

# Study on HI Decomposition Catalyst for Iodine-Sulfur Method for Hydrogen Production

ワシラパン, パンクラウイ

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氏 名 : Wachirapun Punkrawee

論 文 名 : Study on HI Decomposition Catalyst for Iodine-Sulfur Method for Hydrogen Production (水素製造のためのヨウ素-硫黄法における HI 分解触媒に関する研究)

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### 論 文 内 容 の 要 旨

SI (sulfur-iodide) thermochemical water splitting cycle is expecting as an effective method for large-scale hydrogen production without greenhouse gas emission since only heat energy is required. However, HI decomposition, where hydrogen is generated, requires catalyst to increase reaction rate close to equilibrium conversion. Moreover, the activity and stability of present catalysts are insufficient. Therefore, the high activity and stability catalyst will be investigated.

In Chapter 1, current state of catalyst used for HI decomposition is introduced and the issues of HI decomposition catalyst are make clearer. The objective of this study is to develop highly active and stable catalyst for HI decomposition.

In Chapter 2, Cu added to Pt-CeO<sub>2</sub>/activated carbon (M563) catalytic was the most effective for increasing the activity of HI decomposition. At temperature higher than 573 K, this catalyst show catalytic activity which was close to an equilibrium conversion. Cu addition increased Pt dispersion by anchoring effects. Therefore, Cu additive in the catalyst increases activity and stability. Due to I<sub>2</sub> absorption on the catalyst at first 20 h experimental time, HI conversion was greater than that of the equilibrium conversion. However, after 20 h, stable HI conversion reaching to the equilibrium one was sustained over 100h.

In Chapter 3, Pt-TiO<sub>2</sub> loaded on M563 was studied as a highly active and stable catalyst. Although the activity of TiO<sub>2</sub> loaded catalyst was slightly lower than that of CeO<sub>2</sub> loaded one, the higher stability against HI decomposition was achieved. The HI conversion closing to

equilibrium conversion was sustained over 65 h examined. Moreover, effects of noble metal addition on Pt was studied. It was found that Pt-Rh bimetallic system was a highly active and stable to HI decomposition. According to scanning transmission electron microscope (STEM) result, the presence of Rh results in the better dispersion of Pt. Consequently, the better Pt dispersion explains the respectively high catalytic performance. The high dispersion state of Pt was sustained after HI decomposition at 773 K. The PtI<sub>4</sub> formation during HI decomposition was suggested by X-ray photoelectron spectroscopy (XPS) measurement. Eventually, the effect of Rh addition to the catalytic stability could be explained by the high chemical stability of Rh against iodine. Almost equilibrium HI conversion on Pt-Rh-TiO<sub>2</sub>/M563 was sustained over 300 h.

In Chapter 4, active site on carbon for HI decomposition reaction was studied by using four type of activated carbon catalysts. HI adsorption, Raman spectroscopy, and kinetic analysis were used for analysis of active site. It was suggested that number of HI adsorption site on carbon was an important parameter for high HI decomposition and porous structure was crucial factor for achieving high HI decomposition activity. According to kinetic analysis, reaction step of HI decomposition on activated carbon was well agreed with Langmuir-Hinshelwood mechanism in which dissociation of HI into H<sub>ad</sub> and I<sub>ad</sub> was the rate determining step. Therefore, increasing in HI adsorption site was important for achieving the high HI decomposition activity.

In chapter 5, results of this thesis were summarized. The future prospect of HI decomposition catalyst and also SI method were discussed.