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Study on Electromagnetic Properties of Graphite/Graphene/Silver-Coated Copper Powder Monolayer Coated Composites

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Abstract

With the rapid development of electronic technology and military techniques, electromagnetic protection materials are becoming more and more significant to people. Harmful electromagnetic radiation not only affects the normal operation of electronic equipments and military security, but also has a serious impact on human health. At present, using absorbing and shielding materials are effective means to reduce the harm of electromagnetic waves. In this project, graphite, graphene and silver-coated copper powder coated composites were prepared using PU-2540 polyurethane and adopting a coating process for the substrate on plain polyester/cotton fabric. The controlled variable method was used to prepare and study the electromagnetic properties of single-layer coating composites with different functional particle contents. The result showed that within the frequency range of 0.01GHz~1.0GHz, when the total mass of functional particles was 48% relative to that of the polyurethane, the value of the real part of the dielectric constant of the sample remained the largest and its polarization ability was the strongest. Within the frequency range of 0.08GHz~1.0GHz, when the content of functional particles was 24% relative to that of the polyurethane, the value of the imaginary part of the dielectric constant and the loss of the tangent value of the sample kept the maximum, and the loss and attenuation ability with respect to electromagnetic waves were both the strongest. Within the frequency range of 1.3GHz~2.0GHz, when the content of functional particles was 36% relative to that of the polyurethane, the value of the reflection loss of the sample was -26.93dB, and the minimum value of the reflection loss was obtained at a frequency of 1.6GHz, at the moment of which, the absorbing property of the sample was the best.

Keywords

graphite, graphene, silver-coated copper powder, coated composites, electromagnetic properties.

1. Introduction

Electromagnetic protection materials can be produced either by structure design or coating. Coating is easily applicable to most of the current materials that are worth studying from a cost, processing and performance aspect, and it easy to service at a later period [1-3]. According to the absorbing and shielding mechanism of the coating, the properties mainly depend on the functional particles, layer number and layer thickness of the electromagnetic protection materials [4-6].

Graphite is a kind of elemental carbon, belonging to the hexagonal crystal system, [7] which has stable chemical properties, corrosion resistance, and does not react easily to acids and bases [8]. The special structure of graphite determines that it has the following properties: high temperature resistance, strong plasticity, good thermal shock resistance, as well as good conductivity and thermal conductivity [9-10]. Graphite, one of the earliest absorbent materials, is used to fill a layer of airplane skin to absorb radar waves [11]. Graphene is a two-dimensional nanomaterial composed of carbon six-membered rings with a honeycomb lattice structure [12]. The special structure makes it have excellent electrical and mechanical properties, such as high conductivity, [13] a light weight and high tensile strength [14-17]. With its large specific surface area and high carrier concentration, graphene is a lightweight electromagnetic protection material with great development potential [18-19]. Silver-coated copper powder is a new kind of material obtained by coating silver on a copper core surface. Silver has ultra-high conductivity, but is expensive and prone to "migration" at high temperatures and humidity. The conductivity of copper is close to that of silver, but it is easy to oxidize and its conductivity to decrease [20]. The conductivity of silver-coated copper powder is similar to that of silver and copper, and it has high cost performance and avoids the defects of silver ion migration and copper oxidation [21]. There are many methods for preparing silver-coated copper powder, among which electroless plating is the main method for preparing silver-coated copper powder due to its simple process and low cost.

This project used plain polyester-cotton fabric as the base cloth, PU-2540 polyurethane as the matrix, and graphite, graphene and silver-coated copper powder as the functional particles, using the coating process to prepare graphite/graphene/silver-coated copper powder monolayer coating composites. The effects of different functional particle content on the electromagnetic properties of monolayer coated composites were studied by using the method of control variables.

2. Experimental part

2.1. Main experimental materials and chemicals

The main experimental materials were as follows: polyester-cotton fabric (plain woven fabric), provided by Baoji Changyuan Industry Trade Co., Ltd. The

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Experimental materials	Specification	The manufacturer
Graphite powder	Q/HG3991-88	Tianjin Fengchuan Chemical Reagent Technology Co., Ltd
Graphene	5～15μm	Tianjin Kairuisi Fine Chemical Co., Ltd
Silver-coated copper powder	P-30	Shenzhen Changxinda Shielding Material Co., Ltd
Polyurethane	PU-2540	Guangzhou Yuheng Environmental Protection Materials Co., Ltd
Thickener	7011	Guangzhou Dianmu Composite Materials Business Department
Slurry defoaming agent	5020	Jiangsu Hengyu Chemical Industry Group Co., Ltd

Notes: loose packing density of silver-coated copper powders: 1.6~1.8g/cm^3; reference paint resistance: 0.02Ω·cm; distribution of the particle size: D10: ≥15.0μm, D50: 26.7~27.3μm, D90: ≤50μm; appearance: flake, silver white.

Table 1. Main experimental chemicals

The Instrument	Specification	The manufacturer
High-temperature blast drying oven	DGG-9148A	Shanghai Aozhen Instrument Manufacturing Co., Ltd
Coating machine	LTE	Werner Mathis, Switzerland
Digital viscometer	SNB-2	Shanghai Hengping Instrument And Meter Factory
Digital fabric thickness gauge	YG141D	Laizhou Electronic Instrument Co., Ltd
Dielectric spectrometer	BDS50	Novocontrol Gmbh, Germany
Vector network analyzer	ZNB40	Rohde Schwarz, Germany

Table 2. Main test instruments

main experimental chemicals are shown in Table 1.

2.2. Main experimental instruments

Main experimental instruments are shown in Table 2.

2.3. Experimental steps

Single-layer coated composites with different contents of functional particles were prepared according to the following procedure:

1. The base cloth was cut to the size of 25×45cm, fixed on a needle plate clip with a certain tension, the needle plate clip was then installed on the coating machine, and the coating thickness was set at 1mm;

2. (graphite, graphene, silver-coated copper powders) were weighed according to mass percentages of 0%, 12%, 24%, 36%, 48% and 60%, respectively, relative to the polyurethane, and the ratio of functional particles (graphite: graphene:silver-coated copper powder) amounted to 6: 3: 1;

3. 80g of the polyurethane was weighed in a beaker and then mixed on an agitator (600 rpm);

4. Functional particles in the following order: graphene, graphite and silver-coated copper powder (the molar mass from small to large), were added;

5. Then rotating speed was reduced, the beaker stirred for 5 min (500 rpm), and the defoaming agent (30 drops) and thickening agent (viscosity - about 30000 mPa·s) were added;

6. The rotating speed was increased and the beaker stirred for 30 min (2000 rpm);

7. The viscosity was measured with a digital viscometer;

8. The product prepared was smeared on a base cloth and dried at 80°C for 30min.

2.4. Test indicators

(1) Test of the dielectric constant

According to Standard SJ20512-1995: "The test method for the complex dielectric constant and complex permeability of microwave high-loss solid materials" [22], the dielectric constant of the sample was tested by a BDS50 dielectric spectrometer. A circular electrode plate with a diameter of 2.5cm and a clamping distance of 16453A was selected along with a testing frequency of 0.1GHz<f<1.0GHz. After setting, the sample (size 2×2cm) was placed between the upper and lower electrodes of the fixture to test the dielectric constant (the real and imaginary parts and loss tangent value) of the sample. After the test, the required data were saved.

(2) Test of the reflection loss

The reflection loss of the sample was measured by a ZN40 vector network analyzer. The test line was connected to the host machine using a test fixture; the test frequency band was set as 0.1GHz<f<3.0GHz, and the sample was put into a coaxial fixture after the setting. The standard metal plate was covered on the coaxial fixture; the reflection loss of the sample then tested, and the data required after the test were saved. When RL≤-10dB, 90% of electromagnetic waves experiencing the loss were absorbed.

(3) Test of shielding effectiveness

According to GJB6190-2008: "The measurement method for the shielding effectiveness of electromagnetic shielding materials" [23-24], a ZN40 vector network analyzer was used to test the shielding effectiveness of the samples. The test line was connected to the host machine using a test fixture; the test frequency band was set as 0.1GHz<f<3.0GHz, next the sample was put between the coaxial fixture after setting, the shielding performance of the sample then tested, and the data required were saved after the test was completed. When SE≥20 dB, 90% of electromagnetic waves was shielded.

3. Result analysis and discussion

3.1. The influence of the content of functional particles on the electromagnetic properties of the single-layer coated composites

In order to study the influence of the content of functional particles (graphite, graphene, silver-coated copper powder) on the dielectric constant (real and imaginary parts, loss tangent value), reflection loss and shielding effectiveness of the single-layer coated composites, six types of different contents of functional particles of the single-layer coated composites were prepared changing the content of functional particles (mass percentages relative to the polyurethane were 0%, 12%, 24%, 36%, 48 % and 60%) on the plain polyester fabric. Specific process parameters are shown in Table 3.

(1) The influence of the content of functional particles on the dielectric constant of the single-layer coated composites

Single-layer coated composites with different contents of functional particles were prepared and values of the dielectric constant were tested (test frequency range - 0.01GHz<f<1.0GHz), which is shown in Figures 1, 2 and 3.

It can be seen from Figure 1 that within the frequency range of 0.01GHz~1.0GHz, with increasing frequencies of the applied electric field, the value of the real part of the dielectric constant of each sample decreased, and the polarization ability with respect to electromagnetic waves also decreased, which may be because the speed of the dielectric polarization of the sample failed to keep pace with the changing speed of the electric field, resulting in the relaxation phenomenon. When the frequency was 0.01GHz, the value of the real part of the dielectric constant for each sample was the largest, and the magnetic field generated inside the absorbing material had the strongest polarization ability with respect to the applied magnetic field. Within the frequency range of 0.01GHz~0.3GHz, when the content of functional particles was 60 % relative to the polyurethane, the value of the real part of the dielectric constant of the sample decreased the fastest with increasing frequencies of the applied electric field, followed by values

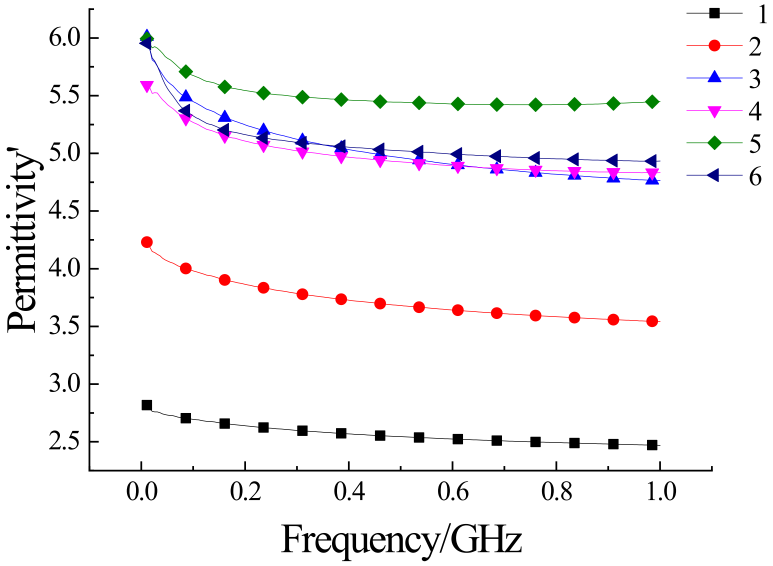

Fig. 1. Influence of the content of functional particles on the real part of the dielectric constant of the single-layer coated composites

N umber	Content of functional particles (%)	Ratio of the graphite/graphene/ silver-coated copper powder	Stirring time (min)	Viscosity (mPa·s)	Thickness (mm)	Drying temperature (℃)	Drying time (min)
1	0	6:3:1	30	30000	1.0	80	30
2	12	6:3:1	30	30000	1.0	80	30
3	24	6:3:1	30	30000	1.0	80	30
4	36	6:3:1	30	30000	1.0	80	30
5	48	6:3:1	30	30000	1.0	80	30
6	60	6:3:1	30	30000	1.0	80	30

Table 3. Process parameters of different contents of functional particles

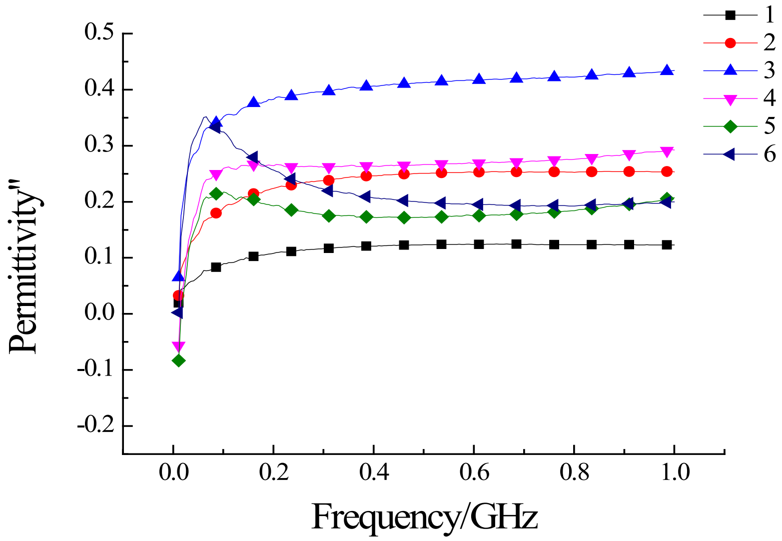

Fig. 2. Influence of the content of functional particles on the imaginary part of single-layer coated composites

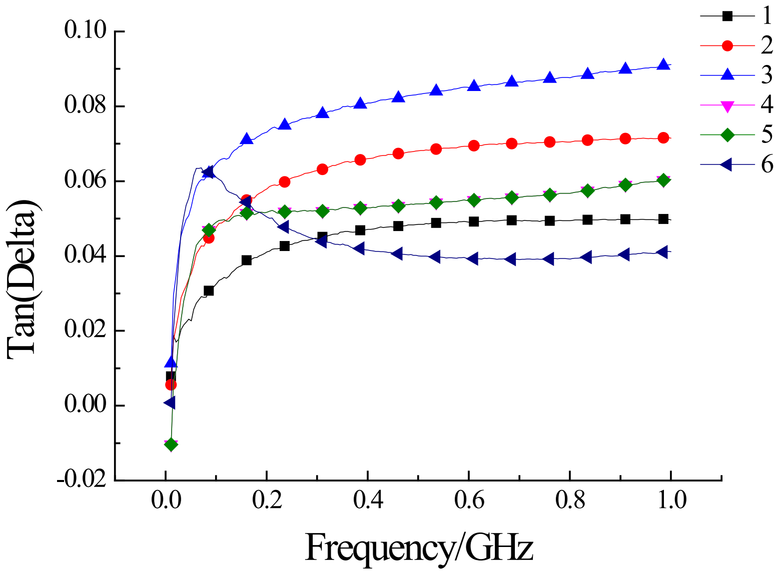

Fig. 3. Influence of the content of functional particles on the loss tangent value of the single-layer coated composites

of samples with contents of functional particles of 24%, 36%, 48%, 12% and 0% relative to polyurethane. Within the frequency range of 0.01GHz~0.3GHz, when the content of functional particles relative to the polyurethane was 48%, the value of the real part of the dielectric constant of sample was the largest, and its ability with respect to polarising electromagnetic waves was the strongest, followed by values of samples with contents of functional particles of 24%, 60%, 48%, 12% and 0% relative to polyurethane in turn. Within the frequency range of 0.3GHz~0.6GHz, the value of the real part of the dielectric constant of samples was the largest when the content of functional particles relative to the polyurethane was 48%, and its polarization ability with respect to electromagnetic waves was the strongest, followed by values of samples with contents of functional particles of 60%, 24%, 36%, 12% and 0% relative to polyurethane. Within the frequency range of 0.6GHz~1.0GHz, the value of the real part of the dielectric constant of sample was the largest when the content of functional particles relative to the polyurethane was 48%, and its polarization ability to electromagnetic waves, was the strongest, followed by values of samples with contents of functional particles of 60 %, 36 %, 24 %, 12 % and 0 % relative to polyurethane. That is, within the frequency range of 0.6GHz~1.0GHz, when the content of functional particles relative to the polyurethane was 48 % and the ratio of the graphite, graphene and silver-coated copper powder was 6:3:1, the value of the real part of the dielectric constant for the sample remained the largest, and its polarization ability to electromagnetic waves was the strongest.

As can be seen from Figure 2, within the frequency range of 0.01GHz~1.0GHz, when contents of functional particles of the sample relative to the polyurethane were 0 %, 12 %, 24 %, 36 % respectively, the value of the imaginary part with increasing frequencies of the applied electric field gradually increased, while when contents of functional particles relative to the polyurethane were 48 % and 60 %, the value of the imaginary part of the dielectric constant for samples decreased after first increasing; but overall values of the imaginary part of the dielectric constant all increased gradually, and its loss ability with respect to electromagnetic waves gradually enhanced. Within the frequency range of 0.01GHz~0.05GHz, when the content of functional particles was 60% relative to the polyurethane, the value of the imaginary part of the dielectric constant for the sample increased with increasing frequencies of the applied electric field, and the increase in speed was the fastest, followed by values of samples with contents of functional particles of 24%, 36%, 48%, 12% and 0% relative to the polyurethane in turn. Within the frequency range of 0.05GHz~1.0GHz, when the content of functional particles of samples relative to the polyurethane was 48%, the value of the imaginary part of the dielectric constant decreased in a small range with increasing frequencies of the applied electric field,. When the content of functional particles of samples relative to the polyurethane was 60%, the value of the imaginary part of the dielectric constant greatly decreased with increasing frequencies of the applied electric field, while values of the imaginary part of the dielectric constant of the rest of samples all increased. And when the content of functional particles of samples relative to the polyurethane was 24%, the value of the imaginary part of the dielectric constant greatly increased with increasing frequencies of the applied electric field. Within the frequency range of 0.01GHz~0.05GHz, when the content of functional particles of samples relative to the polyurethane was 60%, the value of the imaginary part for the dielectric constant of the sample was the largest, and its loss ability with respect to electromagnetic waves was the strongest, followed by values of samples with contents of functional particles of 24%, 36%, 48%, 12% and 0% relative to polyurethane in turn. Within the frequency range of 0.05GHz~0.1GHz, when the content of functional particles of samples relative to the polyurethane was 24%, the value of the imaginary part for the dielectric constant of the sample was the largest, and its loss ability with respect to electromagnetic waves was the strongest, followed by values of samples with contents of functional particles of 60%, 36%, 48%, 12% and 0% relative to polyurethane in turn. Within the frequency range of 0.15GHz~0.25GHz, when the content of functional particles of samples relative to the polyurethane was 24%, the value of the imaginary part of the dielectric constant for the sample was the largest, and its loss ability with respect to electromagnetic waves was the strongest, followed by values of samples with contents of functional particles of 36%, 60%, 12%, 48% and 0% relative to polyurethane in turn. Within the frequency range of 0.25GHz~1.0GHz, when the content of functional particles of samples relative to the polyurethane was 24%, the value of the imaginary part of the dielectric constant of the sample was the largest, and its loss ability with respect to electromagnetic waves was the strongest, followed by values of samples with contents of functional particles of 36%, 12%, 60%, 48% and 0% relative to the polyurethane in turn. That is, within the frequency range of 0.01GHz~0.05GHz, with increasing frequencies of the applied electric field, when the total mass of the graphite, graphene and silver-coated copper powder relative to the polyurethane was 60% and at a ratio of 6:3:1, the coating had the largest value of the imaginary part of the dielectric constant for the sample and the strongest loss ability with respect to electromagnetic waves. Within the frequency range of 0.05GHz~1.0GHz, with increasing frequencies of the applied electric field, when the total mass of the graphite, graphene and silver-coated copper powder relative to the polyurethane was 24% and at a ratio of 6:3:1, the coating had the largest value of the imaginary part of the dielectric constant for the sample and the strongest loss ability with respect to electromagnetic waves. Within the frequency range of 0.01GHz~1.0GHz, along with increasing frequencies of the applied electric field, when contents of functional particles relative to the polyurethane were 48% and 60%, values of the imaginary part of the dielectric constant decreased after first increasing, and the loss ability with respect to electromagnetic waves showed an increasing and then decreasing trend. Values of the imaginary part of the dielectric constant of samples when contents of functional particles relative to the polyurethane were 0%, 12%, 24% and 36% showed an increasing trend all the time, with the increase range being the largest at the beginning, and then decreasing to some extent. The loss ability with respect to electromagnetic waves showed an increasing trend.

As can be seen from Figure 3, within the frequency range of 0.01GHz~1.0GHz, with increasing frequencies of the electric field, loss tangent values of each sample increased to some extent, and the attenuation ability with respect to electromagnetic waves strengthened gradually. When the content of functional particles relative to the polyurethane was 60%, the loss tangent value of samples first increased rapidly, then slowly decreased, and finally the same size remained, while the loss tangent values of the rest of samples increased rapidly first and then increased slowly. When the frequency of the applied electric field was 0.01GHz, the loss tangent value of each sample was at the minimum with the minimum attenuation ability. When the frequency of the applied electric field was 1.0GHz and contents of functional particles relative to the polyurethane were 0%, 12%, 24%, 36% and 48%, respectively, the loss tangent value of samples reached the maximum value, and the attenuation ability of the magnetic field produced in the coating with respect to the applied magnetic field was the strongest. When the frequency of the applied electric field was 0.03GHz and the content of functional particles relative to the polyurethane was 60%, the loss tangent value of the sample was the largest, and the attenuation ability with respect to electromagnetic waves at this time was the strongest. Within the frequency range of 0.01GHz~0.09GHz, when the content of functional particles was 24% relative to the polyurethane, the loss tangent value of the sample increased with increasing frequencies of the applied electric field, and speed increase was the fastest, followed by values of samples with contents of functional particles of 48%, 36%, 12% and 0% relative to the polyurethane in turn. Within the frequency range of 0.09GHz~0.12GHz, when the content of functional particles was 24% relative to the polyurethane, the loss tangent value of the sample was the largest, followed by values of samples with contents of functional particles of 60%, 48%, 36%, 12% and 0% relative to the polyurethane in turn. Within the frequency range of 0.12GHz~0.17GHz, when the content of functional particles was 24% relative to the polyurethane, the loss tangent value of the sample was the largest, followed by values of samples with contents of functional particles of 60%, 12%, 48%, 36% and 0% relative to the polyurethane in turn. Within the frequency range of 0.17GHz~0.3GHz, when the content of functional particles was 24% relative to the polyurethane, the loss tangent value of the sample was the largest, followed by values of samples with contents of functional particles of 12%, 48%, 36%, 60% and 0% relative to the polyurethane in turn. Within the frequency range of 0.3GHz~1.0GHz, when the content of functional particles was 24% relative to the polyurethane, the loss tangent value of the sample was the largest, followed by values of samples with contents of functional particles of 12%, 48%, 36%, 0% and 60% relative to the polyurethane in turn.

In general, within the frequency range of 0.01GHz~1.0GHz, when the total mass of the graphite, graphene and silver-coated copper powder relative to the polyurethane was 24% and at a ratio of 6:3:1, the loss tangent value of the coating was better than that of other samples, and its attenuation ability with respect to electromagnetic waves was the strongest.

(2) The influence of the content of functional particles on the reflection loss of the single-layer coated composites

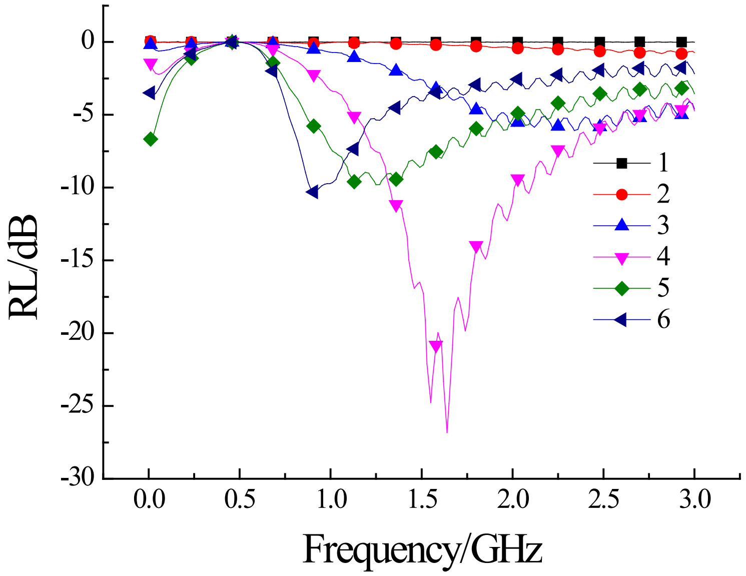

Fig. 4. Influence of the content of functional particles on the reflection loss of the single-layer coated composites

Single-layer coated composites with different contents of functional particles were prepared and values of the reflection loss tested (the test frequency range was 0.01GHz<f<3.0GHz), which is shown in Figure 4.

As can be seen from Figure 4, within the frequency range of 0.5GHz~3.0GHz, with increasing frequencies of the applied electric field, values of the reflection loss of samples all decreased gradually when contents of functional particles were 0% and 12% relative to the polyurethane, and the reduction range was not large. Within the frequency range of 0.8GHz~1.3GHz, the value of the reflection loss of the sample was -10.31dB≤RL≤-5.0dB when the content of functional particles was 24% relative to the polyurethane, and the minimum value was obtained at a frequency of 0.9 GHz. Within the frequency range of 1.3GHz~2.0GHz, the value of the reflection loss of the sample was -26.93dB≤RL≤-10.0dB when the content of functional particles was 36% relative to polyurethane, and the minimum value was obtained at a frequency of 1.6GHz. Within the frequency range of 0.8GHz~2.2GHz, the value of the reflection loss of the sample was -9.9dB≤RL≤-5.0dB when the content of functional particles was 48% relative to the polyurethane, and the minimum value was obtained at a frequency of 1.2GHz. Within the frequency range of 0.5GHz~3.0GHz, with increasing frequencies of the applied electric field, the value of the reflection loss of the sample first decreased and then slowly increased when the content of functional particles was 60% relative to the polyurethane, and the minimum value of the reflection loss of RL= -6.17dB was obtained at a frequency of 2.4GHz.

In conclusion, within the frequency range of 0.5GHz~3.0GHz, with increasing contents of functional particles, the wave peak of the reflection loss peaks moved to a low frequency. When the total mass of the graphite, graphene and silver-coated copper powder relative to the polyurethane was 36% and at a ratio of 6:3:1, the value of the reflection loss for the coating was better than that of other samples, thus increasing the content of functional particles within a certain range, which is conducive to improving the absorbing properties. However, when the total mass of functional particles relative to the polyurethane exceeded 36%, the absorbing property of samples declined.

(3) The influence of the content of functional particles on the shielding effectiveness of the single-layer coated composites

Single-layer coated composites with different contents of functional particles were prepared and values of the shielding effectiveness were tested (the test frequency range was 0.01GHz<f<3.0GHz), which is shown in Figure 5.

As can be seen in Figure 5: within the frequency range of 0.01GHz~3.0GHz, with increasing frequencies of the applied electric field, the shielding-attenuation value of each sample increased after first decreasing, The shielding-attenuation value of the sample was the minimum when the content of functional particles was 0% and 12%, and the attenuation ability with respect to electromagnetic waves was the minimum, followed by shielding-attenuation values of remaining samples with contents of functional particles of 24%, 36%, 48% and 60% relative to the polyurethane in turn. Within the frequency range of 0.6GHz~3.0GHz, with increasing frequencies of the applied electric field, the value of the shielding effectiveness of each sample increased slightly, and the attenuation ability with respect to electromagnetic waves increased. Therefore, at a frequency of 0.01GHz, the maximum shielding-attenuation value was achieved. When the frequency was 0.01GHz, maximum shielding-attenuation values of each sample were 39.93 dB, 38.00 dB, 39.55 dB, 48.35 dB, 55.03dB and 66.38 dB, respectively. This may be

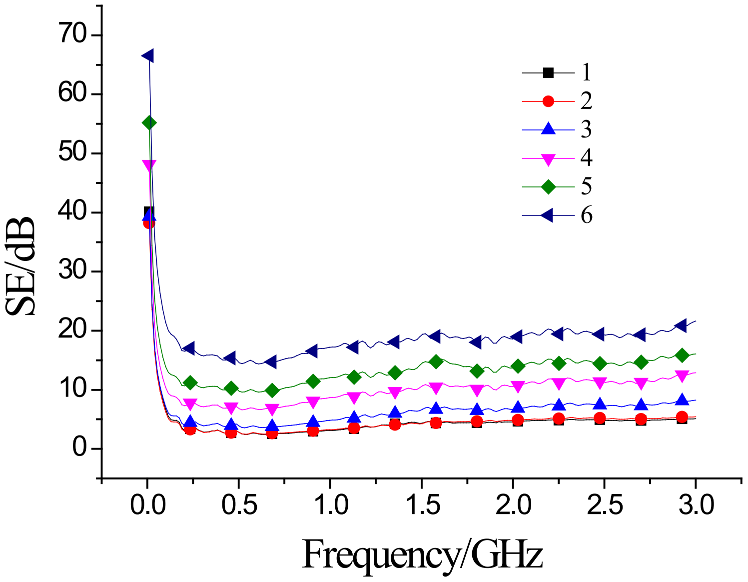

Fig. 5. Influence of the content of functional particles on the shielding effectiveness of the single-layer coated composites

because the conductivity of the coating material was mainly co-determined by the connection of conductive particles into chains, the formation of a conductive network, the spacing of conductive particles, and conductive channels formed by electrons passing through the thin polymer layer. The content of the filler directly determined the formation of the conductive network and conductive channel. As the content of silver-coated copper powders increased gradually, the conductivity of the sample increased and its shielding effectiveness increased [25].

In conclusion, within the frequency range of 0.01GHz~3.0GHz, with increasing frequencies of the applied electric field, when the total mass of the graphite, graphene and silver-coated copper powder relative to the polyurethane was 60% and at a ratio of 6:3:1, the value of the shielding effectiveness of the coating was better than that of other samples.

4. Conclusion

Within the frequency range of 0.01GHz~1.0GHz, an appropriate increase in the content of functional particles can increase the dielectric constant (real and imaginary parts and loss tangent value) of the sample with respect to electromagnetic waves, but when the content exceeds 36% relative to the polyurethane, the dielectric constant of the sample decreases. Within the frequency range of 0.1GHz~3.0GHz, an appropriate increase in the content of functional particles can increase the reflection loss of electromagnetic waves, but when the total mass of functional particles exceeds 36% relative to the polyurethane, the sample's absorbing property will decline, which will affect the sample's absorbing property.

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