論文の内容の要旨

Effects of Operating Parameters on Charcoal yield, Energy Conversion Efficiency and Charcoal Production Rate in High-Pressure Flaming Carbonization of Lignocellulosic Biomass

（リグノセルロース系バイオマスの高圧炭化における操作条件が炭化物収率、エネルギー変換効率及び炭化物生成速度に及ぼす影響）

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Biomass is important renewable fuel that has prospects for reducing dependence on fossil fuels. In several countries, large amount of forestry residues and agricultural wastes remain unused. This biomass can be converted into several forms of valuable energy. Carbonization to produce solid charcoal was focused in this study since the process is relatively simple and less expensive compared with other energy conversion technologies. However, current charcoal production methods are time-consuming and inefficient. High pressure has been reported to favor carbonization process but the roles of pressure have not been clarified and more investigation is strongly required to achieve effective application of this novel technology.

This study aimed to investigate effects of operating parameters in high-pressure flaming carbonization, including pressure, airflow rate and feedstock properties on carbonization performance which is determined by charcoal yield, charcoal production rate and energy conversion efficiency. Flaming carbonization was initiated by ignition at the bottom of feedstock. Upward propagation of flame in limited oxygen transforms feedstock into charcoal. The experiments were conducted in laboratory-scale reactor. Roles of pressure and mechanisms in flaming carbonization were clarified based on experimental results and theory. Consequently, a numerical model was developed in order to quantitatively describe the mechanisms involved in flaming carbonization and to predict the process performance at specified conditions. The contribution of this high-pressure process to environment was evaluated based on energy conversion efficiency and emission of by-products. Finally, the economic comparison was made in order to evaluate the feasibility of this process and benefit of high pressure.

The carbonization experiments were conducted under well-controlled conditions, basically in restricted oxygen. Effects of airflow rate were examined at pressure of 0.9 MPa and varied airflows in range of 1.0-4.0 g/min, using Japanese cypress wood. Airflow rates determined temperature and charcoal yields. Low airflow rate offered higher charcoal yield and energy conversion efficiency. Too high airflow was not favorable for carbonization since abundance of oxygen caused intense combustion, which raised temperature and reduced charcoal yield. Charcoal from a slow airflow run contained relatively high volatile matter content as a result of lower temperature and heating rates.

The investigation of pressure effects was divided into 2 parts; 0.5-1.0 MPa using Japanese cypress and 0.5-3.0 MPa using oak wood. Pyrolysis and combustion reactions, which occur simultaneously in flaming carbonization, seem to determine the sustainability of flame and the carbonization rate. Flaming carbonization could not progress when both airflow and pressure were low because oxygen became insufficient in flaming zone and it limited the combustion kinetics and heat to continue pyrolysis reaction. High operating pressure allowed flaming carbonization to propagate in low airflow conditions. The results from Japanese cypress carbonization at pressure 0.5-1.0 MPa showed the trend of improvement in charcoal yields and energy conversion efficiency at higher pressure.

The carbonization of oak wood under pressure 0.5-3.0 MPa revealed that higher pressure up to 2.4 MPa remarkably increased charcoal yield and energy conversion efficiency at the certain airflow rate. The secondary charcoal formation, which is regarded as a main mechanism to raise charcoal yields, could have been promoted at elevated pressure, resulting in more charcoal and less volatiles. Abundant oxygen with limited volatiles at higher pressure could have caused fluctuation in peak temperature and carbonization rate with increasing pressure. It is because an intensity of partial oxidation was controlled by concentration of oxygen and combustible volatiles in the gas phase. Suppressing of volatiles release at high pressure caused slight decrease in fixed carbon content and heating value of charcoal. Nevertheless, remarkable improvement in energy conversion efficiency and charcoal production rate were regarded as the great benefit of higher pressures. Further increasing pressure above 2.4 MPa did not improve energy conversion efficiency. Thus, pressure of 2.4 MPa is supposed to be optimum pressure under the conditions in this study.

Feedstock properties namely size, moisture content and feedstock species, strongly affected the carbonization performance. Charcoal yield and energy conversion efficiency considerably increased in larger particles because prolonged vapor-phase residence time inside large particles facilitates the secondary charcoal formation. Comparable char production rates that appear in large and small feedstock suggested that size reduction is not strongly required prior to carbonization.

Moist feedstock, containing 21% moisture, apparently reduced reaction temperature, charcoal yield, energy conversion efficiency, char production rate and fixed carbon content of charcoal. Large amount of wood was burnt out to provide heat for moisture evaporation. Large increase of steam in gas phase may have diluted the combustible gases, decelerated combustion reaction, and decreased temperature and fixed carbon content of charcoal.
To study effect of feedstock species, several kinds of lignocellulosic biomass were employed, namely coconut shell, oak wood, corncob and rice husk. Flame propagation in high-density biomass bed was slow because more energy and long time were required to heat and initiate pyrolysis in high-density particles. Long existence of flame at each layer raised the fixed carbon content as well as heating value of charcoal. On the other hand, carbonization of biomass with very low bed density was unsuccessful due to limited supply of combustible vapors. Energy conversion efficiency was comparable among various feedstock species but fixed carbon content on dry ash-free basis was extremely high in charcoal from high-density biomass.

The numerical model was developed to quantitatively describe various phenomena occurring in flaming carbonization. Profiles of calculated temperature, reaction kinetics and gas concentration show the progress of flaming front propagation. The sequence and extent of each reaction were elucidated by the numerical model. The model provided satisfactory agreement with the experimental data. The improved charcoal yield at high pressure is caused by secondary charcoal formation which becomes apparent when oxygen is depleted. The model helps to predict the dynamic behavior of flaming carbonization at specified conditions, and it would be a useful tool for defining suitable operating conditions to attain high efficiency of the high-pressure flaming carbonizer.

High pressure considerably reduced the emission factor of gas and liquid products. However, the production of gas and liquid was inevitable and they would harm the environment if discharged with no post-treatment. Considerable amount of carbon and energy retained in gas and liquid suggested their potential use either as fuel for drying feedstock or as material for other processes. Economic comparison made for 1.0 and 2.4 MPa runs verified the improved profitability of higher pressure.

In summary, high pressure could improve charcoal yield, energy conversion efficiency and charcoal production rate. Under certain conditions, there is the optimum pressure which is supposed to be 2.4 MPa in this study. The performance of flaming carbonization could be adjusted by manipulation of airflow rates and pretreatment of feedstock. High-pressure flaming carbonization could be a beneficial alternative for bioenergy conversion. The process could carbonize various kinds of agricultural wastes, minimize pollutant emission to the environment and improve profitability to manufacturers.