Brain-resembling superabsorbent magnetic hydrogel based on starch for controlled drug release

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Introduction

Smart hydrogels are a particular class of sophisticated absorbable materials with enhanced properties that can be designed to a specific application. These hydrogels are able to undergo changes in their environments in response to external stimuli such as light, enzymatic degradation, exposition to a remote magnetic field, and so on. Starch, the second most abundant carbohydrate polymer in nature (next to cellulose), has been widely applied in food industry, owing to its great importance in the human diet, and in pharmaceutical sector, owing to its appealing properties such as biocompatibility, susceptibility to microbial degradation, and lost cost. The main focus of this work is on developing magnetic field-sensitive hydrogels from starch for controlled drug release systems. This approach consisted of incorporating magnetic particles as magnetite (Fe$_3$O$_4$) into a biomaterial-based device in which starch is the key constituent. We also focused evidencing the actual 3D network polymer structure of this polysaccharide in the swollen hydrogel. Starch was vinyl-functionalized with the use of glycidyl methacrylate (GMA) for further radical UV-induced polymerization-crosslinking reaction with acrylic acid (AAc) and N,N-dimethylacrylamide (DMAAm) in the presence of Fe$_3$O$_4$.

Result and Discussion

Iodine test revealed the actual 3D network structure of starch within the swollen composite. The configuration of the starch network resembled with that of a brain, as shown in Figure 1. FTIR spectroscopy of the hydrogel composites indicated interaction between the carboxylic groups from hydrogel and the iron ions from Fe$_3$O$_4$. The formation of the hydrogel composites also was evidenced by wide-angle X-rays diffraction (WAXD) and energy dispersive X-rays (EDS) spectroscopies. Minimal fragments of the swollen composite allowed the study of its actual morphology by TEM imaging. Stick-type structures were observed in the composite, result of a water-equilibrated structural configuration (obtained in a swollen state) of carboxyl groups of AAc coordinated to iron ions of Fe$_3$O$_4$.

Figura 1. Photos of starch-based water-swollen hydrogels after iodine test showing actual 3D network structure of starch in the hydrogel.

The albumin release mechanism of the hydrogel without Fe$_3$O$_4$ is governed by macromolecular relaxation. In the hydrogel composites, the albumin release was driven by macromolecular relaxation, but with a strong tendency to anomalous transport, because of both tortuosity effect and attenuation of the anion-anion electrostatic repulsion forces. On the other hand, the albumin release became more dependent on anomalous transport with an applied magnetic field, which intensified the tortuosity effect.

Conclusions

Brain-resembling superabsorbent hydrogel composites for drug delivery were developed via UV-induced copolymerization-crosslinking of vinyl-modified starch with AAc and N,N-dimethylacrylamide DMAAm in the presence of Fe$_3$O$_4$ particles.

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