Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Graphene hybrid materials?: The role of graphene materials in the final structure of hydrogels

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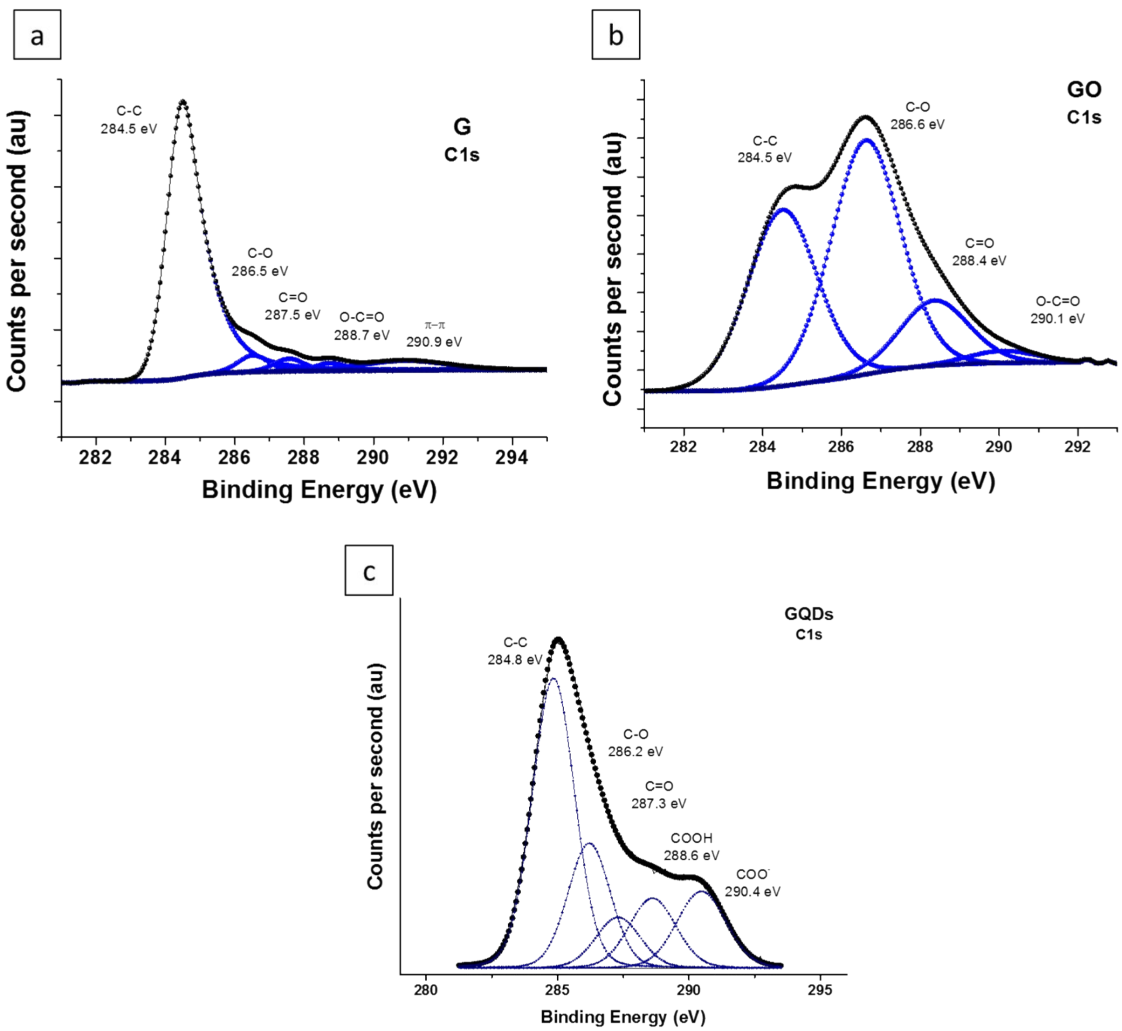


Fig. S1. C 1s Core Spectra of a) Graphene, b) Graphene Oxide and c) Graphene Quantum Dots.

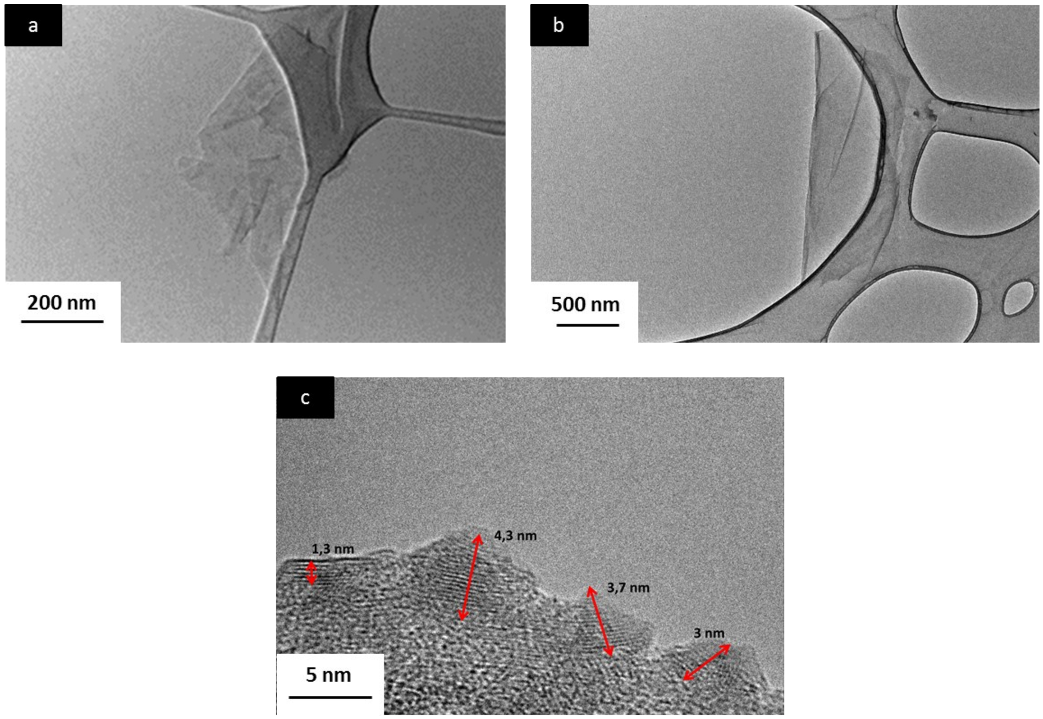
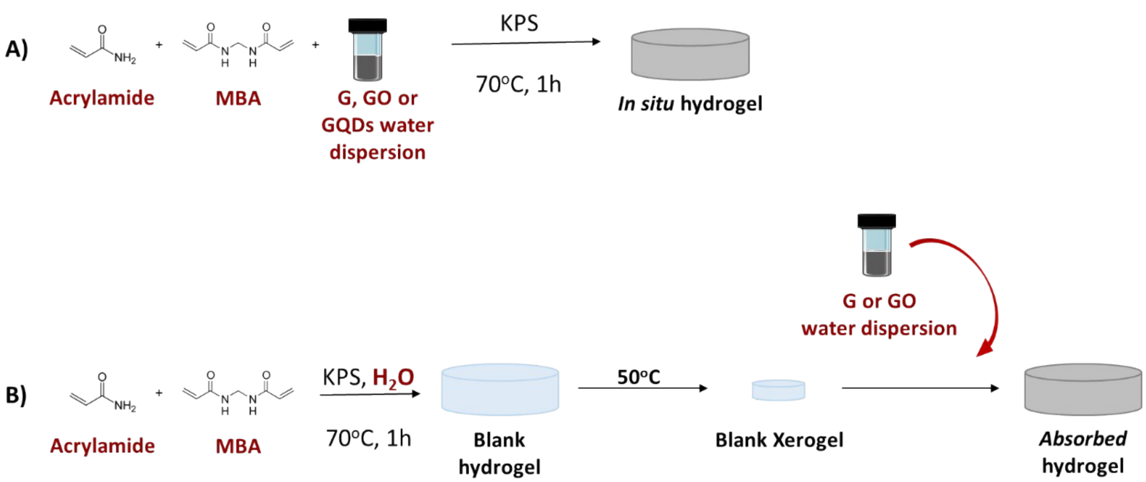


Fig. S2. TEM and HRTEM images of a) Graphene, b) Graphene Oxide and c) Graphene Quantum Dots.



Scheme S1. A) Synthesis of the *in situ* materials. B) Synthesis of the *absorbed* hydrogels.

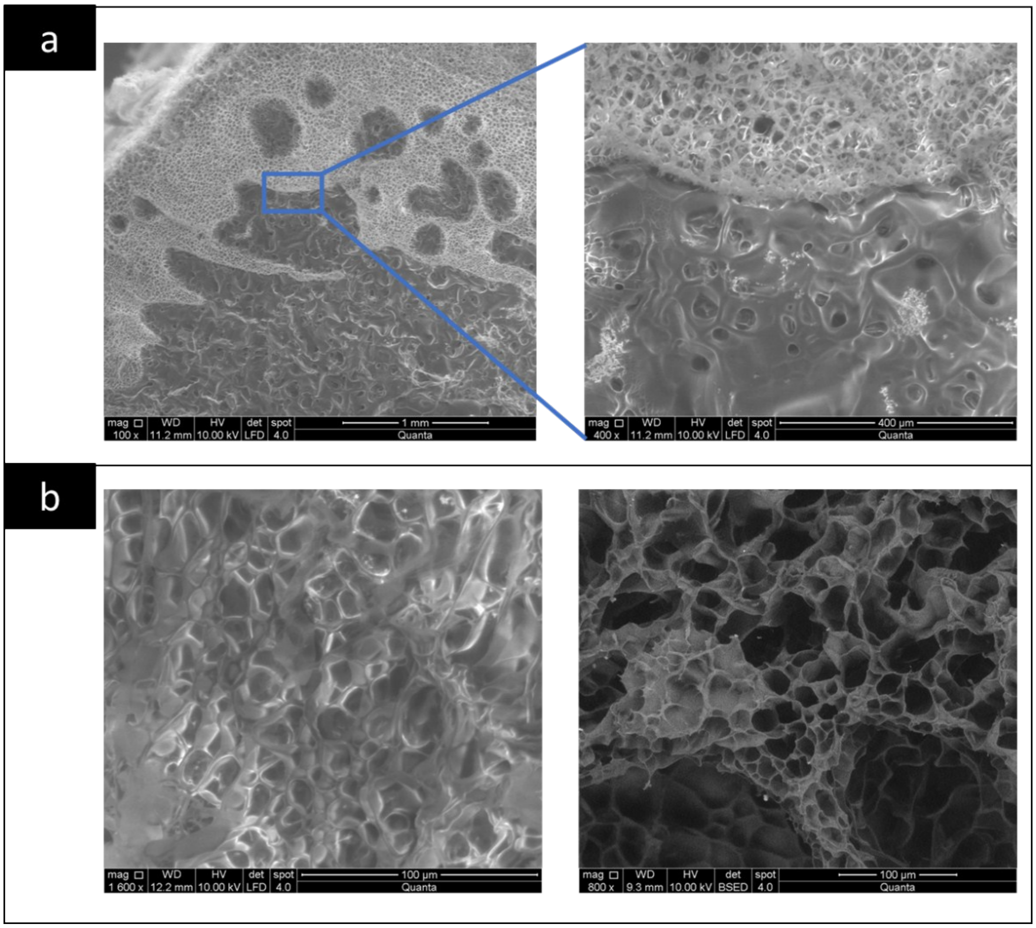


Fig. S3. SEM images of a) 0.2-AMGO and b) 2-AMGO

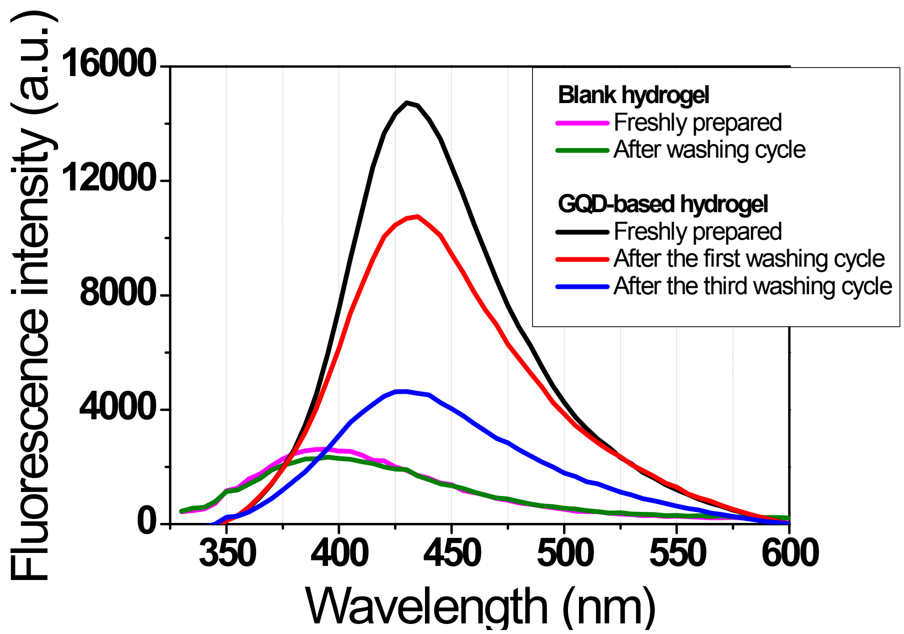


Fig. S4. PL comparison between the GQD-based hydrogel and the blank hydrogel.

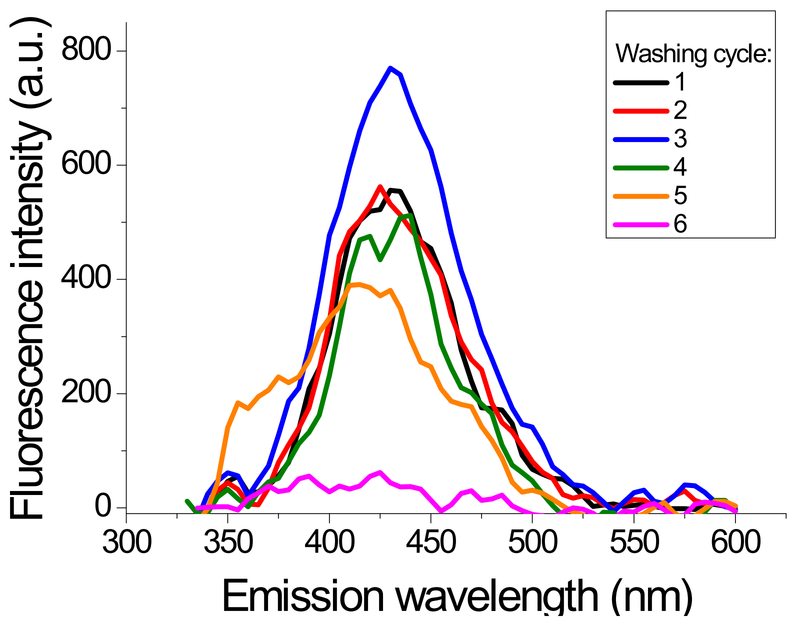


Fig. S5. PL intensity of the washing water from GQDs-based hydrogels washing cycles.

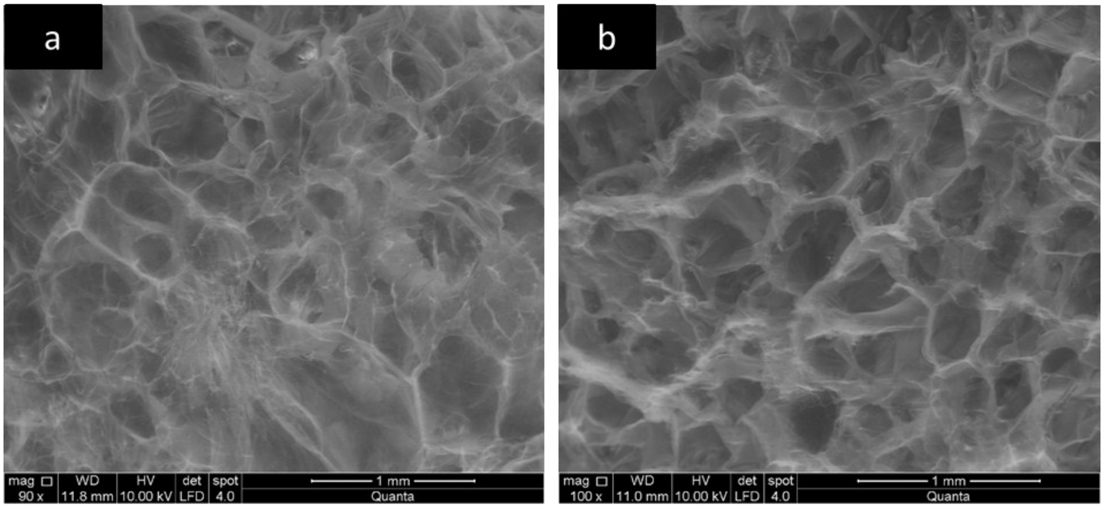


Fig. S6. SEM images of GQDs-based hydrogels a) before and b) after the washing cycles.

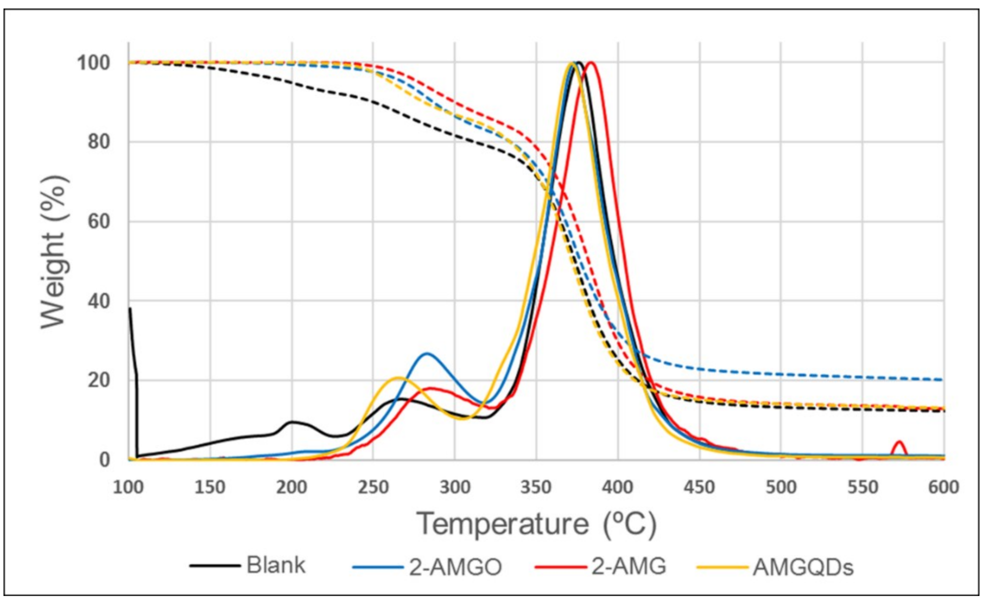


Fig. S7. TGA spectra for the blank (black), 2-AMG (red), 2-AMGO (blue) and AMGQDs (yellow) hydrogels.

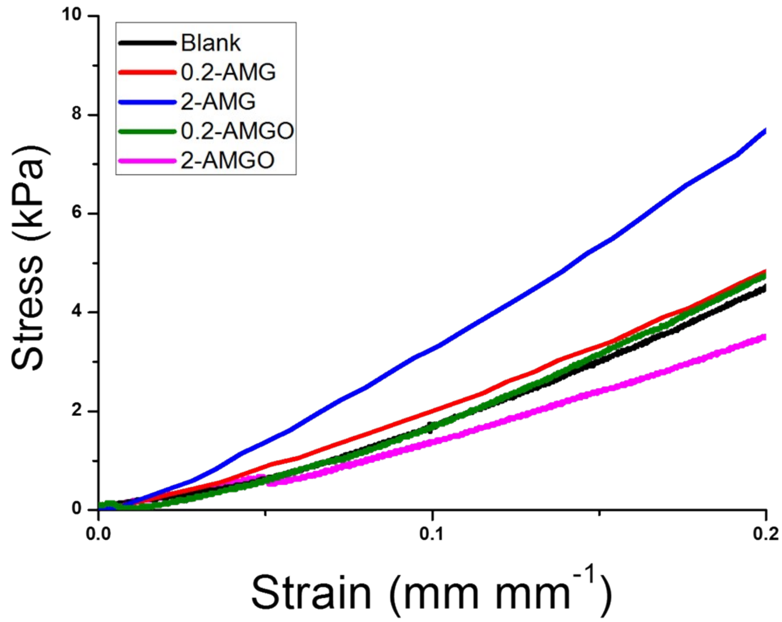


Fig. S8. Stress/Strain curves until 20% of strain of blank (black), 0.2-AMG (red), 2-AMG (blue), 0.2-AMGO (green) and 2-AMGO (pink) hydrogels.

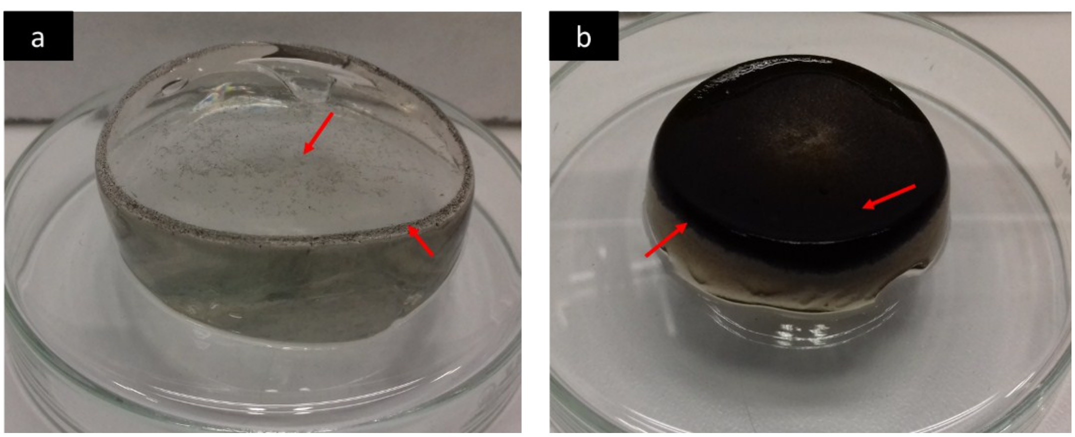


Fig. S9. Digital picture of the *in situ* GO-based hydrogels when they are swollen until the maximum swelling degree. A) 0.2 mg mL^{-1}, b) 2 mg mL^{-1}. Aggregates are highlighted by the arrows.

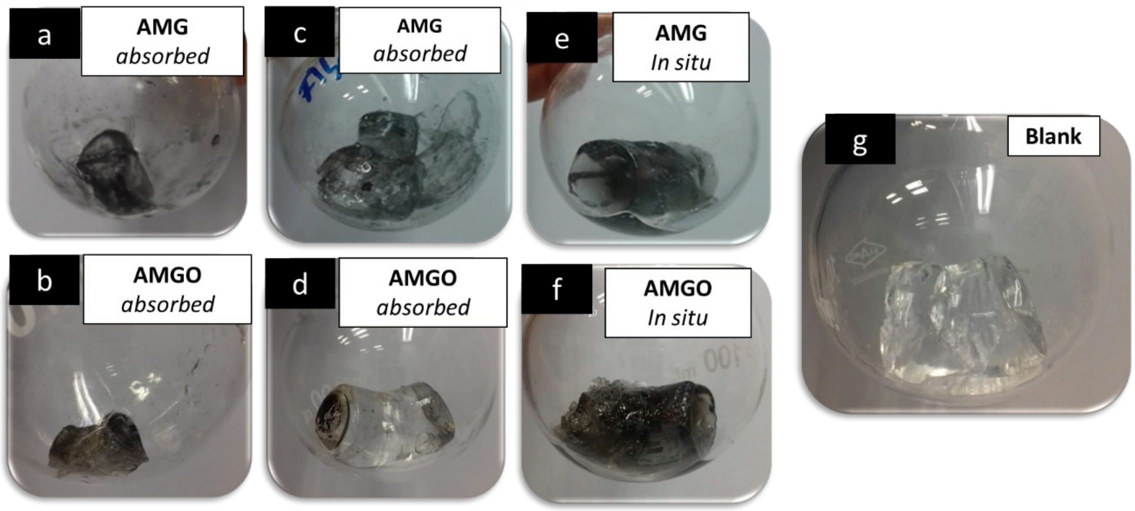


Fig. S10. Partially released of a) G from 0.2-AMG *absorbed* hydrogel. b) GO from 0.2-AMGO *absorbed* hydrogel. Final appearance after three deswelling/swelling cycles of: c) 0.2-AMG *absorbed* hydrogels, d) 0.2-AMGO *absorbed* hydrogels, e) 0.2-AMG *in situ* hydrogels, f) 0.2-AMGO *in situ* hydrogels and g) blank hydrogels.

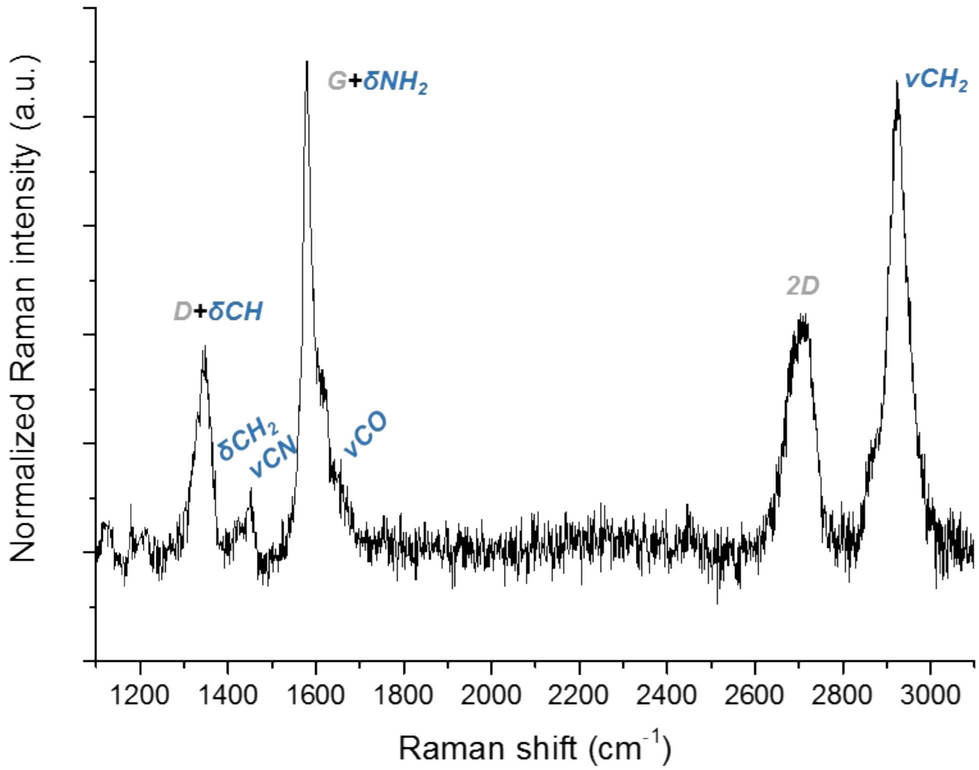


Fig. S11. Representative Raman spectrum of 0.2-AMG, in which both the Raman bands of both the neat hydrogel matrix (~1300 cm^{-1}, δCH; ~1400 cm^{-1} δCH_2; ~1430 cm^{-1} νCN; ~1600 cm^{-1} δNH_2; ~1660 cm^{-1} νCO and ~2900 cm^{-1} νCH_2) and the ones of graphene (~1340 cm^{-1}, D-band; ~1590 cm^{-1}, G-band; ~2700 cm^{-1}, 2D-band), are observed.

Table S1. Comparison of available literature approaches considering, the nanomaterial : monomer ratio, properties and advantages of the final hydrogels.

Nanomaterial	Monomer	Nanomaterial : monomer ratio	Approach	Advantages	Reference
Functionalized graphene flakes	Acrylic acid	~ 0.05	Radical polymerization	Enhancement of compressive strength and elastic modulus.	Polym., 2013, 54, 3921–3930.
Few layer graphene	Methacrylic acid	from 0.0002 to 0.0008	Radical polymerization	Improvement of the mechanical, electrical, and thermal properties.	Healthc. Mater., 2014, 3, 1334–1343.
PAM-stabilized graphene	Acrylamide	~ 5	Physically cross-linked hydrogels.	Higher electrical conductivity and thermal stability.	ACS Appl. Mater. Interfaces, 2013, 5, 8633–8640.
Few layer graphene	Acrylamide	from 0.001 to 0.01	Radical polymerization	Support of the growth of cultured brain cells.	Sci. Rep.,2017, 7, 10942.
Graphene oxide	*N*-isopropyl acrylamide	~1.76	Radical polymerization	Stronger hydrogels.	Carbon 2013, 62, 117 – 126.
Graphene oxide	Vinyl alcohol	~0.5	Physically cross-linked hydrogels.	pH-sensitive hybrid for drug delivery.	Chem. Commun. 2010, 46, 2376–2378.
Graphene quantum dots	9-anthracene methyloxy carbonyl amino acid	~0.125	Physically cross-linked hydrogels.	GQDs tuned the mechanical and self-healing properties of the gels.	RSC Adv., 2016, 6, 54793–54800.