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ABSTRACT:

Multiscale Structural Characterization of Biocompatible Poly(trimethylene carbonate) Photoreticulated Networks – From Conception to Mechanical Behavior

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Photo-cross-linked biodegradable polymer networks are highly interesting materials for use in biomedical applications compared to current alternatives (PCL, PLA or chitosan), since a wider range of physical and mechanical properties, crosslinking densities, and degradation characteristics can be obtained [1-3]. In this work we consider Poly(trimethylene carbonate) (PTMC), a synthetic biodegradable, biocompatible, amorphous, and flexible polymer. By photo-cross-linking PTMC macromers, tough, tear-resistant networks can be obtained [1-3]. Thus, PTMC networks have been investigated for applications such as meniscus [2] and bone [3] implants, and microvascular networks. The application of PTMC networks depends largely on their mechanical behavior [1,2] which strongly depends on the network morphology. For these materials, these structure-property relationships have been successfully investigated by combining macroscopic tensile and Dynamic Mechanical Analyses (DMA) mechanical characterizations with calorimetric (DSC) measurements, and in particular molecular-scale Time-Domain Double-Quantum (DQ) 1H NMR analyses. Firstly, PTMC networks with macromer molecular weights between 3 and 40 kg/mol, were studied. A linear correlation between the crosslink density measured by DMA and NMR was observed, prompting the conclusion that the macroscopic mechanical behavior of crosslinked PTMCs is directly linked to their intrinsic network structure [4]. Then PTMC networks obtained by solvent casting were studied. Networks from macromers with molecular weights of 3, 18 and 32 kg/mol were prepared with various solvent contents. It was shown that when the solvent content increases, the crosslink density measured by DMA and NMR decreases [5]. Moreover, for the PTMC with a molecular weight of 18 kg/mol, its mechanical toughening behavior was greatly increased without damaging its Young's modulus when the solvent content increased. This was attributed to a synergetic effect of the presence of free and dangling chains in the network [5]. These same PTMC networks were considered to produce hybrid semi-interpenetrative networks with chitosan. Our multiscale approach showed that it was possible to pilot the hybrid materials thermomechanical behavior by varying the macromer mass of PTMC [6]. Finally, PTMC networks were also evaluated via equibiaxial tensile tests, which showed a good correlation with DMA as regards the inner structure of PTMC [7]. These results allow a better understanding of PTMC materials and the tailoring of their properties to enhance their potential use in biocompatible applications, achieved by combining macroscopic techniques with solid-state NMR through a robust scientific approach.

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