Enhancing the effective thermal conductivity of liquid with dilute suspensions of nano particles[†]

Bu-Xuan Wang[1*], Le-Ping Zhou[1], Xiao-Feng Peng[1], Xin-Xin Zhang[2]

1. Thermal Engineering Department, Tsinghua University, Beijing, 100084, China

2. Thermal Engineering Department, Beijing University of Science and Technology, Beijing, 100083, China

Abstract

The effective thermal conductivity of dilute CuO nanoparticle suspensions in liquid, with particle mass fraction below 2%, or volumetric fraction much lower than 0.5%, to avoid the possible direct contact of particles, was measured with designed equipment. At the same time, SDBS (sodium dodecyl benzene sulphonate), with mass fraction of 2%, was added as surfactant to the host liquid for further improving the distribution of nanoparticles. With the effective medium approximation and the fractal theory for describing nanoparticle clusters and their size distribution, a predicting cluster method is proposed, and analysis will be presented to predict the effect of clustering particle size and surface adsorption on effective thermal conductivity of liquid.

Keywords: effective thermal conductivity; nanoparticle suspension; interfacial surface adsorption; size effect; cluster; fractal

1. Introduction

The effect of particle inclusions on the effective thermal conductivity of liquid has attracted more and more interest experimentally and theoretically since J.C. Maxwell proposed his fundamental idea in 1873 [1]. However, solid particles may settle out of the suspensions and deposit on heating / cooling wall surface. Recently, as the development of nanotechnology, a novel approach with nano-sized particles in suspensions was proposed and terminologized specially as “nanofluid” by S.U.S. Choi [2] of Argonne National Laboratory of USA in 1995. Several explanations of enhancing thermal conductivity of nanoparticles suspensions have been summarized by Keblinski et al. [3] in 2002. They concluded that the enhancement, especially anomalous increase with metallic nanoparticles inclusion, could be contributed mainly to the liquid laying at liquid / particle interface and the effect of nanoparticles clustering. However, their analyses need to be quantified from experimental data.

Our recent experiments also show the effective thermal conductivity enhancement of ethanol with 25nm SiO_2 particles inclusions, and the clustering of particles into percolating pattern was observed through STM (scanning tunnel microscopic) photos [4]. It was believed that clustering could prominently affect the enhancement. As the measurement was made by thermal-probe method, the effect of liquid convection is hard to be avoided. So, we conducted new experiment based on quasi-steady state method to exclude the effect of local convection [5]. 50nm CuO particle inclusions, with mass fraction below 2%, or volumetric fraction lower than 0.5% suspended in deionized water, was used as the testing medium to avoid the possible direct-contact particle clustering. SDBS (sodium dodecyl benzene sulphonate), with mass fraction of 2%, was added as surfactant, to further improve the distribution of particles in deionized water. As we reported previously, to consider clustering effect would be necessary for modeling the effective thermal conductivity of nanoparticles’ suspensions.

The fractal theory was proposed by Mandelbrot [6], by which Pitchumani et al. [7] firstly researched the effective thermal conductivity of unidirectional fibrous composites. Yu et al. [8~9] obtained a fractal description of effective dielectric coefficient of composite material using effective medium approximation and fractal theory. But as we know, few in open literatures reports to use the fractal theory in describing the cluster of nanoparticle suspensions to predict the effective thermal conductivity.

[†] Paper submitted to be presented at 15th Symposium on Thermophysical Properties.

[*] Corresponding author. Email addresses: bxwang@mail.tsinghua.edu.cn.

In this paper, we will introduce briefly the effective medium theory and the concept of fractal dimension for nanoparticle clusters in liquid, and try to establish a fractal model for predicting the effective thermal conductivity of nanoparticle suspensions with the spatial description of clusters and the consideration of particle size and surface adsorption effect.

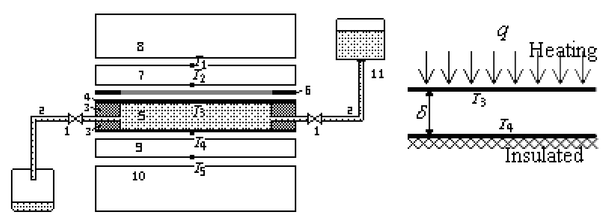
2. Experiment

The experiments were conducted on an apparatus shown in Fig. 1, which was specially designed to suit the quasi-steady condition and could simultaneously measure thermal conductivity and specific heat of testing material in principle. The testing suspension is kept in its original uniform temperature, T_0, before being heated. The analytical solution is given by Carslaw and Jaeger [10] as

$$k = q\delta/(2\Delta T), \quad (1)$$

$$c_p = q/[\rho l(dt/d\tau)], \quad (2)$$

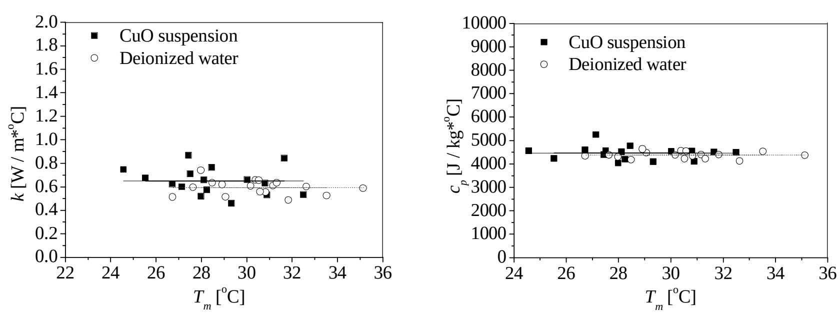
where k is effective thermal conductivity of liquid with particle inclusions, c_p is effective specific heat of the suspension, q is the constant heat flux from the heating surface, δ is thickness of sample, $\Delta T = (T_3 - T_4)$ is the temperature difference between heating surface and insulated bottom surface at quasi-steady state, corresponding to Fourior number greater than 0.55. The cylindrical container for testing medium is 160mm inside diameter and 9mm deep. The Rayleigh number, Ra, of testing medium is controlled less than 10^3, so that liquid convection could be actually neglected. The estimated uncertainty for measured value of k and c_p are $\pm 2.9\%$ and $\pm 3.8\%$, respectively. The testing liquid with CuO nanoparticles inclusion, $\phi > 0$, were prepared by applying supersonic wave for long-time and no visible sediment was found. Besides, we added SDBS (sodium dodecyl benzene sulphonate), 2% by mass fraction, as the surfactant to further improve the distribution of particles in deionized water, and thus, to avoid the direct contact of CuO particles.



1.valve 2.ducting tube 3.supporter (insulator) 4.aluminum sheet 5.sample liquid

6.plane heater 7.9.heat-loss measuring layer 8.10.insulator 11.reservoir

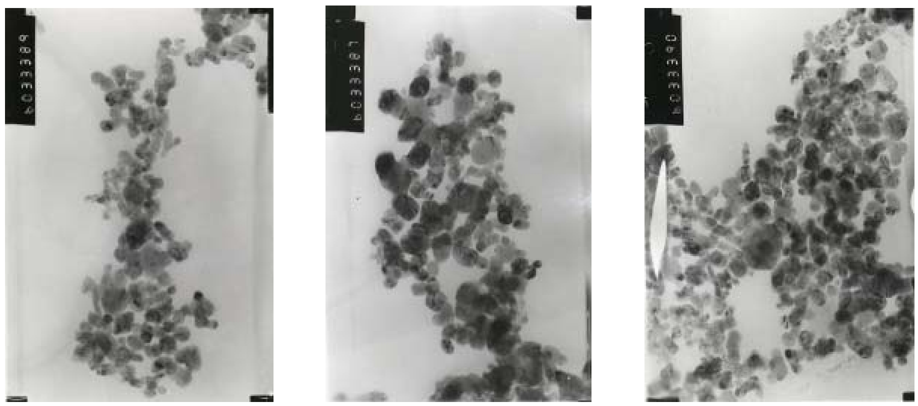
Fig. 1. Measuring apparatus



(a) effective thermal conductivity (b) effective specific heat

Fig.2. Measured value of deionized water and CuO nanoparticle suspension

Our experimental set-up checked well with measurements for thermal conductivity and specific heat of deionized water and ethanol, i.e., $\phi = 0$, at temperature around 300K. Fig.2 shows the measured effective thermal conductivity and effective specific heat for deionized water and CuO nanoparticle suspension with mass fraction of 0.5%, where T_m is mean temperature of suspension, and the solid line and dash line stand for mean value of corresponding properties of suspension and water, respectively. The other results for suspensions with different mass fraction of CuO nanoparticle also exhibit the same trend and are not listed here any more.



(a) mass fraction of nanoparticles, 0.02% (b) 0.04% (c) 0.06%

Fig.3. TEM photos of testing medium

The electron microscopic photos of suspensions, with 50nm CuO particle inclusions (mass fraction: 0.02%, 0.04% and 0.06%, corresponding to volume fraction: 0.13%, 0.25% and 0.38%, respectively), are also photographed and shown in Fig. 3. As can be seen, particle clustering may happen even for low concentration of nanoparticles in liquid. The clustering effect on effective thermal conductivity will be considered later in section 5. Meanwhile, we should consider the clustering effect on effective thermal conductivity only, since it may not affect the value of specific heat which is a thermodynamic property.

3. Basics for theoretical analysis

In effective medium theory, the Maxwell-Garnett's self-consistent approximation (MG) model [1] and the Bruggeman approach [11] were commonly used to treat the effective transport coefficient of mixture and composites. The MG model fits well with experimental data where dilute and randomly distributed components are included in a homogeneous host medium, and the particles are considered to be isolated in the host medium, without interactions existed among them. For two-component entity of spherical-particle suspensions, the MG model [1] gives

$$\frac{k}{k_f} = \frac{(1-\phi)(k_p + 2k_f) + 3\phi k_p}{(1-\phi)(k_p + 2k_f) + 3\phi k_f}, \tag{3}$$

where k_f is thermal conductivity of host medium, k_p is thermal conductivity of particle, and ϕ is volume fraction of particles. The MG model is applicable to suspensions with low-concentration particle inclusions.

The Bruggeman model with mean field approach is used to analysis the interactions among the randomly distributed. For a binary mixture of homogeneous spherical inclusions, the Bruggeman model [11] gives

$$\phi\left(\frac{k_p - k}{k_p + 2k}\right) + (1-\phi)\left(\frac{k_f - k}{k_f + 2k}\right) = 0, \tag{4}$$

and the solution of above quadratic equation is given as:

$$k = (3\phi - 1)k_p + [3(1-\phi) - 1)]k_f + \sqrt{\Delta}, \tag{5}$$

$$\Delta = (3\phi - 1)^2 k_p^2 + [3(1-\phi) - 1)]^2 k_f^2 + 2[2 + 9\phi(1-\phi)]k_p k_f \,. \quad (6)$$

The Bruggeman model has no limitation on the concentration of inclusions, and can be used for particle percolation in suspensions. For low particle-concentration suspension, the Bruggeman model shows almost the same result as the MG model will give. For a particle percolation situation or when the particle concentration is sufficiently high, the MG model fails to predict precisely the experimental results, while the Bruggeman model can still fits well with experimental data.

In addition to MG model and Bruggeman model, many other models that extended these two models were proposed. The three-component Core-Shell-Medium (CSM) model [12] deduced from the MG approximation has considered the adsorption process on the particle surface. The Rayleigh model [13] did not concern the effect of particle interaction, but for particle of small radius, its accuracy is relatively higher than MG model. The Cichocki-Felderhof (CF) model [14] was deduced with statistical method and considered the interaction between particles of same radius. The Monecke model [15] discarded the physical topology technique of effective medium theory, deduced on the assumption that the effective thermal conductivity equals to an interpolation between the extreme limits of its components.

Table 1. Comparison of the calculated value of k / k_f using various models

Particle volume fraction, %	MG model[1]	CSM model[12]	Rayleigh model[13]	CF model[14]	Monecke model[15]	Experimental results[5]
0.1	1.00262	1.00192	1.00262	1.00266	1.00262	1.0982
0.2	1.00526	1.00386	1.00524	1.00539	1.00525	1.1252
0.3	1.00791	1.00582	1.00787	1.0082	1.00788	1.13984
0.4	1.01057	1.00781	1.01051	1.01108	1.01051	1.16996
0.5	1.01324	1.00982	1.01314	1.01404	1.01316	1.11238
0.6	1.01593	1.01185	1.01579	1.01708	1.01581	1.10531

We compare these models with experimental results for suspension of CuO nanoparticles (50nm) in deionized water [5] in Table 1. All these models function as the same in dilute limit, yet none of them explains well with our experimental data [5].

4. The particle size effect and particle surface adsorption

To explain the anomalous increase of effective thermal conductivity for liquid with nanoparticle inclusions, the effect of particle size and particle surface adsorption of liquid need to be considered [3]. Without consideration of radiation, the heat carriers in nanoparticles include only phonons and electrons. The transport regimes for these heat carriers were established by Chen [16], according to the relation between mean free path of heat carriers and length scale of nanostructures. When the mean free path of heat carriers is comparable with the size of nanoparticles, i.e., 10~100nm, the Boltzmann equation is applicable for describing the heat transferring process. Hence, using the relaxation time approximation method [17], the effective thermal conductivity of nonmetallic nanoparticles can be approximated as

$$k_p = \frac{3a^*/4}{3a^*/4+1} k_b \,, \quad (7)$$

where k_b is (bulk) thermal conductivity of particle, $a^* = a/l$ is the nondimensional radius, and l is mean free path of phonons. For metallic nanoparticles, the effective thermal conductivity can be achieved, provided that the Wiedemann-Franz Law still holds when temperature is much higher than Debye temperature. The size effect on the phonon-electron coupling factor is also negligible within the above-mentioned regime [18]. A cubic decreasing law was found in effective electric conductivity for particles smaller than 500nm [19]. Thus, when the relaxation times of electron and phonon are comparable, the following equation can be used for effective thermal conductivity of metallic nanoparticles:

$$k_p = \left(\frac{2a}{5\cdot 10^{-6}}\right)^3 k_b \cdot \tag{8}$$

Besides, the adsorption of liquid molecules on particle surface is thought to be a monolayer one. The way of molecule allocation on particle surface is commonly considered to be a hexagonal closed-packed style. From the Langmuir formula of monolayer adsorption of molecules, the thickness of the adsorption layer can be expressed as [20]

$$t = \frac{1}{\sqrt{3}}\left(\frac{4M}{\rho_f N_A}\right)^{1/3}, \tag{9}$$

where M is molecular weight of liquid, ρ_f is density of liquid, and N_A is Avogadro constant (6.023× 10^{23}/mol). Since the monolayer always occurs in conjunction with the particle sphere, they are completely correlated [21], and hence, the effective thermal conductivity of the nanoparticle can be considered to be the total thermal conductivity of these two substances [22]:

$$k_{cp} = k_{ad}\,\frac{(k_p + 2k_{ad}) + 2A^3(k_p - k_{ad})}{(k_p + 2k_{ad}) - A^3(k_p - k_{ad})}, \tag{10}$$

where $A = 1 - t/(t+a)$, k_{ad} is effective thermal conductivity of the adsorption layer. With the consideration of surface adsorption, we should substitute (a+t), $[(a+t)/a]^3\phi$ and k_{cp} for a, ϕ and k_p, respectively, in Eqs. (3)~(10). The value of k_{ad} is hard to be predicted, but from Eq. (10), we can take k_{cp} as first approximation, and thus the calculated results will stand for the upper bound of enhancement for effective thermal conductivity of liquid with nanoparticles inclusion.

Table 2. Comparison of MG model, extended MG model and experimental results

Particle volume fraction, %	MG model[1]	Extended MG model [22]	Experimental results[5]
0.1	1.003	1.004	1.0982
0.2	1.006	1.00799	1.1252
0.3	1.009	1.01199	1.13984
0.4	1.012	1.01599	1.16996
0.5	1.015	1.01998	1.11238
0.6	1.018	1.02398	1.10531

Table 3. Data for calculation

Copper oxide		Deionized water	
Average radius	a=25nm	Thickness of adsorption monolayer	t=2.8nm
Mean free path of phonons	l=14nm		
Density	ρ_p=6310kg/m^3	Density	ρ_f=996kg/m^3
Thermal conductivity	k_p=32.9W/m/K	Thermal conductivity	k_f=0.613W/m/K

The extended MG model, with consideration of particle size effect and particle surface adsorption of liquid, can thus be deduced using the above equations [22]. The corresponding calculated results are compared with MG model and experimental results, and are listed in Table 2. Data used for calculation are listed in Table 3. However, the extended MG model that considers both effects improves little from MG model and the need for further consideration of particle clustering still remains to explain our experimental results.

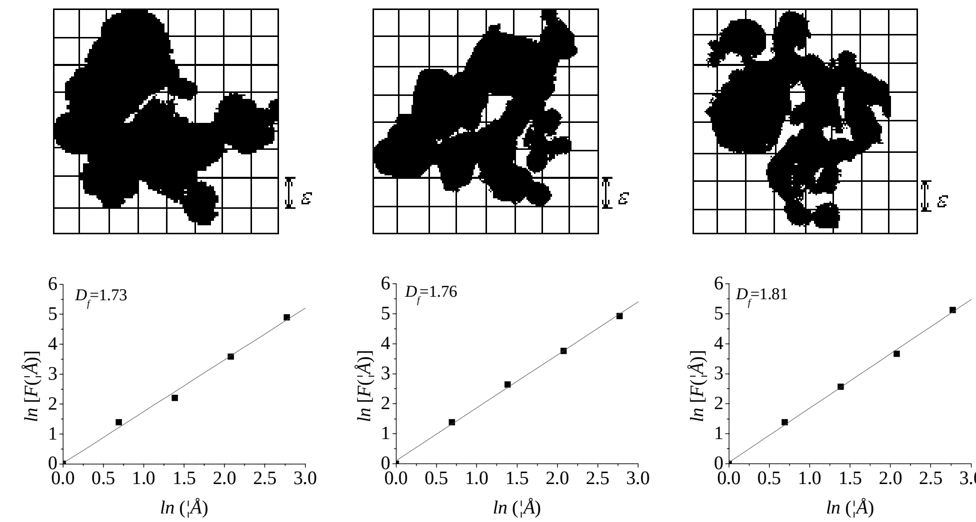
5. A fractal model proposed

To incorporate clustering effect of nanoparticles, a fractal description for cluster distribution is used to predict the effective thermal conductivity of nanoparticle clusters that show some kind of self-comparability in suspensions. Then, with spatial distribution of nanoparticle clusters, the effective thermal conductivity of suspensions is expected to be obtained, using the multi-component MG model proposed by Wood and Ashcroft [23].

As Havlin and Ben-Avraham [24] have figured out, the radius distribution of nanoparticles and the spatial distribution of nanoparticles in suspension have both shown some kind of self-comparability. In scaling theory, the fractal dimension, D_f, can be used for describing this character, or fractals. It is established through a scalar with unit ε. If the volume (area, particle numbers, etc.) of the fractal is $F(\varepsilon)$, then D_f can be decided through the following expression:

$$F(\varepsilon) = C\varepsilon^{D_f}, \tag{11}$$

where C is a shape factor that is independent of ε. The section views of intercepted nanoparticles clusters and the calculation of fractal dimensions are shown respectively on the top and at the bottom of Fig.4. Using Eq. (11), the fractal dimensions of corresponding clusters can be obtained to be 1.73, 1.76 and 1.81, respectively.



(a) Mass fraction=0.02% (b) Mass fraction=0.04% (c) Mass fraction=0.06%

Fig.4. The calculation of fractal dimension of section area of clusters

The enhancement in effective thermal conductivity for liquid with nanoparticle inclusions relates directly with the particle interaction and clustering process. It is reasonable to consider that host liquid and inclusions of percolating cluster compose the nanoparticle suspension. Thus, when using Eq. (1), k_p will be replaced by the effective thermal conductivity of nanoparticle clusters, $k_{cl}(r)$, predicted by Bruggeman model.

Provided that different sizes of clusters, r, have formed in suspension due to the interaction of nanoparticles of equal radius, a, the volume fraction of nanoparticles in cluster can be obtained from fractal theory [25,26]:

$$f(r) = (r/a)^{D_f - 3}, \tag{12}$$

Since particles percolated in liquid, the Bruggeman approach [11], with substituting $f(r)$ for ϕ into Eqs. (4) and (5), can be used to calculate the effective thermal conductivity of clusters, $k_{cl} = k_{cl}(r)$.

For isolated clusters, their spatial distribution can be approximated through statistical method. When particle volume can be expressed as $V = Hr^m$, in which H and m are constants about shape factors of particles, the following log normal distribution function can approximately be used to describe $n(r)$ [23]:

$$n(r) = \frac{1}{r\sqrt{2\pi}\ln\sigma}\exp\{-[\frac{\ln(r/\bar{r})}{\sqrt{2\pi}\ln\sigma}]^2\}, \tag{13}$$

where $\bar{r}$ is the geometric mean radius, σ the standard deviation. The value of $\bar{r}$ can be substituted approximately with the average radii, a, and σ can take the classic value of 1.5.

Using the multi-component MG model proposed by Wood and Ashcroft [23] and further considering the effect of particle clustering and clusters distribution, we can thus obtain the effective thermal conductivity of suspension with nanoparticle inclusions. Substituting the effective thermal conductivity of clusters, $k_{cl}(r)$, and the radius distribution function, $n(r)$, into the modified MG equation, the effective thermal conductivity of liquid with nanoparticle suspension, k, can be expressed as:

$$\frac{k}{k_f}=\frac{(1-\phi)+3\phi\int_0^\infty \frac{k_{cl}(r)n(r)}{k_{cl}(r)+2k_f}dr}{(1-\phi)+3\phi\int_0^\infty \frac{k_f n(r)}{k_{cl}(r)+2k_f}dr}. \tag{14}$$

This equation is the proposed fractal model deduced for predicting of effective thermal conductivity of liquid with nanoparticles inclusion.

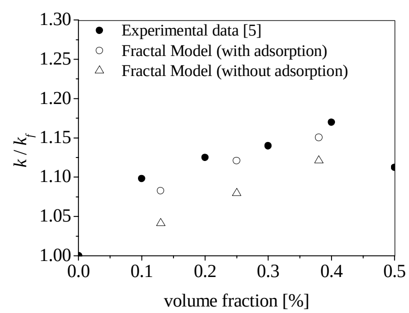


Fig. 5. Comparison of experimental data and predicting results using fractal model with or without consideration on particle size effect and surface adsorption

Taken fractal dimension of clusters to be the calculated value in Fig. 4, and used the proposed fractal model, the consistency between predictive value and experimental data [5] for effective thermal conductivity enhancement of 50nm CuO particle suspensions can be obtained, as shown in Fig. 5. The proposed fractal model fits well with experimental data when the particle concentration is less than 0.5%. Beyond this dilute limit, the possible deposition effect may have to be considered, which is difficult for predicting transport coefficients, e.g., thermal conductivity. At the same time, an obvious decrease in k/k_f is observed if the adsorption effect is not considered, and thus, the packed liquid molecules on the particle / liquid interface contribute significantly to the enhancement of effective thermal conductivity of liquid.

6. Conclusions

A fractal model, which involves applying and improving of the effective medium theory, is proposed for predicting the effective thermal conductivity of liquid with dilute suspension of nonmetallic nanoparticles. The proposed fractal model predicts well the trend for variation of the effective thermal conductivity of liquid with dilute suspension of nanoparticles, and fits successfully with our experimental data for 50nm CuO particles suspension in deionized water when $\phi < 0.5\%$. The calculated result also shows that the predictive calculation of effective thermal conductivity is complicated. Further work would be needed, especially for metallic nanoparticle inclusions.

Though the effective thermal conductivity of nanoparticle suspensions can be predicted successfully through the proposed fractal model, the predictive calculation is still complicated in applying and improving the effective medium theory. Also, the spatial distribution of nanoparticle clusters should be carefully concerned and described. To improve this method, the actual spatial distribution of nanoparticles clusters in liquid needs to be further studied. And the effects of particle size and particle shape also need to be included. For metallic nanoparticles, the effect of particle size still remains to be considered.

Acknowledgement

The authors acknowledge the financial support from the National Natural Science Foundation of China (with Grant Number 59995550-3).

References

1. J. C. Maxwell, Treatise on Electricity and Magnetism, Oxford, Oxford University Press, 1873
2. Choi, S.U.S, enhancing thermal conductivity of fluids with nanoparticles, In Developments and Applications of Non-Newtonian Flows, edited by D.A.Siginer, H.P.Wang, New York, ASME, FED-Vol.231/MD-Vol.66, pp.99-105., 1995
3. P.Keblinski, S.R.Phillpot, S.U.-S.Choi, J.A.Eastman, Mechanisms of Heat Flow in Suspensions of Nano-Sized Particles (Nanofluids), Int. J. Heat Mass Transfer, Vol.45, No.4, pp.855-863, 2002
4. B.X.Wang, H.Li, X.F.Peng, Research on the Heat Conduction Enhancement for Liquid with Nanoparticles Suspensions, General Paper (G-1), present at the Int. Sympo. Therm. Sci. Eng. (TSE 2002), Oct. 23-26, Beijing, 2002
5. L.P.Zhou, B.X.Wang, Experimental Research on the Thermophysical Properties of Nanoparticle Suspensions Using the Quasi-Steady State Method, (in Chinese), in Ann. Proc. Chinese Eng. Thermophys., Shanghai, pp.889-892, 2002
6. B.B.Mandelbrot, The Fractal Geometry of Nature, W.H.Freeman Publisher, San Francisco, 1982
7. R.Pitchumani, S.C.Yao, Correlation of Thermal Conductivities of Unidirectional Fibrous Composites Using Local Fractal Techniques, J. Heat Transfer, Vol.113, pp.788-796, 1991
8. K.W.Yu, Effective Nonlinear Response of Fractal Clusters, Phys. Rev. B, Vol.49, No.14, pp.9989-9992, 1994
9. K.W.Yu, E.M.Y.Chan, Y.C.Chu, G.Q.Gu, Enhanced Nonlinear Response of Fractal Clusters, Phys. Rev. B, Vol.51, No.17, pp.11416-11423, 1995
10. H.S.Carslaw, J.C.Jaeger, Conduction of Heat in Solids, Oxford University Press, 1959 ed., pp.112-115, 1959
11. D.A.G.Bruggeman, Berechnung Verschiedener Physikalischer Konstanten von Heterogenen Substanzen, I. Dielektrizitatskonstanten und Leitfahigkeiten der Mischkorper aus Isotropen Substanzen. Annalen der Physik. Leipzig, Vol.24, pp.636-679, 1935
12. J.E.Spanier, I.P.Herman, Use of Hybrid Phenomenological and Statistical Effective-Medium Theories of Dielectric Functions to Model the Infrared Reflectance of Porous SiC Films, Phys. Rev. B, Vol.61, No.15, pp.10437-10450, 2000
13. J.W.S.Rayleigh, On the Influence of Obstacles Arranged in Rectangular Order upon the Properties of the Medium, Philos. Mag., Vol.34 ,pp.481-502, 1892
14. B.Cichocki, B.U.Felderhof, Dielectric Constant of Polarizable, Nonpolar Fluids and Suspensions, J Stat. Phys., Vol.53, No.1/2, pp.499-521, 1988
15. J.Monecke, Microstructure Dependence of Material Properties of Composites, Phys. Status Solidi B, Vol.154, pp.805-813, 1989
16. G.Chen, Particularities of Heat Conduction in Nanostructures, J. Nanoparticle Research, Vol.2, pp.199-204, 2000
17. G.Chen, Nonlocal and Nonequilibrium Heat Conduction in the Vicinity of Nanoparticles, ASME J. Heat Transfer, Vol.118, No.11, pp.539-545, 1996
18. T.Q.Qiu, C.L.Tien, Size Effects on Nonequilibrium Laser Heating of Metal Films, ASME J. Heat Transfer, Vol.115, pp.842-847, 1993
19. G.Nimtz, P.Marquardt, H.Gleiter, Size-Induced Metal-Insulator Transition in Metals and Semiconductors, J. Crystal Growth, Vol.86, pp.66-71, 1988
20. J.M.Yan, Q.Y.Zhang, J.Q.Gao, Adsorption and Agglomeration − Surface and Porosity of Solid, (in Chinese), Science Press, Beijing, 1986
21. A.W.Adamson, Physical Chemistry of Surfaces, Wiley, New York, 5th ed., 1990
22. B.X. Wang, L.P. Zhou, X.F. Peng, The Particle Size Effect and Surface Adsorption on Effective Thermal Conductivity of Liquid with Nano Nonmetallic Particle Suspension, paper to be presented at FOMMS 2003, Foundation of Molecular Modeling and Simulation, Keystone, Colorado, United States, July 6-11, 2003
23. D.M.Wood, N.W.Ashcroft, Effective Medium Theory of Optical Properties of Small Particle Composites, Philos. Mag., Vol35, No.2, pp.269-280, 1977
24. S.Havlin, D.Ben-Avraham, Diffusion in Disordered Media, Adv., Phys., Vol.36, No.6, pp.695-798, 1987
25. P.M.Hui, D.Stroud, Complex Dielectric Response of Metal-Particle Clusters, Phys. Rev. B, Vol.33, No.4, pp.2163-2169, 1990
26. P.M.Hui, D.Stroud, Effective Linear and Nonlinear Response of Fractal Clusters, Phys. Rev. B, Vol.49, No.17, pp.2163-2169, 1994