Analytical technique on proton transport resistance in support pores in PEFC catalyst layer

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Catalysts with metal particles supported in primary pores of mesoporous carbon have been developed in recent years to prevent catalyst poisoning by ionomers. There is concern that proton transport resistance in a catalyst layer composed of such a catalyst will be high because protons cannot be conducted in primary pores via ionomers. Therefore, it is important to understand the effect of the mesopores for designing the catalyst layer. In this study, in order to evaluate the proton transport resistance inside the primary pores, we devised an AC impedance analysis technique based on a transmission line model including the resistance component. Additionally, we verified the technique by applying it to catalyst layers of various supports.

Fig.1 shows the equivalent circuit of the proton conduction path for electric double-layer charge/discharge in the catalyst layer. The conduction paths in the direction of the catalyst layer film thickness and in the direction of the support pore depth are both expressed in the transmission line circuit. The impedance of this equivalent circuit is represented by Equation 1.
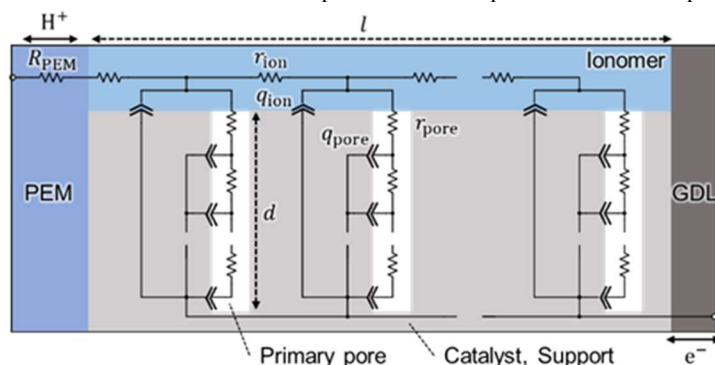

Fig. 1   Equivalent circuit of the catalyst layer composed of meso-porous carbon and ionomer.

$$Z = R_{PEM} + \sqrt{r_{ion}\left(\frac{1}{(j\omega)^\alpha q_{ion} + \frac{1}{z_{pore}}}\right)} \coth l \sqrt{r_{ion}\left((j\omega)^\alpha q_{ion} + \frac{1}{z_{pore}}\right)} \quad \text{Eq. 1}$$

$$z_{pore} = \sqrt{\frac{r_{pore}}{(j\omega)^\alpha q_{pore}}} \coth d \sqrt{r_{pore}(j\omega)^\alpha q_{pore}}$$
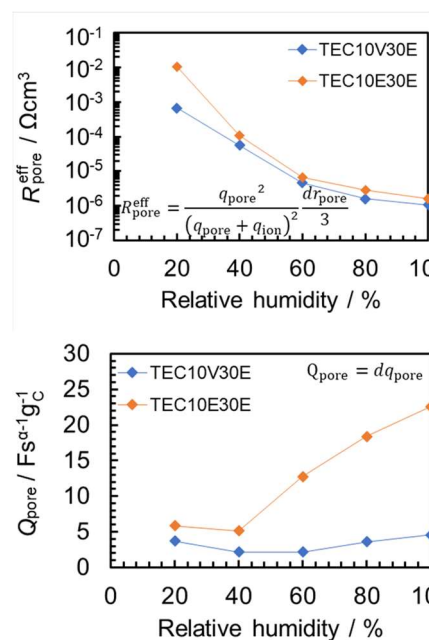

Fig. 2 Humidity dependence of the (a) effective resistance and (b) CPE in primary pores of a catalyst support.

AC impedance measurements were conducted on catalyst layers composed of TEC10V30E (Tanaka Kikinzoku Kogyo) with a small amount of support pores or TEC10E30E (Tanaka Kikinzoku Kogyo) with a large amount of support pores and Nafion™ D2020 (Chemous) ionomer. The temperature of the cell was set to 40 ° C. N_2 was supplied to the working electrode and H_2 was supplied to the counter electrode. In order to eliminate electrochemical phenomena not considered in the equivalent circuit such as oxidation of crossover hydrogen, CO was adsorbed on the platinum catalyst during measurement. The parameters of the effective resistance and CPE were evaluated by fitting the experimental impedance with the above model calculation. The analysis was verified by humidity dependence of the parameters.

The parameters related to the support pores are shown in Fig.2. The effective resistance in the support pores (Fig. 2a) is larger in TEC10E30E than in TEC10V30E. The difference tends to increase with lowering humidity. The CPE of the support pores (Fig. 2b) show a tendency to increase with increasing humidity in TEC10E30E, while TEC10V30E has a small dependence on humidity. These trends are considered to reflect that water adsorption and capillary condensation facilitate the proton transport and increase the available surface area. It can be said that the parameters obtained by our analysis technique are qualitatively valid.