Swelling Dynamics in Ultrathin Protein Films

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Understanding early-time swelling in protein thin films is critical for designing responsive soft actuators. We have demonstrated protein-based actuators with tunable mesoscale structure, where controlled crystallinity enables precise regulation of actuation speed and magnitude under vapor exposure [1]. To uncover the underlying mechanisms, we employ time-resolved synchrotron X-ray reflectivity (XRR) with simultaneous angle- and energy-dispersion to quantitatively probe the swelling dynamics of ultrathin protein films upon exposure to controlled humidity. The ultra-fast XRR at the PF-AR Beamline NE-7A, Photon Factory, Japan, provides sub-second temporal resolution of the evolving film structure during early exposure times. The measurements reveal an interplay between solvent diffusion and polymer relaxation. Interestingly, as relative humidity increases, the system undergoes a transition analogous to a glass transition at ambient temperature, driven by water-induced softening rather than thermal activation. Importantly, it is also found that the protein's intrinsic secondary structure preferentially affects diffusion, suggesting that molecular-scale organization controls macroscopic transport. Additionally, spectroscopic ellipsometry studies track the swelling pathway of protein films dependent on thickness and refractive index to further elucidate diffusion pathways. These findings provide new insight into the mechanisms governing moisture-induced swelling in protein thin films and demonstrate the capability of time-resolved XRR for probing ultrafast structural evolution. These findings and their implications will be discussed in detail.

References

[1] Siraj, S. A., Kumar, V., & Satapathy, D. K. (2025). Langmuir, 41(32), 21553-21561.