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A theoretical investigation of the optical properties of metal nanoparticles in water for photo thermal conversion enhancement

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ABSTRACT

Nanofluids are used in photo thermal absorbers such as direct absorption solar collectors (DASC) to improve the absorption and heat transfer. A range of experimental studies have investigated the photo thermal conversion in nanofluids based on metal nanoparticles. The photo thermal conversion efficiency depends mainly on the ability of the metal nanoparticle based nanofluid to absorb light which in turn is governed by the particle localized surface plasmon resonance (LSPR). In this article we investigate the optical properties of metal nanoparticles in water, and based on this we judge their suitability for use in water based nanofluids. We simulate the optical properties and provide an overview of the absorption, scattering and extinction properties of solid and spherical aluminum, silver, copper and gold nanoparticles suspended in water. We study nanoparticle diameters between 20 and 140 nm for the wavelength range 300–1350 nm (200–1250 nm for aluminum). We compare our simulation results with the available experimental results presented in the literature, and find good agreement.

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1. Introduction

In the chase for cleaner, more efficient energy harvesting, solar energy is an appealing source, being both abundant and free. Between electrical, chemical, and thermal solar energy conversion, thermal is the more simple and straightforward choice [1]. Common photo thermal collectors absorb light utilizing a black body, and transfer the heat to a fluid. The fluid is then transported from the point of absorption to be used directly for heating, or to a generator such as a steam engine to produce electricity. Some examples are flat plate collectors, parabolic troughs and power towers [2]. The collector efficiency is limited by how effectively the heat may be transfered from the black body to the fluid [3]. This limitation can be circumvented by making the liquid absorb the light directly. Such a device is called a direct absorption solar collector (DASC) [4]. Nanofluids may be introduced in non-DASC devices also, such as parabolic troughs [5]. In such cases, a nanofluid may replace the fluid in order to increase absorption in the liquid. Nanoparticles in a liquid will transfer the absorbed energy to the liquid more quickly than that of an idealized black body pipe, since nanoparticles will have a larger surface area in contact with the liquid. Also, the thermal conductivity of the nanoparticles increases with decreasing particle size [6], improving the photo thermal conversion efficiency η.

The photo thermal conversion efficiency depends on the geometric configuration, specific heat, and convection heat transfer, but it is ultimately limited by the optical absorbtion of the nanofluid [7]. In the following we will focus on water as the liquid medium. Water itself is highly transparent and not a great absorber [8]. Metal nanoparticles absorb light through the localized surface plasmon resonance (LSPR). LSPR occurs when the electric field of a photon causes the metal electrons to oscillate, and the photon frequency is matched to cause an amplification of the oscillation, determined by the nanoparticle size and shape, and the dielectric properties of the surrounding medium [9].

In this paper, the absorption of a single particle is presented as a cross section σ_{abs} [nm^2]. The absorption coefficient β_{abs} of a nanofluid is given as

$$\beta_{abs} = \sigma_{abs} N \tag{1}$$

where N is the number of nanoparticles per unit volume. The absorption efficiency Q_{abs} is given as

$$Q_{abs} = \frac{\sigma_{abs}}{A_{np}} \tag{2}$$

where A_{np} is the cross sectional area of the particle (πr^2 for a sphere) [10]. At resonance, the nanoparticle can absorb light from an area which is larger than it's physical area making the efficiency greater

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Nomenclature

Abbreviations

AM 1.5	solar spectral irradiance for air mass 1.5 (W m^{-2} nm^{-1})
DASC	direct absorption solar collector
FDTD	finite-difference time-domain
FWHM	full width half maximum
IR	infrared, 1050 nm and longer
LSPR	localized surface plasmon resonance
NIR	near infrared, 700–2500 nm
PML	perfectly matched layer
VIS	visible, 390–700 nm
wl	wavelength (nm)

Symbols

β	coefficient
ϵ	permittivity (m^{-3} kg^{-1} s^4 A^2)
η	photo thermal conversion efficiency
μ	permeability (kg m s^{-2} A^{-2})
σ	cross section (nm^2)
A	area (nm^2)
c	specific heat (m^2 K^{-1} s^{-2})
d	diameter (nm)
E	electric field (kg m s^{-3} A^{-1})
H	magnetic field (A m^{-1})
I	intensity (W m^{-2})
m	mass (kg)
N	number density (particles/nm^3)
P	power (W)
Q	efficiency
t	time (s)
T	temperature (K)
z	zenith angle (°)

Subscripts

abs	absorption
exp	experiments in literature
ext	extinction
l	liquid
lum	lumerical simulation
np	nanoparticle
sca	scattering
sim	simulations in literature

than 1. Eqs. (1) and (2) are valid for the scattering and extinction cross sections, σ_{sca} and σ_{ext} as well, and are related to the absorption cross section by the following definition [10]:

$$\sigma_{ext} = \sigma_{abs} + \sigma_{sca} \tag{3}$$

In the context of photo thermal conversion for nanofluids, scattered light can be absorbed by neighboring nanoparticles. For DASC applications, there is therefore no immediate need to minimize scattering. However, light must be absorbed in order to be converted to heat, so the main objective is to maximize the absorption efficiency. The absorbance spectra of nanofluids are usually measured experimentally using a transmission spectrometer, which in reality measures the extinction. For small nanoparticles, usually less than 30–40 nm (though this is material dependent), scattering is negligible and the terms extinction and absorption are interchangeable [11]. As the nanoparticles becomes larger however, scattering becomes the dominant process.

One should keep in mind that a high absorption efficiency is not necessarily the same as a high photo thermal conversion efficiency, η. η is given as

$$\eta = \frac{(c_l m_l + c_{np} m_{np})}{IA} \frac{\Delta T}{\Delta t} \tag{4}$$

where c_l and c_{np} is specific heat of the liquid and particle respectively, m_l and m_{np} is mass of the liquid and particle respectively, I is solar intensity, A is the illuminated area of the fluid, and ΔT is the temperature rise in the time interval Δt [12]. Some materials may re-emit the absorbed light at longer wavelengths (fluorescence), and some materials may have low thermal conduction despite a high absorption efficiency, thus yielding a low ΔT. For example, Chen et al. observe that the photo thermal conversion efficiency seems to decrease slightly for increasingly larger gold nanoparticles, when investigating spherical gold nanoparticles in water [7].

Theoretical calculations of optical nanostructures can be performed using for example Mie theory [9], Raleigh scattering method [10], or finite difference [13] or finite element simulations in the time or frequency domain, depending on the system one wishes to investigate. In the Raleigh scattering method, the electric field is assumed to be constant over the entire particle, and it is therefore only valid for particles much smaller than the wavelength of radiation. For larger particles, Mie theory can be applied. Mie theory describes the interaction of an electromagnetic plane wave with a homogeneous sphere. For non-spherical particles, discretization of Maxwell's equations may be appropriate. For transient behavior of the electromagnetic field, a time domain approach is desirable. Frequency domain calculations are particularly useful when studying periodic structures. In the following, four examples of Raleigh scattering calculations are given. Metal nanoparticles dispersed in water have been investigated by Saidur et al. [11], who investigated aluminum nanoparticles in the range 1–20 nm suspended in water, and found the absorption peak at approximately 300 nm. Taylor et al. [14] found the extinction peak for 40–200 nm silver spheres in water to be at 400 nm using Maxwell-Garnett modeling. Gorji and Ranjbar investigated silver spheres and found an extinction peak at approximately 390 nm, but did not get corresponding experimental results for 20 nm diameter spheres in water [15]. Khlebtsov et al. investigated 15 nm gold, and 15 and 60 nm silver spheres and found extinction peaks at 515, 380 and 412 nm respectively [16].

In this paper we simulate the absorption and scattering of a solid metal nanoparticle dispersed in water by solving Maxwell's equations using the finite-difference time-domain (FDTD) method in the commercial software package Lumerical [17].

$$\begin{aligned} \frac{\vec{H}\left(t+\frac{\Delta t}{2}\right) - \vec{H}\left(t-\frac{\Delta t}{2}\right)}{\Delta t} &= -\frac{1}{\mu}\nabla \times \vec{E}(t) \\ \frac{\vec{E}(t+\Delta t) - \vec{E}(t)}{\Delta t} &= \frac{1}{\varepsilon}\nabla \times \vec{H}\left(t+\frac{\Delta t}{2}\right) \end{aligned} \tag{5}$$

Eq. (5) show the one dimensional Maxwell's equations solved in the time domain using finite difference approximation [13], where E is the electric field and H is the magnetic field, μ and ϵ are the permeability and permittivity respectively, t is time, and Δt is the time step (i.e. $t = \Delta t n$, where n is an integer). For every increasing time step, the values of E and H are updated from the previous ones. A light source is an area where the electric and magnetic field

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