Contribution ID: **429** Type: **Poster Presentation**

Gradient hydrogel based on self-filtration

Wednesday, 24 May 2023 10:30 (1h 30m)

Understanding and mastering gelation of biopolymer is important for controlling the structure of the gels and open the way to the design of micro-objects with a gradient of physical properties. In this study, we investigated the gelation between a calcium solution and a suspension of biopolymers. We showed that spontaneous osmotic flow through the gel controls its formation and can be used to a create concentration gradients.

For this purpose, we developed a quasi-bidimensional cell in which the suspension of aggregates at mass fractions below its gel point is brought into contact with a calcium solution. The diffusion of calcium into the system allows the biopolymer to gel. We observed the propagation of the gelation front by microscopy. This front followed the classical diffusion-reaction laws, with a short-time regime limited by the reaction, and a long-time regime limited by the diffusion. We also used fluorescently labelled polymers to quantify the local concentration during the diffusion of calcium in the aggregate suspension.

Very surprisingly, the gelation mechanism induced a solvent flow from the biopolymer suspension to the calcium solution. This flow was characterized by the tracking of micrometric particles. A consequence of this flow was a local increase of the polymer concentration in the gelation front. We explain this phenomenon by the difference in chemical potential between the aggregates' suspension and the calcium solution. However, by analogy with osmotic phenomena in U-tubes, this implies that the forming gel acted as a semi-permeable membrane for calcium ions.

We illustrated the interest of his osmotic phenomenon to produce fibers based on protein fractal aggregates with a core-shell structure. The suspension of aggregates was co-injected with a solution of calcium chloride. Sol-gel transition of the suspension was induced by diffusion of calcium ions in the jet and ionic cross-linking of the proteins. The production of these fibers required a precise control of both hydrodynamic and physicochemical conditions, that were systematically investigated. Hydrodynamic instabilities competed with the gelation kinetics. Increasing the calcium concentration, several regimes were observed: swollen, dispersed, and shrunk fibers. In the first regime, homogeneous fibers were obtained. In the last one, osmotic phenomena led to a spontaneous core-shell structure with a dense shell.

Participation

In-Person

References

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Session Classification: Poster

Track Classification: (MS04) Swelling and shrinking porous media