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Polymer Brushes Derived from Polymer Single Crystals: Synthesis, Characterization and Applications.

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Abstract: Polymer brushes are polymer chains that are densely tethered to a substrate or interface. Different from free polymer chains, these tethered brushes exhibit unique chain conformation and behavior, and can be widely used for different applications due to their special properties. Traditional methods for polymer brush synthesis include grafting-to and grafting-from, both with intrinsic drawbacks. Here, a novel method using polymer single crystals (PSCs) as templating was employed to synthesize polymer brushes. Polymer single crystals are quasi 2-D lamellae structure with functional chain ends distributed on crystal surface. Due to the regularity of chain packing in crystal, the synthesized polymer brushes could have precisely controllable grafting density and tethering point. Complex brush architectures could be achieved by programmable growth of PSCs.

In this thesis, a serious of polymer brushes with various chain compositions/architectures were synthesized via polymer single crystal templating method. The first example is the synthesis of poly(ethylene oxide) (PEO) gradient brushes. The grafting density of PEO brushes was precisely tuned by co-crystallization of end-dissimilar PEO polymers and various gradient patterns could be obtained by programable crystal growth procedures. In a second example, poly(L-lactic acid)-b-poly(ethylene oxide) (PLLA-b-PEO) block-copolymer brushes were synthesized by triblock copolymer single crystal with a tethering segment to achieve a high grafting efficiency. In the third example, binary PEO/PS mixed brushes were synthesized using PLA stereocomplex single crystal as template. The stereocomplex structure prevents the formation of microdomains of the two immiscible polymers and guarantees a uniform mixing of the two polymers. The resulting mixed PEO/PS brushes exhibit characteristic phase structure upon treated with different solvents. Finally, patterned poly(4-vinylpyridine) brushes on PEO single crystals were used to assemble AuNPs into free standing frame structure with precisely tunable sizes. The AuNP frame exhibits special optical property, which is not observed in free AuNPs.