"TREE"-D PRINTING OF CELLULOSE-BASED HYDROGELS BY PHOTOPOLYMERIZATION

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The digital light processing (DLP) is a rapid, clean 3d printing technology that enables the construction of complex parts by the photopolymerization of a liquid formulation. The development of new, natural inks is crucial to expand DLP applications and to comply with the eco-sustainability's requirements. Different biosourced polymers have been modified and recently proposed for DLP printing of 3D shaped hydrogels [1], and the possibility to tune their mechanical properties can enlarge their range of application. Herein, we developed natural-based, composite hydrogel inks for DLP printing, composed entirely of cellulosic-material. The properties of acrylated-carboxymethyl cellulose (M-CMC) hydrogels [2] were modified by the addition of cellulose nanocrystal (CNC) extracted from empty oil palm fruit bunches. CNC were also modified to impart photocurable, methacrylic groups. M-CMC, pristine CNC/M-CMC and methacrylated CNC (M-CNC)/M-CMC hydrogels were tested to evaluate the effects of both the reinforcers and their functionalization. The formulations' rheological properties and photoreactivity were investigated in order to select the most suitable compositions for DLP-3d printing. Either the presence of CNCs or that of M-CNC does not affect the photoreactivity; instead, it endows the cured hydrogels with greater storage modulus. The nano-reinforcers assist the formation of a more rigid network, with improved mechanical properties and printability but reduced swelling ability. The three hydrogels, that show pH-sensitive behavior, were assessed for their suitability for 3d-printing of various geometries. So far as we know, this is the first work that reports the successful photopolymerization-based 3d printing of "all-cellulose" hydrogels.

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[1] Voet, V. S. D., Guit, J. & Loos, K. Sustainable Photopolymers in 3D Printing: A Review on Biobased, Biodegradable, and Recyclable Alternatives. *Macromol. Rapid Commun.* **42**, 1–11 (2021).

[2] Melilli, G. *et al.* DLP 3D printing meets lignocellulosic biopolymers: Carboxymethyl cellulose inks for 3D biocompatible hydrogels. *Polymers (Basel).* **12**, 1–11 (2020).