

Supporting information

Correlation between Ionic Conductivity and Mechanical Properties of Solid-like PEO Polymer Electrolyte

Agathe Naboulsi^{1,2,3}, Ronan Chometon^{2,3,4}, François Ribot², Giao Nguyen¹, Christel Laberty-Robert^{2,3}, Odile Fichet¹

¹CY Cergy Paris Université, LPPI, F-95000 Cergy, France

²Sorbonne Université, CNRS, Laboratoire Chimie de la Matière Condensée de Paris, LCMCP, 4 Place Jussieu, 75005 Paris, France

³RS2E, Réseau Français sur le Stockage Electrochimique de l'Energie, CNRS 3459, 80039 Cedex 1 Amiens, France

⁴CSE, Collège de France, 4 Place Marcellin Berthelot, 75005 Paris, France

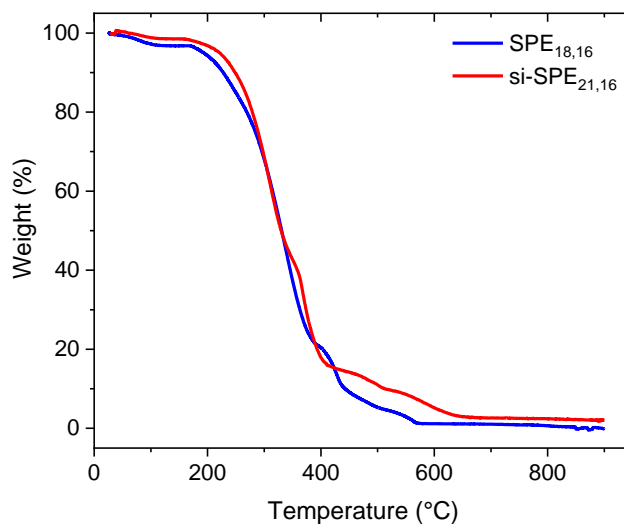


Figure SI-1. Thermogravimetric analysis of SPE_{18,16} and si-SPE_{21,16} under air atmosphere at 10°C/min.

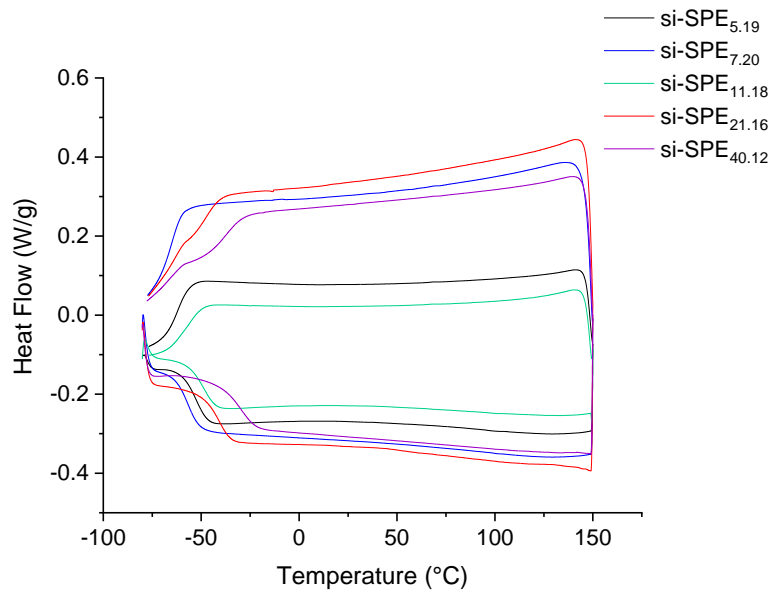


Figure SI-2. First DSC scans thermograms recorded on different si-SPE samples showing the absence of polymerization exotherm

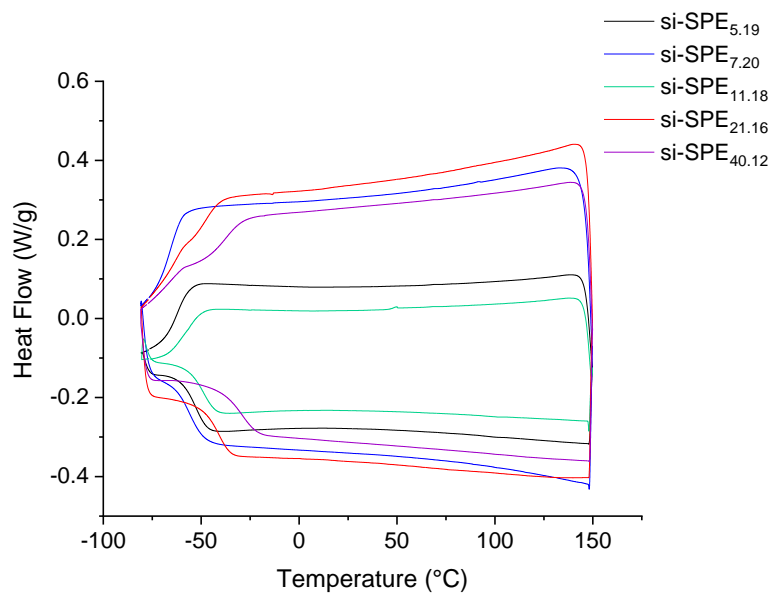


Figure SI-3. Second DSC scans recorded on different si-SPE samples showing the absence of crystallization exotherm

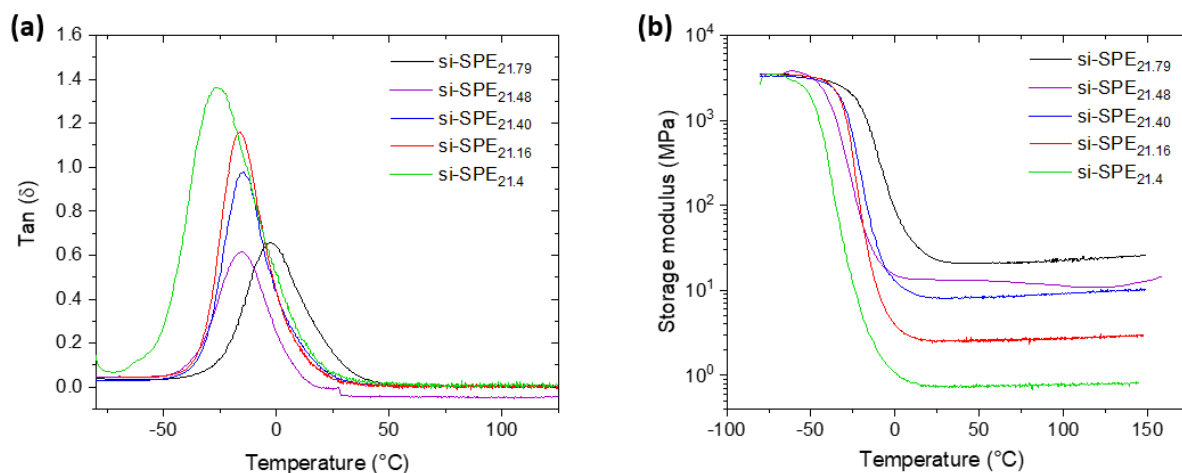


Figure SI-4. $\tan\delta$ (a) and storage modulus (E') (b) as function of temperature for various *si-SPE*_{21,y} materials showing the absence of phase separation.

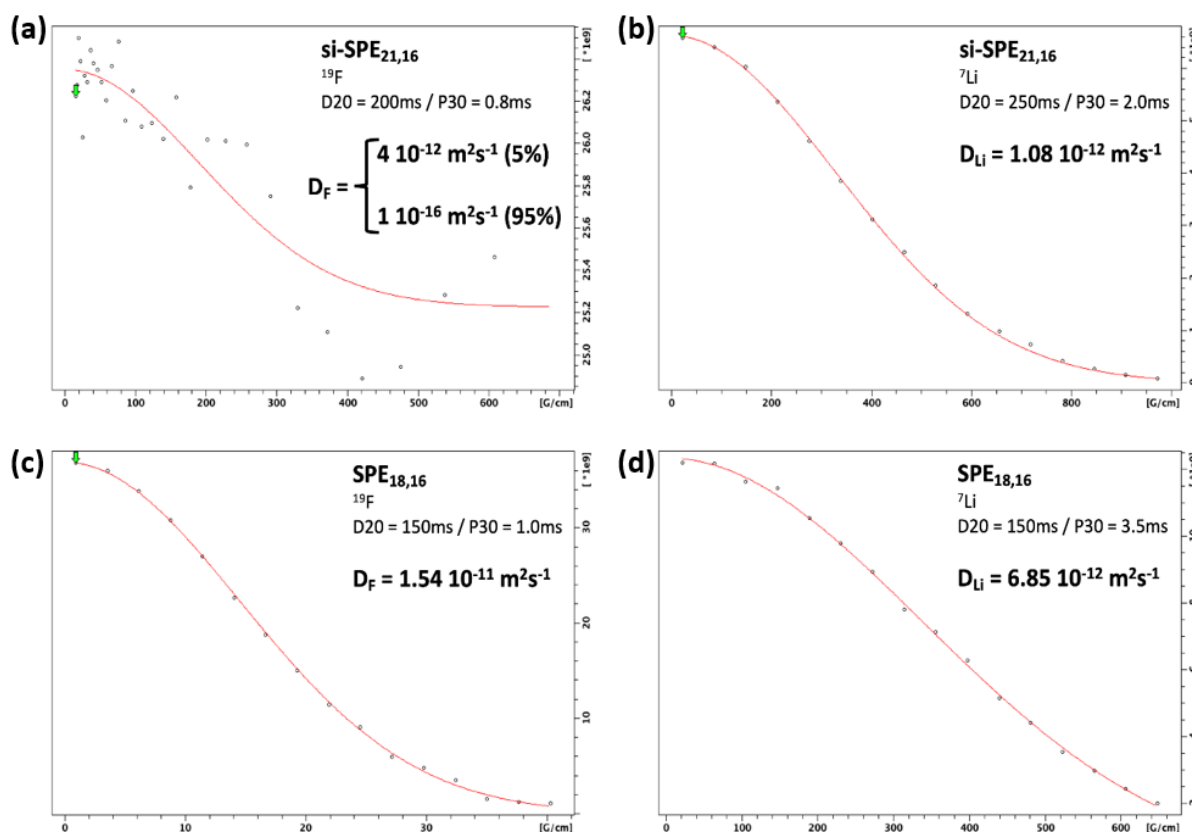


Figure SI-5. NMR experiments: Attenuation curves for (a), (c) ¹⁹F signal and (b), (d) ⁷Li signal for *si-SPE*_{21,16} and *SPE*_{18,16} respectively.

The attenuation curve observed for the ¹⁹F signal in *si-SPE*_{21,16} (Figure SI-5a) shows almost no decay (~5%). It was yet fitted with 2 components to account for this weak decay. The major component corresponds to a diffusion coefficient that was set at $1.0 \times 10^{-16} \text{ m}^2 \cdot \text{s}^{-1}$. It is associated to "immobile" species. The minor component corresponds to a diffusion coefficient of $\sim 4 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$. This small amount of weakly mobile species could correspond to short oligomers or ends of chains.

Table SI-6. Comparison of lithium-ion transport number values for single-ion conducting electrolytes reported in literature.

Authors/Ref	Polymer	Solvent	t^+
Nguyen and al. ¹	Li-PVDF	DMF	0.87 ± 0.3
Bouchet and al. ²	P(STFSiLi)-b-PEO-b-P(STFSiLi)	Dioxane	0.85
Porcarelli and al. ³	LiMTFSI-PEGM-PEGDM	PC	0.84-0.91
Borzutzki and al. ⁴	Polysulfonamide (with -C(CF ₃)) / PVdF-HFP	THF	0.9
Porcarelli and al. ⁵	poly(PEGM)-b-poly(LiMTFSI)	DMF	0.83
Feng and al. ⁶	Li[PSTFSI-co-MPEGA]	DMF or MeOH	0.94
Rohan and al. ⁷	MTF-Li/PVDF-HFP	DMF	0.88
Zhang and al. ⁸	lithium poly (bisphenol A borate) / PEO	MeCN	0.92-0.96
Humbeck and al. ⁹	Tetraarylborate polymer	DMSO, DMF, MeOH	0.89-0.93
Rolland and al. ¹⁰	PS-b-P(MALi-co-POEGMA)	THF/MeOH	0.84
Pan and al. ¹¹	Lithiated poly(bis(4-carbonyl benzene sulfonyl)imide-co-bis(4-amino benzene sulfonyl)imide) / PVDF-HFP	NMP	0.92
Xu and al. ¹²	lithium poly (glutaric acid aluminate)	DMF	0.8
Shi and al. ¹³	PEO-PFSiLi	MeCN	0.9
Inceoglu and al. ¹⁴	PEO-PSLiTFSI	DMF	0.95
Nguyen and al. ¹⁵	phenylsulfonyl(trifluoromethylsulfonyl)imide lithium salt / PEO	MeCN/H ₂ O	0.90-0.92
Li and al. ¹⁶	Lithiated mimic neurons in PEGMA	Acetone	0.974

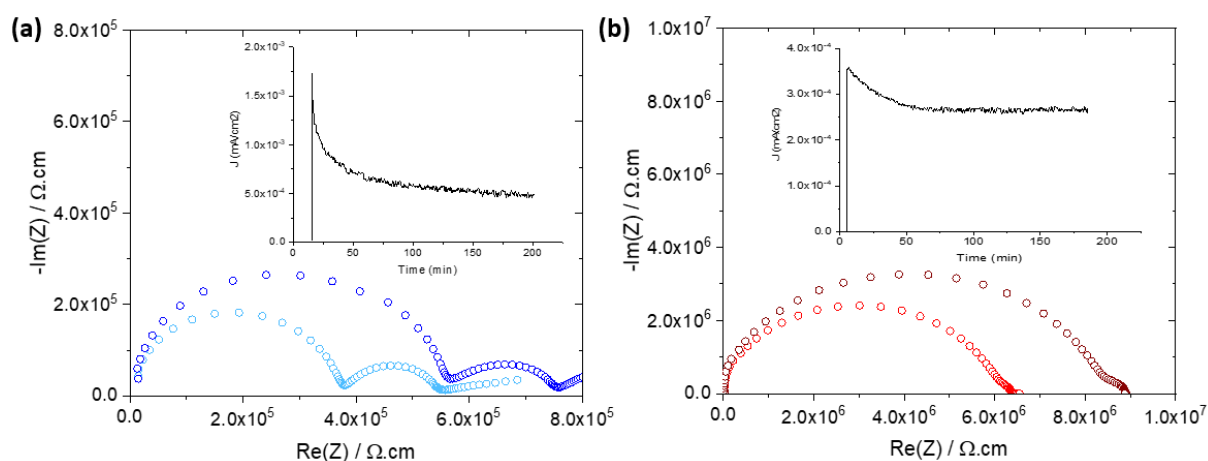


Figure SI-7. Determination of the Li transport numbers by Bruce and Vincent method of (a) Li/SPE_{18,16}/Li and (b) Li/si-SPE_{21,16}/Li with polarization experiment and EIS spectra before (red) and after (black) polarization.

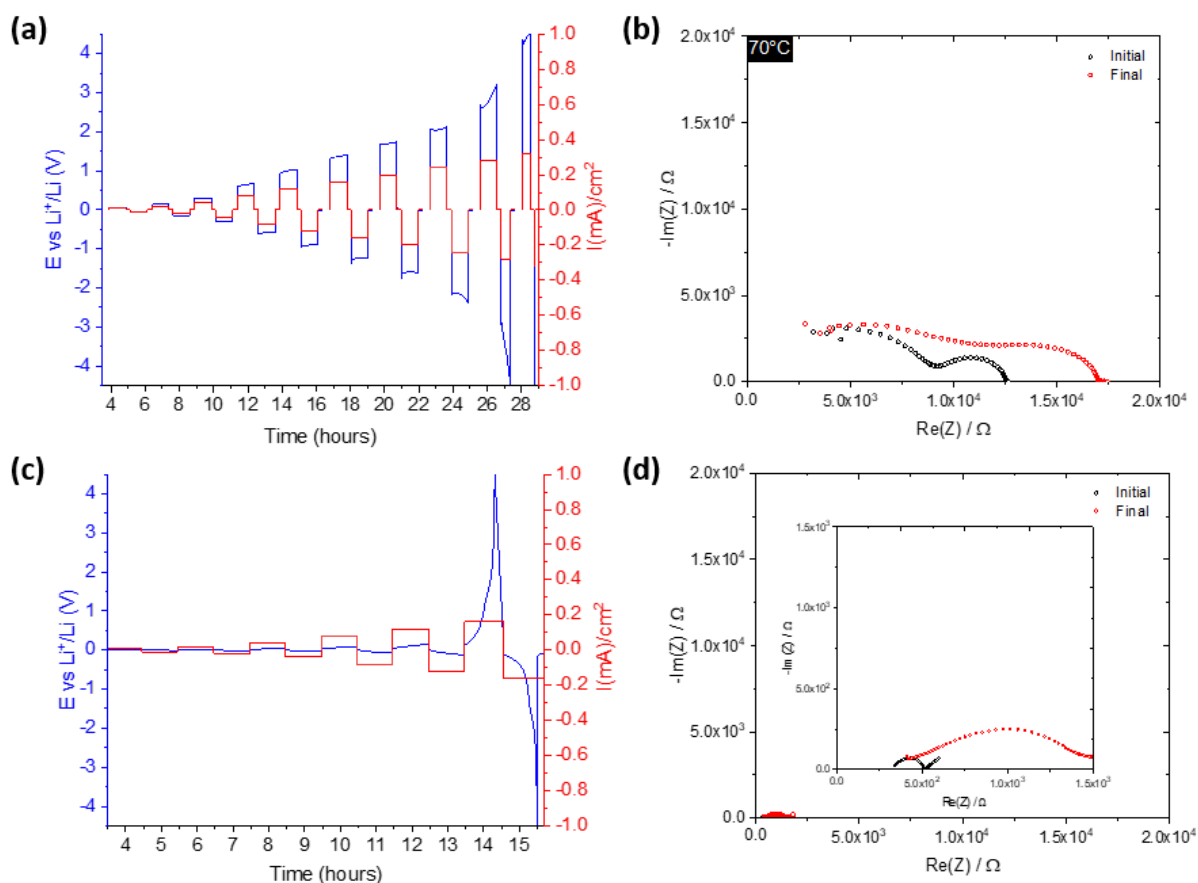


Figure SI-8. CCD determination at 70°C of (a) Li/si-SPE_{21,16}/Li and (c) Li/SPE_{18,16}/Li and (b), (d) Nyquist plots of respective electrolyte before CCD determination (black) and after CCD determination (red) cycling.

The starting current density was 0.01 mA cm⁻² and the current was subsequently doubled gradually to 0.08 mA/cm² and after ramped by 0.04 mA/cm² steps. The plating and stripping were performed respectively for 1h and separated by a rest period of 5 min.

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