主　論　文　の　要　約

論文題目　**Study on elucidation of behaviors and reaction mechanisms of biomass gasification for enhanced energy conversion**
(エネルギー変換効率向上のためのバイオマスガス化挙動と反応機構解明に関する研究)

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論　文　内　容　の　要　約

In this study, the objective is to elucidate the behaviors of oxygen-enriched air biomass gasification from the aspects of both macro and micro aspects with the goal of promoting its commercialization. The study was inspired by the "hydrogen society" vision proposed by Japan, in which the effectively utilizing by-product oxygen has been problematic due to the limitation of regional distribution. To address this obstacle, the gasification of woody biomass using oxygen-enriched air in a small-scale auto-thermal downdraft packed-bed gasifier was proposed to fulfill the by-product oxygen utilization on a regional level, providing a more efficient and localized solution. On top of that, we further clarified the effects of gasifying agents and ash particles on the biomass char gasification reactivity, the rate-determining step in the global biomass gasification process.

In chapter 1, the foundational context for this study was laid by delving into several critical aspects. It began by offering an overview of the current state of global energy consumption, highlighting trends, challenges, and the growing need for sustainable energy solutions. Renewable energy resources were also examined, with a particular focus on the properties of biomass resources, including composition, type, and availability, which are essential for understanding its potential as an energy source. In addition, we provided a comprehensive literature review of thermochemical conversion processes, such as gasification, pyrolysis, and combustion, detailing their mechanisms, efficiencies, and applicability in energy generation. The chapter culminates with a thorough discussion on various types of gasifiers from the aspects of design principles, operational characteristics, and suitability for different types of biomass feedstock. We've introduced how the packed bed gasifiers work in the modes of updraft and downdraft in detail. This introductory chapter thus sets the stage for a deeper exploration of biomass gasification and its potential role in addressing global energy challenges.

In chapter 2, the focus was an in-depth exploration of O_2-enriched air biomass gasification, a pivotal aspect of this study. This chapter comprehensively investigates the impact of using oxygen-enriched air in the gasification process within a small-scale packed-bed gasifier. Key parameters such as the height of the packed bed and the temperature distribution within the gasifier are meticulously analyzed to understand their influence on the gasification process.

The composition and concentration of syngas produced under these conditions are examined, with particular attention to the lower heating value (LHV), which is a critical indicator of the energy content of the syngas. Additionally, the chapter evaluates the tar content in the produced gas, a significant factor in determining the cleanliness and usability of the syngas. The efficiency of the process is assessed in terms of cold gas efficiency (CGE) and carbon conversion rates, providing insights into the effectiveness and environmental impact of the O_2-enriched air gasification method. The quality of the produced syngas, as measured by its lower heating value (LHV), improved from 3.85 to 4.66 MJ/Nm^3, tar content dropped from 0.81 to 0.17 g/Nm^3, and the cold gas efficiency (CGE) rose from 81.22% to 90.69% when the O_2 concentration was raised from 21% to 25%. Thermochemical calculations were further performed using FactSage 8.1 to explore the essence of O_2-enriched effect. These good performances validated the feasibility and potential advantages of using oxygen-enriched air in biomass gasification, especially in small-scale applications, offering a promising avenue for sustainable and efficient energy production.

In chapter 3, kinetics and mechanisms of biomass char gasification in the mixed CO_2 and H_2O atmosphere were studied. The effects of temperature (750–1300 °C) and gasifying agents (CO_2, H_2O, and their mixture) on the gasification characteristics of biomass char were experimentally studied using a batch-type vertical tube furnace. Based on these results, the mechanism has been clarified and an assumption of char active sites was proposed to explain the mechanism transition phenomenon. Chapter 3 delves into the intricate kinetics and mechanisms of biomass char gasification, particularly in atmospheres comprising mixed CO_2 and H_2O. The chapter presents a detailed experimental study conducted within a batch-type vertical tube furnace, where the impact of varying temperatures, ranging from 750 to 1300 °C, is scrutinized. This study also investigates how different gasifying agents, specifically CO_2, H_2O, and their mixtures, influence the gasification characteristics of biomass char. Through these experimental results, the chapter provides a clear elucidation of the underlying gasification behaviors. The gasification behaviors in the atmospheres of CO_2, H_2O, and their mixtures fall into two categories: controlled by chemical reactions and by diffusion. Additionally, a transition in the mechanism was noted in the chemical reaction control regime, shifting from an additive to a competitive mechanism. The analysis of syngas further revealed that in the competitive mechanism, the reactions between char and both CO_2 and H_2O were suppressed. A significant part of the chapter is dedicated to the hypothesis regarding the active sites on biomass char, which are believed to play a pivotal role in the gasification process. The hypothesis suggests that the transition is predominantly due to a shortage of these active sites, leading to a competition between CO2 and H2O molecules for the limited active sites available. Moreover, it was noted that in the competitive mechanism, neither CO_2 nor H_2O showed a distinct advantage in accessing these shared active sites. This hypothesis has explained the observed transition phenomenon in the gasification mechanism, offering a novel perspective on how different temperature conditions and gasifying agents interact with biomass char. The insights gained from this study not only enhance the understanding of biomass gasification kinetics but also have practical implications for optimizing gasification processes in mixed CO_2 and H_2O environments, contributing to more efficient and sustainable biomass energy conversion strategies.

In chapter 4, a comprehensive investigation into the behaviors of ash particles and alkali and alkaline earth metallic (AAEM) compounds during the char gasification process was conducted. The chapter focuses on understanding how ash particles and AAEM compounds, both on the char surface and within their internal cavities, react and transform during gasification. Our findings indicate that the proportions of char particles with ash area ratios of 30-60% and 60%-100% are significantly higher in H_2O gasification than in CO_2 gasification at the same conversion. This observation suggests differing reaction models: the catalytic random pore model for H_2O gasification and the catalytic shrinking core model for CO_2 gasification, which were confirmed by regression calculation. Further investigation into the effects of AAEMs, conducted through EDX analysis, revealed distinct behaviors of Na in the two gasifying agents. In H_2O gasification, Na showed a tendency to aggregate, while in CO_2 gasification, it appeared more dispersed. This contrast also implies that the agglomeration of Na could be associated with the formation of char particle fragmentation, a result of pore structure destruction specific to H_2O gasification. Additionally, the AAEMs inside the char particle cavities in CO_2 gasification suggests a lack of catalytic effect from Na in this context. The chapter delves into the interactions of these compounds with the gasifying agents, and how they affect the structural and compositional changes in the biomass char.

In chapter 5, we summarized the conclusions of all the chapters above and provided future research plans. As discussed in chapter 4, it remains unclear what is the relationship between physical structure inside the char and on the char surface, how the micro-physical structure inside the char evolves and how the micro-physical structure contributes to these catalytic effects. Unraveling these questions is particularly crucial in the context of fluidized bed gasifiers, where the influence of micro-physical structure on catalytic gasification activity is pronounced due to the use of powder feedstock. Furthermore, a cogeneration system combining Distributed Energy Resource Systems (DERSS) with O_2-enriched biomass gasification in this small-scale packed-bed gasifier was proposed, aligning with broader environmental and energy sustainability goals.

In summary, the study has elucidated the behaviors of O_2-enriched air biomass gasification in a small-scale, auto-thermal, packed-bed gasifier, examining both macro and micro aspects. The macro aspects encompass performance evaluation, while the micro aspects focus on char gasification. Special attention is given to the mechanisms of char gasification and the effects of ash to offer instructions for paraments optimization. Overall, the whole study is expected to accelerate industrialization of oxygen-enriched air biomass gasification using small-scale, auto-thermal, packed-bed gasifier.