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RESEARCH ARTICLE - MECHANICAL ENGINEERING

Stability of Aqueous Nanofluids Containing PVP-Coated Silver Nanoparticles

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Abstract Nanofluids have opened a new arena for researchers in the field of heat transfer with their exceptional heat transfer characteristics. Enhanced thermal conductivity and improved stability are the principal advantages of nanofluids for its applications in heat transfer. This paper presents an experimental investigation on the stability of silver–water nanofluids prepared by dispersing 0.1 % volume fraction of polyvinylpyrrolidone-coated silver nanoparticles in distilled water with and without the addition of surfactants. The surfactants used in the present study are polyvinylpyrrolidone and sodium dodecyl sulfate. The stability of the nanofluids was estimated from sedimentation time, pH value, zeta potential and particle size distribution. Thermal conductivity of the nanofluids was measured by thermal property analyzer. It has been found that the stability of nanofluids is influenced predominantly by the size of the particle and the surfactant characteristics. The stability of nanofluid increases with the decrease in the size of nanoparticles. Also, the stability increases with sodium dodecyl sulfate as surfactant as against polyvinylpyrrolidone. However, enhancement in the thermal conductivity is found to be higher with polyvinylpyrrolidone than with sodium dodecyl sulfate.

Keywords Nanofluid · Silver nanoparticles · Stability · Zeta potential

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1 Introduction

Nanofluid is an emerging heat transfer fluid consisting of nanoscale particles suspended in any conventional heat transfer fluids such as water, oil or ethylene glycol. By dispersing very low concentration of nanophase particles ($<1\,\%$ volume fraction) in the traditional heat transfer fluids, the thermal conductivity of the fluid is increased which enhances the overall heat transfer performance of the fluid [1]. These improved characteristics of nanofluids make them potentially useful in the application of electronics, transportation, nuclear reactors, biomedical engineering, heating, ventilation and air-conditioning, energy-efficient heat transfer equipments [2] and electrical engineering [3].

The stability of a nanofluid affects its thermal conductivity. Better dispersion of the nanoparticles in the fluids increases the surface area and surface activity, thus increasing its thermal conductivity. Addition of nanoparticles not only enhances the thermal conductivity, but also leads to agglomeration. This causes less stability to the fluid and a drop in the thermal conductivity [4,5]. Therefore, suppressing the aggregation of the nanoparticles in the nanofluid has become a major issue in the primary level of research for heat transfer applications. Many investigations have been carried out in addressing the issues related to the stability of the nanofluids [2,4–13] and the thermal conductivity enhancement [14–19]. Experimental studies on the nanofluids have shown that the amount and the charge of nanoparticles in the nanofluid and the interaction between the nanoparticles and the dispersant influence the stability of the nanofluid directly. Addition of surfactants, agitation and changing the pH value of the suspension are the various techniques adopted for reducing particle agglomeration for better stability [2]. In spite of extensive research carried out on investigating an effective technique

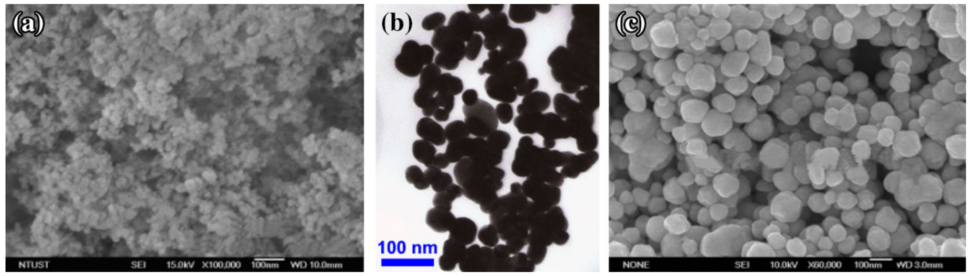


Fig. 1 TEM images of PVP-coated silver nanoparticles, **a** 20 nm, **b** 30–50 nm, and **c** 50–80 nm

for reducing agglomeration, the issue is still not completely addressed.

A considerable amount of research has been done on the numerical and experimental investigations on the enhancement of thermal conductivity using nanofluids. Also, several researchers have studied the heat transfer performance with the nanofluids in natural and forced convection with metal and metal oxide nanoparticles [20–23]. Several studies indicate that nanofluids with metallic nanoparticles have better heat transfer performance than metal oxide nanofluids [22, 23]. Silver is a common and safe material, and it is considered as a suitable material in heat transfer applications with its high thermal conductivity of the order of 429 W/m-K at room temperature [14]. On the other hand, the thermal conductivity of water is higher among other conventional heat transfer fluids. Also, water is historically used as a cooling fluid in most of the thermal devices like heat exchangers, internal combustion engines and solar collectors. In the present study, polyvinylpyrrolidone-coated silver nanoparticles were dispersed in distilled water. Polyvinylpyrrolidone (PVP) and sodium dodecyl sulfate (SDS) were used as surfactants for stabilizing the nanofluids. The effect of particle size, surfactant characteristics and pH value of nanofluid on the stability of silver–water nanofluids has been experimentally investigated using various techniques like sedimentation method, pH measurement, zeta potential and particle size distribution. The effect of particle volume fraction and the surfactant characteristics on the enhancement of the thermal conductivity has also been investigated. Most of the earlier researches were made on the particle volume fractions above 1 %. In the present study, a far smaller particle volume fraction of 0.1 % is considered to study its influence on the stability and thermal conductivity of the nanofluids.

2 Experimental

2.1 Materials

Silver nanoparticles of average sizes 20, 30–50 and 50–80 nm, 99.9 % purity, spherical shape, coated with 0.2 w % polyvinylpyrrolidone (PVP) were purchased from US Research Nanomaterials, Inc. (Houston, USA). Analytical-grade PVP and SDS bought from Sigma-Aldrich Chemicals Ltd were used as received without further purification. A 100-kW, 40-KHz ultrasonic bath of 31 capacity, magnetic stirrer (1MLH) and Shimadzu make electronic balance were used for the preparation of the nanofluids.

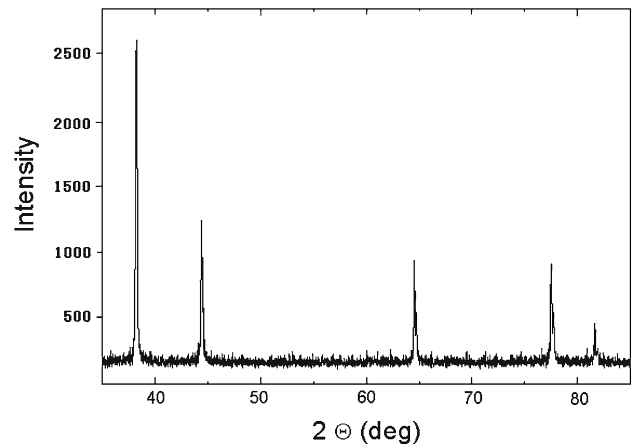


Fig. 2 XRD image of PVP-coated silver nanoparticles

2.2 Characterization

The TEM images of the three sizes of PVP-coated silver nanoparticles and XRD image were provided by the vendor. TEM images shown in Fig. 1 depict the size and morphology of PVP-coated silver nanoparticles. XRD image shown in Fig. 2 confirms the metallic structure of PVP-coated silver nanoparticles. Density of PVP-coated silver nanoparticles is 10.5 g/cm^3. The average molecular weight of polyvinylpyrrolidone (PVP) and sodium dodecyl sulfate (SDS) used as surfactant are 40,000 and 288.38, respectively.

2.3 Dispersion of Surfactants

Based on the extensive literature survey, among the wide range of surfactants available, PVP and SDS are chosen as good surfactants. By mass, 1 % of PVP and SDS were mixed separately in 100 ml of distilled water. In order to evaluate the dispersion nature of the surfactants, the absorbance of the two surfactants was measured with UV–vis spectropho-

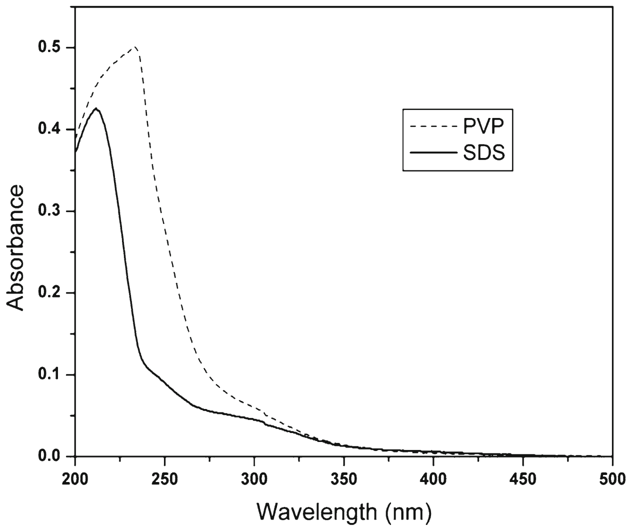


Fig. 3 UV–vis spectrum of PVP and SDS in water suspension

tometer (UV-2450, SHIMADZU, JAPAN). Figure 3 shows that the peak absorbance of PVP and SDS in water suspensions appears at 233 and 211 nm, respectively. The peak absorbance at lower wavelengths shows the better dispersion of the surfactants in distilled water.

2.4 Preparation of Nanofluids

Stable suspensions of nanoparticles in traditional heat transfer fluids are produced by one-step method and two-step method. Two-step method is considered to be the economical method for the large-scale production of the nanofluids for heat transfer applications. Therefore, the two-step method has been employed in the present study. PVP-coated nanoparticles avoid oxidation of metal nanoparticles. Accurately weighed PVP-coated silver nanoparticles and surfactants were dispersed in 100 ml of distilled water to make sample nanofluids. Initially, it was stirred for 30 min in the magnetic stirrer followed by ultrasonication in the ultrasonic bath for 3 h. The use of mechanical stirring and ultrasonication is to break down the agglomeration of the nanoparticles and to keep the suspension stable for a longer time.

3 Stability of Nanofluids

3.1 Sedimentation Technique

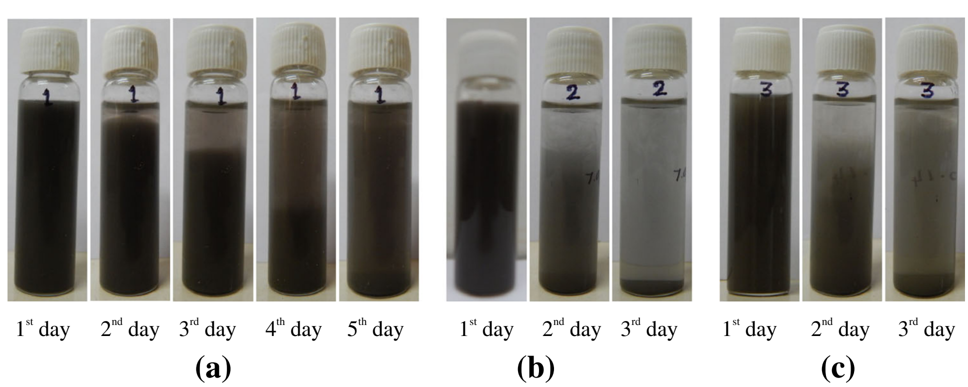
Sedimentation technique is one of the simple techniques used for the measurement of stability of the nanofluids [24]. A known volume of nanofluid sample is kept in a small glass container and photographed at regular time interval. The first image was taken immediately after the preparation of sample and it continued until the nanoparticles settled completely in the container. The samples of silver–water nanofluid were prepared with the three test sizes (20, 30–50 and 50–80 nm) without the addition of surfactants. The volume fraction of the nanoparticles was limited to 0.1 %. The sample nanofluids were kept in glass containers of 10 ml capacity. The sedimentation of the nanoparticles was photographed with a camera every 24 h till the particles settled down completely in the container.

From Fig. 4, it is observed that the nanoparticles of sizes 30–50 and 50–80 nm settled almost within 24 h. But the settling of particles of 20 nm sizes started on the second day slightly and they completely settled only on the fifth day. Hence it is clear that the size of the particles plays an important role in the stability of the nanofluids. The relation between sedimentation velocity and size of the particles in stationery state is given by Stokes law [1].

$$V = \frac{2R^2}{9\mu}(\rho_P - \rho_L)\,g \tag{1}$$

where V is the sedimentation velocity of the particles; R is the radius of the particles; μ is the viscosity of the liquid medium; ρ_p and ρ_L are the density of the particles and liquid medium, respectively; g is the acceleration due to gravity. Enhancement of the stability of the nanofluid can be achieved by reducing the sedimentation velocity of the nanoparticles. According to Stokes, the sedimentation

Fig. 4 Photographs of the silver–water nanofluid without surfactant of nanoparticles size, **a** 20 nm, **b** 30–50 nm, **c** 50–80 nm



velocity can be reduced by using smaller particles. However, the smaller the nanoparticles, the higher will be the surface energy and the possibility of agglomeration. Therefore, the main challenge in preparing the nanofluid is a kind of trade-off between decreasing the sedimentation velocity and suppressing agglomeration simultaneously. One way of suppressing agglomeration without affecting the sedimentation velocity is the addition of surfactants to the nanofluids. To study the effects of surfactant on the stability, PVP-coated silver nanoparticle of 20 nm is chosen since it is more stable than the other two nanoparticles. Two separate samples of silver–water nanofluids were prepared with 0.1 % volume fraction of 20 nm PVP-coated silver nanoparticles with PVP and SDS as surfactants. The concentration of surfactants in the fluid normally varies from 10 to 50 % (1/10–1/2) of the nanoparticles added by mass. Here the concentrations of the surfactants were limited to 10 and 20 % mass fraction of the nanoparticles added in the base fluid.

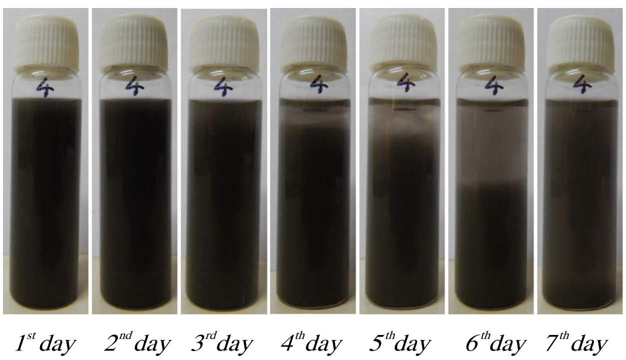


Fig. 5 Photographs of nanofluid with 20 nm silver nanoparticles and PVP (10 %)

The above figures show the photographs of the silver–water nanofluids with two different concentrations of PVP and SDS as surfactants. From Fig. 5, it is observed that PVP-coated silver nanoparticles started to settle in the nanofluid with 10 % PVP on the third day of preparation and they completely settled on the seventh day. On the other hand, the nanoparticles in the nanofluid with 10 % SDS started to settle on the fourth day and they completely settled only on the tenth day of preparation (Fig. 6). Similarly from Fig. 7, it is observed that PVP-coated silver nanoparticles started to settle in the nanofluid with 20 % PVP on the fourth day of preparation and they completely settled on the ninth day but the nanoparticles in the nanofluid with 20 % SDS started to settle on the fifth day and they completely settled only on the eleventh day of preparation (Fig. 8). Also, from Figs. 5, 6, 7 to 8, it is clear that the addition of surfactant as well as increase in the concentration of surfactant enhances the stability of nanofluids. The nanofluids with SDS are more stable than the fluid with PVP.

Fig. 6 Photographs of nanofluid with 20 nm silver nanoparticles and SDS (10 %)

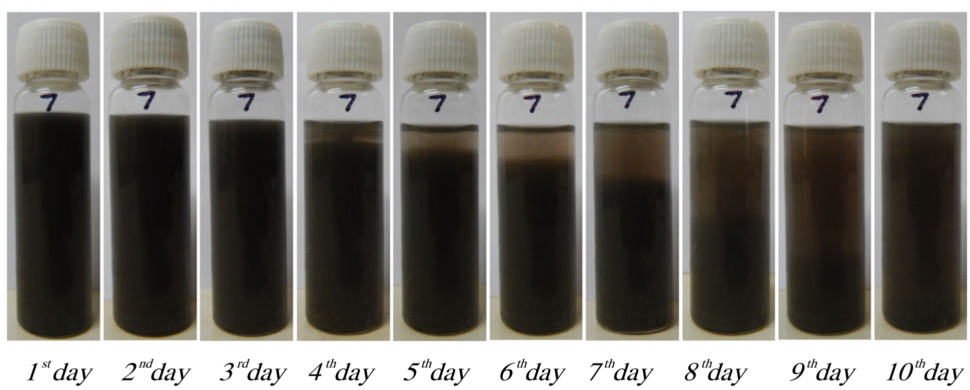
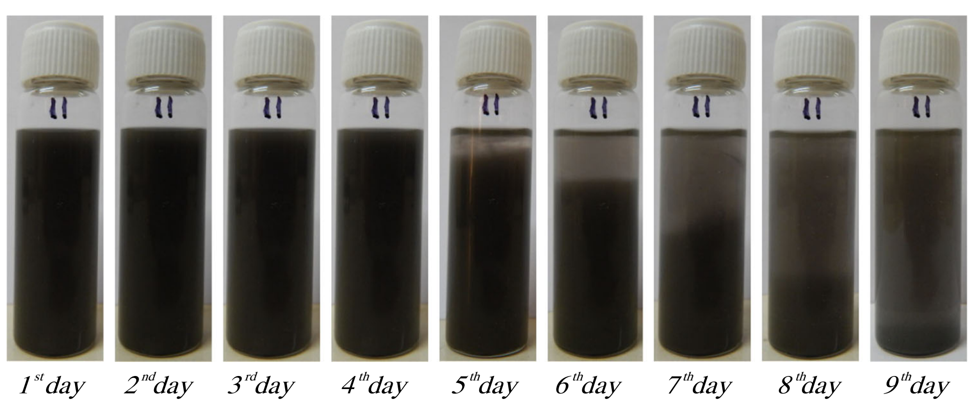


Fig. 7 Photographs of nanofluid with 20 nm silver nanoparticles and PVP (20 %)



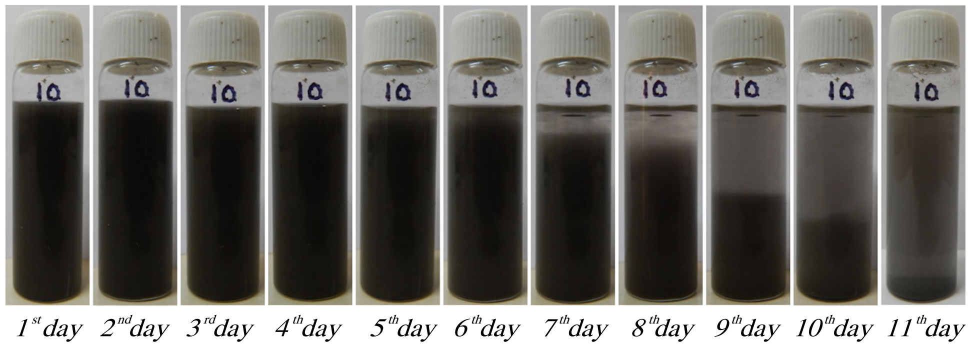


Fig. 8 Photographs of nanofluid with 20 nm silver nanoparticles and SDS (20 %)

3.2 pH Measurement

The pH values of the suspensions have direct effect on the stability enhancement of the nanofluids. The pH values of the nanofluids prepared with and without surfactant were measured using a digital pH meter. The data are presented in Table 1.

From Table 1, it is observed that pH value of the nanofluids without the surfactant increases drastically on the next day itself and it continues till the particles are completely settled. Once they are settled, the pH value remains unchanged. However, pH value of the nanofluids with the addition of surfactant increases just slightly. It remains unchanged after the nanoparticles are settled completely. Also, it is clear that pH values of the nanofluid with surfactant are lower than that without surfactants. The lower the pH value, the higher is the stability of the nanofluid. The decrease in pH value of the nanofluid increases the hydration forces among the particles. It results in the better mobility of the nanoparticles in the nanofluid enhancing the heat transfer process. The pH value of the nanofluids should be low for better heat transfer [25,26]. The pH value of the nanofluid with SDS is lower than PVP. Hence SDS shows improved stability than PVP.

3.3 Zeta Potential and Particle Size Distribution

Zeta potential is an index of the magnitude of the interaction between colloidal particles. Also, it is a measure of dispersion stability of the nanofluid. Higher values of zeta potential indicate more stable dispersion. Particles with zeta potentials more positive than +30 mV or more negative than −30 mV are normally considered stable. The zeta potential and average particle size of the nanofluids were measured with dynamic light scattering system (Zetasizer Nano-ZS, Malvern Instruments Inc., UK).

The zeta potential and particle size distribution of silver–water nanofluids with and without surfactants are presented in Table 2. From the table, it is observed that the zeta potential of the nanofluids prepared without surfactants is lower than −30 mV. It shows that the nanofluids without surfactant are unstable. The zeta potential of the nanofluids with 10 and 20 % of PVP is −29.4 and −30 mV, respectively. However, the zeta potential of the nanofluids with 10 and 20 % of SDS is −32.8 and −34 mV, respectively. The zeta potential of the nanofluid with SDS is higher than PVP.

Zetasizer measures the hydrodynamic size of the particles. According to Stokes–Einstein equation, the average particle size of the nanoparticles in the nanofluid is higher than the actual size [27]. However the average particle sizes of the nanoparticles in the nanofluids with the surfactant are smaller than that without surfactant. The increase in the average particle size of the PVP-coated silver nanoparticles in the nanofluids without the surfactant is due to the agglomeration of the nanoparticles. The average particle size of the nanofluid with SDS is lower than PVP. Also, the increase in the concentration of the surfactant leads to decrease in the average particle size of the nanoparticles in the nanofluids. The lower the average particle size of the nanoparticles in the nanofluid, the higher is its stability. The zeta potential of the nanofluid increases with the decrease in average particle size of the nanoparticles. From the measurements of the zeta potential and average particle size of the nanofluids, it is understood that SDS provides more stability to the nanofluid than PVP. Also, the stability of the nanofluids enhances with the concentration of the surfactants.

The three techniques adopted in the present study show that the stability increases with the addition of surfactant as well as the concentration of surfactant. Among the two surfactants used, SDS has shown relatively higher stability than PVP. SDS is an anionic surfactant, whereas PVP is a neutral surfactant. The anionic surfactant measures high zeta potential compared to neutral surfactant. Higher zeta potential of the nanofluids indicates its good stability. The major drawback in using SDS as surfactant is foam production that obstructs the fluid flow in applications like heat exchangers.

4 Thermal Conductivity of Nanofluid

As the size of the nanoparticles is smaller, the nanofluid will be more stable. Thermal conductivity measurement was carried out on a silver–water nanofluid sample with

Table 1 pH of silver–water nanofluids

Size of Ag NP (nm)	Volume concentration of Ag NP (%)	Surfactant added	Concentration of surfactant (% by mass of NP added)	Value of pH on the no. of day after preparation										
				1	2	3	4	5	6	7	8	9	10	11
20	0.1	–	–	6.93	7.19	7.36	7.41	7.45	Nanoparticles settled					
30–50	0.1	–	–	7.17	7.47	7.50	7.5	7.56	Nanoparticles settled					
50–80	0.1	–	–	7.19	7.75	7.95	7.97	7.96	Nanoparticles settled					
20	0.1	PVP	10	6.81	6.82	6.86	6.87	6.90	6.92	6.95	Nanoparticles settled			
20	0.1	SDS	10	6.56	6.56	6.58	6.60	6.65	6.68	6.72	6.76	6.81	6.86	
20	0.1	PVP	20	6.67	6.69	6.70	6.71	6.73	6.76	6.80	6.82	6.86	NP settled	
20	0.1	SDS	20	6.50	6.51	6.53	6.54	6.57	6.59	6.62	6.65	6.67	6.70	6.73

20 nm PVP-coated nanoparticles. The volume fraction of the nanoparticles was varied from 0.01 to 0.1 %. The concentration of PVP and SDS was 10 %, by mass of the nanoparticles added. The thermal conductivity of the nanofluids was measured with KD2 Pro thermal analyzer (Decagon Devices, Inc., USA) which is based on transient hot-wire technique. Initially, the device was calibrated using distilled water. The thermal conductivity of distilled water was measured as 0.6196 W/mK at 309 °C which is in good agreement with the reference data of 0.613 W/mK with the error of ±1 %. The sample nanofluid was taken in a glass container, and it was kept inside the circulating system of constant temperature water bath (JEIO Tech, Korea, Capacity: 51, Temperature Range: −259 °C to +1509 °C, Temperature stability: ±0.05/0.09 °C) to maintain constant temperature during the experiment. The bath temperature was maintained at 30 °C.

Figure 9 shows the variation of thermal conductivity of silver–water nanofluids with respect to volume fraction of PVP-coated silver nanoparticles with and without surfactant. The thermal conductivity of the nanofluid without surfactant increases from 12.72 to 54.33 % for the variation of volume fraction from 0.01 to 0.1 %. With PVP as surfactant, the thermal conductivity of the nanofluid increases from 12.43 to 52.19 %, whereas it increases from 12.14 to 49.52 % with SDS. The enhancement of thermal conductivity of the nanofluid without surfactant is more than surfactant. Addition of surfactant slightly lowers the thermal conductivity enhancement of the nanofluids. However, the thermal conductivity of the nanofluid with PVP is higher than SDS. This is due to the higher thermal properties of PVP compared to SDS. PVP has good thermal stability [28]. Prolonged heating of SDS at 40 °C or greater causes decomposition of alkyl sulfates into fatty alcohols and sodium sulfate. This indicates less thermal stability of SDS.

5 Conclusions

Silver–water nanofluids have been prepared by dispersing PVP-coated silver nanoparticles in distilled water with and without the addition of surfactants. PVP and SDS were used as surfactants. The stability of the nanofluids was estimated by sedimentation techniques, pH measurement, zeta potential and particle size distribution. The effects of size of the nanoparticles and the addition of surfactants on the stability of nanofluids were studied. The characteristics and concentration of surfactant influence the stability of the nanofluids. The stability of nanofluids with SDS is found to be better than PVP. But the addition of surfactant influences the thermal conductivity enhancement of the nanofluids. However, the enhancement of thermal conductivity of the nanofluids with PVP is higher than SDS. Addition of SDS in water produces more foam compared to PVP which finds a lim-

Table 2 Zeta potential and average particle size of silver–water nanofluids

S. no.	Size of Ag NP (nm)	Concentration of Ag NP (%)	Surfactant added	Concentration of surfactant (% by mass of NP added)	Zeta potential (mV)	Average particle size (nm)
1	20	0.1	–	–	−22.7	108.4
2	30–50	0.1	–	–	−19.9	140.9
3	50–80	0.1	–	–	−14.9	195.7
4	20	0.1	PVP	10	−29.4	98.15
5	20	0.1	SDS	10	−32.8	91.35
6	20	0.1	PVP	20	−30.0	97.28
7	20	0.1	SDS	20	−34.0	80.75

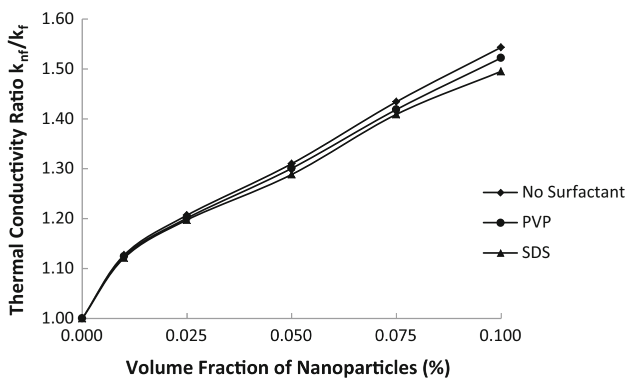


Fig. 9 Thermal conductivity of silver–water nanofluids

itation for its application in heat exchangers. The present study reveals that PVP will be a viable surfactant for the silver–water nanofluids, especially for applications in heat exchangers.

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