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Comparative Study of Rheological Properties of Polyvinyl Alcohol and Polyethylene Glycol

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Abstract. Hydrogels are promising biomaterials for diverse applications that require studying their rheological properties. While some properties of hydrogels have been investigated, their comparative analysis for a deeper understanding of their rheological properties is required to determine their mechanical behavior. Polyvinyl alcohol (PVA) and polyethylene glycol (PEG) are among the hydrogels with diverse applications in engineering. This study aims to provide comparative data on their rheological properties. Both PVA and PEG showed steady shear viscosity as their viscosity did not show a huge change with time. Their shear viscosity increased with shear strain. PEG showed more shear thickening behavior than PVA. While the shear viscosity of PVA reached a plateau, that of PEG continued to increase. This was attributed to the sensitivity of PEG to its deformation because of the junction separations after the application of mechanical force on the polymer. Furthermore, the slow increase in the shear viscosity of both polymers was observed with the increase of the shear rate. This increase was 2.4 % for PVA and 8.7 % PEG, respectively. As these polymers are among the candidates for the preparation of nanocomposites, the results of this study can provide the required information for their applications in engineering.

Keywords: rheology, hydrogels, polyvinyl alcohol, polyethylene glycol, mechanical engineering.

1 Introduction

Polyvinyl alcohol (PVA) and polyethylene glycol (PEG) are among the polymers that absorb water during their preparation. These hydrogels are transparent materials that can be prepared by dissolving in water [1–4] and contain a lot of water in their total mass due to their hydrophilic functional groups attached to their backbone [5–8]. The biocompatibility of PVA and PEG has made them excellent candidates for medical applications for drug delivery systems and tissue engineering [9–14].

Three types of hydrogels are used in engineering based on their chemical composition: natural, synthetic and hybrid hydrogels [10]. This classification is made based on the cross-linking in the hydrogels [15, 16]. The type and degree of crosslinking determine swelling, elastic modulus, and transport of molecules in hydrogels [17]. Further classification of hydrogels depends on their ionic charge, structure, and preparation methods [16, 17]. Their network structure should be controlled to get an appropriate design and characterization, which impact the degradation of hydrogel scaffolds, diffusion of bioactive

molecules, and migration of cells through the network [16, 17].

Freezing without chemical crosslinking is an appropriate method for the preparation of PVA hydrogels. Ring-opening polymerization of ethylene oxide cyclic monomers, radiation crosslinking of PEG, or free radical polymerization of PEG macromers are common methods for preparing PEG-based hydrogels [18–20].

Diverse methods have been applied for the functionalization of hydrogels. Three-component reaction, polyelectrolyte multilayer microencapsulation, and microwave-assisted functionalization have been applied to functionalize PEG and PVA hydrogels [21–23].

Chain polymerization of hydrogels is used for the preparation of heterogeneous network structures containing dense crosslinking regions [9]. For performing the step-growth polymerization, a crosslinker or comonomer is used to react with the terminal functional groups of the hydrogel macromers to produce greater network structure homogeneity [9].

The applications of PVA and PEG are numerous in engineering. These hydrogels are appropriate candidates for coating nanomaterials in order to reduce their natural reactivity and maintain their physical properties [24]. The coating has two advantages: it can shield the nanomaterial from the surrounding environment and help functionalize the nanomaterial by attaching chemical groups to its surface [25].

Superparamagnetic iron oxide nanoparticles and graphene oxide nanoribbons are among the nanomaterials studied, previously [26–31]. These nanomaterials can be coated with hydrogels. Previous studies have shown that the physicochemical properties of these nanomaterials could be modified because of their coating [27, 30, 32]. The obtained nanocomposites have various applications in engineering. Therefore, their coating with PEG and PVA was performed in previous studies.

In this study, we compare the rheological properties of PVA and PEG. We investigate herein the variation of rheological parameters of these hydrogels. The consideration of the hydrophilic networks of these polymers allow for a better understanding of their rheological properties. To our knowledge, no comparative study has been carried out on the rheological properties of these hydrogels.

2 Research Methodology

2.1 Chemicals

PVA (Mw 89,000-98,000) and PEG (Mw 8000) were purchased from Sigma Aldrich and ThermoFisher Scientific, respectively.

2.2 Preparation of hydrogels

The hydrogel solutions were prepared as described previously [27, 30]. Briefly, a total of 0.4 g of PVA was dissolved in deionized water (10 mL) and mixed during heating at 90 °C for 2 h and at 140 °C for 4 h [27].

A total of 4.8 g of PEG was dissolved in deionized water (20 mL) and mixed during 15 minutes at room temperature [30].

2.3 Rheological measurements

The rheological properties of the solutions of PVA and PEG in millipore water were studied using an Anton Paar MCR-302 rheometer.

5 mL of each sample were used for rheological assessments. The circular rheological measurements were performed in triplicate at room temperature [27].

2.4 Statistical analysis

The QtiPlot software was used for the data analysis. The mean values and standard deviations were obtained for the calculations with this software [7].

The statistical significance of data was checked after the calculations [28]. As the obtained data were close to each other, their standard deviation values were small.

3 Results

Figure 1 shows the viscosity of PVA and PEG versus shear rate. The shear rate was measured between 0 and 1 000 s⁻¹.

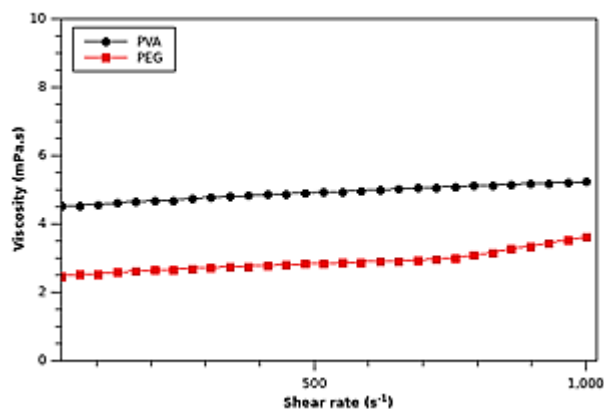


Figure 1 – Viscosity of PVA and PEG versus shear rate

As shown in Figure 1, the viscosity of PVA and PEG increased slowly with the increase of shear rate. Moreover, the increase of viscosity for PEG was more than that of PVA. The first polymer had an increase of 8.7 % in viscosity, whereas the second one had an increase of 2.4 % at shear rate of 1 000 s⁻¹.

Figure 2 shows the change of viscosity of both polymers versus shear strain.

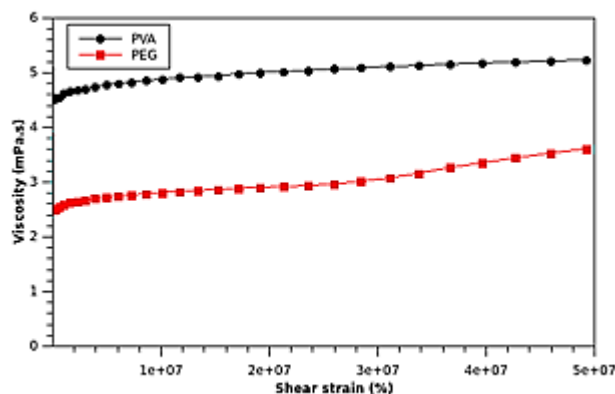


Figure 2 – Viscosity of PVA and PEG versus shear strain

As seen in this figure, the viscosity of PVA and PEG increased with shear strain. The shear-thickening behavior of PEG was more than that of PVA. The viscosity of both polymers reached a plateau. However, PEG reached to the plateau at lower shear strains than PVA. Moreover, PEG showed a second increase of viscosity at high shear strains, whereas PVA did not significantly increase after the plateau. This was attributed to the sensitivity of PEG to its deformation because of the junction separations after the application of mechanical force on the polymer.

The viscosity of PVA and PEG versus time curves are displayed in Figure 3.

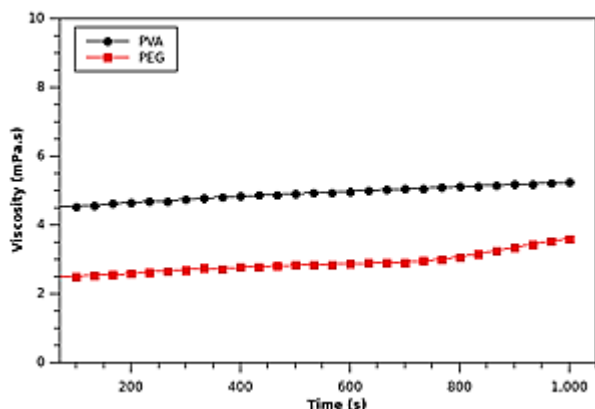


Figure 3 – Viscosity of PVA and PEG versus time

As shown in this figure, the viscosity of both polymers increased with time. Although the viscosity increase of PVA with time was small and reached a plateau, that of PEG was significant and continued at 1 000 s. This shows that when shear stress is applied on these polymers, the effect of the viscosity increase with time for PEG is more than that of PVA. This may be attributed to the difference of the position of the hydrophilic OH group in the structures of these polymers. The presence of this functional group bound to the chain of PEG makes a hindrance for the application of high shear stress on the polymer, whereas this effect is much less in PVA because its OH group is inside its chain, which makes less increase of the viscosity of this second polymer when more shear stress is applied with the increase of time.

The viscosity values of PVA and PEG versus time are presented in Table 1.

Table 1 – Viscosity of PVA and PEG (mPa·s) versus time

Polymer	Time, s		
	200	600	1 000
PVA	4.66 ± 0.03	4.98 ± 0.01	5.24 ± 0.02
PEG	2.62 ± 0.04	2.89 ± 0.02	3.61 ± 0.01

Comparing the results presented in the above table, the increase values of 0.32 mPa·s and 0.26 mPa·s in viscosity were observed for PVA between 600 seconds and 200 s, and between 1 000 s and 600 s, respectively. Whereas in the second time span more viscosity, which was 0.72 mPa·s was observed for PEG compared to the increase of viscosity in the first time span, which was 0.27 mPa·s. This result showed that the viscosity of PEG at higher shear stress in the second time span increased more than its viscosity at lower shear stress in the first time span, whereas PVA showed the inverse effect in these time spans. These results were consistent with the presented figures.

Figure 4 shows torque versus shear strain.

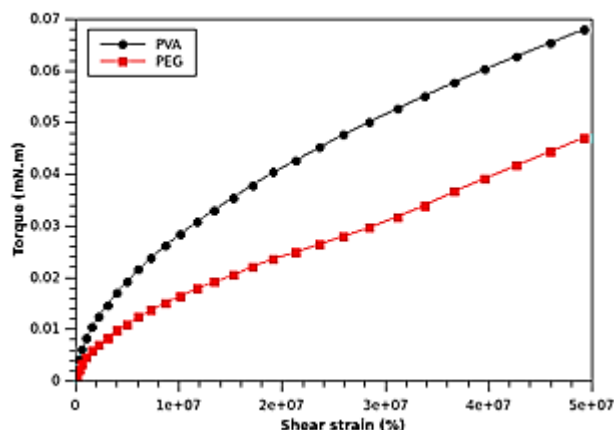


Figure 4 – Torque versus shear strain

The torque values increased with time for both polymers. This was due to their high elasticity that made the torque increase with time. It is worth noting that at the same shear strain, the torque value was more for PVA than PEG. This was consistent with the fact that the initial viscosity of PVA as shown in Figure 2 was more than that of PEG.

The changes in the torque values for PVA and PEG versus time are displayed in Figure 5.

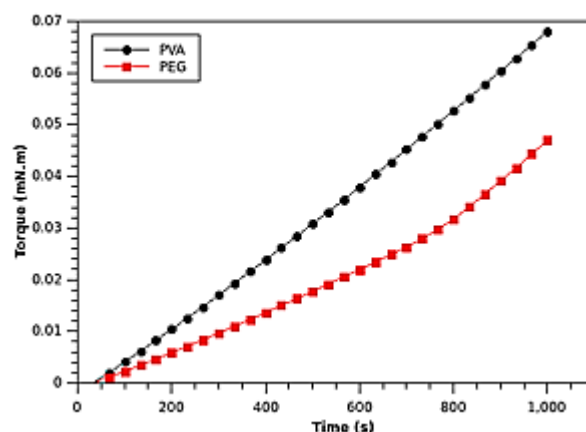


Figure 5 – Torque versus time

This figure shows that the torque values increased with time for both polymers. However, the torque increase was more rapid for PVA than for PEG. This indicates that the torque changes versus time for PVA and PEG were not the same as the first polymer showed a steeper line compared to the second one.

Comparing the torque changes for PVA and PEG versus time showed that this change for the first polymer was linear, whereas it was a curve for the second one. In other words, the torque values when applied for PEG did not change linearly with time. This could correspond to the difference in the structures of these polymers.

Figure 6 shows the shear stress versus shear rate curves of PVA and PEG.

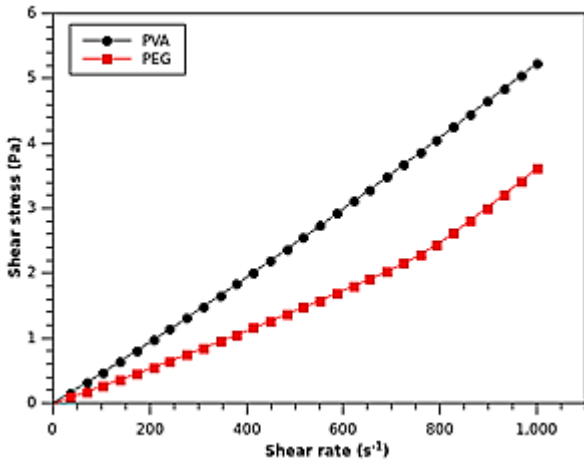


Figure 6 – Shear stress of PVA and PEG versus shear rate

As observed in this figure, the same shear stress corresponds to a greater shear rate for PEG than PVA. This is because the flow velocity gradient in PEG is more than that of PVA when the same shear stress is applied to each polymer solution.

Figure 7 shows the shear stress versus shear strain curves of both polymers.

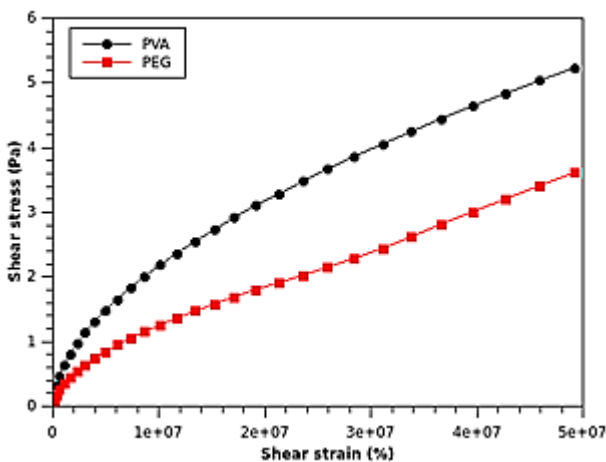


Figure 7 – Shear stress of PVA and PEG versus shear strain

As shown in this figure, less shear stress is required for the same quantity of the deformation of PEG in comparison with that of PVA. In other words, the deformation of PEG is easier than that of PVA as more shear stress is required for the deformation of the second polymer in comparison with the first one. This is attributed to the differences in these polymers' chemical structures and molecular weights.

The shear stress values of PVA and PEG versus shear strain are presented in Table 2.

The data in the above table show that the shear strain increase of polymers from $1 \cdot 10^6$ to $2 \cdot 10^7$ would require more shear stress on PVA than that on PEG.

Table 2 – Shear stress (Pa) of PVA and PEG versus shear strain

Polymer	Shear strain, %		
	$1 \cdot 10^6$	$2 \cdot 10^7$	$4 \cdot 10^7$
PVA	0.64 ± 0.01	3.29 ± 0.01	4.84 ± 0.02
PEG	0.36 ± 0.01	1.91 ± 0.01	3.21 ± 0.01

The same phenomenon was observed for the shear strain increase from $2 \cdot 10^7$ to $4 \cdot 10^7$. This is attributed to the differences in these hydrogels' molecular weights and structures that can determine the shear effect [33]. These comparative data can be used to improve the mechanical properties of these materials for the preparation of blends and composites.

4 Discussion

The results presented in this paper provide important new insights to the comparative rheological properties of PVA and PEG. Previous investigations on the rheological properties of these polymers showed that up to certain shear rates, these polymers would behave as Newtonian liquids. These studies indicated that at high shear rates, viscosity instant flow cessation occurred in these polymers [34–37]. However, no study has reported the comparative analysis of the rheological properties of these polymers.

Shear modulus is a parameter that reflects the ability of polymer to withstand the force applied and resist to the deformation. This parameter is obtained from the division of shear stress by shear strain [38, 39]. In the current paper, the shear modulus values of both PVA and PEG were very small and they decreased with time for both polymers. We expect to observe that this parameter would decrease with the increase of temperature as the deformation of polymers could happen by applying less amount of shear stress with the increase of temperature. In a further investigation, we will study the effect of temperature on the rheological properties of these polymers.

The molecular weight and chain branching are the structural factors that can impact the rheological behavior of polymers. Low molecular weight polymers display Newtonian behavior due to a tumbling or sliding of the molecules over one another during applying a shear stress [40]. Moreover, the ratio of the response time of the polymer to observation or experimental time depends on the molecular weight of the sample and the temperature [40]. The physicochemical properties of some of the materials that were applied in engineering were investigated previously [41–45]. More investigations are required to determine the effect of PVA and PEG on these materials regarding their preparation with polymers.

The composites of PVA and PEG with metals and non-metals are of significant interest in engineering as these hydrogels can improve materials' workability and water retention [46–50]. It has been observed that the viscosity of gel would increase with increasing the concentration of metals when a constant concentration of PEG was used in the samples. An investigation showed that the reduction of the gel viscosity was necessary in

order to control the concentration of metal ions in water-glycol solutions [34]. The rheological properties of dried composite membranes can provide information on the mechanical resistance and tolerance to pressure applied during preparing materials with hydrogels [50]. The structural recovery tests can be conducted by applying different shear rates for getting information on the sample preparation after deposition conditions of mixtures and the change in their viscosity [51]. Some compositions can influence pH, particularly those obtained in a weakly alkaline environment with higher values of relative viscosity [52]. Crosslinking in the composites can improve light shielding, visible light transparency, better mechanical and water vapor barrier properties, and thermal stability [53]. Rheological and electrical assessments of PVA-based composites can give information on the formation of ordered liquid-crystal structures in the polymer matrix [54]. When dimethyl sulfoxide with water is used to dissolve PEG, the system could become more heterogeneous because of the hydrogel structure produced by weakly bonded water molecules. The network structure impacts the rheological properties as the reorganization of hydrogen bonding could contribute to the transitions of the macroscopic viscoelasticity [55].

The rheological properties of PVA and PEG are of interest when they are mixed with other materials in composites. The non-Newtonian behavior of PVA is maintained when it is mixed with graphene oxide nanoribbons [27]. It has been shown that the rheological properties of nanocomposites of PVA and cellulose nanocrystals had limited molecular weight dependence [56]. Lactose-filled PEG composites were also studied concerning their processing temperature. It was reported that decreasing the process temperature could lead to an increase in shear viscosity. Moreover, the temperature dependence of shear viscosity decreased with the increase of shear rate [57]. Another study revealed that introducing silica nanoparticles into PEG-based composites that are used as membranes for separating gases could affect their rheological properties [58, 59]. The study of the viscoelastic behavior of PEG-poly(lactic acid) composites showed that these materials could have a higher magnitude of storage modulus than pure polymers [60]. Another study showed the increase of viscosity due to loading additives in the biphasic polymer solution, which did not change in further growth of the filler concentration [61]. The addition of clay to PEG could change its Newtonian flow behavior to thixotropic shear thinning when its structure broke down by mechanical shearing, and its viscosity decreased with time under conditions of constant shear rate [62].

Blends also are of interest in engineering. A study showed that the viscosity of pure PVA solutions would increase slowly as they age, and the aging of the PVA/PEG blends would be much faster than those of pure polymers [50]. Another study revealed that a blend having viscoelastic behavior could remain stable upon storage [51]. It has been shown that long polymers could interconnect several particles, acting as cross-links which

could explain the mechanism of the enhancement of viscosity [52]. The physical crosslinking methods have been widely applied in materials science because there has been no need to introduce new chemicals, complicating their use in the biomedical environment [53]. Another important aspect of the preparation of blends concerns the phase separation. A study revealed that the presence of PEG forces the PVA solution to separate phase. The authors proposed that this phenomenon could help the formation of gel [63]. Crosslinking has a positive impact on the preparation of PVA and PVA-PEG blends as their rheological properties could reveal their viscoelastic behavior and stability upon storage [64, 65]. Adding PEG to PVA can counteract the reduction of the water content in the polymer matrix to make the blend more appropriate for biomedical applications [66].

The other important issues in preparing materials with PVA and PEG hydrogels are discussed below. The presence of gel with abnormal rheological properties in the glycol regeneration systems is a problem for this purpose because it may cause filters to clog at critical shear rates [35]. High molecular weight PEG can increase the elasticity and stability of shear thickening fluids [67–69]. It is worth noting that the interchain hydrogen bonds and shear-induced orientation can affect the rheological properties of PEG solutions in water. The application can break the intrachain and interchain hydrogen bonds of shear stress, and new interchain hydrogen bonding can be created simultaneously. The viscoelasticity of these systems depends on the reorganization of hydrogen bonding [70–73].

As discussed above, temperature and crosslinking can impact the rheological properties of PVA and PEG. More investigations are required to determine the impact of these factors on the comparative analysis of the rheological properties of these hydrogels for the preparation of composites and blends.

5 Conclusions

This study shows that the rheological properties of PVA and PEG are similar to each other at low shear rates. However, at high shear rates, they behave differently. Moreover, the increase in the shear strain values requires more shear stress on PVA than on PEG.

These results are attributed to the differences in these polymers' network structures and molecular weights. Some issues, such as the viscoelasticity of PVA and PEG and the temperature change that affect the drug delivery output when these hydrogels are used for coating drug vectors, must be addressed in further studies for the development of their biomedical applications.

These investigations will allow better control of the physicochemical properties of these hydrogels for these applications.

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