

# Electrochemical hydrogenation of toluene to methylcyclohexane by non-Pt cathode in the hydrogen storage system

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H<sub>2</sub> energy has been paid attention to use renewable energy efficiently. In order to storage H<sub>2</sub> in a large scale and transport H<sub>2</sub> for a long distance, the organic hydride process which is based on hydrogenation/dehydrogenation of toluene (TL)-methylcyclohexane (MCH) is a promising method.[1] H<sub>2</sub>, produced by the water electrolysis, is stored as MCH by the hydrogenation of TL. MCH, which is liquid under ambient conditions, has advantage to transport and store. H<sub>2</sub> can be reproduced by the dehydrogenation of MCH. As the alternative to catalytic hydrogenation, electrohydrogenation of TL to MCH using 50 wt% Pt/Ketjenblack (KB) cathode has been paid attention.[2] In the system, water is oxidized on anode, and TL is reduced to MCH on cathode (Fig. 1). Because TL can be hydrogenated to MCH with water in 1-step, an energy conversion efficiency would be high. How to suppress evolution of H<sub>2</sub> is important point. The 50 wt% Pt/KB in known as very active cathode, however, there are no reports focusing on non-Pt electrocatalysts. Thus, the purpose of this research is to develop non-Pt electrocatalyst for the electrohydrogenation of TL to MCH.

KB-supported precious metal electrocatalysts were prepared by the conventional impregnation method and H<sub>2</sub> reduction at 673 K. Electrocatalyst ink, which is prepared from the electrocatalyst, Nafion<sup>®</sup> and acetone, was casted on carbon paper (GDL-25BC) to prepare electrodes. Membrane electrode assembly (MEA) was fabricated by hot-pressing the cathode, Nafion<sup>®</sup> 117 membrane and the anode. The MEA is set in an electrolysis cell, and TL and water were

added to cathode and anode, respectively (Fig 1). Galvanostatic electrolysis at 50 mA cm<sup>-2</sup> was carried out.

Fig. 2 shows faradic efficiency of MCH formation (FE(MCH), selectivity in electrolysis) at the 1~20 wt% M/KB cathodes. The 20 wt% Pt/KB showed a low FE(MCH) of 75%. It was to be noted that 20 wt% Ru/KB, 20 wt% Rh/KB and 20 wt% Ir/KB cathodes showed approximately 100% FEs(MCH). In contrast, 20 wt% Pd/KB and 20 wt% Au/KB showed ≈ 0% FE(MCH). When metal loadings were decreased to 1 wt%, FEs(MCH) decreased for all cathodes. However, the Ru/KB cathode showed excellent electrohydrogenation activities at lower loading. At 1 wt%, a 71% FE(MCH) of Ru/KB is far higher than that of Rh/KB (10%), Ir/KB (2%) and Pt/KB (2%). We found that Ru/KB cathode is very active for the electrohydrogenation of TL to MCH even at a low metal loading (1wt%).

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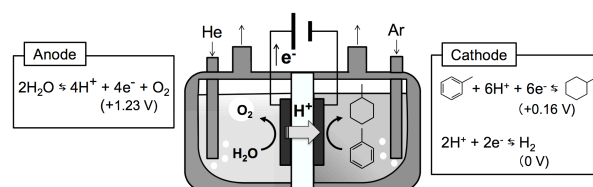


Fig. 1 Schematic diagram of the electrolyzer for the electrohydrogenation of TL to MCH.

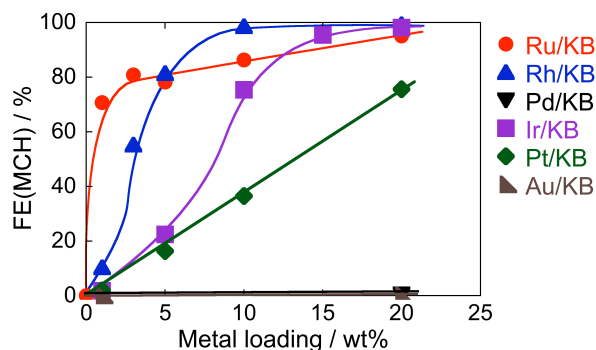


Fig. 2 Effect of metal loadings on faradic efficiency of MCH formation at the electrohydrogenation of TL (50 mA cm<sup>-2</sup>).

## REFERENCES

- [1] Y. Okada, *J. Jpn. Inst. Energy*, 91 (2012) 473.
- [2] S. Mitsushima, *et al.*, *Electrocatalysis*, 7 (2016) 127.