Structure, Heat, and Time: The Dynamic Life of Protein–Phospholipid of Oil-Water Interfaces

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Phospholipids (PLs) and proteins such as β-lactoglobulin (β-LG) are key natural emulsifiers in food and pharmaceutical systems. PLs adsorb rapidly and form compact monolayers, whereas β-LG builds viscoelastic protein networks that provide long-term stability. When used together, their interfacial interactions depend strongly on the molecular structure of the PL and on processing conditions, leading to either co-adsorption or competitive displacement. Most studies focus on long-time interfacial behaviour, while in practice droplet formation and early stabilisation occur within milliseconds. To fully understand emulsion stability across time and processing, it is thus essential to bridge these regimes from rapid adsorption during emulsification to slower rearrangements and heat-induced transformations.

Here, we investigated β-LG combined with saturated (PC 18:0) or unsaturated (PC 18:1) phosphatidylcholines using complementary techniques that capture both equilibrium and dynamic behaviour. Interfacial rheology, ζ-potential, SAXS, μDSC, and CLSM were complemented by microfluidic droplet coalescence experiments probing adsorption and stabilisation at sub-second timescales across 20–90 °C.

On very short time scales (ms-s), combining β-LG with PLs markedly enhanced droplet stability compared to either component alone, revealing a clear synergistic effect against coalescence, most pronounced at elevated temperatures. Below 75 °C, saturated PC 18:0 promoted partial unfolding and co-assembly of β-LG at the interface, yielding cohesive and highly viscoelastic films and increased emulsion stability over time. In contrast, unsaturated PC 18:1 progressively displaced β-LG, leading to less elastic interfaces and reduced long-term emulsion stability. At higher temperatures, interfacial multilayer formation occurred irrespective of PL type, although β-LG retained more structure in the presence of the saturated PC 18:0.

These findings provide molecular-level insight into how interfacial organisation evolves across time and temperature scales, offering design principles for selecting emulsifier combinations and processing conditions to create heat-resilient emulsions.

Keywords:
Emulsion Stability, Mixed Interfaces, Phase transition, Displacement, Competitive Adsorption

Acknowledgements:
This IGF Project (01IF23169N) of the FEI is supported within the programme for promoting the Industrial Collective Research (IGF) of the Federal Ministry of Economic Affairs and Climate Action (BMWK), based on a resolution of the German Parliament.