Supporting Information

Tailor-Made Polymorphs and Nanostructure Strategy to Construct Self-Reinforced Nonswelling High-Strength Bacterial Cellulose Hydrogels

Minghao Zhang, Shiyan Chen, Nan Sheng, Jingjing Yao, Baoxiu Wang, Zhuotong Wu, Huaping Wang**

State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, College of Materials Science and Engineering, Donghua University, Shanghai 201620, PR China

Email addresses: chensy@dhu.edu.cn; wanghp@dhu.edu.cn

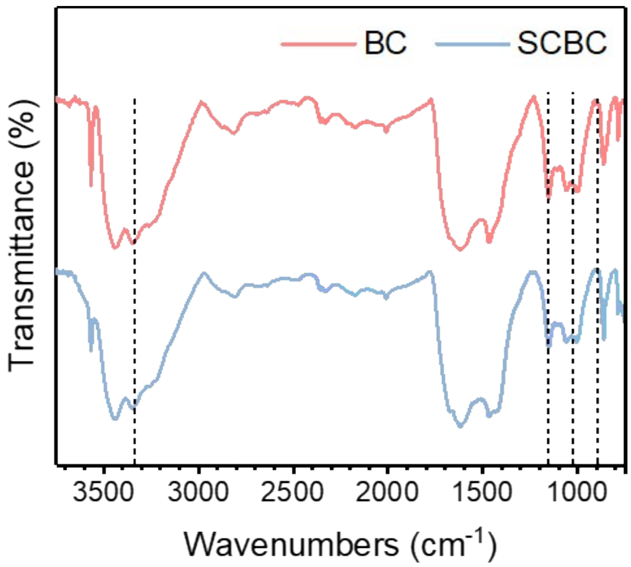

Fig. S1. FT-IR spectra of BC and SCBC hydrogel.

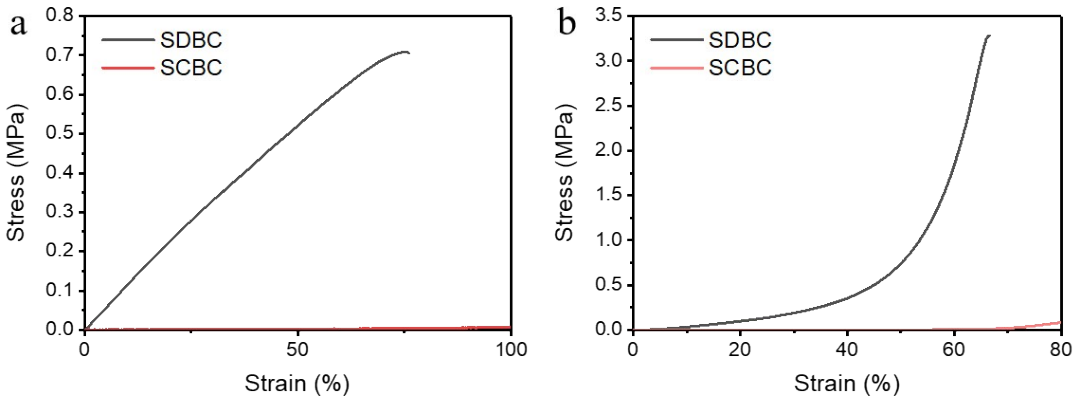

Fig. S2. (a) Tensile (b) compressive stress-stain curves of SCBC and SDBC.

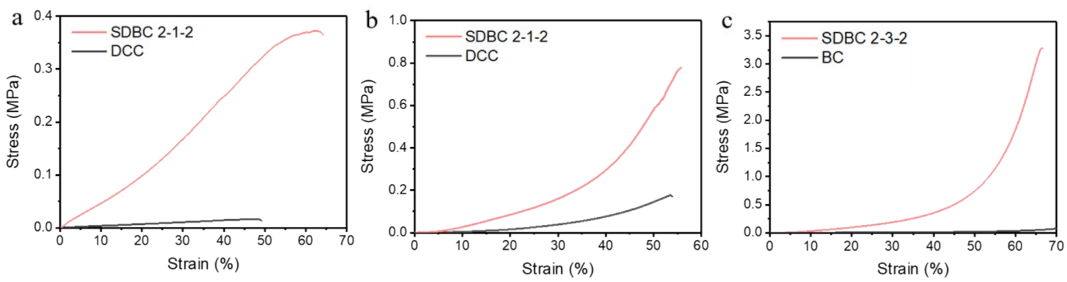

Fig. S3. (a) Tensile (b) Compressive stress-stain curves of SDBC 2-1-2 and DCC hydrogels. (c) Compressive stress-stain curves of SDBC 2-3-2 and BC hydrogels.

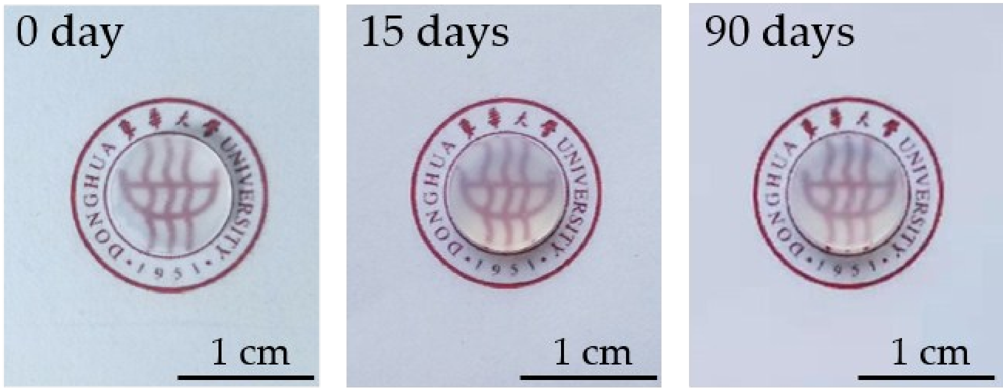

Fig. S4. Snapshots of SDBC hydrogel after 15 days and 90 days soaking in water compared with original hydrogel.

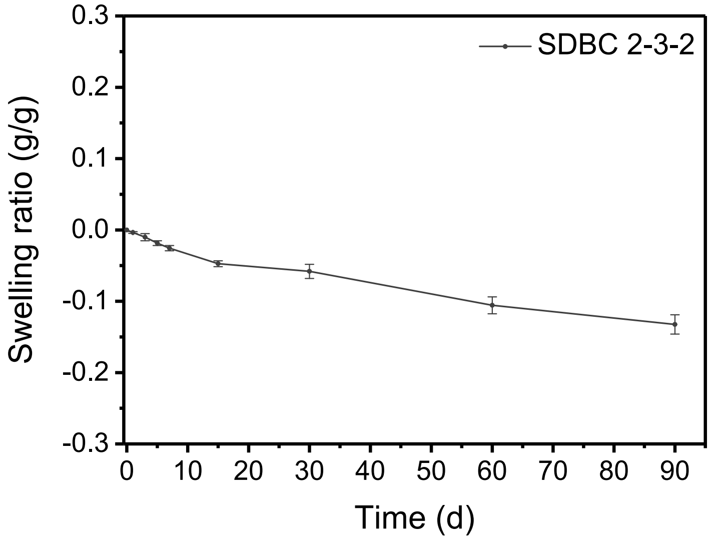

Fig. S5. Swelling ratio (g/g) curve of SDBC 2-3-2 hydrogel.

Contrary to the swelling behavior, we observed the quality decrease of the hydrogels with the time went on. The reason may come from the degradation of hydrogels to a certain extent which is in line with the need for degradation of medical materials.

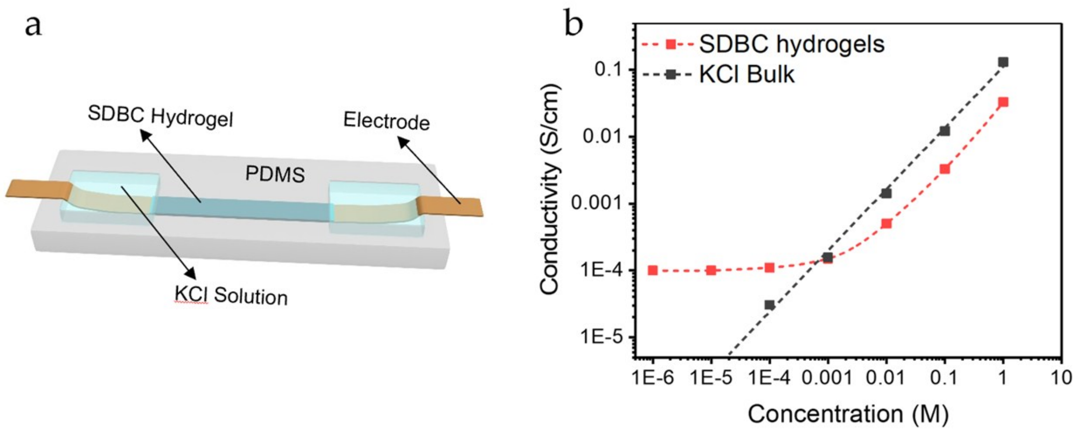

Fig. S6. (a) Schematic of the ionic conductivity measurement of the nanofluidic devices. (b)The ionic conductivity of the SDBC 2-3-2 hydrogel and KCl bulk at different KCl concentrations.

Nanofluid ion transport phenomena occur in many physiological activities, such as the movement of muscles and signal transmission in neurons.[1] The ionic conductivity of the SDBC hydrogel and KCl bulk at different content of KCl was determined and plotted in Fig. S6. The ionic conductivity of SDBC is close to that of the bulk KCl solution when the KCl concentration is higher than 0.01 M, which is linearly proportional to the KCl concentration. However, when the KCl concentration is lower than 0.01 M, the conductivity gradually stabilizes at 1×10^{-4} $S\cdot cm^{-1}$, which regardless of the KCl concentration. The high ionic conductivity at low electrolyte concentrations demonstrates that SDBC hydrogels have typical nanofluid ion transport behavior. This phenomenon is related to the surface charge densities of the SDBC hydrogels and the diameter of the nanochannel inside it.[2] The SDBC hydrogels contains a large amount of BC nanofibers, and nanochannels of 3-5 nm or less are formed between the microfibrils in the nanofibers.[3] These nanochannels facilitate ion transport, while the surface groups with the negative charge contribute to high positive ion selectivity, due to plenty of hydroxyl groups in the molecular chain of cellulose, which can be confirmed by the zeta potential of −6.5 mV (at pH value of 6.3). So SDBC hydrogels can be used as robust, flexible, mechanically stable and biocompatible nanofluidic devices for biorelated applications, such as biosensing, and implantable ionic devices.

Table S1. Different ratio of ECH:AGU of SDBC hydrogels with the same content of LiOH and urea in preparing process.

	M, ECH: AGU	Urea (wt%)	LiOH (wt%)
SDBC 1-3-2	0.52	25	4.5
SDBC 2-3-2	1.04	25	4.5
SDBC 3-3-2	2.07	25	4.5
SDBC 4-3-2	4.14	25	4.5
SDBC 5-3-2	8.28	25	4.5

Table S2. Different urea content of SDBC hydrogels with the same content of LiOH and ratio of ECH:AGU in preparing process.

	M, ECH: AGU	Urea (wt%)	LiOH (wt%)
SDBC 2-1-2	1.04	15	4.5
SDBC 2-2-2	1.04	20	4.5
SDBC 2-3-2	1.04	25	4.5
SDBC 2-4-2	1.04	30	4.5
SDBC 2-5-2	1.04	35	4.5

Table S3. Different LiOH content of SDBC hydrogels with the same content of urea and ratio of ECH:AGU in preparing process.

	M, ECH: AGU	Urea (wt%)	LiOH (wt%)
SDBC 2-3-1	1.04	25	4
SDBC 2-3-2	1.04	25	4.5
SDBC 2-3-3	1.04	25	5
SDBC 2-3-4	1.04	25	5.5

Table S4. Physical properties of SDBC hydrogels at different urea content.

		Tension			Compression		
	W_{H2O} (wt%)	σ (MPa)	ε (%)	E (MPa)	σ (MPa)	ε (%)	E (MPa)
SDBC 2-1-2	94.9	0.37	62	0.47	0.82	62	0.58
SDBC 2-2-2	93.7	0.44	73	0.81	1.08	61	0.59
SDBC 2-3-2	91.4	0.72	74	1.23	3.17	67	0.67
SDBC 2-4-2	92.0	0.35	103	0.63	2.19	64	0.75
SDBC 2-5-2	93.9	0.26	87	0.42	0.74	53	0.65

Table S5. Physical properties of SDBC hydrogels at different LiOH content.

		Tension			Compression		
	W_{H2O} (wt%)	σ (MPa)	ε (%)	E (MPa)	σ (MPa)	ε (%)	E (MPa)
SDBC 2-3-1	94.1	0.25	89	0.33	1.04	62	0.48
SDBC 2-3-2	91.4	0.72	74	1.23	3.17	67	0.67
SDBC 2-3-3	91.5	0.26	91	0.49	1.21	51	1.01
SDBC 2-3-4	91.1	0.24	90	0.44	0.93	51	0.88

Table S6. Physical properties of SDBC hydrogels at different ratio of ECH/AGU.

		Tension			Compression		
	W_{H2O}(wt%)	σ (MPa)	ε (%)	E (MPa)	σ (MPa)	ε (%)	E (MPa)
SDBC 1-3-2	92.6	0.30	65	0.59	1.58	52	0.94
SDBC 2-3-2	91.4	0.72	74	1.23	3.17	67	0.67
SDBC 3-3-2	93.4	0.16	58	0.09	1.63	61	0.38
SDBC 4-3-2	95.0	0.13	66	0.08	0.89	59	0.15
SDBC 5-3-2	95.1	0.12	77	0.02	0.59	42	0.13

REFERENCES

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2. T. Li, S. X. Li, W. Kong, C. Chen, E. Hitz, C. Jia, J. Dai, X. Zhang, R. Briber, Z. Siwy, M. Reed and L. Hu, *Sci. Adv.*, 2019, **5**, eaau4238.
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