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Experimental investigation of conduction and convection heat transfer properties of a novel nanofluid based on carbon quantum dots

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ABSTRACT

So far, many studies have been conducted on heat transfer nanofluids and various nanofluids have been synthesized and evaluated by different nanoparticles. In the present research, the use of biodegradable carbon quantum dots (CQDs) to synthesize heat transfer nanofluids was investigated for the first time. In fact, CQDs are a new generation of carbon nanoparticles and one of the advantages of which is their very small size that facilitates the prepared of nanofluids at very low concentrations with high stability. In the present research, CQDs were synthesized based on microwave method using commercial ammonium hydrogen-citrate as precursor. The nanofluid samples were synthesized based on car radiator coolant and CQDs at the concentrations of 100, 200, 500, and 1000 ppm. Thermal conductivity (k) and convection heat transfer (h) coefficients were investigated as the main features of the fluid's heat transfer characteristics. The obtained results for 200-ppm concentration indicated the improvement of k and h by 5.7% and 16.2%compared to the base fluid, respectively. Besides, the synthesized nanofluids had also significant stability and very low cost which are of great importance for industrial applications. Finally, the heat transfer process in the 200-ppm nanofluid was simulated by Ansys Fluent software.

1. Introduction

Nanofluids are the suspensions obtained by mixing solid nanoparticles with a liquid fluid, which could improve the properties or lead to novel properties. Over the past two decades, numerous scientific studies have been conducted on nanofluids [1–27]. The general objective of these studies has been to investigate the effect of various carbon, metal, metal oxide, and polymer nanoparticles, such as Al_2O_3, CuO, TiO_2, SiC, TiC, Ag, Cu, Au, Fe, C_{60}, MWCNT, SWCNT, graphene, etc., on various fluids such as water, ethylene glycol, lubricant oils, liquid fuels, as well as a variety of heat transfer fluids. Although significant progress has been achieved of in this field, there are concerns on the potential risks of the products and applications of this technology [28–32]. Since metal and metal oxide nanoparticles might have toxic effects on the environment, the use of biodegradable carbon nanoparticles can partly reduce such concerns.

Carbon quantum dots, as a new generation of carbon nanoparticles, have exhibited a great potential for being used as multi-purpose nanomaterials for a wide range of applications. CQDs are a newly emerged class of carbon nanomaterials that have gained much interest among researchers. Beside fluorescent and optical properties, CQDs have favorable advantages such as low toxicity, environmentally friendly, low cost, and simple synthesis methods [33–36]. Furthermore, the possibility of passivation and surface functionalization of CQDs facilitates controlling their physicochemical properties. Since being accidently discovered by Xu et al. [37] during separation and purification of single-walled nanotubes, CQDs have found several applications in the fields of chemical sensors, biosensors, biological imaging, nano-medicine, photocatalyst, and electrical catalysts [38–42].

Among the challenges in the field of nanofluids, lack of long-term stability, base fluid properties variations, high cost of nanomaterials, and possible environmental risks can be mentioned. Thus, the present research is aimed to introduce the biodegradable carbon quantum dots as a new type of nanoparticles to overcome the problems of nanofluids. Accordingly, for the first time in the present research, biodegradable carbon quantum dots was used to synthesize heat transfer nanofluids

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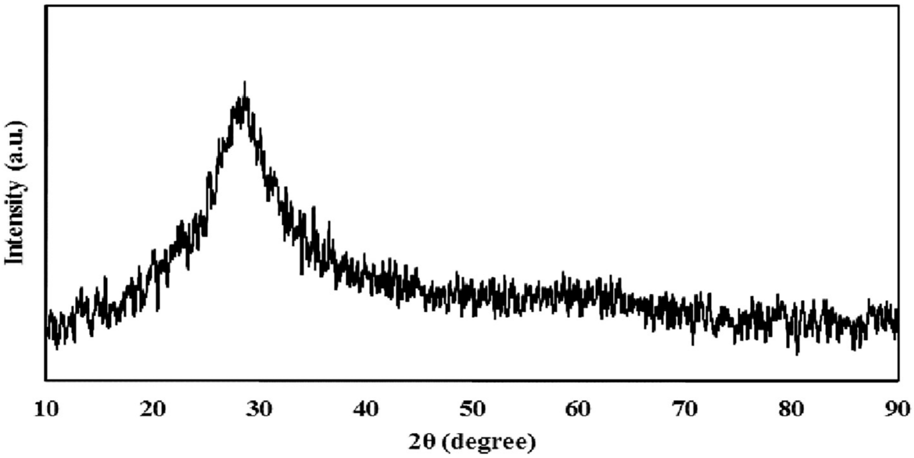

Fig. 1. X-ray diffraction (XRD) pattern of CQDs.

and the heat transfer capability of the radiator coolant-CQDs nanofluids was investigated.

2. Material and methods

2.1. Synthesis and characterization of CQD nanoparticles

First, the CQD nanoparticles were synthesized using microwave method. For this purpose, 2 g of commercial ammonium hydrogen-citrate precursor ($C_6H_{14}N_2O_7$) was completely dissolved in 10 mL of deionized water. Then, the suspension was put into a domestic microwave at 180 °C for 8 min. After the completion of reaction, the obtained dried product was cooled down to ambient temperature. The final product contained 1.65 g of CQDs collected from the reaction container. The X-ray powder diffraction (XRD) of the synthesized CQDs is shown in Fig. 1, which indicates the amorphous nature along with very low crystallinity of CQDs. Fig. 2 shows the infrared spectrum of CQDs, which has a completely different pattern from bulk graphite or other merely carbonic structures. With regard to the presence of oxygen and nitrogen heteroatoms in the structure of CQDs, the absorptions associated with tensile and bending vibrations special for the bonds formed between carbon and heteroatoms are predictable. With regard to Fig. 2, the absorption band of 1730 cm^{-1} region is related to C═O tensile vibration. Wide absorption in 2700–3600 cm^{-1} region is due to the presence of O—H groups related to HO—C═O or C-OH; however, it seems that the N—H tensile absorption has also been overlapped in this region. Specification absorption of C—O has also appeared in 1192 cm^{-1} region corresponding to tensile C—N bond. Tensile absorptions related to C═C and N—H bending are also observed in 1635–1680 cm^{-1} region.

Also, spectrum of the CQDs' emission under 360-nm excitation wavelength is shown in Fig. 3. As can be seen, the emission wavelength at around 460 nm, is in the range of blue spectrum. The image specified at the right corner of Fig. 3 represents the fluorescence property of CQDs compared to the base solvent (deionized water) under UV lamp with wavelength of 365 nm.

The size of CQD nanoparticles was investigated using DLS and HRTEM tests. Figs. 4 and 5 show the results of DLS test and HRTEM imaging of the CQDs, respectively. As can be seen, the mean size of the synthesized nanoparticles is estimated to be about 1.5 ± 0.5 nm.

2.2. Preparing CQDs nanofluids

In the present research, the CQDs-based nanofluid samples were synthesized using the car radiator coolant (CRC) used in the vehicles as the base fluid. In fact, due to the importance of heat transfer in vehicles and internal combustion engines (ICE), one of the objectives of the present research was to improve heat transfer capability of radiator coolant. The radiator coolant was prepared by mixing Caspian Antifreeze (Foumanchimie-under licence of Caspian International Ltd. England) and distilled water at the ratio of 1:1. The CRC-CQDs nanofluid samples were synthesized at four different nanoparticle concentrations, including 100, 200, 500, and 1000 ppm. The nanoparticles within the base fluid were stabilized using merely the bath ultrasonic (P120h. Elmasonic. Germany) and no chemical surfactant was used. For this purpose, a certain amount of CQDs was added to the base fluid and, then, the sample container was put in the bath ultrasonic at the frequency of 37 KHz for 5 min.

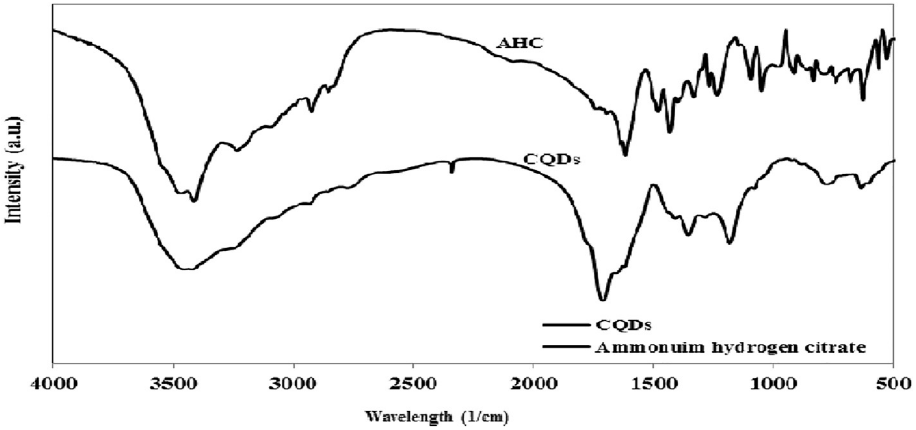

Fig. 2. Infra-red spectra of CQDs and ammonium hydrogen citrate.

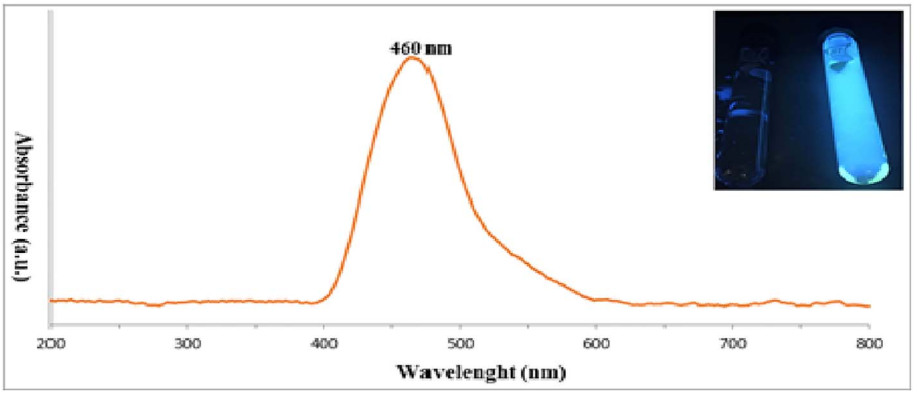

Fig. 3. Photoluminescence spectrum curve of CQDs. (For interpretation of the references to color, the reader is referred to the web version of this article.)

2.3. Measuring heat transfer properties

Thermal conductivity (k) and convection heat transfer (h) coefficients were investigated as two determinant parameters of the fluids' convection heat transferability. Thermal conductivity of the base fluid and the nanofluid samples were measured using KD2-Pro (Decagon devices, USA) at the room temperature of 25 °Cr.·Moreover, in order to measure heat transfer coefficient of the samples, the laboratory system was designed. As shown in Fig. 6, this system consisted of different components, including test section, pump, fluid reservoir, shell-tube exchanger, and circulator. The test component is composed of a straight copper pipe with the inner diameter of 11.42 mm and length of 1 m. The copper pipe is covered by an element heated by AC current and the ceramic insulation with the thickness of 150 mm prevented heat loss. Temperature of the tube surface was measured using 5 thermocouples of K type, and the input and output temperature of the fluid mass were measured by 2 other thermocouples. The flow rate could be adjusted at 1.15–6 L/min. Besides, the fluid was cooled using a shell-tube exchanger and circulator. Finally, the convection heat transfer coefficient was calculated using the following Eqs. (1) and (2):

$$h(x) = \frac{q''}{T_s(x) - T_m(x)} \tag{1}$$

$$q = m^{\cdot}.\ C_p(T_{out} - T_{in}) \tag{2}$$

Where T_s is the average temperature of pipe wall, T_m is the fluid temperature, C_p is the thermal capacity, $m^{\cdot}$ is the fluid mass flow rate, and T_{out} and T_{in} are fluid-copper pipe input and output temperatures, respectively. Also, $q^{''}$ is the constant thermal flux obtained by dividing q thermal flux by the area surrounding the copper pipe.

It should be noted that there are many resources for uncertainty analysis including the tools of measurement, data recording, and data analysis in the experimental works. The set-up uncertainty used in this research has been previously calculated by Askari et al. [17]. Who have reported the value of set-up uncertainty equal to nearly 5.9% based on their precise calculations.

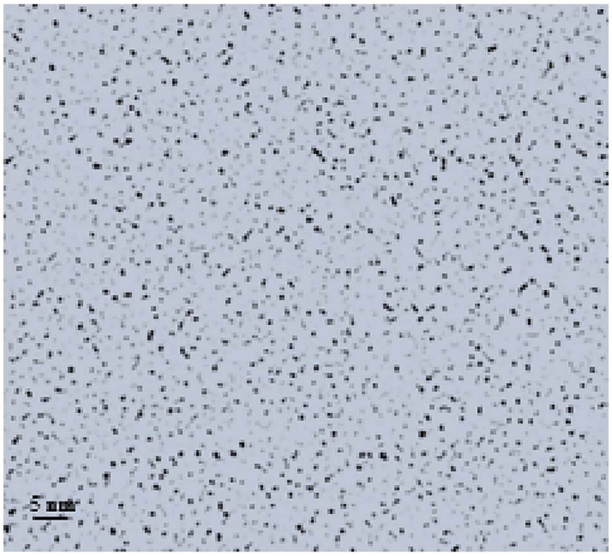

Fig. 5. High resolution transmission electron microscopy (HRTEM) image of CQDs.

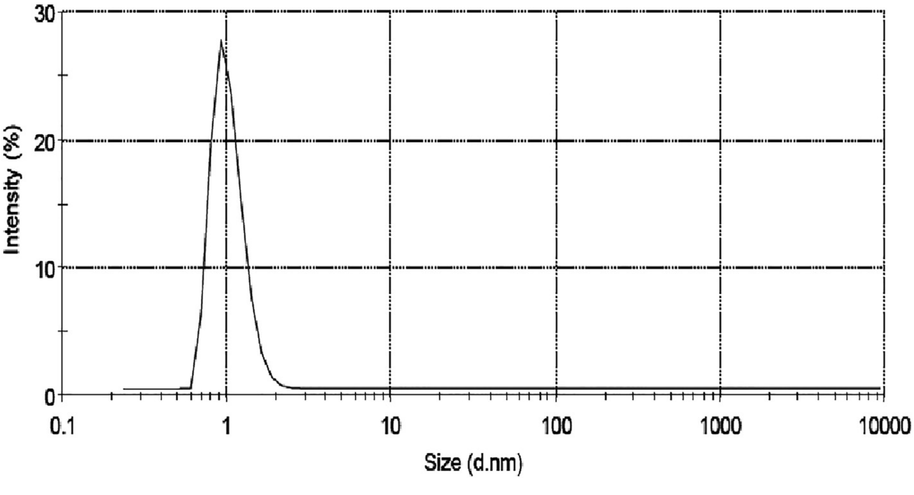

Fig. 4. Particle size distribution of CQDs (DLS analysis).

Fig. 6. Conduction measurement setup (A), Convection measurement setup (B).

3. Result and discussion

3.1. Preparing nanofluids

Preparing stable nanofluids is not as simple as normal liquid-solid mixtures. A highly important problem that disturbs the ideas of synthesizing nanofluids is the agglomeration of the nanoparticles as well as deposition in the base fluid. Due to the strong Van der Waals interactions, nanoparticles tend to stick together and get agglomerated, which can damage the mechanical systems. It should be noted that some of the common methods of stabilizing nanoparticles within the base fluid, such as pH variation or use of chemical surfactants, might cause damage or corrosion of mechanical surfaces. In general, the long-term stability of nanofluids is one of the major factors for their industrial applications, which should be met as much as possible. Concentration of nanoparticles is one of the critical factors affecting the nanofluid's stability. The more concentration of nanoparticles, may lead to more agglomeration and deposition of nanoparticles. Therefore, one of the solutions for increasing the nanofluids' stability is to reduce the nanoparticles' concentration.

Due to the high concentration of functional groups on the surface of CQDs, they have excellent stability within the CRC base fluid. On the other hand, due to their small size (0.5–3 nm), they provide the possibility for synthesizing efficient nanofluids with very low concentrations (100–500 ppm). In this research, the CRC-CQDs nanofluid samples were synthesized at 4 different concentrations of 100, 200, 500, and 1000 ppm in order to investigate the effect of nanoparticle concentration on their stability as well as heat transfer performance. Stability of the samples was investigated by two methods. First, Zeta potential test was carried out, the result of which is shown in Fig. 7. As can be seen, the Zeta potential value was found to be about-36 mv, indicating high stability of the CRC-CQDs nanofluid. Besides, the negative value indicated the presence of amine groups contributing to the suspension's stability.

In the second test, static stability of the samples was investigated. For this purpose, the samples were poured into transparent glassy containers and kept in a totally static place. Superficial changes of the samples were evaluated visually over time. Fig. 8 shows the samples after 6 months, indicating that no nanoparticle deposition occurred. Another important point is that due to the low concentration and high stability of the nanoparticles, the rheological properties of the base fluid will not change considerably, which can prevent damage and dysfunction of the mechanical systems.

It should be noted that for the stabilization of CQD nanoparticles inside the base fluid, the container was placed inside the bath ultrasonic with the frequency of 37 kHz at the ambient temperature of 25 °C for just 5 min, without any addition of chemical surfactants or other additives. Hence, it can be concluded that due to the very low synthesis costs and excellent stability in the base fluid, CQDs are known as an appropriate choice for industrial applications.

3.2. Thermal conductivity

It has been confirmed that the rate of heat transfer can be increased by reducing the size of nanoparticles. Brownian motion of nanoparticles and layered fluid around the nanoparticles are known as popular theories for describing the effect of particle size on the heat transfer of nanofluids. In fact, higher interface between nanoparticles and fluid molecules is due to the smaller size of carbon nanoparticles. Since heat transfer occurs between the fluid-nanoparticle interface, reducing the particle size would improve the fluid's heat transfer capability. On the other hand, Brownian motion of the nanoparticles, as the main mechanism for increasing the heat transfer capability, would be also increased by reducing the nanoparticles size.

Thermal conductivity coefficient (k) was investigated in this research as one of the determinant factors of the fluids' heat transfer

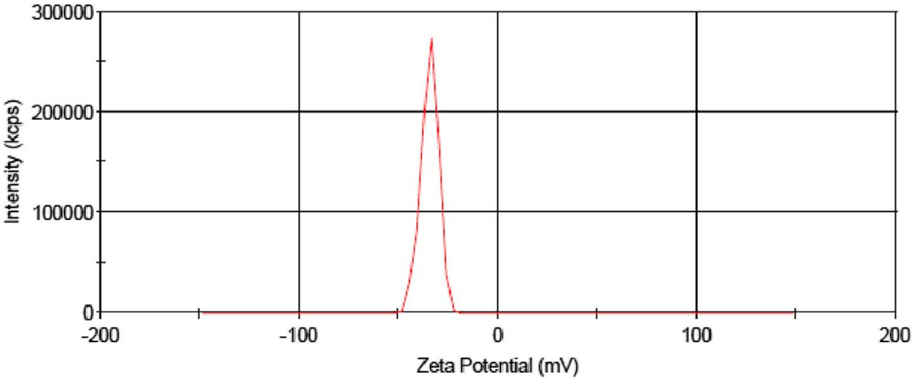

Fig. 7. Zeta potential of the CRC-CQDs nanofluid containing 500 ppm of CQDs.

Fig. 8. Samples of CRC-CQDs after 6 months, (a) base fluid and (b–e) nanofluids by different concentrations of 100–1000 ppm.

Table 1
The effect of CQDs concentration on thermal conductivity of nano fluids.

Concentration of CQDs (ppm)	Weight (wt%)	Thermal Conductivity (W/mK)	Enhancement (%)
0	0	0.368	–
100	0.01	0.382	3.8
200	0.02	0.389	5.7
500	0.05	0.395	7.3
1000	0.1	0.387	5.1

capability. Thermal conductivity of the base fluid and CRC-CQDs samples was measured in four concentrations at 25 °C using KD2-Pro (Decagon devices, USA). The obtained results are shown in Table 1. Accordingly, the maximum improvement of thermal conductivity of the nanofluid compared to the base fluid was 7.3% for the concentration of500-ppm sample. However, if the improvement of k were considered relative to the nanoparticle's concentration, then the 100 ppm sample would have a better status. The notable point in the results was that up to the concentration of 500 ppm, the k variation had an increasing trend, but it was reduced at the concentration of 1000 ppm, which could be due to the nanoparticles' agglomeration at high concentrations, which decreases the nanofluid's performance.

3.3. *Convection heat transfer coefficient*

Heat transfer fluids are among the most widely used materials in various industries. The purpose using radiator coolant in vehicles is to eliminate the excessive heat caused by combustion and friction between the components in the engine. On average, nearly 60–80% of the engine excessive heat is absorbed by the radiator coolant and transferred to the outside environment. The use of nanofluids can improve the heat transfer efficiency in vehicles which consequently, reduce the energy consumption. Although thermal conductivity is one of the major heat transfer properties, nevertheless accurate judgment on the nanofluids' heat transfer performance requires investigating their convection heat transfer behavior.

Since convection heat transfer coefficient is a flow-related property, it depends not only on the boundary conditions in the laboratory system, but also on the Reynolds number. At first, in order to exhibit the precision and accuracy of the data obtained from measuring the convective heat transfer coefficient, the obtained data were validated through comparing with Gnielinski model. The Gnielinski equation for the turbulent flow is as Eq. (3) [17].

$$Nu = \frac{\frac{f}{8}(\mathrm{Re}-1000)\,\mathrm{Pr}}{1 + 12.7\left(\frac{f}{8}\right)^{0.5}\left(pr^{\frac{2}{3}} - 1\right)} \tag{3}$$

In Eq. (4), Prandtl number and parameter f are calculated through Eqs. (4) and (5). Where μ, C_p and K are viscosity, heat capacity and thermal conductivity, respectively.

$$\mathrm{Pr} = \frac{\mu C_P}{K} \tag{4}$$

$$f = \frac{1}{(1.82\log_{10}\mathrm{Re}-1.64)^2} \tag{5}$$

Fig. 9 shows the results of the validation process which was performed using the data of the deionized water's convective heat transfer coefficient. As it can be seen, there is a good relationship between the experimental data and the Gnielinski model's data; thus, it can be concluded that it would be possible to measure the convective heat transfer coefficient of various fluids using this set-up, and the obtained data would be of acceptable precision and accuracy.

In this research, the convection heat transfer coefficient (h) of the car radiator coolant and CRC-CQDs samples was examined in four different concentrations at ambient temperature (25 °C) and different Reynolds numbers. The obtained results are shown in Fig. 10. Accordingly, in all the samples, increasing the Reynolds number led to the increased value of h; on the other hand, addition of the nanoparticles to the base fluid improved the fluid's heat transfer coefficient. Further investigations of the results showed that the increasing trend of h was ascending up to the concentration of 200 ppm, but it became descending after reaching the concentration of 500 ppm. This could be due to agglomeration of CQDs at high concentrations which leads to

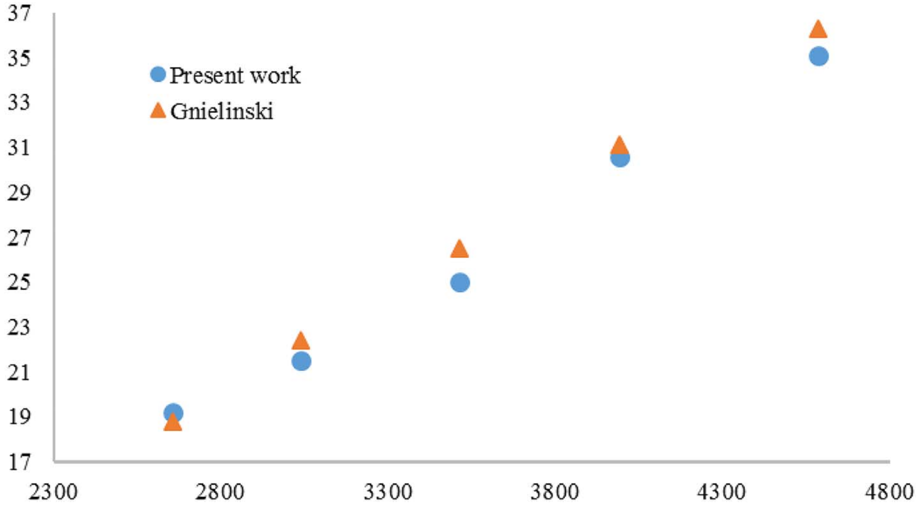

Fig. 9. Validation of experimental set-up as a function of Reynolds number for case of deionized water.

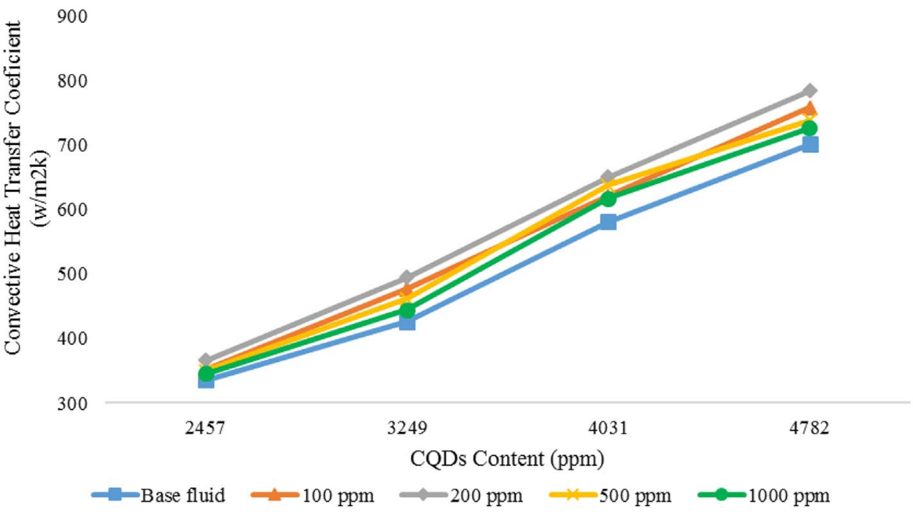

Fig. 10. The trend of changes for convection heat transfer coefficient relative to the Reynolds number in different concentration of the nanoparticles.

Table 2
Convective heat transfer coefficient in 200 ppm concentration.

Re	h (W/m^2K)		Enhancement (%)
	Pure water	Nano fluid	
2457	336	366	8.9
3249	426	495	16.2
4031	581	650	11.87
4782	701	784	11.84

reducing the performance of nanofluid. Because the number of CQDs per unit volume of the fluid is very large due to their small size, and thus a slight increase in the concentration of CQDs resulted in agglomeration of the particles. Therefore, compared to other conventional nanoparticles, low concentration of CQDs indicated better heat transfer performance suggesting high potential of carbon dots to be used as novel nanoparticles for preparation of nanofluids.

As mentioned earlier, the 200-ppm sample had the best function compared to the base fluid. Table 2 shows the heat transfer behavior of nanofluid at different Reynolds numbers and the concentration of 200 ppm. The maximum increase of h compared to the base fluid was found to be about 16.2% in the Reynolds number of 3249.

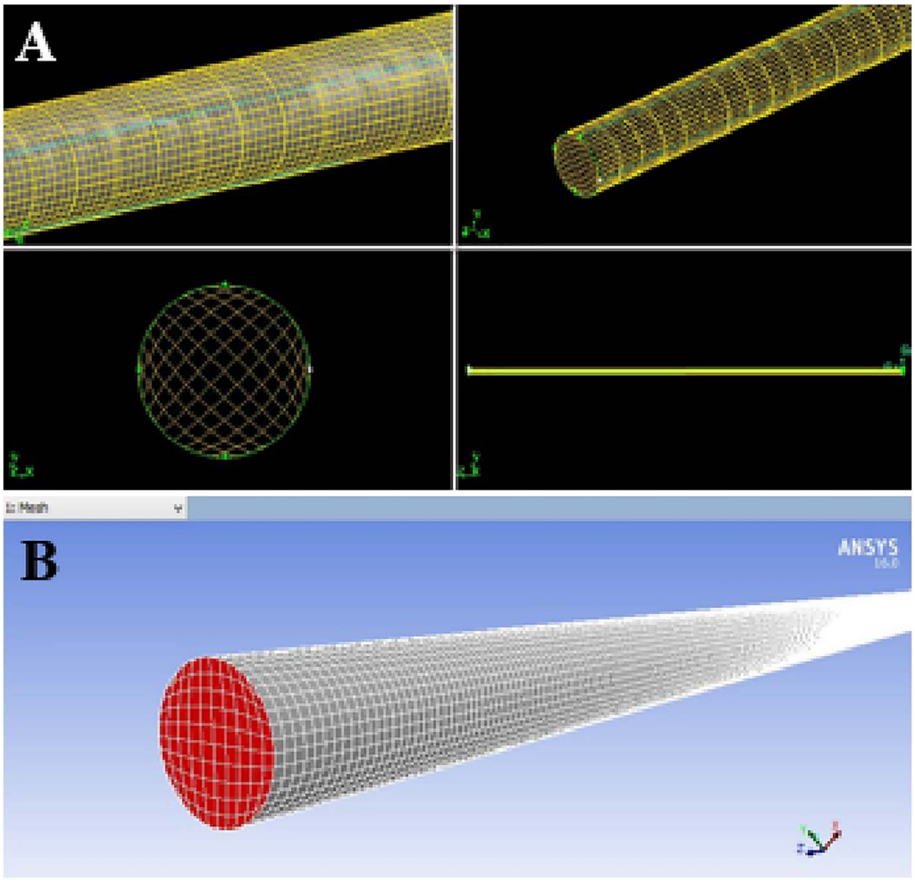

Fig. 11. Geometric modeling of test section by using software: (A) Gambit (B) Ansys Fluent.

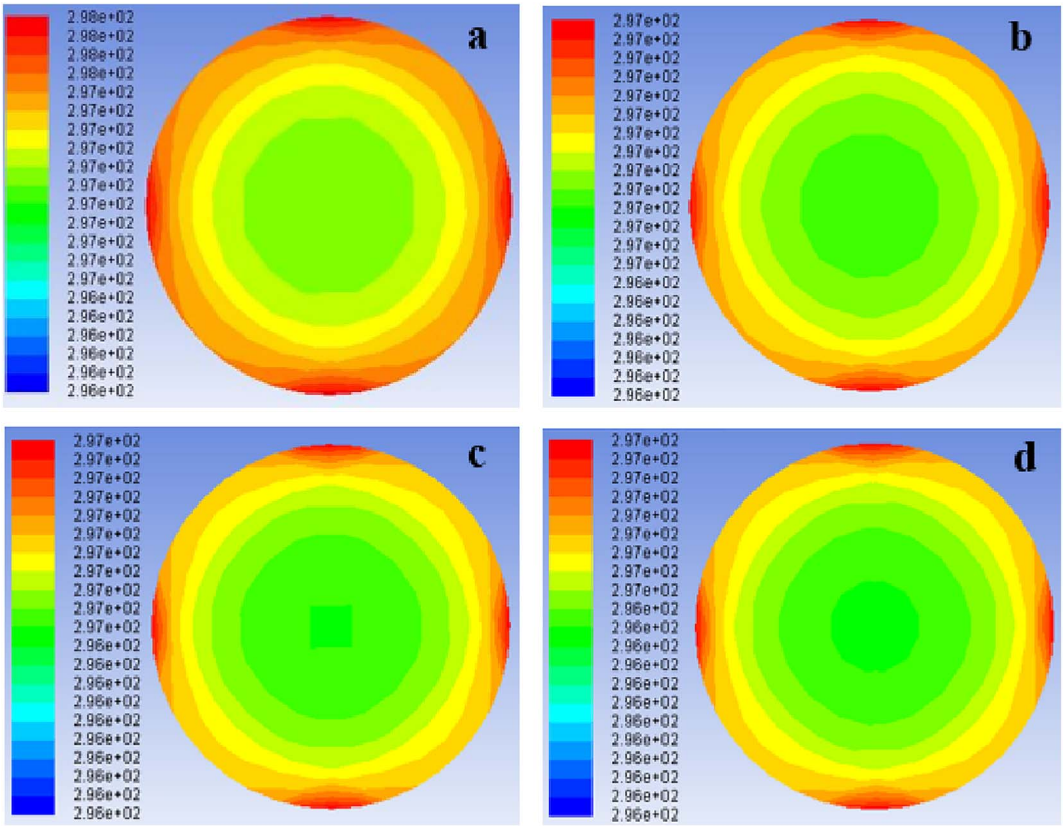

Fig. 12. Temperature contours on the outlet face in Reynolds: (a) 2457, (b) 3249, (c) 4031, (d) 4782.

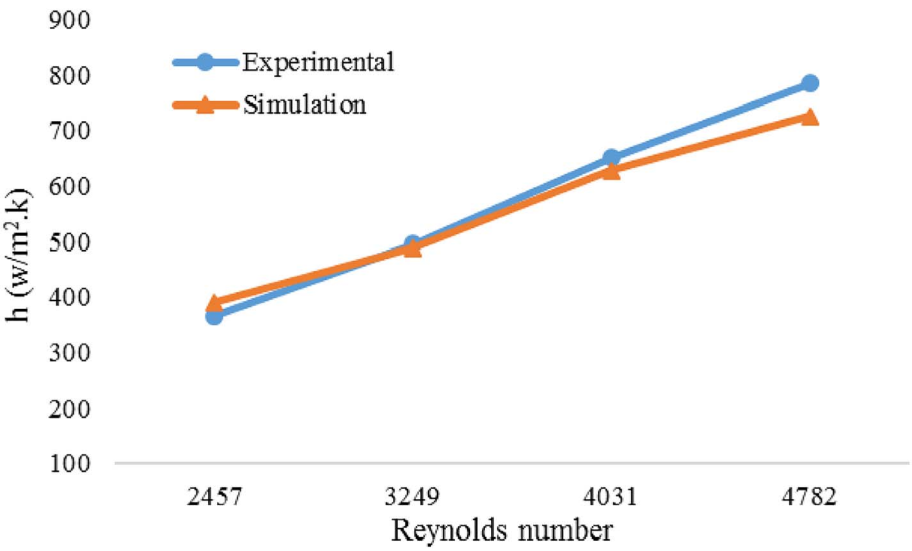

Fig. 13. Comparing the simulation and experimental results for h.

3.4. Heat transfer simulation

The results obtained in Table 2 for the 200-ppm nanofluid sample was simulated in Fluent software. At the first step, the test component of system used for measuring the convection heat transfer coefficient (based on Fig. 6-B) was modeled and meshed by Gambit 2.4.6 software (Fig. 11-A). Then, the geometric model was imported in Ansys Fluent 16 software and the boundary conditions were applied (Fig. 11-B). The simulation process was carried out based on a single-phase mode in turbulent flow conditions using k-epsilon model. Furthermore, to begin the solution, the SIMPLE method was used in the second order upwind mode. The problem was solved in the Steady state at four different Reynolds numbers based on the physical properties of the 200-ppmnanofluid.

To investigate the obtained results, the temperature contours were used on the copper pipe's outlet face, which are appropriately shown in Fig. 12. As can be observed, as expected, increasing the Reynolds number led to the reduction of temperature on the outlet face. The simulation results for convection heat transfer coefficient are shown in Fig. 13. As can be seen, the obtained values from the simulation for the average convection heat transfer coefficient throughout the tube had a slight difference from the experimental values.

4. Conclusion

The main goal of this research is to introduce nanofluids with high stability, appropriate heat transfer capability, cost-effective and environmentally friendly. In the present research, the CQD nanoparticles were used to synthesize the heat transfer nanofluids in order to reduce the possible environmental risks caused by the use of metal and metal oxide nanoparticles. Carbon quantum dot nanoparticles were synthesized using commercial ammonium hydrogen-citrate ($C_6H_{14}N_2O_7$) as precursor. CQD nanoparticles were dispersed at four various concentrations in the CRC as the base fluid without using any surfactant. Investigating the nanofluids' stability indicated their excellent stability. Furthermore, the thermal conductivity and convection heat transfer coefficients of the nanofluid samples were thoroughly investigated. The results indicated 4–7% and 9–16% improvement in thermal conductivity and convection heat transfer coefficients of the CRC-CQDs samples compared to the base fluid, respectively. Further investigations on the obtained results showed that the CQDs had better performance at low concentrations (less than 500 ppm).Because, due to their small size, by increasing the concentration of nanoparticles, the number of the particles was drastically increased per unit volume, resulting in more agglomeration. Finally, the CRC-CQDs nanofluid at the concentration of 200 ppm can be introduced as the best sample in the present research because of low concentration, high stability, low cost and good heat transfer properties.

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