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Swelling Dynamics of Cross-Linked Poly(acrylic acid) and Neutralized Poly(acrylate-*co*-acrylic acid) in Aqueous Solutions of (Hydroxypropyl)cellulose

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ABSTRACT: When a polyelectrolyte gel is placed into a (hydroxypropyl)cellulose (HPC) solution, there is a change in the local HPC concentration around the swelling particle. The HPC concentration profile development around the swelling gel is recorded as a function of time by optical microscopy, using a novel technique involving an isotropic-to-anisotropic phase transition, with a spatial resolution of a few microns. Using this technique the kinetics of swelling of poly(acrylic acid) (PAAc) and neutralized poly(acrylate-*co*-acrylic acid) (PAA) gels when placed in aqueous (hydroxypropyl)cellulose solutions and the HPC concentration redistributions in these systems were studied. A 75% neutralized PAA gel swells by sorbing mainly water, leaving HPC outside. At the beginning of the swelling, the HPC concentration gradient can be as high as 2% μm^{-1}, with a transient HPC concentration of 80%. By contrast, in the case of the PAAc gel, the HPC concentration inside the particle is greater than outside at equilibrium. The final equilibrium is attained in two steps. During the first the solvent penetrates into the particle, producing a large temporary increase of HPC concentration outside the gel. During the second step HPC slowly penetrates the hydrogel, due to preferential interactions between PAAc and HPC, as shown by capillary viscometry for the corresponding linear polymer mixtures.

Introduction

Polyelectrolyte hydrogels can be used as models in the biomechanical area, such as the study of electrophysiological phenomena, the behavior of muscles, etc. Hydrogels may also be used for modeling dissipative structures and biological systems. They are also widely used as superabsorbents and thickening additives.

For all these reasons it is important to understand the gel and the state of the surrounding medium during swelling. Many theoretical and experimental studies have been performed on the hydrogel behavior in solutions of low and high molecular weight solutes. Most of them are limited to the description of the final equilibrium of the charged network plus solution system, but a few describe the kinetics of the hydrogel swelling and contraction as a function of pH and/or ionic strength of the surrounding solution[1−3] or the time dependence of poly(methacrylic acid), PMA, membrane contraction when exposed to solutions of polymers which complex with PMA.[4]

When a dry gel is immersed into a polymer solution, its concentration varies before attaining an equilibrium polymer concentration inside and outside the gel. A theoretical discussion of possible mechanisms has been given by Brochard.[5] She examined the possibility of having linear chains entering a gel. The theoretical approach is limited to the case of a polymer network in a solution of the same polymer, but the discussion on the dynamics has a general validity. She noted that if a swollen gel is placed into a polymer solution, and if the polymer can penetrate the gel, this should occur in two stages. First, the network swells with the solvent which is composed of small, mobile molecules. Later, polymer chains diffuse into the gel. She suggested that this must be a slow process because the driving force (the osmotic pressure difference) and the diffusion constant of the chains are low.

To our knowledge, however, no one has considered the linear polymer concentration changes in the solution near the hydrogel sample during swelling. These changes will have a large influence on the kinetics of the gel swelling and/or contraction and on the final equilibrium. The lack of experimental data may be due to the difficulties of recording such concentration changes around a hydrogel. One of the problems is that a spatial resolution of the concentration measurements on the order of a micron must be achieved.

Recently[6] we showed that it is possible to take advantage of an isotropic-to-liquid crystalline transition of the polymer solution into which the gel is placed to monitor the time dependence of the polymer concentration changes. A dry gel particle is placed in an isotropic aqueous (hydroxypropyl)cellulose (HPC) solution between two glass plates. The hydrogel sorbs water from the polymer solution, and this leads to a local increase of the HPC concentration around the particle. The HPC solution becomes anisotropic (and thus birefringent) when its concentration exceeds 42% due to a liquid crystalline phase transition. The growth and disappearance of the trace of the liquid crystalline phase, i.e. the appearance of a birefringent layer around the hydrogel, are monitored by optical microscopy.

The objectives of this paper are to study the HPC concentration profiles around the swelling gel and their dynamics for two different hydrogels based on cross-linked polyacrylic acid or cross-linked 75% neutralized poly(acrylate-*co*-acrylic acid) (PAA), placed in aqueous HPC solutions. The equilibrium properties of these two systems will be studied and compared. In the case of the 75% neutralized PAA the HPC concentration was found to be larger outside the gel. By contrast, for the poly(acrylic acid) (PAAc) gel the concentration of HPC is higher inside the gel than outside. In this case a two-stage process occurs, a fast one involving only the motion of the solvent molecules and a slow one involving the motion of the linear polymer. In order to understand the difference found for the two systems, the

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viscosity of the corresponding linear polymer/HPC/water mixtures was studied.

Experimental Section

Materials. The hydrogel sample used in this study is a 75% neutralized PAA cross-linked by *N,N′*-methylenebisacrylamide (0.06% mole fraction of the cross-linking agent with respect to the monomer) kindly provided and characterized by Atochem, Centre d'Application de Levallois, France. The initial state is a powder of dry gel particles with a size of about 1 mm. Each particle is an agglomerate of small spherical parts.

The hydrogel based on cross-linked polyacrylic acid was obtained from the 75% neutralized PAA. The 75% neutralized PAA gel was placed in a hydrochloric acid solution where the sodium ions were replaced by hydrogen ones, then precipitated in acetone, and dried to constant weight in vacuum in the presence of P_2O_5. As for the initial 75% neutralized PAA gel, the poly(acrylic acid) gel was a powder of agglomerated small spherical particles. The equilibrium degree of swelling in water is 400 g/g for the PAA hydrogel and 70 g/g for the poly(acrylic acid) hydrogel.

Aqueous solutions of HPC (Klucel type E, molecular weight 6×10^4 as given by manufacturer), kindly provided by Aqualon-Hercules, France, were prepared by mixing HPC and water. The concentrations are given by weight. Below 42%, the solutions are isotropic. Above 42%, they are in a liquid crystalline phase.

Methods. To investigate the hydrogel interaction with the concentrated HPC solution, a dry gel particle was placed in a very large amount of a 40% HPC solution between two glass plates under an optical microscope. The pH of the solution was controlled and fixed at 7. For monitoring the swelling of the particle and the HPC concentration changes, the procedure described in ref 6 for the 75% neutralized PAA hydrogel was repeated. It is based on the fact that a local increase of the HPC concentration induces an isotropic liquid-to-liquid crystal phase transition when the HPC concentration becomes greater than 42%. The induced birefringence can be measured and related to the HPC concentration (see the following paragraph).

The equilibrium HPC concentration redistribution in the presence of the 75% neutralized PAA or the PAAc hydrogels was also studied. Different amounts of dry hydrogel were added to a fixed volume of HPC solution of given concentrations. When the equilibrium state was reached, the HPC concentration outside the gel was measured by refractometry (see the following paragraph). In order to compare the results obtained for different samples, reduced coordinates are used. The reduced initial HPC concentration is C_{HPC}^0/C_{gel}, and the reduced final HPC concentration is C_{HPC}^1/C_{gel}, with C_{gel} being the hydrogel concentration. All the concentrations are specified as molarities of monomer residues.

The dependence of the degree of swelling of the two hydrogel samples on the initial HPC concentration was measured. A swelling ratio α is defined as $\alpha = m(C_{HPC}^0)/m_0$, where $m(C_{HPC}^0)$ is the weight of the hydrogel at equilibrium after interaction with a HPC solution of initial concentration C_{HPC}^0 and m_0 is the equilibrium gel weight in pure water.

The interactions between PAAc or 75% neutralized PAA linear polymers with HPC in water were studied by viscometry, using a poly(acrylic acid) sample (with $M = 1.2 \times 10^5$ obtained by measuring intrinsic viscosity in 2 N NaOH solution) and a 75% neutralized PAA obtained from PAAc by adding the proper amount of NaOH. The specific viscosity η_{sp} was measured with a Ubbelohde viscometer at 25 °C for the two mixtures (PAAc + HPC or 75% neutralized PAA + HPC) in water, with different compositions of the two components.

The dependence of the specific viscosity on polymer concentration for each single polymer in water was also measured. Applying a mixing rule described below, the additive curves for both systems were calculated. The presence and the character of the deviation of the experimental curve from the calculated additive one for each system (PAAc + HPC or 75% neutralized PAA + HPC) depend on the existence and the structure of interpolymer complexes that may be formed.

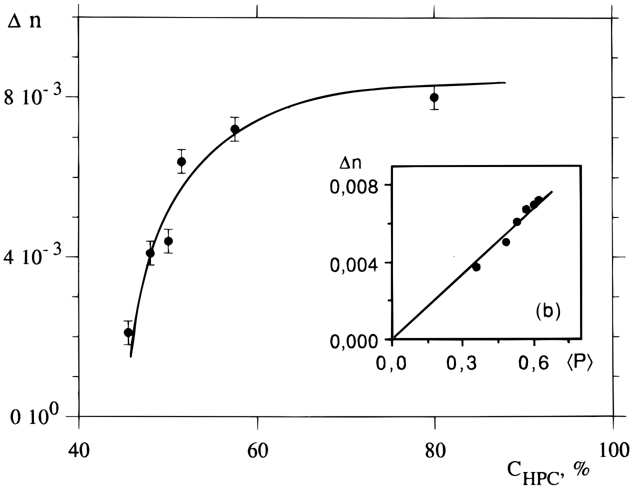

Figure 1. Calibration curve for hydroxypropylcellulose in water: birefringence Δn versus concentration C_{HPC}. Inset: birefringence versus order parameter $\langle P \rangle$.

Measurements of HPC Concentration. At equilibrium, when it is possible to extract a gel particle from HPC solution, the measurements of HPC concentrations were performed by refractometry, using a calibration curve of the refractive index as a function of the HPC concentration.

For cases where the dynamics of the concentration changes are studied, it is not possible to extract the sample. HPC concentrations above 42% were measured from the birefringent colored layer developed around the gel particle.

Using a birefringent gypsum plate, it can be shown that the larger refractive index of the birefringent layer is in the direction perpendicular to the gel surface, which means that the liquid crystalline director is also perpendicular to the gel surface. The birefringent layer has a sequence of colors starting from the gel surface. Each color corresponds to a given light path δ.[7] As the thickness of the layer is fixed and known, one can obtain the birefringence Δn. The variation of Δn from the gel surface can be measured with a very good spatial resolution of about a few microns.

The birefringent HPC layer is without visible defects, in a form of a monodomain.[6] It is thus necessary to relate Δn and HPC concentration in the same well-ordered state. For this, the two refractive indices n_r and n_t were measured for well-aligned HPC solutions at different concentrations with the help of an Abbe refractometer. The concentrated HPC solutions were prepared using the method described in ref 8. Well-aligned HPC solutions were obtained by shearing the solution directly on the plate of refractometer. The Δn versus HPC concentration is given in Figure 1. In order to check the validity of this method, this result was compared to the order parameter $\langle P \rangle$ of the same solutions obtained by X-ray scattering under high shear.[9] Since a linear dependence between Δn and $\langle P \rangle$ is obtained, as shown in the inset of Figure 1, Figure 1 can be used as a valid calibration curve for measuring the HPC concentration around a swelling particle.

Results and Discussion

HPC Concentration Distribution around a Swelling 75% Neutralized PAA Gel. An example of the HPC concentration distribution near the 75% neutralized PAA hydrogel (the time of swelling is 8 min) is shown in Figure 2, *d* being the distance in the HPC solution starting from the boundary of the particle. The horizontal lines give the thickness of each colored zone and thus the mean value of the HPC concentration corresponding to each color. The zone closest to the gel can have a very high HPC concentration, near 80%.

Such graphs were obtained for several swelling times starting from the beginning of the swelling up to the

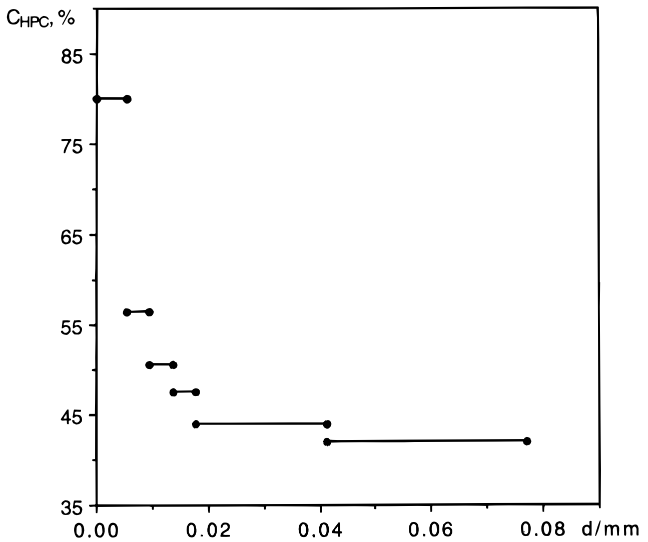

Figure 2. HPC concentration (C_{HPC}) versus distance *d* from the gel boundary (time of swelling 8 min).

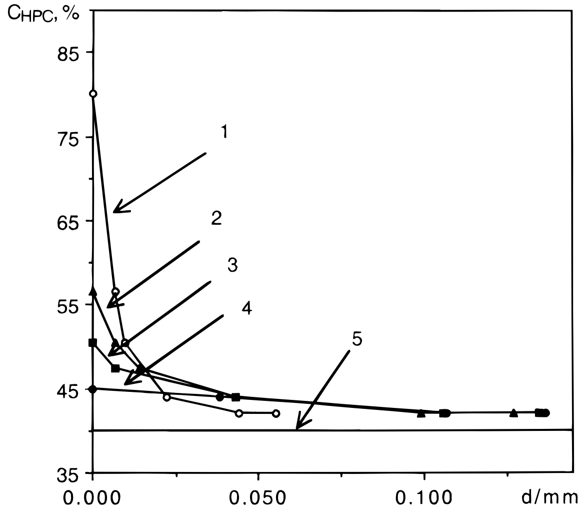

Figure 3. HPC concentration (C_{HPC}) versus distance *d* from the gel boundary for different times after the beginning of swelling, 5 min (1), 35 min (2), 52 min (3), and 122 min (4). The horizontal line 5 corresponds to the initial and final state, with a HPC concentration of 40%.

time when all the colors disappeared. The development of the HPC concentration profile is given in Figure 3 at four different swelling times. The points correspond to the mean values of the HPC concentration in each zone. The boundary between the biphasic and the isotropic areas is shown by the last dot on each curve. The straight line, denoted as 5 in Figure 3, corresponds to the initial and the final concentration of the HPC solution (40%).

Figure 3 shows that at the beginning of swelling the birefringent area is not wide (less than 0.05 mm) with a sharp decrease of the HPC concentration (curve 1). The concentration gradient is then very large, up to 2% $\mu\mathrm{m}^{-1}$. The birefringent layer becomes larger with time, with a disappearance of the concentrated regions and a reduction of the concentration gradient (curves 2 and 3). The final stage of the process is the decrease of the width of the birefringent area (curve 4, Figure 3) and the slow smoothing of the HPC concentration to the initial 40% value. This may last for several hours because of the low diffusion coefficient of the linear HPC chain in a very viscous medium.

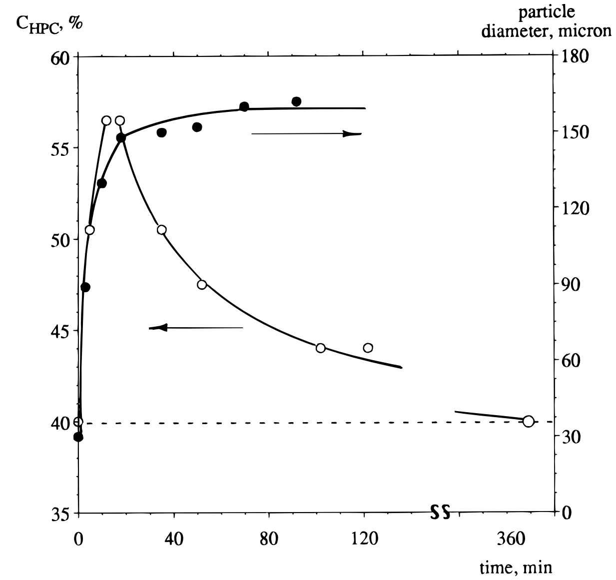

Figure 4. The development of the HPC concentration (C_{HPC}) as a function of time at a fixed distance ($d = 0.011$ mm) from the hydrogel particle. The concentration reaches an equilibrium concentration (40%) after a very long time (about 6 h). The dashed line corresponds to the initial and final HPC concentration of 40%. The diameter of the swelling gel particle (in micrometers) is also plotted as a function of time.

Another way of presenting these data is to plot the evolution of the HPC concentration with time at a given distance *d* from the gel. An example at $d = 0.011$ mm is given in Figure 4. It clearly shows how the concentration increases with time, passing through a maximum and then decreasing when the gel stops to swell. In the same figure the swelling of the gel particle is plotted as a function of time. It can be seen that the HPC concentration passes through a maximum when the gel swells to equilibrium. When the gel stops swelling, the HPC concentration goes back slowly to the initial steady-state concentration.

HPC Concentration Changes around a Swelling PAAc Gel. Changes in the appearance of a PAAc hydrogel particle placed in a large volume of a 40% HPC solution are shown in Figure 5. In Figure 5a, the beginning of the process is the same as for the 75% neutralized PAA hydrogel: the PAAc hydrogel swells by sorbing water, and this induces a local increase of the HPC concentration around the particle. This is clearly seen through the appearance of a birefringent layer, showing that the HPC concentration in this area is greater than 42%. As the degree of swelling of the PAAc gel in water is almost 6 times lower than for the 75% neutralized PAA gel, its swelling in the aqueous HPC solution (and thus the local increase of the HPC concentration) is not so pronounced. When the PAAc gel is swollen to equilibrium, diffusion smoothes the HPC concentration (Figure 5b) and the birefringent layer disappears. All this is similar to the 75% neutralized PAA gel behavior in concentrated HPC solutions described above.

The main difference between the two systems occurs after this first process. Contrary to the 75% neutralized PAA gel + HPC system where at equilibrium the hydrogel + polymer solution becomes everywhere fully isotropic, the PAAc hydrogel sorbs HPC macromolecules from the solution in a second stage and a liquid crystalline phase transition takes place inside the particle. This is shown in Figure 5c, where the gel particle is birefringent, floating in an isotropic HPC

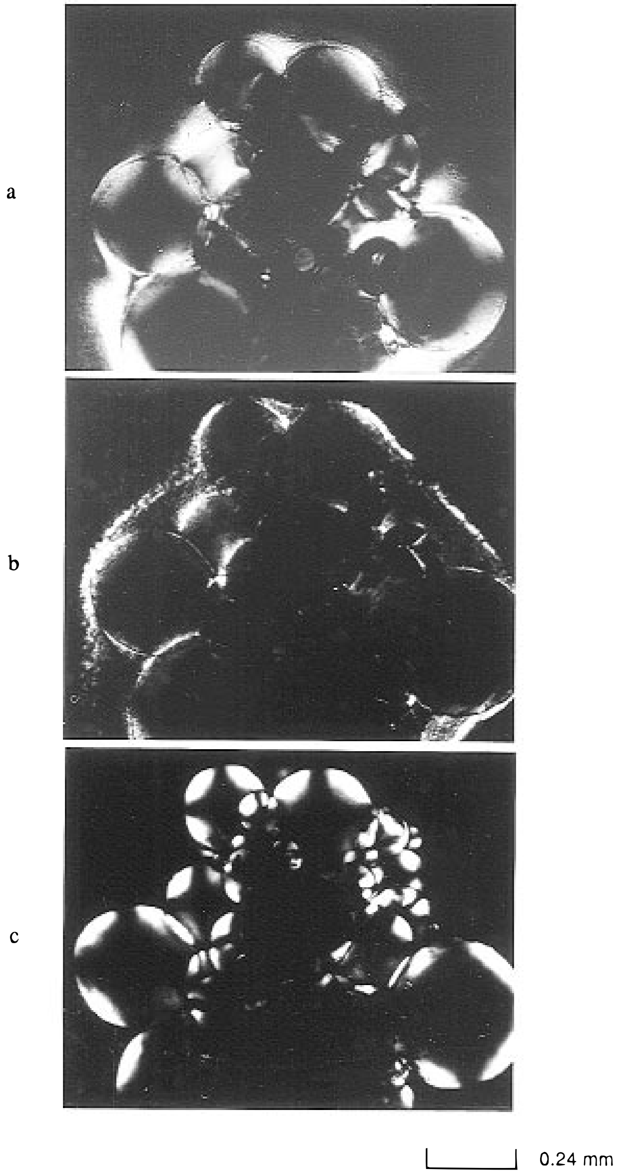

Figure 5. Optical micrographs taken between crossed polarizers of the cross-linked PAAc particle swelling in an aqueous 40% HPC solution: (a) $t = 12$ min after immersing of the dry gel in the HPC solution; (b) $t = 51$ min; (c) $t = 5$ h.

solution. This is the stable final equilibrium situation. Thus for PAAc hydrogels, the HPC concentration is higher inside the gel than outside.

It is known that in gels exposed to solutions of linear polymer, the linear polymer penetrates the gel. Theoretically this was predicted by Brochard,[5] Vasilevskaya and Khokhlov,[10] and Khokhlov and Kramarenko[11] and experimentally shown by Kabanov et al.[12] and Osada and Sato.[13] The linear polymer concentration inside the gel as predicted by theories is always smaller than that outside. In addition, to our knowledge there are no experimental results describing the kinetics of concentration changes of a linear polymer around a swelling gel.

What we show here is that the polymer concentration inside the gel may be greater than the initial polymer concentration. In addition, starting from a dry gel, the final equilibrium state is attained in two stages. First the swelling of the gel is mainly due to solvent penetration, with a large temporary local increase of the polymer concentration outside the gel. In the second stage the linear polymer slowly penetrates inside the gel.

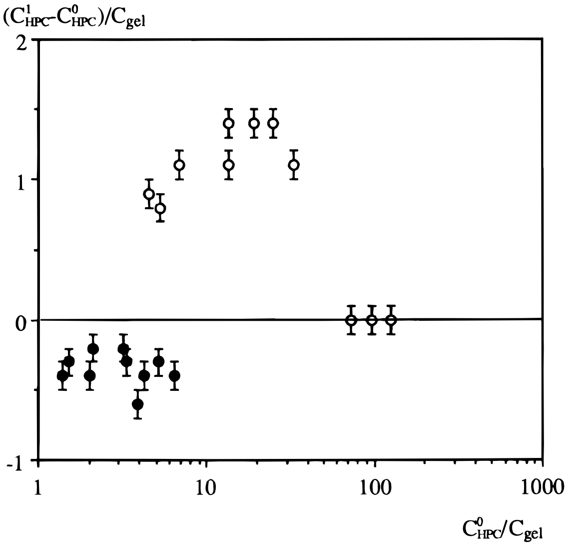

Figure 6. HPC concentration redistribution in the system hydrogel + HPC at equilibrium. Cross-linked PAAc (dark circles) and cross-linked 75% neutralized PAA (opened circles). Solid line corresponds to the absence of sorption or desorption of HPC by the gel.

HPC Concentration Redistribution at Equilibrium. The observed difference between the two systems can be readily seen when measuring the HPC concentration redistribution in the presence of the hydrogel at equilibrium. To study this, a certain amount of dry gel particles is placed in a finite volume of HPC solution. The HPC concentration outside the gel after the equilibrium was attained, $C_{HPC}{}^1$, is compared with the initial HPC concentration $C_{HPC}{}^0$.

The results are given on Figure 6. In these coordinates the horizontal line corresponds to neither sorption nor desorption of HPC macromolecules by the hydrogel. From Figure 6 it is clear that for the 75% neutralized PAA gel all the experimental points are above the straight line, i.e. $C_{HPC}{}^1 > C_{HPC}{}^0$. The 75% neutralized PAA gel is sorbing mainly water, thus increasing the outside HPC concentration (for example, when the initial HPC concentration is 40%, the solution becomes fully anisotropic everywhere in the equilibrium state). However, when the gel concentration in the system hydrogel + solution is very low compared to $C_{HPC}{}^0$, e.g. $C_{HPC}{}^0/C_{gel} > 100$, the difference in $C_{HPC}{}^0$ and $C_{HPC}{}^1$ is too small to be detectable.

By contrast, the PAAc gel is sorbing HPC from the solution, and thus the polymer concentration outside decreases from its initial value (see Figure 6, dark circles). For example, after the interaction with the 30% HPC solution, the PAAc gel particles in the equilibrium state become birefringent, i.e. the HPC concentration inside the gel is greater than 42%.

Degree of Swelling. The ability of preferentially sorbing either water or the linear polymer is reflected by the different swelling of the hydrogel samples in HPC solutions. The ratio of the swelling of the gel in the presence or absence of interaction with HPC on the initial reduced HPC concentration is given in Figure 7 (curve 1, swelling of the 75% neutralized PAA gel; curve 2, swelling of the PAAc gel; $\alpha = 1$ is swelling in water at equilibrium). For the 75% neutralized PAA gel, the results show that when the HPC concentration is less than or equal to the 75% neutralized PAA concentration, its degree of swelling is almost the same as in pure

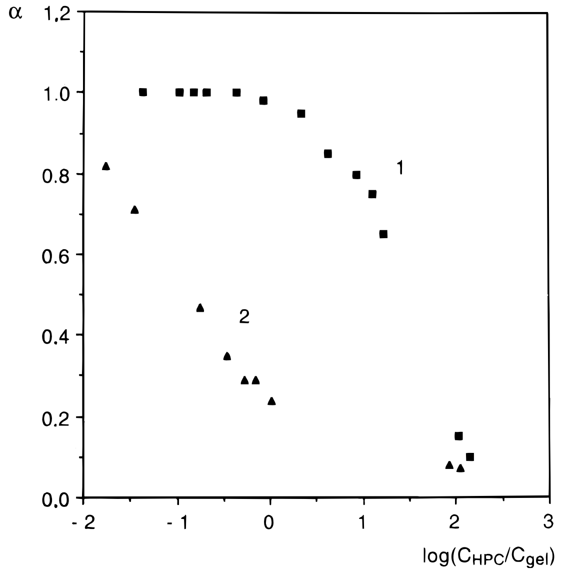

Figure 7. Dependence of α for the 75% neutralized PAA hydrogel (1) and PAAc hydrogel (2) on the reduced initial HPC concentration C_{HPC}^0/C_{gel}.

water. The 75% neutralized PAA hydrogel swelling decreases only when C_{HPC}^0 is more than 10 times higher than C_{gel}. At high concentrations, it has a degree of swelling like a noncharged polymer network.

By contrast to this behavior, the PAAc gel swells quite weakly in the HPC solutions even at low linear polymer concentrations. The decrease of its degree of swelling compared with the one in water begins when $C_{HPC}^0/C_{gel} < 0.1$.

Specific Viscosity. In order to understand the difference between the 75% neutralized PAA and PAAc hydrogel behavior in HPC solutions, the interaction between the corresponding linear polymers and HPC was investigated. Since the formation of interpolymer complex leads to conformational changes in the mixed polymers, the measurements of the viscosity of the diluted linear polymer/HPC mixtures can give useful information about their interactions.[14,15] Thus, pairs of linear flexible polymers such as PAAc + poly(oxyethylene), PAAc + poly(vinyl alcohol), poly(methacrylic acid) poly(oxyethylene), etc. form interpolymer complexes due to cooperative hydrogen bonding between the undissociated carboxylic and OH groups.[13,15-17] The structure of the complex formed by a flexible and a semirigid polymer (for example, PAAc + (hydroxyethyl)cellulose[18]) has some peculiarities, but the main features in the complex formation are the same.

In Figure 8a,b the dependence of the specific viscosity of the PAAc + HPC (Figure 8a) and 75% neutralized PAA + HPC (Figure 8b) mixtures on the composition is given. They are compared with the specific viscosity which would be obtained if these two polymers made an independent contribution to η_{sp}, i.e. the sum of the viscosities of each polymer solution with the concentration they have in the mixture.

In both cases there is a difference between the experimental and the additive curves. In the PAAc + HPC system, one can suppose that a complex of the "zip" mechanism type is formed:[17] the mixture is turbid and its viscosity is lower than the corresponding additive one. In the presence of a low molecular weight acid this mixture precipitates. This suggests that in PAAc + HPC the chains of each polymer prefer to interact with each other than with the solvent, forming an interpolymer complex.

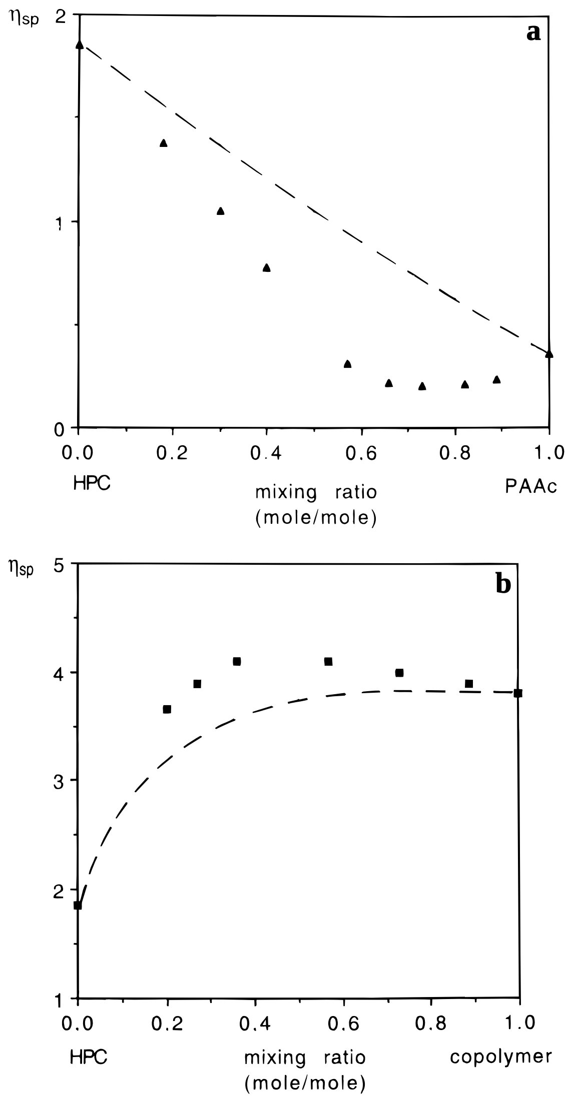

Figure 8. Dependence of the specific viscosity of the mixture on composition. The initial concentration of the components were 0.05 mol/L. The calculated additive curves for both mixtures are given by dashed lines: (a) aqueous solutions of linear PAAc + HPC; (b) aqueous solutions of linear poly(0.75 acrylate-*co*-0.25 acrylic acid) + HPC.

In the mixture 75% neutralized PAA + HPC, not more than 25 mol % of the 75% neutralized PAA chain may take part in complex formation, probably through hydrogen bonding. The viscosity of this mixture is higher than the corresponding additive one because the interactions of the charged carboxyl groups of the 75% neutralized PAA with water are more pronounced. We have suggested that the polymer association leads here to the formation of a branched structure. Similar results were obtained for pairs of linear flexible polymers.[16,19] To investigate these points is by itself interesting and will be studied in the future.

Conclusions

Hydrogels based on cross-linked 75% neutralized PAA and poly(acrylic acid) immersed into an aqueous (hydroxypropyl)cellulose solution show a very different behavior, due to preferential interaction between the poly(acrylic acid) and HPC, as shown by viscometry for the corresponding linear polymer mixtures.

The swelling of 75% neutralized PAA hydrogels in HPC aqueous solutions is accompanied by a large temporary increase of the HPC concentration around the gel particle. This highly concentrated HPC layer is very viscous and acts as a diffusion barrier despite its small thickness. It is at the origin of the long times required for reaching the HPC distribution equilibruim.

The cross-linked PAAc swells less than the cross-linked 75% neutralized PAA, but at equilibrium the concentration of HPC is greater *inside* the gel than outside. The PAAc gel swells in two steps. The first step is similar to the swelling behavior of the 75% neutralized PAA gel. The PAAc hydrogel swells by absorbing mainly water, leaving a HPC-rich layer outside. In a second step, HPC concentrates inside the PAAc gel; therefore, at equilibrium the HPC concentration inside the gel is larger than outside.

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