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Hydrodynamic size of DNA/cationic gemini surfactant complex as a function of surfactant structure

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Abstract. The present study deals with the determination of hydrodynamic size of DNA/cationic gemini surfactant complex in sodium bromide solution using the dynamic light scattering method. Cationic gemini surfactants with polymethylene spacer of variable length were used for the interaction with DNA. The scattering experiments were performed at constant DNA and sodium bromide concentrations and variable surfactant concentration in the premicellar and micellar regions as a function of surfactant spacer length. It was found that the DNA conformation strongly depends on the polymethylene spacer length as well as on the surfactant concentration relative to the surfactant critical micelle concentration. Gemini surfactant molecules with 4 methylene groups in the spacer were found to be the least efficient DNA compacting agent in the region above the surfactant *cmc*. Gemini molecules with the shortest spacer length (2 methylene groups) and the longest spacer length (8 methylene groups) investigated showed the most efficient DNA compaction ability.

Key words: Gemini surfactant — DNA — Hydrodynamic diameter — Micelle — Surfactant spacer

Abbreviations: 12-*s*-12, alkanediyl-α-ω-bis(dimethyldodecylammonium bromide); DMAO, dodecyldimethylamine oxide; *c,* surfactant concentration; *cac,* critical aggregation concentration; *cmc,* critical micelle concentration; d_{h0}, hydrodynamic diameter extrapolated to zero scattering angle; *D,* translation diffusion coefficient; D_0, translation diffusion coefficient extrapolated to zero scattering angle.

Introduction

Cationic amphiphiles are able to compact and stabilize DNA through attractive electrostatic interactions between surfactant cations and anionic phosphate groups of DNA as well as the hydrophobic interaction among chains of surfactant molecules. The important role of cationic surfactant molecules in a complex with DNA is to protect it from the effect of endogenous nucleases. The hydrophobic parts of surfactant molecules may also allow DNA to escape from endosome through endosomal membrane.

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DNA/conventional cationic surfactant complex

Binding of single chain ammonium surfactant ions with various chain length (dodecyltrimethylammonium and tetradecyltrimethylammonium ion) by DNA was investigated by cationic surfactant-selective electrodes. The data revealed that both investigated cations can bind to DNA at a very low equilibrium concentration and the longer chain surfactant ion (tetradecyltrimethylammonium ion) is more easily bound by DNA indicating the importance of the hydrophobic interaction (Hayakawa et al. 1983). As indicated by the results of surfactant ion-selective electrode measurements, the binding of DNA to hexadecyltrimethylammonium bromide followed two-stage first-order kinetics for denatured (single-stranded) DNA and three-stage first-order kinetics for native DNA (Maulik et al. 1998). The collapsing effect from extended DNA coils to

compacted globules upon the addition of single chain cationic surfactant was observed by fluorescence microscopy and dynamic light scattering techniques (Dias et al. 2004). From the small-angle X-ray scattering measurements it follows that DNA/single chain cationic surfactants complexes form hexagonal structures for longer chain surfactants (dodecyl-, tetradecyl- and hexadecyltrimethylammonium bromides) (Miguel et al. 2003).

DNA/dodecyltrimethylammonium bromide/NaBr phase diagrams were studied in ethanol/water solution as a function of surfactant concentration. High ethanol content results in the complex precipitation from the solution. It was found that dodecyltrimethylammonium ions were exchanged by Na ions in this precipitate (McLoughlin et al. 2000). Complexation of DNA with dodecyldimethylamine oxide (DMAO) takes place at a critical degree of DMAO protonation and is therefore primarily controlled by the cationic/non-ionic DMAO form ratio and not by the ratio DNA/surfactant (Wang et al. 2001).

At the interfaces, complexation of DNA and dodecyltrimethylammonium bromide occurs even below the critical aggregation concentration (*cac*) and results in the neutralization of surfactant ions by phosphate groups of free DNA chains. Above the critical aggregate concentration, cooperative binding of surfactant on DNA occurs which leads to the formation of thick surface layers (McLoughlin and Langevin 2004). For the DNA complex with dioctadodecyltrimethylammonium bromide (one head, two alkyl chains) at the air/water interface, the surface pressure vs. area isotherm of the cationic monolayer is shifted to larger molecular areas and the transition between liquid expanded and liquid condensed state of the monolayer disappears upon the addition of DNA (Cárdenas et al. 2005a).

On hydrophobic solid surfaces, the presence of the cationic surfactant leads to the increase in the DNA amount adsorbed onto the solid surface as well as to the significant compaction of DNA layer (Cárdenas et al. 2005b). DNA complexed with hexadecyltrimethylammonium bromide shows the synergistic increase in the amount adsorbed on hydrophobic silica surface as compared to the adsorbed amounts of individual surfactant and DNA (Cárdenas et al. 2003). It was also found that neither DNA molecular weight nor its conformation influences the adsorption of DNA/surfactant complex on the hydrophobic surface (Cárdenas et al. 2003).

DNA/double chain gemini cationic surfactant complex

Gemini surfactants are a class of amphiphilic molecules containing two head groups and two aliphatic chains linked by a rigid or flexible spacer at the level of head groups. They show very potent physicochemical and biological properties relative to corresponding conventional (single chain, single head group) surfactants. The gemini surfactants with a polymethylene spacer denoted as *m*-*s*-*m*, where *m* means the number of carbon atoms in the alkyl chains and *s* is the number of carbon atoms in the spacer, show minimum of compaction efficiency (as a function of variable spacer length) in the complex with DNA at the spacer length $s = 6$ and the fixed alkyl chain lengths (Karlsson et al. 2002). The same results were found at the air/water interface where the spacer length $s = 6$ was found as the turning point of the DNA/12-*s*-12 complex in terms of the spacer conformation. The extrapolated molecular area and collapse pressure of the monolayer show maximum and minimum value, respectively, at $s = 6$ which is believed to be caused by the spacer acquiring the U-shape conformation due to its bending into the air phase (Chen et al. 2002). The fluorescence study of DNA/12-3-12 gemini surfactant showed only small dependence of the *cac* of the complex on the electrolyte (NaBr) concentration (Zhao et al. 2007). From the isothermal titration microcalorimetry experiments it follows that the surfactant dissymmetry (non-equal length of both alkyl chains in a gemini surfactant molecule) shows noticeable effect on the interaction of DNA and gemini surfactant (Jiang et al. 2005). The *cac* decreases with the increased dissymmetry ratio m_1/m_2 (m_1 and m_2 are the chain lengths of both alkyl chains in surfactant molecule), so does the Gibbs free energy change indicating more spontaneous aggregation with the increasing m_1/m_2 value (Jiang et al. 2005). The small-angle X-ray scattering experiments performed with the DNA/*m*-4-*m* gemini surfactant complex revealed the long-range organization of DNA complexed with the long-chain gemini surfactants (m = 12–16) (Uhríková et al. 2005).

Complexes of DNA and gemini surfactants with lipids form lamellar lipid bilayers intercalated with parallel DNA strands (Uhríková et al. 2002). Behavior of nucleo-gemini surfactants at the air-water interface (gemini surfactants with the bromide counterions exchanged with nucleotides) was studied (Wang et al. 2005). Nucleoamphiphiles formed by the complexation of cationic gemini *m*-2-*m* surfactants with ethylene spacer and anionic nucleotides – uracil 5'-monophosphate or adenine 5'-monophosphate were investigated by surface pressure measurements, Brewster angle microscopy and FTIR-ATR (Fourier transform infrared-attenuated total reflectance) spectroscopy. Although no pH value is specified in the reference, the conditions were chosen so that two nucleic acids form a complex with single gemini molecule. Fluid solutions of nucleo-gemini surfactants indicate intensive aggregation and transition to hydrogel state upon addition of complementary DNA bases or other nucleo-gemini surfactants with complementary bases (Wang et al. 2005). A new cationic divalent surfactant based on arginine (arginine-N-lauroyl amide dihydrochloride) formed vesicles with anionic surfactant (sodium hexadecylsulfate) and its interaction in vesicular form with DNA was investigated

(Rosa et al. 2007). It was found that the phase behavior of DNA/catanionic vesicle complex is similar to that of DNA complexed with conventional cationic surfactants with the advantage of non-toxicity and suitability for real biological applications (Rosa et al. 2007). The present study is focused on the detailed determination of the hydrodynamic size of DNA/12-*s*-12 gemini surfactant complex as a function of surfactant spacer length throughout the premicellar and micellar region of surfactant concentrations to get better knowledge of DNA/gemini surfactant interactions with the view of prospective application of the DNA/gemini surfactant complex in gene therapy.

Materials and Methods

Gemini surfactants alkanediyl-α-ω-bis(dimethyldodecylam monium bromide) $C_{12}H_{25}$-$(CH_3)_2N^+$-$(CH_2)_s$-$N^+(CH_3)_2$-$C_{12}H_{25}$·$2Br^-$ (hereinafter referred to as 12-*s*-12, *s* is the number of carbon atoms in the spacer) were prepared by the reaction of tertiary diamine with 1-bromoalkane as described previously (Imam et al. 1983). The product was purified by multiple crystallization from acetone-methanol mixture. Thin layer chromatography and elemental analysis confirmed the identity of the compounds.

DNA from salmon sperm (Fluka, molecular weight 3 kbp) was dissolved in 2×10^{-3} mol/l NaBr solution at the concentration 0.5×10^{-3} mol/l bp. In the DNA/NaBr stock solution, gemini surfactants 12-*s*-12 (*s* = 2, 4, 6, 8) were dissolved at the following concentration relative to the surfactant critical micelle concentration (*cmc*) in 0.002 mol/l NaBr solution: 0.01 *cmc*, 0.1 *cmc*, *cmc* and 10 *cmc*. Surfactant *cmc* values in 0.002 mol/l NaBr solution previously determined from surface tension measurements were found in the region 1.02×10^{-4}–1.52×10^{-4} mol/l for spacer lengths *s* = 2 to 8.

Dynamic light scattering

A Brookhaven light scattering system (BI 9000 AT digital correlator, 200 SM goniometer and argon laser 514.5 nm wavelength) was used for the dynamic light scattering measurements. Scattered intensity was registered in the angular range 20–135° at 25°C. Solutions for the light scattering experiments were prepared using deionized water which was additionally filtered for mechanical impurities through the syringe filters with 0.8 μm pore size.

The autocorrelation curve was analyzed by the method of cumulants up to the second cumulant. The slope Γ of the autocorrelation function at zero correlation time is expressed as $\Gamma = D\,q^2$, where *D* is translational diffusion coefficient of aggregate. Scattering vector *q* is equal to $4\,\pi\,n/\lambda\,\sin^2\,\theta/2$ where θ is scattering angle, λ is wavelength of the incident laser beam, and *n* is refractive index of solvent. Translational diffusion coefficient D_0 is given by the extrapolation of the quantity Γ/q^2 to zero scattering angle

$$D_0 = \lim_{q \to 0} \Gamma / q^2 \tag{1}$$

Hydrodynamic diameter d_{h0} extrapolated to zero scattering angle is calculated from the Stokes-Einstein formula

$$d_{h0} = \frac{k\,T}{3\,\pi\,\eta\,D_0} \tag{2}$$

η is solvent viscosity, k is the Boltzmann constant and *T* is absolute temperature. For each surfactant and at each scattering angle, 5 independent measurements and calculations of the autocorrelation function were carried out. Mean values and standard deviations of Γ/q^2, D_0, and d_{h0} were calculated. Particle size spectra were calculated based on the inverse Laplace transformation from autocorrelation function using the constrained regularization program for inverting noisy linear algebraic and integral equations – Contin algorithm (Provencher 1982).

Due to the large number of data to be processed, a custom application software written in Visual Basic was used for automated data format conversion from measurement files.

Results and Discussion

It is known from previous studies of DNA-cationic surfactant systems based on fluorescence microscopy (Karlsson et al. 2002; Dias et al. 2008; Gaweda et al. 2008) and dynamic light scattering experiments (Dias et al. 2005; Gaweda et al. 2008) that DNA undergoes compaction from extended coil to globular state due to the presence of cationic macroions in the solution. This DNA compaction occurs at a certain surfactant concentration roughly corresponding to the *cac* which was found to be 4–5 μmol/l for DNA/hexadecyltrimethylammonium bromide (CTAB) system (Dias et al. 2008). It was also determined for DNA/gemini 12-*s*-12 surfactant system (Karlsson et al. 2002) by means of fluorescence microscopy with the approximate values 0.047, 0.25, 0.48, 0.39 μmol/l, for gemini surfactants 12-2-12, 12-4-12, 12-6-12, 12-8-12, respectively.

In Fig. 1, the dependence of the quantity Γ/q^2 as a function of scattering vector *q* is shown for the DNA/12-*s*-12 complex at different surfactant concentrations relative to surfactant *cmc*. It should be noted that *cmc* values of surfactants in 0.002 mol/l NaBr aqueous solution are smaller (almost by one order of magnitude, see the experimental section and results published by Danino et al. (1995)) than those of gemini surfactants in electrolyte-free aqueous solution. This results from screening of electrostatic repulsion of surfactant

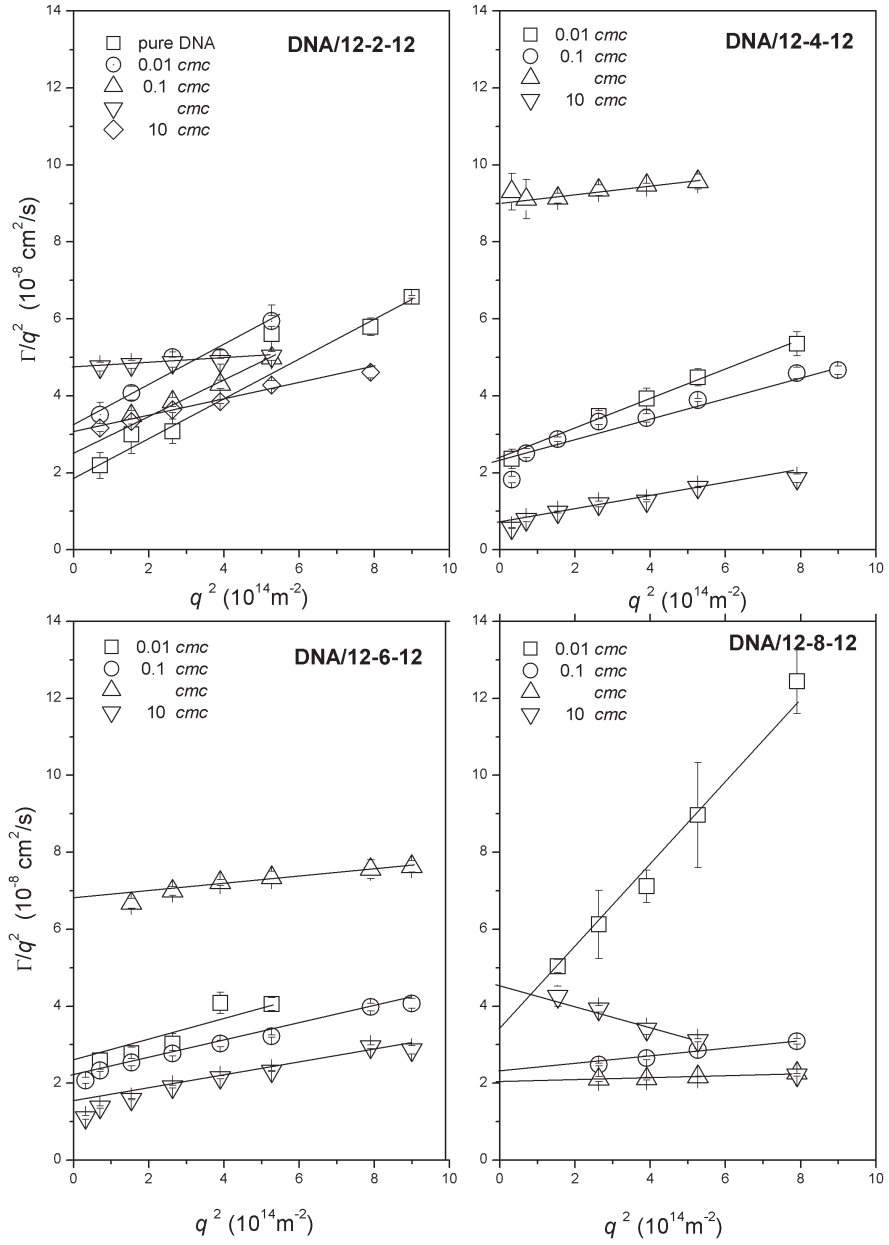

Figure 1. Dependence of Γ/q^2 vs q^2 (Γ – slope of the autocorrelation function at zero correlation time, q – scattering vector) of the DNA/12-*s*-12 complex for the following values of surfactant concentration 0.01*cmc*, 0.1 *cmc*, *cmc*, 10 *cmc* and surfactant spacer number values s = 2, 4, 6, 8.

heads due to the presence of electrolyte ions in the solution. The quantity Γ/q^2 extrapolated to zero scattering vector provides the value of D_0 (Eq. (1)). Hydrodynamic diameter calculated from the Stokes-Einstein equation (Eq. (2)) is plotted in Fig. 2 and the data for D_0, and d_{h0} are shown in Table 1. The slopes of individual Γ/q^2 lines plotted in Fig. 1 are shown in Fig. 3 and Table 1 as a function of surfactant concentration.

Region of premicellar surfactant concentration

The smallest surfactant concentration used in the investigated DNA/surfactant complex was 0.01 *cmc* which represents concentrations from 1.52×10^{-6} to 1.02×10^{-6} mol/l for gemini surfactants 12-*s*-12 with the spacer length s = 2, 4 ,6, 8. These values are above the region of DNA/12-*s*-12 *cac* for surfactants with the stated spacer length, as

Table 1. Values of diffusion coefficient D_0, hydrodynamic diameter d_{h0} extrapolated to zero scattering angle and the slope $d\Gamma/q^2/dq^2$ of DNA/12-*s*-12 complex as a function of the surfactant spacer number *s* and surfactant concentration *c*/*cmc*

DNA/12-*s*-12	*c*/*cmc*	*c* (10^{-4} mol/l)	D_0 (10^{-8} cm^2/s)	d_{h0} (nm)	$d\Gamma/q^2/dq^2$ (10^{-4} cm^4/s)
DNA	–	–	2.05	242	0.52
DNA/12-2-12	0.01	0.015	3.57	139	0.38
	0.1	0.151	3.14	158	0.29
	1.0	1.51	4.72	105	0.06
	10.0	15.10	3.05	163	0.21
DNA/12-4-12	0.01	0.013	2.54	195	0.36
	0.1	0.13	2.41	206	0.28
	1.0	1.31	9.13	54	0.08
	10.0	13.10	0.74	669	0.15
DNA/12-6-12	0.01	0.013	2.43	205	0.23
	0.1	0.127	2.17	229	0.22
	1.0	1.27	6.90	72	0.08
	10.0	12.70	1.39	356	0.18
DNA/12-8-12	0.01	0.010	2.97	167	1.17
	0.1	0.102	2.20	225	0.12
	1.0	1.02	1.98	251	0.04
	10.0	10.20	4.74	105	–0.32

shown in the text above (Karlsson et al. 2002). DNA chains are compacted and interactions between DNA globules and non-micellized gemini molecules are present in this surfactant concentration region. The concentration dependence of the complex hydrodynamic size is rather weak in this region of surfactant concentration (Fig. 2). In other words, no further compaction of DNA globules is achieved upon the concentration increase of non-micellized gemini molecules in the solution. Also, there is minimum difference in the DNA/surfactant complex hydrodynamic size observed as a function of the spacer number *s* at 0.01 *cmc*. At 0.1 *cmc*, significantly lower size was found for DNA/12-2-12 complex which means more efficient DNA compaction as compared to gemini molecules with other spacer numbers (Fig. 2). This result generally corresponds with the finding of most efficient DNA compaction by gemini molecules with short spacers (*s* = 2, 3) (Karlsson et al. 2002). The molecule spacer is so short that DNA compaction results from DNA interaction with quasi-single head surfactant molecule with double positive charge which can more efficiently (without steric limitations resulting from various molecule conformations predetermined by its spacer geometry) adsorb on phosphate groups of DNA chains.

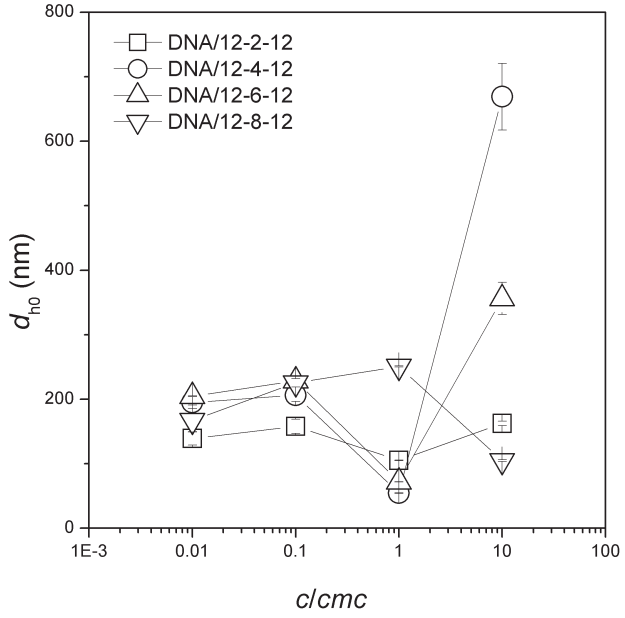

Figure 2. Dependence of hydrodynamic diameter d_{h0} extrapolated to zero scattering angle of the DNA/12-*s*-12 complex as a function of the surfactant concentration *c*/*cmc* for surfactant spacer number values *s* = 2, 4, 6, 8.

Region of critical micelle concentration

Significant DNA compaction was observed for gemini molecules with the spacer length *s* = 2, 4, 6 (Figs. 1 and 3). The mean hydrodynamic diameter of DNA compacted by 12-*s*-12 gemini molecules at the surfactant *cmc* is in the region 50–100 nm for spacers *s* = 2, 4, 6 (Table 1). The method of

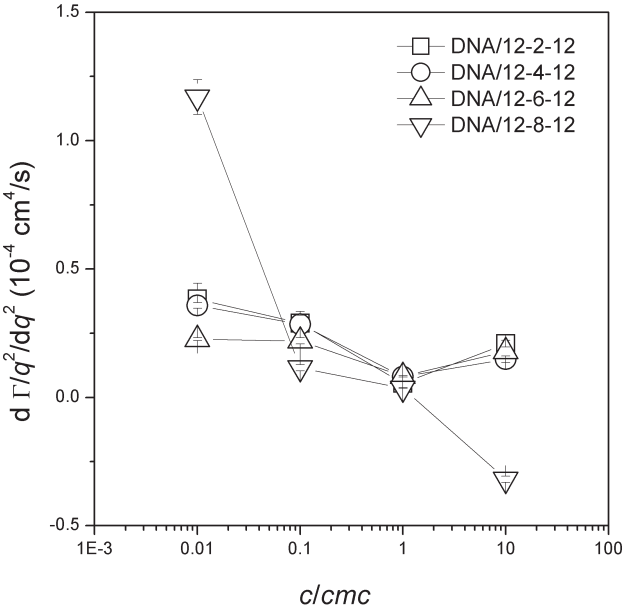

Figure 3. The slope $d\Gamma/q^2/dq^2$ from the dependence plotted in Fig. 1 as a function of the surfactant concentration *c*/*cmc* for surfactant spacer number values *s* = 2, 4, 6, 8.

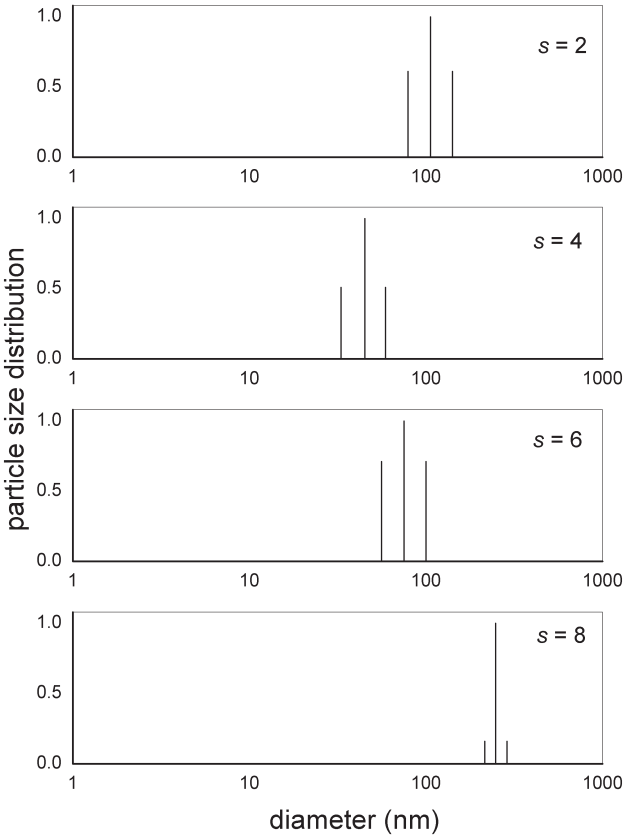

Figure 4. Particle size distribution of DNA/12-*s*-12 complex at surfactant *cmc* for gemini surfactants with the spacer length *s* = 2, 4, 6, 8.

cumulants (as a result of Taylor series expansion of logarithm of the autocorrelation function) giving only mean values of diffusion coefficient and hence, of aggregate size, provides no information on aggregate size polydispersity. Therefore, numerical solutions based on the inverse Laplace transformation (Contin algorithm) were applied to obtain the particle size spectra. The spectra were recorded at each scattering angle. For the sake of simplicity, only spectra at the scattering angle 90° are shown for *s* = 2, 4, 6, 8. For the DNA complex with 12-2-12, 12-4-12, 12-6-12, and 12-8-12, the size peak value was found at 106, 45, 75, and 248 nm, respectively (Fig. 4). The size spectra are unimodal indicating the presence of only single particle size in the system. The values of particle peaks roughly correspond to the hydrodynamic size values provided by the cumulants method (Table 1).

Region of surfactant micelles

In this region of surfactant concentration ten times above the *cmc*, a noticeable increase in hydrodynamic size of the DNA/12-*s*-12 complex can be observed for gemini molecules with the spacer length *s* = 4, 6. Again, almost no increase in size was found for the DNA/12-2-12 complex at this high surfactant concentration which indicates efficient compaction ability of 12-2-12 throughout the premicellar and micellar concentration region. The largest hydrodynamic size of the complex was observed for gemini molecules with the spacer length *s* = 4 (Fig. 2). The large mean hydrodynamic size provided by the cumulants evaluation method may indicate decompaction of DNA globules for this gemini surfactant. Particle size spectra (Fig. 5) show for *s* = 4 bimodal size distribution with two peaks. The size values may indicate coexistence of uncompacted DNA coils several hundreds of nanometers large in size which were observed by the dynamic light scattering method (Dias et al. 2005) and of some kind of aggregates with intermediate size between 12-4-12 gemini micelles in aqueous electrolyte-free solution with the diameter lower than 10 nm (Pisárčik et al. 1998) and compacted DNA globules of 100 nm size (Dias et al. 2005). It should be brought to mind that the addition of NaBr electrolyte to surfactant solution decreases not only its *cmc* value but also promotes micelle growth (and also causes the sphere-to-rod micellar shape transition at high electrolyte concentration) which may result in larger micelle sizes in electrolyte solution, as compared to those in electrolyte-free aqueous solution. Therefore, we assume that the first peak in the DNA/12-4-12 particle size spectrum could be attributed to free gemini

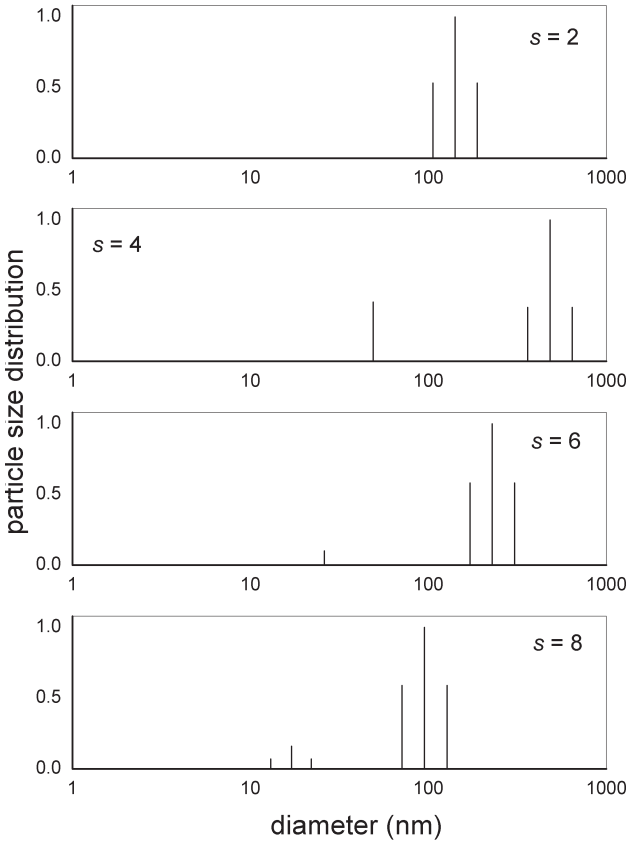

Figure 5. Particle size distribution of DNA/12-*s*-12 complex at the surfactant concentration 10 *cmc* for gemini surfactants with the spacer length *s* = 2, 4, 6, 8.

micelles which are formed in the solution at the cost of interaction of gemini molecules with DNA which would lead to its efficient compaction. The diminishing peak of free micelles can be observed also for DNA/12-6-12 and DNA/12-8-12 complexes. Due to their spacer rigidity, gemini molecules with intermediate spacer length (12-4-12 and 12-6-12) act as the "worst" DNA compacting agents at high surfactant concentration which corresponds with previous results from fluorescence microscopy (Karlsson et al. 2002) and measurements at the air/water interface (Chen et al. 2002).

12-8-12

As seen in Figs. 2 and 3, the DNA/12-8-12 complex shows different behavior to that found for DNA with other investigated gemini surfactants. Two observations can be made. First, there is no decrease in size of the DNA/12-8-12 complex at the *cmc* observed and second, the decrease in the complex size is found with the increasing surfactant concentration in the micellar region, as opposed to the DNA decompaction tendency caused by gemini molecules with the spacer length *s* = 4, 6. Also, the slope of Γ/q^2 vs. q^2 changes from the positive value (premicellar region) through zero (*cmc* region) to the negative value (micellar region). This different behavior may be attributed to the increased interaction between 12-8-12 surfactant bisammonium cations and phosphate groups on DNA chains which results in effective DNA compaction even in the region above the surfactant *cmc* where gemini molecules with stiff spacer of intermediate length fail to compact DNA (Fig. 2). The increased interaction of 12-8-12 with DNA may be due to the better availability of bisammonium cations for phosphate groups which results from a more flexible spacer. This behavior corresponds with the observed efficient DNA compaction ability for 12-*s*-12 gemini molecules with either short spacers (*s* = 2, 3) or with spacers which are long enough (*s* = 8, 10) to efficiently interact with DNA (Karlsson et al. 2002).

Conclusions

The presented dynamic light scattering study of DNA/12-*s*-12 cationic gemini surfactant showed various efficiency of gemini surfactant molecules on DNA compaction as a function of surfactant spacer length. Whereas gemini molecules with all spacer lengths investigated are able to compact DNA in their premicellar region, significant differences in the DNA compaction efficiency occur in the micellar region. Gemini surfactant molecules with 4 methylene groups in the spacer were found to be the least efficient DNA compacting agent in the region above the surfactant *cmc*. Gemini molecules with the shortest spacer length (2 methylene groups) and the longest (8 methylene groups) spacer length investigated showed the most efficient DNA compaction ability which is to relate to spacer geometry and stiffness. This supports the importance of structure, charge distribution and geometry of gemini surfactant molecule when interacting with oppositely charged polyelectrolyte.

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References

Cárdenas M., Braem A., Nylander T., Lindman B. (2003): DNA Compaction at Hydrophobic surfaces induced by a cationic amphiphile. Langmuir **19,** 7712–7718; doi:10.1021/la026747f

Cárdenas M., Nylander T., Jönnson B., Lindman B. (2005a): The interaction between DNA and cationic lipid films at the air-water interface. J. Colloid Interface Sci. **286,** 166–175; doi:10.1016/j.jcis.2005.01.008

Cárdenas M., Nylander T., Lindman B. (2005b): DNA and cationic surfactants at solid surfaces. Colloids Surf., A **270–271,** 33–43; doi:10.1016/j.colsurfa.2005.05.034

Chen X., Wang J., Shen N., Luo Y., Lin L., Liu M., Thomas R. K. (2002): Gemini surfactant/DNA complex monolayers at the air-water interface: effect of surfactant structure on the assembly, stability, and topography of monolayers. Langmuir **18,** 6222–6228; doi:10.1021/la025600l

Danino D., Talmon Y., Zana R. (1995): Alkanediyl-α,ω-bis(dimethyla lkylammonium bromide) surfactants (dimeric surfactants). 5. Aggregation and microstructure in aqueous solutions. Langmuir **11,** 1448–1456; doi:10.1021/la00005a008

Dias R. S., Pais A. A. C. C., da Miguel M. G., Lindman B. (2004): DNA and surfactant in bulk and at interfaces. Colloids Surf., A **250,** 115–131; doi:10.1016/j.colsurfa.2004.07.026

Dias R. S., Innerlohinger J., Glatter O., Miguel M. G., Lindman B. (2005): Coil-globule transition of DNA molecules induced by cationic surfactants: a dynamic light scattering study. J. Phys. Chem. B **105,** 10458–10463; doi:10.1021/jp0444464

Dias R. S., Magno L. M., Valente A. J. M., Das D., Das P. K., Maiti S., Miguel M. G., Lindman B. (2008): Interaction between DNA and cationic surfactants: effect of DNA conformation and surfactant headgroup. J. Phys. Chem. B **112,** 14446–14452; doi:10.1021/jp8027935

Gaweda S., Morán C., Pais A. A. C. C., Dias R. S., Schillén K., Lindman B., Miguel M. G. (2008): Cationic agents for DNA compaction. J. Colloid Interface Sci. **323,** 75–83; doi:10.1016/j.jcis.2008.04.006

Hayakawa K., Santerre J. P., Kwak J. C. T. (1983): The binding of cationic surfactant by DNA. Biophys. Chem. **17,** 175–181; doi:10.1016/0301-4622(83)87001-X

Imam T., Devínsky F., Lacko I., Krasnec L. (1983): Preparation and antimicrobial activity of some new bisquaternary ammonium salts. Pharmazie **38,** 308–310

Jiang N., Wang J., Wang Y., Yan H., Thomas R. K. (2005): Microcalorimetric study on the interaction of dissymmetric gemini surfactants with DNA. J. Colloid Interface Sci. **284,** 759–764; doi:10.1016/j.jcis.2004.10.055

Karlsson L., van Eijk M. C. P., Södermann O. (2002): Compaction of DNA by gemini surfactants: effects of surfactant architecture. J. Colloid Interface Sci. **252,** 290–296; doi:10.1006/jcis.2002.8477

Maulik S., Chattoraj D. K., Moulik S. P. (1998): Biopolymer-surfactant interaction: 3. Kinetics of binding of cetyltrimethylammonium bromide to deoxyribonucleic acid. Colloids Surf., B **11,** 57–65

McLoughlin D. M., McManus J. J., Gorelov A. V., Dawson K. A. (2000): DNA complexes with cationic surfactant in mixed solvents: the influence of excess surfactant and salt. Prog. Coll. Polym. Sci. **115,** 186–191; doi:10.1007/3-540-46545-6_38

McLoughlin D., Langevin D. (2004): Surface complexation of DNA with cationic surfactant. Colloids Surf., A **250,** 79–87; doi:10.1016/j.colsurfa.2004.04.096

Miguel M. G., Pais A. A. C. C., Dias R. S., Leal C., Rosa M., Lindman B. (2003): DNA-cationic amphiphile interactions. Colloids Surf., A **228,** 43–55; doi:10.1016/S0927-7757(03)00334-0

Pisárčik M., Dubničková M., Devínsky F., Lacko I., Škvarla J.(1998): Dynamic light scattering and electrokinetic potential of bis(quarternary ammonium bromide) surfactant micelles as the function of the alkyl chain length. Colloids Surf., A **143,** 69–75; doi:10.1016/S0927-7757(98)00497-X

Provencher S. W. (1982): CONTIN: A general purpose constrained regularization program for inverting noisy linear algebraic and integral equations. Comput. Phys. Commun. **27,** 229–238; doi:10.1016/0010-4655(82)90174-6

Rosa M., Miguel M. G., Lindman B. (2007): DNA encapsulation by biocompatible catanionic vesicles. J. Colloid Interface Sci. **312,** 87–97; doi:10.1016/j.jcis.2006.07.084

Uhríková D., Rapp G., Balgavý P. (2002): Condensed lamellar phase in ternary DNA-DLPC-cationic gemini surfactant system: a small-angle synchrotron X-ray diffraction study. Bioelectrochemistry **58,** 87–95; doi:10.1016/S1567-5394(02)00122-6

Uhríková D., Zajac I., Dubničková M., Pisárčik M., Funari S. S., Rapp G., Balgavý P. (2005): Interaction of gemini surfactants butane-1,4-diyl-bis(alkyldimethylammonium bromide) with DNA. Colloids Surf., B **42,** 59–68

Wang Y., Dubin P. L., Zhang H. (2001): Interaction of DNA with cationic micelles: effects of micelle surface charge density, micelle shape, and ionic strength on complexation and DNA collapse. Langmuir **17,** 1670–1673; doi:10.1021/la0010673

Wang Y., Desbat B., Manet S., Aimé C., Labrot T., Oda R. (2005): Aggregation behaviors of gemini nucleotide at the air-water interface and in solutions induced by adenine-uracil interaction. J. Colloid Interface Sci. **283,** 555–564; doi:10.1016/j.jcis.2004.09.003

Zhao X., Shang Y., Liu H., Hu Y. (2007): Complexation of DNA with cationic gemini surfactant in aqueous solution. J. Colloid Interface Sci. **314,** 478–483; doi:10.1016/j.jcis.2007.04.059

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