Mechanical properties of hydrogels based on "physically" cross-linked double polymer networks of HM-PAA and a polysaccharide

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The problem of development of new high-strength soft materials is of great current interest. During the last years, investigation of hydrogels with high mechanical properties has drawn much attention. It is known that hydrogels based on one cross-linked polymer network in most cases have limited mechanical performance. An approach to dramatically enhance the mechanical properties of hydrogels consists in the creation of so-called double networks [1], which are two interpenetrating polymer networks with strongly contrasting properties. However, an essential drawback of chemically cross-linked double networks is the degradation of their mechanical properties after first deformation due to the irreversible breaking of the first network during deformation. In the recent years, some examples of fully "physically" cross-linked double networks have appeared [2]. However, most of them (e.g. with the first polysaccharide network cross-linked by metal ions) barely show fast and full recovery of mechanical properties due to low ability of cross-links to recover. The aim of this work is the development and study of fully "physically" cross-linked double networks comprised of a polysaccharide network cross-linked by easily recoverable dynamic covalent bonds and a hydrophobically modified polyacrylamide (HM-PAA) network.

Double network hydrogels were synthesized by a convenient one-pot method: first polysaccharide network was obtained, in which monomers and initiator for the synthesis of HM-PAA were dispersed. Then, second HM-PAA network was photopolymerized inside the existing first network. It was shown that as-prepared gels have mechanical properties (tensile elastic modulus and elongation at break) higher than those for the corresponding single networks. They show large hysteresis in compression cycles, which is explained by the breaking of dynamic covalent bonds that serve as cross-links in the first network, and the recovery of mechanical properties is full and proceeds very fast (in seconds) due to the recovery of dynamic covalent bonds. As a result of their fully "physical" labile character, double networks show self-healing properties after cut. Such double networks, which combine high mechanical strength with self-recovery and self-healing properties, are very promising for use as functional materials, e.g. in soft robotics, biomedical applications etc.

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References

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