Jestr

Journal of Engineering Science and Technology Review 6 (12 (2013) 160-163

Research Article

JOURNAL OF
Engineering Science and Technology Review

www.jestr.org

Nano-Structured Si/C/N Composite Powder Produced by Radio Frequency Induction Plasma and its Microwave Absorbing Properties

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Received 15 May 2013; Accepted 25 July 2013

Abstract

Nano-structured Si/C/N composite powders were produced by radio frequency (RF) induction plasma utilizing methyltrichlorosilane (CH_3SiCl_3) and nitrogen (N_2) as precursors. The micro-morphology, chemical composition, functional group structure and absorbing properties of the samples were investigated. The results indicate that the size of Si/C/N particles is 0~50 nm with homogeneous distribution. The Si/C/N composites mainly consist of SiC (40.97%) and Si_3N_4 (51.03%). The Fourier transform infrared (FT-IR) spectroscopy characterization shows that the composites have stronger Si-C (780 cm-1) and Si-N (950 cm-1) absorbing bands. Moreover, the composites possess good microwave absorbing properties.

Keywords: Si/C/N, Nanocomposite powder,Radio frequency plasma,Absorbing properties

1. Introduction

Absorbing materials play an important role in military and civil application. Progress in the engineering of antennas and electromagnetic shielding systems depends crucially on the development of materials absorbing electromagnetic radiation in a broad range, from meter to millimeter waves[1][2]. The high absorbance of such materials must be combined with low reflectance in the same frequency range[3].

Various kinds of materials can be used for absorbing materials, including metal-alloys[4-6], ceramic, oxides, composites and transition metal sulfide absorbing materials. Nano-structured absorbing materials have received steadily growing interest because of their fascinating properties like wide absorbing frequency, low density and good compatibility, compared with the bulk or microsized counterparts. The increased surface area, number of dangling bond atoms and unsaturated co-ordination on surface leads to interface polarization, multiple scatter and absorbing more microwave[7] [8]. It is proposed that the nano-structured composite absorbing materials are the future research tendency of absorbing materials.

There are many methods for nano-structured materials preparation, among which thermal plasma is a novel technique. During the process, thermal plasma is normally generated by direct current (DC) arc or inductively coupled radio frequency (RF) discharge [9-11]. It can be described as a high enthalpy flame with extremely high temperature fields (1000–20000 ºC) and a wide range of velocity fields from several m/s to supersonic values. Since the resultant huge enthalpy can be realized for various kinds of plasma forming gases and easily controlled by electricity, thermal plasmas have been expected to facilitate not only fast chemical reactions but also rapid heat transfer in a variety of synthetic routes for nano-structured materials.

Silicon carbide matrix composite would be a good candidate for microwave absorbing materials. Especially, it has been reported that Si/C/N and Si/C/N/O nano composites have good absorbing properties in the wave range of centimeters to millimeters[12-14]. However, there are few reports on the preparation of nano-structured Si/C/N composite powders by RF technique. Therefore, this work is aimed at exploring the feasibility of produce nano-structured Si/C/N powders by RF plasma. Moreover, the microwave absorbing properties were investigated.

2. Experimental

The nano-structured Si/C/N composite powder was synthesized from methyltrichlorosilane (CH_3SiCl_3, 99.0% purity), nitrogen (N_2, 99.999% purity) and hydrogen (H_2, 99.999% purity) in a RF torch system. The reactant gas composition was optimized by adjusting the amounts of CH_3SiCl_3, H_2 and N_2. The temperature of the RF plasma reactor was detected by a pyrometer and the temperature was maintained at 3600–4800ºC.

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ISSN: 1791-2377

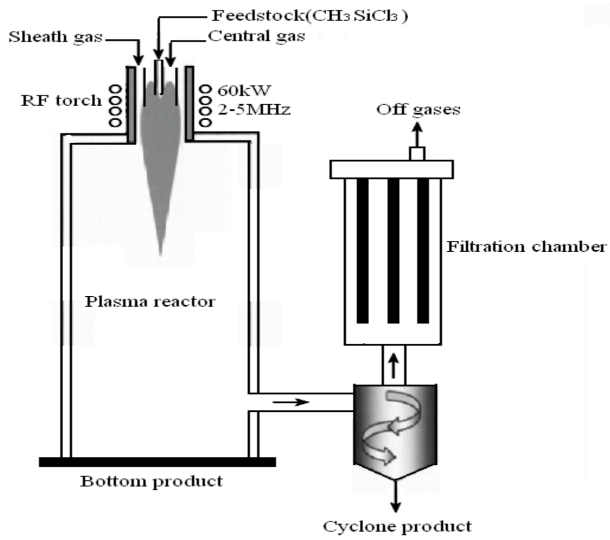

Fig.1 Schematic illustration of R.F. induction plasma

Table 1 Experimental parameters for R.F. induction plasma nano-structured Si/C/N composite powders

Central gas and flow rate	Argon-20slpm
Sheath gas and flow rate	85%Nitrogen+15%Hydrogen-100slpm
Tip of injection probe position	5 cm within coil region
Liquid feeding rate-CH_3SiCl_3	32.5g/min
Plate power	52kW
Chamber	8kPa

Scattering electron microscope (SEM) was taken to investigate the micro-morphology of the nano-structured Si/C/N composite powders prepared by RF plasma. The images were observed under an acceleration voltage of 5.0kV with a HITACHI S-4300 scanning electron microscope. The chemical composition of the Si/C/N composite powder was determined with a nitrogen analyzer (LECO-TN114) for the nitrogen content, with an oxygen analyzer (LECO-RO316) for the oxygen content, and with a carbon analyzer (LECO-CS334) for the carbon content. The silicon content was calculated as the remaining. Surface functional groups of the sample are investigated by Fourier transform infrared (FT-IR) spectroscopy system (Thermo Scientific Nicolet 6700, America). The microwave absorbing property of the nano-structured Si/C/N composites within the frequency range of 8.2–12.4 GHz was investigated utilizing Agilent 8722ES microwave vector network analyzer.

3. Results and discussions

The SEM micrograph of the Si/C/N composites powders is shown in Fig.2. From Fig.2, it can be seen that most of the Si/C/N particles produced by RF plasma are in the size of ~50nm. The size of these particles is relatively uniform, but there is some agglomeration. During the RF plasma process, the temperature is very high and the nano-structured particles have higher surface energy, which would result in the agglomerations.

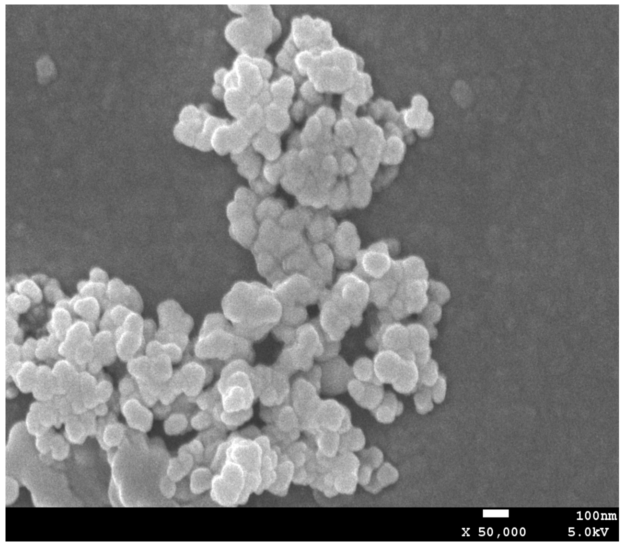

Fig.2 SEM micrograph of the nano-structured Si/C/N composite powders.

For absorbing materials, the composition plays the most important role on the absorbing properties. The chemical composition of the Si/C/N powders is shown in Table 2.

Table 2 The chemical composition of the Si/C/N powders

Element	wt.%
Si	58.98
Ct	14.82
Cf	6.55
N	19.91
O	0.16

According to the composition, the chemical constituent of the composites can be calculated. It is simply assumed that the Nitrogen element is in the form of Si3N4，the Carbon element is in the forms of SiC and free carbon (Cf) and the Oxygen element is in the forms of SiO2 and adsorbed oxygen (O2) on the powder surface, respectively. Therefore, the calculated chemical constituent of the composites is listed in Table 3.

Table 3 The chemical constituent of the Si/C/N composites

Constituent	wt.%
SiO_2	0.30
SiC	40.97
Si_3N_4	51.03
Cf	7.55
O_2	0.05

Factually, the Nitrogen element may be existed in a much more form. Some Nitrogen atom can dope in the lattice of SiC. In Si/C/N absorbing materials, some Carbon atoms in SiC are replaced by Nitrogen, resulting in negatively charged cavities. Under the electric field, the electromagnetic wave can be consumed by polarization relaxation of cavities[12]. For the reason, the strength of electromagnetic wave is weakened and the absorbance is enhanced.

Fig. 3 shows the FT-IR spectra of the Si/C/N powders. The corresponding functional groups have been marked in the figure. It indicates that the specimen have some functional groups, such as Si-O-Si, Si-N, Si-C, C-OH, Si-H, Si-CH_3, C-O, C-H. These assignments were made based on the assignments and references reported by Zhou et. al [15]. The major difference between the FT-IR spectrums recorded in this work and that of Zhou et. al is the presence of the strong broad absorption band at 967 cm-1 peak found here. This band may be assigned to both Si-N and Si-C combined. The Si-C (780 cm-1) and Si-N (950 cm-1) bands in bulk are generally reported to be broad.

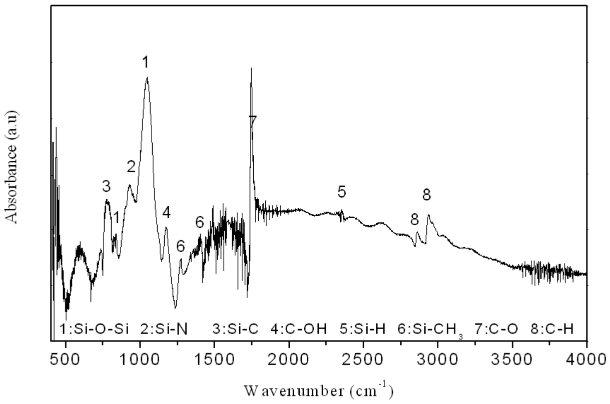

Fig. 3 FT-IR spectra of the nano-structured Si/C/N composites

It is thought that the absorbing properties of materials are mainly affected by the complex permittivity and permeability. The electromagnet wave energies (τ) absorbed per unit volume can be described as formula:

$$\tau = \frac{1}{2} \times \frac{1}{4\pi} \times \left(\varepsilon_0 \varepsilon'' |E| + \mu_0 \mu'' |H|^2 \right)$$

Where ε_0 is the vacuum permittivity, ε'' is the imaginary part of complex permittivity, E is the electric field vector of electromagnetic wave, μ_0 is the vacuum permeability, μ'' is the imaginary part of complex permeability, H is the magnetic field vector of electromagnetic wave. From formula (1), it can be seen that the absorbing properties are mainly depend on the imaginary parts of complex permittivity (ε'') and permeability (μ''). To enhance the imaginary parts of complex permittivity (ε'') and permeability (μ'') is an effective way to improve the absorbing properties.

The electromagnetic parameters of the nano-structured Si/C/N composites are shown in Table 4. From Table 4, it is obvious that the imaginary parts of complex permittivity (ε'') and permeability (μ'') of the Si/C/N composites increase obviously with the dielectric loss increasing gradually over the frequencies ranging from 8.2 GHz to 12.4 GHz. The increase of dielectric loss is due to the dipolar polarization. In addition, it can be seen that the complex permittivity (ε'') of composites is as high as 5.78–4.26 in the tested frequency range.

Table 4 The electromagnetic parameters of the Si/C/N composites

Frequency/GHz	ε’’	μ’’
8.2	5.78	0.01
8.6	5.54	0.01
9.0	5.32	0.01
9.4	5.15	0.01
9.8	5.02	0.01
10.2	4.89	0.01
10.6	4.75	0.01
11.0	4.62	0.01
11.4	4.50	0.01
11.8	4.39	0.02
12.4	4.26	0.02

Based on the electromagnetic parameters of the Si/C/N composites, the relationship between reflection coefficient (X-band) and the thickness of the absorber can be simulated (see in Fig.4) by software named FLUENT 6.3. From Fig.4, it can be seen that with the increasing thickness of the absorber, the reflection coefficient is reduced obviously. Moreover, with the increasing frequency, the reflection coefficient is also reduced at the same thickness.

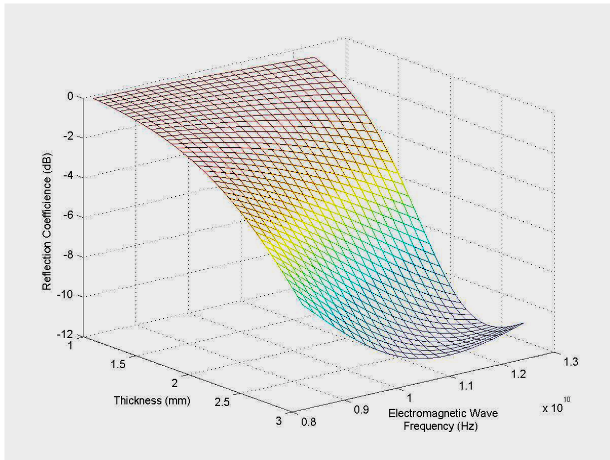

Fig.4 The relationship between reflection coefficient (X-band) and the thickness of the absorber

When the particle size of materials distributes in the nano level, the electronic energy level of nanoparticles was split by quantum effects, and the split level spacing is in the energy range corresponding to the microwave (10-2–10-5 eV), which would cause new microwave absorbing effects. Meanwhile, due to nanoparticles have small size, large surface area, a high proportion of surface atoms and increased dangling bonds, the sectional polarization and multiple scattering becomes important mechanism for microwave absorbing. When the size of the nanoparticles is equal to or smaller than the wavelength, de Broglie wavelength, the coherence length of the superconducting state or the penetration depth of magnetic field, the crystal periodic boundary conditions will be destroyed and the atom density near the particle surface layer of the amorphous nanoparticles is reduced, resulting in material characteristics like sound, light, electricity, magnetism, thermodynamics become abnormal, such as the increase in light absorption, magnetic enhancement, etc[8][12]., which makes the development of multi-band electromagnetic wave absorbing materials becomes feasible.

4. Conclusion

In summary, nano-structured Si/C/N composite powders were successfully produced by radio frequency (RF) induction plasma utilizing methyltrichlorosilane (CH_3SiCl_3) and nitrogen (N_2) as precursors. The micro-morphology, chemical composition, functional group structure and absorbing properties of the samples were investigated. The results indicate that the size of Si/C/N particles is ~50 nm with homogeneous distribution. The Si/C/N composites mainly consist of SiC (40.97%) and Si_3N_4 (51.03%) and the composites have stronger Si-C (780 cm-1) and Si-N (950 cm-1) absorbing bands. Moreover, the composites possess good microwave absorbing properties.

Acknowledgement

This work was Supported by the National Natural Science Foundation of China(No.51205330)

References

1. Petrov,V.-M., Gagulin,V.-V., "Microwave absorbing materials", Inorganic Materials37(2), 2001,pp.93–98.
2. Zhang,H._-T.,Zhang,J.-S.,Zhang,H.-Y.,"Numerical predictions for radar absorbing silicon carbide foams using a finite integration technique with a perfect boundary approximation",Smart Materials and Structures15(3), 2006,pp.759-766.
3. N.Y. Mendoza, B.M. Goortani and P. Proulx. "Numerical simulation of silica nanoparticles production in an RF plasma reactor: effect of quench", Journal of Materials Science and Engineering C, 2007, 27(5-8):1265-1269.
4. Mouchon,E., Colomban,P.,"Microwave absorbent: preparation, mechanical properties and RF-microwave conductivity of SiC (and/or mullite) fibre reinforced Nasicon matrix composites ", Materials Science, 31(2) 1996,pp.323−334.
5. Kagawa ,Y., Matsumura,K., Iba,H., et al. "Potential of short Si-Ti-C-O fiber-reinforced epoxy matrix composite as electromagneticwave absorbing material",Materials Science42(4), 2007,pp.1116−1121.
6. Liu,H.-T.,, Cheng,H.-F., Wang, J.,et al. "Dielectric properties of the SiC fiber-reinforced SiC matrix composites with the CVD SiC interphases",Journal of Alloys and Compounds491(1-2),2010,pp.248-251.
7. Sharma, R., Agarwala,R.-C. and Agarwala, V., "Development of radar absorbing nano crystals by microwave irradiation",Materials Letters62(15). 2008,pp.2233-2236
8. Chatrchyan,S., Khachatryan,V., Sirunyan, A. -M. et al. "Observation of a new boson at a mass of 125 GeV with the CMS experiment at the LHC",Physics Letters B716(1), 2012,pp.30-60.
9. Ding,D.-H.,Zhou.W.-CH.,Zhang,B., et al. " Complex permittivity and microwave absorbing properties of SiC fiber woven fabrics",Journal of Materials Science46(8), 2010,pp.2709-2714.
10. Zhao,D.-L.,Luo,F and Zhou,W.-CH.,"Microwave absorbing property and complex permittivity of nano SiC particles doped with nitrogen",Journal of Alloys and Compounds490(1-2), 2010,pp.190-194
11. Chiu,SH.-CH.,Yuh.-CH,.,Li,Y.-Y.,"High electromagnetic wave absorption performance of silicon carbide nanowires in the gigahertz range", The Journal of Physical Chemistry C114(4), 2010,pp.1947-1952
12. Zhao,D. –L.,Zhao,H.-SH.,Zhou,W.-CH.,"Dielectric properties of nano Si/C/N composite powder and nano SiC powder at high frequencies ",Physica E9(4) ,2001,pp.679-685.
13. Eick,B.-M.,Youngblood.J.-P.,"SiC nanofibers by pyrolysis of electrospun preceramic polymers",Journal of Materials Science44(1), 2009,pp. 160-165
14. Courric, S., Tran,V. -H., " The electromagnetic properties of blends of poly(p-phenylene-vinylene) derivatives",Polymers for Advanced Technologies11(6),2000,pp.273-279.
15. Zhou,Y.-P., Probst, D., Thissen,A., et al. "Hard silicon carbonitride films obtained by RF-plasma-enhanced chemical vapour deposition using the single-source precursor bis(trimethylsilyl) carbodiimide",Journal of the European Ceramic Society26(8), 2006,PP.1325-1335.