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審査研究科	生命環境科学研究科		
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論 文 の 要 旨

Abstract of thesis

Gasification is the most effective means to convert biomass into high energy content gases. Unfortunately, producer gases often contain tars, trace contaminants and particulates with relatively low H₂ levels. Consequently, the objective of this research is mainly to (1) use synthesized catalysts to catalytical steam reforming of model tar compounds; (2) use commercial catalysts to test the hot cleaning system for reducing tar content; and (3) enrich H₂ production through combination of hot cleaning system and water-gas shift reactors. Therefore, the produced gas can be efficiently used.

This research was divided into three tasks. Task (1) included catalytic steam reforming for tar (toluene and benzene) catalytic conversion using new synthesized Ni-based catalysts with olivine as a carrier. Three catalysts (Ni/olivine, Ni-Ce/Olivine and Ni-Ce-Mg/Olivine) were prepared by wet impregnation in this task with benzene/toluene as model tar compounds. Catalytic steam reforming of benzene and toluene was performed in a bench scale fixed bed reactor at temperatures between 700 and 830 °C using a molar ratio of steam/carbon of 3 to 5. Task (2) utilized catalytic hot cleaning in two-bed system (guard and catalytic reactor) from fluidized gasifier to remove tars. The system consisted of a guard bed and catalytic reactor to treat the producer gas from an air blown, fluidized bed biomass gasifier. A slipstream was drawn immediately downstream of the cyclone. Task (3) involved hydrogen production from a combination of hot cleaning and water-gas shift system (high temperature and low temperature bed). Overall, the typical techniques were used for catalyst characterization including X-Ray diffraction, X-ray photoelectron spectroscopy, Fourier transform infrared, scanning electron microscopy, BET, thermogravimetric and mercury porosimetry analysis, etc.

The dissertation is divided into 6 chapters. In the chapter 1, the author gave a literature review on the previous studies relating to biomass gasification, its products, tar and cleaning treatment. The author addressed the existing

problems of cleaning treatment of produced gas, difficulties of tar removal and low hydrogen contents and pointed out the advantages of catalytic hot cleaning technology of tar. The methods, key contents and goals of each chapter are also presented. Then the author proposed research objectives at the end of this chapter. In the chapters 2 and 3, the author investigated new synthesized catalysts for model tar conversion. The results indicated that 3.0% NiO/olivine doped with 1.0% CeO₂ as the most promising catalyst based on catalytic activity and its resistance to coking. Cerium oxide is thought to promote the catalytic activity of nickel through a redox mechanism and resist the deposition of the carbon. In addition, the use of MgO dopant in Ni-Ce/olivine catalyst was evaluated and results indicates that Ni-Ce-Mg/olivine catalysts could improve the resistance to carbon deposition, enhance energy gas yield and resist 10 ppm H₂S poison at 100 mL min⁻¹ for up to 400 min. In the chapter 4, the author explored the hot cleaning system of tar from real biomass gasification. The guard bed used dolomite to crack the heavy tars and absorb H₂S. The catalytic reactor was used to evaluate three commercial Ni-based catalysts. The system was effective in eliminating heavy tars (> 99% destruction efficiency) and in increasing H₂ concentration by 6–11 vol%. Space velocity had little effect on gas composition while increasing temperature boosted hydrogen yield and reduced light hydrocarbons (CH₄ and C₂H₄), suggesting tar destruction is controlled by chemical kinetics. In the chapter 5, the author investigated the enrichment of H₂ production through the combination of hot cleaning system and water-gas shift reactors. The results indicated that steam reforming of tars and light hydrocarbons and reacting steam with carbon monoxide via the water-gas shift reaction could increase hydrogen content in the producer gas to 27-30 vol% through biomass gasification. In general, H₂ production was quantified as a function of temperature, space velocity and steam/gas ratio. Finally, in the chapter 6, the author summarizes the overall conclusions and future research perspectives. It includes improved gasification technologies, optimized operating conditions, innovative catalysts for tar catalytic conversion and enhanced H₂ production. A large-scale pilot study and thermal and biochemical hybrid system of syngas fermentation are recommended for future research.

審査の要旨 Abstract of assessment result

The originality of this research has been demonstrated in each of the three tasks conducted by the author. Concerning the new doped Ni-based catalysts for steam reforming model tar, the author synthesized new catalysts of both Ni-Ce/Olivine and Ni-Ce-Mg/Olivine. The dopant of CeOx improved coke resistance by redox mechanism. The Ni-Ce-Mg/Olivine catalyst provided a better Ni dispersion, high toluene conversion (93%) and resistant to H₂S poison. As for the hot cleaning system, the author used hot cleaning system for catalytic tar removal as well as for enhancing H₂ production. The results were satisfactory as it could achieve > 99% destruction of heavy tars and increasing H₂ concentration by 6-11 vol%. For H₂ production from biomass gasification, this research was the first attempt to use the combination of hot cleaning system and water-gas shift reactors for enhancing H₂ production. By using different reactor setups under various operating conditions, the goal of each task was met. Furthermore, the author used various techniques to characterize catalysts to explain why and how the better performance of these catalysts and subsequent deactivation were achieved. In addition, the literature review is excellent leading to the rationale for this research, and the structure of thesis is sound. Overall, the contents of the thesis meet the quality of Ph.D. thesis.

The final examination committee conducted a meeting as a final examination on 19 January, 2018. The applicant provided an overview of dissertation, addressed questions and comments raised during Q&A session. All of the committee members reached a final decision that the applicant has passed the final examination.

Therefore, the final examination committee approved that the applicant is qualified to be awarded the degree of Doctor of Philosophy in Environmental Studies.