

### Solid-state alkaline fuel cell electrode using layered double hydroxide as ion conductor

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#### Introduction

All solid state alkaline fuel cells (SAFCs) without liquid alkaline medium have attracted attention because of several unique benefits including the usage of non-noble metal catalysts along with the restriction to carbonate precipitation in electrode when air is supplied in the cathode. Recent studies almost focused on the anion exchange membrane (AEM) design aimed to high hydroxide ion conductivity at low relative humidity (*RH*) and considerable durability at elevated temperature, which has not been achieved yet.<sup>1</sup> Moreover, the studies on the electrode ionomer for SAFC are extremely rare. As the same problem as AEMs, a serious degradation of anion exchange ionomer will occur by temperature increase and hydroxyl attack at high pH. Furthermore, the ionomer swells easily under high *RH*, which results in the difficulty of gas diffusion onto the cell reaction site.

To solve the issues above, inorganic ion conductor is considered as a promising candidate with high mechanical and thermal durability. Layered double hydroxide (LDH) is one of it. LDHs are anionic clay and the general formula for LDHs is  $[M^{II}_{1-x}M^{III}_x(OH)_2]^{x+}(A^{n-})_{x/n}mH_2O$ , where  $M^{II}$  is a divalent cation such as  $Mg^{2+}$ ,  $Zn^{2+}$ , etc., and  $M^{III}$  is a trivalent cation such as  $Al^{3+}$ , and  $Fe^{3+}$ , etc.,  $A^{n-}$  is an anion such as  $CO_3^{2-}$ ,  $Cl^-$ ,  $NO_3^-$ . LDH can also be delaminated to a single layer to extend the surface. The LDH size can be controlled by the pH, temperature, and time during synthesis process. The ion conductivity of LDH has been studied in the previous research as  $10^{-3}$  S  $cm^{-1}$ .<sup>2</sup> However, using LDH as an ion conductor for the  $H_2/O_2$  fuel cell test has not been achieved.

In this study, LDH and delaminated LDH (dLDH) were synthesized as ion conductor in electrode. Cell performance test was successfully operated with different LDH/dLDH:Pt/C ratio in catalyst layer. The relationship between electrode structure and LDH electrode cell performance was explored for the first time.

#### Experiment

Mg-Al  $NO_3^-$  LDH was synthesized by hydrothermal coprecipitation method under pH = 9.5 and 80 °C for 4 hours. Then the layered LDH was delaminated in formamide solution for 12 hours at 80 °C with  $N_2$  bubbling. The resulted LDH/dLDH was analyzed by XRD and TEM investigation. The commercial Pt/C catalyst was mixed with LDH/dLDH powder and immersed into ethanol to form the catalyst paste with the different weight ratio (Pt:C:LDH/dLDH = 1:1:0, 1:1:1, 1:1:2, 1:1:4, 1:1:10). The catalyst paste was sprayed on the two sides of a pore-filling membrane synthesized by 3-Methacryloylamino propyl trimethylammonium chloride (MAPTAC) (50 wt% aqueous solution). The Pt loading ratio for each electrode is around 0.03 mg  $cm^{-2}$ . *IV* test was performed at 50 °C with the humidified  $H_2/O_2$  supplied.

#### Results and discussion

The synthesized LDH particle was confirmed by XRD

pattern. The layered structure was confirmed by (003) and (006) surface. The particle size was almost around 50 nm. The cell performance results of LDH/dLDH electrode are shown in figure 1 and 2. From the result, it was figured out that cell performance test was successful by using LDH/dLDH ion conductor. Without any LDH/dLDH, the cell performance was very poor because of the bad formation of ion conducting path in the electrode. For LDH electrode, the cell performance increased with the elevated LDH loading ratio. It was considered that the ion conducting path can be easily constructed with large LDH loading. However, for dLDH electrode, an opposite result was obtained that the cell performance decreased when loading more dLDH into the electrode because delaminated LDH was easily restacked when formamide solution removed. During this process, large number of Pt/C particles was clamped into LDH layers and electron conducting path was difficult to be formed. To confirm the electron conducting resistance, Ohm meter was used to measure the electrode. It was found that in high dLDH loading ratio electrode (Pt:C:dLDH = 1:1:4), the electron conducting resistance was 30 times larger than that in LDH electrode (Pt:C:LDH = 1:1:4). It was indicated that to improve the cell performance of SAFC by using LDH/dLDH electrode, electron conducting resistance cannot be ignored. The method to reduce electron conducting resistance should be considered in future works.

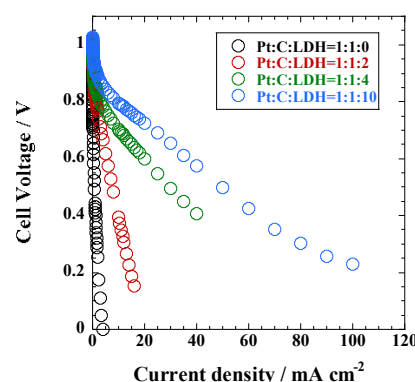


Fig.1. *IV* curve with different Pt:C:LDH ratio

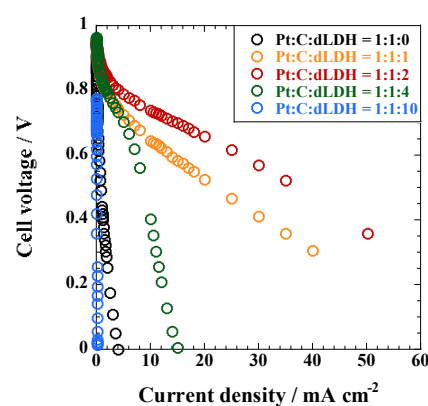


Fig.2. *IV* curve with different Pt:C:dLDH ratio (dLDH stands for delaminated LDH)

#### Reference

1. J. R. Varcoe et al., *Electrochem. Commun.*, 8 (2006) 839–843
2. T. Tamaki et al., *Electrochem. Commun.*, 25 (2012) 50-53