

Supplementary Information

Antifouling Hydrogel Film Based on Sandwich Array for Salivary Glucose Monitoring

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Fig. S1

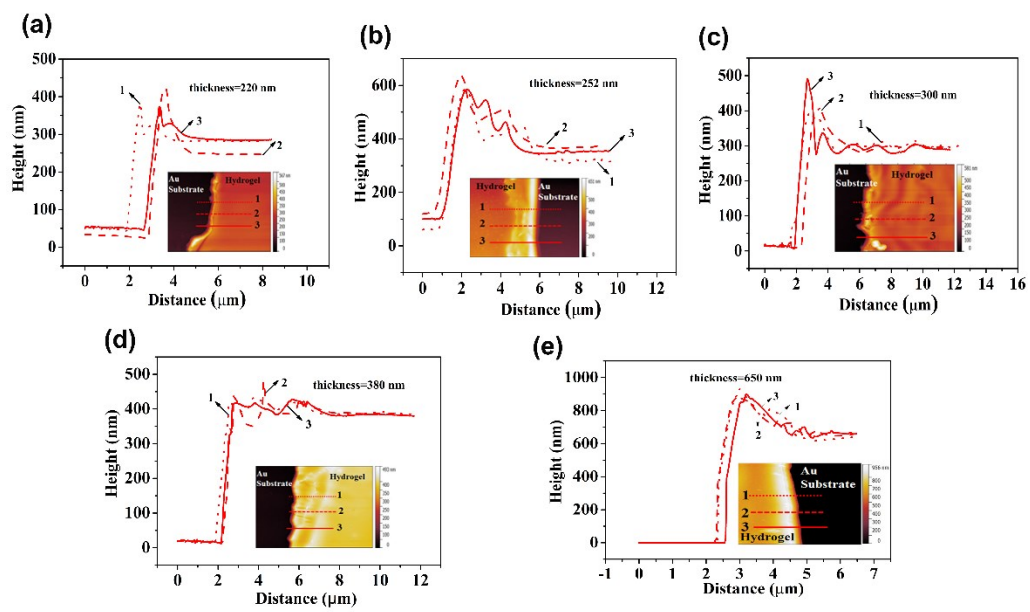


Fig. S1 The different thickness of HFSA obtained at different spinning speed.

Fig. S2

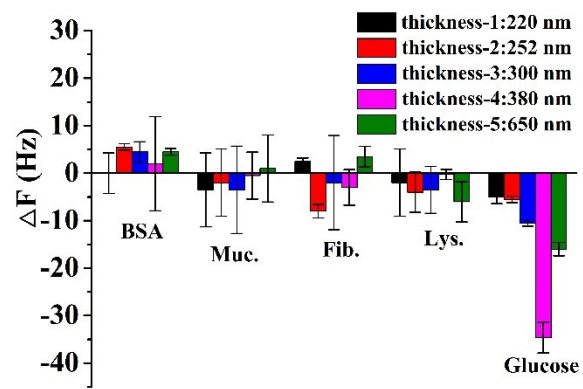


Fig. S2 Effect of thickness of HFSa on protein resistance and glucose sensitivity.

Fig. S3

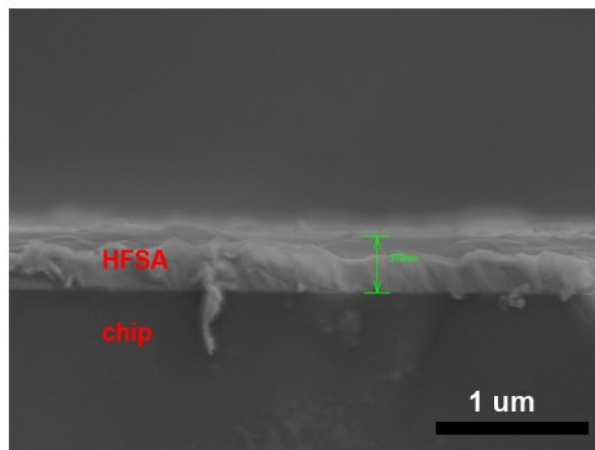


Fig. S3 The thickness of HFSA obtained by SEM.

Fig. S4

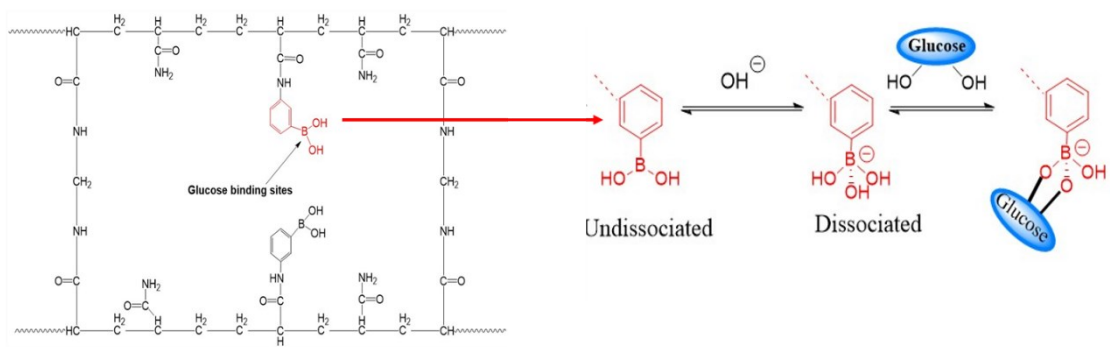


Fig. S4 The mechanism of glucose recognition between phenylboronic acid and glucose molecules

Fig. S5

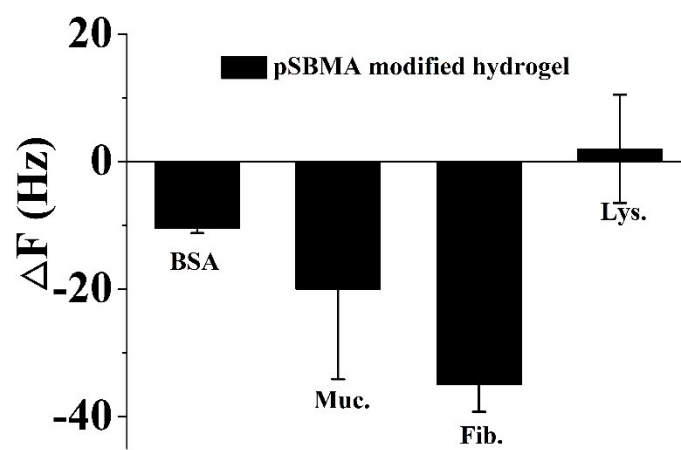


Fig. S5 The protein resistance of pSBMA modified hydrogel at polymerization time of 30 min.

Fig. S6

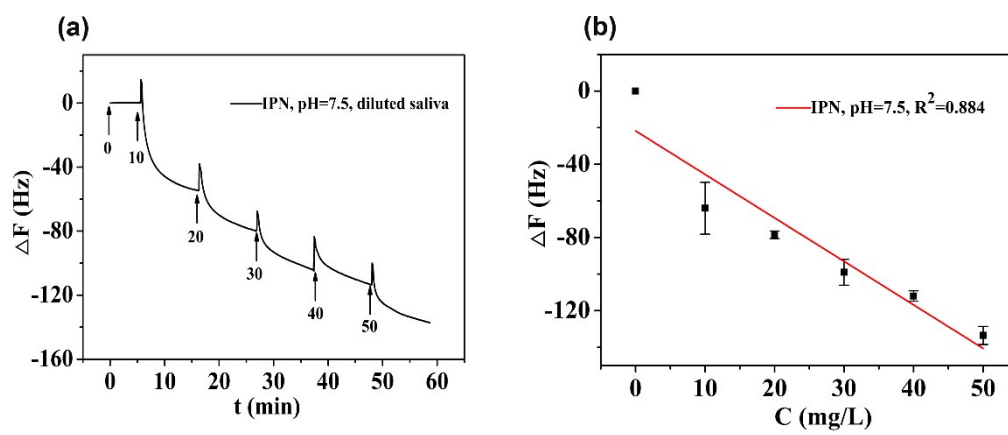


Fig. S6 (a) The detection of glucose in diluted saliva by IPN hydrogel film-coated QCM sensor. (b) Relationship between frequency shift and glucose concentration.

Fig. S7

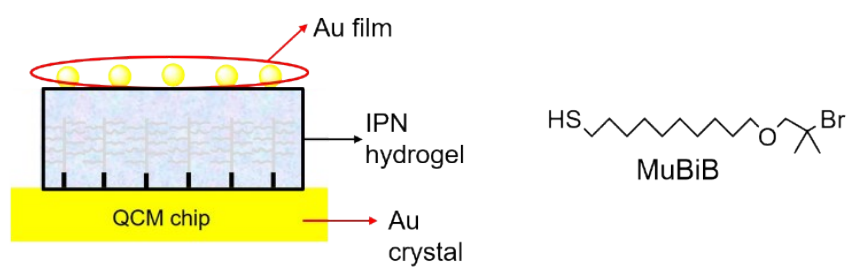


Fig. S7 Au film-coated IPN hydrogel

Fig. S8

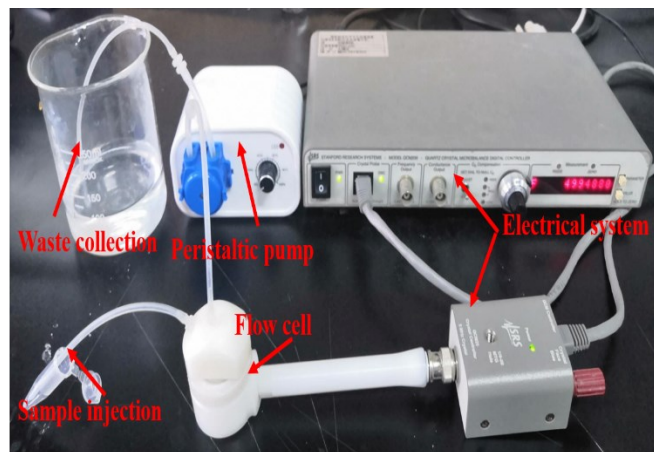


Fig. S8 The photograph of system

Table S1**Table S1.** Elemental surface composition of initiator on the IPN hydrogel and HFSA determined from XPS.

Sample	Element (atom %)						
	C	O	S	N	Br	B	Au
Initiator on the IPN hydrogel	40.963	7.83	2.882	-----	2.631	-----	48.575
HFSA	60.324	15.821	4.347	3.437	-----	-----	16.07

Table S2**Table S2.** The % RSD of IPN hydrogel film after eight association–dissociation cycles.

Film	Glucose level (mg/L)	Mean of glucose response (Hz)	Standard deviations	% RSD
IPN hydrogel	10	7.9	0.2	2.5%
HFSA	10	11.4	0.49	4.3%

Table S3**Table S3.** Elemental surface composition of IPN hydrogel determined from XPS.

Sample	Element (atom %)						
	C	O	S	N	Br	B	Au
IPN hydrogel	69.623	16.733	0.162	12.167	-----	1.315	-----

Table S4

Table S4 Comparison of analytical properties of the QCM sensor with previous reports for glucose detection

Glucose-responsive material	Detection range	Limit of detection	Response time	Specimen	Reference
PBA	10 – 5994 mg/L	-----	100 s	7.5 PBS	1
ConA	1.8 – 1350 mg/L	0.9 mg/L (3 δ)	-----	Distilled water, cattle serum	2
PBA	900 – 9000 mg/L	-----	-----	9.0 PBS	3
CP	1.8 – 3600 mg/L	-----	30 min	7.4 PBS, 10% human serum	4
PBA	1 – 36 mg/L	1 mg/L	5 min	7.5 PBS	5
PBA	0 – 50 mg/L	10 mg/L	2 min	7.5 PBS, 10% saliva, 1% diluted serum	6
PBA	0 – 160 mg/L	3 mg/L	10 min	PBS, artificial saliva	7
PBA	0 – 40 mg/L	5 mg/L	5 min	PBS, 50% saliva	8
PBA	0 – 50 mg/L	3 mg/L	5 min	PBS, 10% saliva	Our work

Reference:

1. Q. Dou, D. B. Hu, H. K. Gao, Y. M. Zhang, A. K. Yetisen, H. D. Butt, J. Wang, G. J. Nie and Q. Dai, *RSC Adv.*, 2017, 7, 41384-42390.
2. D. P. Tang, Q. F. Li, J. Tang, B. L. Su and G. N. Chen, *Anal. Chim. Acta.*, 2011, 686, 144-149.
3. C. Sugnaux, H. A. Klok, *Macromol. Rapid Commun*, 2014, 35, 1402-1407.
4. C. Li, X. Chen, F. Y. Zhang, X. X. He, G. Z. Fang, J. F. Liu and S. Wang, *Anal. Chem.*, 2017, 89, 10431-10438.
5. Z. Z. Zhang, Q. Dou, S. W. Wang, D. B. Hu, X. D. Guo, B. X. Liao, Z. P. Zhao, H. L. Liu and Q. Dai, *J. Mater. Chem. C*, 2020, 8, 9655-9662.
6. Z. Z. Zhang, Q. Dou, S. W. Wang, D. B. Hu, B. Yang, Z. P. Zhao, H. L. Liu, Q. Dai, *Nanoscale*, 2020, 12, 22787-22797.
7. Q. Dou, Z. F. Zhang, Y. X. Wang, S. W. Wang, D. B. Hu, Z. P. Zhao, H. L. Liu and Q. Dai, *ACS Appl. Mater. Interfaces*, 2020, 12, 34190-34197.
8. Q. Dou, S. W. Wang, Z. F. Zhang, Y. X. Wang, Z. P. Zhao, H. J. Guo, H. L. Liu and Q. Dai, *Nanoscale*, 2020, 12, 19317-19324.