Supporting information

Predictive Zeta Potential Measurement Method Applicable to Nonaqueous Solvents in High-Concentration Dispersion Systems for the System of LiClO₄–Propylene Carbonate Solution and LiCoO₂ Powder Sheet[†]

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Fig. S1. Schematic diagram of the polypropylene self-made cell

Raw data of streaming potential measurement by streaming potential method fitting methods and results

The raw data of transient response of streaming potential and predictive calculated streaming potential is provided as Excel files. The worksheet is changed for each measured solution. From the left column, the time since the liquid started to flow (*t*), streaming potential (*E*), operation (how many pressurization or depressurization operations), and *p* are shown. For the fitted data that calculates E_{∞} , column *E* is colored yellow. The fitting to calculate E_{∞} is performed twice for each of the pressurization and depressurization processes as recorded in the Excel files for the predictive streaming potential measurement.



Fig. S2. The transient response of the streaming potential of the $LiCoO_2$ sheet in a 10 mmol L^{-1} $LiClO_4$ aqueous solution (pH 5.86). Rawdata is saved in Excel file.



Fig. S3. (a) The result of approximating the numerical value $(E(t) - E_{\infty})/(E_0 - E_{\infty})$ that standardizes the difference from the initial value E_0 with the exponential function exp $(-t/\tau)$ by the Gauss-Newton approximation method in a 10 mmol L⁻¹ LiClO₄ aqueous solution. (b) The relationship between the external pressure and the streaming potential in the part where the 200-700 hPa pressure step was repeated four times in a 10 mmol L⁻¹ LiClO₄ aqueous solution.

Discussion of ζ potential in LiClO₄ aqueous solution

The relationship between the external pressure and the streaming potential of the 10 mmol L⁻¹ LiClO₄ aqueous solutions adjusted to each pH obtained by the same procedure is shown in Fig. S4 (a). For pH adjustment, the aqueous solution which dissolves LiClO4 at a concentration of 10 mmol L⁻¹ and HClO₄ at a concentration of 0.1 mol L⁻¹, or dissolves LiClO₄ at a concentration of 10 mmol L⁻¹ and LiOH at a concentration of 0.1 mol L⁻¹ were used. Since the external pressure and the streaming potential showed a good linear relationship in each pH solution, the pH dependence of the ζ potential calculated from this slope is shown in Fig. S4 (b). (From this figure, it was confirmed that the ζ potential value on the surface of the LCO sheet in the LiClO₄ aqueous solution changed from a positive value to a negative value as the pH increased, and showed an isoelectric point at a pH of about 3. Fig. S4 (c) shows the pH dependence of the ζ potential of the LiCoO₂ powder in a 10 mmol L⁻¹ LiClO₄ aqueous solution measured by electrophoresis. The electrophoresis method used was ELS-Z2K from Otsuka Electronics. The pH was adjusted by the same method as that measured by the streaming potential method. As a result, the isoelectric point of the zeta potential measured by the electrophoresis method was about 3, which was the same as the result measured by the streaming potential method. This gives reliability to the numerical values measured by the current streaming potential method





Fig. S4. (a) The relationship between the external pressure and the streaming potential of the LCO sheet in 10 mmol L⁻¹ LiClO₄ aqueous solutions adjusted to each pH. (b) The pH dependence of ζ potential of the LCO sheet in 10 mmol L⁻¹ LiClO₄ aqueous solution obtained by streaming potential method. (c) The pH dependence of ζ potential of the LCO sheet in 10 mmol L⁻¹ LiClO₄ aqueous solutions was obtained by electrophoresis.

Variation of ζ potential in LiClO₄ aqueous solution with electrolyte concentration

In addition, the relationship between the external pressure and the streaming potential of the 1.0 mol L^{-1} , 2.0 mol L^{-1} , and 3.0 mol L^{-1} LiClO₄ aqueous solutions obtained by the same procedure is shown in Fig. S5 (a). Even when the concentration was changed, the external pressure and the streaming potential showed a good linear relationship. The concentration dependence of the ζ potential is shown in Fig. S5 (b). It was confirmed that the ζ potential had a negative value at any concentration, and the sign of the ζ potential did not change depending on the salt concentration.



Fig. S5. (a) The relationship between the external pressure and the streaming potential of the LCO sheet in 1.0 mol L⁻¹, 2.0 mol L⁻¹, and 3.0 mol L⁻¹ LiClO₄ aqueous solutions. (b) The concentration dependence of the ζ potential of the LCO sheet in 1.0 mol L⁻¹, 2.0 mol L⁻¹, and 3.0 mol L⁻¹ LiClO₄ aqueous solutions.



Fig. S6. The results of an exponential approximation for the streaming potential of the LCO sheet in (a) 0.5 mol L^{-1} , (b) 1.5 mol L^{-1} , (c) 2.0 mol L^{-1} , and (d) 3.0 mol L^{-1} LiClO₄ PC solution.