Heteromultivalent Carbohydrate-Based Supramolecular Hydrogels to Mimic the Extracellular Matrix

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The pursuit of designing and synthesizing complex systems that closely resemble the highly glycosylated extracellular matrix (ECM) has gained significant interest in various fields: 3D cultures, drug delivery and tissue engineering [1]. Supramolecular hydrogels hold great potential for achieving this goal. However, the development of these hydrogels has been hindered by a lack of knowledge behind the fundamental parameters governing their hierarchical self-assembly. Furthermore, the limited examples of carbohydrate-based hydrogels in the literature predominantly involve homomultivalent presentation of a single carbohydrate, which falls short of replicating the complex heteromultivalent nature of the ECM [2].

To address these challenges, this project aims to synthesize biocompatible supramolecular hydrogels that emulate the highly and heterogeneously glycosylated ECM through a hierarchical supramolecular self-assembly approach, by employing rationally designed neoglycolipids. Various photopolymerizable neoglycolipids with distinct sugar headgroups, such as α-D-mannose, β-D-galactose, β-D-glucose, and β-lactose, have been synthesized. The hierarchical self-organization of these neoglycolipids into different hydrogel structures has been thoroughly characterized. Importantly, the resulting hydrogels exhibit multiple interactions with fluorescent lectins specific to the exposed sugars, highlighting their heteromultivalency.

In addition, hybrids of hydrogels and glyconanoring-coated carbon nanotubes have been developed to enhance the mechanical properties of the constructs.

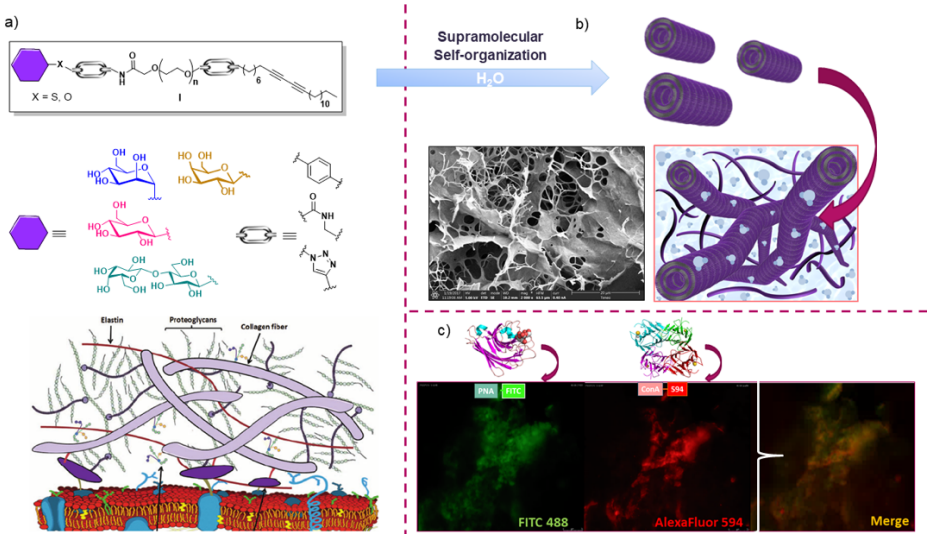

Figure 1. (a) Neoglycolipids. (b) Supramolecular characterization. (c) Heteromultivalency.

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