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ARTICLE

Preparation of Graphene-Coated Aluminum Composite Powder by Dispersing Graphene Aerosol Through Electrical Explosion

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Abstract: A new method to produce graphene-coated aluminum composite powder by graphene aerosol prepared by electrical explosion was proposed. This method solves the problems of uneven dispersion and weak interfacial bonding of graphene in the metal composite powder without damaging the intrinsic properties of graphene. The graphene aerosol of specific content was prepared by the electrical explosion, and the obtained graphene had two structures, namely the flake graphene and the gel graphene. The graphene had 5 – 8 layers, and the graphene aerosol was uniformly dispersed in the chamber. Then, the graphene aerosols were mixed with spherical aluminum particles by stirring under the air flow, and the graphene-coated aluminum composite powders with different graphene contents were prepared. When the content of graphene aerosol is 1.5wt%, the graphene is uniformly dispersed in the aluminum composite powder and the graphene sheets adhere to the metal particles. Finally, the mechanism of in-situ formation of graphene-coated metal composite powder by dispersing of graphene aerosols was analyzed.

Key words: electrical explosion; graphene aerosol; aerosol agglomeration; composite powder

Graphene has attracted extensive attention due to its excellent mechanical and electrical properties[1] in the fields of polymer, metal, ceramic, and graphene composites, showing great potential in large-scale application[2–4]. In the graphene metal composites, graphene is usually added to the matrix as reinforcement to improve the properties[5]. However, due to the π-π Van der Waals force and the electrostatic and polar interactions between graphene sheets, graphene is easy to aggregate when it combines with metal. Besides, the unstable and uneven dispersion of graphene in the metal composites degrades the effect induced by graphene[6]. Therefore, the graphene agglomeration seriously restricts its application in metal composites. Generally, the mechanical properties and corrosion resistance of composites can be improved after the graphene addition, regardless of the even/uneven dispersion of graphene in the metal composites. However, the enhancement is more obvious with the uniform dispersion of graphene[7]. Therefore, the performance of graphene-reinforced metal composites is directly related to the dispersion of graphene.

The dispersion methods of graphene in metal composites include solid phase dispersion[8], liquid phase dispersion[9], and solid-liquid combination[10]. High-energy ball milling is a commonly used solid-phase dispersion method to improve the dispersion of graphene by reducing the surface energy difference between graphene and metal powder[11]. However, this method may destroy the integrity of graphene in the mixing process, therefore reducing the performance of graphene in metal composites. Liquid phase dispersion mainly involves the dispersion of graphene in the water or organic solvents by ultrasound and the mixture between graphene and liquid metal[12]. Various dispersants are added into the solution to prevent the interactions between graphene sheets, thereby resulting in a relatively stable performance of graphene. However, the conjugated structure of the graphene surface can be damaged by the dispersion methods, which degrades the chemical purity of the composites. In addition, the ultrasound

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may lead to the graphene defects, so the application duration of ultrasound should be strictly controlled. Besides, the large-scale composites can be hardly produced through ultrasound. The solid-liquid method involves the graphene dispersion by ultrasound and the mixture of graphene and metal powder by ball milling[13–14]. This method can improve the dispersion of graphene and the bonding degree between graphene and metal powder, but the process is complex and hard to apply in large-scale production. Therefore, novel methods for the uniform dispersion of graphene in metal composites are urgently required.

The electrical explosion method involves the exertion of a large instantaneous current onto a conductor in a few microseconds. The conductor melts under joule heating. Then the superheated molten particles are further vaporized. The explosive product diffuses rapidly with the shock wave in the medium environment and forms nano-powder in the medium after cooling[15]. Currently, the electrical explosion is a common method for the graphene preparation because of its easy operation and high efficiency. Gao et al[16] obtained the graphene from graphite rods and graphite paper in water by the electrical explosion method. Wang et al[17] prepared the graphene by electrical explosion in a tube under argon atmosphere. After the high-speed explosion, the graphite products collide with the rigid wall, causing the graphite to evenly break off and forming the uniformly dispersed graphene aerosol. The relatively small diameter of graphene prepared by electrical explosion method is conducive to good bonding between the composite matrix and graphene. Thus, the electrical explosion method is quite suitable for graphene reinforcement in the composite materials. The uneven dispersion and weak interface bonding between graphene and metal composite can be solved without destroying the inherent properties of graphene[18].

In this research, the graphene-coated aluminum composite powder was produced by dispersing graphene aerosol which was prepared by electrical explosion. A self-designed device for preparation of graphene-coated aluminum composite powders was developed. Based on the theory of aerosol mechanics, the mixing test of graphene aerosol was carried out with aluminum powder as matrix material. The distribution characteristics of graphene deposition on the surface of aluminum powder were analyzed. The mechanism of in-situ dispersion of graphene aerosol on aluminum composites was analyzed.

1 Experiment

Fig. 1 shows the schematic diagram of preparation process of the graphene-coated metal composite powder. Firstly, the graphite was sent into the explosion-constraint channel, namely detonation tube, which was open at one end and closed at the other end. Two discharge electrodes connected to the pulse energy storage capacitor and introduced large current into the graphite through the gas discharge mode, so the graphite was heated and exploded. In the confined channel, the explosion products of graphite underwent high pressure, high temperature, and shock wave, then changed into graphene, and were sprayed out from the detonation tube. Thus, the uniformly dispersed graphene was obtained and suspended in the protective atmosphere to form the aerosol. By controlling the process of electrical explosion, the specific amount of graphene aerosol was obtained.

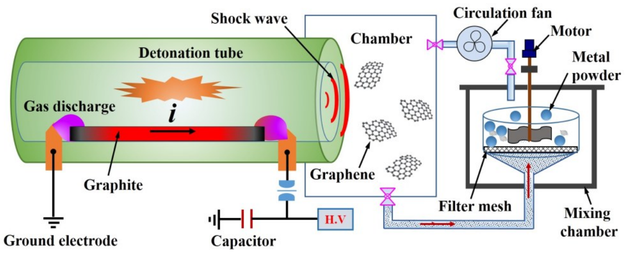

Fig.1 Schematic diagram of preparation process of graphene-coated metal composite powder

The mixed metal powder was evenly spread on the filter mesh in advance, and the size of the filter mesh was smaller than that of the mixed metal particle. After a certain amount of graphene aerosol was obtained in the aerosol chamber, the circulating fan started. Meanwhile, the motor was turned on, and thus the dispersion blades started to stir the metal powder. Graphene aerosol particles passed through the filter along with circulating air flow. The graphene was stably adsorbed on the surface of metal particles by intercept capturing, inertial deposition, and diffusion deposition. The incorporation amount of graphene was calculated by weighing the metal powder before and after the experiment.

The experiment device was similar to that in Ref. [17]. In order to improve the yield of graphene aerosol, the rigid wall was removed. Therefore, the experiment parameters of graphene preparation were optimized in this research. The products were characterized by scanning electron microscope (SEM) coupled with energy dispersive spectrometer (EDS), transmission electron microscope (TEM), atomic force microscope (AFM), and Raman spectroscope. The initial charging voltage was 14 kV, the capacitance was 8.88 μF, and the inner diameter of the detonation tube was 3 mm. Fig.2 shows SEM morphologies of graphite powder and aluminum powder. The size of graphite powder was 23 μm, and the amount of graphite powder for one electric explosion was 8 mg. The graphite powder was pressed into the graphite strips and put into the detonation tube for electrical explosion. Spherical aluminum particles were selected for composite mixing tests. The average particle size of aluminum powder was 15 μm.

After the electrical explosion, the graphene aerosol was uniformly suspended in the argon medium. Various substrates were placed at the chamber bottom, and the graphene was deposited on the substrate surface.

2 Results and Discussion

2.1 Characterization of graphene aerosol

Fig.3 shows SEM morphology of the graphene prepared by

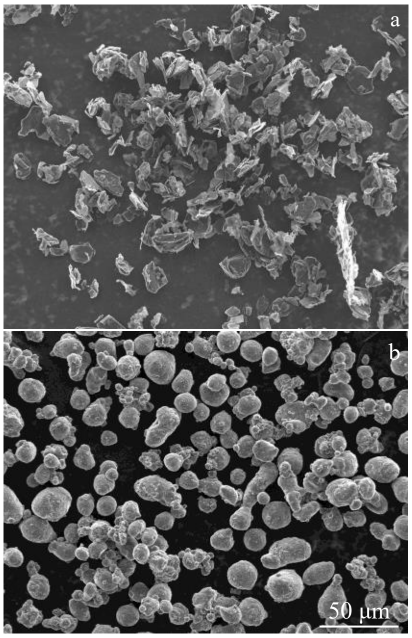

Fig.2 SEM morphologies of graphite powder (a) and aluminum powder (b)

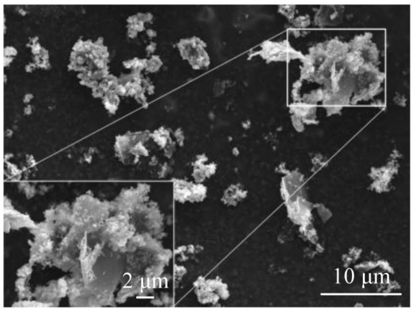

Fig.3 SEM morphology of graphene prepared by electrical explosion

electrical explosion. No large flake graphite can be observed in the graphene after electrical explosion, but a small amount of thin flake and floccule graphene appears. The floccule graphene is agglomerated and partially adheres to the flake graphene, forming the graphene aggregates, which is mainly related to the aerosol characteristics. The aerosol collides with each other, then is agglomerated in the sedimentation process, and finally is deposited on the conductive substrate under the action of gravity.

Fig. 4 shows TEM morphologies of graphene prepared by electrical explosion and the high-resolution transmission electron microscope (HRTEM) was used. Generally, the obtained graphene has two structures: the flake graphene and the gel graphene. The flake graphene (Fig. 4a) has a smooth lamellar structure, and some graphene sheets are bent with obvious folds. The bending and folding structures are mainly caused by the thermodynamic instability of two-dimensional materials. During the electrical explosion, the graphene sheet is bent by the shock wave, resulting in the crimp-lamellar graphene and the agglomeration of graphene sheet. The flake graphene size is 0.1–5.0 μm. As shown in Fig.4b, the stacked layers of graphene can be clearly observed, and the distance between each layer is 0.3–0.4 nm. The number of graphene layers can be identified as 5 by the number of stacked layers at graphene edges. Moreover, the flake graphene in Fig. 4a has good transparency because the obtained graphene is very thin. According to Fig.4c, the small-size graphene is agglomerated, thereby forming the gelatinous aerosol[19].

Fig. 5a shows AFM image of the graphene prepared by electrical explosion. It is found that the graphene size is 0.02–2.00 μm, and the graphene shape is irregular. The relatively large graphene sheets were selected for thickness measurement, as indicated by the blue lines in Fig. 5a. The corresponding thickness distributions of graphene sheet 1 and graphene sheet 2 are shown in Fig. 5b and 5c, respectively. The length of the graphene sheet 1 is about 472 nm and the thickness is 3.6–4.8 nm. The theoretical thickness of single atomic layer of graphite is about 0.34 nm. Therefore, it can be calculated that the graphene sheet 1 has 10–14 graphene layers. The thickness of graphene sheet 2 is 3.9–4.6 nm, inferring that the graphene sheet 2 has 11–13 graphene layers. Because the substrate is not flat and the sampling is random, certain error exists in the measured results.

2.2 Graphene-coated aluminum composites from graphene aerosol

After the electrical explosion, the graphene aerosol particles are constantly in irregular motion: they collide with each other and stick together into large particles. To prevent the

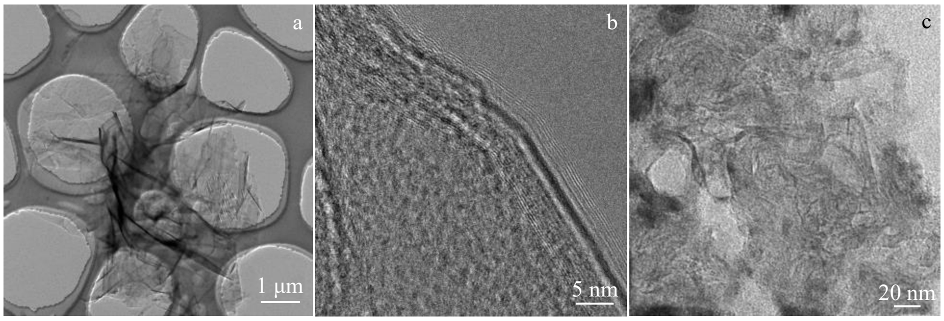

Fig.4 TEM morphologies of flake graphene (a) and gel graphene (c) prepared by electrical explosion; HRTEM morphology of flake graphene (b)

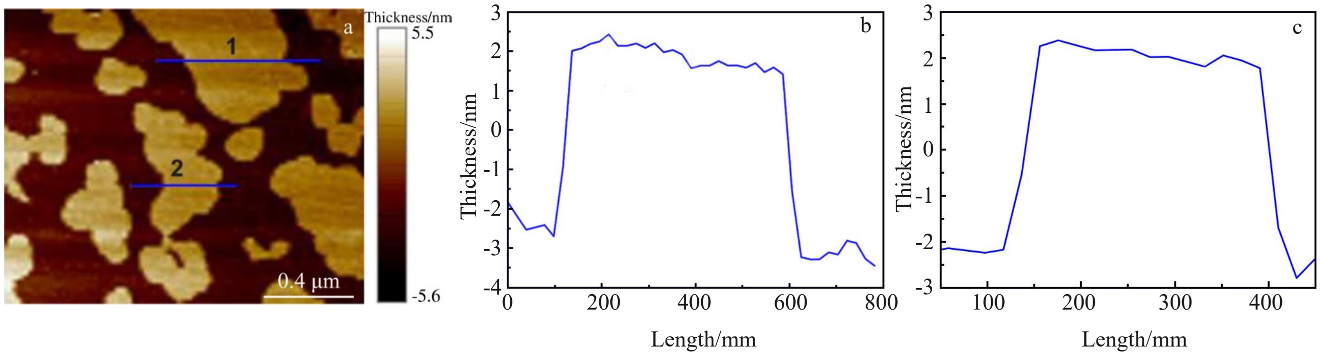

Fig.5 AFM image of graphene (a); thickness distributions of sheet graphene 1 (b) and sheet graphene 2 (c) in Fig.5a

coagulation of graphene aerosol, the mixture of metal powder and graphene aerosol should be conducted immediately after the electrical explosion.

In this research, 5.0wt%, 3.0wt%, and 1.5wt% graphene aerosol was used for in-situ deposition on aluminum composite powder. Fig. 6 shows SEM morphologies of graphene-coated aluminum composite powder after mixture with different graphene contents. It can be seen that some aluminum particles adhere to the graphene sheets, and some graphene sheets with large diameter are not completely wrapped or adhere to other graphene sheets. Some graphene sheets form a connection with the edge of aluminum particles, and others do not adhere to the aluminum particles. When the content of graphene aerosol in the mixture is 5.0wt%, as shown in Fig. 6a, some free graphene sheets exist, indicating that the composite bonding is inferior between the aluminum particle and graphene. In addition, when the graphene content is large (5wt%), the aggregation of graphene in the composite is serious, and the graphene easily sticks to each other due to the uneven dispersion of graphene. When the content of graphene aerosol in the mixture is 3.0wt%, slight agglomeration of graphene occurs, the graphene dispersion is better, and no free graphite sheets can be observed. When the content of graphene aerosol in the mixture is 1.5wt%, no obvious agglomeration of graphene can be observed, and the graphene is dispersed evenly in aluminum composite.

Fig. 7 shows EDS element distributions of the graphene-coated aluminum composite powders after mixture with 1.5wt% graphene aerosol. As shown in Fig.7c, the graphene is well dispersed in the aluminum composite. The graphene with small size can better adhere to the particle surface.

With increasing the graphene aerosol content in the mixture, the aggregation of graphene in aluminum composite powder gradually is more obvious. Therefore, the content of graphene aerosol in aluminum composite powder should be controlled in a suitable range. Gao et al[20] reported the maximum content of graphene for uniform dispersion in aluminum composite powder by ball milling method. When the average diameter of aluminum powder is 13 μm and the number of graphene layers is 8, the maximum content of graphene for uniform dispersion in aluminum composite powder is 0.5wt%. When the graphene content exceeds 0.5wt%, the graphene overlaps on the surface of aluminum powder. Therefore, the maximum content of graphene deposited on the surface of aluminum powder can be calculated by Eq.(1), as follows:

$$\varphi = \frac{4\pi (D/2)^2 t}{\frac{4}{3}\pi (D/2)^3} = \frac{6t}{D} \tag{1}$$

where φ is the critical content of graphene flakes for uniform dispersion in aluminum composite powder (wt%); t is the thickness of graphene sheet (nm); D is the diameter of aluminum particles (nm). Thus, the smaller the diameter of aluminum particles, the more the graphene can be uniformly dispersed on aluminum composite powder. The thickness of graphene sheets prepared by electrical explosion method is about 2.4 nm. Therefore, the maximum content of graphene sheets is only 0.11wt%, which is far less than the content of

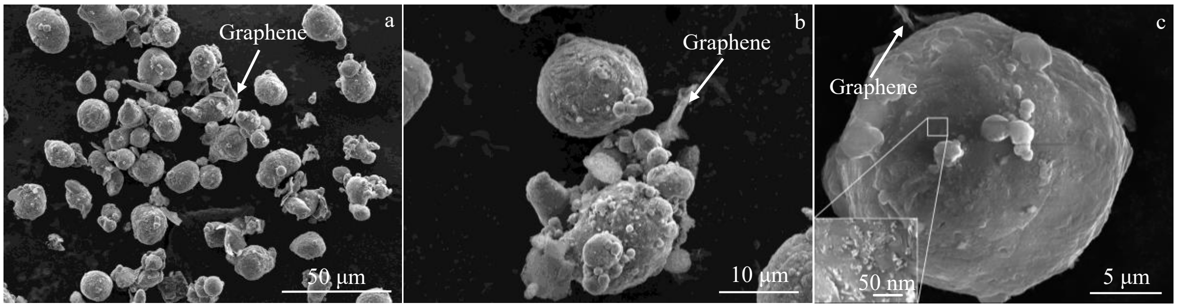

Fig.6 SEM morphologies of graphene-coated aluminum composite powders after mixture with 5.0wt% (a), 3.0wt% (b), and 1.5wt% (c) graphene aerosol

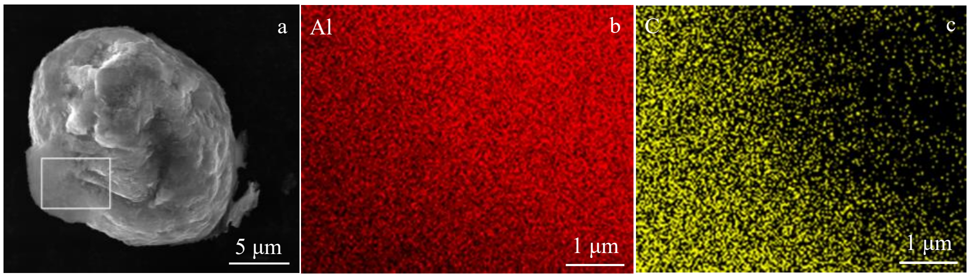

Fig.7 SEM morphology of graphene-coated aluminum composite powders after mixture with 1.5wt% graphene aerosol (a); EDS element distributions of Al (b) and C (c) for rectangular area in Fig.7a

graphene aerosol for the in-situ deposition of graphene on aluminum composite powder.

2.3 Formation mechanism of graphene-coated aluminum composites from graphene aerosol

According to the graphene aerosol characteristics and aerosol dynamics, the mechanism of aerosol mixing to form graphene-coated composite materials is as follows. Firstly, the graphene aerosol flows around the obstacle immersed in the fluid. The particle deviates from the obstacle and accelerates, but the large particles cannot immediately adapt to the local change of airflow velocity. Thus, a velocity difference occurs between the suspended particle and the surrounding gas, and the inertial forces drive the particle to further move. However, the deflected airflow tends to push the particle away from the obstacle. Therefore, the particle motion depends on the competition result between inertial forces and fluid resistance. Most metal particles spread on the filter mesh are placed vertically along the airflow direction. When the airflow passes through, the airflow streamline near the metal particles is bent[21]. Under the action of inertial force, the aerosol particles are deposited on the metal particle surface not along the bent airflow line, as indicated by the moving track of graphene sheet 1 in Fig. 8. With increasing the graphene sheet size and the airflow velocity, the inertial sedimentation effect is also increased.

Secondly, when the graphene aerosol particles pass near the metal particles, they approach the metal particles by the diffusion, inertia, gravity, and electricity. During the stirring process, a large number of electrons are enriched on the surface of aluminum particles along the transfer direction of charge, and some ions can adsorb graphene. The graphene can fully adhere to the aluminum particles and the bonding force is improved, resulting in the graphene-coated aluminum composite powder. As a result, the graphene sheet is captured by the metal particles during the airflow[22], as indicated by the moving track of graphene sheet 2 in Fig.8.

Thirdly, due to the Brownian motion, the motion trajectory of graphene sheet is inconsistent with the airflow direction, and the graphene sheet particles can diffuse to metal particles and then be deposited onto the surface of metal particles, as indicated by the graphene sheet 3 in Fig. 8. The smaller the diameter of the graphene sheet, the more significant the Brownian motion of the graphene sheet in the diffusion process, and the better the diffusion deposition.

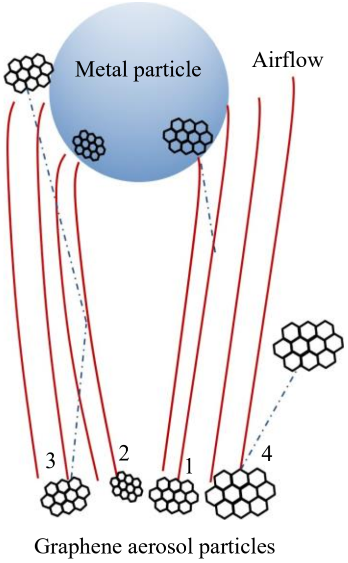

Fig.8 Schematic diagram of formation mechanism of graphene-coated aluminum composites by graphene aerosol

Finally, the graphene aerosol particles have a certain sedimentation rate due to the gravity. Therefore, the trajectory of aerosol particles deviates from the airflow direction, forming free graphene sheets, as indicated by the graphene sheet 4 in Fig.8.

3 Conclusions

1) The graphene prepared by electrical explosion has two types: the flake graphene and the gel graphene. The graphene has 5–8 layers and is uniformly dispersed in the chamber in the form of aerosol. The aerosol graphene easily collides with each other and can be agglomerated after electrical explosion.

2) Graphene sheets with small diameters can easily adhere to the metal particles, while the graphene sheets with large diameters adhere to the edges of metal particles. With increasing the graphene aerosol content in the mixture, the graphene is agglomerated.

3) Graphene aerosols are dispersed on the surface of metal particles by inertial deposition, interception capture, and diffusion deposition.

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电爆法制备石墨烯气溶胶用于石墨烯包覆铝复合粉体

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摘　要：提出了一种通过电爆法制备石墨烯气溶胶并将其用于石墨烯包覆铝复合粉体的新方法，在不破坏石墨烯本征特性的前提下，解决了石墨烯在金属复合粉体中分散不均匀、界面结合弱的问题。利用电爆法制备了一定浓度的石墨烯气溶胶，得到的石墨烯有2种结构：片状和凝胶状。石墨烯层数为5~8层，石墨烯气溶胶均匀分散在腔内。另外，在气流的作用下，通过搅拌将石墨烯气溶胶与球形铝粉颗粒混合，制备了石墨烯含量不同的石墨烯包覆铝复合粉体。当石墨烯气溶胶含量为1.5%（质量分数）时，石墨烯均匀分散在复合材料中，石墨烯片粘附包裹在金属颗粒上。最后，分析了石墨烯气溶胶原位混合形成石墨烯金属复合粉体的机理。

关键词：电爆法；石墨烯气溶胶；气溶胶凝并；复合粉体

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