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A simple method to improve dielectric property of SiC powder

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Aluminum-doped silicon carbide powders were synthesized via solid state reaction of Si/C system at 1600□ in Ar atmosphere, using aluminum powder as the dopant, which were investigated by X-ray diffraction (XRD) and scanning electronic microscope (SEM). The electric permittivities of the prepared powders were determined in the frequency range of 8.2–12.4 GHz. The dielectric real part ε' and imaginary part ε'' of undoped powder have minimum values (ε' = 5.5–5.3, ε'' = 0.23–0.20), and increase with increasing aluminum content. The mechanism of dielectric loss by doping has been discussed.

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Keywords: Silicon carbide, Dielectric property, Solid state reaction

1. Introduction

Silicon carbide has many excellent properties, e.g. high strength and hardness, good corrosion resistance, and high thermal stability and thermal conductivity; it is often considered as the most important carbide, which has wide applications from structural materials to electric devices at elevated temperatures [1-3]. In recent years, increasing levels of electromagnetic pollution (due to the use of electronic and telecommunication systems) has recently focused attention on the absorbing material, especially SiC materials, in the microwave range. Compared with the other absorbing material, such as spinel ferrites, hexagonal ferrites, and metallic magnetic materials, etc., which are magnetic absorbers with lower Curie temperature, SiC are dielectric loss material, which has been used in the high temperature components of aircraft.[4]

However, the pure SiC material presents the poor absorbing ability in the gigahertz (GHz) band range. Therefore, Li et al. have prepared Ni-single-doped (*p*-type doping) SiC powders using the mechanically activated self-propagating high-temperature synthesis method and dielectric property of doped SiC powder presents better than that of the pure SiC powder.[5] Li and Luo et al. have obtained the B and Al-single-doped (*p*-type doping) SiC powders using the *sol–gel* method and thermal diffusion, and presented the better dielectric properties owing to the bound holes generated by the B and Al-doping.[6,7] Jin et al. have synthesized the Al-single-doped (*p*-type doping) SiC using microwave method, which shows also the better dielectric property of SiC in the same frequency range.[8] Zhao and Jiao et al. have prepared the N-single-doped (*n*-type doping) SiC powder by laser synthesis and chemical vapor deposition, respectively, which presents the better dielectric property in the frequency range of 8.2-12.4GHz [9,10].

It can be seen that the dielectric property of SiC powder has been improve by the *p*-type or *n*-type doping. So, in the paper, the Al-single-doped SiC powder has been prepared by the simplest method, solid state reaction. The dielectric property has been determined in the frequency range of 8.2 to 12.4GHz. The mechanism of dielectric loss has been discussed.

2. Experimental procedure

Silicon powder (99% pure, mean particle size of 20 mm; Tianjin Kermel Chemical Reagents Development Centre, China) and carbon black (99% pure, particle size of 20–40 nm; Jiaozuo Chemical Co. Ltd., China) were used as reactant materials. Aluminum powder (99% pure, mean particle size of 20 mm; Shanpu Chemical Co. Ltd., Shanghai, China) was used as doping source. The reactant powders of Al, Si and C were weighed out in molar rations of 0:1:1, 0.05:0.95:1 and 0.1:0.9:1, respectively. The powder batches were mixed in ethanol for 20 h and then dried at 60℃. The mixed powders were poured into a graphite crucible and calcined at 1600 ^{o}C for 15 minutes in a 0.1 MPa Ar atmosphere inside a resistance heating graphite furnace.Additionally, because the excess carbon in CS products will affect the accuracy of Raman spectra and dielectric property, theprepared powders were fired at 650 ℃ in air atmosphere for 1 h to remove the excess carbon.

The CS products were identified by X-ray diffraction (XRD, X'Pert PRO MPD, Cu Ka). The 99.99%Si (a = 5.43088Å) was used as inner standard. The lattice constant of prepared SiC powders was calculated by software Jade 5.0. The morphology of the CS powders was investigated using scanning electronic microscope (SEM, JSM-6360LV, JEOL, Tokyo, Japan). Because of the no dielectric loss of paraffin, the samples for dielectric parameters measurement at room temperature were prepared by mixing the produced powders with paraffin in a mass ratio of 20:80, and then the mixtures were molded into a brass flange to fabricate rectangular composite samples with the dimensions of 10.16 mm (width)×22.86 mm (length)×2 mm (thickness). The dielectric parameters were carried out by waveguide technique with mode TE10 in the frequency range of 8.2–12.4 GHz, with the prepared samples set in a brass holder in which fills the waveguide. After being calibrated with an intermediate of a short circuit and blank holder, reflection and transmission coefficients were obtained by PNA network analyzer (Agilent Technologies E8362B, Palo Alto, CA), and then both the real and imaginary parts of the permittivity were given.

3. Results and discussion

Fig. 1 shows the XRD patterns of the prepared powders doped by different Al content. It can be seen that the SiC powders have been synthesized at 1600 °C although the reaction of Si and C has the poor energy. In the XRD pattern of undoped sample, all peaks correspond to the β-SiC phase and no second phase has been observed. It indicates that the prepared undoped powder is pure β-SiC phase. For the doped samples, except for β-SiC phase the α-SiC phase has also been found. It may be that the Al-doping has generated the transformation of β-SiC to α-SiC. In addition, for the 10%Al doped powders, the Al_2O_3 phase has also been observed. It is possible that the content of Al in SiC lattice reaches the solid solubility limit and the Al_2O_3 has been generated by the reaction of excess Al and adsorbed oxygen.

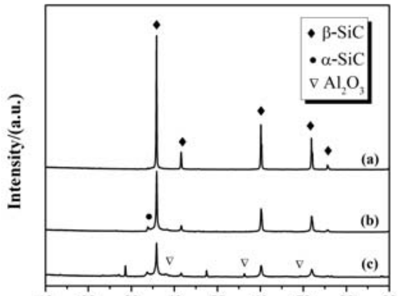

Fig. 1. XRD patterns of the prepared powders doped by different Al content (a) 0%; (b) 5%; (c) 10%.

Furthermore, the lattice constant corresponding to undoped β-SiC is only 4.346Å, which is much less than the standard value of β-SiC(4.358Å). It is due to the formation of carbon antisites [11]. So, the prepared product is C-enriched SiC powder which is a more suitable material for *p*-type doping, e.g. acceptor doping with Al [12]. It has been observed that compared with undoped β-SiC, all the peaks of doped SiC shift in the direction of lower 2θ, suggesting the increase of the lattice constant a of β-SiC due to the increase of interplanar distance d according to the Bragg formula. The lattice constants a corresponding to β-SiC doped with 5% and 10% Al are 4.355 and 4.359Å, respectively. Because the covalent radius of Al (1.18Å) is greater than that of Si (1.11Å), the increase of lattice constant is caused possibly by the Al substitution on Si of SiC lattice. So the more substitution, the higher lattice constant is.

The SEM images of the prepared powders doped by different Al content have been shown in Fig. 2. It can be seen that for the undoped sample it present better crystallization and the mean particle sized is about 1-2 μm. However, for the Al-doped samples, the mean particle size is about 0.1μm, which is due to the fact that the Al doping affects the crystallization of SiC and retards the crystalline growth.

To examine in more detail the chemical composition of prepared powders with different Al contents, the EDS analysis was performed, as shown in Fig. 3. For undoped product only C and Si elements are observed and the molar ratio of C to Si is about 1.3, which indicates the undoped product is C-enriched SiC powder. However, for the doped products the Al element has been also observed besides C, Si, and O. In addition, the Al content increases and Si content decreases with increasing Al content, as shown in Table 1. It indicates that the more Si atoms are substitutes by the more Al atoms, thus resulting in the increase of lattice constant of SiC, which is in agreement with results in Fig. 1.

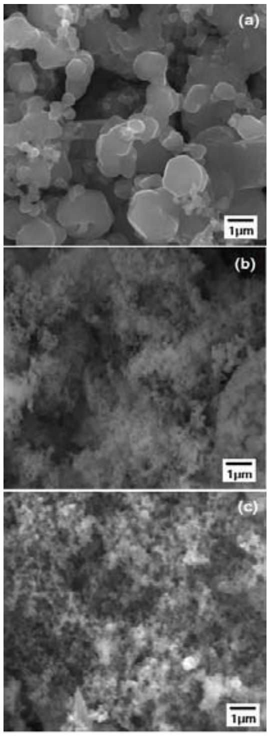

Fig. 2. SEM images of the prepared powders doped by different Al content (a) 0%; (b) 5%; (c) 10%.

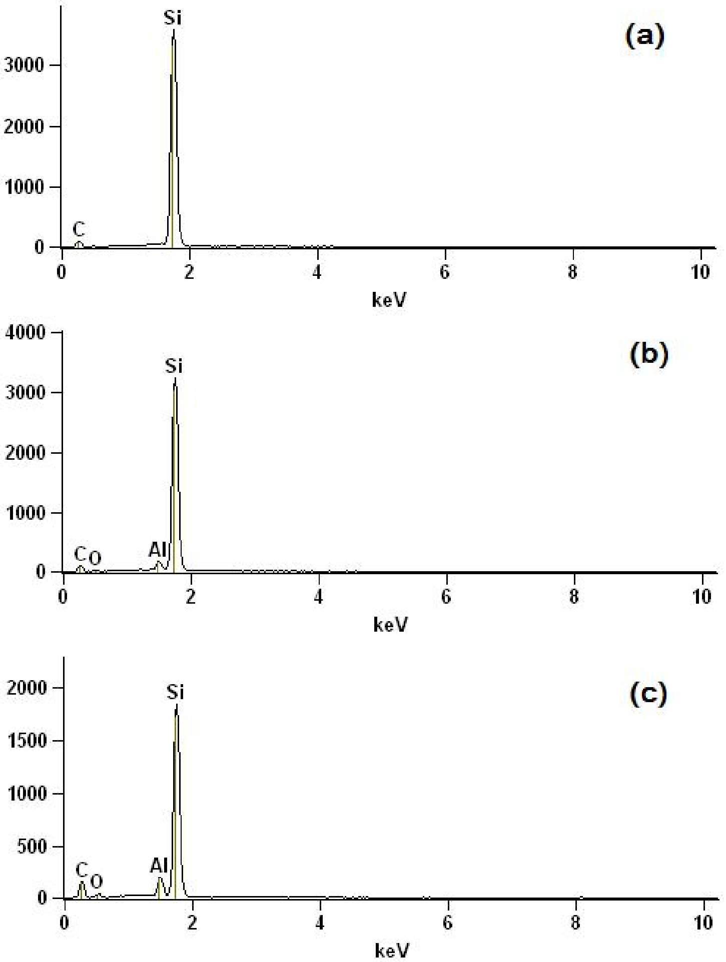

Fig. 3. EDS patterns of the prepared powders doped by different Al content (a) 0%; (b) 5%; (c) 10%.

Table 1. Atom ratios of prepared SiC powders.

Samples	**C**	**Si**	**Al**	**O**
(a)	57.37	42.63	0	0
(b)	61.54	37.09	0.87	0.50
(c)	68.79	27.70	1.68	1.83

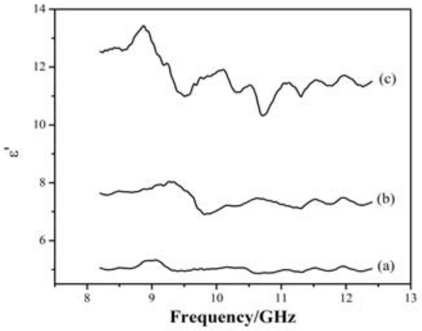

Fig. 4. Dielectric real part ε' as a function of frequency in the range of 8.2-12.4 GHz for the prepared powders doped by different Al content (a) 0%; (b) 5%; (c) 10%.

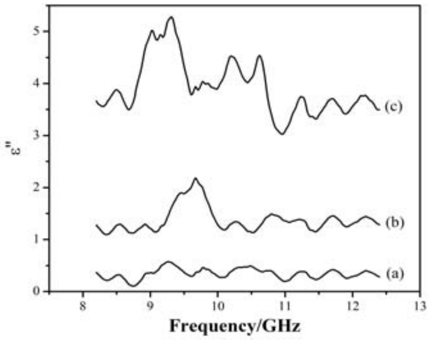

Fig. 5. Dielectric imaginary part ε'' as a function of frequency in the range of 8.2-12.4 GHz for the prepared powders doped by different Al content (a) 0%; (b) 5%; (c) 10%.

Figs. 4 and 5 show the dielectric real part ε' and imaginary part ε'' as a function of frequency in the range of 8.2–12.4 GHz for the prepared powders with 0, 5 and 10% Al, respectively. For the undoped SiC powder, the ε' and ε'' are 5.2-5.0 and 0.5-0.1, respectively. However, Compared with the undoped product, for the 10%Al doped SiC powder both of them have reached maximum values, 13.4–10.3 and 5.3–3.0, respectively, and were improved by about 15%. Although there is a few amounts of Al_2O_3 in the product (c), the ε' and ε'' are very small and there is almost no influence on the ε' and ε'' of doped SiC powder. Therefore, the ε' and ε'' increase significantly with increasing Al content. Furthermore, when Al dopant content is 10% the ε' and ε'' of sample show the better frequency dispersion behavior, which is that complex permittivity and loss decrease with increasing frequency. In this work, the prepared products are *p*-type SiC powders by Al doping. The Al atoms enter the SiC lattice and substitute the Si of SiC lattice and there will exist bound holes around Al_{Si} defects in the SiC crystal [6]. Under the alternating electromagnetic field, these bound holes will migrate to and fro to form relaxation polarization and loss, thus leading to higher ε' and ε'', respectively.

4. Conclusions

The Al-doped SiC powders have been synthesized successfully by solid state reaction method. The prepared undoped product is C-enrich SiC powder and the lattice constants of doped SiC powders increase with increasing Al dopant content due to the Al-doping. The particle sizes of prepared SiC powders decrease significantly with the increase of Al dopant content. The dielectric real part ε' and imaginary part ε'' of SiC powder doped by 10%Al have the maximum values and present the best dielectric loss. The β-SiC powders doped with N have higher ε' and ε'' due to the relaxation polarization and loss of bound hole from Al_{Si} defects, respectively.

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