



Thermal conductivity studies of novel nanofluids based on metallic silver decorated mesoporous silica nanoparticles



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ABSTRACT

In the present study, the mesoporous structure of silica ($m\text{SiO}_2$) nanoparticles as well as hemiaminal grafted $m\text{SiO}_2$ decorated by metallic silver ($\text{Ag}/m\text{SiO}_2$) has been used for the preparation of glycerol based nanofluids. Structural and morphological characterization of the synthesized products have been carried out using Fourier transform infrared spectroscopy (FTIR), scanning electron microscope (SEM), X-ray diffraction (XRD), UV–vis spectroscopy, inductively coupled plasma (ICP) and N_2 adsorption–desorption isotherms. The thermal conductivity and viscosity of the nanofluids have been measured as a function of temperature for various weight fractions and silver concentrations of $m\text{SiO}_2$ and $\text{Ag}/m\text{SiO}_2$ nanoparticles, respectively. The results show that the thermal conductivity of the nanofluids increase up to 9.24% as the weight fraction of $m\text{SiO}_2$ increases up to 4 wt%. Also, increasing the percent of the silver decorated $m\text{SiO}_2$ ($\text{Ag}/m\text{SiO}_2$) up to 2.98% caused an enhancement in the thermal conductivity of the base fluid up to 10.95%. Furthermore, the results show that the nanofluids have Newtonian behavior in the tested temperature range for various concentrations of nanoparticles.

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1. Introduction

In recent decades, in order to save energy and raw materials and with regard to economic and environmental issues, many efforts have been undertaken to build efficient heat exchange devices. The common trend to address this problem is to enlarge the surface of the heat sink by, for example, incorporating micro-channels into the heat transfer structure [1]. But this approach requires an undesirable increase in the size of the thermal device systems. Furthermore, the heat transfer can be enhanced passively by the increasing thermal conductivity of the fluids. It is well known that metals and metal oxides have much higher thermal conductivity than conventional heat transfer fluids such as water, ethylene glycol, and engine oil [2]. Therefore, researchers have tried to increase the thermal conductivity of base fluids by suspending micro or larger sized solid particles in the fluids since Maxwell's theoretical work was published more than 100 years ago [3]. But using suspensions containing micro or millimeter-sized particles will be exposed to many difficulties, such as sedimentation of particles, corrosion of transport equipments and higher pumping power requirements [4]. In the past two decades with the development of nanotechnology, researchers are facing a new

approach to improve the heat transfer properties by applying nanofluids. Nanofluids are a colloidal mixture of nanoparticles (1–100 nm) and a base liquid, which have been described by Choi [5]. The surface/volume ratio of nanoparticles is 1000 times larger than microparticles. The use of nanometer-sized particles with large specific area not only improves the heat transfer but also according to the Stokes theory provides higher stabilization against the particle sedimentation avoiding the clogging of the heat sink [6]. Recently, nanofluids have attracted great interest due to the reports of greatly enhanced thermal properties. The thermal properties of different nanofluids, including thermal conductivity, viscosity, specific heat, convective heat transfer coefficient and critical heat flux have been studied extensively [7]. Among all of these properties, thermal conductivity has gained the most attention and it is believed to be the most important parameter responsible for the enhanced heat transfer. Many researchers have reported experimental studies on the thermal conductivity of nanofluids containing Al_2O_3 , TiO_2 , SiO_2 , CuO , Cu , Ag , Au , Pd , etc. nanoparticles [8–12]. Among various nanoparticles, silver nanoparticles have been considered by many researchers due to their very high thermal conductivity [13–16]. It is well known that the thermal conductivity of silver at room temperature is 700 times greater than that of water, and 3000 times greater than that of engine oil. Also, due to the high thermal and chemical stability and the electrical insulation that silica offers, silica containing nanofluids can be very important to certain industries where

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cooling is required for example in high voltage applications [17]. Although several experimental and theoretical research have focused on silica nanofluids [18–29], but only a few have focused on the synthesis of mesoporous silica nanofluids and their relationship with the enhancement heat transfer properties [30,31]. Mesoporous silica ($mSiO_2$), which was produced by Mobil Corporation Laboratories in 1992, has received great attention due to its uniform mesopores with high specific surface area in applications such as medicine, biosensors, imaging and catalysis [32]. Since heat transfer is a surface phenomenon at the particle–fluid interface, its magnitude will increase with an increase in the surface area of the particles. Therefore, due to increased surface area of nanoparticles, enhancement in the thermal conductivity of nanofluids containing porous nanoparticles is expected. Amrollahi et al. [30] studied on water based $mSiO_2$ nanofluids. They dispersed $mSiO_2$ nanoparticles in water using an ultrasonic probe and showed that after twenty-four hours sonification, the nanofluid containing 2.5 vol% $mSiO_2$ nanoparticles resulted in a 7% increase in the thermal conductivity of the based fluid. Nikkam et al. [31] prepared aqueous dispersions of mesoporous silica in the range of 1–6 wt% by adjusting the pH of the nanofluids under ultrasonic treatment. Measurement of the thermal conductivity of the nanofluids at different temperatures showed that the maximum thermal conductivity enhancement of $\sim 4.1\%$ was obtained for 6 wt% at 60 °C. The effectiveness of nanofluids coolants depends on the flow mode (laminar or turbulent) and can be estimated based on fluid dynamic equations. Viscosity is an important properties to determine nanofluids dynamic and heat transfer properties. Limited investigations are reported on the viscosity of mesoporous silica nanofluids [31]. In this work, we have synthesized $mSiO_2$ nanoparticles and a hybrid structure of $mSiO_2$ decorated with silver nanoparticles ($Ag/mSiO_2$). Furthermore, the effects of temperature and weight fraction of synthesized nanoparticles on the thermal conductivity and viscosity of glycerol based nanofluids have been investigated. Among the various heat transfer fluids, glycerol is desirable for a number of reasons including their high heat capacity, very low freezing point and compatibility with biological and environmental perspective. On the other hand, within the biodiesel production, glycerol occurs in large quantity as a natural byproduct. Currently, at least 300,000 tons of excess glycerol is globally burned for energy production [33]. Therefore, the use of glycerol-based coolants is economically affordable. To our knowledge, this is the first report on the thermal conductivity of glycerol based suspensions containing mesoporous silica nanoparticles. The questions which arise are whether our proposed methods are general and if it is possible to develop modified mesoporous silica with better properties and performance?

2. Experimental

2.1. Materials and reagents

All the reagents used in this work were analytical grade and purchased from Merck Co., including tetraethyl orthosilicate (TEOS 99%), hexadecyltrimethylammonium bromide (HTAB 98%), ammonium hydroxide (NH_4OH 29–30%), formaldehyde solution (HCHO 37%), ethanol (EtOH 99.9%), 3-aminopropyltriethoxysilane (APTS 98%) and silver nitrate ($AgNO_3$ 99.8%).

2.2. Synthesis of mesoporous silica nanoparticles ($mSiO_2$)

$mSiO_2$ was synthesized as reported previously [34] with a slight modification. As a typical experiment, 0.5 g HTAB was dissolved in 250 mL deionized water under magnetic stirring. After the solution turned clear, the pH of the solution was adjusted to approximately

11.5 with the addition of ammonium hydroxide solution. Then, the temperature was raised to 80 °C and 2.5 mL TEOS was injected to the above solution with an injection rate 30 mL h⁻¹. Stirring (450 rpm) was continued for 2 h. After a 24 h aging time, the product was washed with water and ethanol and dried at room temperature. The removal of HTAB was performed by calcination at 550 °C for 6 h (heating rate of 1 °C/min) in air.

2.3. Functionalization of $mSiO_2$ with hemiaminal group

The hemiaminal groups were produced by grafting APTS groups on the $mSiO_2$ and then letting the amine groups react with HCHO [35]. In a typical example, 0.8 g of $mSiO_2$ was stirred in a solution with various amounts of APTS (with aspect molar ratio's of APTS/ $mSiO_2$ equal to 0.25 and 0.42 denoted as $mSiO_2-NH_2-0.25$ and $mSiO_2-NH_2-0.42$, respectively) in 26 mL of ethanol for 6 h at 65 °C to graft its surface with primary amine groups, resulting in $mSiO_2$ -Amine. The resulting sample was dried under ambient conditions, and then it was suspended in 26 mL of 37% HCHO solution at 40 °C for 1 h to produce a white colored hemiaminal-functionalized sample denoted as $mSiO_2$ -hemiaminal.

2.4. Synthesis of $mSiO_2$ nanoparticles decorated with Ag nanoparticles ($Ag/mSiO_2$)

Ag nanoparticles were decorated on mesoporous silica ($Ag/mSiO_2$) by the reduction of $AgNO_3$ solution using the hemiaminal reducing agent which is anchored on the surface and the channel walls of $mSiO_2$. The dried $mSiO_2$ -hemiaminal powder was dispersed in 160 mL aqueous solution of 5 mM $AgNO_3$ and stirred for 1 h at 40 °C. The obtained products were separated by filtration and washed with deionized water, followed by drying at room temperature.

2.5. Preparation of nanofluids

The nanofluids were prepared by dispersing certain amounts of $mSiO_2$ and $Ag/mSiO_2$ nanoparticles in known quantities of glycerol with the help of a magnetic stirrer for around 3 h and then ultrasonating the suspension for 1 h using a 280 W ultrasonicator.

2.6. Characterization of synthesized nanoparticles and thermal conductivity

The morphology of the synthesized particles was observed by scanning electron microscopy (SEM) (Hitachi, S4160). Crystal structure analyses were done using an X-ray diffractometer (XRD) (Xpert Philips, PW 3040/60) with Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$). The FTIR spectra were performed using a Fourier transform infrared spectrometer (FTIR, Bruker ALPHA) in the range of 400–4000 cm⁻¹. N_2 adsorption–desorption isotherms were measured on a NOVA STATION B instrument at 77 K. Specific surface areas were calculated according to the Brunauer–Emmett–Teller (BET) method, and the pore-size distribution were calculated using desorption branch of the isotherm by the Barrett–Joyner–Halenda (BJH) method. UV–vis spectra were recorded on a Perkin-Elmer Lambda 25 spectrophotometer. Inductively coupled plasma (ICP) data were provided by Varian vista-PRO. The thermal conductivity of the nanofluids was measured by using a KD2-Pro thermal properties analyzer (Decagon Devices, USA), based on the transient hot wire method. The probe sensors used for these measurements are of 60 mm length and 1.3 mm diameter (KS-1). In order to study the temperature effect on the thermal conductivity of nanofluids, a thermostat bath was used to ensure that all the measurements are at constant temperature. The viscosity of nanofluids was measured

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