Chapter 6

Electrical Conductances of Sodium Polystyrenesulphonate in 2-Ethoxyethanol–Water Mixed Solvent Media in the Presence of Sodium Chloride at 308.15, 313.15, 318.15 and 323.15 K

6.1 Introduction

We have reported the results of conductivity measurements on salt-free solutions of sodium polystyrenesulphonate in 2-ethoxyethanol–water mixtures in Chapter 4. The addition of salts to a polyelectrolyte solution might change its conductivity behaviour considerably, and hence studies on the conductivity of polyelectrolytes in the presence of a salt might help elucidate polyion-counterion interactions in polyelectrolyte solutions with added salts.

Although the polyelectrolyte conductivities have been well understood in salt-free solutions[1] using the scaling description for the configuration of a polyion chain according to Dobrynin *et al.*,[2] the situation is quite unsatisfactory for salt-containing polyelectrolyte solutions.

Devore and Manning[3] first attempted to describe the electric transport properties of polyelectrolyte solutions containing a simple salt using the Manning counterion condensation theory[4] without much success. In view of the inadequacy of the Manning theory of the electrical transport of salt-containing polyelectrolyte solutions, a phenomenological treatment of the results of conductance experiments in terms of the additivity (commonly known as the "primitive additivity") of contributions of the polyelectrolyte and the simple salt to the total specific conductance was made.[5-9]

Traditionally, this approach takes the form of an assumed additivity of the specific conductance of the polyelectrolyte and of the salt, which gives the specific conductance (κ) of the polyelectrolyte in a salt solution through the equation,

$$\kappa = \kappa_P + \kappa_S \qquad (1)$$

where κ_P is the specific conductance of the polyelectrolyte in the absence of a simple salt and κ_S is the specific conductance of the simple salt in the absence of polyelectrolyte.

However, earlier investigations[5-9] suggest that the experimentally obtained specific conductances for salt-containing polyelectrolyte solutions do not, in general, agree with those predicted by simple additivity, Eq. (1).

Ander *et al.*[5,6] modified the 'primitive' additivity by taking into account the Debye-Hückel interactions between the polyion and the salt ions to give the polyelectrolyte specific conductance in a polyelectrolyte-salt solution as

$$\kappa = \kappa_p + \kappa_S\left(D_2 / D_2^0\right) \tag{2}$$

where D_2 and D_2^0 are the coion self-diffusion coefficients in a salt-containing polyelectrolyte solution and in an infinitely dilute polyelectrolyte-free salt solution, respectively. The ratio of self-diffusion coefficients D_2 / D_2^0 has been used as a quantitative measure[4] of the effective interaction of uncondensed small ions in the presence of the polyelectrolyte, and hence the effective specific conductance of the added simple salt would be $\kappa_s\left(D_2 / D_2^0\right)$.

Although the "modified" additivity has been shown to be somewhat better than the "primitive" one, departures from the experimental results are still prominent.[5-9] Later Bordi *et al.*[10] evaluated equivalent conductances for a hydrophilic polyion in presence of a salt in the light of the scaling approach[2] and compared with the experimental values. The agreement is rather good although a quantitative description is still awaiting.

In this chapter, a simple model is introduced to analyze the conductivity of polyelectrolyte in the presence of an added electrolyte based on scaling theory for the conductivity of polyelectrolyte solutions.[1] This model has been extensively tested with data on sodium polystyrenesulphonate in the presence of sodium chloride in 2-ethoxyethanol–water mixed solvent media at different temperatures. The data set used here considers a number of parameters *e.g.*, relative permittivity of the medium, temperature and concentration of the added salt. Moreover, three decades of concentration of the polyelectrolyte were covered. Very good quantitative agreement with only one adjustable parameter has been observed.

6.2 Theory

Here we introduce a simple equation for describing the conductivity behaviour of polyelectrolyte in salt solutions following the model for the electrical conductivity of solutions of polyelectrolytes without salt proposed by Colby *et al.*[1] using the scaling description for the configuration of a polyion chain according to Dobrynin *et al.*[2]

In semidilute solutions, the polyion chain is modeled as a random walk of N_ξ correlation blobs of size ξ_0, each of them containing g monomers. Each blob bears an electric charge $q_\xi = zefg$ (z being the counterion valence and e is the electronic charge) and the complete chain, of contour length $L = N_\xi \xi_0$, bears a charge $Q_p = N_\xi q_\xi = zefgN_\xi$, where f is the fraction of uncondensed counterions. Due to the strong electrostatic interactions within each correlation blob, the chain is a fully extended conformation of g_e electrostatic blobs of size ξ_e. This means that for length scales less than ξ_0, the electrostatic interactions dominate (and the chain is a fully extended conformation of electrostatic blobs of size ξ_e), and for length scales greater than ξ_0, the electrostatic interactions are screened and the chain is a random walk of correlation blobs of size ξ_0.

Following Colby *et al.*,[1] the specific conductivity of a salt-free polyelectrolyte solution (κ_p) is given by

$$\kappa_p = fc_p\left[\lambda_c^0 + \frac{c_p \xi_0^2 e^2 f}{3\pi\eta_0}\ln\left(\frac{\xi_0}{\xi_e}\right)\right] \tag{3}$$

where c_p is the number density of monomers, λ_c^0 the limiting equivalent conductivity of the counterions and η_0 the coefficient of the viscosity of the medium.

The interactions between the polyion and the counterions will be modified in the presence of an electrolyte and this will result in a different level of counterion condensation, *i.e.*, in a different value of f (designated as f'). The effective specific conductivity due to the polyelectrolyte in the presence of a simple salt can be expressed as

$$\kappa_{p(eff)} = f'c_p\left[\lambda_c^0 + \frac{c_p \xi_0^2 e^2 f'}{3\pi\eta_0}\ln\left(\frac{\xi_0}{\xi_e}\right)\right] \tag{4}$$

Thus the total specific conductivity (κ) of a polyelectrolyte solution with added simple electrolyte should be equal to the sum of the specific conductivity of the simple salt in the absence of a polyelectrolyte and the effective specific conductivity due to the polyelectrolyte in the presence of a simple salt and is given by

$$\kappa = \kappa_S + f'c_p\left[\lambda_c^0 + \frac{c_p \xi_0^2 e^2 f'}{3\pi\eta_0}\ln\left(\frac{\xi_0}{\xi_e}\right)\right] \tag{5}$$

This equation, Eq. (5), has one adjustable parameter f' and this could be obtained by the method of a least squares fit of the experimental specific conductivity of the polyelectrolyte solution (κ) in presence of a salt to Eq. (5) using the measured specific conductance (κ_s) of the salt in the absence of the polyelectrolyte. This value of f' takes care of the changed polyion-counterion interactions under the influence of the added salt. The second term in Eq. (5) is the actual contribution of the polyelectrolyte species towards the total specific conductivity in the presence of an added salt.

6.3 Experimental

2-Ethoxyethanol (G. R. E. Merck) was distilled with phosphorous pentoxide and then redistilled over calcium hydride. The purified solvent had a density of 0.92497 $g \cdot cm^{-3}$ and a co-efficient of viscosity of 1.8277 $mPa \cdot s$ at 298.15 K; these values are in good agreement with literature values.[11] Triply distilled water with a specific conductance of *ca.* 10^{-6} $S \cdot cm^{-1}$ at 308.15 K was used for the preparation of the mixed solvents. The relative permittivities of 2-ethoxyethanol–water mixtures at the experimental temperatures were obtained with the equations as described in the literature[12] using the literature density and relative permittivity data of the pure solvents[11,13] and the densities of the mixed solvents are given in Table 4.1(Chapter 4).

The sodium salt of polystyrenesulphonic acid employed in these investigations was purchased from the Aldrich Chemical Company. The average molecular weight of the sample was *ca.* 70000 and a degree of sulphonation of 1.0 and it was purified by dialysis.[14,15] The molecular weight reported by the manufacturer agreed well with that determined in the present study obtained in presence of 0.05 M sodium chloride (NaCl) at 298.15 K using the Mark-Houwink relationship,[16] $[\eta] = 1.39\text{x}10^{-4} M^{0.72}$, where $[\eta]$ is the intrinsic viscosity, and M is the average molecular weight. The absorption coefficient of the sodium polystyrenesulfonate solutions used at 261 nm, which is considered to be a characteristic indicator of the sample purity,[15] is found to be 400 $dm^3 \cdot cm^{-1} \cdot mol^{-1}$. Spectroscopic examination of the polyelectrolyte sample using this criterion was employed periodically to substantiate the sample purity. Sodium chloride (Fluka) was of puriss grade. This was dried *in vacuo* for a prolonged period immediately before use and was used without further purification.

Conductance measurements were carried out on a Pye-Unicam PW 9509 conductivity meter at a frequency of 2000 Hz using a dip-type cell with a cell constant of 1.15 cm^{-1} and having an uncertainty of 0.01 %. The cell was calibrated by the method of Lind and co-workers[17] using aqueous potassium chloride solution. The measurements were made in a water bath maintained within ± 0.01 K of the desired temperature. The details of the experimental procedure have been described earlier.[18,19] Several independent solutions were prepared and runs were performed to ensure the reproducibility of the results.

In order to avoid moisture pickup, all solutions were prepared in a dehumidified room with utmost care. In all cases, the experiments were performed at least in three replicates. The experimental uncertainties in density, viscosity, and conductivity were always within 0.02 %, 0.80 %, and 0.03 %, respectively.

6.4 Results and Discussion

The experimental values of specific conductivities (κ) of sodium polystyrenesulphonate in the presence of varying concentrations sodium chloride in 2-ethoxyethanol–water mixtures have been listed as a function of the equivalent polyelectrolyte concentration (c_p) at 308.15, 313.15, 318.15 and 323.15 K in Table 5.1. The specific conductivities of sodium polystyrenesulphonate as a function of the polymer concentration at given temperatures and solvent compositions in 2-ethoxyethanol–water mixtures with varying amounts of added NaCl are shown in the Figures 6.1 to 6.12. Figures 6.13 and 6.14, on the other hand, demonstrate the influence of temperature on the specific conductivity versus the polymer concentration profiles. From these figures and also from Table 6.1, it is apparent that for all the solutions studied the specific conductivities increase with polyelectrolyte concentration. Addition of salt increases the specific condutances of the polyelectrolyte-salt solutions as expected. The specific conductances of the polyelectrolyte-salt systems are, in general, found to decrease with increasing amount of 2-ethoxyethanol in the mixed solvent media (shown in the representative Figure 6.15).

Since the present 2-ethoxyethanol–water mixtures are poor solvents for the uncharged polymer polystyrene, the electrostatic blob is collapsed into a dense globule, we use a value of 5 A^0 as the effective monomer size (b) as suggested by Colby *et al.*[1]

Under poor solvent conditions, the electrostatic blob size (ξ_e) and the correlation blob size (ξ_0) are given by[1]

$$\xi_e = b(f^{*2}\xi)^{-1/3} \quad (6)$$

$$\xi_0 = (cb)^{-1/2}(f^{*2}\xi)^{-1/3} \quad (7)$$

The specific conductivity values of the polyelectrolyte-salt system as a function of polyelectrolyte concentration in a given solvent medium at a given temperature and for a given salt concentration were fitted to Eq. (5) by the method of least-squares analysis. The best-fitted f' values along with the standard deviations are reported in Table 6.1. In the Figures 6.1 to 6.12 we compare the calculated specific conductivities using the f' values obtained in the semidilute regime (reported in Table 6.1) with those obtained experimentally. From the standard deviations recorded in Table 6.1, as well as from an inspection of these figures, it is directly evident that the present method of analysis reproduced the experimental results even in dilute solutions quite satisfactorily. It should be noted that this vigorous test of the proposed model has been performed with 36 sets of data considering the effect of medium, temperature and concentration of the added simple salt. We have, thus, been able to develop a simple model in describing the specific conductivity behaviour of polyelectrolyte-solutions with added salt.

6.5 Conclusions

The electrical conductances of the solutions of sodium polystyrenesulphonate in 2-ethoxyethanol–water mixed solvent media containing 25, 40 and 50 mass percent of 2-ethoxyethanol have been reported at 308.15, 313.15, 318.15 and 323.15 K in the presence of sodium chloride. The conductance data have been analyzed on the basis of a simple equation with only one adjustable parameter developed in the present study following the model for the electrical conductivity of solutions of semidilute polyelectrolytes without salt proposed by Colby *et al.*[1] using the scaling description for the configuration of a polyion chain according to Dobrynin *et al.*[2] Excellent agreement between the experimental results and those obtained using Eq. (5) has always been observed. We expect that the model proposed here provides a universal description of the specific conductivities of polyelectrolyte solutions in the presence of an added electrolyte.

6.6 References

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Table 6.1 Fraction of Uncondensed Counterions (f') and the Standard Deviations (σ) of Fit for Sodium Polystyrenesulphonate in the Presence of NaCl at Temperatures (308.15, 313.15, 318.15 and 323.15) K in 2-Ethoxyethanol–Water Mixed Solvent Media as Obtained According to Eq. (5)

T (K)	Mass % of Cosolvent	c_{NaCl} ($mol \cdot L^{-1}$)	f'	$\sigma \times 10^6$
308.15	25	1×10^{-4}	0.40	3.09
		1×10^{-3}	0.38	4.59
		1×10^{-2}	0.34	10.06
	40	1×10^{-4}	0.25	1.14
		1×10^{-3}	0.23	1.82
		1×10^{-2}	0.21	2.37
	50	1×10^{-4}	0.25	0.74
		1×10^{-3}	0.24	1.52
		1×10^{-2}	0.21	2.91
313.15	25	1×10^{-4}	0.40	5.19
		1×10^{-3}	0.35	6.29
		1×10^{-2}	0.33	6.82
	40	1×10^{-4}	0.23	3.40
		1×10^{-3}	0.21	4.01
		1×10^{-2}	0.18	3.77
	50	1×10^{-4}	0.25	1.13
		1×10^{-3}	0.23	1.65
		1×10^{-2}	0.18	5.60
318.15	25	1×10^{-4}	0.38	3.84
		1×10^{-3}	0.35	4.26
		1×10^{-2}	0.32	5.67
	40	1×10^{-4}	0.25	2.45
		1×10^{-3}	0.22	3.62
		1×10^{-2}	0.17	8.48
	50	1×10^{-4}	0.23	1.69
		1×10^{-3}	0.21	2.76
		1×10^{-2}	0.18	5.08
323.15	25	1×10^{-4}	0.34	6.33
		1×10^{-3}	0.31	7.65

Table 6.1 (Continued)

T (K)	Mass % of Cosolvent	c_{NaCl} $(mol \cdot L^{-1})$	f'	$\sigma \times 10^6$
		1×10^{-2}	0.28	7.57
	40	1×10^{-4}	0.24	4.87
		1×10^{-3}	0.22	5.61
		1×10^{-2}	0.20	7.69
	50	1×10^{-4}	0.23	2.32
		1×10^{-3}	0.22	3.90
		1×10^{-2}	0.19	5.10

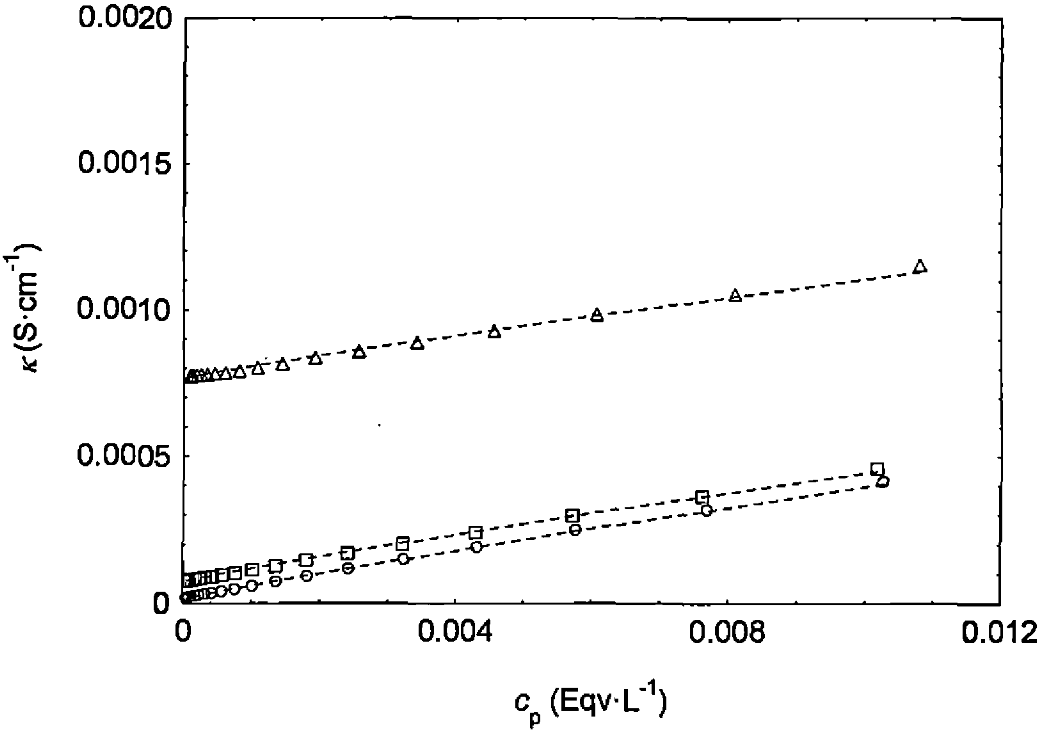

Figure 6.1 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 308.15 K in a 2-ethoxyethanol–water mixture with 25 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

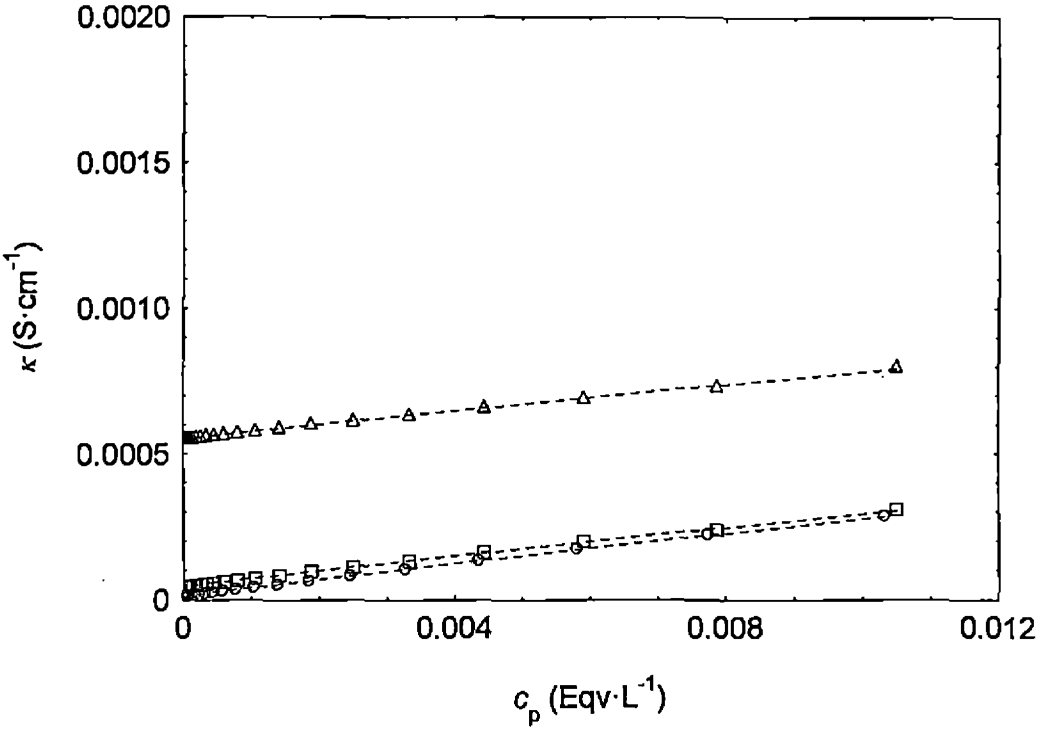

Figure 6.2 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 308.15 K in a 2-ethoxyethanol–water mixture with 40 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol \cdot L^{-1}$ NaCl respectively (see text).

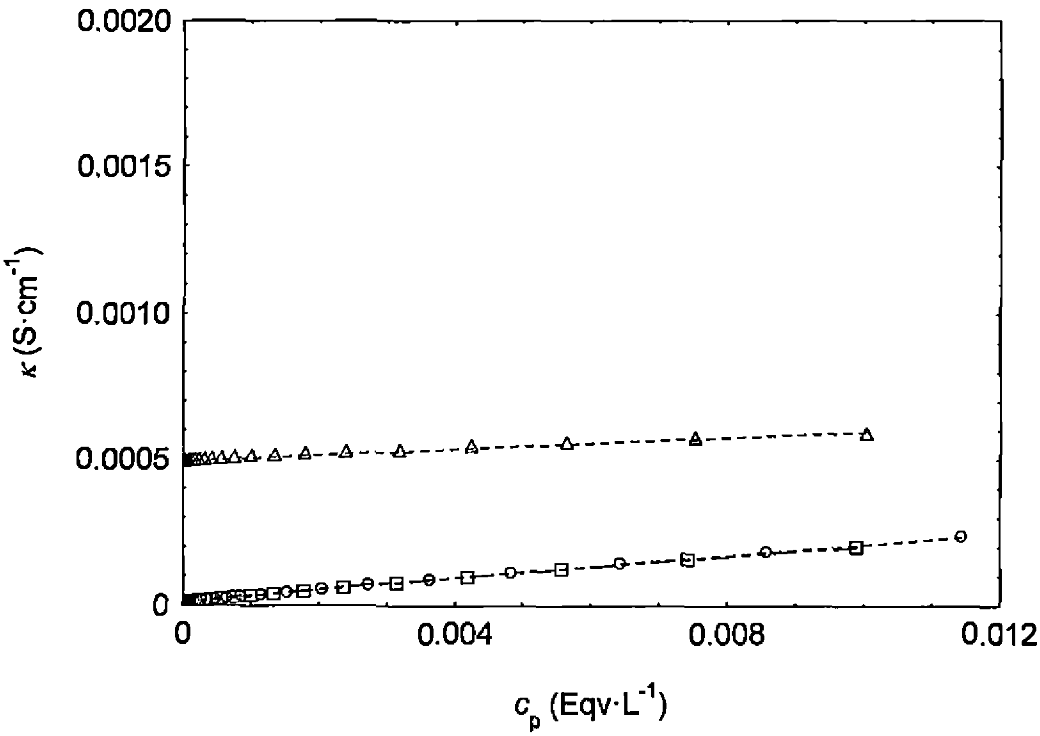

Figure 6.3 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 308.15 K in a 2-ethoxyethanol–water mixture with 50 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol \cdot L^{-1}$ NaCl respectively (see text).

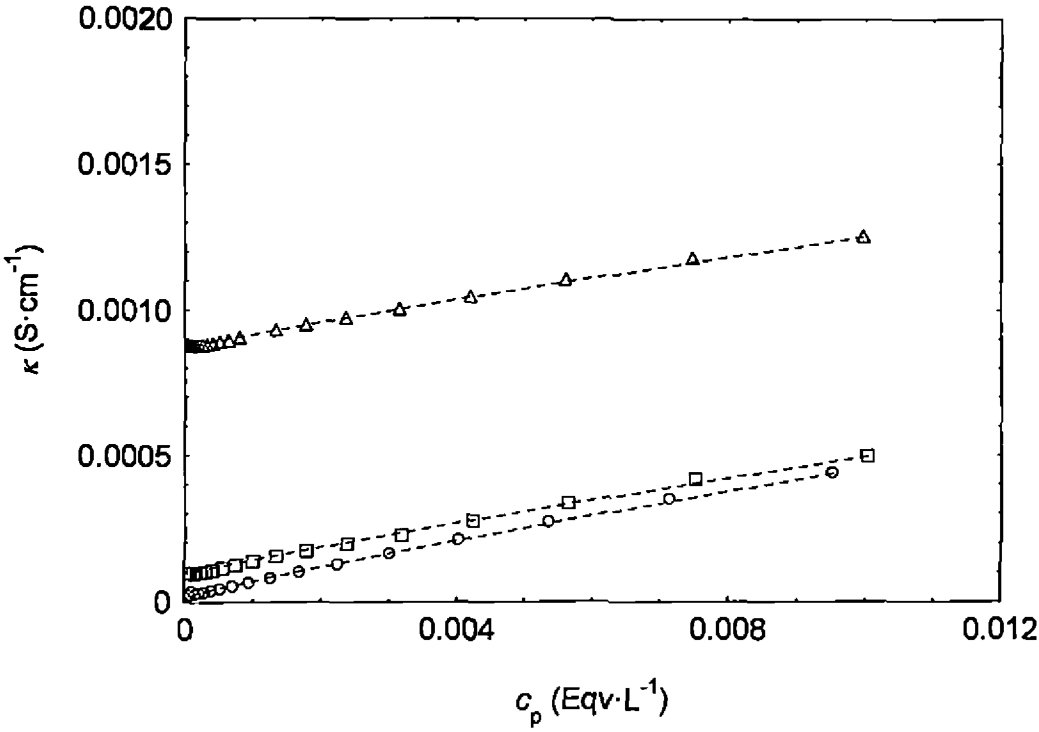

Figure 6.4 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 313.15 K in a 2-ethoxyethanol–water mixture with 25 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

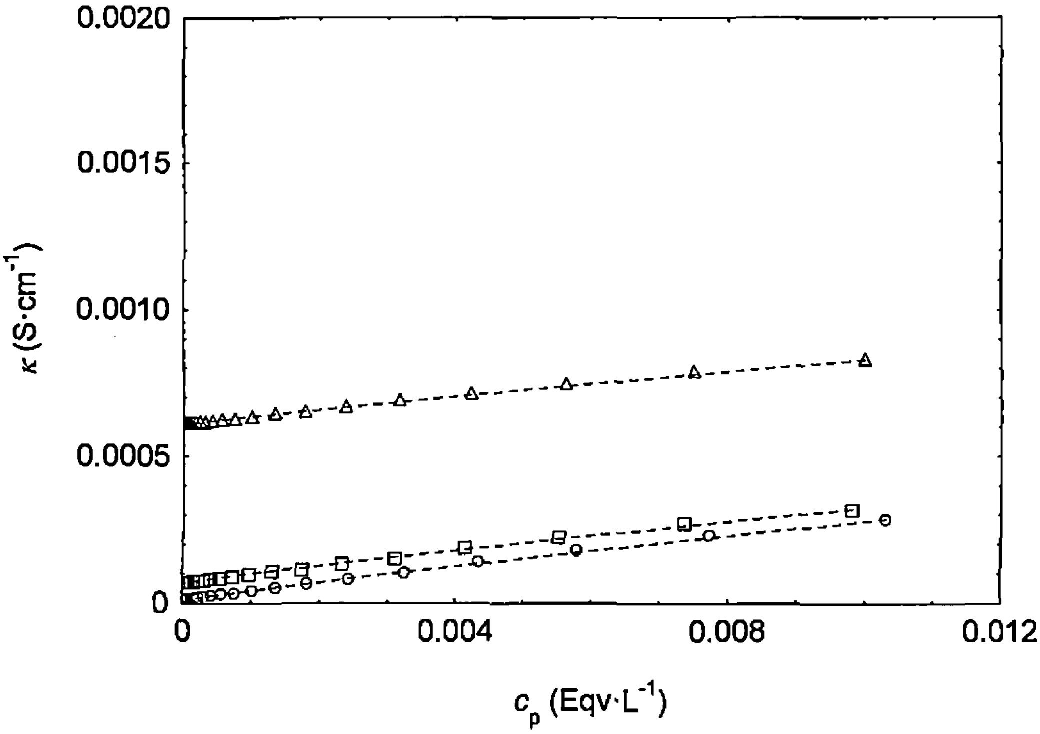

Figure 6.5 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the *polymer concentration* (c_p) *at 313.15* K *in a* 2-ethoxyethanol–water mixture with 40 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol \cdot L^{-1}$ NaCl respectively (see text).

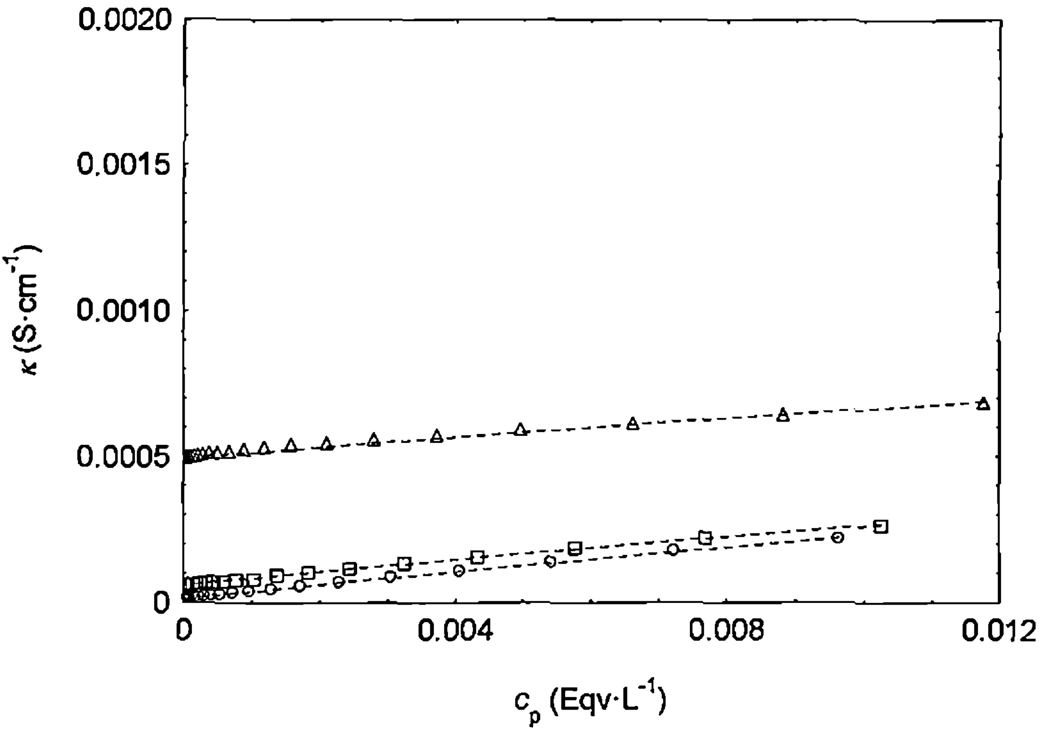

Figure 6.6 Specific conductivities (κ) of sodium polystyresulphonate as a function of the polymer concentration (c_p) at 313.15 K in a 2-ethoxyethanol–water mixture with 50 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

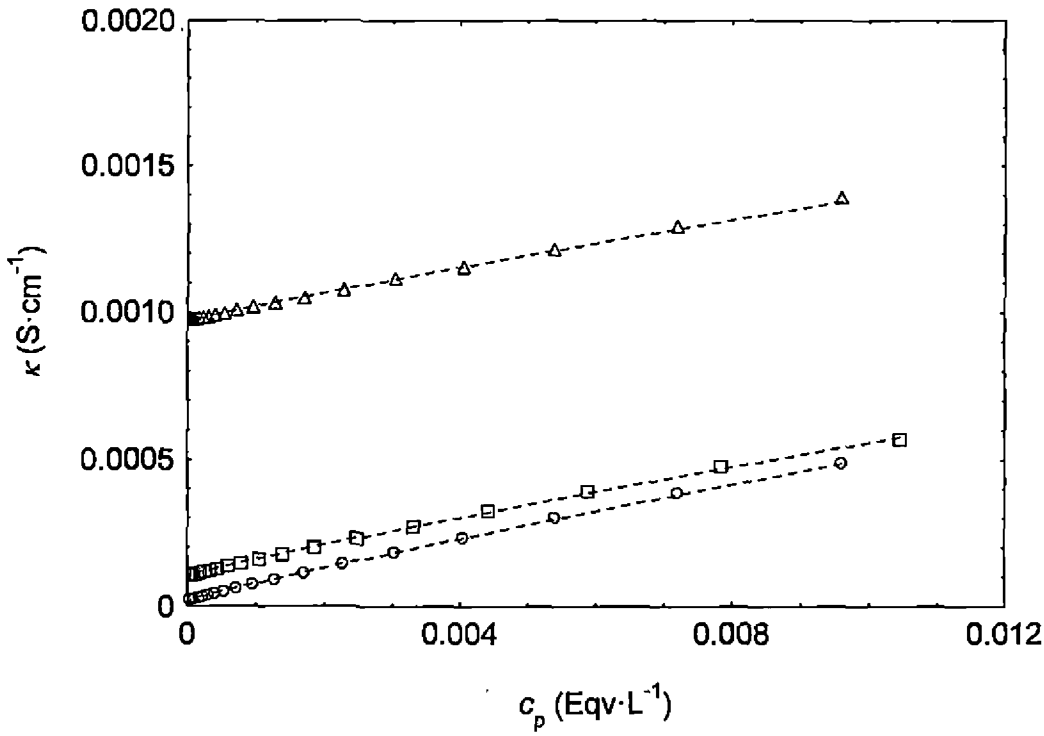

Figure 6.7 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 318.15 K in a 2-ethoxyethanol–water mixture with 25 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

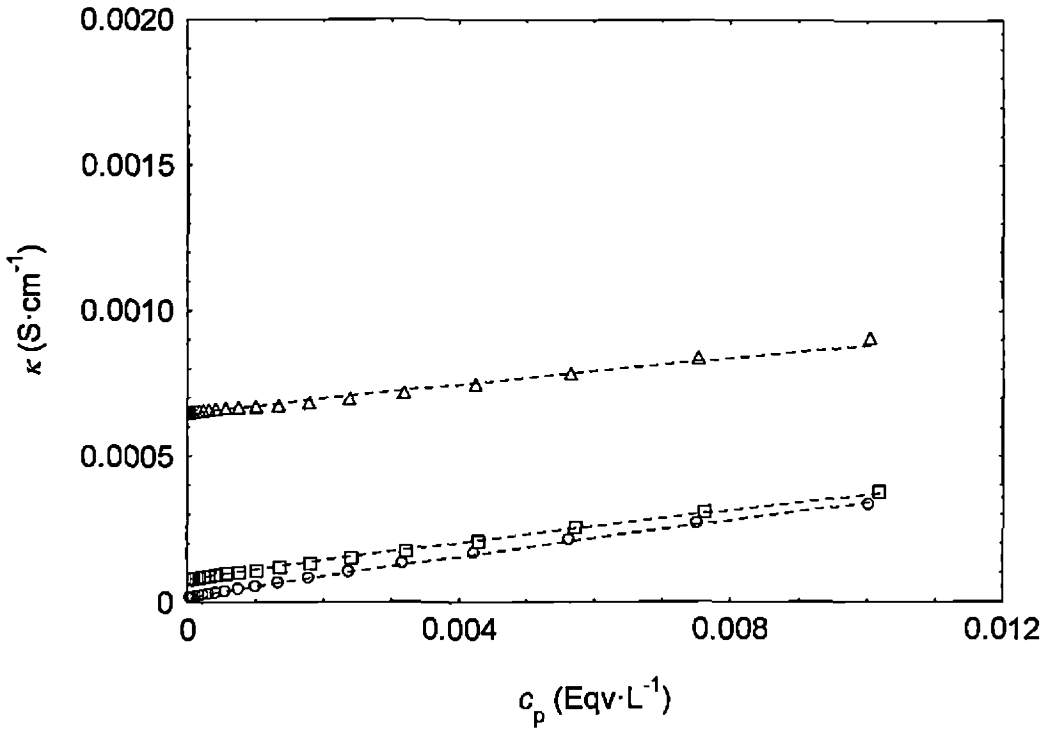

Figure 6.8 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 318.15 K in a 2-ethoxyethanol–water mixture with 40 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol \cdot L^{-1}$ NaCl respectively (see text).

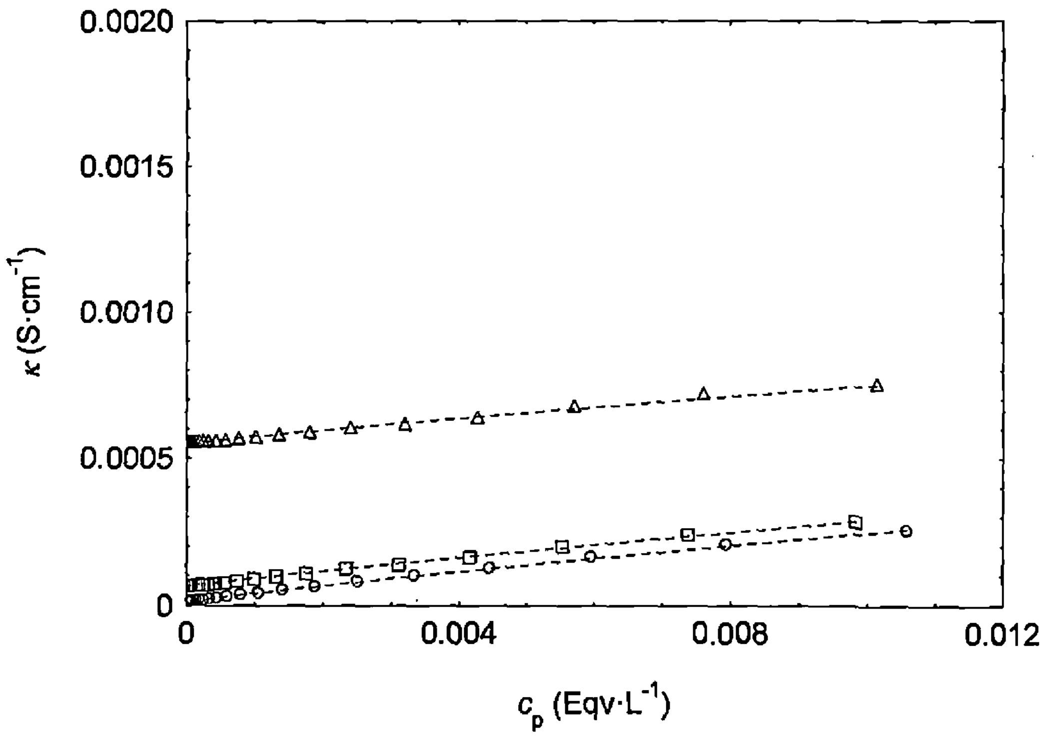

Figure 6.9 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 318.15 K in a 2-ethoxyethanol–water mixture with 50 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

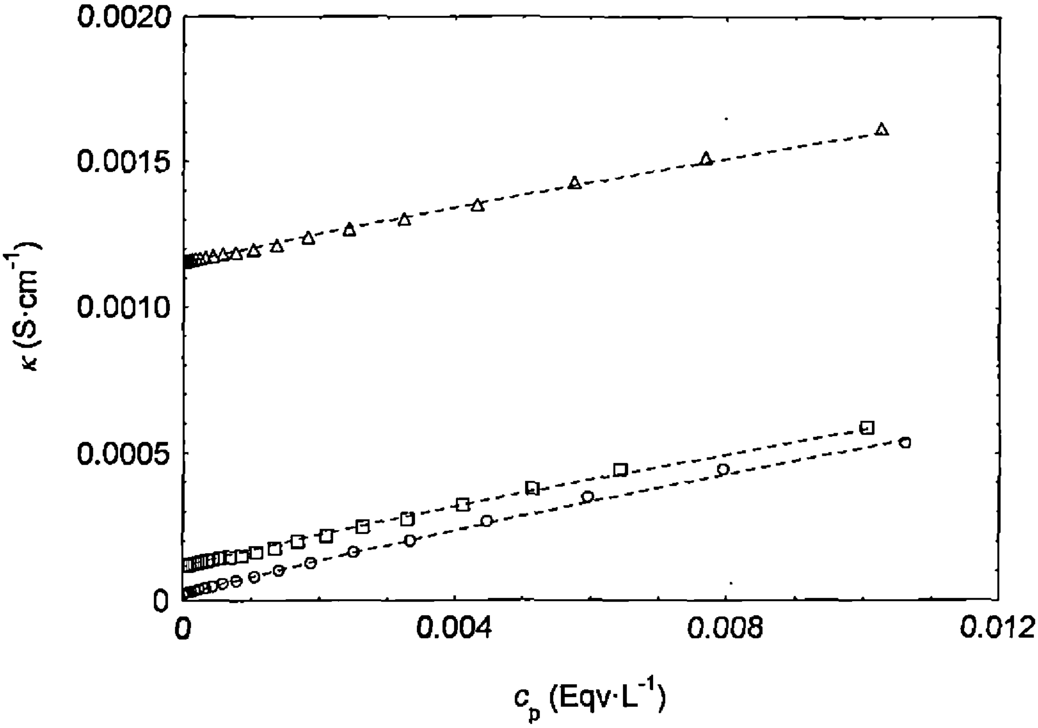

Figure 6.10 Specific conductivities (κ) of sodium polystyresulphonate as a function of the polymer concentration (c_p) at 323.15 K in a 2-ethoxyethanol–water mixture with 25 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

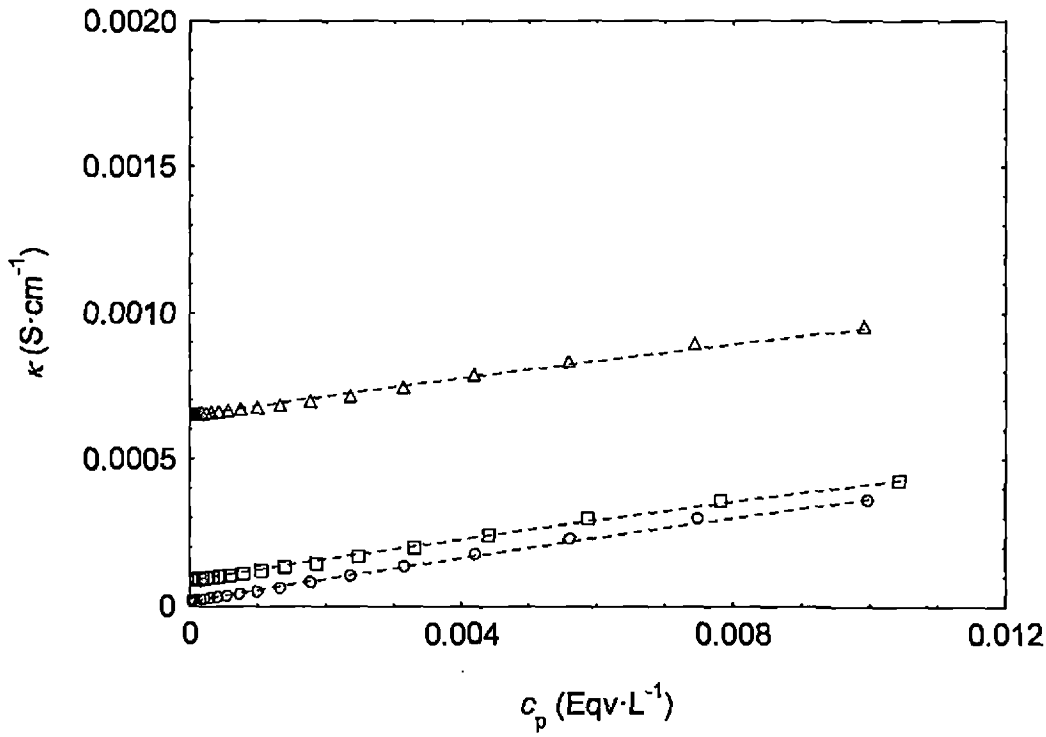

Figure 6.11 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 323.15 K in a 2-ethoxyethanol–water mixture with 40 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

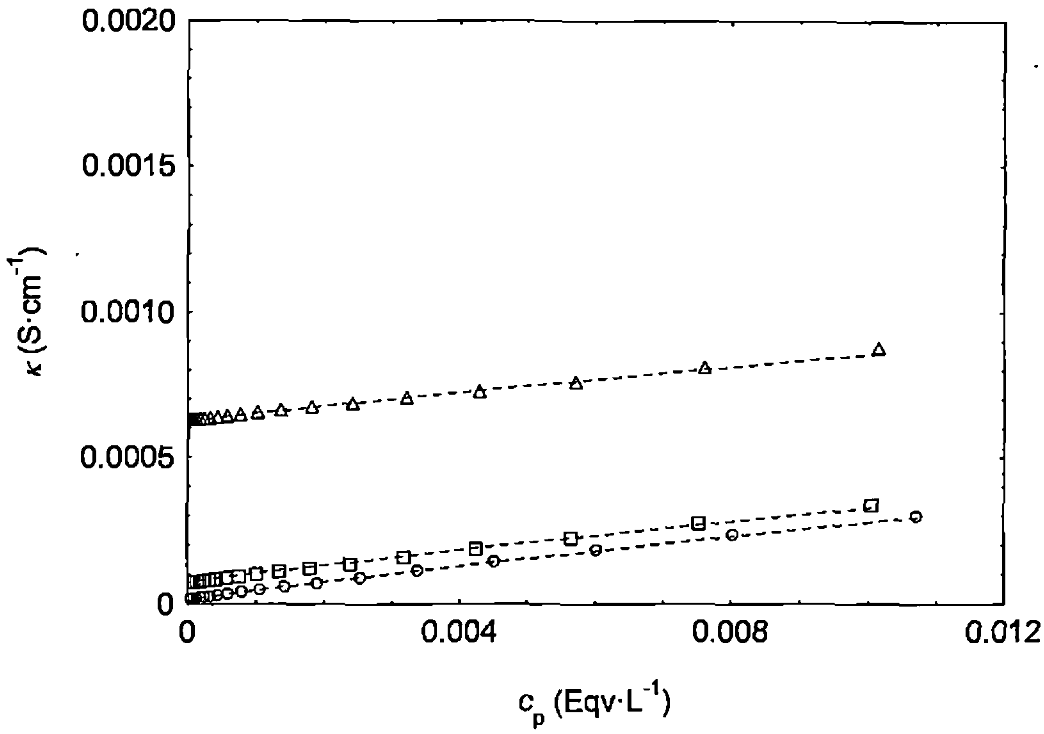

Figure 6.12 Specific conductivities (κ) of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at 323.15 K in a 2-ethoxyethanol–water mixture with 50 mass percent of 2-ethoxyethanol. Experimental: Open symbols represent experimental values whereas the dashed lines are according to Eq. (5). Circles, squares and triangles represent the polyelectrolyte solutions in presence of (1×10^{-4}, 1×10^{-3}, and 1×10^{-2}) $mol\cdot L^{-1}$ NaCl respectively (see text).

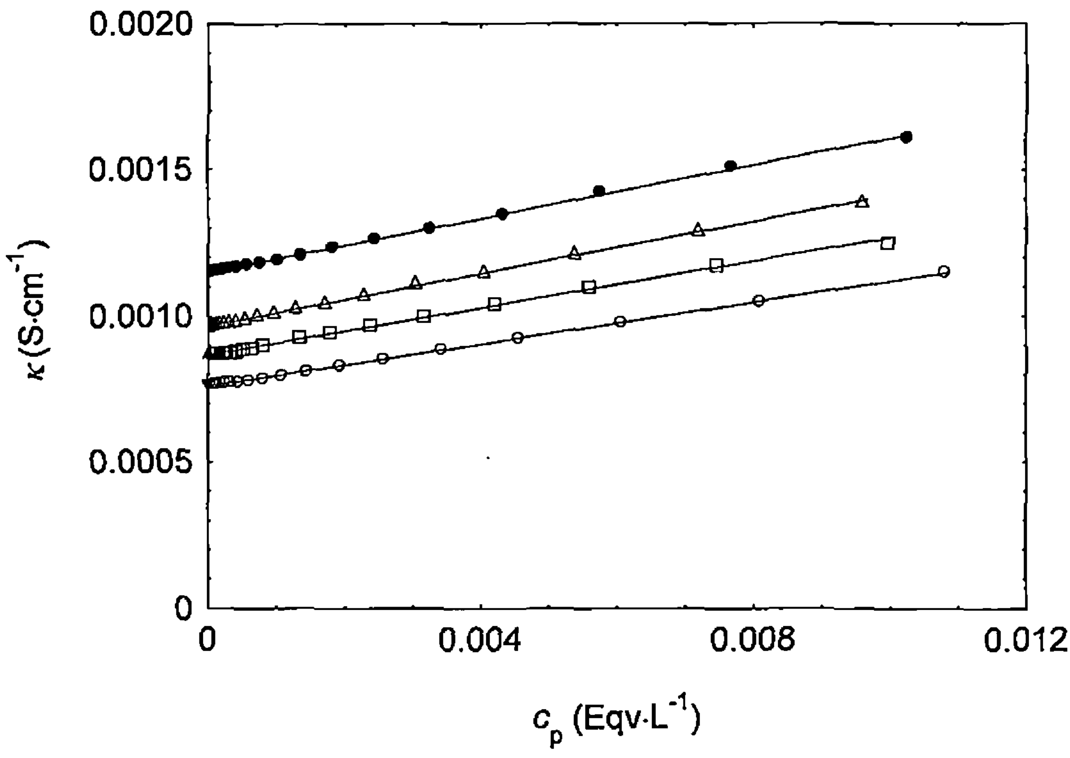

Figure 6.13 Specific conductivities of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at temperatures 308.15 K (○), 313.15 K (□), 318.15 K (Δ) and 323.15 K (•) in a 2-ethoxyethanol–water mixture with 25 mass percent of 2-ethoxyethanol in the presence of 1×10^{-2} mol · L^{-1} NaCl. Lines are used to guide the eye.

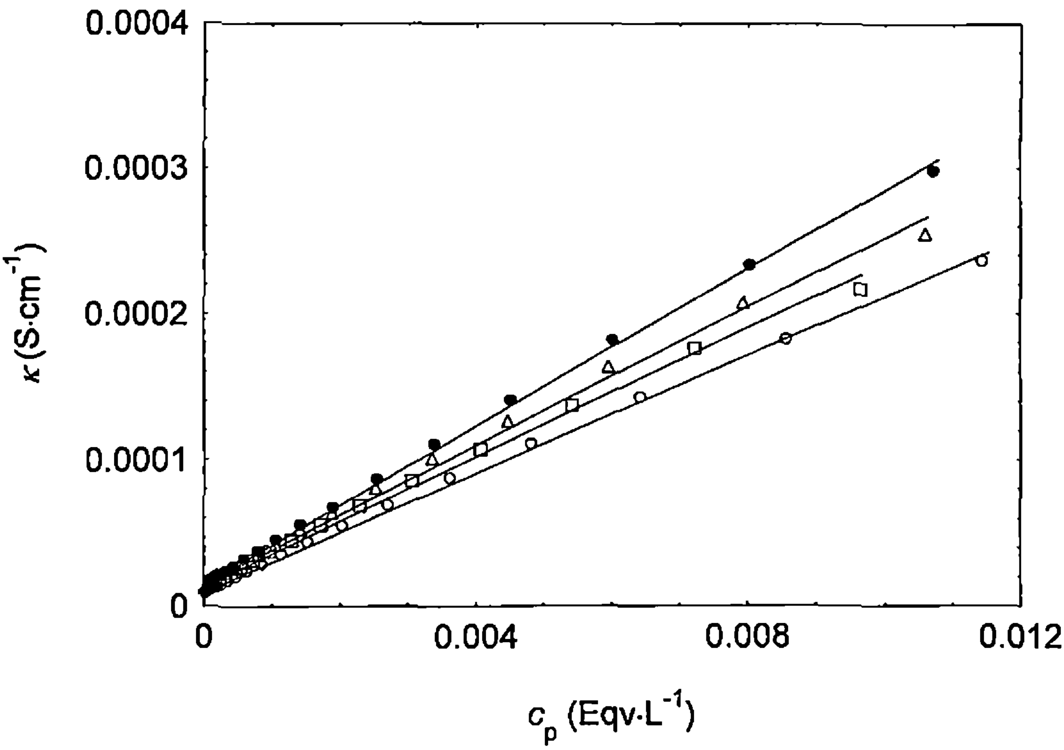

Figure 6.14 Specific conductivities of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at temperatures 308.15 K (○), 313.15 K (□), 318.15 K (Δ) and 323.15 K (•) in a 2-ethoxyethanol–water mixture with 50 mass percent of 2-ethoxyethanol in the presence of 1×10^{-4} mol·L^{-1} NaCl. Lines are used to guide the eye.

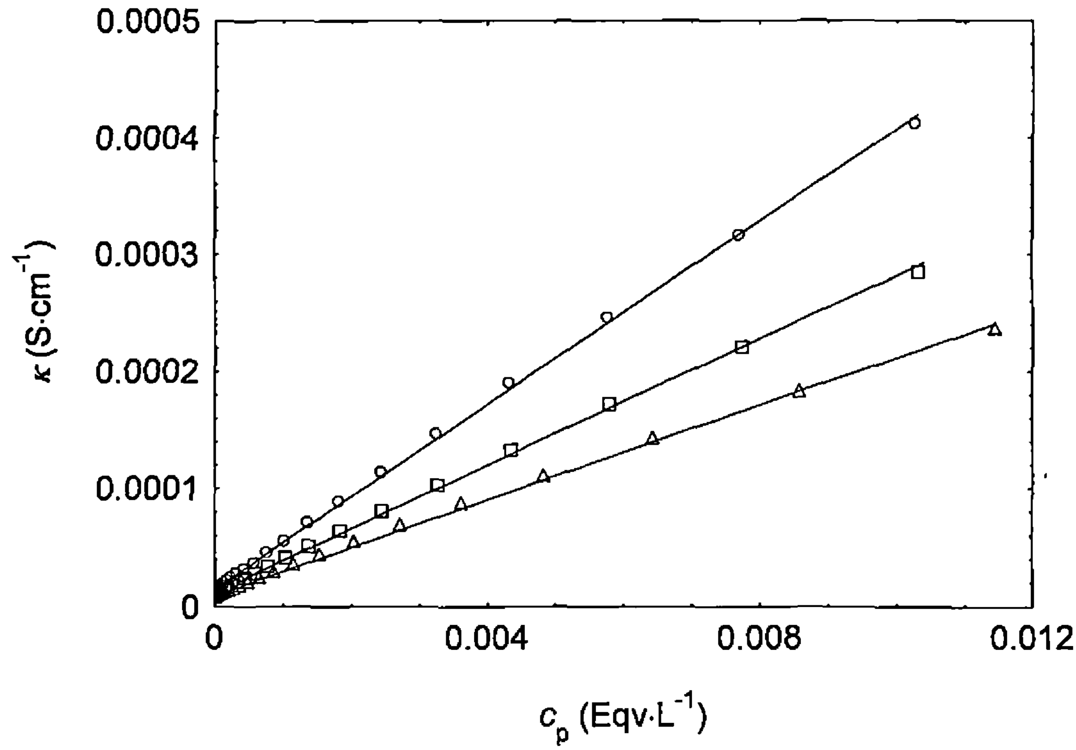

Figure 6.15 Specific conductivities of sodium polystyrenesulphonate as a function of the polymer concentration (c_p) at a temperature of 308.15 K in a 2-ethoxyethanol–water mixture with 25 (○), 40 (□) and 50 (Δ) mass percent of 2-ethoxyethanol in the presence of 1×10^{-4} mol·L^{-1} NaCl. Lines are used to guide the eye.