constituent phases. Thus, as the

annealing temperature is less than Curie temperature of Co_{23}B_6 phase and higher than Curie temperature of the amorphous phase, the anisotropy induced by a magnetic field applied during nanocrystallization (Fig. 2-4) primarily originates from the Co_{23}B_6 grains, which results in an abrupt increase in K_u after magnetic annealing at 510 °C (Fig. 5).

The average anisotropy constant $\langle K \rangle$ of a coupled multiphase system with anisotropies randomly oriented on a scale smaller than a magnetic correlation length L_{ex} can be described by [20]:

$$\langle K \rangle = \sqrt{K_u^2 + \sum_v x_v \beta_v^2 K_{1,v}^2 (D_v/L_{ex})^3}, \quad (7)$$

where K_u denotes a uniaxial anisotropy, which is uniform on a scale much larger than L_{ex} . The random contributions are represented by the local anisotropy constants $K_{1,v}$, the grain sizes D_v and the volume fractions x_v of the individual structural phases labelled by the index v . The parameters β_v mainly involve conventions used to define the anisotropy constants for different symmetries. Therefore, two different parameters lead to an increase in $\langle K \rangle$. First, increasing K_u as a result of higher temperature annealing (Fig. 8) and nano-crystallization of the sample (Fig. 2-4), which results in contribution of the local anisotropy constant of the crystalline phases to the average anisotropy constant, eq. 7. Thus, the local density of induced anisotropy in the residual amorphous phase becomes negligible or considerably smaller than that in the primary bcc phase and the anisotropy induced by annealing at $T_{C,amorph} < T_{ma} < T_{C,Co_{23}B_6}$, in a magnetic field applied during nanocrystallization, mainly due to the bcc grains of Co_{23}B_6 phase.

The subsequent decrease and then increase in K_u with increasing annealing temperature to 525 and 550°C can be related to the formation of octahedral crystals of Co_2B and specially superlattice structure of orthorhombic Co_3B in annealed sample at 550°C. The induction of an extra magnetic anisotropy in a crystal by the directional ordering of solute atoms is due to the change in the crystal symmetry. This means that the effectiveness of the solute atoms on the induction of K_u depends on the type of solid solution and its crystalline system. This effect can be seen also in the completely different shape of the hysteresis loop of the sample annealed at 550°C and the high coercivity of this sample (Figure 6) which is due to limited formation of Co_3B as a semi-hard magnetic phase with the anisotropy energy of 507 kJ/m³ [21]. In other words, the almost perfectly rectangular or flat hysteresis loops after field annealing at 510, 515 and 525°C indicate that field-

induced anisotropy clearly dominates over the residual contributions from the random magneto-crystalline anisotropies. On the contrary, domination of extrinsic magnetocrystalline anisotropy over field-induced anisotropy is noticeable in the sample which is magnetically annealed at 550°C, and is due to strong magneto-crystalline anisotropy of Co₃B and its higher relative concentration.

Therefore, although the grain size of the Co₂₃B₆ in the ribbons annealed at 525 and 550 °C remains almost unchanged and based on random anisotropy model [20], it is expected that ferromagnetic exchange interaction between these small grains force the magnetic moments more and more to align parallel, simultaneously precipitation of semi-hard Co₃B phase can significantly deteriorate the soft magnetic properties in the sample annealed at 550 °C. Thus, a very small volume fraction of a phase with high anisotropy can change the magnetic behaviour of the whole sample.

Transverse magnetic annealing at a temperature of 510°C results in rather a substantial decrease in permeability (Fig. 8). However, annealing at higher temperatures does not further decrement the permeability. In particular, permeability depends sensitively on the angle between the applied magnetic field and macroscopic anisotropy direction. As the samples are magnetized along their length, during DC magnetic measurements, and this direction is perpendicular to the induced K_u axis, the permeability is determined by magnetization rotation and, hence, is inversely proportional to the induced anisotropy energy [22] (Fig. 8) :

$$\mu = \frac{J_s^2}{2\mu_0 K_u}, \quad (8)$$

where J_s is the average saturation polarization of the material. Figure 9 shows the change in the coercivity of samples as a function of magnetic annealing temperature. The coercivity of samples starts from 2.9 A/m for as-quenched ribbons and ends at 355 A/m for ribbons magnetically annealed at 550°C (Fig. 6 and 9). Limited crystallization of Co₂₃B₆ phase in the sample annealed at 510°C (Fig. 4a and 4b) results in increased coercivity due to magneto-crystalline anisotropy. In addition, coercivity is directly related to the average anisotropy constant $\langle K \rangle$ by [22]:

$$H_c = p_c \frac{\langle K \rangle}{J_s}, \quad (9)$$

where p_c and p_μ are dimensionless pre-factors of the order of unity and μ_0 is the vacuum permeability. Thus, by increasing the average anisotropy constant, eq. 7, H_c of the ribbons increases (Fig. 9). Furthermore, the substantial increase in the coercivity of magnetic annealed sample at 550°C compared to the one magnetically annealed at 525°C can be due to the formation of Co_3B as a semi-hard magnetic phase.

Figure 9 also shows the change in the saturation magnetization of ribbons versus magnetic annealing temperature. First, there is an increase from 0.84 to more than 1 T after magnetic annealing the amorphous ribbons at 510°C. However, further increase in magnetic annealing temperature results in an abrupt decrease in saturation magnetization of the samples. The evolution of saturation magnetization can be evaluated by the magnitude of magnetic moment of formed phases during magnetic annealing. The decrement about 40 % is easily accounted for by the amount of crystalline phases as evidenced by Fig. 3b.

First, as Cobalt atoms occupy four different types of special positions in the cubic Co_{23}B_6 structure with unique symmetries and neighboring atomic configurations, an effective localized moment can be attributed to each type of site. Ohodnicki et al. [6] showed that in Co_{23}B_6 structure, the largest local moments are observed at the 4a and 8c sites. As has been pointed out previously in theoretical calculations for the $(\text{Fe,Co})_{23}\text{B}_6$ structures [23], large local moments are observed for some of the transition-metal atoms at these sites (Fe at 4b sites = $2.64 \mu_B$, Fe at 8c sites = $2.60 \mu_B$, Co at 8c sites = $1.91 \mu_B$ relative to those calculated for pure bcc Fe = $2.17 \mu_B$ and fcc Co = $1.60 \mu_B$). Thus, larger local moments of Cobalt atoms in Co_{23}B_6 atomic structure compared to Cobalt atoms in pure Cobalt results in an increase in M_s of magnetic annealed sample at 510°C compared to as-quenched sample. Further magnetic annealing results in the formation of Co_2B phase with a magnetic moment of $0.76 \mu_{B/f.a.}$ leading to substantial decrease in M_s of magnetically annealed samples (Fig. 9).

3.3. AC magnetic properties

The total power loss of different samples measured at three different frequencies are decomposed into three different power loss mechanisms; hysteresis, eddy current and anomalous, based on the analysis borrowed from reference [11]. As the latest has the main contribution to the total power loss, more than 90% [24–26], and magnetic annealing is to decrease this main mechanism of power loss, the trend of this type of power loss is investigated in more details as follows. Fig. 9 represents anomalous loss behavior of different samples as a function of magnetic induction at different frequencies; 50 kHz, 500 kHz and 1 MHz. As mentioned before, the anomalous loss is dominant and its frequency dispersion is in a good agreement with the total power loss behavior of these samples.

The anomalous loss of nanocrystalline ribbons is higher than as-quenched ribbons at 50 kHz (Fig. 9a). However, upon increasing the frequency to 500 kHz, the performance of $T_{ma} = 525^\circ\text{C}$ sample is better than as-quenched sample, which may be due to the fact that the contribution of anomalous loss is more significant at higher frequencies. Magnetic annealing as a method of decreasing this kind of power loss, becomes even more effective. Thus, in addition to $T_{ma} = 525^\circ\text{C}$ sample, nanocrystalline ribbons annealed at 525°C also show lower anomalous loss compared to the as-quenched sample at 1 MHz. As a case in point, magnetic annealing at 525°C decreases the anomalous loss of as-quenched ribbons by 70%, at 1 MHz and 5 mT. Although more anisotropy is induced in the samples with $T_{ma} = 510$ and 515°C , it seems that sufficient amount of anisotropy energy is induced in the sample magnetically annealed at 525°C . At higher temperatures, the crystallization of secondary phase leads to a worse compromise between permeability and power loss.

Additionally, according to Herzer's theory [27], the relationship between total power loss (P_t) and the total eddy current loss (P_{ec}), which includes classic and excess eddy current loss has the form of:

$$P_t \approx P_{ec} \left[1 + \frac{w^2}{(w \cos \beta + t)^2} \frac{m^2}{1 - m^2} \right], \quad (10)$$

where w denotes the average domain width, t is the ribbon thickness, β is the out-of-plane angle of the magnetization vector and m denotes the average magnetisation component along the ribbon axis normalised to the saturation magnetisation. First, by changing the mechanism of

magnetization from domain wall motion to domain rotation, the movement of domain walls is limited substantially resulting in lower excess eddy current loss or anomalous loss (Fig. 10b and c). Finally, the demagnetization energy is lower in samples with narrower domain width (w) (Fig. 5) and as can be concluded from eq. 5, the total power loss decreases in such samples due to the prohibited domain propagation. Thus, different mechanisms are effective on decrement of total power loss during transverse magnetic annealing.

4. Conclusions

In this work we develop a better understanding of nanocrystallization, phase transformation and soft magnetic behavior of Co-based melt-spun ribbons through magnetic annealing at temperatures between the Curie points of the amorphous and the main nanocrystalline phase. XRD and TEM are used to investigate the structure of the ribbons magnetically annealed at different temperatures. In addition, structural features are correlated with magnetic properties measured by Kerr microscopy, magnetization and thermomagnetic measurements. Magnetic annealing at 510°C results in the formation of metastable Co_{23}B_6 phase which has not been anticipated by thermodynamic models, but further due to kinetics. Thus Co_{23}B_6 nucleates at lower temperatures compared to Co_2B and Co_3B phases, owing to its lower nucleation activation energy. The almost perfectly rectangular hysteresis loops after field annealing at 510, 515 and 525°C indicate that field-induced anisotropy dominates over the residual contributions from magneto-crystalline anisotropy. However, domination of magnetocrystalline anisotropy over field-induced anisotropy is noticeable in the samples which are magnetically annealed at 550°C due to formation of Co_3B , a semi-hard magnetic phase. The anomalous loss, as the main mechanism of power loss can be reduced by 70 % by annealing at 525°C in a transverse magnetic field, which results in the promotion of relative contribution of domain rotation over domain wall motion. Another contributing factor is the decrement of domain width by increasing the magnetic annealing temperature and it contributes to further decrease in anomalous loss by promoting pinning of domain walls. Therefore, nanocrystalline Co-based systems offer an interesting engineering tradeoff between the extreme properties of amorphous and fully crystalline soft magnets.

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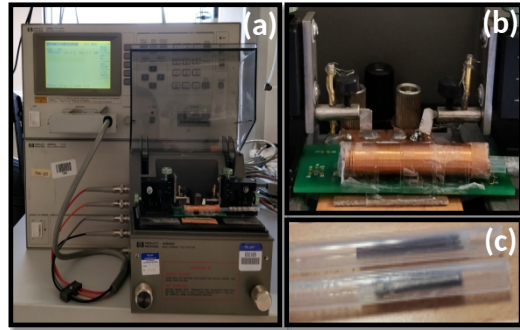


Figure 1. Power loss measurement set-up; (a) complete set-up, (b) solenoid with 150 turns of wire of guage 28 to produce coherent magnetic field at the center, and (c) sample holders.

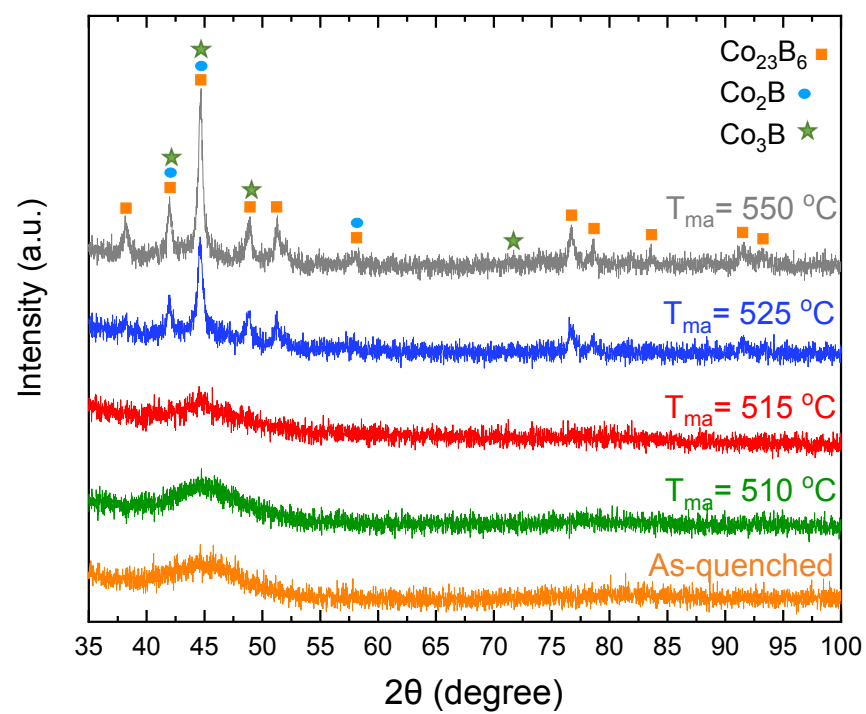


Figure 2. XRD patterns of as-quenched and magnetic annealed samples at 510, 515, 525 and 550°C.

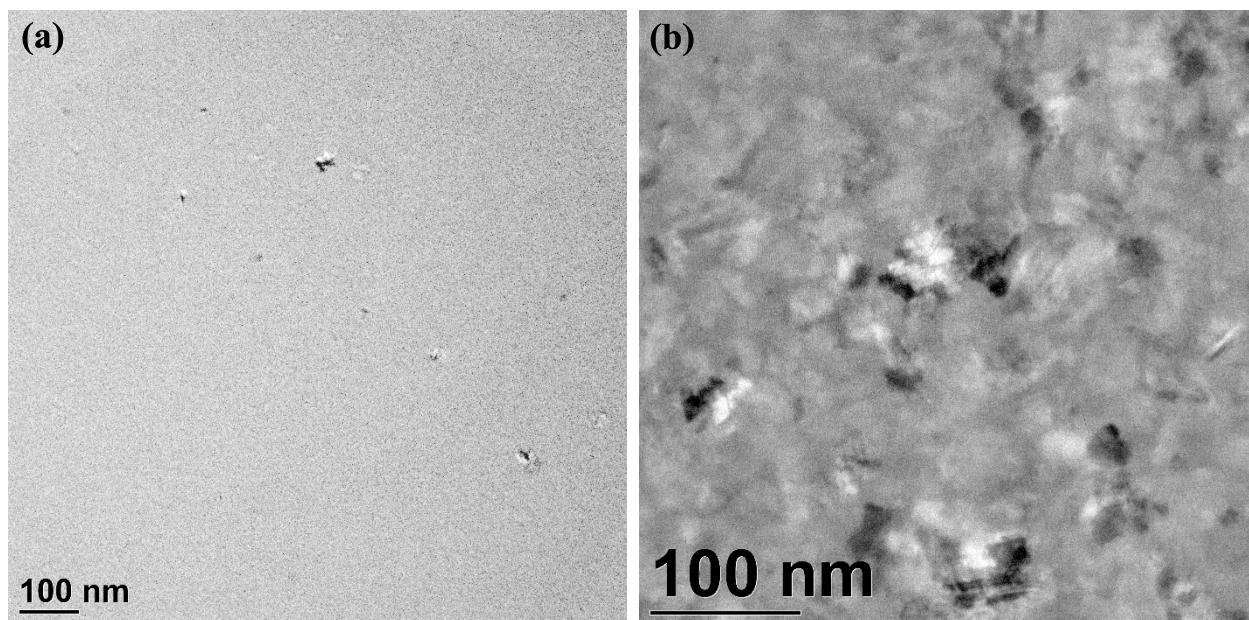


Figure 3. TEM micrographs of magnetic annealed samples at:(a) 510°C and (b) 525°C.

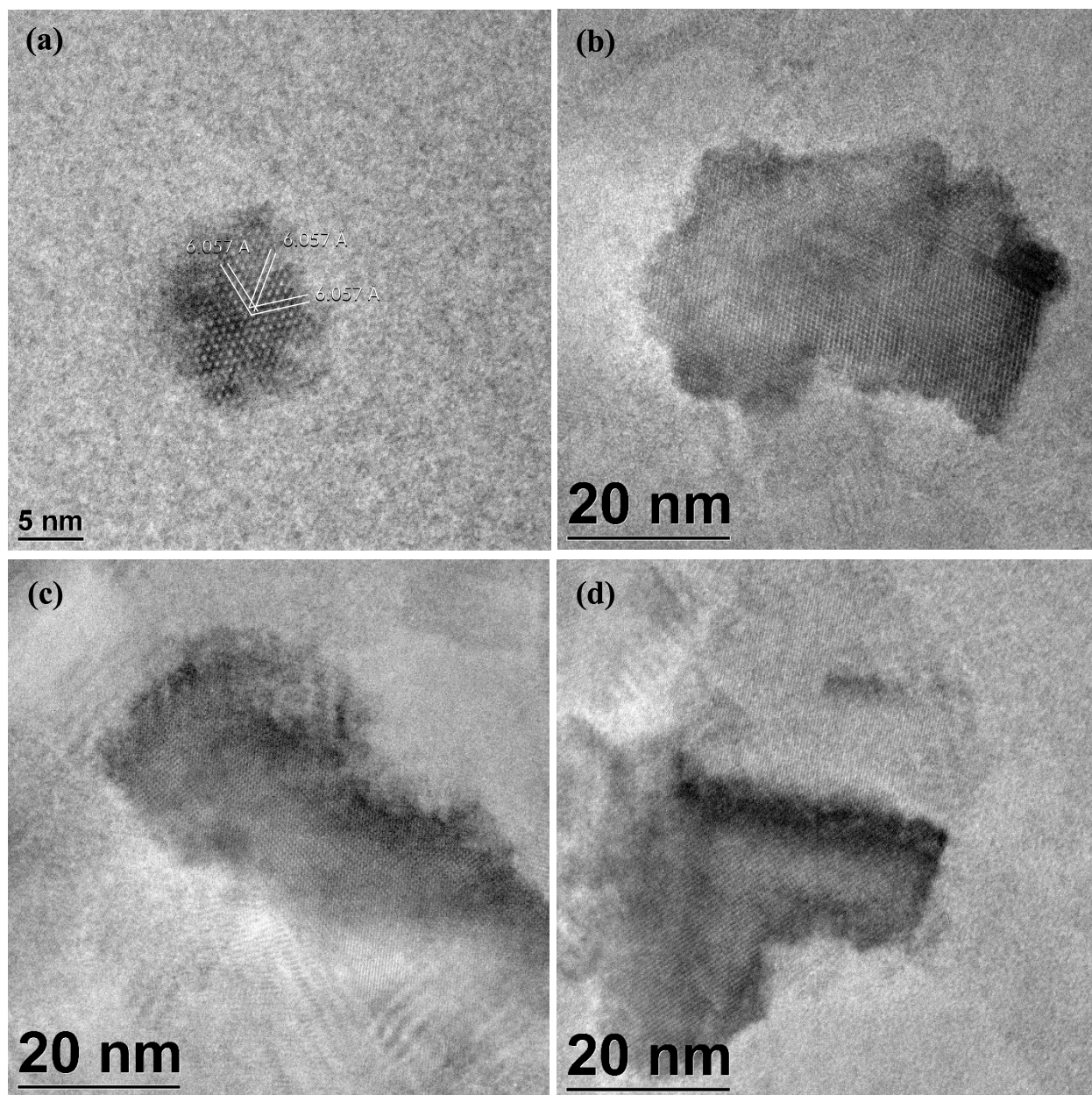


Figure 4. TEM micrographs of {111} planes in Co_{23}B_6 in magnetic annealed samples at: (a) 510°C and (b) 525°C; and (c) {100} and (d) {110} planes of Co_2B in magnetic annealed sample at 525°C.

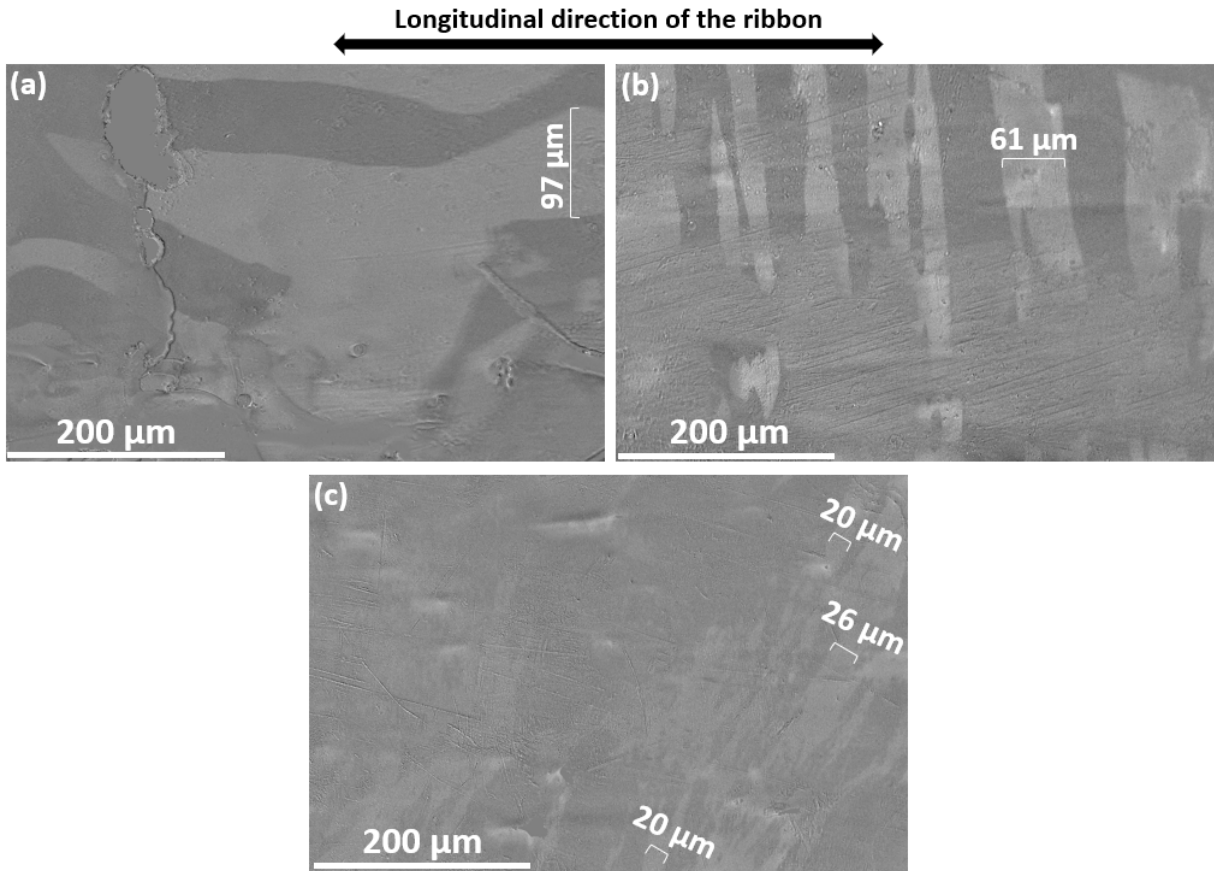


Figure 5. Domain images during the magnetization of the (a) as-quenched, magnetically annealed at (a) 510 and (b) 525 $^{\circ}\text{C}$ ribbons at $\mu_0 H = 0$ T.

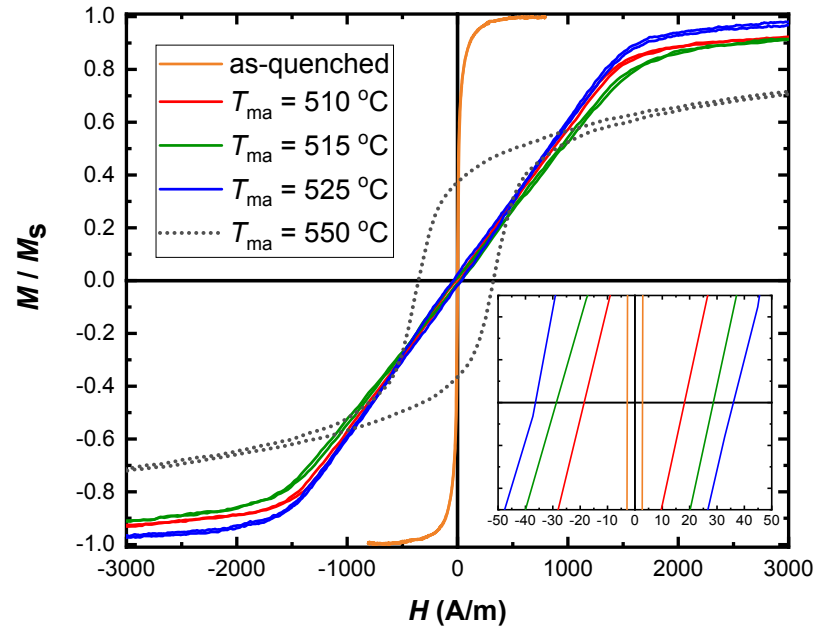


Figure 6. Hysteresis loops of as-quenched and magnetic annealed ribbons at 510, 515, 525 and 550°C.

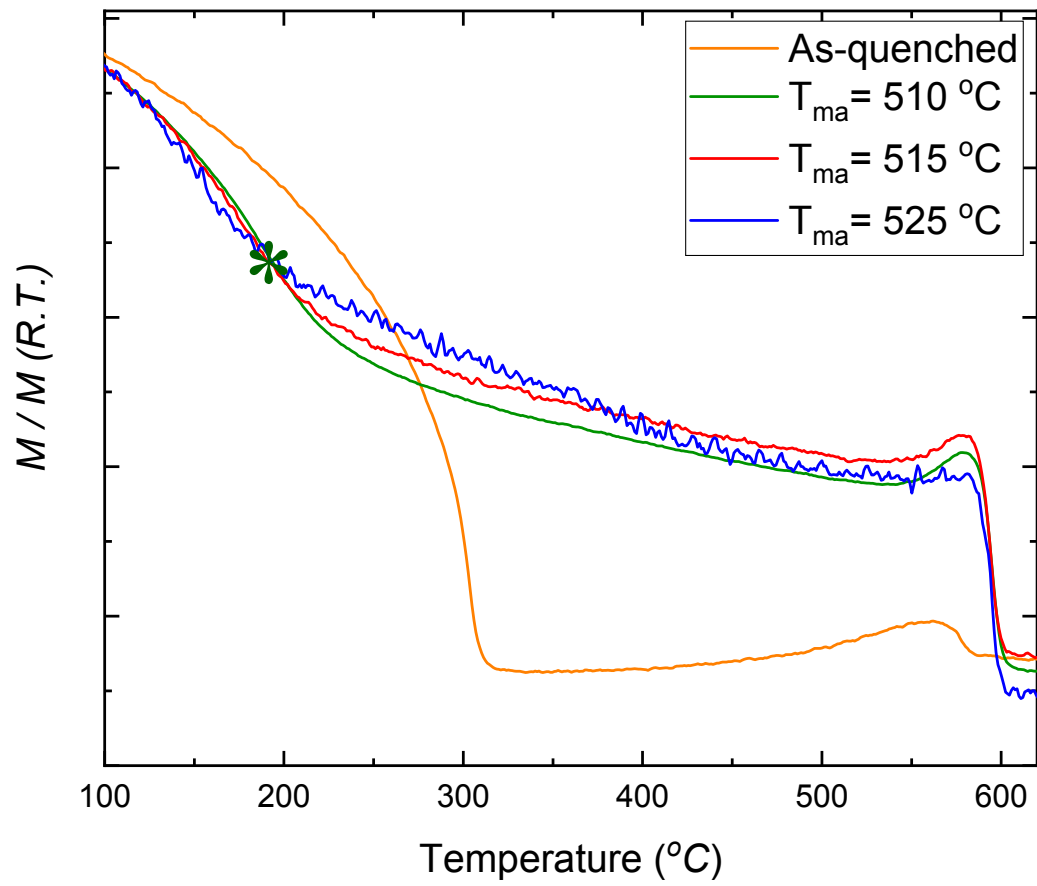


Figure 7. Temperature dependence of normalized magnetization for as-quenched and magnetically annealed samples.

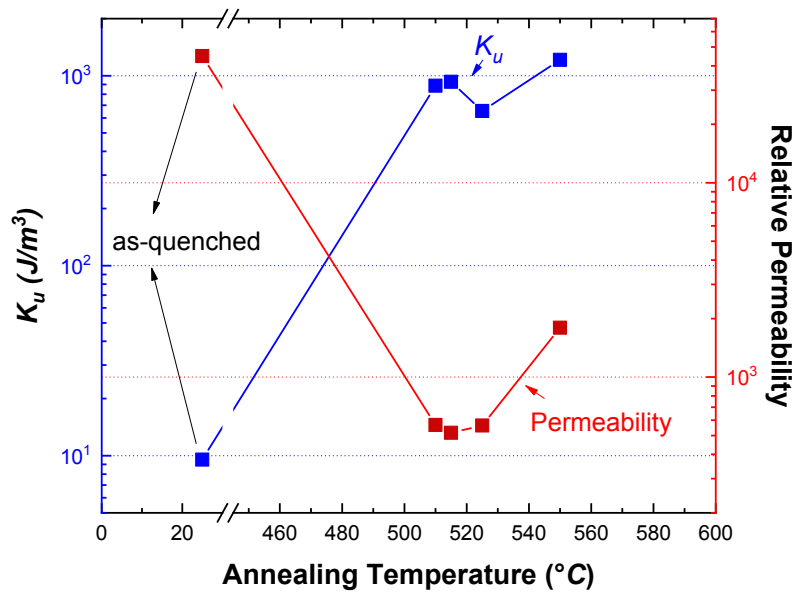


Figure 8. Induced anisotropy energy and relative permeability of magnetic annealed ribbons as a function of annealing temperature (the size of the symbols represents the measurements uncertainty).

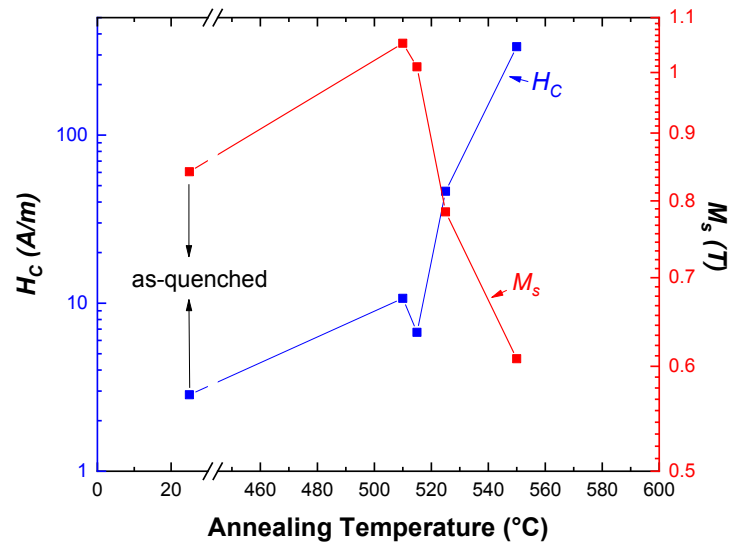


Figure 9. Coercivity and saturation magnetization of magnetic annealed ribbons versus magnetic annealing temperature.

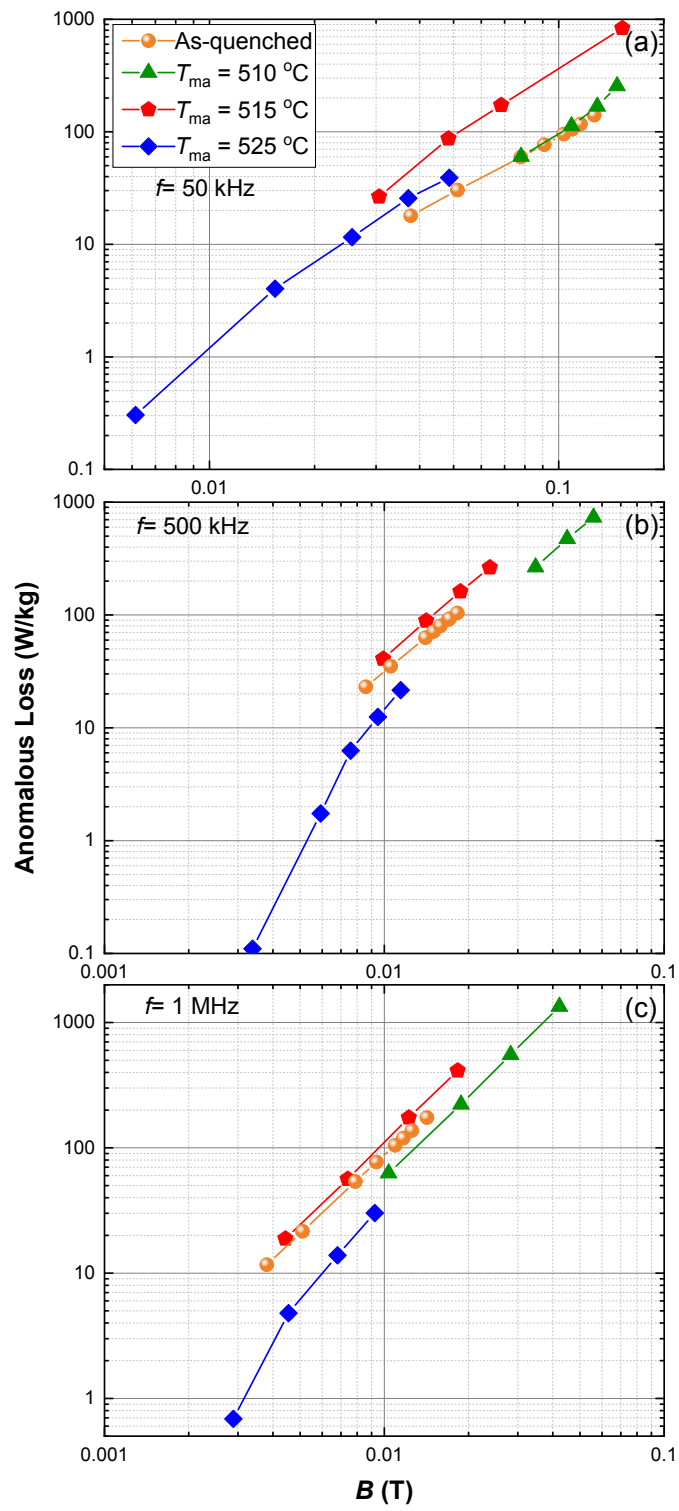


Figure 10. Anomalous loss of as-quenched and magnetic annealed ribbons as a function of magnetic induction at (a) 50 kHz, (b) 500 kHz and (c) 1 MHz.

