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Biodegradable nanocomposite hydrogel structures with enhanced mechanical properties prepared by photo-crosslinking solutions of poly(trimethylene carbonate)–poly(ethylene glycol)–poly(trimethylene carbonate) macromonomers and nanoclay particles ☆

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ABSTRACT

Soft hydrogels with elasticity modulus values lower than 100 kPa that are tough and biodegradable are of great interest in medicine and in tissue engineering applications. We have developed a series of soft hydrogel structures from different methacrylate-functionalized triblock copolymers of poly(ethylene glycol) (PEG) with poly(trimethylene carbonate) (PTMC) by photo-crosslinking aqueous solutions of the macromonomers in 2.5 and 5 wt.% colloidal dispersions of clay nanoparticles (Laponite XLG). The length of the PTMC blocks of the macromonomers and the clay content determined the physicomechanical properties of the obtained hydrogels. While an increase in the PTMC block length in the macromonomers from 0.2 to 5 kg/mol resulted in a decrease in the gel content, the addition of 5 wt.% Laponite nanoclay to the crosslinking solution lead to very high gel contents of the hydrogels of more than 95%. The effect of PTMC block length on the mechanical properties of the hydrogels was not as pronounced, and soft gels with a compressive modulus of less than 15 kPa and toughness values of 25 kJ m^{-3} were obtained. However, the addition of 5 wt.% Laponite nanoclay to the formulations considerably increased the compressive modulus and resilience of the hydrogels; swollen nanocomposite networks with compressive modulus and toughness values of up to 67 kPa and 200 kJ m^{-3}, respectively, could then be obtained. The prepared hydrogels were shown to be enzymatically degradable by cholesterol esterase and by the action of macrophages. With an increase in PTMC block length in the hydrogels, the rates of mass loss increased, while the incorporated Laponite nanoclay suppressed degradation. Nanocomposite hydrogel structures with a designed gyroid pore network architecture were prepared by stereolithography. Furthermore, in the swollen state the porous gyroid structures were mechanically stable and the pore network remained fully open and interconnected.

1. Introduction

Hydrogels are hydrophilic networks that can take up very large amounts of water. Their biocompatibility and their consistency (which resembles the extracellular matrix (ECM)) make these materials very interesting for numerous applications in medicine and biomedical engineering [1]. In particular, biodegradable hydrogels are interesting materials from which to prepare matrices for cell encapsulation or scaffolding structures that support the adhesion and proliferation of cells. It has been demonstrated that variations in the rigidity of these highly compliant hydrogels can be a determining factor in regulating the cellular processes that determine the morphology, motility and differentiation of (mesenchymal stem) cells [2,3].

Despite their many advantageous properties, the brittleness and fragility of hydrogels has limited their use in load-bearing applications such as in the regeneration of cartilage, tendon, dermis and arterial walls [4]. While many natural hydrogel structures combine a high uptake of body fluids with low elasticity modulus, high elongation at break and high toughness, most synthetic hydrogels that are currently used in medical applications and tissue engineering easily fracture under only mild loading conditions.

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