Investigation of the thermophysical properties of ethylene glycol based nanofluids containing decorated carbon nanotubes with silver and copper nanoparticles by passing time

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Abstract
In this study, functionalized carbon nanotubes decorated with silver and copper nanoparticles (fCNTs-Ag, fCNTs-Cu) with mass ratios of 2% and 4% have been used to prepare ethylene glycol (EG) based nanofluids with different concentrations (0.1, 0.25 and 0.5 wt.%). The main goal of this research is the investigation of the effect of passing time on the thermal conductivity and stability of EG-based nanofluids. The thermal conductivity of samples was measured by a Decagon KD2 Pro thermal analyzer device for 90 days. The results showed that in all samples, the thermal conductivity of nanofluids was increased with increasing the concentration of nanoadditives. In addition, it was surprisingly observed that the thermal conductivity of samples was increased by passing time exponentially. The stability of the samples was investigated using UV-vis spectroscopy for 60 days. It was observed that the stability of nanofluids containing fCNTs-Cu is higher than the stability of nanofluids containing fCNTs-Ag.

Keywords: thermal conductivity, hybrid nanofluids, stability, UV-vis spectroscopy, KD2 Pro thermal analyzer device.

1. Introduction

Increasing heat transfer efficiency has a special importance in human life because it causes an increase in the availability of energy for future demands. Also, due to the increasing global energy demand for the consumption of fossil fuels, which has caused major environmental crises such as climate change, improving the heat transfer process can reduce thermal losses [1-2]. Thermal conductivity is one of the most important factors in the thermal performance of a heat transfer fluid. Among traditional heat transfer fluids (ethylene glycol, oil, water, etc.), water has the highest thermal conductivity, but it is less than the thermal conductivity of most metals, metal oxides and carbon nanotubes. Because the solids have higher thermal conductivity than the fluids, dispersing solid nanoparticles into the fluids can improve the thermal conductivity of a sample, which is called a nanofluid. It is expected, with the large aspect ratio (surface to volume ratio) and other unique properties of nanoparticles, the thermal conductivity and heat transfer coefficient of the nanofluids were substantially enhanced [1-4]. According to previous research, it has been found that the various parameters such as particle size and shape, pH of the fluids, surfactant, solvent type, hydrogen bonding, temperature, base fluids and the alignment of the nanoparticles utilized (carbon nanotubes, Graphene, and metal oxides nanoparticles) have affected the thermal conductivity of the nanofluids directly and can either increase or decrease the thermal conductivity [2-6]. Nanofluids have different applications, such as industrial cooling, smart fluids, solar energy, nuclear reactor, phase change heat transfer, sensing and imaging, brake fluids, detergent, microchip cooling and automotive cooling [6-8]. If more than one nanoparticle or more than one base fluid was used to make the nanofluid, a hybrid nanofluid is prepared which is expected to have improved thermophysical properties. Different combinations of nanoparticles and base fluids were used to prepare hybrid nanofluids by various research groups [9-12]. Carbon nanotubes are a suitable choice for the preparation of nanofluids due to their unique properties, such as a very large aspect ratio, high flexibility and strength, and very high thermal conductivity. Also, silver and copper nanoparticles can improve the thermal conductivity of base fluids due to their high thermal conductivity coefficient. Various research groups have investigated the prepration of hybrid nanofluids containing carbon nanotubes, silver nanoparticles, and copper nanoparticles. Jha et al. [13] synthesized Cu decorated MWCNTs based on nanofluids in deionized water (DI water) and ethylene glycol (EG) without any surfactant. They observed a 35.3% and 10.1% increase in thermal conductivity with respect to DI water and EG at a concentration of 0.03 vol%. Amiri et al. [14] investigated the stability, viscosity, shear stress and thermal conductivity of multiwalled carbon nanotubes in the presence of gum Arabic (GA), cysteine (Cys) and silver (Ag), which were named respectively as (MWCNTs-GA), (MWCNTs-Cys) and (MWCNTs-Ag). The results showed that the covalent functionalization by Ag is more effective than noncovalent functionalization. They found that the sequence of thermal conductivity at constant temperature and concentration was MWCNTs-Ag > MWCNTs-Cys > MWCNTs-GA > Ag/water > water. Jha et al. [15] investigated the thermal and electrical conductivity behavior of ZnO and Al_2O_3 decorated multiwalled carbon nanotubes (MWCNTs) nanofluids with deionized water (DI water) and ethylene glycol (EG) as base fluids. They observed a linear behavior in the thermal conductivity enhancements with volumetric fraction of ZnO-MWCNTs and Al_2O_3 -MWCNTs nanocomposite and temperature. The increase in thermal conductivity corresponding to pure base fluid is about 11% and 9.7% for ZnO-MWCNTs and Al_2O_3 -MWCNTs for DI water-based nanofluids. Similarly, 8.5 and 6.8% enhancement has been observed with ZnO-MWCNTs and Al_2O_3 -MWCNTs, EG based nanofluids respectively. Farbod et al. [16] investigated the thermal conductivity enhancement of water-based nanofluids containing pristine and functionalized MWCNTs decorated with Ag nanoparticles with mass ratios of 1%, 2% and 4% at different temperatures (20, 30, 40 and 50 ˚C). They measured the maximum enhancement of 20.4% for the sample containing functionalized MWCNTs decorated with 4 wt.% Ag at 40 ˚C. Gu et al. [17] prepared water-based nanofluid containing carbon nanotubes (CNTs) decorated with Ag nanoparticles (Ag/CNTs). They observed that the thermal conductivity enhancement increases with the thermal filler loading and the decorative quantity of Ag nanoparticles. They measured the maximum thermal conductivity enhancement up to 21% with Ag/CNTs loading at 3.0 wt% in suspension as the quantity of Ag nanoparticles decorated on CNTs was 1.0 mol%. Shanbedi et al. [18] investigated the thermo-physical and hydrodynamic properties of heat transfer nanofluids containing Cu-decorated MWCNTs, Fe-decorated MWCNTs, and Ni-decorated MWCNTs. They observed a significant increase in the thermal and electrical conductivities of heat transfer nanofluids containing metal nanoparticles-based MWCNTs. In our previous work [19], the stability and effect of passing time and temperature on the thermal conductivity of CNTs-ethylene glycol (EG) nanofluids were investigated. It was found that immediately after nanofluid preparation not too much increase in thermal conductivity was observed, but the nanofluid aging had a great influence on the improvement of the thermal conductivity, as after 65 days, about a 50% increase was observed. In order to investigate the effect of aging and fCNTs decorated with nanoparticles on the thermophysical properties of ethylene glycol-based nanofluids, in this work, fCNTs decorated with Ag and Cu nanoparticles with mass ratios of 2% and 4% have been used to prepare ethylene glycol-based nanofluids with different concentrations (0.1, 0.25, and 0.5 wt.%). Thermal conductivity and stability of the samples were investigated for 60 days, and the results are given in detail in this article.

2. Materials and Methods

2.1 Decoration of functionalized carbon nanotubes with silver nanoparticles (fCNTs-Ag):

The chemical reduction method was used to decorate functionalized carbon nanotubes (fCNTs) with silver nitrate (AgNO3) as the source of Ag nanoparticles and sodium borohydride (NaBH4) as a reducer. To decorate fCNTs with 4% Ag nanoparticles, 0.096 gr of fCNTs was added to 50 mL deionized water and the suspension was sonicated for one hour. 111 ml of NaBH4 solution (0.002 M) was added to 37 ml of AgNO3 solution (0.001 M) and the resulting solution was added to the sonicated fCNTs solution under cold bath (6-10 °C) drop by drop in 3 minutes. Then the sample was washed and centrifuged 5 times, and it was dried at 70 ˚C for 24 hours. A similar method was used to decorate fCNTs with 2% Ag nanoparticles. The amounts of NaBH4 and AgNO3 solutions were half of the amounts corresponding to 4% Ag nanoparticles.

2.2. Decoration of functionalized carbon nanotubes with copper nanoparticles (fCNTs-Cu):

Similar to the previous step, the chemical reduction method was used to decorate functionalized carbon nanotubes (fCNTs) with copper nanoparticles. Copper (II) sulfate ($CuSO_4$) was used as a source of copper nanoparticles, sodium borohydride ($NaBH_4$) and trisodium citrate ($Na_3C_6H_5O_7$) were used as reducers. To decorate fCNTs with 4% Cu nanoparticles, 0.096 gr of fCNTs was added to 50 mL deionized water and the suspension was sonicated for one hour. 1 ml of $Na_3C_6H_5O_7$ solution (0.1 M) and 100 ml of $NaBH_4$ solution (0.001 M) was added to 1.57 ml of $CuSO_4$ solution (0.001 M) and the resulting solution was added to the sonicated fCNTs solution under cold bath (6-10 °C) drop by drop in 3 minutes. Then the sample was washed and centrifuged 5 times, and it was dried at 70 ˚C for 24 hours. A similar method was used to decorate fCNTs with 2% Cu nanoparticles. The amounts of $Na_3C_6H_5O_7$, $NaBH_4$ and $CuSO_4$ solutions were half of the amounts corresponding to 4% Cu nanoparticles.

2.3. Preparation of nanofluids:

A two-step method was used to prepare nanofluids. According to the desired concentration to prepare each sample, a specific mass of nanoadditives (fCNTs and decorated fCNTs with 2% and 4% of Ag and Cu nanoparticles) was added to a specific volume of the base fluid (ethylene glycol). Then the sample was sonicated for 2 hours to obtain a uniform solution. Nanofluids with different concentrations (0.1, 0.25 and 0.5 wt.%) for all nanoadditives were prepared. Each sample was divided into two separate parts. One part was kept inside the capped tube in a closed and stationary water bath in order to measure UV-vis spectrum and the other part was kept inside the other tube to measure the thermal conductivity. To measure the thermal conductivity of samples, a transient hot wire method and the KD2 Pro thermal properties analyzer manufactured by Decagon Devices Inc., were used. In addition, to measuring thermal conductivity, this device also calculates the error coefficient. If the error coefficient is less than 0.01, it means that the measurement conditions are suitable and the thermal conductivity value will be acceptable. To ensure the accuracy of the thermal conductivity values, each measurement was repeated 7-10 times and the average of the measurements was reported. In order to investigate the stability of samples, UV-vis spectroscopy was used. For each stability measurement, 100 µl of solution was taken from the surface of the sample. Then it was diluted with deionized water with a certain ratio and was sonicated for a few minutes. The above process was repeated for all samples for 60 days.

3. Results and discussion:

3.1. Characterization of samples:

Figure 1 shows the X-ray diffraction pattern (XRD) of fCNTs and fCNTs decorated with 2% and 4% of Ag nanoparticles (fCNTs, fCNTs-2%Ag and fCNTs-4%Ag). As can be seen, for the decorated samples, in addition to the CNTs peaks (about 26˚), the Ag peaks can also be observed, which indicates the presence of this element in the samples. In order to investigate the morphology of the samples, a field emission scanning electron microscope (FESEM) and transmission electron microscope (TEM) were used. Figure 2 shows the FESEM and figure 3 shows the TEM images of fCNTs-2%Ag and fCNTs-4%Ag. As can be seen, silver nanoparticles were placed on the surface of carbon nanotubes. Figure 4a and b show the X-ray energy diffraction spectroscopy (EDX) related to fCNTs-2%Ag and fCNTs-4%Ag. The results showed that carbon, oxygen and silver elements are present in the sample. Also, the weight percentage of each element can be observed in the tables. As can be seen, the atomic percentage of silver in the fCNTs-4%Ag sample is higher than that in the fCNTs-2%Ag sample.

Figure 5 shows the X-ray diffraction pattern (XRD) of fCNTs and fCNTs decorated with 2% and 4% of Cu nanoparticles (fCNTs, fCNTs-2%Cu and fCNTs-4%Cu). As can be seen, for the decorated samples, in addition to the CNTs peaks (about 26˚), the Cu peaks can also be observed, which indicates the presence of this element in the samples. Figure 6 shows the FESEM images of fCNTs-2%Cu and fCNTs-4%Cu. As can be seen, copper nanoparticles were placed on the surface of carbon nanotubes. Figure 7a and b show the X-ray energy diffraction spectroscopy (EDX) related to fCNTs-2%Cu and fCNTs-4%Cu. The results showed that carbon, oxygen and copper elements are present in the sample. Also, the weight percentage of each element can be observed in the tables.

3.2. Investigation of the thermal conductivity of ethylene glycol-based nanofluids containing fCNTs, fCNTs-Ag and fCNTs-Cu by passing time:

Figure 9 shows the thermal conductivity of ethylene glycol-based nanofluids containing fCNTs and fCNTs decorated with Ag and Cu nanoparticles as a function of passing time for 90 days. All samples were prepared with concentrations of 0.1, 0.25 and 0.5 wt.% and all measurements were made at 25°C. Measuring the thermal conductivity of each sample was repeated 7–10 times and the average value was reported. As can be seen for all samples, the thermal conductivity of EG-fCNTs nanofluids is higher than the thermal conductivity of pure ethylene glycol (0.254 W/m.K). It is also observed that increasing the concentration of nanoadditives (fCNTs, fCNTs-2%Ag, fCNTs-4%Ag, fCNTs-2%Cu and fCNTs-4%Cu) has increased the thermal conductivity of nanofluids as the highest increasing percentage of thermal conductivity is related to the 0.5 wt.% samples. The results show that for all samples, decorated fCNTs with Ag and Cu nanoparticles has increased the thermal conductivity of nanofluids. Also, increasing the mass ratios of Ag and Cu nanoparticles has caused a slight increase in the thermal conductivity of nanofluids as the 4% decorated samples have higher thermal conductivity than the 2% decorated samples. For all decorated samples, the results showed that nanofluids containing fCNTs decorated with Ag nanoparticles have higher thermal conductivity than nanofluids containing fCNTs decorated with Cu nanoparticles. This result can be attributed to the higher thermal conductivity of silver (429 W/m. K) compared to copper (401 W/m. K). The highest increase in thermal conductivity was related to the nanofluid containing fCNTs-4%Ag and a concentration of 0.5 wt.%. This increase was 11% (immediately after preparing the sample) and 21.3% (90 days after preparing the sample). Investigation of the effect of aging on the thermal conductivity values of samples for 90 days showed that the thermal conductivity of samples increased over passing time. As can be seen in all samples, this increase is not linear and has an exponential behavior. This result is in agreement with our previous work [19]. Since the thermal conductivity of pure ethylene glycol does not change with passing time, the significant increase in the thermal conductivity of EG-fCNTs nanofluids by aging can be attributed to the bonding and aggregating the ethylene glycol molecules and fCNTs and forming bigger particles. One can say that during the ultrasonic process, some chemical bonds are broken, and new bonds are formed between fCNTs and ethylene glycol molecules by passing time. This phenomenon can be attributed to the solid-like nanolayer that forms around the nanoparticles in the solution [19-24]. This nanolayer acts as a bridge between the nanoparticle and the base fluid, and it leads to an increase in the thermal conductivity of EG-fCNTs nanofluids. On the other hand, by increasing the thickness of this nanolayer, the thermal conductivity of the sample is increased. So, the significant increase in the thermal conductivity of samples by aging can be explained as follows: at the beginning of the nanofluid preparation, the thickness of a nanolayer is thin and by passing time, its thickness increases and the thermal conductivity of samples increases.

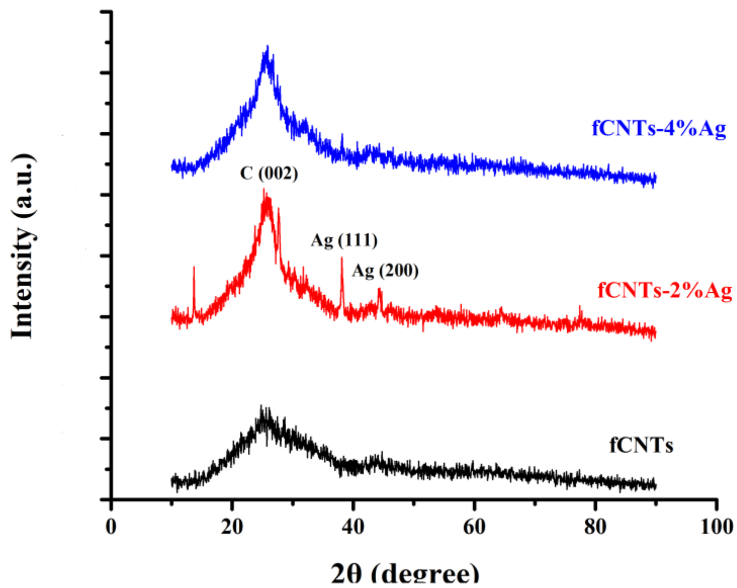

Fig1: X-ray diffraction pattern (XRD) of fCNTs and fCNTs decorated with 2% and 4% of Ag nanoparticles.

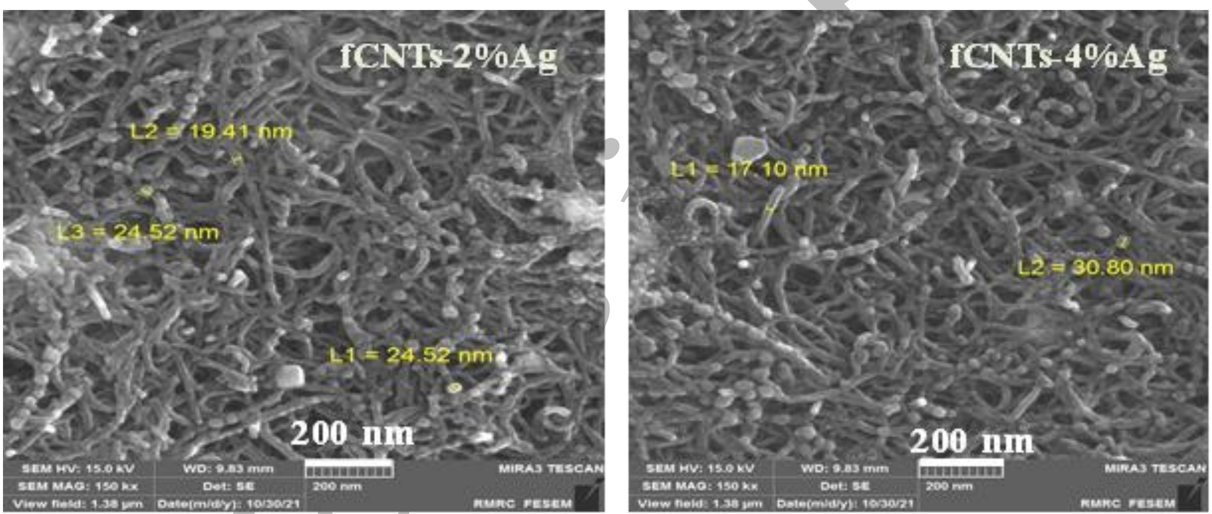

Fig2: FESEM images of fCNTs decorated with 2% and 4% of Ag nanoparticles.

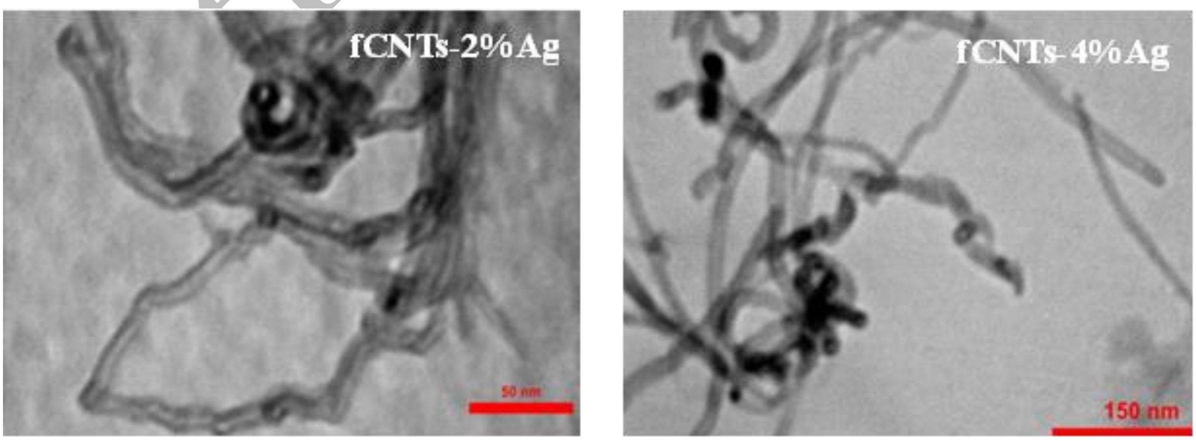

Fig3: TEM images of fCNTs decorated with 2% and 4% of Ag nanoparticles.

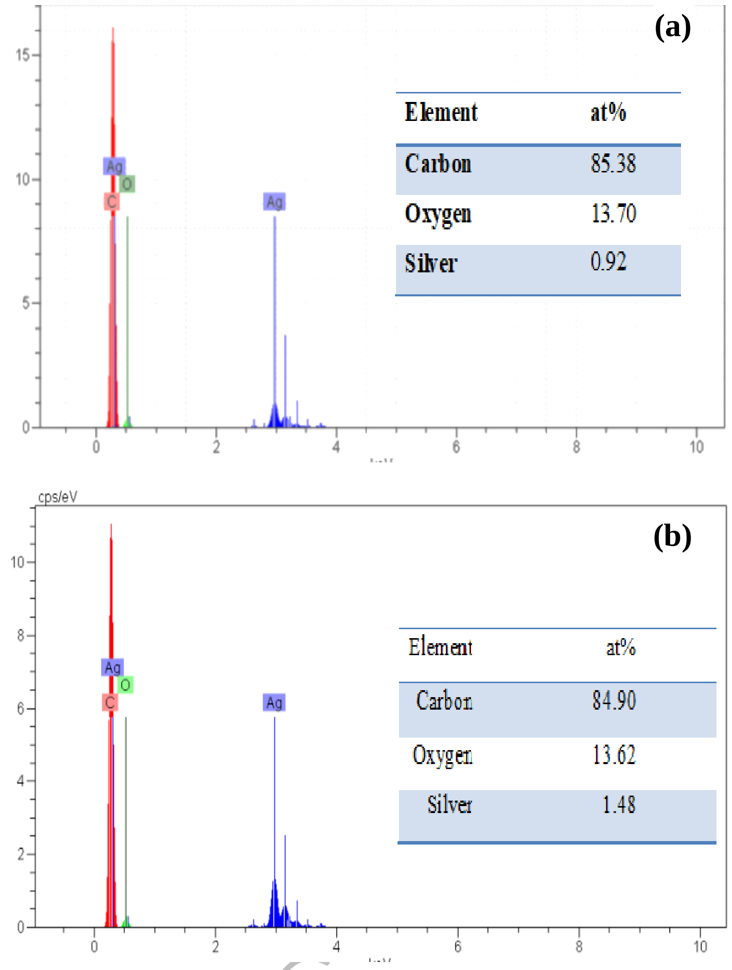

Fig4: EDX patterns of fCNTs decorated with (a) 2% and (b) 4% of Ag nanoparticles.

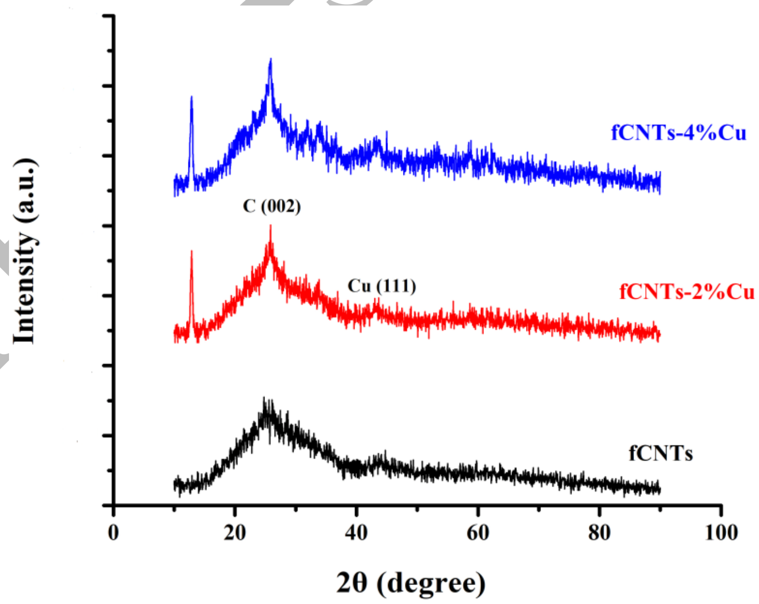

Fig5: X-ray diffraction pattern (XRD) of fCNTs and fCNTs decorated with 2% and 4% of Cu nanoparticles.

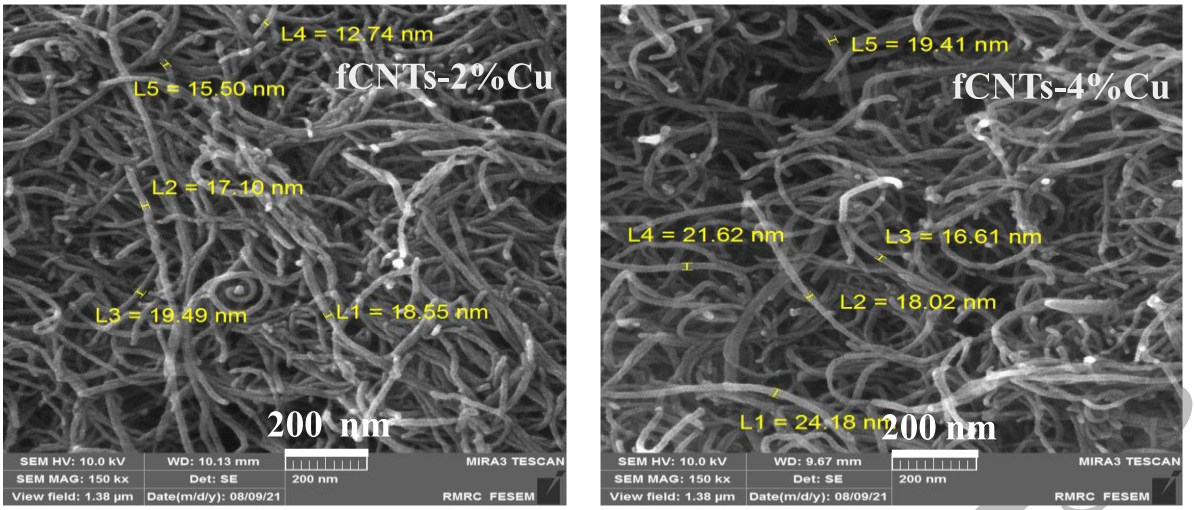

Fig6: FESEM images of fCNTs decorated with 2% and 4% of Cu nanoparticles.

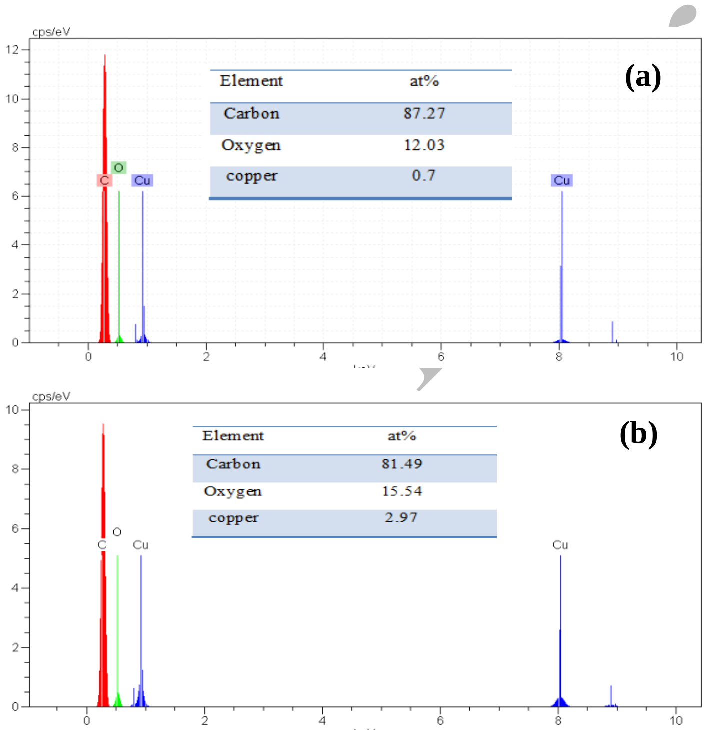

Fig7: EDX patterns of fCNTs decorated with (a) 2% and (b) 4% of Cu nanoparticles.

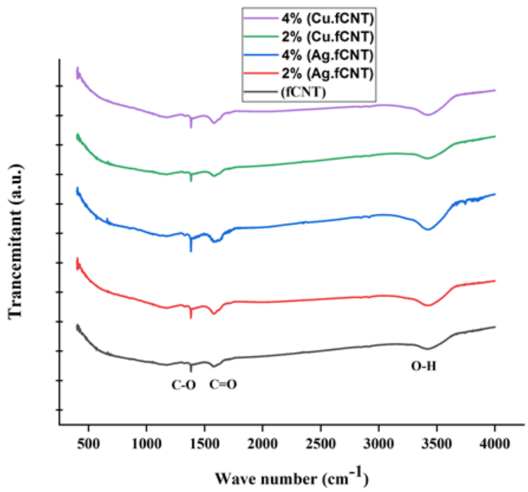

Fig8: The FTIR spectrum of fCNTs and fCNTs decorated with 2% and 4% of Ag and Cu nanoparticles.

3.3. Investigation of the stability of ethylene glycol-based nanofluids containing fCNTs, fCNTs-Ag and fCNTs-Cu by passing time:

UV-vis spectroscopy was used to investigate the stability of the samples. After preparation of each sample, one part of it was placed inside the capped tube in a closed and stationary water bath immediately. To obtain any absorption spectrum, 100 µl of solution was taken from the surface of the sample by a sampler to prevent confusion and distortion in the sample. Then it was diluted with a certain amount of deionized water and was sonicated for a few minutes. The above process was repeated for all samples for 60 days. Due to their hydrophobic properties, carbon nanotubes do not disperse in aqueous solutions properly. Functionalization of the surface of carbon nanotubes improves their dispersion in aqueous solutions. Because functionalized carbon nanotubes have a surface charge, they will have a mutual repulsion effect on each other, so stability of the sample increases. Figures 10 and 11 show the UV-vis spectrum of ethylene glycol-based nanofluids containing fCNTs decorated with Ag and Cu nanoparticles for 60 days and different concentrations of 0.1, 0.25 and 0.5 wt.%. The stability of the sample was investigated by considering the intensity of the absorption peak on different days. The reduction time of the intensity of the absorption peak up to half of its initial intensity was considered. The longer reduction time means the more stability of the sample. As can be seen, for nanofluids containing fCNTs-4%Ag about 35 days and for nanofluids containing fCNTs-2%Ag about 32 days after preparation of the samples, the intensity of the absorption peak reached about half of its initial intensity. This amount was about 43 days for nanofluids containing fCNTs-4%Cu and about 45 days for nanofluids containing fCNTs-2%Cu. The results showed that the stability of nanofluids containing fCNTs-Cu is higher than the stability of nanofluids containing fCNTs-Ag. One can say, this result is due to the higher density of silver compared to copper.

4. Conclusion:

In this study, the effect of passing time on the thermal conductivity and stability of ethylene glycol-based nanofluids containing functionalized carbon nanotubes and also functionalized carbon nanotubes decorated with silver and copper nanoparticles were investigated. The mass ratios of silver and copper nanoparticles were 2 and 4% and the concentrations of nanofluids were 0.1, 0.25 and 0.5wt.%. The results showed that ethylene glycol-based nanofluids containing fCNTs, fCNTs-Ag and fCNTs-Cu have higher thermal conductivity compared to pure ethylene glycol. In all samples, increasing the concentration of nanoadditives has increased the thermal conductivity of the sample. It was observed that the nanofluids containing fCNTs-Ag and fCNTs-Cu have higher thermal conductivity than the nanofluids containing fCNTs. The biggest increase in thermal conductivity was 11% (immediately after preparation of the sample) for nanofluids containing fCNTs-4%Ag and 0.5 wt.%. Also, in all samples, the thermal conductivity of nanofluids increased by passing time exponentially, as 90 days after preparation of the sample, the thermal conductivity of nanofluids containing fCNTs-4%Ag and 0.5 wt.% reached 21.3%. UV-vis spectroscopy was used to investigate the stability of the samples. The results showed that the stability of nanofluids containing fCNTs-Cu is higher than the stability of nanofluids containing fCNTs-Ag. One can say this result is due to the higher density of silver compared to copper.

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Conflict of interest

The authors declared no potential conflicts of interest with respect to the research, authorship and publication of this article.

Data Availability Statements

The datasets generated and/or analyzed during the current study are available from the corresponding author on reasonable request.

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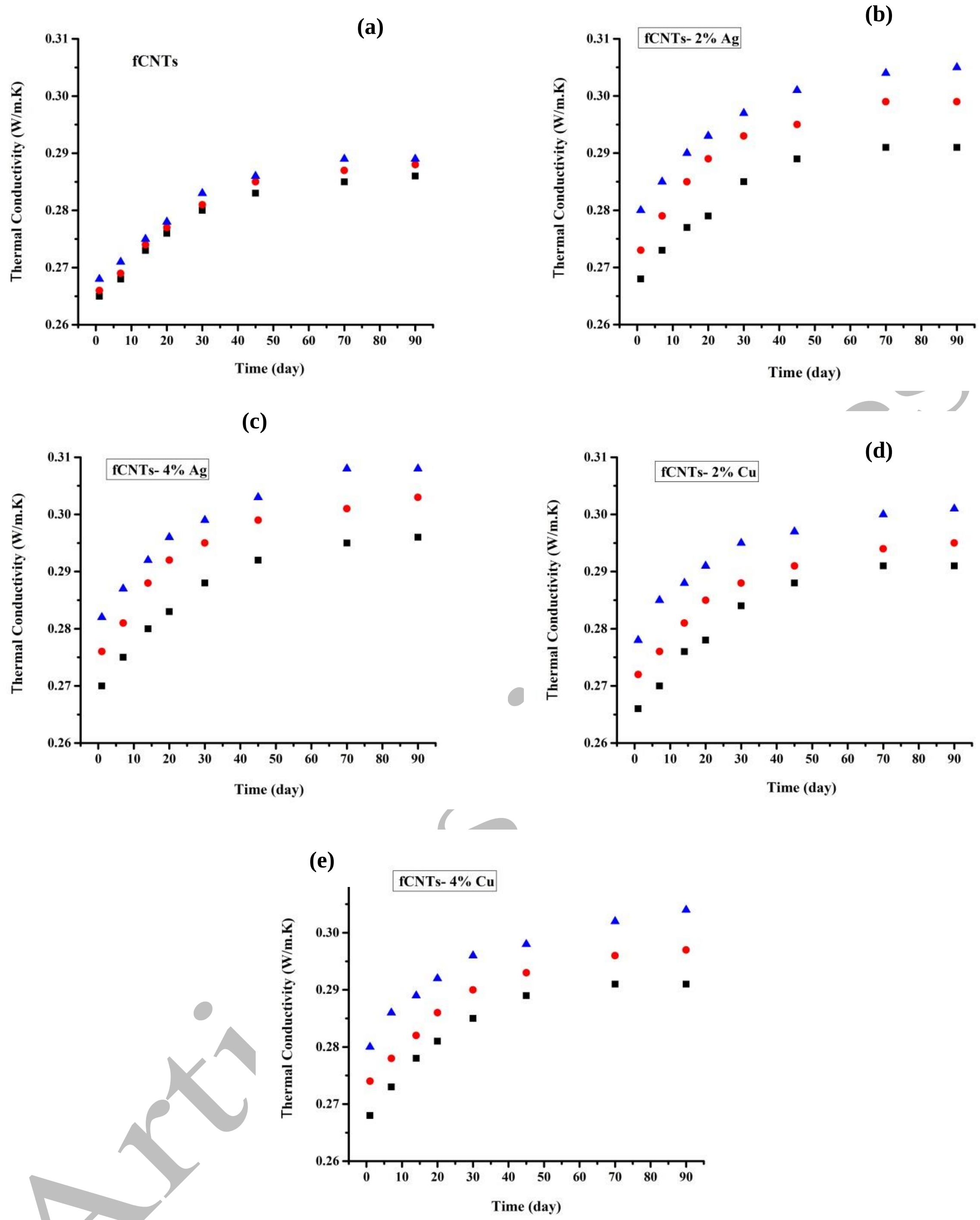

Fig9: The thermal conductivity of ethylene glycol-based nanofluids containing a) fCNTs, b) fCNTs-2%Ag, c) fCNTs-4%Ag, d) fCNTs-2%Cu and e) fCNTs-4%Cu with concentrations of 0.1 (■), 0.25 (●) and 0.5 (▲) wt.% .

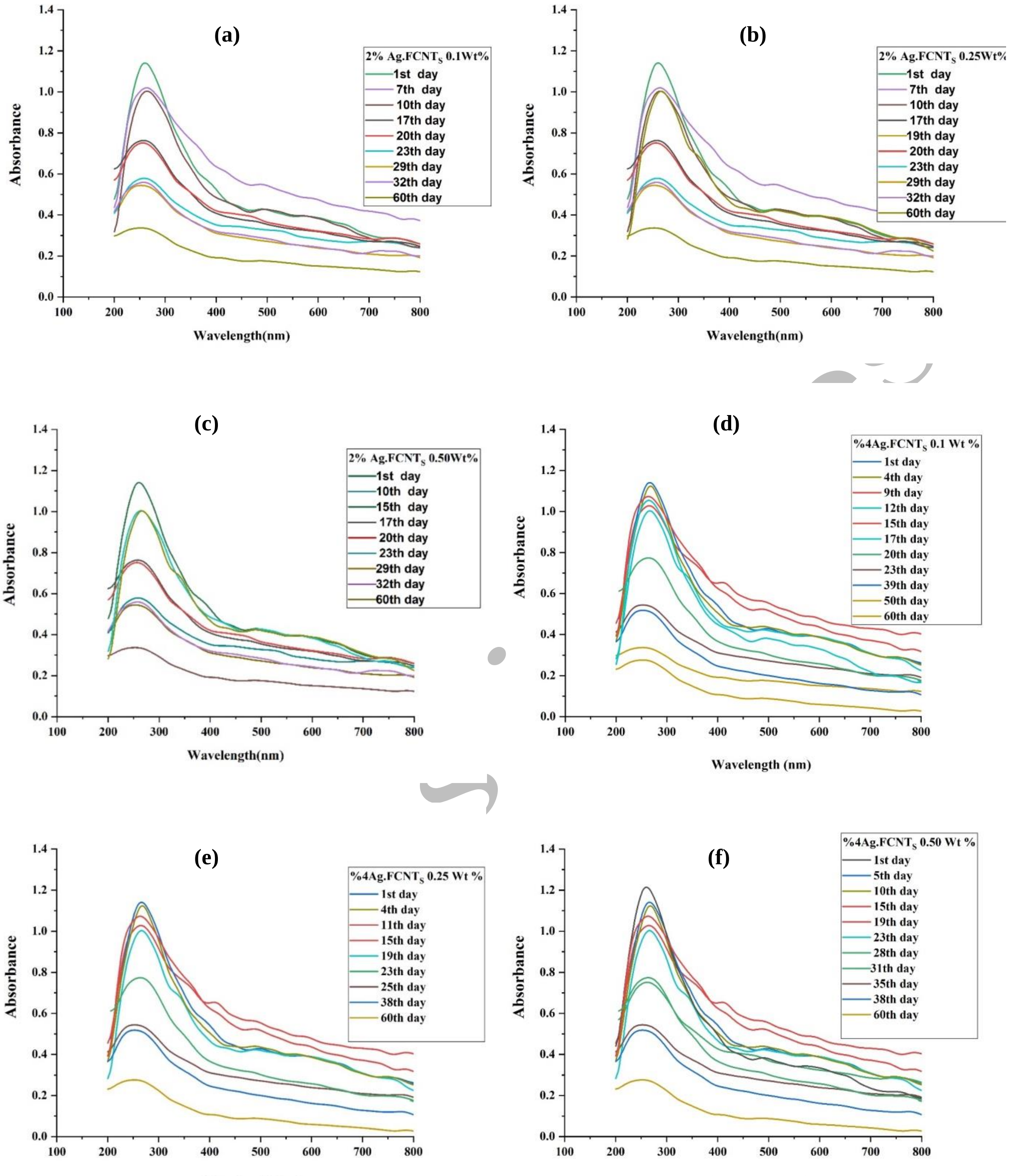

Fig10: UV-vis spectrums of ethylene glycol-based nanofluids containing a) fCNTs-2%Ag- 0.1 wt.%, b) fCNTs-2%Ag- 0.25 wt.%, c) fCNTs-2%Ag- 0.5 wt.%, d) fCNTs-4%Ag- 0.1 wt.%, e) fCNTs-4%Ag- 0.25 wt.% and f) fCNTs-4%Ag- 0.5 wt.%.

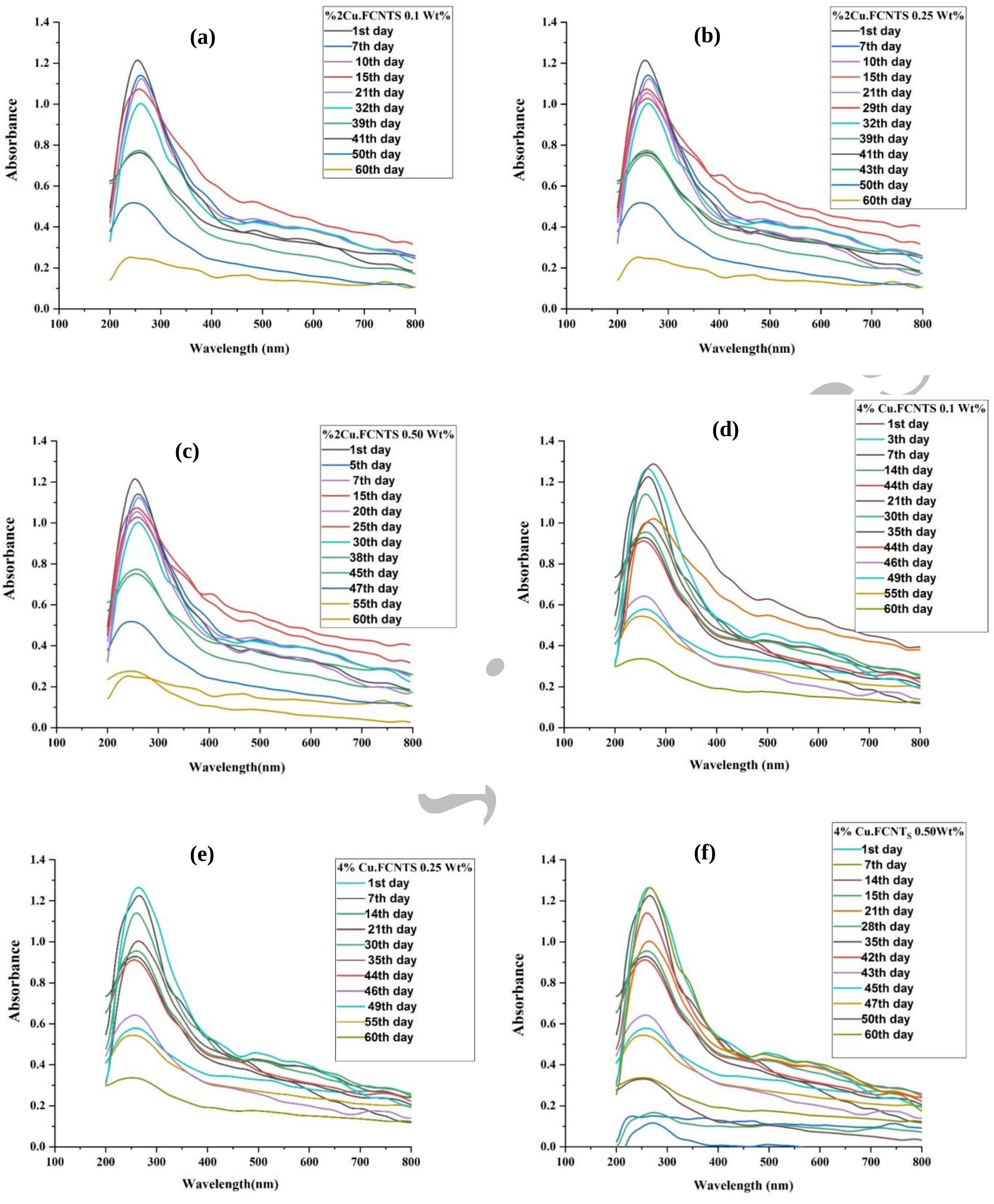

Fig11: UV-vis spectrums of ethylene glycol-based nanofluids containing a) fCNTs-2%Cu- 0.1 wt.%, b) fCNTs-2%Cu - 0.25 wt.%, c) fCNTs-2%Cu - 0.5 wt.%, d) fCNTs-4%Cu - 0.1 wt.%, e) fCNTs-4%Cu - 0.25 wt.% and f) fCNTs-4%Cu - 0.5 wt.%.

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