Article

Not peer-reviewed version

Enhancing the Squareness and Bi-phase Magnetic Switching of Co2FeSi Microwires for Sensing Application

Mohamed Salaheldeen * , Asma Wederni , Mihail Ipatov , Valentina Zhukova , Ricardo Lopez Anton * , Arkadi Zhukov

Posted Date: 6 April 2023

doi: 10.20944/preprints202304.0071.v1

Keywords: Heusler alloys; Glass-coated microwires; Multi-step magnetic behavior; Sensing Applications

Article

Enhancing the Squareness and Bi-Phase Magnetic Switching of Co_2FeSi Microwires for Sensing Application

Mohamed Salaheldeen [1,2,3,4,*], Asma Wederni [1,2,4], Mihail Ipatov [1,2], Valentina Zhukova [1,2,4], R. Lopez Anton [6,*] and Arcady Zhukov [1,2,4,5]

1 Departamento de Polímeros y Materiales Avanzados, Facultad Química, Universidad del País Vasco,UPV/EHU, 20018 San Sebastián, Spain
2 Departamento de Física Aplicada, EIG, Universidad del País Vasco, UPV/EHU, 20018 San Sebastián, Spain
3 Physics Department, Faculty of Science, Sohag University, Sohag 82524, Egypt
4 EHU Quantum Center, University of the Basque Country, UPV/EHU, 20018 San Sebastián, Spain
5 IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain
6 Instituto Regional de Investigación Científica Aplicada (IRICA) and Department of Applied Physics, University of Castilla-La Mancha, 13071, Ciudad Real, Spain
* Correspondence: mohamed.salaheldeenmohamed@ehu.eus (M.S.); ricardo.lopez@uclm.es (R.L.A.)

Abstract: In current study we have obtained Co_2FeSi-glass coated microwires with different geometrical aspect ratio, $\rho = d/D_{tot}$ (diameter of metallic nucleus, d and total diameter, D_{tot}). The structure and magnetic properties are investigated at a wide range of temperature. XRD analysis illustrates a notable changing in the microstructure by increasing the aspect ratio of Co_2FeSi glass coated microwires. Amorphous structure is detected for the sample with the lowest aspect ratio (ρ = 0.23), whereas a growth of crystalline structure is observed in the other samples (aspect ratio ρ = 0.30 and 0.43). This change at the microstructure properties correlates with dramatic changing in magnetic properties. For the sample with the lowest ρ -ratio, non-perfect square loops are obtained with low normalized remanent magnetization. A notable enhancement in the squareness and coercivity are obtained by increasing ρ -ratio. Changing the internal stresses strongly affects the microstructure, resulting in a complex magnetic reversal process. The thermomagnetic curves show large irreversibility for the Co_2FeSi with low ρ -ratio. Meanwhile, if we increase the ρ -ratio, the sample shows perfect ferromagnetic behavior without irreversibility. The current result illustrates the ability to control the microstructure and magnetic properties of Co_2FeSi-glass-coated microwires by changing only their geometric properties without performing any addition heat treatment. The modification of geometric parameters of Co_2FeSi glass-coated microwires allows to obtain microwires which exhibit an unusual magnetization behavior that offers opportunities to understand the phenomena of various types of magnetic domain structures, which is essentially helpful for designing sensing devices based on thermal magnetization switching.

Keywords: heusler alloys; glass-coated microwires; multi-step magnetic behavior; sensing applications

1. Introduction

The use of ferromagnetic materials in spintronic applications has garnered increasing attention in recent years due to their unique magnetic properties that enable the control and manipulation of spin currents. Among the different types of ferromagnetic materials, micro/nano-structured materials have emerged as a promising candidate for enhancing spintronic devices' performance [1–9]. One of the most promising multidisciplinary research fields is spintronics, which enables the creation of the next-generation of nano & micro devices with improved processing and memory capability while consuming less power [10]. To address the various required criteria, such as high spin polarization or high Curie temperature, *Tc*, a new generation of materials with multifunction uses has to be created [11]. The Heusler compounds are one of the potential smart materials with desirable characteristics for spintronic and magneto-electronic applications [12]. Perfect lattice matching with

 doi:10.20944/preprints202304.0071.v1

major substrates, high *Tc* above room temperature, and intermetallic controllability for spin density of states at the Fermi energy level—where approximately 100% of spin polarized near the Fermi level is reported [11–15]—are just a few of their advantages.

In terms of half-metallic alloys, Co_2-based full-Heusler compounds are among the most promising ones because of their high thermal stability, high Curie temperatures (Tc ≈1100 K) for the bulk sample, high magnetic moment (~ 6 μ_B/f.u.), and low Gilbert damping constant (α = 0.004) [14,16,17]. They also have exotic transport properties, an electric structure determined by ab initio calculations [16], and high magnetic moments. The significant anomalous Hall effect that Co_2-based Heusler compounds exhibit because to the enormous Berry curvature associated with their band structure is also of current interest [16,18]. These factors have the scientific community interested in Co-based full-Heusler alloys. These types of alloys are therefore extensively researched in several configurations, including nanoparticles [19], thin films [15,17,20], and nano/micro wires [21–24]. It is important to note that the fabrication of Heusler alloys nanoparticles and thin films faces numerous difficulties for application purposes, including the high cost of preparation methods, chemical composition inhomogeneity, and ease of oxidation from the perspective of proper atomic ordering and material chemistry [20]. The diffusion of substrate atoms into the film results in the existence of atomic disorder and phase separations, which are commonly observed [25], in addition to the lattice mismatch between the alloy and the substrate. Furthermore, in order to start the requisite structural ordering, the arc-melted or thin-film formed Heusler alloys need lengthy, high temperature annealing procedures [26].

Magnetic wires research has received a lot of interest during the last several decades [27]. The focus is on amorphous magnetic wires, which can exhibit unusual magnetic features such as spontaneous magnetic bistability or the Giant magnetoimpedance phenomenon [27,28]. Several manufacturing processes involving fast solidification can be used to create magnetic wires containing amorphous and/or nanocrystalline phases [27,28]. Nevertheless, only the Taylor-Ulitovsky manufacturing approach allows the preparation of magnetic microwires with the widest diameter range (from 0.2 to 100 μm) [27,28]. Such microwires are composites consisting of metallic nucleus (with $0.2 \leq d \leq 100$ μm) usually comprised of iron, cobalt, nickel or their alloys, covered by thin, flexible and insulating glass (typically Pyrex or Duran) coating (typically with thickness from 0.5 to 10 μm) [27,28]. As a result, the prospective applications of glass-coated microwires in sensing, actuation, and biomedical engineering have been expanded. The insulating and flexible glass coating protects the microwires from oxidation, corrosion, and other environmental factors while simultaneously giving them outstanding mechanical stability. Moreover, the glass layer and the magnetically flexible amorphous metallic nucleus provide high sensitivity to external stimuli including magnetic fields, temperature fluctuations, and mechanical stress. [27–35]. Such sensitivity is connected to the ferromagnetic origin of the metallic nucleus, which responds to the applied stimulus. Innovative sensors that monitor magnetic fields, temperature, and stress have been developed using glass-coated microwires for a range of applications [27–29]. Additionally, they have shown potential characteristics for actuators and in medical applications, including as cancer treatment and medicine administration. Future technological advancements can use glass-coated microwires because of their distinctive combination of properties [27,29].

In this article, we report an attempt to prepare Co_2FeSi glass-coated microwires with variable geometrical aspect ratios $\rho = d/D_{tot}$ (being *d*-diameter of metallic nucleus and D_{tot} - total diameter). The fabrication method was chosen because of the intriguing magneto-structural behavior of glass-coated microwires derived from Heusler alloys, as well as functional properties of glass-coated microwires such as superior mechanical properties, insulating, thin and flexible glass-coating, and thin dimensionality [27,29–35]. As a result, we have prepared Co_2FeSi glass-coated microwires using the Taylor-Ulitovsky procedure, which is detailed previously [27,36,37]. The Taylor-Ulitovsky approach, which has been utilized since the 1960s [36], is one of the current fabrication methods used to make Heusler alloys glass-coated microwires with a wide variety of geometric characteristics [21,22,24,27–35]. The primary benefit of this low-cost technique is that it allows the production of thin and long (a few kilometers long) microwires with a wide diameter range (d- values ranging from 0.2

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

to 100 µm) at high speeds (up to a few hundred meters per minute) [36–38]. This process is also used to prepare glass-coated microwires with excellent mechanical properties [21,39–41]. Glass coating on microwires can give us with extra benefits such as better insulation and environmental protection. Moreover, the availability of a biocompatible thin, flexible, insulating, and highly transparent glass coating may aid biological applications [29,42,43]. As a result, Heusler microwires based on Co_2FeSi are a potentially smart material with applications in a wide range of devices. To the best of our knowledge, up to date no one has reported on the production and structural, mechanical, or magnetic characterization of Co_2FeSi-based glass-covered Heusler microwires with varied ρ - ratios, as well as the investigation of its influence on magneto-structure behavior.

2. Materials and Methods

Arc melting is a method of manufacturing Co_2FeSi alloys that involves melting the precursor components together in an electric arc furnace. Typically, the following procedures are used to create Co_2FeSi alloys by arc melting: i) preparing the precursor ingredients. The precursor elements for the Co_2FeSi alloy are weighed and deposited in a graphite crucible, containing cobalt (99.99%), iron (99.9%), and silicon (99.99%). ii) The materials melting. The crucible containing the precursor materials is put in an electric arc furnace, and an electrical current is fed through the materials to start the melting process in a vacuum and argon atmosphere. The furnace temperature is precisely regulated to ensure that the ingredients melt and mix equally. iii) The cooling and solidification processes. The crucible is withdrawn from the furnace and allowed to cool once the components have melted and combined. The Co_2FeSi alloy (ingot) is created as the ingredients consolidate. This process were then repeated five times to achieve perfect homogeneity and a homogeneous microstructure. Once the Co_2FeSi alloy has solidified and formed an ingot, the ingot is used to prepare Co_2FeSi glass-coated microwires using the Taylor-Ulitovsky process. As described in the introduction, the Taylor-Ulitovsky preparation technique offers significant benefits over alternative procedures for manufacturing glass-coated microwires. One advantage is that it enables the fabrication of microwires with very thin glass coatings, generally up to a few micrometers thick. This thin covering permits the electrical and magnetic characteristics of the microwire metallic nucleus to be preserved, making the resultant microwires valuable for a wide range of applications. Many prior publications [21,22,24,27–35] explain the manufacturing method in detail. A glass capillary was produced and filled with molten Co2FeSi alloy after a high frequency inductor heated an ingot over its melting temperature. The diameter of the metallic nuclei, d, was then determined by varying the speed of wire drowning and the rotation of the pick-up bobbin. The manufactured microwire is sent via a coolant stream to complete the fast melt quenching process.

We used Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray (EDX) (JEOL-6610LV, JEOL Ltd., Tokyo, Japan) to determine the aspect ϱ -ratio of Co_2FeSi glass-coated microwires samples and its related nominal chemical composition.

The XRD structure analysis was carried on by using X-ray diffraction (XRD) BRUKER (D8 Advance, Bruker AXS GmbH, Karlsruhe, Germany).

The magnetic behavior was studied through two different ways: hysteresis loops at temperatures between 5 and 350 K, and thermomagnetic curves following three different protocols, zero field cooling (ZFC), field cooling (FC) and field heating (FH) at low magnetic field (H = 200 Oe). All magnetization curves were measured using a PPMS (Physical Property Magnetic System, Quantum Design Inc., San Diego, CA) vibrating-sample magnetometer at temperatures, T, between 5 and 400 K for ZFC, FC and FH magnetic curves. For the hysterics loops we only focus on the in-plane configuration where the applied magnetic field is parallel to the wire axis. The results are provided in terms of the normalized magnetization, M/M_{5K}, where M_{5K} is the magnetic moment obtained at 5 K to avoid the misleading of the errors estimation in the estimation of the magnetization saturation values.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

3. Results

3.1. Analysis of Chemical and Structural Data

The geometries (ϱ-ratios) and chemical compositions of prepared samples are shown in Table 1. Using the EDX data from Table 1, it was revealed that the metallic nucleus composition differed considerably from the stoichiometric one (Co_2FeSi). The features of the preparation process, which involved alloy melting and casting, were the cause of this slight variation. To quantify the difference, we checked at the nominal composition for 8 sites as illustrated in Figure 1a. An atomic average of $Co_{44}Fe_{23}Si_{33}$ was used to confirm that the true 2:1 ratio of Co and Fe applied in all sites. A high Si ratio was found because of the interfacial layer that exists between the metallic nucleus and the glass covering.

Table 1. The geometrical parameters and average (Av.) of atomic percentage of Co, Fe and Si elemental composition in Co_2FeSi glass-coated microwires.

Sample	Aspect ratio (ρ)	Chemical composition
$GCMW_A$	0.23	$Co_{44}Fe_{23}Si_{33}$
$GCMW_B$	0.30	$Co_{44}Fe_{23}Si_{33}$
$GCMW_C$	0.43	$Co_{44}Fe_{23}Si_{33}$

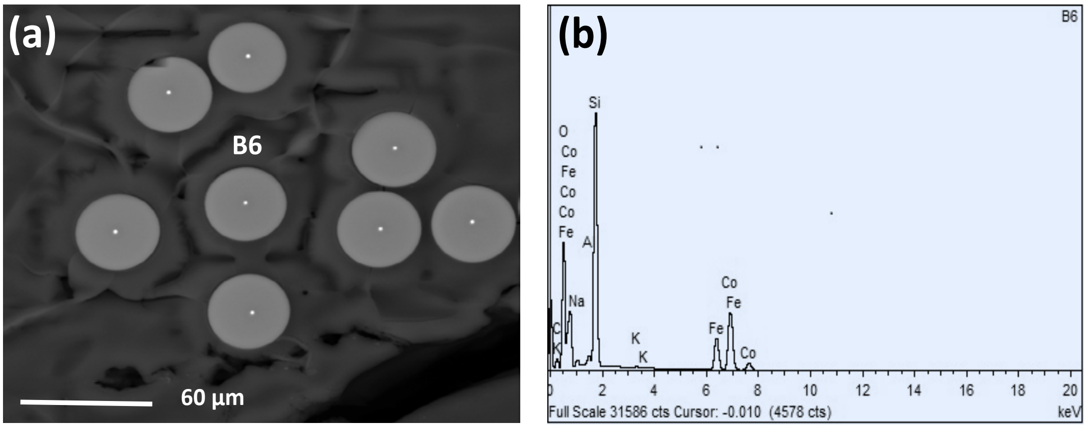

Figure 1. The cross section of selected Co_2FeSi glass-coated microwires with aspect ratio 0.30 images (a) and the chemical composition spectra of EDX of one of the points (b).

In order to study the order state of our produced Co_2FeSi Glass coated microwires, and to elucidate the effect of the aspect ratio modification on the crystalline structure, XRD structure analysis was carried on by using X-ray diffraction (XRD).

As illustrated in Figure 2 the changing in the geometric ρ-ratio has a strong influence on the structure of Co_2FeSi glass-coated microwires. For the sample with the lowest ρ- ratio, i.e. $\rho = 0.25$, the sample shows an amorphous structure where no crystalline peaks is detected. The wide halo at $2\Theta = 22.3°$ is related to the glass coating layer, as reported in our previous works [21,22,24,27–35]. By increasing the geometric aspect ratio a crystalline structure of metallic nucleus becomes evident with a notable peak at $2\Theta = 46.2°$, attributed to the (220) reflection. Further increase of geometric ρ-ratio results in the perfect crystalline structure of studied samples, where the crystalline peak intensity increases and additional peak appears at $2\Theta = 85.4°$, corresponding to the (422) reflection. The analysis of XRD profiles of the two crystalline Co_2FeSi samples, i.e. $GCMW_C$ (P = 0.30) and $GCMW_B$ (P = 0.43), indicates an A2 single-phase structure with a small tetragonal distortion (traces of tetragonal martensite phase), and a broadened peak around 22° attributed to an amorphous state for $GCMW_C$ and mixed $L2_1$ or B2 phases with amorphous state for $GCMW_B$ sample [34,35].

The (220) and (422) reflections in $GCMW_C$ sample are split due to some tetragonal distortions of the crystal lattice. A similar phenomenon was seen and discussed elsewhere [44]. It is known, that a

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

split in the brag diffraction patterns lead to a small distortion of the crystalline structure [45]. The absence of a (400) peak around 85°, which is expected to be present in the A2 structure, increases the possibility that the crystallites are too fine to be detected by X-rays, as reported elsewhere [46]. In addition, the absence of some peaks can be caused by a similar scattering factor of the constituent elements (Co, Fe, and Si) [47]. Otherwise, according to the theoretical outcomes of Zhang et al., the disordered A2 state is more energetically preferable than those of the ordered $L2_1$ or B2 phases [48,49]. Nevertheless, the well-defined and sharp diffraction patterns in this sample ($GCMW_C$ sample) indicate a high crystallinity, as-compared with the other two XRD spectrum. As the the development of traces of secondary phase (tetragonal martensite) can affect the magnetic behavior, this will be explored in more details in the following sections.

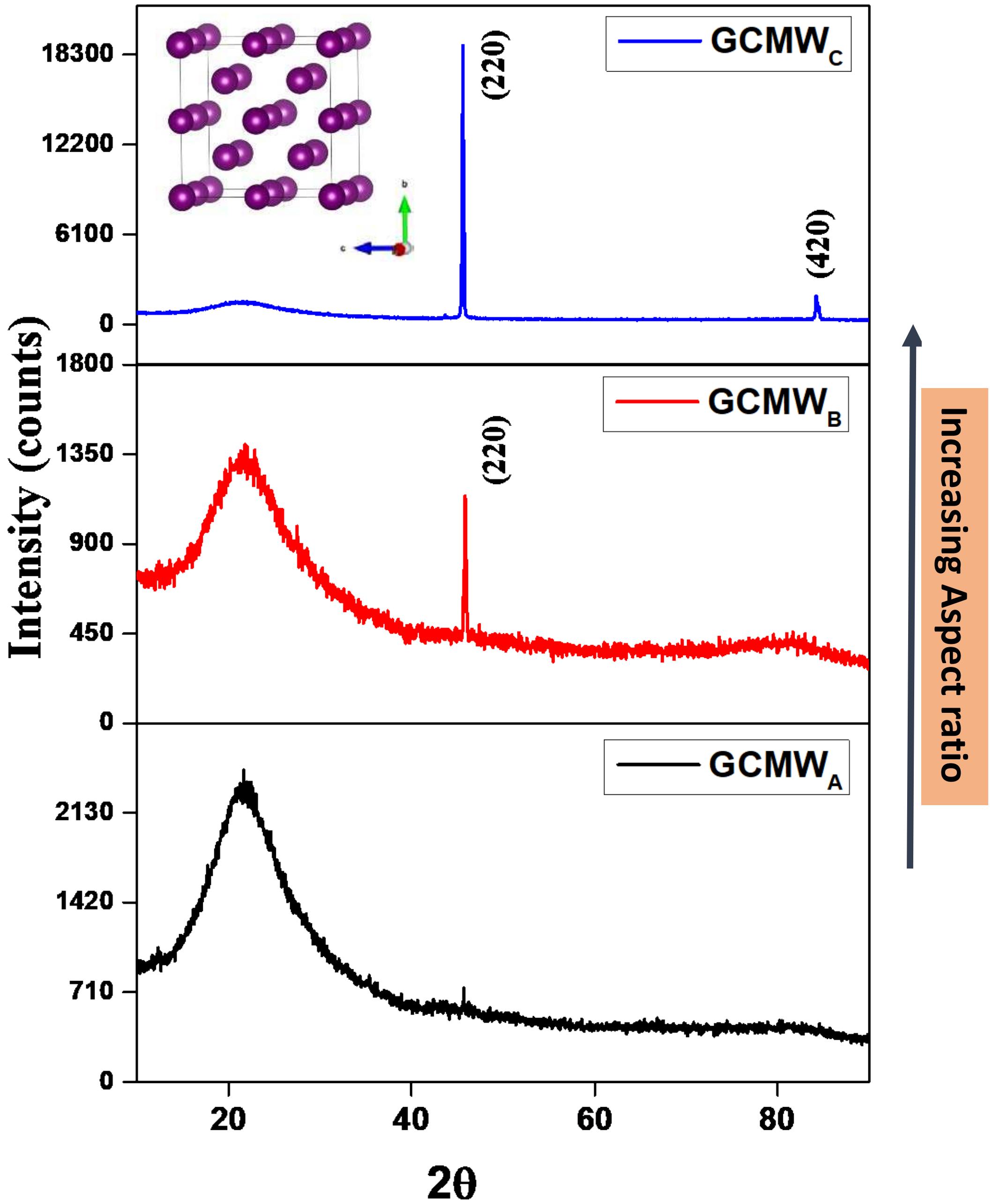

Figure 2. XRD analysis of Co_2FeSi glass-coated microwires with different aspect ratio measured at room temperature. The inset of Figure 2 indicates the A2-type cubic structure.

We estimated the lattice parameters of the two crystalline Co_2FeSi glass-coated microwires and then we employed the Debye- Scherrer's equation, as presented in our previous work [23], to investigate the microstructure of Co_2FeSi in greater depth. Using this methodology, we can estimate the average grain size, D_g, associated to the principal peaks, which is approximately 37.6 nm and 45.8 nm for $GCMW_B$ and $GCMW_C$ of Co_2FeSi microwires, respectively as illustrated in Table 2.

Table 2. The average grain size and lattice parameters of Co_2FeSi glass-coated microwires with different aspect ratio.

Sample	Average grain size (nm)	Lattice parameters
$GCMW_A$	-	-
$GCMW_B$	45.8	5.63
$GCMW_C$	37.6	2.81

3.2. Magnetic Characterization

3.2.1. Room Temperature Magnetic Properties

Figure 3 shows the magnetic hysteresis loops of Co_2FeSi glass-coated microwires with different ϱ- ratios, obtained at room temperature with an applied magnetic field parallel to the microwire axis. All samples exhibit typical ferromagnetic behaviour, due to the high Curie point of Co_2FeSi alloy greater than 1100 K [46]. The sample with low ϱ -ratio exhibits soft magnetic properties with coercivity, Hc, around 14 Oe and non-square hysteresis loop shape (Figure 3a). However, the sample with the largest ϱ -ratio shows almost perfectly square hysteresis loops with higher Hc (about 87 Oe), than Co_2FeSi with low ϱ -ratio (see Figure 3b and 3c). In addition, the hysteresis loop shows multistep magnetic behavior (indicated with arrows in figure 3c). The almost square hysteresis loops for the $GCMW_C$ microwire with normalized remanent, Mr, near to 0.96 indicates the axial character of magnetic anisotropy with easy axis of magnetization along the direction of applied magnetic field. Thus, the increase in ϱ - ratio affects the magnetocrysttaline ansiotropy and its direction has the same direction of (220) and (420), as illustrated in the structural section. However, in the sample $GCMW_B$ with crystaline structure, non-perfectly square loops is observed. Such change in the hysteresis loop shape must be related to the presence of the considerable amount of amorphous phase beside the disordered B2 or little ordered $L2_1$ structures. In our previous work at the same alloys, but with low ϱ – ratio (ϱ = 0.26), the enhancement of the magnetocrystlline anisotropy, the squareness and coercivity of Co_2FeSi glass coated microwires after annealling was observed [21,22,24]. As we illustated in our previous work, the main two factors affecting the magnetic anisotropy behavior in Heusler based glass- coated mixcrowires are unixial magnetic anisotropy and cubic magnetocrystalline anisotropy [21,22,24]. By increasing the ϱ-ratio an enhacement in the crystalline phase content correlates with the magnetic properties modification, i.e. the main factor controlling the magnetic anistropy is the cubic magnetocrystalline anistropy. Unfortunately, currently we are not able to measure this type of anisotropy experimentaly, but the perfectly square loop indicates its stronge effect for the $GCMW_B$ and $GCMW_C$ samples. As seen in Figure 4, the $GCMW_C$ sample shows the highest anisotropy field H_k, coercivity H_c and normalized remanent M_r.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

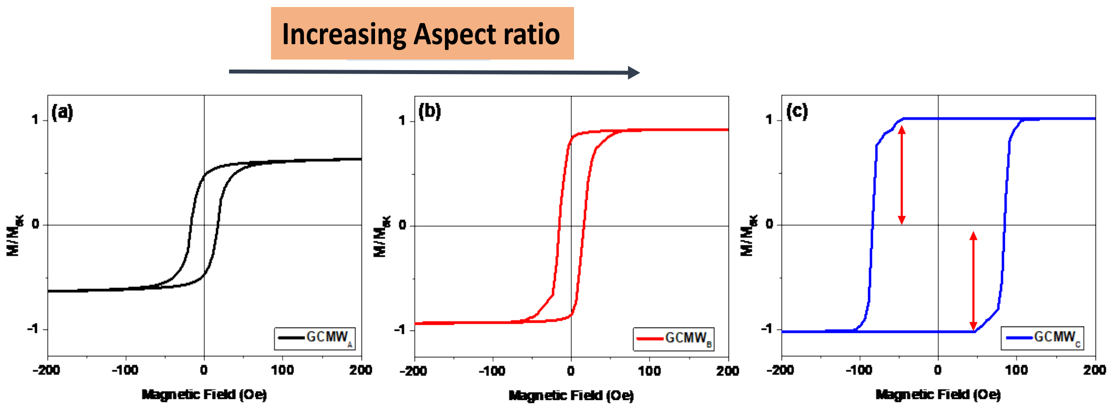

Figure 3. Room temperature hysteresis loops for Co_2FeSi glass-coated microwires (a) $GCMW_A$, (b) $GCMW_B$ and (c) $GCMW_C$. The arrows in Figure 3 (c) pinpoints the multistep magnetic behavior.

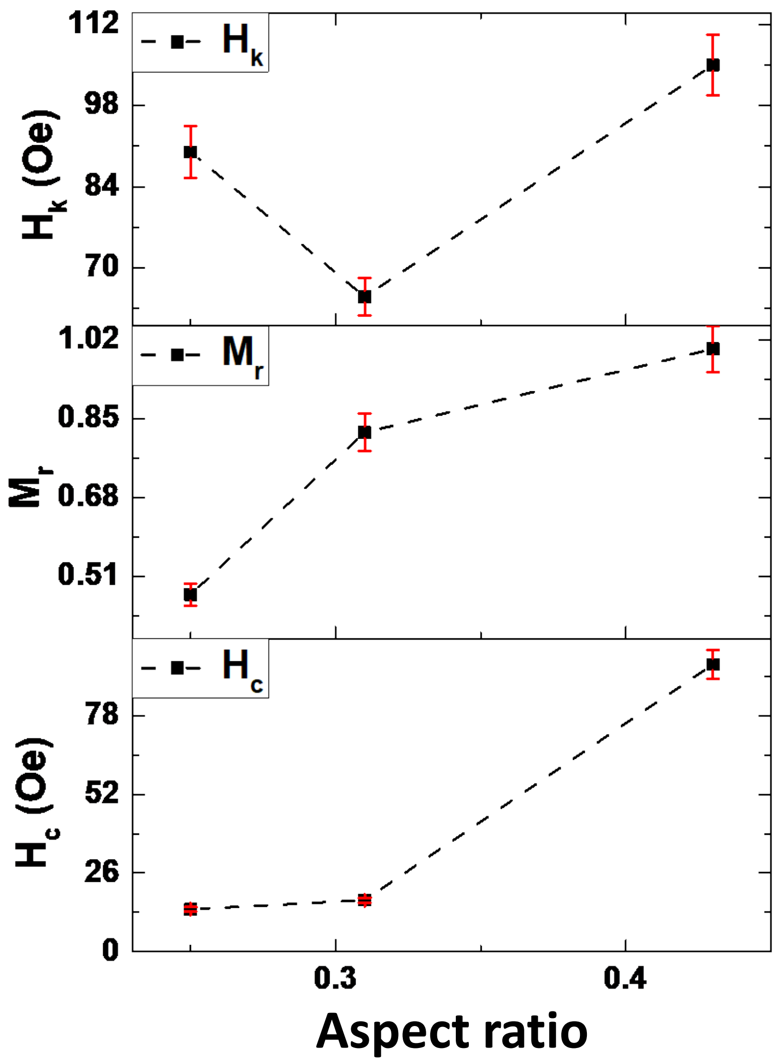

Figure 4. Aspect ratio dependence on coercivity (H_c), normalized remanence (M_r), and in –plane anisotropy field (H_k) of Co_2FeSi glass-coated microwires (lines for eye guide).

3.2.2. Thermomagnetic Properties

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

It is worth noting that the ferromagnetic materials temperature stability is a crucial characteristic for their possible applications in spintronic and sensing devices. Hence, for a wide range of measurement temperatures, 5-350 K, we investigated the magnetic behavior of Co_2FeSi glass coated microwires with different ϱ- ratios. The shape of the loops follows the same trend observed at room temperature: non-square for the $GCMW_A$ sample, quite square for the $GCMW_B$ one, and almost square for the $GCMW_C$ one (loops not shown) In Figure 5, the evolution of H_c and M_r with the temperature is shown. This behavior demonstrates that for the $GCMW_C$ sample, cubic magnetocrystalline anisotropy prevails up to 350 K.

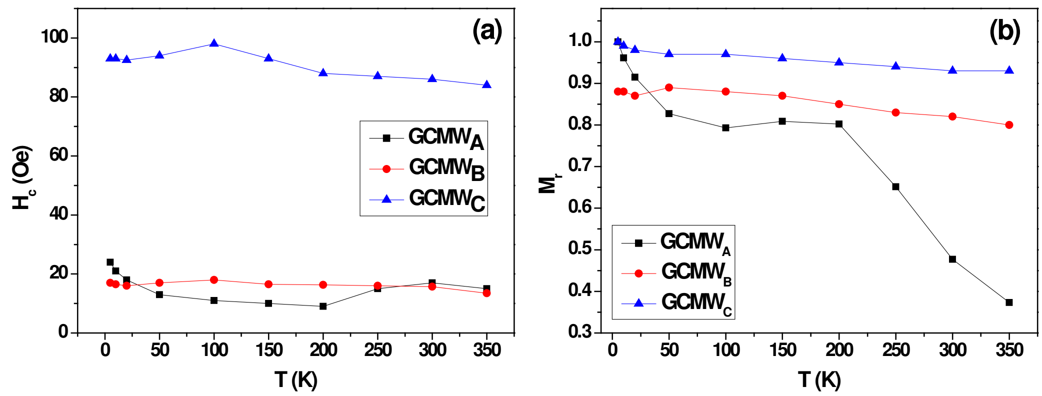

Figure 5. Temperature dependence of the coercivity (a) and normalized remanence (b) of Co_2FeSi glass-coated microwires with different aspect ratio (lines for eye guide).

By analyzing the hysteresis loops measured at temperature range , 5–350 K of Co_2FeSi glass-coated microwires with different ϱ- ratio, an interesting magnetic behavior is found for both the temperature dependence of H_c and of the normalized remanence, M_r. $GCMW_C$ sample shows the highest value of the coercivity at the all measuring range of temperature range, with an average value of H_c 6 times higher than those of the $GCMW_A$ and $GCMW_B$ samples. Both $GCMW_A$ and $GCMW_B$ samples show quite similer values of the H_c where the different between the average value of coercivity is about 2 Oe. By estimating the differences of the coercivity (ΔHc) between the maximum value of coercivity (**$H_{c\ (max)}$**) and the lowest value of the coercivity (**$H_{c\ (min)}$**) for all samples we pretend to show its stability with temperature. The samples with a clear crystalline phase, $GCMW_B$ and $GCMW_C$ samples, show higher temperature stability than the amorphous $GCMW_A$ sample: Hence the ΔHc is 3.5 and 9 Oe for $GCMW_B$ and $GCMW_C$ samples, respectivly, whereas ΔHc is 15 Oe for the $GCMW_A$ one. The magnetic stability is more clear in the case of Mr tendency with temperature of Co_2FeSi glass-coated microwires with different ϱ - ratios. As shown in Figure 5b, both $GCMW_B$ and $GCMW_C$ samples show high stability with temperature, with ΔM_r 0.05 and 0.06, respectivily (see Table 3). Meanwhile, the behavior of Mr of $GCMW_A$ is rather different, comparing to the other samples with higher ϱ -ratios, where a monotonic increase with decreasing the temperature has been observed.

Table 3. The geometrical parameters and average (Av.) of Co_2FeSi glass-coated microwires with different aspect ratio.

Sample	ΔH_c ($H_{c\ (max)}$ - $H_{c\ (min)}$)	ΔM_r ($M_{r\ (max)}$ – $M_{r\ (min)}$)
$GCMW_A$	15 ± 2 Oe	0.7 ± 0.1
$GCMW_B$	3.5 ± 0.5 Oe	0.06 ± 0.01
$GCMW_C$	9 ± 2 Oe	0.05 ± 0.01

Figure 6 shows the complete thermomagnetic behavior of Co_2FeSi glass-coated microwires with different ϱ -ratios. We performed the ZFC, FC and FH magnetic temperature dependence to check any possible phase transition. Thus, the measurements were performed at low magnetic field 200 Oe. For $GCMW_A$ sample, the ZFC, FC and FH magnetizations curves show non-homogonous behavior, beside an irreversible magnetic behavior at T = 150 K. Such irreversibility has been observed in our

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

previous work dealing with Co_2FeSi-based glass-coated microwires with aspect ratio ρ = 0.26, (see [21,22,24].). In this works we have illustrated that the irreversibility enhanced by preforming annealing at 873 K and 973 K for 1 h. The induced martensitic transition and the changing in the internal stresses associated with the glass-covering layer with temperature allow to control the irreversibility behavior. The interesting point for Co_2FeSi-glass-coated microwires with the lowest ρ-ratio, i.e., ρ = 0.23, is that its blocking temperature is observed at T = 150 K, like the Co_2FeSi-glass-coated microwires with ρ = 0.26 [21,22,24]. For $GCMW_A$ sample, which is totally in amorphous state (see Figure 2), the main reason for the irreversibility behavior is the strong internal stress induced by the glass covering layer. For $GCMW_B$ and $GCMW_C$ sample, i.e., increasing ρ -ratio, the irreversibility behavior disappeared and the usual ferromagnetic behavior is observed with homogenous ZFC, FC and FH magnetic curves. The homogenous magnetization curves are due to the induced crystal structure with A2-type and B2 or $L2_1$ cubic structure for $GCMW_C$ and $GCMW_B$, respectively.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

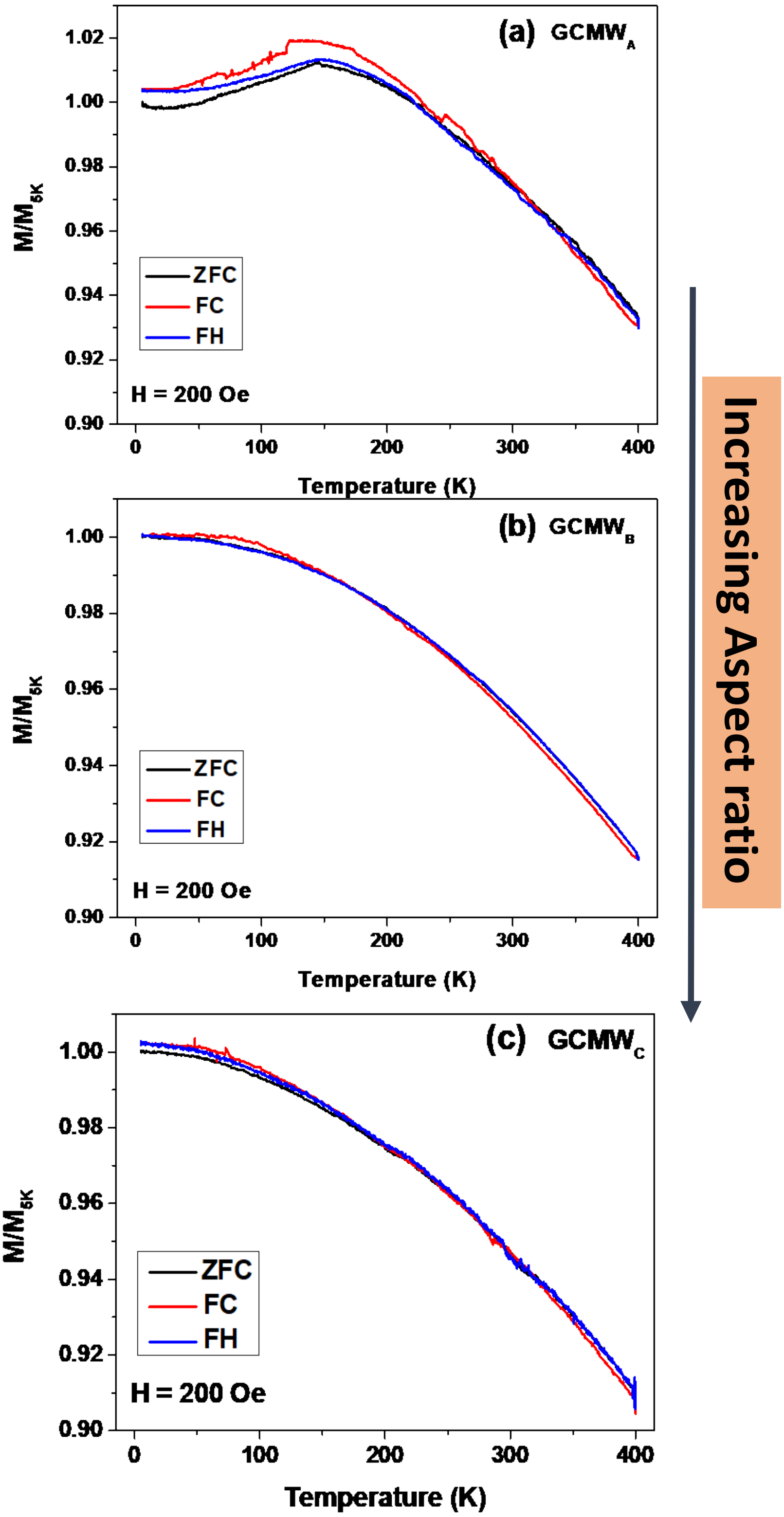

Figure 6. Temperature dependence of magnetization measured for Co_2FeSi glass-coated microwires (a) $GCMW_A$, (b) $GCMW_B$ and (c) $GCMW_C$ with applied external magnetic field 200 Oe.

We believe that increasing the ρ - ratio of Co_2FeSi glass-coating microwires affects recrystallization, atomic ordering, and stress reduction. Furthermore, for samples with a high ρ -ratio, the induced $L2_1$/B2 and A2 cubic structure types generate a strong magneto crystalline anisotropy, explaining the the behavior of magnetic properties such as H_c, M_r, H_k, and thermomagnetic curves with temperature. In fact, as shown in several previous publication, the internal stresses values are affected by the ρ – ratio: the lower the ρ – ratio, the higher the internal stresses related to the presence of the glass-coating [50–52]. On the other hand, the glass-coating thermal conductivity can affect the quenching rate of the metallic nucleus: lower quenching rate must be at the origin of higher crystallinity of the microwires with relatively thick glass-coating [27].

Resuming, Co_2FeSi-glass-coated microwires are an excellent candidate for a wide range of industrial applications, and specially for sensors, due to their perfect squared loops at a wide temperature range and homogenous thermomagnetic behavior with temperature.

4. Conclusions

In summary, we have fabricated Co_2FeSi glass-coated microwires with different geometrical aspect ratios. A strong influence of the geometric aspect ratio on the magnetic and structural properties is observed and discussed. The increase in the aspect ratio correlates with the increasing degree of crystallinity. Promising coercivity and normalized remnant stability with temperature are found for Co_2FeSi-glass coated microwires with a high aspect ratio. For the sample with the lowest aspect ratio, the thermomagnetic curves show large irreversibility with a blocking temperature of T = 150 K. The induced crystal structure of the A2-type gives rise to a high cubic magnetocrystalline anisotropy that controls the magnetic behavior of Co_2FeSi glass-coated microwires and makes it a suitable candidate for magnetic sensing.

Author Contributions: Conceptualization, M.S. and A.Z.; methodology, V.Z.; validation, M.S., V.Z. and A.Z.; formal analysis, M.S and A.W.; investigation, M.S., A.W, and A.Z.; resources, V.Z. and A.Z.; data curation, M.I; writing—original draft preparation, M.S., A.W. and A.Z.; writing—review and editing, M.S., R.A.L and A.Z.; visualization, M.S., A.W., and M.I; supervision, R.A.L and A.Z.; project administration, V.Z. and A.Z.; funding acquisition, V.Z., and A.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Spanish MICIN, under PID2022-141373NBI00, by EU under "INFINITE" (Horizon Europe) project and by the Government of the Basque Country, under PUE_2021_1_0009 and Elkartek (MINERVA, ZE-KONP and MAGAF) projects and by under the scheme of "Ayuda a Grupos Consolidados" (Ref.: IT1670-22). MS wish to acknowledge the funding within the Maria Zambrano contract by the Spanish Ministerio de Universidades and European Union –Next Generation EU ("Financiado por la Unión Europea-Next Generation EU"). We also wish to thank the administration of the University of the Basque Country, which not only provides very limited funding, but even expropriates the resources received by the research group from private companies for the research activities of the group. Such interference helps keep us on our toes.

Institutional Review Board Statement: "Not applicable.".

Informed Consent Statement: "Not applicable.".

Data Availability Statement: "Not applicable.".

Acknowledgments: The authors are thankful for the technical and human support provided by SGIker of UPV/EHU (Medidas Magnéticas Gipuzkoa) and European funding (ERDF and ESF).

Conflicts of Interest: "The authors declare no conflict of interest.".

References

1. Li, T.; Pickel, A.D.; Yao, Y.; Chen, Y.; Zeng, Y.; Lacey, S.D.; Li, Y.; Wang, Y.; Dai, J.; Wang, Y.; et al. Thermoelectric properties and performance of flexible reduced graphene oxide films up to 3,000 K. *Nat. Energy* **2018**, *3*, 148–156.
2. Hoop, M.; Ribeiro, A.S.; Rösch, D.; Weinand, P.; Mendes, N.; Mushtaq, F.; Chen, X.Z.; Shen, Y.; Pujante, C.F.; Puigmartí-Luis, J.; et al. Mobile Magnetic Nanocatalysts for Bioorthogonal Targeted Cancer Therapy. *Adv. Funct. Mater.* **2018**, *28*, 1705920.
3. Salaheldeen, M.; Vega, V.; Fernández, A.; Prida, V.M. Anomalous In-Plane Coercivity Behaviour in Hexagonal Arrangements of Ferromagnetic Antidot Thin Films. *J. Magn. Magn. Mater*. **2019**, 491, 165572.
4. Salaheldeen, M.; Nafady, A.; Abu-Dief, A.M.; Díaz Crespo, R.; Fernández-García, M.P.; Andrés, J.P.; López Antón, R.; Blanco, J.A.; Álvarez-Alonso, P. Enhancement of Exchange Bias and Perpendicular Magnetic

Anisotropy in CoO/Co Multilayer Thin Films by Tuning the Alumina Template Nanohole Size. *Nanomaterials* **2022**, *12*, 2544. https://doi.org/10.3390/nano12152544
5. Salaheldeen, M.; Martínez-Goyeneche, L.; Álvarez-Alonso, P.; Fernández, A. Enhancement the Perpendicular Magnetic Anisotropy of Nanopatterned Hard/Soft Bilayer Magnetic Antidot Arrays for Spintronic Application. *Nanotechnology* **2020**, 31, 485708.
6. Skjærvø, S.H.; Marrows, C.H.; Stamps, R.L.; Heyderman, L.J. Advances in artificial spin ice. *Nat. Rev. Phys*. **2019**, 2, 13–28.
7. Maniv, E.; Murphy, R.A.; Haley, S.C.; Doyle, S.; John, C.; Maniv, A.; Ramakrishna, S.K.; Tang, Y.-L.; Ercius, P.; Ramesh, R.; et al. Exchange bias due to coupling between coexisting antiferromagnetic and spin-glass orders. *Nat. Phys*. **2021**, *17*, 525–530.
8. Salaheldeen, M.; Abu-Dief, A.M.; Martínez-Goyeneche, L.; Alzahrani, S.O.; Alkhatib, F.; Álvarez-Alonso, P.; Blanco, J.Á. Dependence of the Magnetization Process on the Thickness of $Fe_{70}Pd_{30}$ Nanostructured Thin Film. *Materials* **2020**, 13, 5788. https://doi.org/10.3390/ma13245788.
9. Salaheldeen, M.; Méndez, M.; Vega, V.; Fernández, A.; Prida, V.M. Tuning Nanohole Sizes in Ni Hexagonal Antidot Arrays: Large Perpendicular Magnetic Anisotropy for Spintronic Applications. *ACS Appl. Nano Mater*. **2019**, 2, 1866–1875.
10. Elphick, K.; Frost, W.; Samiepour, M.; Kubota, T.; Takanashi, K.; Sukegawa, H.; Mitani, S.; Hirohata, A. Heusler Alloys for Spintronic Devices: Review on Recent Development and Future Perspectives. *Sci. Technol. Adv. Mate*. **2021**, 22, 235–271, doi:10.1080/14686996.2020.1812364.
11. Sheron Tavares; Kesong Yang; Marc A. Meyers. Heusler alloys: Past, properties, new alloys, and prospects. *Progress in Materials Science*, **2023**, 132, 101017, http://doi.org/10.1016/j.pmatsci.2022.101017.
12. De Groot, R.A.; Mueller, F.M.; Engen, P.G.V.; Buschow, K.H.J. New Class of Materials: Half-Metallic Ferromagnets. *Phys. Rev. Lett*. **1983**, *50*, 2024.
13. Bai, Z.; Shen, L.E.I.; Han, G.; Feng, Y.P. Data Storage: Review of Heusler Compounds. *Spin* **2012**, 2, 1230006, doi:10.1142/S201032471230006X.
14. Balke, B.; Wurmehl, S.; Fecher, G.H.; Felser, C.; Kübler, J. Rational Design of New Materials for Spintronics: Co_2FeZ (Z=Al, Ga, Si, Ge). *Sci Technol Adv Mater* **2008**, 9, doi:10.1088/1468-6996/9/1/014102.
15. Hirohata, A.; Sagar, J.; Lari, L; et al. Heusler-alloy films for spintronic devices. *Appl. Phys. A*, **2013** 111, 423–430, https://doi.org/10.1007/s00339-013-7679-2.
16. Li, P.; Koo, J.; Ning, W.; Li, J.; Miao, L.; Min, L.; Zhu, Y.; Wang, Y.; Alem, N.; Liu, C.X.; et al. Giant Room Temperature Anomalous Hall Effect and Tunable Topology in a Ferromagnetic Topological Semimetal Co_2MnAl. *Nat. Commun*. **2020**, 11, 1–8, doi:10.1038/s41467-020-17174-9.
17. Jourdan, M.; Minár, J.; Braun, J.; Kronenberg, A.; Chadov, S.; Balke, B.; Gloskovskii, A.; Kolbe, M.; Elmers, H.J.; Schönhense, G.; et al. Direct Observation of Half-Metallicity in the Heusler Compound Co_2MnSi. *Nat. Commun*. **2014**, 5, 3974
18. Guillemard, C.; Petit-Watelot, S.; Pasquier, L.; Pierre, D.; Ghanbaja, J.; RojasSánchez, J.C.; Bataille, A.; Rault, J.; le Fèvre, P.; Bertran, F.; et al. Ultralow Magnetic Damping in Co_2Mn-Based Heusler Compounds: Promising Materials for Spintronics. *Phys. Rev. Appl*. **2019**, 11, 064009.
19. Nehla, P.; Ulrich, C.; Dhaka, R.S. Investigation of the Structural, Electronic, Transport and Magnetic Properties of Co_2FeGa Heusler Alloy Nanoparticles. *J. Alloys Compd*. **2019**, *776*, 379–386.
20. Patra, N.; Prajapat, C.L.; Babu, P.D.; Rai, S.; Kumar, S.; Jha, S.N.; Bhattacharyya, D. Pulsed Laser Deposited Co_2FeSi Heusler Alloy Thin Films: Effect of Different Thermal Growth Processes. *J. Alloys Compd*. **2019**, *804*, 470–485.
21. Salaheldeen, M.; Ipatov, M.; Zhukova, V.; García-Gomez, A.; Gonzalez, J.; Zhukov, A. Preparation and magnetic properties of Co_2-based Heusler alloy glass-coated microwires with high Curie temperature. *AIP Adv*. **2023**, 13, 025325.
22. Salaheldeen, M.; Garcia, A.; Corte-Leon, P.; Ipatov, M.; Zhukova, V.; Zhukov, A. Unveiling the Effect of Annealing on Magnetic Properties of Nanocrystalline Half-Metallic Heusler Co_2FeSi Alloy Glass-Coated Microwires. *J. Mater. Res. Technol*. **2022**, 20, 4161–4172.
23. Salaheldeen, M.; Ipatov, M.; Corte-Leon, P.; Zhukova, V.; Zhukov, A. Effect of Annealing on the Magnetic Properties of Co_2MnSi-Based Heusler Alloy Glass-Coated Microwires. *Metals* **2023**, 13, 412. https://doi.org/10.3390/met13020412.
24. Salaheldeen, M.; Garcia-Gomez, A.; Ipatov, M.; Corte-Leon, P.; Zhukova, V.; Blanco, J.M.; Zhukov, A. Fabrication and MagnetoStructural Properties of Co_2-Based Heusler Alloy Glass-Coated Microwires with High Curie Temperature. *Chemosensors* **2022**, 10, doi:10.3390/chemosensors10060225.
25. Khovaylo, V.V.; Rodionova, V.V.; Shevyrtalov, S.N.; Novosad, V. Magnetocaloric Effect in "Reduced" Dimensions: Thin Films, Ribbons, and Microwires of Heusler Alloys and Related Compounds. *Phys. Status Solidi* **2014**, *251*, 2104–2113.

Preprints (www.preprints.org) | NOT PEER-REVIEWED | Posted: 6 April 2023 doi:10.20944/preprints202304.0071.v1

26. Belmeguenai, M.; Tuzcuoglu, H.; Gabor, M.S.; Petrisor, T.; Tiusan, C.; Zighem, F.; Chérif, S.M.; Moch, P. Co_2FeAl Heusler Thin Films Grown on Si and MgO Substrates: Annealing Temperature Effect. *J. Appl. Phys.* **2014**, *115*, 043918.
27. Zhukov, A.; Corte-Leon, P.; Gonzalez-Legarreta, L.; Ipatov, M.; Blanco, J.M.; Gonzalez, A.; Zhukova, V. Advanced Functional Magnetic Microwires for Technological Applications. *J. Phys. D Appl. Phys*. **2022**, 55, 253003, doi:10.1088/1361-6463/AC4FD7.
28. H. Chiriac H and T.A. Ovari, Amorphous glass-covered magnetic wires: Preparation, properties, applications. *Prog. Mater. Sci*., **1996**, 40, 333-407
29. Mitxelena-Iribarren, J. Campisi, I. Martínez de Apellániz, S. Lizarbe-Sancha, S. Arana, V. Zhukova, M. Mujika, A. Zhukov, Glass-coated ferromagnetic microwire-induced magnetic hyperthermia for in vitro cancer cell treatment, *Mater. Sci. Eng. C* **2020**, 106 , 110261, Doi: 10.1016/j.msec.2019.110261.
30. Salaheldeen, M.; Garcia-Gomez, A.; Corte-León, P.; Gonzalez, A.; Ipatov, M.; Zhukova, V.; Gonzalez, J.M.; López Antón, R.; Zhukov, A. Manipulation of Magnetic and Structure Properties of Ni_2FeSi Glass-Coated Microwires by Annealing. *J Alloys Compd* **2023**, 942, 169026, https://doi.org/10.1016/j.jallcom.2023.169026.
31. Salaheldeen, M.; Talaat, A.; Ipatov, M.; Zhukova, V.; Zhukov, A. Preparation and Magneto-Structural Investigation of Nanocrystalline CoMn-Based Heusler Alloy Glass-Coated Microwires. *Processes*, **2022**, 10, 2248, doi:10.3390/PR10112248.
32. Salaheldeen, M.; Garcia-Gomez, A.; Corte-Leon, P.; Ipatov, M.; Zhukova, V.; Gonzalez, J.; Zhukov, A. Anomalous Magnetic Behavior in Half-Metallic Heusler Co_2FeSi Alloy Glass-Coated Microwires with High Curie Temperature. *J Alloys Compd* **2022**, 923, 166379, doi:10.1016/J.JALLCOM.2022.166379.
33. Hennel, M.; Varga, M.; Frolova, L.; Nalevanko, S.; Ibarra-Gaytán, P.; Vidyasagar, R.; Sarkar, P.; Dzubinska, A.; Galdun, L.; Ryba, T.; et al. Heusler-Based Cylindrical Micro- and Nanowires. *Phys. Status Solidi A* **2022**, 219, 2100657.
34. Salaheldeen, M.; Wederni, A.; Ipatov, M.; Gonzalez, J.; Zhukova, V.; Zhukov, A. Elucidation of the Strong Effect of the Annealing and the Magnetic Field on the Magnetic Properties of Ni_2-Based Heusler Microwires. Crystals 2022, 12, 1755.https://doi.org/10.3390/cryst12121755.
35. Salaheldeen, M.; Wederni, A.; Ipatov, M.; Zhukova, V.; Zhukov, A. Preparation and Magneto-Structural Investigation of High Ordered (L21 Structure) Co_2MnGe Microwires. Preprints.org 2023, 2023020494. https://doi.org/10.20944/preprints202302.0494.v1.
36. A.F. Cobeño, A. Zhukov, A.R. de Arellano - Lopez, F. Elías. J.M. Blanco, V. Larin and J. González, "Physical properties of nearly zero magnetostriction Co-rich glass-coated amorphous microwires" J. Mater. Res. 14 (1999) 3775-3783, doi: https://doi.org/10.1088/0022-3727/32/24/308
37. Ulitovsky AV, Maianski I M, Avramenco A I 1960 Method of continuous casting of glass coated microwire, *Patent* No 128427 (USSR), 15.05.60, Bulletin, No 10, p. 14
38. Zhukov, A.; Ipatov, M.; Talaat, A.; Blanco, J.M.; Hernando, B.; Gonzalez-Legarreta, L.; Suñol, J.J.; Zhukova, V. Correlation of Crystalline Structure with Magnetic and Transport Properties of Glass-Coated Microwires. *Crystals* **2017**, *7*, 41, doi:10.3390/cryst7020041.
39. Chiriac, H.; Lupu, N.; Stoian, G.; Ababei, G.; Corodeanu, S.; Óvári, T.A. Ultrathin Nanocrystalline Magnetic Wires. *Crystals* **2017**, *7*, 48, doi:10.3390/CRYST7020048.
40. Corte-Leon, P.; Zhukova, V.; Ipatov, M.; Blanco, J.M.; González, J. Churyukanova, M.; Taskaev, S.; Zhukov, A. The effect of annealing on magnetic properties of "Thick" microwires. *J. Alloys Compd*. **2020**, *831*, 150992.
41. Goto, T.; Nagano, M.; Wehara, N. Mechanical properties of amorphous $Fe_{80}P_{16}C_3B_1$ filament produced by glass-coated melt spinning. *Trans. JIM* **1977**, *18*, 759–764.
42. Zhukova, V.; Cobeño, A.F.; Zhukov, A.; de Arellano Lopez, A.R.; López-Pombero, S.; Blanco, J.M.; Larin, V.; Gonzalez, J. Correlation between magnetic and mechanical properties of devitrified glass-coated $Fe_{71.8}Cu_1Nb_{3.1}Si_{15}B_{9.1}$ microwires. *J. Magn. Magn. Mater*. **2002**, *249*, 79–84.
43. Talaat, A.; Alonso, J.; Zhukova, V.; Garaio, E.; García, J.A.; Srikanth, H.; Phan, M.H.; Zhukov, A. Ferromagnetic Glass-Coated Microwires with Good Heating Properties for Magnetic Hyperthermia. *Sci. Rep*. **2016**, 6, 39300.
44. Kozejova, D.; Fecova, L.; Klein, P.; Sabol, R.; Hudak, R.; Sulla, I.; Mudronova, D.; Galik, J.; Varga, R. Biomedical applications of glass-coated microwires. *J. Magn. Magn. Mater.* **2019**, *470*, 2-5.
45. Kudryavtsev, Y. V.; Lee, Y.P.; Lee, N.N.; Huang, M.D. Effect of Structural Disordering on the Magnetic, Magneto-Optical and Optical Properties of the Ni2MnIn Heusler Alloy Films. *Mater. Sci. Forum.* **2005**, 480–481, 623–628, doi:10.4028/WWW.SCIENTIFIC.NET/MSF.480-481.623.
46. Dubowik, J.; Kudryavtsev, Y.; Lee, Y.P.; Lee, N.N.; Hong, B.S. INFLUENCE OF STRUCTURAL ORDER ON MAGNETIC PROPERTIES OF Ni_2MnIn HEUSLER ALLOY FILMS. *Mol. Phys. Report.* **2004**, 40, 55–61.
47. Wang, K.; Xu, Z.; Fu, X.; Lu, Z.; Xiong, R. Magnetic and Structural Properties of Sputtered Thick Co_2FeSi Alloy Films. *J. Magn. Magn. Mater*. **2023**, 570, 170557, doi:10.1016/J.JMMM.2023.170557.

48. Zhang, X.; Han, L.; Dehm, G.; Liebscher, C.H. Microstructure and Physical Properties of Dual-Phase Soft Magnetic Fe-Co-Ti-Ge *Alloys. J. Alloys Compd*. **2023**, 945, 169282, doi:10.1016/J.JALLCOM.2023.169282.
49. Zhang, H.G.; Song, B.T.; Chen, J.; Yue, M.; Liu, E.K.; Wang, W.H.; Wu, G.H. Magnetization Variation in Fe–Cr-Ga System. *Intermetallics* **2019**, 113, 106580, doi:10.1016/J.INTERMET.2019.106580.
50. Xiao, A.; Zhou, Z.; Qian, Y.; Wang, X. Direct Observation of Evolution from Amorphous Phase to Strain Glass. *Mater*. **2022**, Vol. 15, Page 7900 2022, 15, 7900, doi:10.3390/MA15227900.
51. Zhukov, J. Gonzalez, A. Torcunov, E. Pina, M.J. Prieto, A.F. Cobeño, J.M. Blanco, V. Larin and S. Baranov, Ferromagnetic resonance and Structure of Fe-based glass-coated Microwires, J. Magn. Magn. Mater., 203 (1999) 238-240.
52. H. Chiriac, T.A. Ovari, Gh. Pop, F. Barariu, Internal stresses in highly magnetostrictive glass-covered amorphous wires, J. Magn. Magn. Mater., 160 (1996) 237-238.