

# ChemSusChem

Supporting Information

## **Non-Isocyanate Poly(hydroxyurethane)-Based Networks as Solid Polymer Electrolytes for Lithium Metal Batteries**

Ashish Raj, Bruno Grignard, Christophe Detrembleur, and Jean-François Gohy\*

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### Non-isocyanate poly(hydroxyurethane)-based networks as solid polymer electrolytes for lithium metal batteries

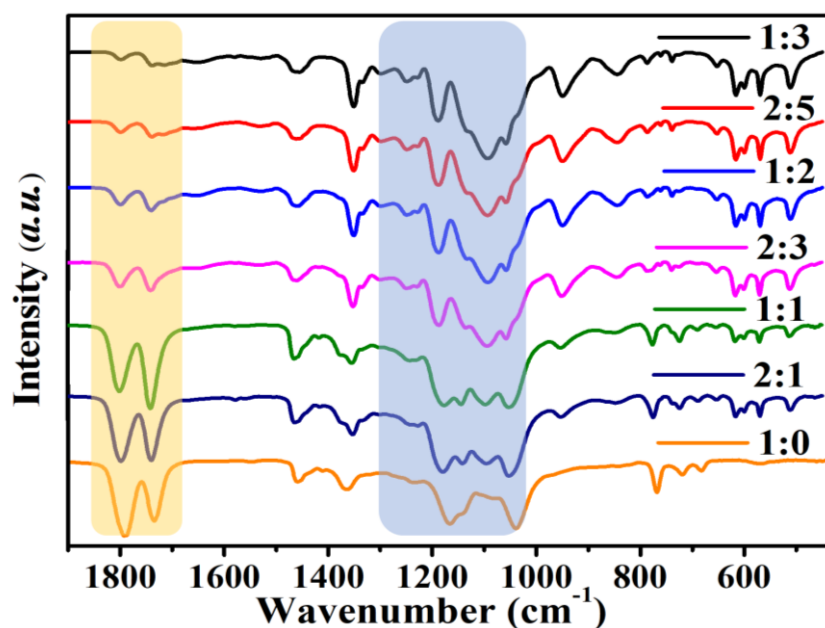
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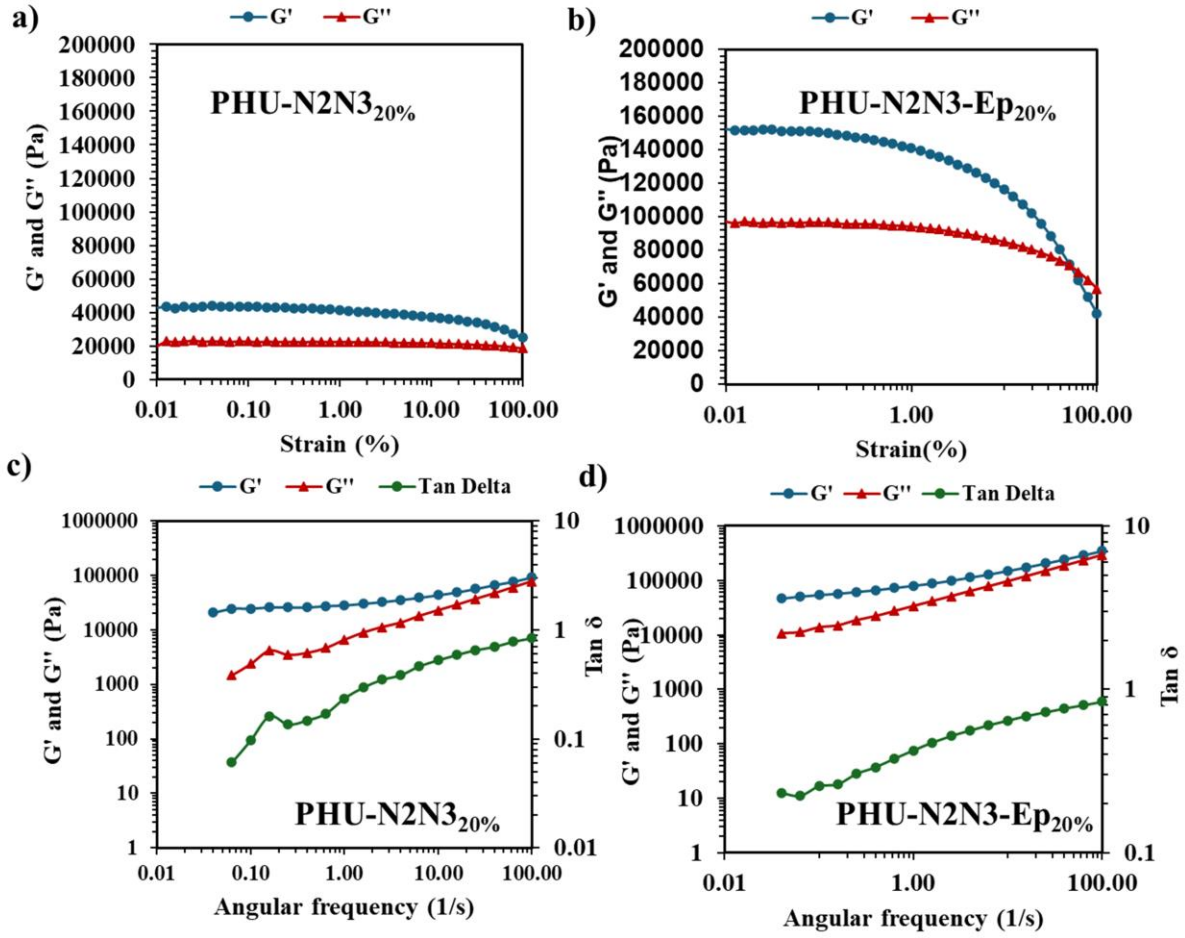
[b] Center for Education and Research on Macromolecules (CERM), CESAM Research Unit,  
University of Liège,  
Allée du 6 août, Building B6A, 4000 Liège, Belgium

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University of Liège,  
13 Allée du 6 août, Building B6A, 4000 Liège, Belgium

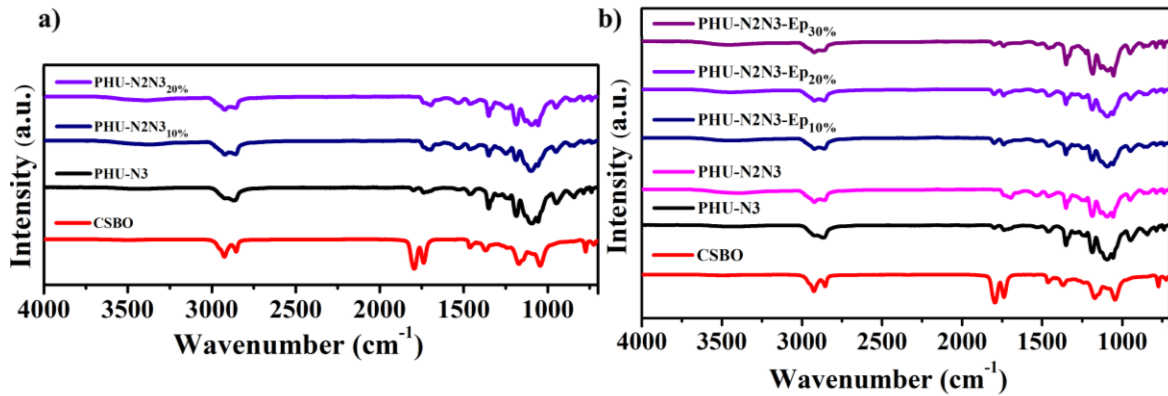
[d] WEL Research Institute, Avenue Pasteur, 6, 1300 Wavre, Belgium



**Figure S1:** IR spectra of PHU-N3 with respective formulations y:z corresponding to CSBO:N3 ratio in mol/mol.



**Figure S2:** Assessment of mechanical properties of PHU-N2N3 a) and c) and PHU-N2N3-Ep, b) and d) by rotational rheology. a) and b): strain sweep test at a constant angular frequency of 10/s at 21°C. c) and d): frequency sweep test at a constant strain of 0.1% at 21°C.



**Figure S3:** IR spectra of PHU-N2N3, a) and PHU-N2N3-Ep b) with respective formulations in comparison to PHU and CSBO.

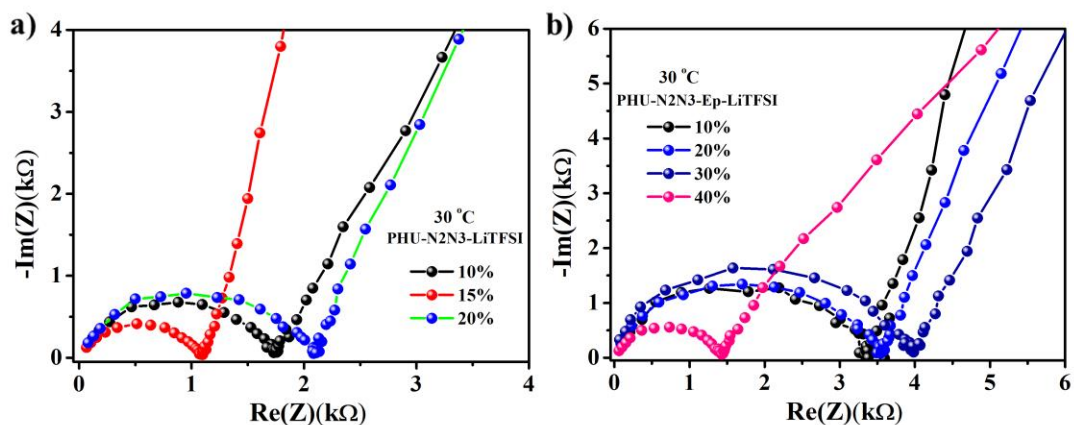


Figure S4: Nyquist plots of SS|PHU-SPE|SS symmetric cell at 30 °C: a) PHU-N2N3-LiTFSI and b) PHU-N2N3-Ep-LiTFSI.

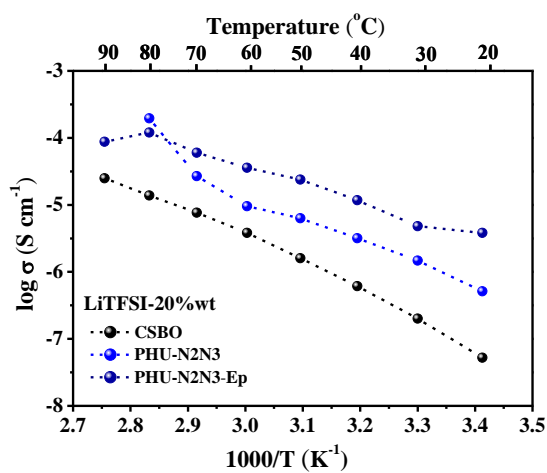


Figure S5: Variable temperature conductivity of PHUs in comparison to CSBO with 20 wt% of LiTFSI.

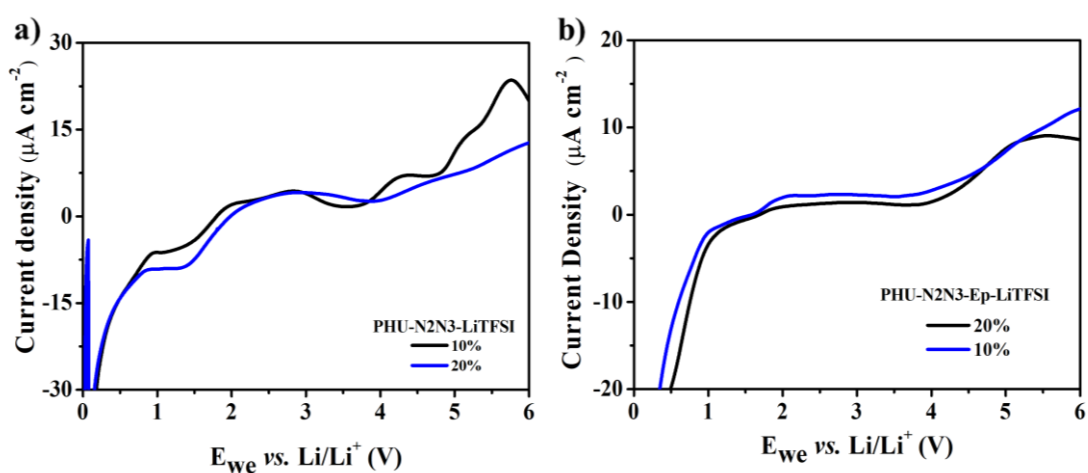
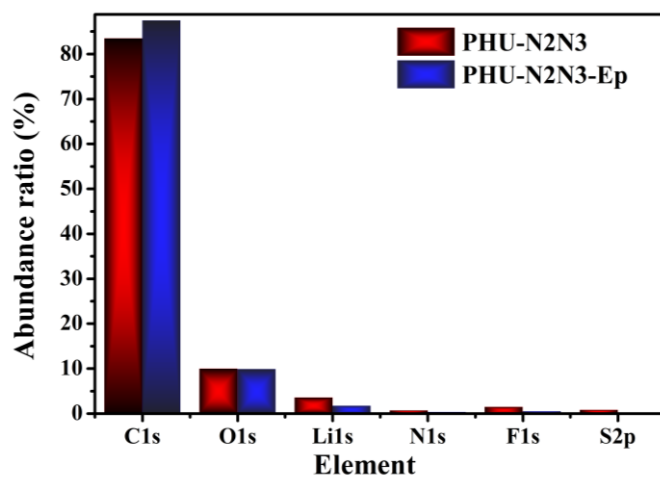
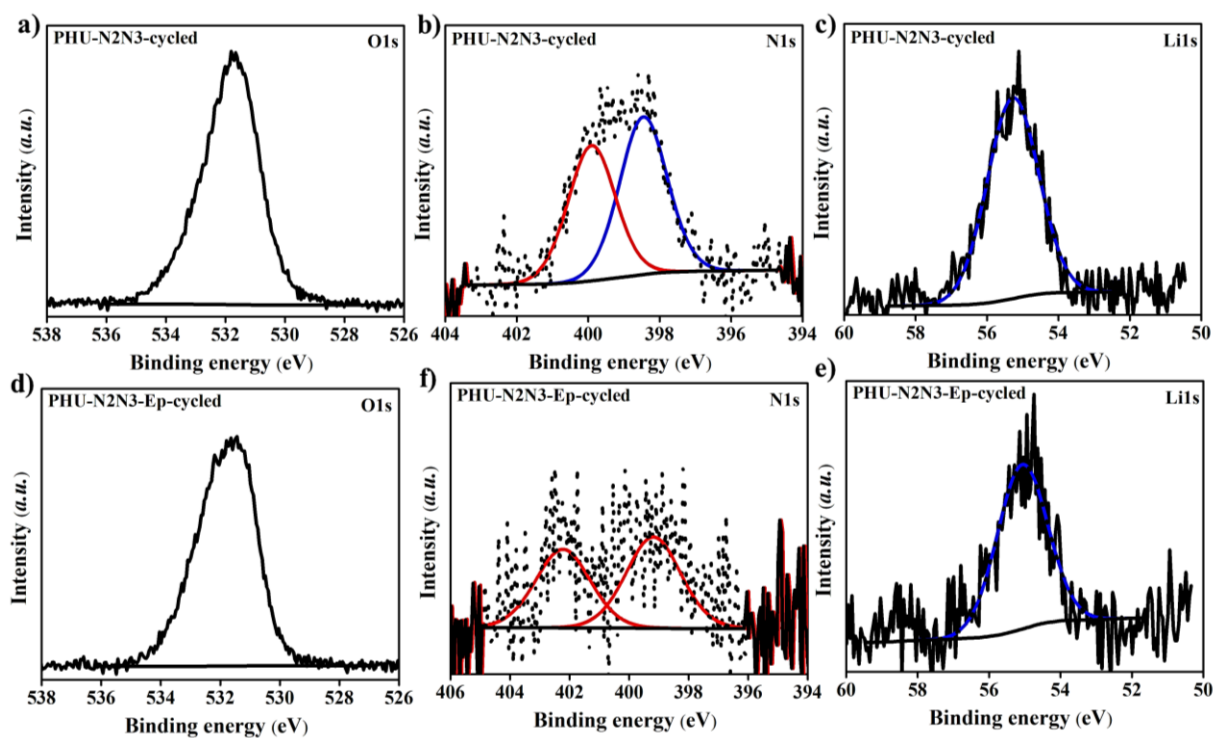


Figure S6: Linear sweep voltammetry at 60 °C of a) PHU-N2N3-LiTFSI and b) PHU-N2N3-Ep-LiTFSI.



**Figure S7:** Bar chart of abundance ratio % as result of quantification via XPS on cycled PHU SPE membranes with Li-Li symmetric cells for stripping and plating.



**Figure S8:** HR-XPS of a) and d) oxygen (O1s), b) and e) nitrogen (N1s), c) and f) lithium (L1s) of cycled PHU SPE membranes after stripping and plating with Li-Li cells.

**Table S1:** Quantification table of XPS analysis of cycled PHU SPE membranes indicating, binding energy (BE), full width at half maximum (FWHM), and abundance ratio (A%).

		F 1s			O 1s	N 1s			C 1s					S 2p <sub>3/2</sub>	Li 1s
		F-C	F-Li	F total		NH <sup>+</sup> -(C,H)	N-(C,H)	N total	O-C=O/C O <sub>3</sub> <sup>-2</sup>	O-C-O/C=O	C-(O,N)	C-(C,H)	C total		
PHU-N2N3-film-cycled	BE	688.2	684.6		531.8	399.9	398.5		288.8	287.9	286.3	284.9			55.3
	FWHM	1.8	1.8		2.1	1.6	1.6		1.3	1.3	1.3	1.3			1.7
	A%	1.2	0.2	1.5	9.9	0.3	0.4	0.7	3.0	1.2	7.3	72.0	83.5	0.8	3.6
PHU-N2N3-Ep-film-cycled	BE	688.2	684.5		531.8	399.8	398.4		288.5	287.8	286.4	284.8			55.2
	FWHM	1.9	1.9		2.1	1.4	1.4		1.2	1.2	1.2	1.2			1.6
	A%	1.0	0.2	1.3	7.2	0.3	0.3	0.6	2.1	0.2	6.0	79.3	87.6	0.6	2.7

