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Neutron irradiation sensitivity of thermal conductivity for Al₂O₃ nanofluids

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Keywords: neutron-irradiation, nanofluids, thermal conductivity, sensitivity analysis

Abstract

In this work, the thermal conductivity of Al₂O₃ nanofluids has been investigated for the sensitivity towards neutron irradiation. The solution combustion method has been used for the synthesis of Al₂O₃ nanoparticles that have been used for the preparation of the nanofluids. Prepared nanofluids have been neutron-irradiated for 7 and 14 days. Dynamic Light Scattering, Scanning Electron Microscopy, and Ultraviolet-Visible Spectroscopy have been used to ascertain the change in properties before and after neutron-irradiation. Thermal conductivity has been measured for un-irradiated and neutron-irradiated nanofluids at 30 °C using a KD2 pro thermal properties analyzer. The decrease in thermal conductivity has been observed after neutron-irradiation that further decreases with increased duration of exposure and concentration of nanoparticles. 5 and 10% decrease in thermal conductivity has been recorded after 7 and 14 days of neutron irradiation for change in concentration from 0 to 2 volume percent. Neutron-irradiation sensitivity analysis revealed that heat transfer characteristics are sensitive at higher concentrations and during initial exposure of neutron-irradiations.

1. Introduction

Nanofluids have been widely investigated for the heat transfer characteristics [1–6] and reported as future generation material for numerous applications [7–10]. The metal oxide nanoparticles significantly enhance the heat-carrying capacity of conventional fluids [11–14]. Al₂O₃ [15], CuO [16], Fe₂O₃ [17], TiO₂ [18], *etc* based nanofluids show improved performance in terms of an increase in the thermal conductivity. Nanofluids can be of great use in nuclear reactors for the power production industry [19]. For advanced nuclear reactors, new materials, fuel, cooling fluids, and more efficient heat transfer processes are required and therefore these are the topics of great interest [20, 21].

There are many environmental factors that can affect the performance of nanofluids [22]. Materials used in nuclear reactors are in continuous contact of radiations which can affect the material properties and must be evaluated. Pinho *et al* [23] investigated the effect of the gamma irradiations on the thermophysical properties of ZrO₂ nanofluids and observed a significant decrease in the thermal conductivity of nanofluids after exposure to gamma radiation. Nesvizhevsky [24] studied the interaction of neutrons with nanoparticles and observed a decrease in energy transfer. Eggers *et al* [25] shown reduced heat loss by performing irradiation and energetic analysis of nanofluid-based volumetric absorbers for concentrated solar power. Konobeyev *et al* [26] developed a model for atomic displacement cross-sections for neutron irradiation of materials and established the formation of defects due to irradiation. A significant reduction in mechanical strength after neutron irradiation has also been reported [27].

Senor *et al* [28] evaluated the effect of neutron irradiations on the thermal conductivity of SiC based nanocomposites and suggested that irradiation-induced defects result in the degradation in thermal

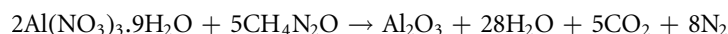
conductivity. Rocha *et al* [29, 30] suggested that nanofluids have a negligible impact on neutron transport in the core due to the low concentration of nanoparticles. They emphasized on proper nanoparticles selection for better compatibility with the chemical and irradiation environment of the reactor. However, there is a need to investigate the neutron-irradiation sensitivity of the thermal conductivity of nanofluids for different durations of the exposure.

In this study, the effect of neutron irradiation has been evaluated on the thermal conductivity of Al_2O_3 nanofluids. Al_2O_3 nanoparticles have been synthesized using the solution combustion method and used for the preparation of nanofluids in distilled water. These nanofluids have been exposed to neutron-irradiation for different durations. Un-irradiated and neutron-irradiated nanofluids have been characterized using Dynamic Light Scattering, Scanning Electron Microscopy, and Ultraviolet-Visible Spectroscopy to ascertain changes in characteristics due to irradiation. The thermal conductivity of un-irradiated and neutron-irradiated nanofluids has been measured and reported in the paper. Neutron-irradiation sensitivity of thermal conductivity has also been calculated. These results will be helpful in understanding the implications of utilizing nanofluids in nuclear reactors.

2. Materials and method

2.1. Synthesis of Al_2O_3 nanoparticles

Al_2O_3 nanoparticles have been synthesized using the procedure adopted from our previous work [31]. Precisa (XB 220 A model) weighing balance, having a precision of 0.0001 g, has been used for all the weight measurements. The stoichiometry amount of the base chemicals required for the synthesis has been calculated using molecular weight. To synthesize Al_2O_3 nanoparticles, 7.5026 g aluminum nitrate has been dissolved in 10 ml distilled water using magnetic stirrer. 3.003 g urea has been added to this solution through vigorous stirring. The solution has been heated to obtain a transparent sticky gel. The gel has been first dehydrated at 100 °C for 2 h and then combusted at 1000 °C for 2 h. The combusted sample has been ground to obtain a fine powder of Al_2O_3 nanoparticles. Following is the equation showing the formation of Al_2O_3 Nanoparticles:



2.2. Preparation of Al_2O_3 nanofluids

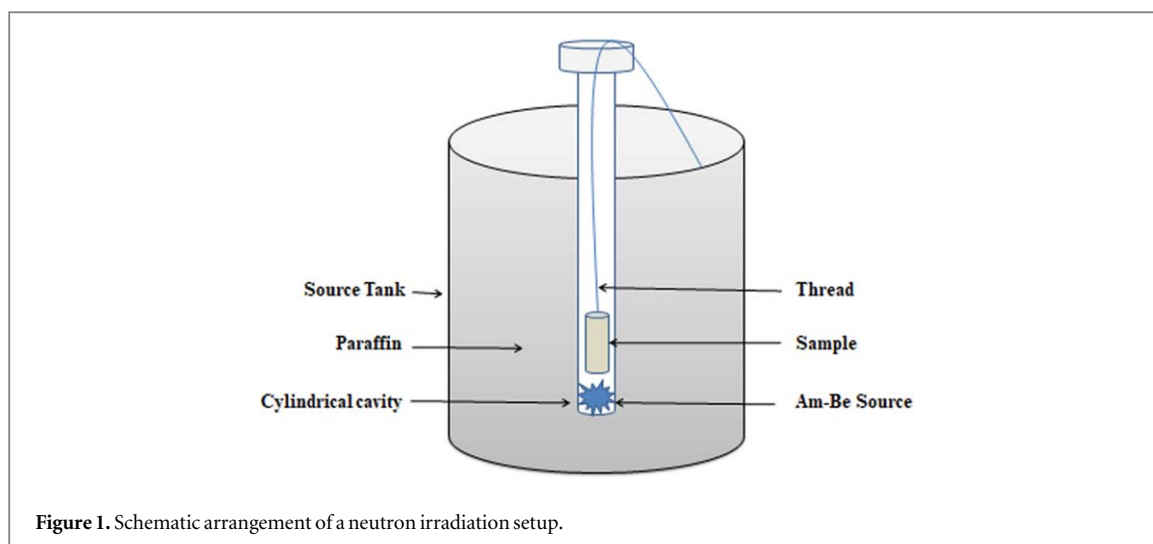
Al_2O_3 nanofluids of 0.25, 0.50, 0.75, 1.00, 1.25, 1.50, 1.75 and 2.00 volume % concentrations have been prepared in distilled water (Polisher; Biopak) base fluid using a two-step approach from the synthesized Al_2O_3 nanoparticles. The required amount of nanoparticles for the preparation of different volume concentration nanofluids have been calculated using the law of mixture formula. Nanofluids of desired concentrations have been prepared by suspending the required amount of synthesized Al_2O_3 nanoparticles in the base fluids. The suspensions have been stirred for 1 h using magnetic stirrer (Tarsons; SPINOT), sonicated for 30 min using probe ultrasonic processor (Electrosonic; E1-250 W) and undergone ultrasonic vibrations for 90 min using water bath ultrasonicator (Toshcon; SW4), so as to obtain homogenous stable nanofluids suspension.

2.3. Neutron irradiation of nanofluids

The prepared nanofluid samples have been irradiated by neutron flux using the setup of Meena *et al* [32]. Radioisotopic Am-Be neutron source has been used for irradiation as it produces highly stable flux and is an efficient option for aqueous samples. Figure 1 shows a schematic of the neutron source setup used for the irradiation of nanofluids. The neutron irradiation setup consists of a neutron source tank which is a 0.5 cm thick steel cylinder (diameter 92 cm and height 122 cm) filled with paraffin. This source tank consists of a central cylindrical cavity of 20 cm diameter inside of which the neutron source was placed. The nanofluid samples to be irradiated have been placed inside the central cylindrical cavity near the neutron source.

2.4. Measurement of thermal conductivity

The thermal conductivity of un-irradiated and neutron-irradiated nanofluids samples after exposure of 7 and 14 days has been measured at 30 °C using KD2 Pro Thermal Properties Analyzer of Decagon Devices, which is based on the transient line heat source method. To measure the thermal conductivity, KS-1 sensor of needle diameter 1.3 mm and length 60 mm have been inserted vertically in nanofluid containers of 50 ml volume, 30 mm diameter and 120 mm length that has been placed upside down. Dimensions of the container are large enough to be considered infinite as compared to the sensor needle. The performance of the sensor has been verified with the standard glycerin sample supplied with the KD2 Pro. Measured thermal conductivity of the



standard glycerin sample has been $0.289 \text{ W m}^{-1} \cdot \text{K}$ which is comparable to $0.285 \text{ W m}^{-1} \cdot \text{K}$ as reported by the manufacturer.

Average of three measurements of thermal conductivity has been taken for each sample to ensure the accuracy and consistency of the results. A time gap of 30 min has been kept between consequent measurements to nullify the effect of temperature increase in the vicinity of the probe due to transient heat. Results hence obtained have found to be consistent and reproducible.

3. Results and discussion

3.1. Nanofluids stability analysis

The stability of the prepared nanofluids has been analyzed after 7 and 14 days. Figure 2 shows images of Al₂O₃ nanofluids as prepared, after 7 days and after 14 days. No visible sedimentation shows the absence of aggregation and agglomeration of nanoparticles. Table 1 shows the results of the thermal conductivity measurement of un-irradiated Al₂O₃ nanofluids as prepared, after 7 days and after 14 days which are within the accuracy region. Figure 2 along with table 1 verifies the stability of prepared nanofluids.

3.2. Characterization of Al₂O₃ nanofluids

The un-irradiated and neutron-irradiated Al₂O₃ nanofluids have been characterized using Dynamic Light Scattering (DLS, Malvern, Nano-ZS), Scanning Electron Microscopy (SEM, Carl-Zeiss, EVO-18) and UV-visible Spectroscopy (UV-vis, Thermo Fisher, Multiskan) to ascertain stability, size and absorption characteristics. Al₂O₃ nanofluids of 0.25 volume % concentration have been used for the characterization studies.

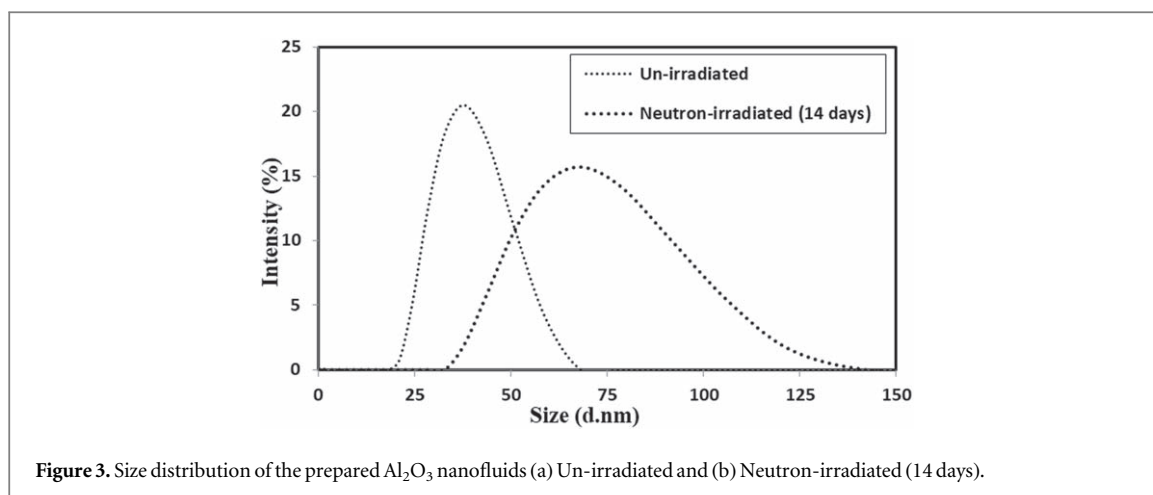


Figure 3. Size distribution of the prepared Al_2O_3 nanofluids (a) Un-irradiated and (b) Neutron-irradiated (14 days).

Table 1. Thermal conductivity of Un-Irradiated Al_2O_3 Nanofluids after 7 and 14 days.

Volume Percent (vol. %)	Thermal conductivity (W/(m.K)) of Un-Irradiated Al_2O_3 nanofluids		
	As prepared	After 7 days	After 14 days
0	0.614	0.613	0.614
0.25	0.626	0.624	0.624
0.5	0.636	0.635	0.633
0.75	0.646	0.646	0.644
1	0.657	0.654	0.655
1.25	0.668	0.666	0.664
1.5	0.678	0.674	0.673
1.75	0.685	0.682	0.682
2	0.695	0.692	0.691

3.2.1. DLS

Figure 3 shows the size distribution of un-irradiated and neutron-irradiated Al_2O_3 nanofluids. Weighted average particle size for un-irradiated Al_2O_3 nanofluids is 44 nm whereas 70 nm for the neutron-irradiated Al_2O_3 nanofluids. An increase in average size indicates agglomeration and aggregation after neutron irradiation.

The zeta potential values, 35.3 mV and 34.2 mV, have been obtained for the un-irradiated and neutron-irradiated Al_2O_3 nanofluids after 14 days, respectively (figure 4). Shoulder like behavior in zeta potential graph of the neutron-irradiated sample may be attributed to the slight presence of another population arising out due to the change in properties of the sample under the influence of the neutron-irradiation. Longer tails in the hydrodynamic size distribution of neutron-irradiated samples also emphasize the minute presence of relatively larger-sized particles, forming another population, resulting in the shoulder like behavior in zeta potential graph.

3.2.2. SEM

Figure 5 shows the images of Al_2O_3 nanofluids before and after the neutron-irradiation. Nanosize distribution of synthesized nanoparticles in the prepared nanofluids is evident from the figure. For un-irradiated Al_2O_3 nanofluids, the average particle size is 45 nm with a standard deviation of 2 nm. Figure 5(a) also shows the uniform distribution of nanoparticles in the nanofluids with no aggregation. Figure 5(b) shows the formation of a few clumps with an average particle size of 73 nm with a standard deviation of 3 nm for the neutron-irradiated nanofluids.

3.2.3. UV-vis

Figure 6 shows the absorption spectra of un-irradiated and neutron-irradiated nanofluids. For un-irradiated Al_2O_3 nanofluids, absorption maximum appears at 359 nm and for neutron-irradiated nanofluids, the respective absorption maximum appears at 366 nm. The shift in absorption maximum wavelength indicates an

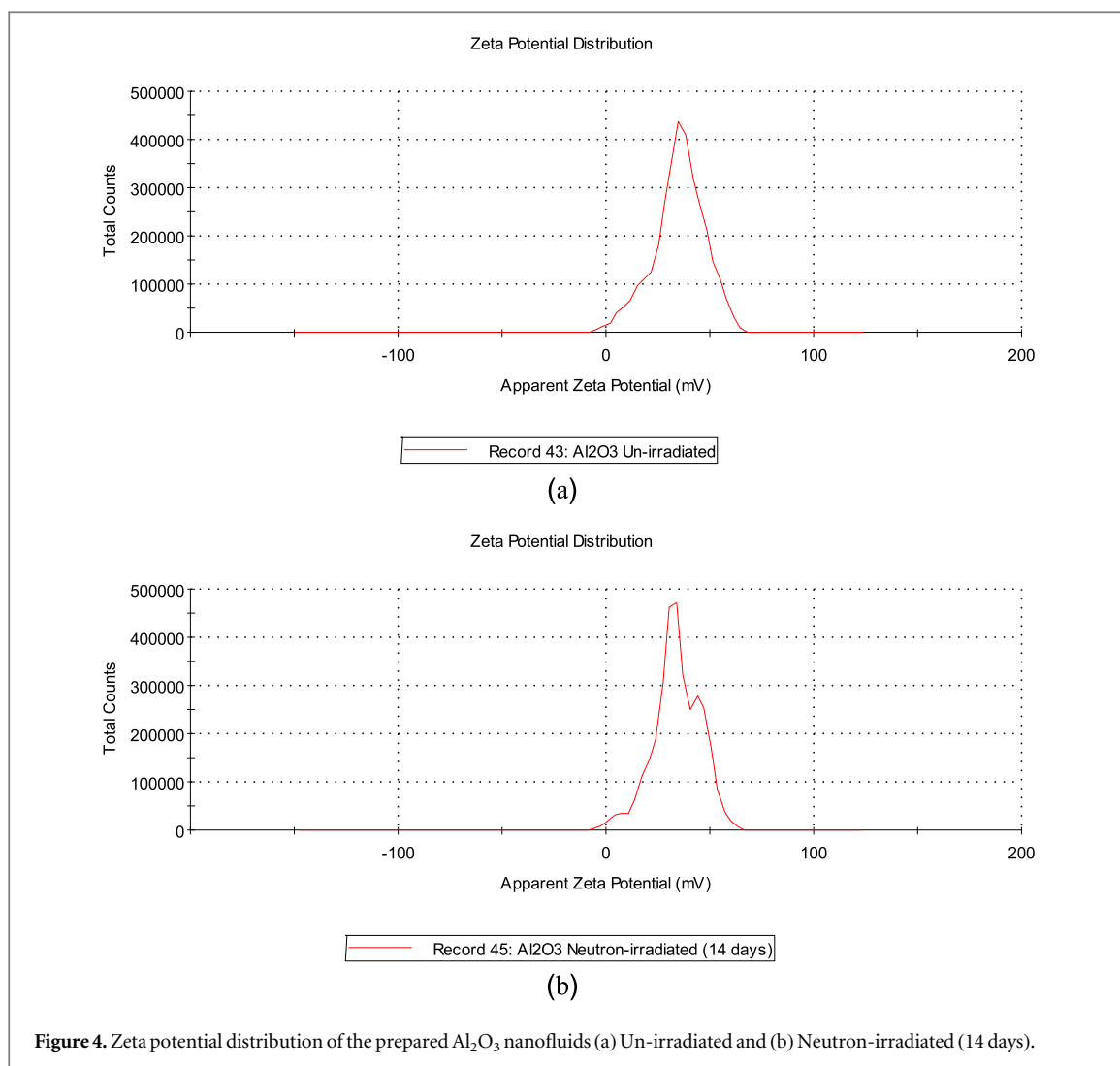


Figure 4. Zeta potential distribution of the prepared Al_2O_3 nanofluids (a) Un-irradiated and (b) Neutron-irradiated (14 days).

increase in the size that in turn indicates the agglomeration or aggregation of nanoparticles after neutron-irradiation.

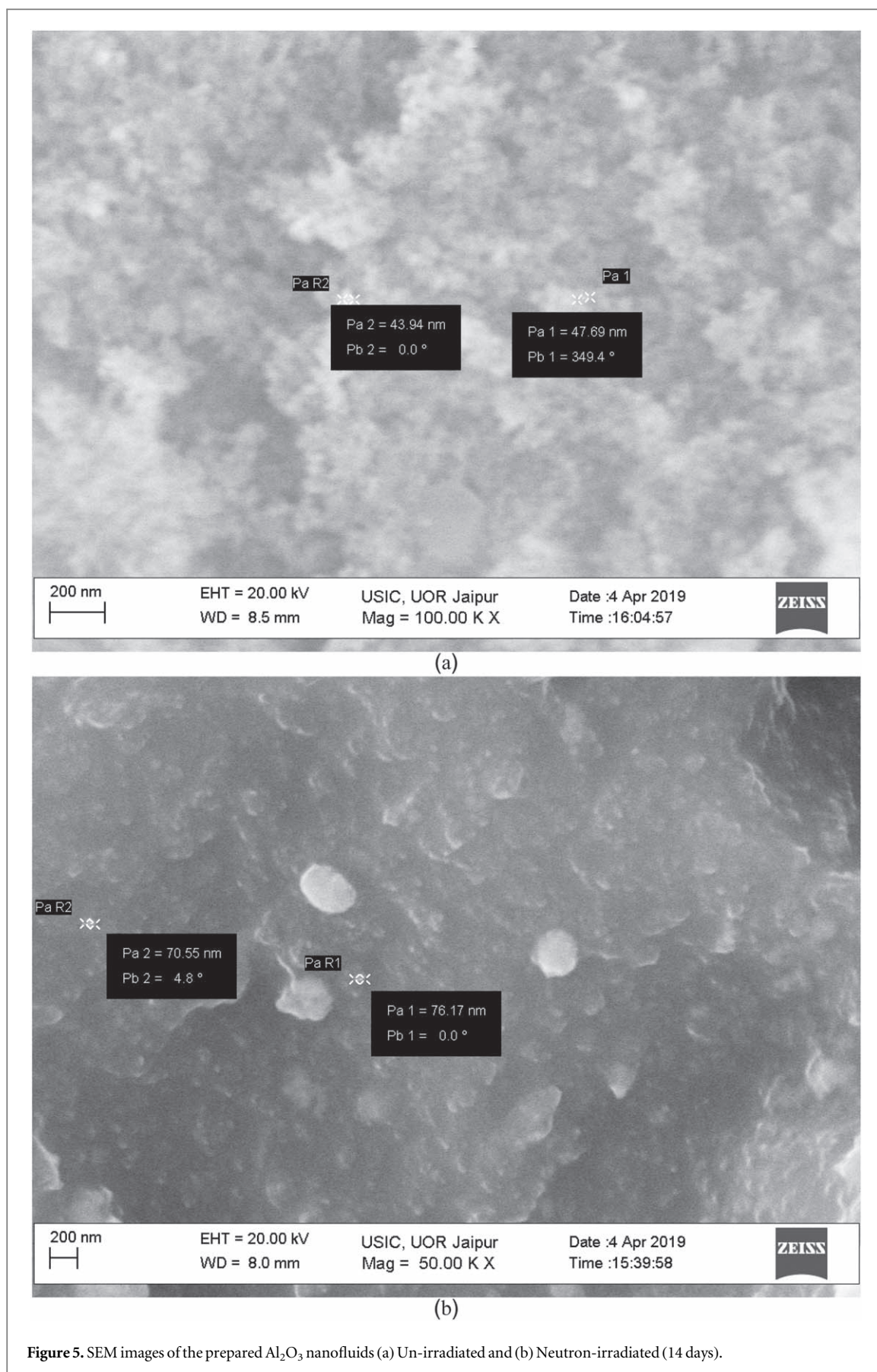
Calculated bandgap energy using the Tauc plot (figure 7) is 3.17 and 3.11 eV for un-irradiated and neutron-irradiated nanofluids. The slight reduction in energy level favors more absorption of excited valance band electrons.

3.3. Thermal conductivity measurement analysis

Thermal conductivity measurements show a significant change in thermal conductivity with a change in concentration of nanoparticles and/or exposure to neutron-irradiations. Figure 8 shows the change in thermal conductivity with an increase in the concentration of Al_2O_3 nanoparticles in distilled water base fluids for un-irradiated and neutron-irradiated samples. Results show that the thermal conductivity of un-irradiated Al_2O_3 nanofluids is higher as compared to distilled water that further increases with an increase in the concentration of Al_2O_3 nanoparticles in the base fluid. An increase of 13% in thermal conductivity has been observed for the increase in the concentration of nanofluids from 0 to 2 volume percent.

Xie *et al* [33] demonstrated that the thermal conductivity enhancement of nanofluids is influenced by multifaceted factors including the volume fraction of the dispersed nanoparticles, the tested temperature, the thermal conductivity of the base fluid, the size of the dispersed nanoparticles, the pretreatment process, and the additives of the fluids [34–37]. Various models show that the enhancement in thermal conductivity is the combined effect of nanomaterial properties *viz.* size, shape, agglomeration and medium properties [38–40].

Figure 8 also shows a decrease in the thermal conductivity of nanofluids that has been exposed to neutron-irradiation. Radiation affects the material's electrical, thermal, structural, physical and mechanical properties. A change in the material properties is evident from SEM images (figure 5). DLS size distributions (figure 3) exhibit the increase in weighted average particle size along with the long tails for the neutron-irradiated samples. An increase in the average size indicates agglomeration after neutron-irradiation and it has been well established



that agglomeration leads to a decrease in thermal conductivity of nanofluids. With the increase in nanoparticle concentration, the thermal conductivity increases for un-irradiated nanofluids but decreases for neutron-irradiated nanofluids. The observation also supports that neutron irradiation tends to increase agglomeration

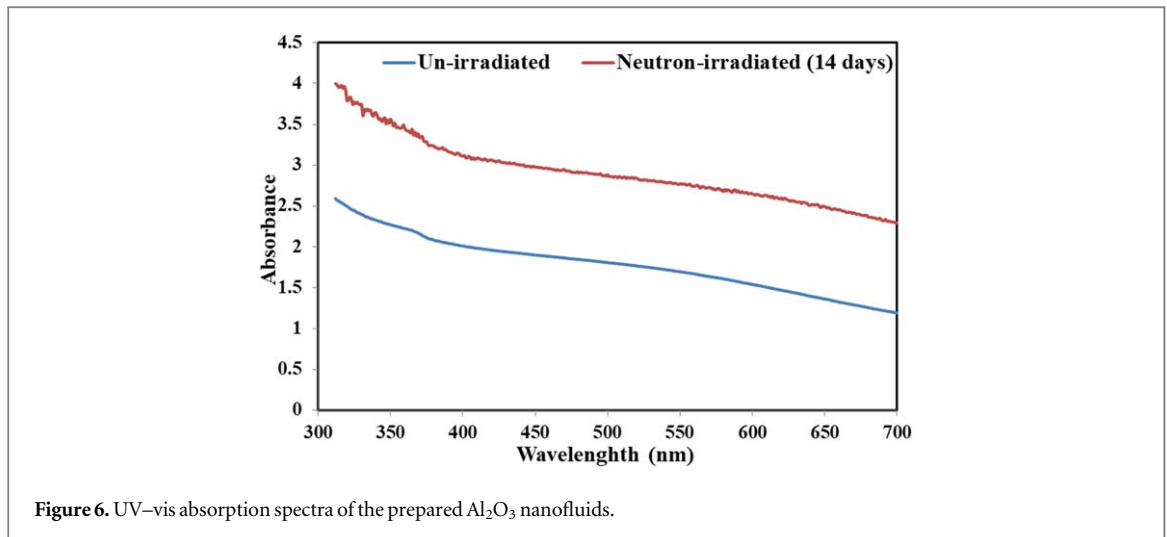


Figure 6. UV-vis absorption spectra of the prepared Al_2O_3 nanofluids.

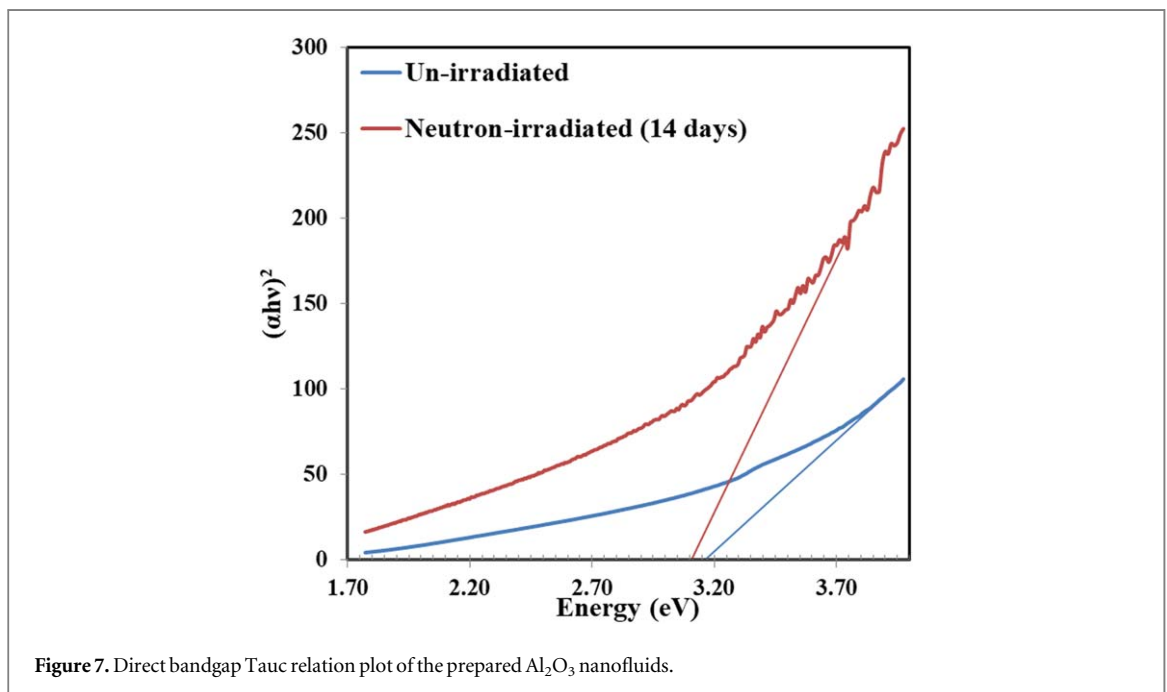


Figure 7. Direct bandgap Tauc relation plot of the prepared Al_2O_3 nanofluids.

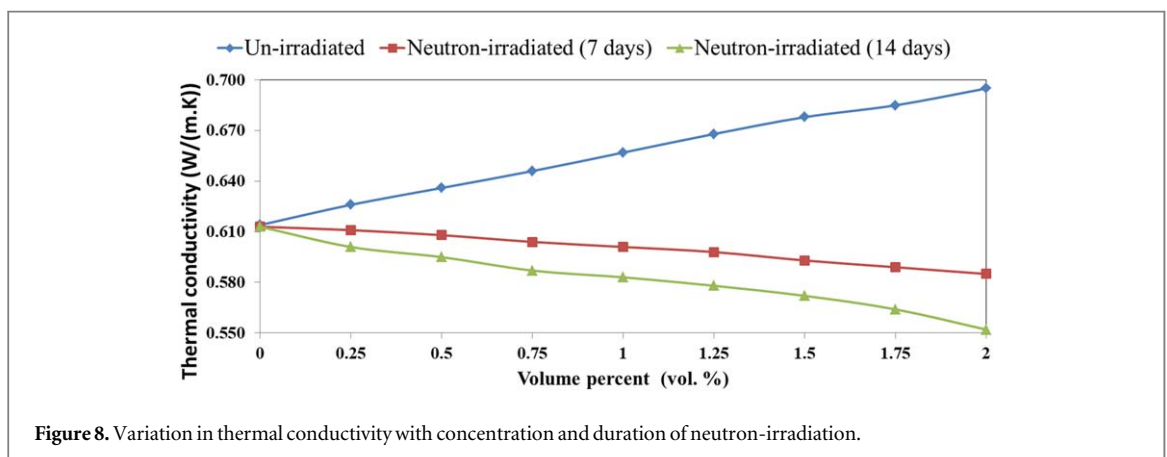


Figure 8. Variation in thermal conductivity with concentration and duration of neutron-irradiation.

and this agglomeration is more prominent at higher concentrations resulting in the larger decrease in thermal conductivity at higher concentrations. It has also been suggested that the bombardment of neutrons on a material induce defects [28] and vacancies [41] apparent from UV-vis spectra (figure 6). Further, the decrease in

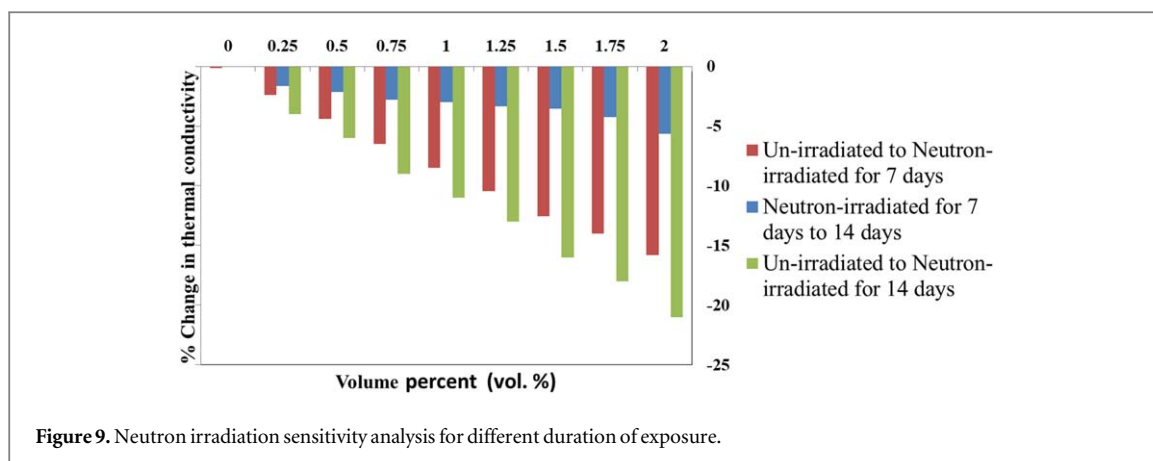


Figure 9. Neutron irradiation sensitivity analysis for different duration of exposure.

thermal conductivity is more prominent with increased duration of radiation exposure. For an increase in concentration from 0 to 2 volume percent, 5 and 10% decrease in thermal conductivity have been obtained for Al_2O_3 nanofluids exposed to neutron-irradiation for 7 and 14 days, respectively.

3.4. Neutron irradiation sensitivity analysis

The sensitivity of a quantity with a change in different parameters is evaluated in a sensitivity analysis. Here neutron-irradiation exposure duration has been changed and respective thermal conductivity sensitivity has been evaluated. Figure 9 shows the sensitivity of thermal conductivity for different duration of exposure of neutron-irradiation. Sensitivity analysis reveals that the decrease in thermal conductivity is highly sensitive to neutron-irradiation during initial exposure of 7 days which is also higher at higher concentration.

4. Conclusions

In the present study, the thermal conductivity of Al_2O_3 nanofluids has been investigated for the neutron-irradiation sensitivity. Following are the important inferences drawn from the results of the study:

- (1) The thermal conductivity of neutron-irradiated Al_2O_3 nanofluids is lowered as compared to un-irradiated nanofluids that further decreases with increased duration of exposure. For a change in concentration from 0 to 2 volume percent, 5 and 10% decrease in thermal conductivity has been observed for Al_2O_3 nanofluids exposed to neutron-irradiation for 7 and 14 days, respectively.
- (2) For neutron-irradiated Al_2O_3 nanofluids heat carrying capacity also decreases with increasing concentration contrary to un-irradiated nanofluids where thermal conductivity increases with increasing concentration.
- (3) Neutron irradiation sensitivity analysis revealed that heat transfer characteristics are more sensitive at higher concentrations. It can also be inferred that neutron irradiation sensitivity is most prominent at initial exposure and this sensitivity decreases with increased duration of exposure.

The results of the study emphasize the need for a detailed investigation of the applicability of nanofluids in nuclear reactor systems.

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