VISCOSITY OF HYDROXYPROPYL CELLULOSE SOLUTIONS IN THE PRESENCE OF POLY(*N*-VINYLPYRROLIDONE)

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Dedicated to the memory of
Acad. Bogdan C. Simionescu (1948 – 2024)

The paper reports viscosity data for aqueous solutions of hydroxypropyl cellulose (HPC) in the presence of poly(*N*-vinylpyrrolidone) (PVP) in dilute, semidilute and concentrated regimes. The two polymers are miscible over the entire range of composition. In dilute solution, mixed isolated coils with more extended chain conformation are formed and binary polymer-polymer interactions are favored when a small amount of PVP is added to HPC solutions. The viscosity increases in semidilute and concentrated regimes due to chain entanglements and a pronounced synergistic effect was observed upon the addition of a small amount of PVP to the HPC isotropic solution. The intermolecular hydrogen bonding between the hydroxyl groups of HPC and the carbonyl groups of PVP influences the overall flow behavior.

Keywords: hydroxypropyl cellulose, poly(*N*-vinylpyrrolidone), aqueous solution, viscosity, intermolecular interactions

INTRODUCTION

Hydroxypropyl cellulose (HPC) is one of the most studied cellulose derivatives, considered a model system for understanding the rheological behavior of liquid crystalline polymers.[1,2] The rheology of anisotropic HPC aqueous solutions was thoroughly investigated[3,4] and the phase separation effect on flow behavior was evidenced in static and in flow conditions.[5–7]

HPC is an inexpensive polymer with high potential for application in pharmaceutical, cosmetic and food domains.[8–10] The hydroxypropyl moieties grafted to the main macromolecular chain of cellulose confer thermosensitivity to the polymer chains in aqueous solution. The lower critical solution temperature (LCST) is located around 41 °C,[3,11] depending on the molar substitution number of HPC. Above LCST, in a narrow temperature range (between 41 °C and 44 °C), the HPC chains self-associate and form dynamically metastable aggregates, which further form gel microspheres by crosslinking these associated structures.[12]

Mixtures of HPC with other water-soluble polymers lead to changes in phase transition occurence, influencing the rheological behavior. These features are important, especially for the design of new materials with various functionalities and improved properties. As for example, by adding 1% poly(acrylic acid) to HPC solution, LCST decreases considerably. The hydrogen bonding between the two polymer chains determines the formation of an interpolymer complex, influenced by environmental pH and molecular weight of the polymers.[13,14]

In the present study, the viscosity of HPC aqueous solutions was investigated at 25 °C in dilute, semidilute and concentrated isotropic solutions. The main interest was to evidence the viscosity changes by the addition of small amounts (up to 20%) of a second water-soluble

polymer, poly(*N*-vinylpyrrolidone) (PVP). PVP is nontoxic and presents excellent biocompatibility and ability to form complexes with both hydrophobic and hydrophilic polymers (including cellulose derivatives) and associated structures with numerous active substances.[15,16] This complexing ability of PVP allows for the tailoring of the dissolution profiles of poor water-soluble polymers, inhibits precipitation, increases the bioavailability of poorly water-soluble drugs and controls their delivery.[10,15]

EXPERIMENTAL

Klucel™ hydroxypropylcellulose (HPC) sample used in this study was supplied by Aqualon (Klucel). The molar substitution number (the average number of moles of hydroxypropyl substituent per mole of anhydroglucose residue) is around 3, according to the manufacturer.[17] Poly(*N*-vinylpyrrolidone) (PVP) sample was purchased from Sigma-Aldrich. Both polymers were dissolved in distilled water at room temperature using a magnetic stirrer.

Stock solutions of HPC and PVP of different concentrations were firstly prepared and then mixtures of different HPC/PVP compositions were achieved by mixing the solutions of the two polymers of identical concentration. w_{HPC} and w_{PVP} represent the weight fractions of HPC and PVP in the polymer mixture, respectively.

For the dilute solutions of HPC, PVP and their mixtures, the viscometric measurements were carried out at 25 °C by using an Ubbelohde viscometer, capillary type 0a with the diameter of 0.53 mm, in combination with an automatic measurement system (Lauda Instruments, Germany). The flow time for water, t_o, was 184.44 s.

The rheological measurements were performed at 25 °C with a MCR 302 Anton Paar rheometer, equipped with a Peltier system for temperature control. The viscosity of semidilute solutions was determined with a concentric cylinder system CC27. For concentrated solutions, a plate-plate geometry was used (diameter of the upper plate = 50 mm; gap = 500 μm). The shear viscosity measurements were carried out in the stationary flow regime, for shear rates in the range 0.01 s^{-1} – 1000 s^{-1}.

RESULTS AND DISCUSSION

Models used to describe the viscometric behavior of HPC-based solutions

Two models were selected to determine the intrinsic viscosity, $[\eta]$, of HPC, PVP and HPC/PVP aqueous solutions: the classical Huggins equation,[18] generally applied for neutral polymers in dilute solution, and the new Wolf approach,[19] which can be applied over a large domain of concentrations.[20]

The Huggins method (Equation (1)) can be applied to experimental data obtained for neutral polymers in dilute solution:[18]

$$\eta_{sp}/c = [\eta] + k_H[\eta]^2 c \quad (1)$$

where η_{sp} is the specific viscosity, $\eta_{sp} = \eta_{rel} - 1$, $\eta_{rel} = t/t_o$ is the relative viscosity, t and t_o represent the flow times of polymer solution and solvent, respectively, k_H is the Huggins constant, c is the solute concentration. In this study, c denotes the total concentration of dissolved macromolecules.

For Newtonian fluids, in the limit of infinite dilution, the viscometric behavior can be described by the model developed recently by Wolf:[19]

$$\ln \eta_{rel} = \frac{\tilde{c} + \alpha \tilde{c}^2}{1 + \beta \tilde{c} + \gamma \tilde{c}^2} \quad (2)$$

where the dimensionless parameter $\tilde{c} = c[\eta]$ is the reduced polymer concentration; α, β and γ are system-specific parameters. α takes into account the friction between the macromolecules and depends on the sample composition. The parameters β and γ reflect the changes of the free volume in polymer solution as compared to the pure solvent. Very often, only two of the three parameters suffice for modeling the viscosity as a function of polymer concentration, and in most cases α can be considered zero.

The new Wolf approach proved to be very versatile and it can be applied to any type of macromolecule (charged or neutral), including polymer mixtures in solution, over a large domain of concentrations, from the infinite dilution up to the melt state.[20,21]

Viscosity of binary aqueous solutions at infinite dilution

Figure 1 presents the concentration dependences of η_{sp}/c and ln η_{rel} for HPC and PVP solutions in water at 25°C. It can be observed that the experimental data for the pure polymer solutions fit well with the Huggins and Wolf models. The obtained values for the intrinsic viscosity and viscometric parameters using Equations (1) and (2) are given in Table 1.

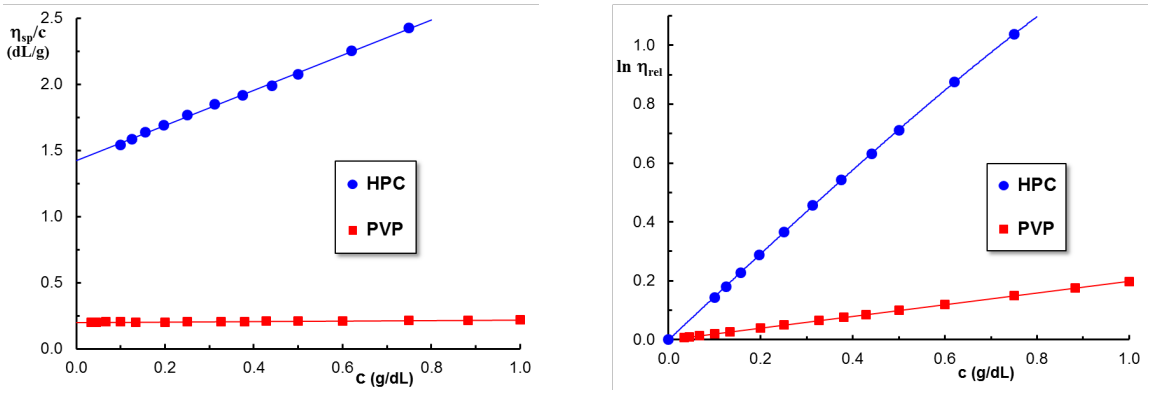


Figure 1: Plots of η_{sp}/c and ln η_{rel} as a function of concentration for HPC and PVP solutions in water at 25 °C

Table 1
Viscometric parameters for HPC, PVP and their mixtures obtained by fitting the experimental data according to Huggins (Eq. (1)) and Wolf (Eq. (2)) models. The parameter α (from Eq. (2)) was set to zero in all cases

w_{HPC}	w_{PVP}	[η] (dL/g) (Eq. (1))	k_H (Eq. (1))	[η] (dL/g) (Eq. (2))	β (Eq. (2))	γ (Eq. (2))	$[\eta]_{add}$ (dL/g) (Eq. (5))	c^* (g/dL) (Eq. (8))
1.00	0	1.3328	0.6378	1.3031	0.0475	0.0386	1.3031	0.5909
0.99	0.01	1.4788	0.5502	1.3754	0.0540	0.0678	1.2921	0.5598
0.97	0.03	1.5697	0.5288	1.4326	-0.1160	0.1780	1.2701	0.5375
0.95	0.05	1.4062	0.5874	1.4282	-0.1144	0.1798	1.2480	0.5391
0.925	0.075	1.4025	0.5598	1.4005	-0.1090	0.1867	1.2205	0.5498
0.90	0.10	1.3991	0.5492	1.3719	-0.1068	0.2075	1.1930	0.5613
0.88	0.12	1.3545	0.5812	1.3391	-0.1125	0.2122	1.1709	0.5750
0.85	0.15	1.266	0.6518	1.2674	-0.1003	0.2099	1.1379	0.6075
0.80	0.20	1.1477	0.7251	1.1542	-0.1075	0.2056	1.0828	0.6671
0.50	0.50	0.8500	1.0470	0.8301	0.0015	0.1567	0.7524	0.9276
0.25	0.75	0.5325	1.8726	0.5325	0.1022	0.0987	0.4770	1.4460
0	1.00	0.2309	0.4379	0.2016	0.1327	0	0.2016	3.8194

The viscosity of HPC with different molecular weights in aqueous solutions was reported by different authors.[2,4,6] For Klucel-type HPC in water at 25 °C, Law and Kayes[23] established the following dependence between the intrinsic viscosity and the molecular weight (M):

$$[\eta] = 6.25\times10^{-5} M^{0.84} \quad \text{(dL/g)} \qquad (3)$$

The molecular weight of HPC was calculated with Equation (3) and intrinsic viscosity values determined experimentally with the Wolf model ([η] = 1.3031 dL/g), resulting M = 1.4×10^5 g/mol.

For fractioned PVP samples with the molecular weights from 2.8×10^4 g/mol to 5.12×10^5 g/mol in aqueous solution at 25 °C, the following Mark-Houwink relationship was reported:[24]

$$[\eta] = 3.9\times10^{-5} M^{0.82} \quad \text{(dL/g)} \qquad (4)$$

By using Equation (4) and the determined value of [η] at 25 °C (0.2016 dL/g), the molecular weight of PVP was calculated as M = 4×10^4 g/mol.

The high exponent values from Equations (3) and (4) suggest that water is a good solvent for both polymers (HPC and PVP) under these thermodynamic conditions. Generally, the macromolecules containing both polar hydrophilic and hydrophobic groups are fully soluble in water at room temperature. Heating of these homogeneous solutions may cause phase separation and the phase diagram presents a lower critical solution temperature (LCST).[25] HPC and PVP exhibit LCSTs in aqueous solutions: LCST is about 41 °C for HPC[11,12,26,27] and around 70 °C for PVP.[28] This is particularly important for generating thermosensitive hydrogels. Water becomes a marginal solvent as the temperature increases and the driving force for the network formation is the enhancement of the hydrophobic interactions.[29]

Viscosity of ternary HPC/PVP aqueous solutions at infinite dilution

Figures 2 and 3 show the viscometric plots obtained according to Huggins and Wolf methods for solutions of different HPC/PVP compositions (w_{PVP} represents the weight fraction of PVP in the HPC/PVP mixture). We carefully examined the solution viscosity of HPC solutions when a small amount of PVP is added. In order to avoid the data overlapping in the region of low PVP content, not all curves were presented in Figures 2 and 3. The parameters obtained from the evaluation of experimental data are listed in Table 1. For all ternary systems, the experimental values of $[\eta]$ are higher than those calculated from the additive rule:

$$[\eta]_{add} = w_{HPC} \cdot [\eta]_{HPC} + w_{PVP} \cdot [\eta]_{PVP} \quad (5)$$

The Huggins constant is higher than 0.5 for all HPC-containing systems, suggesting a tendency of aggregation due to favorable polymer-polymer intermolecular interactions.

Figure 4 illustrates the change in intrinsic viscosity when PVP is gradually added to the HPC solution. A positive deviation from the additive rule was observed over the whole range of composition (Fig. 4a) and it is higher in the region of low PVP content ($w_{PVP} \leq 0.15$, Fig. 4b).

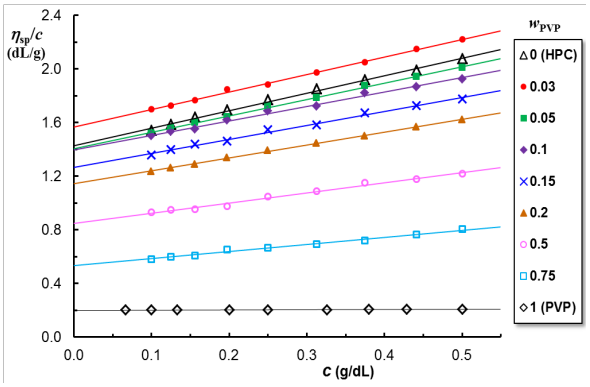


Figure 2: Huggins plots (according to Eq. (1)) for HPC/PVP solutions of different polymer compositions (water, 25 °C)

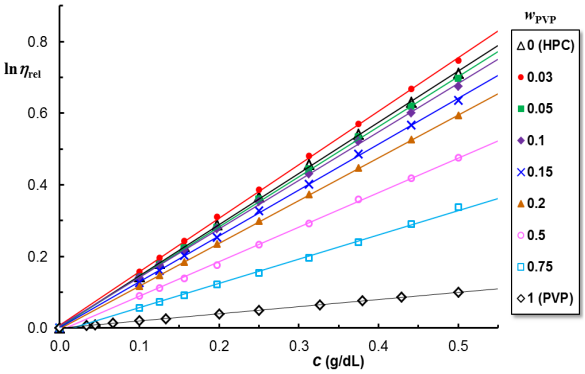


Figure 3: Wolf plots (according to Eq. (2)) for HPC/PVP solutions of different polymer compositions (water, 25 °C)

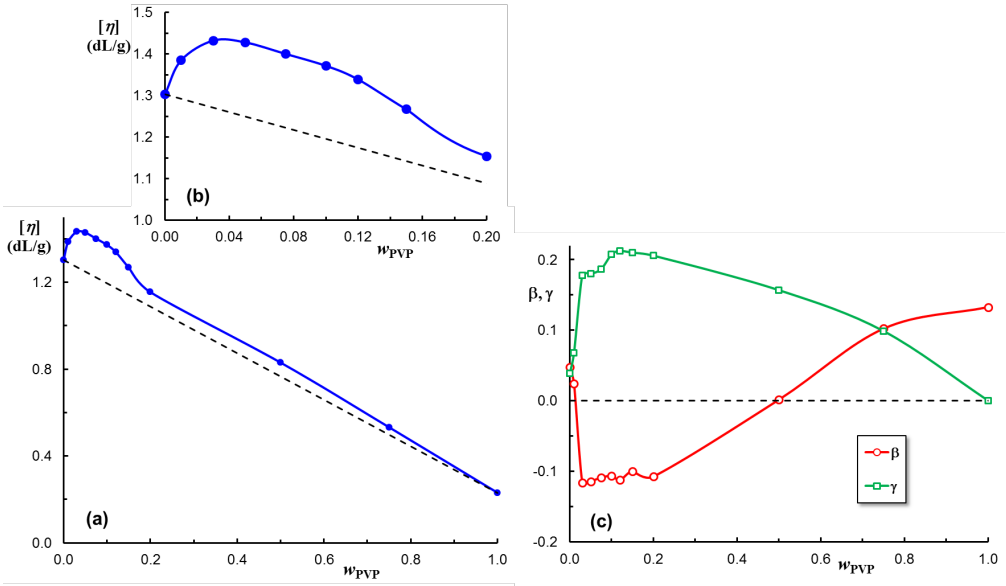


Figure 4: Intrinsic viscosity (a,b) and interaction parameters (c) as a function of HPC/PVP composition (water, 25 °C)

The parameters $\beta < 0$ and $\gamma > 0$ in the region of composition where HPC represents the majority of macromolecules in the polymer mixture, suggesting that strong interactions are established between the unlike chains when PVP is added to the HPC solution. We can assume that the

preferred intermolecular contacts between HPC and PVP macromolecules represent the driving force that favors the formation of mixed isolated coils with a more extended chain conformation, determining the viscosity increase. The H-bond interactions are exhibited between the C=O groups of PVP and –OH groups of HPC (Scheme 1). In addition, other intra- and intermolecular interactions between –OH groups of HPC can occur. The existence of strong interactions between HPC and PVP was also evidenced in solid state by differential scanning calorimetry, infrared spectroscopy, X-ray diffraction and scanning electron microscopy.[30]

For a molecular interpretation of the viscometric parameters α, β and γ, the series expansion as a function of $\tilde{c}$ was written up to the fourth power:[22]

$$\ln \eta_{rel} = \tilde{c} + (\alpha - \beta)\,\tilde{c}^2 + (\beta^2 - \alpha\beta - \gamma)\tilde{c}^3 - (\beta^3 - \alpha\beta^2 - 2\beta\gamma + \alpha\gamma)\tilde{c}^4 \ldots$$

$$\ln \eta_{rel} = \tilde{c} + M_2\,\tilde{c}^2 + M_3\,\tilde{c}^3 - M_4\,\tilde{c}^4 \ldots \quad (6)$$

The factors M_2, M_3 and M_4 reveal the contributions of binary, ternary and quaternary interactions between the macromolecules in solution and they can be evaluated from the viscometric parameters α, β and γ according to:

$$M_2 = (\alpha - \beta)$$

$$M_3 = \beta^2 - \alpha\beta - \gamma$$

$$M_4 = \beta^3 - \alpha\beta^2 - 2\beta\gamma + \alpha\gamma \quad (7)$$

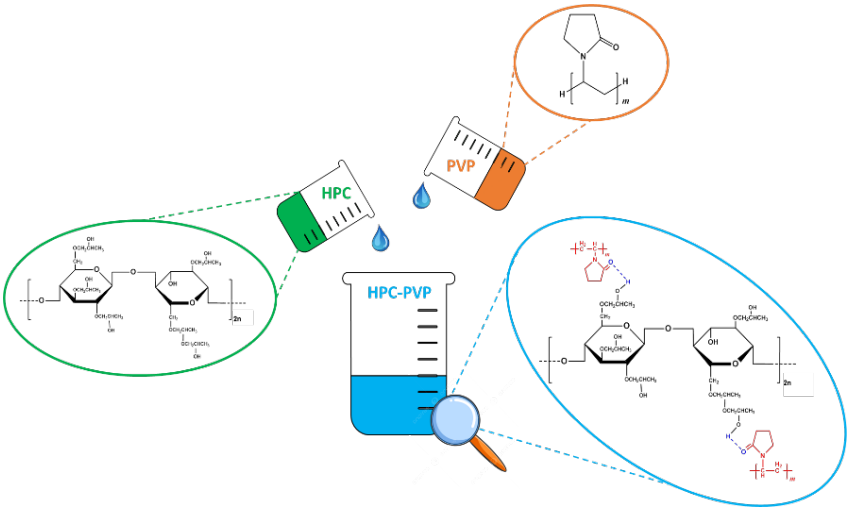
Figure 5 shows the dependence of M_2, M_3 and M_4 on HPC/PVP composition. For HPC-rich solutions, strong binary interactions are established, whereas for high PVP content, they are not preponderant. Ternary and quaternary interactions are less favored for low PVP content, but they become more important for high PVP content. However, the overall experimentally measurable effect is an increase in viscosity (Fig. 4), suggesting the existence of supramolecular formations that induce an increase in flow resistance. Previously, it was shown that HPC/PVP mixtures are miscible over the entire range of compositions in both solution and solid state.[30] We were focused on the effect of gradual PVP addition on the behavior of HPC solutions and we can conclude that the probability of mixed isolated coils formation is higher for $w_{PVP} \leq 0.2$.

Viscosity of semidilute solutions of HPC/PVP mixtures

The intrinsic viscosity determined in dilute solutions is correlated with the hydrodynamic volume occupied by a single macromolecular coil. As the polymer concentration increases, the polymer coils begin to touch one another, causing an increase in viscosity. The critical overlapping concentration, c*, which separates the dilute–semidilute regimes of concentration, can be calculated by using the Graessley model:[31]

$$c^* = 0.77/[\eta] \quad (8)$$

According to the data shown in Table 1, the c* values for all HPC/PVP mixtures are below 4 g/dL. Thus, a concentration of 5 g/dL belongs to the semidilute solution regime, regardless of the polymer composition.



Scheme 1: Illustration of intermolecular interactions between HPC and PVP

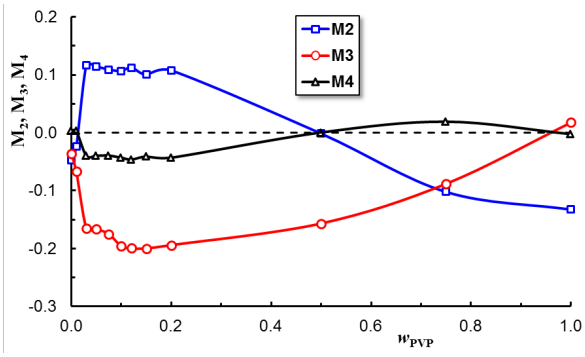


Figure 5: M_i (Eqs. (6) and (7)) for HPC/PVP mixtures of different compositions

The shear viscosities for semidilute solutions were determined using concentric cylinder geometry and the experimental values were not influenced by the applied shear rate ($\dot{\gamma}$).

Figure 6 presents the dependence of Newtonian viscosity on HPC/PVP composition for semidilute aqueous solutions of 5 g/dL at 25 °C. As it was observed for the dilute solutions, positive deviations from the additive rule were also registered.

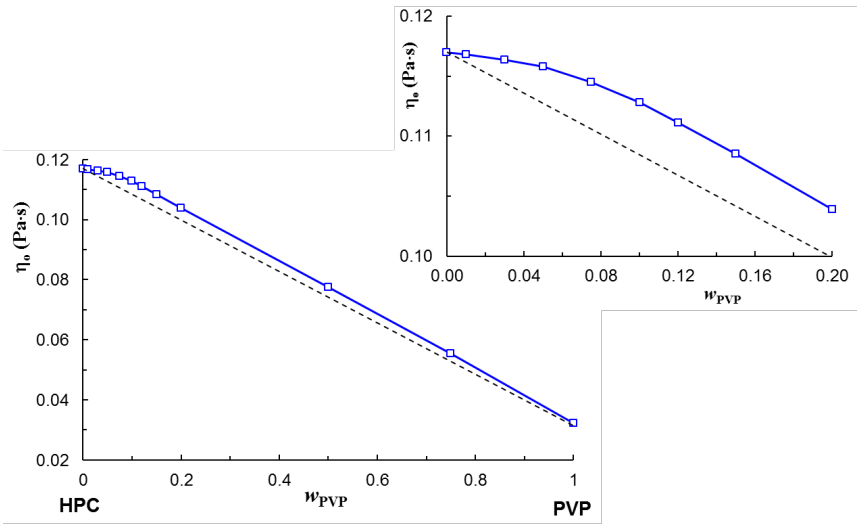


Figure 6: Newtonian viscosity as a function of HPC/PVP composition for 5 g/dL polymer content (water, 25 °C). The inset shows the detail for $w_{PVP} \leq 0.2$

Shear viscosity of concentrated isotropic HPC solutions in the presence of PVP

Upon increasing the concentration even more, still in the semidilute regime, the macromolecular chains start to entangle. Their lateral motion is strongly restricted, and the chains can nearly only move longitudinally, greatly increasing their viscosity. The total polymer concentration selected in this study for the rheological investigation of HPC/PVP mixtures was 30 g/dL, well above the entanglement concentration, which is usually several times higher than c*.[4,32]

For concentrated HPC/PVP solutions, the flow viscosity was determined as a function of shear rate ($\dot{\gamma}$), by using a plane-plane geometry. The Newtonian plateau was registered for low $\dot{\gamma}$ values (below 1 ÷ 30 s^{-1}, Fig. 7), when the relaxation time (τ) of the deformed macromolecules is shorter than the flow characteristic time and the solution preserves an isotropic entangled structure.[4] By increasing $\dot{\gamma}$, the macromolecules have no time to fully relax and they tend to orient along the flow direction, determining a pseudoplastic behavior, when the viscosity depends on the applied shear rate.

In the entangled state, similar to other semiflexible polysaccharides, the specific

viscosity obeys a power law as a function of degree of polymerization (N) and concentration of HPC:[27]

$$\eta_{sp} \sim N^{3.9} c^{4.2} \quad (9)$$

This dependence of viscosity on chain length and polymer concentration differs from that of synthetic flexible polymers (such as PVP) in thermodynamic good solvents:[32,33]

$$\eta_{sp} \sim N^{3.4} c^{3.5} \quad (10)$$

In a physical network of "bridges",[34] the mobility of the macromolecules is limited, favoring the intermolecular interactions (Scheme 1). As a consequence, the viscosity of HPC/PVP mixtures increases.

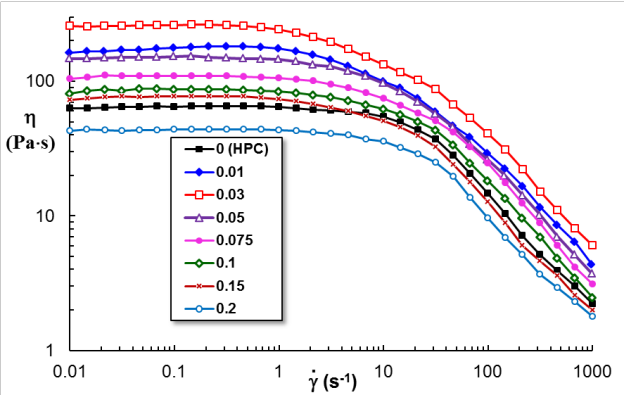


Figure 7: Flow curves registered at 25 °C for 30 g/dL HPC aqueous solutions in the presence of different content of PVP

In order to quantify the changes in the apparent viscosity of the HPC solution resulting from PVP addition, the following ratio was calculated:

$$R = \frac{\eta_{mixture} - \eta_{HPC}}{\eta_{HPC}} \quad (11)$$

Figure 8 shows the parameter R dependence on $\dot{\gamma}$ values for different HPC/PVP compositions. Positive values were obtained up to 10% PVP added into HPC solution and the highest effect was registered for 3% PVP due to interactions between unlike macromolecules leading to intermolecular complex formation (Scheme 1).

Another observation that can be pointed out from Figure 8 is the R maxima registered at high shear rates for concentrations lower than 10%. For shear rates between 100 s^{-1} and 500 s^{-1}, the HPC/PVP complex exhibits an increased resistance to flow, as compared with pure HPC solution. We can suppose that the polymer solutions form clusters at rest or during shear, being inhomogeneous on the mesoscopic scale.[35] In the absence of external forces, segmental clusters are formed, resulting from the chain connectivity (tendency to minimize Gibbs energy).[36] Under shear conditions, molecular clusters can appear and their size increases by shear (as a result of entropy minimization).[35] Thus, HPC and PVP macromolecules/segments may flow together due to the strong interactions established between them.

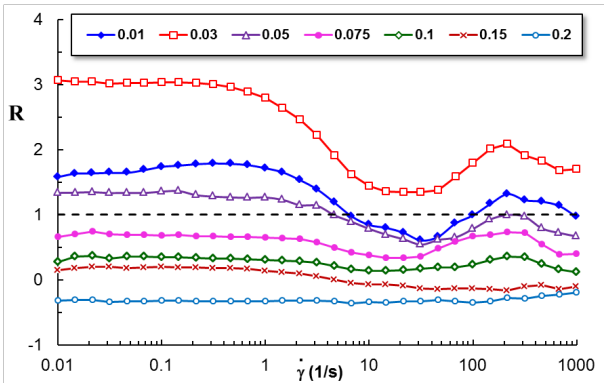


Figure 8: Changes of apparent viscosity (reflected by parameter R, Eq. (11)) by addition of PVP into HPC solution (total polymer concentration of 30 g/dL)

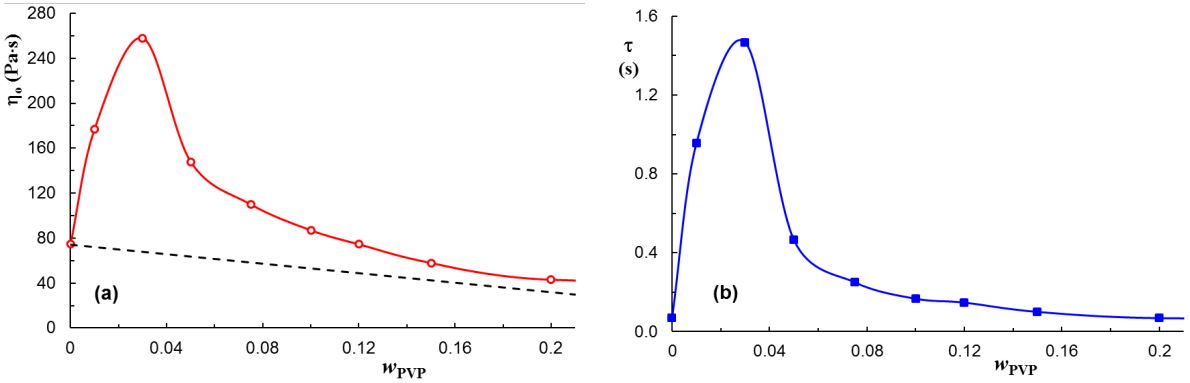


Figure 9: Influence of PVP addition on (a) Newtonian viscosity and (b) relaxation time of HPC solution (water, 25 °C, total polymer concentration of 30 g/dL)

Similar to intrinsic viscosity, the Newtonian viscosity (obtained at low shear rates) and the longest relaxation time (τ) present a maximum value for the HPC/PVP mixture with 3% PVP (Fig. 9). HPC contains in its structure a large amount of proton-donating hydroxyl groups, whereas PVP macromolecules present proton-accepting carbonyl groups, able to form intermolecular hydrogen bonding with HPC chains. The hydroxypropyl groups of HPC considerably influence the hydrophobic/hydrophilic balance at increasing temperatures, determining a phase transition that can lead further to polymer segregation due to a deswelling effect and the formation of a thermosensitive network.[14,37]

This behavior can be exploited in the additive manufacturing[38] or pharmaceutical field, especially for drug delivery applications.[10,15,39,40] Furthermore, the addition of a small amount of PVP into HPC-based systems enhances the solubility profile of macromolecules and improves the drug loading and delivery.[15,39,40] It was shown that PVP addition into low molecular weight HPC matrices enhances the release profile of pilocarpine, increasing the diffusion rate of the water-soluble drug.[39]

HPC/PVP mixtures form nanofibers, which are appropriate for cutaneous applications.[41] The selection of a suitable composition with an optimum viscosity is crucial to obtain uniform, smooth and beadless electrospun nanofibers. Generally, the PVP addition influences the crystallinity and morphology of biopolymer films, with an impact on their mechanical, thermal and optical properties.[42] On the other hand, the addition of cellulose or cellulose derivatives into PVP films or electrospun nanofibers improves the mechanical properties and thermal stability of the composites.[43,44]

CONCLUSION

The addition of low amounts of PVP to HPC solutions could be considered beneficial for many applications, both polymers being suitable for biomedical applications. The intermolecular hydrogen bonding between the proton-donating hydroxyl groups of HPC and proton-accepting carbonyl groups of PVP is favorable for low PVP content (up to 15%) and involves the formation of mixed coils in dilute solution. Also, the formation of HPC/PVP complexes in semidilute and concentrated solutions presents the advantage that they are able to increase viscosity at very low PVP concentrations, without chemical crosslinking.

ACKNOWLEDGEMENTS: The authors thank Ashland Aqualon (Klucel) for kindly providing the HPC sample.

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