# **Electronic Supplementary Information (ESI)**

### Continuous Amperometric Hydrogen Gas Sensing in Ionic Liquids<sup>†</sup>

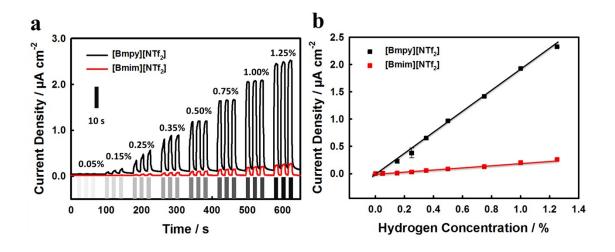
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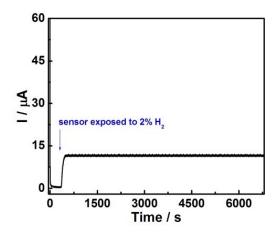
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Figure S1 summarized the IL-based amperometric hydrogen sensor responses to hydrogen when the gas sampling times is 10 s. Table S1 describes the different analytical methods for quantitative analysis of hydrogen concentrations. Figure S2~S4 detailed the three linear regression models based on the data sets of Figure 6, respectively. Figure S5~S6 and Table S2 summarized different hydrogen gas sampling times and the analytical figure of merits for hydrogen detection using the reported IL-based amperometric hydrogen sensor.



**Figure S1A.** (a) continuous sensing hydrogen using the amperometric hydrogen sensors in [Bmpy][NTf<sub>2</sub>] (black) and [Bmim][NTf<sub>2</sub>] (red), constant potential at E=+0.4 V vs. Fc<sup>+</sup>/Fc is the potential used for the hydrogen sensing, sampling time is 10 seconds for each measurement. (b) Calibration curves for concentration range in 0.05% ~ 1.25% hydrogen in two types of IL-based sensors at 10 second exposure time in Figure a.



**Figure S1B**. Constant potential chronoamperometric recording of sensing response toward 2% hydrogen exposure starting at the 300<sup>th</sup> second (marked by the downward arrow). Electrolyte: [Bmpy][NTf<sub>2</sub>]; flowrate: 100 sccm.

Method	Detection range	Speed of respond	Sensitivity	LOD/LOQ	Ref
Solid electrolyte amperometric H <sub>2</sub> sensor	10 ~ 40 ppm	< 1 min	N/A	10 ppm	21
Capacitive H <sub>2</sub> sensor	40 ~ 400 ppm	N/A	N/A	40 ppm	23
Optical H <sub>2</sub> sensor	0.5% ~ 4.0%	15 s	N/A	0.5%	22
Aqueous based electrochemical H <sub>2</sub> sensor	0.4% ~ 4.0%	<1s	0.16 µA/ppm	0.4%	27
Ionic liquid based electrochemical H <sub>2</sub> sensor	0.5% ~ 1.25%	<1s	3.08 µA cm <sup>-2</sup> % <sup>-1</sup>	0.5%	Current work

# Table S1. Parameters summary of different hydrogen sensors

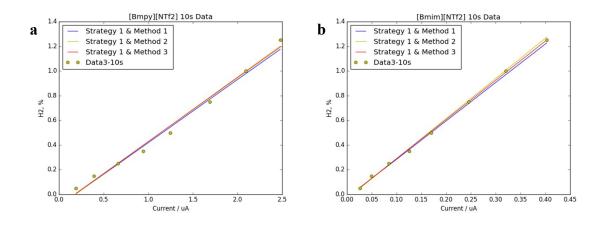
 Table S2. Data analysis approaches

Methods	Description	Training Data	Testing
			Data
Method 1	Train one model on Data1-10s and Data2-10s	Data1-10s, Data2-	Data3-10s
	and test on Data3-10s	10s	
Method 2	Train two models on Data1-10s and Data2-10s,	Data1-10s, Data2-	Data3-10s
	compute a new model based on the difference	10s	
	between the two models and the model for		
	Data2-10s.		
Method 3	Train one model on Data3-10s	Data3-10s	Data3-10s

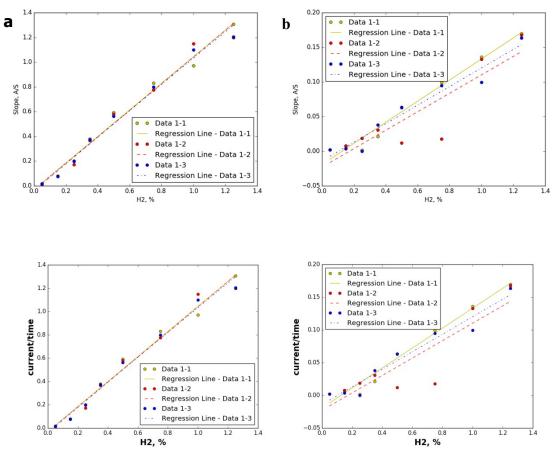
Strategy 1: Predicting concentration based on summit of current

Strategy 2: Predicting the concentration based on current/time

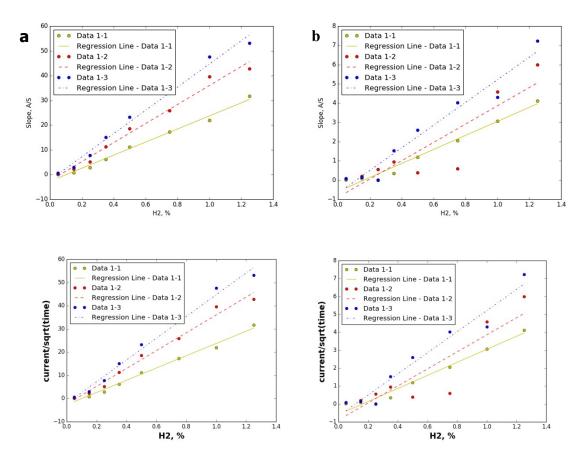
Strategy 3: Predicting the concentration based on (current/square root of time)



**Figure S2.** Curve for predicting hydrogen concentration based on summit of current by using strategy 1 in (a) [Bmpy][NTf<sub>2</sub>], (b) [Bmim][NTf<sub>2</sub>].



**Figure S3.** Curve for predicting hydrogen concentration based on current/time by using strategy 2 in (a) [Bmpy][NTf<sub>2</sub>], (b) [Bmim][NTf<sub>2</sub>].



**Figure S4.** Curve for predicting hydrogen concentration based on (current/square root of time) by using strategy 3 in (a) [Bmpy][NTf<sub>2</sub>]; (b) [Bmim][NTf<sub>2</sub>].

### Reference

1. Z. Wang, M. Guo, G. A. Baker, J. R. Stetter, L. Lin, A. J. Mason and X. Zeng, *Analyst*, 2014, **139**, 5140-5147.

2. Y. A. Tang, X. W. Chi, S. Z. Zoub and X. Q. Zeng, *Nanoscale*, 2016, **8**, 5771-5779.