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ORIGINAL CONTRIBUTION

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Isothermal titration calorimetric studies of surfactant interactions with negatively charged, 'hairy' latex nanoparticles

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Abstract Isothermal titration calorimetry (ITC) measurements of the mixture of the cationic surfactant cetyl trimethyl ammonium bromide (CTAB) with negatively charged, 'hairy' copolymer latices (poly-(2,3-epoxypropylmethacrylate-*co*-methacrylic acid) in different ratio) at high water excess indicate a monomer adsorption mechanism of CTAB by the polymer particles. The number of adsorbed CTAB molecules at saturation corresponds approximately to the number of negative elementary charges bound at the surface of the latices. The mixing enthalpy is the sum of demicellization and sorption enthalpies. At 25 °C for CTAB the demicellization enthalpy amounts to 10 kJ/mol, whereas the adsorption enthalpy varies from −7 kJ/mol (surface charge density of the latices $\sigma = -0.37$ C/m^2) to +3 kJ/mol ($\sigma = -0.085$ C/m^2). The hydrodynamic radius R_H of the latex particles upon titration of cationic detergent and salt (NaBr) decreases by about 2 nm until the onset of aggregation near the isoelectric point. Titration of nonionic or anionic detergents has much less influence on the hydrodynamic radius and produces no measurable adsorption heat. The results are consistent within a model of latex particles with extended negatively charged polymer chains interacting predominantly via Coulombic forces with detergents.

Keywords ITC · Epoxypropyl methacrylate · Detergent · Adsorption enthalpy · Demicellization

Introduction

Noncovalent bonding of small molecules to charged surfaces of larger colloids (proteins, latices, quantum dots) in solutions finds various applications, the latest among them in the bioconjugation to quantum dot probes [1]. One variant of binding a label to the carboxylated cap of a quantum dot is the sorption of a positively charged leucine zipper peptide via electrostatic interaction with the negatively charged cap. Latex nanoparticles with drugs bonded to their surface are able to penetrate the blood-brain barrier (BBB) [2, 3] in animals which has been proved by electron microscopic analysis [4]. Therefore, the nature and strength of the bonding of molecules to the surface of latex particles is of considerable interest.

Although electrostatic dominance in noncovalent bonding of small charged molecules to oppositely charged surfaces seems to be clear, there are, to our knowledge, only a few papers dealing with quantitative evaluation of corresponding binding enthalpies. In this paper, we study the sorption of a nonionic, a cationic and an anionic surfactant on polymer latices with variable negative surface charge density by means of

Isothermal Titration Calorimetry (ITC). To illustrate the power of ITC, let us regard the quasi-binary mixture of N_D molecules of a surfactant D with $N_L$latex binding sites L (N_L is usually unknown at the beginning of the experiment). For the number N_W of water molecules in the mixture we assume $N_W > > N_D, N_L$. At the beginning of experiment, the (reduced) mole fraction of D in the solution is given by $X = \frac{N_D}{N}$ with $N = N_D + N_L$. Now, a small volume V_S of the mixture D/L at the same temperature, but with mole fraction $X_S = \frac{N_{DS}}{N_S} \neq X$ with $N_S = N_{DS} + N_{LS} < < N$ is added (injected using a syringe) to the initial mixture (volume V and mole fraction X). The mole fraction after the injection (i.e., after mixing V at X with $V_S$at X_S) is given by $X' \approx X + (X_S - X) \cdot \epsilon$, where $\varepsilon = \frac{N_S}{N} \ll 1$. To keep the temperature after mixing constant, per $N_S$injected molecules/sites the heat $q_{ITC}(X, X_S) = \frac{(N+N_S)\cdot H(X') - N \cdot H(X) - N_S \cdot H(X_S)}{N_S}$ has to be added to the mixture and is measured in the ITC-experiment. $H(X)$ is the molar enthalpy of the mixture at mole fraction X (at given temperature and pressure). Using the expressions given above, one obtains for the central quantity in ITC the formula [5]

$$q_{ITC}(X, X_S) = H(X) - H(X_S) + (X_S - X)\frac{dH(X)}{dX} \tag{1}$$

Adding surfactants to latices, one has to take into account heat effects related both to aggregation (micellization) and to adsorption. The simplest model for the molar enthalpy of an aqueous surfactant solution (without latex) with critical micelle concentration $CMC < < 1$ mol/l assumes $H(X < X_c) = 0$ with $X \approx C/$(55.5 mol/l) and $Xc \approx CMC/$(55.5 mol/l). For surfactant concentrations C above the CMC, where monomers and micelles coexist in the solution, the molar enthalpy increases linearly with the mole fraction X of the surfactant:

$$H(X > X_c) = \frac{X - X_c}{1 - X_c} H_{mic} \tag{2}$$

where H_{mic} is the molar micellization enthalpy, i.e., the difference in molar enthalpies of micelles and monomers. Within the frame of this simple model, isothermal titration of surfactant solution with mole fraction $X_S > Xc$ to a sample containing monomers at mole fraction $X < X_c$gives, according to Eq. (1):

$$q_{ITC}(X < X_c, X_S > X_c) = -\frac{X_S - X_c}{1 - X_c} H_{mic} \tag{3}$$

and

$$q_{ITC}(X < X_c, X_S < X_c) = q_{ITC}(X > X_c, X_S > X_c) = 0 \tag{4}$$

If the experimental q_{ITC} values obey Eqs. (3) and (4), we call the behavior of the surfactant 'ideal'. Deviations from ideality are caused, e.g., by interactions between monomers, micelles, by significant dilution enthalpies of monomer and micelles and by a size distribution of micelles. They manifest themselves in a smeared monomer-micelle transition and significant X-dependence of q_{ITC} over a larger X interval.

In general, the measurement of q_{ITC} values provides a method for the thermodynamic characterization of micelle formation of single surfactants [6, 7] or surfactant mixtures [7] and of surfactant sorption on polymers [8, 9]. However, the interpretation of q_{ITC} values for binding reactions is difficult and is usually only qualitative, due to the many linked equilibria that may contribute to the overall heat effect. Excess enthalpy functions have been derived [5] from q_{ITC} values in mixtures of the phospholipid POPC and detergents of the type $C_{12}EO_n$ with $n = 3$–6. Dynamic light scattering (DLS) was used to determine changes in latex particle size upon adsorption of detergents [10, 11] and cellulose [12]. The structure of physically adsorbed polymer layers [13] and polyelectrolyte multilayers [14] as well as the structure of the colloid particles [16, 17] itself has been studied by small-angle scattering of X-ray and neutrons. Calorimetric studies of binding interactions between ionic surfactants and polyethylene glycol [9], poly(oxypropylene)-poly(oxyethylene)-poly(oxypropylene) (PPO-PEO-PPO) triblock copolymers [18, 19] and polystyrene latex particles [15] have been performed.

However, the quantitative distinction between hydrophobic and columbic binding of a surfactant to another particle has not been sufficiently elaborated. In this study, as surfactants, we have chosen the anionic surfactant sodium-dodecyl-sulfate ($C_{12}H_{25}NaO_4S$, SDS), the cationic surfactant cetyl-trimethyl-ammonium-bromide ($C_{19}H_{42}NBr$, CTAB), and the non-ionic surfactant decylhexaglycol ($C_{22}H_{46}O_7$, $C_{10}E_6$). As substrates, we used a series of hydrophilic, 'hairy' copolymer latex particles ('Nfg-EPS'), consisting of glycidyl-methacrylate copolymerized with different amounts of methacrylic acid. By applying the steady in-flow method, (poly-)methacrylic acid should be enriched at the particle surface in form of water-soluble chains.

Experimental

Chemicals and sample preparation All chemicals were used as received. The solutions were prepared with ultrapure water from a Millipore unit. This water was also used in the ultrafiltration process for latex purification.

A series of carboxylated polyglycidyl methacrylate latices was prepared by a semi-batch emulsion polymerization in a 250-mL-double wall glass reactor (HWS Mainz), equipped with glass anchor stirrer, nitrogen inlet, reflux condenser and dropping funnel. 0.2 g sodium-dodecyl-sulfate (SDS, Serva) were dissolved in 156 mL water, 0.108 g potassium peroxydisulfate (Sigma) in 20 mL. The SDS solution was given into the

Table 1 Samples used in this study. R_H is the hydrodynamic radius of the particles determined by DLS. The surface charge density has been measured by titration to the point of zero charge using 0.1 mmol/l P-DADMAC solution [22]. In column 5, the surface charge density σ given in column 4 has been converted into amount of elementary charges e per g latex using the formula $\sigma/(\mathrm{mol}\cdot e/\mathrm{g}) = \sigma/(\mathrm{C/m^2})\cdot\frac{3}{\rho\cdot R\cdot F}$ with particle mass density ρ (in column 3) and Faraday constant F. Molar adsorption enthalpy H_{ads} and saturation coverage for CTAB upon EPS-particle surfaces is given in column 6 and column 7, respectively

Latex	R_H [nm]	ρ [g/cm^3]	σ [C/m^2]	σ [mmol e/g]	H_{ads} [kJ/mol]	Saturation coverage [(mmol CTAB)/(g EPS)]
EPS1	66.4 ± 0.5	1.39	0.085	0.029	3 ± 1.5	0.05 ± 0.01
EPS3	71.4 ± 0.5	1.40	0.22	0.068	0 ± 1.5	0.08 ± 0.02
EPS5	71.5 ± 0.5	1.42	0.37	0.113	−7 ± 1.5	0.20 ± 0.03

reactor, the amount $Y_{EP} = 18$ g of 2,3-epoxypropyl methacrylate (Fluka) were added. The emulsion was purged with nitrogen under stirring (350/min) for 20 min at ambient temperature. Then, while heating up to polymerization temperature (60 °C), purging with nitrogen was continued. The potassium peroxydisulfate solution was given via the dropping funnel. A variable amount Y_S of the carboxylating agent, methacrylic acid (Fluka) ($Y_S = 0.4$ g for EPS1, $Y_S = 1.2$ g for EPS3, and $Y_S = 2.0$ g for EPS5), was dissolved in ultra-pure water and filled up with it to a total volume of 20 mL (the finally resulting copolymers Y_{EP} (2,3-epoxypropyl methacrylate) + Y_S (methacrylic acid) have been denoted by EPS1, EPS3, and EPS5). This solution was sucked into a 20-mL-plastic syringe (B Braun) which was fixed on an infusion pump (Typ 610 B.S, Medipan, Warszawa). After 10 min at 60 °C, the polymerization was started by fast inflow of the potassium peroxydisulfate solution from the dropping funnel into the monomer emulsion in the reactor. At the same time, the infusion pump was started. Over a period of 2.2 h, it admixed the methacrylic acid solution constantly to the polymerizing latex. After 6 h, the polymerization was finished by stopping the nitrogen stream and cooling down the latex. Purification and characterization of the dispersions were described in detail [20]. Ion exchange of the purified latices was performed according to [21]. All molar data related to 196 mL total water volume in the reactor. The hydrodynamic radii and the surface charge density of the particles purified by ultrafiltration are given in Table 1. The nonionic detergent $C_{10}E_6$ and the anionic detergent SDS were purchased from Fluka and the cationic detergent CTAB from Merck.

Isothermal Titration Calorimetry ITC experiments were carried out with an MCS ITC, MicroCal, USA. Micellar surfactant solutions were placed in a 250-µL continuously stirred syringe at 400 rpm. The sample cell has a volume of 1.345 mL and injections were made with volumes of 5 to 20 µL in intervals of 3 to 10 min. Both samples in cell and in syringe were degassed for at least 10 min before titration. Each experiment was carried out at 25.0 ± 0.02 °C where the temperature was controlled using a Haake oil bath.

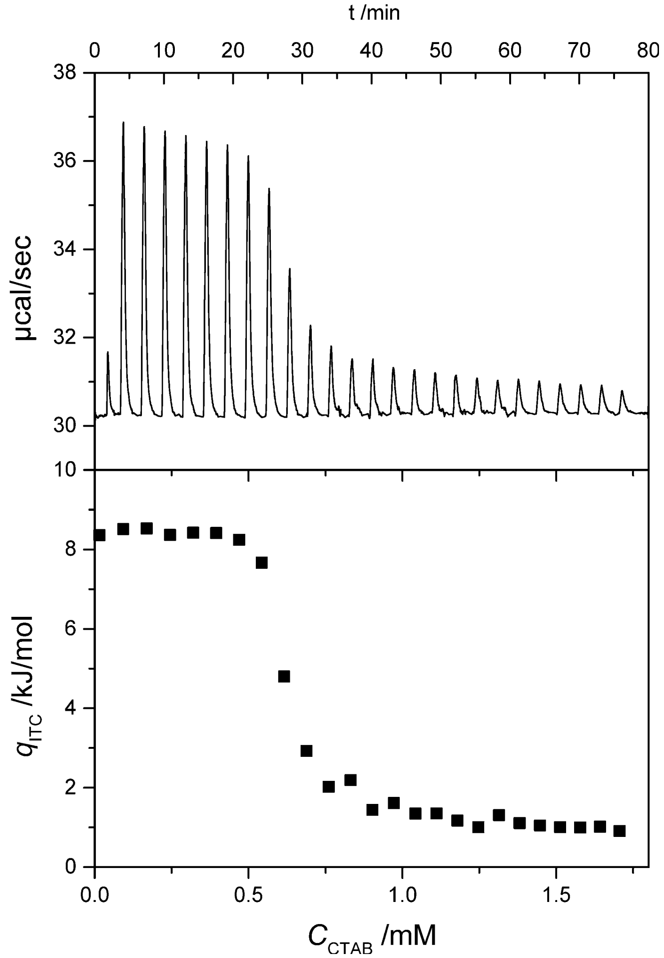

Fig. 1 (*above*) Heat power in the calorimeter mixing cell during injections of CTAB at concentration 10.4 mmol/l into water of increasing CTAB concentration, C_{CTAB}, vs time (first injection 2 µL, followed by injections of 10 µL). The temperature in the cell is held constant at 25 °C. (*below*) q_{ITC} vs concentration C_{CTAB} of CTAB in the cell for the set of injections displayed above

Dynamic Light Scattering DLS measurements were made using an ALV/DLS-5000 compact goniometer system (Langen, Germany) with a Nd-YAG-Laser at 532 nm wavelength as light source. The scattering cells (10-mL cylindrical ampoules) were immersed in a thermostated bath of toluene at 25 °C (Julabo, Germany). Concentra-

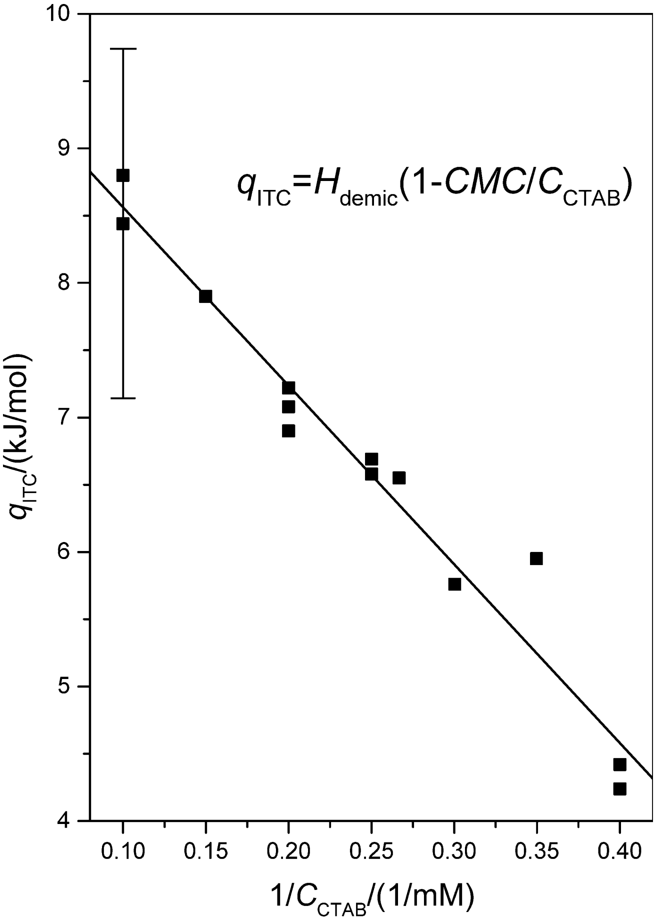

Fig. 2 q_{ITC} of CTAB into H_2O at 25°C vs inverse concentration of CTAB in the syringe, $1/C_{CTAB}$. We obtain H_{demic} of 9.9 kJ/mol and *CMC* of 1.3 mmol/l by linear fit. *Error bars* are given only for the first points

tion of EPS in the samples was in general 100 mg/L. Measurements were made at 13 angles between 30° and 150°. Correlograms were produced for a time of 60 s at each angular position. Diffusion coefficients were determined by a cumulant fit of second order to each correlogram. Hydrodynamic radii were calculated from the diffusion coefficient using Stokes-Einstein equation together with the water viscosity at 25 °C.

Results and discussion

Aqueous solutions of CTAB

In Fig. 1, the raw ITC data (mixing heat power vs time) together with the integrated signals (q_{ITC} per injected mole vs concentration) for titration of CTAB into water at 25 °C are shown. From the linear behavior of the q_{ITC}-plot vs inverse concentration of CTAB in the mixture, $1/C_{CTAB}$(Fig. 2) we derive an approximate "ideal" behavior of the molar enthalpy $H(X)$ of the mixture, i.e., for $C_{CTAB} < CMC$ the value of $H(X)$ can be put to zero and for $C_{CTAB} > CMC$ the value of $H(X)$ increases linearly with the mole fraction X of CTAB in the mixture. Within the idealized model, the fit at 25 °C gives for the molar demicellization enthalpy $H_{demic} = (9.9 \pm 1.5)$ kJ/mol and for the critical micelle concentration the value $CMC = (1.3 \pm 0.2)$ mmol/l.

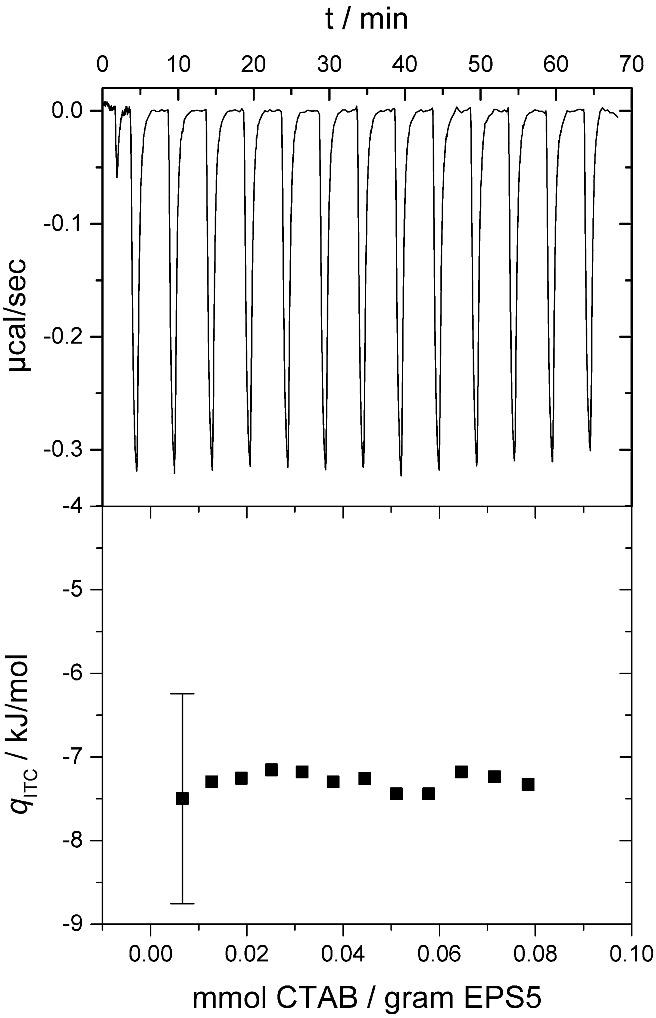

Fig. 3 Raw data and q_{ITC} vs ratio of CTAB to EPS5 for a set of 20-µL injections of an aqueous 0.5 mmol/l CTAB solution into 1.25 g/L EPS5 at 25 °C. The adsorption is exothermic ($H_{ads} \approx (-7 \pm 1.5)$ kJ/mol)

Detergents and EPS-copolymer lattices

Titration of aqueous solutions of CTAB below the *CMC* into EPS-dispersions should reveal the adsorption enthalpy of monomers by EPS. In Fig. 3 the titration curves in case of $C_{CTAB} = 0.5$ mmol/l and $C_{EPS5} = 10$ g/L deliver the molar adsorption enthalpy of $H_{ads} \approx -7$ kJ/mol. CTAB solutions above the *CMC* contain both monomers and micelles. If, upon titration of CTAB at concentration $C_{CTAB} > CMC$ to EPS-latices, only monomers are adsorbed, the observed q_{ITC} values should be the sum of demicellization and the monomer adsorption enthalpies, i.e., at the beginning of the titration the equation

$$q_{ITC}(C_{CTAB}) = \begin{cases} H_{ads} \text{ for } C_{CTAB} < CMC \\ H_{demic}(1 - CMC/C_{CTAB}) + H_{ads} \text{ for } C_{CTAB} > CMC \end{cases} \quad (5)$$

should be valid, where C_{CTAB} is the CTAB concentration in the syringe. In Fig. 4, the measured q_{ITC} values for titration of CTAB at concentrations $C_{CTAB} = 10$ mmol/l, 5 mmol/l, 2.5 mmol/l, and 0.5 mmol/l into aqueous EPS-latices are presented. The error bars (shown only for the first points) were obtained by repeating the measurements three to five times and averaging. They indicate the approximate standard deviation of a single measurement. Using Eq. (5), from these data the approximate CTAB-monomer adsorption enthalpies H_{ads} of (-7.0 ± 1.5) kJ/mol for EPS5, (0 ± 1.5) kJ/mol for EPS3, and $(+3 \pm 1.5)$ kJ/mol for EPS1 can be derived analyzing the beginning of the titration curves (i.e., at vanishing coverage of EPS-surfaces by CTAB). These values are listed in Table 1. At saturation of surface coverage, the q_{ITC} values upon titrating CTAB with concentration C_{CTAB} into EPS-dispersions should approximate those of titration into water. In Fig. 5 the titration curves of CTAB with $C_{CTAB} = 2.5$ mmol/l into EPS1, EPS3, and EPS5 dispersions together with the 2.5 mmol/l CTAB titration curve into pure water are depicted. We obtain from these curves the saturation values$\theta_{sat} \approx (0.05 \pm 0.01)$, (0.08 ± 0.02), and (0.20 ± 0.03) mmol CTAB/g EPS, for

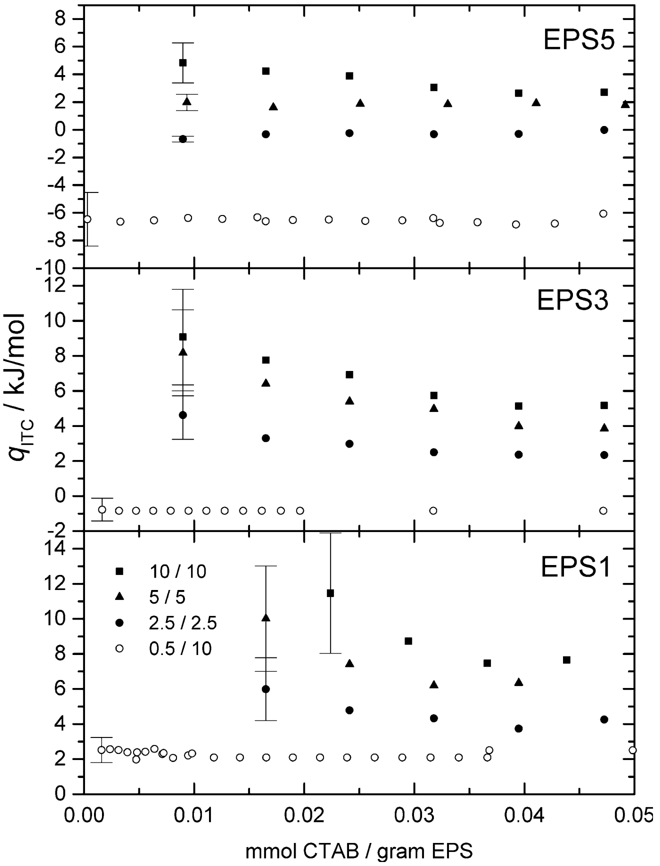

Fig. 4 Titration heat of micelles (concentration of CTAB in syringe $C_{CTAB} = 10$ mmol/l, 5 mmol/l, and 2.5 mmol/l, *solid symbols*) of CTAB solutions with EPS5, EPS3 and EPS1 (concentration of EPS in calorimeter cell 10 g/L, 5 g/L, and 2.5 g/L), respectively, and titration heat of monomers (0.5 mmol/l, *open circles*) with EPS5, EPS3, and EPS1 (10 g/L). *Error bars* are given only for the first points. Note that the abscissa denotes the ratio of the amount of CTAB to the amount of EPS in the calorimeter cell, whereas in Eq. (5)C_{CTAB} denotes the concentration of CTAB in the syringe

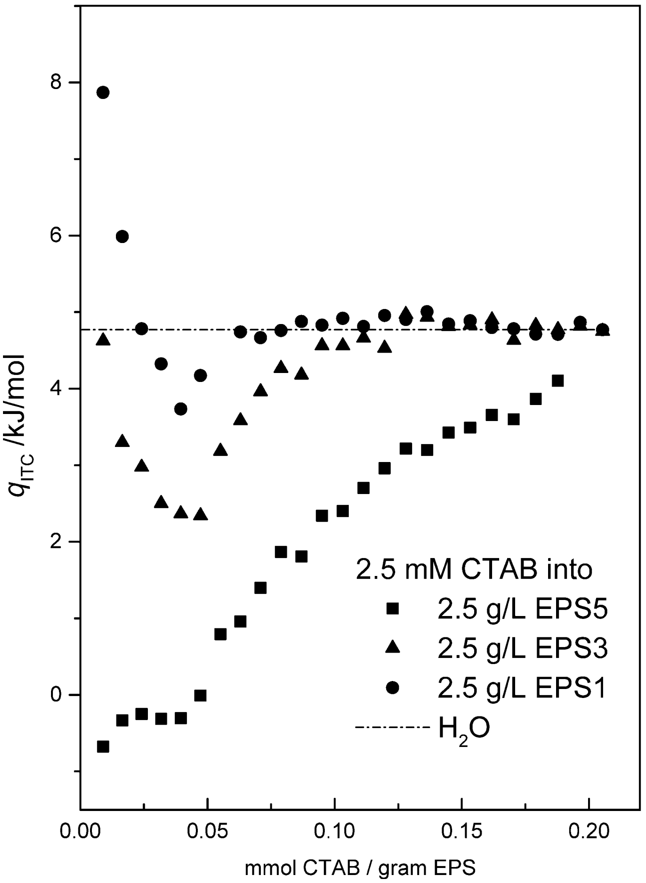

Fig. 5 Titration of 2.5 mmol/l CTAB into 2.5 g/L EPS5, EPS3, and EPS1. The *dashed line* indicates the heat q_{ITC} of 2.5 mmol/l CTAB into pure water (cf. Fig. 2). From these data, approximate saturation values for surface coverage of the latices of 0.05, 0.08, and 0.2 for EPS1, EPS3, and EPS5, respectively, are obtained (cf. Table 1)

EPS1, EPS3, and EPS5, respectively. These saturation values are proportional to the total amount of negative charge at the surface of the latices (cf. Table 1). For EPS5, the saturation value corresponds to a nearly dense packing of CTAB on the surface.

Repeating the above described titration experiments with the anionic detergent SDS and the nonionic detergent $C_{10}E_6$ yields nearly vanishing adsorption enthalpies for all EPS-latices (Fig. 6).

The calorimetric behavior of the detergents interacting with EPS-latices described in the previous section corresponds to the results obtained by DLS measurements. The hydrodynamic radius of the EPS-particles upon mixing with the cationic detergent CTAB decreases, and the mixing of EPS-latex with the anionic SDS and nonionic $C_{10}E_6$ results in a negligible influence on the radius. In Fig. 7, this behavior is documented for EPS5. In Fig. 8 the decrease of the hydrodynamic radii of EPS1 and EPS3 latices upon adding CTAB to the solution is shown. The line drawn in Fig. 7 is a linear fit to the values of the hydrodynamic radius of EPS5 (solid circles) vs ratio of CTAB amount to EPS5 mass in the solution. The negative slope of this line is approximately the same that is obtained in case of added NaBr (solid squares). Therefore, we conclude that the cations added to the solution screen the negative charges in the polymeric chains at the surface of the particles which leads to their contraction.

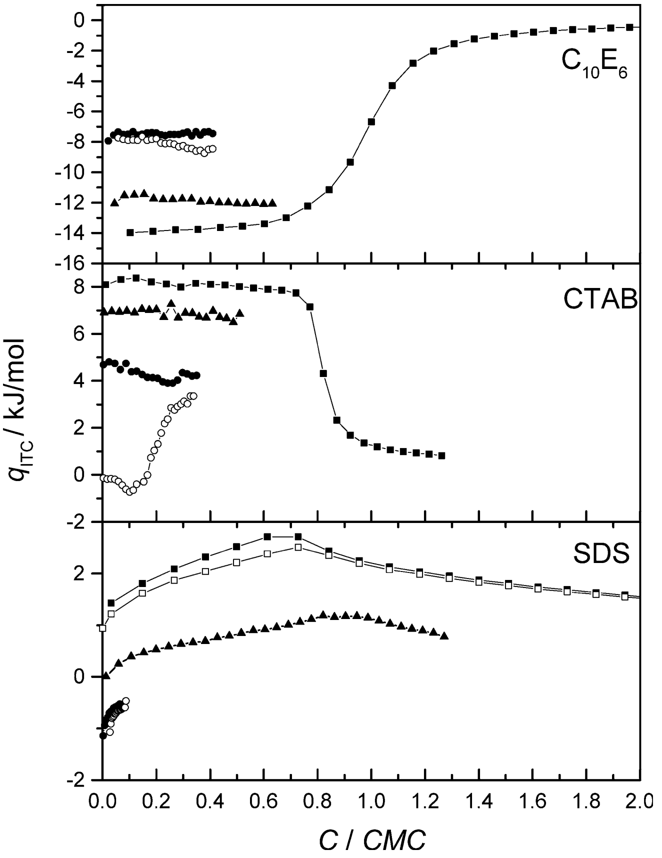

Fig. 6 Comparison of three surfactants titrating at 25 °C into pure water and into aqueous EPS-latex.$C_{10}E_6$: 10 (*squares*), 5 (*upright triangles*), and 2.5 (*solid circles*) mmol/l $C_{10}E_6$into water and 2.5 (*open circles*) mmol/l $C_{10}E_6$ into 5 g/L EPS1. Within experimental error no adsorption heat was observed.*CTAB*: 10 (*squares*), 5 (*upright triangles*), and 2.5 (*solid circles*) mmol/l CTAB into water and 2.5 mmol/l (*open circles*) CTAB into 5 g/L EPS5. One can clearly observe a nonvanishing adsorption heat.*SDS*: 200 (*solid squares*), 80 (*upright triangles*), and 5 (*solid circles*) mmol/l SDS into water and 200 (*open squares*), 5 (*open circle*) mmol/l SDS into 5 g/L EPS3. Within experimental error no adsorption heat was observed. For the plot,*CMC*-values of 1 mmol/l, 1.3 mmol/l, and 9.4 mmol/l for $C_{10}E_6$, CTAB, and SDS have been assumed, respectively

Conclusion

In aqueous solution, the negatively charged hydrophilic 'hairs' of the EPS-copolymer latices (poly-(2,3-epoxypropyl methacrylate-*co*-methacrylic acid) in different

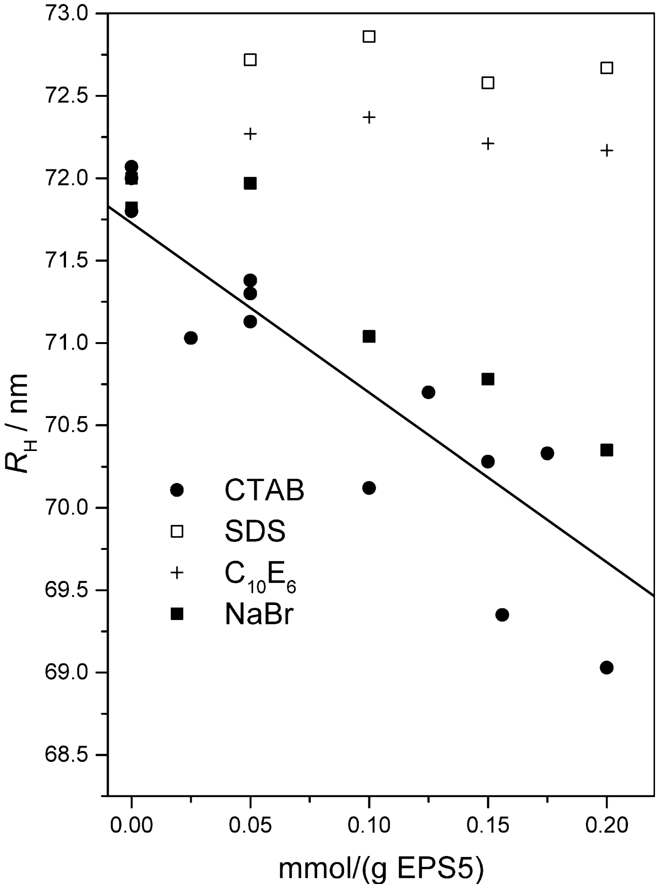

Fig. 7 Hydrodynamic radius R_H of latex EPS5 at 25 °C in different environments: CTAB (*solid circles*), SDS (*open squares*), $C_{10}E_6$ (*crosses*). and NaBr (*solid squares*). For concentrations above 0.2 mmol/g aggregation of the latex particles is observed. The *line*indicates the decrease of the radius of EPS5-particles in CTAB and NaBr with increasing concentration. SDS and $C_{10}E_6$ do not change the hydrodynamic radius significantly and do not lead to aggregation

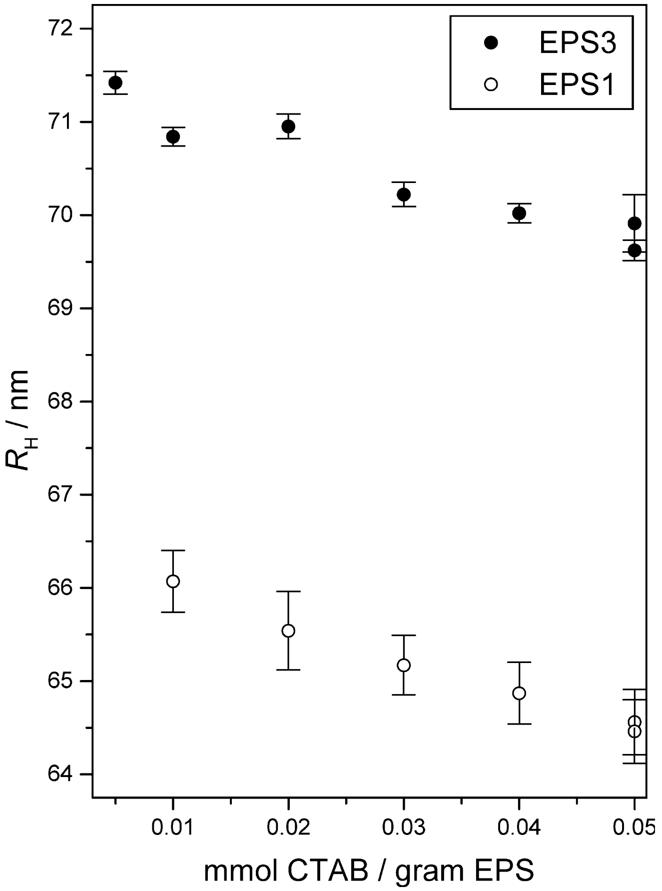

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Fig. 8 Hydrodynamic radius R_H of EPS1 and EPS3 latices at 25 °C vs molar amount of CTAB per gram EPS-particles. A decrease of R_H with increasing amount of CTAB is observed, and near and above saturation aggregation of particles is observed. (not shown). The *error bars* correspond to the standard deviation of the mean hydrodynamic radius (the average of the values for R_H obtained at the different scattering angles of a given sample)

ratio) are extended off the latex core because of mutual Coulombic repulsion. The release of the Coulomb-stress in the surface network by intercalating cations leads to a contraction of the 'hairs' and, correspondingly, to a decrease of the hydrodynamic radius of the latices. Nonionic and anionic detergents like $C_{10}E_6$ and SDS do not exhibit adsorption enthalpies on EPS-surfaces and in the region of concentrations far below their *CMC* do not change the hydrodynamic radius of EPS-latices. However, for cationic detergent CTAB, adsorption enthalpies in the range from +3 kJ/mol at low surface charge to −7 kJ/mol at high surface charge density of EPS-particles were measured. These values can be explained assuming predominant Coulombic adsorption at high surface charge density and an increasing contribution of hydrophobic interaction with decreasing surface charge of the latices. Indeed, adsorption of detergent monomers on the latex surface moves them from aqueous surrounding to a more or less hydrocarbon environment. Initiated by Coulombic attraction, the monomers after adsorption will be surrounded by an increasing amount of hydrocarbons with decreasing surface charge. Therefore, the enthalpy of adsorbed monomers will be concomitantly increased. To prove the proposed structural model, small angle X-ray or neutron scattering experiments should be performed.

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