Synthesis, Reactivity, and pH-Responsive Assembly of New Double Hydrophilic Block Copolymers of Carboxymethyldextran and Poly(Ethylene Glycol)

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Abstract:

Double hydrophilic block copolymers (DHBC) were prepared by end-to-end coupling of two biocompatible water-soluble homopolymers: the polysaccharide dextran (Mw 8300 or 14,700 g mol□1) and □-amino poly(ethylene glycol) (PEG-NH2, Mw 3000 or 7000 g mol□1). The synthesis involved, first, specific oxidation of the dextran terminal aldehyde group and, second, covalent linkage of PEG-NH2 via a lactone aminolysis reaction. The diblock copolymers dextran-PEG (DEX-PEG) were converted in high yield into the corresponding carboxymethyldextran-PEG (CMD-PEG) derivatives with control over the degree of substitution, from 30 to 85 mol% CH2COOH groups per glucopyranosyl unit. Further modifications of a CMD-PEG block copolymer led to N-(2-aminoethyl)carbamidomethyldextran-PEG yielding a pair of oppositely-charged DHBC of identical charge density, chain length, and neutral block/charged block content. The properties of CMD-PEG in aqueous solutions were studied by static and dynamic light scattering as a function of solution pH, providing evidence of the pH-sensitive assembly of the copolymers driven by inter- and intra-chain hydrogen-bond formation.

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