

Studies on Selective and Simultaneous Production of Charcoal and Light Oil by Pyrolysis of Biomass with Recycling of Heavy Oil

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論 文 内 容 の 要 旨

With rapid growth of energy demand and serious shortage of fossil fuels, biomass has been receiving attention because of its renewability, common availability and carbon neutrality. Pyrolysis is a simplest thermochemical process to convert biomass to gas, carbonized solid (biochar) and liquid (bio-oil). The biochar is a carbon rich solid and expected as a high quality fuel for combustion and gasification, reducing agent, soil enhancer, etc. Increasing or maximizing the biochar yield has been an important technical subject, while the yield in the existing pyrolysis processes is determined virtually by operating variables such as peak temperature, heating rate and pressure. The bio-oil has been utilized but exclusively as an alternative to petroleum derived heavy oil. This is mainly because a substantial portion of the bio-oil consists of high mass component that, hereafter termed heavy oil, is transformed into coke/char (*i.e.*, evaporation residue) upon heating.

This study proposes a novel pyrolysis of biomass, which simultaneously enables production of biochar at increased or maximized yield and selective production of light bio-oil without evaporation residue. While the formation of heavy oil is inevitable regardless of operating variables of the pyrolysis, the

proposed pyrolysis can avoid discharge of the heavy oil from the process by its full and internal recycling to the pyrolysis. The heavy oil is captured by the feedstock biomass at downstream of the pyrolyzer or therein, recycled being carried by the feedstock, and then converted into biochar, light oil or gas. The principles of the heavy oil capture are condensation into macropores of the feedstock and sorption (i.e., absorption and adsorption) into a flexible polymer-like matrix of the feedstock. The concept of the proposed pyrolysis and the main purpose of this study are presented in Chapter 1, a major part of which consists of a critical review of existing studies on the biomass pyrolysis toward production of the biochar and/or bio-oil.

Chapter 2 proposes a pyrolysis process that is equipped with a heavy oil sorber at the pyrolyzer downstream, reports details of the experimental method and results, and discusses on the results. Ten sequential pyrolysis runs were performed for simulating a continuous processing. In each run of pyrolysis, heavy oil vapor was formed in a horizontal screw conveyer pyrolyzer at 500 °C and captured by a sorber that consisted of a fixed bed of the feedstock, chipped cedar, at 110 °C. The heavy-oil-loaded feedstock was then subjected to the next run. The sorber allowed the light oil and steam (pyrolytic water) to pass through itself and to be condensed downstream. The pyrolysis reached a steady state until the 7th run at a heavy-oil/feedstock mass ratio around 40%, which corresponded to once-through conversion of the recycled of heavy oil as high as 40%. The heavy oil recycling increased the biochar mass yield by a factor of 1.3 mainly due to self-charring of the recycled heavy oil. The light oil consisted mainly of organic compounds with carbon number of 2–12 and contained less than 0.2 wt% of residue after evaporation. The concept of “feedstock-mediated recycling of heavy oil” was thus demonstrated with neither a catalyst nor chemical additive.

Chapter 3 proposes internal and full recycling of the heavy oil in an updraft moving bed pyrolyzer that is designed so that its top part plays a role of the heavy oil sober. Continuous pyrolysis was simulated with a vertical moving bed reactor consisting of a train of 22 containers, each of which had been charged with the feedstock (pine, mallee, cherry and rice husk). The results demonstrated the full recycling of the heavy oil and resulting selective production of light oil that was free from evaporation residue and consisted of organic compounds with carbon number no greater than 13. Moreover, the heavy oil recycling increased the biochar yield from the pine by a factor over 1.4. Not only the self-charring of the heavy oil and its co-carbonization with the feedstock but also chemical deposition of vaporous heavy oil onto the pyrolyzing solid caused such significant increase in the biochar yield. In other words, vapor-solid contact in the moving bed pyrolyzer successfully promoted the conversion of heavy oil, thereby maximizing the biochar yield.

Chapter 4 further discusses on the characteristics of the heavy oil recycling and its effect on properties of the biochar and bio-oil. Detailed product analyses showed that the heavy oil recycled into the pyrolyzer was converted into biochar, water and light oil, but insignificantly to non-condensable light gases, strongly suggesting importance of water-forming cross-linking for the biochar formation from the heavy oil, and

also heavy-to-light oil transformation with little gas formation. Detailed gas chromatography mass spectrometry analysis of the light oil showed that cellulose-derived levoglucosan, the most abundant compound in general biomass pyrolysis, was fully decomposed and incorporated into the biochar. On the other hand, the yields of furans and phenols, such as 5-methylfurfural, furfural, phenol, 4-methylphenol, increased by 2–4 times with the help of heavy oil recycling.

Chapter 5 summarizes the results of this study drawing general conclusions. Future perspective of the biomass pyrolysis and its effective integration into the biomass energy chain are also described.

論文審査の結果の要旨

本論文は、蒸発残さを含まない軽質成分のみで構成される液成分の選択的取得ならびに炭化物収率の大幅な向上を同時に達成するバイオマスの熱分解法を提案し、同法の有効性を実験結果に基づいて示すとともに、重質液成分のリサイクル反応機構を明らかにしたものであり、次世代のバイオマス転換技術開発に有用な知見を与えるばかりでなく、炭素資源利用に関する化学反応工学への学問的寄与が明らかである。よって、本論文は博士（工学）の学位論文に値するものと認める。