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Dielectric properties of Al-doped Ti_3SiC_2 as a novel microwave absorbing material

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ABSTRACT

Al-doped Ti_3SiC_2 powders were synthesized by solid state reaction under a vacuum atmosphere from Ti/Si/TiC powders, with an optimum Al doping content. Results showed that the Ti_3SiC_2 major phase could be generated at a temperature as low as 1250 °C through Al doping, and that the doped powders had a relatively narrow particle size distribution with a good dispersibility. The formation of $Ti_3Si_{1-x}Al_xC_2$ solid solution was further proved by XPS. The microwave dielectric parameters and reflection loss of the prepared Al-doped Ti_3SiC_2 samples were determined in the frequency range of 8.2–12.4 GHz. It was found that the sample synthesized at 1350 °C showed the high values in both the imaginary part of permittivity ε'' and dielectric loss tan δ, which were 4.39–7.32 and 0.57–0.78, respectively. For the microwave absorbing coating with a 2.6 mm thickness of the sample, a better reflection loss of almost below −12 dB was obtained in the whole frequency range of 8.2–12.4 GHz.

1. Introduction

In recent years, microwave absorbing materials have been paid much attention because of the demand of military stealth technologies, which have been also developed for the elimination of electromagnetic wave contamination from the extensive applications of electronic devices [1–3]. Presently, the microwave absorbing materials are usually categorized into two types: microwave absorbing coating materials and monolithic structural microwave absorbing materials [4]. Both the materials need dielectric and/or magnetic absorbers to dissipate incident electromagnetic energy into heat through dielectric or magnetic loss [5,6].

Among the microwave absorber materials, the magnetic absorbers, such as ferrites [7] and carbonyl iron [8], have been widely applied over the past decades. However, the ferrites and carbonyl-iron have a higher density and an inferior absorbing characteristic at the higher temperature environment due to the limitation of Curie temperature [9,10]. For dielectric absorbers, the carbon-based materials with light weight, e. g. the carbon black, carbon fiber and nanotube, and graphite, have been extensively investigated, which showed good microwave absorbing properties arising from the dielectric loss [11,12]. Additionally, many studies have focused on magnetic/dielectric composite systems to broaden further the microwave absorbing frequency band. Namely, the ferrites and carbonyl iron are combined with the dielectric absorbers, such as the conducting polymer, carbon nanotube and graphene [13–15]. Although all the above involved dielectric materials have the potential applications in microwave absorption, they are also unsuitable for employing at the high temperature environment because of their lower oxidation resistance. In order to overcome these problems, the carbides with the good oxidation resistance including SiC [16] and Ti_3SiC_2 [5] have been studied for the microwave absorbing application.

The layered ternary carbide Ti_3SiC_2 has been considered as a potential structure/functional material because of its combined properties, such as low density, high modulus, good thermal and electrical conductivity, good oxidation resistance and easy machinability [17], in which the good oxidation resistance and electrical conductivity render Ti_3SiC_2 to become a potential microwave absorbing material used at a high temperature environment. Our previous studies demonstrated that Ti_3SiC_2 had the microwave absorbing characteristic which was closely relevant with its purity [5]. Zou et al. [18] reported that the purity of Ti_3SiC_2 material could be improved by Al doping. Based on this, we sequentially studied the microwave dielectric properties of Al-doped Ti_3SiC_2. As a result, Al doping improved the microwave dielectric properties of Ti_3SiC_2, in which the refection loss was less than −10 dB within a frequency width of 2 GHz [19]. Nevertheless, the microwave dielectric properties of Ti_3SiC_2 further improved by choosing an optimum Al doping content has not been addressed yet. In this study, we reported the better microwave dielectric properties of Ti_3SiC_2 doped with an optimum Al content, and discussed the mechanism of the dielectric properties improved by Al doping in the frequency range of 8.2–12.4 GHz.

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2. Experimental

Titanium powder (99% in purity, mean particle size of 75 μm), silicon powder (99% in purity, mean particle size of 40 μm) and titanium carbide powder (99% in purity, mean particle size of 75μm) were used as the starting materials, and aluminium powder (99% in purity, mean particle size of 40 μm) was used as the dopant. The powders of Ti/Si/Al/TiC were weight out in a mole ratio of n_{Ti}:n_{Si}:n_{Al}:n_{TiC}=2:1.8:0.4:3. For comparison, the powders of Ti/Si/TiC were also weight out in a mole ratio of n_{Ti}:n_{Si}:n_{TiC}=2:2:3, which did not contain the Al dopant. Then, the powder bathes were mixed in ethanol for 6 h using planetary milling with zirconia ball media and dried at 60 °C. Finally, the dried mixtures were placed into the vacuum sintering furnace and calcined at the temperatures of 1250–1400 °C for 2 h to prepare the Al-doped Ti_3SiC_2 powders, with a heating rate of 10 °C/min and a vacuum degree of about 0.1 Pa.

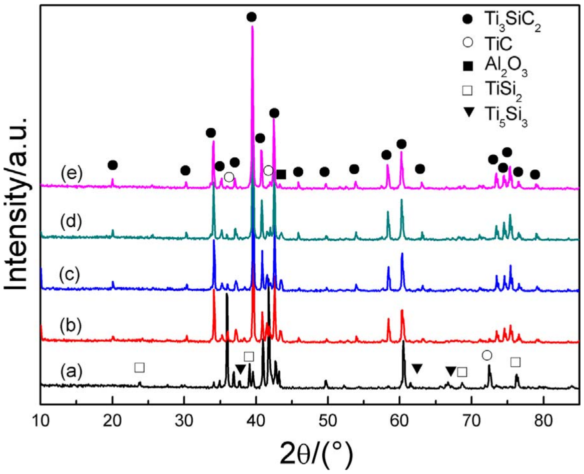

Fig. 1. XRD patterns of the Al-doped Ti_3SiC_2 powders synthesized at different temperatures: (a) undoped powder at 1250 °C; (b) Al-doped powder at 1250 °C; (c) Al-doped powder at 1300 °C; (d) Al-doped powder at 1350 °C; (e) Al-doped powder at 1400 °C.

The crystalline phases of the as-prepared powders were identified by X-ray diffraction (XRD, DX-1000, Fangyuan instrument Co. Ltd., China) with Cu K_α radiation. The morphologies of the powders were observed by scanning electron microscopy (SEM, JSM-6360LV, JEOL, Japan), and the chemical compositions were determined by X-ray photoelectron spectroscopy (XPS, K-Alpha, Thermo Scientific) using Al K_α X-ray source.

The samples for dielectric parameter measurements at room temperature were prepared by mixing the prepared powders with the molten paraffin in a mass ratio of 1:1. The mixtures were molded into a brass flange to fabricate the rectangular composite samples with the dimension of 22.86 mm(Length)×10.16 mm(Width)× 2.26 mm(Thickness). The dielectric parameters of the samples were determined by PNA vector network analyzer (Agilent Technologies E8362B, Palo Alto, CA) with a waveguide technique in the frequency range of 8.2–12.4 GHz.

3. Results and discussion

Fig. 1 shows the XRD patterns of the unoped and Al-doped Ti_3SiC_2 powders synthesized at the temperature of 1250–1400 °C from Ti/Si/

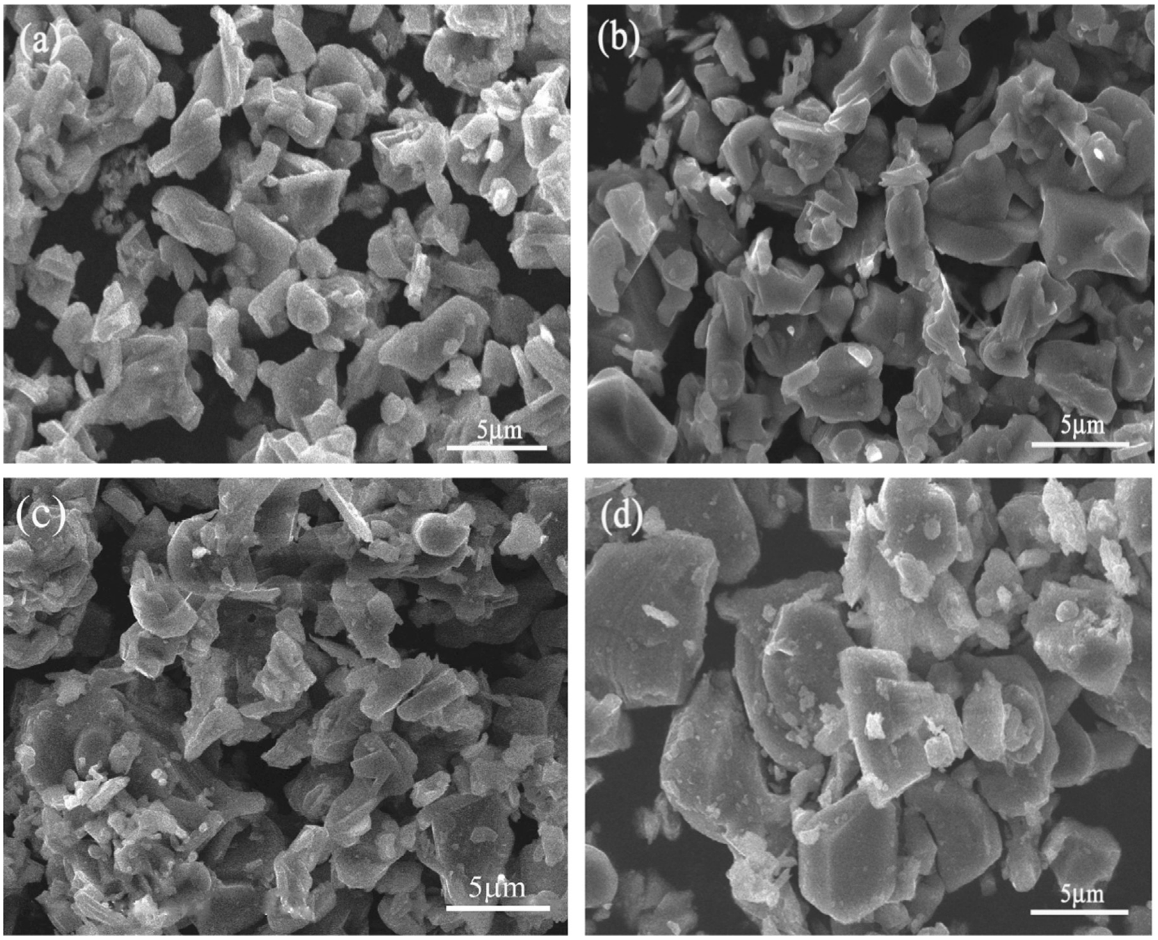

Fig. 2. SEM images of the Al-doped powders synthesized at different temperatures: (a)1250 °C; (b)1300 °C; (c)1350 °C; (d)1400 °C.