

Microwave extinction characteristics of nanoparticle aggregates

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Abstract. Structure of nanoparticle aggregates plays an important role in microwave extinction capacity. The diffusion-limited aggregation model (DLA) for fractal growth is utilized to explore the possible structures of nanoparticle aggregates by computer simulation. Based on the discrete dipole approximation (DDA) method, the microwave extinction performance by different nano-carborundum aggregates is numerically analyzed. The effects of the particle quantity, original diameter, fractal structure, as well as orientation on microwave extinction are investigated, and also the extinction characteristics of aggregates are compared with the spherical nanoparticle in the same volume. Numerical results give out that proper aggregation of nanoparticle is beneficial to microwave extinction capacity, and the microwave extinction cross section by aggregated granules is better than that of the spherical solid one in the same volume.

1. Introduction

Absorbing composite coating, prepared by mixing the nano-absorbent and matrix harmoniously, is one of the effective ways to reduce the radar cross section (RCS) of object. While fabricating composite coating, filler granules inevitably collide with each other, and then congregate to aggregates with fractal structure [1, 2]. The scattering and absorption characteristics of nanoparticle aggregates are of great significance for researches on microwave absorption mechanism. Because of nanoparticles aggregating randomly, the actual fractal structure is complicated and changeable, which has a significant impact on the extinction performance of aggregates. It is obvious that the performance cannot be investigated according to the monomer theory [3]. There are lots of researches on clusters of given fractal structure [4-6]. Nevertheless, Studies on extinction characteristics associated with the actual complex structures have been carried on infrequently [7, 8].

The fractal theory is a promising approach for study on extinction performance of nanoparticle clusters. The diffusion-limited aggregation (DLA) is easy and feasible to simulate the self-similar fractal growth by the way of solving simple kinematics and kinetic equations. The fractal structures simulated by this model are in good agreement with the experimental results [9, 10]. According to the rule of three dimensional DLA, under the conditions of nearest neighbour and subordinate neighbour, the fractal aggregates are all satisfied with the scale invariance and self similarity, and have the same fractal dimension (about 2.48) despite their growth in different nearby conditions and different



appearance. It is shown that the fractal dimension of cluster is not associated with the structure of lattice in the small range of particle number. Thus DLA is a kind of potent method for exploring the structure and growth process of clusters. However the interaction between electromagnetic wave and particle aggregates is very complex, the Maxwell Equations often possessing analytic solution can only be solved numerically. The discrete dipole approximation (DDA) is a general method to calculate electromagnetic scattering and absorption of particles of arbitrary geometry and composition. As a good method for numerical solution, the DDA possesses several features, such as simple procedure, high efficient iterative algorithm, fast computing speed, and so on. Therefore, it has been widely utilized to investigate scattering and absorption performance of particles [11-14].

In this paper, the fractal aggregation model DLA is utilized to simulate the possible structures of spherical nanoparticle clusters. Then the impacts of the aggregation states on microwave extinction performance are numerically analyzed based on the DDA. The results would provide profitable theoretical references for the application of nanoparticles in microwave absorbing composites.

2. Theory of discrete dipole approximation

The way of calculating extinction performance of nanoparticle with arbitrary shape by DDA method is as present. The DDA replaces the solid particle by a set of N point dipoles, with the spacing between the dipoles far small than the wavelength. Thus the research on the actual particle is displaced to that on these small dipole groups. It is supposed that the relative permittivity of the solid particle is $\varepsilon_r(\bar{r})$. The position vector of the j electric dipole is \bar{r}_j , and the polarization α_j . The electric dipole moment can be expressed as follow.

$$\bar{P}_j = \alpha_j \bar{E}_j(\bar{r}) \quad (1)$$

Where $\bar{E}_j(\bar{r})$ is the total domain electric field on the dipole at \bar{r}_j , including the electric field of the incident wave and the dipole electric field formed by other dipoles on the location.

$$\bar{E}_j = \bar{E}_0 \exp(i\vec{k} \cdot \bar{r}_j - i\omega t) - \sum_{i \neq j} \bar{A}_{ji} \bar{P}_i \quad (2)$$

Here \bar{E}_0 denotes the field of the incident wave, and $k=\omega/c$ the incident wave vector. The interaction matrix is \bar{A}_{ji} [11].

$$\bar{A}_{ji} \bar{P}_i = \frac{\exp(ikr_{ji})}{r_{ji}^3} \left\{ k^2 \bar{r}_{ji} \times (\bar{r}_{ji} \times \bar{P}_i) + \frac{(1-jkr_{ji})}{r_{ji}^2} [r_{ji}^2 \bar{P}_i - 3r_{ji}(\bar{r}_{ji} \cdot \bar{P}_i)] \right\} \quad (j \neq i) \quad (3)$$

Where $\bar{r}_{ji} = \bar{r}_j - \bar{r}_i$. For a system containing N point dipoles, \bar{A}_{ji} is a $3N \times 3N$ symmetric complex matrix. While $j = i$ and $\bar{A}_{jj} = \alpha_j^{-1}$, Equation of electric dipole moment \bar{P}_j can be gained according to above formulas.

$$\sum_{i=1}^N \bar{A}_{ji} \bar{P}_i = \bar{E}_{inc,j} \quad (j=1, 2, \dots, N) \quad (4)$$

This is a $3N$ -dimension linear complex vector equation, which can be solved by the fast Fourier transform and the complex conjugate gradient technique. For the sake of calculating \bar{P}_j accurately, the appropriate dipolar polarizability α_j should be determined according to the dielectric parameters of the solid particle. In the case of $|m|kd < 1$, the calculating results obtained from the DDA are good agreement with that by other theory based on lattice dispersion relation (LDR) [15]. Thereby the LDR expression is selected as the type of polarization.

$$\alpha_{LDR} = \frac{\alpha_{CMR}}{1 + \frac{\alpha_{CMR}}{d^3} \left[(b_1 + m^2 b_2 + m^2 b_3 \sum_{j=1}^3 (\hat{a}_j \hat{e}_j)^2) (kd)^2 - \frac{2}{3} i (kd)^3 \right]} \quad (5)$$

Where $b_1=-1.8915316$, $b_2=0.1648469$, $b_3=-1.7700004$ and $\alpha_{CMR} = 3d^3(\varepsilon_r - 1)/[4\pi(\varepsilon_r + 2)]$. \hat{a} and \hat{e} presents the unit vector of the incident direction and the direction of the electric field vector, respectively. Here m denotes the complex refractive index of the particle, and the square of m is equal to ε_r .

Once Eq. (4) has been solved for the unknown polarizations \bar{P}_j , the extinction, absorption and scattering cross sections C_{ext} , C_{abs} and C_{sca} can be evaluated as follows [11].

$$C_{ext} = \frac{4\pi k}{|E_0|^2} \sum_{j=1}^N \text{Im}[\bar{E}_{inc,j}^* \cdot \bar{P}_j] \quad (6)$$

$$C_{abs} = \frac{4\pi k}{|E_0|^2} \sum_{j=1}^N \left\{ \text{Im}[\bar{P}_j \cdot (\alpha_j^{-1})^* \cdot \bar{P}_j^*] - \frac{2}{3} k^3 |\bar{P}_j|^2 \right\} \quad (7)$$

$$C_{sca} = \frac{k^4}{|E_{inc}|^2} \int d\Omega \left| \sum_{j=1}^N [\bar{P}_j - \hat{n}(\hat{n} \cdot \bar{P}_j)] \exp(-ik\hat{n} \cdot \bar{r}_j) \right|^2 \quad (8)$$

Where \hat{n} denotes the unit vector of the scattering direction, and $d\Omega$ the solid angle.

Based on aggregation structure generated by the DLA, the extinction capacity of radar wave by nanoparticle aggregates could be investigated by the available computer code DDSCAT [16]. Here the source program has been re-compiled and added the parts of results processing and output.

3. Results and discussion

Taking the nano-carborundum for an example, the influences of the particle number, original diameter, fractal structure and orientation on microwave extinction characteristics are investigated. Here the relative permittivity of SiC is cited from reference [17], namely $\varepsilon_r = \varepsilon_\infty + (\varepsilon_0 - \varepsilon_\infty) \cdot [1 + (j2\pi f\tau)^a]^{-1}$.

Where f presents electromagnetic frequency, $\varepsilon_0=82.4$, $\varepsilon_\infty=4.1$, $\tau=5.9 \times 10^{-11}$ and $a=0.52$. Because the way of attenuating electromagnetic wave is due to the joint action by plenty of granules in composite, the calculated extinction parameters are replaced by the corresponding ones per unit volume, which makes the calculated results more reasonable.

3.1. Different extinction performance between aggregates and solid particle with the equal volume

Extinction performance of two different nano-carborundum aggregates has been compared with that of spherical solid particle in the equal volume. Wherein the number of granules is $N=100$ with particle diameter $a=80\text{nm}$ in one aggregates, and $N=50$ with $a=80\text{nm}$ in another ones. Fractal structure of aggregates is generated by the DLA. Relationship between extinction performance of the three scatters and the incident wave frequency has been investigated by the DDA. The numerical results are shown in figure 1. As can be seen from the graphs, extinction performance of nanoparticle aggregates is much better than that of the solid one in the same volume.

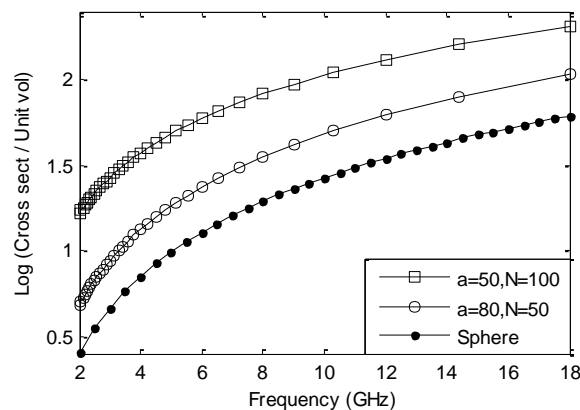


Figure 1. Extinction performance of aggregates and solid particles with the equal volume

3.2. Impact of original particle size on extinction performance

No matter what method is utilized to fabricate nanometer granules, the particle size of original granules is hardly single, but a certain distribution. So it is necessary to investigate the impact of original diameter on microwave extinction. Here the quantity of nano-carborundum in clusters is supposed as $N=100$. Figure 2 presents the relationship between extinction performance and original particle size in aggregates at different frequency. It is obvious that extinction performance of aggregates has nothing to do with the original particle size in the range of radar wave band. This conclusion is similar to that of single nanoparticle. It is worth to note that the result is drawn from the case of unit volume of particle.

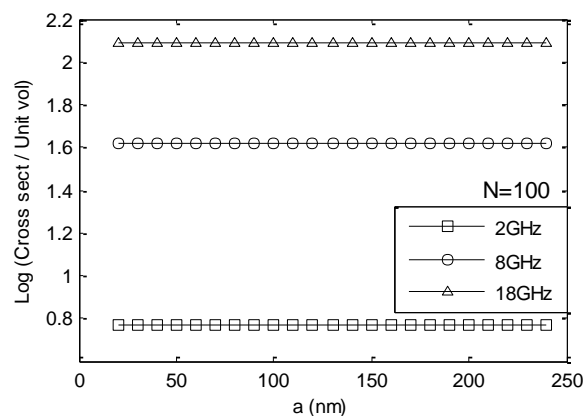


Figure 2. Relationship between extinction performance and original particle size in aggregates

3.3. Impact of particle quantity in aggregates on extinction performance

In general, particle quantity in aggregates is random, ranging from a few to more than thousands. Here the particle diameter is supposed as $a=50\text{nm}$. Figure 3 displays the relationship between extinction performance and particle quantity in aggregates. When the number of aggregated granules is less than 60, extinction cross section increases along with enhancing the particle quantity. When the number of aggregated granules is more than 70, extinction cross section has little attenuation while increasing the particle quantity. Too much number of aggregated particles would cause that aggregates develop to excessive large volume and the absorbent particles distribute unsymmetrical in composite coating, which may induce high conductivity and reflectivity locally. Therefore, proper aggregation of nanometer granules is beneficial to microwave extinction performance.

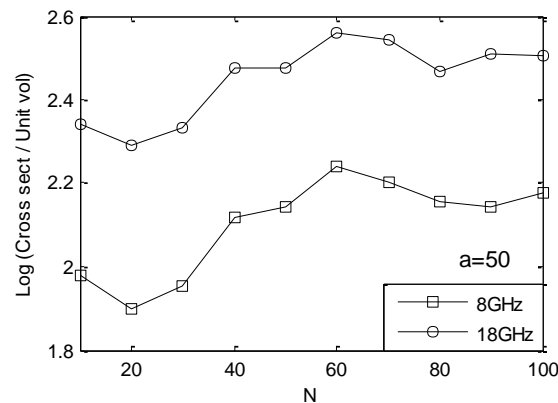


Figure 3. Relationship between extinction performance and particle quantity in aggregates

3.4. Impact of aggregate fractal structure and orientation on extinction performance

Figure 4 presents three different fractal structures with 100 aggregated nano-carborundum particles. Here it is supposed that Θ denotes the angle between the wave vector and the unit vector along with the principal axis of inertia. Figure 5 displays the relationship between extinction cross section and the angle at 12GHz. Numerical results indicate that diverse fractal structures lead to different extinction capacity even though these aggregates contains equal number of particles. With the increase of the angle between the incident wave and the principal axis of inertia of aggregates, extinction performance of each aggregates exhibits similar trend. When the direction of the principal axis of inertia of aggregates approaches parallels to that of incident wave, extinction capacity falls into decline. And the vertical relationship between the two directions would lead to the best extinction performance.

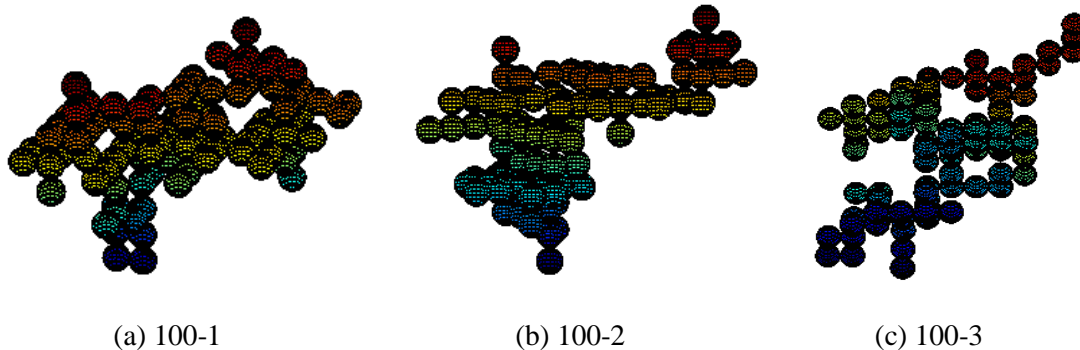


Figure 4. Fractal structures of nanoparticle aggregates at $N=100$

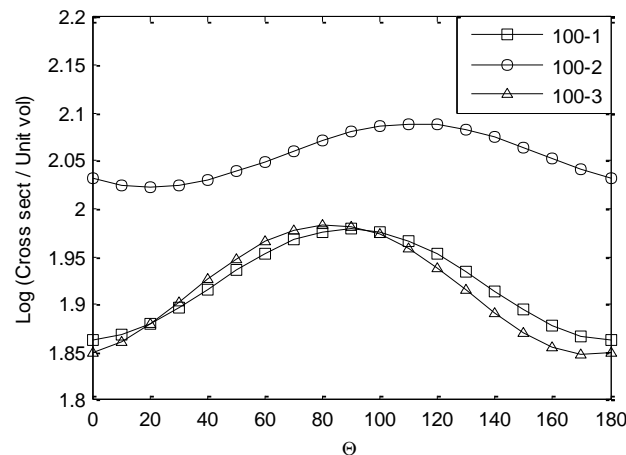


Figure 5. Relationship between extinction performance and the angle of incident wave

4. Conclusions

The DLA model for fractal growth is utilized to investigate nanoparticle aggregates. Possible structures of nanoparticle clusters are generated by computer simulation. Based on the DDA method, microwave extinction characteristics of different nano-carborundum aggregates are numerically analyzed. The analysis of calculated data shows that microwave extinction capacity of nanoparticle aggregates is better than that of spherical solid one in the same volume. Extinction performance of aggregates is associated with the quantity of granules but independent on the original particle size in it. Proper aggregation of nanoparticle is beneficial to microwave extinction capacity. Given identical original particle size and granule quantity, different fractal structure and orientation relative to the incident wave would result in different extinction performance. When the direction of the principal axis of inertia of aggregates approaches vertical to that of incident wave, extinction capacity is the best. And the parallel relationship between the two directions will lead to the worst extinction performance.

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