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Supplementary Information

Healable Green Hydrogen Bonded Networks for Circuit Repair, Wearable Sensor and Flexible Electronic Device

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Experimental Section

1. Materials

Amylopectin (from maize), ethanol, ehylene glycol, glycerol, aceton, chloroform, n-hexane, 1-methyl-3-octylimidazolium chloride, 1-butyl-3-methylimidazolium bromide, sodium chloride (NaCl), potassium chloride (KCl), calcium chloride (CaCl₂), amhydrous ferric chloride (FeCl₃), sodium sulfate (Na₂SO₄), sodium phosphate (Na₃PO₄), copper particles, zinc particles, starch (from wheat, unmodified), gluten were purchased from Sigma-Aldrich. Electroluminescent ZnS:Cu powders were purchased from Shanghai KPT company. Silver nanowires, with diameter of 30 ± 5 nm and length ranged from 100 to 200 µm, were purchased from Nanjing XFNANO materials Tech Co., Ltd. Commercial plain flours were purchased from Smooth-On, Inc. Unless otherwise specified, reagents were used as received without further purification.

2. Fabrication of green structurally dynamic materials (GHBN)

3 g salt and 4 g amylopectin were first mixed in a beaker by a glass rod. Then, they were mixed with 3 g water, and stirred into a homogenous solution. Before heating the beaker under 60°C for 5 min in a water bath, a piece of fresher film was covered on the top of the beaker to prevent the water evaporation. The beaker was moved out of the water bath and naturally cooled to 25°C (room temperature). Finally, a homogenous GHBN in soft matter state could be prepared.

3. Optimized temperature range in the water bath (Fig. S1)

We prepared the samples with the same ratio (Sample 24, 4 g amylopectin, 3 g NaCl and 3 g water, more details can be found in the Table S2,3) but under different temperatures (T) in the water bath process. After being heated for 5 minutes and naturally cooled to 25 °C (room temperature), the resulting products could be classified into three types (Fig. S1): a liquid state (T < 55°C), a homogenous soft matter state (55°C < T < 65°C), and a too sticky state (T > 65°C). Therefore, the optimized temperature range in the water bath was chosen as 60 \pm 5°C.

4. Computational modeling (Fig. 1,3 and Fig. S3,4)

4.1 Molecular dynamics (MD) simulation

In this study, we have employed a molecular dynamics (MD) simulation with use of the Discover module in the commercial software Materials Studio. The canonical ensemble (NVT) with the COMPASS (Condensed-phase Optimized Molecular Potentials for Atomistic Simulation Studies) force field was used to calculate the potential energy of the system considered, in which the parameters derived from ab initio data can be applied to predict the gas-phase and condensed-phase properties of organic and inorganic materials^[1].

The amylopectin clusters were modeled from each individual amylopectin with 3 repeated subunits. The computational model consists of two amylopectin clusters with diameters of 3 nm, consisting of 2000 water molecules or 302 chloroform molecules. The two clusters were placed at a distance of 5 Å, and at least 15 Å distance from the periodic boundaries. The simulation was run at a constant temperature of 298 K (25°C) for 250 ps, with a time step of 1 fs. The healing was simulated by removing the surface solvent molecules, and cutting the aggregated cluster into two equal parts, relocating the two clusters to the same as the foregoing simulation. The calculation of hydrogen bonds was based on the position of the molecules after the motion became stable.

The motion of the molecules became stable within 100 ps, which was used to calculate the diffusivity by measuring the gradient from the plot of the mean squared displacement (MSD) against time. The diffusivity, $D\alpha$, is given by the following equation:

$$D_{a} = \frac{1}{6 \cdot N_{a}} \lim_{t \to \infty} \frac{d}{dt} \sum_{i=1}^{N_{a}} \left\{ r_{i}(t) - r_{i}(0) \right\}^{2}$$

Where N is the number of diffusive atoms, and r is the position vector of the atom in the system. Since the MSD value can be averaged over the number of atoms, the equation simplifies to $D\alpha = a/6$. The data for the concentration profiles were collected from the stabilized system between 210 and 250 ps for statistical and structural analysis.

4.2 Density Functional Theory (DFT) simulation

Density Functional Theory (DFT) simulations were conducted using the commercial software Materials Studio with the CASTEP module.^[2] GGA (generalized gradient approximation) was employed for the exchange–correlation. The self-consistent ground state cell optimization was performed by density mixing scheme using an ultra-soft pseudo-potentials (USP) plane-wave basis set, with a cut-off energy of 380 eV and thermal broadening of 0.1 eV. The Monkhorst-Pack was used to determine the k-points, which was fixed at 0.04 Å⁻¹ for all calculations.

The surface simulations were performed using a slab model with periodic boundary conditions. A cell of 15×15×35Å was constructed with a single optimized glucose

(Glucopyranose) molecule model placed at the bottom. Then, the a single optimized solvent molecule (water/chloroform) was placed ~ 2.0 Å from glucose model at the position of the oxygen atom in the glucopyranose ring. The hydrogen bonds was calculated based on the atoms position after the geometry optimization.

The adsorption energy values of gluocose molecule to different solvent molecules were calculated using the following formula:

 $E_{ads} = [E_{(glucose/solvent)} - (E_{glucose} + E_{solvent})]$

Where E_{chem} is the adsorption energy, $E_{(glucose/solvent)}$ is the total energy after adsorption, $E_{glucose}$ is the total energy of the glucose molecule, and $E_{solvent}$ is the total energy of water or chloroform molecules after optimization. A negative binding energy indicates favorable adsorption.

5. The influence of different organic solvents (Table S1, Fig. S5)

Different types of organic solvents were used to replace the role of water in this GHBN approach. Generally, 3 g organic solvent and 4 g amylopectin were mixed together, heated under 60°C for 5 min in a water bath and naturally cooled to 25°C (room temperature). As shown in Table S1, the polar solvents, such like ethanol, ethylene glycol, glycerol and acetone, contained the oxygen atoms that can form hydrogen bonds with hydroxyl groups of amylopectins, similar to that of water. However, the resulting products were either phase separation or easy to dry since these kinds of alcohols were volatile solvents (Case I, Fig. S5a). Chloroform is another kind of polar solvent without electronegative oxygen atom. Thus, chloroform was unable to form hydrogen bonds among themselves, leaving powder-like and easily broken mixture (Case II, Fig. S5c). We also used non-polar solvent and charged ionic liquids to replace water. Unfortunately, the mixtures were all powder-like similar to that by using chloroform.

6. Measuring the Young's modulus of as-prepared GHBN (Figure 2b, Table S2, Figure S6)

We measured 13 samples with different amylopectin/water/salt ratios in region (III) (Fig. 2). All the samples were shaped into 10 mm (Width) $\times 1$ mm (Thickness) $\times 50$ mm (Length). The testing condition was at room temperature 25°C and the humidity was 30-35%. The samples were fixed on a tension tester (BOSE ElectroForce 3200 Series III Test Instruments) and left 1.0 cm length to stretch (Fig. S6b). The stretching speed was 0.5 mm/s. The values of Young's modulus of samples with different ratios were listed in Table S2. The Young's

modulus results were varied from 1305 Pa (Sample 24) to 198 Pa (Sample 31). To better show the relationship between the Young's modulus and the component ratio, we distinguished the results in different color depths (Fig. 2b, Fig. S6a). The Young's modulus of Sample 24, 44, 53 and 45 were more than 1000 Pa. The modulus of Sample 27 was litter lower than dark yellow region, only about 950 Pa, while the modulus of samples in region with lightest yellow color were less than 500 Pa. The values of Young's modulus results were calculated from the gradient of the stress-strain curves. Three representative types of curves could be found in Fig. S6c-e.

7. Measuring the conductivity of as-prepared GHBN (Fig. 2b, Table S3, Fig. S7)

Since the GHBN were ionic conductors, we measured the conductivity of 13 samples with different amylopectin/water/salt ratios in region (III) (Fig. 2) through an alternating current (AC) impedance method. The measuring system and the instrument was PARSTAT 2273 (Princeton Applied Research) (Fig. S7b). Current ranging was 200 mA, and the frequency range was from 1 Hz to 1 MHz. The samples was cut into 5 mm (Width) \times 5 mm (Length) \times 1mm (Thickness), and sandwiched by copper tapes (Fig. S7c,d). The testing condition was at room temperature 25 °C and the humidity was 30-35%. The conductivities of samples were listed in Table S3, and distinguished in different color depths in Fig. S7a. The AC resistances were varied from 0.2 to nearly 0 S/m. The samples could be classified into 3 types based on their AC resistance. The values of conductivities were calculated from the impedance curves. Three representative types of curves could be found in Fig. S7e.

8. Antiseptic function of NaCl (Fig. S8)

In this GHBN system, the employment of high-concentration salt not only makes GHBN to be conductive but also plays an antiseptic role in preventing the GHBN from bacterial breeding. The GHBN with/without NaCl (the weights of amylopectin and water were the same) were stored into glass beakers, and covered by a film of fresher film (Fig. S8). After placed at room temperature, under the humidity of 30-35% for 5 days, the GHBN without NaCl was full of germs while the counterpart with NaCl was still in a good condition.

9. Other naturally renewable powders instead of amylopectin (Table S4, Fig. S9)

We compared the properties of the mixtures fabricated by different naturally renewable powders, rather than the amylopectin. As shown in Table S4, amylose, gluten and commercial plain flours which contained amylose, amylopectin and gluten were used to fabricate GHBN instead of amylopectin. The preparation processes were similar to that by using the amylopectin.

10. The influence of working temperature (Fig. S10)

We compared the conductivities of the GHBN under different working temperature (Fig. S10). We used a hot plate to control the working temperature. The working temperature was changed from 20°C to 70°C, and the according conductivities were recorded by the AC impedance method.

11. The influence of different salts (Figure S11)

We compared the influences of cations and anions on the conductivities of GHBN by replacing the NaCl by other inorganic salts (Fig. S11). For the cations, the concentrations of NaCl, KCl, CaCl₂ and FeCl₃ were fixed at 4.6 mol/L since KCl exhibited a saturated concentration to this value. For the anions, the concentration of the NaCl, Na₂SO₄ and Na₃PO₄ were 0.74 mol/L since Na₃PO₄ exhibited a saturated concentration to this value. The according conductivities were recorded by the AC impedance method. The testing condition was at room temperature 25°C and the humidity was 30-35%.

12. The influence of silver nanowires (Fig. S12)

Different amounts of silver nanowires, with diameter of 30 ± 5 nm and length ranged from 100 to 200 µm (purchased from Nanjing XFNANO materials Tech Co., Ltd), were used to replace the role of NaCl in the GHBN fabrication. The preparation processes were similar to that by using the amylopectin. The according conductivities were recorded by the AC impedance method. The testing condition was at room temperature 25 °C and the humidity was 30-35%.

13. Sealing to protect the GHBN (Fig. S13)

A commercial available silicone elastomer, Ecoflex 00-30 was used as the polymeric layer to protect the GHBN from the water evaporation. The silicone elastomer liquid was prepared with one part Ecoflex 00-30 A and one part Ecoflex 00-30 B (Smooth-On). The liquid was dropped onto the GHBN bulk and cured under 25°C overnight. Owing to the existence of this polymeric coating, the GHBN bulk can remain their mechanical as well as electrical properties for more than 180 days. The according conductivities were recorded by the AC

impedance method. The testing condition was at room temperature 25°C and the humidity was 30-35%.

14. Healing study (Fig. 3)

A GHBN piece, with diameter of ~2 mm and length of ~10 cm, was linked into a direct current (DC) circuit and the voltage was 1V to prevent possible electrochemical reactions or water electrolysis.^[3] The testing condition was at room temperature 25°C and the humidity was 30-35%. An open-circuit current appeared when the GHBN piece was completely cut off. When the fresh cut surfaces of these two fragments were brought into contact, the current increased and remained close to its initial value. The electrical healing process was just within 2-3 s. The cutting/healing cycles were repeated more than 100 times and the GHBN remained their healing property. For a visual demonstration, a GHBN piece was linked into a DC circuit in series with a fan and a LED.

15. Fabrication of GHBN based switch and electrical wires (Fig. 5b,c and Fig. S15)

A single-pole double-throw switch was fabricated by hand with the assistance of a mould. The programmed GHBN electrical wires located on 3D spherical surfaces were performed by injecting the GHBN through a 10 mL syringe by hand.

16. Fabrication of GHBN based flexible battery (Fig. 5d-f, Fig. S17-19)

The anode of flexible battery was made as follows: one GHBN bulk was homogenously mixed with 50 wt% copper (Cu) microparticles with the diameter of ~75 μ m (purchased from Sigma-Aldrich). The anion of flexible battery was made as follows: one GHBN bulk was homogenously mixed with 50 wt% zinc (Zn) microparticles with the diameter of ~5 μ m (purchased from Sigma-Aldrich). Then these two blocks were pushed together, yielding a flexible Cu-Zn battery.

The potential of separated anode or cathode part was 0 V (Fig. S16a,b). However, the potential increased to 0.57 V when the two parts were contacted. The potential can be raised by a series connection of the GHBN based flexible battery. To prevent the existence of inner electric potential, each flexible battery was connected by a copper tape.

17. Fabrication of GHBN based flexible electroluminescent light (Fig. 5g,h, Fig. S20,S21)

The flexible electroluminescent device was assembled by three layers: two GHBN layers on the top /bottom parts, and an emissive layer in the middle part. The fabrication of middle emissive layer can be found as follows:^[4] PDMS 184 liquid (Dow Corning 10:1) and ZnS:Cu

microparticles (purchased from Shanghai KPT company) were mixed in a weight ratio of 1:1. Then, the opaque solution was spin-coated onto a polystyrene Petri dish with a speed of 1000 r/s for 60 seconds. The PDMS/ZnS:Cu gel was fully solidified under 80 °C for 2 hours and its thickness was ~56 µm. Finally, it can be peeled off and used. To make the electroluminescent device to release the light, the thickness of GHBN should not be too thick, about 1 mm. Copper wires were used to electrically connect the GHBN layers. The input power of this flexible electroluminescent device was purchased from the Shanghai KPT company. The power device can transfer the DC field of two commercial batteries to an AC field with increased voltage.

18. Characterizations

The mechanical measurements were performed by BOSE ElectroForce 3200 Series III Test Instruments. The AC impedance investigations were measured by a PARSTAT 2273Advanced Electrochemical System, Princeton Applied Research. Environmental scanning electron microscope (ESEM) images and elemental analysis were performed by using the HITACHI TM3030 Tabletop SEM. Voltage of GHBN based batteries was measured by an Agilent U1251B Handheld Digital Multimeter. Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectra were recorded on a Bruker Equinox 55 spectrometer.



Fig. S1 Optimized temperature range in the preparation process. When the temperature was lower than 55° C or higher than 65° C, the amylopectin/water/salt ternary mixture was in a liquid state or formed a very sticky gel, which was not suitable for use in further electrical application. A homogenous soft matter state could be obtained when the temperature was $55\sim65^{\circ}$ C, see the green region.



Fig. S2 Infrared spectra are difficult to observe the formation of dynamic hydrogen bonds in this GHBN strategy. ATR-FTIR Spectra of amylopectin and the amylopectin/water/salt ternary mixture. The absorption peak ranged from 2900 to 3600 cm⁻¹ cannot attribute to the formation of hydrogen bonds bridging hydroxyl groups of amylopectin and those of water molecules, since water itself contains a plenty of hydrogen bonds in this region.



Fig. S3 Water, rather than chloroform, can form physically cross-linked hydrogen bonds with hydroxyl groups of the glucopyranose unit. Density functional theory (DFT) computational modeling of the interaction between glucopyranose molecules and (a) water or (b) chloroform molecules. Physically cross-linked hydrogen bonds can exist among hydroxyl groups of the glucopyranose and those of water molecules. However, lowly electronegative chlorine atoms of chloroform were unable to form hydrogen bonds with hydroxyl groups of the glucopyranose. (c) The values of chemisorption energy of water (blue in color) and chloroform (green in color) when met with the amylopectin.



Fig. S4 Chloroform molecules cannot build a hydrogen bonded network with amylopectin molecules. (a) Schematic illustration of proposed interactions between chloroform and amylopectin molecules. Lowly electronegative chlorine atoms of chloroform were unable to form hydrogen bonds among themselves, leaving two separated amylopectin clusters. Thus, the mixture of amylopectin and chloroform cannot serve as dynamic materials. (b) Molecular dynamics (MD) computational modeling of two amylopectin clusters in the chloroform matrix separated with a gap of 0.5 nm in a $10 \times 10 \times 10$ nm³ cell at 0 ps and 100 ps, respectively. Only part of amylopectin molecules contacted together, which was contributed by the intermolecular hydrogen bonds between the hydroxyl groups of amylopectin molecules. (c) Computational modeling of few intramolecular hydrogen bonds among themselves, yielding the failure of building a hydrogen bonded network. (d) The dependence of displacement on the time in the MD modeling. The curves were obtained by calculating the

displacement of amylopectin clusters in water (blue in color), chloroform (green in color) and vacuum (grey in color) matrix. (e) The values of diffusivities of amylopectin clusters in water (blue in color), chloroform (green in color) and vacuum (grey in color) matrix. The diffusivities were calculated according to the gradient of the curves in (d).

	Solvent species	Chemical formula	Possibility	Result	Digit
			to	after	al
			generate	mixing	image
			hydrogen	with	S
			bonding	amylopect	
			with	in	
			amylopect		
			in		
Polar Solvent	Ethanol	ОН	Yes	Phase	
	Acetone	o	Yes	separation	
	Ethylene Glycol	ноон	Yes		Case
	Glycerol	НООН	Yes	Easy to dry	
	Chloroform	CI CI CI	No	Powder- like	Case (II)
Non- polar Solvent	n-hexone	\sim	No	Powder- like	Case (II)
Ionic Liquid	1-Methyl-3- octylimi- dazolium chloride		No	Powder-	Case
	1-Butyl-3- methylimida- zolium bromide	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~	No	like	(II)

Table S1. Diverse organic solvents mixed with amylopectin



Fig. S5 Other organic solvents are difficult to build stable structurally dynamic materials when mixed with amylopectins. Representative photographs of the mixtures consisting of amylopectins and (a,b) ethanol or (c,d) chloroform solvents, rather than water. The mixture that consisted of amylopectins and ethanol was easy to dry due to the intrinsic volatile nature of this organic solvent. The mixture that consisted of amylopectins and chloroform just showed a powder like status due to mismatched molecular interaction.

Sample NO.	Amylopectin	NaCl percentage	Water	Young's Modulus
	percentage (%)	(%)	percentage	(Pa)
			(%)	
24	40	30	30	1,305
44	45	20	35	1,225
53	33	33	34	1,124
45	35	30	35	1,081
27	50	10	40	943
54	50	5	45	475
18	30	30	40	338
59	50	0	50	326
60	60	0	40	310
19	30	40	30	226
16	45	23	32	218
23	40	20	40	210
31	28	42	30	198

Table S2. Young's modulus of diverse samples

(Note: The size of the samples was 50 mm x 10 mm x 1 mm; the testing condition was at room temperature, ~ 25°C and the humidity was ~ 35%)



Fig. S6 Mechanical study of GHBN. (a) Diagrams of mechanical properties of GHBN in a homogenous soft matter state. The values are marked by a range of color depths. The detailed value and component of each point can be found in the Table S2. (b) Representative photograph of a sample being measured by a tension tester. Representative stress-stain curves of (c) sample 24, (d) sample 27, and \in sample 54, respectively. The sizes of the samples are 50 mm x 10 mm x 1 mm; the testing condition was at room temperature, ~ 25°C and the humidity was ~ 35%.

Sample NO.	Amylopectin	NaCl percentage	Water	Resistance
	percentage (%)	(%)	percentage	(S m ⁻¹)
			(%)	
19	30	40	30	0.197
24	40	30	30	0.174
18	30	30	40	0.172
16	45	23	32	0.172
31	28	42	30	0.171
23	40	20	40	0.169
53	33	33	34	0.151
27	50	10	40	0.146
45	35	30	35	0.145
44	45	20	35	0.142
54	50	5	45	0.046
60	60	0	40	N/A
59	50	0	50	N/A

 Table S3. Resistance of diverse samples

(Note: The size of the samples was 5 mm x 5 mm x 1 mm; the testing condition was at room temperature, ~ 25°C and the humidity was ~ 35%)



Fig. S7 Electrical study of GHBN. (a) Diagrams of electrical properties of GHBN in a homogenous soft matter state. The values are marked by a range of color depths. The detailed value and component of each point can be found in the Table S3. (b) Representative photograph of a sample being measured by an electrochemical system. (c) Photograph and (d) schematic illustration of the details for measuring the sample. (e) Representative impedance curves of sample 24, sample 27, and sample 54, respectively. The size of the samples was 5 mm x 5 mm x 1 mm, the testing condition was at room temperature, ~25°C and the humidity was ~35%. All the impedance measurements were under an alternating field (AC) to prevent the appearance of electrochemical reactions or water electrolysis.



Fig. S8 Salt not only makes GHBN to be conductive but also plays an antiseptic role in preventing the GHBN from bacterial breeding. Representative photographs of GHBN (a) with and (b) without salt after stored at room temperature for 5 days. The beakers were covered by a layer of fresher film.

Powder species	Possibility to	Result after mixing	Digital images
	generate hydrogen	with water and salt	
	bonding with water		
Amylose	Yes	Powder-like, easy to	Case I
		break	
Gluten	Yes	Powder-like, easy to	Case II
		break	
Commercial plain flours	Yes	Homogeneous	Case III
(contains amylose,			
amylopectin and gluten)			

 Table S4. Diverse renewable powders mixed with water and salt.



Fig. S9 Attempts to use other renewable powders to build GHBN. Photographs of the mixtures consisting of (a) amyloses, (b) glutens, and (c) commercial flours that contains amylose, amylopectin, gluten and other additives. Amyloses or glutens just enabled the mixtures to show a powder like status, while the commercial flour yielded mixture exhibited a homogenous and elastic state. (d) Photograph of the above-mentioned mixtures placed in the bottle. The mixture using amyloses showed a phase separation phenomenon.



Fig. S10 The conductivity of GHBN increased when the working temperature raised. The dependence of conductivity of GHBN on the working temperature.



Fig. S11 The conductivity of GHBN can be improved by changing the types of salts. (a) The dependence of conductivity of GHBN on the salts with diverse cations. The concentration of the salt solution was fixed at 4.6 mol/L to ensure all the salts were completely dissolved in the water. (b) The dependence of conductivity of GHBN on the salts with diverse anions. The concentration of the salt solution was fixed at 0.74 mol/L to ensure all the salts were completely dissolved in the salts were completely dissolved in the salt solution was fixed at 0.74 mol/L to ensure all the salts were completely dissolved in the water. The testing condition was at room temperature, $\sim 25^{\circ}$ C and the humidity was $\sim 35\%$. Representative photographs of the mixtures consisting of salts with diverse (c) cations and (d) anions.



Fig. S12 The conductivity of GHBN increased by raising the quantity of silver nanowires. The dependence of conductivity of GHBN on the quantity of silver nanowires. Error bars represent standard deviation from five samples per weight fraction. The inset images are the environmental scanning electron microscope (ESEM) images of samples within different weight fraction. The testing condition was at room temperature, ~25°C and the humidity was ~ 35%.



Fig. S13 Sealing is a better way to prevent the evaporation of water in the GHBN. (a) Schematic illustration of the process of sealing a GHBN bulk. The Ecofelx polymeric liquid was carefully dropped onto a piece of GHBN to form a transparent cover. With the evaporation of organic solvents and further solidification of the Ecoflex, a thin protecting layer could be formed upon the GHBN. (b) Representative photographs of the sealing process. (c) Cross-sectional ESEM image of the Ecoflex covered GHBN. The thickness of the Ecoflex layer was around 5 micrometers. (d) The dependence of the conductivity of GHBN on the storage time. Owing to the existence of the polymeric layer, the water in the GHBN cannot evaporate, yielding a stale conductivity for more than 180 days. The testing condition was at room temperature, ~25°C and the humidity was ~35%.



Fig. S14 GHBN can heal in both mechanical and electrical fields. The values of (a) Young's modulus and (b) conductivity of the GHBN before (orange in color) and after (purple in color) healing. The samples were cut then closely put together for more than 5 minutes before testing. The testing condition was at room temperature, ~25°C and the humidity was ~35%.



Fig. S15 Flexibility of GHBN to serve as programmed electrical wires. (a-d) Representative photographs of the GHBN electric wires placed upon 3D spherical surfaces with on-demand patterns in series with a LED light. The LED kept lighted, indicating the GHBN were able to serve as the electric wires.



Fig. S16 Electrical properties of the stretchable Ecoflex sealed GHBN wire. (a) Plots of resistance change of Ecoflex sealed GHBN wire for the applied strain in the range from 3% to 400%. (b) The life-time test under a strain of 100% at a frequency of 0.25 Hz. Left part: The resistance change curves were recorded after each 1000 cycles and 200 cycles of data are shown in each recording. Right part: The magnified view of the part of $\Delta R/R_0$ -*t* curve after 5000 loading-releasing cycles. (c) Plots of resistance change of the Ecoflex sealed GHBN wire as a function of time (input strain: 30%) for diverse frequencies including 0.04, 0.08 and 0.25 Hz.

GHBN-based soft battery



Fig. S17 The fabrication of GHBN based soft battery. (a) Schematic illustration of the fabrication process of GHBN based soft battery. Two pieces of GHBN were mixed with 50 wt% copper (Cu) or zinc (Zn) microparticles (MPs), respectively. Then these two blocks were pushed together, yielding a flexible Cu-Zn battery. Cu/Zn microparticles served as cathode/anode, whereas GHBN played a role as the soft solid electrolyte. (b) ESEM images and elemental analysis of GHBN incorporated with Cu MPs. Cu-MP/amylopectin/NaCl, represented by copper/carbon/sodium, are marked in red, blue and yellow, respectively. (c) ESEM images and elemental analysis of GHBN incorporated with Zn MPs. Zn-MP/amylopectin/NaCl, represented by zinc/carbon/sodium, are marked in green, blue and yellow, respectively.



Fig. S18 GHBN based soft battery can be cut and connected to increase the potential. (a-i) Representative photographs of cutting then connecting the GHBN based battery. The potential of one GHBN based battery was \sim 0.57 V. The potential of a series connection of eight GHBN batteries could reach as high as 4.16 V. The bottom part of each image is the schematic illustration of each process.



Fig. S19 A series connection of GHBN based battery can increase the power voltage. The dependence of the power voltage on the number of series connected GHBN based battery. The testing condition was at room temperature, $\sim 25^{\circ}$ C and the humidity was $\sim 35\%$.



Fig. S20 Cross-sectional ESEM image of the ZnS:Cu emissive layer. The ZnS:Cu microparticles were homogenously dispersed in the PDMS layer and the thickness of this layer was \sim 56 μ m.



Fig. S21 A GHBN based flexible electroluminescent light. (a) Schematic illustration of a GHBN based flexible electroluminescent light. The device was composed of the bottom and top GHBN conductors with sandwiched a \sim 56 µm ZnS:Cu/Ecoflex emissive layer. Photograph of a flexible electroluminescent device in a (b) off and (c) on state. The electroluminescent region sandwiched between top/bottom GHBN layers can emit a blue luminescent light when applied a high-frequency alternating voltage. The relative humidity was \sim 35% and the temperature was 25°C.

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