Strong and thermostable polymeric graphene/silica textile for lightweight practical microwave absorption composites

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18

Graphical Abstract

19

20 Novel lightweight polymeric graphene/silica textile composites present excellent overall
 21 performance including highly mechanical robustness, sufficient thermal stability and broadband
 22 absorption, allowing them to serve as the scalable materials for practical microwave-absorption
 23 applications.

24

Abstract

Since various criteria including effective absorption intensity, broad absorption bandwidth, sufficient mechanical properties and excellent stability are required to meet the benchmark in practical service, here we propose a novel strategy to fabricate strong and thermostable polymeric graphene/silica textile composites for practical microwave absorption applications. For achieving homogeneous configuration, a unique silica textile coupled with freeze-drying method is employed as the critical factor in the formation of the polymeric composites, allowing reduced graphene oxide (RGO) to in situ form three-dimensional conductive frameworks. With the co-existence of silica textiles and thermoset polymers, such integrated bi-matrices offer the composites with substantial reinforcement in the mechanical properties and thermal stability. As expected, the as-fabricated lightweight composites ($\sim 1 \text{ g/cm}^3$) present strong tensile strength of 40 MPa and high thermal stability beyond 225 °C (95% mass retention in air), coupled with a microwave absorption peak at -36 dB and effective absorption in the full X-band at a low filler loading (4.1 wt% RGO). Implication of the results suggests that the excellent overall performance allows such novel polymeric composites for practical microwave absorption applications.

Key words: graphene framework, silica textile, polymeric composite, thermostable, strong, microwave absorption

Introduction

The materials of electromagnetic absorption and attenuation have attracted increasing attention owing to their significant roles in absorbing and diminishing undesirable electromagnetic irradiations from the electronic devices and communication apparatus.^[1-7] For the practical applications such as high-speed aircrafts and shuttles in the aerospace industry, various critical factors are highly concerned in the structural components, including broadband absorption capacity for avoiding signal detection, strong mechanical properties for resisting deformation under stress, sufficient thermal stability for maintaining stable operation under high-speed transport and etc.^[4-6,8-12] As a consequence, combination of both functional and structural criteria requires the microwave absorption layers to hold outstanding overall performance, allowing them to reach the benchmark that is capable for practical circumstances.

For the pursuit of broadband absorption, recent advances suggest that carbon-based composites of various morphologies have presented outstanding capability in the microwave absorption, with many advantages including light weight, low cost, easy processability, excellent corrosion resistance, and etc.^[7,13, 14] Due to the unique two-dimensional (2D) nanostructures and tunable electrical conductivity, graphene nanosheets of large aspect ratios possess exclusive features in establishing three-dimensional (3D) conductive networks for generating leaking current, enabling to deliver effective electromagnetic energy conversion and attenuation.^[6,15] In the past years, explorative efforts have been paid to the development of graphene-based heterofillers, aiming to offering tunable complex permittivity and permeability to meet the impedance matching conditions.^[16] For typical examples, Chen et al. have recently prepared reduced graphene oxide (RGO)/Ni heterostructures, and the corresponding wax-based composites possessed a maximum reflection loss -17 dB with presence of 60 wt% filler

loading.^[17] Similarly, Fu et al. have reported that the NiFe₂O₄ nanorods/RGO heterofillers exhibited microwave absorption intensity up to -29.2 dB at 60 wt% filler loading.^[18] Zhang et al. have synthesized iron oxides/RGO fillers, and their wax-based composites with filler loading of 50 wt% presented a microwave absorption peak around -21 dB in the 2-18 GHz.^[19] Likewise, graphene-based hybrid fillers with a variety of hierarchical structures have also been studied.^[20-22] However, it is noted that the studies that mainly focus on controllable design of hierarchical fillers generally suffer from some critical issues. As is well known, magnetic nanofillers would primarily contribute to the magnetic loss in the relatively lower frequency regions in GHz. Their contribution in X band (8.2-12.4 GHz) and Ku band (12.4 GHz-18 GHz) is largely on the interfacial control for the dielectric loss in the graphene-based hybrid fillers.^[13,23] On the other hand, the presence of the secondary non-conductive heterofillers would usually increase the filler loading in the composites, owing to their impacts on the generation of effective 3D conductive networks for leaking current. Compared to the carbon-based heterofillers, bare carbon fillers are found to be more promising as the effective fillers for microwave absorption composites since they are generally found to deliver highly effective microwave absorption performance at filler loadings below 10 wt%.^[8,13,16,24-26]

Polymers and ceramics are ideal matrices in designing robust composites of strong mechanical properties. Generally, ceramic-based composites require sintering processes for achieving the dense and strong feature, and thus a portion of carbon-based fillers may not survive at extremely high-temperature treatments. Apparently, polymeric composites of more facile processing appear more practical and thus are attracting extensive explorations in recent studies.^[27-29] For instance, Singh et al. fabricated RGO/nitrile butadiene rubber (RGO/NBR) composites by mechanical mixing RGO and NBR in the xylene solution, and the composites

with 10 wt% RGO exhibited maximum absorption intensity up to -57 dB and a with a wide effective bandwidth from 7.5 to 12 GHz in the 2-18 GHz investigated region.^[28] Wang and coworkers have processed the RGO/MnFe₂O₄ hybrids into polyvinylidene fluoride (PVDF) via mechanical mixing with subsequent hot press. Due to the synergetic effect between graphene-based fillers and PVDF matrices, the composites with filler loading of 5 wt% can reach -29.0 dB with effective absorption range from 8.00 to 12.88 GHz.^[29]

High-temperature service requires the composites to be endowed with thermal stability in the harsh environment, specifically under high-speed transportation.^[5,30,31] In the recent attempts made by Cao and coworkers, SiO₂ nanoscale powders have been widely utilized for processing carbon/SiO₂ ceramic composites with the conventional ceramic processing routes.^[32-35] Although the as-prepared carbon/SiO₂ composites are able to offer effective microwave absorption at high temperature, the high density (dense ceramic-based composites generally >2 g/cm³), poor processability and mechanical brittleness of the ceramic composites fabricated from powders are also the concerns in practical applications.

With the consideration of substantial improvements in the filler loading, microwave absorption bandwidth, mechanical properties and thermal stability, here we present a novel strong and thermally stable polymeric graphene/silica textile composite for effective microwave absorption. With the assistance of freeze-drying approach and unique silica textiles, GO nanosheets is allowed to in situ generate homogenous 3D RGO conductive networks in the polymeric composites. More importantly, such silica textiles play significant roles in the considerable enhancement of the overall performance. The lightweight polymeric graphene/silica textile composites (density ~1.0 g/cm³) at a low filler loading of 4.1 wt% show excellent overall performance, including strong tensile strength (~40 MPa), improved thermal stability (225 °C in

air with 95% mass retention) and peak absorption up to -36 dB with effective absorption at full X-band. The exceptional features and advantages coupled with associated opportunities have been discussed.

2. Experimental section

Graphene oxide: Graphene oxide aqueous solution was prepared according to the modified Hummers method.^[36] Briefly, commercial graphite powders (2 g) and NaNO₃ (1 g) were added into the concentrated sulphuric acid (120 ml) in an ice bath, and the mixture was kept at 0 °C for 1 h. Subsequently, KMnO₄ (6 g) powders were gently added into the above mixture. (Caution: fast mixing may result in explosion). Until the stirring was applied for 2h, the mixture was heated and kept at 30 °C for 0.5 h. Then, deionized water (150 mL) was slowly added, and H₂O₂ solution (5%, 50 mL) was subsequently introduced. Upon the completion of the action, the final solution was washed with deionized water and HCl (5%), and the GO aqueous solution was achieved. **RGO/silica textile:** Initially, commercial silica textile (thickness ~0.25 mm, density ~0.7 g/cm³, where the strength and modulus of a single silica fiber in the textile is 3700 MPa and 91.6 GPa, respectively.) was washed with acetone and water under sonication for 1h, followed by drying in a vacuum oven. A stripe of the cleansed silica textile was then folded for three times with two ends fixed, with aiming to achieve ~2 mm in thickness. The folded silica textile was immersed in the mixture solution of GO and hydroquinone (GO: hydroquinone=1:5 wt/wt) under sonication for 2 h. It is noted that the height of the GO mixture solution was approximately the same to the thickness of the folded silica textile. Until stable mixture solution was achieved, the mixture was then sealed and heated up to 100 °C for 12 h, allowing GO for in situ self-assembling into RGO networks inside the folded silica textile with the presence of hydroquinone.

The as-fabricated samples were then washed with deionized water, followed by freeze-drying overnight.

RGO/silica textile/PF composites: The as-prepared RGO/silica textile were immersed into the phenol formaldehyde (PF) resin/ethanol (EtOH) solution (PF%=30 wt%). Until 2 h immersion, the mixture was then transferred into an oven, and was heated to 100 °C for resin curing (~6 h). The dried polymeric samples (RGO/silica textile/PF composite) were cut into different size for further characterizations. Using different initial GO concentrations (5, 7.5 and 10 mg/ml), the corresponding composites of different RGO loadings were obtained. According to the mass track in the entire processing, the final RGO loadings in the three as-prepared RGO/silica textile/PF composites were estimated to be around 1.3, 2.8 and 4.1 wt% (0.5, 1.0, 1.5 vol% as volume fraction), assigned as Composite-1, Composite-2 and Composite-3, respectively. Accordingly, Composite-1, Composite-2 and Composite-3 approximately contained PF of 28.2, 25.9 and 23.2 wt% (14.9, 13.1, 12.1 vol% as volume fraction), with 70.5, 71.3 and 72.7 wt% (84.6, 85.9, 86.4 vol% as volume fraction) in silica textile, respectively. The density of all the samples was estimated around 0.96~1.05 g/cm³.

Characterizations: X-ray powder diffraction (XRD) was applied on a PANalytical X' Pert PRO MPD diffraction system. Field-emission scanning electron microscopy (SEM) was performed on a ZEISS supra 55 system. In the measurement of surface resistivity, the samples were positioned on the classical four-probe setup (D41-11E/ZM, Sevenstar System). Three different spots on each sample were taken, and the average values were acquired to determine the surface resistivity. Static in-plane tensile tests were performed on a mechanical analyzer (TA-XT Plus system, SMS). In the measurement, the polymeric samples were cut into cuboids (1 × 4 × 0.2 cm³) and the two ends were fixed between stainless steel clamps with a tensile speed of 2

mm/min at room temperature. The silica textile sample was folded into a dimension of $(2 \times 4 \times 0.2 \text{ cm}^3)$, 8-layer stacked in thickness). Thermogravimetric analysis (TGA) was carried out with a TGA type instrument (Mettler Toledo, Switzerland) from 30 to 900 °C with a heating rate of 5 °C/min in air.

Complex permittivity and microwave absorption: All the testing samples were processed into dimensions of $22.86 \times 10.16 \times 2 \text{ mm}^3$. The complex permittivity (including real and imaginary permittivity) was measured on an Anritsu 37269D vector network analyzer with the wave guide method in X-band (8.2-12.4 GHz). Microwave absorption curves were obtained using the measured complex permittivity. The reflection loss (R_L), namely the term for evaluating microwave absorption performance of the absorbing layers on the ideal metal plane,^[33] was achieved by the relation^[32-35]

$$R_L = 20 \log(|Z_{in} - 1| / |Z_{in} + 1|), \quad (1)$$

where the normalized input impedance (Z_{in}) of absorption layer is given as^[32-35]

$$Z_{in} = (\mu_r / \epsilon_r)^{1/2} \tanh[j2\pi f d (\mu_r \epsilon_r)^{1/2} / c], \quad (2)$$

where c is the light velocity, f the microwave frequency, d the thickness of the absorber, ϵ_r and μ_r the complex permittivity and permeability of the samples, respectively. In all the composites, the complex permeability was approximately to be 1 due to the weak magnetic properties.^[32-35]

3. Results and discussions

In the typical preparation of the RGO/silica textile/PF composites (Figure 1), a piece of silica textile was folded into layered stacked structure for achieving the thick 3D textile architecture. The folded textile was subsequently immersed into the mixture of GO solution and hydroquinone, followed by thermal treatment to in situ form the 3D RGO framework inside the

3D textile architecture. Upon freeze-drying for maintaining the homogeneous configuration, the dried RGO/textile hybrid was then immersed into the PF resin solution and was thermally cured to fabricate the robust RGO/silica textile/PF composites. As highlighted in the scheme (Figure 1), the presence of silica textile fibers is mainly responsible for the prevention of RGO from aggregation in the entire process.

As shown in Figure 2, a silica textile (0.25 mm in thickness) of excellent flexibility (Figure 2a and 2b) was folded for three times to obtain the 8-layered stacked architecture with a thickness of 2 mm (Figure 2c and 2d). By cutting the redundant part, the thermally cured RGO/silica textile/PF composites were processed into the target size for subsequent tests. As exhibited in Figure 2g and 2h, rough top and cross-section views of the RGO/silica textile/PF composites suggest that the composites have well retained the textural and layer-stacking features of the folded silica textile.

Figure 1 Scheme of the procedures for RGO/silica textile/PF composites.

Figure 2 Photos of a stripe of silica textile (a, b), folded silica textile (c, d), and as-fabricated RGO/silica textile/PF (e, f); Top and cross-section views of the RGO/silica textile/PF (g, h).

Figure 3 gives the typical SEM images of the samples. Representative top views of the silica textile, RGO/textile and RGO/silica textile/PF composites are demonstrated in Figure 3a, 3e and 3i, respectively. Apparently, no significant change was observed in the silica textile as expected. Meanwhile, the corresponding cross-section views of the silica textile, RGO/textile and RGO/silica textile/PF composites are presented in Figure 3b-3d, Figure 3f-3h and Figure 3j-3l, respectively. In addition to the textural feature of silica textile, RGO sheets were found to be well dispersed in the textural structure (Figure f), indicating that the presence of silica textile has played a significant role in preventing the RGO from unexpected aggregation in the formation of the RGO framework (Figure 3g and 3h). According to the observation in Figure 3k and 3l, the cross-section views of the RGO/silica textile/PF composites suggest the polymerization of the PF resin in the as-fabricated composites.

Figure 3 Top views of the textile (a), RGO/textile (e) and RGO/silica textile/PF (i); Cross-section views of the textile (b-d), RGO/textile (f-h) and RGO/silica textile/PF (j-l).

The samples were carried out with various techniques for more characterizations. X-ray diffraction results show the typical broadened peak around 24° in the textile, which is attributed to silica.^[37] The (002) plane of the graphitic feature (with peak around 26°) indicates the conversion of GO into RGO in the thermal treatment. The broadened peaks observed in the RGO/silica textile/PF composites imply the as-fabricated composites have presented all the representative feature of all the RGO, silica textile and PF resin as expectation. The samples were applied to the tensile measurement, and the results were shown in Figure 4b, which suggests that the as-fabricated RGO/silica textile/PF composite apparently holds a strong tensile strength in the range of 39~44 MPa. Among the three composites, Composite-2 exhibited the optimal tensile strength due to the synergetic effects based on the coexistence of the both silica textile and RGO sheets in the polymeric matrices. The surface resistivity of the composites was acquired by the classical four-probe method, and the results in Figure 4c exhibit that the Composites-3 with the highest RGO loading presented the lowest surface resistivity (average $\sim 50 \Omega/\square$) as expected.

Figure 4 XRD spectra (a), tensile strength (b) and surface resistivity (c) of the samples as marked. The start * in (b) indicates that the value could be underestimated since the cross-section area of the textile (without stretching) was larger than that upon tensile measurement.

The as-fabricated samples with the dimensions of $22.68 \times 10.16 \times 2 \text{ mm}^3$ were directly applied to measure the electromagnetic parameters for obtaining the corresponding complex permittivity. The composites fabricated with initial different GO concentrations 5, 7.5 and 10 mg/ml are denoted as composite-1 (1.3 wt% RGO), composite-2 (2.8 wt% RGO) and composite-3 (4.1 wt% RGO), respectively. As exhibited in Figure 5, both the real and imaginary permittivity has been found to increase with the increased GO concentration used in the fabrication. In the investigated frequency region, the largest values are estimated to be around 4.75~3.75 and 3.25~2.47 in the real and imaginary permittivity, respectively. The enhanced real permittivity is mainly associated with the polarization in the RGO,^[6,12,15] and meanwhile the improved imaginary permittivity is attributed to the leaking current induced in the conductive RGO framework.^[6,12,15] It is observed that the composites fabricated with 10 mg/ml GO solution present slightly enhancement in the complex permittivity, compared to the samples with 7.5 mg/ml GO solution (Figure 5). Such result is mainly due to the fact that both the composites with 7.5 mg/ml and 10 mg/ml GO solutions possess the similar RGO framework established in the RGO/silica textile/PF composites. This observation should be understood by the fact that the configuration may approach the up-limit for the silica textile in preventing RGO aggregation, since the surface resistivity of the Composite-2 (average $\sim 72 \text{ } \Omega/\square$) and Composite-3 (average $\sim 50 \text{ } \Omega/\square$) was on the same order (Figure 4c). As shown in Figure 5c and 5f, the tangent loss, namely the ratio of imaginary part to real part, indicate the competition of the polarization and electric loss in the composites.

Figure 5 Real (a), imaginary permittivity (b) and tangent loss (c) of the samples marked; Real (e), imaginary permittivity (e) and tangent loss (f) at various frequencies.

Figure 6 Reflection loss plots of Composite-1 (a and d), Composite-2 (b and e) and Composite-3 (c and f).

Microwave absorption performance was achieved based on the measured complex permittivity. As demonstrated in Figure 6a-6c, 3D plots exhibit the relationship of reflection loss associated with frequency and thickness. The observation in Figure 6b and 6c suggests that the composite-2 and composite-3 have presented similar 3D curve coupled with effective microwave absorption band covering the entire X-band at thickness of 4.0 mm, due to their similar complex permittivity. Additionally, the peaks values found in the 3D curve indicate that the optimal thickness of the composites is around 3.5 mm. As shown in Figure 6e and 6f, the reflection loss at different thicknesses implies that composite-3 fabricated with 10 mg/ml GO solution delivers much enhanced reflection loss peak approaching -37 dB at thickness of 3.3 mm, which is much greater than the value found in the composite-2 (~-28 dB) at the same thickness. Although it seems that Composite-3 and Composite-2 have the similar permittivity from 12 to 12.4 GHz, the tangent loss (Figure 5c and 5f) shows the differences of these two samples, which explains the differences observed in the reflection loss peaks in the range of 12-12.4 GHz (Figure 6c and 6f). Apparently, the effective absorption band would be extended to higher frequency owing to the band extension feature caused by the absorption peak.^[22,23] Such thickness is well consistent with the results in the 3D plots (Figure 6e and 6f). Moreover, implication of the microwave absorption performance suggests that the composites with more than 7.5 mg/ml GO solution possess the broadband absorption in the full X-band, while the composite with 10 mg/ml GO solution that has larger complex permittivity enables the absorption peak to achieve a greater intensity, indicating the realization of improved impedance matching between the composites and free space.^[22,34,38]

Figure 7 TGA curves of the samples as marked.

For evaluating the thermostability of the composites, the samples were applied to thermogravimetric analysis (TGA) in air with a temperature increment of 5 °C/min. As manifested in Figure 7a, the silica textile shows excellent weight retention larger than 95% up to 900 °C, and both thermally cured PF resin and RGO have 90% weight retention higher than 200 °C in air. More details of the TGA results were given in Figure 7b, showing that thermally cured PF and RGO present 95% weight retention at 150 °C and 225 °C, respectively. As a consequence, the RGO/silica textile/PF composite exhibits the weight retentions of 95%, 96%, 97% and 98% at temperatures of 225 °C, 182 °C, 157 °C and 132 °C, respectively. The results indicate that such robust polymeric composites of excellent thermostability higher than 130 °C (>98% weight retention) are allowed to be utilized as high-temperature absorption in certain high-speed vehicles and aircrafts. For the applications in the more critical environment (e.g. missiles and space-shuttles), the service temperature could be extended to even higher with the presence of

thermal barrier coatings. Obviously, further improvements could be made via using much higher thermostable polymers (e.g. polyimide) instead of PF resins in the formation of polymeric composites, and thereby the flexible composites of enhanced thermostable composites are able to be envisaged.^[39]

According to the comparison with the state of the art in graphene-based microwave absorption composites (Table 1), various advantageous features could be achieved in the as-fabricated RGO/silica textile/PF composite. (1) In the work of graphene-based heterofillers, introduction of magnetic heterostructures is generally found to increase the filler loading due to the graphene aggregations in the chemical synthesis.^[13] In some typical examples, the filler loadings could even reach larger than 50 wt%,^[17-19] which is far higher than 4.1 wt% in the as-fabricated RGO/silica textile/PF composites here (Table 1). Therefore, utilization of bare carbon fillers appears more promising in the low filler-loading microwave absorption composites, consistent with the observation in the previous report.^[8,13,16,24,26] Compared to the polymeric graphene-based composites in the previous report, it is noted that employment of the exclusive silica textiles plays as the effective spacers for preventing the RGO away from aggregations, while they substantially contribute to the enhanced mechanical properties and thermostability. With the subsequent freeze-drying process, the homogeneous configuration could be well retained, enabling the composites to deliver efficient absorption at a low filler loading (4.1 wt%). (2) As a widely used matrix, wax is usually employed to prepare the composites with the purpose of demonstrating the microwave absorption performance (Table 1).^[17-21,24,40-42] It is noticeable that the utilization of wax-based composites is mainly for highlighting the performance from the fillers. However, the strategy for preparing RGO/silica textile/PF composites in this work has well demonstrated an effective processing route with excellent scalability for practical

320 applications (Table 1). (3) Generally speaking, the effective absorption bandwidth is considered
321 as a more critical factor than the absorption peak intensity in many practical cases, and the
322 attempts on the extension of effective absorption bandwidth have been largely made via
323 controlling the filler morphology and performance. Recently, RGO-based 3D structures for
324 enhancing internal reflections have shown outstanding microwave absorption performance
325 owing to the well preservation of 3D electrically conductive networks.^[6,24,42] In the design of
326 RGO/silica textile/PF composites, employment of 3D silica textile plays the significant role in
327 the reservation of 3D graphene framework for generating efficient channels for internal
328 reflection loss (Figure 8a). Thus, a microwave absorption peak up to -36 dB along with an
329 effective absorption region in the full X-band is achieved. (4) As is well known, polymers are
330 excellent matrices for fabricating mechanically robust composites.^[24,28,29,43-45] In the approach for
331 RGO/silica textile/PF composites, silica textile structures, on the other hand, are introduced as
332 the strong architecture in the reinforcement of the mechanical properties. The synergetic effects
333 caused by the established RGO-PF-fiber multi-phase interfaces enable the RGO/silica textile/PF
334 composites to hold strong tensile strength (40 MPa) (Figure 8b). Different from the cases of
335 using wax matrices (examples in Table 1), the RGO/silica textile/PF composites here could be
336 directly extended for practical applications, specifically for the circumstances where sufficient
337 mechanical strength is a critical requirement in the microwave absorption structures. (5) In the
338 recently years, the composites for high-temperature service are emerging in the aerospace and
339 related industry, which requires the composites to supply with sufficient stability under the harsh
340 temperature.^[32-35] In the previous work, silica-based composite ceramics embedded with carbon
341 nanomaterials have been fabricated, exhibiting considerable microwave absorption
342 performance.^[32-34] Apparently, employment of lightweight silica textiles (0.7 g/cm^3) into strong

thermoset polymers seems more advantages than the brittle and denser carbon/silica ceramic composites (density $> 2 \text{ g/cm}^3$), particularly in the much reinforced mechanical properties, light weight and universal processability. As a consequence, such silica textiles introduced in this work present crucial impacts on the lightweight polymeric graphene-based composites ($\sim 1 \text{ g/cm}^3$ in density), including prevention of 3D RGO framework from aggregation, improvement in the mechanical strength and promotion of the thermal stability for the composites. As illustrated in Figure 8c, contribution of PF, textiles and RGO in the overall performance indicates that such universal strategy shows outstanding efficiency for combing the advantages of each component into one ideal unit, which is able to be extended to scalable practical microwave-absorption applications in stealth technology, satellite communication, remote sensing, and other fields.

Figure 8 Schemes of reflection loss (a) and enhanced mechanism for mechanical strength (b) in the RGO/silica textile/PF composites; advantageous features from RGO, silica textile and PF, allowing the RGO/silica textile/PF composite to be endowed with the excellent overall performance for practical microwave absorption applications.

359 Table 1 Overall performance comparison of the typical graphene-based composites.

Graphene-based composites (Matrices)	Filler loadings	Mechanical properties	Thermal stability	RL peak/span of RL >10 dB (Investigated regions)	Performance from fillers or composites	Refs
3D RGO framework (silica textile/PF)	4.1 wt%	40 MPa (tensile strength)	225 °C in air with 95% mass retention	-36 dB / 8.2-12.4 GHz (8.2-12.4 GHz)	Composite	This work
RGO/Ni (paraffin wax)	60 wt%	—	—	-17 dB / 3.0–4.0 GHz, 12.0–14.0 GHz (2-18 GHz)	Filler	[17]
RGO/Fe ₃ O ₄ (paraffin wax)	40 wt%	—	—	-27 dB / 4.5–6.5 (2-18 GHz)	Filler	[40]
NiFe ₂ O ₄ nanorod–graphene (paraffin wax)	60 wt%	—	—	-44.6 dB / 6.5-9.2 GHz (2-18 GHz)	Filler	[18]
SiO ₂ @Fe ₃ O ₄ core/shell nanorod	20 wt%	—	—	-31.9 dB / 5-8 GHz (2-18 GHz)	Filler	[41]

array/graphene

(paraffin wax)

3D graphene– Fe ₃ O ₄	–	–	–	-27 dB / 7-10 GHz (2-18 GHz)	Composite	[46]
RGO (paraffin wax)	2 wt%	–	–	-42.4 dB/ 9.44–15.58 (2-18 GHz)	Filler	[24]
RGO/polypyrr ole	15 wt%	–	–	-54.4 dB/ 10-17 GHz (2-18 GHz)	Filler	[42]
RGO@Fe ₃ O ₄ N C@C@MnO ₂ (paraffin wax)	25 wt%	–	–	-38.8 dB/ 12.3 to 17.7 (2-18 GHz)	Filler	[20]
graphene@Fe ₃ O ₄ @SiO ₂ @Ni O (paraffin wax)	25 wt%	–	–	-52 dB / 12-17 GHz (2-18 GHz)	Filler	[21]
iron oxides/RGO (paraffin wax)	50 wt%	–	–	-21 dB/ 7-10.5 GHz (2-18 GHz)	Filler	[19]
RGO (NBR)	10 wt%	–	–	-57 dB / 7.5–12 GHz	Composite	[28]

				(2-18 GHz)		
RGO	7 wt%	—	>200 °C in	-42 dB/ 8.2-12.4 GHz	Composite	[32]
(silica)			air	(8.2-12.4 GHz)		
				-20 dB/ 4.5-6.5 GHz, 16.2-18 GHz	Composite	[43]
GN-Fe ₃ O ₄	30 wt%	—	—	(0.1-18 GHz)		
(epoxy)						
			>100 °C in	-29 dB / 8.0-12.88 GHz	Composite	[29]
RGO-MnFe ₂ O ₄	5 wt%	—	air with	(2-18 GHz)		
(PVDF)			99% mass retention			
			>200 °C in	-37 dB / 2-5 GHz	Composite	[47]
carbon	77 wt%	—	O ₂ with	(2-18 GHz)		
nanotubes/Fe ₃			95% mass			
O ₄	Fe ₃ O ₄		retention			
(RGO)						
				-37 dB/ 3-4 GHz, 10.3-12 GHz	Composite	[48]
porous	30 wt%	—	—	(2-18 GHz)		
Fe ₃ O ₄ @ZnO						
sphere/RGO				-25 dB/ 11.5-18 GHz	Filler	[49]
(epoxy)				(2-18 GHz)		
3D RGO/	10 wt%	—	—			
ZnO						
(paraffin wax)						

RGO@Fe ₃ O ₄				- 44.2 dB/		
@C@polyaniline	25 wt%	—	—	9.5-15.5 GHz	Filler	[50]
(paraffin wax)				(2-18 GHz)		
α -Co/GN				47.5 dB/		
(paraffin wax)	60 wt%	—	—	9.2–14.0 GHz	Filler	[51]
				(2-18 GHz)		

Conclusions

In summary, novel lightweight RGO/silica textile/PF composites toward practical microwave absorption applications were demonstrated in this work. Owing to the critical contribution from the silica textile, the composites with a low filler loading (4.1 wt%) presented robust tensile strength of 40 MPa and high thermal stability beyond 225 °C (95% mass retention in air). Meanwhile, the microwave absorption intensity was observed up to -36 dB and the effective absorption covered in the entire X-band. The results suggest that the excellent overall performance achieved in the RGO/silica textile/PF composites enable them to be sufficient in practical microwave absorption applications. Additionally, the strategy proposed in current study also demonstrates a facile and universal route for scalable fabrication of graphene-based composites, which could be extended in plenty of fields.

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