Relating residual stress and microstructure to mechanical and
giant magneto-impedance properties in cold-drawn
Co-based amorphous microwires

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Received 2 June 2012; received in revised form 19 June 2012; accepted 22 June 2012
Available online 10 August 2012

Abstract

The effect of cold-drawing on the tensile property and giant magneto-impedance (GMI) effect of melt-extracted Co-based amorphous microwires was evaluated through detailed analyses of the distribution of residual stress and microstructural evolution. The tensile ductility and tensile strength increased gradually with cross-sectional area reduction ratio \((R)\) until 51\%, and decreased with further deformation. The microwire with \(R = 51\%\) exhibits the highest tensile ductility of 1.09\% and tensile strength of 4320 MPa. Structural and thermodynamic analyses reveal that it is the mechanical deformation rather than thermal activation that induces the precipitation of nanocrystals and arrests the quick extension of shear bands leading to the enhanced ductility. Interestingly, the GMI effect also attains the maximum value of 160\% at 10 MHz when \(R = 51\%\) (30\% larger than that of the as-cast wires), before decreasing with further cold-drawing. Such an identical evolution trend of both tensile and GMI properties can be ascribed to two underlying mechanisms: the generation of longitudinal and circumferential residual stresses and the growth of deformation-induced nanocrystals during cold-drawing. The role of residual stress is established herein not only as a trigger to accelerate the amorphous-to-nanocrystalline phase transformation but also as a decisive contributor to the mechanical and GMI performance. The unique simultaneous improvement of both mechanical and GMI properties of cold-drawn Co-based microwires opens up new possibilities for a variety of engineering applications, such as high-performance magnetic, stress and biological sensors.

Keywords: Amorphous microwire; Melt extraction; Cold drawing; Mechanical property; Giant magneto-impedance

1. Introduction

Amorphous alloys, which have no crystalline structure, often yield a superior elastic limit, high fracture strength and excellent magnetic properties, and are promising materials to meet the increasing demand of structure–functional integration materials [1–6]. However, their use in engineering applications is limited by their inferior plastic deformation, which is typically concentrated in extremely thin (10–20 nm) shear bands, resulting in a catastrophic failure [7]. A variety of approaches capable of retarding the sudden propagation of shear bands, such as stress or chemical inhomogeneity, phase separation at the micro- or nano-scale, and synthetic composites, have been proposed to enhance the ductility of amorphous alloys [8–10]. For instance, a new Ti-based metallic-glass-matrix composite, which includes the ductile dendrites within the glass matrix, demonstrates a significantly improved tensile ductility [10]. “Work-hardenable” ductile bulk metallic glass with a ductility of 18\%
was obtained by introducing an atomic-scale inhomogeneity to induce multiplication of shear bands [11].

In another perspective, it is well established that the deformation behavior of the amorphous alloys exhibits an obvious size effect: typically the smaller the specimen, the softer it behaves [12]. Especially, when the sample size is below micro- or nano-scale, there is a significant uniform elongation and/or extensive necking in the unconstrained condition [13–17]. Thus, it predicts that the introduction of nano- or micro-scale inhomogeneities into a fine-sized amorphous sample will potentially lead to a remarkable improvement of its mechanical performance. Such a nanocrystal-embedded microstructure was reported recently by Sergueeva and Branagan in glass-covered Fe-based amorphous microwire through the spinodal decomposition method, yielding a tensile ductility of 8% and a tensile strength up to 4.8 GPa [18–19].

Compared with other conventional composition adjustments and heat-activated treatment of amorphous alloys, mechanical deformation can introduce inhomogeneous features, e.g., it can soften and harden regions or result in microstructural change of the amorphous matrix, for the benefit of its mechanical characteristics [20,21]. The different hardness regions were achieved in a Zr-based amorphous alloy by channel-die compression (CDC), resulting in an increase in compressive deformation from 0.2% for the as-cast sample to ~4% for the CDC material [20]. A much larger improvement in plastic deformation of ZrTiCuNiBe bulk metallic glasses was achieved from 0.5% up to 15% due to the introduction of microstructure inhomogeneities upon cold-rolling at room temperature [21]. Similar results have also been reported in lateral pre-compression [22], or imprinting processes [23] to limit the fast propagation of the main shear band, generating a high compressive or tensile ductility of monolithic bulk metallic glasses. Among those mechanical processes, the conventional cold-drawing process has generated much interest due to its simplicity and manageable stress level via area reduction ratio and thus preferred for the pre-mechanical treatment of small-sized symmetric amorphous microwires. It has been reported that cold-drawn wire exhibited a fracture stress that is 5–8% larger than that of undrawn wire with a plastic strain of 0.7% [24], which is accounted for by the interactions between numerous shear bands in pre-deformed areas. Wu et al. [25] reported an improvement of tensile fracture strength of cold-drawn Co-based in-rotating-water spinning wire by removing surface flaws and generating compressive residual stress, which also led to improved fracture reliability. Their recent work on cold-drawing of Co-based melt-extracted amorphous wires revealed a nanocrystallization behavior at ambient temperature that led to a larger tensile ductility of 1.64% accompanied with a high tensile strength exceeding 4000 MPa [26].

In addition to the structural applications relying on its mechanical properties, the amorphous magnetic microwire also has distinctively positioned itself as an important sensing element for a variety of sensor applications, capitalizing on its singular giant magneto-impedance (GMI) effect. By definition, the GMI effect refers to a large change in impedance for a conductor carrying AC current subjected to an external DC magnetic field [27], which is attributed to the skin effect compounded with a dynamical magnetization under DC magnetic fields. In the case of microwires, it is acknowledged that they typically possess a unique bamboo domain structure consisting of an outer shell with circumferential anisotropy and an inner core with axial anisotropy [28]. It is the multiple interactions of the outer shell domains of the wire with the circular field generated by the high-frequency current and applied DC magnetic field that are responsible for the GMI performance. As the microwire is amorphous, its exact configuration of domain structure is determined by the magnetoelastic anisotropy arising from the coupling of negative magnetostriction and residual stresses formed during the fabrication process and post-processing. It is therefore crucial to understand and further to manipulate the residual stress in terms of magnitude, orientation and distribution in order to modulate a desirable GMI effect for targeted sensor applications. The cold-drawing process offers such an ideal way to manipulate the residual stresses and hence the magnetoelastic energy, conditioning suitable configuration of the magnetic domains structure to enhance the GMI properties of amorphous wires. Among the few studies dedicated to the GMI effect of cold-drawn wires [29,30], a large improvement of GMI ratio from 22% to 90% was achieved via cold-drawing amorphous wires produced by the in-rotating-water rapid quenching technique, which can be attributed to the induced magnetoelastic anisotropy in the surface layer of the wire [29]. Hu et al. [30] argued that there existed a transformation of the domain structure from longitudinal to the circular in “hard-drawn” wires, leading to the improvement of its GMI properties.

These works either focus on the residual stress analyses in the as-cast state [31,32] or GMI performance of cold-drawn wires [29,30,33]; however, a clear correlation between the cold-drawn induced stress state and the GMI effect remains unproven. To the best of our knowledge, there is no publication that considers the effect of cold-drawn induced nanocrystals on the GMI characteristics, let alone alongside the effect of residual stresses. In this context, the present work appears to be the first comprehensive study on the influence of cold-drawing on the composition, microstructural evolution, tensile and magnetic properties of the melt-extracted amorphous microwires, and provides an in-depth analysis of the role of cold-drawing-induced residual stresses and nanocrystallization on their mechanical and GMI properties. The results obtained make this conventional drawing technique and the use of processed microwires for sensing applications extremely promising.

2. Experimental details

The master ingots with a normal composition of Co_{68.15}Fe_{4.35}Si_{12.25}B_{15.25} were prepared by arc-melting pure
Co (99.99%), Fe (99.9%), Si (99.99%), and B (99.7%) elements in an argon atmosphere and sucking into an 8 mm in diameter copper mould for further extraction processing. The continuous near-circular CoFeSiB wire with a diameter of \( \sim 60 \mu m \) was fabricated by a modified melt extraction method, as schematically shown in Fig. 1a [34]. The diameter of the extracted wires was reduced step by step through a number of drawing processes using diamond dies without any intermitting annealing (Fig. 1b). The cross-section area reduction ratio, \( R = (D_0^2 - D_1^2)/D_0^2 \), where \( D_1 \) is the diameter after drawing and \( D_0 \) is the original diameter, was controlled within 4% per step.

The amorphous state of the cold-drawn samples with different \( R \) values was identified by a D/max-rB 12KW X-ray diffractometer (XRD) with Cu K\( \alpha \) radiation. Micro-tension experiments of wires with different area reductions were performed using a 10 N Instron 3343 universal testing machine at a constant strain rate of \( 4.2 \times 10^{-4} \) s\(^{-1} \). Tensile samples were prepared following the ASTM standard D3379-75 with a gauge length of 10 mm. The geometry of the extracted wires as well as fracture morphology was studied by scanning electron microscopy (SEM, Quanta 200FEG). Transmission electron microscopy (TEM) specimens were prepared by ion milling at an ion-energy of 4.5 keV and the microstructural change was examined using a high-resolution TEM Tecnai G\(^2\) F30, equipped with a high-angle annular detector dark-field scanning transmission electron microscope (HAADF-STEM).

The magnetic impedance for the amorphous wire with different \( R \) values was measured with an HP 4192A precision impedance analyzer in the frequency range of 0.1–20 MHz. The impedance \( (Z) \) was measured by the four-point method in which a given AC current flows along the magnetic microwire and a voltage is picked up at its ends. A detailed description of the measurement facility and process can be found elsewhere [35]. The obtained GMI ratio \( \Delta Z/Z(\%) \) is defined as [36]:

\[
\Delta Z/Z(\%) = \frac{Z_{\text{(H)}} - Z_{\text{(max)}}}{Z_{\text{(max)}}} \times 100\%
\]

where \( Z_{\text{(H)}} \) and \( Z_{\text{(max)}} \) are the impedance magnitudes of the microwire in the measured external magnetic field and maximum measurement magnetic field, respectively.

3. Results

3.1. Drawing effects on the macroscopic properties

Co\(_{68.15}\)Fe\(_{4.35}\)Si\(_{12.25}\)B\(_{15.25}\) metallic wires were easily drawn to a cross-sectional area reduction of \( \sim 75\% \) without rupture. The outer surface of the cold-drawn wire with different \( R \) values is shown in Fig. 2a. All of the drawing wires exhibit smooth surfaces without any visible scratches, while the grooves and fluctuations in the as-quenched wire, as indicated by the arrows, can be seen occasionally on the surface. Note that the existence of these flaws results in a severe deterioration of their mechanical properties. After the drawing process, it can be seen that the grooves and flaws were removed; wires became more circular and round. Fig. 2b presents X-ray diffraction patterns of the as-quenched and cold-drawn samples. Most of these samples exhibit consistent feature, i.e., a broad diffuse halo except for a small crystal peak overshadowed by the background noise for the 64\% drawn wires, indicating that cold-drawing is an effective way to reduce the diameter of the melt-extracted microwire without changing its macro-scale structure constitute. This is especially important for the sensing application of wires, since they prefer to be thinned down to a fine diameter in meeting the requirement of miniaturization but without compromising their mechanical integrity. It should be noted that the conventional XRD technique is unable to detect phases with a content of less than \( \sim 5 \) wt.\% (depending on crystal symmetry); the microscale structural change (if any) induced during
cold-drawing process needs to be further clarified by electron microscopy.

Compositional inhomogeneity generated during different quenching experiences can also alter the mechanical performance to a great deal. Fig. 3 shows the chemical composition profile of the as-cast and \( R = 51\% \) wires; the B element is not listed here because of its small atomic number. Horizontal axis values refer to the distance from the concave to the free surface for the as-cast and the diameter of the drawn wire, as illustrated in the inset of Fig. 3. For the as-cast wire, there exists obvious element segregation from the copper-connected concave to the free surface, depending on the different cooling rate. The Co content is richer near the copper-connected surface and decreases steadily towards the free surface; Si content increases slowly to the free surface while Fe concentration remains almost constant. For the drawn wire, meanwhile, all these elements are distributed uniformly throughout the whole cross-section. Therefore, cold-drawing can effectively reduce the composition segregation and improve macro-scale chemistry homogenization. As demonstrated by Liu et al. [37], the respective changes of hardness and elastic modulus of \( \sim 0.15 \) GPa and 10 GPa, respectively, were reported for \( \text{Zr}_{50}\text{Cu}_{50} \) bulk metallic glass along the radial direction; this was caused by the introduction of excess frozen-in defects resulting from the different cooling rate.

### 3.2. Tensile property and fracture morphology

The effect of cold-drawing on the mechanical performance of melt-extracted amorphous wires was investigated and the representative tensile stress–strain curves for microwires with different \( R \) values are presented in Fig. 4. The cold-drawn wires exhibit noticeable tensile plasticity as compared with the as-quenched sample (a). The tensile strain as well as tensile strength increase gradually with cross-section reduction until \( R = 51\% \) and then decrease with further deformation. Cold-drawn microwires with \( R = 51\% \) exhibit the highest tensile strength of 4320 MPa and a maximum tensile ductility of 1.09\% that is a slightly lower than our previous reported value (1.64\%), measured using a dynamic mechanical analyzer [26]. It should be noted that the curves in Fig. 4a–c possess similar elastic
modulus, as calculated from the slope of these curves, while the modulus become slightly larger for samples in Fig. 4d–f, indicating somewhat different structures of these two groups of wires.

To verify the wire’s usefulness as a fine engineering material, it is necessary to evaluate its mechanical properties in the context of prevalent materials of the same scale. The engineering stress–strain curve of the present 51% cold-drawn wire together with a series of Zr-based [38,39], Pd-based [24,40], Fe-based [41], Mg-based [42], Ni-based [43,44] and Co-based amorphous microwires [45] are compiled and presented in Fig 5a. It can be clearly seen that, compared with other microwires fabricated by in-rotating-water quenching, glass coating and even conventional melt extraction methods, the present microwire exhibits the largest fracture absorption energy (fracture toughness), as characterized by the area underneath the stress–strain curves. Fig. 5b compares the tensile stress vs. tensile strain data for all of those above-mentioned wires and the microwire in the present study exhibits the highest tensile strength and the largest tensile ductility of all discussed samples.

Further to tensile properties, typical fracture features are also examined (Fig. 6a–f) for the microwires before and after drawing. All of the fracture surfaces consist of two regions: a relatively featureless zone caused by shear slip and a vein pattern produced by the rupture of the remaining section after the initial shear displacement. It is generally believed that the generation of multi-shear bands can effectively impede the sudden propagation of the main shear band and improve the plasticity. However, in the present study, the plasticity of the cold-drawn wires can also be improved while the number of shear bands near the fracture region is reduced as the drawing process continues; indeed fewer shear band steps were observed on the surface, especially for the sample with $R = 51\%$.

Fig. 6f shows a high magnification of the vein pattern of the $R = 51\%$ wire, where the smooth tearing morphology – and hence a viscous-like deformation, indicating the activation of flow process can be clearly seen on the fracture surface.

3.3. TEM analysis of microstructural evolution

First and foremost, micro-scale structural changes during the drawing process exert a significant effect on the high mechanical performance of cold-drawn microwires. The high tensile strength and plausible tensile ductility in the cold-drawn microwires led us to examine the internal structure change during the drawing process in more detail. Fig. 7a–c gives the high-resolution transmission electron microscopy (HRTEM) images for the as-cast, $R = 51\%$ and $R = 64\%$ cold-drawn wires. No contrast and lattice fringe can be detected in Fig. 7a, demonstrating that the as-cast wire is fully amorphous without any nanocrystals, which is also verified by the corresponding selected-area electron diffraction (SAED) pattern (inset in Fig. 7a). Compared with the as-cast structure, isolated nanocrystallites with an average size of 4 nm distributed homogeneously in the amorphous matrix were observed for the $R = 51\%$ sample (Fig. 7b). The degree of nanocrystallization becomes higher and more obvious for the $R = 64\%$ wire, as seen in Fig. 7c. Fig. 7d shows the high-angle annular detector dark-field (HAADF) image of the $R = 51\%$ wire. The readily seen bright and dark contrast indicates the microscale non-uniform distribution of the constructed elements after drawing. These inhomogeneities embedded in the amorphous matrix are believed to arrest the fast extension of shear bands and stabilize the sample against the catastrophic failure, resulting in the enhanced ductility.

3.4. Giant magneto-impedance for wires with different $R$ values

The dependence of the GMI ratios at 10 MHz and 15 MHz as a function of applied axial magnetic field $H$, for different $R$ values, i.e., as-cast, 19%, 36%, 51%, 64%, is presented in Fig. 8. One can see that, at both frequencies, all the wires including the as-cast wire show double-peak
features. The shape of curves and the amplitude of GMI ratio \((\Delta Z/Z)\) as well as the anisotropy field \((H_k)\) at which the maximum GMI ratio occurs depend intimately on the cross-sectional area reduction. To clearly elucidate the role of each drawing step on the GMI characteristics of micro-wires, we plotted the field dependence of maximum GMI ratio \((\Delta Z/Z)_{\text{max}}\) and anisotropy field \((H_k)\). The \((\Delta Z/Z)_{\text{max}}\) decreased dramatically after the initial drawing process; for example, from 134% to 88% at 10 MHz for \(R = 19\%\). This interesting feature is consistent with that reported by Chiriac et al. [29], but they did not explain the reason for this feature. This will be elucidated in the later section. With further deformation, the \((\Delta Z/Z)_{\text{max}}\) starts to increase and reaches a maximum of 160% at 7 Oe for \(R = 51\%\) before decreasing again with increasing \(R\) up to 64%. The anisotropy field \((H_k)\) undergoes a rapid increase from 1 Oe to 5 Oe at 10 MHz after the first drawing step before a relatively small increase of 2 Oe with further drawing. Afterwards the anisotropy field levels off at 7 Oe. Overall the simple cold-drawing process proves to be capable of enhancing the GMI ratio by 30% and the anisotropy field by a factor of 7 in comparison with those of the as-cast wire, respectively. The effect of each cold-drawing step remains the same at both frequencies. Yet it should be noted that at 15 MHz the maximum improvement of the GMI ratio is 44%, much larger than 26% at 10 MHz, whereas the maximum increase of anisotropy field is quite similar, i.e., 6 and 6.6 Oe, respectively. Such a simultaneous large improvement of maximum GMI ratio and anisotropy field is particularly important for the magnetic sensing application which requires a strong response and a wide measurement range.

4. Discussions

4.1. Residual stress state during cold-drawing process

As a first step, we have considered the drawing process where a friction coefficient \(f\) exists between the microwire and the die with a normal stress of \(r_n\). The friction stress \(f r_n\) opposes the motion of the microwire through the die. In analogy to the drawing process of metallic rods and tubes [46], when amorphous wires were drawn through the diamond die, as shown in Fig. 9, the equilibrium of forces in the \(x\) direction can be expressed as:

\[
\frac{1}{4} \pi (\sigma_x + d_{\sigma_x})(D + d_D)^2 = \frac{1}{4} \pi \sigma_x D^2 - \pi D \sigma_x (f + \tan \alpha) dx
\]

where \(\alpha\) is the angle of diamond dies, and \(\sigma_x\) and \(D\) represent the axis stress and diameter at position \(x\), respectively. Since \(d_D = 2 \tan \alpha dx\) and according to the Tresca yield
criterion \([47,48]\), \(\sigma_x + \sigma_n = \sigma_y\) here \(\sigma_y\) is the yield strength of the extracted wire. Thus Eq. (2) can be reduced to:

\[
\frac{d\sigma_x}{(B - 1)\sigma_x - B\sigma_y} = 2\frac{dD}{D} \tag{3}
\]

where \(B = 1 + \frac{f}{\tan \alpha}\) is constant and related to the geometry of the die. Thus Eq. (3) can be integrated directly as follows:

\[
\frac{1}{B - 1} \ln [(B - 1)\sigma_x - B\sigma_y] = 2\ln D + C \tag{4}
\]

Here \(C\) is a constant, and can be identified with the initial condition, i.e. \(D = D_0, \sigma_x = 0\); we can then have:

\[
\frac{1}{B - 1} \ln [-B\sigma_y] = 2\ln D_0 + C \tag{5}
\]

After Eq. (4) minus Eq. (5), the axial and circumferential drawing stress can be given by:

\[
\sigma_x = \frac{B}{B - 1} \sigma_y \left[ 1 - \left( \frac{D}{D_0} \right)^{2(B - 1)} \right] \tag{6}
\]

\[
\sigma_n = \frac{B}{B - 1} \sigma_y \left[ \left( \frac{D}{D_0} \right)^{2(B - 1)} - 1 \right] \tag{7}
\]

Because of its low deformability, the area reduction ratio of Co-based metallic wires was limited to 4% per each step, thus the stress of the wires when extracted out of the diamond die can be calculated as: \(\sigma_x = 0.115\sigma_y\), \(\sigma_n = 0.885\sigma_y\), respectively. Bear in mind that the compressive surface residual stress is beneficial for the increase of hardness and fracture strength, while the tensile residual stress has the opposite effect [25,49–51]. Here in the present case \(\sigma_n \gg \sigma_x\); this suggests that the circumferential compressive stress benefits the mechanical performance of the drawn wires.
4.2. Nanocrystallization mechanism during the drawing process

The features obtained from HAADF and HRTEM images provide direct evidence that homogeneous precipitation of nanocrystallites has taken place to some extent during the cold-drawing process. Since amorphous wires are fabricated by rapid solidification technology, the chemistry and stress distribution are not perfectly uniform in the as-cast condition, as shown in Fig. 3a. These inhomogeneities are expected to act as nucleation sites of nanocrystallites and tend to transform into a lower-energy configuration under external thermal or stress activation [26].

One possible reason for the nanocrystals formation is the temperature rise during the drawing process, since Lewandowski et al. [52] reported that the temperature can increase up to hundreds of degrees during the deformation of metallic glass. Compared with their adiabatic shearing model [52], in the present work, the magnitude and the distribution of temperature during the wire-drawing process depend on the initial temperature of the wire and die, heat generation due to plastic deformation and, most importantly, the friction at the die–material interface. The temperature increase during the drawing process can be established by [53]:

\[
\Delta T = \eta \frac{W}{\rho c} = \eta \int_0^l \frac{\sigma x dx}{\rho c}
\]  

(8)
where $\eta$ is the efficiency of temperature conversion, and often takes a value 0.9 for the wet wire drawing, $\rho$ is the material density ($7.7 \times 10^3$ kg m$^{-3}$), $c$ is the specific heat of the material ($2.18 \times 10^3$ J kg$^{-1}$ K$^{-1}$) and $\epsilon_f$ is final axial strain in the wire at the exit section, and can be expressed as: $\epsilon_f = 2 \ln \frac{D_0}{D_1}$. The temperature rise at the exit section of the die can be calculated from the following equation:

$$\Delta T = 2 \frac{B\eta \sigma_y}{B - 1} \frac{\ln \frac{D_0}{D_1}}{\rho c} \left[ 1 - \left( \frac{D_1}{D_0} \right)^{2(\beta - 1)} \right]$$

Taking into account the real drawing process, where $\alpha = 4-6^\circ$, and $f = 0.135$ for the experiment measurement lubricated by oil, the temperature rise at the exit section of the die is $\sim 0.93$ K, which is much lower than the wire’s glass transition or crystallization temperature, implying another mechanism for the nanocrystal formation, which is likely to be stress-induced crystallization during the drawing process. In this respect, the relationship between the change in the energy barrier for nucleation ($\Delta G_m$) required to form a critical sized nucleus and the hydrostatic pressure ($P$) can be expressed as [54]:

$$\left( \frac{\partial (\Delta G_m^*)}{\partial P} \right) = - \frac{64 \pi a^3}{3 \left( \Delta G_m + E + P \Delta V \right)}$$

where $\Delta G_m$ is the molar free energy change, i.e. the driving force, for an amorphous-to-crystalline phase transformation, $E$ is the elastic energy induced by a volume change during the phase transformation, $\gamma$ is the interfacial free energy between the crystalline and the amorphous phases, and $\Delta V$ is the volume change associated with the formation of a crystalline nucleus. The values for $\Delta G_m$ and $\Delta V$ are both negative and the elastic strain energy ($E$) is negligible. Therefore, the value of $(\partial (\Delta G_m^*)/\partial P)_T$ is also negative, indicating that the energy barrier for nanocrystallites nucleation decreases with increasing hydrostatic pressure. The circumference stress is $\sim 0.885 \sigma_y$, as calculated in Section 4.1; this is much higher than the longitudinal stress and likely to promote the phase transformation during the drawing process.

### 4.3. Effect of residual stress and nanocrystallization on the GMI effect

Compared with the as-cast microwire, melt-extracted microwires after cold-drawing exhibit a higher GMI ratio except for an initial drop for the $R = 19\%$ sample. The distribution of stress generated in the quenching process and the added external stress throughout the wire volume as well as the microstructural evolution during drawing process play decisive roles in the determination of its magnetization configuration, and hence its magnetic performance. In this section, we discuss the effect of the longitudinal tensile and circumferential compressive residual stresses generated during the drawing process and the size of those mechanical-induced nanocrystals on the evolution of magnetic domain configuration. The magnetic anisotropy of amorphous wires is mainly determined by magnetoelastic interactions due to the absence of magnetocrystalline anisotropy (for $R < 51\%$) and negligible shape anisotropy.

![Fig. 10](image_url)

Fig. 10. (a) Radial dependencies of residual stress tensor components in Co-base amorphous microwire, and the corresponding domain structure for the (b) as-quenched, and (c) cold-drawn samples.
The magnetoelastic energy density of an amorphous wire can be formulated as [55]:

\[
w_{\text{m-el}} = -\frac{3}{2} \lambda_s (\sigma^{(q)}_{rr} x^2 + \sigma^{(q)}_{qy} x y + \sigma^{(q)}_{zz} x^2) \tag{11}
\]

where \( \lambda_s \) is the saturation magnetostriction constant, \( \sigma^{(q)}_i \) are the diagonal components of the residual stress tensor in cylindrical coordinates \( (r, \varphi, z) \); and \( x \) are the components of the unit magnetization vector. The residual quenching stress throughout the wire is assumed to be a function of reduced wire radius \( x/(r R_0) \), and can be reasonably approximated by means of the following simplified relations [56]:

\[
\begin{align*}
\sigma^{(q)}_{rr}/\sigma_y &= a (1 - x^2) \tag{12} \\
\sigma^{(q)}_{qy}/\sigma_y &= a (1 - 3x^2) \tag{13} \\
\sigma^{(q)}_{zz}/\sigma_y &= b (1 - 2x^2) \tag{14}
\end{align*}
\]

where \( a \) and \( b \) are the constants relevant to the material property.

Fig. 10a illustrates such radial dependence of residual stress. There exists an intersection at \( x_1 \) for \( \sigma^{(q)}_{rr}/\sigma_y \) and \( \sigma^{(q)}_{zz}/\sigma_y \). The inner core and outer shell are then classified at this point for an as-prepared amorphous wire with negative magnetostriction, and \( x_1 \) is the inner core radius, where the easy axis is along the wire due to \( \sigma^{(q)}_{zz}/\sigma_y < \sigma^{(q)}_{rr}/\sigma_y \); for outer surface \( (r > x_1) \), \( \sigma^{(q)}_{zz}/\sigma_y > \sigma^{(q)}_{rr}/\sigma_y \) results in a circumferential anisotropy. The corresponding core–shell domain structure for the as-cast wire is shown in Fig. 10b, consisting of the outer shell circular domains and inner core axial domains. For the cold-drawn wires, as discussed in Section 4.1, the existence of circumferential compressive stress and longitudinal tensile stress will cause a change in the volume fraction of outer shell circular domains and inner core axial domains, i.e., the outer shell domains become larger and the inner core domains are reduced accordingly, as depicted in Fig. 10c, but the domain configuration remains unchanged. This can be mathematically interpreted as the movement of intersection \( x_1 \) for \( \sigma^{(q)}_{rr}/\sigma_y \) and \( \sigma^{(q)}_{zz}/\sigma_y \). As demonstrated in a previous study [57], a larger GMI effect in Co-rich amorphous wire was achieved by an external axial tensile stress due to the rearrangement of the domain wall induced by tensile stress and the increase in circular anisotropy and permeability. The coupling effect of external axial tensile stress and residual quenched axial stress \( \sigma_{zz} \) will cause the intersection \( x_1 \) to move to the left, increasing the outer shell circular domains. In analogy, the circumferential compressive residual stress will cause a smaller \( \sigma_{qy} \) similarly resulting in the intersection moving slightly to the left, i.e. point \( x'_1 \). As such, the axial residual tensile stress and circumferential compressive stress induced by the cold-drawing process together increase the volume of the outer shell and hence the circumferential permeability, giving rise to an improved GMI ratio.

Based on the above analyses, one would expect a monolithic increase of GMI ratio with a further drawing process. However, Fig. 8 shows a complex field dependence of maximum GMI ratio. Thus the residual stress can only explain the increase of GMI from \( R = 19\% \) to \( 51\% \). To understand the large reduction of GMI for the initial drawing steps, one needs to consider the imperfect geometries of wires, which are expected to act as defects to cause stress concentration at the surface during the cold-drawing process, as schematically shown in Fig. 11. Along with the SEM images in Fig. 2a, it can be seen that the inhomogeneous surface of as-cast wire is forced into several stress-concentrated local regions, which will result in the inhomogeneities of magnetoanisotropy energy in the surface and deteriorate the soft magnetic properties. As solely a surface related property, the GMI effect is reduced sharply.

In another perspective, as discussed in Section 4.2, mechanical-induced nano-scale crystals formed during the cold-drawing process have a significant effect on the GMI properties. Isolated small sized nanocrystals of less than 4 nm embedded in the amorphous matrix for the 51\% drawn sample yields a maximum GMI ratio of \( \sim 160\% \) at 10 MHz. This phenomenon can be attributed to the residual stress generated during the drawing process, as the effect of crystalline anisotropy is negligible in this case. When the deformation is beyond the observed critical point of \( R = 51\% \), we can see from Fig. 7c that the size of these Co-rich nanocrystals becomes larger, with a diameter exceeding 10 nm, and these crystals nearly touch with each other. The existence of these large-size nanocrystals causes an increase of magnetocrystalline anisotropy and magnetic hardness [58], resulting in a deterioration of the soft magnetic property and hence the reduction of the GMI ratio.

The last point deals with the effect of deformation on the anisotropy field which is governed by the magnetostriction constant \( \lambda_s \), saturation magnetization \( M_s \), and the difference between \( \sigma_{zz} \) and \( \sigma_{qy} \), and can be expressed as [32]:

\[
\begin{align*}
\phi(x) &= \phi(R_0) + \phi(R) \\
&= \phi(0.5R_0) + \phi(R/1.5R_0) \\
&= \phi(0.5R_0) + \phi(R/1.5R_0)
\end{align*}
\]
\[ H_z = \frac{3\lambda_s}{M_s} (\sigma_{zz} - \sigma_{pp}) \] (15)

As the difference between \( \sigma_{zz} \) and \( \sigma_{pp} \) increases with the cold-drawing process as shown in Fig. 10, the anisotropy field then increases accordingly. However, instead of a linear increase with increasing reduction area, the anisotropy field became constant after \( R = 36\% \). In a reverse way of reasoning, this suggests that the difference between \( \sigma_{zz} \) and \( \sigma_{pp} \) could become smaller. Indeed, as shown in a previous study [25], the circumferential stress exhibits a peak feature with increasing reduction area, i.e., the stress increases first and then decreases. This explains the observed evolution of anisotropy field with the wire area reduction. It is desirable to conduct a similar quantitative analysis for the present wires, which will be addressed in future work.

5. Conclusions

The effect of cold-drawing on the compositional and microstructural variations has been systematically examined and correlated to the mechanical and GMI properties of melt-extracted Co-based microwires. The Co-based microwires can be successfully cold-drawn with up to 75% cross-sectional area reduction. Tensile ductility, tensile strength as well as the GMI effect of the drawn wires increased with cold-drawing and reached a peak of 1.09%, 4320 MPa and 160%, respectively, at 51% cross-sectional area reduction and followed by a reduction with further deformation. Both HRTEM and HAADF observations confirm the presence of mechanically induced (rather than thermally activated) nano-sized crystallites precipitated in the amorphous matrix during drawing and can stabilize the shear bands and arrest its fast propagation, leading to an enhanced ductility. The residual stress not only accelerates the amorphous-to-nanocrystalline phase transformation but also contributes to the mechanical and magnetic properties. Both GMI ratio and anisotropy field are significantly improved after cold-drawing. The complex dependence of GMI characteristics on cold-drawing has been elucidated by a full consideration of residual stress and nanocrystalline structure, as well as geometrical defects. The present study demonstrates that with the precipitation of fine-sized nanocrystals and the generation of large circumferential compressive stress, the drawing process can be exploited for simultaneous improvement of both mechanical and soft magnetic properties.

Before closing, it is worth mentioning the important implications and the future work of broad interest. As the present study establishes a fundamental correlation between the residual stress, microstructure and the mechanical and magnetic properties of ferromagnetic microwires, the potential for performance improvement remains to be explored by the manipulation of residual stress and microstructural control, for example, through various annealing methods. Technically, maximizing improvement in the soft magnetic properties of wires without compromising their mechanical performance, such as post-treatment, multilayer electrodeposition on the drawing wire, as well as the optimization of drawing process are of much interest for their practical applications.

Acknowledgements

Mr. H. Wang is grateful for the financial support from the Short-doctoral visiting scholar program of Harbin Institute of Technology for his study at the University of Bristol, UK, and thanks his co-workers, in particular Guoqiang Wang, Jingshun Liu, Dongming Chen, and Lun-yong Zhang, for useful discussions. FXQ would like to acknowledge the financial support from EPSRC institutional sponsorship.

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