Carbon 118 (2017) 86–97

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

A universal permittivity-attenuation evaluation diagram for accelerating design of dielectric-based microwave absorption materials: A case of graphene-based composites

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ARTICLE INFO

Article history:
Received 25 January 2017
Received in revised form
4 March 2017
Accepted 5 March 2017
Available online 6 March 2017

Keywords:
Graphene
Polymer composites
Complex permittivity
Microwave absorption

ABSTRACT

Since Materials Genome Initiative has been proposed for demonstrating promising approaches in achieving high-performance materials, here we employ such concept to present a Materials Genome Initiative inspired universal permittivity-attenuation evaluation diagram (PAED) for accelerating design of highly efficient dielectric-based microwave absorption composites. Initially, parameter sweeping has been applied to screening the best microwave absorption parameters based on the absorption-permittivity mode, aiming to creating a novel PAED. For well demonstrating the validity of such diagram, experiments are applied to scalable fabricate various graphene-based composites with a simple approach, achieving strong microwave absorption (−57 dB with full-band qualified absorption in X-band) and considerably high radar cross scattering reduction (24.25 $\mathrm{dBm^2}$ at 90°) with the guide of the proposed diagram. Noticeably, implication of the verification results from experiments, previous literature and simulation indicates validity of such novel PAED, and thus it highlights a new general roadmap for rationally designing high-performance microwave absorption.

1. Introduction

To help businesses discover, develop, and deploy new materials twice as fast, the Materials Genome Initiative was proposed at the White House in 2011, with objectives of integrating experiments, computation and theory for facilitating access to materials data and equipping next-generation materials workforce. With assistant of calculation and experiments, Materials Genome Initiative concept was expanded to incorporating the important parameters of nanomaterials into the roadmap.

Lightweight composites of high-performance microwave absorption properties promise great applications with examples ranging from cutting-edge electronics to specific devices in aerospace and communication industries [1–5]. Among the pursued carbon nanotube- (CNT) and graphene-based materials, their composites, particularly polymeric ones, have attracted extensive development efforts for their appropriate dielectric and magnetic properties [6–12]. For microwave absorption in stealth technology, the absorption coatings demand more effective absorption towards incident electromagnetic waves, which has been identified as a major bottleneck in terms of performance and reliability improvements. In principle, the general approach for fabricating microwave-absorption composites involves the preparation of dried powders and further dispersion into the matrices, with aim to creating moderate three-dimensional (3D) absorbing networks in the insulating matrices [9–11,13,14]. On the basis of the concept, incorporation of carbon materials of electric loss along with magnetic loss materials has been largely considered to prepare effective carbon/magnetic hybrid fillers in the past decades [15–22]. For recent work as examples, Wen et al. have fabricated multi-walled

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http://dx.doi.org/10.1016/j.carbon.2017.03.016
0008-6223/

CNT/Fe (MWCNT/Fe), MWCNT/Co and MWCNT/Ni hybrid structures into epoxy composites (60 wt% filler loading), showing enhanced absorption peaks greater than 30 dB [18]. More recently, Wang and coworkers reported that the wax composites of MWCNT/Fe_3O_4@ZnO heterotrimers (50 wt% filler loading) possessed effective microwave absorption in the whole X-band (8.2–12.4 GHz) [19]. Likewise, Sun et al. studied the Fe_3O_4/reduced graphene oxide (RGO) hybrids by fabricating wax-based composites, which exhibited a reflection loss up to 27 dB at 40 wt% filler loading [15]. Chen et al. prepared Ni/RGO hybrids (60 wt% filler loading), which presented a highest absorption performance up to 17 dB in the investigated range [16].

Interestingly, recent studies have shown that utilization of bare carbon fillers (without hybridization with a second phase or magnetic material) appears unique advantages for achieving low filler-loading composites of high-performance microwave absorption if a certain appropriate approach is applied in the processing [4,9,12,23,24]. For instance, Bai et al. has utilized a viscous polyethylene oxide (PEO) solution to serve as the polymeric matrix and dispersing agent to prevent RGO from aggregation upon in situ reduction [12]. The resulting homogeneous RGO/PEO composites with only 5 wt% RGO loading presented highly effective microwave absorption up to 40 dB [12], indicating that the bare RGO nanosheets with a homogeneously dispersed fashion at much lower filler loadings could provide competitive absorption performance in comparison with other graphene/magnetic hybrids [15–22]. Very recently, the preparation of 3D nanoscale graphene-based aerogels have considerably lowered the filler loading for acquiring qualified microwave absorption performance, owing to the generation of a more fashion for creating effective electromagnetic energy conversion channels in the composites [4,7,24]. Thus, these results imply that bare carbon should be more ideal as the efficient fillers for large-scale fabricating lightweight microwave absorption composites, specifically for those applied at higher frequency range due to the much suppressed magnetic response ability.

Although significant experimental efforts have been made in the past decades, very few attempts have been concerned on understanding how the complex permittivity impacts the microwave absorption. Generally speaking, the plots for microwave absorption performance, the term of reflection loss (RL), are achieved from the complex permittivity via the relations [25,26].

$$Z_{in} = \sqrt{\frac{\mu_r}{\varepsilon_r}}\tanh\left[j\frac{2\pi}{c}\sqrt{\mu_r\varepsilon_r}fd\right], \tag{1}$$

$$R_L = 20\log\frac{|Z_{in}-1|}{|Z_{in}+1|}, \tag{2}$$

where Z_{in} is the normalized input impedance, c the light velocity, f the frequency, d the thickness of the absorber, ε_r and μ_r the complex permittivity and permeability of the samples, respectively. In practice, the calculated RL results enable to reveal the absorption performance with regard of frequency and thickness. However, no obvious correlations could be observed from the complex permittivity to microwave absorption performance, and meanwhile the effects of complex permittivity on the microwave absorption performance could not be confirmed according to the RL plots. Apparently, these uncertainties required more experimental efforts to optimize the electromagnetic parameters for acquiring the qualified performance, which would impact the further development of high-performance microwave absorption and thus a rational way for achieving the ideal performance is highly needed.

As illustrated in Fig. 1, here we employed the Materials Genome Initiative concept for designing high-performance microwave absorption based on the parameter sweeping, with purpose of establishing Materials Genome Initiative inspired permittivity-attenuation evaluation diagram (PAED) via screening the best microwave absorption performance. Meanwhile, experimental fabrication has also been utilized for scalable producing various polymeric composites with different features. Subsequently, complex permittivity from the as-fabricated composites and typical previous report has been screened on the PAED to prove the validity. Further verification from CST simulation has been also applied. The combined verification results exhibit that microwave absorption properties are in good agreement with the prediction from the PAED, suggesting a powerful strategy for tuning microwave absorption performance from dielectric absorbing materials in practical application.

2. Experimental section

2.1. Parameter sweeping using absorption-permittivity model

Parameter sweeping of microwave absorption was obtained by using absorption-permittivity model. According to the Equation, $RL = 20\log(|Z_{in}-1|/|Z_{in}+1|)$, the absorption-permittivity model was described as $Abs\,(RL) = F(\varepsilon', \varepsilon'', d, f)$, where the terms of ε' and d were set as the given constant ($\varepsilon' = 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25$ and 30; $d = 2$, 3, 4 and 5 mm), and thus such Equation could be simplified as $Abs\,(RL) = F'(\varepsilon'', f)$, ($0 < \varepsilon'' < \varepsilon'$; $8.2\text{ GHz} < f < 12.4\text{ GHz}$).

2.2. Establishment of PAED

The PAED was established based on the parameter sweeping results, followed by screening the best microwave absorption points in each case. Subsequently, the real and imaginary permittivity of the selected points were recorded for data output. In the re-built real permittivity-imaginary permittivity plots, the values of the selected points were highlights for establishing the PAED at different material thickness.

2.3. Material fabrication

The polymeric composites, including soft and tough features coupled with RGO-based dielectric fashion, were fabricated for verifying the PAED. Initially, GO aqueous solution was prepared based on the modified Hummers method.[30] In brief, graphite (2 g) and $NaNO_3$ powders (1 g) were dispersed into the concentrated H_2SO_4 (120 Ml) in an ice bath, allowing the mixture to be kept at 0 °C for 1 h. Subsequently, $KMnO_4$ (6 g) powders were added slowly. (Caution: fast mixing may result in explosion). Upon vigorously stirring for 2 h, the mixture was heated and kept at 30 °C for 0.5 h. Then, amount of water (150 mL) was added dropwise, followed by adding H_2O_2 solution (5%, 50 mL). The resulting solution was washed with water and HCl (5%) to obtain the GO aqueous solution.

In the typical process of soft PPN-RGO composites, a piece of commercial polypropylene nonwoven (PPN) without any treatment was immersed in the in the mixture solution of GO and hydroquinone (GO: hydroquinone = 1:5 wt/wt) under sonication for 2 h. The amount of GO used was determined by the volume of the PPN. After stable mixture solution was achieved, the extra mixture solution above the PPN was removed. Subsequently, the mixture was then sealed and heated up to 100 °C for 12 h, allowing GO for in situ self-assembling into RGO interconnections. Until the completion of heating, the resulting sample was unsealed and then washed with water, followed by freeze-drying.

For fabricating tough PPN-RGO-PF composites, the as-prepared