Supplementary Information

Carboxylated polyimide separator with excellent lithium ion transport properties for high-power densities lithium-ion battery

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Figure S1. Optical photographs of (a) PI-0 separator and (b) PI-3 separator.



Figure S2. TGA of PI-0 and PI-3 separators



Figure S3. The calculation model of lithium ion transport rate across (a) PI separator,

(b) PI-COOH separator.

According to Newman's equations¹ which describes mass transport, ion gradients, and potential drops in the electrolyte, the relationship between the ion transport rate and ion conductivity can be concluded from the following equations:

i = nqsv(1)

 $i \propto \sigma(2)$

where the i is current density, n is the ion concentration, q is the electron charge, s is the pore area, v is the ion transport rate and σ is the ion conductivity. Thus, for the PI-0 separator (Figure S3(a)), the following equation can be inferred:

$$n_1v_1 + n_2v_2 = k\sigma_1(3)$$

where the n_1 and n_2 are respectively the lithium ion concentration and hexafluorophosphate ion concentration, v_1 and v_2 are respectively lithium ion transport rate and hexafluorophosphate ion transport rate, σ_1 is the ion conductivity of the PI-0 separator and k is a constant related with the q, s and electric field intensity. In addition, the relation between the ion transport rate and transference number can be described by:

$$\frac{n_1 v_1}{n_1 v_1 + n_2 v_2} = t_1$$
(4)

where t₁ is the transference number of the PI separator. Thus, the result can be obtained: $v_1 = 0.39k$ $v_2 = 1.76k$

For the PI-COOH separator (PI-1 separator as representative) (Figure S3(b)), the similar equation can also be given by:

$$n_{1}v_{1} + n_{2}v_{2} + n_{3}v_{3} = k\sigma_{2} (5)$$

$$\frac{n_{1}v_{1} + n_{3}v_{3}}{n_{1}v_{1} + n_{2}v_{2} + n_{3}v_{3}} = t_{2} (6)$$

where n_1 and n_2 is respectively the lithium ion concentration and bulk hexafluorophosphate ion concentration in the PI-1 separator in the pore (slow transport channel), n_3 is the lithium ion concentration on the surface of the PI-1 separator (fast transport channel), v_3 is the transport rate of lithium ion on the surface of the PI-1 separator, σ_2 is the ion conductivity of the PI-1 separator and t_2 is the transference number of the PI-1 separator. Thus, the result can be obtained:

$$v_3 = 2.43k$$

Similarly, the results for the other PI-COOH separator can also be obtained:

$$v_4 = 2.49k$$

 $v_5 = 2.58k$

where v_4 and v_5 are the lithium ion transport rates on the surface of the PI-2 and PI-3, respectively.

References

1. M. Doyle, T. F. Fuller and J. Newman, Journal of the Electrochemical Society, 1993,

140, 1526-1533.