

Final Report on the results of OTKA/NKFIH K 108646 project entitled: "Nonequilibrium aspects of polyelectrolyte based nanoassemblies"

The present project focused on the systematic investigations of aqueous nanoassemblies of polyelectrolytes (PE) formed in the presence of oppositely charged surfactants (S) and other additives. In accordance with the original research plan, the work was divided into four work packages including *i)* the study of the trapped nonequilibrium aggregate formation; *ii)* the understanding of role of polyelectrolyte chemistry; *iii)* the development of novel stable, composite nanoparticles and *iv)* the elaboration of the link between the nonequilibrium bulk and interfacial association of PE/S mixtures including biomacromolecules. Similarly, our final report is also organized into four parts as shown in the subsequent sections. We should note, however, that according to the decision of the OTKA KM1 Jury, only roughly the half of the originally planned budget was allocated to the OTKA 108646 K project. Specifically, the purchase of the planned Nanoparticle Tracking Analysis (NTA) instrument was not supported. To achieve the primarily goals of the proposal, a high intensity laser source was ordered and installed for the extension of the already existing Dynamic Light Scattering (DLS) equipment (with the written permission of the OTKA KM1 Jury). Thus, instead of the NTA, highly accurate DLS and turbidity measurements were utilized for the monitoring of the aggregation processes and the mean size of the polyelectrolyte nanoassemblies with oppositely charged compounds. Nevertheless, some limited number of NTA measurements were carried out for the characterization of polyelectrolyte/surfactant aggregate formation, within a new collaboration with J. Selyehó University. However, the main body of the research has been carried out in the principal scientist's laboratory.

WP1. Controlling the Size, Composition and Stability of Nonequilibrium PE/S Aggregates

1.1 Impact of electrolyte and polymer concentration gradients on poly(diallyldimethylammonium chloride) (PDADMAC)/sodium dodecyl sulfate (SDS) association. ([1], O/1, P/2)

In the first work package, the focus was on the detection of potential compositional heterogeneity of oppositely charged polyelectrolyte/surfactant complexes and its consequences on the nonequilibrium phase properties. For this aim the association of poly(diallyldimethylammonium chloride) (PDADMAC) and sodium dodecyl sulfate (SDS) was investigated systematically in the presence of NaCl through dynamic light scattering (DLS, 3D LS), turbidity and electrophoretic mobility measurements. For the preparation of the mixtures three different mixing procedures were applied. At first, we mixed equal volumes of dilute polyelectrolyte and surfactant solutions at constant concentration of NaCl. In the second and third methods, concentrated polyelectrolyte solution was mixed with dilute surfactant solution, where both or only the polyelectrolyte solution contained NaCl. We have found, that the aggregation of PDADMAC/SDS assemblies was largely dependent on the mixing procedures at surfactant excess, where the use of concentrated polyelectrolyte resulted in large PE/S aggregates. In polyelectrolyte excess, however, the impact of mixing methods diminished. In this concentration range, highly reproducible changes in the turbidity, scattered intensity and mean size of the assemblies were observable over a long time period (from hours to days, see characteristic examples in Figure 1) in sharp contrast with the negligible time dependence of these parameters at surfactant excess. This phenomenon was not reported before, and we attribute it to a specific and slow exchange process initiated by the inhomogeneous distribution of surfactants among the polyelectrolyte chains of different lengths.

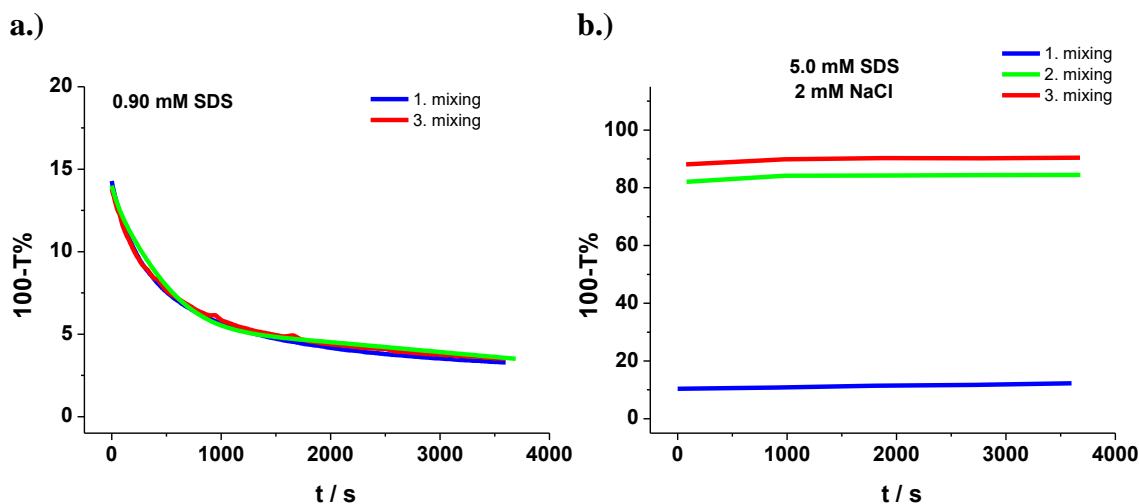


Figure 1. Variation of the turbidity with time of PDADMAC/SDS systems (in the presence of 2 mM NaCl) prepared by different mixing protocols at **a.)** polyelectrolyte excess (0.90 mM SDS) and **b.)** surfactant excess (5 mM SDS). The width of the lines is commensurable with the standard error of turbidity measurements ($c_{\text{PDADMAC}}=250 \text{ mg} \cdot \text{dm}^{-3}$).

The compositional heterogeneity of the PE/S assemblies is also supported by the bimodal distribution of the electrophoretic mobility data (see Figure 2). The results indicate that slightly charged large aggregates may coexist with highly charged small polyion chains with a low amount of bound surfactants. Our observations clearly show that depending on the composition, the local concentration gradients or the polydispersity of the polyelectrolyte can significantly affect the nonequilibrium features as well as the dynamics of PE/S assembly formation.

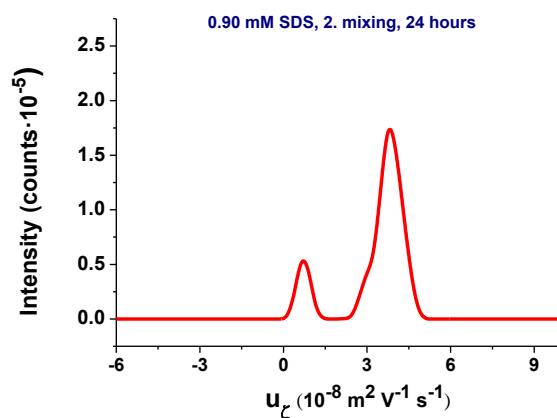


Figure 2. Bimodal electrophoretic mobility distribution of PDADMAC/SDS assemblies measured 24 hours after their preparation at 0.90 mM SDS and $250 \text{ mg} \cdot \text{dm}^{-3}$ PDADMAC final concentrations.

1.2 Effect of dilution on nonequilibrium aggregate formation of sodium poly(styrene sulfonate) (PSS) and dodecyltrimethylammonium bromide (DTAB) in the presence of salt. ([2], O/3, P/2)

The polyelectrolyte/surfactant mixtures play a distinguished role in the efficacy of shampoos and toiletries, through the deposition of PE/S precipitates on the hair surface upon dilution of the formulations. The classical interpretation of this dilution/deposition phenomenon is related to the equilibrium phase properties of the system e.g. to a composition change during which

the system enters into the two-phase region. Our studies, however, clearly indicated that the phase properties of polyelectrolyte/surfactant mixtures could also be affected by the applied solution preparation protocols and electrolyte additives. In the present project, we have studied the impact of dilution on the nonequilibrium aggregate formation in PSS/DTAB/NaCl mixtures using electrophoretic mobility, turbidity, density, SAXS and ATR-IR measurements. Mixtures prepared with a 100 fold dilution of concentrated PSS/DTAB/NaCl solutions in water were compared with those systems made by rapid 1:1 volume mixing of dilute PSS/NaCl and DTAB/NaCl solutions. The study revealed that the phase-separation concentration range as well as the composition, morphology and visual appearance of the precipitates was significantly different in the two cases. The photos as well as the SAXS and ATR-IR data are consistent with the formation of more amorphous, sticky precipitates upon dilution of the PSS/DTAB/NaCl concentrate as compared to the systems prepared at the same final concentrations of the compounds via mixing of the dilute polyelectrolyte and surfactant solutions (see Figure 3).

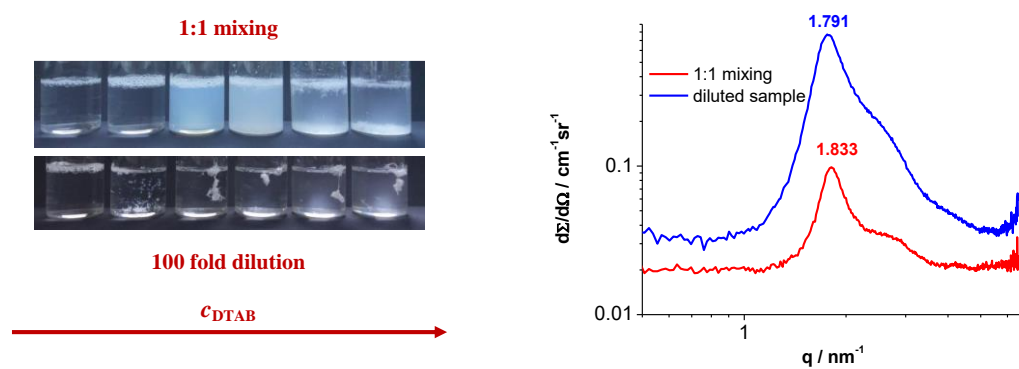


Figure 3. Visual appearance and one dimensional scattering patterns of PSS/DTAB/NaCl precipitates prepared by the different methods; $c_{\text{DTAB}} = 5 \text{ mM}$, $c_{\text{PSS}} = 500 \text{ ppm}$, $c_{\text{NaCl}} = 6 \text{ mM}$; $t = 25 \text{ }^{\circ}\text{C}$.

Summarizing, our observations clearly demonstrate that, in addition to the equilibrium phase properties, the dilution/deposition process is also strongly related to the formation of nonequilibrium PE/S aggregates which can be used to modulate the efficiency of various commercial applications.

WP2. Impact of the Nature of Polyelectrolyte Charge on the Nonequilibrium Aggregate Formation

2.1 Thermodynamic stability of the aqueous mixtures of sodium poly(vinyl sulfate)-poly(vinyl alcohol) (PVAS) copolymer (10.6 mole% vinyl sulfate monomers) with dodecyl (DTAB) or hexadecyltrimethylammonium bromide (CTAB) ([6], O/5, P/4)

The impact of the slow- and the fast- mixing protocols was studied systematically on the turbidity, electrophoretic mobility and size of PVAS/CTAB complexes at different ionic strengths. We proposed a stepwise cationic surfactant binding mechanism onto the PVAS molecules (see Figure 4a). At surfactant concentrations below charge neutralization, the cationic surfactant molecules bind to the vinyl sulfate groups, whereas at larger concentrations, they could adsorb onto the surface of the charge neutralized PVAS/CTAB assemblies. Thus, at small excess of the surfactant, charge stabilized dispersions of PVAS/CTAB nanophases can be prepared via the application of rapid-mixing, whereas the slow-mixing procedure leads to phase separation. However, in sharp contrast to the earlier investigated homopolyelectrolyte/surfactant mixtures, a further increase of the surfactant

concentration led to an additional CTAB binding on the nonionic units of PVAS. This binding leads to the reswelling of the individual polyion/surfactant complexes and their resolubilization into a thermodynamically stable solution. It was also observed that the nonequilibrium states were absent for the PVAS/DTAB mixtures due to the low extent of charge reversal of PE/S aggregates as well as for the PVAS/CTAB mixtures at high ionic strengths where the charge stabilization mechanism did not work.

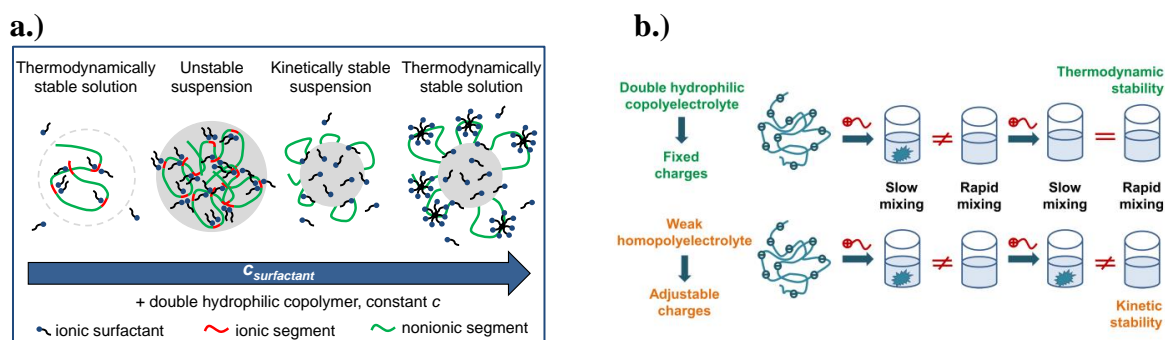


Figure 4. a.) Schematic representation of the binding mechanism of CTAB on PVAS [6]. b.) The impact of the type of polyion charge on the nonequilibrium PE/S association [5].

2.2 Nonequilibrium association in aqueous PVAS/CTAB and poly(acrylic acid) (PAA)/CTAB mixtures ([5], O/5, P/4)

In the second part of this workpackage, the investigations were extended for PVAS samples from 10.6 to 2.35 mole% of vinyl sulfate monomers and their association with CTAB was compared with poly(acrylic acid)/CTAB complexation at similar low (initial) charge densities of these polyions. We have observed a marked difference in the nonequilibrium behavior of PVAS/CTAB and PAA/CTAB systems in the absence of added supporting electrolyte (see Figure 4b). In the case of the weak polyelectrolyte PAA, the binding of CTAB onto the carboxylate groups increases its ionization degree. In addition, there are additional intermolecular hydrogen bonds between the neutral carboxylic groups of the PAA/CTAB complexes. This leads to decreased solubility of the polyion/surfactant complexes and to the formation of charge stabilized colloidal dispersions at surfactant excess via the rapid homogenization of the mixtures. In contrast, the double hydrophilic PVAS molecules cannot adjust their low charge density and thus the amount of the bound cationic amphiphile. Furthermore, there is an additional surfactant binding step onto the nonionic vinyl alcohol units. This results in the reswelling of the PVAS/CTAB complexes and thus the disappearance of kinetically arrested states at appropriately large surfactant excess. The nonequilibrium concentration range further reduces with decreasing charge of the PVAS copolymer. These observations clearly reveal that the nonequilibrium features of PE/S mixtures are crucially dependent on the charge regulation capability of the polyelectrolytes as well as on their chemical composition.

WP3. Preparation of Stable Novel Nanoparticles with Controlled Size and Structure

3.1 Association between poly(diallyldimethylammonium chloride) (PDADMAC) and sodium dodecyl sulfate (SDS) in the presence of dodecyl maltoside ($C_{12}G_2$) ([8], [9], O/5, O/6, P/5)

In the first part of the work, the potential route of the synthesis of PDADMAC/SDS/ $C_{12}G_2$ nanoparticles was investigated using electrophoretic mobility, turbidity and fluorescence spectroscopic measurements. We have demonstrated that the addition of nonionic amphiphiles could destabilize the charge-stabilized colloidal dispersions of PDADMAC/SDS

nanoparticles even if a rapid mixing protocol is applied for the solution preparation. This finding is attributed to the complex exchange processes which slow down the attainment of the final composition of the mixed surfactant adsorbed layer on the PDADMAC/SDS/C₁₂G₂ nanoparticles. However, at the same concentration range stable dispersions of polyion/mixed surfactant nanoparticles was produced through the application of a new two-step solution preparation technique (see Figure 5a). This method is based on the order of addition effect of the two types of surfactant and is likely to be utilized in future scientific and industrial applications. In order to explore the fine structure of PDADMAC/SDS/C₁₂G₂ polyion/mixed surfactant nanoparticles, small-angle X-ray scattering (SAXS) measurements were also carried out in collaboration with Jan Skov Pedersen group (Aarhus University, Denmark). It has been shown that the SDS binds in the form of small cylindrical surfactant micelles, even at low SDS-to-PDADMAC ratios, in contrast to the globular amphiphile structures usually predicted in this composition range of PE/S systems. These anisometric surfactant self-assemblies mimic the basic building units of the hexagonal structure of the PDADMAC/SDS precipitate and/or suspensions formed at charge equivalence or at higher SDS-to-PDADMAC ratios. The addition of dodecyl maltoside does not lead to significant changes in the structure of PDADMAC/SDS complexes or their aggregates (see Figure 5b). The only exception is for samples at SDS-to-PDADMAC ratios close to charge neutralization and at high concentrations of C₁₂G₂ where the precipitate forms a multiphasic or distorted hexagonal structure.

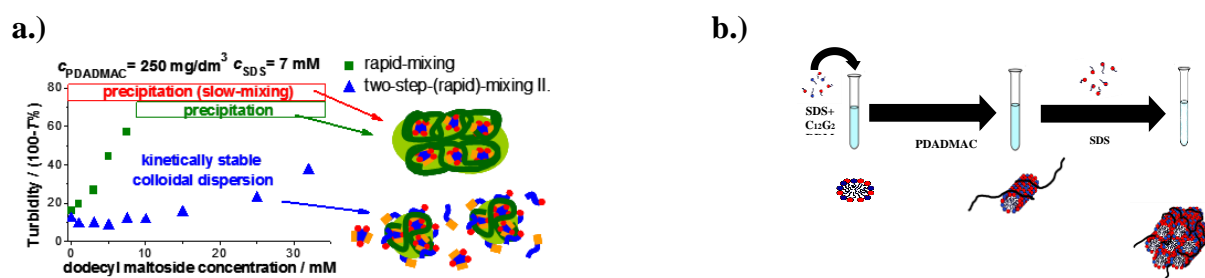


Figure 5. *a.)* The impact of the order of addition of the solution components on the stability of polyion/mixed surfactant nanoparticles [9]. *b.)* Formation of anisometric PDADMAC/SDS/C₁₂G₂ nanoassemblies [8].

3.2 Novel mixed nanoassemblies of C₁₂G₂, and the polyanion PSS with the cationic surfactants CTAB or DTAB ([7], O/6, O/7, P/5)

It has been shown that the nonionic surfactant additive has two distinct impacts on the PSS/CTAB complexation. At low C₁₂G₂ concentrations, the synergistic binding of the cationic and nonionic surfactants to the PSS molecules is the major effect enhancing the kinetically stable concentration range of overcharged polyion/surfactant nanoparticle dispersions. At large excess of dodecyl maltoside, however, another impact of the nonionic surfactant additive dominates, i.e. the stripping of the ionic amphiphile from the PSS/mixed surfactant complexes into the mixed micelles, which reduces the surface charge of the PSS/CTAB/C₁₂G₂ nanoparticles and thus destabilizes their dispersion. At even larger nonionic surfactant concentrations, the reduced binding of CTAB leads to the redissolution of the precipitate. In contrast, neither the destabilization nor the resolubilization effects of the added dodecyl maltoside were observed for the PSS/DTAB system due to the much lower driving force of DTAB binding compared to CTAB (see Figure 6).

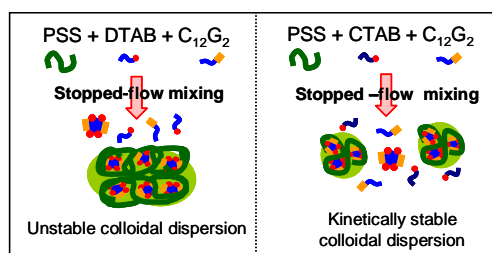


Figure 6. Comparison of the effect of dodecyl maltoside on the nonequilibrium association of PSS with DTAB and CTAB [7].

3.3 Novel assemblies of gold nanoparticles, poly(ethylene imine) (PEI) and sodium alkyl sulfates ([4], O/2, O/4, P/1, P/3)

As it was emphasized in the original proposal, there is a compelling need to prepare novel soft matter nanoparticles with well-defined size and composition as well as with appropriate hydrophobicity and internal structure. Building on the results of WP1 and WP2, novel nanocomposites, containing gold nanoparticles (NPs) embedded in polyelectrolyte/surfactant matrix, were developed as an unplanned but exciting outcome of the project. Specifically, we have shown that branched PEI molecules and SDS, without additional reducing components, can be utilized for the synthesis of gold nanohybrids with polyamine/anionic surfactant shell around the gold core. The charge and hydrophobicity of this layer as well as the interparticle interactions and size of the NPs can be adequately adjusted through the variation of polymer/surfactant ratios, provided that the well-defined rapid-mixing method is used (see Figure 7). These findings may be further utilized for the controlled synthesis of aqueous dispersions and solid assemblies of noble metal NPs with hierarchical structure and tunable properties. Investigations towards the synthesis of these types of nanosystems are already in progress.

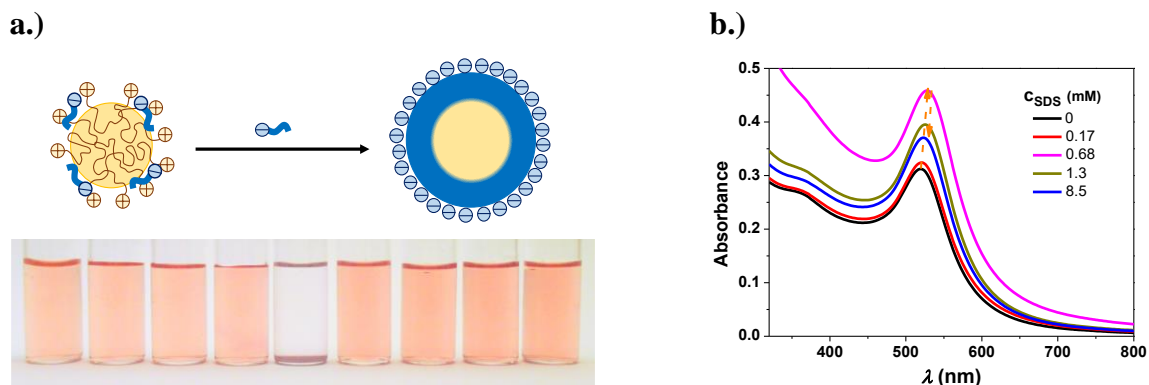


Figure 7. a.) Visual observation and illustration and b.) UV-VIS spectra of gold/PEI/SDS nanocomposites [4].

WP4. Nonequilibrium Association of Biomacromolecules and Surfactants at the Air/Water Interface

4.1 Impact of nonequilibrium aggregates on the properties and structure of PDADMAC/SDS layers on the free aqueous interface ([10], O/5)

In this work package, the research was started with the investigations at the air/water interface of PDADMAC/SDS mixtures. It has been shown that the surface properties of these systems can be categorized with respect to the dynamic changes in the bulk phase behavior, the bulk composition, and the sample history. First, bulk PDADMAC/SDS aggregates can

spontaneously interact with the adsorption layer and are retained in it most readily for positively charged aggregates with an expanded structure. In a second distinct mechanism, aggregates inherently become trapped at the interface during its creation and lateral flocculation occurs. This irreversible process is most pronounced for aggregates with the lowest charge. A third mechanism involves the deposition of aggregates at interfaces due to their transport under gravity. The prevalence of each mechanism critically depends on different factors and experimental conditions, which are outlined systematically in this study.

4.2 Structure and Morphology of the adsorbed Lysozyme layers at the Air–Water Interface ([3], O/5)

In the second part of these studies, the adsorption kinetics of the lysozyme biomacromolecule at the air–water interface from dilute solutions was investigated systematically in the absence and presence of strong denaturants, urea and guanidine hydrochloride (GuHCl) using neutron reflectometry, Brewster angle microscopy (BAM) and interfacial rheology measurements. It was revealed that the thickness of the adsorption layer in the absence of added denaturants is less than the short axial length of the lysozyme molecule, which indicates significant deformation of the globules. Two-dimensional elongated aggregates in the surface layer merge over time to form an extensive network at the approach to steady state (see Figure 8). Addition of denaturants results in the acceleration of adsorption and the increase of the adsorption layer thickness. Both effects may be connected to an increase of the effective total volume of macromolecules due to the changes of their tertiary structure, i.e., the formation of molten globules under the influence of urea and the partial unfolding of globules under the influence of GuHCl. In the former case, the increase of the globule hydrophobicity leads to cooperative aggregation in the surface layer during adsorption. Unlike the case of solutions without denaturants, the surface aggregates are short and wormlike, their size does not change with time, and they do not merge to form an extensive network at the approach to steady state. The addition of surfactants affects both the tertiary structures of lysozyme and the nature of its aggregates in the adsorbed layer which needs further investigations.

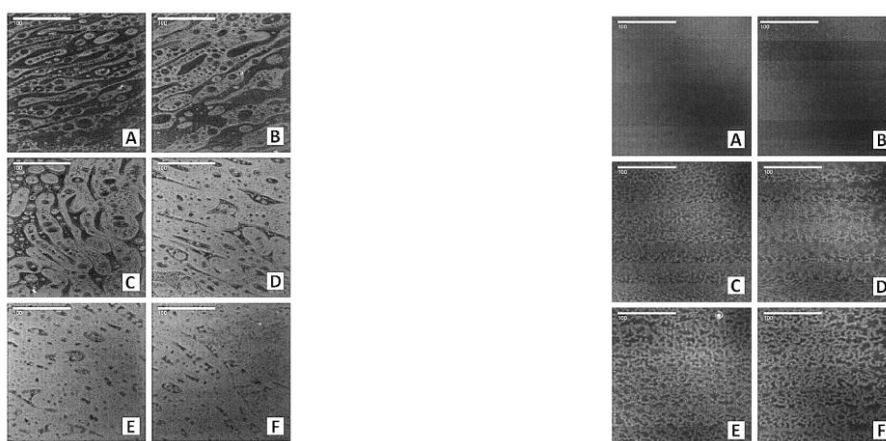


Figure 8. Left: BAM images of an adsorbed layer from 3.5 μM lysozyme solution after (A) 0, (B) 2, (C) 5, (D) 15, (E) 60 and (F) 120 min. **Right:** BAM images of adsorbed lysozyme layer from 3.5 μM solution in 5 M urea after (A) 0, (B) 10, (C) 20, (D) 40, (E) 60 and (F) 120 min. [3].

General conclusions and future perspectives

The present research clarified the crucial role of concentration gradients in the formation of nonequilibrium polyelectrolytes(PE)/surfactant(S) assemblies. On one hand, the polyelectrolyte concentration gradients could largely enhance the size and compositional heterogeneity of the formed nonequilibrium aggregates. On the other hand, the electrolyte concentration gradients, present during a significant dilution of a PE/S/salt formulation, could lead precipitates with remarkably different morphology compared to the mixing of dilute polyelectrolyte and surfactant solutions. In an additional part our work, we have revealed that with reducing ratio of the ionic/nonionic units of copolymer polyelectrolytes as well as via the application of fixed polyion charges instead of adjustable ones, the nonequilibrium concentration range of PE/S systems considerably decreases. Based on this knowledge, we have also succeeded to synthesize novel polyion/mixed surfactant nanoparticles as well as new types of gold nanohybrids embedded in a matrix of polyelectrolyte/surfactant assemblies. Finally, it was shown that the bulk PE/S and protein aggregates could largely affect the air/water interface via their sedimentation and entrapment in the adsorbed layer as well as through their specific surface interactions. The results of the project are expected to be further exploited towards new directions of nanoscience research as well as to next generation home and personal applications.

Publication of the project results

Scientific papers

- [1] Bali K.; Varga Zs.; Kardos A.; Mészáros R.: Nonequilibrium Association of Oppositely Charged Macromolecules and Amphiphiles. Manuscript under preparation.
- [2] Bali K.; Varga Zs.; Kardos A.; Varga I.; Domján A.; Wacha A.; Bóta A.; Mihály J.; Gilányi T.; Mészáros R.: Effect of Dilution on the Nonequilibrium Aggregate Formation in Sodium Poly(styrenesulfonate)/Dodecyltrimethylammonium Bromide Mixtures with Added Salt. To be submitted to **THE JOURNAL OF PHYSICAL CHEMISTRY B**
- [3] Campbell R.; Tummino A.; Varga I.; Milyaeva O.; Tichonov M.; Lin S.-Y.; Laux V.; Forsyth V.; Noskov B.: Adsorption of Denaturated Lysozyme at the Air–Water Interface: Structure and Morphology. Submitted to **LANGMUIR**, Status: Minor revision
- [4] Bali K.; Sáfrán Gy.; Pécz B.; Mészáros R.: Preparation of Gold Nanocomposites with Tunable Charge and Hydrophobicity via the Application of Polymer/Surfactant Complexation. **ACS OMEGA**, 2, 8709-8716 (2017) (new open access ACS journal, free publication upon personal invitation from Luiz Liz Marzan coeditor)
- [5] Bodnár K.; Szarka K.; Nagy M.; Mészáros R.: Effect of the Charge Regulation Behavior of Polyelectrolytes on their Nonequilibrium Complexation with Oppositely Charged Surfactants. **THE JOURNAL OF PHYSICAL CHEMISTRY B** 120, 12720-12729. (2016)
- [6] Bodnár K.; Fegyver E.; Nagy M.; Mészáros R.: Impact of Polyelectrolyte Chemistry on the Thermodynamic Stability of Oppositely Charged Macromolecule/Surfactant Mixtures. **LANGMUIR**, 32, 1259-1268. (2016)
- [7] Fegyver E.; Mészáros R.: Complexation between Sodium Poly(styrenesulfonate) and Alkyltrimethylammonium Bromides in the Presence of Dodecyl Maltoside. **THE JOURNAL OF PHYSICAL CHEMISTRY B**, 119, 5336-5346. (2015)

- [8] Plazzotta B.; Fegyver E.; Mészáros R.; Pedersen J. S.: Anisometric Polyelectrolyte/Mixed Surfactant Nanoassemblies Formed by the Association of Poly(diallyldimethylammonium chloride) with Sodium Dodecyl Sulfate and Dodecyl Maltoside. **LANGMUIR**, 31, 7242-7250. (2015)
- [9] Fegyver E.; Mészáros R.: Fine-Tuning of The Nonequilibrium Behavior of Oppositely Charged Macromolecule/Surfactant Mixtures via the Addition of Nonionic Amphiphiles. **LANGMUIR**, 30, 15114-15126. (2014)
- [10] Campbell R. A.; Yanez Arteta M.; Angus-Smyth A.; Nylander T.; Noskov B. A.; Varga I.: Direct Impact of Nonequilibrium Aggregates on the Structure and Morphology of Pdadmac/SDS Layers at the Air/Water Interface. **LANGMUIR**, 30, 8664-8674. (2014)

4.2 Oral presentations

- O/1. Bali K.; Varga Zs.; Kardos A.; Varga I.; Mészáros R.: “Nonequilibrium Phase Properties of Oppositely Charged Polyelectrolyte/Surfactant Systems: The Effect of Concentration Gradients”, 11th Conference on Colloid Chemistry (11CCC), Eger, Hungary, **2018.05.28-30.**, **accepted oral presentation.**
- O/2. Bali K.; Mészáros R.: “Surface Modification of Polyamine-Capped Gold Nanoparticles: Competition between Au(III) Reduction and Surfactant Binding” 16th Conference of the International Association of Colloid and Interface Scientists (IACIS 2018), Rotterdam, Netherlands, **2018.05.21-25.**, **accepted oral presentation**
- O/3. Bali K.; Varga Zs.; Kardos A.; Varga I.; Domján A.; Wacha A.; Bóta A.; Mihály J.; Mészáros R.: “Effect of Dilution on the Nonequilibrium Properties of Poly(Styrenesulfonate)/Dodecyltrimethylammonium Bromide System” 16th Conference of the International Association of Colloid and Interface Scientists (IACIS 2018), Rotterdam, Netherlands, **2018.05.21-25**, **accepted oral presentation**
- O/4. Mészáros R.; Bali K.: “Application of Polymer/Surfactant Complexation for the Preparation of Gold Nanoparticle Assemblies with Tunable Hydrophobicity”, 7th International Colloids Conference, Sitges, Barcelona, Spain, **2017.06.18-21.**
- O/5. Campbell R.; Varga I.: “General Physical Description of the Behavior of Oppositely Charged Polyelectrolyte/Surfactant Mixtures at the Air/Water Interface”, 31st Conference of the European Colloid and Interface Society, (ECIS 2017), Madrid, **2017.09.03-08.**
- O/6. Mészáros R.: “Nonequilibrium Association of Oppositely Charged Macromolecules and Amphiphiles”, Scientific Seminar, Department of Physical and Theoretical Chemistry, Comenius University, **invited lecture**, Bratislava, Slovakia, **2016.11.13.**
- O/7. Fegyver E., Mészáros R.: “Nonequilibrium Aspects of Polyion/Mixed Surfactant Complexation”, 5th International Colloids Conference, Amsterdam, Netherlands, **2015.06.21-24.**

4.3 Posters

- P/1. Bali K.; Mészáros R.: “Influence of Added Anionic Surfactant on the Polyamine Induced Reduction Process of Gold(III)-Ions”, 11th Conference on Colloid Chemistry (11CCC), Eger, Hungary, **2018.05.28-30.**, **accepted abstract**
- P/2. Bali K.; Varga Zs.; Kardos A.; Varga I.; Mészáros R.: “Effect of Electrolyte Concentration Gradient on the Phase Properties of Oppositely Charged Polyelectrolyte/Surfactant Mixtures”, 7th International Colloids Conference, Sitges, Barcelona, Spain, **2017.06.18-21.**

- P/3. Bali K.; Mészáros R.: *“Preparation of Polyamine/Anionic Surfactant/Gold Nanocomposites: Effect of Temperature, pH and Preparation Method”*, 7th International Colloids Conference, Sitges, Barcelona, Spain, **2017.06.18-21**.
- P/4. Bodnár K.; Fegyver E.; Nagy M.; Mészáros R.: *“Effect of the Charge Regulation Behavior and Chemistry of Polyelectrolytes on their Nonequilibrium Complexation with Oppositely Charged Surfactants”*, 30th Conference of the European Colloid and Interface Society, (ECIS 2016), Rome, **2016.09.04-09**.
- P/5. Mészáros R.; Fegyver E.: *“Nonequilibrium Complexation of Oppositely Charged Polyelectrolytes and Surfactants in the Presence of Nonionic Amphiphiles”*, 29th Conference of the European Colloid and Interface Society, (ECIS 2015), Bordeaux, France, **2015.09.02-05**.

Budapest, 2018.03.28.

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