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Sandwich structured lightweight carbon black/cement composites for X-band electromagnetic wave absorption and thermal insulation

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Abstract. In order to develop a lightweight cement based composite with dual-function of EM wave absorption and thermal insulation, sandwich structure was designed to achieve excellent EM absorption capacity, and expanded perlite and carbon black were used as lightweight aggregates and EM absorbent, respectively. The EM absorption properties were studied by arching reflected method, and the results indicate that the sandwich structure design can obviously enhance the EM absorption capacity of cement composites. The sandwich structured composites exhibit better EM absorption properties than the traditional gradient multi-layer structured composites. The reflection loss of three-layer and four-layer sandwich structured composites can be less than -10 dB and -15 dB in the whole X-band. The thermal conductivity test results shows that the developed cement composites possess thermal insulation function.

Keywords: composite materials; layered structure; microwave absorption; sandwich structures; thermal conductivity

1. Introduction

Nowadays, electromagnetic (EM) pollution has been becoming an increasingly serious issue, which can do harm to human health, decrease sensitivity of precision equipment, and even cause information leakage (Gupta and Tai 2019). The application of EM absorbing materials is the most effective method to protect EM pollution, thus the development of EM absorbing materials with high performance has become more and more important and urgent.

Cement-based composite, which exhibits excellent mechanical properties and durability, is commonly used in civil fields and military fortifications. However, its EM absorption capacity is unsatisfactory. In general, introduction of dielectric or magnetic loss fillers is a feasible way to adjust the EM properties and enhance the EM absorption capacity of cement materials, and the most commonly used fillers include carbon black (Dai *et al*. 2010), carbon nano tube (Nam *et al*. 2018), helical carbon fiber (Xie *et al*. 2018), ferrites (He *et al*. 2018), industrial byproduct (Bantsis *et al*. 2012) etc.. But, ideal EM absorption cannot be obtained by adding fillers into cement merely, because of the impedance mismatch.

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Structure design is an effective means to optimize the EM absorbing properties of cement materials. Lv *et al*. (2015) developed a porous structure foam concrete, the test results indicate that the RL values are between -10 dB and -13 dB in X-band, when the sample thickness is 30 mm. The porous structure cement composites fabricated by porous aggregates, such as expanded perlite (Li *et al*. 2011), expanded polystyrene (Guan *et al*. 2007), hollow glass beads (Lv *et al*. 2018), etc., exhibit acceptable EM wave absorption performance as well. The double layer cement-based composite shows ideal EM absorption property in X-band, the bandwidth of -10 dB reflection loss (RL) reaches nearly 3GHz (Zhang and Sun 2010). Sandwich structure is an effective EM absorbing structure, however, the sandwich structured cement composites for EM absorption has been rarely reported.

In recent years, functional integration has been the main trend of building materials development, and the lightweight components has also become the main focus of building material design. In this study, a lightweight sandwich structured cement composite was fabricated by introducing light aggregates and carbon black (CB) absorbent, and the EM wave absorption property of the composite was studied in X-band, as well as its thermal insulation performance. The proposed sandwich structured cement composite can be a strong candidate for an EM wave absorbing and thermal insulation building material.

2. Materials and methods

2.1 Raw materials

The cementitious material used in this study was ordinary Type P.O 42.5 R Portland cement, which was produced by Tangshan Quanhe Cement Co. Ltd., China. Its specific area and ignition loss are 385 m^2/kg and 0.6%, respectively, and its oxide compositions are given in Table 1. Nano-sized carbon black (CB), produced by Tianjin Jinqiushi Chemical Industrial Co. Ltd., China, was used as absorbing agent. Expanded perlite (EP) with the bulk density of 150 kg/m^3 and the average diameters of 1 mm was used as aggregates, and its chemical compositions are shown in Table 2. In addition, CB dispersing agent (4050A, Winbos New Materials Co., Ltd., China) and water reducing agent (101A^+, China Building Materials Academy) were needed as well.

2.2 Sample preparations

The CB absorbent and CB dispersing agent was first dissolved in half of total amount of water, and then dispersed in an ultrasound disperser for 30 min to obtain CB solution. The weight ratio of CB and dispersing agent was selected as 4:1. The prepared CB solution and remaining water were mixed with cement in a mortar mixer for 30 s at a low speed, and then the aggregates were added into the mixer and mixed for another 90 s in a high speed.

For the single layer sample, the prepared mortar was added into an oiled mould, and then vibrated for 30s to remove air bubbles. For the multi-layer sample, the mortar with different CB and aggregate contents were poured into the moulds layer by layer according to the multi-layered design. The samples were removed from the moulds after 24 h and cured in a moist room for 28 days. The size of the samples for EM absorbing properties measurement were 180 mm×180 mm (the thickness of each sample is listed in Table 3), and the size of the samples for thermal conductivity test were300mm×300 mm×10 mm. The composition design of samples is shown in Table 3, and the photo of prepared multi-layer samples are shown in Fig. 1.

Table 1 Oxide compositions of the P.O 42.5 R cement

Chemical compositions	CaO	SiO	Al_2O_3	Fe_2O_3	SO_3	MgO
Content/ wt.%	62.64	23.85	6.12	3.26	2.32	1.81

Table 2 Chemical compositions of expanded perlite

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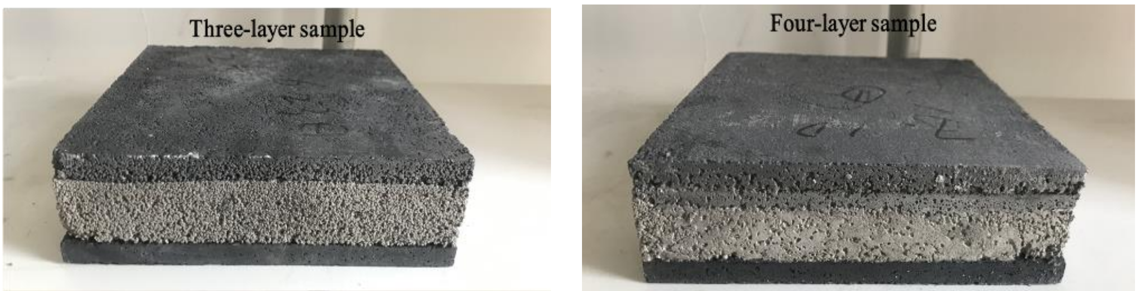

Fig. 1 The picture of multilayered cement mortar specimen

Table 3 Composition design of the samples

No.	CB content (wt.%)				EP content (vol.%)				Thickness (mm)				Density (g/cm^3)
Single layer samples													
1#	0				0				20				1.41
2#	1				0				20				1.39
3#	1				60				20				0.49
Multi-layer samples													
	1st layer	2nd layer	3rd layer	4th layer	1st layer	2nd layer	3rd layer	4th layer	1st layer	2nd layer	3rd layer	4th layer	
4#	1.30	0	3.80	-	60	60	40	-	60	60	40	-	0.48
5#	1.30	0	5.40	-	60	60	40	-	60	60	40	-	0.48
6#	0.65	0	3.80	-	60	60	40	-	60	60	40	-	0.43
7#	1.30	0	3.80	-	60	60	40	-	60	60	40	-	0.51
8#	1.10	0	2.00	4.30	60	60	60	30	10	25	5	10	0.47
9#	1.10	0.50	0	4.30	60	60	60	30	10	5	25	10	0.46
10#	0.60	1.10	1.60	4.30	70	60	45	30	20	10	10	10	0.44

2.3 Testing methods

The morphology of EP and CB particles were obtained by means of scanning electron microscopy (SEM, Quanta200) and transmission electron microscopy (TEM, Tecnai F20 ST).

The EM parameters of CB and EP were tested by the coaxial transmission/reflection method in 2-18 GHz. The sample preparation method is as follows: the testing materials were mixed with dissolved paraffin (with a 30% filler volume concentration), and then compressed into toroidal shaped with 7mm outer diameter, 3.04mm inner diameter, and 3mm thicknesses. The EM wave absorption performance were studied by arch reflecting method in 8-12 GHz, according to the standard JC/T 2499-2018 (Test method for electromagnetic wave absorbing property of building materials). The testing system should be calibrated and the sample should be drying before measurement, in order to make sure of the accuracy of the testing results. The test equipment for EM parameters and EM absorption performance test include an Agilent 5234A vector network analyzer, standard horn antennas, Agilent 85071E testing software, and coaxial airline clamp.

Thermal conductivity was tested by a DR-PD120A heat conduction and storage coefficient tester, according to the national standard GB/T 10249-2008 (Thermal insulation-Determination of steady-state thermal resistance and related properties-Guarded hot plate apparatus).

3. Results and discussion

3.1 Microstructure and EM parameters of carbon black and expanded perlite

The microstructures of CB particles are revealed by TEM image as shown in Fig. 2, from which it can be observed that the CB used in this study are spherical and their diameters are less than 100 nm. Meanwhile, the botryoidally shaped multi-pore secondary structures are formed due to aggregation of CB particles. The nanometer CB particles have large specific surface area and more dangling bonds, which can be prone to interface polarization and cause multi-scattering and reflection to absorbing more incident waves. In addition, the quantum size effect of nanometer particles can degrade the electron energy level, which is in the range of microwave energy, forming a new channel for incident attenuation. Therefore, the nanoscale CB exhibits higher EM attenuation capacity than ordinary lager scale carbon materials, such as graphite (Rusly *et al*. 2018) and carbon fiber (Pei *et al*. 2018).

The SEM micrograph of EP particle is presented in Fig. 3, which reveals that the EP is roughly spherical with a diameter of approximate 1mm, and the particle is filled with cellular through-hole structure. Thus, the EP particles can be regarded as porous materials, which can increase the porosity of cement matrix, providing more EM wave transmission path and improving the impedance condition of cement composites. In addition, the spherical EP particles coated by cement matrix and CB particles can cause multi-scattering and reflecting of incident waves, enhancing the EM absorption ability of cement composites. The porous structured EP particles can also improve the thermal insulation properties of cement composites due to the increase of porosity.

The EM characteristics of CB and EP are investigated by a vector network analyzer, and the results are shown in Fig. 4 and 5. As shown in Fig. 4, the real part of permittivity (ε') of CB is in the range of 7 ~ 18, while the imaginary part (ε'') is in the range of 6 ~ 23, as illustrated in Fig. 5. The real and imaginary parts of its permeability are nearly 1 and 0, respectively. Therefore, CB is a kind of strong dielectric loss material and the composites composed of CB can attenuate EM waves by tunnel effect, damping vibration, polarization and contact conduction of CB particles, transforming the EM energy into heat energy (Anna *et al*. 2006) and (Ghosh *et al*. 2020).

3.2 Microwave absorption performance of single layer cement composites

The EM absorption performance of single layer cement mortar are illustrated in Fig. 6. It can be seen that the pure cement exhibits a certain EM absorption capacity in X-band, and the reflection

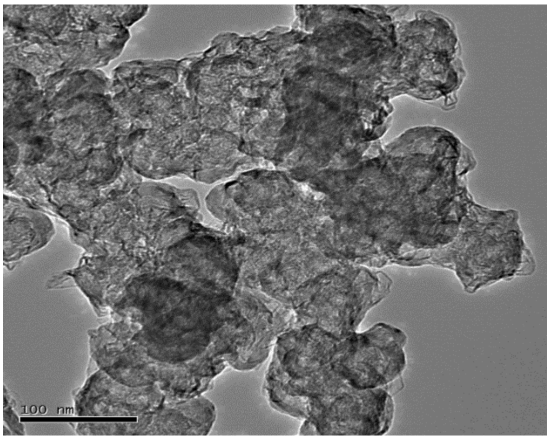

Fig. 2 TEM image of carbon black

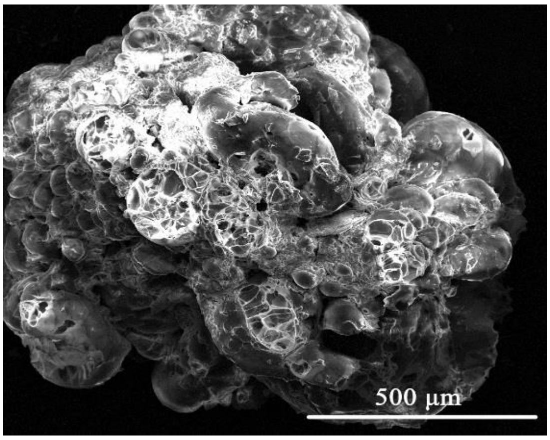

Fig. 3 SEM image of expanded perlite

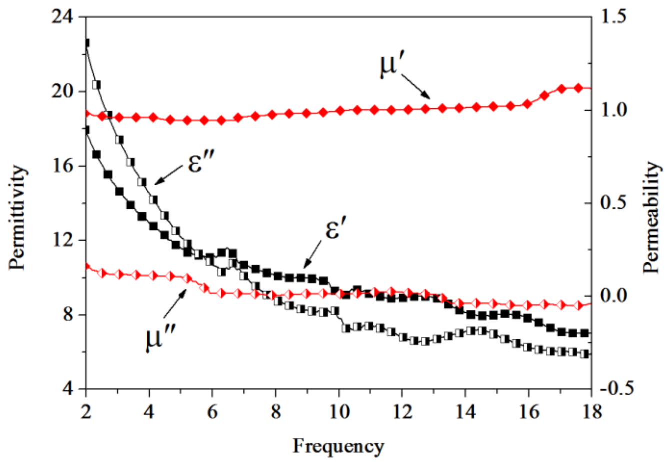

Fig. 4 EM parameter of carbon black

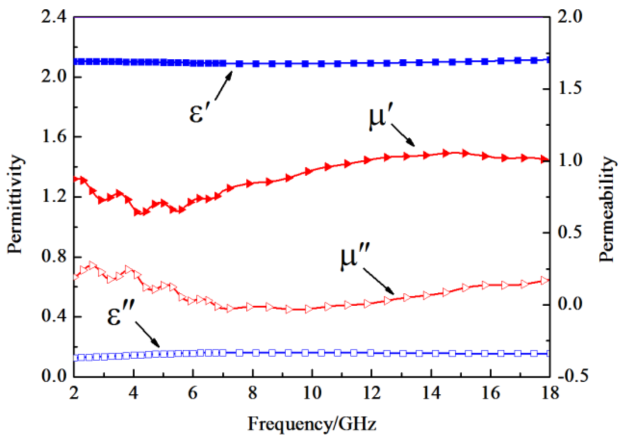

Fig. 5 EM parameter of expanded perlite

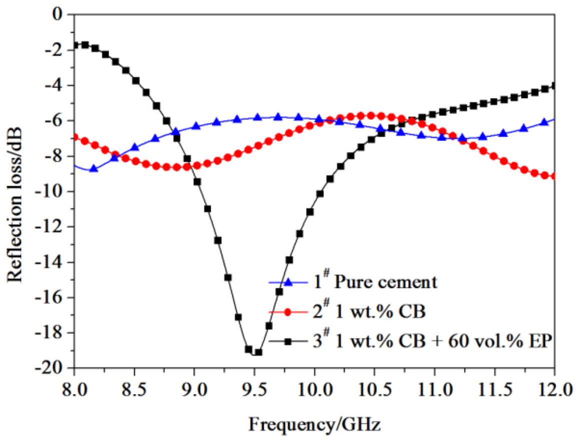

Fig. 6 EM absorption performance of single layer cement mortars

loss (RL) is in the range of -5 dB ~ -9 dB, the main reason for the EM attenuation of cement can be attributed to the different kinds of metal oxide contain in it. The addition of 1 wt.% CB into cement matrix can slightly improve the EM absorption performance, but the RL also cannot reach -10 dB in X-band. When the cement mortar contains 1 wt.% CB and 60 vol.% EP, an absorption peak of -19 dB RL is obtained in 9.5 GHz, and the RL values are less than -10 dB in 9.1-10.1 GHz.

CB is a kind of dielectric loss type absorbent, possessing high permittivity. The introduction of CB can increase the permittivity of cement composites, improving its EM attenuation capacity. However, the unilateral increase of permittivity will lead to the impedance mismatch between the composites and free space, causing heavy reflection of incident waves. Thus, the effect of CB on improving the EM absorption performance of cement composites is limited.

As its low permittivity and porous characteristic, the impedance condition of CB/cement composites can be improved by the addition of EP, meanwhile, the incident waves can more easily penetrate the composites, leading to the intense absorption peak by the interference effect (Xie *et al*. 2016). But the extensive addition of EP will decrease the permittivity of the composites, which can reduce the EM loss ability. Therefore, the EM absorption performance of single-layered cement composites cannot be significantly improved, due to the contradiction between EM loss capacity and impedance matching cannot be solved.

3.3 Microwave absorption performance of sandwich structured cement composites

Multilayer structure design is an effective method to improve the impedance condition of EM absorbing materials, due to the impedance gradient characteristic of multilayer structure can reduce the incident wave reflection (Danlée *et al*. 2014). We designed a sandwich structure to further improve the EM absorption properties of cement materials. For the sandwich structure, except the electrical loss of CB and destructive interference between incident and reflected waves, an EM wave resonator also can be formed by the wave-transparent interlayer, providing an additional EM attenuation path.

The RL of the prepared three-layer sandwich structure cement mortar are illustrated in Fig. 7 and Fig. 8. It can be seen from Fig. 7 that the three-layer sandwich structure cement composites exhibit excellent EM absorption properties in X-band, which can be affected obviously by the variety of CB contents of top and bottom surface layer. When the CB content of bottom layer is 5.4%, the absorption peak of RL curve becomes weaker, just like sample 5#. When the CB content of bottom layer is high (5.4%), the conductivity will be increase obviously, leading to the reduction of skin depth (Kwak *et al*. 2007). Thus, the incident wave cannot penetrate the bottom layer, increasing the wave reflection, further weakening the EM absorption ability. Compared with 4# and 6# samples, it is clearly that the top surface layer contain 1.3% CB shows stronger EM absorption ability in the whole X-band, the RL can be nearly lower than -12 dB and the minimum RL reaches -23 dB. That is because the EM attenuation capacity of surface layer contains 1.3% CB is stronger than that with 0.65% CB.

The RL of cement composites also can be influenced by the thickness of sandwich layer, as shown in Fig. 8. It can be observed that when the thickness of sandwich layer decrease from 30mm to 20mm, the shape of the RL curve changes from a “V” shape to a “W” shape in the X-band, the absorption peaks with RL of -21 dB and -17 dB are obtained at 8.4 GHz and 11.2 GHz, respectively. However, the EM absorption performance of the sample with 30 mm thickness sandwich layer can be better than that with 20 mm sandwich layer in 8.8-12 GHz. So, the effective absorption frequency ranges of sandwich structure cement composites can be adjusted by varying the sandwich thickness.

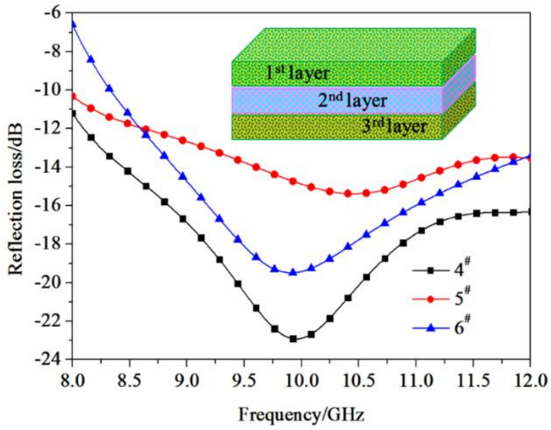

Fig. 7 Effect of CB content on the RL of three layer sandwich structured cement composites

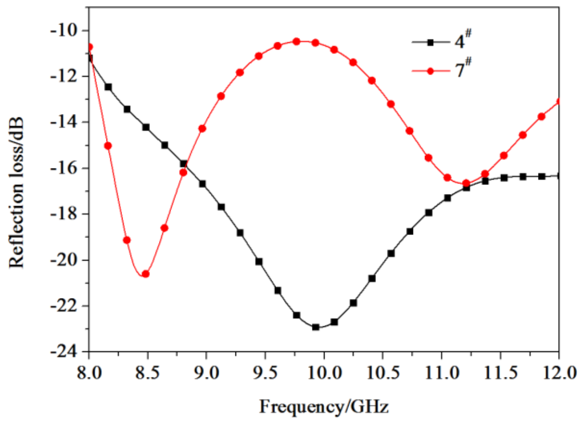

Fig. 8 Effect of interlayer thickness on the RL of three layer sandwich structured cement composites

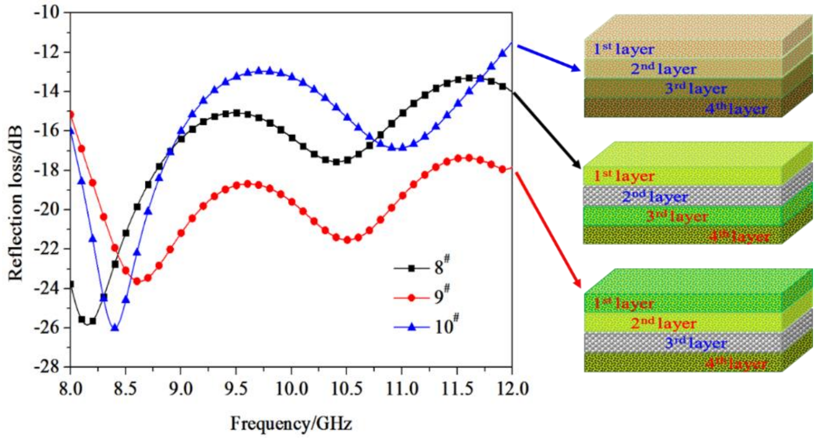

Fig. 9 The EM absorption properties of four layer gradient (10#) and sandwich (8#, 9#) structured cement composites

It also can be seen that the EM absorption capacity can be significant influenced by the variation of sandwich layer position. When the sandwich layer is located in the second layer, the RL of 8# sample is under -13 dB in the whole X-band, and the minimum RL reaches nearly -26 dB. However, if the sandwich layer is located in the third layer, the RL of 9# sample can be lower than -15 dB in X-band, which is much better than 8#. The design of two layers gradient above the sandwich layer can further reduce the reflection of incident waves, making more incident waves propagate into the sandwich structure and be attenuated by electrical loss of CB, resonance of sandwich layer, and interference effect. Therefore, the 9# sample exhibits much better EM absorption properties than 8# sample.

In Ren *et al*. 2020, the RL of the fabricated double-layered cement composites with 30mm thickness and 3 wt.% carbon powders can less than -10 dB in X-band, but the RL value cannot reach -15 dB, as well as a three-layered cement absorber (Zhang and Sun 2012). Thus, compared to the traditional layer structured cement based absorber, the sandwich structured absorber fabricated in this study exhibits better microwave absorbing performance.

3.4 Thermal insulation property of the cement composites

In order to verify the thermal insulation property of the lightweight cement composites design in this work, we prepared a specific sample to test its thermal conductivity. The sample of the tested lightweight cement composites contains 1 wt.% CB absorbent and 60 vol.% EP lightweight aggregate, and its heat conductivity coefficient is 0.073 W/(m·K), which can meet the standard value (≤ 0.085 W/(m·K)) specified in the national standard GB/T 20473-2006 (Dry-mixed thermal insulating composition for building) and can be used as thermal insulation building materials.

4. Conclusions

In this work, sandwich structured lightweight carbon black/cement composites integrated EM wave absorption and thermal insulation properties were fabricated, and the EM absorption properties were investigated by arch reflecting method in X-band.

• The sandwich structured cement composites exhibit higher EM absorption performance than traditional gradient multi-layer structure. The RL of three-layer sandwich structured composites can be lower than -10 dB in the whole X-band, which can be further decreased to be less than -15 dB by the design of four-layer sandwich structure.

• The thermal conductivity of the lightweight cement composites is 0.073 W/(m·K).

• This sandwich structured cement composite can be a candidate for X-band EM absorption and thermal insulation building material.

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