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Elucidation of the Strong Effect of the Annealing and the Magnetic Field on the Magnetic Properties of Ni_2-Based Heusler Microwires

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Abstract: We study the effect of annealing and the applied magnetic field from 50 Oe to 20 kOe on the magneto-structural behavior of Ni_2FeSi-based Heusler microwires fabricated by using Taylor-Ulitovsky technique. Using the XRD analysis, a strong effect of annealing, manifested as the development of the crystallization process, was observed. The average grain size and crystalline phase content of annealed sample increase from 21.3 nm and 34% to 32.8 nm and 79%, respectively, as-compared to the as-prepared one. In addition, upon annealing, phase transforms into a monoclinic martensitic structure with a modulation of 10 M, which cannot be found in the as-prepared sample. Concerning the magnetic properties, both samples show ferromagnetic behavior below and above the room temperature, where the Curie temperature of Ni_2FeSi is higher than the room temperature. The induced secondary phases have a noticeable effect on the magnetic behavior of the annealed sample, where a high normalized saturation magnetization (NM_s) and low normalized reduced remenance ($M_r = M/M_{5K}$), compared to the as-prepared have been detected. Additionally, the coercivity of annealed sample shows one flipping point at 155 K where its behavior changes with temperature. Meanwhile, the as-prepared sample show two flipped point at 205 K and 55 K. A mismatch between field cooling (FC) and field heating (FH) magnetization curves with temperature has been detected for annealed sample at low applied magnetic field. The difference in magnetic and structure behavior of Ni_2FeSi microwires sample is discussed considering the effect of induced internal stresses by the presence of a glass coating and the recrystallization and stresses relaxation upon annealing.

Keywords: Heusler alloys; glass-coated microwires; magnetic field; annealing; Taylor-Ulitovsky

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1. Introduction

Micro/nanostructured ferromagnetic materials with different physical forms gained especial consideration of researchers due to their possible applications in the field of spintronics, magneto optics, and thermoelectricity application [1–5].

Heusler made the important discovery that ferromagnetic alloys, often known as Heusler alloys, can be created from nonmagnetic materials in the 20th century. More than a thousand Heusler alloys are being studied because of their exceptional electronic, magnetic, mechanical, and electrical capabilities that can be observed by this family of materials [6]. Perfect lattice matching with various substrates, variable Curie temperature, Tc, and intermetallic controllability for spin density of states at the Fermi energy level, where approximately 100% of spin polarization near the Fermi level is recorded, are some of the outstanding benefits and remarkable features [7–13]. Thus, Heusler alloys are a strong contender for the upcoming wave of multifunction spintronic applications due to these benefits [7–9,12].

The development of high spin polarization in the highly ordered $L2_1$ crystal phase structure is one of the most important characteristics demonstrated by half and full metallic Heusler alloys. Heusler alloys can appear in two crystalline phases: the high symmetry austenite phase, with the simplest structure presented by a cubic $L2_1$ or B2 structure, and the less symmetrical martensite phase which can present a tetragonal, monoclinic, or orthorhombic structure (with or without structural modulation) [9–13].

In order to obtain the necessary structural ordering i.e., $L2_1$ and prevent the formation of disordered structures, such as B2, A2, and DO3, which may arise during the alloy manufacturing process, the half and full metallic Heusler alloys prepared by physical vapor deposition, i.e., thin films forms, ball milling, or by arc melting, are crucial [14,15]. To avoid the formation of unneeded phase structures, lengthy annealing times at high temperatures are used. To alleviate the aforementioned drawbacks, Heusler alloy manufacturing has recently switched to a quick quenching process [16,17]. The advantage of the rapid quenching process is that it allows to fabricate various amorphous and crystalline materials in various forms, such as ribbons and microwires, quickly, easily, simply, and in one step [18,19].

The magneto-mechanical features of Ni-based Heusler alloys, which include magnetic field induced superelasticity and magnetic shape memory effect, are remarkable and promising for applications in sensing and data storage [20,21]. Additionally, a significant spin polarization was expected by theoretical calculations for Ni_2Fe-based Heusler alloys [14]. A fresh study on the simple way for making thin Heusler wires into spintronic devices is sought from a business standpoint. Racetrack memory, domain wall logic, and oscillators are only a few examples of the numerous spintronic applications based on tiny magnetic wires [22–24]. A thin magnetic wire with strong spin polarization is necessary for all of these applications.

Due to their reduced dimensions, freedom to customize and manufacture their magnetic, electric, mechanical, and structural characteristics, Heusler alloy-based microwires have promising characteristics [19,25]. The Taylor-Ulitovsky method, known since the 1960s, is one of the quick quenching procedures used to produce Heusler alloys glass-coated microwires. Using this technique, glass-coated microwires with metallic nucleus varying in diameter from 0.5 to 100 μm may be produced [19,26]. The major advantage of this low-cost method is that it enables quick (up to a few hundred meters/min) manufacturing of thin and long (a few kilometers) microwires with an extended geometric range. This approach also yields glass-coated microwires with improved mechanical characteristics [27]. Additionally, the availability of a biocompatible thin, flexible, insulating, and highly transparent glass covering would be beneficial for biomedical applications [27–29]. As a result, the Ni_2-based Heusler microwires are a potential smart material for a variety of device applications. The manufacturing, structural, mechanical, and magnetic characterization of Ni_2-based Heusler glass-covered microwires have not yet been extensively studied, as far as we are aware. To show their potential uses in cutting-edge microspintronics, the structural and magnetic characteristics of Ni_2FeSi microwires will therefore be the main focus of the present work.

In the current article, we describe a structural and magnetic characterization of Ni_2FeSi alloy microwires with an emphasis on how the annealing time affect the physical (magnetic and structure) characteristics of these alloys. The magnetic behavior during heating and cooling in the temperature range of 5 K to 400 K and magnetic field (50 Oe to 20 kOe) are given particular consideration. We demonstrate that distinct magnetic phases with magnetic properties that do not present in the as-prepared samples are produced during annealing conditions. In addition, different magnetic behavior was observed depending on the external applied magnetic field and the temperature. The annealed sample shows gradual uniform magnetic dependence by increasing the applied magnetic field. Ni_2FeSi can be used for spintronic applications by fine-tuning its physical characteristics under annealing conditions.

2. Materials and Methods

The Ni_2FeSi ingot has been prepared by melting high purity Ni (99.99%), Fe (99.99%), and Si (99.99%) supplied by Technoamorf S.R.L. Co., (TURKU, Finland), in a traditional arc furnace with argon as the environment to prevent oxide formation during the melting process. To produce an alloy with high homogeny, the melting procedure was repeated five times. Then, using the EDX/SEM setup as described in our earlier study, we evaluated the chemical composition [30,31]. After validating the chemical composition, we prepared the glass-coated microwire using the Taylor-Ulitovsky. More information on the process of glass-coated microwires preparation was previously documented and discussed elsewhere [26,32–34]. The total Ni_2FeSi glass-coated microwire diameter (metallic nucleus and Duran glass coating), D, is around 20 µm, while the inner metallic nucleus diameter, d, is 9 µm. The Ni_2FeSi microwire was fabricated and then annealed for one hour at 973 K. The structure analysis and chemical composition of the metallic nucleus have been performed using EDX/SEM, as previously reported elsewhere [30,35]. PPMS (Physical Property Magnetic System, Quantum Design Inc., San Diego, CA, USA) was used to study the magnetic properties at temperatures between 5 and 400 K and a variety of applied magnetic fields (H = 50 Oe to 20 kOe). The results are provided in terms of the normalized magnetization, M/M_{5K}, where M_{5K} is the magnetic moment measured at 5 K with a magnetic field equal to 20 kOe. The microwire bunch was employed for magnetic measurements revealing relative changes of magnetization.

3. Results

3.1. Chemeical and Structure Analysis

To check the chemical composition of Ni_2FeSi glass-coated microwires we performed EDX/SEM analysis and the output results are listed in Table 1. The composition of the metallic nucleus was found to be somewhat different from the stoichiometric one using the EDX data from Table 1 (Ni_2FeSi). This slight variance was due to the peculiarities of the preparation procedure, which included alloy melting and casting. We examined the nominal composition for 10 locations to determine the amount of difference. The actual 2:1 ratio for Ni and Fe was verified for all locations, with an atomic average $Ni_{44}Fe_{23}Si_{33}$. Because of the interfacial layer between the glass covering and the metallic nucleus, a high Si ratio was detected.

Table 1. Atomic percentage of Ni, Fe and Si elemental composition in Ni_2FeSi glass-coated microwires.

EDX Spectrum	Ni (at %)	Fe (at %)	Si (at %)
Average	44	23	33

To study the effect of annealing condition on the structure properties of Ni_2FeSi glass-coated microwires, we performed the XRD analysis for as-prepared and annealed Ni_2FeSi microwires samples.

Figure 1 illustrates the XRD analysis of as prepared and annealed Ni_2FeSi samples. The XRD measurements was carried out at room temperature. As shown in Figure 1, a noticeable change in the structure characterization is observed. First, both diffractograms display a wide halo at $2\theta \approx 23°$, related to the contribution of amorphous glass coating as reported in our previous works [30,31,35]. The as-prepared diffractogram shows strong single XRD peak at $2\theta \approx 46°$ as a (220) reflection peak. The presence of the (220) and (111) superlattice reflections confirms as the ordered of $L2_1$ cubic structure. The lattice parameter, a, is 0.578 nm with space group Fm-3m.

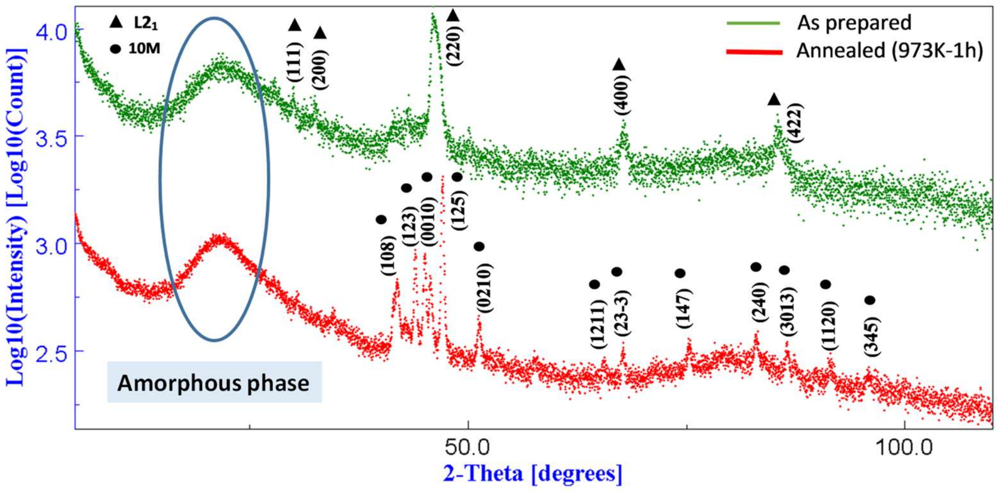

Figure 1. XRD spectra of as-prepared and annealed Ni_2FeSi glass-coated microwires samples.

For annealed sample (at 973 K for 1 h), several peaks are observed in comparison to the as-prepared one. The reflection peaks are recognized to be a monoclinic structure with modulation. Thus, XRD patterns of Ni_2FeSi alloy annealed at 973K for 1 h and measured at room temperature demonstrates a modulated martensitic phase, which present a five-layered monoclinic 10 M structure, with cell parameter: a = 0.514 nm, b = 0.499 nm, c = 2.506 nm, and β = 92.26°. Consequently, by increasing the temperature, the cubic high temperature parent austenite phase transforms into a monoclinic martensitic structure with modulation 10 M. In other words, when applying an external stress (temperature change in our case), the martensitic domains move and permit the creation of a large macroscopic deformations in the sample. This deformation does not require a huge amount of energy because only the domain walls move [36].

Mostly, austenite to martensite undergoes a solid–solid transformation, displaying a first-order structural transformation and leading to a homogeneous deformation of the structure mainly made by distortion [37]. This transition is able to be displacive because it is diffusion less (without displacement of sets of atoms).

To estimate the amount of change in the structure of Ni_2FeSi microwires under the annealing condition, we calculated the crystalline phase content and average of the grain size using the equation reported in our previous work [30,35]. We observed the average crystallite size and the crystalline phase content of the annealed sample increase from 21.3 nm and 34% to 32.8 nm and 79%, respectively, as compared to the as-prepared one.

3.2. Magnetic Properties

As we mentioned in the experimental part, the magnetic properties has been investigated by using PPMS at a wide temperature, T, (5–400 K) range and applied magnetic field, H, (50–20,000 Oe). In our investigation, we focused on the magnetization, M, measurements parallel to the wires axis where the easy magnetization axis is expected. In addition, we performed a normalization of the magnetization for all magnetic measurements to magnetization value at 5K, M/M_{5K} ratio (M_{5K} is the highest magnetic moment measuring at 5K) to avoid the expected errors with evaluation of the magnetization saturation (related to the composite character of studied microwires) of the annealed and the as-prepared samples, where a small errors in the calculation may lead to the misunderstanding of the major differences of the magnetic properties between the as-prepared and the annealed sample. The M/M_{5K}-H loops shown in Figure 2 illustrate the evolution of the magnetic behavior upon variation the temperature for the as-prepared and annealed samples. All samples show ferromagnetic ordering as their Curie temperatures are above the room temperature. From the comparison of the M/M_{5K}-H loops of as-prepared (black loops) and annealed (red loops), we can deduce that the M/M_{5K}-H loops of the as-prepared samples show more squared shape than those of the annealed samples. In addition, the normalized magnetization saturation of annealed samples is observed at higher magnetic field and higher M/M_{5K} value in current case as compared to the as-prepared samples for measuring temperature from 305 to 55 K (see Figure 2a–e). While the normalized saturation magnetization of as-prepared loops became higher than that of the annealed one at 5 K (see Figure 2f). Additionally, the axial magnetic anisotropy field shows the same tendency as the saturation field. These observations can be attributed to an onset of different magnetic phase for the annealed sample, which does not exist in the as-prepared sample. Indeed, the change in the saturation field and axial magnetic anisotropy field are strongly related to the change in the structure i.e., change in the magnetic response for both as-prepared and annealed Ni_2FeSi microwires. Additionally, internal stresses relaxation upon annealing can also affect M/M_{5K}-H loops character.

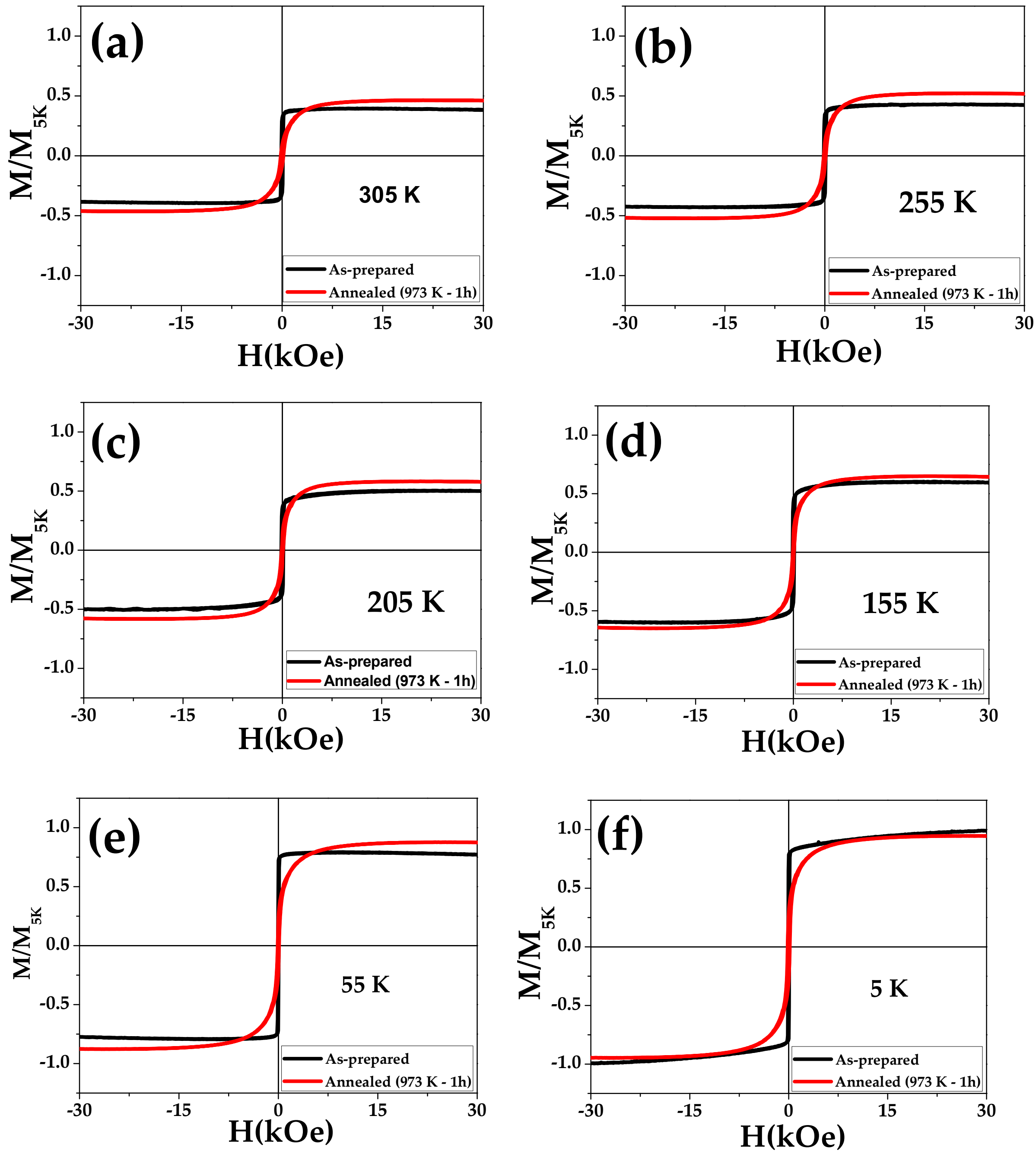

Figure 2. Magnetization curves M/M_{5K} (H) of as-prepared and annealed Ni_2FeSi glass-coated microwires measured at maximum field 30 kOe.

More details on the magnetic properties can be extracted from the M-H loops for the as-prepared and annealed samples. In Figure 3, we plotted the behavior of the coercivity, reduced remanence, (M_r = M/M_{5K}), and the normalized saturation magnetization values (NM_s), defined as the saturated value of M/M_{5K}(H) loops for as-prepared and annealed samples with variation in the temperature. For the temperature dependence of magnetic properties both the as-prepared and annealed sample show interesting magnetic behavior. As indicated in the Figure 3a, the annealed sample show higher coercivity, H_c, than the as-prepared sample at room temperature. However, the H_c sharply decreases when T decreases, reaching the lowest value at T = 155 K and then starts to increase with a further decrease in the temperature reaching the maximum value at T = 5 K. Different scenario has been reported for the coercivity dependence with temperature for as-prepared sample, where a monotonic increase with decreasing the temperature from 305 to 255 K has been observed. Then the coercivity starts to decrease with decreasing temperature from 255 to 55 K. Finally, it increases with a decrease in the temperature from 55 to 5 K, i.e., in the as-prepared sample two filliped points at 255 K and 55 K are observed, where the coercivity tendency with temperature changes. While the annealed sample shows one filliped point at T = 155 K. The unusual behavior of the coercivity with temperature has been reported previously in another Heusler-based glass-coating microwires [30,31,35]. In addition, the M_r of annealed sample with the temperature shows a sharp drop in its values compared to the as-prepared sample as illustrated in Figure 3b. The sharp drops in the M_r can be related to the growth of the out-of-axis magnetization of microwires. Unfortunately, at current moment we do not have the possibility to evaluate the angle of the magnetization tilting for the annealed sample. Here, we want to underline the strong effect of the annealing on the magnetic behavior of Ni_2FeSi glass-coated microwires as compared to non-annealed sample. Moreover, as we mentioned above, the normalized saturation magnetization dependence with the temperature for both studied samples is shown in Figure 3c. As we can see from Figure 3, both samples behave in a similar way, showing the NM_s increase by decreasing the temperature, usually observed in the ferromagnetic materials. On the other hand, annealed sample shows higher NM_s values for almost the whole temperature range, as shown in Figure 3c. It is worth mentioning that the temperature dependence of the magnetization can provide useful information on short-range atomic arrangements in even disordered magnetic materials [38–40]. Thus, the "flattening" of the temperature dependence of magnetization, typically observed in amorphous alloys [38–40], is commonly attributed to fluctuations in the exchange interactions typical for the amorphous alloys. Accordingly, higher NM_s values must be related to the devitrification of amorphous matrix and atopic disorder decrease upon annealing.

The main point in the anomalous changing of the axial coercivity, reduced remanence, anisotropy field, and saturation magnetization of Heusler-based glass-coated microwires is the strong mechanical stress induced during the preparation of glass-coating microwires [41]. In addition, the internal and external mechanical stresses are very sensitive to temperature. In the current case, we deal with Ni_2FeSi glass-coated microwires with different microstructures, as explained in the structure part in this manuscript. The annealing condition strongly changes the microstructure properties by inducing additional phase structure with different magnetic properties (see Figures 2 and 3).

As discussed elsewhere, field cooling (FC) and field heating (FH) are powerful tools for studies of nanostructured magnetic materials [18,35]. Accordingly, we performed field cooling (FC) and field heating protocols at different applied magnetic field to evaluate the behavior of studied samples under low and high external magnetic field.

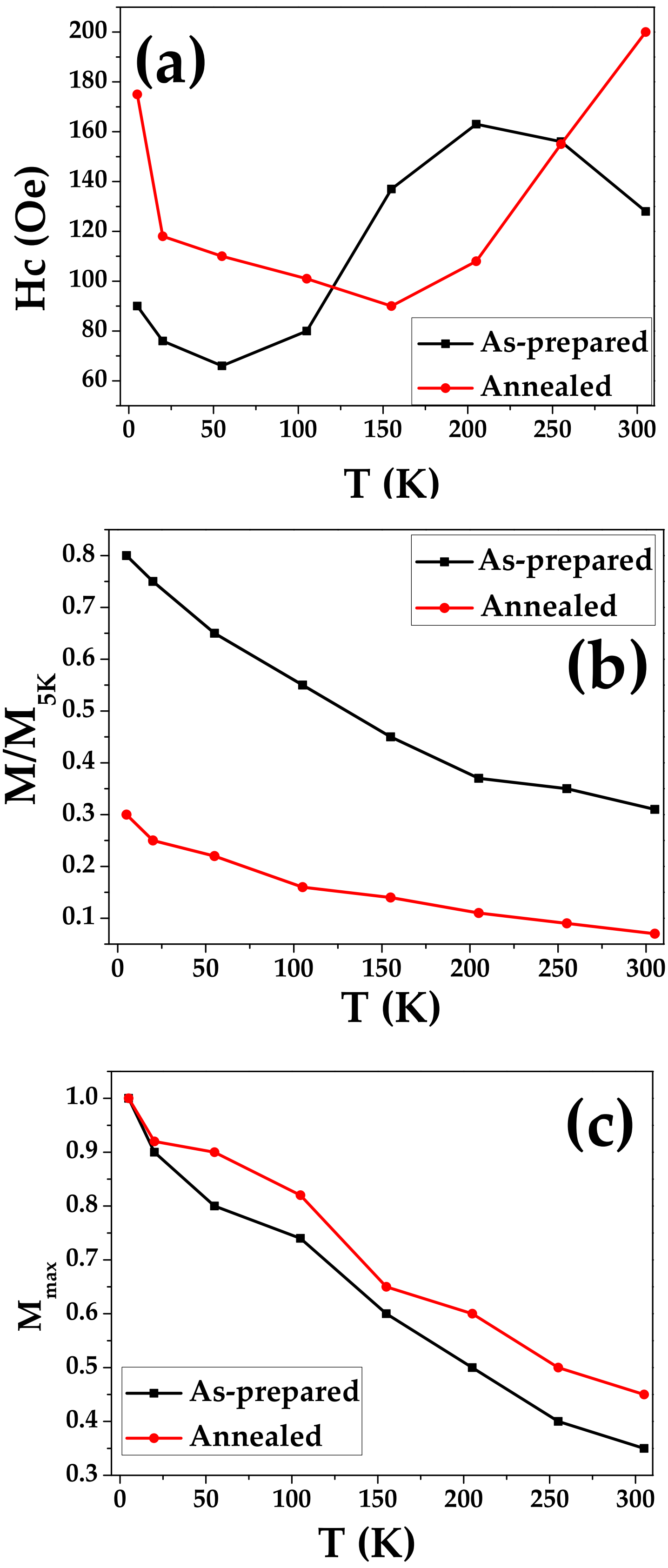

Figure 3. Temperature dependencies of coercivity (**a**), normalized remanence (**b**), and normalized saturation magnetization i.e., NM_s (**c**) of Ni_2FeSi glass-coated microwires, as-prepared and annealed at 973 K (1 h) (lines are just an eye guide).

Figure 4 described the temperature dependence of magnetization, M/M_{5K}, of the as-prepared and annealed samples at applied low magnetic field from 50 Oe to 200 Oe. As indicated in Figure 4, a noticeable difference in the magnetization M/M_{5K} vs. T (K) between the as-prepared and annealed samples is found. The as-prepared sample shows a regular M/M_{5K} (T) dependence typical for ferromagnetic materials: the M/M_{5K} ratio increases by decreasing the temperature. In addition, the FC and FH magnetization curves show almost perfect matching from 400 K to 100 K at H = 50 Oe and perfect matching from 400 K to 5 K at H = 200 Oe. Usually, a considerable dependence of magnetization curves (particularly magnetization values) on magnetic field is linked to the magnetic and atomic disorder, typically observed in rapidly quenched Heusler alloys [39]. Additionally, rapid melt quenching of metallic nucleus surrounded by the glass-coating with rather different thermal expansion coefficients involves the onset of large internal stresses ranging from 100 to 1000 MPa, distributed in a complex way inside the metallic nucleus [17,38,40,41]. Accordingly, the small mismatching between the FC and FH for the as-prepared sample below 100 K can be originated by a change in the magnitude of the internal stresses, which can affect the magnetic anisotropy. The interesting part is that in as-prepared sample this kind of the mismatching disappeared at the applied magnetic field (H = 200 Oe), i.e., the external magnetic field works against the internal mechanical stress induced during the fabrication process. For annealed sample, a strong mismatching between the FC and FH is observed at a whole range of measuring temperature. In addition, the FC and FH magnetization curves show multistep magnetic curves with different slopes (see Figure 4a,b).

This mismatching between FC and FH and the multistep magnetization cures gradually disappeared upon increasing the applied magnetic field, as observed in Figure 5a–c. Such a mismatching in glass-coating microwires Heusler alloys can be related to the change in the magnetic phase content, i.e., to the recrystallization process (see Figure 1) as well as to the phase transition from the disordered crystalline structure, changing in the strength of the internal mechanical stress and the non-perfect chemical composition distribution in the alloy [41–43].

Noteworthy, the magnetization behavior of the annealed sample is strongly affected by the temperature and the magnetic field than that of as-prepared sample, where the as-prepared sample shows a poor variation with the temperature and the external magnetic field. This indicated that the as-prepared sample has a quite stable thermal stability to the temperature and the external magnetic field. Meanwhile, the magnetization of annealed sample is more sensitive to the temperature and applied magnetic field. This is due do the increase in the $L2_1$ phase crystalline content, which presents a high sensitivity to the temperature and the magnetic field. As seen at Figure 5, the FC and FH magnetization curves have a different response by changing the external magnetic field.

As shown above, during the annealing, the precipitation of the crystalline phase from the amorphous precursor takes place. Such microstructure evolution and the change in magnetic properties brought on by annealing are strongly correlated [44]. In fact, the so-called "nanocrystalline materials," which are two-phase systems with nanocrystalline grains randomly dispersed in an amorphous phase, can exhibit improved magnetic softness. It was possible to successfully explain the magnetic softness of such materials by taking into account the correlation between the average crystalline size D and the exchange correlation length, L. Better magnetic softness can be attained when the macroscopic magnetic anisotropy levels out (when L >> D) [45,46]. But the process of recrystallization, which involves increasing both the crystalline phase concentration and the average crystalline size D, is typically connected to the magnetic hardening of such materials [32,45,46]. When it comes to glass-coated microwires in particular, devitrified microwires can preserve rectangular hysteresis loops during the early stages of devitrification, although magnetic hardening is frequently seen as the crystallization process progresses [26,32,47].

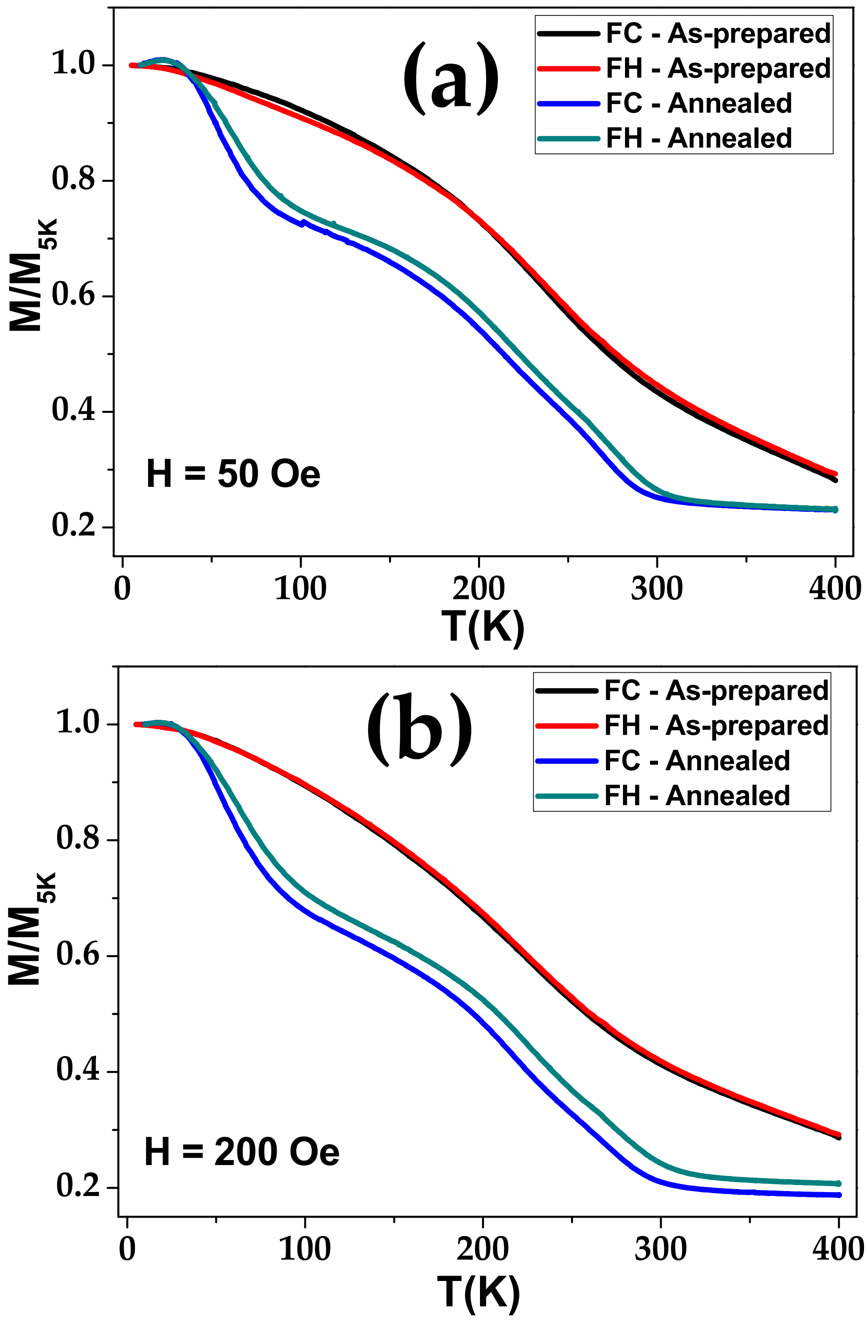

Figure 4. Temperature dependence of magnetization measured for as prepared Ni_2FeSi glass-coated microwires with applied external magnetic field (**a**) H = 50 Oe and (**b**) H = 200 Oe.

Therefore, the observed magnetic behavior, where H_c, M_r, FC, and FH vary with temperature, is confirmed by the correlation between the crystalline structures and magnetic properties of the annealed samples. Additionally, the change in the micromagnetic structure caused by the internal stress is responsible for the modest variation in magnetic properties of annealed sample. It would be prudent to preform additional study to learn more about the Ni_2FeSi samples' micromagnetic structure once they have been produced and annealed. Last but not least, we think that annealing at 973 K for one hour causes recrystallization, atomic ordering, and a decrease in internal stresses. Additionally, the anomalous magnetic behavior of annealed Ni_2FeSi glass-coated microwires can be attributed to the onset of two distinct magnetic phases, each with distinct magnetic anisotropies.

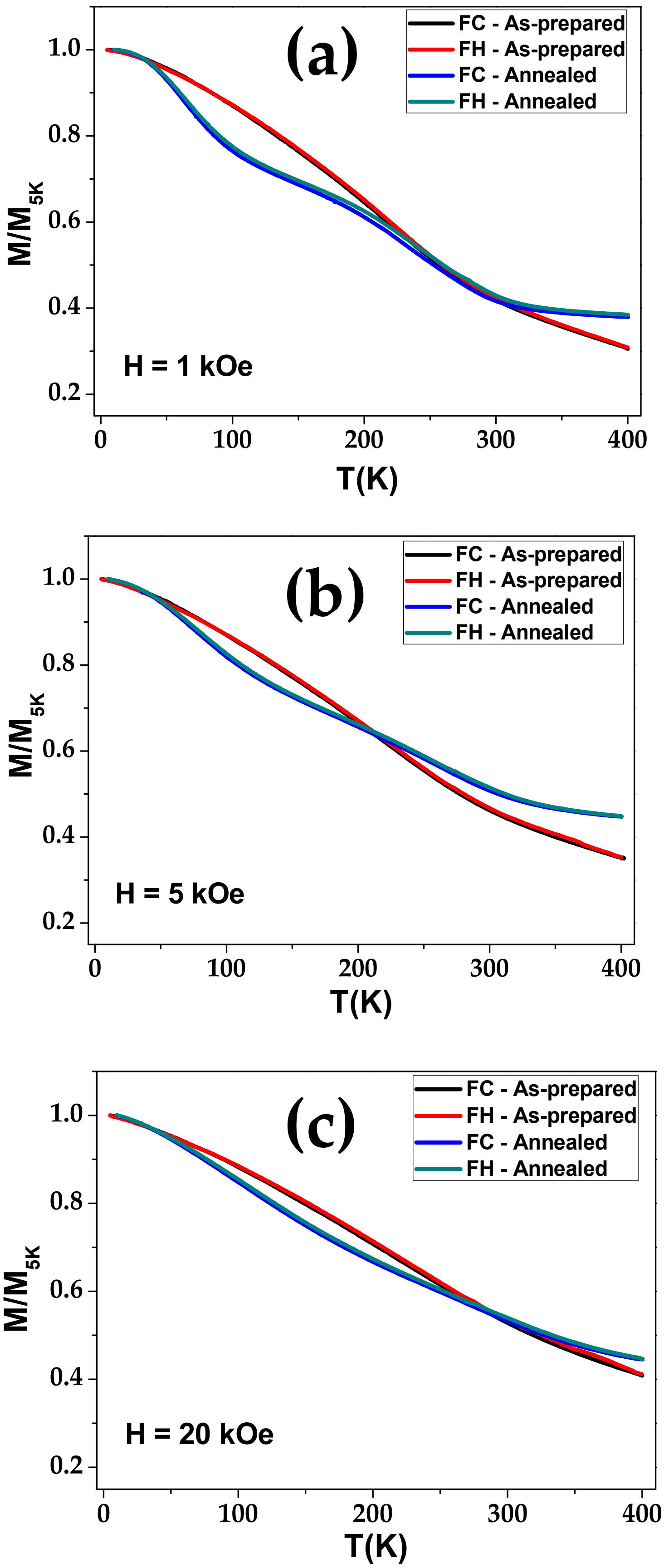

Figure 5. Temperature dependence of magnetization measured for annealed Ni_2FeSi glass-coated microwires with applied external magnetic field (**a**) H = 1 kOe, (**b**) H = 5 kOe, and (**c**) H = 20 kOe.

4. Conclusions

In conclusion, we report on the effect of annealing and the magnetic field on the magnetic properties of Ni_2FeSi glass-coated microwires. The annealing induces a transformation from cubic high temperature parent austenite phase transforms into a monoclinic martensitic structure with modulation 10 M besides to the enhancement of the crystalline phase content from 34% to 79% as compared to the as-prepared sample. The changing in the structure has a strong effect on the magnetic properties of the annealed sample. The hysteresis loops for annealed sample show vanishing reduced remanence at room temperature. Meanwhile higher normalized saturation magnetization is observed for annealed sample at temperature range from 305 K to 55 K. The FC and FH magnetic curves of annealed sample show multistep magnetic behavior with different slopes and magnitude can be modify gradually by changing of external magnetic field. Experimental results discussed considering the devitrification of the amorphous precursor, internal stresses relaxation, and recrystallization process. Observed findings demonstrate how the magnetic field and annealing have a significant impact on the magnetic characteristics of Ni_2FeSi glass coated microwires.

Author Contributions: Conceptualization, M.S. and A.Z.; methodology, V.Z.; validation, M.S., V.Z. and A.Z.; formal analysis, M.S.; investigation, M.S., A.Z. and V.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., J.G. and A.Z.; visualization, M.S., A.W. and M.I.; supervision, A.Z.; project administration, V.Z., J.G. and A.Z.; funding acquisition, V.Z., J.G. and A.Z. All authors have read and agreed to the published version of the manuscript.

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

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