SANS study for degradation behavior of catalyst membrane in fuel cells

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1. Introduction

The catalyst membrane used in fuel cells consists of a platinum catalyst supported on the surface of a carbon support; the platinum is coated with an ionomer (conductive polymer), which enables the membrane to function as a catalyst. A major change for fuel-cell durability is the degradation of the catalyst layer during long-term operation, which leads to a decline in fuel-cell performance. The degradation of the catalyst layer is generally attributed to carbon corrosion resulting from carbon oxidation. However, the mechanisms by which carbon oxidation induces structural changes in the ionomer and platinum, leading to catalyst membrane degradation, remain poorly understood.

Structural analyses using small-angle neutron scattering (SANS) have demonstrated that the spatial distribution of the ionomer within the catalyst membrane is closely related to fuel-cell performance [1,2]. Building on these findings, this study aims to employ SANS to elucidate the complex structural evolution of the catalyst membrane during degradation. By exploiting H/D contrast variation in neutron scattering, the voids within the catalyst membrane were filled with an appropriate solvent, allowing the carbon and ionomer structures to be selectively isolated and their structural evolution to be analyzed independently.

2. Experiment

Two types of Pt-supported carbon with different internal structures were used to fabricate a catalyst membrane by mixing them with an ionomer. Accelerated degradation tests were then performed to obtain catalyst membranes with different degradation states.

A mixture of octane and d-octane was subsequently introduced into the dried catalyst membrane to fill the pore space, with the mixing ratio adjusted to match the scattering length density (SLD) of the void phases to that of either the carbon support or the ionomer. SANS measurements were performed for each sample at sample-to-detector distances of 1 and 8 m, covering a wide q range.

3. Results and Discussions

The SANS profiles shown in Fig. 1 were obtained under ionomer contrast-matching conditions and reveal the evolution of the carbon phase during degradation. Notably, the fractal characteristics of the carbon structure changed significantly between 3 and 6 h of degradation. By independently analyzing the evolution of the carbon and ionomer phases, the complex structural evolution underlying catalyst-membrane degradation was investigated. The obtained insights are expected to provide design principles for durable catalyst membranes, contributing to the development of fuel cells with improved long-term performance.

[1] M. Harada *et al.*, ACS Omega **6**, 15257 (2021).
[2] M. Harada *et al.*, ACS Appl. Mater. Interfaces. **15**, 42594 (2023).

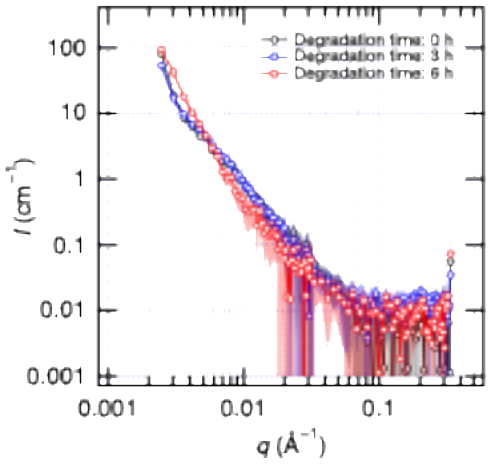

Fig. 1. SANS profiles of carbon in the catalyst membrane at different degradation times.