Design and synthesis of ionically cross-linked graphene oxide-reinforced hydrogel.

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Hydrogels have attracted significant attention due to their high water content, tuneable structure, and excellent biocompatibility, making them promising materials for biomedical engineering, tissue scaffolding, and environmental remediation. [1] However, conventional hydrogels sometimes exhibit poor mechanical strength, limited structural stability, and restricted functional versatility, which constrain their use in demanding applications. One approach to overcome these limitations involves incorporating nanomaterials as reinforcing agents within hydrogel matrices. Among these, graphene oxide (GO) is particularly attractive due to its highly oxidized nature and high specific surface area. This provides a large and functional oxygen-rich interface that promotes strong interactions with polymer networks. Such interactions allow enhanced surface contact between the two components, further enhancing the hydrogel mechanical strength, elasticity and responsiveness, while maintain high water retention capabilities. However, achieving uniform dispersions of GO nanosheets within polymer matrices remains a significant challenge, despite being critical for fully optimising the mechanical and functional properties of hydrogels. [2]

A powerful strategy to overcome this limitation, which has been partially explored in the context of nanocomposites, [3-5] is to exploit GO's acidic functional groups and pair them with hydrogels of complementary ionic character, allowing strong electrostatic interactions to form at their interface. These interactions are expected to significantly improve GO dispersion, enhance compatibility between organic and inorganic phases, and strengthen interfacial adhesion. [4] In addition, homopolar surface charges are anticipated to induce electrostatic repulsion, improving GO colloidal stability and enabling greater structural control in the resulting Nanoscale Ionic Hydrogels (NIHs). [6] Collectively, this approach is expected to yield GO-based NIHs with improved mechanical properties and enhanced performance, effectively moving beyond the concept of a traditional polymer matrix toward a single-phase hybrid system. [7]

Herein, a GO-reinforced poly(2-hydroxyethyl methacrylate-co-2-(dimethylamino)ethyl methacrylate) (HEMA–DMAEMA) hydrogel is synthesised to maximise electrostatic interactions between the basic amine groups of the polymer network and the carboxylic acid functionalities of GO. To further enhance such interactions, GO nanosheets were pre-treated with succinic anhydride to introduce additional carboxylic acid groups at their edges (C-GO).

Preliminary results indicate improved dispersion of C-GO within the pre-polymer mixture, suggesting enhanced compatibility between DMAEMA monomers and C-GO sheets compared to standard GO. Ongoing spectroscopic characterisation will help elucidate and assess interfacial interactions (Infrared and Raman Spectroscopy), surface chemistry and chemical environment (X-Ray Photoelectron Spectroscopy), and nanoscale dispersion and morphology (Scanning Electron Microscopy). Moreover, rheological assessment will provide further insight into the system's structure-property relationships, allowing us to link this design approach to the resulting mechanical performance and network architecture of the hydrogel.

Keywords:
Hydrogels, Graphene oxide, Hybrid materials, Nanocomposites.

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