The Microstructure and Magnetic Properties of FePt Alloys Prepared by Spark Plasma Sintering

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Abstract: $Fe_{50}Pt_{50}$ alloys were prepared by melting, milling and spark plasma sintering (SPS), the microstructure and magnetic properties of the alloys were analyzed. The results show that the alloy will transform into $L1_0$ ordered fcc phase from disordered fcc phase during sintering. The degree of order and the coercivity of $Fe_{50}Pt_{50}$ alloys decreases as the SPS temperature rises from 700℃ to 900℃. A maximum magnetic energy product of 50.81 kJ/m^3 can be obtained for the alloy sintered at 800℃.

Key words: metal materials; FePt alloy; spark plasma sintering; coercivity; XRD
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放电等离子体烧结 FePt 合金的微观结构和磁学性能

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摘 要：采用熔炼、机械破碎和放电等离子体烧结法制备了 $Fe_{50}Pt_{50}$ 合金，研究了合金的微观结构和磁学性能。结果表明在烧结过程中合金中无序的面心立方相向有序的 $L1_0$ 有序相转变，$Fe_{50}Pt_{50}$ 合金的有序度和矫顽力随着放电等离子体烧结温度的升高而降低；在 800℃烧结时，可获得的最大磁能积为 50.81 kJ/m^3。

关键词：金属材料；FePt 合金；放电等离子烧结；矫顽力；X 射线衍射

FePt alloys in the ordered face centered tetragonal structure (fct) have been widely investigated, due to its merits such as: (a) a high magnetocrystalline anisotropy (K_u≈7×10^{7}erg/cc), with an atomic composition near equal atomic ratio of $Fe_{50}Pt_{50}$ (for the atomic ratio of Pt, within 35~55), which make it the candidate material for ultrahigh data storage application. The grain size of a $L1_0$-ordered FePt nanogranular films can be reduced to 3~4 nm while still thermally stable in magnetic properties at room temperature, which makes it almost the best candidate for perpendicular magnetic recording (PMR) media with the recording density of 1 Tbits/in^2 or even higher[1-3]; (b) the combination of high coercive force and magnetic energy product $(BH)_{Max}$, makes it to be considered a good substitute for the existing rare earth based permanent magnets and potentially applied in some special area such as micro generator and MAGMAS (Magnetic Micro Actuators and Systems); And also (c) excellent mechanical strength, biocompatibility, corrosion resistance and magnetic properties, which make it suitable for dental applications[4].

The ordering of FePt films are widely researched

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by many authors using different methods including annealing[5], intermediate layer deposition[6], third elements addition[7-8] and the changing of matrix materials[9-10]. However, for the bulk FePt alloy, it's hard to control the ordering process. According to the Fe-Pt equilibrium phase diagram, the disorder-order transform from an A1 (fcc) phase to a $L1_0$ (fct) phase will proceed at temperature lower than 1300℃ for $Fe_{50}Pt_{50}$ even during quenching. So it's hard to control the ordering process and hence the magnetic properties of the alloy. Spark plasma sintering (SPS) is known as a sintering and consolidation technique[4, 11]. It can sinter and compact the specimen within a short time by application of a high current and compressor force. In this paper, we report the micro- structure and magnetic properties of FePt bulk magnets prepared by melting, mechanically milling and SPS.

1 Experiment

Alloy ingot of composition $Fe_{50}Pt_{50}$ was prepared from 99.95% purity iron and 99.99% platinum by vacuum induction melting. The as-cast alloy was cast in the form of a cylinder with the diameter of 10 mm. The ingot was then crushed into small pieces and mechanically milled into fine powder and sieve with a 200 mesh screen. The powder was then filled into a graphite mold and pressed by spark plasma sintering (SPS) at different temperatures. The following are the experimental conditions: the sintering temperature are 700℃, 800℃, 900℃, the pressure and time are 10 kN and 10 min respectively. The dimension of the samples is 10mm in diameter. The constituent phases in milled powders and SPS samples were examined by X-ray diffraction (XRD, Ritaku TTR III) using Cu K_α radiation. The magnetic properties of the SPS bulks were evaluated using a permanent magnet tester (NIM-2000F, National Institute of Metrology, China) at room temperature.

2 Results and discussion

2.1 XRD results

Fig.1 is the XRD patterns of as cast and as milled samples respectively. It can be seen from Fig.1 that both samples are Fe(Pt) solution, namely fcc phase, no peak of ordered phase are visible. During the process of melting and casting, disordered fcc FePt phase are presented instead of ordered fct FePt phase, that means disordered phase are more stable on this situation which forms during the cooling process. After mechanical milling, the peaks of Fe(Pt) phase are notably widen, that means grain refining or micro-strain or both of them exist in the alloy during mechanical milling. The software Jade was used to estimate the grain refining and micro-strain level of both of the as-cast and the as-milled samples. Hall method was used during the calculation. From the profile fitting results, if both grain size and strain was considered, the grain size of both samples will large than 100nm, which are contradicting with the method. As a result, only strain was considered during the fitting. The micro-strain are 0.301% and 0.749% obtained for the as-cast and as-milled samples respectively, which disclose the fact of strain accumulation during the milling process.

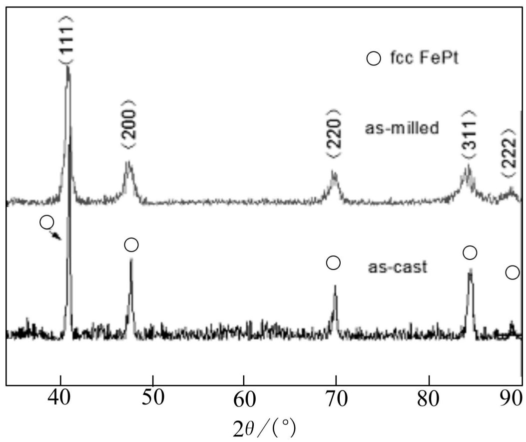

Fig.1 XRD patterns of FePt alloy on as cast and as milled samples

图 1 FePt 合金铸态及球磨态样品的 X 射线衍射图

The XRD patterns of FePt alloys processed by spark plasma sintering are shown in Fig.2. It can be seen from Fig.2 that the ordered fct $L1_0$ FePt phase appears after SPS. The degree of the distortion of the ordered fct phase can be estimated by the apparent splitting of the (200), (002), (220), (202), (311) and (113) reflections, which originated from the (200), (220) and (311) reflections of the fcc phase. And obvious (001) and other $L1_0$ feature peaks are visible on the patterns. The results show that the $L1_0$ ordering was undergoing during the SPS process.

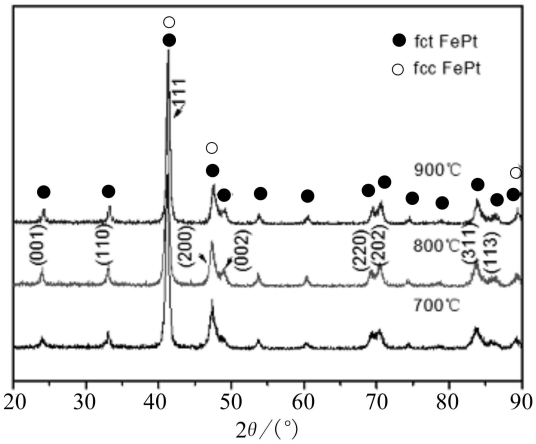

Fig.2 XRD patterns of FePt alloys processed by spark plasma sintering

图 2 放电等离子体烧结 FePt 合金的 X 射线衍射图

The order degree of the $Fe_{50}Pt_{50}$ alloys can be obtained according to the equation below[12]:

$$S^2=(1-c/a)/[1-(c/a)_s] \tag{1}$$

Where S is the degree of order, $(c/a)_s$ is the c/a value of the fully ordered FePt alloy, which is 0.956. Fig.3 shows the axial ratio c/a and the order degree S of $Fe_{50}Pt_{50}$ alloys SPSed at different temperatures. The result shows that the order degree of the $Fe_{50}Pt_{50}$ alloys decreases with the SPS temperature rising. A S=0.895 are obtained at 700℃. While the axial ratio a/c increases with the SPS temperature.

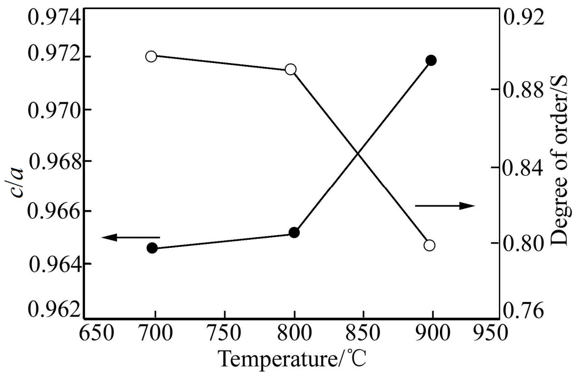

Fig.3 Axial ratio *c*/*a* and degree of order of $Fe_{50}Pt_{50}$ alloys SPSed at different temperatures

图 3 不同温度下 SPS 的 $Fe_{50}Pt_{50}$ 合金轴比及有序度曲线

2.2 Magnetic properties

Fig.4 shows hysteresis loops of the SPS $Fe_{50}Pt_{50}$ alloy sintered at different temperatures. The magnetic properties of the alloys are summarized in Tab.1. It can be seen from Fig.4 and Tab.1 that the H_c of FePt alloy decrease from 398.4 to 192.7 kA/m with sintering temperature increasing. The relationship between coercivity and anisotropy constant can be expressed as[12]:

$$H_c=K_k/(\mu_0 M_{sm}) \tag{2}$$

Where M_{sm} is the saturation magnetization of soft magnetic phase and K_k is the anisotropy constant of the hard magnetic phase. As mentioned above, the degree of order of fct phase decreases with the SPS temperature rising, while the degree of order is closely related to the anisotropy constant. The higher the anisotropy constant, the larger the H_c value will be. Another reason is when the sintering temperature increase, the grain size of the alloy may increase, and this can decrease the possibility of nucleation of ordered fct phase from the disordered fcc phase, and hence of reversed domains.

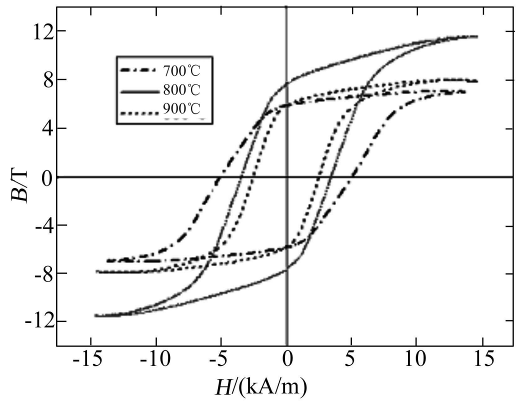

Fig.4 Hysteresis loops of the SPS $Fe_{50}Pt_{50}$ alloy sintering at different temperatures

图 4 不同温度下 SPS 的 $Fe_{50}Pt_{50}$ 合金磁滞回线

Tab.1 Properties of FePt alloy spsed at different temperatures

表 1 在不同温度下放电等离子体烧结的 FePt 合金的磁学性能

SPS temperature/℃	H_c/(kA/m)	$(BH)_{max}$/(kJ/m^3)	M_r/M_s
700	398.4	45.42	0.83
800	273.8	50.81	0.67
900	197.2	28.91	0.72

It can be seen from Fig.4 that the sample SPSed at 700℃ exhibit the best squareness, which are characterized to be magnetically isotropic; the sample SPSed at 800℃ has the worse squareness , which also means a more strong magnetic anisotropic. Both the squareness and the isotropic of the sample SPSed at 900℃ are in between. For all the three hysteresis loops, the shape are smooth, it is typical for a single component system. But for the M_r/M_s in Tab.1, a value higher than 0.70 indicate the existence of the exchange coupling between the hard and soft magnetic phases[13]. The hard magnetic phase is with high coercivity and low magnetic permeability. On the contrary, the soft magnetic phase is with low coercivity and high magnetic. Normally fcc FePt alloy which are soft magnetic phase are more likely formed by sintering, and fct phase which are hard magnetic phase are harder to get. This means during the SPS process the amount of soft magnetic phase may decrease at sample sintered at 700℃ and have a minimum percentage near this temperature and would increases with the increase of sintering temperature to 800℃. The percentage of soft magnetic phase achieve the highest amount at 900℃.For the application of FePt alloy, one can modification on the preparation process to obtain the requiring properties. For example, if high coercivity are required, the sample pressed at 700℃ is better. If both coercivity and permeability are needed, then the sample sintered at 800℃ are better, because the magnetic energy product is the best among all three.

3 Conclusion

(1) Spark plasma sintering can promote the ordering of fct FePt from fcc FePt.

(2) The order degree of $Fe_{50}Pt_{50}$ alloys decreases with the increase of SPS temperature from 700℃ to 900℃, a degree of order of 0.895 are obtained at 700℃.

(3) The coercivity of $Fe_{50}Pt_{50}$ alloy decreases with increase of SPS temperature, the reason should be the increase of anisotropy constant and grain size.

(4) A maximum magnetic energy product of 50.81 kJ/m^3 can be obtained for alloy sintered at 800℃.

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