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ARTICLE

Microwave Properties of RE(Nd, Tb)FeCoB Alloy System

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Abstract: The RE(Nd, Tb)FeCoB magnetic powders were prepared by arc melting, high energy ball milling and a partial oxidation treatment method. A X-ray diffraction device and a vector network analyzer were used to analyze the phase structure and microwave absorbing properties of the powders. The results of B addition to the microwave absorbing performances in the NdFeCo alloy, indicate that $Nd_2Fe_{14}B$ phase will precipitate and the relative content of the α-Fe phase increases with the increasing of B in $Nd_{10.53}Fe_{77.84}Co_{11.63}$ alloy powder. When Nd is replaced by the heavy rare earth Tb in NdFeCoB alloy, the powders consist of α-Fe, $Tb_2Fe_{14}B$, Tb_2Fe_{17} and a small amount of Tb_2O_3 phases. $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder has a minimum of absorption peak frequency, which is –9.5 dB at 4.5 GHz. After substituting Tb for Nb in $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ alloy the absorption peak frequency increases to 6.3 GHz, but the reflectivity value is reduced to –11 dB.

Key words: NdFeCoB alloy; microwave absorption properties; microstructure; high energy ball milling

The application of electromagnetic technology can create the material civilization, but at the same time it brings people into a world full of electromagnetic radiation. The electromagnetic wave radiation emission from the working electronic instruments, not only interfere the normal work of other electronic instruments, but also threat the health of human body[1-3]. A lot of research work have been done to reduce the harm of the electromagnetic radiation, and people find that using of magnetic absorbing materials to guide the electromagnetic wave is a good idea. Through the resonance, a large number of the electromagnetic wave radiation energy can be absorbed. Coupling the electromagnetic wave energy into heat energy is a very good way to inhibit and interfere electromagnetic wave radiation. Generally, the working frequency band of the mobile communication, the electronic equipment, and radar systems are in the range of about 1~6 GHz, but so far the absorption peak frequencies of the majority absorbing materials are above 6 GHz when the absorbing layer thickness less than 2 mm[4-7]. So researching the absorbing materials with thin thickness and low absorption peak frequency becomes one of today's hotspots. In this work，$Nd_2(Fe, Co)_{17}$ alloy was used as the basic material. With the purpose of reducing material absorption peak frequency, B element was added into the alloy to change the phase structure, and then heavy rare earth Tb was used to replace light rare earth Nd to explore the effect on the phase structure and microwave absorbing properties.

1 Experiment

The $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{100-x}B_x$ (x=0, 1, 3, 5, mole fraction, %) samples were prepared by Fe, Co, Nd, Tb (purity not less than 99.50%) and Fe-B alloy. They were smelted in a vacuum arc furnace under the protection of high purity argon (99.99%), then the samples underwent a homogenization heat treatment at 1050 °C for 48 h under vacuum environment, and were milled 72 h under the protection of gasoline using planet ball mill (QM-lSP), whose speed was 250 r/min. The mass ratio of the balls to the powers was 15:1. The powder underwent oxidation treatment at 100 °C for 2 h and the XRD device (D8 ADVANCE) was used for phase analysis. The well-prepared powders were mixed with paraffin in 4:1 ratio (mass ratio). The mixture was prepared into a coaxial ring with a thickness of 3.5 mm, the inside and outside diameters of which were 3 and 7 mm, respectively. The complex

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permeability and the complex permittivity in the frequency range of 2~18 GHz, which were used to calculate the reflectivity of the sample, were measured with HP8722ES microwave vector network analyzer.

2 Results and Discussion

2.1 Effects of B on the microwave absorbing performances in NdFeCo alloy

Fig.1 is the XRD patterns of $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{100-x}B_x$ (x=0, 1, 3, 5) powders. It is demonstrated that $Nd_{10.53}Fe_{77.84}Co_{11.63}$ powders are mainly composed of α-Fe, Nd_2Fe_{17}, a small amount of Fe_2Nd and Nd_2O_3 phases, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{99}B$ powders mainly consist of α-Fe, Nd_2Fe_{17}, a small amount of $Nd_2Fe_{14}B$ and Nd_2O_3 phases, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powders are mainly composed of α-Fe, $Nd_2Fe_{14}B$, Nd_2Fe_{17}, and a small amount of Nd_2O_3 phases, and $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{95}B_5$ powders mainly consist of α-Fe, a small amount of $Nd_2Fe_{14}B$ and Nd_2O_3 phases, and some crystal is turned into non-equilibrium organization. With the adding of B in $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{100-x}B_x$ powders, the relative content of the α-Fe phase increases, so the addition of B element has a great influence on the phase composition of the $Nd_{10.53}Fe_{77.84}Co_{11.63}$ powders.

Fig.2 shows the electromagnetism parameters of the NdFeCoB powders with different contents of B element in the frequency range of 2~18 GHz. As seen from Fig.2a and 2b, the NdFeCo powder has a high complex dielectric constant real part ε' and an imaginary part ε'' in 2~7 GHz when 3% of the B element is added. But as it reaches 5%, the complex dielectric constant real part ε' declines. The reason may be that a part of the crystal forms a non-equilibrium organization, and makes the electrical conductivity reduce, thus complex dielectric constant real part ε' value declining; at the same time, the freedom energy ΔE reduces, as a result the complex dielectric constant imaginary part ε'' rises. From Fig.2c it can be seen that with adding 3% of B, the complex permeability real part μ' and the imaginary part μ'' of NdFeCo powder reduces obviously in 2~7 GHz, and this may be due to that a large number of $Nd_2Fe_{14}B$ phase precipitate and the relative content of α-Fe increases, which lead to the increase of the saturated magnetization (M_s), so the complex permeability (real part μ' and imaginary part μ'') reduces. But when the content of B reaches 5%, the part of non-equilibrium organization is formed, and the saturated magnetization (M_s) of the powder is decreased; as a consequence the complex permeability (real part μ' and imaginary part μ'') rises[8].

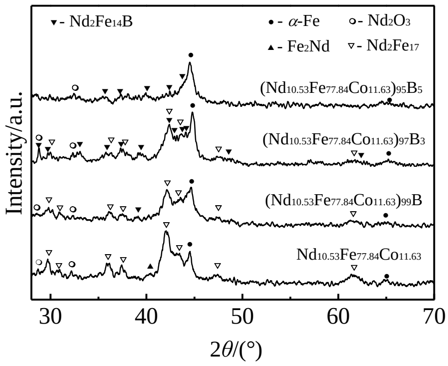

Fig.1 XRD patterns of $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{100-x}B_x$ (x=0, 1, 3, 5) powders

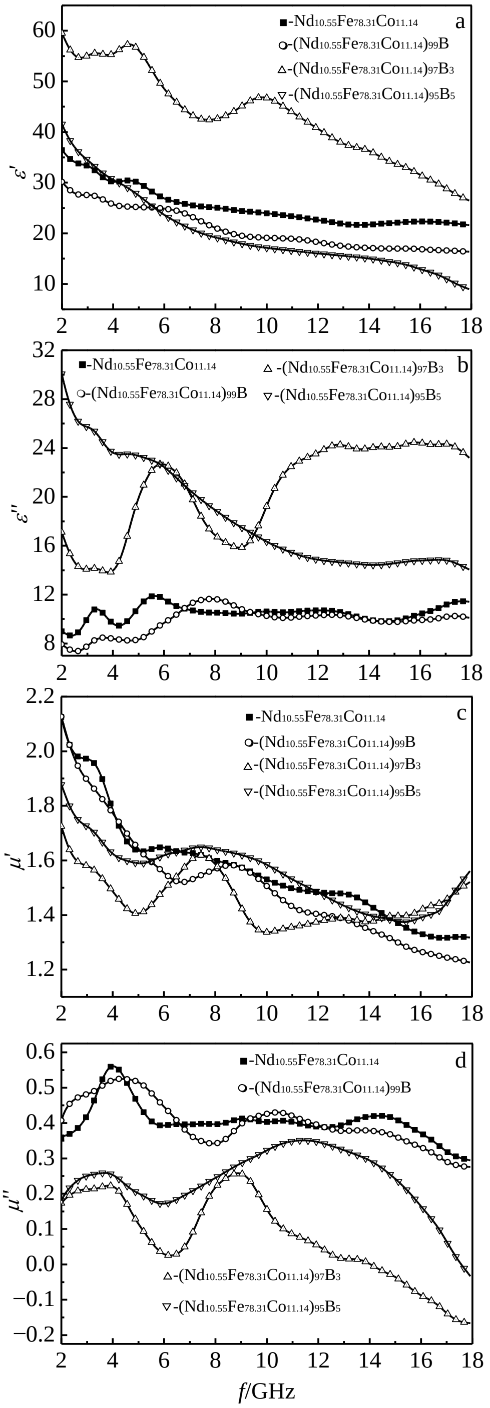

Fig.2 Electromagnetic parameters of $(Nd_{10.55}Fe_{78.31}Co_{11.14})_{100-x}B_x$ (x=0, 1, 3, 5) powders: (a) curves of ε' vs f, (b) curves of ε'' vs f, (c) curves of μ' vs f, and (d) curves of μ'' vs f

According to the equivalent of transmission line theory, the reflection coefficient for coated single-layer electromagnetic microwave absorbing materials can be deduced as:

$$R = 20\lg(|Z-1)/(Z+1)| \qquad (1)$$

where, $Z=\sqrt{\mu_r/\varepsilon_r}\tanh(j2\pi fd\sqrt{\mu_r\varepsilon_r}/c)$,

ε_r, μ_r and d are the relative dielectric constant, the relative permeability and the thickness of the absorbing material, respectively, f for electromagnetic wave frequency, c for electromagnetic wave in free space velocity, and j for imaginary unit. The reflectivity (R) of wave-absorbing materials is calculated by formula (1).

As shown in Fig.3, when x=0, 1, 3, 5, the minimum reflectance and the absorption peak frequency are –9.6 dB at 6.2 GHz, –9.4 dB at 6.5 GHz, –9.5 dB at 4.5 GHz and –6.2 dB at 6.8 GHz, respectively, when the thickness d=1.8 mm. The lowest absorption peak frequency is declined to 4.5 GHz as the addition of B element is 3%. The reason is that a great quantity of $Nd_2Fe_{14}B$ phase precipitate and the saturation magnetization (M_s) of $Nd_2Fe_{14}B$ phase is larger than that of Nd_2Fe_{17} phase; at the same time the relative content of α-Fe phase increased also makes the saturation magnetization (M_s) increase. According to $f_r \propto 1/M_S^2$, if the saturation magnetization (M_s) of the powder increases, the absorption peak frequency will decrease. When the content of B reaches 5%, a lot of non-equilibrium organization are formed, and the saturation magnetization (M_s) of the powders reduces as a result of the absorption peak frequency rising to 6.8 GHz.

2.2 Effects of Tb on microwave absorption properties of NdFeCoB alloy

The previous section discussed the impact of B on NdFeCo alloy and achieved some beneficial results. So this part is going to discuss the organization structure and microwave absorption characteristics changing by other rare earth elements replace Nd. Through the comparative analysis, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ was chosen as the research object, and the changing of organization structure and the absorbing properties were studied when using heavy rare earth Tb substitution of Nd. $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ (mole fraction, %) alloy was obtained as the same experiment method as previous.

Fig.4 is the XRD patterns of $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ (RE=Nd,Tb) powders, and the patterns show that $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder is mainly composed of α-Fe, $RE_2Fe_{14}B$, RE_2Fe_{17} and a small amount of RE_2O_3 phase.

Fig.5 shows the curves of the complex permittivity and the complex permeability versus frequency of the $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ (RE = Nd, Tb) powders in 2~18 GHz band.

The graph shows that, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ has higher complex dielectric constant real part ε', this may be mainly related to the fact that the electrical conductivity of Nd is greater than that of Tb's and the complex dielectric constant real part ε' is proportional to the square of the conductivity, thus complex dielectric constant real part ε' of the $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder is higher than that of the $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$'s[8-11]. $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ alloy has high complex dielectric constant imaginary part ε'' in the frequency band of 2~18 GHz. In band of 2~4 GHz, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ has higher complex permeability (real part μ' and imaginary part μ''); along with the frequency increase, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ complex permeability real part μ' will be lower than that of $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$; when frequency is more than about 6 GHz, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ complex permeability real part μ' is again higher than that of $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ real part μ'; but when the frequency is higher than about 4 GHz, $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ complex permeability imaginary part μ'' will be lower than that of $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ complex permeability imaginary part μ''.

According to the formula (1) the reflectivity of wave-absorbing materials can be calculated. As shown in Fig.6, the least reflectivity values of $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ and $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powders are about –9.5 dB at 4.5 GHz and –11 dB at 6.3 GHz when the thickness d=1.8 mm. The main reason why the absorption peak frequency of $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder is lower than that of $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder is that the M_s of $Nd_2Fe_{14}B$ phase in $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder is bigger than that of $Tb_2Fe_{14}B$ phase in $(Tb_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ and according to the relationship

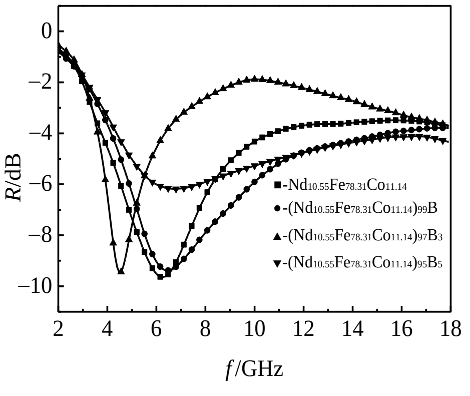

Fig.3 Reflectivity of $(Nd_{10.55}Fe_{78.31}Co_{11.14})_{100-x}B_x$ (x=0, 1, 3, 5) powder

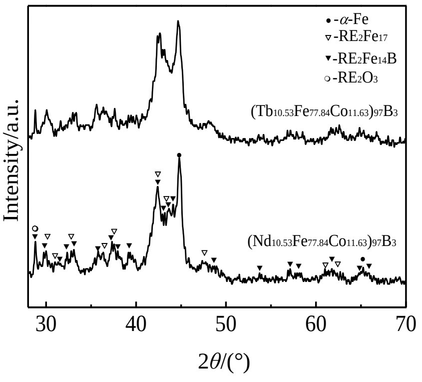

Fig.4 XRD patterns of $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ (RE=Nd, Tb) powders

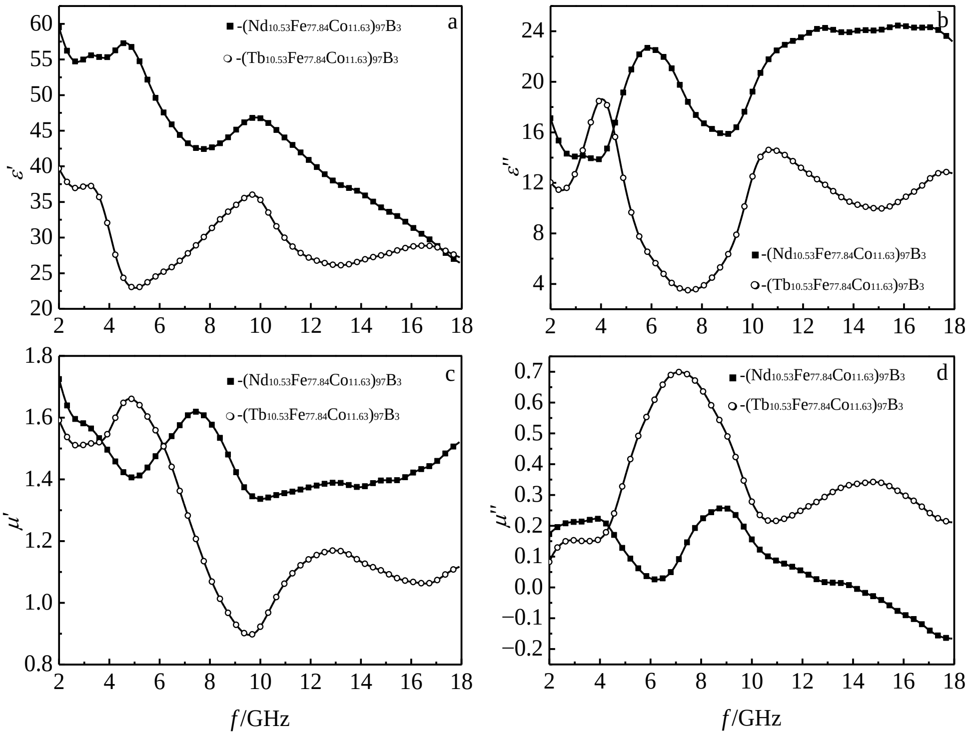

Fig.5 Electromagnetic parameters of $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$(RE=Nd, Tb) powders: (a) curves of ε' vs f, (b) curves of ε'' vs f, (c) curves of μ' vs f, and (d) curves of μ'' vs f

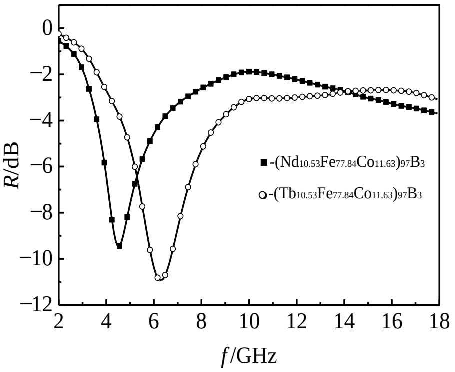

Fig.6 Reflectivity of $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$(RE=Nd, Tb) powder

$f_r \propto 1/M_s^2$, the absorption peak frequency of the $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ powder will decrease[8-10].

3 Conclusions

1) $Nd_2Fe_{14}B$ phase will precipitate when B is added into $Nd_{10.53}Fe_{77.84}Co_{11.63}$ alloy, and the relative content of the α-Fe phase increases. $(RE_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$(RE=Nd, Tb) powders are mainly composed of α-Fe, $RE_2Fe_{14}B$, RE_2Fe_{17} and a small amount of RE_2O_3 phases.

2) When 3% of B is added into $Nd_{10.53}Fe_{77.84}Co_{11.63}$ alloy, the powder has a minimum of absorption peak frequency. When the coating thickness d=1.8 mm, the absorption peak value of the powders reaches –9.5 dB at 4.5 GHz.

3) The absorption peaks of the $(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$ alloy powders shift towards high frequency when Nd is replaced by heavy rare earth Tb.

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RE(Nd, Tb)FeCoB 合金体系的微波吸收特性研究

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摘　要：采用熔炼、高能球磨、微氧化热处理工艺，制备RE(Nd, Tb)FeCoB粉体，借助X射线衍射仪和网络矢量分析仪等，研究不同B含量的NdFeCo粉体组织结构和微波吸收特性，以及用重稀土Tb取代NdFeCoB合金中的Nd后粉体的组织结构和微波吸收特性。结果发现：在$Nd_{10.53}Fe_{77.84}Co_{11.63}$合金中加入B元素后会析出$Nd_2Fe_{14}B$相，而且，粉体中$\alpha$-Fe相的相对含量随着B元素含量的增加而增加；用重稀土Tb取代NdFeCoB合金中的Nd后粉体主要由α-Fe、$Tb_2Fe_{14}B$、Tb_2Fe_{17}以及少量的Tb_2O_3相组成；$(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$粉体具有最低的吸收峰频率，其反射率最小值和吸收峰频率分别为–9.5 dB和4.5 GHz；用重稀土Tb取代$(Nd_{10.53}Fe_{77.84}Co_{11.63})_{97}B_3$合金中的Nd后，粉体的吸收峰频率升高到6.3 GHz，但反射率最小值降低到–11 dB。

关键词：NdFeCoB；微波吸收特性；组织结构；高能球磨

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