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Investigation on optical theoretical models of SiO2 nanofluids

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***Abstract*— Photovoltaic and photovoltaic heat technology can be coupled by nanofluid-based solar direct absorption thermal collector. It is an important means to improve the efficiency of comprehensive utilization of solar energy. Nanofluids optical theory plays an important role in the development of new photovoltaic-thermal experimental platform. In addition, study on optical characteristics of nanofluids is still in the initial stage. As a result, it is of great significance for the study of the law and mechanism of it. In this paper, the Rayleigh scattering model and the Mie scattering model are used to analyze the critical optical characteristics -- transmittances of the nanofluids. Furthermore, the consistency between different theoretical models and experimental datum is studied by contrast verification between experiments and theory calculation. The conclusion shows that Mie scattering model performs better than Rayleigh scattering model and expresses a better applicability in the development of photovoltaic thermal experimental platform. This theoretical study on optical characteristics of nanofluids is expected to prompt on the application of nanotechnology in the field of solar energy. It is also expected to improve the efficiency of comprehensive utilization of solar energy.**

Keywords— nanofluids; rayleigh scattering model; mie scattering model; transmittance

I. INTRODUCTION

Recently, more and more researches attach great importance on the technology of nanofluids which weigh a lot in increasing areas [1]. Existed experiments have proved that nanofluids have a good potential on optics. To look deeply, nanoparticles attribute to the excellent optical property of nanofluid to a great extent. Compared to the optical properties of bulk materials, nanofluids perform better as a result of the nanoparticles' effect of nanoparticles, the nanoparticles' interface arrangement of atoms and quantum effects of nanomaterials [2]. Therefore, the key of analyzing nanofluids' optical properties lies in effective calculation of nanoparticles' optical properties. Popular methods for such calculation include Rayleigh scattering model, Mei model, effective medium approach, discrete dipole approximation and T matrix approach and so on.

Among them, Rayleigh scattering model and Mei model have been used widely in lab and college for their simplicity. Rayleigh scattering model is named by Lord. Rayleigh, who is well-known for investigation on the interaction between sun light and atmosphere [3]. Tyagi was the first to apply it in calculating nanoparticles' optical properties [4]. Mie scattering model was suggested by Gustav Mie in 1908 [12], which works well in resolving homogeneous spherical single particles' optical properties. In the analysis of nanofluids' optical properties, domestic and foreign scholars have done a lot of researches up to now. For Rayleigh scattering model, Mu Lijuan, a master in Shanghai Institute of Electric Power, used the self-prepared SiO2 nanofluids and TiO2 nanofluids to analyze what effects particle size, mass fraction and optical path put on transmittance. This thesis [5] involves comparison between the experimental values of two nanofluids' transmittances and the theoretical analysis by Rayleigh scattering model, while lacks numerical analysis of constraints on theoretical models. Otanicar [6], Saidur [7] used Rayleigh scattering model to design direct absorption solar thermal collector based on nanofluid. The former one explored the effects of different volume fractions of carbon nanotubes, graphene and silver nanoparticles on the collector efficiency. The latter one focused on the effect of the particle size and volume fraction of alumina nanoparticles on the alumina-water nanofluid extinction coefficient. All in all, the two researches have not involved the analysis of transmission. As for Mie theory, Mahendia [8] investigated Cu-polyvinyl alcohol nanofluid absorption spectra with the use of Mie theory. Moreira [9] and others used Mie theory to calculate and predict Ag-water and Au-water nanofluids' extinction coefficients. Neither of them have step in the correlation researches on metal oxide nanofluids' transmittances.

In this thesis, we analyzes the accuracy of two models in calculating the transmittances of SiO2-water nanofluids. We also provide suggestions on how to choose calculation model in order to save workload and propose the change of two models' relative deviations with the wavelength of incident light. The optical properties of nanofluids investigated in our study are supposed to be a fundamental work in the field of solar thermal utilization [10]. In this filed, previous studies [6] also indicate that the transmittances of nanofluids directly affect the efficiencies of thermal collectors. Thus, our study is expected to be meaningful and worthwhile for related researchers.

II. ANALYSES OF NANO-OPTICAL THEORY

A. Overview

The electromagnetic theory which proposed by Maxwell in 1865 can be used to explain the reflection, transmission of plane wave in an interface or a certain-thickness dielectric layer. It can also be used to explain incidence, scattering and other properties of the light, regarded as electromagnetic wave, which interacts with a particle surface. The interaction between the light and the particle is related to the size parameter of the particle which is relative to the wavelength of the incident light. The dimension parameter is defined as follows:

$$\alpha = \frac{\pi D}{\lambda} \tag{1}$$

In Eq. (1), D is the equivalent diameter of the particle and λ refers to the wavelength of the incident light. According to the size parameter, the following three regions can be defined [11]; (1) $\alpha \ll 1$, it is well-known as Rayleigh scattering region where the particle size is sufficiently smaller than the wavelength; (2) $\alpha \approx 1$, it is generously called Mie scattering model region where the particle diameter and wavelength are of the same order of magnitude [12]; (3) $\alpha \gg 1$, it belongs to the geometric optics where the particle size is large enough that the particle surface can be treated as a general surface when solving such issue.

The above models are based on solutions to Maxwell equation. For a specific type of nanoparticle, the simplification of Maxwell equation is different resulting in different equation of the optical parameters of nanoparticles.

B. Classification of Theoretical Models

For the Rayleigh scattering model, when the size of the particle (spherical or non-spherical) is much smaller than the wavelength of the incident radiation, the number of wavelength in the vacuum is approximately zero and the retardation effect is ignored [13] so that the background scattering is negligible. If the incident light is regarded as a plane wave and the particle is regarded as a homogeneous, isotropic sphere, the model of the interaction between the electromagnetic wave and the particle can be simplified to a dipole model in a uniform electrostatic field [14]. If the condition $\alpha \ll 1$ is satisfied, phase deviation is negligible and the additional field can be quickly transferred to the interior of the particle [15]. Therefore, the high order terms of size parameter in the calculation formula of Stokes parameter can be neglected [14]. Such statements simplify the calculation of particle scattering field.

On the base of two above assumptions, the simplified optical efficiencies can be obtained by the following equations [15]:

$$Q_{e\lambda} = 4\alpha \,\mathrm{Im}\{\frac{m^2-1}{m^2+2}[1+\frac{\alpha^2}{15}(\frac{m^2-1}{m^2+2})\frac{m^4+27m^2+38}{2m^2+3}]\}+\frac{8}{3}\alpha^4 \,\mathrm{Re}|(\frac{m^2-1}{m^2+2})|^2 \tag{2}$$

$$Q_{s\lambda} = \frac{8}{3}\alpha^4 \,\mathrm{Re}|(\frac{m^2-1}{m^2+2})|^2 \tag{3}$$

In Eq. (2) and (3), α refers to the dimension parameter of the particle and m is named as the optical constant of nanoparticles. It is defined as $m = n - ki$ in which the real part n is the refractive index and the imaginary part k is the absorption index [11].

Assuming the base fluid is non-absorption fluid $Q_{e\lambda} = Q_{s\lambda} + Q_{a\lambda}$ can be applied [14]. For homogeneous, isotropic particles which satisfy $\alpha \ll 1$ and $|m|\alpha \ll 1$, we can get the simple expression of $Q_{a\lambda}$:

$$Q_{a\lambda} = 4\alpha \,\mathrm{Im}\{\frac{m^2-1}{m^2+2}\} \tag{4}$$

Eq. (2), (3) and (4) have been cited in many related theses. In addition, as a result of the above equations, for the homogeneous isotropic spherical particles, particle optical properties are only relevant to the particle size parameters and particle optical constant m.

For some nanoparticles, conditions such as $\alpha \ll 1$ a and $|m|\alpha \ll 1$ are difficult to meet due to the Brownian motion and the intermolecular force so that the Rayleigh scattering model is not suitable in such situation. Therefore, we will introduce another model used in our work which is called Mie scattering model.

Generously speaking, Mie theory is the solution to Maxwell equation for spherical particles. We suggest those spherical particles are isotropic in Rayleigh scattering model which means that phase function calculated here is independent of the scattering direction and the degree of polarization of the scattered light is zero. These assumptions [14] are proper for sufficient small nanoparticles. In the case that particle diameter has the same order of magnitude with wavelength, they are no longer valid. In Mie scattering model, the incident field is exponentially expanded by an infinite number of vector harmonic functions and the expansion function contains the angle function with the scattering angle as a variable. It is obvious that this model takes the effect of different scattering directions on scattering field calculation into consideration. Therefore, Mie scattering model is suitable for the calculation of such particles.

In Mie scattering model, optical efficiencies can be expressed as Eq. (5) and (6) [15]:

$$Q_{e\lambda} = \frac{2}{\alpha^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}(a_n + b_n) \tag{5}$$

$$Q_{s\lambda} = \frac{2}{\alpha^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2) \tag{6}$$

If the base fluid is non-absorption fluid, the relationship between three optical efficiencies is supposed to be $Q_{e\lambda} = Q_{s\lambda} + Q_{a\lambda}$, from which we can get the expression of $Q_{a\lambda}$, a_n and b_n showed in Eq. (5) and (6). α refers to the dimension parameter of the particle. Meanwhile, a_n and b_n are called Mie coefficiency. Considering the permeability of particles is

consistent with the surrounding medium, Mie coefficiencies can be calculated by the following equations [15]:

$$a_n = \frac{\psi_n(\alpha)\psi_n'(\mathrm{m}\alpha) - \mathrm{m}\psi_n'(\alpha)\psi_n(\mathrm{m}\alpha)}{\xi_n(\alpha)\psi_n'(\mathrm{m}\alpha) - \mathrm{m}\xi_n'(\alpha)\psi_n(\mathrm{m}\alpha)} \tag{7}$$

$$b_n = \frac{m\psi_n(\alpha)\psi_n'(\mathrm{m}\alpha) - \psi_n'(\alpha)\psi_n(\mathrm{m}\alpha)}{m\xi_n(\alpha)\psi_n'(\mathrm{m}\alpha) - \xi_n'(\alpha)\psi_n(\mathrm{m}\alpha)} \tag{8}$$

As it is shown in Eq. (7) and (8), $\psi_n(\alpha)$ and $\xi_n(\alpha)$ are called Riccatti-Bessel functions in which recursive method should be applied for calculation. Specific mathematical calculation process can refer to the literature [16].

III. Calculation of the Optical Properties of Nanofluids

A. Calculation of Extinction Coefficient of Particle System

The models described above are aimed at single particle. In fact, the nanofluids studied here contain not only single nanoparticle but a particle system composed of numerous nanoparticles.

When the light incidents to the nanofluid, it is partially absorbed by the fluid, partially absorbed and scattered by the nanoparticles in the particle system. The rest transmits through the particle system. Among them, the particle system has an effect especially on the incident light attenuation by scattering.

According to Maxwell's theory, regarding incident light as an electromagnetic wave, there are three kinds of scattering caused by the particle system shown in Fig. 1 (1) diffraction without interacting with particles, (2) reflection from the particle surface and (3) internal refraction of the particles.

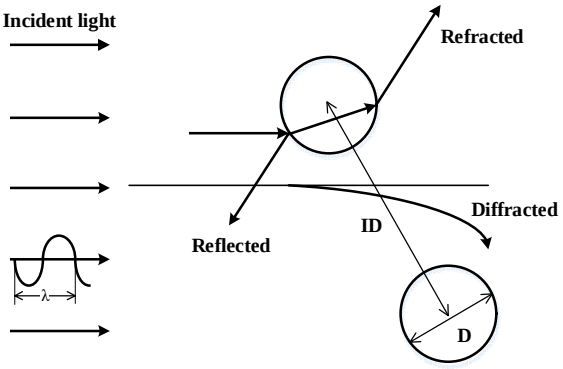


Fig. 1. Interaction between electromagnetic waves and spherical particles [11].

For a particle system, parameters to determine whether it is in the non-independent scattering region are the following: (1) size parameter α, (2) volume fraction f_v and (3) interparticle distance to the wavelength $\frac{ID}{\lambda}$. Refering to Fig. 2, we can learn that for particle systems with volume fraction less than 0.6% and interparticle distance to the wavelength greater than 0.5, it is in the individual scattering region. In addition, Siegel and Howell have proposed that the Eq. (9) should be satisfied for the individual scattering regions [17]:

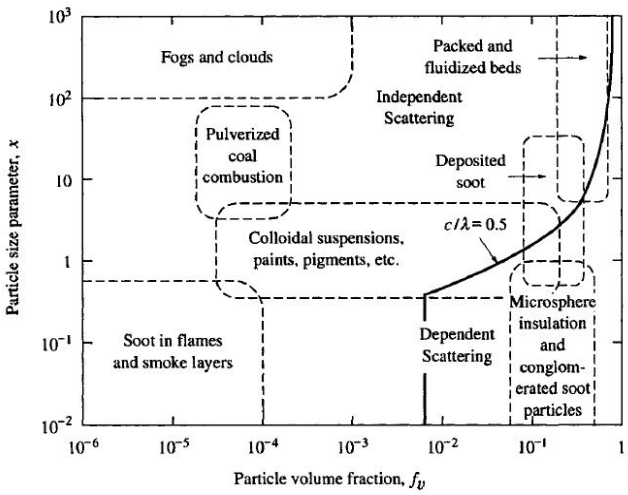


Fig. 2. Scattering regime map for independent and dependent scattering [19].

$$ID + 0.1D > \frac{\lambda}{2} \tag{9}$$

In the Eq. (9), D refers to the equivalent diameter of the particle. λ refers to the wavelength of incident light and interparticle distance can be calculated from Eq. (10) [18]:

$$ID = 2.77Df_v^{-3.081} \tag{10}$$

To sum up, as for a particle system which satisfies the condition of independent scattering, the extinction coefficient of the particle system can be calculated by the following simplified equation:

$$\mu_{e\lambda,\,p} = \frac{3}{2}\frac{fvQ_{e\lambda}}{D} \tag{11}$$

In the Eq. (11), $\mu_{e\lambda,p}$ refers to the extinction coefficient of the particle system. $Q_{e\lambda}$ refers to the extinction coefficient of single nanoparticle and f_v refers to the volume fraction of the nanoparticle system.

B. Calculation of Nanofluid Optics.

The extinction coefficient of particle system mentioned above is obtained on the assumption that the base fluid is completely transparent. Since the base fluid of most nanofluids has non-negligible attenuation characteristics so that the influence of the base fluid on the extinction coefficient of the nanofluids needs to be considered. For simplicity, we only consider the nanofluid dispersion system which meets the requirements of independent scattering region. In such system, the extinction coefficient of nanofluid can be considered as a simple addition of the absorption coefficient of base fluid to the absorption coefficient of particle system, as shown in Eq. (12):

$$\mu_{e\lambda,\,np} = \mu_{e\lambda,\,p} + \mu_{e\lambda,\,f} \tag{12}$$

In the Eq. 12, $\mu_{e\lambda,p}$ refers to the extinction coefficient of base fluid. Considering the base liquid as a pure liquid, for the calculation of attenuation characteristics, the scattering can ignored and the attenuation is mainly due to absorption [20]. Therefore, the extinction coefficient of the base liquid is calculated as shown in Eq. (13):

$$\mu_{e\lambda,\,f} = \frac{4\pi k}{\lambda} \tag{13}$$

In the Eq. (13), k refers to the absorption index of the base fluid and λ is the wavelength of incident light.

The nanofluid mainly investigated here is SiO2-water nanofluid. In recent years, the study on the optical properties of water is becoming more and more sufficient [21, 22] so that calculation and analysis of SiO2-water nanofluid transmittance in this thesis are based on the data on water absorption coefficient from existing work.

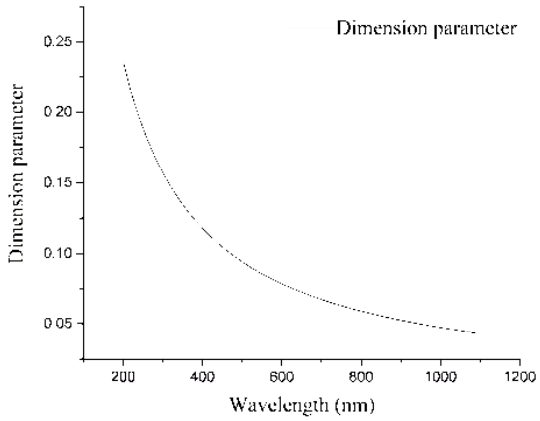
Based on Eq. (13), the relationship between the extinction coefficient of nanofluids and the transmittance of nanofluids can be obtained by Lambert-Beer's law:

$$T = \frac{I}{I_o} = e^{-L\mu_{e\lambda,np}} \tag{14}$$

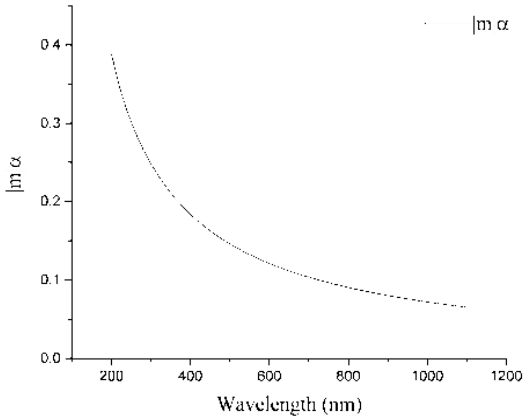
In the Eq. (14), I_0 refers to the intensity of the incident light. I refer to the intensity of the transmitted light. Besides, $\mu_{e\lambda,p}$ refers to the extinction coefficient of base fluid and L refers to L refers to optical path difference.

IV. Comparison of Theory Models with Experimental Data on Nanofluids Transmittance

In order to evaluate the applicability and accuracy of two models in predicting optical properties of SiO2-water nanofluids, numerical simulation by MATLAB are used and experiments are made for further comparison in our study.

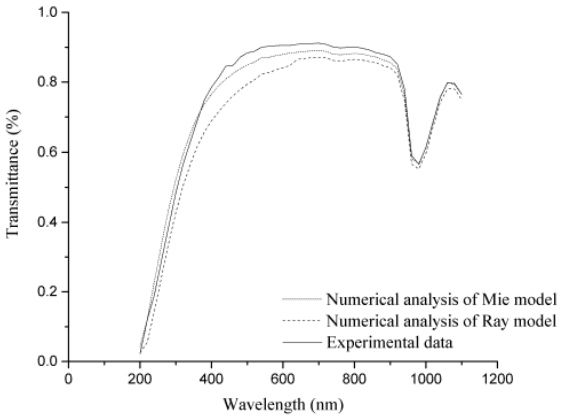


a) Approximation standard I for Rayleigh scattering model

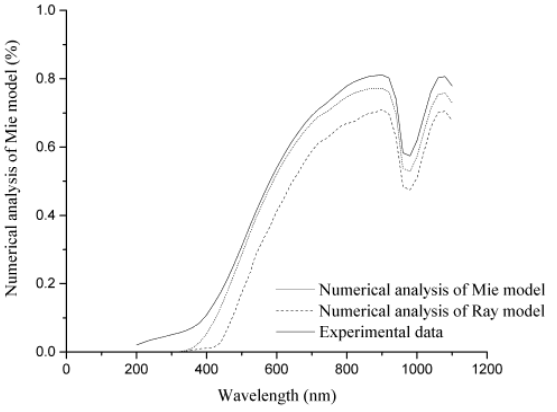


b) Approximation standard II for Rayleigh scattering model

Fig. 3. The relationship between wavelength and approximation standards for Rayleigh scattering model.



a) volume fraction 0.1%



b) volume fraction 1%

Fig. 4. Transmittance of SiO2 nanofluid for Rayleigh scattering model, Mie scattering model and experiments.

The relations between the wavelength and approximate standards for Rayleigh scattering model are shown in Fig. 3. Besides, the calculation results of the SiO2-water nanofluid's transmittance by Rayleigh scattering model, Mie scattering theory and experiments are shown in Fig. 4. From Fig. 3 and Fig. 4a, one can conclude that if the SiO2 nanofluid with particle size of 15nm has a volume fraction lower than 0.6% [22] and two approximate standards are small enough to satisfy Rayleigh scattering model's approximate conditions $\alpha \ll 1$ and $|m|\alpha \ll 1$, the difference between results calculated by Rayleigh scattering model and experimental data is relatively small. Meanwhile, it is suitable to apply Rayleigh scattering model for predicting the transmittance of the SiO2- water nanofluid.

As shown in Fig. 3, it is obvious that with the decrease of the wavelength, the approximate standards for Rayleigh scattering model are sufficiently large contributing to a failure of satisfying the two approximate conditions. Further analysis from Fig. 4a shows that in the small wavelength region, the results calculated from Rayleigh scattering model express an obvious difference compared with experimental datum, which determines that Rayleigh scattering model isn't suitable for prediction. However, Mie scattering theory gives a good prediction on the nanofluid's transmittance compared with experimental datum. Besides it can be concluded from Fig. 4b that when the volume fraction of the SiO2-water nanofluid is larger than 0.6%, results calculated from Mie scattering model agrees with the experimental data well. In contrast, Rayleigh scattering model shows deviation obviously. Further analysis shows that the volume fraction effects most whether it locates in the wavelength regions satisfying two approximate conditions or not. In this situation, it is more proper to choose Mie scattering model.

In order to investigate the relation between results calculated from two models and experimental datum intensively, we select three representative wavelengths and calculate devia- tions which is shown in TABLE I. From TABLE I, one can find that when the wavelength is larger than 700nm, the deviations between two models and experiment are both small. When the wavelength is less than 700nm, the deviations of two models show differences which indicates that Rayleigh scattering model performs well in a wavelength region. Notably, deviations from Mie scattering model among selected wavelengths are consistently acceptable.

TABLE 1. NUMERICAL VALUES OF TWO MODELS AND EXPERIMENTAL DATA.

Wavelength [nm] / Volume fraction	Rayleigh scattering model			Mie scattering model			Experiments		
	300	700	1100	300	700	1100	300	700	1100
0.1%	0.43	0.87	0.75	0.52	0.89	0.76	0.48	0.91	0.76
1%	2.0E-05	0.59	0.68	1.1E-04	0.64	0.73	0.05	0.69	0.78

Based on TABLE I, the relative deviations between two models and experiment are calculated which is shown in TABLE II. From TABLE II, for Rayleigh scattering model, one can find that the deviation with the volume fraction of 0.1% seems more acceptable than the deviation with the volume fraction of 1%. For Mie scattering model, the effect of the volume fraction is not as obvious as Rayleigh scattering model. As for the effect of wavelength, both models shows bad predictions at the wavelength of 300nm. The deviations from two models is the smallest when the wavelength is close to 1100nm and rapidly increases with the decrease of the wavelength. By comparing figures and tables, for the SiO2-water nanofluid with particle sizes of 15nm, we suggest that 700nm is a beginning wavelength where two models predictive capability express an considerable difference. The certain value of the wavelength will be figured out in our follow-up work.

TABLE 2. RELATIVE DEVIATION BETWEEN TWO MODELS.

Wavelength [nm] / Volume fraction	Rayleigh scattering model			Mie scattering model		
	300	700	1100	300	700	1100
0.1%	10.42%	4.40%	1.32%	8.33%	2.20%	0.00%
1%	100.00%	14.49%	12.82%	100.00%	7.25%	6.41%

Based on the above analyses, one can conclude that the Mie scattering model surely has a better prediction. Meanwhile, only if the volume fraction is less than 0.6%; $\alpha \ll 1$ and $|m|\alpha \ll 1$ are satisfied, Rayleigh scattering model can predict well. When it comes to reality application, researchers shall choose different prediction models with different experimental conditions in order to reduce the computational workload under certain accuracy.

In addition to the effect of the wavelength and the volume fraction analyzed here, particle sizes; types of nanoparticles and different base fluids also affect the choice of the theoretical model for the calculation of nanofluids' optical properties. These effects are not the main contents of our study but will be discussed in the follow-up work.

V. CONCLUSION

In this thesis, we firstly introduce two theoretical models for calculating nanofluids optical properties. The theoretical bases and calculation processes of Rayleigh scattering model and Mie scattering model are analyzed in detail. Secondly, using the numerical calculation function of MATLAB, we calculate SiO2-water nanofluids transmittances by two models. By comparing the theoretical values and the experimental datum, we suggest the applicable scopes of two models which goes as: (1) if the volume fraction of the nanofluid is less than 0.6% and the approximate conditions of Rayleigh scattering model $\alpha \ll 1$ and $|m|\alpha \ll 1$ are satisfied, Rayleigh scattering model is preferable for the reduce of workload. (2) if the volume fraction of the nanofluid is larger than 0.6%, Mie scattering model is more suitable. (3) for the SiO2-water nanofluid with particle size of 15nm, we suggest that 700nm is a beginning wavelength where two models predictive capability express an obvious difference. The conclusions proposed by this thesis are expected to provide some help to those who are related in the research of this field.

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