

Supporting Information

Chemical Analysis Using NMR and GPC. Proton nuclear magnetic resonance spectra (^1H NMR) were recorded on Bruker Avance-400 (400 MHz) and -500 (500 MHz). GPC elution operated at 40 °C with three Agilent PL HFIPgel columns (300 × 7.5 mm) and equipped with RI and UV/Vis detection. GPC was performed in a 1,1,1-trifluoroethanol (TFE) (with 0.02 M sodium trifluoroacetate) mobile phase, using poly(methyl methacrylate) as calibration standards, and operating at 0.75 mL/min. Samples were passed through a 0.4 μm filter prior to GPC analysis.

Table S1. GPC characterizations of zwitterionic polymers.

polymer	theo. M_n^a	TFE GPC ^b	
		M_n	PDI
PMPC ₁₀₇	31,600	34,900	1.19
PMPC ₂₀₀	59,000	61,300	1.21
PMBP ₉₆	32,400	27,300	1.37
PMBP ₁₆₄	55,300	38,800	1.32
PMPC ₂₇ - <i>b</i> - PMBP ₂₄	16,100	32,300	1.25

^a Determined by ^1H NMR. ^b PMMA standards.

Guinier plot analysis and bimodal solutions. Radius of gyration of the larger PMBP structures in solutions was determined using the Guinier plot (Guinier 1955). In this approximation, reliable only in dilute solutions and for the condition $qR_G \ll 1$, $\ln R = R_0 + \frac{R_G^2}{3} + \dots$. Since these solutions are bimodal, the intensity contribution, R , from the larger structures has to be evaluated. This is possible using either CONTIN analysis or multiple exponential fits. In either case Eq. 4 at $t \rightarrow 0$ will have a contribution from all diffusive species (Brown 1993):

$$\lim_{t \rightarrow 0} g_1(\Gamma, t) = \int_0^\infty w(\Gamma) d\Gamma \quad (1)$$

If there are two distinct distributions, $\int_0^\infty w(\Gamma) d\Gamma = \int_0^a w(\Gamma) d\Gamma + \int_a^b w(\Gamma) d\Gamma = w_1 + w_2$, where w_1 and w_2 are the total weights for each distribution.

Or for a double exponential fit (i.e. $g_1(\Gamma, t) = a_1 e^{\Gamma_1 t} + a_2 e^{\Gamma_2 t}$), it is simply the pre-factor for each exponential decay (Brown 1993):

$$\lim_{t \rightarrow 0} g_1(\Gamma, t) = a_1 + a_2 \quad (2)$$

The total weights are proportional to the intensity contribution from each species, so that the intensity for a bimodal system can be decomposed in the following way:

$$I(q)_{total} = I(q)_1 + I(q)_2 \quad (3)$$

$$I(q)_1 = I(q)_{total} \left(\frac{w_1}{w_1 + w_2} \right) \quad \text{or} \quad I(q)_1 = I(q)_{total} \left(\frac{a_1}{a_1 + a_2} \right) \quad (4)$$

And similarly:

$$I(q)_2 = I(q)_{total} \left(\frac{w_2}{w_1 + w_2} \right) \quad \text{or} \quad I(q)_2 = I(q)_{total} \left(\frac{a_2}{a_1 + a_2} \right) \quad (5)$$

Static Light Scattering. The same laser and set up were used for both static and dynamic light scattering. In a standard Zimm analysis, the inverse Rayleigh ratio is $R = \frac{I_s r^2}{I_{total} V}$, where I_s is the scattering intensity, I_{total} is the total intensity, r is the distance to the detector, and V is the scattering volume. The optical constant is $K = \frac{n^2 \left(\frac{\partial n}{\partial c} \right)^2}{N_A \lambda^4}$, where N_A is Avogadro's number, and $\frac{\partial n}{\partial c}$ is the change in the refractive index with concentration. For PMPC, $\frac{\partial n}{\partial c} = 0.145 \text{ g}^{-1} \text{ cm}^3$ as measured by Matsuda *et al.*

Pure Water Salt Concentrations. It must be mentioned that there is a residual salt concentration of 4E-6 M, based on conductivity measurements on our samples. The polyelectrolyte does not contribute to this salt concentration, as it is electrically neutral.

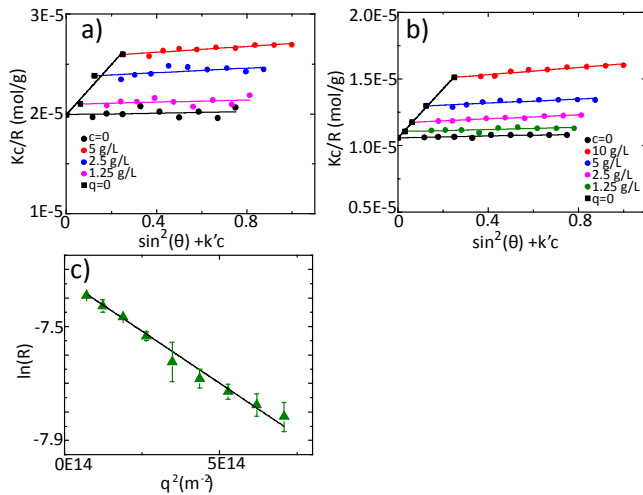


Figure S1. Static Light Scattering analysis of polyzwitterions. a) Zimm plot for PMPC₁₀₇. b) Zimm plot for PMPC 200. c) Guinier Plot for the intensity contribution from RH₂ in PMBP₁₆₄ solutions. R_g values are obtained from the slopes of the linear fits.

Table S2. R_h of PMPC₂₀₀ with a concentration of 5 g/L in water at varied NaCl salt concentrations.

NaCl Conc.	R _h (nm)
4 μM	5.07
0.75 mM	5.07
7.5 mM	5
75 mM	5
1.5 M	4.98

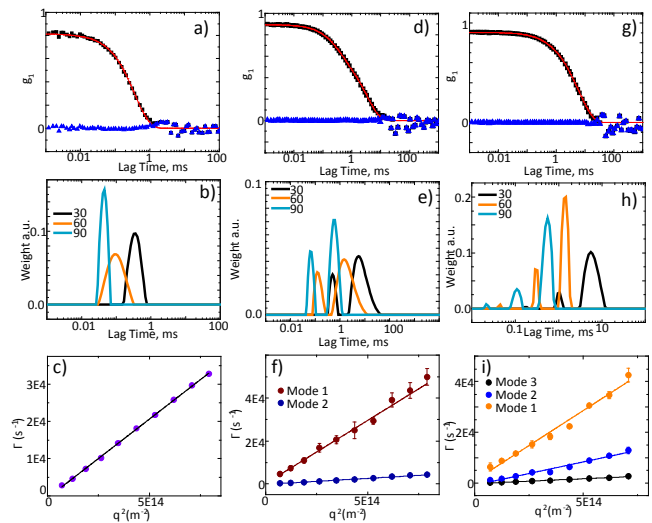


Figure S2. Dynamic light scattering analysis by CONTIN and multiple exponentials. a) Correlation function (black) g_1 , single-exponential fit (red). The fit agrees well, as shown by the residuals (blue). b) CONTIN analysis of correlation function in a) for three different angles. In agreement with exponential fitting, we observe only one distribution of time scales for each angle. c) Inverse relaxation time Γ vs q^2 is linear within the errors. From the slope, we calculate the diffusion coefficient. d) Correlation function (black) g_1 , double-exponential fit (red). The fit agrees well, as shown by the residuals (blue). e) CONTIN analysis of correlation function in d) for three different angles. In agreement with exponential fitting, we observe two distributions of time scales for each angle. f) Inverse relaxation times Γ vs q^2 are linear within the errors for both modes. From the slope, we calculate the diffusion coefficients. g) Correlation function (black) g_1 , triple-exponential fit (red). The fit agrees well, as shown by the residuals (blue). h) CONTIN analysis of correlation function in g) for three different angles. In agreement with exponential fitting, we observe three distribution of time scales for each angle. i) Inverse relaxation times Γ vs q^2 are linear within the errors for all three modes. From the slope, we calculate the diffusion coefficients.

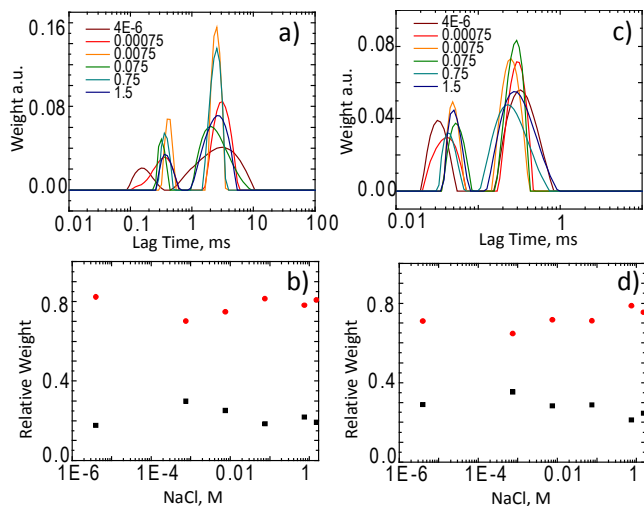


Figure S3. CONTIN relaxation time distributions of 5 g/L PMBP164 solutions as a function of NaCl concentration. a) Overlaid distributions for 30°. b) Relative weights of the modes at 30° as a function of added NaCl. The relative weights do not vary significantly. Mode 1 contributes about 70-82%, Mode 2 contributes about 18-30%. c) Overlaid distributions for 90°. d) The relative weights do not vary significantly as a function of NaCl concentration. Mode 1 contributes about 65-78%, Mode 2 contributes about 22-35%.

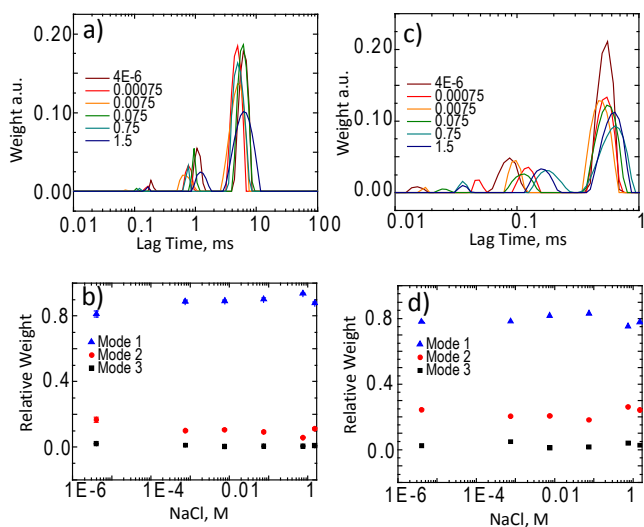


Figure S4. CONTIN relaxation time distributions of 1.25 g/L PMBP-PMPC solutions as a function of NaCl concentration. a) Overlaid distributions for 30°. b) Relative weights of the modes at 30° as a function of added NaCl. The relative weights do not vary significantly. Mode 1 contributes about 82-90%, Mode 2 contributes about 6-11%, and Mode 3 0.3-1%. The errors are standard deviations of three runs. c) Overlaid distributions for 90°. d) The relative weights do not vary significantly as a function of NaCl concentration. Mode 1 contrib-

utes about 78-83%, Mode 2 contributes about 18-26%, and Mode 3 1.2-4.0%.

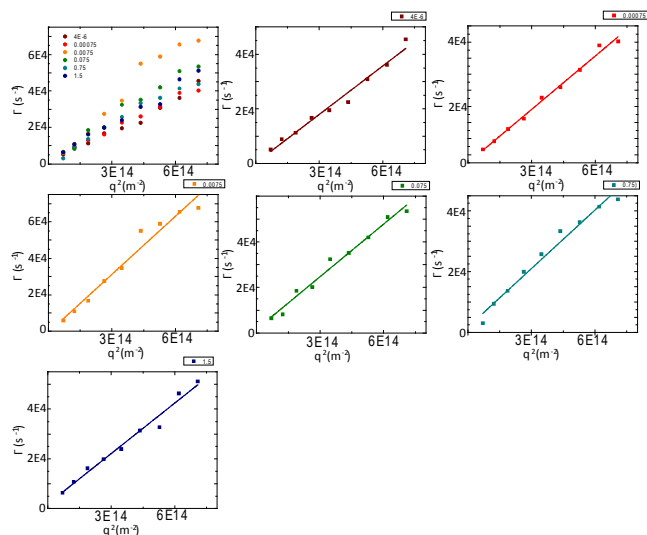


Figure S5. Fastest mode seen in PMPC-PMBP diblock solutions. Diffusion coefficient analysis as a function of NaCl. There is considerable noise in the data because the peak only contributes between 1-3% to the correlation function.