

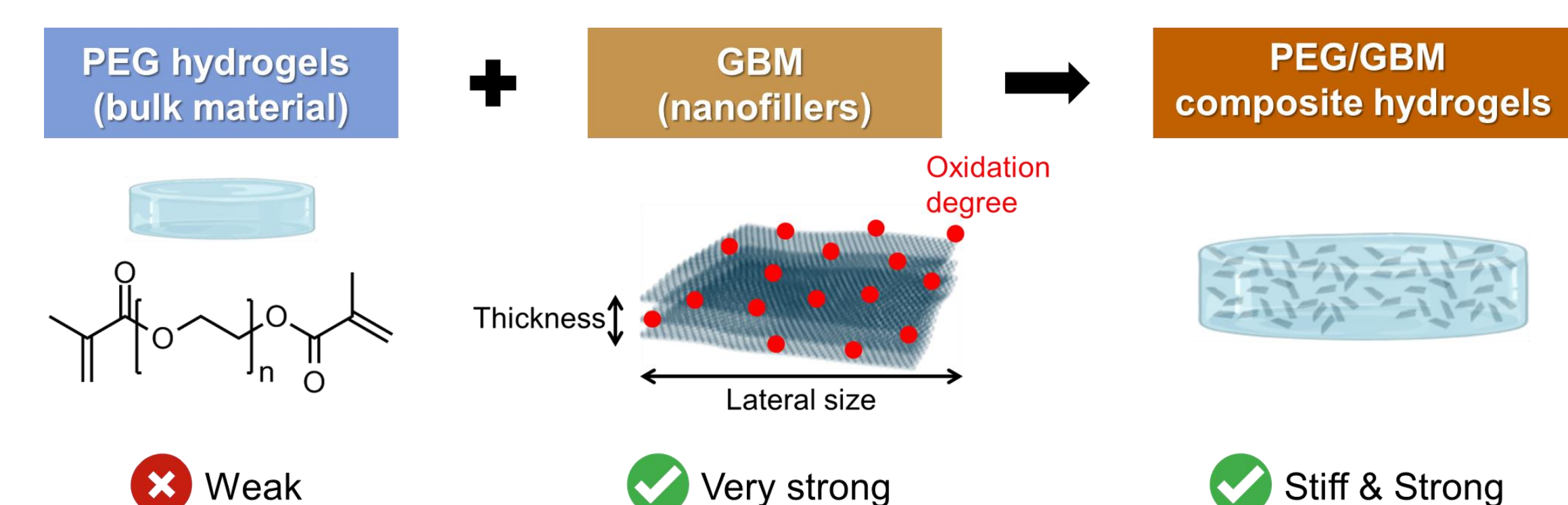
Exploring Graphene-Based Materials As New Nanofillers To Reinforce Poly(Ethylene Glycol) Hydrogels For Load-Bearing Applications

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Introduction & Objectives

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. Graphene is a single-layer honeycomb lattice of sp²-bonded carbon atoms. It is the thinnest yet strongest material ever measured [1]. Graphene-based materials (GBM), particularly oxidized forms – which are reportedly less cytotoxic than reduced ones [2] – have been proposed as nanofillers for hydrogels [3,4]. The goal of this work is to explore the use of different GBM as nanofillers to reinforce PEG hydrogels. We aim to understand the influence of GBM parameters (thickness, lateral size, oxidation degree or form) on the tensile properties of composite hydrogels.



Nanofillers: single-layer and few-layer graphene

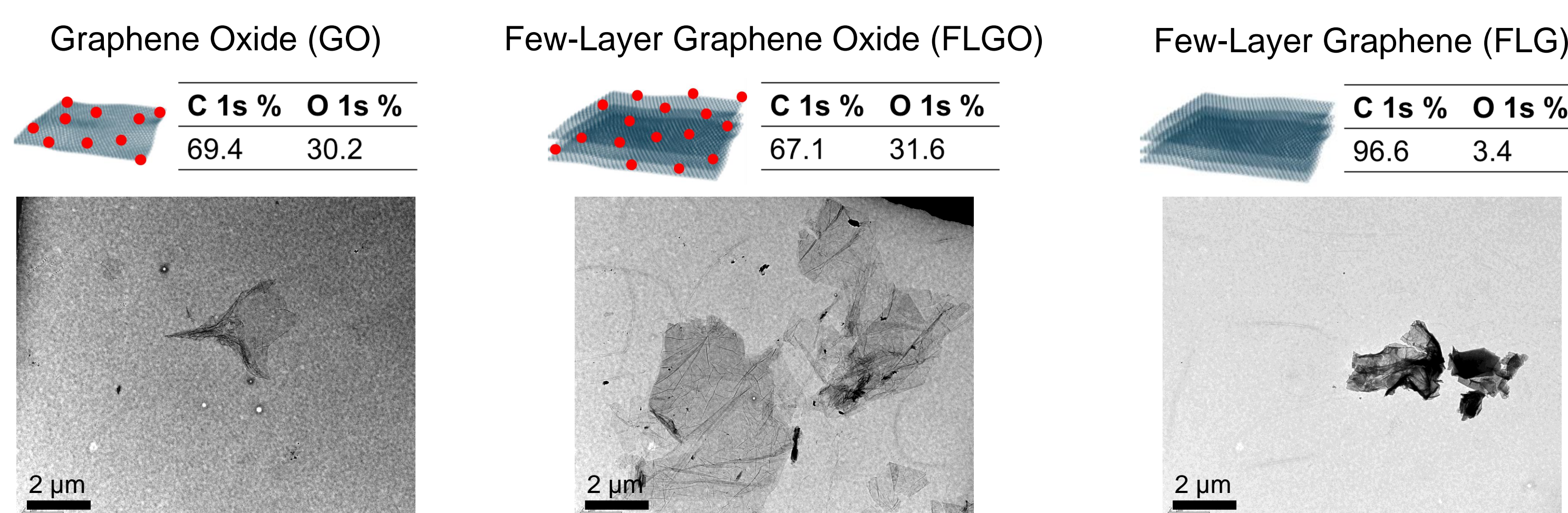
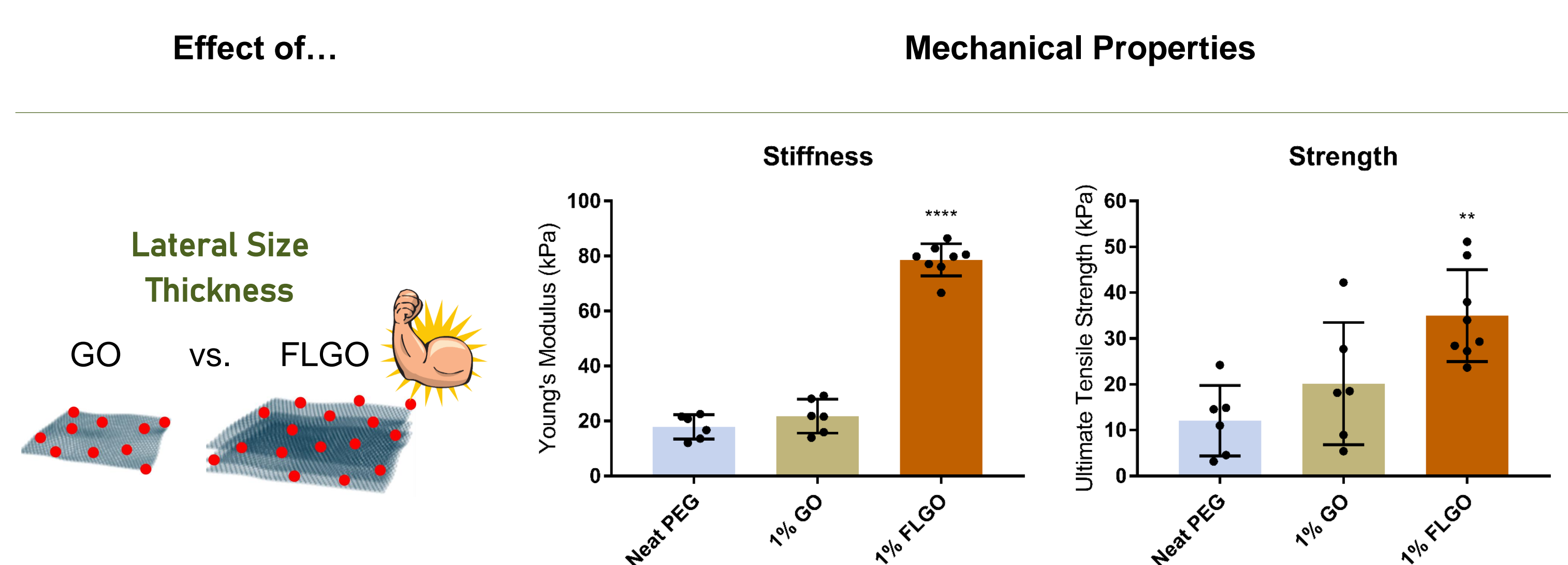


Fig.1 – X-ray photoelectron spectroscopy (XPS) analysis revealing the atomic percentage (carbon and oxygen) of GO, FLGO and FLG; TEM images of GO, FLGO and FLG; scale bar: 2 μm. GO and FLGO were synthesized from graphite powder and FLG, respectively, by modified Hummer's method.

Effect of GBM thickness, lateral size, oxidation degree or form



The incorporation of 1 wt% FLGO causes a 339% increase in stiffness and 190% increase in strength.

Mechanical properties of PEG/GBM composite hydrogels

Effect of GO concentration

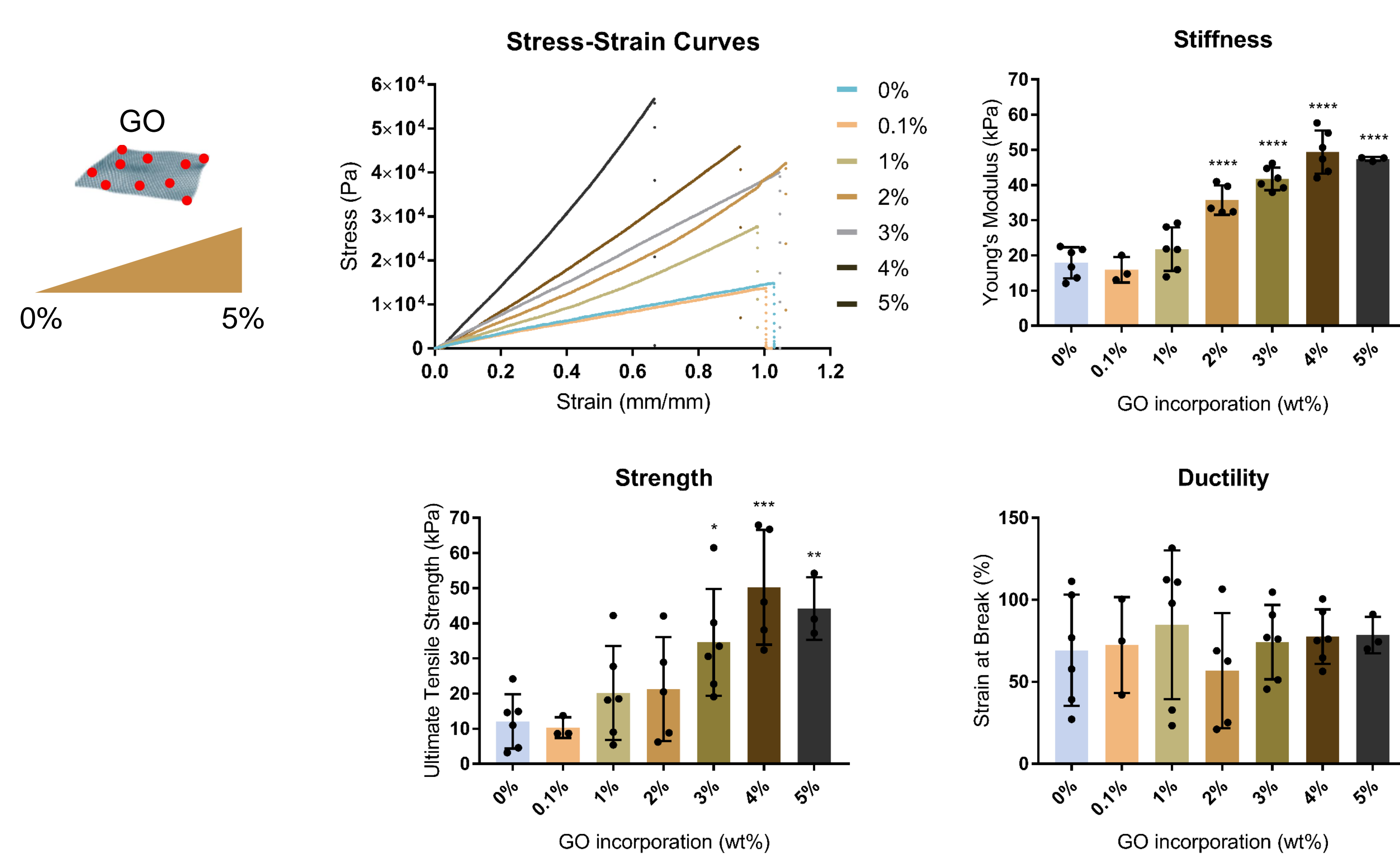
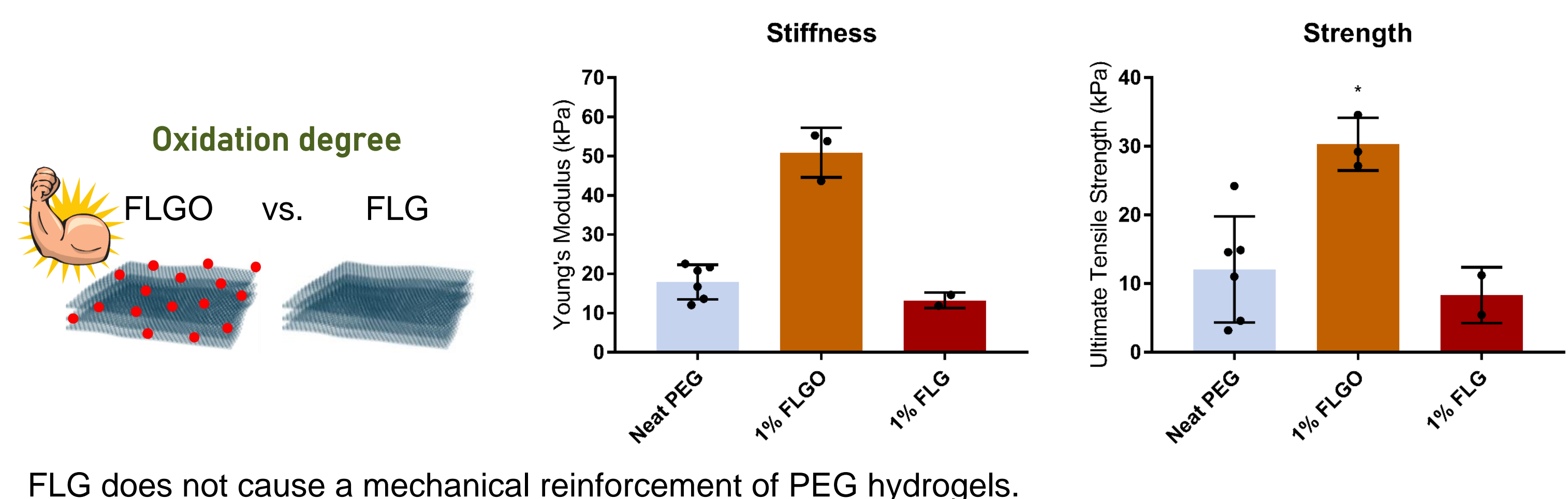
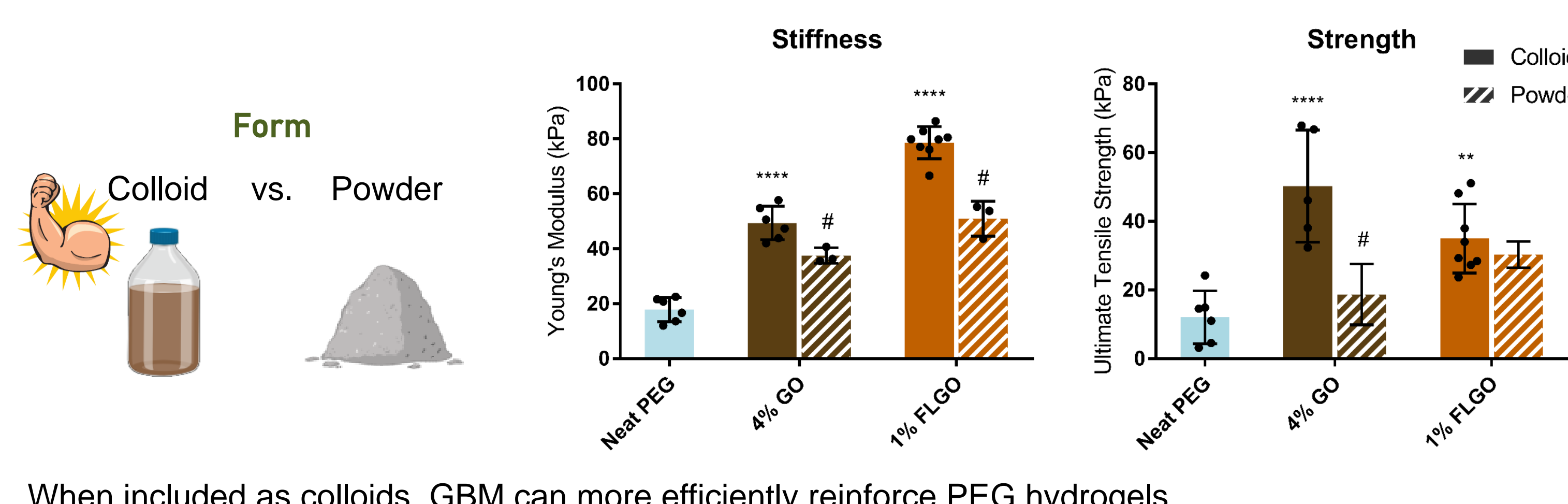


Fig.2 – Stress-strain curves, Young's modulus, ultimate tensile strength and strain at break of composite hydrogels with 15 wt% PEG dimethacrylate (MW= 8 kDa) and 0-5 wt% of GO, evaluated according to ASTM D882-02 (hydrated thin films) using a TA.XTplus Texture Analyser. Data shown as mean ± SD. One-way ANOVA, * p < 0.05, ** p < 0.01, *** p < 0.001, **** p < 0.0001 vs. 0% GO (neat PEG hydrogels).



FLG does not cause a mechanical reinforcement of PEG hydrogels.



When included as colloids, GBM can more efficiently reinforce PEG hydrogels.

Fig.3 –Young's modulus and ultimate tensile strength of hydrated composite hydrogels with 15 wt% PEG (MW= 8 kDa) and GBM (as indicated). Data shown as mean ± SD. One-way ANOVA for lateral size/thickness; Kruskal-Wallis test for oxidation degree; * p < 0.05 ** p < 0.01, **** p < 0.0001 vs. neat PEG hydrogels; Mann-Whitney test between powder and colloid, for GO and FLGO separately, # p < 0.05.

Take-Home Message

- Oxidized forms of graphene (GO and FLGO), especially when added as colloids, can be used as nanofillers to reinforce PEG hydrogels.
- The incorporation of GBM with larger lateral size (i.e. FLGO) results in stiffer and stronger composite hydrogels, given their larger surface area to which PEG chains can transfer stress during deformation.

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References

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