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Thermomechanical effect on magnetic behaviors of antiferromagnetic Mn-Fe(Cu) alloy

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Abstract: The effect of thermomechanical treatment on the magnetic properties of $Mn_{85.5}Fe_{9.0}Cu_{0.5}$ alloy was studied by use of a materials testing machine, a vibrating sample magnetometer, an X-ray diffractometer, a homogeneously and adjustably magnetic field and strain gauges. The results show that the orientation of fct phase and magnetic domains is affected by the thermomechanical treatment. When the compressive strain of thermomechanical treatment is -1.2%, the magnetic-field-induced strain reaches the highest value in the adapted situation.

Key words: thermomechanical treatment; antiferromagnetic Mn-Fe(Cu) alloy; magnetic-field-induced strain; magnetization curve

1 Introduction

High damping[1], narrow hysteresis two-way shape memory effect (TWSME)[2] and magnetic-fieldcontrolled shape memory effect[3–4] were successively found in a martensitic antiferromagnetic Mn-Fe(Cu) alloy. Especially, Mn-Fe(Cu) alloy can be potential practical multi-functional antiferromagnetic materials with a magnetic-field-induced strain at room temperature and in a polycrystalline alloy. PENG et al[3–4] suggested that the effect of driving force of the magnetic shape memory of the antiferromagnetic alloy attributes to the decrease of free energy through the moving for martensitic twin variants. WANG et al[5] promoted a Landua model to prove the assumption that the twin is just the antiferromagnetic domain in the alloy.

In order to obtain favorably orientated martensitic twins and large strains, thermomechanical treatment is often applied in temperature-controlled shape memory alloys. Recently, thermomechanical treatment has also been used in magnetic-field-controlled shape memory alloys, whose mechanism is related to the movement of twin boundaries, as well as temperature-controlled shape memory alloys. ULLAKKO et al[6] obtained a magnetic-field-induced strain (MFIS) of 4% in a near-stoichiometric ferromagnetic polycrystalline Ni₂MnGa alloy by thermomechanical treatment above $A_{\rm f}$ temperature, which is much higher than that without this treatment.

Usually, the formation of the domains in antiferromagnets is related to local fluctuations of the antiferromagnetic order at the Neel temperature with subsequent pinning of the domain walls by lattice imperfections. While the formation of domain in ferromagnets is promoted by the reduction of magnetostatic energy, the antiferromagnet possesses no local magnetization in the absence of an external field and there seems to be no obvious counterbalance to the increase of the free energy produced by the domain walls[7]. In other words, antiferromagnetic domains are more sensitive to temperature and external stress than ferromagnetic ones[7–8].

Thus, in this work, materials testing machine, vibrating sample magnetometer (VSM), X-ray diffractometer (XRD), Gouy magnetic balance, homogeneously and adjustably magnetic field and strain gauges were applied to testing the effect of thermomechanical treatment on

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MFIS and magnetization curve for the alloy.

2 Experimental

Mn-Fe(Cu) alloys were prepared by mediumfrequency induction melting under an argon atmosphere from electrolytic manganese (99.99%) and iron (99.99%). A amount of 5% Cu (molar fraction) was added to stabilize the γ -phase, hindering the γ β transformation in quenching. The addition of copper of less than 5% to the Mn-Fe alloy has no effect on the structure if the ratio of iron to manganese is equal[9]. The composition of the alloy is Mn_{85.5}Fe_{9.5}Cu_{5.0}, determined by energy-dispersive spectrometer system using a standard calibration method. The ingots were annealed and forged into rectangular specimens with dimensions of 150 mm \times 20 mm \times 20 mm and heated at 1 233 K and then guenched in water. Specimens were cut from the ingots for the experiments. The differential scanning calorimetry (DSC) (TA Instruments DSC 2910) results show that the A_s , A_f , M_s and $M_{\rm f}$ temperatures of the quenched sample are 422, 470, 462 and 428 K, respectively. Its Neel temperature was determined by Gouy magnetic balance (MB-2) under the applied magnetic field of 1.2 T. Transmission electron microscopy (TEM) results show that the morphologies of the alloy at room temperature are twins with fct martensitic structure with lamellas of about 200 nm in width, and selected-area electron diffraction (SAD) data display that its twin planes lie in {011} planes[4-5].

The specimens were heated at 473 K and undertaken with different compressive strains, -0.8%, -1.2%, -2.4% and -18.6%, respectively, until they were water-cooled temperature to room for the thermomechanical treatment by use of materials testing machine (Shimadzu AG-100KNA). The sample without thermomechanical treatment was titled as ε_{M-s} :0. The others were titled as ε_{M-S} :thermomechancal strain. Magnetization curve was investigated by VSM (JDM-13). The X-ray diffractometer (D/max 2550V XRD) with Cu K_{α} radiation was used to investigate the orientation of the thermomechanical specimens. There exists apparent pre-stress effect in the alloy[4]. MFISs along the magnetic field with pre-compressive stresses of -0.18, -0.36, -0.54 and -0.91 MPa, respectively, under the applied magnetic field strength of 1.0 T were conducted with strain gauges and magnetic field.

3 Results and discussion

Fig.1 shows the dependence of magnetic susceptibility on temperature for the Mn-Fe(Cu) alloy. Compared with the ferromagnetism, the magnetic susceptibility of Mn-Fe(Cu) alloy is very weak, which is



Fig.1 Magnetic susceptibility of Mn-Fe(Cu) alloy

less than 1×10^{-5} g⁻¹. However, there is a slight peak at 463 K, at which the magnetic order of alloy changes from paramagnetism to antiferromagnetism. 463 K could be determined as T_N temperature, which is in accordance with value in Ref.[9] measured by neutron diffraction. The result indicates that the Mn-Fe(Cu) alloy is antiferromagnetic at room temperature.

The magnetic structure in antiferromagnetic Mn-Fe (Cu) alloy was collinear and the magnetic moments were along the tetragonal axis (c-axis)[9]. There existed coupling between its martensitic twin and antiferromagnetic domains in the alloy[5]. ZHANG showed that there was a positive MFIS of 1.6% under an applied field of 3.8 T in the alloy. However, both the domain-wall motion in an antiferromagnets[3, 10] and the dislocation motion at the twin boundaries in the alloy[4, 11] needed a threshold field to overcome the spin reorientation via antiferromagnetic domain-wall motion and structure reorientation via twin dislocations. A prestress could play the same role as a threshold field to initiate the twin motion to obtain MFIS[4]. Therefore, prestresses were applied to testing the MFIS in the alloy.

Fig.2 shows the MFISs at different prestresses before and after the thermomechanical treatment with the different compressive strains. The results show that the thermomechanical treatment and the prestress affect the MFIS. The prestress affects the MFIS evidently. The higher the prestress, the larger the MFIS. The effect of thermomechanical compressive strain is not apparent. However, the compressive strain of -1.2% in thermomechanical treatment has the best MFIS in the adapted situations. It reaches 0.19% of pure MFIS at the prestress of 0.91 MPa under the applied magnetic field of 0.9 T. MFISs of the other thermomechanical treatments are not good. They are not yet as large as those of non-thermomechancial treatment samples.



Fig.2 Relationship of MFIS and prestress under 1.0 T for Mn-Fe(Cu) alloy

Fig.3 shows the XRD patterns with different thermomechanical compressive strains at room temperature. The main peaks, both {200} and {220}, had split into two peaks, showing that their microstructure is fct phase. Compared with the powder XRD pattern of sample without thermomechanical treatment, the relative intensity of the peak (111) vs (200) of thermomechanical samples with compressive strains of -1.2% and -18.6% changes, showing difference of the orientation of the thermomechanical samples.



Fig.3 XRD patterns of Mn-Fe(Cu) alloy before and after thermomechanical treatment

Antiferromagnet is a kind of weak magnetic substances. Their magnetization curves are much lower than those of ferromagnets (about $10^{-2}-10^{-3}$ order lower). So, it needs much higher magnetic field strength to obtain saturation intensity[12–15]. Fig.4 shows magnetization curves along the magnetic field of the alloy before and after thermomechanical treatment. Their figurations are as the same as the above mentioned. Under the magnetic field strength of 1.8 T, the magnetic



Fig.4 Dependence of magnetization on field of alloy before and after thermomechanical treatment

tizations are not saturated. However, there is obvious difference between the two curves under the applied magnetic field of 1.0 T. For the magnetic susceptibility, $\chi = \frac{dM}{dH}$, the magnetic susceptibility after the thermomechanical treatment of -1.2% is higher than that without thermomechanical treatment. For single-crystal antiferromagnets, their magnetic susceptibility perpendicular to magnetic field, χ_{\perp} , is much higher than that parallel to magnetic field, $\chi_{//}$. For these polycrystals, the thermomechanical treatment increases the magnetization strength. In another words, the antiferromagnetic domains whose antiferromagnetic vector is perpendicular to magnetic field are more than magnetic those parallel to field after the thermomechanical treatment, which leads to the increase of magnetization strength. For the magnetic structure of alloys was collinear and the magnetic moments were along the tetragonal axis (c-axis)[9], there exists coupling between its martensitic twin and antiferromagnetic domains in the alloy[5]. It can be deduced that the orientation of the magnetic domains is affected by the thermomechanical treatment for the alloy. In the alloy, because of the coupling between transformation and antiferromagnetic

martensitic transformation and antiferromagnetic transition, its martensitic twins could be regarded as its antiferromagnetic domains [1, 4–5]. Its MFISs are closely connected with its structure state and magnetic state. The thermomechanical treatment changes the XRD pattern and the magnetization curves are shown in Fig.3 and Fig.4, which means that the orientation of the fct phase and magnetic domains changes. Accordingly, the MFISs are influenced by the thermomechanical treatment. Among all the thermomechanical treatments, the compressive strain of -1.2% owns the best MFISs in the adapted situations.

4 Conclusions

1) The changes of MFIS, XRD and magnetization curve before and after thermomechanical treatment in an antiferromagnetic alloy were studied. The results indicate that the thermomechanical treatment affects the orientation of fct phase and magnetic domains, by which its MFISs change.

2) Among all the compressive strains of the thermomechanical treatment, the compressive strain of -1.2% reaches the largest MFIS of 0.19%, under the applied magnetic field of 0.9 T.

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