

# Exploring Graphene-Based Materials As New Nanofillers To Reinforce Poly(Ethylene Glycol) Hydrogels For Load-Bearing Applications

**Helena Ferreira**

Patrícia Henriques, Andreia Trindade Pereira, Inês Castro Gonçalves

*Instituto de Inovação e Investigação em Saúde (i3S), Universidade do Porto, Rua Alfredo Allen 208, 4200-135, Porto, Portugal*

*Instituto de Engenharia Biomédica (INEB), Universidade do Porto, Rua Alfredo Allen 208, 4200-135, Porto, Portugal*

helenaferrera@i3s.up.pt

## ABSTRACT

Poly(ethylene glycol) (PEG) hydrogels are biocompatible and highly resistant to protein adsorption, serving as a “blank slate” with tunable physicochemical properties. However, hydrogels are typically very weak, preventing their use in load-bearing applications, e.g. for cartilage, intervertebral disc, blood vessels or cardiac valves. Several strategies have been proposed for their mechanical reinforcement, such as the incorporation of nanofillers [1].

Graphene is a single-layer honeycomb lattice of sp<sup>2</sup>-bonded carbon atoms with outstanding stability, mechanical and electroconductive properties. It is the thinnest yet strongest material ever measured [2]. Graphene-based materials (GBM), particularly oxidized forms – which are reportedly less cytotoxic than reduced ones [3] – have been proposed as nanofillers for hydrogels [4, 5].

The goal of this work is to explore the use of different GBM as nanofillers to reinforce PEG hydrogels, turning them into stiffer and stronger hydrogels. We aim to understand the influence of different GBM parameters, such as concentration, thickness, lateral size or oxidation degree, in the mechanical properties of PEG/GBM composite hydrogels.

Graphene oxide (GO) and few-layer graphene oxidized (FLGO) were synthesized from graphite powder and FLG, respectively, by modified Hummer’s method. According to XPS analysis, GO contained 30.2% of oxygen atoms, while FLGO contained 31.6% oxygen atoms, which confirms the oxidation of graphite and FLG, respectively. TEM images show GO sheets with a lateral size of < 3 μm, while FLGO sheets were ~5 μm large.

PEG/GBM composite hydrogels were produced by *in situ* crosslinking of a precursor solution containing PEG dimethacrylate macromers (8 kDa, 15 wt%) and different concentrations of dispersed GO, FLG or FLGO. Tensile tests show that composite hydrogels with ≥2 wt% GO are significantly stiffer and stronger than neat PEG hydrogels. The higher the GO concentration, the greater the stiffness and strength appear to be (in the 2-4 wt% range). The incorporation of 1 wt% FLGO in PEG hydrogels results in an increase of Young’s modulus from 17.9±4.4 kPa to 78.7±5.8 kPa. In fact, the tensile properties of 1 wt% FLGO hydrogels were comparable to those of 4 wt% GO hydrogels. FLGO sheets have a larger surface area with which PEG chains can interact, possibly providing more interface to transfer stress from the polymer network to the nanofillers (vs. GO sheets). Regarding the oxidation degree, FLG appears to not reinforce neat PEG hydrogels. The low oxidation of FLG prevents its interaction with PEG chains, as expected. The reinforcement is also greater when GO and FLGO are incorporated in the hydrogel’s precursor solution as a colloid than as a powder.

The development of such composites paves the way for the application of hydrogels as versatile as PEG in load-bearing tissues.

## REFERENCES

- [1] Kaniewska, Karbarz, Katz, *Applied Materials Today*, no. 20 (2020) pp. 100776.
- [2] Lee, Wei, Kysar *et al.*, *Science*, no. 5887 (2008) pp. 385.
- [3] Pinto, Gonçalves, Magalhães, *Colloids and Surfaces B: Biointerfaces*, no. 111 (2013) pp. 188.
- [4] Jang, Hong, Cha, *Journal of the Mechanical Behavior of Biomedical Materials*, no. 69 (2017) pp. 282.
- [5] Pereira, Henriques, Costa *et al.*, *Composites Science and Technology*, no. 184 (2019).

## ACKNOWLEDGMENTS

Funding from Projects POCI-01-0145-FEDER-032431, PTDC/CTM-COM/32431/2017, UID/BIM/04293/2019.