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Research paper

Theoretical study on effective thermal conductivity of salt/expanded graphite composite material by using fractal method

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HIGHLIGHTS

- Study on effective thermal conductivity of salt/EG composite by a fractal method.
- Multiple three-phase fractal units based on Serpinski carpet form a fractal model.
- Using this model gives a description of microstructures of salt/EG composite.
- Turning fractal units into resistance networks can predict thermal conductivity.
- Comparisons between model results and experimental data show a good agreement.

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ABSTRACT

Fractal geometry has the advantage of describing the macroscopic behaviors of complex systems. Based on statistical self-similar theory, a theoretical study on the effective thermal conductivity of salt/expanded graphite (EG) composite material is conducted by a fractal approach. To allow the demonstration of more microstructural details, multiple fractal units which can represent three-phase media are employed to describe the complex microstructures of the composite. Turning these units into corresponding thermal resistance networks gives a set of predicted effective thermal conductivities that are fully comparable with the existing experimental results. Moreover, by use of this approach, factors that affect the heat conduction performance of the composite are analyzed, and it is found that the quantity of EG plays a dominant role, whereas the distribution of EG needs further attentions.

1. Introduction

Salt/expanded graphite (EG) composite is a potential phase change material (PCM) for thermal energy storage. Owing to its large heat storage capacity, excellent heat transfer ability and low investment cost [1,2], this kind of materials is of great interest in many fields. The European project DISTOR has conducted several researches on the development of PCM-storage system integrated with $NaNO_3/KNO_3$-EG composite for solar thermal power plants [3,4]. A series of nitrates/EG composites were prepared by Lopez et al. [5,6] and introduced into waste heat recovering during industrial processes. Study on thermal properties of the salt/EG composite has great significance in improving the heat storage/release performances of the composite. The thermal conductivity, which is an important thermal physical parameter, therefore, acts as a vital factor for these applications.

Over the past decade, the effective thermal conductivities of various salt/EG composites have been extensively studied by many researchers. Techniques such as transient hot-wire, transient plane source and flash laser become indispensable for experimental determinations [5,7,8]. Also, theoretical thermal conductivity predictions to interpret the experimental results are developed. According to the report from Xiao and his coworkers [9], "quadratic parallel" (QP) model is adopted to calculate the effective thermal conductivities of nitrates/EG composites in original powder state. The structures of alkali nitrate salt/EG composite fabricated by infiltration are idealized as simple laminates and treated with parallel and series models for estimation of the effective thermal conductivity [7]. Realistically, however, the salt/EG composites are multi-phase complex materials [10]. Their overall heat transfer performances are influenced by the thermal conductivities, volume fractions and distributions of all

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components forming the materials. The QP model, which is one of the weighted average models [11,12], only considers the former two factors. The effects of the real size and distribution of each component are not discussed in depth. The laminate models take the microstructure related effects into consideration. But their applications are limited to the infiltrated composites with layered structures. When it comes to the salt/EG composites prepared by a simple physical mixing method, none of the above mentioned models can predict the effective thermal conductivity effectively without a proper analysis of the disordered and irregular microstructures of these composites.

Fractal is a word proposed by Mandeldort [13] to describe the objects that exhibit repeating patterns at different scales. Because of the discoveries that various aspects of microstructures of materials can be considered to be fractals, fractal theory is applicable to predict the overall macroscopic behaviors of the complex materials. Pitchumani et al. [14] are the first to adopt the fractal theory in the research of the effective thermal conductivity of unidirectional fibrous composites. Then, Yu et al. [15,16] and Kou et al. [17] use different methods based on the fractal theory to determine the effective thermal conductivities of saturated/unsaturated porous soils, and the results agree well with the existing experimental data [18,19]. Utilizations of fractal models also are successfully extended to the analysis of the effective thermal conductivities of liquid with nanoparticles [20], wood perpendicular to fibres [21], biological media embedded with randomly distributed vascular trees [22], insulating concrete [23], porous fibrous media [24] and polyurethane [25]. Such recent progresses in applications of the fractal theory open a route toward the development of a novel and better method for calculation of the effective thermal conductivity of the salt/EG composite.

In this work, we try to predict the effective thermal conductivity of the salt/EG composite through a fractal approach. Firstly, the distribution patterns of the multiple phases in the composite material are analyzed by use of the fractal theory. Then a fractal model based on the combination of different three-phase fractal units is established. Secondly, thermal resistance method is introduced to calculate the effective thermal conductivity of this fractal model. Finally, the results obtained from the present model are compared with the data from both the experiments and the QP model.

2. Microstructure of the salt/EG composite

2.1. Experimental observation

Before applying the fractal concept to the studies on the salt/EG composite, it is necessary to analyze the material's microstructures. As a member of multi-phase complex materials, the composite generally consists of salt crystals, graphite layers and void space. For simplicity, the latter two constituents are considered as a porous EG matrix, and the composite, thus, becomes a mixture of the salt particles and this matrix. Fig. 1 displays the microstructures of $LiNO_3$/KCl-EG composite as an example. This kind of the salt/EG composite materials was prepared in our previous work [8] and the pictures were taken by a scanning electron microscope (SEM, S-3700N, Hitachi Inc., JNP). In order to distinguish the inorganic salt and extra-salt parts, we use white arrows to point out the salt particles.

As it is seen in Fig. 1(a), the numerous salt crystals with different sizes and shapes disperse randomly in the porous EG matrix with a magnification of 1500 times. When this material is magnified 500 times, the random distribution patterns of the salt particles with bigger size formed by several or tens of salt crystals are observed. Fig. 1(c) is the image at the magnification of 40. The composite particles also exhibit a disordered and irregular character. Such similar phenomena found in the salt/EG composite material when different length scales are chosen imply the self-similarity of the fractals from a statistical point of view. Therefore, using self-similar scaling laws to predict the properties of the salt/EG composite is possible in our case.

2.2. Fractal analysis

It is known that statistical fractals only show self-similar characteristics in a certain range of length scale. The statistical self-similar regions in the salt/EG composite have lower and upper limits. Assuming that the minimum and maximum diameters of the particles in a self-similar region are represented by λ_{min} and λ_{max}, the value of $\lambda_{min}/\lambda_{max}$ indicates the range of length scale where statistical self-similarity exists. Based on Yu's studies on the complex materials [26], Eq. (1) gives the relationship among the fractal dimension D_f, volume fraction of the porous EG matrix φ_m and $\lambda_{min}/\lambda_{max}$ for statistical self-similar regions in the salt/EG composite

$$D_f = 2 - \frac{\ln \varphi_m}{\ln(\lambda_{min}/\lambda_{max})}. \tag{1}$$

The corresponding curves of D_f versus φ_m at different values of $\lambda_{min}/\lambda_{max}$ are also depicted in Fig. 2. With the rise of the volume fraction of the porous EG matrix, the fractal dimension exhibits an increasing trend. The value of D_f will tend to a maximum as φ_m reaches the value of 1. In the same figure, an impact that $\lambda_{min}/\lambda_{max}$ has made on the D_f-φ_m curve should be noticed. Because many materials are characterized by the presence of more than one statistical self-similar region [23,27], taking all the self-similar regions with different lower and upper limits into consideration will result in different fractal dimensions for modeling use. This circumstance makes fully displaying the composite's whole microstructures difficult when a single simple fractal unit is adopted.

To obtain a better fractal description for the complex materials, recently, the employment of multiple fractal units is found to be a useful approach. In Pia's models, it is assumed that the multiphase complex material is a mixture of several independent two-phase porous components. Each component shows self-similarity in a unique fractal region and can be described by a fractal unit. Therefore, combining several fractal units together can simulate the whole structures of the complex material. By adopting such assumption, two different fractal units representing cement matrix and wooden aggregate are employed to reproduce the same microstructures of insulating concrete [23]. An intermingled fractal units model also is established in Pia's later work to describe the same type of complex systems [27]. Besides, another method to analyze the multi-phase complex material by using different fractal units is to divide the structures of the multiphase system into two or more self-similar regions directly. Assuming that each region where the multiple phases distribute can be represented by a unique fractal unit, the whole structures of the complex material can be described by combining these fractal units together. Nevertheless, this approach only is applicable to two-phase systems because challenges still remain with respect to extending a fractal unit to multi-phase cases. In the present work, we attempt to employ multiple fractal units to model the salt/EG composite material with the latter method. By introducing an analogy between the salt/EG composite and the unsaturated soil based on Ma's models [18], a three-phase fractal unit of the type Serpinski carpet is formed. The detailed description of the fractal model for the salt/EG composite will be given in Section 3.