SUMMARY AND CONCLUSION

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Interfacial and micellization behavior of (C_{12}AAS)Na_2-HTAB mixtures were studied using different physicochemical techniques. CMC values gradually increase with increasing proportion of (C_{12}AAS)Na_2, indicating increased hydrophobicity between the oppositely charged surfactants. Negative Gibbs free energy of micellization indicates the spontaneity of the micellization process. With increasing mole % of $\alpha_{(C_{12}AAS)_2Na_2}$, surface pressure at the CMC (π_{CMC}) passes through minima due to strong synergestic interaction between the surfactant components. Oppositely-charged surfactants can achieve proximities to each other through the head group interaction and interact mainly at the micellar surface. Limiting molecular area of the mixed surfactant systems at the air-liquid interface gradually decrease and the surface excess values increase with increasing mole fraction of $\alpha_{(C_{12}AAS)_2Na_2}$. Conductance studies show that maximum number of the surfactant molecules are in their dissociated forms near the CMC; so the micellar surface charge density is higher. Micellar size gradually increases with decreasing α_{AAS}, due to the preferential accumulation of HTAB in the micelle. Rheological studies provide additional information on the internal structure of mixed surfactant aggregates. During micellization, maximum number of HTAB molecules cooperate on the micellar surface leading to the formation of closed packed aggregates. Oppositely charged surfactants in their mixed states can form different viscous and gelatinous entities. Viscosity studies reveal thixotropic nature of the mixed surfactant systems where viscosity do not change spontaneously with increasing $\alpha_{(C_{12}AAS)_2Na_2}$,with few exceptions. Surfactant mixtures form different types of aggregates; so the knowledge on the surface morphology by phase contrast, polarization optical microscopic (POM), fluorescence microscopic (FM) and field emission scanning electronic microscopy (FE-SEM) studies are considered to be essential. Studies on the interfacial and micellization behavior of mixed surfactants are expected to provide new insights and can be used as drug delivery systems, especially in dermatological formulations and to synthesize otherwise water insoluble inorganic nanoparticle synthesis.

The theoretical investigation has simplified the molecular thermodynamics-related theory for micelle formation by mixed surfactants systems. Different working models to predict *CMC* of non-ideal binary surfactants as well as specific interactions were compared. The different models could provide reasonable quantitative predictions on the different micellization parameters for cationic-anionic mixed surfactant systems. The simplified "working models" employed herein could act as valuable preliminary screening tools in the design and selection of non-ideal surfactant mixtures for practical applications. Synergistic interaction behavior of $(C_{12}AAS)Na_2$-HTAB mixtures were assessed using Rubingh's, Rosen, Motomura and SPB models. The experimental *CMC* values are lower than the predicted values calculated from Clint formalism, indicating non-ideality in the mixing behavior. *CMC* values gradually increase with the increasing proportion of $(C_{12}AAS)Na_2$. Oppositely charged surfactants can localize in vicinity to each other and interact mainly at the micellar surface. The two carboxylate groups of $(C_{12}AAS)Na_2$ repel each other, the extent of which is minimized by HTAB through electrostatic attraction; thus the micellar size gradually decreases with increasing $\alpha_{(C_{12}AAS)_2Na_2}$. A maximum number of HTAB molecules become available on the micellar surface, leading to the formation of closed packed micellar structures. The binary mixtures show significant synergism (negative β^R value). With increasing hydrophobicity of the spacer, synergistic interactions between the surfactant components also increase. With increasing $\alpha_{(C_{12}AAS)_2Na_2}$, the magnitude of β^R decreases, X_1^{ideal}, X_1 (Rubingh model) and X_1^{σ} (Rosen model) gradually increase with an increase of $\alpha_{(C_{12}AAS)_2Na_2}$. The magnitude of β^{σ} at air-water interface gradually decreases with increasing $\alpha_{(C_{12}AAS)_2Na_2}$and follows the order: $C_{12}MalNa_2$+HTAB > $C_{12}AspNa_2$+HTAB > $C_{12}GluNa_2$+HTAB. ΔG_m values are more negative than ΔG_m^{ideal} which indicate that the micellization process is spontaneous. Mixed micellization of $(C_{12}AAS)Na_2$+HTAB is enthalpy driven where ΔH_m values decrease with increasing $\alpha_{(C_{12}AAS)_2Na_2}$. It is believed that theoretical investigations of mixed micelles of such binary surfactants can provide new insights, which will eventually help in understanding the bulk and interfacial activities of mixed surfactant systems. Strong synergistic interaction between the oppositely charged surfactants can result in the formation of liquid crystal,

viscous gels and even vesicles. However, further theoretical investigations employing molecular dynamics could support the propositions made here, in the future. Oppositely charged mixed surfactants form different types of aggregates as stated earlier; so the knowledge on the surface morphology by phase contrast, polarization optical microscopy (POM), fluorescence microscopy (FM), field emission scanning electron microscopy (FE-SEM) and small angle X-ray scattering (SAXS) studies are considered to be essential.

Micro-structure of $(C_{12}AAS)Na_2$+HTAB aggregates were investigated by combined phase manifestation, optical and electron microscopic studies. Texture of the liquid crystals formed by the surfactant aggregates depend on the concentration and composition. Energetics of phase transition processes were evaluated by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) studies. Cytotoxicity could be correlated with the viscosity of the gels. Gels impart insignificant skin irritation although they possess substantial antibacterial activities that project its potential as dermal drug delivery systems. However further *in vitro* and *in vivo* studies by incorporating appropriate drugs into the gels are necessary and being considered as the future perspectives.

Solid and solution properties of bivalent metallosurfactants of $(C_{12}AAS)Na_2$ are characterized by different physicochemical processes. Formations of coacervates were ensured by NMR and FTIR as well as layered structures were supported by XRD studies. Phase transition and liquid crystalline nature of the solid complexes were confirmed from TGA and DSC studies. Lift-off area (A_0) and minimum molecular area (A_{min}) values increase with increasing $\alpha_{(C_{12}AAS)_2M_2}$ due to the formation of expanded monolayer whereas the non spontaneous mixing behaviors are confirmed with the higher negative ΔG_{ex}^0 values. BAM images demonstrate qualitative information on the texture of mixed monolayer as function of composition. TEM images confirmed the formation of spherical vesicles and the results were correlated with DLS studies. Hydrodynamic diameter and negative zeta potential of the vesicles gradually increase with increasing $\alpha_{(C_{12}AAS)_2M_2}$, due to the hydrophobic interaction between oppositely charged surfactants.

Cytotoxicity studies indicate that liposomes are almost nontoxic. Results suggest that $(Na_2AAS)_2M_2$+ SPC mixed liposome can safely be used as potential drug carrier. In order to further explore the cell viability, the formulations were subjected to *in vitro* studies under biological condition, which reveal that the hybrid vesicles could act as promising vehicles for different drugs with controlled and prolonged release. Studies on molecular dynamics simulation and associative interaction through different physicochemical processes, *viz.*, SANS, SAXS and cryo-TEM studies are the future perspectives. Also such systems are expected to have superior properties as potent vectors as drug delivery.