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Photothermal Characteristics of Magnetic Nanofluids for Solar Thermal Applications

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***Abstract:* Photothermal characteristics of nanofluids are of special importance in the design and performance analysis of solar thermal collectors with nanofluids as heat transfer media. The present work investigated the photothermal characteristics of the magnetite (Fe_3O_4) based nanofluids in zero and applied magnetic fields. Stable kerosene and water based magnetite nanofluids were prepared *via* a coprecipitation method and their photothermal characteristics were measured under direct sun. Effects of magnetite particle volume fraction and magnetic field on the temperature enhancement as function of irradiation time were analyzed. The results showed that the photothermal characteristics could be enhanced with magnetite nanoparticles used in nanofluids at lower particle volume fraction. In addition the, the photothermal characteristics of magnetic nanofluids (MNFs) were found to be related to magnetic field.**

***Keywords:* Photothermal characteristics; Magnetic nanofluids; solar collectors, solar thermal applications, Magnetic field.**

1. INTRODUCTION

Recently, the volumetric radiative heat transfer provided by nanofluids has shown increasing interests as an effective way of solar thermal energy conversion [1-6]. The use of these composite fluids in solar collectors such as Direct Absorbing Solar Collectors (DASCs) could lead to higher effective emissivity and /or absorptivity relative to conventional area-based surface radiative heat transfer affecting their thermal efficiency [1, 7]. As the solar radiation interacts with participating media such as nanofluids, attenuation of the solar radiant energy takes place through absorption and or scattering mechanisms leading to its conversion into the thermal energy. The collected energy could be utilized for water heating applications, electricity generation, biomass gasification etc.

In addition, the thermal effectiveness of the solar collectors could be affected by the thermophysical properties of the used nanofluids such as the thermal conductivity, heat capacity, viscosity etc. The thermal effectiveness of the systems with nanofluids is therefore related to the characteristics of the nanoparticles, viz., the morphology, chemical composition, the volume fraction etc.[8].

Magnetic nanofluids (MNFs), suspension of magnetic nanoparticles in nonmagnetic carrier fluids, constitute a special class of nanofluids which exhibit both magnetic and fluids properties[9]. When used in presence of moderate magnetic fields, MNFs exhibit a high thermal conductivity which makes them of crucial interest in thermal energy applications. Some experimental[10-12] and theoretical investigations[9, 13] on the thermal conductivity of MNFs have shown that the thermal conductivity of MNFs is higher than that of the common heat transfer fluids (such as water, engine oil, etc.) both in zero and applied magnetic fields. some few studies have proved an enhanced spectral absorption of MNFs for white light ($\lambda=500\ nm$- $\lambda=600\ nm$) showing their potential application in solar thermal applications[1, 14]. The use of MNFs as heat transfer media in solar thermal systems could offer a possibility of enhancing their efficiency through radiative properties (absorption, scattering, etc.) and thermophysical properties such as thermal conductivity enhancement, heat capacity etc. [1, 9]. However, theoretical and experimental investigations on the photothermal characteristics (viz. absorptivity and emissivity) of the MNFs are needed to design and characterize these systems both in zero and applied magnetic fields. This paper investigated the enhancement of photothermal characteristics of magnetite (Fe_3O_4) based MNFs in zero and external magnetic fields. Effects of particle volume fraction and magnetic field were analysed for water and kerosene based magnetite nanofluids.

II. EXPERIMENTAL

A. Magnetite nanoparticles synthesis

Magnetite (Fe_3O_4) nanoparticles used in this study were synthesized *via* a co-precipitation method. $FeCl_2.4H_2O$ and $FeCl_3.6H_2O$ in a mole ratio of 1:2 were mixed in a round bottom flask.

The mixture was stirred vigorously and a stoichiometric quantity of sodium hydroxide solution was added progressively. Then, the precipitation process occurred immediately after about 10 minutes, promoting a color change in the solution to dark-black which is the characteristic color of the magnetite. The magnetite (Fe_3O_4) nanoparticles could be formed according to the following equation:

$$FeCl_2 + 2FeCl_3 + 8NaOH \rightarrow Fe_3O_4 + 4H_2O + 8NaCl \quad (1)$$

After the reaction was complete, the synthesized magnetite nanoparticles were washed several times with deionized water and alcohol and dried under vacuum for 4 hours at a temperature of 80^0C before characterization.

B. Preparation of magnetic nanofluids

a) Kerosene based magnetic nanofluids

In this study, kerosene and water based magnetite nanofluids were used. Kerosene based magnetite nanofluids were prepared *via* a bottom up technique[15]. The magnetite nanoparticles were firstly treated with oleic acid ($CH_3(CH_2)_7CH{=}CH(CH_2)_7COO^-$) and then MNFs with different volume fractions were obtained by dispersing oleic acid (OA)-coated magnetite nanoparticles in engine oil. An excess of oleic acid was soaked in the reaction flask immediately after the magnetite nanoparticles synthesis. The mixture was heated to about 90^0C, and stirred at 200rpm for one hour. During the particle surface modification process the pH was kept between 9 and10. After one hour, the mixture was cooled and acidified to p^H=5 using nitric acid (HNO_3). The oleate ions ($CH_3(CH_2)_7CH{=}CH(CH_2)_7COO^-$) produced could chemisorb strongly on the magnetite nanoparticles and the precipitate could coagulate and fall out of the solution[16]. After particle surface modification, the OA-coated magnetite nanoparticles were separated from the solution using a permanent magnet, washed 5 times with alcohol and deionized water, respectively, and dried under vacuum at 80^0C for 5 hours. The MNFs with different volume fractions were obtained by dispersing the OA-coated magnetite nanoparticles in kerosene. The particle volume fractions were calculated by the following formula [12, 13]:

$$\varphi_p = \frac{\omega_p \times \rho_f}{\omega_p \times \rho_f + \rho_p \left(1 - \omega_p\right)} \quad (2)$$

where ω_p is the weight particle fraction, ρ_p and ρ_f are the densities of dispersed phase (i.e., Fe_3O_4) and carrier fluid, respectively.

The dispersion was performed using a mode PT18-10 ultra-turrax homogenizer (Kika-Werk, Germany). Dried OA-coated NPs were mixed with kerosene, stirred for 10 minutes and ultrasonicated for 1 hour. After ultrasonication, the suspension was further stirred vigorously for 30 minutes and the obtained MNFs samples were collected.

b) Water based magnetic nanofluids

The water based MNFs were prepared by dispersing citric acid-coated magnetite nanoparticles in deionized water. Magnetic nanoparticles were mixed with deionized water and ultrasonicated for 30 minutes to break the possible aggregates/agglomerates due to the magnetic and the surface energy relevant to this particle size. 10% of citric acid was added slowly and stirred at 200rpm for 1 hour. The surface modification process was performed at a p^H=10.

After the surface modification, the product was cooled at room temperature and the citric acid-coated Fe_3O_4 nanoparticles were separated from the solution using a permanent magnet. In order to avoid any contaminant ions, the product was washed for several times with alcohol and deionized water, respectively. MNFs with different volume fractions were obtained by dispersing the citric acid-coated magnetite nanoparticles in deionized water at a p^H=5. The citric acid could bind to magnetite particles surface by chemidsorption of the carboxylate (i.e., citrate) ions. The surface structure created by later ions could create some steric and or coulombian inter-particle repulsion that can overcome the attractions of Fe_3O_4 nanoparticles due to magnetic and van der Waals interactions[16, 17] ensuring the stability of the prepared MNFs.

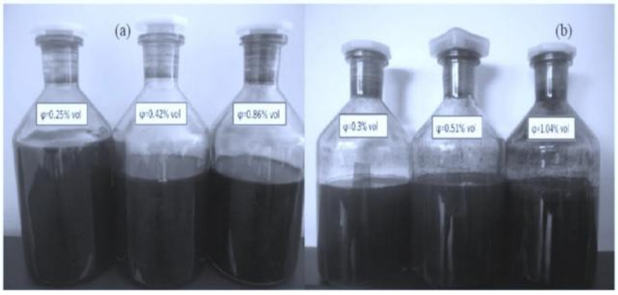

Fig. 1 Representative sample of the prepared (a) kerosene and (b) water based MNFs

The representative samples of the prepared MNFs are presented on fig. 1. All the samples appeared stable for one month. Obviously, the MNFs with a lower particle volume fraction could be more stable due to the inter-particle separation.

B. Photothermal characteristics measurement

In this study, the temperature and temperature enhancement were recorded using a mode 4-2 digital temperature meter with a Pt100 probe (PHYWE, Germany). To analyse the photothermal characteristics of MNFs was designed as shown of the figure below (fig. 2). The measurements were directly conducted under the sun at the site of the College of Science and Technology, University of Rwanda (Kigali Campus) which is located at 1.9439^0 S and 30.0594^0 E. To collect the maximum solar irradiance, the measuring device was mounted at 30^0 south. This design allowed a magnetic field $\vec{B}$ perpendicular to the direction of the sunlight. Due to the number of samples and the weather conditions, measurements were conducted within three consecutive days and these effects were taken into consideration by analysing both the recorded temperatures and the temperature enhancement. MNFs were sealed in quartz tubes (d =26 mm, h=150 mm). The tubes were arranged in an insulation box. Insulation materials were put under and between the tubes. To ensure the same endothermic and heat transfer area each tube was filled with MNFs of the same amount[17].

Temperature of the MNFs were measured and recorded in real time with Pt100 probes immersed in the samples and a horizontal magnetic field was generated by a pair of FeNdB-magnets (see fig. 2(b)). The magnetic field intensity, which was measured by a digital teslameter (PHYWE, Germany), was adjusted by varying the distance between the magnets. All the measurements were performed between 23 ^{0}C and 27 ^{0}C.

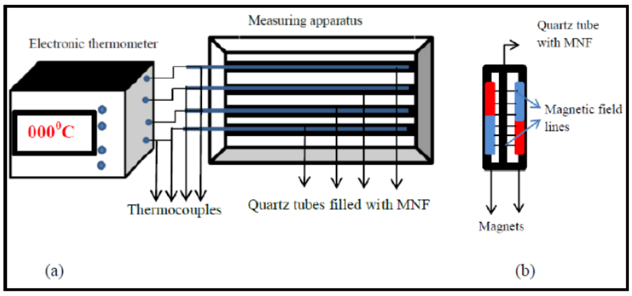

Figure 2 MNFs Photothermal characteristics measurements system: (a) Measuring apparatus and (b) Arrangement for photothermal characteristics measurement in magnetic field

III. RESULTS AND DISCUSSION

A. Structural analysis of magnetic nanoparticles

The phase composition and the physical particle size of synthesized product were examined by a model D-37079 Göttingen diffractometer with a Cu Kα radiation ($1.54178\ nm$) (PHYWE System GmbH & Co. KG Robert-Bosch-Breite 10).

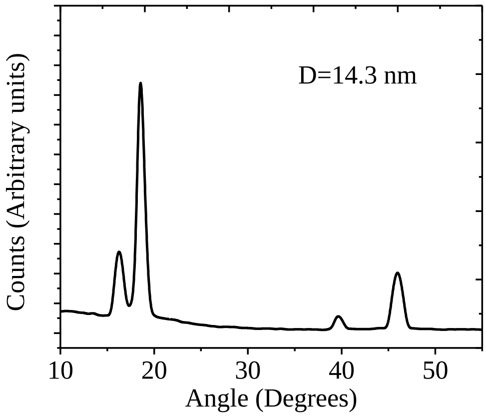

Fig. 3 X-ray diffraction patterns of synthesized magnetite nanoparticles

The samples were continuously scanned in the range of range of 10-60^0 and the obtained patterns were analyzed using a mode MDI Jade 5. Fig. 2 shows the XRD pattern of the synthesized magnetite nanoparticles.

Broad picks typical for nanoparticle were observed and all the reflection peaks were well indexed with FCC spinel structure without impurity picks. The crystallite size *D* of the samples was calculated using the well-known Sherrer's equation:

$$D = \frac{K\lambda}{B\cos\theta} \tag{3}$$

with $K = 0.9$ the shape factor, *1.54178nm* the wavelength of the X-ray beam used, B is the peak width and θ the Bragg angle, respectively. The estimated crystallite size (i.e. Average physics size) for the prepared sample was *14.3nm*.

B. Photothermal characteristics of MNFs

Photothermal effect refers to the photo-excitation of a material resulting in the production of thermal energy (Heat). In this work, the temperature increase and the corresponding temperature enhancements were analysed in zero and applied magnetic field. As aforementioned, the measurements were conducted under direct sun and the temperature changes in MNFs were recorded as function of solar irradiation time. Effects of volume fraction in zero and applied MNFs were analysed. The temperature enhancement was calculated as the difference of recorded temperature in MNFs and that recorded in the corresponding carrier fluids at a given time.

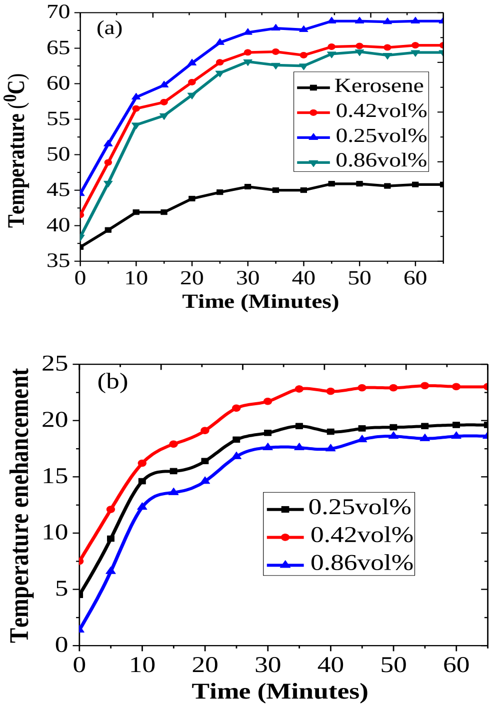

Fig. 4 Photothermal characteristics of kerosene based MNFs: (a) Temperature and (b) Temperature enhancement as function of solar irradiation time at different volume fractions.

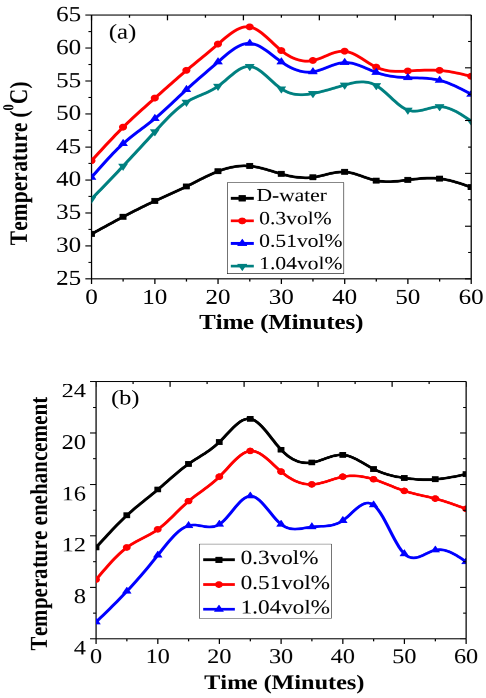

Figure 5 Photothermal characteristics of water based MNFs: (a) Temperature and (b) Temperature enhancement as function of solar irradiation time at different volume fractions

a) Effect of volume fraction

Figures 4(a), 4(b) show the temperatures and the corresponding temperature enhancements as function of irradiation time in kerosene based MNFs at different volume fractions, respectively. It is seen that the temperature of MNFs increases more rapidly than that of the corresponding carrier fluids (i.e., kerosene). The same trend was observed in water based MNFs. Figures 5(a) and 5(b) show the temperatures and the corresponding temperature enhancements as function of irradiation time in water based MNFs at different volume fractions, respectively. Clearly, the temperature increases with decreasing the volume fraction and the highest temperatures recorded were 66.8 ^{0}C at a volume fraction of 0.25 vol% (for the kerosene based MNF) and 63.2 ^{0}C at a volume fraction of 0.3 vol% (for water based MNFs), respectively. Obviously, the addition of magnetite nanoparticles in different carrier fluids could improve their thermal energy absorption properties leading to a rapid increase of the temperature as function of irradiation time[18]. Magnetite nanoparticles could absorb almost all solar energy in the volume of the MNFs and disseminate in the surrounding medium [14, 18, 19]. The rapid increase of temperature in MNFs could also be attributed to the improved thermal conductivity of MNFs compared to the corresponding carrier fluids [10, 11, 20, 21].

Clearly, the temperature increases with decreasing the volume fraction and the highest temperatures recorded were 66.8 ^{0}C at a volume fraction of 0.25 vol% (for the kerosene based MNF) and 63.2 ^{0}C at a volume fraction of 0.3 vol% (for water based MNFs), respectively.

3.2.2 Effect of magnetic field

To analyze the effect of magnetic field on the Photothermal properties, the temperature enhancement were recorded within 30 minutes at different magnetic fields for both water and kerosene based MNFs. Figures 6(a), 6(b) show the recorded temperature enhancement in $\varphi_p = 1.04$ vol% and $\varphi_p = 0.3$ vol% water based MNFs as function of time, respectively. It is seen that the temperature enhancement increases with the increase of magnetic field perpendicular to the solar irradiance direction. Obviously, the highest temperature enhancement was recorded at lower particle volume fraction. The increase of temperature enhancement with the increase of magnetic field intensity was also observed in kerosene based MNFs. Figures 7(a) and 7(b) show the recorded temperature enhancements in $\varphi_p = 0.86\% vol$ and $\varphi_p = 0.25$ vol% kerosene based MNFs. The applied magnetic field could enhance both the thermal conductivity and the optical properties of MNFs leading to improved photothermal characteristics. In the presence of magnetic field, the highest temperature enhancement was 24.7 for water based MNFs and 22.3 for kerosene based MNFs at particle volume fractions of 1.04 vol% and 0.86 vol% respectively.

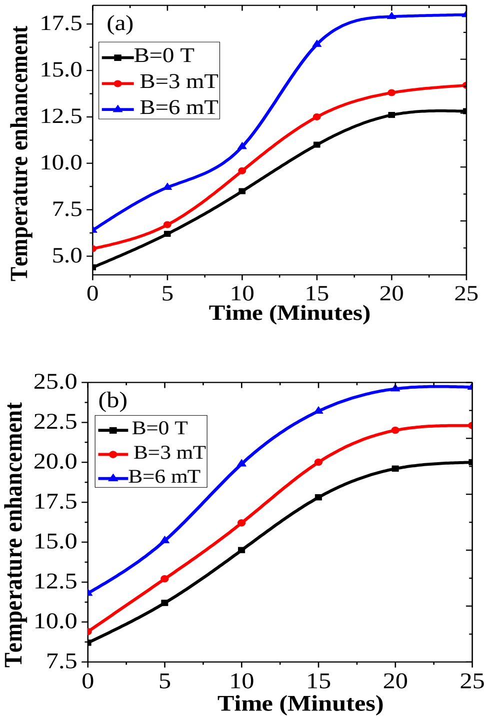

Fig. 6 Temperature enhancement as function of solar irradiation time in (a) $\varphi_p = 1.04$ vol% and (b) $\varphi_p = 0.3$ vol% water based MNFs at different magnetic field intensities.

This observation was in agreement with previous work on DASC using conventional (nonmagnetic) nanofluids as heat transfer media. As the particle volume fraction of MNPs increases, the amount of solar radiation penetrating in the fluid reduces due to multiple scattering and reflections on the particles.

At a higher particle volume fraction, the solar irradiance could be absorbed on the fluids surface rather than in volume, reducing the temperature enhancement of MNFs [3, 4].

When a magnetic field perpendicular to the sunlight direction was applied, a further increase in temperature enhancement was observed. Under the same conditions of particle size and magnetic field, the temperature enhancement increased with the decreasing particle volume fraction and higher temperature enhancements were obtained at $\varphi_p = 0.25$ vol% and $\varphi_p = 0.3$vol% for water and kerosene based MNFs, respectively (see Figures 6 and 7). The plausible cause of the increase of the temperature enhancement with increasing magnetic field could be the formation of the chain-like magnetic nanoparticles formation in MNFs[21].

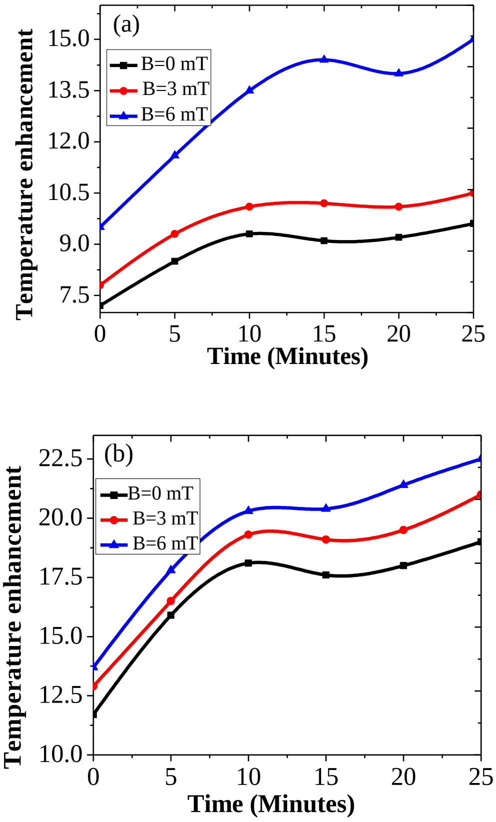

Figure 7 Temperature enhancements in (a) $\varphi_p = 0.86$ vol% and (b) $\varphi_p = 0.86$ vol% kerosene based MNFs as function of irradiation time at different magnetic field intensities.

The application of magnetic field could result in the compression of the space occupied by magnetic nanoparticles, leading to an enhancement of absorption of the incident irradiance in the volume of the MNF [1, 6, 13, 21]. These findings suggest that the use of both magnetic field and MNFs in solar thermal collector could contribute cooperatively to the efficiency solar thermal collectors.

IV. CONCLUSION

Photothermal properties of magnetic water and kerosene based MNFs could be enhanced when an applied magnetic field was perpendicular to the sunlight direction. Magnetite nanoparticles were prepared *via* a co-precipitation method and stable MNFs with different volume fractions were prepared by dispersing OA-coated and citric acid-coated magnetic nanoparticles in kerosene and water, respectively. Effects particle volume fraction and externally applied magnetic field on the photothermal characteristics were analysed. The temperature enhancement increased with the increase of magnetic field intensity and an optimum enhancement was achieved at 6mT. In addition, higher photothermal characteristics were observed at lower particle volume fraction in the magnetite nanofluids both in zero and applied magnetic fields. These results showed a great potential of using MNFs in solar thermal collectors as heat transfer media.

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