

**The role of nickel-iron based layered double hydroxide on crystallinity, ion conductivity, thermal and mechanical properties on poly(ethylene-oxide) solid electrolyte**

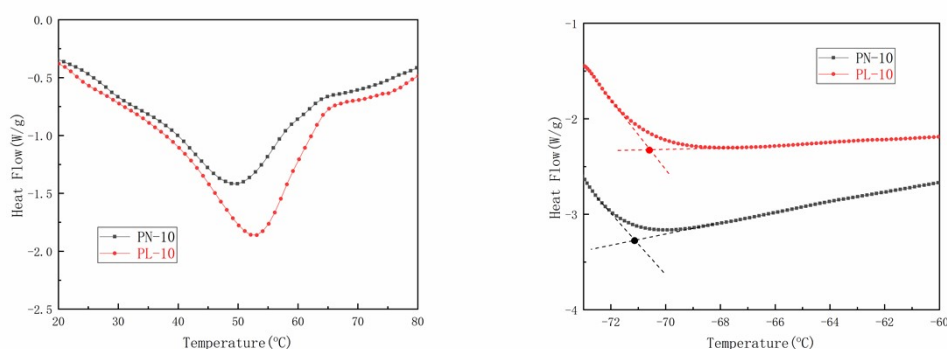
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## Synthesis of lithium montmorillonite (LM)

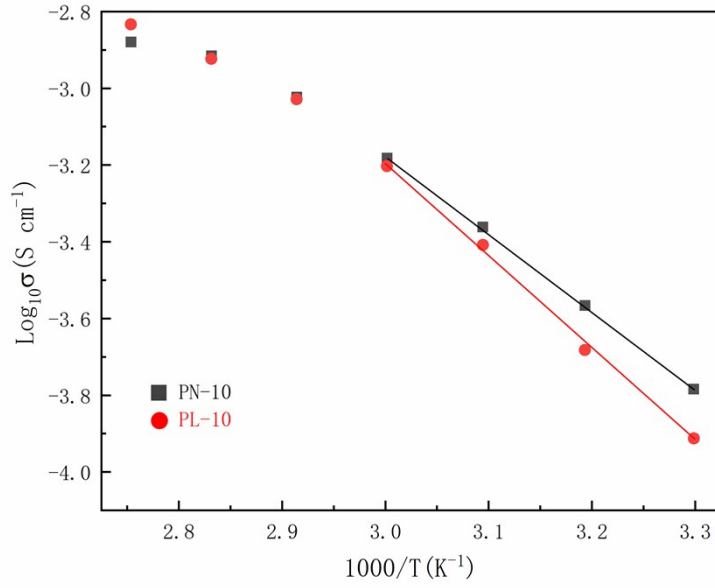
The montmorillonite and lithium chloride were added to distilled water at a mass ratio of 1: 2 to form a suspension by stirring. The ion exchange between the montmorillonite layers was performed by stirring at 60 °C for 24 h. After that, the suspension was centrifuged and the upper liquid was removed. Then, the precipitate was dried at 80 °C for 24 h after washing precipitate for 5 times. The LM powder was obtained after grinding and screening the dry precipitation with a 200 mesh sieve.



**Fig. S1.** DSC thermograms of PN-10 and PL-10

**Table S1.** Zeta potential of NILDH and lithium montmorillonite

Sample	ZP (mV)
NILDH	9.89
lithium montmorillonite	-14.6



**Fig. S2.** The influence of temperature on ionic conductivity of PN-10 and PL-10

**Table S2.** The values of parameters for Equation S1 and the corresponding calculated values of lithium-ion transference number ( $t_{Li^+}$ ).

Samples	$I_0$ ( $\mu A$ )	$I_s$ ( $\mu A$ )	$R_i$ ( $k\Omega$ )	$R_f$ ( $k\Omega$ )	$R_0$ ( $k\Omega$ )	$R_s$ ( $k\Omega$ )	$\Delta V$ (mV)	$t_{Li^+}$
PEO/LiTFSI	2.263	1.524	4.728	5.189	19.965	21.212	50	0.20
PN-10	4.910	3.725	0.874	0.978	9.757	11.630	50	0.27

$$t_{Li^+} = \frac{I_s R_f [\Delta V - I_0 R_0]}{I_0 R_i [\Delta V - I_s R_s]} \quad (S1)$$

where  $I_0$  and  $I_s$  are the initial and steady-state current determined by the DC polarization, respectively;  $R_f$  and  $R_i$  are the initial and final resistances of the electrolytes;  $R_0$  and  $R_s$  are the interface resistance measured by AC impedance before

and after polarization;  $\Delta V$  is the DC potential applied on the cell.