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Supporting Information

for *Adv. Mater. Interfaces,* DOI: 10.1002/admi.202000509

Repulsion of Polar Gels From Water: Hydration-Triggered Actuation, Self-Folding, and 3D Fabrication

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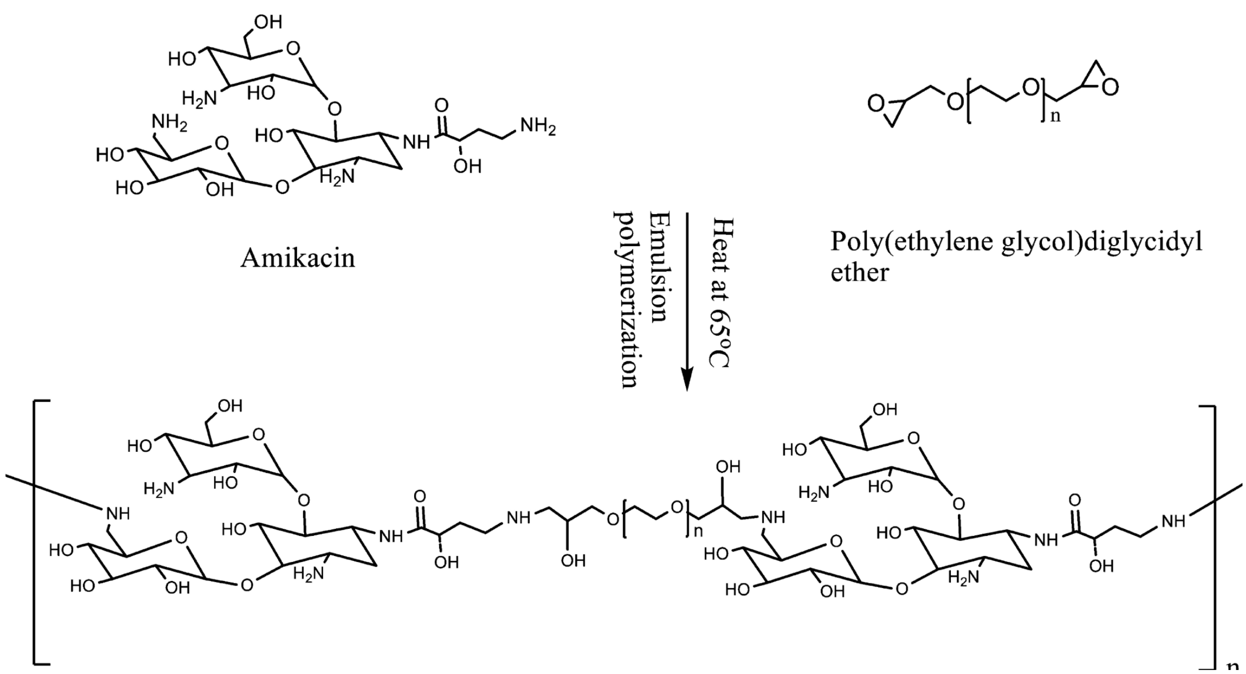

Protonation of amines within Amikagels under acidic conditions

$$[R - NH_2]_{gel} + [H^+]_{aq} \rightarrow [R - NH_3{}^+]_{gel}$$

Deprotonation of amines within Amikagels under highly basic conditions

$$[R - NH_3{}^+]_{gel} + [OH^-]_{aq} \rightarrow [R - NH_2]_{gel} + H_2O$$

Figure S1. *Reaction between amines in amikacin, an aminoglycoside antibiotic, with epoxide groups present in poly(ethylene glycol) diglycidyl ether (PEGDE). Figure adapted from [1] with permission. Protonation and deprotonation equations for amines present in Amikagels are also shown.*

Actuation of Amikagels on Different Solvents

To study the actuation of Amikagels in presence of different solvents, AM-PEGDE films were placed on acetone, which triggered the actuation behavior of the gels, leading to a 3D confirmation (**Figure S1**). Acetone is a polar solvent and could trigger the actuation of Amikagels. However, the actuation on acetone was slower than that on water. Placing the AM-PEGDE films on hexane, a non-polar aprotic solvent, did not result in actuation (**Figure S2**). The lack of electrostatic repulsion/attraction between hexane and AM-PEGDE films is responsible for the lack of actuation on this solvent.

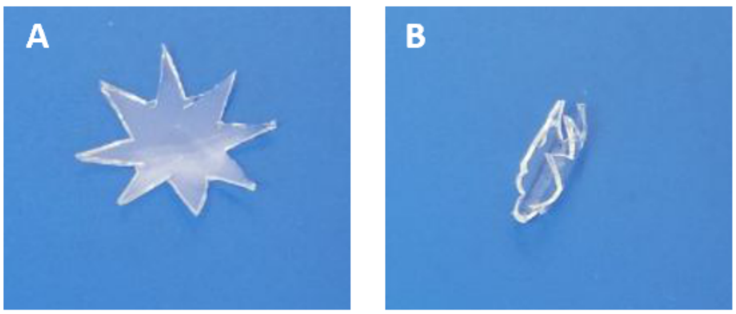

Figure S2. *AM-PEGDE on acetone:* ***A.*** *t=0 (before placing on acetone) and* ***B.*** *3 minute after placing on acetone.*

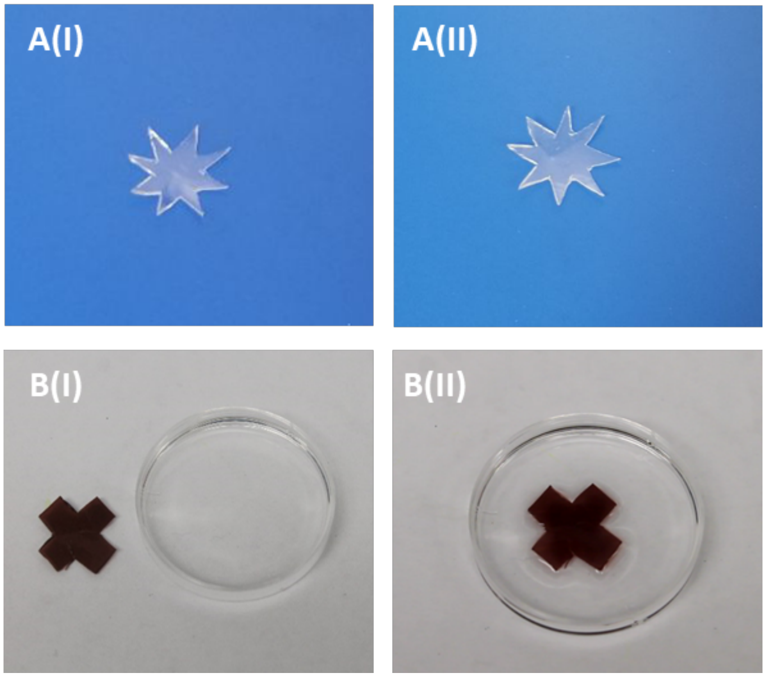

Figure S3. *(**A**). AM-PEGDE on hexane.* ***(I).*** *time=0 (before placing on hexane) and* ***(II).*** *5 minute after placing on hexane. (**B**). AM-PEGDE-GNR nanocomposites* ***(I).*** *time t=0 (before placing on the hexane) and* ***(II).*** *3 minutes after placing on hexane.*

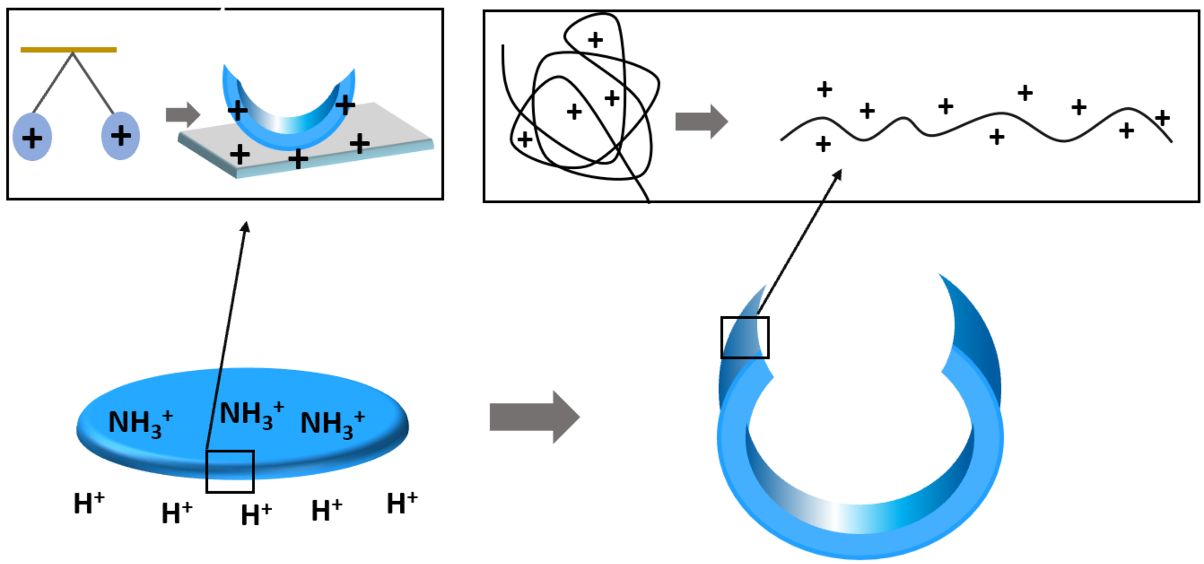

Figure S4. *Hydration triggered self-folding and actuation behavior of Amikagels upon placing on polar solvents. Protonation of amines induces repulsion thus minimizing contact area, which in turn, triggers the actuation of the gel away from aqueous / polar solvents.*

Mechanical Characterization

Tensile strengths of the different Amikagel films were determined using a TA.plus Texture Analyzer at a constant strain rate 10^{-3} s^{-1} at room temperature. The thickness and diameter of the films were measured with calipers. The maximum force (N) reached in a tension test was divided by the cross-section area (m^2) in order to obtain the ultimate tensile strength (UTS) in Pascals (Pa).

$$UTS\ (Pa) = \frac{Maximum\ \ Force\ \ (N)}{Area\ \ (m^2)}$$

The area underneath the stress–strain (σ–ε) curve up to fracture was calculated and considered as modulus of toughness[2]. Three independent specimens were analyzed for each condition and ultimate tensile strength and modulus of toughness values were calculated as the average value of these three measurements. The ultimate tensile strength (UTS) for AM-PEGDE was ~3 times higher than that of AM-BDDE. BDDE-based films show less flexibility and toughness compared to PEGDE-based films likely because of the shorter chain of the former. The average distance between two neighboring cross-links or the network mesh size is lower in AM-BDDE compared to AM-PEGDE. Incorporating GNRs within Amikagel films increases the UTS and toughness of the resulting nanocomposite films, which is along expected lines.

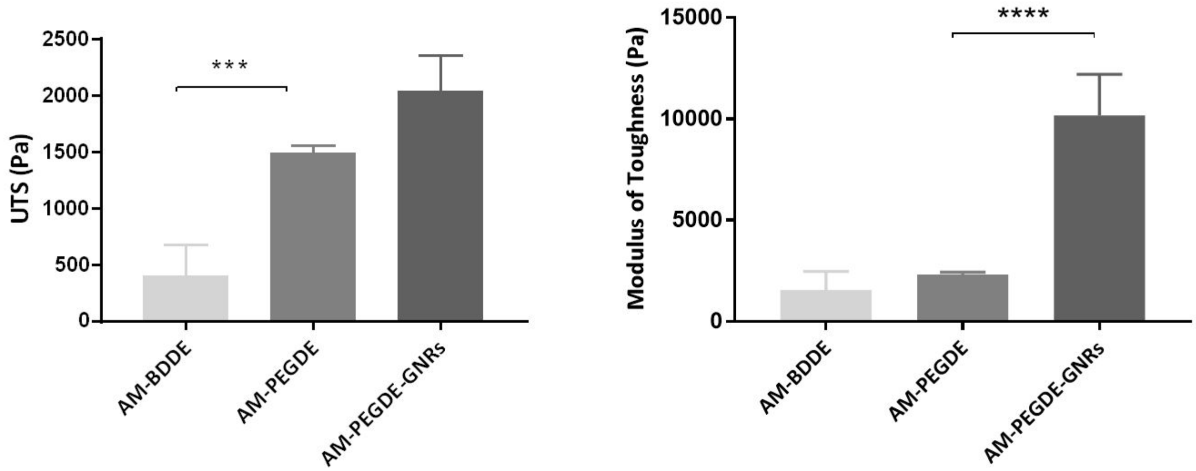

Figure S5. (A). *Ultimate tensile strength (UTS) and* ***(B).*** *modulus of toughness for AM-BDDE, AM-PEGDE and AM-PEGDE-GNR (1 wt%) films. Statistical analysis was determine using one-way ANOVA and Tukey test (n=3 for each condition, *p<0.05, **p<0.01, ***p<0.001, ****p<0.0001).*

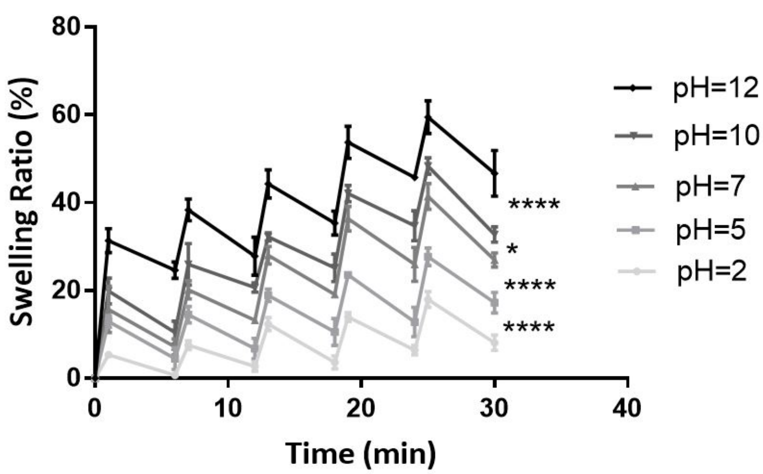

Figure S6. *Swelling / deswelling ratios for AM-PEGDE Amikagels. Statistical analysis was carried out using one-way ANOVA and Tukey test (n=3 for each condition, *p<0.05, **p<0.01, ***p<0.001, ****p<0.0001).*

REFERENCES

1. Grandhi, T. S.; Mallik, A.; Lin, N.; Miryala, B.; Potta, T.; Tian, Y.; Rege, K., Aminoglycoside antibiotic-derived anion-exchange microbeads for plasmid DNA binding and in situ DNA capture. *ACS Appl Mater Interfaces* **2014,** *6* (21), 18577-89.
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