The 10th EuChemS Chemistry Congress

12 - 16 July 2026

Antwerp, Belgium

Innovative Materials (IM)

IM - D4 - P22

Systematic functionalization of silk sericin via acetylation: A design of experiments approach toward sustainable biomaterials

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Silk sericin, the glue-like protein that binds fibroin fibers within silk, is generated in large quantities as an industrial byproduct and discarded despite its rich chemical functionality. Its intrinsic properties, including biocompatibility, biodegradability, antioxidant activity, and antibacterial behavior, make it a promising candidate for bio-based materials; however, its effective valorization remains limited by the lack of a quantitative understanding of the relationship between chemical modification and structural evolution. Here, a quantitative framework is established to investigate and control sericin functionalization using acetylation as a model reaction, which was selected for its ability to tune the degree of functionalization and secondary structure organization while introducing a biodegradable ester functionality. A Design of Experiments (DoE) approach was employed to systematically assess the effects of acetyl chloride equivalents, initial sericin concentration, and lithium chloride content, including their interaction effects. Fourier-transform infrared (FTIR) spectroscopy was advanced as a quantitative, model-integrated analytical approach. Band deconvolution across the Amide I region (1780–1580 cm^{-1}) enabled the extraction of two complementary descriptors, defined as the functionalization index (FI) and β-sheet index (BI), derived from band areas and intensities. The resulting models exhibited strong predictive capability and statistical significance ($p < 0.01$), with good agreement between experimental and predicted responses. Notably, the system displayed pronounced non-linear behavior governed by significant quadratic and interaction effects, revealing a multidimensional response surface. These findings demonstrate a direct coupling between chemical reactivity and protein structural organization, enabling predictive control over sericin modification. Overall, this work introduces a reproducible and quantitative methodology for linking reaction parameters to both chemical and structural outcomes, supporting the development of sericin as a sustainable, high-value biomaterial platform.

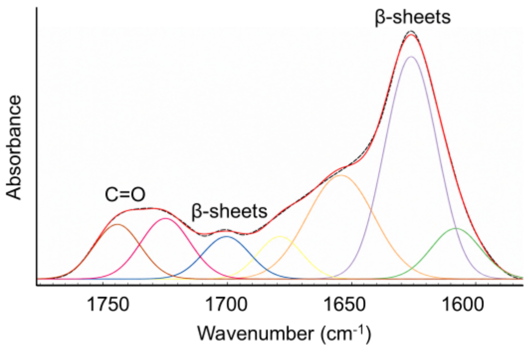

Fig.1. FTIR band deconvolution illustrating the extraction of functionalization and β-sheet structural descriptors.

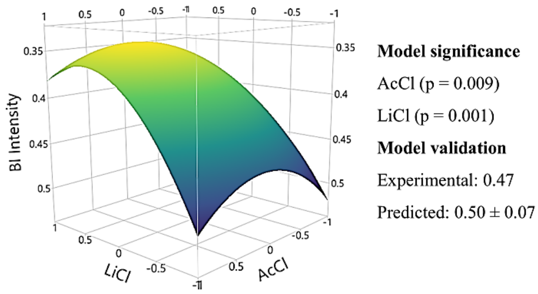

Fig. 2. Response surface of BI (intensity) highlighting the effects of AcCl and LiCl, with model significance and validation.