Rheology of Polyelectrolyte Solutions: 
Current Understanding and Perspectives 

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Polyelectrolytes with ionic groups are abundant in nature and essential in life. Counterions can be released into a solution and leave charges on polyelectrolyte chains, making the solution properties significantly different from those of electrically neutral polymers. A lot of scientific efforts have been made to elucidate the effect of electrostatic interactions on the conformation and material properties of polyelectrolyte solutions. However, unfortunately, our understanding is still far away from being perfect, and continuous efforts need to be provided by researchers from different aspects. This short review will highlight the research progress made in the past few years on the conformation and viscoelastic properties of polyelectrolyte solutions.

Key Words: Polyelectrolyte / Solution rheology / Electrostatic screening / Ionic liquid / Polymerized ionic liquid

1. INTRODUCTION

When dissociative groups are introduced onto polymer repeating units, counterions can be released into a solution and leave charges on a polymer chain. Such charged polymers in solution have been firstly named as polyelectrolytes by Fuoss,\(^1\) and can be found in both natural (e.g., DNA and polysaccharides) and synthetic (e.g., poly(acrylic acid) and poly(sodium styrene sulfonate)) polymers. Polyelectrolytes are often divided into the following two groups depending on the nature of dissociative groups.\(^1\) Weak polyelectrolytes are macromolecules possessing basic or acidic groups which are protonated or deprotonated depending on the solution pH, while strong polyelectrolytes describe macromolecules that consist of salt ions. Note that the term of weak and strong is sometimes used to characterize polyelectrolytes having different charge fractions.\(^2\) There is also the case that polyelectrolytes have both positive and negative charges on the same chain. Such polymers are called as polyampholytes,\(^3\) and many proteins belong to this category. On the other hand, when two oppositely charged polyelectrolytes are mixed in a solution, coacervates can be formed to make the solution viscoelastic, and their properties are one of the hot topics in the field of the molecular rheology although not discussed in this review.\(^4-6\)

The unique conformation and resulting rheological properties for polyelectrolyte solutions have been recognized since the beginning of the 20th century. For example, Sackur reported the variation of the viscosity of a protein solution with the addition of bases and salts in 1902.\(^17\) Nowadays, it is widely accepted that these unique features of polyelectrolyte solutions are originated from electrostatic interactions acting between the repeating units with dissociated counterions. As a result, extensive experimental\(^7-37\) and theoretical\(^38-57\) studies have been conducted over the past century in order to understand accurately and precisely the effect of the electrostatic interactions on the conformation and viscoelastic properties of polyelectrolytes in solution. However, as discussed below, there still remain many active research subjects due to the conflicting information reported on the experimental results in comparison to theoretical predictions. Nevertheless, polyelectrolytes are surrounding us in applications, such as rheological thickeners for foods,\(^58\) adhesives for dental materials,\(^59\) colloid stabilizers in inks and concretes,\(^60, 61\) desalination membranes,\(^62\) coating materials for drug delivery.\(^63\)

It should be also emphasized that polymerized ionic liquids, consisting of bulky cations and anions weakly coordinated, have been attracting research interests as a new class of polyelectrolytes in the past two decades.\(^64\) Since polymerized ionic liquids can dissolve into organic solvents with low dielectric constants (e.g., THF and methanol)\(^65-67\) as well as ion concentrated solutions (e.g., ionic liquids)\(^68, 69\) how electrostatic interactions act on polyelectrolyte chains in such solvents is another interesting subject matter to be addressed.

Several review articles have been published to summarize the latest progress in the polyelectrolyte science at each
turning point. The most recent one was provided in 2020 by Dobrynin who reviewed the conformation and dynamics of polyelectrolyte solutions at polymer concentration ranges spanning from dilute to entangled regimes. Therefore, the subjects of debate for polyelectrolyte solutions which were not described by Dobrynin’s review will be introduced in this short review.

2. POLYELECTROLYTE SOLUTIONS IN THE UNENTANGLED REGIME

A long-recognized but still poorly understood behavior for polyelectrolyte solutions involves the Fuoss law in which the specific viscosity is proportional to the square root of polymer concentration in the semifluid unentangled (SU) regime. Dobrynin et al. explained this unique trend theoretically by proposing the electrostatic blob model that accounts for the effect of the electrostatic interactions on the polyelectrolyte conformation by using the Debye-Hückel theory. Applying the Rouse-Zimm model for unentangled polymer solutions, they showed that the specific viscosity $\eta_{sp,SU}$ for polyelectrolyte SU solutions in good solvents was predicted as

$$\eta_{sp,SU} \propto N c_p^{1/2} b_B^{1/2} \left( 1 + \frac{2 \Delta c_b}{c_p} \right)^{1/2}$$

where $N$, $b$, $c_p$, and $c_s$ are the degree of polymerization, the monomer size, the molar concentration of monomers, and that of additive salts. $A$ is the number of monomers between uncondensed charges. In other words, $A$ characterizes the number of charged units on a polyelectrolyte chain, which depends on the type of polyelectrolytes and solvents. $B = b^2/(l_B^2)$, $l_B$ is the dimensionless contour length parameter, where $l_B = e^2/(4\pi \varepsilon_r \varepsilon_0 k_BT)$ is the Bjerrum length defining a length at which the electrostatic energy between two charges becomes comparable to the thermal energy. Here, $e$, $\varepsilon_r$, $\varepsilon_0$, $k_B$, and $T$ are the elementary charge, the dielectric constant of solvent, the vacuum permittivity, the Boltzmann constant, and the absolute temperature. Using the relation for the terminal modulus $G = \eta/\tau_{SU} = 1000 c_p RT/N$ of unentangled polymer solutions, the relaxation time of polyelectrolyte SU solutions can be derived as

$$\tau_{SU} \propto \eta_s N^2 c_p^{1/2} b_B^{1/2} \left( 1 + \frac{2 \Delta c_b}{c_p} \right)^{1/2}$$

where $\eta_s$ is the solvent viscosity. Thus, the relaxation time for polyelectrolyte solutions in the SU regime is predicted to decrease with increasing polymer concentrations, in good agreement with the experimental data. Conceptually speaking, this is due to the chain shrinkage caused by the self-screening of charge effects by polyelectrolyte ions. As a result, the polyelectrolyte chain size decreases faster than the chain size of neutral polymers, leading to a decrease in the relaxation time with the increasing polymer concentration. However, as technology progresses and the number of polyelectrolyte studies increases, researchers found some polyelectrolytes which showed the polymer concentration dependence of $\eta_{sp}$ deviating from the classical Fuoss law in the SU regime.

Figure 1 shows the dependence of $\eta_{sp}$ on $c_p$ for several polyelectrolyte solutions reported in literature. Since the values of $\eta_{sp}$ are plotted against $c_p$ normalized by the overlap monomer concentration $c_p^*$ for each polyelectrolyte, the $\eta_{sp}$ data for $c_p/c_p^* > 1$ lie in the SU regime until $c_p$ reaches the next critical polymer concentration, i.e., the entanglement polymer concentration above which solutions potentially belong to the semifluid entangled regime. In Fig. 1, the specific viscosity for solutions of poly(sodium styrene sulfonate) (NaPSS), poly(sodium acrylate) (NaPAA), and N-methyl-2-vinylpyridinium chloride random copolymer (PMVP-Cl) exhibits a power-law dependence of $\eta_{sp} \propto c_p^{0.5}$, in good agreement with Dobrynin’s scaling prediction (dashed line in Fig. 1 given by Eq. 1). On the other hand, for solutions of sodium carboxymethyl cellulose (NaCMC), sodium hyaluronate (NaHA), Chitosan, and hyaluronate...
3. POLYCELESTROLYTE SOLUTIONS IN THE ENTANGLED REGIME

While an extensive study has been conducted for unentangled polyelectrolyte solutions, there is only a limited number of studies dealing with the entanglement dynamics of polyelectrolytes in solution.\textsuperscript{11,12,16,24,27,31,32,36} There are potentially two problems hindering the experimental analysis to investigate the effect of the electrostatic interactions on the solution rheology of entangled polyelectrolyte solutions. One is the difficulty of measuring the responses characteristic for entangled solutions, such as the rubbery-plastic plateau regime at high frequencies. With conventional rheometers, the measurable rheological response of polyelectrolyte solutions is restricted to that in the terminal regime observed at low frequencies since polyelectrolytes can dissolve only in low-viscosity solvents, such as water. For this reason, the zero-shear viscosity has been routinely analyzed and compared with the scaling predictions.\textsuperscript{12,16,24,27,31,32} The other is related to the width of the polyelectrolyte semiflexible entangled (SE) regime where the electrostatic interactions affect the viscoelastic properties of polyelectrolyte solutions. Dobrynin et al.\textsuperscript{42} defined the polyelectrolyte SE regime in the concentration range of \(c_e \leq c_p < c_D\), with \(c_e\) the entanglement concentration at which entanglements start to form; and \(c_D\) a critical polymer concentration at which the electrostatic screening length is equal to the electrostatic blob size. Polyelectrolyte solutions at \(c_p \geq c_D\) behave like neutral polymers so their solution properties are predicted to follow the scaling laws of neutral polymers in the SE regime. Since the polyelectrolyte SE regime is typically narrow, a quantitative scaling analysis of semiflexible entangled solutions of polyelectrolytes tend to become challenging. For example, considering that the transition of the polymer concentration regime takes place gradually, the experimental results with narrow SE regimes might over- or underestimate the scaling exponent for the polyelectrolyte SE regime due to different exponents in the surrounding regimes.\textsuperscript{27} As a result, existing experimental studies reached different conclusions regarding the validity of Dobrynin’s scaling model for salt-free polyelectrolyte solutions in the SE regime.

Table I summarizes the scaling laws of the key viscoelastic parameters for semidilute entangled solutions of both salt-free polyelectrolytes and neutral polymers in good solvents.\textsuperscript{42} The tube model for neutral polymers was applied to derive the scaling laws for polyelectrolyte SE solutions.\textsuperscript{75} Among them, the plateau modulus \(G_e\), the Rouse time of an entanglement strand \(\tau_e\), and the reptation time \(\tau_{rep}\) are specific for entangled polymer solutions. The use of the relaxation time ratio \(\tau_{rep}/\tau_e\) was proposed to eliminate the effect of the retarded solvent dynamics on the polymer relaxation times which may occur at high polymer concentrations due to the cooperative motions of solvent molecules with polymer chains.\textsuperscript{36,76} It is interesting to note that the reptation time of polyelectrolyte SE solutions is predicted to be independent of the polymer concentration at the salt-free condition.

The first doubt on the applicability about the tube model to polyelectrolyte systems was raised by Lopez.\textsuperscript{31,32} He reported that while the predicted power-law dependence of \(\eta_{sp}\) on \(c_p\) was observed in the SE regime, the dependence of \(c_e\) on the degree of polymerization \(N\) was much weaker than the one expected. The obtained scaling was \(c_e \propto N^{-0.77}\), rather identical to that for neutral polymers in good solvents. In order to evaluate the dependence of \(G_e\), \(\tau_e\), and \(\tau_{rep}\) on \(c_p\), Han and Colby\textsuperscript{50} dissolved a high molecular weight poly(cesium styrene sulfonate) (CsPSS; \(M_n = 2.83 \times 10^6\) Da) in a glass-forming glycerol, and estimated the values of \(G_e\), \(\tau_e\), and \(\tau_{rep}\) from the master curve.

Table I: Summary of scaling laws of the specific viscosity \(\eta_{sp}\), the entanglement concentration \(c_e\), the plateau modulus \(G_e\), the Rouse time of an entanglement strand \(\tau_e\), and the reptation time \(\tau_{rep}\) for salt-free polyelectrolytes and neutral polymer in good solvents. More details on the scaling law can be found in the original article by Dobrynin et al.\textsuperscript{52}

| Semidilute Entangled (SE) Regime |                 |                 |
|---------------------------------|-----------------|-----------------|
|                                | salt-free polyelectrolytes | neutral polymers |
| \(\eta_{sp}\)                   | \(N^{3.3}c^{1.5}\) | \(N^{3.9}c^{2.9}\) |
| \(c_e\)                         | \(N^{-2}\)       | \(N^{-0.76}\)   |
| \(G_e\)                         | \(N^{0.0}c^{1.5}\) | \(N^{0.2}c^{2.3}\) |
| \(\tau_e\)                      | \(N^{0.0}c^{-1.5}\) | \(N^{0.9}c^{-2.3}\) |
| \(\tau_{rep}\)                  | \(N^{3.3}c^{0}\) | \(N^{3.3}c^{1.6}\) |
| \(\tau_{rep}/\tau_e\)           | \(N^{3.3}c^{1.5}\) | \(N^{3.9}c^{3.9}\) |
of the complex modulus constructed by applying the time-temperature superposition (TTS) principle. Fig. 2 shows the literature report about the dependence of \( \tau_{\text{rep}} / \tau_e \) and \( G_e \) on \( c_p \) normalized by the entanglement concentration, i.e., \( c_p / c_e \), for CsPSS solutions in glycerol.\(^{36}\) The value of \( \tau_{\text{rep}} / \tau_e \) and \( G_e \) were scaled as \( \tau_{\text{rep}} / \tau_e \propto c_p^4 \) and \( G_e \propto c_p^{2.3} \), respectively, with respect to \( c_p \), in good agreement with the scaling predictions for neutral polymers in the SE regime. Thus, they concluded that salt-free polyelectrolyte solutions behaved like solutions of neutral polymers in the entangled concentration regime.

Although the rheological data provided in literature and their analysis seem to be reliable and accurate, it should be noted that the number of charged units on a polyelectrolyte chain (i.e., 1/A) may decrease with decreasing \( \varepsilon \) of solvents in accordance with the Manning counterion condensation theory.\(^{39}\) Based on Dobrynin’s scaling model, smaller the value of 1/A, smaller the value of \( c_D \) and larger the value of \( c_e \), narrowing the polyelectrolyte PE regime as \( \varepsilon \) decreases. Eventually, the polyelectrolyte solution might reach the worst case that \( c_e \) is larger than \( c_D \), indicating that the properties of the polyelectrolyte solution in the entangled regime follow those for neutral polymers at any \( c_p \). In fact, Dobrynin and Jacobs\(^{56}\) have argued this point in their recent paper, and showed that polyelectrolytes with \( N > 10^4 \) are required to observe the effect of the electrostatic interactions on the viscoelastic properties of polyelectrolyte SE solutions.

Following Dobrynin and Jacobs’ suggestion, Matsumoto et al.\(^{77}\) recently investigated the entanglement properties of aqueous polyelectrolyte solutions containing a high molecular weight NaPSS (\( M_u = 3.16 \times 10^6 \) Da, \( N = 1.53 \times 10^4 \)) by using diffusing wave spectroscopy (DWS). The DWS is a dynamic light scattering technique for turbid samples, and enables the measurement of the complex modulus of viscoelastic materials at frequencies as large as \( 10^7 \text{ rad s}^{-1} \).\(^{78}\) Fig. 3(a) shows the dependence of the complex modulus \( G' \) on the angular frequency \( \omega \) for a NaPSS solution at \( c_p = 0.297 \text{ M} \) measured at 25°C by using DWS. Here, monodisperse polystyrene particles of 211 nm in diameter were suspended as tracer particles in the NaPSS solution for the DWS experiment. In Fig. 3(a), they observed a full width of the rubbery-plateau regime, evidenced by the two crossovers of \( G' \) and \( G'' \) at high and low frequencies. Thus, using the DWS technique, the values of \( G_e \),

![Fig. 2](image1.png)

**Fig. 2** Dependence of the plateau modulus \( G_e \) and the ratio of the reptation time \( \tau_{\text{rep}} \) to the Rouse time of an entanglement strand \( \tau_r \) on the monomer concentration normalized by the entanglement polymer concentration, i.e., \( c_p / c_e \), for solutions of poly(cesium styrene sulfonate) (CsPSS) in glycerol at 25°C. Data are provided from ref. 36.

![Fig. 3](image2.png)

**Fig. 3** (a) The complex modulus is plotted against angular frequencies for an aqueous solution of NaPSS at \( c_p = 0.297 \text{ M} \) measured at 25°C by using DWS. The test solution is seeded by monodisperse polystyrene (PS) tracer particles of 211 nm in diameter at 0.3 wt%. Green arrows guide the determination of \( G_e \), \( \tau_{\text{rep}} \), and \( \tau_r \). (b) Dependence of the plateau modulus \( G_e \) and the ratio of the reptation time \( \tau_{\text{rep}} \) to the Rouse time of an entanglement strand \( \tau_r \) on the monomer concentration \( c_p \) for solutions of NaPSS in water at 25°C. Dashed lines represent the scaling prediction of \( G_e \propto c_p^{4.0} \) and \( \tau_{\text{rep}} / \tau_r \propto c_p^{1.5} \) for salt-free polyelectrolyte solutions in the SE regime. Data are provided from ref. 77.
τ_{rep} and τ_e were successfully estimated without applying the TTS principle, which is usually required for the measurement via bulk rheometers. Fig. 3(b) plots the literature values of G′ and τ_{rep}/τ_e as a function of c_p. Both G′ and τ_{rep}/τ_e increased with increasing c_p with a power of 1.5, in good agreement with the scaling prediction of G′ and τ_{rep}/τ_e for salt-free polyelectrolyte SE solutions. The DWS microelectrode study indicates that the concept of the tube model for neutral polymers can be applicable to polyelectrolyte systems. However, further in-depth study of different types of polyelectrolytes is required towards a comprehensive understanding of the charge effect on the entanglement dynamics of polyelectrolytes.

4. POLYMERIZED IONIC LIQUIDS IN IONIC LIQUID SOLUTIONS

The final topic of this short review involves the solution rheology of polymerized ionic liquids (PILs). Ionic liquids (ILs) are molten salts consisting of bulky cations or anions weakly coordinated.79) Owing to regulated electrostatic interactions acting between ionic liquid ions, many ILs can exist in the liquid state at room temperature. Accordingly, ILs have attracted research attention for use as a plasticizer to tune the mechanical properties of polymer-based materials, similar to the role of ordinary solvents.

PILs are macromolecules possessing the ionic liquid structure on their repeating units. Since PILs can retain some advantageous features of ILs (e.g., thermal/electrochemical stability, high ionic conductivity, the ability to absorb carbon dioxide), they are considered as a promising candidate for electrolytes in batteries, membranes for gas filtration as well as stabilizers for nanoparticles.80) The performance of PIL-based materials can be further improved by adding, for example, ILs as solvents due to the increased mobility of polymer chains.81) In this context, understanding the viscoelastic properties of PILs in IL solutions are of great importance to optimize PIL-based material design and resulting performance.

The viscoelastic behavior of PILs in IL solutions resemble that for polyelectrolytes in salt solutions, such as NaPSS in aqueous solutions of sodium chloride. Therefore, how ionic liquid ions screen the charges on a PIL chain is a research question to be answered. For ordinary polyelectrolytes in salt solutions, the Debye-Hückel (DH) theory is often employed to predict the degree of charge screening at a given ion concentration. However, the DH theory is expected to be inapplicable at high IL concentrations since it assumes solutions with low salt content in the absence of ion-ion interactions. In fact, Smith et al.82) measured experimentally the screening length in IL solutions by using surface force apparatus, and showed that the screening length increased with increasing IL concentrations at high IL concentrations, following the decrease in the screening length in accordance with the prediction by the DH theory at low IL concentrations. The observed increase in the screening length at high IL concentrations is unexpected, and named as the charge underscreening effect to specify such a unique screening behavior.83)

Motivated by the above observation, Matsumoto et al.84,85) conducted systematic rheological characterizations of a PIL, poly-(1-butyl-3-vinylimidazolium bis(trifluoromethanesulfonyl)imide) (PC_4-TFSI), in a mixture of an IL, 1-butyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (Bmim-TFSI), and a non-ionic solvent, dimethylformamide (DMF). Specifically, they evaluated the specific viscosity η_sp and the longest relaxation time λ while varying IL concentrations c_IL at a fixed monomer concentration c_p in both dilute and semidilute unentangled regimes. Fig. 4 shows an example of the dependence of η_sp on c_IL for a PIL solution at c_p = 0.25 M in the semidilute unentangled regime. The value of η_sp remained a constant for c_IL < 3.0 × 10^{-3} M and started to decrease with increasing c_IL, exhibiting a typical charge screening behavior for polyelectrolyte solutions. However, η_sp reached a minimum at c_IL ~ 2 M, and started to increase when c_IL > 2 M, suggesting the charge underscreening effect on the viscosity of PILs in IL solutions.

In order to explain the observed non-monotonic dependence of η_sp on c_IL, Matsumoto et al.85) proposed a new charge screening model, given by

\[ η_{sp, SU}^{mod} = K_1 N \left( \frac{b}{B} \right)^{9/4} r_p^{5/4} (c_p \tau_B^{mod})^{3/2} + η_{sp, SU}^{int}, \]

where K_1 is the scaling prefactor. The first term of the right-hand side of Eq. 3 represents the charge underscreening effect in IL solutions in which the modified screening length \( r_B^{mod} \) controls the electrostatic interaction on PIL chains. The value of \( r_B^{mod} \) for polyelectrolyte solutions was expressed as

\[ r_B^{mod} \sim r_B \left( 1 + \frac{a^3}{r_B^3} \right), \]

where \( r_B = \left[ \frac{1000 N_A c_p}{b} \left( 1 + \frac{2 \pi c_p}{c_p} \right) \right]^{-1/2} \) is the screening length for polyelectrolyte solutions at low salt concentrations, while a is the ion diameter of salts. On the other hand, the second term of the right-hand side of Eq. 3 represents the complete charge screening at high IL concentrations. They introduced
this term based on an idea that an extended polyelectrolyte chain in a salt-free solution can shrink until its original chain size before influenced by electrostatic interactions. Fig. 4 compared the dependence of the measured $\eta_{sp}$ on $c_{IL}$ with the model prediction of $\eta_{sp, SU}$ (Eq. 1) proposed by Dobrynin et al. and that of $\eta_{sp, mod}$ (Eq. 3) proposed by Matsumoto et al. The predicted curve of $\eta_{sp, SU}$ agreed well with the measured $\eta_{sp}$ only at low IL concentrations, and deviated significantly for $c_{IL} > 0.1$ M. In particular, the upturn observed at $c_{IL} \sim 2$ M cannot be described by Dobrynin’s prediction based on the DH theory. On the other hand, the predicted curve of $\eta_{sp, mod}$ by Matsumoto’s charge screening model explained successfully the measured $\eta_{sp}$ over the measured $c_{IL}$ range.

From the above observation, it can be seen that PILs are a type of polyelectrolytes, but the knowledge of ordinary polyelectrolytes cannot be readily adopted to describe the properties of PIL solutions. For example, Matsumoto’s charge screening model predicts the effect of the ion diameter $a$ on the solution rheology of PILs in IL solutions. Josef and Guterman$^{67}$ reported that a peak was observed in the SAXS profile when dissolving their PIL in DMF, but it disappeared when adding methanol in the mixture of PIL and DMF. This result indicates that the solvent type affects the number of dissociated ions on a PIL chain even in salt-free solutions. A number of PILs with different chemical structures have been synthesized for the sake of advanced materials for various applications. However, unfortunately, there still remain a lot of subjects to be discussed in terms of the physical properties of PILs.

5. CONCLUSIONS AND OUTLOOK

Despite a dramatic progress in the polymer science, it is interesting to find that there remain a lot of active subjects on the conformation and viscoelastic properties of polyelectrolyte solutions. This is due predominantly to the complexity of the electrostatic interactions acting together with other interactions, such as hydrodynamic interactions and polymer-polymer/polymer-solvent interactions, in polyelectrolyte solutions. As a result, many conflicting results have been observed between experimental results and theoretical predictions.

The electrostatic blob model laid the foundations to understand the effect of the electrostatic interactions on the conformation and viscoelastic properties of polyelectrolyte solutions. In fact, the blob model successfully captured several characteristic features observed in the viscosity, osmotic pressure, and light scattering of polyelectrolytes both in salt-free and salt solutions. However, as the technology progresses, researchers found that some polyelectrolytes showed a significant deviation from the predictions by the electrostatic blob model. In the semidilute unentangled regime, polysaccharides often exhibited a stronger power-law dependence of the solution viscosity on the polymer concentration. The predicted entanglement dynamics of polyelectrolyte solutions has not been observed in many polyelectrolyte systems through conventional rheometers, while microrheology showed the entanglement properties in good agreement with those predicted by the electrostatic blob model. Polymerized ionic liquids can dissolve in ion-saturated solvents (e.g., ionic liquids), revealing an unexpected charge screening effect on the conformation of polyelectrolytes (i.e., chain re-expansion due to the charge underscreening). Although not introduced in the main text, the effect of the salt type, known as the Hofmeister series (or the ion specific effect), would be also an important question to be addressed.

Polyelectrolytes are ubiquitous in nature and have been used so far in various types of applications, such as food products, cosmetics, and oil recovery. Cellulose derivatives are a good example of natural polyelectrolytes and can potentially serve as an unlimited polymer source on the earth in the future. The global action for the sustainable development goals by the United Nations will boost the need of such a natural source of polymeric materials. Within this context, it is urgently

![Fig. 4 Dependence of the specific viscosity $\eta_{sp}$ at 25°C on the Bmim-TFSI concentration $c_{IL}$ for PC_{2}-TFSI solutions at $c_{p} = 0.25$ M. Blue-dashed curve represents the scaling law of $\eta_{sp, SU}$ for polyelectrolyte SU solutions (Eq. 1). Red-solid curve represents the predicted curve of $\eta_{sp, mod}$ based on Matsumoto’s charge screening model for polyelectrolyte SU solutions (Eq. 3). Here, $A = 2$, $b = a = 0.49$ nm, $n_{IL} = 6.0$, $e_{IL} = e_{DMF}(1 - 0.23c_{IL} + 1.5 \times 10^{-2}c_{IL}^{2})$, and $c_{IL} = c_{res} + c_{IL}$ were used in the curve fitting. The residual salt concentration $c_{res}$ in DMF is expressed as $c_{res} = (9.24 \times 10^{-6} + 0.009c_{sp})$ M, while the dielectric constant $e_{IL} = 38.3$ of DMF is measured using an impedance spectroscopy. Data are taken from ref. 69.](image-url)
required to provide a way to predict accurately the performance of polyelectrolyte-based materials. This review could not cover all the important issues present in the polyelectrolyte science, but the contents described in this review will motivate polymer scientists towards a comprehensive understanding of polyelectrolytes.

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