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Effect of electron injection on tar profiles generated from oxidative pyrolysis of biomass and plastic

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ABSTRACT

This study reports the component of tar produced from oxidative pyrolysis of cellulose and polypropylene with electron-injected air using a laboratory-scale reactor. This experiment was conducted using a cylindrical fixed bed reactor with a fixed bed. The feedstock tested in this experiment was cellulose and polypropylene. During experiments, the feedstock was heated by an electric heater to 600, or 700 °C with or without electron- injection into the air. Under all experimental conditions, oxidative pyrolysis occurred immediately after the start of the experiment. The analysis results of absorbed liquid tar samples show significant differences were observed between with or without electron injection. Electron injection gave a clear impact on tar production during the oxidative pyrolysis of cellulose and polypropylene decomposition.

Keywords: pyrolysis, oxidative pyrolysis, cellulose, polypropylene, tar reduction

1. INTRODUCTION

The establishment of pyrolysis and gasification technologies will lead to a reduction in the use of fossil fuels. Effective use of energy from biomass materials and plastic wastes is essential for building a sustainable society, and research on pyrolysis and gasification technologies is underway to achieve this goal. One of the challenges of existing pyrolysis and gasification technologies is to deal with tar. Tar is a highly viscous and flammable liquid that is mixed with the gas generated during pyrolysis and gasification and discharged. Tar has become a detriment to industrialization by clogging pipes and adversely affecting engine generators. Various technologies to reduce and remove tar have been studied [1], but no method has yet been established that is economically inexpensive and can be used universally for all materials. Therefore, there is a need for further efficiency improvements and new treatment technologies.

Recently, a new pyrolysis gas activation technique, the electron injection method into pyrolysis reaction was reported [2][3]. This method can activate a pyrolysis reaction with partial combustion by simply injecting electrons. This method can be installed directly in the reactor where the pyrolysis reaction is taking place, and does not require any additional equipment other than an electron generator and an electron injection mechanism, and thus has the potential to activate the pyrolysis reaction at a lower cost than existing methods of activating pyrolysis. In this study, the authors investigated the effects of electron injection into the air on the oxidative pyrolysis of cellulose and polypropylene in regards to tar generation.

2. MATERIAL AND METHODS

2.1 Material

In this study, river sand was used as a supporting material of the fixed bed reactor. Feedstocks were used Cellulose and Polypropylene. Cellulose was used as particles and sheets. The particle cellulose was manufactured by FUJIFILM Wako Pure Chemical Corporation (CAS RN® 9004-34-6) and sheet cellulose was manufactured by ADVANTEC TOYO, LTD. Polypropylene was manufactured by Japan Polypropylene Corporation (NOVATEC-PP, BC03C). When the air was used as the injection gas, pure air of G3 was used.

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2.2 Experimental methods and conditions

The schematic diagram of the experimental reactor is shown in Fig.1. An electron generator that was used in this experiment was ITM-F201 device, produced by Andes Electric Company Japan.

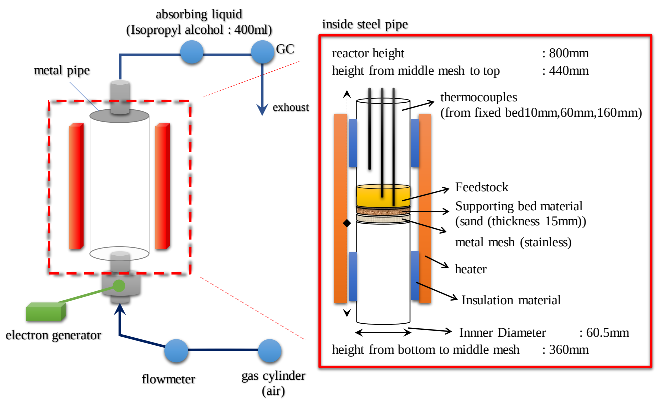

Fig.1 Schematic diagram of the experimental equipment

Electron was injected into the air using ten electron generators before the air was supplied into the reactor. When an electron was not injected, only air was supplied. Experimental conditions are summarized in Table.1.

Table 1 Experimental conditions

Case	1	2	3	4	5	6	7	8
experiment ID	Cell 600 OFF	Cell 600 ON10	Cell 700 OFF	Cell 700 ON10	PP 600 OFF	PP 600 ON10	PP 700 OFF	PP 700 ON10
Number of electron generator	0	10	0	10	0	10	0	10
Sample size	3	3	3	3	3	3	3	3
Temperature ℃	600		700		600		700	
Feedstock	Cellulose				Polypropylene			
Feedstock weight g	30							
Carrier gas	Air							
gas flow rate L/min	0.100							

The reactor was heated to the target temperature (600°C or 700°C) and then the feedstock was thrown into the reactor from top. In the experiment using cellulose as the feedstock, the particulate cellulose was wrapped in sheets of cellulose to prevent dust explosion and blowback. The experiment ID was defined by “temperature + number of electron generators.” For example, 600OFF, and 600ON10 indicate that the temperature was 600°C without electron injection, the temperature was 600°C with the usage of 10 electron generators.

The air was fed from the bottom of the reactor at the flow rate of 0.100 L/min, which was insufficient for complete combustion within 180 minutes. Therefore, Cellulose and Polypropylene were thermally decomposed by oxidative pyrolysis in this study. During the experiments, tar in the syngas was collected by impinger bottles filled with 400 ml isopropyl alcohol (IPA) and cooled in an ice bath. The syngas composition was measured by GC (490 Micro GC System, United State) after tar collection. The weight of tar in the syngas was calculated by measuring the weight of IPA before and after the experiments. The number of repetitions of the experiment was set to three for each experimental condition.

2.3 Characterization of products

A In this study, syngas composition (H_2, O_2, CO, CO_2, CH_4, C_2H_4, C_2H_6, C_3H_6, and C_3H_8) was measured continuously. The tar collected from the syngas was analyzed by the gas chromatography-mass spectroscopy (GCMS-QP2020 NX, Japan) equipped with a capillary column (SH-Rxi-5 Sil MS, length: 30 m, internal diameter: 250 lm, film-thickness: 0.25 lm).

2.4 Results and discussion

2.4.1 Syngas composition

The time change of the concentration of oxygen in the syngas is shown in Fig.2. The oxygen concentration decreased to less than 1% within at least 10 minutes from the start of the experiment. Thereafter, the reaction proceeded under the oxygen-depleted condition for at least 80 minutes at all temperature conditions.

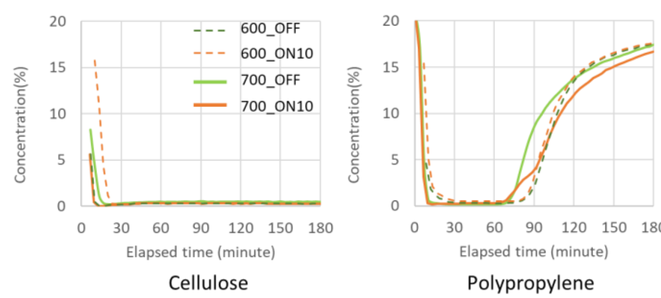

Fig.2 Concentration of oxygen in the syngas

The time changes of the production rate of H_2, CH_4, C_2H_6 calculated from its concentration and the N_2 concentration as well as the N_2 flow rate at 600°C and 700°C are shown in Fig.3,4, respectively. When comparing electron ON/OFF, Comparing the electron ON/OFF, the cellulose experiment showed a statistical advantage at both temperatures. Similarly, in the polypropylene experiment, a statistical advantage was observed at 700°C. At 600°C, a difference was observed in C_2H_6. The higher the temperature, the greater the effect of injecting electrons, and the effect was greater for cellulose than for polypropylene.

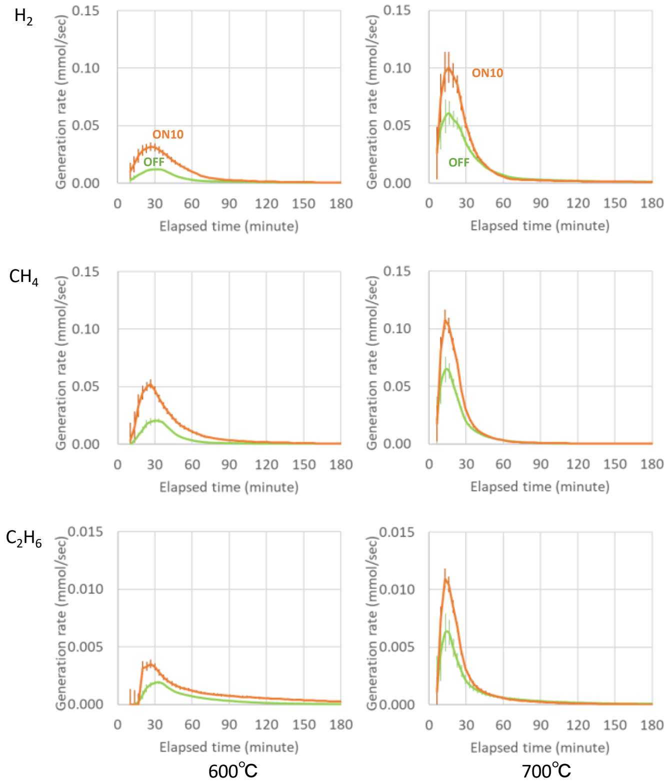

Fig.3 Generation rate of H_2, CH_4, C_2H_6 (Cellulose)

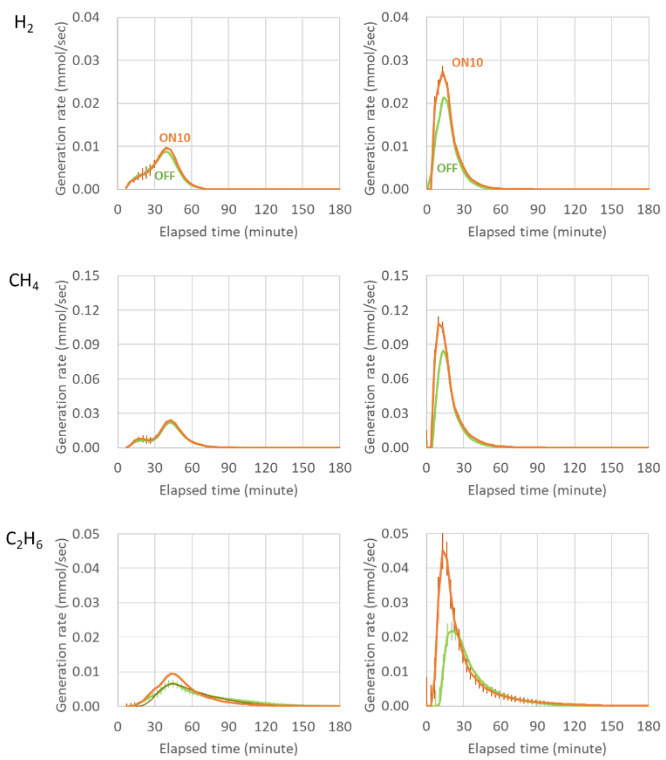

Fig.4 Generation rate of H_2, CH_4, C_2H_6 (Polypropylene)

2.4.2 Tar

Fig.5 shows the tar concentration in the syngas when the feedstocks was cellulose, and the tar concentration decreased with electron injection at both 600°C and 700°C. Both temperatures showed a statistical advantage.

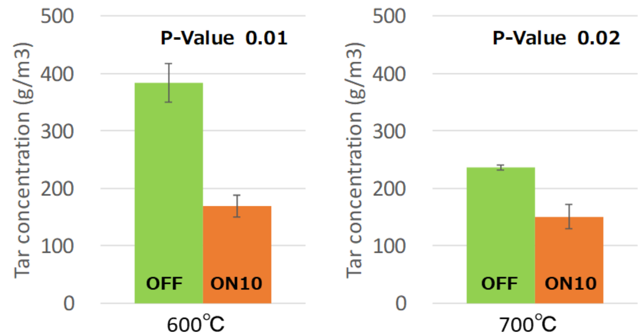

Fig.5 Tar concentration in the syngas (cellulose)

Similarly, the concentration of tar in the syngas of polypropylene was also reduced by the electron injection. As shown in Fig.6, the p-value<0.05 was obtained at 700°C, confirming the statistical superiority difference. However, at 600°C, although the tar concentration decreased on average with electron injection, there was no statistical difference.

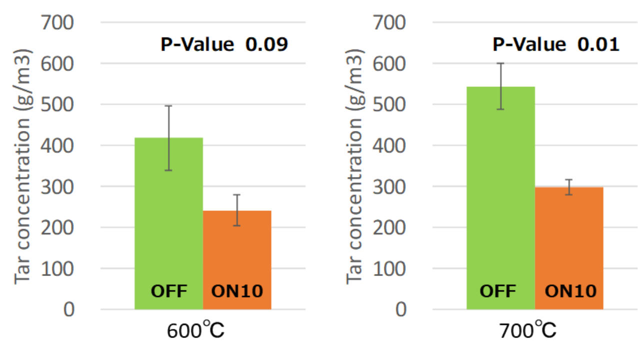

Fig.6 Tar concentration in the syngas (Polypropylene)

The results of GC/MS analysis are shown next. Fig.7 shows Cell700OFF and Cell700ON10 in one figure. Similarly, Fig.8 shows PP700OFF and PP700ON10 combined in one figure. As shown in Fig.7 and 8, the major peaks were detected in the same way regardless of the electron injection.

Fig7. two spectra on a single screen at 700°C (cellulose)

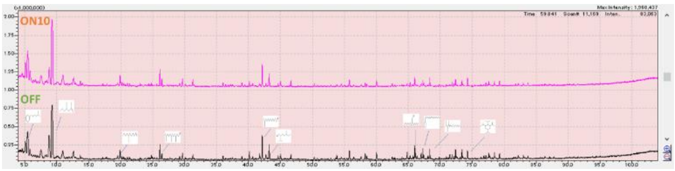

Fig.8. two spectra on a single screen at 700°C (Polypropylene)

On the other hand, the height of each peak changed due to the effect of electron injection. A comparison of the height of the major peaks is shown in Fig. 9. The height of the electron OFF was calculated as 100%.

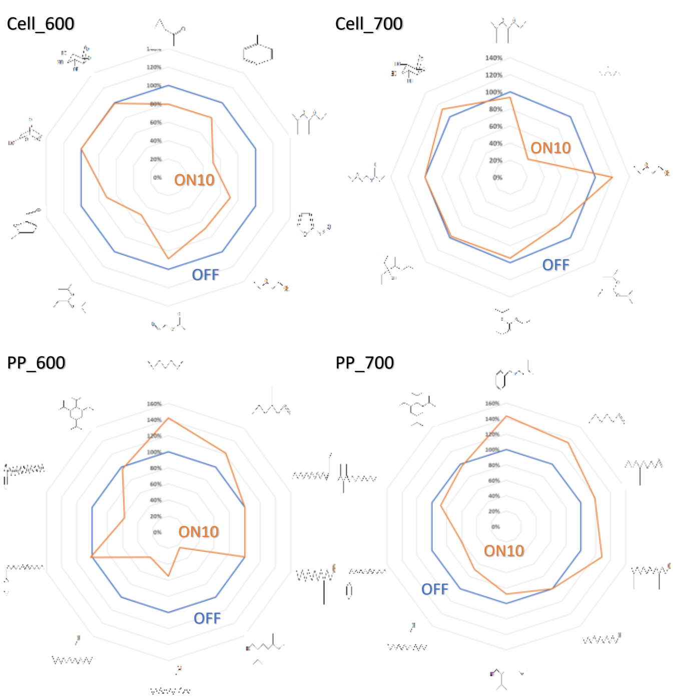

Fig.9 Intensity ratio of major peaks

In the case of cellulose, the trend was different between 600°C and 700°C. At 600°C, the height of the main peak decreased with overall electron implantation.

On the other hand, at 700°C, certain substances either decreased or increased, while other substances showed little difference. When the feedstock was polypropylene, the amount of material with a relatively small molecular weight increased and the amount of material with a large molecular weight decreased when electrons were injected. This trend was consistent regardless of the temperature.

In plasma modification, elementary reactions in which H_2O, O_2, and N_2 are activated by the presence of electrons, and elementary reactions in which electrons react directly with organic compounds have also been proposed. [4][5] Although the temperature of the reaction field was different, it is possible that a similar reaction occurred in this study. Tamer et al. found a good agreement between the experimental and simulation results for the oxidation pyrolysis reaction when electrons were injected. [2] In the present study, the increase in the rate of pyrolysis gas production was also confirmed, and the trend was consistent.

2.5 Conclusions

In this study, the effect of electron injection on the tar produced in the oxidative pyrolysis reaction was experimentally confirmed using a fixed-bed experimental equipment. Cellulose, a biomass material, and polypropylene, a petroleum-derived material, were used as raw materials. The results showed that the presence of electrons in the oxidative pyrolysis reaction reduced the amount of tar produced and changed its composition.

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