Supporting Information

Polymer Grafted Graphitic Carbon Nitride as Precursors for Reinforced Lubricant Hydrogels

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Experimental:

Preparation of g-CN:[1] 2.0 g of cyanuric acid (C) and 2.0 g of melamine (M) were mixed with 40 mL distilled water and shaked overnight. The mixture was centrifuged at 6000 rpm for 10 minutes and the precipitate dried at 60 °C under vacuum overnight. The dried product was transferred into a capped crucible and put into N_2 protected oven at 550 °C for 4 hours, with a heating rate of 2.3 °C /min^{-1}. The product is obtained as yellow solid. g-CN must be well grinded prior to use.

Synthesis of Tough DMA Hydrogels (touB): 1 g g-CN pre3h was added to 1 g water, 1 g DMA, 100 mg MBA and mixed for 10 minutes. Subsequently, the mixture was flushed with

nitrogen for 3 minutes and the mixture was put between two 50 W LED daylight sources (20 cm apart from each other) to initiate gelation which takes place in 1 hour. EG (14 wt.%) can be removed via washing with water as described in the manuscript.

Synthesis of Tough DMA Hydrogels (touC): 1 g g-CN pre3h was added to with 2 g water, 1 g DMA, 100 mg MBA and mixed for 10 minutes. Subsequently, the mixture was flushed with nitrogen for 3 minutes and the mixture was put between two 50 W LED daylight sources (20 cm apart from each other) to initiate gelation which takes place in 1 hour. EG (10 wt.%) can be removed via washing with water as described in the manuscript.

Reference DMA Hydrogel Without g-CN (touRef): Mixture composed of 450 mg EG, 450 mg water and 80 mg DMA was used to replicate the g-CN based prepolymer. The EG/water/DMAsolution was added to with 1 g water, 2 g DMA, 100 mg MBA, 20 mg ascorbic acid and mixedfor 10 minutes. 0.2 mL H_2O_2 solution was added to the mixture and gelation took place in 10 minutes. EG (10 wt.%) can be removed via washing with water as described in manuscript.

Synthesis of Reference DMA Hydrogel For Lubricant Hydrogels (lubB): 1 g g-CN pre3h was added to 2 g water, 1.8 g DMA, 75 mg MBA and mixed for 10 minutes. Subsequently, the mixture was flushed with nitrogen for 3 minutes and the mixture was put between two 50 W LED daylight sources (20 cm apart from each other) to initiate gelation which takes place in 1 hour. EG can be removed via washing with water as described in the manuscript.

Synthesis of Reference SPMA Hydrogel For Lubricant Hydrogels (lubC): 1 g g-CN pre3h was added to 2 g water, 1.2 g SPMA, 75 mg MBA and mixed well for 10 minutes. Subsequently, the mixture was flushed with nitrogen for 3 minutes and the mixture was put between two 50 W LED daylight sources (20 cm apart from each other) to initiate gelation which takes place in 1 hour. EG can be removed via washing with water as described in the manuscript.

Reference Lubricant Hydrogels Without g-CN (lubRef): Mixture composed of 450 mg EG, 450 mg water and 80 mg DMA was used to replicate the content of g-CN based prepolymer. The EG/water/DMA solution was added to 2 g water, 1 g DMA, 0.8 g SPMA, 75 mg MBA, 20 mg ascorbic acid and mixed for 10 minutes. 0.2 mL H_2O_2 solution was added to initiate gelation

which takes place in 1 hour. EG can be removed via washing with water as described in the manuscript.

Figure S1. Digital image of MCR Tribometer set-up for tribological measurement, 3 cylindrical hydrogel samples were clamped to a T-PID/44 holder (top) and slided over a hydrogel sample prepared in a petri dish (below).

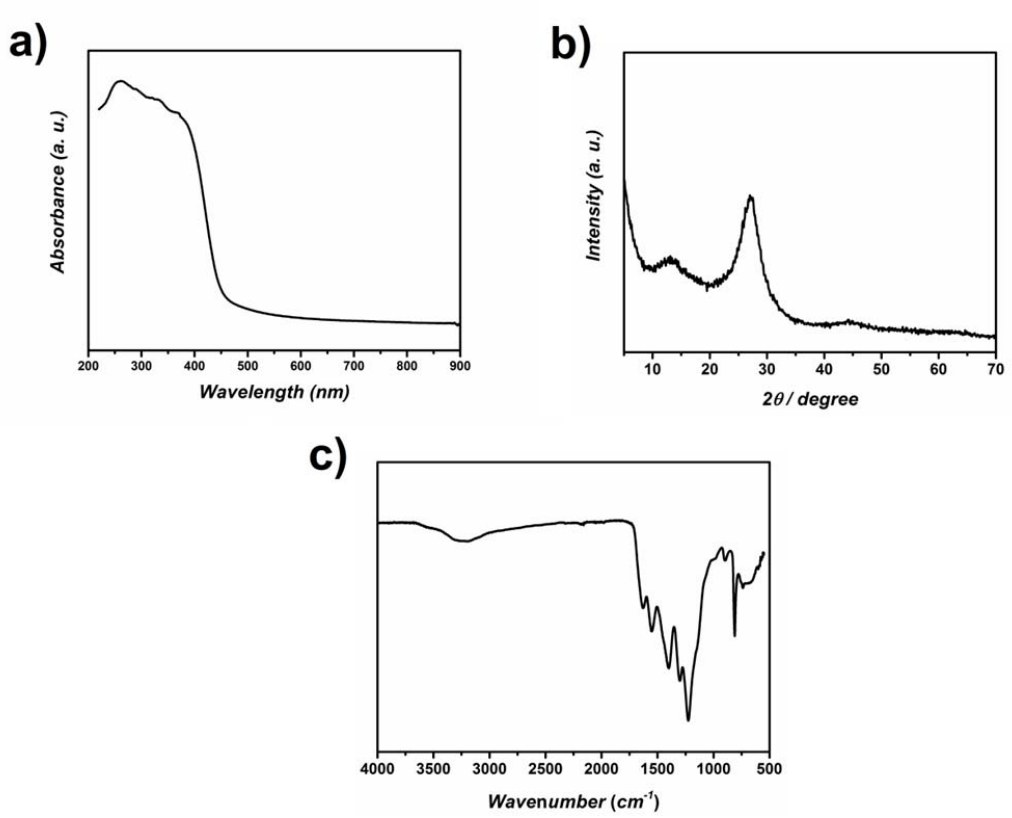

Figure S2. a) UV-Vis spectrum, b) powder XRD profile and c) FT-IR results of synthesized g-CN.

Table S1. Surface zeta potential and C:N ratio of synthesized g-CN.

	Surface zeta potential (mV)	**C:N ratio**
g-CN	-30.6	0.6110

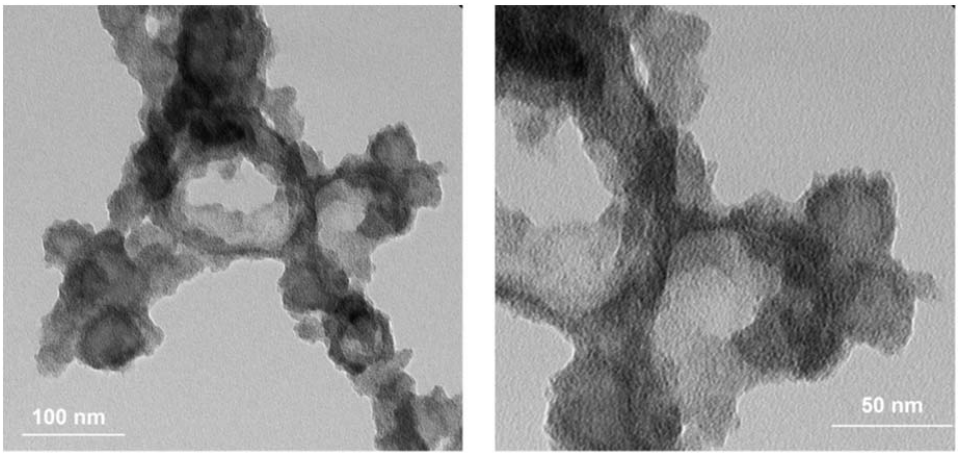

Figure S3. TEM images of g-CN pre3h.

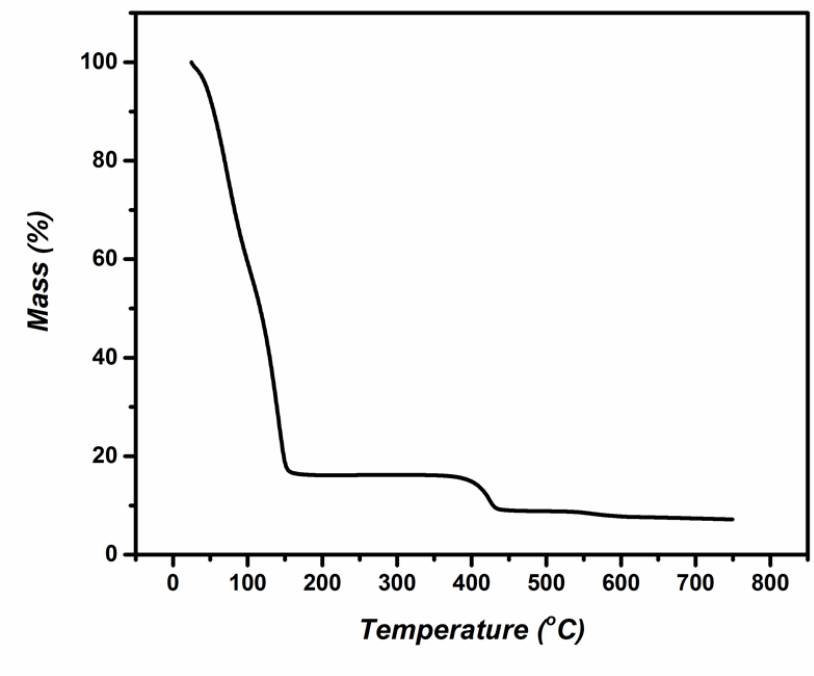

Figure S4. TGA diagram of g-CN pre3h.

Table S2. Surface zeta potential and C:N ratio of isolated particles from g-CN pre3h.

	Surface zeta potential (mV)	**C:N ratio**
g-CN pre3h (isolated powder)	-33.2	1.748

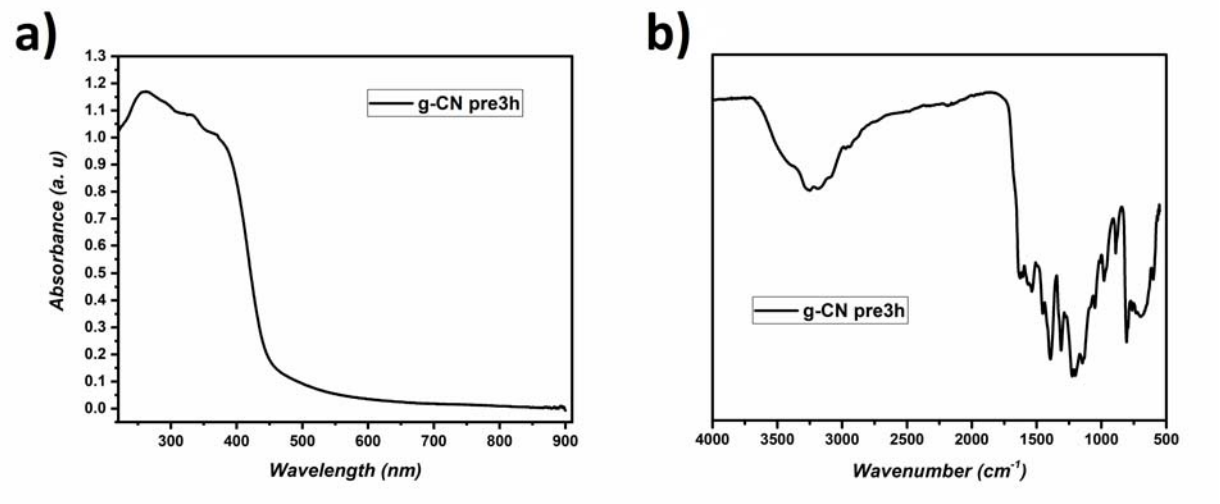

Figure S5. a) Solid state UV-Vis and b) FT-IR spectra of isolated powders from g-CN pre3h precursor.

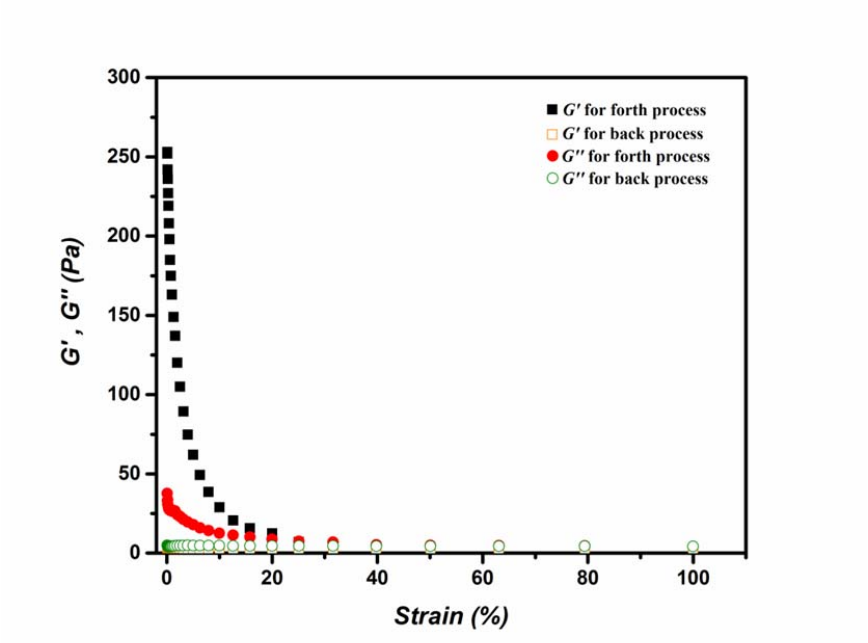

Figure S6. Rheology result of g-CN pre1h, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

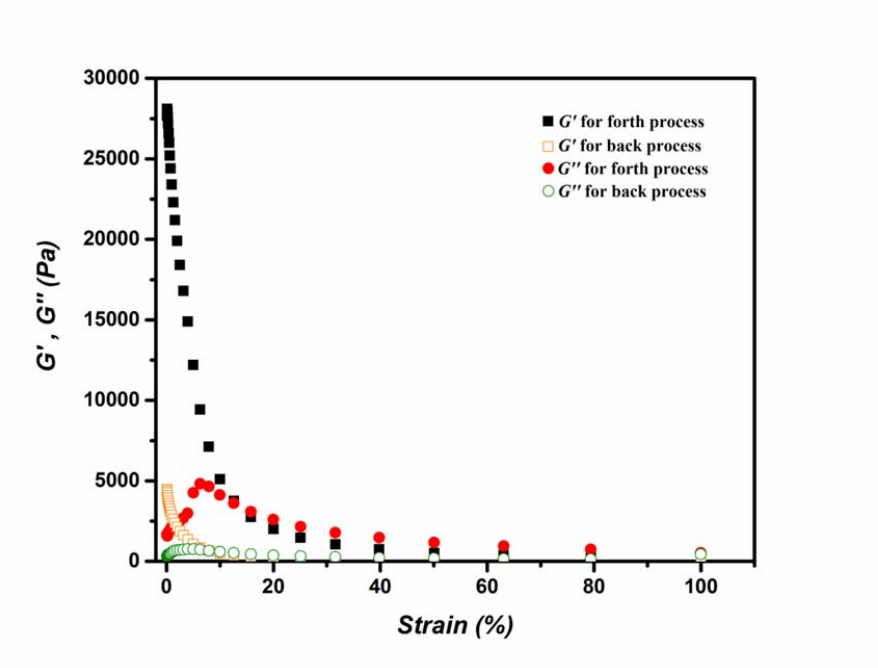

Figure S7. Rheology result of g-CN pre6h, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

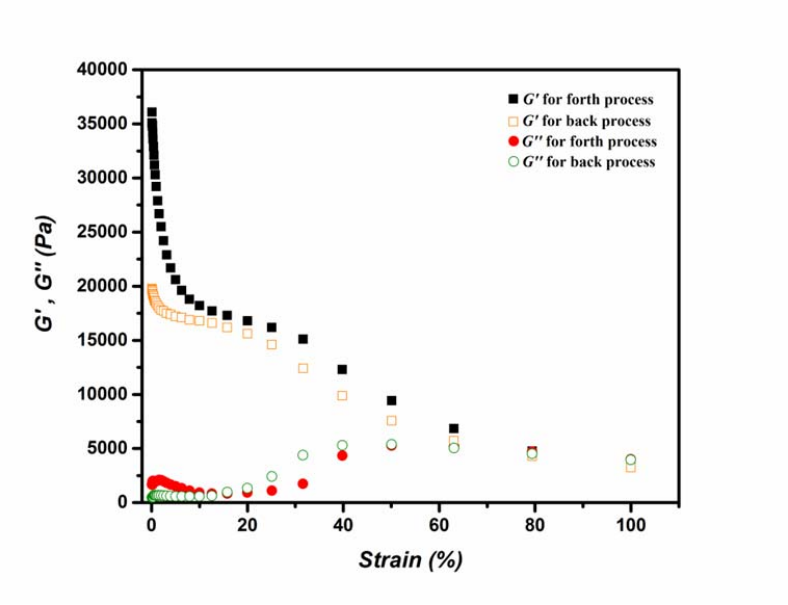

Figure S8. Rheology result of g-CN pre12h, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

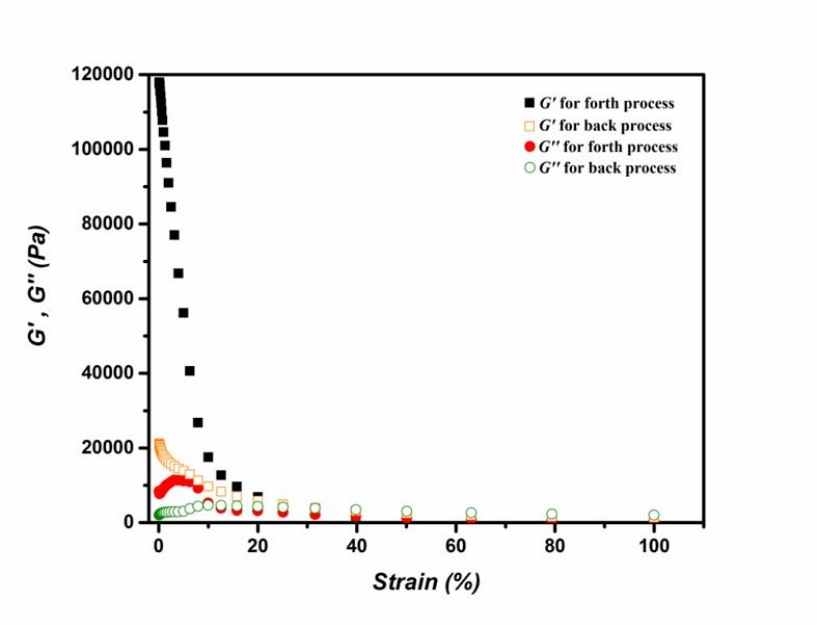

Figure S9. Rheology result of g-CN 2pre3h, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

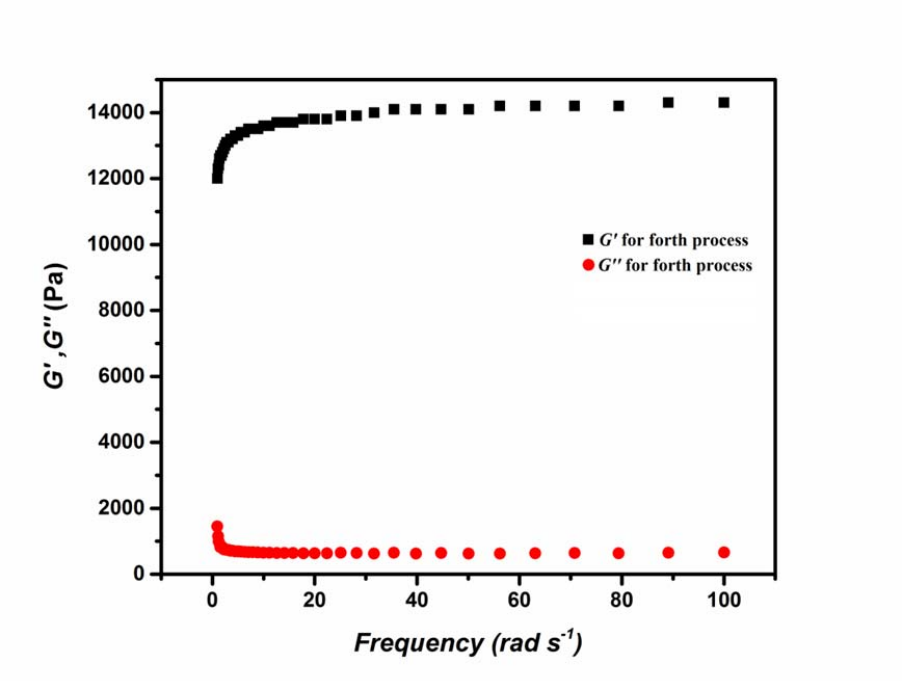

Figure S10. Rheology result of g-CN pre3h, storage (*G*', black squares) and loss modulus (*G*'', red circles) against frequency.

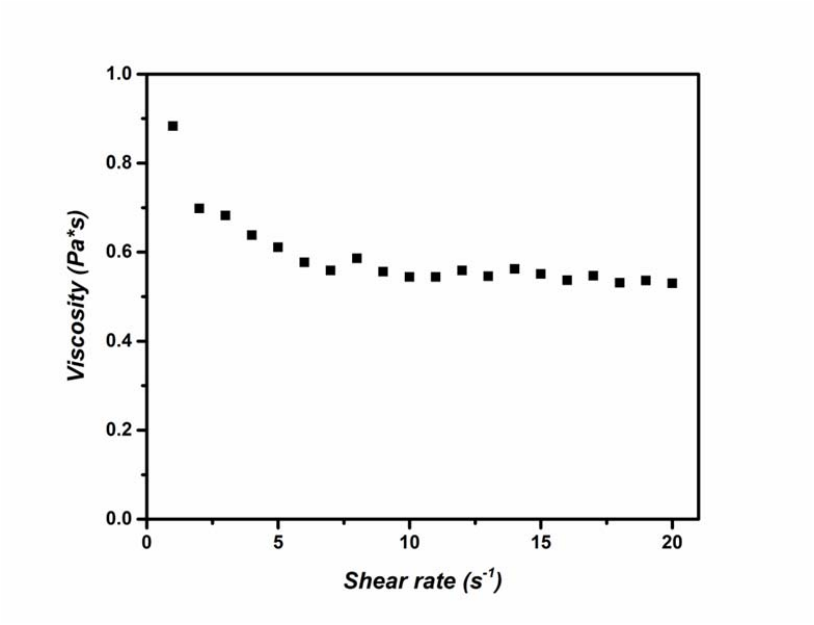

Figure S11. Viscosity profile of g-CN pre1h against shear rate between 1-20 s^{-1}.

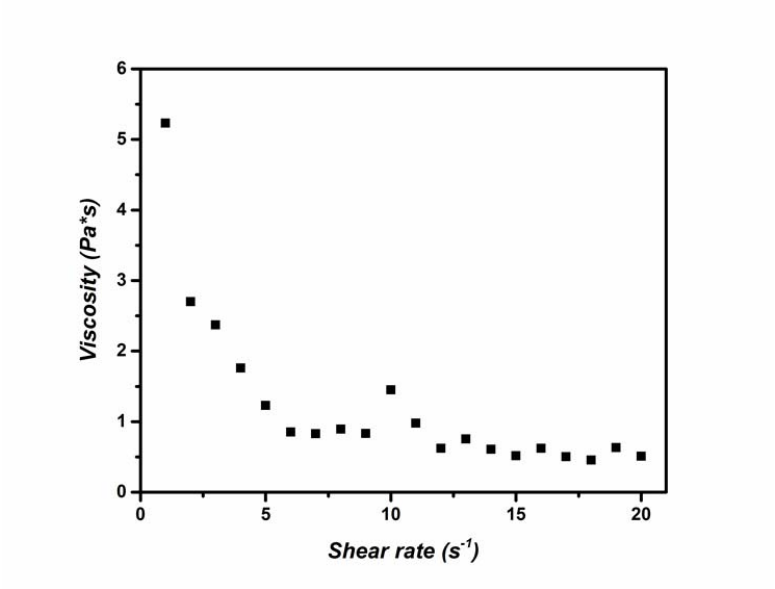

Figure S12. Viscosity profile of g-CN pre3h against shear rate between 1-20 s^{-1}.

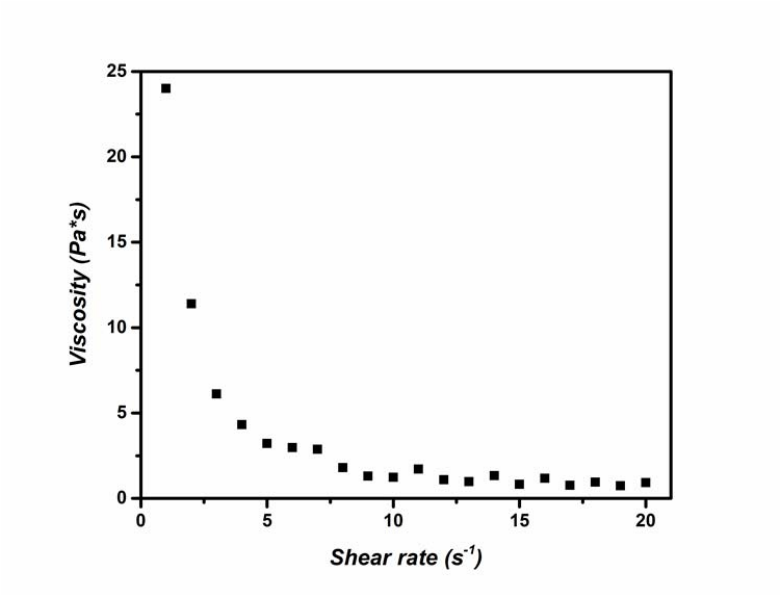

Figure S13. Viscosity profile of g-CN pre6h against shear rate between 1-20 s^{-1}.

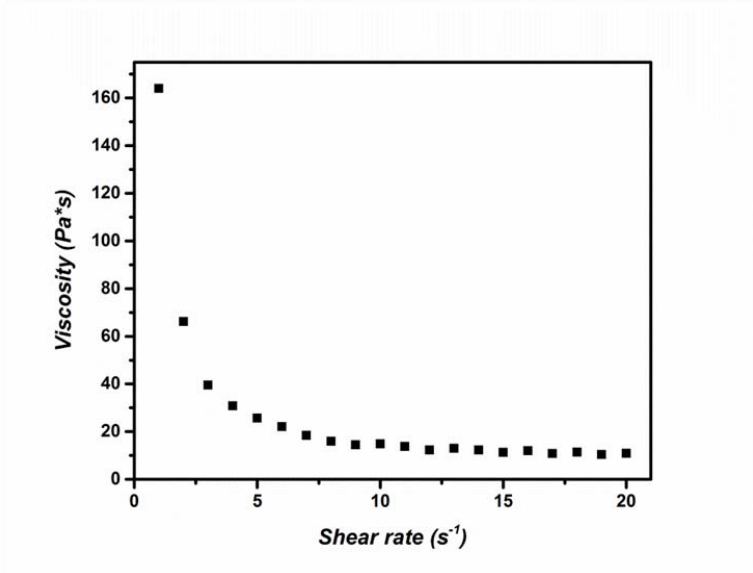

Figure S14. Viscosity profile of g-CN pre12h against shear rate between 1-20 s^{-1}.

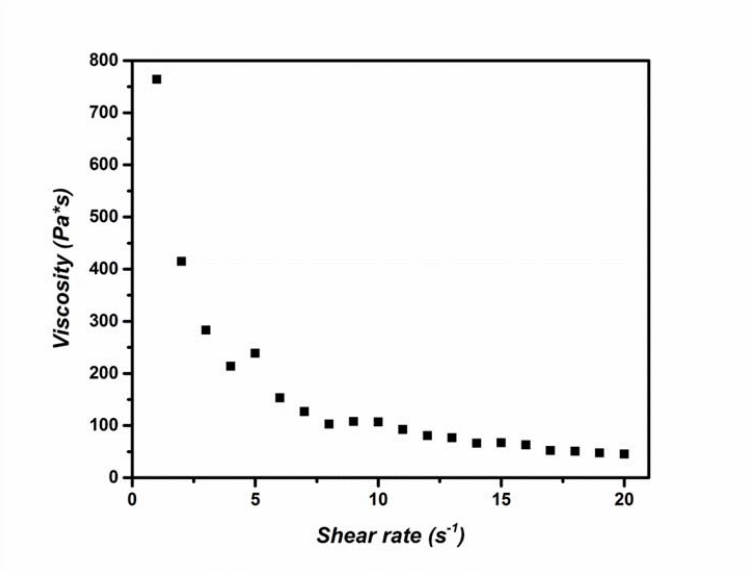

Figure S15. Viscosity profile of g-CN 2pre3h against shear rate between 1-20 s^{-1}.

Figure S16. g-CN pre3h after standing still for 2 months.

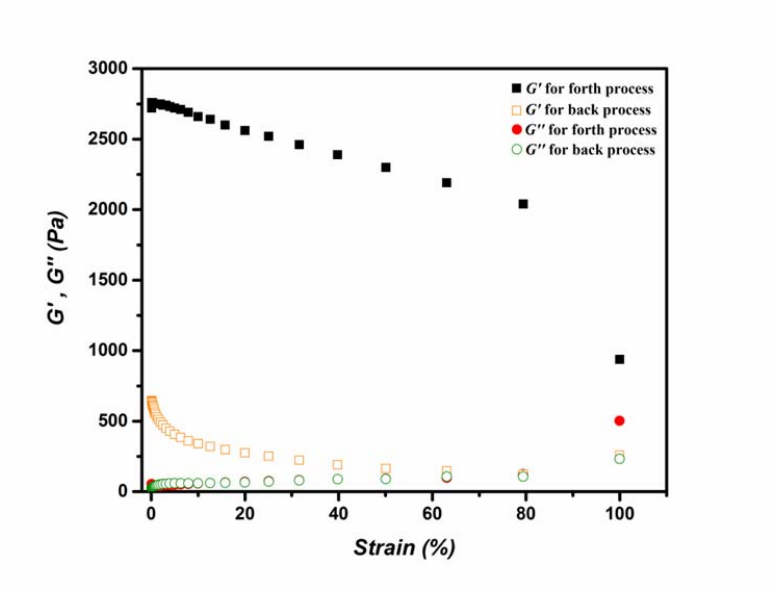

Figure S17. Rheology result of g-CN pre3h:water in 1:0.25 ratio, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

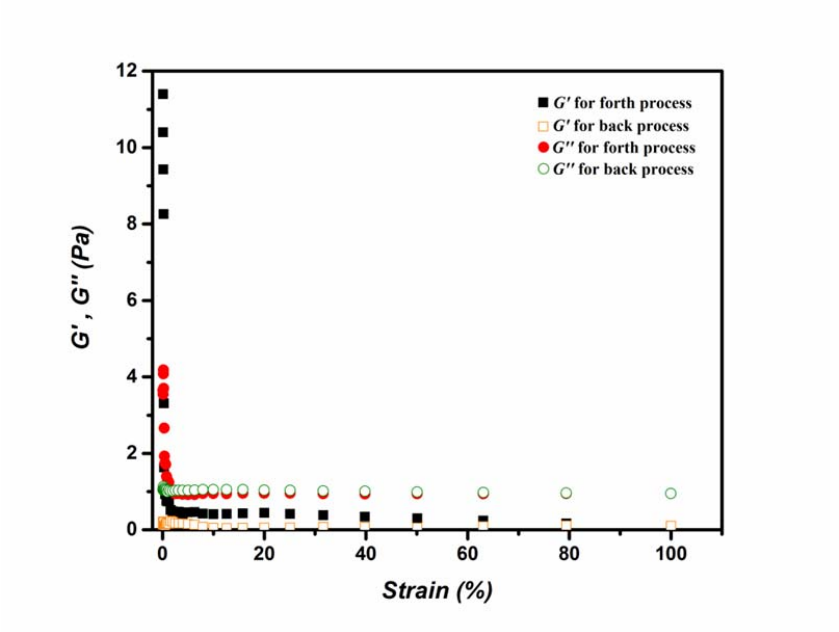

Figure S18. Rheology result of g-CN pre3h:water in 1:0.5 ratio, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

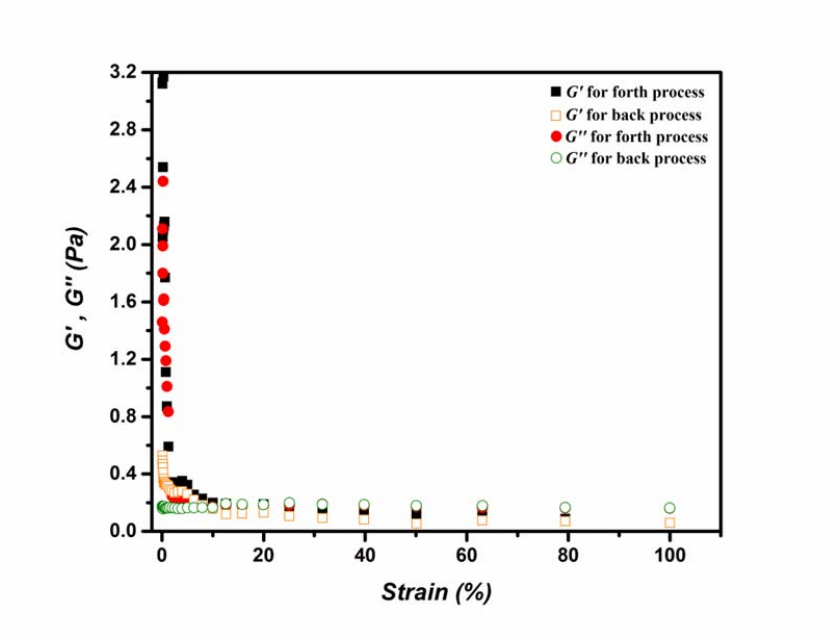

Figure S19. Rheology result of g-CN pre3h:water in 1:0.75 ratio, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

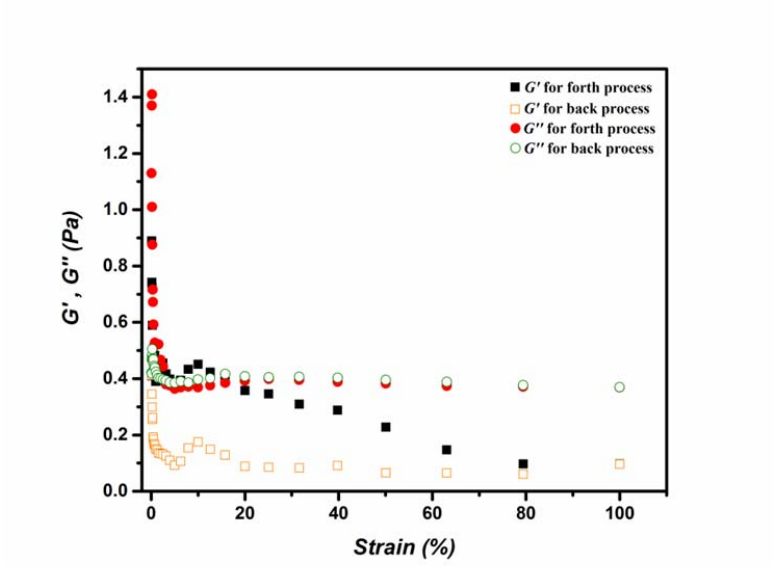

Figure S20. Rheology result of g-CN pre3h:water in 1:1 ratio, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

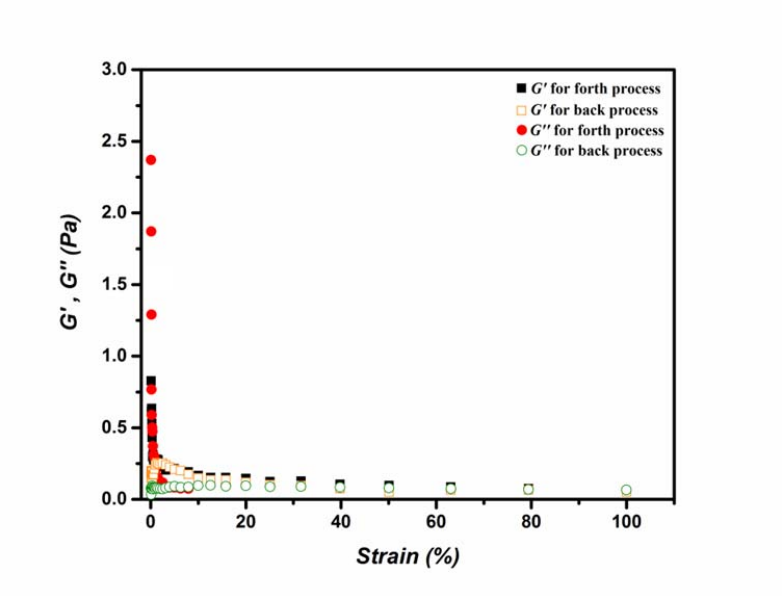

Figure S21. Rheology result of g-CN pre3h:water in 1:1.5 ratio, storage (*G*', black and orange squares) and loss modulus (*G*'', red and green circles) against strain, back (open) and forth (filled) process.

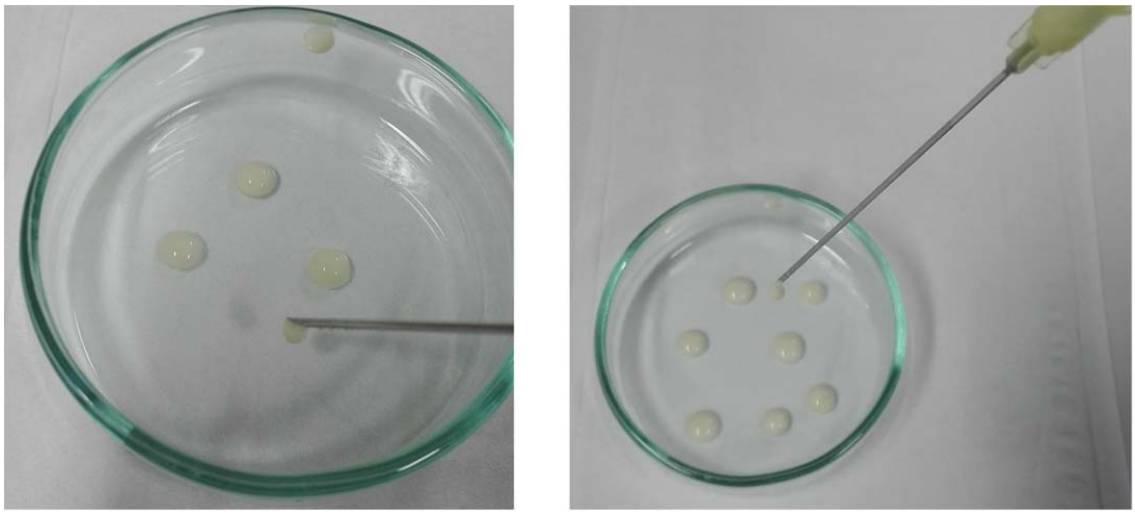

Figure S22. Image showing injection of g-CN pre3h through needle.

Table S3. Amounts of precursor, water, monomer and crosslinker for exemplary organohydrogel synthesis.

Sample	Precursor (g-CN pre3h) amount (wt.%)	Water amount (wt.%)	DMA amount (wt.%)	MBA amount (wt.%)
touA	25	25	47.5	2.5
touB	32.5	32.5	32	3
touC	22	38	38	2
touRef	25[a]	25	47.5	2.5

[a] Reference solution (450 mg EG, 450 mg water and 80 mg DMA) was utilized

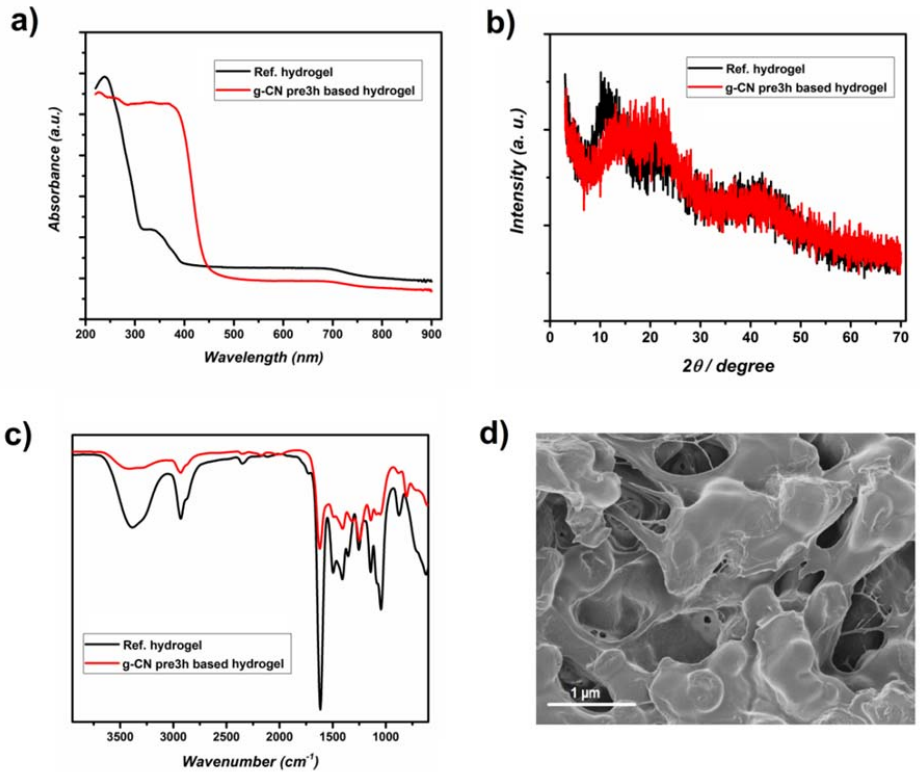

Figure S23. Solid state analysis of freeze dried reference and g-CN pre3h based tough hydrogel (touA) a) solid state UV spectra, b) powder XRD, c) FT-IR and d) SEM image of g-CN pre3h based tough hydrogel.

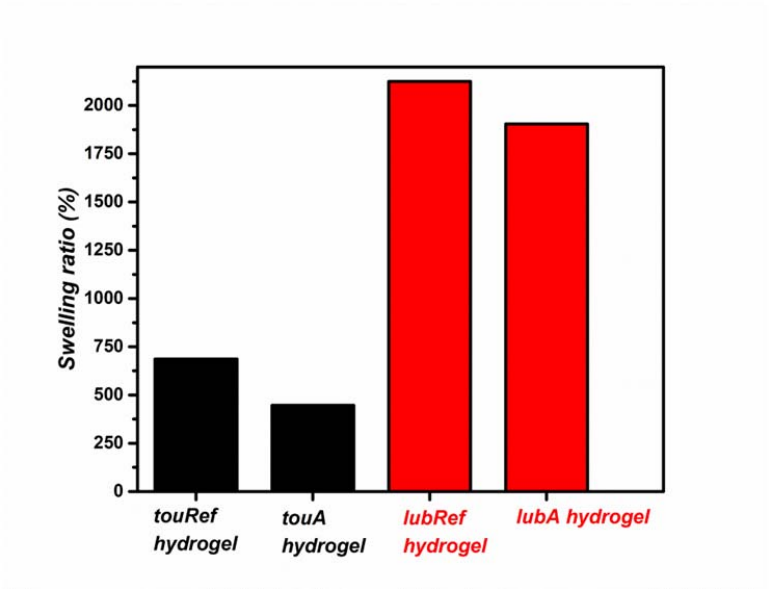

Figure S24. Swelling ratios of reference and g-CN pre3h based tough and lubricant hydrogels.

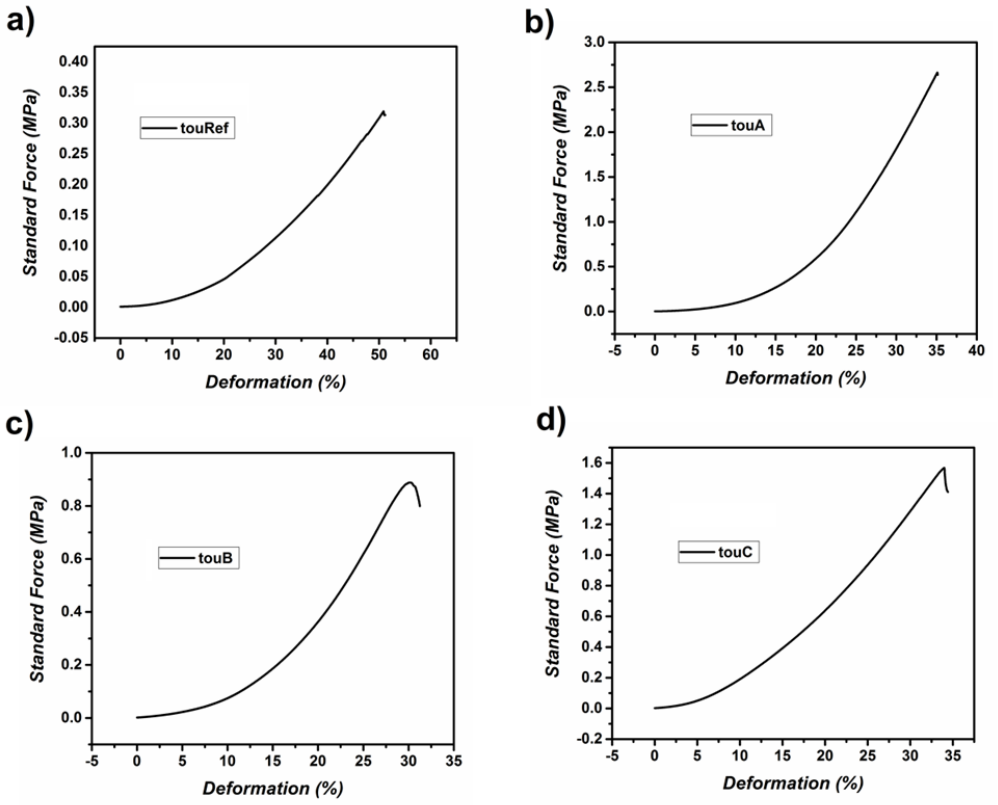

Figure S25. Compression profiles of a) touRef, b) touA, c) touB and d) touC organohydrogels.

Table S4. Elastic moduli values of tough organohydrogels and hydrogels.

Specimen	E_{mod} (-10% before break, MPa)	Compression (-10% before break, %)	E_{mod} at break (MPa)	Compression at break (%)	Maximum stress (MPa)
tou Ref	1.17	34	1.51	44	0.36
touA	11.9	24	13.74	34	2.67
touB	3.04	20	4.29	30	0.79
touC	6.01	22	7.37	32	1.51
tou Ref hydrogel	0.94	42	0.72	52	0.14
touA hydrogel	9.94	21	14.53	31	2.36
touB hydrogel	4.40	14	7.75	24	1.07
touC hydrogel	0.51	17	0.59	27	0.12

Table S5. Compositions of lubricant and blank hydrogels.

Sample	Precursor (g-CN pre3h) amount (wt.%)	Water amount (wt.%)	DMA amount (wt.%)	MBA amount (wt.%)	SPMA amount (wt.%)
lubA	20	40	20	1.5	18.5
lubB	20	40	38.5	1.5	-
lubC	24	48	-	1.8	26.2[b]
lubRef	20[a]	40	20	1.5	18.5

[a] Control solution used to synthesize reference organohydrogel, [b] limited to 1.2 g instead of 1.8 g due to solubility.

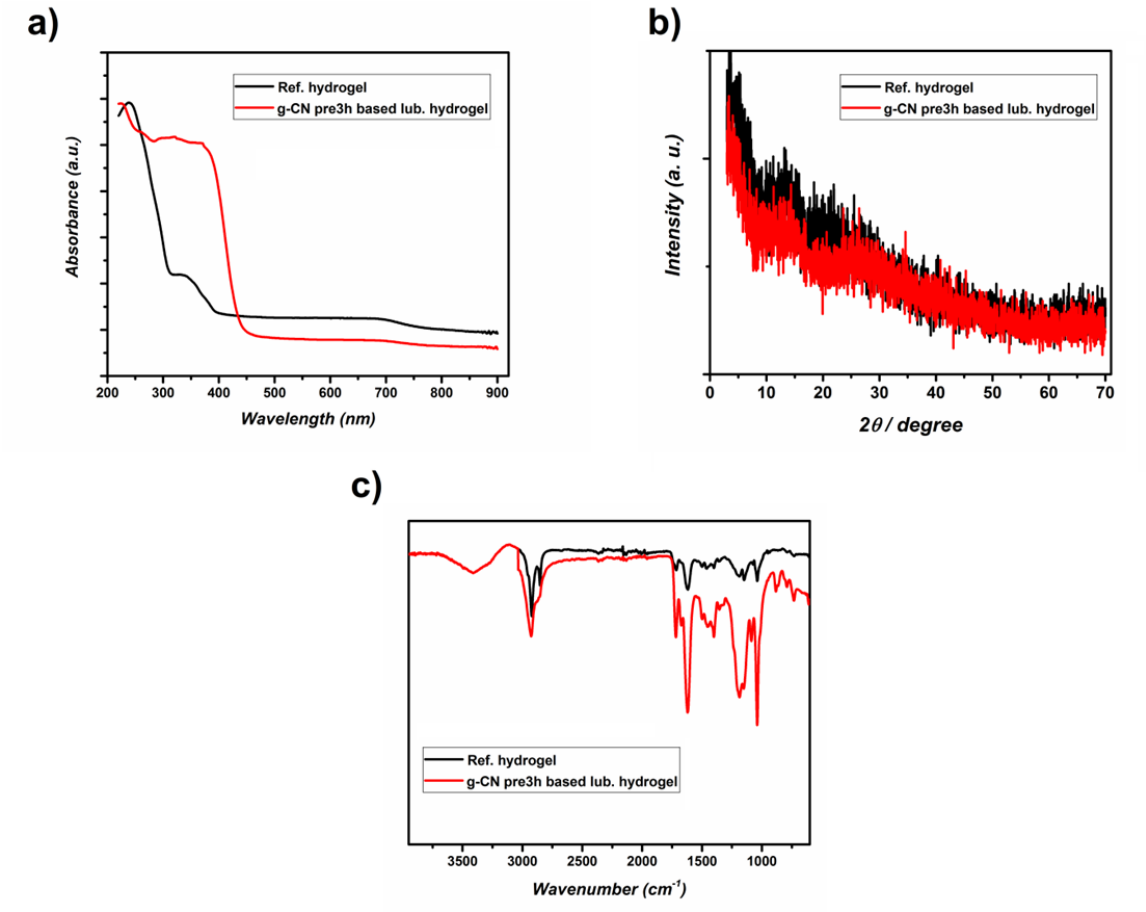

Figure S26. Solid state analysis of freeze dried reference and g-CN pre3h based lubricant hydrogel (lubA) a) solid state UV spectra, b) powder XRD and c) FT-IR spectra.

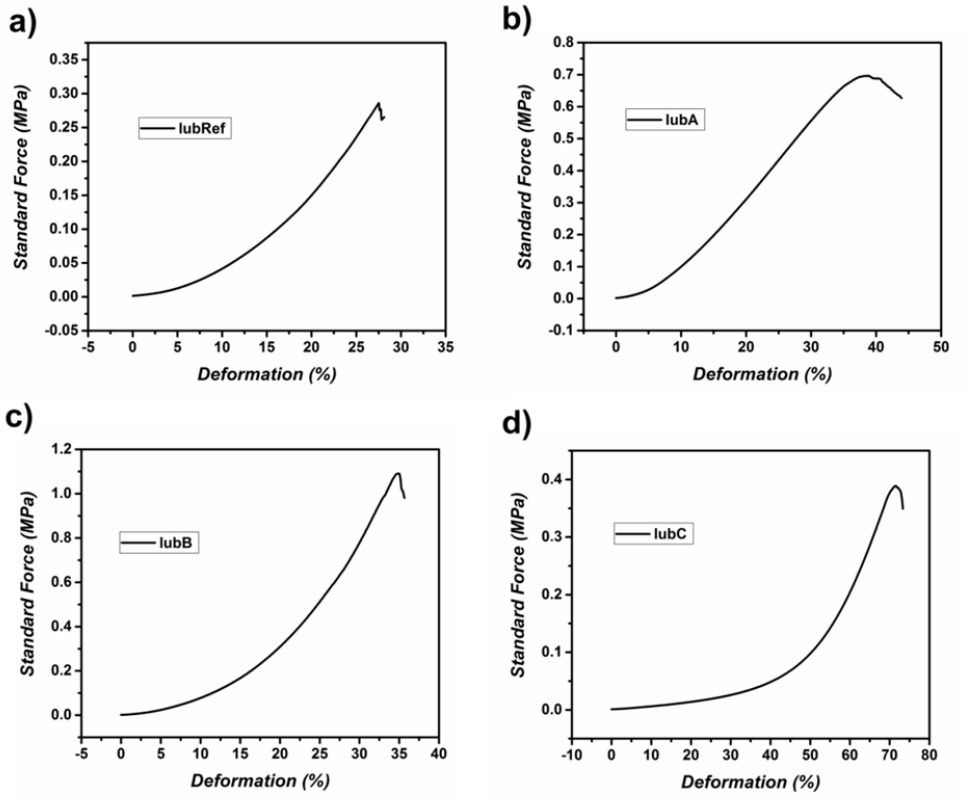

Figure S27. Compression profiles of a) lubRef, b) lubA, c) lubB and d) lubC organohydrogels.

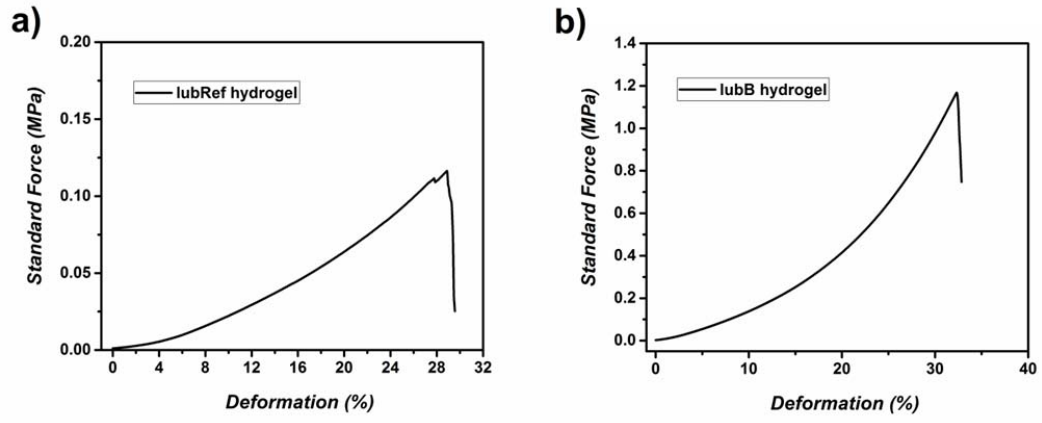

Figure S28. Compression profiles of a) lubRef and b) lubB hydrogels.

Table S6. Elastic moduli values of lubricant organohydrogels and hydrogels.

Specimen	E_{mod} (-10% before break, MPa)	Elongation (-10% before break, %)	E_{mod} at break (MPa)	Elongation at break (%)	Maximum stress (MPa)
lub Ref	0.66	18	1.83	28	0.29
lubA	2.52	23	2.47	33	0.79
lubB	3.10	22	5.49	32	0.96
lubC	1.27	57	1.69	67	0.38
lub Ref hydrogel	1.01	19	1.68	29	0.30
lubA hydrogel	1.51	3	7.96	13	0.47
lubB hydrogel	4.75	18	27.75	28	1.28

Video S1. The video shows two hydrogel monoliths on an inclined plane. After a film of water is added one hydrogel does not move (lubB), while the other one (lubA) slides down.

Video S2. The video shows two hydrogel monoliths on an inclined plane. One hydrogel sticks to the plane (lubB), while the other one (lubA) slides down.

References

1. Shalom, M.; Inal, S.; Fettkenhauer, C.; Neher, D.; Antonietti, M., Improving carbon nitride photocatalysis by supramolecular preorganization of monomers. *J. Am. Chem. Soc.* **2013,** *135* (19), 7118-21.