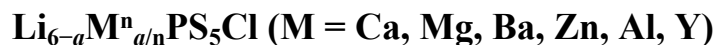


*Supporting Information*

**AC Conductivity Study of Mechanochemical Synthesized Solid Electrolytes of**



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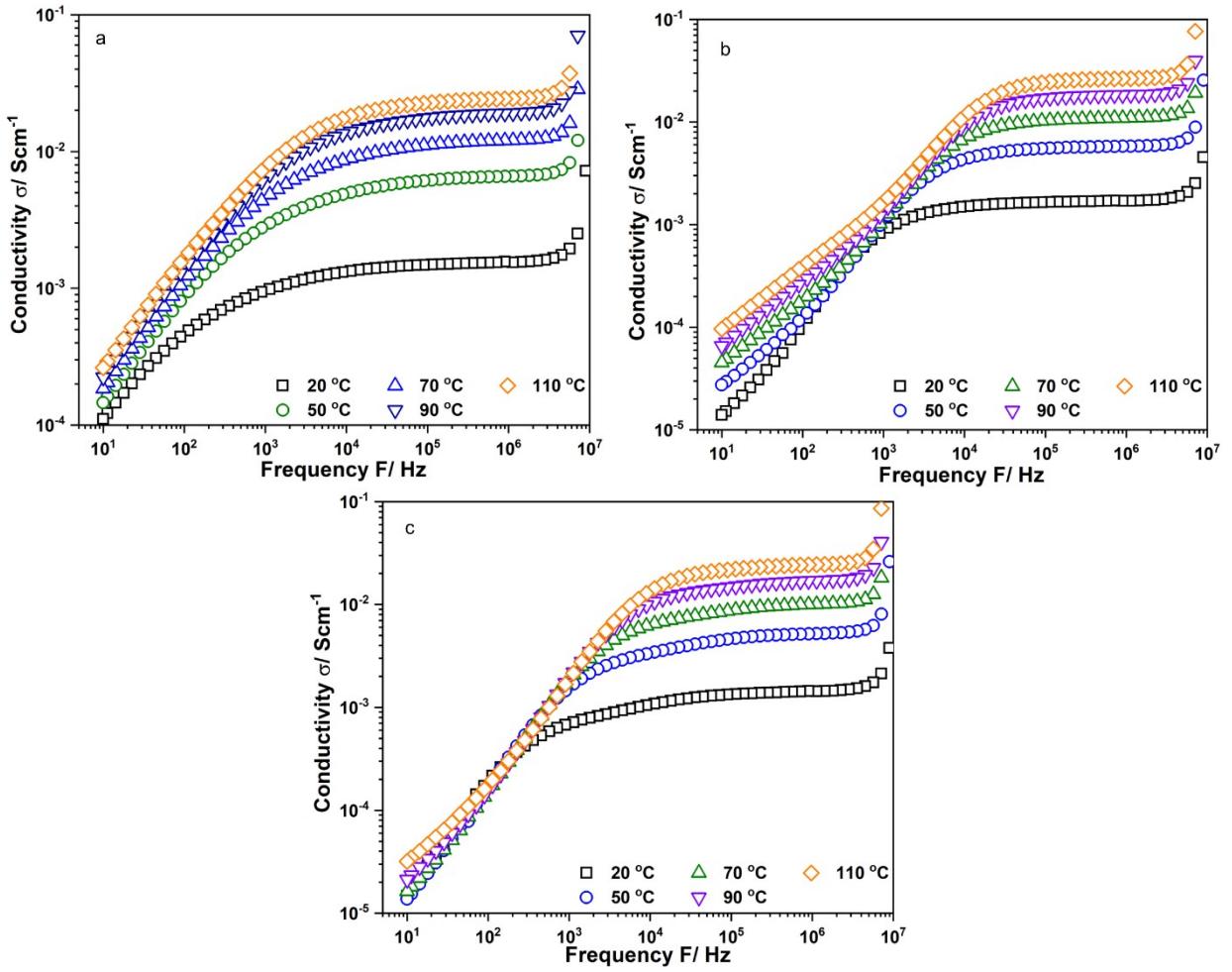


Figure S1: Conductivity isotherms of (a)  $\text{Li}_{5.96}\text{Ca}_{0.002}\text{PS}_5\text{Cl}$ ; (b)  $\text{Li}_{5.9}\text{Ca}_{0.05}\text{PS}_5\text{Cl}$  and (c)  $\text{Li}_{5.8}\text{Ca}_{0.1}\text{PS}_5\text{Cl}$  measured from 10 Hz to 9 MHz.

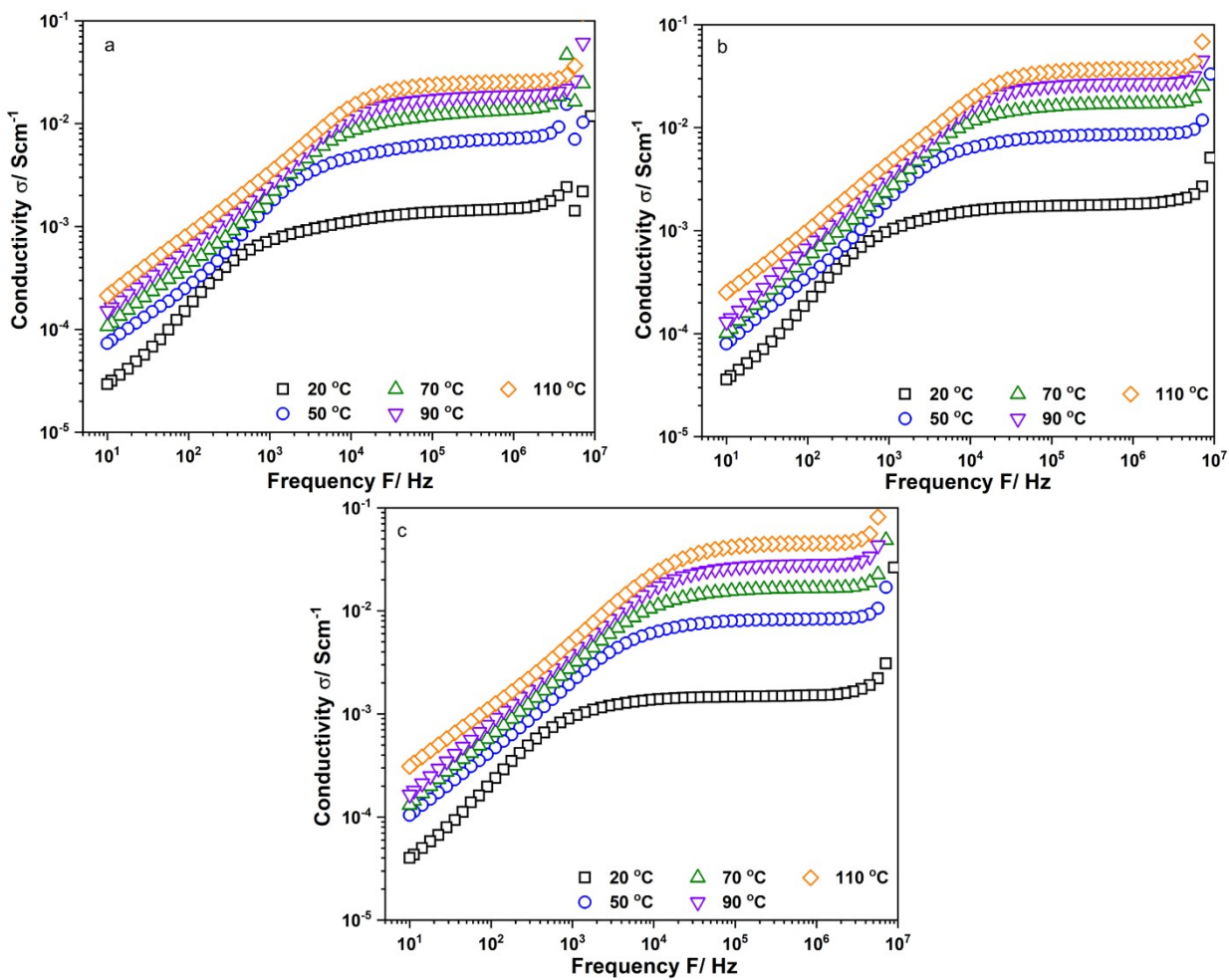


Figure S2: Conductivity isotherms of (a)  $\text{Li}_{5.94}\text{Ba}_{0.03}\text{PS}_5\text{Cl}$ ; (b)  $\text{Li}_{5.94}\text{Zn}_{0.03}\text{PS}_5\text{Cl}$  and (c)  $\text{Li}_{5.94}\text{Y}_{0.02}\text{PS}_5\text{Cl}$  measured from 10 Hz to 9 MHz.

Table S1: The values of A and n in  $\sigma = \sigma_{DC} + A\omega^n$

	A					n				
	20 °C	50 °C	70 °C	90 °C	110 °C	20 °C	50 °C	70 °C	90 °C	110 °C
Li <sub>6</sub> PS <sub>5</sub> Cl	9.41x10 <sup>-6</sup>	8.26x10 <sup>-5</sup>	5.54x10 <sup>-5</sup>	3.63x10 <sup>-5</sup>	4.92x10 <sup>-5</sup>	0.241	0.1442	0.1594	0.2044	0.2122
Li <sub>5.96</sub> Ca <sub>0.002</sub> PS <sub>5</sub> Cl	7.03x10 <sup>-5</sup>	2.22x10 <sup>-3</sup>	3.39x10 <sup>-3</sup>	4.59x10 <sup>-3</sup>	3.73x10 <sup>-3</sup>	0.106	0.045	0.0592	0.0571	0.0663
Li <sub>5.94</sub> Ca <sub>0.03</sub> PS <sub>5</sub> Cl	3.77x10 <sup>-4</sup>	2.56x10 <sup>-3</sup>	3.98x10 <sup>-3</sup>	3.40x10 <sup>-3</sup>	5.35x10 <sup>-3</sup>	0.0528	0.0394	0.0532	0.088	0.0543
Li <sub>5.9</sub> Ca <sub>0.05</sub> PS <sub>5</sub> Cl	2.46x10 <sup>-4</sup>	1.90x10 <sup>-3</sup>	1.55x10 <sup>-3</sup>	3.08x10 <sup>-3</sup>	5.06x10 <sup>-3</sup>	0.0478	0.0341	0.0913	0.0669	0.0629
Li <sub>5.8</sub> Ca <sub>0.1</sub> PS <sub>5</sub> Cl	6.1x10 <sup>-5</sup>	8.64x10 <sup>-4</sup>	1.10x10 <sup>-3</sup>	2.08x10 <sup>-3</sup>	1.54x10 <sup>-3</sup>	0.105	0.0537	0.072	0.065	0.104
Li <sub>5.94</sub> Ba <sub>0.03</sub> PS <sub>5</sub> Cl	1.90x10 <sup>-4</sup>	1.19x10 <sup>-3</sup>	2.00x10 <sup>-3</sup>	4.12x10 <sup>-3</sup>	6.33x10 <sup>-3</sup>	0.0887	0.0914	0.1062	0.0711	0.0543
Li <sub>5.94</sub> Zn <sub>0.03</sub> PS <sub>5</sub> Cl	4.86x10 <sup>-5</sup>	2.06x10 <sup>-3</sup>	3.95x10 <sup>-3</sup>	6.74x10 <sup>-3</sup>	5.84x10 <sup>-3</sup>	0.1346	0.0405	0.0414	0.036	0.07
Li <sub>5.94</sub> Mg <sub>0.03</sub> PS <sub>5</sub> Cl	3.91x10 <sup>-5</sup>	1.4x10 <sup>-4</sup>	1.02x10 <sup>-4</sup>	9.79x10 <sup>-4</sup>	3.04x10 <sup>-3</sup>	0.1109	0.1152	0.0699	0.1213	0.0895
Li <sub>5.94</sub> Al <sub>0.02</sub> PS <sub>5</sub> Cl	1.43x10 <sup>-5</sup>	1.65x10 <sup>-4</sup>	6.25x10 <sup>-5</sup>	2.12x10 <sup>-5</sup>	1.40x10 <sup>-5</sup>	0.1988	0.1425	0.234	0.3356	0.3868
Li <sub>5.94</sub> Y <sub>0.02</sub> PS <sub>5</sub> Cl	1.13x10 <sup>-4</sup>	1.83x10 <sup>-3</sup>	4.34x10 <sup>-3</sup>	8.68x10 <sup>-3</sup>	1.55x10 <sup>-2</sup>	0.0744	0.0434	0.0401	0.0371	0.0327

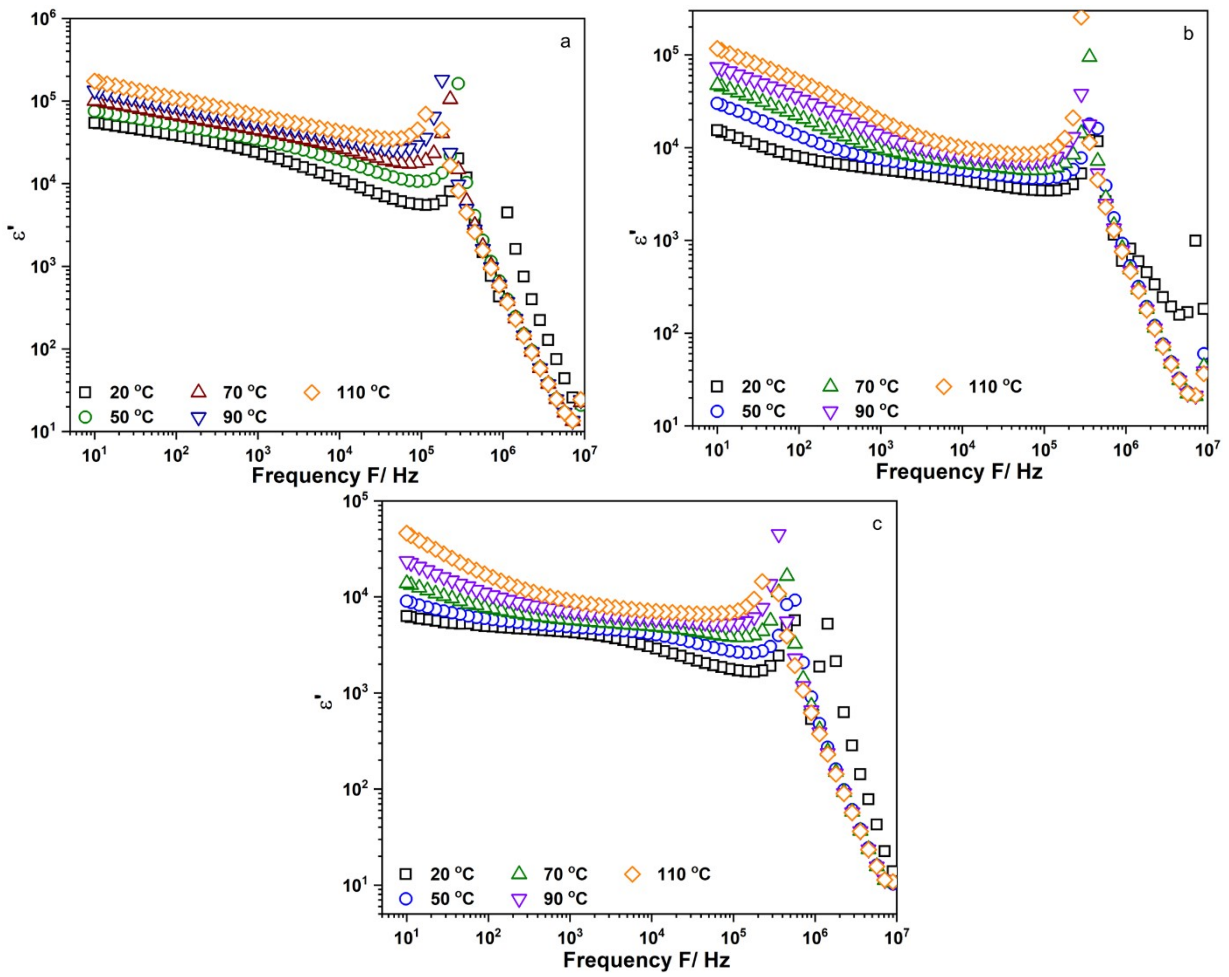


Figure S3: Frequency dependent of the real part of permittivity,  $\epsilon'$ , of (a)  $\text{Li}_{5.96}\text{Ca}_{0.002}\text{PS}_5\text{Cl}$ ; (b)  $\text{Li}_{5.9}\text{Ca}_{0.05}\text{PS}_5\text{Cl}$  and (c)  $\text{Li}_{5.8}\text{Ca}_{0.1}\text{PS}_5\text{Cl}$  measured from 10 Hz to 9 MHz.

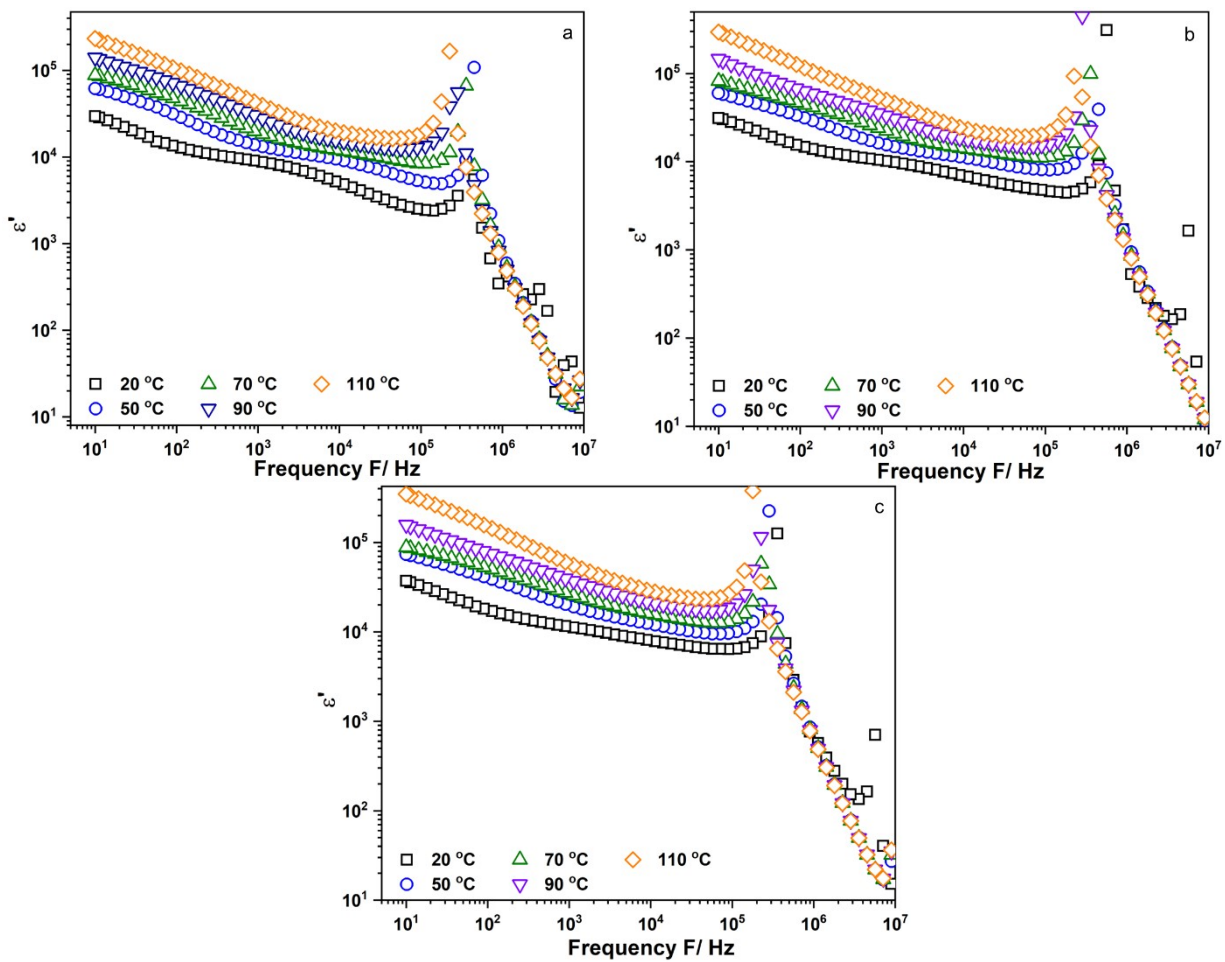


Figure S4: Frequency dependent of the real part of permittivity,  $\epsilon'$ , of (a)  $\text{Li}_{5.94}\text{Ba}_{0.03}\text{PS}_5\text{Cl}$ ; (b)  $\text{Li}_{5.94}\text{Zn}_{0.03}\text{PS}_5\text{Cl}$  and (c)  $\text{Li}_{5.94}\text{Y}_{0.02}\text{PS}_5\text{Cl}$  measured from 10 Hz to 9 MHz.

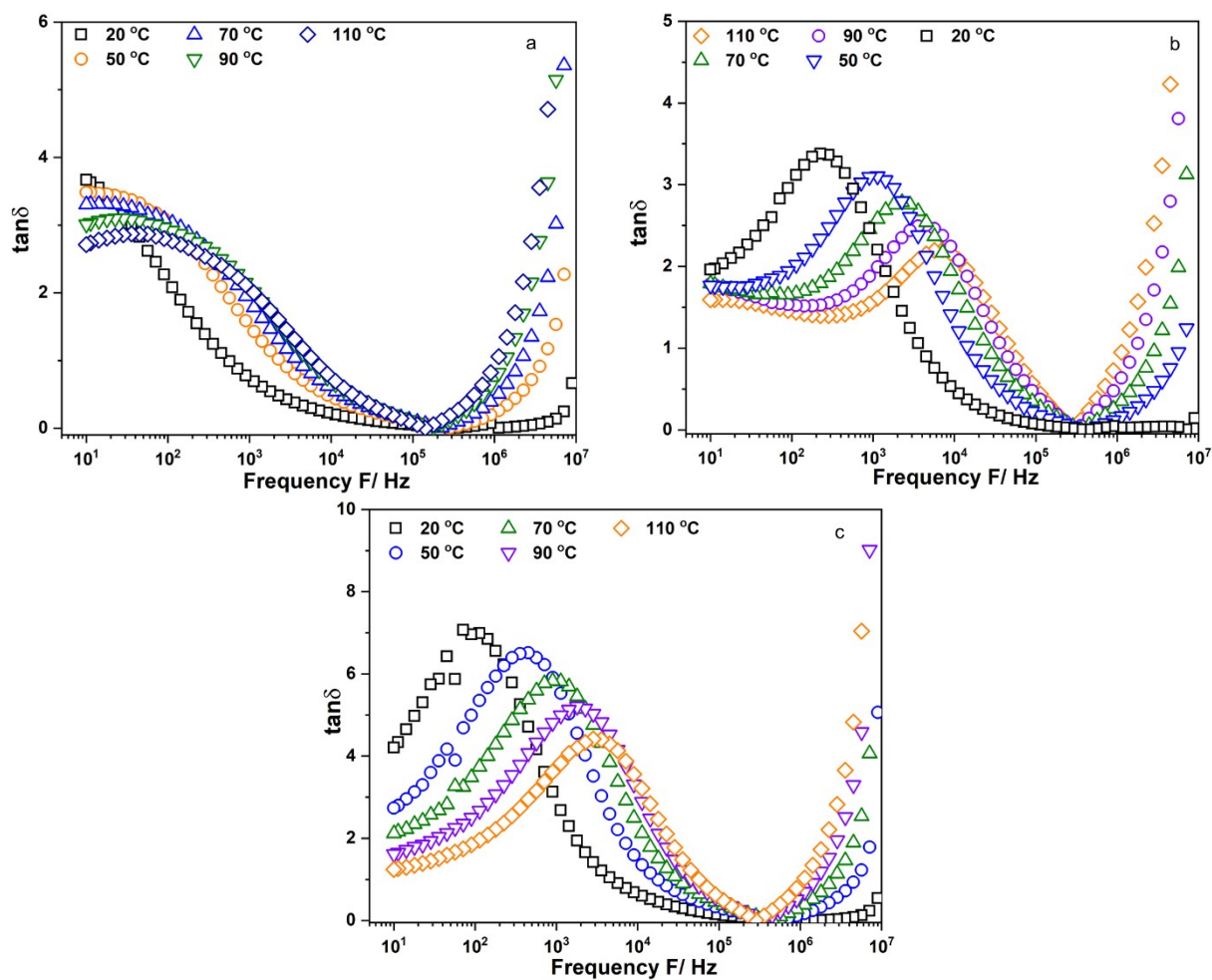


Figure S5: Frequency dependent of the loss factor,  $\tan\delta$ , of (a)  $\text{Li}_{5.96}\text{Ca}_{0.002}\text{PS}_5\text{Cl}$ ; (b)  $\text{Li}_{5.9}\text{Ca}_{0.05}\text{PS}_5\text{Cl}$  and (c)  $\text{Li}_{5.8}\text{Ca}_{0.1}\text{PS}_5\text{Cl}$  measured from 10 Hz to 9 MHz.

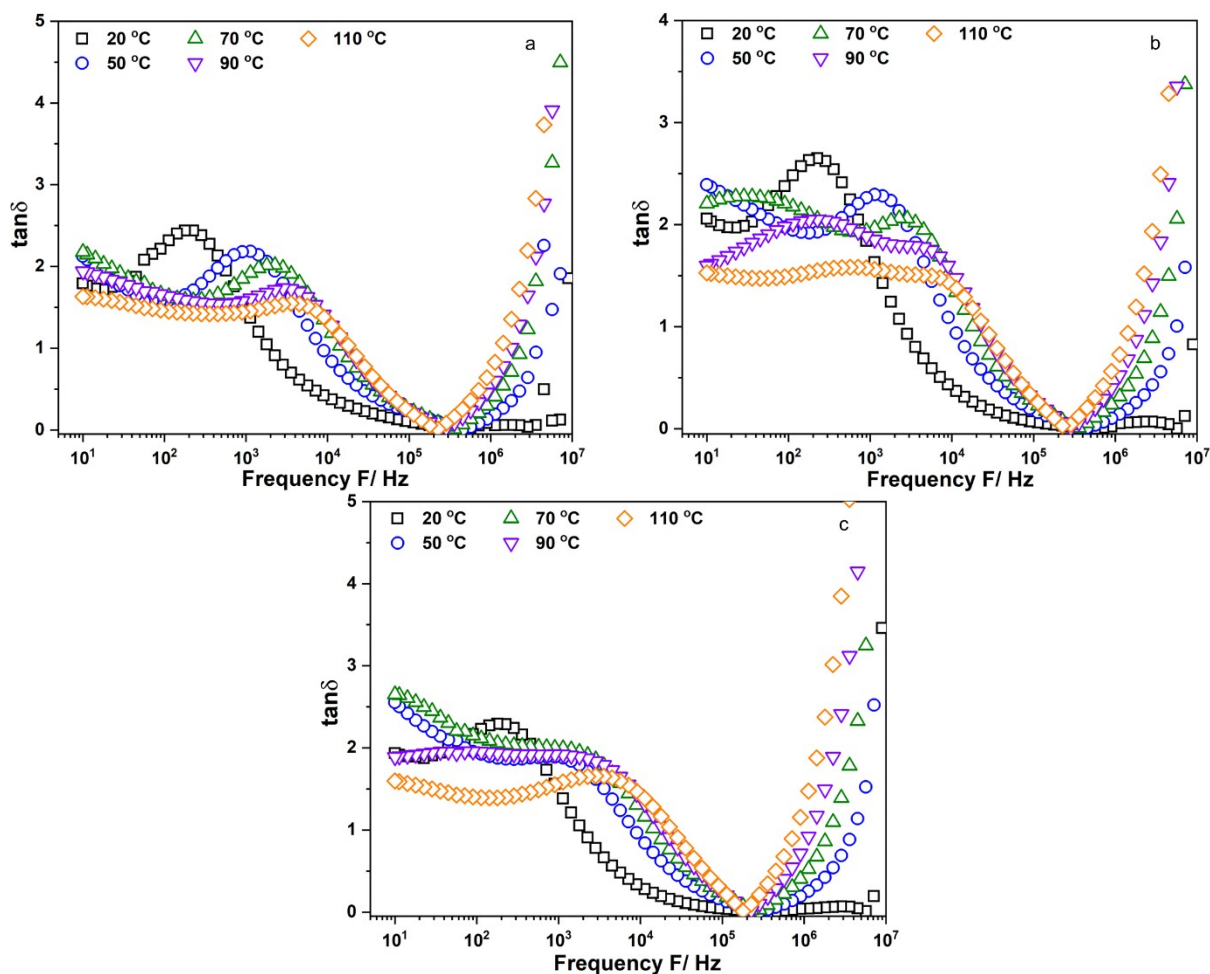


Figure S6: Frequency dependent of the loss factor,  $\tan\delta$ , of (a)  $\text{Li}_{5.94}\text{Ba}_{0.03}\text{PS}_5\text{Cl}$ ; (b)  $\text{Li}_{5.94}\text{Zn}_{0.03}\text{PS}_5\text{Cl}$  and (c)  $\text{Li}_{5.94}\text{Y}_{0.02}\text{PS}_5\text{Cl}$  measured from 10 Hz to 9 MHz.