

Naphthalene destruction performance from tar model compound using a gliding arc plasma reformer

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Abstract—Recycling of various wastes such as sewage sludge requires an energy conversion process like thermal pyrolysis/gasification. During the process, tar and syngas are produced, but the tar brings trouble in pipelines and creates operating problems for the facility. In this study, to investigate naphthalene destruction in a gliding arc plasma reformer, parametric experiments were achieved in the variables that can affect the destruction efficiency. And through the parametric studies, the optimal operating conditions and the results were taken. For the parametric studies, steam input amount (steam/carbon ratio), input discharged power SEI (specific energy input), total feed gas amount, input naphthalene concentration, and electrode length were selected for experiments. Optimal conditions were 2.5 of S/C ratio, 1 kWh/m³ of SEI, 18.4 L/min of total gas amount, 1% of input naphthalene concentration, and 95 mm of electrode length. The corresponding maximum destruction efficiency of naphthalene was 79%, and energy efficiency showed 47 g/kWh.

Key words: Tar Model Compound, Gliding Arc Plasma, Reformer, Thermal Destruction, Pyrolysis/Gasification

INTRODUCTION

Through a thermal decomposition gasification process, wastes like a biomass, solid waste, organic sewerage sludge etc, can be used as an alternative source of energy [1]. The product gas formed from biomass gasification contains the major components CO, H₂, CO₂, CH₄, and N₂, in addition to organic (tars) and inorganic (H₂S, HCl, NH₃, alkali metals) impurities and particulates. This syngas is mostly composed of carbon monoxide and hydrogen, which can be utilized in IGCC (integrated gasification combined cycle), and IGFC (integrated gasification fuel cell), etc [2].

The organic impurities range from low molecular weight hydrocarbons to high molecular weight hydrocarbons. The lower molecular weight hydrocarbons can be used as fuel in gas turbine or engine applications. The higher molecular weight hydrocarbons are collectively known as “tar”. However, tar formation during thermal pyrolysis/gasification is a major problem for adoption. At ambient conditions tar condenses or polymerizes into more complex structures in exit pipes, heat exchangers and particulate filters, leading to choke and attrition, which can result in the decrease of total efficiency and an increase in the cost of the process.

Therefore, the aspect of tar cracking or removal during gas cleaning-up is one of the most important technical uncertainties in implementation of gasification technology [3]. To solve this issue, numerous researches have been conducted, and they are divided into physical and chemical methods. As a physical approach, scrubber, cyclone filter, wet-type electrical dust collector, activated carbon, etc., are available. Regarding chemical methods, catalysis, thermal cracking, partial oxidation, plasma destruction, and so on can be the best possible solutions. A scrubber is suitable for large-sized

plants and for low environmental contamination, but its cost is one of the most significant matters. Catalysis is applicable but at risk to sulfuric, chlorine, and nitrogen compounds, and coke is easy to form. So, recent researchers using plasma have been taking the initiative to overcome these problems. Especially, non-thermal plasma technology should be used to destruct the tar under low pressure or atmospheric state, and power consumption is relatively low [4].

After fundamental study on pulsed non-thermal plasma cracking for tar removal, higher efficiency of tar removal has been exhibited due to the formation of radical in comparison to the existing thermal and catalytic cracking [5]. However, installation cost and short life cycle of pulse power supply is the key for implementation. Besides these studies on the removal of VOCs (volatile organic compounds) like light aromatic tars, such as benzene, toluene, xylene, etc., are required. Reduction technologies of VOCs using plasma are mainly based on corona discharges, dielectric barrier discharges (DBD), gliding arc discharges, etc. These methods show high energy efficiency, and are not affected by the type and concentration of VOCs. This feature gets additional attention from the public [6-8].

However, corona discharge and DBD have a significant effect on reformer flow rate, and density of plasma is relatively low. They can be applied to scientific research, but commercial potential is low. In addition, selectivity during reforming reaction is difficult to control. Meanwhile, gliding arc discharge features quick start-up performance within few seconds, and easier control on reaction. Along with these, higher destruction efficiency and low energy utilization can be achieved. This method is developing as a new energy alternative [9,10].

A gliding arc plasma reformer developed in our reforming study [4] was used for tar destruction. Parametric studies on the factors that can affect the destruction and energy efficiencies of naphthalene were conducted to discover tar destruction characteristics. And through the parametric study, optimal conditions and their results were taken

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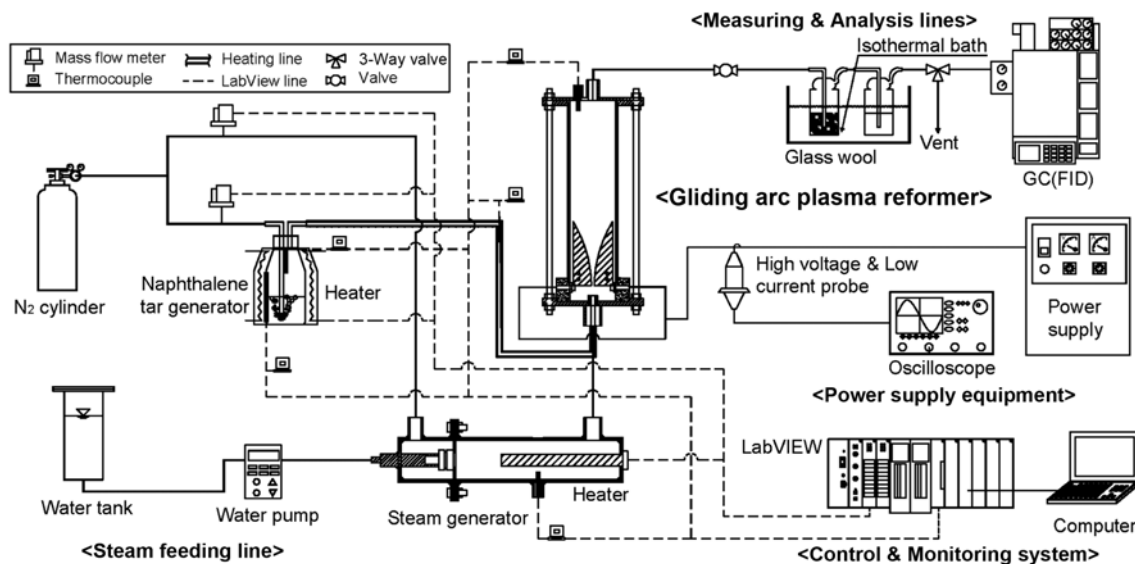


Fig. 1. Schematic of a gliding arc plasma experimental setup.

to guide operating patterns.

EXPERIMENTAL APPARATUS AND METHODS

1. Experimental Equipment

Fig. 1 shows a schematic for a gliding arc plasma experimental setup, which consists of gliding arc plasma reformer, steam feeding line, gas feeding line, power supply equipment, measuring and analysis lines, control and monitoring system. The gliding arc plasma reformer consisted of a quartz cylinder (55 mm in diameter and 200 mm in heights) to see the inside and three conical electrodes which are set at 120 degrees radially. Gas nozzle diameter and electrode gap distance were 1.5 mm and 3 mm, respectively. The electrode length was designed to be changed for 70, 80, and 95 mm. External wall of the plasma reformer was made of a quartz tube for insulation and inspection through a window. Ceramic material (Al_2O_3 wt 96%) was utilized inside the bottom for insulation and electrode fixation.

Steam was fed through a steam generator and feeding pump (KNF, STEPDOS03, Switzerland) with the water converted to steam. Gas feeding line utilized MFC (LINETECH, M3030V, Korea) with 0-1 L/min of range for naphthalene generation with nitrogen. Regarding dilution nitrogen, MFC (BRONKHOST, F201AC-FAC-22-V, Netherlands) with 0-20 L/min of range was adopted. High voltage power supply (Unicon Tech., UAP-15K1A, Korea) was implemented to provide stable plasma discharge into the reformer. As measuring equipment, high voltage probe (Tektronix, P6015, USA) and current probe (Tektronix, A6303, USA) were installed to determine electricity characterization for plasma reformer, and data logger (KIMO, KTT300, USA) was utilized for real-time monitoring of inside temperature. Gas chromatograph (SHIMADZU, GC-14B, Japan) with FID detector and DB-1 column (30 m \times 0.25 mm, 0.25 μm) was used for analysis.

Control and monitoring system utilized LabVIEW (National Instrument LabVIEW 8.6, USA) for water pump, heater of steam generator, and nitrogen gas flow rate. The temperature and experimental

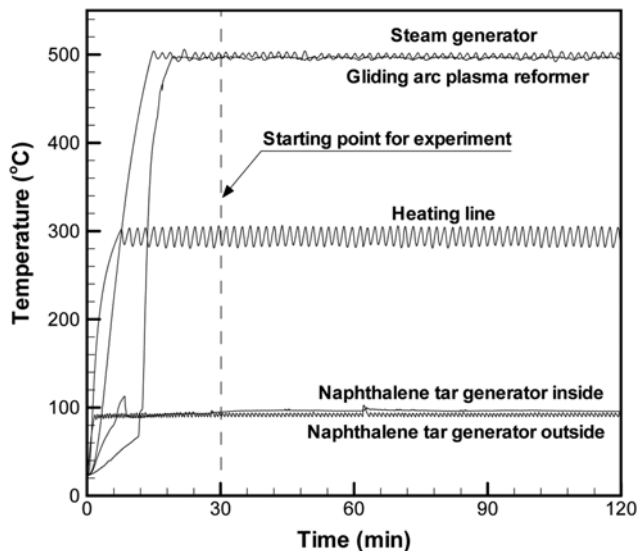


Fig. 2. Initial operation characteristics and stabilization condition.

conditions were continuously monitored with this.

2. Experimental Method

Fig. 2 displays the initial operation characteristics and stabilization condition for each part in the experimental test rig. The tests were achieved after the temperatures were stabilized at each part.

Temperature of the steam generator was set to about 500 °C. Temperature of the naphthalene line was maintained at 300 °C. For the naphthalene tar generator, the outside wall of reactor was set to 90 °C. The internal temperature of the gliding arc plasma reformer was set to 500 °C.

At this state, 0.4 L/min of nitrogen into a naphthalene generator and 8 L/min of nitrogen into a steam generator were introduced. Both gases were mixed after the generator and they entered into the plasma reactor. At the same time, discharge was formed at the gliding arc plasma reformer.

Table 1. Experimental conditions in each parameter

Conditions	Steam/Carbon ratio	Specific energy input (kWh/m ³)	Total gas flow rate (L/min)	Input naphthalene conc. (%)	Electrode length (mm)
Range limits	1.2-4.5	0.77-1.17	10.5-27.6	0.6-1.3	70-95

Sampling of naphthalene tar was made through sampling ports both at inlet and outlet. Sampled gas was analyzed by using a gas chromatograph FID. The GC was set to 200 °C for injector and 280 °C for detector. And oven temperature was raised in 10 °C/min for 40-320 °C, and then 320 °C at the oven temperature was maintained for 20 min.

Parametric studies were achieved in steam to carbon (S/C) ratio, specific energy input (SEI), total feed gas amount, input naphthalene concentration and electrode gap change. The test conditions are shown in Table 1.

3. Data Analysis

3-1. Naphthalene Destruction Efficiency

Destruction efficiency of naphthalene was derived from Eq. (1) [9].

$$\eta_e(\%) = \frac{[\text{Naphthalene}]_{\text{inlet}} - [\text{Naphthalene}]_{\text{outlet}}}{[\text{Naphthalene}]_{\text{inlet}}} \times 100 \quad (1)$$

where, $[\text{Naphthalene}]_{\text{inlet}}$ is input naphthalene concentration (%), and $[\text{Naphthalene}]_{\text{outlet}}$ is naphthalene outlet concentration (%).

3-2. Energy Efficiency

Energy efficiency was calculated using Eq. (2) [8].

$$\eta_e(\text{g/kWh}) = \frac{[\text{Naphthalene}]_{\text{removal}} \times \text{Total gas flow rate}}{\text{Power input}} \times 100 \quad (2)$$

where $[\text{Naphthalene}]_{\text{removal}}$ is a removal concentration of naphthalene (%), total gas flow rate is an input gas amount into a plasma reformer (g/h), and power input is plasma input power (kW).

3-3. Specific Energy Input [8]

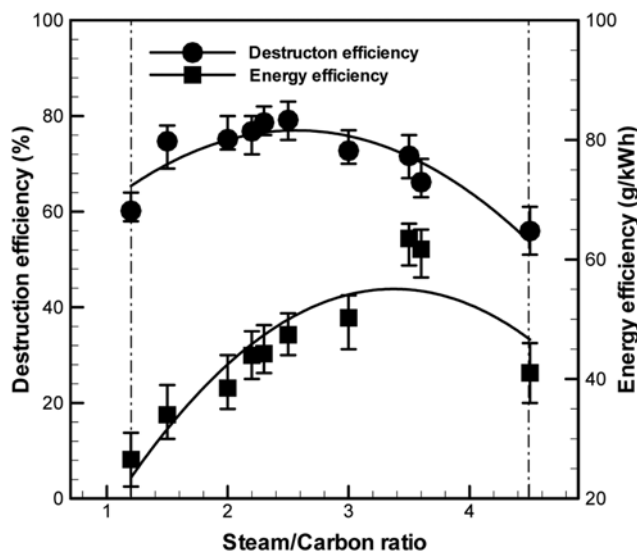
Specific energy input (SEI) is the ratio of an input energy and process gas amount, which is denoted as Eq. (3).

$$\text{SEI (kWh/m}^3) = \frac{\text{Discharge power (kW)}}{\text{Total gas flow rate (m}^3/\text{h)}} \quad (3)$$

where discharge power is power for plasma (kW), and total gas flow rate is amount of gas into a reformer (m³/h).

RESULTS AND DISCUSSION

In this study, a gliding arc plasma reformer was used for the de-

**Fig. 3. Effect of steam feed amount.**

struction of tar which produced during pyrolysis process. Naphthalene as a tar model compound was selected for the test on a parametric study and optimal operation characteristics.

The optimal operating conditions and their results are shown in Table 1. Carbon balance means that naphthalene destructed to almost the decomposed gas which tells some other portion to carbon black. The carbon black was lower 1% due to steam reforming.

1. Effect of Steam Feed Amount

Fig. 3 shows naphthalene destruction and energy efficiency depending on steam flow amount change (S/C ratio). For a plasma reformer, carbon black was formed when S/C ratio was less than 1.2, and more than 4.5 of S/C ratio showed the decrease of temperature at a steam generator because of the increase in water content. Therefore, S/C ratio was changed between 1.2 to 4.5.

Destruction efficiency of naphthalene was gradually increased with S/C ratio, and showed the maximum at 2.5 of S/C ratio as 79% and after this point, it was decreased.

Steam injection on the plasma discharge in the reformer leads to the direct formation of hydroxyl radicals, which react with naphthalene for destruction [11].

Table 2. Optimal operating conditions and their results

Conditions	Steam/Carbon ratio	Specific energy input (kWh/m ³)	Total gas flow rate (L/min)	Feed gas temp. (°C)	Input naphthalene conc. (%)	Electrode length (mm)	Nozzle diameter (mm)	Electrode gap (mm)	
Reference values	2.5	1	18.4	500	3.6	95	1.5	3	
Results	Decomposed gas composition (% , N ₂ excluded)						Carbon balance (%)	Naphthalene destruction efficiency (%)	Energy efficiency (g/kWh)
	CO	CO ₂	C ₂ H ₂	C ₂ H ₆	C ₃ H ₈	H ₂			
	13.4	50.8	1.2	4.8	6.0	23.7	99	79	68

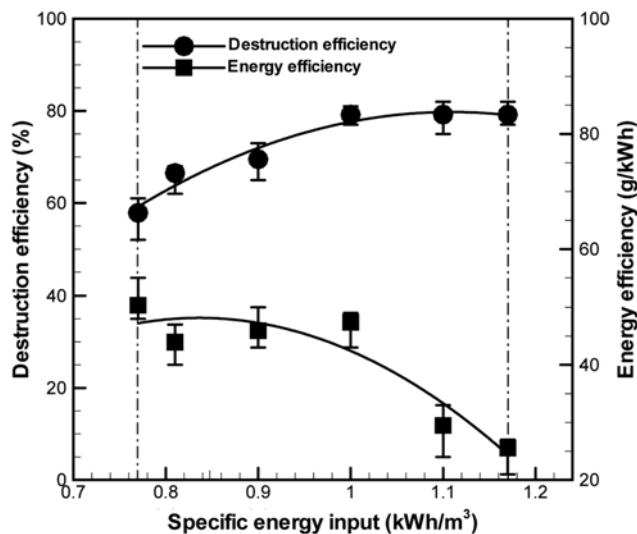


Fig. 4. Effect of specific energy input.



This could be explained the reason why the destruction efficiency should be increased. However, water also has an adverse effect on toluene removal due to its electronegative characteristics. Too many water molecules limit the electron density in the system and quench the activated chemical species. The reason is why the destruction should be decreased. Therefore, controlled steam is essential for naphthalene destruction [8].

In addition, increase in steam input will add the amount of total gas in a reformer, and it does not provide sufficient holding time for naphthalene destruction. It will reduce the naphthalene destruction efficiency. For energy efficiency, S/C ratio means the amount of total gas, and efficiency increases up to 63 g/kWh at 3.5 of S/C ratio. After this point, decrease in holding time from an increased amount of total gas will reduce the efficiency.

2. Effect of Input Discharged Power

Fig. 4 displays the effect of input discharged power, that is, specific energy input (SEI). The SEI change was made between 0.77-1.17 kWh/m³ by changing input discharged power from 0.85 to 1.33 kW. If the SEI was increased, gradual increase in naphthalene destruction efficiency was exhibited, and an average of 79% of destruction efficiency was achieved at higher than 1.0 kWh/m³. High power, i.e., high SEI, gives more available electrons to initiate the destruction of naphthalene by collision. It is a fact that the quantity of electric transfer between electrode increases with increasing applied voltage with fixed geometry of an electrode [12]. In addition, energy efficiency is significantly decreased from 50 g/kWh to 26 g/kWh along with the increase in SEI.

The plasma reformer in this study shows a relatively higher efficiency in comparison to the one from existing research which showed about 70% of naphthalene destruction efficiency along with 2.38 g/kWh of energy efficiency [13].

3. Effect of the Total Feed Gas Amount

Fig. 5 shows the effect of a total feed gas amount. Total feed gas was controlled within the range of 10.5-27.6 L/min.

With larger than 10.5 L/min of total feed gas, intermittent dis-

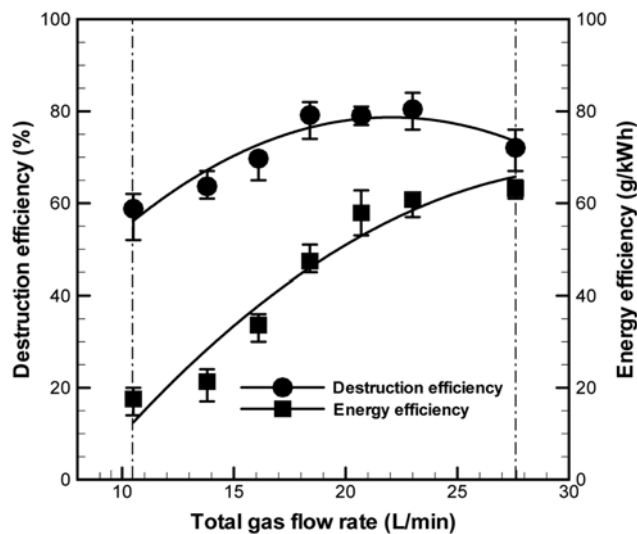


Fig. 5. Effect of total gas flow rate.

charge was formed and stable plasma discharge was made over 16 L/min. Therefore, 10.5 L/min of a total feed gas amount shows 59% of naphthalene destruction efficiency due to the instability of plasma discharge. Greater than 16 L/min achieved more than 70% of the destruction efficiency, and 18.4 L/min displays 79% or higher destruction efficiency.

It may be explained that stable plasma discharge should be needed in proper injecting gas amount (i.e., injection gas velocity) for injection momentum longitudinally to prevent diffusion loss to outside of plasma column.

Passing 23 L/min of a gas amount shows a decrease due to the reduction of holding time at discharge area by increased flow rate of gas within the same size of nozzle diameter. In general, reduction of naphthalene destruction efficiency from an increase in a total feed gas amount is made regardless of chemical structure, and it is related with the reduced electron collision between reactive ion and radical because of a shorter amount of interaction between plasma

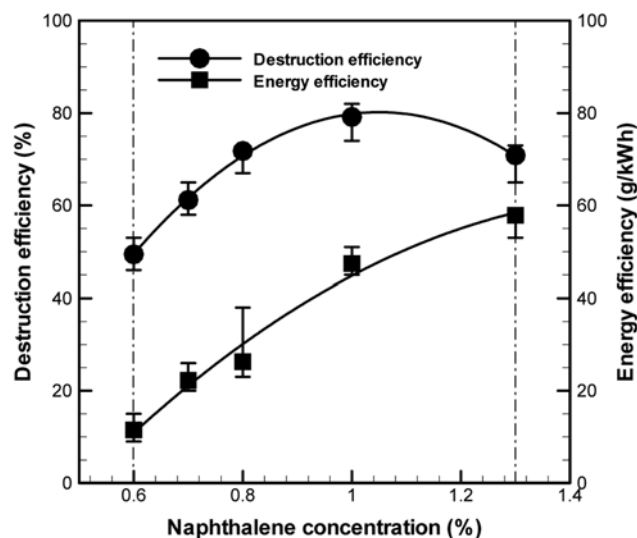


Fig. 6. Effect of input naphthalene concentration.

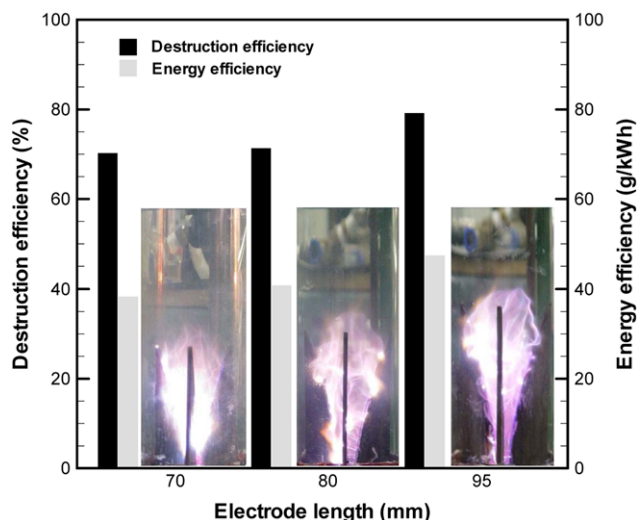


Fig. 7. Effect of various electrode lengths.

discharge area and naphthalene [14].

Regarding energy efficiency, increase in a total feed gas amount shows a significant increase in energy efficiency.

4. Effect of Input Naphthalene Concentration

Fig. 6 shows the effect of input naphthalene concentration. Experiment is conducted by fixing the flow rate of diluting nitrogen at 10 L/min and varying the input naphthalene amount between 0.1-0.5 L/min. As a result, input naphthalene concentration ranges from 0.6-1.3%. Destruction efficiency of naphthalene shows 49%, when input naphthalene is 0.6%. Along with an increase in input naphthalene concentration, destruction efficiency is increased from 1% to 79%. In general, low input concentration shows higher destruction efficiency in comparison to the higher one [8]. When input naphthalene concentration is low (up to 1%), discharge is made intermittently. Gradual increase in input concentration will ensure higher naphthalene destruction efficiency. For more than 1% of input naphthalene concentration, moisture content slightly reduces the efficiency. Regarding energy efficiency, increase in input naphthalene concentration shows a gradual increase in energy from 12 g/kWh to 58 g/kWh.

5. Effect of Electrode Length

Fig. 7 displays the picture of consequences on naphthalene destruction efficiency, energy efficiency, and plasma discharge by changing electrode length. Electrode length is varied within 70, 80, and 95 mm, respectively, and other variables are available in Table 1. Naphthalene destruction efficiency is increased from 70% to 79% when electrode length is increased from 70 to 95 mm. Energy efficiency shows a similar trend of an increase from 38 g/kWh to 47 g/kWh. As shown in the plasma discharge photograph, a longer electrode length shows a large area of discharge, and passing time of gas will be increased. Therefore, naphthalene destruction efficiency and energy efficiency are increased.

CONCLUSION

A gliding arc plasma reformer was used for the destruction exper-

iment in naphthalene, which is a tar model compound. Parametric studies were achieved in S/C ratio, SEI, total feed gas amount, input naphthalene concentration, and electrode length.

Naphthalene destruction efficiencies in steam feed amount, total feed gas amount, and input naphthalene concentration showed the maximum value at a specific point. Energy efficiency in steam feed amount had the maximum value. But in the case of total feed gas amount, the input naphthalene concentration increased with increasing each variable. However, in the case of input discharge power, the destruction efficiency increased with increasing the power, and energy efficiency decreased. Along with the increase in electrode length, the electrode should be as long as possible until discharge is formed.

Through the parametric screening studies, optimum operating condition was taken in showing maximum tar destruction. The conditions are 2.5 of S/C ratio, 1.00 kWh/m³ of SEI, 18.4 L/min of a total feed gas amount, 1% of input naphthalene concentration, 95 mm of electrode length. And at this condition, naphthalene destruction efficiency and energy efficiency were 79% and 47 g/kWh, respectively.

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