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Tough Double Interpenetrating Network Hydrogel with Dual Reinforcement Mechanism

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Tough hydrogels, containing 3D cross-linked polymer networks and large amount of water, are highly rated for their wide applications in bioengineering, soft robotics and agriculture. Double interpenetrating networks show extraordinary toughness compared with single ones. Alginate, as a natural anionic polymer with advantages such as biocompatibility, low toxicity and low cost, could potentially be applied in biomedical applications. Ionic crosslinking methods with divalent cations (e.g., Ca^{2+}) is the most common route to prepare alginate hydrogels under mild conditions. In this work, we mixed hydrophilic polyacrylic acid (PAA) and alginate hydrogel to form a tough double interpenetrating network (DIPN) structure. In order to improve the toughness of the hydrogel, SiO_2 was added and in this way, we successfully achieved toughen DIPN hydrogel with ionic crosslinking and nanocomposites toughening mechanism simultaneously. We have systematically the impact of the amount of Ca^{2+} and SiO_2 in the hydrogel on the mechanical performances. Compared with PAA hydrogel with elongation of 240% and stress to failure at 0.36 MPa, we obtained a alginate (Ca^{2+})-PAA (SiO_2) hydrogel with elongation of 1100% and stress to failure at 0.80 MPa. Our work paves the way for the construction of tough hydrogels with multiple reinforcement methods, with potential applications of biocompatible hydrogels into drug delivery and tissue engineering.