Low magnetostriction in Fe_{100-x}Mn_x (x = 45, 48, 50, 52, 55) alloys

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Abstract. Investigation of the magnetostriction in Fe\textsubscript{100-x}Mn\textsubscript{x} alloys with x = 45, 48, 50, 52 and 55, in as-cast, annealed and cold-rolled states was performed. The magnetostriction was measured by two methods: capacitance dilatometer and strain-gauge bridge. A careful structure characterization was made by XRD. For all samples the magnetostriction measured at room temperature was quasi-zero. Additionally, we found an orthorhombic structure for as-cast and annealed samples. These results are in strong disagreement with those reported in literature. Possible reasons for these disagreements are briefly discussed.

1. Introduction

The magnetostrictive materials are defined as materials which undergo a change in shape (length, volume) due to change in the magnetization state of the material. All magnetically ordered materials exhibit a magnetostriction, however the magnitude depends strongly on the involved atoms as well as the symmetry of the lattice. In 3d-elements such as nickel, iron or cobalt, the magnetostriction is between 10 and 50 ppm depending also on the crystallographic direction. In the mid-20 century a new area began with the discovery of giant (around 1000’s of ppm) magnetostriction in rare earth alloys, e.g. Terfenol-D, that exhibits magnetostriction values up to 2000 ppm at room temperature in acceptable external fields. Pure rare earth elements or different rare earth based alloys exhibited large magnetostriction, but either at very high magnetic fields, or at cryogenic temperatures, or both. Already a well-known new class of magnetostrictive materials called Galfenol exhibits a rather high magnetostriction of the order of 400 ppm (in single crystals) in iron – 19 % gallium and 450 ppm in iron – 28 % Ga-alloys at room temperature [2], however, in very low external magnetic fields.

Recently, Peng \textit{et al.} [1] obtained an unexpectedly large magnetostriction in antiferromagnetic polycrystalline Fe\textsubscript{58}Mn\textsubscript{42} under a moderate applied field of 1 T. They achieved a linear strain of 169 ppm parallel to the field and -581 ppm when the sample is subjected to a compressive stress of 152 MPa. Later, Jingjing Zhang \textit{et al.} [3] obtained an enhanced magnetostriction of 690 ppm at 1350 kA/m in as-cast Fe\textsubscript{50}Mn\textsubscript{50} alloy. However a reduced magnetostriction was obtained after annealing the sample at 1100 °C for 24 h, which for this decrease was attributed to the decomposition of the single
γ-phase in a mixture of fcc γ-phase, hcp ε-phase and bcc α-phase during isothermal heat treatment. The same authors found a giant magnetostriction of 2100 ppm when the ingot of Fe50Mn50 alloy was subjected to 70% cold-rolling and annealed at 600 °C for 1 h [4]. They concluded that the preferred $<1 1 0>$ orientation formed along the rolling direction during the deformation is responsible for the improvement in magnetostriction. Very recently, we found very low magnetostriction, that is around zero, on annealed and cold-rolled Fe$_{100-x}$Mn$_x$ alloys with $x = 38, 42, 46, 50$ and $55$ [5]. In that work we also showed the susceptibility and the magnetization as function of the temperature and the texture analysis processed through software LABOTEX from the XRD measurements (AXS Bruker D8 diffractometer with GADDS area detector NANOSTAR). The linear increase of the magnetization with applied field and slope increase with temperature indicated that the alloys were antiferromagnetically ordered. Additionally, a very low magnetization value around 1 emu/g was measured at a field of 9 T, at room temperature. In cold-rolled samples with Mn content lower or equal to $x = 42$, no significant textures have been observed, while for the samples with $x > 42$ texture components in $\{110\}<1-12>$ or $\{011\}<01-1>$ were observed. However, all average longitudinal and transverse magnetostriction values measured on those Fe-Mn alloys were always around zero irrespective of heat treatment, concentration, texture and rolling reduction.

Because of the unclear situation from previous work, we have been motivated to prepare new samples and repeat the magnetostriction investigations in Fe$_{100-x}$Mn$_x$ alloys with $x > 42$ where we previously found marked textures [5], that is, $x = 45, 48, 50, 52$ and $55$, in the as-cast, annealed and cold-rolled state. Again, a careful structural characterization was achieved by XRD. Moreover, the longitudinal and transverse magnetostriction was measured at room temperature by two methods: micro-capacitance dilatometer and strain-gauge bridge.

2. Experimental techniques
Ingot of polycrystalline Fe$_{100-x}$Mn$_x$ alloys with nominal concentrations of $x = 45, 48, 50, 52$ and $55$ were prepared from high purity (> 99.9 %) elements by using high-frequency induction melting (Hüttinger RF Generator, 30kW, 600 kHz) under argon atmosphere in a copper crucible. Each ingot was re-melted three to four times, to assure the chemical homogeneity. Due to the high vapour pressure of Mn, it cannot be excluded that the real Mn-concentrations of samples were smaller than the nominal ones. Parts of each ingot were subjected to 30 %, 50% and 70% cold-rolling at room temperature (the percentages give the relative change in sample thickness due to cold-rolling).

The X-ray diffraction, XRD, patterns were recorded by means of an Xpert Philips powder diffractometer (Goniometer Philips PW 3050/60) using CuKα$_{1,2}$ radiation in a Bragg Brentano geometry, and a X’Celerator detector. The X-ray generator Philips PW 3040/60 worked at a power of 40 kV and 40 mA and the goniometer was equipped with a graphite monochromator. Diffraction patterns were recorded in the angular range 5° to 135° with a scan step size of 0.02°. Collected data were refined using the Rietveld package TOPAS (Bruker AXS Topas V 2.1) based on the fundamental parameter approach, with diffractometer parameters and wavelength settings adjusted using a LaB$_6$ standard.

Longitudinal and transverse magnetostriction measurements of as-cast, annealed at 1000 °C for 24 h and cold rolled samples were performed at room temperature by a strain gauge method using an AC-bridge (50-kHz bridge; HBM type KWS 85A1), in a pulsed-field magnetometer, which exhibits a maximum magnetic field of 5 T at pulse duration of 50 ms. It is important to mention that we used a half bridge circuit, where one strain gauge is on the sample and a second strain gauge on a non-magnetic dummy. With this set-up, the effect of the magneto-resistance of the strain gauge is balanced. On annealed Fe$_{48}$Mn$_{52}$ alloy, the magnetostriction was also measured by micro-capacitance dilatometer method up to 9 T [6].

3. Experimental Results
Figure 1 shows XRD patterns for as-cast Fe$_{100-x}$Mn$_x$ ($x = 45, 48, 50, 52, 55$) samples. Our results indicate that the phase present in our as-cast and annealed Fe$_{48}$Mn$_{52}$, Fe$_{48}$Mn$_{55}$, Fe$_{50}$Mn$_{50}$ and
Fe$_{45}$Mn$_{55}$ samples is a single orthorhombic hkl $\gamma$-phase instead of cubic fcc $\gamma$-phase, as reported in [2, 3]. Additionally, the average Lorentzian crystallite size of all as-cast and annealed samples is very small, i.e. about 50 nm. The sample with highest content of iron Fe$_{55}$Mn$_{45}$ presents two phases (77% of $\alpha$-Fe and 23% of orthorhombic $\gamma$-phase) already in the as-cast state. In all as cast and annealed samples investigated, no significant textures have been observed. In figure 1(a), XRD pattern corresponding to the orthorhombic phase of Fe-Mn was indexed. The lines corresponding to the cubic structure of Fe is indexed in figure 1(e).

When the samples were subjected to cold-rolling at room temperature, for the samples rolled at 30% and 70%, phase separation in three phases, namely: $\alpha$-Fe, Fe-Mn and MnO, occurred. The detailed analysis of the composition, for example for the Fe$_{48}$Mn$_{52}$ alloy, is shown in figure 2. Due to cold-rolling, the grain size became smaller (an average size about 20 nm) for all samples. For the highest deformation state even an amorphous background was formed (see figure 2(b)). It is also worth to mention the absence of the line corresponding to (222) in the XRD obtained for the sample Fe$_{48}$Mn$_{52}$ annealed and cold-rolled at 50%. In figure 2(b), only MnO was indexed (Fe-Mn and Fe were already indexed in the figure 1(a) and figure 1(e), respectively).

In figure 3, all longitudinal and transverse magnetostriction measurements performed on as-cast and annealed single hkl $\gamma$-phase samples using a strain-gauge method are given. As can be seen, the average longitudinal and transverse magnetostriction values of all samples are around zero up to an applied field of 5 T. On the other hand, the magnetostriction of the as-cast Fe$_{55}$Mn$_{45}$ alloy, which contains 77% of $\alpha$-Fe and 23% of the hkl $\gamma$-phase, corresponds to that of pure $\alpha$-Fe, with a saturation magnetostriction around -6 ppm (see figure 4). Therefore for the further studies this sample was not considered.

Figure 1. XRD patterns of Fe$_{100-x}$Mn$_x$ with $x$ = 45, 48, 50, 52 and 55.
Figure 2. XRD patterns of the heat treated and cold rolled Fe\textsubscript{48}Mn\textsubscript{52} alloy with respective compositions

Figure 3. Longitudinal and transverse magnetostriction measured on the as-cast and annealed Fe\textsubscript{100-x}Mn\textsubscript{x} with x =48, 50, 52 and 55
To investigate the magnetostriction at higher values of the applied field, the longitudinal magnetostriction was measured up to 9 T at 200 K and 300 K on annealed Fe_{48}Mn_{52} alloy using a micro-capacitance dilatometer. The magnetostriction at each temperature was measured three times and the results are shown in figure 5. For both temperatures 200 K and 300 K, the average magnetostriction value lies always around zero even at 9 T. This result is in agreement with the magnetostriction measured with strain-gauge.
The magnetostriction of all as-cast and annealed cold rolled samples is also around zero up to 3 T, independent of the measuring direction with respect to the rolling direction. Also, annealing at 600 °C for 1 h after the deformation process has no effect to the results. As examples, the longitudinal and transverse magnetostrictions measured on Fe_{48}Mn_{52} which were subjected to cold rolling at 30%, 50% and 70% are shown in figure 6. These measurements are in agreement with results presented in our previous work [5], however in strong contradiction to published data from the material Fe_{50}Mn_{50} [4].

4. Discussion
All results, structural and magnetostrictive properties, are in disagreement with the results reported in [3, 4]. In our previous work [5] and also in this one, we found very low magnetostriction in Fe_{100-x}Mn_{x} alloys. One of the reasons for the disagreement could be the difference in structures, that is, our as-cast and annealed samples present an orthorhombic hkl $\gamma$-phase while in [2, 3] a cubic fcc $\gamma$-phase was reported. For our cold-rolled annealed samples appeared impurities. However, the low magnetostriction value in our Fe-Mn alloys seems reasonable keeping in mind that these antiferromagnetic materials exhibited a very low magnetization (around 1 emu/g at 9 T [5]) and generally, the magnetostriction value in magnetic materials is directly related to the magnitude of the magnetization. It is interesting to note that Fe-Mn alloys are antiferromagnetic materials for a broad concentration range indicating that this ordering occurs due to a band structure effect.

It is worth to mention that measuring magnetostriction with strain gauge, a careful caution must be paid on the magnetoresitience of the strain-gauge employed. The active gauge and dummy gauge must be set parallel and connected to form adjacent sides of a Wheatstone bridge in order to cancel a trivial magnetoresistence change of strain-gauge. The wrong cementation and the wrong set and kind of the gauges lead sometimes to an anomalous increase in resistence with increasing of magnetic field.

Figure 7 shows the longitudinal and transverse magnetostriction of Fe-Mn alloys measured with strain gauge where the magnetoresistance effect was not completely compensated. Generally, the intensity of the measured magnetoresistance varies quadratic with the applied field, and is independent of the alloy’ composition of the samples and the direction of the applied field. Therefore, to eliminate the influence of the magnetoresitence on the magnetostriction of the material, the total magnetostriction, $\lambda_{\text{long}} + \lambda_{\text{trans.}}$, must be calculated.
In fact, identical values of longitudinal and transverse “magnetostriction” were obtained for the single hkl \( \gamma \)-phase alloys irrespective of the composition of the samples (see Figure 7(a)). For these samples the total magnetostriction \( \lambda_{\text{long}} \lambda_{\text{trans}} \) is around zero, which is the actual magnetostriction of these samples. For the sample \( \text{Fe}_{55}\text{Mn}_{45} \) alloy, which contains 77\% pure Fe, the total magnetostriction \( \lambda_{\text{long}} \lambda_{\text{trans}} \) is typical of that of \( \alpha \)-Fe (see Figure 7(b)). These results show that the effect of the magnetoresistance of the strain-gauge on magnetostriction measurements can be canceled measuring the resistance applying a magnetic field longitudinally and transversely. However the influence of the magnetoresistance for fields smaller than 1 T as used in [3, 4] is small.

5. Conclusion

A careful investigation of the structure of \( \text{Fe}_{100-x}\text{Mn}_x \) alloys with compositions \( x = 45, 48, 50, 52 \) and 55, in the as-cast, annealed and cold-rolled states was performed refining the XRD data by means of the Rietveld package TOPAS. We obtained a single orthorhombic hkl \( \gamma \)-phase for the as-cast and annealed samples, however three phases (Fe-Mn, \( \alpha \)-Fe and MnO) were formed in coexistence when the samples were subjected to cold rolling. The magnetostriction measured by capacitance dilatometer (up to 9 T) and strain-gauge bridge (up to 5 T) was around zero for all samples investigated, irrespective of the composition, annealing & deformation conditions. These results are in strong disagreement with those reported in [3, 4].

6. References

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