Microwave Absorption Performance of Graphene Nanoplatelets Dispersed SiC

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ABSTRACT

Microwave absorption performance of graphene nanoplatelets based SiC composites have been studied in the present work. Graphene nanoplatelets were dispersed with varying weight fractions viz. 0.5, 1, 2, 2.5 and 3 wt.% in SiC using ultrasonication and ball milling methods. Microwave attenuation behaviour of the prepared composites have been observed in the 2-18 GHz range. The results reflect that dispersion of graphene in SiC proves to be very instrumental in attenuating the incident microwave signals translating into enhanced absorption. The enhancement in the microwave absorption performance is due to the presence of conductive graphene nanoplatelets which is very instrumental in promoting conduction and polarisation losses in the composite translating into enhanced absorption. For 3 wt.% graphene nanoplatelets dispersion in SiC, the value of reflection loss (RL) was obtained as -38.67 dB at 14.67 GHz frequency for 2.2 mm thickness with the corresponding bandwidth of 5 GHz.

Keywords: Microwave absorption; Silicon carbide; Graphene nanoplatelets

1. INTRODUCTION

In this electronic era, microwave absorbing materials have played a very vital role in the areas of electronic communication, health care, and stealth technology[1-3]. Therefore, the development of efficient and cost-effective microwave absorbing materials has grabbed the global attention. This has prompted many researchers to develop microwave absorbers exhibiting enhanced features viz. high reflection loss, wide bandwidth and thin thickness. However, a single material cannot exhibit all these desirable attributes. Among the various microwave absorbers, carbon, graphene, carbon nanotubes (CNTs), ceramics and ferrites materials have been studied and reported to exhibit enhanced microwave absorption properties[4]. CNTs and graphene have unique and excellent properties such as structural stability, thermal, mechanical and electrical properties.

Dielectric properties of the materials or composites dictates the attenuation behaviour in the microwave spectrum. Materials with high dielectric properties exhibits enhanced microwave absorption. Therefore, silicon carbide (SiC) exhibiting remarkable dielectric properties can be combined with other carbonaceous materials to achieve enhanced microwave absorption[5]. Complex permittivity of SiC could be modified with the addition of CNTs and graphene in order to achieve enhanced absorption. Due to the remarkable features of graphene, researchers have utilised its excellent properties in developing efficient and high performance microwave absorber such as graphene dispersed ceramics, ferrites, polymers, and metals[6]. Yang[7], *et al.* synthesised graphene based ferrite composite and achieved a minimum RL of -36.8 dB at 9.9 GHz with 2.9 mm thickness. Wang[8], *et al.* synthesised graphene based flower-like cobalt ferrite composite and achieved a minimum RL of -42 dB at 12.9 GHz with 2 mm absorber thickness. Chen[9], *et al.* studied the microwave attenuation behaviour of graphene and silicone rubber composite and achieved a minimum RL of -32.1 dB at 13.2 GHz with 2 mm absorber thickness. However, one of the major challenges is the agglomeration of graphene particles owing to larger surface area. In this study, the agglomeration tendency of graphene has been tried to resolve using ultrasonication.

In the present work, an effort to tailor and tune the dielectric properties of SiC with varying concentration of graphene nanoplatelets has been made. Here, SiC with excellent dielectric properties is combined with conductive graphene nanoplatelets in order to develop high performance and low-cost microwave absorber.

2. EXPERIMENTAL METHODOLOGY

Graphene nanoplatelets was ultrasonically cleaned and de-agglomerated in acetone medium for 1 hour. These ultrasonically treated graphene nanoplatelets were then dispersed in SiC using a planetary ball mill at 150 rpm for 1 hour. Graphene nanoplatelets were dispersed at varying weight fractions viz. 0.5, 1, 2, 2.5 & 3 wt.% in SiC. Ball to power ratio was taken as 10:1. The sample codes for graphene nanoplatelets dispersed SiC samples with different weight fractions are as given in Table 1.

X-ray diffractometer was used to perform phase analysis. For all the synthesised samples, the intensities of the diffracted peaks corresponding to its diffraction angle (2θ) was recorded. Agilent N5222 PNA series Vector Network Analyser was used to carry out the dielectric measurements of the samples in 2-18

Table 1. Sample codes for Graphene nanoplatelets dispersed Silicon Carbide

Composition	Sample code
Pure SiC	SG0
SiC+0.5 wt.% Graphene nanoplatelets	SG1
SiC+1 wt.% Graphene nanoplatelets	SG2
SiC+2 wt.% Graphene nanoplatelets	SG3
SiC+2.5 wt.% Graphene nanoplatelets	SG4
SiC+3 wt.% Graphene nanoplatelets	SG5

GHz. Reflection loss for the samples was calculated using Eqns. (1 and 2)[10-12].

$$RL \text{ (dB)} = 20 \log |Z_{in} - Z_o / Z_{in} + Z_o| \quad (1)$$

$$Z_{in} = Z_o \sqrt{(\mu_r / \varepsilon_r)} \tanh[j.(2\pi f t_{cal}/c).\sqrt{(\varepsilon_r.\mu_r)}] \quad (2)$$

where, ε_r and μ_r represents the complex permittivity and permeability values respectively, Z_{in} and Z_o represents the characteristic impedance of the absorber and free space respectively, f is the working frequency, c is the speed of the light, and t_{cal} is the absorber thickness. As seen from Eqns (1-2), the value of RL is a function of complex permittivity, permeability, frequency and thickness of the absorbing material.

The dielectric and conduction losses plays a very significant role in microwave attenuation of the incident microwaves. Apart from these major loss mechanisms, the absorber's geometrical aspect viz. thickness also plays an important role on the attenuation of the incident microwave signals. Destructive interference of microwaves due to the geometrical aspect results in the phase cancellation, thus attenuating the reflected microwave signals. The role of geometrical aspect is more pronounced when the thickness of absorber satisfies Eqn (3):

$$t_{sim.} = [nc/4.f_m \sqrt{(|\varepsilon_r \mu_r|)}] \quad (3)$$

where, f_m is the matching frequency corresponding to min. RL, simulated thickness of the sample is represented by t_{sim}, complex permittivity and permeability are represented by ε_r and μ_r respectively at peak frequency, c is the speed of the light, and $n = 1,3,5$, etc.

The imaginary part of complex permittivity according to free electron theory[13-16], is given as:

$$\varepsilon'' = [1 / (2\pi\varepsilon_o \rho f)] \quad (4)$$

where, ε_o and ρ is the free space permittivity and electrical resistivity, respectively. Imaginary part of permittivity is influenced by the electrical resistivity of the composite which is evident from Eqn. (4). As we all know that graphene has good electrical conductivity, therefore, dispersion of graphene nanoplatelets in SiC will affect the imaginary part of permittivity of the composite system, thereby affecting the min. RL characteristics.

3. RESULTS AND DISCUSSION

In this section XRD analysis, dielectric properties and microwave absorption behaviour of the graphene nanoplatelets dispersed SiC composite system has been discussed.

3.1 XRD Analysis

The X-ray diffraction patterns as shown in Fig. 1 depicts the peaks of SiC (Moissanite 6H; ICSD Collection Code:27051) with 2θ values as 35.58, 35.65, 38.13, 59.91, 60.00, 71.74 and 71.79º belonging to (011), (006), (012), (013), (110), (116) and (022) planes respectively. With the dispersion of graphene nanoplatelets, a diffraction peak corresponding to carbon emerges and becomes distinctly evident after 2 wt.% graphene nanoplatelets dispersion. Absence of any other phase indicates that the composite system under study contains only SiC and graphene nanoplatelets.

3.2 Dielectric Properties

The electric and magnetic energy storage capability of a material are represented by the real (ε', μ') parts of complex permittivity and permeability. Subsequently, the losses in electric and magnetic energy are represented by corresponding imaginary (ε'', μ'') parts of complex permittivity and permeability. Figure 2 depicts the nature of real and imaginary part of complex permittivity and permeability curves of dispersed graphene nanoplatelets in SiC with respect to frequency. It is observed from Fig. 2 that with the dispersion of graphene nanoplatelets in SiC, complex permittivity and permeability changes. It is noticed from Fig. 2(a) that the value of ε' varies with the increase in frequency but for the sample SG5 it decreases gradually. The ε'' values shows a gradually increasing trend after 7.5 GHz with respect to frequency for all the samples (except for SG3 which shows a gradually decreasing trend) as shown in Fig. 2(b). The synergistic effect of dielectric SiC particles, conductive graphene nanoplatelets, interfacial polarisation due to the multiple interfaces involved may be the major reasons behind this kind of behaviour. The values of μ' remains almost constant with increase in frequency and displays an increasing trend with the increase in graphene nanoplatelets concentration as indicated in Fig. 2(c) but for sample SG5 it decreases gradually after displaying an almost constant value up to 9 GHz. However, the value of μ'' displays a mixed behaviour with varying graphene nanoplatelets concentration as shown in Fig. 2(d).
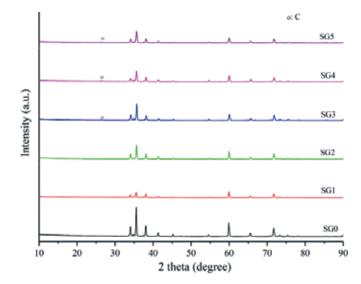

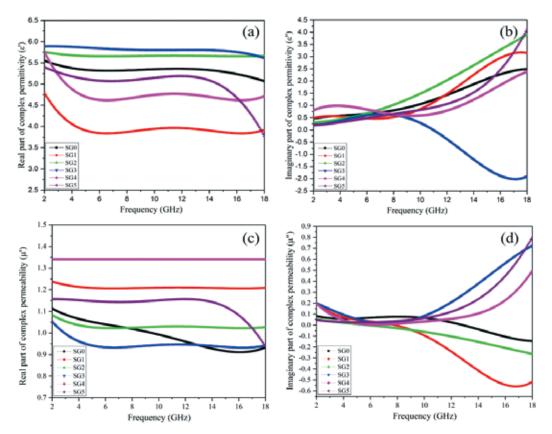**Figure 1. XRD pattern of graphene nanoplatelets dispersed SiC composites.**

Figure 2. (a) Real part of complex permittivity, (b) imaginary part of complex permittivity, (c) real part of complex permeability, and (d) imaginary part of complex permeability with increasing frequency for graphene nanoplatelets dispersed SiC composites.

4. MICROWAVE ABSORPTION BEHAVIOUR

Figure 3 shows the effect of graphene nanoplatelets dispersion on reflection loss with respect to frequency. The values of RL were computed for different thicknesses, however, the data corresponding to minimum reflection loss has been shown.

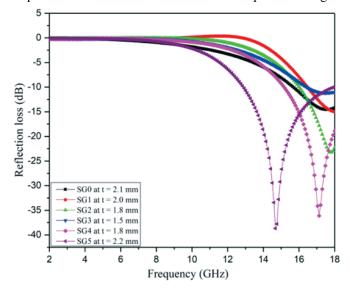It is observed from the results that graphene nanoplatelets dispersion enhance the microwave absorption at higher frequencies owing to the synergistic effect of many mechanisms viz. conduction loss, interfacial polarisation loss, multiple scatterings and reflections. The maximum absorption was observed in sample SG5 at 2.2 mm thickness with minimum reflection loss (RL) value –38.67 dB at 14.67 GHz frequency with 5 GHz bandwidth. The values of minimum RL along with matching thickness and -10 dB bandwidth for all the samples are also listed in Table 2.

It is observed that with the dispersion of very small quantity of graphene nanoplatelets, the microwave attenuation behaviour of the composite is significantly enhanced. A comparative table comparing the microwave absorption performance of the material under study with some of the recently developed graphene based composites has been shown in Table 3. Compared with the reported graphene based composites, graphene nanoplatelets dispersed SiC composites exhibits enhanced absorption.

Figure 3. Reflection loss characteristics of graphene nanoplatelets dispersed silicon carbide composites.

Table 2. Reflection loss of Graphene nanoplatelets dispersed Silicon carbide

Material code	Min. RL (dB)	Frequency (GHz)	Thickness (mm)	-10 dB bandwidth (GHz)
SG0	-14.5	17.5	2.1	2.28
SG1	-15.0	18.0	2.0	1.5
SG2	-23.2	17.7	1.8	2.0
SG3	-11.2	17.47	1.5	1.6
SG4	-36.13	17.12	1.8	2.8
SG5	-38.67	14.67	2.2	5.0

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Table 3. Microwave absorption performance comparative table for Graphene based absorbers

Graphene based materials	Min. RL (dB)	Thickness (mm)	-10 dB bandwidth (GHz)	Ref.
Graphene/PANI	-36.9	3.5	5.3	[17]
Graphene/Fe	-45.0	3.0	4.4	[18]
Graphene/Ni//C	-34.2	1.6	3.2	[19]
3D-graphene/Fe_3O_4	-27	2.0	5.8	[20]
Graphene/SiC	-38.67	2.2	5.0	Present wok

5. CONCLUSIONS

Graphene nanoplatelets were successfully dispersed in SiC using ball milling technique. Phase of SiC remain constant after dispersion of graphene nanoplatelets. The 3 wt.% graphene nanoplatelets dispersion gives maximum microwave absorption of -38.67 dB at 14.67 GHz frequency which is a relatively low loading fraction. A 5 GHz bandwidth was achieved corresponding to -10 dB. This result indicates that the dispersion of graphene nanoplatelets in SiC can prove to be a good candidate for developing high performance and low-cost microwave absorbers economically.

REFERENCES

1. Chen, C.Y.; Pu, N.W.; Liu, Y.M. & Eason, G. Remarkable microwave absorption performance of graphene at a very low loading ratio. *Composite part B: Engineering.* 2017, **114**, 395-403.
2. Zhu, H.L.; & Fan, R.H. Microwave absorption properties of MWCNT-SiC composites synthesized via a low temperature induced reaction. *AIP advances*, 2011, **1**, 32-40.
3. Singh, S.; Shukla, S., Kumar, A. & Singh, D. Influence of Zn dispersion in SiC on electromagnetic wave absorption characteristics. *J. Alloys Compd.,* 2018, **738**, 448-460.
4. Bi, S.; Ma, L.; Mei, B.; Tian, Q.; Liu, C.; Zhong, C. & Xiao, Y. Silicon carbide/carbon nanotube heterostructures: Controllable synthesis, dielectric properties and microwave absorption. *Adv. Power Techn.*, 2014, **25**(4), 1273-1279.
5. Xie, S.; Jin, G.Q.; Meng, S.; Wang, Y.; Qin, Y.; & Guo,X. Microwave absorption properties of in situ grown CNTs/SiC composites. *J. Alloys Compd.*, 2012 **520**, 295-300.
6. Zang, Y.; Xia, S.; Ren, L. Li; Chen, G.Q.; Quan, H.; & Wu, Q. Microwave absorption enhancement of rectangular activated carbon fibers screen composites. *Composites Part B*, 2015 **77**, 371-378.
7. Yang, H.; Ye, T.; Lin, Y.; Zhu, J.; & Wang, F.; Microwave absorbing properties of the ferrite composites based on graphene. *J. Alloys Compd.,* 2016 **683**, 567-574.
8. Wang, S.; Zhao, Y.; Xue, H.; Xie, J.; Feng, C.; Li, H.; Shi, D.; Muhammad, S. & Jiao, Q. Preparation of flower-like $CoFe_2O_4$@graphene composites and their microwave absorbing properties. *Materials Letters*, 2018, **223**, 186-189.
9. Chen, C.Y.; N. Pu,N.W.; Liu, Y.M.; Wu, C.H.; Cheng, T.Y.; Lin, M.H.; Ger, M.D.; Gong, Y.J.; Peng, Y.Y.; Crubb, P.M. & Chen, R.T. Microwave absorption properties of holey graphene/silicone rubber composites. *Composites Part B*, 2018, **135**, 119-128.
10. Kumar, A.; Agarwala, V. & Singh, D. Microwave absorbing behavior of metal dispersed TiO_2 nanocomposites. *Advanced Powder Technology*, 2014, **25**(2), 483-489.
11. Kumar A.; Agarwala V. & Singh D. Effect of Particle Size of $BaFe_{12}O_{19}$ on the Microwave Absorption Characteristics in X-Band. *Progress in Electromagnetics Research M,* 2013, **29**, 223-236.
12. Kumar A.; Agarwala V. & Singh D. Effect of Mg substitution on microwave absorption of $BaFe_{12}O_{19}$. *Advanced Materials Research*, 2012, **585**, 62-66.
13. Ni, X.; Ma, J.; Li, J.; Jiao, D.; Huang, J. & Zhang, X. Microwave characteristics of Co/TiO_2 nanocomposites prepared by mechanochemical synthesis. *Journal of Alloys Compounds*, 2009, **468**, 386-391.
14. Kumar, A.; Singh, S. & Singh, D. Effect of heat treatment on morphology and microwave absorption behavior of milled SiC. *Journal of alloy Compounds,* 2019, **772**, 1017-1023.
15. Kumar A.; Agarwala V. & Singh D. Effect of milling on dielectric and microwave absorption properties of SiC based composites. *Ceramic International,* 2014, **40**(1), 1797-1806.
16. Singh, S.; Sinha, A.; Zunke, R.H.; Kumar, A. & Singh, D. Double layer microwave absorber based on Cu dispersed SiC composites. *Adv. Powder Technol.,* 2018, **29**(9), 2019-2026.
17. Chen, X.N.; Meng, F.C.; Zhou, Z.W.; Tian, X.; Shan L.M. & Zhu S.B. One-step synthesis of graphene/polyaniline hybrids by in situ intercalation polymerization and their electromagnetic properties. *Nanoscale*, 2014, **6**, 8140–8148.
18. Park, K.Y.; Han, J.H.; Lee, S.B. & Yi, J.W. Microwave absorbing hybrid composites containing Ni–Fe coated carbon nanofibers prepared by electroless plating. *Composites Part A-Applied Science and Manufacturing*, 2011, **42**, 573–578.
19. Li, C.; Huang, Y. & Chen, J. Dopamine-assisted one-pot synthesis of graphene@Ni@C composites and their enhanced microwave absorption performance. *Materials Letter*, 2015, **154**,136–139.
20. Hu, C.; Mou, Z.; Lu, G.; Chen, N.; Dong, Z.; Hu, M. & Qu, L. 3D graphene-Fe3O4 nanocomposites with high-performance microwave absorption. *Phys. Chem. Chem. Phys.,* 2013, **15**, 13038–13043.

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He has carried out the experimental work in this work.

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He has carried out some of the initial studies and helped in the experimental part of this work.

Mr Samarjit Singh received his BTech in Mechanical Engineering from Tezpur University and MTech in Materials Science and Engineering from MNNIT Allahabad. He is currently pursuing his PhD in the department of Applied Mechanics, MNNIT Allahabad. He is currently working in the area of Microwave absorbing materials for enhanced microwave absorption.
He has carried out the literature survey and helped in the experiments of the present work.

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The current study has been deemed possible owing to his valuable comments and suggestions.