



Preparation of Soft Magnetic Fe-Ni-Sb-B Nanoalloy by Solid-Solid Reaction

GUOQING ZHONG* and QIN ZHONG

State Key Laboratory Cultivation Base for Nonmetal Composite and Functional Materials, School of Material Science and Engineering, Southwest University of Science and Technology, Mianyang 621010, P.R. China

*Corresponding author: Fax: +86 816 2419201; Tel: +86 816 6088252; E-mail: zgq316@163.com

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The Fe-Ni-Sb-B amorphous alloy has been prepared by a solid-solid chemical reaction of ferric trichloride, nickel chloride, antimony chloride and potassium borohydride powders at room temperature. The inductive couple plasma study indicates that the resultant is composed of Fe 5.38 %, Ni 56.19 %, Sb 29.02 % and B 0.44 %. The XRD and thermal analysis show that the alloy is a non-strict sense amorphous alloy which constituted with NiSb and FeNiB. The TEM image shows the resultant is spheroidicity particles which average size is estimated to be 20-30 nm. The mole ratio of the metal salts has been determined to be 1.0:1.0:0.25 to get the amorphous alloy. The largest saturation magnetization value of the 17.35 emu/g is obtained.

Key Words: Solid-Solid Reaction, Nanoalloy, Amorphous, Magnetism, Characterization.

INTRODUCTION

Nano-size metallic alloys have been shown novel optical, catalytic, electrical and magnetic properties with respect to those of pure metals and corresponding bulk materials¹⁻⁴. Many amorphous alloys, such as Co- and Fe-based amorphous alloys, possess superior magnetic properties compared to their crystalline counterparts because of their unique atomic structures⁵⁻⁸. These amorphous alloys are very promising candidates for using in magnetic devices and have attracted considerable attention in the past several decades. Amorphous alloys can be prepared by various techniques, including liquid quenching, mechanical alloying, vapor deposition and chemical alloying⁹⁻¹¹, etc. The room temperature solid-solid chemical reaction is a very simple method to prepare the metals and alloys nanoparticles. The reaction can be quickly accomplished at room temperature, which is a challenge to those traditional methods.

Antimony is a poor conductor of electricity and heat and has wear resistance and corrosion resistance. In recent years, the study of antimony alloy is mainly focused on anode¹²⁻¹⁴ and Heusler alloys¹⁵⁻¹⁸, but amorphous nanoalloy powder about antimony has been less investigated. Therefore, to study the synthetic reaction of the antimony alloys based on Fe nano-materials at room temperature will be useful not only for studying incompatibility alloy, but also for exploration of the new preparation method of other alloy nanoparticles. In this work, the Fe-Ni-Sb-B amorphous nanoalloy is synthesized by room temperature solid-solid chemical reaction, the

effect of the mole ratio of the metal salts on the crystalline state of the alloys is discussed and the magnetic performance is tested.

EXPERIMENTAL

All the chemical reagents used in the preparation experimental were analytical grade which were purchased from Chengdu Kelong Chemical Reagent Factory of China. Ferric trichloride hexahydrate, nickel chloride hexahydrate and antimony chloride were weighed and placed in an agate mortar and mixed up. The corresponding mole ratio of the metal salts was 1:1:0.25. Then, the white potassium borohydride powder was added into the mixture and the mole ratio of the all metal salts to potassium borohydride was 1:3. Immediately, the mixture became black and released a lot of colorless gas. The released gas was tested with wet pH paper and indicated that the gas was neutral gas. The mixture was grinded carefully about 3 min and 0.5 % of polyvinylpyrrolidone (PVP) of the total mass of the metal salts was added into the agate mortar and the grinding was kept on about 5 min. Then, the resultant was moved to a glass beaker as soon as possible and the resultant was rinsed repeatedly by a large amount of deionized water and collected by a centrifugation for removal of the residual reactants until no chlorine ion in washed water could be tested by the silver nitrate solution. Afterwards the resultant was washed thoroughly by ethanol to remove residual water in the resultant. Finally, the resultant was dried in vacuum at 313 K for 5 h to get the fine black particles.

The composition of the resultant was determined by the inductively coupled plasma-atomic mass spectrometer which was solubilized by a certain amount of conc. nitric acid. To study the thermal stability, the differential scanning calorimetry and thermogravimetry were carried out by a TA Q500 thermal analyzer at a heating rate of 20 °C/min under pure nitrogen atmosphere and the TG-DSC curves of the resultant was shown in Fig. 1. The powder X-ray diffraction patterns of the resultants were recorded by a D/max-II X-ray diffractometer in the diffraction angle range of 3°-80° in Fig. 2. The morphology and the particle sizes of the resultant were studied by a Tecnai G20 (FEI) transmission electron microscopy at 300 kV. The TEM picture of the resultant was given in Fig. 3. The soft magnetic properties of the resultants were investigated by a BKT-4500Z vibrating sample magnetometer.

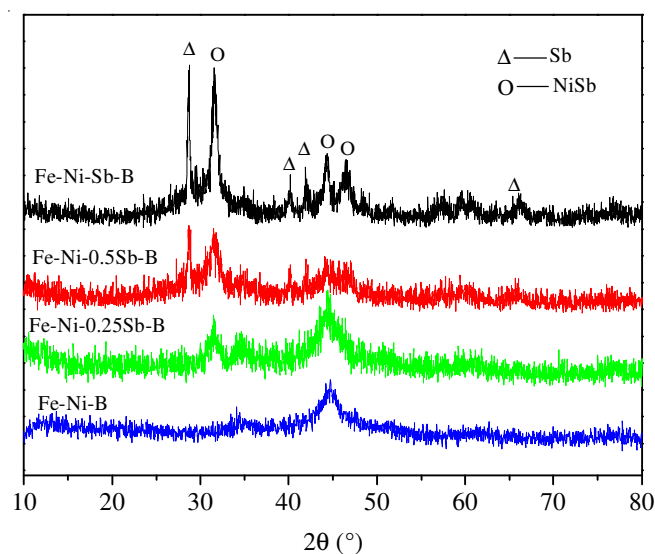


Fig. 1. Powder XRD patterns of Fe-Ni-xSb-B ($x = 1, 0.5, 0.25$ and 0) alloy nanoparticles

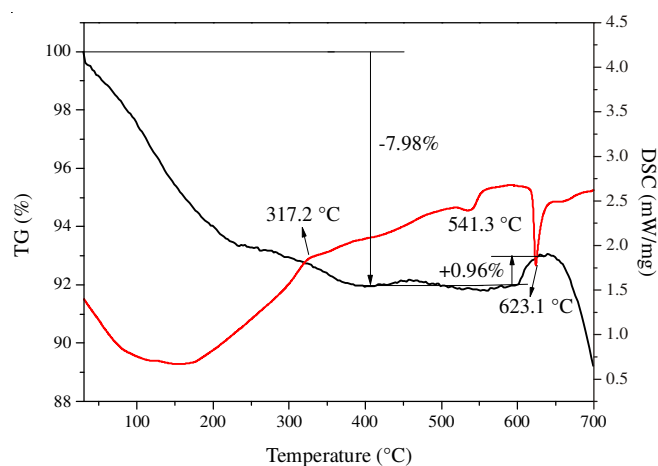


Fig. 2. TG-DSC curves of Fe-Ni-Sb-B

RESULTS AND DISCUSSION

The composition and content of the resultant for $n(\text{Fe}):n(\text{Ni}):n(\text{Sb}) = 1.0:1.0:0.25$ have been tested for Fe 5.38 %, Ni 56.19 %, Sb 29.02 % and B 0.44 % by inductive couple plasma. The total content of iron, nickel, antimony and boron of the resultants is 91.03 %, isn't equal to 100 %. The rest of

difference (8.97 %) should come from the PVP and be oxygenated of the fresh nanoalloy particles and the inference will be discussed in the result of thermal analysis. Combined with the mole ratio of the metal salts added into reaction, the loss of iron element is obvious.

In order to determine the mole ratio of the metal salts for getting the amorphous alloy, the alloys which came from different mole ratio of the metal salts have been prepared. The mole ratio of Fe, Ni and Sb is 1.0:1.0:1.0, 1.0:1.0:0.50 and 1.0:1.0:0.25, respectively. The XRD characteristic peaks of Sb (JCPDS card 35-0732) and NiSb (JCPDS card 65-0935) are observed obviously and they are sharp and strong which come from the mole ratio of the metal salts for 1:1:1 (Fe-Ni-Sb-B). With the decreasing of antimony, the characteristic peaks of Sb weakened to vanished (Fe-Ni-0.25Sb-B, the mole ratio is 1:1:0.25), but the peaks of NiSb is changed from sharp to diffuse. Contrasted with the XRD patterns of the Fe-Ni-B amorphous alloy, the diffuse peaks of NiSb from the resultant still existed. So we conjecture that Fe-Ni-0.25Sb-B may be constituted with NiSb and FeNiB alloys and it is a non-strict sense amorphous nano-alloy. In order to obtain amorphous alloy, the mole ratio of the metal salts is 1:1:0.25.

In order to study the crystallization temperature and determine the ICP results of the alloy, the TG-DSC test was used at a constant heating rate of 20 °C/min in nitrogen. The TG curve shows an observed weight loss (7.98 %) below 400 °C, which comes from the carbonation decomposition of PVP. It is closed with the data (8.97 %) of the inductive couple plasma. The incomplete decomposition of PVP may be causes the rest of difference (about 1 %). There are two endothermic peaks at 541 °C and 623 °C in the DSC curve, but slow decrease in mass is occurred in the TG curve. This indicates that the endothermic peaks do not come from oxidation or volatilization of the resultant, but the two crystallization processes of the resultant. The conclusion determines the conjecture that the resultant should be constituted with NiSb and FeNiB alloys.

The image of TEM is shown in Fig. 3. A statistical analysis of the TEM image shows that the resultant is formed from

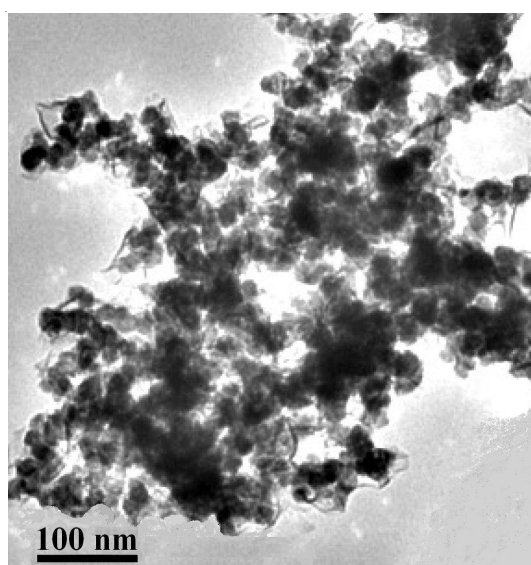


Fig. 3. TEM image of Fe-Ni-Sb-B

TABLE-1
SATURATION MAGNETIZATION, COERCIVITY AND REMANENT MAGNETIZATION OF THE ALLOYS

$n(\text{Fe}):n(\text{Ni}):n(\text{Sb})$	Ni (%)	Saturation magnetization (emu/g)	Coercivity (O_e)	Remanent magnetization (emu/g)
1:1:1	22.88	2.92	89.83	0.83
1:1:0.5	39.93	11.68	134.92	3.19
1:1:0.25	56.19	17.35	150.58	5.13

ultrafine, spheroidicity particles and minor reunion is existed in it. The average size of the nanoparticles is measured from the TEM image and estimated to be 20-30 nm.

Fig. 4(A) illustrates the hysteresis loop of the resultant. The saturation magnetic polarization and coercive force are

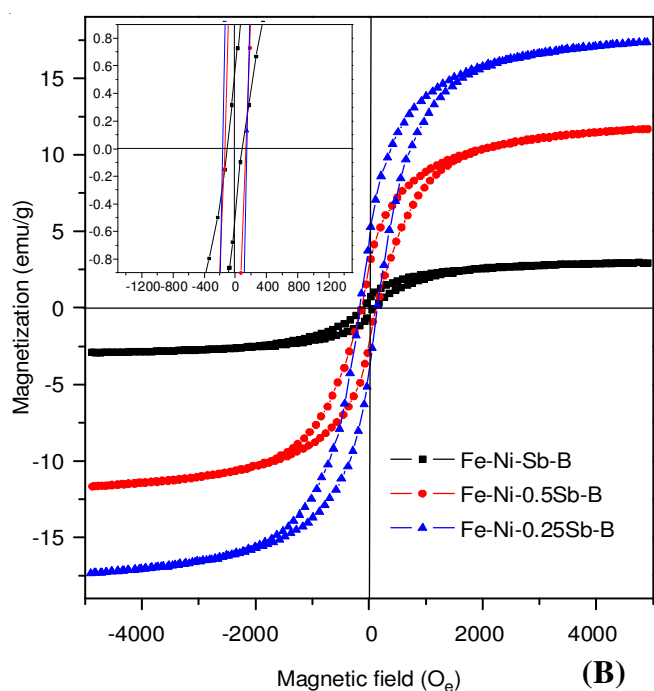
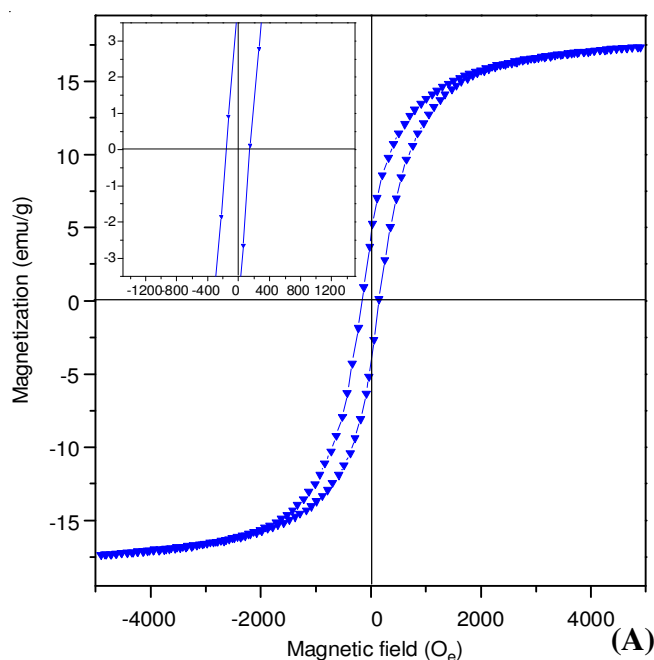


Fig. 4. Magnetic hysteresis curve of the Fe-Ni-Sb-B nanoalloy (A) and the hysteresis loops of the alloys with different mole ratio of the metal salts (B)

shown for 17.35 emu/g and 157.98 O_e . In order to study the effect of magnetic properties of alloy, the hysteresis loops of the alloys with different mole ratio of the metal salts have been tested. The test outcomes have been shown in Fig. 4(B). Table-1 shows that the increase of saturation magnetic polarization causes significant enhancement of the saturation magnetic polarization (M_s) from 2.92 emu/g for Fe-Ni-Sb-B to 17.36 emu/g for Fe-Ni-0.25Sb-B with increasing nickel content. Obtained data of the coercive force (H_c) is 89.83, 134.92 and 150.58 O_e , respectively. The long and narrow of hysteresis loop of Fe-Ni-0.25Sb-B amorphous alloy indicates that the resultant has low coercive force and remanent magnetization. Therefore, Fe-Ni-0.25Sb-B amorphous alloy has superior soft magnetism.

Conclusion

In summary, the Fe-Ni-Sb-B amorphous alloy has been prepared easily by a simple solid-solid chemical reaction method at room temperature. The advantages of this synthetic method are simple, convenient, save energy and environmental friendly and it is in accordance with the requirement of green chemistry. The characterization results indicate that the resultant is consisted of spheroidicity particles which average size is estimated to be 20-30 nm from TEM image. The mole ratio of metal salts has obvious effect to the resultants in crystal structure and magnetism. The alloy is constituted with the NiSb and FeNiB alloys and it is an amorphous nano-alloy. The mole ratio of the metal salts has been determined to be 1.0:1.0:0.25 for obtaining amorphous alloy. The alloy has superior soft magnetism.

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