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Effect of Ti_3SiC_2 addition on microwave absorption property of Ti_3SiC_2/cordierite coatings

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ABSTRACT

Ti_3SiC_2/cordierite (MAS) composite coatings with different Ti_3SiC_2 concentrations were prepared via atmosphere plasma spraying (APS) method. The characteristics of the Ti_3SiC_2/MAS powders and as-sprayed coatings, such as microstructure, and phase constitution, were observed and measured. The effect of the Ti_3SiC_2 addition on the electromagnetic shielding, dielectric, and microwave absorption properties of Ti_3SiC_2/MAS coatings in the frequency of 8.2–12.4 GHz was measured and discussed. The addition of Ti_3SiC_2 can significantly improve the electromagnetic shielding and complex permittivity of the coatings, which can be ascribed to the enhanced polarization effect and electrical conductivity. When the Ti_3SiC_2 content increases to 30 wt.%, the coating shows an excellent microwave absorption property, and the effective absorption bandwidth (≤ -5 dB) can be obtained across the whole measured frequency with a thickness of 1.8 mm. Further increase in Ti_3SiC_2 content led to high permittivity, which is harmful to the impedance match and results in strong reflection and weak absorption.

1. Introduction

Recently, with the rapid development of electronic devices and telecommunication systems, microwave absorbing materials (MAMs) have attracted much attention in commercial and military applications. For military purpose, the development of MAMs applied in the frequency range of 8.2–12.4 GHz (X-band) is necessary [1–5]. The MAM is a special designed material or material system to convert the incident electromagnetic energy into heat by the mechanism of dielectric and/or magnetic loss, aimed to reduce the reflected electromagnetic energy of the target. As an ideal MAM, it should possess light weight, high microwave absorption, broad width, tunable absorption frequency, and multifunctional [6]. Traditional MAMs, such as carbonyl-iron [7,8], ferrites [9], and barium titanate [10], have been used and researched widely because of their properties of high magnetic loss and dielectric loss. Besides, a variety of carbon-based composite materials have been investigated for potential applications as MAMs at room temperature [11–14]. However, the disadvantages of heavyweight and low resistance to elevated temperature restricted their widespread applications.

MAX phase materials (where M denotes an early transition metal, A is a mostly IIIA or IVA group element, and X is either C or N) have been studied extensively because of their combination of numerous salient properties of both metals and ceramics [15]. Ti_3SiC_2 is the typical compound of MAX phase material, with a unique combination of mechanical, electrical, and thermal properties of both metals and ceramics [16–20]. Its room-temperature electrical conductivity is 4.5×10^6 $(\Omega \cdot m)^{-1}$, which is roughly twice that of pure Ti and more than four times that of near-stoichiometric TiC, and thus Ti_3SiC_2 as an excellent conductive filler is suitable for conductive composites [20]. However, there are few published literatures investigating the microwave absorption property of Ti_3SiC_2, especially for plasma sprayed coatings. During the plasma spray process, Ti_3SiC_2 undergoes a high-temperature environment and behavior of Ti_3SiC_2 will have a vital influence on the property of the Ti_3SiC_2 coatings.

This study is focused on the influence of Ti_3SiC_2 addition on the electromagnetic shielding, dielectric, and microwave absorption properties of Ti_3SiC_2/MAS composite coatings. In this study, the Ti_3SiC_2/MAS coatings with different Ti_3SiC_2 addition were fabricated by a micro-plasma spraying system developed by the Second Artillery Engineering College [21]. Atmospheric plasma spraying (APS) is a process that uses plasma arc to melt or soften feedstock, such as metal and ceramics, propels the molten particulates toward a prepared surface, and then forms a functional coating. Compared with other coating preparation technology, APS has the advantage of relatively high deposition rates and efficiency, low cost, and high flexibility [22–24].

The S-parameters, complex permittivity, and permeability of the coatings were investigated in the frequency range of 8.2–12.4 GHz. Furthermore, the reflection loss of the coatings was calculated and the possible microwave absorbing mechanisms were discussed.

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2. Experimental procedure

2.1. Materials

Cordierite (MAS) has a combination properties of low dielectric constant ($\varepsilon = 5$–6), high resistivity ($\rho > 10^{12}$ Ω•m), elevated thermal and chemical stabilities, and very low thermal expansion coefficient ($\alpha =$ 1–2 $\times$ 10^{-6} °C^{-1}), which makes it a potential available material employed in the electronic industry [25,26]. Yi Liu et al. [26] have prepared pure MAS ceramic and found that the real and imaginary part of complex permittivity for pure MAS ceramic were about 4.5 and 0.02, respectively, which indicated that the pure MAS shown weak electromagnetic shielding and microwave absorption properties.

Industrial-grade raw MAS ($Mg_2Al_4Si_5O_{18}$) powders with an average diameter of 50 μm, according to the supplier, were purchased from Hebei Xingtai New Refractory Materials Co. Ltd. (China). A commercial grade of Ti_3SiC_2 powders purchased from Shanghai Yuehuan New Materials Technology Co. Ted. (China) were introduced as microwave absorber in this study. Fig. 1(a) and (b) show the scanning electron microscopy (SEM) photographs of the started materials of MAS and Ti_3SiC_2, respectively.

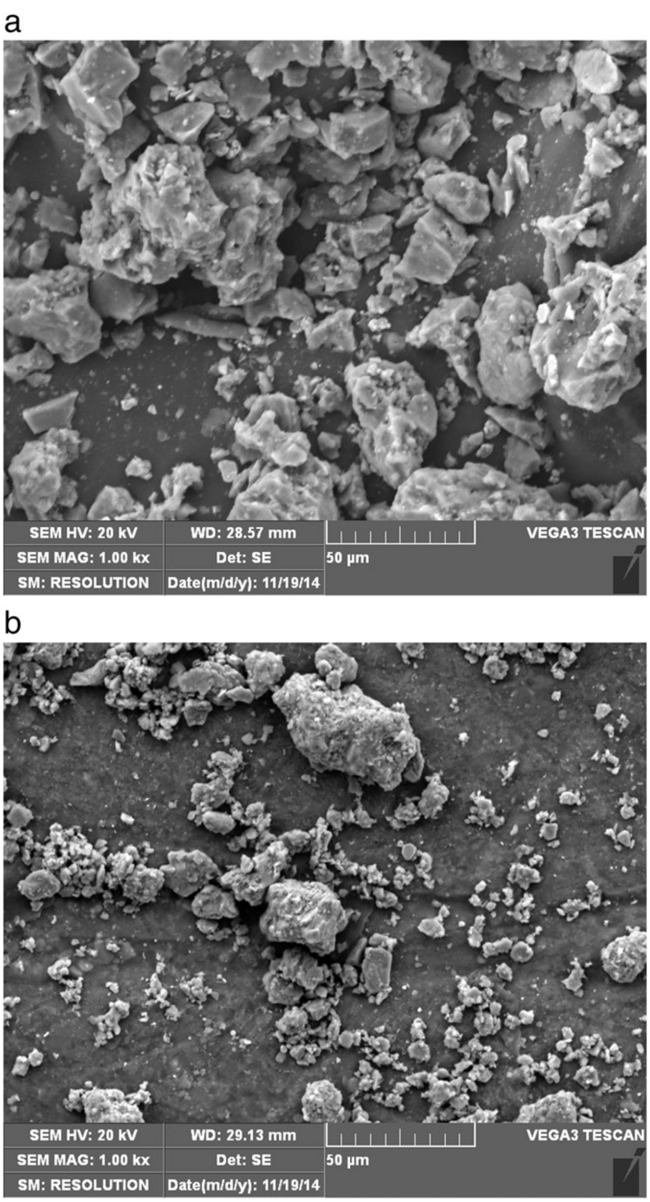

Fig. 1. SEM photographs of (a) the MAS powders, (b) the Ti_3SiC_2 powders.

2.2. Preparation process

The most versatile powder preparation method for thermal spraying is spray drying, which allows any kind of small material particulates to be kept together in a large spherical agglomerate with an organic binder [27]. The spray-drying process was as follows: (i) the Ti_3SiC_2 and MAS powders were blended uniformly to produce viscous slurry with the addition of binder (PVA) and deionized water by planetary wet ball milling (QM-3SP4, Nanjing NanDa Instrument Plant, China). Al_2O_3 balls (6 mm in diameter) were used as the milling media, and the wet ball milling process continued for 3 h with a rotary speed of 300 r/min in order to blend the particles homogeneously. The Ti_3SiC_2 and MAS powders were taken in the weight ratio of 10/90, 15/85, 20/80, 30/70, and 40/60, which were abbreviated as 10TSC, 15TSC, 20TSC, 30TSC, and 40TSC in the figures. Fig. 2 displays the size distribution curve of the ball-milled powders and the size of powders ranges from 2 to 10 μm and centers around of 5 μm.(ii) The slurry of powder mixture was spray dried by a centrifugal spray dryer (LGZ-5, Wuxi Dongsheng Spray-drying Machinery Plant, China). The pump fed the slurry into a centrifugal atomizer and the moisture contained in the droplets evaporated during flight in the stream of cleaned and heated air in the chamber. The parameters of spray drying were listed in Table 1. (iii) The solid particles were collected in the powder collector and dried in the stove at 120 °C for 2 h. Fig. 3 displays the SEM photograph of Ti_3SiC_2/MAS sprayable feedstocks with 30 wt.% Ti_3SiC_2 concentration after spray drying, which exhibits spherical morphology with a diameter range from 40 to 100 μm. The feedstock powders are reconstituted from the fine particles and the surface is relative smooth.

Coating samples were prepared by a micro-plasma spraying system in air atmosphere developed by the Second Artillery Engineering College. Argon and nitrogen were adopted as primary gas and secondary gas at a flow rate of 20 and 3 standard liters per minute (slpm), respectively. The plasma torch power during operation was about 9.8 KW. More details of the plasma spraying parameters were listed in Table 2. The coatings were sprayed on a graphite substrate in a thickness about 2.5 mm and then mechanically removed from the substrate.

2.3. Characterization

The scanning electronic microscope (SEM, Model JSM-6360, Tokyo, Japan) attached with a Links System energy dispersive spectrometer (EDS) was used to observe the morphology and analyze the elemental distribution of coatings. The crystalline phases of the coatings were identified by X-ray diffraction (XRD, Philips, Netherlands) with Cu-K

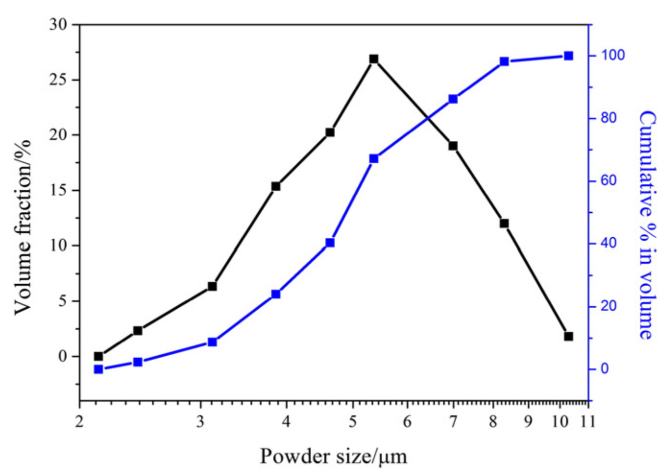

Fig. 2. Size distribution and cumulative curve of the ball-milled powders.