**Casein stabilized interfaces, the role of molecular structure**

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The future of dairy may lie in the development of animal-free milk, providing a more sustainable alternative to traditional dairy. Central in this development is the synthesis of casein by microorganisms, a key protein for dairy functionality. To anticipate the development of microbial casein, we need to answer fundamental questions such as: Are casein micelles necessary? Is casein phosphorylation essential? Can fewer casein variants achieve comparable functionality?

Hence, this research used bovine casein as a benchmark to investigate the effect of casein chemistry on the stabilization of air- and oil-water interfaces. We show that casein micelles are beneficial for stiffer viscoelastic interface at polar oils but individual casein fractions might be a good alternative. The phosphorylation is essential for creating interfacial networks by forming a space-spanning network with calcium. The location of phosphorylation will also affect the interfacial configuration which has a big effect on the sensitivity to calcium. In αs2-, and β-casein, the phosphorylation creates a tail-train configuration which makes them highly calcium sensitive.

With this work we show that casein chemistry critically impacts interfacial viscoelasticity, enabling the tailoring of functional properties through selective use of casein fractions. Microbial casein should focus on phosphorylation to create interfacial networks, but using all four casein fractions in animal-free dairy might not be necessary.