Program in Polymers and Soft Matter

Polymer Seminar - November 4, 2020

Phase-dependent redox properties in marine adhesive proteins

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Nature seems full of contradictions, or so it seems, until one digs a little deeper. Case in point: catecholic 3,4-dihydroxyphenyl-L-alanine (Dopa) residues in mussel foot proteins (mfps) are critical to mussel (*Mytilus californianus*) plaque adhesion, but only when protected from oxidation at the adhesive-substratum interface. Dopa oxidation is thermodynamically favorable in seawater yet oxidized Dopa is barely detectable in plaques. These apparent paradoxes led us to investigate how plaques insulate Dopa-containing mfps against oxidation. Using Dopa as a convenient redox reporter, we show that Dopa-containing mfp3 and mfp6 in phase-separated droplets remain stable despite rapid Dopa oxidation in the surrounding equilibrium solution, even when powerful oxidants (periodate and DPPH) are present. The results suggest that a cohort of oxidation-prone proteins is endowed with phase-dependent redox stability. Moreover, in forming LLPS compartments, Dopa proteins become reservoirs of chemical energy.

[1]Miller, D.R., Spahn, J. & Waite, J.H. J. R. Soc. Interface 2020, 12: 20150614
[2]Valois, E et al., Sci. Adv. 2020; 6: eaaz6486