Small-Angle Scattering Studies of Sodium Dodecyl Sulfate Interactions with Gelatin. 1

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The structural perturbations that result from the interactions between the anionic surfactant sodium dodecyl sulfate and the biopolymer gelatin have been studied using small-angle neutron scattering. In particular, contrast variation has been used to highlight the various components in the mixture. We discuss these data in terms of structural changes associated with the individual components and their interactions with each other. The effect of ionic strength on these structures is also considered.

Introduction

The formation of complexes between anionic surfactants and proteins in aqueous solutions have been well established. In this paper we shall be concerned with the structure of the complexes formed by sodium dodecyl sulfate (SDS) and gelatin. This system has been studied extensively by precipitation,[1] surface tension,[2,3] and viscosity measurements.[4,5] With increasing concentration of surfactant, various different regimes have been identified and interpreted in terms of the degree of association of the surfactant with the biopolymer. The structure of these complexes however is not known.

Gelatin is a biopolymer obtained from the denaturation of collagen. Native collagen consists of rods formed from triple helices which are held together by covalent bonds and during the denaturation process these bonds are destroyed and the helical nature is disrupted. Above 37 °C, gelatin solutions are liquids and the molecules are essentially random coils. When cooled to room temperature, however, solutions exceeding 1% by weight of gelatin change into gels and the chains partially revert back to the triple helix structure. Around neutral pH the gelatin chain is amphoteric and certain groups (e.g., lysine and argenine ≈ 7.5% of residues) will be positively charged, providing the possibility of electrostatic binding with SDS to the polar head group, while others (e.g., glutamic and aspartic acid ≈ 11.8%) will be negatively charged. Other residues are strongly hydrophobic (leucine, isoleucine, methionine, and valine ≈ 6.2%) and NMR results[6] suggest strongly that these sites provide the opportunity for hydrophobic bonding to the SDS apolar tail. The bulk of the gelatin chain is made up of glycine, proline, and hydroxyproline (≈58%), which are weakly hydrophobic, and it is their periodicity along the chain that provides the vehicle for helix formation.

Pezron *et al.*[7] have studied the sol state of gelatin (at 50 °C) in the presence of 0.1 M NaCl using light, small-angle neutron, and X-ray scattering. Their results indicate that in the dilute regime, where the chains are effectively isolated, the persistence length or stiffness of the chain is ≈20 Å and the radius of gyration R_G is ≈350 Å. In the semidilute regime the small-angle scattering could be described by a two-state model in which two length scales were identified. These were a screening length ξ of $\approx R_G/10$ and an inhomogeneity size of $\approx R_G/3$. However, in that study, an extended low Q range (1×10^{-3} Å^{-1}) was available making it possible to see the larger scale structure. A schematic illustration of this structure is given in Figure 1. They also found that the screening length followed a power law of the form $\xi \sim c^{\nu}$ where c is concentration and ν was in the range -0.5 to -0.7, consistent with scaling theory for homopolymers.[8]

Surface tension measurements on gelatin-SDS solutions[2,3] indicate two critical concentrations which lie on either side of the cmc (critical micelle concentration) of pure SDS. The lower concentration break (cmc1) can be nearly an order of magnitude below that of the cmc of pure SDS and has been attributed to induced micellization brought about by a condensation of the surfactant onto the gelatin backbone, *i.e.* micelles bound to the gelatin. This transition is independent of the gelatin concentration. The second transition (cmc2) corresponds to the formation of nonadsorbed micelles. This transition occurs on saturation of the gelatin with micelles and therefore increases with increased gelatin concentration.

The most obvious effect of adding SDS to gelatin solutions, at concentrations greater than cmc1, is a substantial increase in viscosity.[4,5] This increase in viscosity was associated with an increase in the high-frequency modulus, but the characteristic relaxation time was independent of both surfactant and gelatin concentration for a wide range of viscosities and concentrations.

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The viscosity rise would be consistent with the micelles acting as transient cross-linking sites or nodes for the gelatin. However, viscosity is a macroscopic property reflecting relaxation processes in the bulk. It does not *per se* provide direct structural information. Similarly, in other polymer surfactant systems, binding isotherms cannot be simply interpreted in terms of molecular structure, as recent results have shown that virtually identical binding isotherms can give very different viscosities.[9]

The main aim of this paper is to investigate the binding of SDS to gelatin directly, using small-angle neutron scattering (SANS) which can give detailed structural information on a molecular scale. The effects of surfactant concentration and ionic strength on the structure of these systems will be discussed and contrast variation methods will be used to enable the individual components of these mixtures to be studied independently.

Small-Angle Scattering Analysis

Small-angle neutron scattering is an ideal tool with which to study structures in the size range of 5 to 300 Å, and gelatin and SDS together with the complexes they form fall in this range. An advantage of using neutrons in these systems is the ability to suppress selectively the scattering from either component by adjusting their scattering length densities relative to the solvent. For example, the scattering of deuterated SDS (D-SDS) in D_2O is very weak whereas that from gelatin (protonated) is rather strong. (See Table 1.) To interpret scattering data from these systems necessitates the use of structural models, and we shall describe these briefly.

A general two-shell model has been rather successful in many cases for interpreting the scattering from micelles.[10] However, recent improvements of this model which incorporate polydispersity or asymmetry,[11,12] have led to a more realistic description of the structure, at least for SDS. Surface roughness and nonsphericity of the micelles can be incorporated into the two shell model straightforwardly. In this account we shall use a monodisperse elliptic model[13] as it is known that SDS forms rodlike micelles at higher concentrations and it is not unreasonable to assume that there is an ellipsoidal intermediate between spherical and rod structures.

In general, for interacting systems of globular charged micelles, the neutron scattering intensity can be described in terms of an intermicellar structure factor $S'(Q)$ and a shape factor $P(Q)$.[8]

$$I(Q) = N_p P(Q) S'(Q) \quad (1)$$

Q is the momentum transfer vector ($4\pi \sin(\theta/2)\lambda$, where θ is the scattering angle and λ the neutron wavelength) and N_p is the micellar number density. $P(Q) = \langle|F(Q)|^2\rangle$ where $F(Q)$ is the form factor and the brackets represent a spatial average. The $F(Q)$ for monodisperse ellipsoids is identical to that for a sphere, but with an equivalent radius which varies with orientation.[13] $S'(Q)$ is a modified structure factor which can account for the nonsphericity of the micelles and represents an average taken over both the size distribution and orientation.[13] The structure factor is calculated for a given micellar charge and ionic screening using the mean spherical approximation given by Hayter and Penfold,[14] with modifications for low volume fractions and a penetrating background.[15] The model assumes an elliptical core-shell structure in which the core contains, say, 90% of the surfactant hydrocarbon tails. The axial ratio is adjusted so as to keep the minimum core radius less than the fully-stretched length of the hydrocarbon chain (ca. 16.7 Å). This avoids a "hole" at the center of the micelle, apparent in earlier spherical models, and allows a smooth transition to rodlike structures seen with higher concentrations of SDS or salt. The 4–5 Å thick "shell", which makes a significant contribution to the scattering, contains the remainder of the surfactant tails, the surfactant head groups and their associated hydrated counterions (which are constrained to vary according to the charge on the micelle). Further parameters adjusted in each fit are an inverse Debye screening length, and an effective volume fraction for the charged sphere $S(Q)$. The best fit must reproduce the absolute neutron scattering intensity and the known molar volumes of SDS. In the regression analysis, some parameter correlation may be expected, especially between the inverse Debye length (which it is not easy to calculate in the presence of gelatin) and the micellar charge. The resultant fits also depend on some of the assumed parameters including the molar volumes, bulk densities, and degree of hydration.

For the gelatin scattering we shall focus on the semidilute regime and a suitable analysis has been given by Pezron *et al.*[7] In their model, they treat the scattering as a sum of two terms, one originating from the screening length or average mesh size (ξ) and one from larger-scale structural inhomogeneities (ζ). For the first term in good solvency conditions, De Gennes[8] suggests that when $Q < \xi^{-1}$, the scattering intensity is given by

$$I_\xi(Q) = I_\xi(0)/[1 + Q^2\xi^2] \quad (2)$$

where (0) refers to $Q = 0$. For the second regime, the model of Debye and Bueche is used to characterize the extent of the spatial inhomogeneities over a length scale ζ.

$$I_\zeta = I_\zeta(0)/[1 + Q^2{}_\zeta{}^2]^2 \quad (3)$$

The data in this paper were all analyzed using the FISH program[16] at the Rutherford and Appleton Laboratory.

Experimental Section

A series of solutions were prepared using D_2O (Fluorochem), distilled H_2O, and deuterated SDS (D-SDS) which was 98% pure and obtained from MSD. The protonated SDS (H-SDS) was 99% pure and obtained from BDH. The purity of the SDS samples was checked by surface tension measurements. The gelatin (ex Kodak) used was deionized photographic bone gelatin type IV which was alkali processed and has a nominal molecular weight of 1.07×10^5 and a density of 1.4 g/mL. The gelatin has a natural pH of 5.8 and an isoelectric point of 5.0, making the chain slightly negative in solution. The gelatin and SDS were dissolved in water at 323 K for 1 h and then cooled to 298 K. During the experiment the samples were maintained at the required temperature using a water circulation system.

Small-angle neutron scattering experiments were carried out at the Rutherford Appleton Laboratory, Didcot, Oxfordshire, using the small-angle scattering camera LOQ. Samples were measured in 2-mm Helma cells with neutrons of wavelength 2.2 to 10 Å at a sample detector distance of 4.3 m. This gave an effective Q range of 0.006 to 0.2 $Å^{-1}$.

By use of D-SDS and solvent contrast variation with D_2O/H_2O mixtures, it is possible to observe scattering from either gelatin

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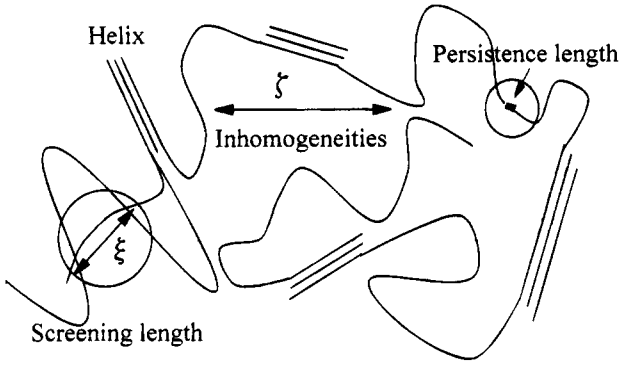

Figure 1. A schematic representation of a gelatin network.

Table 1. Scattering Length Densities

material	$\varrho/10^{-6}$ Å^{-2}
H_2O	−0.56
D_2O	6.39
gelatin	2.11
D-SDS	6.73
H-SDS	0.39

or SDS alone or both together in the mixtures. The scattering length densities of the various components are given in Table 1.

Results and Discussion

Pure Gelatin Solutions. The structure of a gelatin gel is shown schematically in Figure 1. The network structure of the gel is formed by means of linkages through residual triple-helix strands. If these strands are of a significant length, then scattering characteristic of rods should be visible in the data, but no indications are found in the present data. Figure 2 shows a double logarithmic plot of the scattering intensity, $I(Q)$, *vs* Q for a 5% gelatin solution in D_2O at 298 K. This plot is not linear over any substantial part of the Q range measured as might be expected from a dispersion of rigid rods. The scattering can, however, be fitted to a Lorentzian (eq 2) which gives a mesh size ξ of 51 ± 2 Å (Table 2). Over the Q range available, only minimal improvements in the fits were obtained by including the inhomogeneity term (eq 3). This is in contrast to the earlier data of Pezron *et al.*[7] who found a significant improvement in the low Q fit by using both length scales and obtained values of 35 Å for ξ and 135 Å for ζ. Their data, however, were obtained down to a significantly lower Q ($\approx 1 \times 10^{-3}$ Å^{-1}).

Pure SDS Solutions. Figure 3 shows data for 0.18% by weight H-SDS and 2.0% by weight H-SDS in D_2O at 298 K. Below the cmc (0.23 wt % or 8 mM/L) there is no effective scattering, as expected, but above the cmc a strong structure peak occurs. These data are typical of interacting charged particles, where the scattering from the individual particles appears as a peak at low Q because of the structure factor for interparticle correlations. Although the micelle data can be quite well represented by a spherical geometry, the low Q data are fitted rather better by a monodisperse ellipsoid than by polydisperse spheres. The deconvolution of the ellipsoidal fit to these data into a shape factor and structure factor is also shown in the figure. From the model fitting, it was found that the SDS micelle has a slightly elliptical shape, an aggregation number of around 64, and an axial ratio of 1.2 (Table 3). The effective negative charge on the micelle can be interpreted as approximately one-third of the molecules in the micelle not having associated Na^+ counterions. However, both the charge and the Debye length are correlated parameters and their significance should not be overinterpreted.

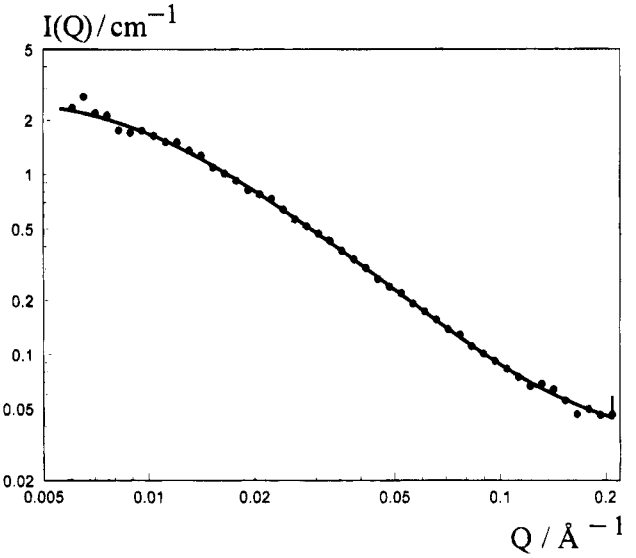

Figure 2. Small-angle scattering from a 5% gelatin gel in D_2O at 298 K. Superimposed is a fit using eq 2.

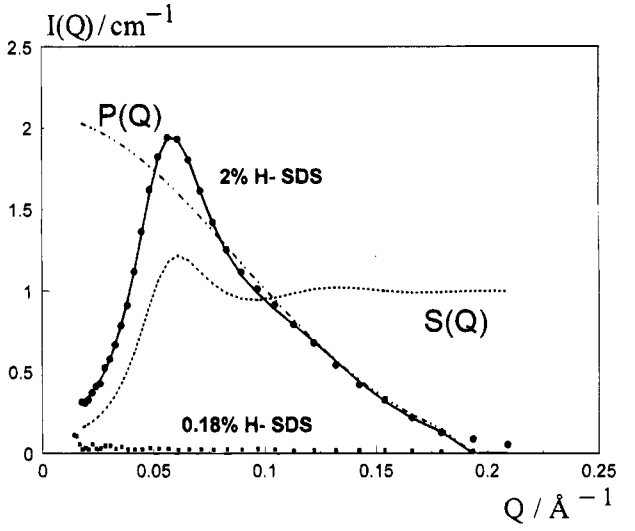

Figure 3. Small-angle scattering for H-SDS solutions in D_2O at 298 K. The solid line represents a fit using the Hayter–Penfold model.

Mixtures of Gelatin and SDS. Figure 4 shows the scattering from 5% gelatin solutions and very small concentrations of added H-SDS, well below the normal cmc in pure D_2O. Interfacial tension data for this system give a cmc1 ≈ 1 mM (0.029 wt %),[1,2] which can be interpreted as *adsorbed* micellization. For the 0.03% H-SDS sample there is evidence of a clear change in shape of the gelatin. At this concentration, adsorbed micelles would only account for a cross section of ≈0.006 cm^{-1}, which would not be visible in the scattering. The data for the 0.03% SDS can therefore be fitted to the same structural models as for pure gelatin and the results are also given in Table 2. There is virtually no difference in the mesh size calculated directly from eq 2 or by adding the inhomogeneity term. The improvement in the statistics is also marginal. The ξ values suggest a collapse of the mesh due to adsorption of SDS, which is at first sight surprising as adsorption effectively increases the net charge on the matrix. However, the binding of SDS to the hydrophobic sites may make some kind of internal bridging with the cationic sites possible as well as micelles bridging across cationic sites.

For the 0.18% SDS sample the same form of scattering as at 0.03% is retained, at least up to $Q \approx 0.05$ Å^{-1}, and

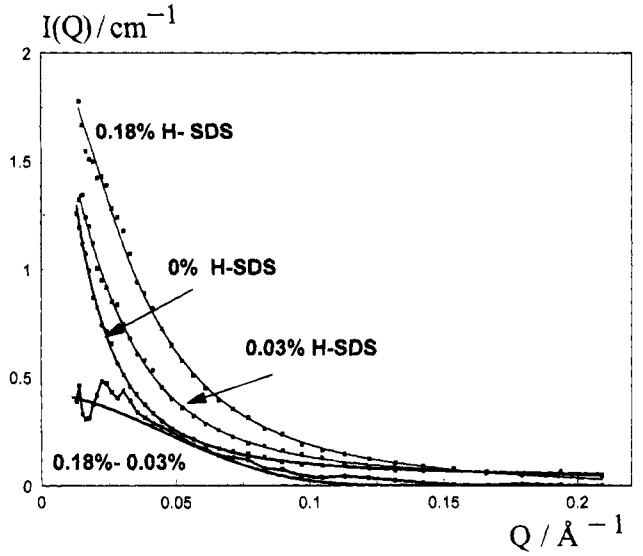

Figure 4. Small-angle scattering from 5% gelatin solutions at 298 K with different concentrations of H-SDS. The solid lines represent numerical fits to the data as described in the text.

Table 2. Mesh Size for Gelatin Solutions

sample	mesh/Å
gelatin (5%)	51 ± 2
gelatin + 0.03% SDS	40 ± 1
gelatin + 0.18% SDS	30 ± 0.5

the enhancement in intensity is compatible with the extra scattering expected from SDS (≈0.2 cm^{-1}). As this SDS concentration is well below cmc2 (≈0.23%) no free micelles are present and any free monomer in solution will be undetectable (see Figure 3). Subtracting these two sets of data should give, at least approximately, the scattering from the adsorbed micelles, assuming that the micelle–gelatin cross-terms are small. The subtracted data are also shown in Figure 4, but the resultant intensity of the low Q limit is rather larger than would be expected for 0.2% H-SDS in D_2O (≈0.12 cm^{-1}). It is tempting to fit these data to a spherical micelle with no structure factor and the radius obtained is 33 ± 2 Å. This value is about 40% larger than for free SDS micelles. Table 2 also includes results from fitting the 0.18% raw data to the mesh model. Here the value obtained for ξ is smaller than for the 0.03% sample suggesting a further collapse of the network has occurred.

Figure 5 shows the scattering from the same system as in Figure 4, but with a much higher surfactant concentration. For comparison, the data from pure gelatin and pure H-SDS are also shown. Several features emerge from these data. The most striking effect is that there is no upturn in intensity at low Q due to the gelatin. This suggests that the gelatin structure has been completely changed by the addition of the SDS and that an $S(Q)$ of a similar form as used to describe the SDS also modifies the gelatin scattering. The H-SDS scattering is also rather different, showing an increased intensity and also a shift in the position of the maximum. This increase in scattering cannot be accounted for by a simple addition of the scattering curves from pure SDS and the gelatin or by a change in contrast of the solution (both solutes being protonated will reduce the effective contrast for the other). The pure SDS data, based on a cmc of 0.23% by weight reflects the fact that 1.77% by weight is in micelles and 0.23% by weight is free. A 5% gelatin solution[2,3] requires about 3% by weight SDS for saturation and therefore all the micelles in the mixed system will be adsorbed. This cannot, in itself, explain the increase in the intensity of

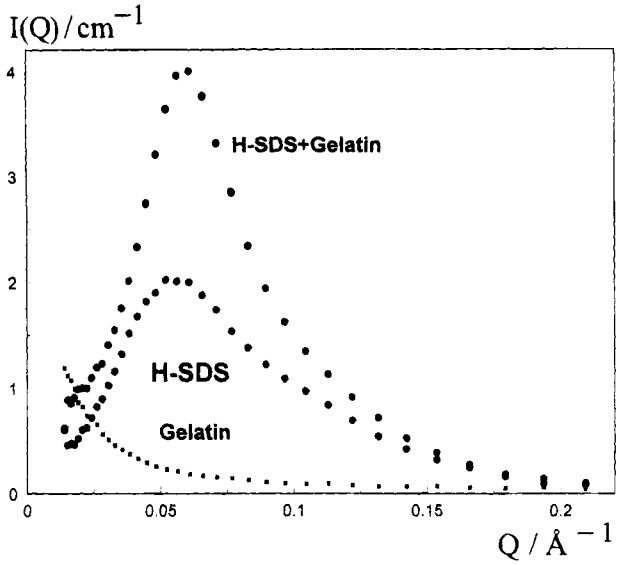

Figure 5. A comparison of the small-angle scattering from a 5% gelatin solution with added 2% H-SDS and with pure 2% H-SDS and 5% gelatin solutions in D_2O at 298 K.

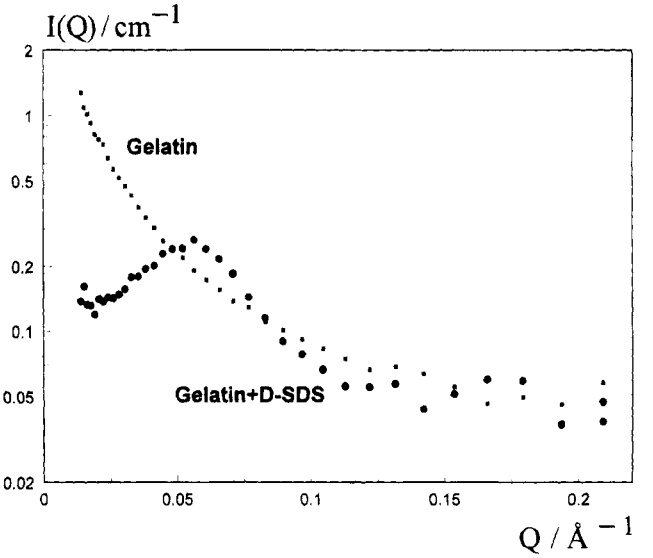

Figure 6. Small-angle scattering from a 5% gelatin solution compared with added D-SDS in D_2O at 298 K.

the structure peak and points to a further contribution from the gelatin due to the strong interaction with SDS. The small shift in the structure peak suggests that the micelles may be closer together but this may be due to a change in the number and or size of the micelles as the amount of free SDS is reduced. The size of the micelles is discussed further below.

The effects discussed above can be examined in more detail by selectively masking the solutes by contrast variation. Inspection of the scattering length densities given in Table 1 shows that it is possible to match either the gelatin scattering length density with the solvent (40% D_2O–60% H_2O) or the 98% D-SDS with pure D_2O.

Figure 6 shows the scattering from gelatin and D-SDS in D_2O. The scattering from the D-SDS is essentially invisible even given a small change in contrast brought about by the gelatin itself. For comparison, the scattering from a pure gelatin solution is also shown. With added surfactant the gelatin scattering changes dramatically, as was indicated in Figure 5, and the low Q scattering due to the mesh is substantially reduced. The data suggest that the gelatin adopts a conformation with a similar

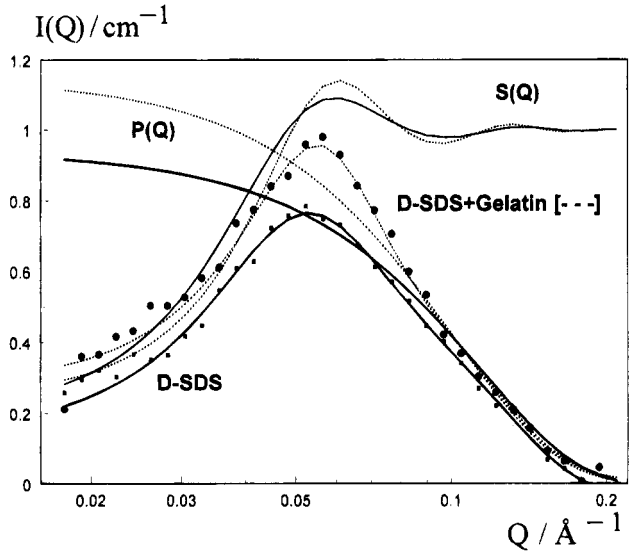

Figure 7. Small-angle scattering from a 2% D-SDS solution with (●) and without (■) gelatin in a solvent mixture of 40% D_2O/60% H_2O which is at contrast match with gelatin and at 298 K. The lines are a deconvolution of a numerical fit to the data into a structure factor and a shape factor. The continuous line is for D-SDS and the dashed line for the mixture.

Table 3. Typical Fitted Parameters to the Hayter–Penfold Model

system	axial ratio	equivalent sphere radius/Å	Debye length/Å	aggregation no.
2% D-SDS	1.2	21 ± 2	25	64 ± 5
2% D-SDS + 5% gelatin	1.6	24 ± 2	25	
2% H-SDS 0.1 M NaCl	1.8	25 ± 2	8	112 ± 10

structure factor to the SDS. This can be thought of as nonscattering holes in the structure formed by the "invisible" micelles. Indeed, the position of the maximum in Figure 6 is similar to that in Figure 5 (≈0.06 Å^{-1}). This dramatic change in the network, presumably due to strong binding of the SDS to the gelatin, corroborates the changes in the gelatin mesh structure seen in Figure 3 where the chains were undersaturated with SDS.

This picture is further elucidated in Figure 7, by comparing the reverse system in which the gelatin is contrast matched to the solvent so that only the D-SDS is visible. For comparison, data for a pure 2% D-SDS solution is also shown. The increase in the height of the structure peak compares with that given in Figure 5 showing the contribution of extra D-SDS micelles formed by adsorption onto gelatin. This increase in intensity, as discussed above, should be directly related to the total SDS present less the normal cmc, which is consistent with the data in the figure.

In order to analyze the data in more detail, the Hayter and Penfold model has been used and the fitted parameters are given in Table 3. Two features are apparent from the fits: the adsorbed micelles appear to be more elliptical and slightly larger than nonadsorbed ones though the error bars on the numbers are significant. This is compatible with the simple model proposed to explain Figure 4.

Figure 7 also shows a deconvolution of the scattering into $S(Q)$ and $P(Q)$. The change in size and shape of the micelles results in a more rapidly decaying $P(Q)$ in the presence of gelatin (i.e., larger micelles). The $S(Q)$ shows that the system has become slightly more concentrated as the average separation of the micelles becomes less. One possible rationalization of the data is the penetration of gelatin into the micelle interior. This picture confirms

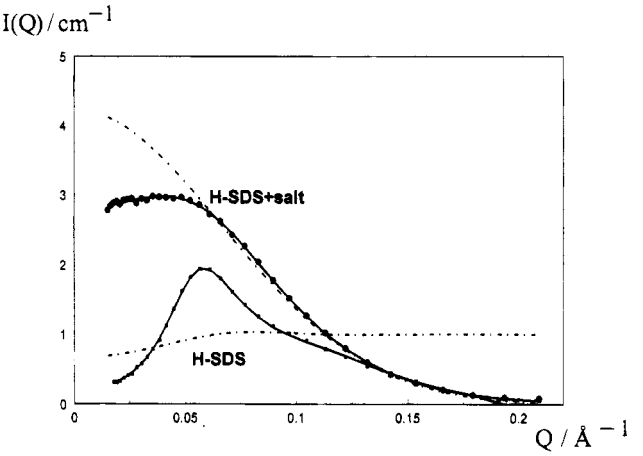

Figure 8. Small-angle scattering of 2% H-SDS solutions in D_2O, with (●) and without (■) 0.1 M NaCl at 298 K. The solid lines are a deconvolution of a numerical fit to the data into a structure factor and a shape factor.

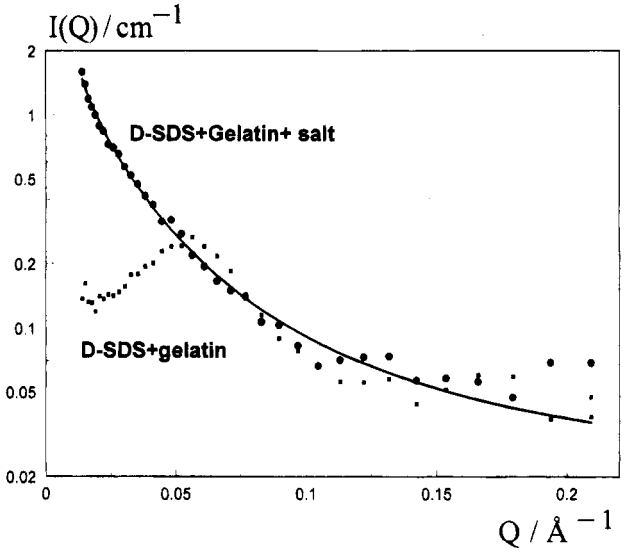

Figure 9. Small-angle scattering from a 2% D-SDS solution and 5% gelatin with (●) and without (■) 0.1 M NaCl in D_2O at 298 K.

the data obtained from the purely protonated system described above, where both solutes were visible.

Effects of Ionic Strength. Figure 8 shows SANS data for 2% H-SDS and 0.1 M NaCl in 40% D_2O/60% H_2O solution. The model fits to the data indicate that the micelles formed in the presence of salt are slightly larger but more elliptical than in the absence of salt (Table 3). The salt screens the intermicellar repulsion, so the interparticle structure factor is much less obvious and appears to peak at higher Q. The scattering is thus dominated by the pure shape factor $P(Q)$ for isolated spheres.

To separate the components in the system, a similar set of data to that described above was obtained from D-SDS in pure D_2O so that only gelatin scattering was visible. The data are shown in Figure 9. The effect of adding D-SDS, as in Figure 6 above, was to produce a structure peak in the gelatin scattering and the addition of salt can be seen to completely remove it. The effect is similar to that shown in Figure 8 as the salt effectively screens the interaction between the D-SDS micelles and to some extent the electrostatic interaction between the gelatin and the D-SDS. The loss of emphasis of the structure factor in Figure 8 also must also ocur in Figure 9 and accounts for the loss of the weaker structure peak of the gelatin. The

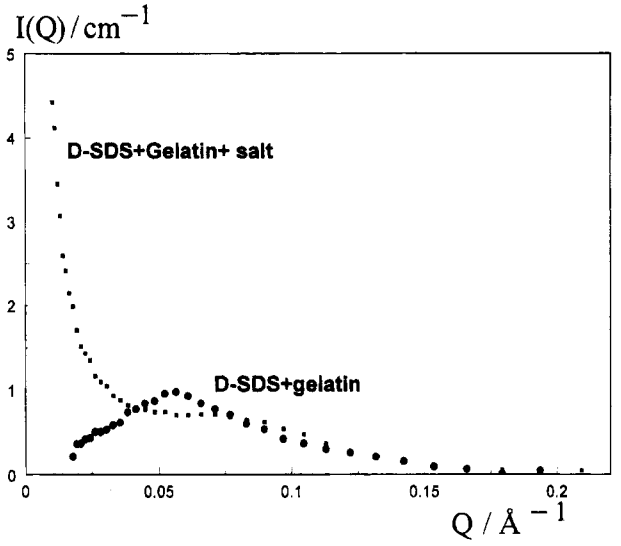

Figure 10. Small-angle scattering from a 2% D-SDS solution and 5% gelatin, with (●) and without (■) 0.1 M NaCl in 40% D_2O/60% H_2O. The sample with salt was at 318 K and without at 298 K.

surfactant is still bound to the gelatin, but the data suggest that the regular spacing of the micelles required to give the strong structure peak is lost and the adsorbed micelles are more disordered along the gelatin chain. Finally, in Figure 10, we show data from D-SDS in 40% D_2O/60% H_2O with 5% gelatin, with and without salt. These results show the effect of the gelatin and salt on the SDS structure directly as a sharp upturn of the data at low Q. The intensity of this scattering is stronger than that for gelatin in pure D_2O (Figure 3) and is a direct result of the very strong binding/aggregation of the gelatin/D-SDS complex. The salt sample was recorded at 318 K but the temperature effects are small.[17]

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Conclusions

SDS micelles are seen to be slightly ellipsoidal in nature with aggregation numbers of approximately 64. The addition of salt to pure SDS solutions increases the size of the micelles and the axial ratio of the ellipses.

Below the cmc the interaction of SDS and gelatin has been observed. At very low SDS concentrations, the SDS appears to bind to the gelatin resulting in a decrease in the gel mesh size.

Above the cmc, the addition of SDS to the system results in a disruption of the gelatin mesh, and the gelatin structure factor follows that of the adsorbed SDS. In the adsorbed state the micelles are larger and more elliptical, though we note similar changes on adding salt to pure SDS. The structure factors suggest that the bound micelles are closer together than in the bulk and that they are strongly bound to the gelatin. It might be conjectured that the SDS micelles act as bridging sites for the gelatin network, which would lead to an increase in the bulk viscosity. On adding NaCl to the SDS–gelatin complex, the SDS structure factor is strongly suppressed and the gelatin scattering more closely resembles that of pure gelatin though the micelles are still bound.

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