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Magnetorheological and Volumetric Properties of Starch and Polyethylene Glycol Solutions in the Presence of NiO Nanoparticles

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The effect of NiO nanoparticles on the rheological and volumetric properties of the dilute solutions of starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 was investigated. To this end, the stable nanofluids should have bee prepared, therefor, the nanoparticles of NiO were added to these solutions and dispersed by a shaker and an ultrasonic bath for making the homogeneous nanofluids. The UV-Vis spectroscopy, zeta potential and dynamic light scattering were used to specify the stability and particle size distribution of colloidal solutions studied. Fluid flow and suspense structure of NiO nanoparticles in the solutions of starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 were studied by measuring the magnetorheological properties at T = 298.15 K. Pseudoplastic (or shear-thinning) behavior was observed for all the suspensions investigated. Bingham plastic, Herschel-Bulkley and Carreau-Yasuda models were applied for modeling the magnetorheological properties of nanofluids. The density values of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 nanofluids were also measured at T = (298.15, 308.15 and 318.15) K. The excess molar volumes were calculated from the density data for highlighting the interparticle interactions occurred in the nanofluids. Singh *et al.* equation was used for fitting the excess molar volume values. The trend of excess molar volumes of nanofluids with concentration indicated that the significant interactions observed in NiO-starch-NaOH-H_2O nanofluid are attractive interactions and in NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 colloidal solutions are van der Waals-type interactions.

Keywords: Nanofluids, NiO nanoparticles, Starch solution, Polyethylene glycol, Magnetorheological property

INTRODUCTION

The dilute colloidal fluids especially magnetic nanofluids consisting of the heat transfer liquids and nanoparticles have a single thermal behavior and great potential for development in the medical applications, mechanical engineering, electronic packing aerospace, *etc.* [1-4]. Nickel oxide, NiO, one of the magnetic nanoparticles, has a variety of applications particularly in magnetically recyclable protein separation system [5,6]. The polymeric carbohydrate starch or amylum consisting of a large number of glucose units can be used in different areas of industry especially in fabrication of some nanoparticles in aqueous solution [7,8]. The low toxicity and water-soluble qualities of some polymers such as poly(ethylene glycol), PEG, make them suitable for industrial applications. PEG and its derivatives are applied as surfactants, excipients in some drugs, lubricants, heat transfer fluid in electronic apparatus, gene therapy vectors, *etc.* [9]. Rheological properties are used to describe the flow behavior of fluids and also deformation of structures made in fluids. Rheological measurements also give the useful information for designing the colloidal solutions in the food industry, paint industry and heat transfer applications [10]. Considering the role of rheological properties in the application of aqueous solutions and suspensions of starch in industry, rheological properties have been the subject of many recent studies [11-14]. In spite of the large applications of NiO nanoparticles, the research activities about colloidal solutions containing NiO nanoparticles are

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scarce. Thermal conductivity enhancement of ethylene-glycol-based nanofluids of NiO nanoparticles was investigated by Gangwar *et al.* [15]. The stability of commercial NiO nanoparticles in water was investigated by Zhang *et al.* [16]. In the recent years, a particular attention has been paid on the polymer based nanofluids. Effect of PEG on rheology and stability of nanocrystalline titania hydrosols was studied by Alphonse *et al.* [17]. Zhang *et al.* were investigated the thermal conductivity of PEG nanofluids containing carbon coated metal nanoparticles [18]. In our previous works, we studied the volumetric, transport and rheological properties and also stability of nanofluids of ZnO-PEG-H_2O [19], ZnO-PEG [20], and Fe_3O_4 nanoparticles coated with oleic acid-PEG [21] and ZnO-PEG-poly(vinyl pyrrolidone) [22].

This work is an attempt in the direction of our previous studies on the stability, magnetorheological and volumetric properties of polymer based nanofluids [22]. In the present work, we use the starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 solutions as the base fluids. Rheological properties and density values for these solutions are measured at different temperatures. Nanoparticles of NiO are added to these solutions and dispersed by an ultrasonic bath for making the homogeneous NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 nanofluids; however, we use the shaker for making the homogeneous NiO-starch-NaOH-H_2O nanofluid. Stability and particle size distribution of the investigated colloidal solutions are studied through the analysis of the ultraviolet-visible (UV-Vis) spectroscopy, and zeta potential and dynamic light scattering. Rheological behaviors of NiO nanoparticles dispersed in starch-NaOH-H_2O are investigated in volume fractions of NiO (φ_1) = 1.17% and 3.36%, and shear rates (γ = 0.01-1000 s^{-1}) at different magnetic fields and 298.15 K. Magnetorheological properties of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 nanofluids are also studied at φ_1 = 1.35% and 4.12% and T = 298.15 K. The magnetorheological properties of nanofluids are correlated by the Bingham plastic [23], Herschel-Bulkley [23] and Carreau-Yasuda [24] models. Interparticle interactions occurred in the investigated nanofluids can be determined by calculating the excess molar volume values. This requires to measure the density data for NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 nanofluids at T = (298.15, 308.15 and 318.15) K. The excess molar volumes are calculated from these data and fitted with Singh *et al.* equation [25].

EXPERIMENTAL SECTION

Materials

In this work, we purchased the bulk NiO from Merck. NiO nanoparticles with a diameter less than 50 nm, minimum mass fraction purity 0.998, product No. [637130] and CAS No. [1313-99-1] with density of 6670 (kg m^{-3}) were purchased from Sigma-Aldrich. NiO nanoparticles were preserved under vacuum for two hours to remove water from the surface of particles then kept them in desiccators under argon atmosphere. Starch (from wheat) for biochemistry, sodium hydroxide, NaOH, with a minimum mass fraction purity 0.99, PEG with molar masses of 400, 2000 and 6000 (g mol^{-1}) were purchased from Merck and applied without any purification. Double distilled, deionized water was used.

Preparation of Nanofluids

For preparation of NiO-starch-NaOH-H_2O nanofluid, the required amount of wheat starch was added to boiling double distilled, deionized water and stirred on magnetic stirrer (ALFA-HS860. IRAN) to make the solution. Then, this solution was cold, and the required amount of NaOH was solved in it, afterward, NiO nanoparticles were dispersed in solution by using the shaker (LABTRON Co. for test tube type LS-100, IRAN) with speed of 3000 rpm for one hour and shaker (IKA-VIBRAX type VXR basic with VX8 universal attachment, Germany) with speed of 400 rpm four 15 h. For preparation of NiO-PEG400-PEG2000 nanofluid, PEGs with molar masses of 400 (g mol^{-1}) and 2000 (g mol^{-1}) with ratio of 275:1 were mixed at 328.15 K to make the homogeneous solution; then this solution was cold and NiO nanoparticles were dispersed in solution by applying the ultrasonic bath (Ultrasonic bath, Grant, Grant instruments (Cambridge) Ltd., England) for seven hours. The NiO-PEG400-PEG6000 nanofluid was also prepared as the same as NiO-PEG400-PEG2000 colloidal solution.

Apparatus

We used the analytical balance (A&D HR-200, A&D company, limited, Japan) with an uncertainty of 0.1 mg for preparing the nanofluids of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000. The UV-Vis spectra were measured with spectrophotometer (Shimadzu UV-1700-Pharma). The particle size distribution of NiO nanoparticles dispersed in PEG, and zeta potential values of nanofluids were measured by dynamic light scattering (DLS, Malvern, Nano ZS, ZEN 3600, England). Anton Paar-Physica rheometer (MCR 300 rheometer using the two plate technique, PP 20/MR) was applied for measuring the magnetorheological properties of the nanofluids. Rheological properties were measured 2-5 s after the shear rate reached to the steady-state condition. Temperature was controlled with a precision of 0.01 K. The parallel disks was separated by a standard gap of 1 mm. A 0.3 ml of the nanofluid was placed between the two parallel disks where the magnetic circuit had been designed around these disks. A sensor can measure the torque and calculate the corresponding force exerted on the moving top plate when the magnetic field changes. The shear stress at a designated point on the plate is then evaluated. A shaft encoder measures the angular rate and the corresponding shear rate. The viscosity value of 85 (mPa s) was measured for PEG with molar mass of 400 (g mol^{-1}) at 298.15 K in zero shear rate using Anton Paar-Physica rheometer; the values of 84.71 and 85.05 have been reported respectively in the literature [26] and obtained by us in our previous work with capillary viscometer [19]. This indicates the good accuracy of rheometer in measuring the viscosity values. Density data were measured using a single-arm capillary pycnometer having a bulb volume of about 5 cm^3 and a capillary bore with an internal diameter of 1 mm at T = (298.15, 308.15 and 318.15) K in which the temperature was controlled with a precision of 0.1 K by a temperature controller (Julabo, MD-18V, Germany). The uncertainty for density measurements was found to be 0.0001 g cm^{-3}.

RESULTS AND DISCUSSION

Experimental Results

The bulk of NiO and NiO nanoparticles were dispersed in starch-NaOH-H_2O to prepare the homogeneous colloidal solutions with NiO weight fraction (w_1) of 0.00003. UV-Vis spectra were recorded for these solutions with passing the time to study the stability of colloidal solutions. The suspensions were kept in the quartz cell without stirring throughout the recording of spectra. The recorded UV-Vis absorption spectra are presented in Fig. 1a. The XRD spectrum of nanoparticles taken by the supplier is also shown in Fig. 1a. The TEM image of NiO nanoparticles in the form of powder taken by the supplier is given in Fig. 1b. Stability of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 nanofluids was also investigated by recording the UV-Vis spectra with passing the time; the weight fraction of NiO nanoparticles (w_1) in these fluids is 0.00005. The UV-Vis absorption spectra are recorded in Figs. 1b and 1c. In these figures, the spectra for colloidal solutions of bulk-NiO-PEG400-PEG2000 and bulk-NiO-PEG400-PEG6000 with w_1 = 0.00005 are also recorded. Analysis of the spectra for NiO nanoparticles in comparison with bulk-NiO recorded in Figs. 1a-1c clarifies that no blue shift and red shift for maximum wavelength of NiO are observed. As far as we know, the optical properties of nanostructures depend on their shapes, sizes and the surrounding environment [27, 28]. In this work, the preparation conditions for colloidal solutions containing NiO nanoparticles are similar to those containing bulk-NiO, therefore, in these colloidal solutions, the UV-Vis maximum wavelength locations are similar to each other. Figures 1a-1c also reveal that the stability for nanofluids of NiO-starch-NaOH-H_2O is very good at least for 30 days, however, the stability for NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 systems is not good as NiO-starch-NaOH-H_2O nanofluid. The zeta potential values were also measured to confirm the stability of the studied nanfluids. The obtained zeta potential values for NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 are respectively 57.4, -6.62 and -5.1 mV. We know that a high positive or high negative value of zeta potential (>30 mV or <-30 mV) proves the stable suspensions [29]. Therefore, in our work, the stability of NiO-starch-NaOH-H_2O nanofluid is better than that of the other nanofluids investigated.

The particle size distributions for nanofluids of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 have also been measured. The determined mean particle sizes are respectively 376.0, 490.6

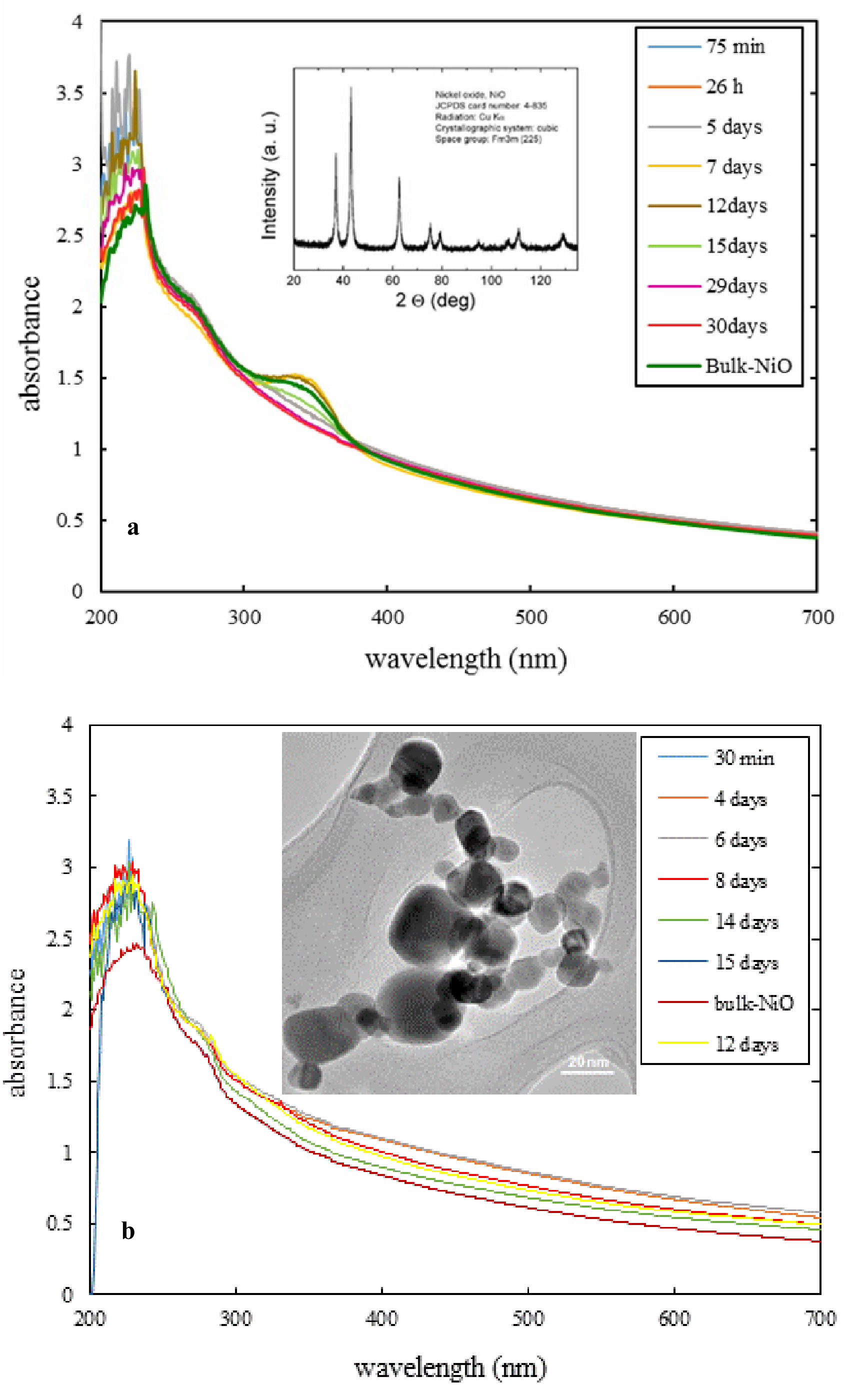


Fig. 1. UV-Vis absorption spectra for nanofluids. 1a: NiO nanoparticles in starch-NaOH-H_2O at NiO weight fraction of 0.00003 and bulk-NiO in starch-NaOH-H_2O at NiO weight fraction of 0.00003 and XRD spectrum of NiO nanoparticles taken by producer. 1b: NiO nanoparticles and bulk-NiO in PEG400-PEG2000 at NiO weight fraction of 0.00005 and TEM image of NiO nanoparticles taken by the supplier. 1c: NiO nanoparticles and bulk-NiO in PEG400-PEG6000 at NiO weight fraction of 0.00005.

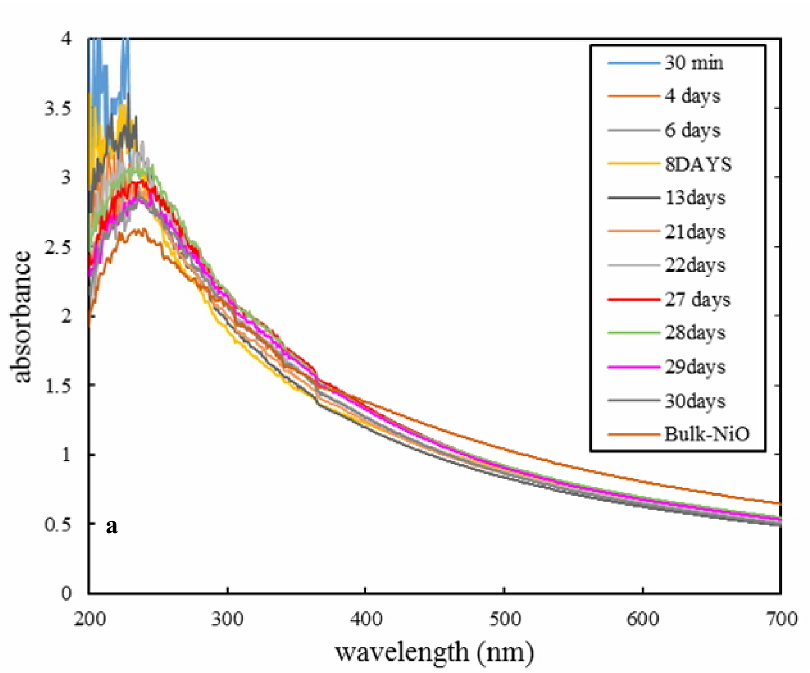


Fig. 1. Continued.

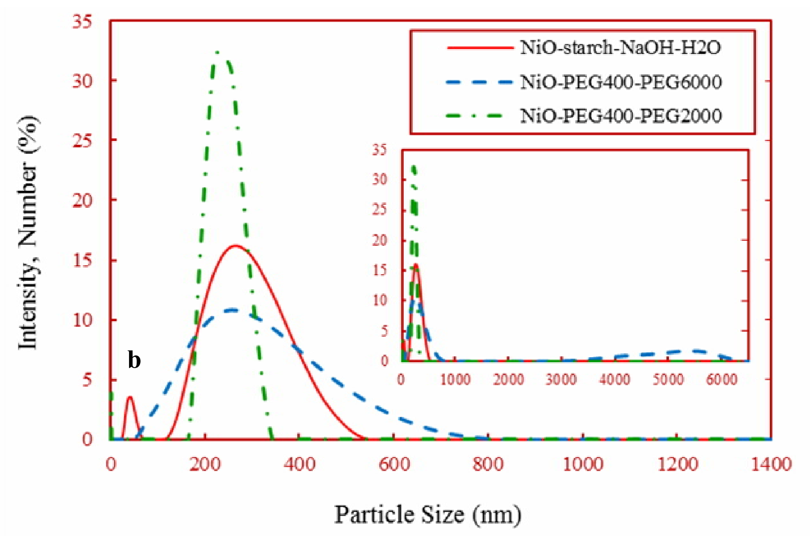


Fig. 2. Particle size distribution for the nanofluids studied.

and 220.9 nm for nanofluids of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000. The particle size distributions for these nanofluids are shown in Fig. 2. The observed large particle sizes for the studied nanoparticles are probably due to the polymer chains over nanoparticles and also aggregation forms of nanoparticles.

The rheological properties of the nanofluids using an external magnetic field have been measured to reveal the

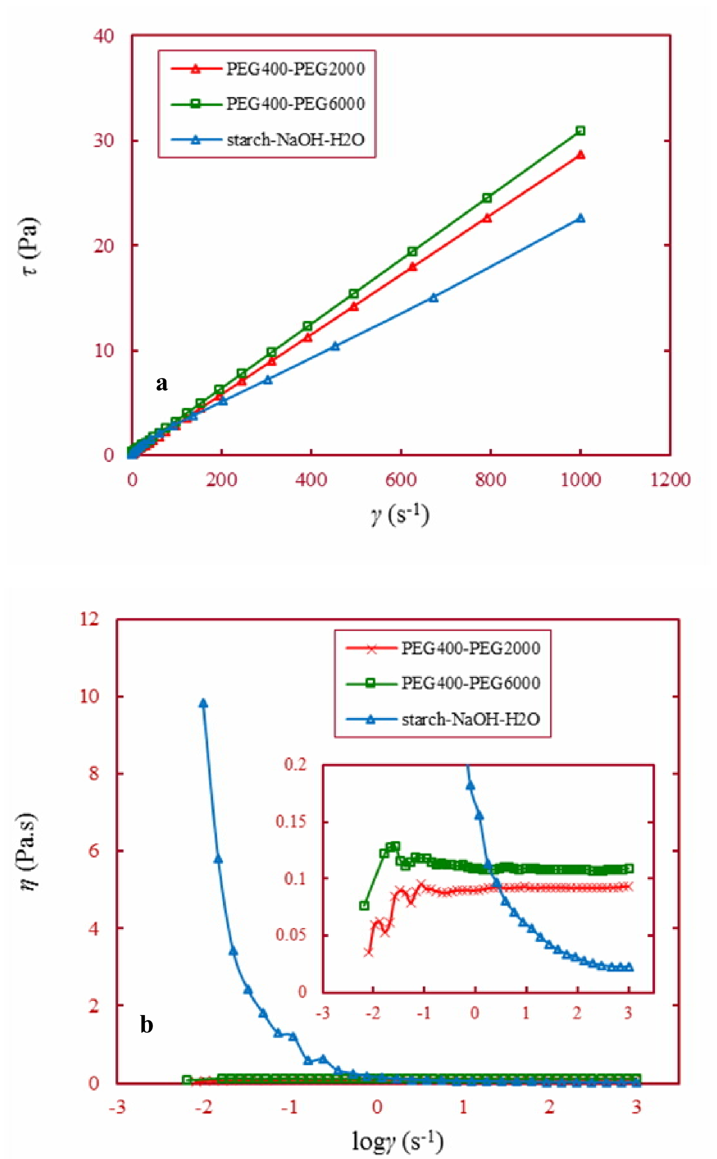


Fig. 3. Rheological properties for the solution of starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 at T = 298.15 K: 3a: Shear stress *vs*. shear rate 3b: Shear viscosity versus shear rate.

effects of NiO nanoparticles on flow behavior of the starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 solutions. There are two types of deviation from Newton's law that are observed in real systems. The most common deviation is shear thinning behavior, where the viscosity of the system decreases as the shear rate is increased. The second deviation is shear thickening behavior where, as the shear rate is increased, the viscosity of the system also increases. This behavior is observed because the system crystallizes under stress and behaves more like a solid than a solution. The presence of suspended particles often affects the viscosity of a solution. In fact, with the right particles, even a Newtonian fluid can exhibit non-Newtonian behavior [30]. Therefore, we prepared the nanofluid of NiO-starch-NaOH-H_2O at φ_1 = 1.17% and 3.36%, and the nanofluids of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 at φ_1 =

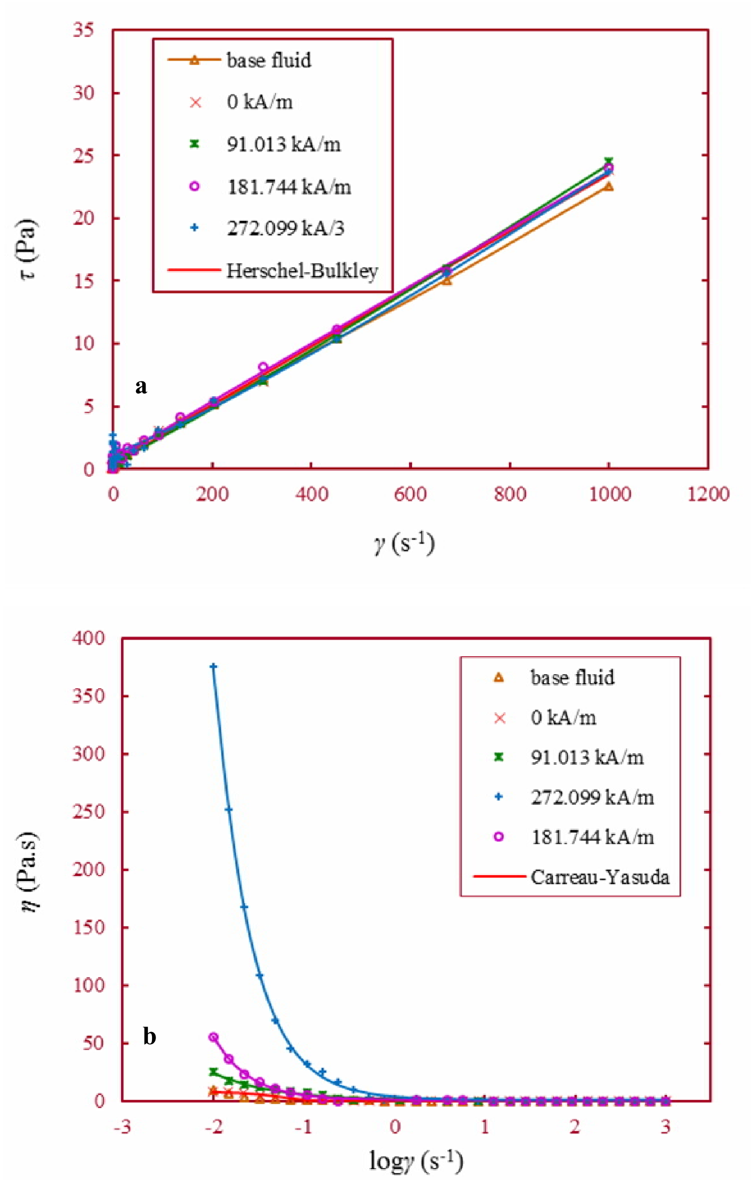
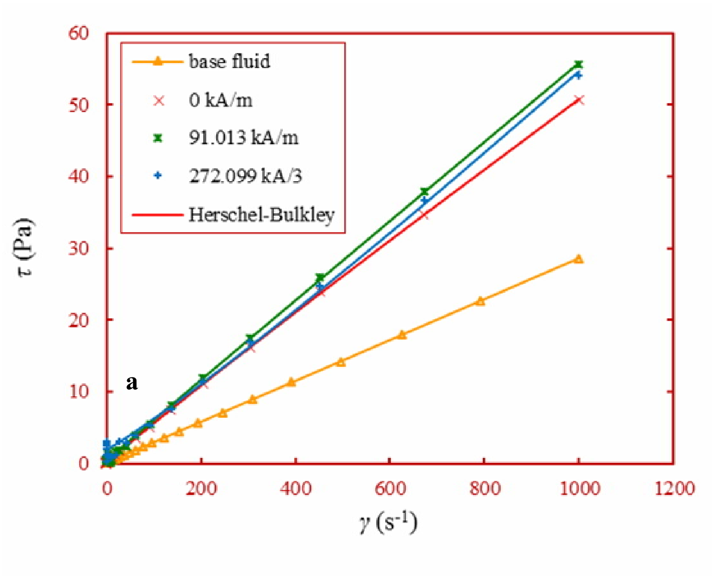


Fig. 4. Rheological properties for nanofluid of NiO-starch-NaOH-H_2O at φ_1 = 3.36%, different magnetic fields and 298.15 K, 4a: Shear stress *vs*. shear rate. 4b: Shear viscosity *vs*. shear rate.

1.35% and 4.12%. The variations of shear stress and viscosity with shear rate increasing at different magnetic fields were measured for the investigated colloidal solutions at 298.15 K. The obtained results are shown in Fig. 3 for the starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 solutions; and in Figs. 4-6 for nanofluids of NiO-starch-NaOH-H_2O at φ_1 = 3.36%, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 at φ_1 = 4.12%. The rheological properties for other investigated nanofluids (NiO-starch-NaOH-H_2O at φ_1 = 1.17%, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 at φ_1 = 1.35%) are given in Figs. S1-S3 as supplementary material. Figures 3-6 and also S1-S3 indicate that the starch-NaOH-H_2O solution shows pseudoplastic (or shear-thinning) behavior which is



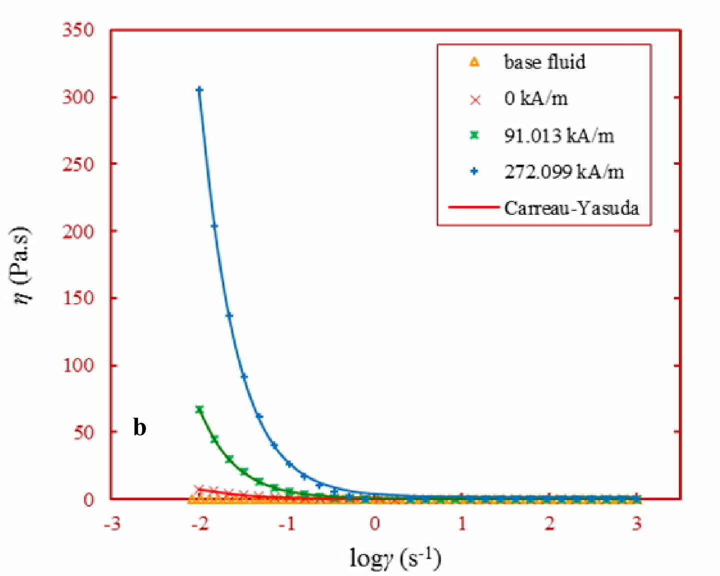


Fig. 5. Rheological properties for the nanofluid of NiO-PEG400-PEG2000 at φ_1 = 4.12%, different magnetic fields and 298.15 K, 5a: Shear stress *vs*. shear rate. 5b: Shear viscosity *vs*. shear rate.

compatible with that observed by Che *et al.* for dilute aqueous solutions of cassava starch [14]; this is due to the reduction in the intermolecular resistance to flow with increasing in shear rate. PEG400-PEG2000 and PEG400-PEG6000 solutions exhibit the shear thickening behavior with initial rise on shear rate; this can be due to the formation of some structures between PEG400 and PEG2000 or PEG6000 at very low values of shear rate. All the suspensions investigated (NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000) show the shear-thinning behavior. This revealed that the resistance to flow was reduced by using the small forces, and the aggregations of NiO nanoparticles or network flocs formed between starch, NaOH, H_2O, PEG and NiO was broken into the smaller flow units with shear rate increasing. In addition, large values for viscosity and shear stress have

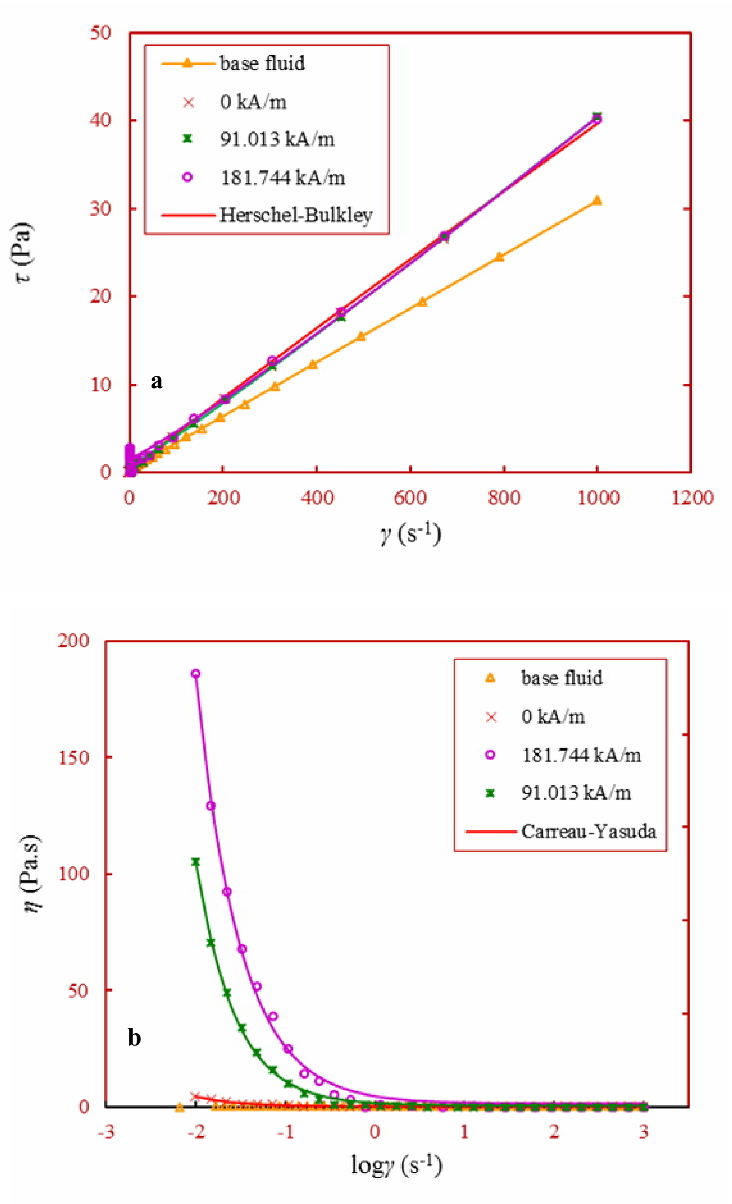


Fig. 6. Rheological properties for the nanofluid of NiO-PEG400-PEG6000 at $\varphi_1 = 4.12\%$, different magnetic fields and 298.15 K, 6a: Shear stress *vs*. shear rate 6b: Shear viscosity versus shear rate.

been observed at higher magnetic fields which can be due to form of chain like structure of nickel oxide. The results obtained from this work are also consistent with those we observed in our previous work for Fe_3O_4 nanoparticles coated with oleic acid-PEG nanofluid [21].

The experimental density (*d*) values for nanofluids of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 and solution of starch-NaOH-H_2O have also been measured at T = (298.15, 308.15 and 318.15) K. The obtained results are collected in Tables 1 and 2. The excess molar volume, V_m^E values, have been calculated by Eq. (1) for characterizing the nonideal behavior of the investigated colloidal solutions of NiO-starch-NaOH-H_2O

Table 1. Density (d) and Excess Molar Volume (V^E_m), for Nanofluid of NiO-Starch-NaOH-H_2O and Solution of Starch-NaOH-H_2O at Different Temperatures[a]

100 w_1[b]	100 φ_1[c]	100 w_2[d]	w_3[f]	d (kg m^{-3})	10^6 V^E_m (m^3 mol^{-1})	100 w_2[d]	100 w_3[f]	d (kg m^{-3})	10^6 V^E_m (m^3 mol^{-1})
		NiO-Starch-NaOH-H_2O					Starch-NaOH-H_2O		
				T = 298.15 K					
0	0	0.6	0.62	1374.3					
0.004	0.001	0.6	0.62	1369.8	-1.132	0.62	0.62	1369.1	-1.127
0.01	0.002	0.6	0.62	1373.0	-1.159	0.62	0.62	1369.5	-1.130
0.03	0.01	0.6	0.62	1379.8	-1.214	0.62	0.62	1370.5	-1.138
0.04	0.01	0.6	0.62	1385.2	-1.257	0.62	0.62	1371.0	-1.141
0.06	0.01	0.6	0.62	1390.1	-1.296	0.62	0.62	1372.8	-1.156
0.13	0.03	0.6	0.62	1391.8	-1.300	0.62	0.62	1374.7	-1.169
0.38	0.08	0.6	0.62	1396.2	-1.303	0.62	0.62	1376.0	-1.169
0.63	0.13	0.6	0.61	1401.6	-1.313	0.62	0.62	1377.9	-1.174
0.88	0.18	0.6	0.61	1405.8	-1.313	0.62	0.62	1379.0	-1.172
1.13	0.24	0.6	0.61	1407.2	-1.290	0.61	0.61	1379.9	-1.169
				T = 308.15 K					
0	0	0.6	0.62	1368.0					
0.004	0.001	0.6	0.62	1368.9	-1.155	0.62	0.62	1367.2	-1.141
0.01	0.002	0.6	0.62	1372.1	-1.181	0.62	0.62	1368.4	-1.151
0.03	0.01	0.6	0.62	1379.3	-1.240	0.62	0.62	1369.5	-1.159
0.04	0.01	0.6	0.62	1383.0	-1.269	0.62	0.62	1370.5	-1.167
0.06	0.01	0.6	0.62	1389.8	-1.323	0.62	0.62	1372.2	-1.181
0.13	0.03	0.6	0.62	1391.4	-1.327	0.62	0.62	1372.7	-1.182
0.38	0.08	0.6	0.62	1395.1	-1.323	0.62	0.62	1373.8	-1.180
0.63	0.13	0.6	0.61	1400.9	-1.337	0.62	0.62	1375.6	-1.184
0.88	0.18	0.6	0.61	1404.3	-1.330	0.62	0.62	1377.3	-1.188
1.13	0.24	0.6	0.61	1405.3	-1.304	0.61	0.61	1378.2	-1.184

Table 1. Continued

				$T = 318.15$ K					
0	0	0.6	0.62	1364.5					
0.004	0.001	0.6	0.62	1364.7	-1.157	0.62	0.62	1367.0	-1.177
0.01	0.002	0.6	0.62	1365.0	-1.159	0.62	0.62	1367.3	-1.180
0.03	0.01	0.6	0.62	1372.6	-1.222	0.62	0.62	1367.9	-1.184
0.04	0.01	0.6	0.62	1379.3	-1.277	0.62	0.62	1368.5	-1.188
0.06	0.01	0.6	0.62	1387.1	-1.339	0.62	0.62	1369.9	-1.200
0.13	0.03	0.6	0.62	1388.9	-1.345	0.62	0.62	1370.4	-1.201
0.38	0.08	0.6	0.62	1392.7	-1.342	0.62	0.62	1371.9	-1.202
0.63	0.13	0.6	0.61	1397.6	-1.348	0.62	0.62	1373.8	-1.207
0.88	0.18	0.6	0.61	1401.7	-1.347	0.62	0.62	1375.0	-1.206
1.13	0.24	0.6	0.61	1403.0	-1.323	0.61	0.61	1376.4	-1.207

[a]Uncertainties for concentration, temperature and density are 0.0001, 0.01 K and 0.0001 g cm^{-3}, respectively. [b]w_1 is weight fraction of NiO nanoparticles. [c]φ_1 is volume fraction of NiO nanoparticles. [d]w_2 is weight fraction of starch. [f]w_3 is weight fraction of NaOH.

and starch-NaOH-H_2O

$$V_m^E = \sum_{i=1}^{4} \left[\frac{p}{d} - \frac{p_i}{d_i}\right] \quad (1)$$

in this equation, p and p_i are the masses of the solution and component i in kg, respectively; d and d_i are the density of solution and density of component i, respectively. Subscripts 1, 2, 3 and 4 stand for NiO nanoparticle, starch, NaOH and H_2O, respectively. We used the value of 6670 (kg m^{-3}) for density of NiO nanoparticle provided by producer. The solutions studied are dilute and away from pure the nanoparticle, thereby, the calculated excess molar volumes are not very sensitive to the density data of nanoparticle at different temperatures, therefore, we can use the value of 6670 (kg m^{-3}) at three investigated temperatures. The calculated V_m^E values for the nanofluid of NiO-starch-NaOH-H_2O and solution of starch-NaOH-H_2O have been tabulated in Table 1 and also shown in Fig. 7. As can been seen from these table and figure, V_m^E values for the aforementioned solution are negative and decrease with

Table 2. Density (d) and Excess Molar Volume (V^E_m), for the Nanofluid of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 at Different Temperatures[a]

100 w_1[b]	100 φ_1[c]	w_2[d]	d (kg m^{-3})	10^6 V^E_m (m^3 mol^{-1})	100 w_1[b]	100 φ_1[c]	w_2[d]	d (kg m^{-3})	10^6 V^E_m (m^3 mol^{-1})
NiO-PEG400-PEG2000					NiO-PEG400-PEG6000				
T = 298.15 K									
0	0	0.9997	1116.2		0	0	0.9997	1116.0	
0.01	0.001	0.9996	1116.8	1.866	0.01	0.001	0.9996	1116.2	2.058
0.02	0.003	0.9995	1117.2	1.761	0.02	0.003	0.9995	1116.4	2.017
0.04	0.01	0.9992	1117.3	1.801	0.04	0.01	0.9992	1116.5	2.057
0.06	0.01	0.9990	1117.5	1.794	0.06	0.01	0.9990	1117.0	1.954
0.09	0.02	0.9987	1117.7	1.817	0.09	0.02	0.9987	1117.2	1.976
0.20	0.03	0.9976	1118.6	1.846	0.20	0.03	0.9976	1118.6	1.846
0.60	0.1	0.9936	1121.7	2.007	0.60	0.1	0.9936	1122.5	1.759
1.01	0.17	0.9986	1125.2	2.046	1.00	0.17	0.9896	1126.3	1.714
1.40	0.24	0.9856	1128.9	2.032	1.40	0.24	0.9856	1130.0	1.707
1.80	0.31	0.9816	1133.0	1.909	1.80	0.31	0.9816	1133.7	1.707
T = 308.15 K									
0	0	0.9997	1107.7		0	0	0.9997	1108.0	
0.01	0.001	0.9996	1109.2	1.699	0.01	0.001	0.9996	1109.7	1.537
0.02	0.003	0.9995	1109.6	1.592	0.02	0.003	0.9995	1110.5	1.300
0.04	0.01	0.9992	1109.8	1.601	0.04	0.01	0.9992	1111.3	1.115
0.06	0.01	0.9990	1110.2	1.529	0.06	0.01	0.9990	1111.7	1.044
0.09	0.02	0.9987	1110.3	1.584	0.09	0.02	0.9987	1112.0	1.036
0.20	0.03	0.9976	1110.9	1.710	0.20	0.03	0.9976	1113.5	0.876
0.60	0.10	0.9936	1113.9	1.901	0.60	0.10	0.9936	1117.5	0.774
1.01	0.17	0.9986	1117.3	1.968	1.00	0.17	0.9896	1121.3	0.744
1.40	0.24	0.9856	1121.0	1.949	1.40	0.24	0.9856	1125.0	0.753
1.80	0.31	0.9816	1125.1	1.821	1.80	0.31	0.9816	1128.7	0.769

Table 2. Continued

				T = 318.15 K					
0	0	0.9997	1099.2		0	0	0.9997	1101.4	
0.01	0.001	0.9996	1101.2	1.668	0.01	0.001	0.9996	1101.9	1.437
0.02	0.002	0.9995	1101.8	1.494	0.02	0.002	0.9995	1102.9	1.132
0.04	0.01	0.9992	1101.9	1.534	0.04	0.01	0.9992	1103.1	1.140
0.06	0.01	0.9990	1102.2	1.495	0.06	0.01	0.9990	1103.6	1.035
0.09	0.01	0.9987	1102.5	1.484	0.09	0.01	0.9987	1104.5	0.830
0.20	0.03	0.9976	1103.3	1.546	0.20	0.03	0.9976	1106.0	0.668
0.60	0.1	0.9936	1106.2	1.769	0.60	0.1	0.9936	1110.5	0.404
1.01	0.17	0.9986	1109.6	1.834	1.00	0.17	0.9896	1114.5	0.315
1.40	0.23	0.9856	1113.3	1.812	1.40	0.23	0.9856	1118.2	0.327
1.80	0.3	0.9816	1117.3	1.708	1.80	0.3	0.9816	1121.8	0.376

[a]Uncertainties for mole fraction, temperature and density are 0.0001, 0.01 K and 0.0001 g cm^{-3}, respectively. [b]w_1 is weight fraction of NiO nanoparticles. [c]φ_1 is volume fraction of NiO nanoparticles. [d]w_2 is weight fraction of PEG400.

temperature increasing. The negative excess molar volumes indicate that a more efficient packing and/or attractive interaction has been occurred when the starch, NaOH and H_2O were mixed. Decreasing the interactions with increasing the temperature is the main reason for the decrease in excess molar volume with temperature enhancement [31,32]. The excess molar volume values for nanofluid of NiO-starch-NaOH-H_2O are more negative than those of the starch-NaOH-H_2O solution, indicating that the attractive interactions also exist between the NiO nanoparticles and starch-NaOH-H_2O solution which are more than interactions occurred among starch, NaOH and H_2O molecules.

The excess molar volume, V_m^E values, have been calculated by Eq. (2) for nanofluids of PEG400-PEG2000 and NiO-PEG400-PEG6000.

$$V_m^E = \sum_{i=1}^{3} x_i M_i [\frac{1}{d} - \frac{1}{d_i}] \quad (2)$$

In this equation, x is the mole fraction, M is the molar mass, and subscripts 1, 2 and 3 stand for NiO nanoparticle, PEG400 and PEG2000 or PEG6000, respectively. The calculated V_m^E values for the nanofluids of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 have been tabulated in Table 2 and also shown in Fig. 7. As can been seen from this table and Fig. 7, V_m^E values for the nanofluids are positive and decrease with temperature increasing. The van der Waals-type interactions which are categorized as dispersion forces can be deduced from the positive values of excess molar volume [31,32]. Therefore, we can conclude that van der Waals-type interactions are dominant in the

Table 3. Parameters of Bingham Plastic Model, Eq. (3), and Herschel-Bulkley Model, Eq. (4), along with Viscosity at High Shear Rate, η_∞, for the Nanofluids Containing NiO Nanoparticles at Different Magnetic Field

φ_1%	τ_y (Pa)	η	R^2	η_∞ (Pa.s)	τ_y (Pa)	K_{HB}	n	R^2
	Bingham plastic model				Herschel-Bulkley model			
				NiO-Starch-NaOH-H_2O at T = 0 kA m^{-1}				
1.17	0.6588	0.0302	0.98847	0.6588	0.24442	0.10265	0.81688	0.99078
3.36	0.30829	0.02337	0.99827	0.30829	0.27126	0.02777	0.97422	0.99984
				NiO-Starch-NaOH-H_2O at T = 91.013 kA m^{-1}				
1.17	0.38904	0.02966	0.99761	0.38904	0.25636	0.04307	0.94438	0.99926
3.36	0.42012	0.0235	0.99771	0.42012	0.51423	0.01549	1.06226	0.99917
				NiO-Starch-NaOH-H_2O at T = 181.744 kA m^{-1}				
1.17	0.59126	0.02959	0.99571	0.59126	0.58132	0.03048	0.9956	0.9999
3.36	0.7109	0.02311	0.99594	0.7109	0.68779	0.02525	0.9868	0.99996
				NiO-Starch-NaOH-H_2O at T = 272.099 kA m^{-1}				
1.17	1.09332	0.028	0.99817	1.09332	1.23174	0.01787	1.06696	0.99906
3.36	1.10427	0.02182	0.97579	1.10427	1.27056	0.00871	1.13732	0.99624
				NiO-Starch-NaOH-H_2O at T = 361.988 kA m^{-1}				
1.17	1.59521	0.02666	0.98034	1.59521	1.75917	0.01455	1.09035	0.99831
				NiO-PEG400-PEG2000 at T = 0 kA m^{-1}				
1.35	0.14246	0.03557	0.99979	0.14246	0.08723	0.04212	0.04212	0.47396
4.12	0.23362	0.0511	0.99963	0.23362	0.11726	0.06525	0.96342	0.99968
				NiO-PEG400-PEG2000at T = 91.013 kA m^{-1}				
4.12	0.44694	0.05553	0.99933	0.44694	0.4183	0.05857	0.99203	0.99999

Table 3. Continued

				NiO-PEG400-PEG2000 at T = 181.744 kA m^{-1}				
1.35	0.40236	0.03582	0.99672	0.40236	0.48651	0.02823	1.03561	0.99972
				NiO-PEG400-PEG2000 at T = 272.099 kA m^{-1}				
4.12	1.67396	0.0517	0.99283	1.67396	1.95904	0.02828	1.09017	0.9983
				NiO-PEG400-PEG2000at T = 361.988 kA m^{-1}				
1.35	0.9564	0.03608	0.99503	0.9564	1.03306	0.02915	1.03185	0.99977
				NiO-PEG400-PEG6000at T = 0 kA m^{-1}				
1.35	0.16257	0.0455	0.99979	0.16257	0.17213	0.05429	0.97316	0.99983
4.12	0.10493	0.03998	0.99977	0.10493	0.18606	0.04849	0.97036	0.99979
				NiO-PEG400-PEG6000 at T = 91.013 kA m^{-1}				
1.35	0.55362	0.04513	0.99903	0.55362	0.59255	0.04092	1.01468	0.99995
4.12	0.57374	0.03918	0.99775	0.57374	0.71283	0.02587	1.06214	0.99917
				NiO-PEG400-PEG6000 at T = 181.744 kA m^{-1}				
1.35	0.92218	0.04477	0.99728	0.92218	1.06166	0.032	1.05021	0.99945
4.12	1.33353	0.03808	0.99044	1.33353	1.54321	0.02035	1.09372	0.99817

nanofluids of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 which decrease with temperature increasing.

Modeling the Experimental Results

Various empirical models such as Bingham plastic and Herschel-Bulkley [23] are often used to determine the shear rate dependency of shear stress; therefore, in our work these models are applied to calculate the yield stress (critical level of stress) as follow:

Bingham plastic model: $\tau = \tau_y + \eta\gamma$ (3)

Herschel-Bulkley model: $\tau = \tau_y + K_{HB}(\gamma)^n$ (4)

where τ_y is the yield-stress parameter, η is the suspension viscosity, and K_{HB} and n are the structure-dependent adjustable parameters. The determined shear stress data were correlated to Eqs. (3) and (4) and the obtained results were collected in Table 3. The solid lines in Figs. 4a-6a and S1a-S3a illustrate the performance of Herschel-Bulkley model in fitting the shear stress values. Table 3 shows that the yield stress or critical level of stress obtained by

Table 4. Parameters of Carreau-Yasuda Model (Eq. (5)) along with Standard Deviation, σ, [a]Obtained from Fitting the Viscosity Values of Nanofluids Containing NiO Nanoparticles

φ_1%	λ	a	n	σ (Pa s)
		NiO-Starch-NaOH-H_2O at T = 0 kA m^{-1}		
1.17	0.00031	0.6851	-758.4	0.482
3.36	0.02742	1.636	-55690	0.427
		NiO-Starch-NaOH-H_2O at T = 91.013 kA m^{-1}		
1.17	103.6	26.82	-2.238	0.901
3.36	137.8	38.64	0.3425	0.873
		NiO-Starch-NaOH-H_2O at T = 181.744 kA m^{-1}		
1.17	97.62	20.45	-0.1997	0.545
3.36	105.9	20.21	-0.09956	0.813
		NiO-Starch-NaOH-H_2O at T = 272.099 kA m^{-1}		
1.17	89.19	8.476	-0.1561	1.385
3.36	103.2	15.92	-0.0714	1.822
		NiO-Starch-NaOH-H_2O at T = 361.988 kA m^{-1}		
1.17	92.55	15.34	-0.1664	4.295
		NiO-PEG400-PEG2000 at T = 0 kA m^{-1}		
1.35	6.113	81.080	-4.117	0.153
4.12	93.75	6.747	0.09063	0.148
		NiO-PEG400-PEG2000at T = 91.013 kA m^{-1}		
4.12	102	26.84	-0.06486	0.446
		NiO-PEG400-PEG2000 at T = 181.744 kA m^{-1}		
1.35	94.85	41.07	-0.1547	0.356
		NiO-PEG400-PEG2000 at T = 272.099 kA m^{-1}		
4.12	99.23	36.39	-0.05862	1.959
		NiO-PEG400-PEG2000at T = 361.988 kA m^{-1}		
1.35	99.92	38.45	-0.02034	0.828

Table 4. Continued

NiO-PEG400-PEG6000at T = 0 kA m^{-1}				
1.35	42.83	1.547	-0.2731	0.146
4.12	111.8	48.08	0.0663	0.112
NiO-PEG400-PEG6000 at T = 91.013 kA m^{-1}				
1.35	104	19.79	-0.04757	0.582
4.12	103.3	25.57	-0.001453	0.972
NiO-PEG400-PEG6000 at T = 181.744 kA m^{-1}				
1.35	100	23.23	-0.0636	0.974
4.12	102.6	61.26	0.1232	2.504

[a] $\sigma = \sqrt{\frac{\sum_{i=1}^{N}(\eta_i^{\exp} - \eta_i^{cal})^2}{N}}$ in which, N is the total number of data.

Herschel-Bulkley model is enhanced with the rise of concentration and magnetic fields. The yield stresses evaluated by Bingham plastic model in some cases are higher than those obtained by Herschel-Bulkley model. This is because of the linear dependency of shear stress on shear rate in Bingham plastic model while in the nanofluids studied deviation from linear dependence is observed.

The Carreau-Yasuda model [24], as the following equation, is aaplied for correlating the visosity values of the investigated nanofluids at each concentration and magnetic field:

$$\eta = \eta_\infty + (\eta_0 - \eta_\infty)[1 + (\lambda\gamma)^a]^{(\frac{n-1}{a})} \tag{5}$$

where η_0 and η_∞ are respectively the viscosity of colloidal solutions at very low and high shear rates. λ, a and n are the parameters of this model. The obtained results are given in Table 4 and illustrated in Figs. 4b-6b and S1b-S3b. As can be seen from these figures and table, the performance of the Carreau-Yasuda model in fitting the viscosity values of the considered nanofluids is good especially at low values of shear rate.

The excess molar volumes for nanofluids of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 and solution of starch-NaOH-H_2O were fitted with Singh *et al.* [25] equation. The Singh *et al.* [25] model is as follows:

$$V^E_{m123} = V^E_{m12} + V^E_{m13} + V^E_{m23} + F_1F_2F_3[A_{123} + B_{123}F_1(F_2 - F_3) + C_{123}F_1^2(F_2 - F_3)^2] \tag{6}$$

In this equation, F is used as the weight fraction in fitting the V^E_m values for nanofluid of NiO-starch-NaOH-H_2O and solution of starch-NaOH-H_2O, while, F is applied as mole fraction in correlating the V^E_m values for nanofluids of NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000. V^E_{m12}, V^E_{m13}, V^E_{m23}, A_{123}, B_{123} and C_{123} are the fitting coefficients of Singh *et al.* equation. The evaluated parameters of Singh *et al.* equation along with the standard deviations are given in Table 5. The fitting quality of these model is also shown in

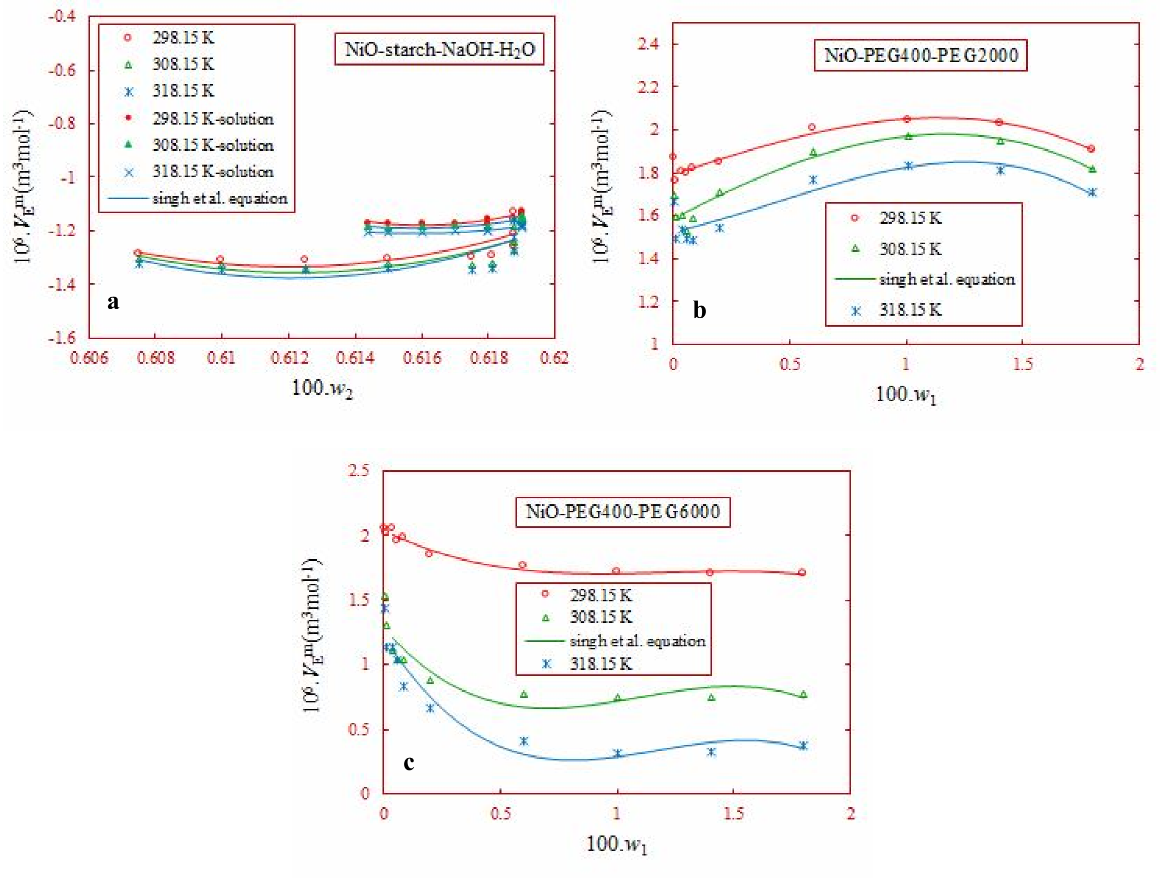


Fig. 7. Experimental and calculated excess molar volume, V_m^E, plotted against concentration (weight fraction of starch (w_2), weight fraction of NiO nanoparticles (w_1)) for nanofluids of NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000 at different temperatures.

Fig. 7. From this figure and Table 5 one can conclude that the performance of the aforementioned model is good in fitting the excess molar volumes of the investigated nanofluids.

CONCLUSIONS

The solutions of starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 were prepared. Rheological properties and density values for these solutions were measured at different temperatures. Nanoparticles of NiO were added to these solutions and dispersed by a shaker and an ultrasonic bath to make homogeneous nanofluids. The UV-Vis spectroscopy, zeta potential and dynamic light scattering were used to specify the stability and particle size distribution of the colloidal solutions studied. Result analysis of these methods revealed that the investigated nanofluid of NiO-starch-NaOH-H_2O is reversibly more stable than other two nanofluids for a long time. Rheological behaviors of NiO nanoparticles dispersed in starch-NaOH-H_2O, PEG400-PEG2000 and PEG400-PEG6000 were investigated in two volumetric solid the attractive interactions exist between NiO nanoparticles and starch-NaOH-H_2O solution which are more than interactions occurring among starch, NaOH and H_2O molecules. In addition, the significant interactions observed in NiO-

Table 5. Parameters of Singh *et al.* Equation (Eq. (6)) along with the Standard Deviations (σ) for the Nanofluids Containing NiO Nanoparticles and Solution of Starch-NaOH-H_2O at Different Temperatures

$10^3\ V^E_{m12}$	$10^4\ V^E_{m13}$	V^E_{m23}	$10^{-5}\ A_{123}$	$10^{-7}\ B_{123}$	$10^{-8}\ C_{123}$	σ
			NiO-Starch-NaOH-H_2O			
			T = 298.15 K			
15.39	-284.6	-1.175	-0.2339	-0.6734	-5.434	σ ($10^6\ V^E_m$) = 0.037
						σ (d/kg m^{-3}) = 4.382
			T = 308.15 K			
7.094	-284.5	-1.189	-0.2381	-0.6873	-5.505	σ ($10^6\ V^E_m$) = 0.037
						σ (d/kg m^{-3}) = 4.417
			T = 318.15 K			
15.39	-284.7	-1.189	-0.2902	-0.8542	-6.986	σ ($10^6\ V^E_m$) = 0.042
						σ (d/kg m^{-3}) = 5.055
			Starch-NaOH-H_2O			
			T = 298.15 K			
6.256	-83.42	-2.319	0.4063	5.419	184.3	σ ($10^6\ V^E_m$) = 0.008
						σ (d/kg.m^{-3}) =0.98
			T = 308.15 K			
6.290	-83.42	2.305	0.2601	3.886	132.7	σ ($10^6\ V^E_m$) = 0.01
						σ (d/kg.m^{-3}) = 1.209
			T = 318.15 K			
6.242	-83.42	-3.769	0.2203	2.754	93.66	σ ($10^6\ V^E_m$) = 0.006
						σ (d/kg m^{-3}) = 0.687
			NiO-PEG400-PEG2000			
			T = 298.15 K			
1796	-0.5735	-0.0000574	0.6973	0.1313	-0.2349	σ ($10^6\ V^E_m$) = 0.027
						σ (d/kg m^{-3}) = 0.085
			T = 308.15 K			
1589	-0.5735	-0.0000574	1.190	0.1253	-0.269	σ ($10^6\ V^E_m$) = 0.048
						σ (d/kg m^{-3}) =0.149
			T = 318.15 K			
1534	-0.1434	-0.0000144	0.1028	0.3747	-0.430	σ ($10^6\ V^E_m$) = 0.054
						σ (d/kg m^{-3}) = 0.166

Table 5. Continued

			NiO-PEG400-PEG6000			
			$T = 298.15$ K			
2541	-2455	-0.2457	-8.075	1.465	-0.8806	$\sigma\,(10^6\,V^E_m) = 0.026$
						$\sigma\,(d/\text{kg m}^{-3}) = 0.081$
			$T = 308.15$ K			
1817	-2523	-0.2525	-20.120	4.579	-3.082	$\sigma\,(10^6\,V^E_m) = 0.105$
						$\sigma\,(d/\text{kg m}^{-3}) = 0.327$
			$T = 318.15$ K			
1757	-2535	-0.2537	-26.16	5.212	-3.23	$\sigma\,(10^6\,V^E_m) = 0.096$
						$\sigma\,(d/\text{kg m}^{-3}) = 0.296$

concentrations at different magnetic fields and shear rates (γ = 0.01-1000 s^{-1}) and T = 298.15 K. Shear thinning behaviors of starch-NaOH-H_2O solution and shear thickening behaviors of solutions of PEG400-PEG2000 and PEG400-PEG6000 were changed to a pseudoplastic (or shear-thinning) behavior for all the suspensions investigated (NiO-starch-NaOH-H_2O, NiO-PEG400-PEG2000 and NiO-PEG400-PEG6000). The trend of excess molar volumes of nanofluids with concentration and temperature indicates that PEG400-PEG2000 and NiO-PEG400-PEG6000 colloidal solutions are van der Waals-type interactions. Bingham plastic, Herschel-Bulkley and Carreau-Yasuda models have successfully been applied for modeling the magnetorheological properties of nanofluids. The excess molar volume values were adequately fitted to the Singh *et al*. equation.

SUPPLEMENTARY MATERIAL AVAILABLE

The Figs. S1-S3 are available free of charge *via* the Internet at http://

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