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Chain Conformation of Linear Polyelectrolyte in Salt Solutions: Sodium Poly(styrene sulfonate) in Potassium Chloride and Sodium Chloride Solutions

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ABSTRACT: Translational diffusion behaviors in infinite dilutions of sodium salts of poly(styrene sulfonate) (NaPSS) were studied by quasi-elastic light scattering as functions of salt concentration and molecular weight. With increasing added salt concentration, contraction of the hydrodynamic radius of NaPSS has been shown to be gradual, e.g., 130–95 Å for a sample with $M = 1.77 \times 10^5$ over a salt concentration range of 0.01–3 M NaCl; this behavior is contrasted to invariance of the radii of a globular protein, BSA, and of a short, rodlike fragment of DNA over the same salt concentration range. Further, NaPSS in 3.1 M KCl and 0.15 M NaCl behaves like a neutral polymer in Θ and good solvents, respectively. In terms of the molecular weight dependence of the translational diffusion coefficient in these ionic conditions, the results are established to be in excellent agreement with theoretical predictions deduced from various calculations by Skolnick and Fixman and by Odijk and Houwaart, provided we make use of the hydrodynamic factors of Akcasu and Han for neutral polymers in Θ and good solvents.

Introduction

This short report has a simple objective. It is to show that a linear synthetic polyelectrolyte responds to solvent ionic conditions in the context of theories of de Gennes et al.,[1] of Skolnick and Fixman,[2] of Odijk,[3] and Odijk and Houwaart[4] while a globular protein and a rodlike fragment of DNA remain impervious to such.

Translation diffusion of neutral polymers in dilute solution has been studied extensively by dynamic light scattering whereby the relationships of the translational diffusion coefficient, D_0, and molecular weight, M, in Θ and good solvents, $D_0 \propto M^{-0.5}$ and $D_0 \propto M^{-0.6}$, respectively, are well established experimentally and theoretically.[5] On the other hand, the situation is somewhat different with linear polyelectrolytes either of synthetic or biological origin.[6] Unlike neutral organic polymers in different solvent conditions, polyelectrolytes must be examined in terms of the solvent ionic conditions by means of varying concentration of added salt, C_s, since one of the crucial, independent variables is ionic strength affecting the chain conformation. For instance, the molecular weight dependence of intrinsic viscosity is sensitively controlled by C_s; the molecular weight exponent of the Mark–Houwink–Sakurada relation has been shown[7,8] to vary over a wide range, 0.45–1.59, reflecting the conformation change of polyelectrolytes. Pursuant to this kind of conformational probes, we have examined the dependences of D_0 on molecular weight, M, and salt concentration, C_s, with the sodium salt of poly(styrene sulfonate) (NaPSS). Relative to C_s dependence on NaPSS, it is contrasted to those of bovine serum albumin (BSA) and a rodlike DNA fragment.

Experimental Section

Materials. Four of the NaPSSs with $M = 1.77 \times 10^5$, 3.54×10^5, 6.9×10^5, and 1.06×10^6 were purchased from Pressure Chemical Co. (Pittsburgh, PA), while four others with $M = 2.54 \times 10^4$, 4.07×10^4, 5.79×10^4, and 1.46 t 10^5 were sulfonated in our laboratory according to Vink's method[9] on parent polystyrene samples purchased from the same vendor. The last one with $M = 2.15 \times 10^5$ was a gift from Daniel Brown of the Polymers Division of the National Bureau of Standards. The molecular weight distribution of the NaPSS samples was characterized by GPC to have $M_w/M_n \approx 1.2$. The degree of sulfonation for all samples was estimated to be around unity, some by elemental analysis for sulfur and others by potentiometric titration. As for the four commercial samples, we take the estimate by Koene and Mandel.[10] Bovine serum albumin (BSA, Pentex Brand Monomer Standard, Miles, Laboratories, Elkhart, IN) as an example of a globular macromolecule has been used. Contamination by dimers in the sample as provided by the vendor to be the monomer standard was once characterized to be very small earlier in this laboratory.[11] A similar conclusion has been drawn by Gabriel and Johnson's group,[12,13] who have used the same brand sample without further purification, contrary to their earlier practice. DNA with 150 ± 10 base pairs was isolated from chicken nucleosome core particles[14] and had the absorbance ratios of $A_{260}/A_{280} = 1.87$ and $A_{260}/A_{230} = 2.12$. The sample purity was assessed by electrophoresis on 7% polyacrylamide slab gels with Hae III restriction fragments of pBR 322 DNA as the marker.

All solutions were prepared with deionized water that was made from house distilled water upon passing it through the millipore Q2 system (Millipore) with one carbon and two ion-exchange filters, and NaCl and KCl were of analytical grade (Merck). Scattering solutions were made dust-free by filtration through 0.2-μm Acrodisc filters. Mononucleosome DNA solutions were dialyzed (4 × 500 mL over a period of 24 h) at 4 °C against buffer solutions of various ionic strengths. DNA concentrations were determined by optical absorption at 260 nm using an extinction coefficient of 20 mg^{-1} mL cm^{-1}. Buffers for BSA and DNA were, respectively, 8 mM phosphates, pH 7.5 at 25 °C, and 2.5 mM Tris, 0.1 mM EDTA, pH 8.95 at 20 °C, and that for NaPSS was 8 mM phosphates except 2 mM phosphates for the least ionic strength medium case.

Method. The instrument used for quasi-elastic light scattering (QELS) was the same one as in the report by Amis et al.[15]. The measurements were performed by using a 64 channel real time autocorrelator (K7025; Malvern Instruments, Ltd., Malvern, Worcestershire, U.K.) connected to a commercial goniometer (Malvern RR103). The autocorrelation function of the scattered light from an argon ion laser (Lexel 75–0.2) at 488-nm wavelength was acquired as a function of scattering angle from 30° to 130° with a selected photomultiplier tube (ITT FW 130) operated in a photon counting mode. The addition of a temperature control coil to the goniometer sample holder allowed temperature regulation in the range 5–35 °C to better than ±0.2 °C. An Apple II microcomputer interfaced to the correlator provided data storage and preliminary analyses. Subsequent detailed fittings to various model functions were performed using an unscaled Marquardt-type nonlinear regression routine on Vax 8600. The normalized intensity autocorrelation function for homodyne detection arising from a single diffusive process with the decay constant Γ can be written as

$$g^{(2)}(t) = 1 + Ae^{-2\Gamma t} \tag{1}$$

with $\Gamma = DQ^2$, where A is an instrumental optical constant, D is the diffusion coefficient, and Q is the magnitude of the scattering wave vector, defined as $(4\pi n/\lambda) \sin (\theta/2)$, with n being the re-

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Table I
Diffusion Coefficients of Different Samples of NaPSS at Three Ionic Conditions

	$10^7 D_0$, cm^2/s, at		
M_w	3.1 M KCl[a]	0.15 M NaCl[b]	2 mM Na_3PO_4 (polymer concn, g/L)[c]
2.54×10^4	9.7 ± 0.3	6.5 ± 0.2	5.9 ± 0.2 (1.0)
4.07×10^4	7.4 ± 0.2	5.0 ± 0.2	5.1 ± 0.4 (0.5)
5.79×10^4	6.4 ± 0.1	3.9 ± 0.1	3.5 ± 0.2 (0.5)
1.46×10^5	3.7 ± 0.1	2.19 ± 0.06	
1.77×10^5	3.84 ± 0.03	2.13 ± 0.06	1.62 ± 0.06 (0.2)
2.15×10^5		1.99 ± 0.08	
3.54×10^5	2.63 ± 0.05	1.31 ± 0.06	1.01 ± 0.07 (0.2)
6.90×10^5	2.00 ± 0.04	0.92 ± 0.01	0.72 ± 0.06 (0.05)
1.06×10^6	1.40 ± 0.04	0.69 ± 0.03	0.52 ± 0.04 (0.01)

[a] 3.1 M KCl and 8 mM phosphates. [b] 0.15 M NaCl and 8 mM phosphates. [c] 2 mM phosphates only; the minimum concentration of polymer used for the diffusion coefficient measurement in each sample is indicated in parentheses.

fractive index of the medium, λ the incident wavelength in vacuo, and θ the scattering angle.

In order to ensure that the diffusion coefficients, D, determined from Γ versus Q^2 plots (for the scattering angle range of 30°–130°), are reduced to D_0 values, we have effected the following for the three systems. For DNA fragment, we have extrapolated to infinite dilution the diffusion coefficient, D, obtained in polymer concentration range 0.1–2.0 g/L at 1 mM NaCl. For the other salt concentrations, 0.2 and 1.0 M, we have explicitly confirmed that the observed diffusion coefficients, D, have already reached D_0 values for the polymer concentration ranges 0.25–2.0 and 0.1–2.0 g/L, respectively. For BSA, our determinations were all made at a protein concentration of 1.0 g/L, which has been well shown repeatedly[16–18] to be low enough to produce D_0 values. In the case of NaPSS, the majority comes from the extrapolation to infinite dilution but not for all molecular weight at all salt concentrations; in particular, at low C_s at 2 mM phosphate (ionic strength 5 mM), we observed a sensitive dependence on polymer concentration and $g^{(2)}(t)$ showed two exponential decays as have been observed by others[10,19]. We refer to D_0 values as those extracted only from the fast mode. In Table I we indicate those values of D that are not explicitly confirmed to be D_0 by specifying the minimum polymer concentration at which D values are determined. In the case of high salt concentrations, their viscosities must be corrected for, and all D_0 values listed are corrected by multiplying relative viscosities[20] over that of pure water at 25 °C. For the sake of brevity, the details of sample preparations and corrections for D_0 values for DNA and NaPSS are deferred to elsewhere.[21,22]

Results and Discussion

Salt Concentration Dependence. In Figure 1 are displayed plots of D_0 versus C_s for all three configurations, BSA as a globule, a DNA fragment as a rod, and NaPSS as a wormlike-coil chain. The salt concentration spans for about 2 $^1/_2$ logarithmic decades for BSA and NaPSS and 3 decades for DNA. Qualitatively, chain contraction of NaPSS and conformational invariance of BSA and the DNA fragment upon increase of C_s are all clearly shown. For BSA, the horizontal line is drawn with $D_0 = 6.0 \times 10^{-7}$ cm^2/s at 25 °C, as has been reported by others,[16–18] where we have ignored the 2–4% increase of D_0 over the ionic strength range since our data precision is about equal to the correction and to the size of the squares in the figure. This amounts to the Stokes radius of 41 Å, which is now a well-established value. Turning to the DNA fragment, we can calculate D_0 with use of the length parameters of Elias and Eden[23] for DNA, namely a rod length L = 510 Å = 150bp × 3.4 Å/bp (bp stands for base pair) and diameter d = 26Å, via Broersma's formula,[24]

$$D_0 = (kT/3\Pi\eta_0 L)[\ln(L/d) + \gamma] \qquad (2)$$

where kT has the usual meaning, η_0 is solvent viscosity,

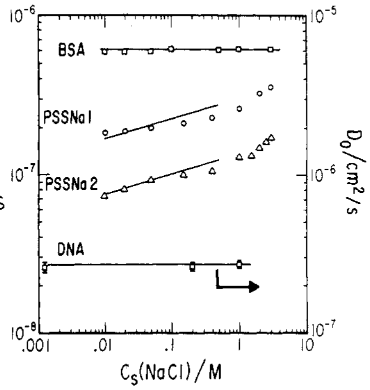

Figure 1. Salt concentration dependences of the translational diffusion coefficients for BSA, NaPSS, and DNA in dilute solution. BSA (Pentex brand monomer standard, 6.6 × 10^4 g/mol) at concentration of 1.0 g/L; PSSNa 1, $M = 1.77 \times 10^5$ g/mol at 0.5 g/L; PSSNa 2, $M = 6.90 \times 10^5$ g/mol at 0.1 g/L; DNA, 150 ± 10 bp at 0.25 g/L. Buffer conditions were 8 mM phosphates at 25 °C for BSA and NaPSS and 2.5 mM Tris, 0.1 mM EDTA, pH 8.95 at 20 °C for DNA. Solid lines and curves are the calculated values (see text).

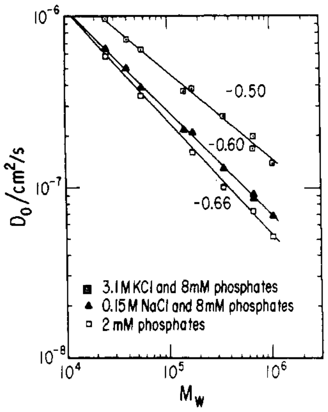

Figure 2. Molecular weight dependence of the translational diffusion coefficients of NaPSS in three ionic conditions; the data are collected in Table I.

and γ is the correction factor for end effect of a hydrodynamic cylinder model.[23,25] Here we choose γ = 0.33 at $L/d \approx 20$, following the corrections to the Broersma's original γ value by Tirado and de la Torre.[26] The calculated D_0 at 20 °C is 2.75 × 10^{-7} cm^2/s, and this is drawn in as the horizontal line over the data points. Agreement is remarkable indeed. Relative to the observed independence of D_0 with respect to C_s, a similar result has been reported earlier to Elias and Eden[23] with the rotational relaxation time over a decade in NaCl concentration, 0.2–2.5 mM, but with several different lengths all still within the rod limit. Hence, our result is in corroboration of theirs with D_0 over a wide range of C_s but with just one molecular weight sample.

Molecular Weight Dependence. Turning to the results for NaPSS, qualitatively, chain contraction with increasing C_s is apparent as stated earlier. We defer to later a quantitative comparison (as indicated by solid curves) with theories until their molecular weight dependences are presented. These in three different ionic conditions are shown in Figure 2. In 3.1 M KCl solution (with 8 mM phosphates) at 25 °C, which has been established to be θ condition for NaPSS by Takahashi et al.,[7] D_0 is shown to scale as $M^{-0.5}$ which is consistent with those behaviors of neutral organic polymers.[5] In 0.15 M NaCl (with 8 mM phosphates), the molecular weight exponent is found to be −0.6, which is indicative of a good solvent behavior in terms of neutral polymers such as polystyrene in toluene,[5] if we assume the electrostatic stiffening to be still small enough of an effect. Upon further decrease in C_s, we find the exponent of −0.66 which may be taken as evidence for chain stiffening at 2 mM phosphates.

In order to analyze these results more quantitatively, we compare them against the scaling relations arrived at by Odijk,[3] Odijk and Houwaart,[4] Skolnick and Fixman,[2] Fixman and Skolnick,[27] and Akcasu and Han.[28] The calculated D_0 is obtained as follows:

1. Calculate the total persistence length after taking into account the electrostatic part.
2. Calculate the unperturbed radius of gyration, $R_{g,0}$, of the wormlike coil model via Benoit and Doty's formula[29] based on the total persistence length.
3. Calculate the perturbed R_g with the excluded volume expansion factor, α_s, following the schemes of Odijk and Houwaart[4] and Fixman and Skolnick.[27]
4. Deduce the corresponding hydrodynamic radius, R_H, with use of the scaling relationship between R_g and R_H found applicable for neutral polymers in θ and good solvents, found by Akcasu and Han.[28]
5. Calculate the D_0 values to be compared with our experimental values.

The relevant equations used in following the above outlined procedure are given below:

1. The total persistence length is

$$L_t = L_p + L_e \tag{3}$$

where L_p is the intrinsic persistence length and L_e is the electrostatic persistence length which is given[4] by

$$L_e = (l_B/4\kappa^2a^2)f[1 - (8/3)Y^{-1} + (e^{-Y}/3)(Y + 5 + 8Y^{-1})] \tag{4}$$

where $Y = \kappa L$ and Bjerrum length, l_B, and Debye–Hückel screening length, κ^{-1}, are defined by

$$l_B = e^2/\epsilon kT \tag{5}$$

and

$$\kappa^2 = 8\pi l_B N_0 I \tag{6}$$

with e being the elementary charge, ϵ the dielectric permittivity of the solvent, I the ionic strength, N_0 Avogadro's number, and a the linear charge spacing along the chains. The factor f is introduced in eq 4 to account for the effective charge on the polyelectrolyte chain, which may be reduced in case of strong interactions with counterions such as that for $a < l_B$, $f = a^2/l_B^2$; this amounts to Manning's counterion condensation.

2. The unperturbed radius of gyration of wormlike coil of Benoit and Doty[29] is given by

$$R_{g,0}^2 = L_t^2[(x/3) - 1 + (2/x) - (2/x^2)(1 - e^{-x})] \tag{7}$$

where $x \equiv L/L_t$ with L being the chain contour length.

3. For the excluded volume parameters due to electrostatic repulsive interactions, we have the schemes of Odijk and Houwaart[4] and of Fixman and Skolnick[27]

$$R_g^2 = \alpha_s^2 R_{g,0}^2 \tag{8}$$

$$\alpha_s^2 = 0.541 + 0.459(1 + 6.04Z_{el})^{0.46} \tag{9}$$

$$Z_{el} = \{3^{3/2}\pi^{1/2}/16\}\kappa^{-1}[\ln (4\pi/l_B\kappa) + \sigma - (1/2)]L^{1/2}L_t^{-3/2} \tag{10}$$

where σ is Euler's number.

4. Akcasu and Han scalings[28] between R_H and R_g are given for Θ and good solvent conditions, repectively, as

$$R_H/R_g = 0.664 \qquad \text{(in } \Theta \text{ solvent)} \tag{11}$$

$$R_H/R_g = 0.537 \qquad \text{(in good solvent)} \tag{12}$$

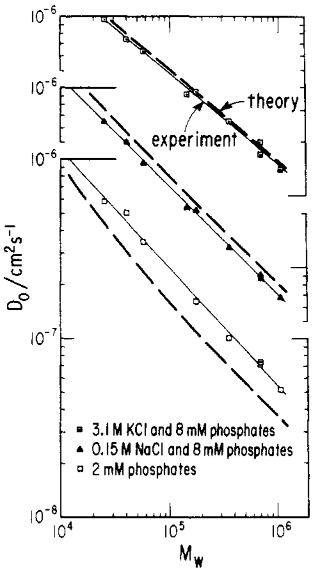

Figure 3. Comparison of D_0 between the experimental values and calculated ones, with the parameters collected in Table II.

Table II
Input and Calculated Parameters for Theoretically Predicted D_0

parameter	3.1 M KCl[a]	0.15 M NaCl[b]	2 mM Na_3PO_4[c]
L_p, Å	12.0	12.0	12.0
a, Å	2.5	2.5	2.5
l_B, Å	7.13	7.13	7.13
R_H/R_g	0.664	0.537	0.537
L_e, Å	0.10	1.75–1.91	21.64–64.47
α_s	1.0	1.5–2.3	1.8–2.2

[a] 3.1 M KCl and 8 mM phosphates; note here that only L_p, a, and R_H/R_g are used to calculate the solid curve in the top of Figure 3 since L_e is practically zero and $\alpha_s = 1$ is the Θ condition criterion. [b] 0.15 M NaCl and 8 mM phosphates. [c] 2 mM phosphates only.

5. Finally, the diffusion coefficient in infinite dilution is computed with the Stokes–Einstein equation,

$$D_0 = kT/6\pi\eta_0 R_H \tag{13}$$

In calculating D_0 values, we use three well-established length parameters, i.e., Bjerrum length of 7.1 Å in water at 25 °C; the linear charge spacing along the chain contour of NaPSS, a = 2.5 Å; and the intrinsic persistence length of NaPSS, L_p = 12 Å, as deduced by Nierlich et al.[30] and Weill and Maret.[31] In Figure 3, such comparisons are shown for all three salt conditions, where the calculated D_0 are given by dashed curves and the experimental data are the same ones shown in Figure 2, except that three plots are vertically displaced for clarity. The input parameters and calculated ones in the above scheme are all collected in Table II. Agreement in the Θ condition is remarkable indeed, particularly in view of the fact that there entered no adjustable parameter and no vertical rescaling of any kind is effected. Thus, NaPSS in 3.1 M KCl with 8 mM phosphates behaves like another neutral polymer at its Θ condition. We should reiterate that in this case there were only two input length parameters, namely L_p and a, since L_e is so small that its contribution to L_t is negligible and no excluded volume effect needs to be considered. As for the case of 0.15 M NaCl with 8 mM phosphates, agreement is still noteworthy although the theoretical prediction overestimates D_0 uniformly by about 10% over the entire molecular weight range. In the last case, in 2 mM phosphates, we have no right to expect an agreement to emerge since we still use the good solvent case scaling between R_H and R_g, i.e., 0.537. More importantly the chain stiffening is seen to be substantial by L_e values calculated to be 22–64 Å (see Table II.). Despite the discrepancy in the absolute magnitude of D_0 between predicted and experimental, the general trend is clearly borne out as predicted by the theories. We must offer a caveat here, however, that a direct comparison with R_g could have yielded better agreement since it would not involve step 4 (R_H/R_g factor) in the above procedure to calculate D_0. We should add that this sort of analysis has already been attempted by Koene and Mandel[10] with some success, although they did not resort to the R_H/R_g factor as we do here. Parenthetically we should add here that if we were to use the results of Le Bret,[32] which are obtained by using the full Poisson–Boltzmann equation instead of the linearized version and are essentially the same as the Fixman[33] results, the comparisons with experiments are only slightly changed in the 2 and 150 mM NaCl cases, whereas they are affected significantly by underestimating the experiment for Θ condition at 3.1 M KCl. The reason is simply that these calculations overestimate L_e in high salt conditions where it should reduce to a negligible attribution. We suggest on this basis that Fixman–Le Bret results are applicable better in low salt conditions as demonstrated by Tricot,[34] but their applicability in high salt conditions needs to be reexamined more carefully.

Returning to Figure 1, solid curves drawn over the data of NaPSS 1 and NaPSS 2 are arrived at by the same procedure but with the R_H/R_g factor at a constant value of 0.537, the good solvent case. Hence, it is likely to be inapplicable above $C_s = 1$ M. For this reason, the comparison is not extended beyond 0.5 M, although the hydrodynamic factor R_H/R_g has been shown to be more or less constant for the C_s range 0.01–1.0 M.[35] It is readily seen that the salt concentration dependence is predicted to be power laws (the "curves" are almost straight lines) whereas the experimental results show upturns at higher concentrations at around 0.5 M. This prediction could be an artifact, arising solely from the use of uniform scaling of R_g to R_H. Nevertheless, it seems safe to conclude that the theories appear to give a semiquantitative agreement with the experiment.

Conclusions

In 3.1 M KCl, NaPSS behaves like a neutral polymer at its Θ condition with persistence length of 12 Å and in 0.15 M NaCl it behaves like a neutral polymer in good solvent. The polyelectrolyte behaviors of chain stiffening with excluded volume effect as salt concentration is lowered are in a semiquantitative agreement with the theories, but with an important provision that our procedure to calculate D_0 entails two types of specific scaling of R_g and R_H, at Θ condition and in good solvent condition. There was no salt concentration dependence of D_0 of globular BSA and a rodlike DNA fragment with 150 base pairs.

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Registry No. NaPSS, 9080-79-9; KCl, 7447-40-7; NaCl, 7647-14-5; Na_3PO_4, 7601-54-9.

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