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ARTICLE

Structure and Microwave Absorption Properties of Nd-Fe-C Alloys

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Abstract: $Nd_{10.2}Fe_{89.8\text{-}x}C_x$ (x=0, 2.6, 5.2, 7.8) powders were prepared by arc smelting and ball milling. The structure and morphology of the powders were investigated by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The electromagnetic parameters of the powders were measured by vector network analyzer (VNA) at room temperature. It is found that the microwave absorbing properties of $Nd_{10.2}Fe_{89.8\text{-}x}C_x$ (x = 0, 2.6, 5.2, 7.8) depend sensitively on the compositions. The minimum absorption peak frequency shifts to the higher region upon the C content. The minimum reflectivity value of $Nd_{10.2}Fe_{84.6}C_{5.2}$ can reach –13.2 dB at 5.2 GHz and the bandwidth of R <–10 dB reaches 1.3 GHz with the best matching thickness of 1.8 mm. The minimum absorption peak shifts towards lower frequency region with increasing absorbing coating thickness. The minimum absorption peak is related to interference loss.

Key words: NdFeC alloy; ball milling; electromagnetic parameter; microwave absorbing property

With the development of mobile phones, local area network, radar systems, microwave darkroom, etc. in recent years, the electromagnetic waves pollution has aroused concerns about our human beings, which makes most countries in the world have devoted a large amount of manpower and material resources to research absorbing materials[1-4]. A material needs high microwave permeability, high magnetic loss, a proper ratio between the permeability and permittivity, and a favorable form of frequency dependence of permeability to possess good absorbing property[5]. Iron, cobalt, nickel and the corresponding metallic alloys have received much attention due to their various applications in permanent magnet and potential application in microwave absorbing devices[6-9]. Particularly, the Fe-based soft magnetic absorbing material ha attracted much attention for its low price and good absorbing property[10]. C as an absorbing material is considerable due to its excellent conductivity, dielectric properties, low density and corrosion resistance, and the low dosage can achieve high attenuation coefficient of electromagnetic wave[11-13]. With the development of technology, rare earth materials have widespread use in aerospace, automotive, electronics, medical, and military[14-16]. Rare earth (RE) alloys and compounds with the unique 3d-4f interactions between transition metals arise much more researcher' attention, because they can enhance microwave absorption properties, broaden the bandwidth and reduce the matching thickness[17].

The inter-metallic compounds of RE-Fe were used as microwave absorbent due to the high saturation magnetization which can improve the microwave absorbing properties of materials[18].

In this work the Nd-Fe-C alloys were investigated by adjusting the component, with the aim of producing microwave absorbing materials that have excellent reflectivity, and low cost, and are suitable for low frequency and

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industrialized production.

1 Experiment

$Nd_{10.2}Fe_{89.8-x}C_x$ (x=0, 2.6, 5.2, 7.8) powders were synthesized with the Nd, Fe and C whose purity is no less than 99.50%, 99.99%, 99.99%, respectively. The alloys of NdFeC were smelted by a vacuum arc furnace under the protection of argon atmosphere. With the purpose of homogeneous diffusion, the smelted alloys were annealed at 900ºC for 7 d in the hermetic vacuum quartz glass tubes, and then quenched in ice-water mixture. We set planet ball mill (QM-3SP2) speed at 300 r/min, used absolute ethanol to protect the sample, controlled the mass ratio of ball to the powder 20:1, and then mechanically milled the powder for 24 h. The phases of powder were examined by X-ray diffraction (XRD, Empyrean PIXcel 3D, Cu Kα). The morphology of the powder was characterized by scanning electron microscopy (SEM, JSM-5610LV). The mass ratio of powder and paraffin was 4:1 with the paraffin as a compatibilizer for the mixture. The $Nd_{10.2}Fe_{89.8-x}C_x$ was made into a toroidal specimen with thickness of about 3.5 mm, inter and outer diameter of the coaxial sample of 3 mm and 7 mm, respectively. The permeability and permittivity of the composite absorbers were measured in the frequency range of 2~12 GHz by vector network analyzer (VNA,Agilent 5230C).

2 Results and Discussion

2.1 Effect of C content on the structure and morphology of Nd-Fe-C

Fig.1 shows the XRD patterns of $Nd_{10.2}Fe_{89.8-x}C_x$ (x=0, 2.6, 5.2, 7.8) alloys with different C contents. According to the XRD results, the peak of α-Fe is detected except for the main phase Nd_2Fe_{17}. Soft-magnetic phase with high permeability and low coercive force has been reported by R. Keefer's et al[19]. It has been reported that the low coercive force is one of necessary conditions for good absorbing materials[20].

Fig.2 shows the amount of larger size particle increases upon the C content increasing. It is found that most $Nd_{10.2}Fe_{89.8-x}C_x$ powders become flaky after ball milling. Flakiness form is considered to be the best shape for absorbing materials, which has been presented by Ref. [21]. According to the morphology of samples we can know that the powders of $Nd_{10.2}Fe_{89.8-x}C_x$ will have a good performance inabsorbing electromagnetic interference (EMI).

2.2 Microwave absorbing properties of Nd-Fe-C alloys

Fig.3 shows the complex permittivity and permeability of $Nd_{10.2}Fe_{89.8-x}C_x$ (x = 0, 2.6, 5.2, 7.8). In Fig.3, the real parts (ε') decrease with an increase of frequency. The ε'' values decline with the C content increasing. The resonance frequency of ε''

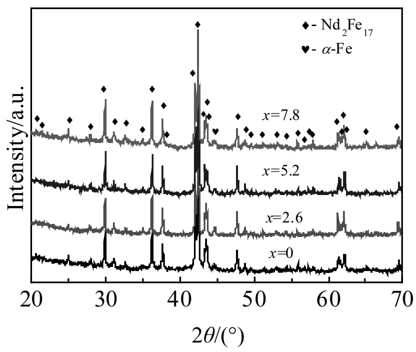

Fig.1 XRD patterns of $Nd_{10.2}Fe_{89.8-x}C_x$ alloys with different contents of C

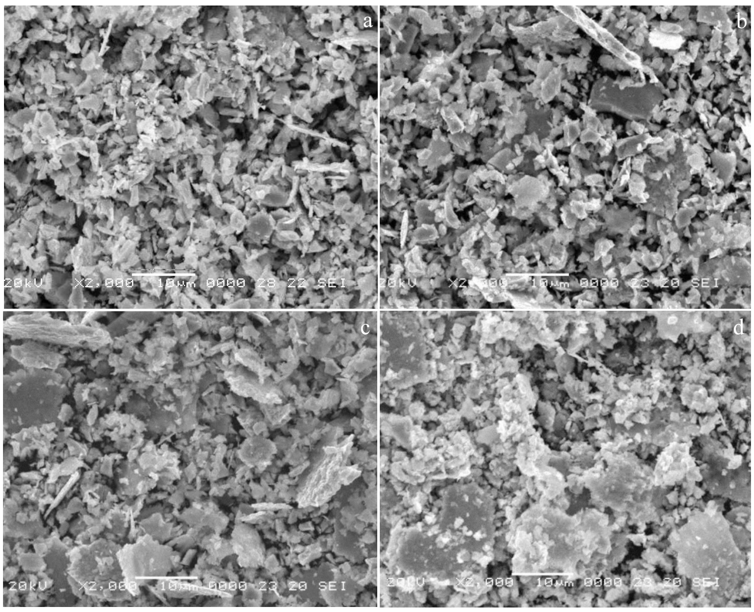

Fig.2 SEM images of $Nd_{10.2}Fe_{89.8-x}C_x$: (a) x=0, (b) x=2.6, (c) x=5.2, and (d) x=7.8

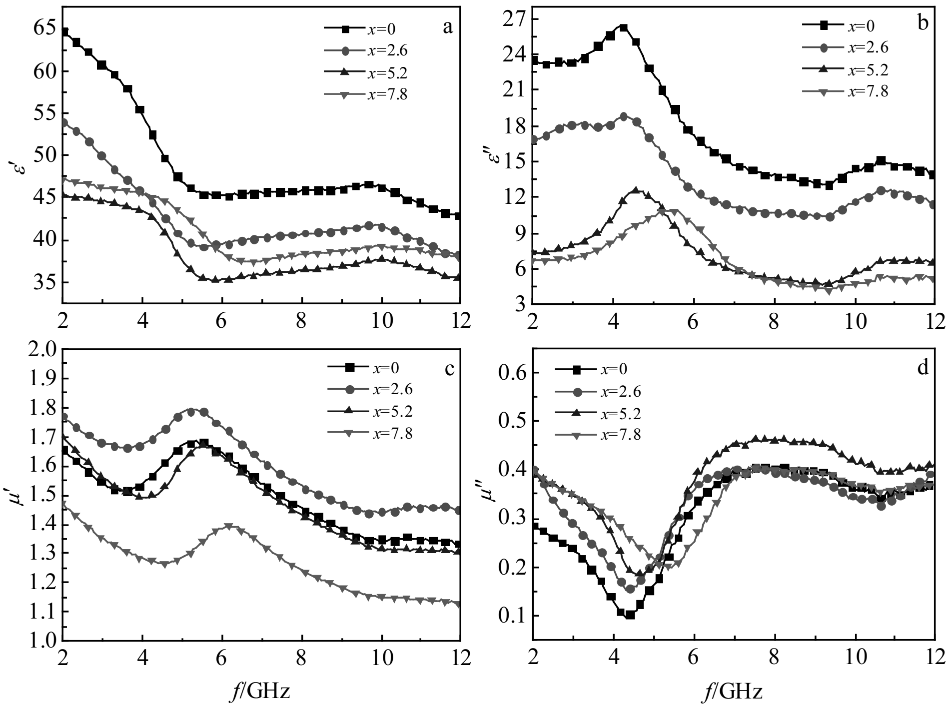

Fig.3 Electromagnetic parameter of $Nd_{10.2}Fe_{89.8-x}C_x$ (x = 0, 2.6, 5.2, 7.8) alloys: (a) ε', (b) ε'', (c) μ', and (d) μ''

shifts to the high frequency region. As shown in Fig.2, the large size particles increases significantly with the increasing amount of C substitution. The large size results in the conductivity increase[22]. According to the investigation of R. Pelster et al[23], resonant frequency of ε'' is proportional to conductivity which leads to the resonant frequency of ε'' shift tohigher frequency region[24]. As shown in Fig.3, the magnetic permeability dispersion also relies on the C concentration. The value of μ' decreases with the frequency enhancement, which may result from natural ferromagnetic resonance and the domain wall motion[25]. The large size particle and amount of flaky particles increase with the increase of C content is shown in Fig.2. The large flaky particles possess low electrical resistivity (ρ); therefore the electrical resistivity (ρ) decreases with the increase of C content[26]. From Ref. [27], we know that M_s decreases with the increase of C content. And μ' is in direct proportion to ρ, Ha and M_s[24]. From Zhang[28], we can know that the Ha (plane anisotropy) increases with the increases of draw ratio. The draw ratio is improved by increasing the particle size. So, when the decrement of ρ and M_s are smaller than the increment of Ha, the value of μ' will increase. Otherwise, the value of μ' will decrease. The dependence of permeability on frequency is complex. The imaginary parts μ'' dispersion curves may be affected by the eddy currents in ferromagnetic materials[29].

According to the generalized transmission line theory, the reflection loss of the powder can be calculated by Eq. (1)[30]:

$$R = 20\lg\left|\frac{\sqrt{\dfrac{\mu_r}{\varepsilon_r}}.\tanh(\mathrm{j}\dfrac{2\pi fd}{c}\sqrt{\mu_r\varepsilon_r})-1}{\sqrt{\dfrac{\mu_r}{\varepsilon_r}}.\tanh(\mathrm{j}\dfrac{2\pi fd}{c}\sqrt{\mu_r\varepsilon_r})+1}\right| \quad (1)$$

where μ_r is relative permeability, ε_r is relative permittivity, f is the frequency of electromagnetic waves, c is wave propagation velocity in free space (or the speed of light), j is the imaginary unit, and d is matching thickness.

The reflectivity R of the samples can be calculated from Eq. (1) with the given thickness and frequency (2~12 GHz), as shown in the absorbing curve of Fig.4. Fig.4 shows that the minimum absorption peak frequency shifts towards higher

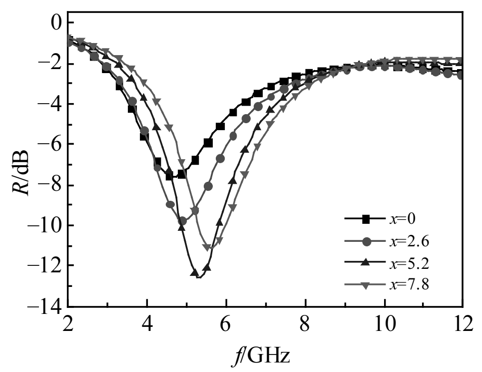

Fig.4 Reflectivity curves of $Nd_{10.2}Fe_{89.8-x}C_x$ (x=0.0, 2.6, 5.2, 7.8) alloys

frequency region upon the C content. With the C content increase the saturation magnetization (M_s) decreases which leads the minimum reflectivity peak value shift to higher frequency region.

In particular, as shown in Table 1, the powders have excellent micro-wave absorption properties when C content increases from x=0.0 to x=7.8. The powder of $Nd_{10.2}Fe_{84.6}C_{5.2}$ has the best properties, the minimum reflectivity value can reach –13.2 dB at 5.2 GHz and the bandwidth of R<–5 dB reach 3.1 GHz with the matching thickness of 1.8 mm. The powder of $Nd_{10.2}Fe_{87.2}C_{2.6}$ also has a good performance in 4.0~6.0 GHz, the minimum reflectivity value can reach –9.8 dB at 5.0 GHz and the bandwidth of R<–5 dB reach 2.2 GHz with the matching thickness of 1.8 mm. It is noteworthy that the powder of $Nd_{10.2}Fe_{82}C_{7.8}$ has good properties in 4.0~6.0 GHz too. The minimum reflectivity is –11.2 dB at 5.8 GHz and the bandwidth of R<–5 dB is 2.3 GHz with the matching thickness of 1.8 mm. Overall, adjusting the C content can make the microwave absorbing materials achieve a good property at different frequencies.

Fig.5 and Table 2 show the minimum reflectivity value of $Nd_{10.2}Fe_{84.6}C_{5.2}$ with different thickness. There is a relationship between the absorber thickness d_m and magnetic loss frequency (f_r) of absorbers by Eq. (2)[31]:

$$d_m = \frac{\lambda_m}{4} = \frac{nc}{4 f_r \sqrt{\mu_r \varepsilon_r}} \quad (n = 1, 3, \ldots) \qquad (2)$$

where d_m is the matching thickness and f_r is the frequency of a minimum absorption peak, ε_r and μ_r are the complex permittivity and permeability at f_r, respectively, and c is the velocity of light[32].

Table 1 Minimum *R* and peak frequencies with different contents of C (*d* = 1.8 mm)

C content, x	0	2.6	5.2	7.8
Minimum R/dB	–7.8	–9.8	–13.2	–11.2
f_m/GHz	4.6	5.0	5.2	5.8
Frequency width of R < –5 dB/GHz	1.8	2.2	3.1	2.3

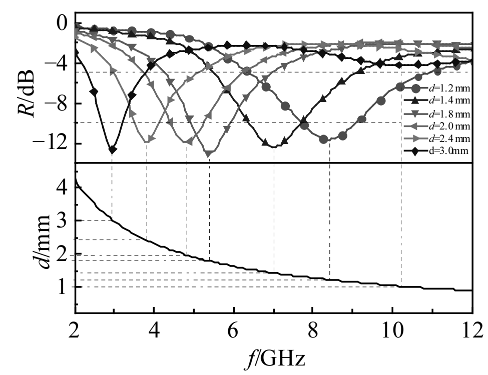

Fig.5 Reflectivity and λ/4 thickness on frequency at various thickness for the $Nd_{10.2}Fe_{84.6}C_{5.2}$

Table 2 Minimum *R* and peak frequencies of $Nd_{10.2}Fe_{84.6}C_{5.2}$ alloy with different thicknesses

d/mm	1.2	1.4	1.8	2.0	2.4	3.0
Minimum R/dB	–11.8	–12.3	–13.2	–12.0	–11.9	–12.8
f_m/GHz	8.5	7.0	5.2	5.0	3.9	3
Frequency width of R<–10 dB/GHz	1.6	1.5	1.3	1.2	0.8	0.6

Apparently, it can explain that the minimum reflectivity peak value shifts to lower frequency with the thickness increase. In the present work and according to Eq. (2), adjusting the thickness can make the microwave absorbing materials achieve a good property at different frequencies. Overall, the $Nd_{10.2}Fe_{84.6}C_{5.2}$ powder has the potential to promote the development of the microwave absorbing materials. On the other hand, Fig.5 shows a relationship between d_m and λ/4. This result reveals that the appearance of the minimum reflectivity peak is caused by the destructive interference[33].

According to Eq. (1) and Eq. (2), we can know that the absorbing mechanism of $Nd_{10.2}Fe_{89.8-x}C_x$ powder on the electromagnetic wave is related to electromagnetic loss within the absorbing coatings and the effect of coating thickness on the interference loss of electromagnetic wave.

3 Conclusions

1) $Nd_{10.2}Fe_{89.8-x}C_x$ (x=0, 2.6, 5.2, 7.8) alloy powder maintain the phase of Nd_2Fe_{17} except for the α-Fe phase.

2) The minimum reflectivity peak value shifts towards a higher frequency region with the C content increase. The powder of $Nd_{10.2}Fe_{84.6}C_{5.2}$ has good properties in 4.0~6.0 GHz, the minimum reflectivity peak value can reach –13.2 dB at 5.2 GHz and the bandwidth of R<–10 dB achieves 1.3 GHz with the best matching thickness of 1.8 mm. The minimum reflectivity peak value of $Nd_{10.2}Fe_{84.6}C_{5.2}$ can be adjustable in 2.0~10.0 GHz by changing the thickness.

3) The absorbing mechanism of $Nd_{10.2}Fe_{89.8-x}C_x$ powder on the electromagnetic wave is related to electromagnetic loss within the absorbing coatings and the effect of coating thickness on the interference loss of electromagnetic wave.

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Nd-Fe-C 合金的结构及其微波吸收特性

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摘 要：采用电弧熔炼及球磨工艺制备出 $Nd_{10.2}Fe_{89.8-x}C_x$ (x=0, 2.6, 5.2, 7.8) 合金微粉，借助 XRD、SEM 和网络矢量分析仪等仪器分别对合金微粉的结构、形貌及其室温下微波吸收性能进行了研究。研究发现，随着 C 含量的增加，$Nd_{10.2}Fe_{89.8-x}C_x$ 合金的最小反射峰频率向高频方向移动，其中 $Nd_{10.2}Fe_{84.6}C_{5.2}$ 合金具有最好的吸波效果。在最佳匹配厚度 1.8 mm 下，$Nd_{10.2}Fe_{84.6}C_{5.2}$ 合金的最小反射损耗在 5.2 GHz 处达到–13.2 dB 左右，反射损耗小于–10 dB 的频带宽度达到了 1.3 GHz。随着匹配厚度的增加，最小反射损耗向低频移动，最小反射损耗与干涉损耗有关。

关键词：NdFeC 合金；球磨；电磁参数；微波吸收特性

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