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Construction and Cross-Linking of Polyelectrolyte Multilayers Based on Polysaccharides as Antifouling Coatings

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Abstract : Marine biofouling is a worldwide problem at vast economic and ecological costs. Historically it was combated with toxic coatings such as tributyltin. As those coatings being banned nowadays, finding environmental friendly antifouling solution has become an urgent topic. In this study antifouling coatings consisted of natural occurring polysaccharides hyaluronic acid (HA), alginic acid (AA), chitosan (Ch) and polyelectrolyte polyethylenimine (PEI) are constructed into polyelectrolyte multilayers (PEMs) in a Layer-by-Layer (LbL) method. LbL PEM construction is a straightforward way to assemble biomacromolecular coatings on surfaces. Advantages about PEM include ease of handling, highly diverse PEM composition, precise control over the thickness and so on. PEMs have been widely employed in medical application and there are numerous studies regarding their protein adsorption, elasticity and cell adhesive properties. With the adjustment of coating composition, termination layer charge, coating morphology and cross-linking method, it is possible to prepare low marine biofouling coatings with PEMs. In this study, using spin coating technology, PEM construction was achieved at smooth multilayers with roughness as low as 2nm rms and highly reproducible thickness around 50nm. To obtain stability in sea water, the multilayers were covalently cross-linked either thermally or chemically. The cross-linking method affected surface energy, which was reflected in water contact angle, thermal cross-linking led to hydrophobic surfaces and chemical cross-linking generated hydrophilic surfaces. The coatings were then evaluated regarding its protein resistance and biological species resistance. While the hydrophobic thermally cross-linked PEM had low resistance towards proteins, the resistance of chemically cross-linked PEM strongly depended on the PEM termination layer and the charge of the protein, opposite charge caused high adsorption and same charge low adsorption, indicating electrostatic interaction plays a crucial role in the protein adsorption processes. Ulva linza was chosen as the biological species for antifouling performance evaluation. Despite of the poor resistance towards protein adsorption, thermally cross-linked PEM showed good resistance against Ulva spores settlement, the chemically cross-linked multilayers showed poor resistance regardless of the termination layer. Marine species adhesion is a complex process, although it involves proteins as bioadhesives, protein resistance its own is not a fully indicator for its antifouling performance. The species will pre select the surface, responding to cues like surface energy, chemistry, or charge and so on. Thus making it difficult for one single factors to determine its antifouling performance. Preparing PEM coating is a comprehensive work involving choosing polyelectrolyte combination, determining termination layer and the method for cross-linking. These decisions will affect PEM properties such as surface energy, charge, which is crucial, since biofouling is a process responding to surface properties in a highly sensitive and dynamic way.

Keywords : hyaluronic acid, polyelectrolyte multilayers, protein resistance, Ulva linza zoospores
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