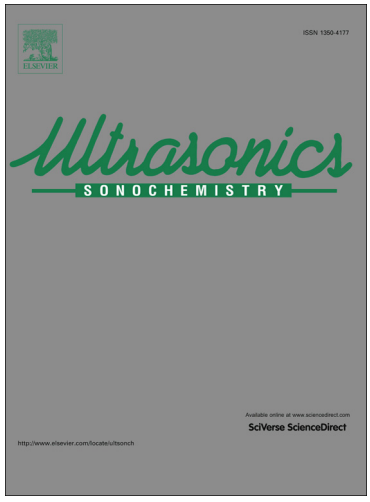

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Chiral betulin-imino-chitosan hydrogels by dynamic covalent sonochemistry

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Abstract

A series of chiral hydrogels was prepared from a homogeneous mixture of chitosan and betulinic aldehyde in different molar ratios, under the effect of ultrasound. The hydrogelation mechanism has been investigated by FTIR and CD spectroscopy, wide angle X-ray diffraction and polarized light microscopy. The morphology of hydrogels was examined by SEM. The swelling ability has been tested in three media of different pH. It was concluded that hydrogelation occurred by different pathways, closely related to the peculiarities of the chitosan-betulin systems. Circular dichroism measurements revealed chiroptical properties of the hydrogels, correlated to their content and crosslinking pathway.

Keywords: hydrogels, chitosan, betulin, dynamic covalent chemistry, chirality

1. Introduction

Biomaterials have been extensively studied in the last decades for their bio-applicability potential as implants, scaffolds for tissue engineering or matrix for targeted drug delivery, but also in other important domains as agriculture, environment protection, or food chemistry [1]. To this aim, natural compounds proved to be the most promising building blocks to create biomaterials due to their potential to preserve the biological properties and to mimic the natural tissues [2]. In this respect, our group developed a new strategy of hydrogelation of the chitosan biopolymer with monoaldehydes of natural provenience to provide hydrogels with eco-design [3-9]. The method is mainly based on the ability of the reversible imine units to self-order into clusters, which play the role of chitosan crosslinkers. In this way, using various aldehydes, biocompatible hydrogels with good mechanical properties [5,6], superporous morphology [7], antitumor activity [6], antifungal properties [8] or efficient luminescence [9] were obtained, paving the way to a new type of multifunctional hydrogels.

A class of natural compounds which lately attracted a lot of interest is that of chiral terpenes, due to their ability to fix biotargets and to induce special supramolecular architectures of the materials containing them [10,11]. An important representative is betulin