THE ROLE OF PHYSICAL CROSS-LINKS IN CHEMICAL CROSS-LINKING OF GELATIN METHACRYLOYL HYDROGELS

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Gelatin methacryloyl (GM) hydrogels are of great interest for various biomedical applications such as drug delivery or tissue engineering. Their mechanical properties are a critical feature for most applications and are typically tuned by e.g. the variation of the degree of methacryloylation (DM). However, prior work also suggests that thermal sample history plays an often overlooked role in that regard. [1] The impact of thermal sample history, substitution, and conformational changes of GM in aqueous solution were examined as well as physical and chemical gelation behavior. To this end, a set of modified gelatins (GM) with varying substitution patterns were investigated *via* circular dichroism (CD) spectroscopy, differential scanning calorimetry (DSC), and *in situ* RT-NIR photorheology. DSC measurements demonstrated a decreased tendency towards physical gelation with increasing DM. CD spectroscopy allowed to correlate this finding with the decreased formation of triple-helical structures in GM of high DM. RT-NIR-photorheology proved the significant increase of hydrogel stiffness for the proposed dual crosslinking mechanism, even for non-physically gelling GM. Interestingly, found double bond conversions do slightly decrease for the dual crosslinking method. [2] We suggest that GM hydrogel stiffness is strongly dependent on the total number of cross-links including physical interactions. Not only are those findings important regarding reproducibility of mechanical properties, but also in terms of their optimization with regard to biomedical applications, where strong hydrogels are a desirable outcome.

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[1] L. Rebers, T. Granse, G. E. M. Tovar, A. Southan and K. Borchers, Gels, 2019, 5, 4.

[2] L. Rebers, R. Reichsöllner, S. Regett, *et al.*, *Sci Rep*, 2021, 11, 3256.