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Development of polymer synthetic systems able to undergo hardening via complex coacervation

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The aim of this project is to synthesize a bioadhesive for soft tissue repair and wound closure: the project is really challenging because a lot of requirements need to be fulfilled. The problem of achieving a strong bond under wet conditions has been solved by many marine organisms, which are able to bond dissimilar materials together under seawater with little if any surface preparation. Complex coacervation, that is the separation of a macromolecular solution composed of two oppositely charged macroions into two immiscible liquid phases, plays an important role in the formation of these adhesives. In particular, both the liquid phases contain huge amounts of water, but the complex coacervate phase contain most or all the charged macromolecules. Bio-inspired adhesives modelled after these examples have been developed: however, despite a good biocompatibility and biodegradability, they often lack the mechanical properties to maintain a strong bond in wet environments.

So, in order to get a more resistant material, some strengthening mechanisms have to be introduced. In order to build an adhesive which can still display a tunable viscoelasticity and self-assembly, providing better mechanical properties, a new strategy is being developed in this project by modifying the chemistry of the polyelectrolytes undergoing complex coacervation. The synthesis of two sets of polylelectrolytes have been successfully achieved in this project.

The second step is the complex coacervation between the synthesized copolymers. Two water solutions of the two polyelectrolytes were mixed in order to have the same concentration of charged groups in the final material. Phase separation immediately took place and, after centrifugation, the polymer concentrated phase sedimented. So, the synthesized polymers have successfully been able to undergo complex coacervation.

The coacervate phase has been characterized by rheology to determine its mechanical properties and by underwater probe-tack test to measure the adhesion properties. Thanks to the chemical modification introduced, the material displays much higher storage and loss modulus compared to the unmodified counterpart and shows much higher work of adhesion when tested in fully submerged conditions.

In conclusion, we were able to synthesize a new set of polyelectrolytes which are able to undergo complex coacervation. The strength and the underwater adhesion energy of the coacervate phase dramatically increases due to the modification introduced. These results match our expectations and show that this material can be a promising candidate for the development of an underwater adhesive for soft tissue repair.

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