

Hydrophilic Macromolecular Immobilisation and Release as a Route to Surface Engineering of High-Performance Materials

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Abstract

Optimising surface properties such as friction and wettability is essential for the design of high-performance polymeric materials used in demanding environments. This study draws on biomimetic design principles to enhance the interfacial behaviour of polymeric substrates through the strategic use of synthetic hydrophilic macromolecules. Three distinct macromolecular surface modification methodologies are presented: matrix elution, surface anchoring, and surface adsorption. Matrix elution involves the incorporation of soluble, uncross-linked polymers (e.g., polyvinyl alcohol) into the bulk matrix, enabling sustained emergence and replenishment at the surface. Surface anchoring utilises the chemical immobilisation of macromolecules (e.g., polyvinyl pyrrolidone) to form irreversibly bound loops and coils at the polymer surface. In contrast, surface adsorption relies on the physical attachment of block copolymers or cellulose derivatives via weak intermolecular forces, generating a highly lubricious, transient boundary layer. Micro-tribological characterisation demonstrates that these approaches fundamentally alter the frictional “fingerprint” of the base polymer. For example, surface-anchored structures achieved up to a 45% reduction in the mean coefficient of friction compared to unmodified substrates, effectively suppressing undesirable stick-slip behaviour. In addition, the macromolecular modifications significantly improve surface wettability by reducing advancing contact angles and hysteresis, thereby establishing a stable hydrophilic interface. Ultimately, modulating polymer surfaces through hydrophilic macromolecular entrapment, anchoring, and release offers a versatile and effective strategy for surface engineering of high-performance materials. A deeper understanding of the mechanisms governing macromolecular migration and persistence at the surface provides a robust framework for developing next-generation polymer networks with tailored interfacial mechanics suitable for a wide range of advanced applications in materials technology.

Keywords: Surface Modification; Hydrophilic Polymers; Biotribology; Macromolecular Release