S1

Supporting Information

Lightweight, Superhydrophobic, Lignin-Based Polyurethane Foams for Underwater Pressure Sensing

Honglong Zhao,^{*a,b*} Xiaozhen Ma,^{*a,b*} Xiaobo Xu,^{*a*} Minghui Cui,^{*a*} Nathan E. Stott,^{*a,b*} Jin

Zhu,^{*a,b*} and Jing Chen^{*a,b*}*

a Key Laboratory of Bio-based Polymeric Materials Technology and Application of Zhejiang Province, Laboratory of Polymers and Composites, Ningbo Institute of Material Technology and Engineering, Chinese Academy of Sciences, Zhejiang, Ningbo 315201, PR China

b University of Chinese Academy of Sciences, Beijing, 100039, PR China



Figure S1: Quantitative 31P NMR spectra of lignin polyol.

The hydroxyl value of the lignin polyol after water removal is 7.5 mmol/g, as shown in Fig.S1.



Figure S2: Images of (a) LPUF and (b) Gr-LPUF.

Fig. S2a is photo of LPUF, Fig.S2b is photo of LPUF.



Figure S3: (a) SEM image of Gr-LPUF (b) Foam pore size and its distribution.

The surface of unmodified Gr-LPUF is comparably smooth. The size and distribution of the foam holes are relatively uniform.



Figure S4: Reflective air bubbles on the surface of a superhydrophobic, conductive LPUF.

When the {{Gr-LPUF}PPy}PDMS foam was put into water, a layer of reflective bubbles on the surfaces of the foam were clearly observed, as shown in Fig. S4.



Figure S5: Cyclic compression curves of {{Gr-LPUF}PPy}PDMS and partial enlarged versus.

{{Gr-LPUF}PPy}PDMS foam also has a very good elastic recovery rate, after 50 repeated compression tests, {{Gr-LPUF}PPy}PDMS foam still has an elastic recovery rate of 93.88% and a maximum compressive strength of 378.45KPa. All of these characteristics are favourable for the preparation of piezoresistive sensors.



Figure S6: Foam resistance before and after PDMS coating process (a) Before coating with PDMS (b) After coating with PDMS.

To explore the effect of PDMS coatings on the electrical conductivity of foams, we measured the electrical resistance of the {Gr-LPUF}PPy foam before and after PDMS dip-coating, from Fig.S6 we can see that the foam electrical resistance increased from $23.24k\Omega$ to $33.58k\Omega$ after immersing PDMS(The size of the foam is 3.5cm*2.5cm*1cm), because the PDMS coating hinders the conductive components from contacting each other, the number of conductive pathways decreases, and the electrical resistance increases. Although the electrical resistance of the foam increased, the PDMS coating has little effect on the variation of material electrical resistance with pressure, foam maintains good sensing properties.

Polymer materials	Conductive material	Sensing range	Pressure sensitivity	Reference
Sodium alginate sponge	Mxene	0-1.85 kPa	2.4 kPa ⁻¹	42
		1.85-20 kPa	3.56 kPa ⁻¹	
		20-66.6 kPa	0.5 kPa ⁻¹	
Polyurethane foam	rGO	0-2.7 kPa	0.022 kPa ⁻¹	40
	MWNTs	2.7-10.8 kPa	0.088 kPa ⁻¹	
		10.8-48.8 kPa	0.034 kPa ⁻¹	
Highly compressible	rGO	0-5 kPa	4.93 kPa ⁻¹	43
wood sponge		5-50 kPa	0.75 kPa ⁻¹	
Polydimethylsiloxane	AgNPs	0-40 kPa	0.0132 kPa ⁻¹	22
sponge	rGO	40-120 kPa	0.0033 kPa ⁻¹	
Commercial melamine-	WS ₂ nanosheet	0-125pa	0.39 kPa ⁻¹	20
formaldehyde sponges		125Pa-5 kPa	0.02 kPa ⁻¹	

Table S1:	A comparison of work in recent ye	ars
-----------	-----------------------------------	-----

S4

High-internal-phase emulsions sponges	Graphene	0-20 kPa	0.083 kPa ⁻¹	38
Rubber sponge	AgNPs	0-9 kPa	0.005 kPa ⁻¹	39
		9-30 kPa	0.0130 kPa ⁻¹	
		30-90 kPa	0.002 kPa ⁻¹	
PDMS foam	CNF	0-1 kPa	0.06 kPa ⁻¹	44
		1-6 kPa	0.08 kPa ⁻¹	
		6-20 kPa	0.01 kPa ⁻¹	
Lignin-based	Graphene	0-25 kPa	6.38 kPa ⁻¹	This work
polyurethane foams	РРу	25-150 kPa	1.14 kPa ⁻¹	
		150-350 kPa	0.48 kPa ⁻¹	

We compare the performance with those of some published hydrophobic foam sensors in recent years, as shown in Table S1, the superhydrophobic conductive foam sensor has a comparably high sensitivity in the range of 0-25 Kpa.



Figure S7: Durability testing at 50% compression deformation underwater for 1000 compression cycles.

We combined the electro-chemical workstation with a 1KN universal material testing machine to compress the superhydrophobic conductive foam sensor underwater for 1,000 cycles of 50% deformation and record the change of the current signal. As shown in Fig. S7, the signal remains stable in 1000 cycles, indicating that the superhydrophobic conductive foam sensor also has excellent stability.



Figure S8: Water depth detection by SCFS.

As can be seen in Fig. S8, I/I_o rose immediately the moment the foam came into contact with the water while the electrical signal remained stable when the water addition was temporarily stopped. Throughout the process, the I/I_o curve follows a trapezoidal upward trend. When the water level change, the SCFS shows a clear signal change, and the change in current reflects the change in depth of the water in real time.



Figure S9: Digestion process of superhydrophobic, conductive LPUF in alkaline 0.5 mol/L NaOH methanolic solution at 60°C.

As shown in Fig. S9, 0.045 g of superhydrophobic, conducting LPUF was put into a 0.5 mol/L sodium hydroxide methanol solution while placed in an oven at 60°C. The solution started to turn light yellow after one hour and then turned dark brown after 3 hours with no visible solid materials, yielding complete alkaline digestion of the foam.