Towards active bijels: Prussian-blue stabilized bicontinuous emulsions as electrode modifiers

João Victor Mattioni [a], Kalil Cristhian Figueiredo Toledo [a], Denise Freitas Siqueira Petri [a]

[a] Department of Fundamental Chemistry, Institute of Chemistry, University of São Paulo. 05508-000 São Paulo, SP, Brazil

joaomattioni@iq.usp.br

Bicontinuous interfacially jammed emulsion gels (bijels) are a class of soft materials composed of two interpenetrating and immiscible liquid phases arranged in a bicontinuous morphology, generated through spinodal decomposition. Their formation rely on the adsorption of colloidal particles at the liquid–liquid interface, where the particles become jammed and form a mechanically robust barrier between the two continuous domains. This structure is only achieved when the particles exhibit neutral wettability with respect to both liquid phases, thereby preventing preferential partitioning into either phase.

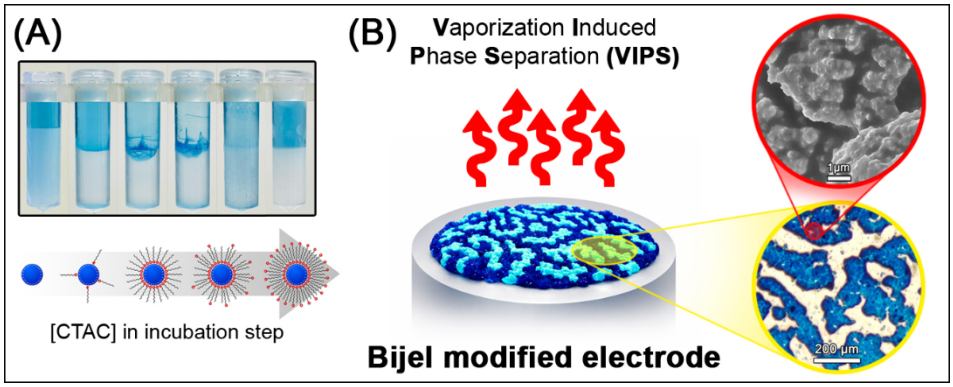
Due to their high interfacial area and interconnected transport pathways, they emerge as a promising platform for the design of functional materials in areas such as catalysis. However, most reported systems employ silica as stabilizing particles, while the use of electroactive nanoparticles remains comparatively unexplored.

Prussian blue is a mixed valence coordination polymer consisting of Fe(III) and Fe(II) cations interconnected by cyanide bridges within a three-dimensional cubic crystalline lattice. It exhibits well defined electrochemical activity arising from intervalence charge transfer between Fe(II) and Fe(III) centers, which promotes electron transport through the Fe(II)–CN–Fe(III) framework. Under an applied potential, the material undergoes reversible redox interconversions, enabling its use in electroactive and charge transport related applications.

Citrate-stabilized Prussian blue nanoparticles (26 ± 7 nm) were synthesized by direct mixing of iron(III) chloride and potassium hexacyanoferrate(II) in the presence of citric acid at 60 °C and purified by dialysis. Surface hydrophobicity was tuned by incubating the nanoparticles with increasing concentrations of the cationic surfactant cetyltrimethylammonium chloride (CTAC) and the resulting interfacial behavior was assessed by zeta potential measurements and qualitative visual tests. The optimal condition was selected to enable interfacial stabilization of a bicontinuous structure formed between water and 1,6-hexanediol diacrylate through vaporization induced phase separation (VIPS) in a ternary mixture containing ethanol as a compatibilizing solvent.

When drop cast onto planar conductive substrates, the resulting bijel preserves interconnected bicontinuous channels, as confirmed by optical and scanning electron microscopy. These results demonstrate that Prussian blue nanoparticles can act not only as interfacial stabilizers, but also as electroactive building blocks for bicontinuous architectures, opening a route toward bijel based electrode coatings with coupled structural and charge transport functionality.

Keywords:
bijel, bicontinuous emulsion, nanoparticles at interfaces, Prussian blue, surface wettability



(A) Surface wettability modulation of Prussian blue nanoparticles with increasing CTAC content. (B) Bijel structured modified electrode surface fabricated through vaporization induced phase separation (VIPS).

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