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Agarose based multifunctional materials: Evaluation of thixotropy, self-healability and stretchability

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ABSTRACT

This paper reports a microwave assisted one pot facile synthesis of ester derivatives of agarose (Agr-GA_{Est}) through chemical reaction of agarose (Agr) with gallic acid (GA), an organic acid found in many plants employing carbodiimide chemistry. Agr-GA_{Est} was characterised by FT-IR, ^{13}C NMR spectroscopy, thermogravimetric analysis (TGA), DMA measurements, scanning electron microscopy (SEM), UV–vis spectrophotometry and rheometry. The native agarose was insoluble in ethylene glycol, but Agr-GA_{Est} (obtained at an optimized molar ratio of 1: 0.5) formed good quality gel ($\tan\delta \sim 0.1$) at 4% (*w/v*) concentration. The gel thus obtained exhibited substantial degree of thixotropy (hysteresis loop area = 38.73%), rapid self-healing ability (12 min) upon complete cleavage of the gel and excellent stretching ability (>20 times of its original length). These types of multifunctional gels would find applications in food and personal health care industries.

1. Introduction

Multi-functional materials that show reversible properties such as self-healing are known as dynamic materials and are inspired by biological systems, where any damage got self-repaired (Phadke et al., 2012). These type of materials often has superior mechanical properties and finds multiple applications (Lehn, 2005). Most of the polymer based gel materials show dual properties such as liquid like flow and elastic behaviour and during their physical interactions, suitable polymeric entanglements lead to the solid like properties (Urban, 2012). These remarkable properties of gels make them very good candidates to induce tailor made functional properties such as self-healing or solvent responsive healing (Sharma, Mondal, Mukesh, & Prasad, 2013). Cleavage of the macromolecular chains leads to the formation of free radicals or functional groups such as —C=C—, —COOH, —NH_2, —OH, —Si—O, *etc*., and chain mobility or diffusion in the segments bring reactive groups in contact with each other resulting repairing of physical network (Yang & Urban, 2013). There are several reports on self-healing polymeric gel systems fabricated mainly by cross-linking with groups containing —NH—CO— moiety capable of forming reversible covalent bonds upon cleavage (Hager, Greil, Leyens, van der Zwaag, & Schubert, 2010; Kushner, Vossler, Williams, & Guan, 2009). In oppose to the conventional approach of introducing groups capable to induce self-healing, Imato, Nishihara, Kanehara, Amamoto, Takahara, and Otsuka (2012) have prepared a polymer gel by the reversible formation of diarylbibenzofuranone cross-linker from the dimerization of stable arylbezofuranone, which self-healed without any external stimuli (Imato et al., 2012). Although different kinds of interactions have been exploited to design polysaccharide-based physical networks such as hydrogen bonds (Braccini, Rodríguez-Carvajal, & Pérez, 2005), ionic (Chung et al., 2002), host–guest recognition (Charlot, Auzély-Velty, & Rinaudo, 2003), non-covalent interactions in polysaccharide ion gels (Sharma, Mondal, Mukesh, & Prasad, 2013), but there are no report on the studies on non-covalent bond triggered self-healing ability of functionalized polysaccharide including agarose based materials and healing induced by solvents.

Highly stretchable hydrogels are of great interest in tissue engineering and for bio-medical applications. Ionically crosslinked alginate and covalently crosslinked polyacrylamide gels are reported to have stretchability more than 20 times of its original length (Sun et al., 2012). The prior arts revealed that there are no reports on the multifunctional materials based on seaweed derived polysaccharides. These encourage us to take the study on

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