

# Deposition enhanced by coacervation in mixtures of chitosan and a non-ionic sugar-based surfactant

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## SUMMARY

This study explores the use of chitosan–alkyl polyglucoside (APG) mixtures as sustainable ingredients for 2-in-1 shampoo formulations. By varying surfactant concentration and ionic strength at a constant chitosan level, we observed distinct phase behaviors influenced primarily by APG concentration. The system exhibited transparent phases at low and high surfactant levels and turbid mixtures at intermediate concentrations, attributed to coacervation—essential for conditioning deposit formation. Ionic strength modulated these transitions but did not alter the overall phase pattern. Importantly, coacervate regions significantly enhanced deposition, confirming the potential of chitosan–APG systems to deliver effective conditioning. These findings support the development of eco-friendly hair care products and advance understanding of biopolymer–surfactant interactions.

## METHODS

### Chemicals:

- Alkyl polyglucoside (APG), a 60% aqueous solution of an equimolar mixture of caprylyl and capryl glucosides (Oramix GC-110), was supplied by Safic-Alcan (Spain).
- Chitosan (CHI), with a molecular weight of 100–300 kDa and ~90% degree of deacetylation, was obtained from Thermo Fisher Scientific (USA).
- Glacial acetic acid and NaCl ( $\geq 99.5\%$  purity) were purchased from Merck (Germany). Ultrapure Milli-Q water (resistivity  $>18 \text{ M}\Omega\text{-cm}$ ) was used throughout.

### Sample preparation:

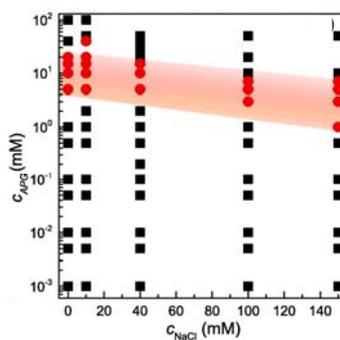
The preparation of the mixtures of chitosan and APG requires the sequential addition of the required volumes of chitosan and APG stock solution into a 25 mL flask, followed by the dilution with a diluted aqueous acetic acid solution at pH 4.5 and fixed NaCl concentration. To ensure the homogenization of the mixtures, they were subjected to mild stirring (1000 rpm) for 24 h.

### Experimental Methods:

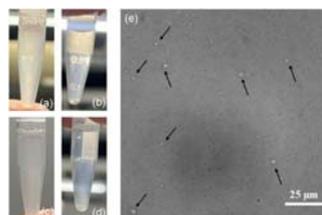
- QCM-D:** Quartz crystal microbalance with dissipation monitoring (QSense Explorer) was used to assess layer deposition on negatively charged gold surfaces. Frequency and dissipation shifts were analyzed using a viscoelastic single-layer model.
- Electrophoretic Mobility:** Zeta potential was inferred from Laser Doppler velocimetry (Zetasizer Nano ZS, Malvern Instruments).
- UV-Vis Spectroscopy:** Absorbance and turbidity (at 450 nm) were measured using a Jasco V-730 spectrophotometer.
- Centrifugation:** Performed at 14,000 rpm for 1 h to analyze phase separation.
- Microscopy:** Optical images were obtained using a Nikon Eclipse 80i microscope (50 $\times$  objective) with a Hamamatsu CMOS camera.

## RESULTS & DISCUSSION

### Phase behaviour:

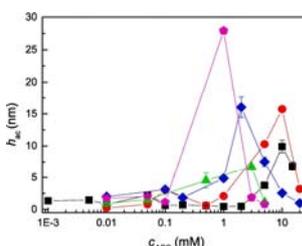


The phase behavior of chitosan–APG mixtures is generally stable across varying ionic strengths, but increasing NaCl concentration causes a shift in the coacervation region toward lower APG concentrations. This occurs because added salt screens the electrostatic repulsion between chitosan chains, promoting direct hydrogen bonding with APG. This "salting-out" effect enhances polymer–surfactant interactions, enabling coacervate formation at lower surfactant concentrations.



Initially, the dispersions exhibit a noticeable turbidity, indicative of a heterogeneous system with dispersed aggregates. Upon centrifugation, a distinct white sediment forms at the bottom of the vials, which suggests phase separation and the settling of a denser component. This behavior is consistent with the formation of complex coacervates. Coacervation process is evidenced by numerous micron-sized droplets dispersed in the continuous medium.

### Adsorption of solid surfaces:



The adsorption behavior of chitosan–APG reveals three distinct regions that align with the system's phase behavior. At low APG concentrations: adsorption is minimal. In the coacervation region, deposition sharply increases, reaching a maximum that coincides with peak. Beyond this region, deposition drops.

## CONCLUSIONS

This study shows that coacervation in polyelectrolyte–surfactant systems can occur without direct electrostatic interactions, driven instead by hydrogen bonding enhanced at higher ionic strengths. These findings enable tunable deposition through ionic conditions and have broad applications in fields like drug delivery and personal care. Future work should explore real-world conditions.

## REFERENCES

- A. Puente-Santamaría, J. N. Molina-Basurto, E. Gerardin, F. Ortega, R.G. Rubio, E. Guzmán, Hydrogen bond-driven interactions between chitosan and biobased surfactants: A study of bulk behavior and surface adsorption, *Journal of Molecular Liquids*, 425 (2025) 127259.