Application of ZnO/TiO$_2$ Nanocomposite for the Improvement of Heat Transfer Coefficient in Tube Heat Exchangers

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ABSTRACT
The potential of nanofluids for the improvement of heat transfer coefficient in various heat exchange equipment has been considered and studied as a major application during recent decades. In this research, heat transfer coefficient of ZnO, TiO$_2$ and ZnO/TiO$_2$ nanofluids in a shell and tube heat exchanger has been studied experimentally. ZnO nanoparticle was synthesized through precipitation technique. Sol-gel and mechanical techniques were employed to synthesize the ZnO/TiO$_2$ nano-composite. Particle size analysis (PSA), XRD, FTIR and FE-SEM techniques were used to characterize ZnO/TiO$_2$ nano-composite. Based on the results, the nanofluid heat transfer coefficient was enhanced by increasing the nanofluid concentration and temperature. The heat transfer coefficient of TiO$_2$ was higher than that of ZnO nanofluid and the heat transfer coefficients of nano-composites were higher than that of ZnO and TiO$_2$ nanofluids and the base fluid. Also, the heat transfer coefficient and the overall heat transfer coefficient were increased 3.79 to 9.09 times and 4.27 to 9.14, respectively by increasing the nano-composite content.

Keywords: Heat Transfer Coefficient, Nanocomposite, Nanofluid, Shell and Tube Heat Exchanger
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INTRODUCTION
Heat exchangers are among the most important unit operations in processing industries playing an important role in the management of energy requirements. In order to increase the efficiency and improve the rate of heat transfer in heat exchangers, various techniques such as increasing the heat transfer area and improvement of heat transfer coefficient are proposed. Higher heat transfer coefficient fluids can replace conventional fluids (e.g. water and ethylene glycol), which have led to many research studies during the last decade. Nanofluids are among the very recent alternatives for this purpose. Addition of nano-scale particles into conventional base fluids can cause a significant enhancement in thermal properties [1, 2]. Several Nanoparticles are synthesized and added to various base fluids in order to investigate their effect on thermal properties such as TiO$_2$, ZnO, Al$_2$O$_3$, and CuO among many others.

Many research activities have been performed on this subject during the last decade. Madhesh et al. investigated the convective heat transfer, pressure drop, friction factor and the rheological properties of Cu-TiO$_2$ nanofluid in a heat exchanger [3]. The results showed an increase in Nusselt number by 52%, the convective heat transfer coefficient by 49% and the overall heat transfer coefficient by 68%. Farajollahi et al. measured thermal transfer properties of water/Al$_2$O$_3$ and water/TiO$_2$ nanofluids in a shell and tube heat exchanger under turbulent flow conditions [4]. They studied the effect of Peclet number and nanofluid concentrated on heat transfer characteristics. Based on the results, the heat transfer behavior is generally improved. Also, heat transfer characteristics of

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TiO₂ nanofluid with an optimum concentration (0.3 vol. %) and a certain peclent number (20000-60000) was higher than Al₂O₃ nanofluid. Anoop et al. reported the effect of SiO₂/water nanofluid on the overall heat transfer coefficient and pressure drop in plate and shell and tube heat exchangers [5]. The results revealed that heat transfer coefficient was a function of flow rate and nanofluid concentration in both heat exchangers. Albadr et al. investigated the effect of Al₂O₃ concentration on the convective heat transfer and friction factor in a shell and tube heat exchanger under counter-current and turbulent flow conditions [6]. The results indicated that the heat transfer coefficient was enhanced by increasing the nanofluid concentration and mass flow rate. Lotfi et al. studied the effect of MWCNT/water nanofluid heat transfer in a shell and tube heat exchanger [7]. The results showed an increase in the heat transfer coefficient compared with the base fluid (water). Zeinali Heris et al. compared the convective heat transfer of CuO/water and Al₂O₃/water nanofluids through a circular tube under laminar flow conditions with constant wall temperature [8]. The results indicated that the increase in nanofluid concentration and peclent number increased the heat transfer coefficient. Also, the enhanced heat transfer of Al₂O₃/water nanofluid was higher than that of CuO/water. Sajadi et al. reported the heat transfer and pressure drop characteristics of ZnO/water nanofluid in a circular tube heat exchanger under constant wall temperature conditions [9]. The results indicated that nanoparticle concentration can increase heat transfer coefficient, overall thermal performance and pressure drop. Tasi et al. investigated the heat transfer of Au/water nanofluids [10]. Experimental results demonstrated that under identical conditions, nanofluid results in a significant reduction in thermal resistance of tubing compared to the base fluid. Ghozatloo et al. studied the enhancement of water/graphene nanofluid heat transfer in a shell and tube heat exchanger under laminar flow conditions [11]. The results indicated that the convective heat transfer coefficient was enhanced by increasing nanofluid concentration and temperature. Godson et al. studied the heat transfer of Ag/water nanofluid in a shell and tube exchanger [12]. They studied the influence of inlet temperature, Reynolds number, nanofluid concentration and mass flow rate on LMTD, effectiveness, convection heat transfer and pressure drop. The results showed that enhancement in the heat transfer coefficient, pressure drop, LMTD and effectiveness is performed by increasing nanofluid concentration and Reynold's number.

Based on the results reported in the literature, TiO₂ is a promising material for heat transfer improvement which is previously studied as a photo-catalyst [13, 14] and membrane material both as support [15] and the top-layer [16]. So, this study is aimed at the improvement of TiO₂ in a nanocomposite structure with ZnO which is also a promising material for heat transfer coefficient enhancement. ZnO/TiO₂ nanocomposite is synthesized through various techniques and evaluated in several applications such as solar cells [17], photo-catalysts [18], sensors [19] and many others. In this study, sol-gel and mechanical techniques are implemented for the synthesis and preparation of ZnO/TiO₂ nanocomposite. Corresponding nanofluids were prepared and used in a shell-tube heat exchanger for investigating the effect of concentration on the heat transfer and overall heat transfer coefficient at various temperatures. It is expected that ZnO/TiO₂ nanocomposite can be a good choice for heat transfer enhancement in tube heat exchangers.

**EXPERIMENTAL**

**Materials and methods**

TiO₂ nanoparticles were purchased from Tecnan Co. The average particle size and specific surface area was 10-15 nm and 100-150 m²/g, respectively. Zinc acetate, isopropyl alcohol, NaOH, ammonium hydroxide, ethanol and methanol were purchased from Merck Co. All solutions were prepared using deionized water.

**Synthesis of ZnO nanoparticles**

ZnO nanoparticles were synthesized through precipitation technique. 0.1 g of zinc acetate and isopropyl alcohol were mixed at 65 °C for 15 min by magnetic stirrer. Then the mixture was added to 12 ml of isopropyl alcohol placed in a water bath at 0 °C. 15 ml of NaOH (0.05M) in the water bath (0°C) was added drop-wise into the final solution and stirred at a high stirring rate. The produced solution was maintained under 65 °C for 5min. Finally, it was washed using ethanol, methanol and water 3 times and then cooled at the ambient temperature [20].

**Synthesis of ZnO/TiO₂ nanocomposite**

**Preparation of ZnO sol**

1ml of ammonium hydroxide was mixed with 100ml of zinc chloride (0.2 M) until a white precipitate of zinc hydroxide was formed. Adding
more ammonium hydroxide (1 ml) leads to the dissolution of the precipitate [21].

Preparation of TiO2 sol

1 ml of titanium isopropoxide was dissolved into isopropyl alcohol (32 ml) and mixed with magnetic stirring for 2 h. 0.05 ml of citric acid was added to the final solution to prepare a uniform sol [13, 14]. ZnO and TiO2 sols were mixed to obtain the nanocomposite in a 50-50 molar ratio and then sonicated for 5 min. For drying the ultimate product, it was kept at room temperature for 72 h. At last, the prepared nanocomposite was calcined at various temperatures (500 °C, 600 °C & 800 °C) for 1 h. TiO2/ZnO nanocomposites with 25-75 mol%, 50-50 mol% and 75-25 mol% proportions were obtained by mixing appropriate proportions of the as-prepared sols.

Preparation of ZnO/TiO2 nanocomposite by mechanical technique

In order to prepare the nanocomposite by mechanical method, ZnO and TiO2 nanoparticles with 25-75 mol%, 50-50 mol% and 75-25 mol% were mixed with ethanol for 15 min by a high speed magnetic stirrer and then sonicated for 30 min. The mixture was filtered and washed with water and ethanol 3 times and dried at room temperature for 24 h. Finally, the obtained nanopowder was sintered at 550 °C (with a 4.7 °C/min. rate) for 2 h.

Preparation of ZnO and TiO2 nanofluids

To prepare ZnO and TiO2 and ZnO/TiO2 nanofluids, nanoparticles with 0.001, 0.01 and 0.1 wt. % were added to deionized water (2:l) and stirred for 15-20 min. At last, the mixture was sonicated (100 W) for 45 min to disperse the nanoparticles in the base fluid.

Heat exchange experiments

To study the effect of nanofluid on heat exchange characteristics, an experimental setup was designed. The heat exchanger is a shell and tube type with counter-current flow, including both heating and cooling loops. The nanofluid flowing through the shell consists of a coolant roll and deionized water through the tubing was the heating medium. The cooling loop consists of a pump, hot fluid tank, heater, temperature controller and measurement of tube temperature inlet and outlet systems. The cooling loop consists of a pump, nanofluid reservoir tank, temperature controller and the measurement of shell temperature inlet and outlet systems. The shell and tube were made of pyrex and copper, respectively. The details of the system can be found elsewhere [22]. To measure the heat transfer coefficient (h) and the overall heat transfer coefficient (U), experiments were carried out in this heat exchanger with counter-current flow at two temperatures (50 °C and 60 °C) for water/water, water/nanoparticle and water/nanocomposite systems.

RESULTS AND DISCUSSION

Characterization

XRD Analysis

The XRD pattern of ZnO/TiO2 nano-composite is given in Fig. 1. The sharp diffraction peaks show the nano-crystalline structure. Sharp characteristic peaks are observed at 2θ = 25.5, 27.5, 32.5, 35.5, 41.5, 44.5, 48.5 and 57 for the ZnO-TiO2 nanocomposite [23-25]. Compared with XRD patterns for ZnO and rutile TiO2 phase, whole peaks are shifted before 2θ = 60 while higher 2θ peaks are also observed in ZnO and TiO2 separately. The characteristic peaks of each phase are also seen in the nanocomposite while other peaks disappeared which show a change of morphology in the nanocomposite.

Fig.1. XRD patterns for ZnO/TiO2 (30/50 mol %) nano-composite, ZnO and TiO2 (Rutile).
Particle size analysis (PSA)

The ZnO/TiO$_2$ nano-composite is characterized by particle size analyzer (Horiba LB-360) (0.001-6μm). The PSD of TiO$_2$/ZnO nano-composite is given in Fig. 2 which shows an average particle size of 40nm with a uniform size distribution.

FTIR analysis

The FTIR spectrum of the synthesized nanocomposite was recorded in the range of 400–4000 cm$^{-1}$ shown in Fig. 3. The peaks at 3600 cm$^{-1}$ and 1607 cm$^{-1}$ were attributed to OH stretching and bending of water, respectively. The band at around 1406 cm$^{-1}$ was attributed to the vibration mode of Ti–O bond. The absorption peaks at ~ 600 cm$^{-1}$ are also related to Zn–O bonding. The peak at 974 cm$^{-1}$ is responsible for Ti–O bonds [26].

SEM micrograph

Fig. 4 represents the SEM micrograph of the ZnO/TiO$_2$ nano-composite. Based on the Figure, semi-spherical nanoparticles are observed in the ~100 nm size range and smaller.

Heat Exchange Experiments

Effect of nanofluid concentration

Figs. 5 and 6 show the effect of TiO$_2$ and ZnO nanofluid concentration on the heat transfer.
coefficient and the overall heat transfer coefficient in the heat exchanger at 50ºC. The results indicate that the heat transfer coefficient is increased by increasing the nanofluid concentration due to the enhanced thermal conductivity and reduced specific heat capacity and thermal boundary layer thickness. The heat transfer coefficient and the overall heat transfer coefficient are 0.5865 W/m²°C and 0.5895 W/m²°C for the water/water system, respectively. A significant increase is observed using nanofluids in the shell side. The heat transfer coefficient of TiO₂ nanofluid is usually higher than that of ZnO, except for 0.1 wt. % which may be due to the high stability of ZnO at higher concentrations. It must be mentioned that nanofluid stability is a function of several factors such as size, size distribution, solvent type, temperature, zeta potential and many others which must be calculated per case. Usually, heat transfer coefficients are enhanced by adding stable nanoparticles to the base fluid and this continues up to the concentration that instabilities are observed which results in the reduction of heat transfer coefficient thereupon. The same trend is also observed for the overall heat transfer coefficient.

Table 1 shows the heat transfer coefficient and overall heat transfer coefficient enhancement compared with the base (water/water) fluid. The maximum heat transfer coefficients occurred at 0.01 wt. % of ZnO and TiO₂ in their corresponding nanofluids. Higher than 0.01 wt.% a slight enhancement occurs in the heat transfer coefficient in both nanofluids which can be ignored due to economic reasons. Experiments performed higher than 0.1 wt. % show that heat transfer coefficient reduction is observed due to agglomeration.
Fig. 6. Effect of TiO$_2$ and ZnO nanofluid concentrate on the overall heat transfer coefficient.

Fig. 7. Effect of temperature on the heat transfer coefficient.

Fig. 8. Effect of temperature on the overall heat transfer coefficient.
Effect of temperature

The effect of temperature on heat transfer coefficient and overall heat transfer coefficient are shown in Figs. 7 and 8. As the temperature is increased, the heat transfer coefficient in raised at all nanofluid concentrations. Heat transfer coefficient variations of TiO$_2$ and ZnO nanofluid in comparison with the concentrations at 60 ºC is higher than that of 50 ºC. The variations of heat transfer coefficient of ZnO nanofluid are higher than that of TiO$_2$ nanofluid.

The heat transfer coefficient enhancement compared to the original system (water/water) is given in Table 2. The maximum heat transfer coefficient of nanofluid is occurred at 25/75 mol% of ZnO/TiO$_2$ nano-composite.

Effect of nanocomposite concentration

The heat transfer coefficient and the overall heat transfer coefficient of the nanofluids obtained from the nanocomposite prepared by sol-gel and mechanical techniques with 75-25%, 50-50% and 25-75% of ratios of TiO$_2$/ZnO are illustrated in Figs. 9 and 10, respectively. The results reveal that by increasing the nanocomposite concentration, heat transfer and overall heat transfer coefficients are increased, which is apparently higher than that of ZnO and TiO$_2$ nanoparticles separately. Based on the results, the heat transfer coefficients of nanofluids obtained from the nanocomposite by sol-gel is higher than that of the prepared nanocomposite by mechanical technique which is possibly due to higher crystallinity of the product from precipitation compared with the mechanical method.

CONCLUSIONS

In this work, ZnO nanoparticle and ZnO/TiO$_2$ nanocomposite were synthesized via precipitation and sol-gel techniques. The nanofluid is prepared and characterized by PSA, XRD, FTIR and FE-SEM analyses. Effect of nanofluid concentration and temperature on heat transfer coefficient and overall heat transfer of the nanofluid in shell-tube heat exchanger are investigated. The results show that:

- The heat transfer and overall heat transfer coefficients of nanofluids are higher than that of the base fluid enhanced by nanofluid concentration.
- The nanofluid heat transfer coefficient variations

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<th>Table 2. Heat transfer coefficient enhancement compared to the base fluids.</th>
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<td>Heat transfer coefficient (W/m$^2$C), Overall heat transfer coefficient (W/m$^2$C)</td>
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<td>ZnO/TiO$_2$ ratio: 25-75%, 50-50%, 75-25%</td>
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<td>Nanofluid conc. (wt. %)</td>
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at 60 °C are higher than that of 50 °C.
- The heat transfer coefficients of TiO$_2$ nanofluid are higher than that of ZnO but the heat transfer coefficients variations of ZnO nanofluid are higher than that of TiO$_2$.
- The heat transfer coefficients of nanocomposites are higher than that ZnO and TiO$_2$ nanofluids and the base fluid. The highest heat transfer coefficients occur at the ZnO/TiO$_2$ = 25/75% molar ratio.

CONFLICT OF INTEREST
The authors declare that there is no conflict of interests regarding the publication of this manuscript.

REFERENCES