Supporting Information

Three-Dimensional Graphene Structure for Healable Flexible Electronics Based on Diels-Alder Chemistry

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1. The synthesis of furfuryl-contained linear polyurethane (FLPU) and DAPU

10 g of DGEBA was mixed and heated with 5.7 g of furfurylamine (in the stoichiometric ratio 1:2 molar) and reacted in 20 mL methanol at 60 °C for 6 h. At the end of the reaction, the product (the component with bi-furan and bi-hydroxyl, 2F2OH) was obtained by drying the sample under vacuum until constant weigh was reached.

In order to prepare FLPU, 1.0 g of MDI was dispersed in 10.0 g anhydrous DMF and stirred at room temperature for 30 min, at first. Then 4.0 g PTMEG2000 in 20.0 g anhydrous DMF was dropped into the above solution and reacted at 80 °C for 2 h in N₂ atmosphere. Afterwards, 1.07 g of 2F2OH dispersed in 5 g anhydrous DMF was added and the resultant solution was reacted at 80 °C for another 2 h in N₂ atmosphere which resulted in the solution of furfuryl-contained linear PU (FLPU).

And the covalently cross-linked DAPU was prepared by adding 0.754 g BMI into FLPU solution and reacted at 65 $^{\circ}$ C for 5 h.

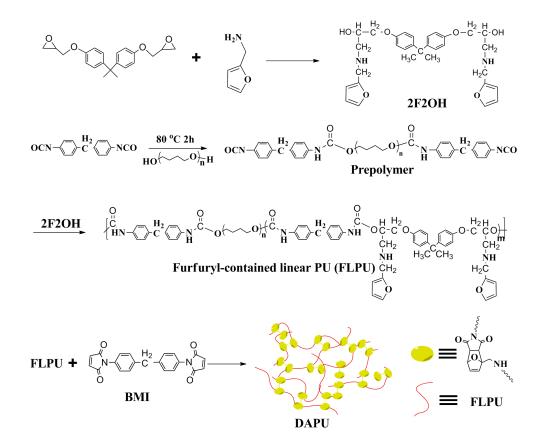


Figure S1. The synthetic procedures of FLPU and DAPU.

2. The optical images of GO, FAGS and FAGS/DAPU

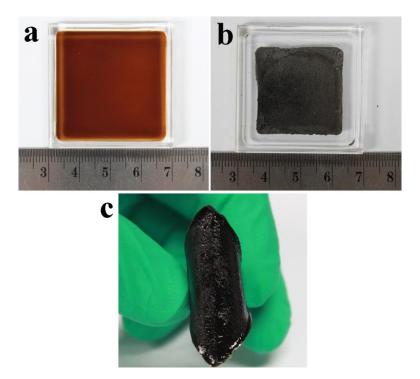


Figure S2. The optical images of GO solutions (a), FAGS (b) and FAGS/DAPU (c).

3. AFM images of the GO sheets.

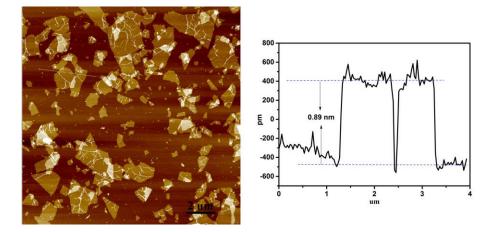
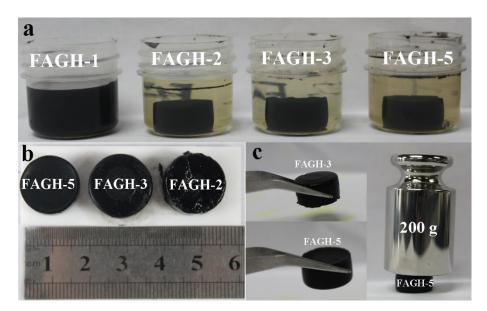


Figure S3. The AFM image of the as-prepared GO sheets.



4. The pictures of graphene hydrogels prepared by furfurylamine

Figure S4. The pictures of graphene hydrogels prepared by furfurylamine (FAGHs). (a) FAGHs after the self-assembly process, (b) photos of FAGH-2, FAGH-3 and FAGH-5 after washing, (c) the mechanical property of FAGH-3 and FAGH-5.

5. The SEM images of FAGS/DAPU'

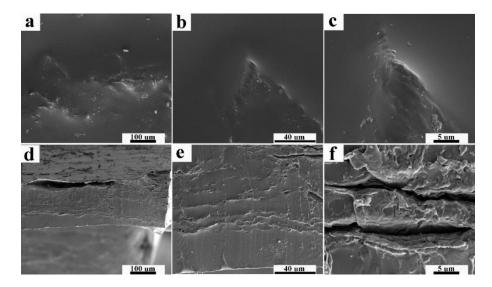


Figure S5. SEM images of FAGS/DAPU': (a-c) top view of FAGS/DAPU' at different magnifications; (d-f) cross-sectional view of FAGS/DAPU' at different magnifications

6. The healing process of FAGS/DAPU with heat

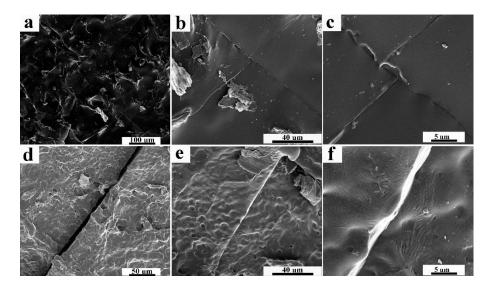


Figure S6. Visual inspection of the healing process of FAGS/DAPU with heat: (a, d) fissures cut by a knife, (b-c and e-f) surface of the sample after healing with heat at different magnifications

7. The healing process of FAGS/DAPU by microwave

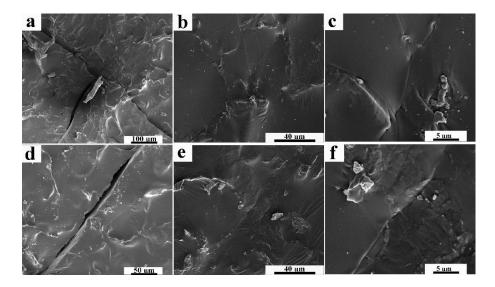


Figure S7. Visual inspection of the healing process of FAGS/DAPU by microwave: (a, d) fissures were cut by a knife, (b-c and e-f) surface of the sample after healing at different magnifications