

## Development of process model of a rotary kiln for volatile organic compound recovery from coconut shell

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**Abstract**—The volatile organic compounds (VOCs) contained in coconut shell are wasted in the carbonization process of coconut shell due to the difficulty of recovery. The VOCs recovery is useful and necessary, because the VOCs are a sustainable energy source, and the recovery is an economically feasible project. A simulation model of the VOC recovery process from coconut shell using a rotary kiln is developed to investigate the process characteristics and the role of model parameters. The model includes the energy and material balances for the processing solid and the gas in the kiln. The validity of the proposed model is partially examined with the experimental results. From the simulation, the dominant heat transfer mechanism is determined for the understanding of the process operation. In addition, the optimal operating conditions of the rotary kiln are found for the use in the design and control of the kiln.

Key words: Process Model, VOC Recovery, Rotary Kiln, Heat Transfer

### INTRODUCTION

Carbonization has a long history because charcoal, its product, has been a favorite fuel for cooking. In the metallurgical industry a large amount of charcoal is used as the most useful reductant. Carbonization is a relatively simple process, high temperature treatment without air supply. Though it has a long history and a wide variety of applications, not many studies have been conducted until various new feed materials, such as solid waste and sewage sludge have been introduced. The carbonization of these new materials is no longer a simple heating process because of various components contained in the materials. The conventional feeds of carbonization are mostly wood and coal. In addition, the recent rise of energy cost in industries and the reduction of carbon dioxide emission as a solution of the global warming has invited many possible eco-friendly sources of new energy. The recovery of volatile organic compounds (VOCs) from coconut shell carbonization fits these requirements. Conventionally the coconut shell has been carbonized in a simple hearth with partially smothering environment, and the produced VOCs in the process escaped to the air. No recovery has been attempted due to the small scale of the process and lack of necessity. By introducing a large-scale carbonization process, the valuable VOCs can be recovered for a possible energy source. Because the VOCs contain many different fatty acids having a wide range of boiling points, the temperature control of the processing equipment, a rotary kiln, is very important. Especially, the design and insulation of the conduit transporting the recovered VOCs in gas phase between processing equipments cause many difficulties.

Charcoal from coconut shell is widely used in the production of activated carbon, and large amounts of VOCs are wasted during the charcoal production. Because most carbonization processes are small and the coconut shell is heavy for its transportation, the coco-

nut shell is simply carbonized in an empty, half-open drum. Though the wasted amount of VOCs from the raw coconut shell ranges from 62.5% to 74.4% according to the carbonization temperature [1], the VOCs have not been recovered until recently. Many attempts have been made to recover the VOCs and to use them in the processes consuming large amounts of energy, such as steam boilers and power generation plants.

The production of activated carbon uses the charcoal as a raw material, and the production plants of the activated carbon have been located in the developed countries consuming most of the activated carbon. Therefore, the transportation cost of the raw material from tropical countries producing the coconut limits shipping the raw coconut shell. Instead, the processed coconut charcoal about one-third in weight of the coconut shell has been used as the raw material due to the transportation cost. Recently, production plants of activated carbon have been built in the tropical countries cultivating coconut trees, and the production of the activated carbon is rising. In the carbon activation process fuel oil has been used as an energy source to elevate the gas temperature in the rotary kiln, but the recovered VOCs can replace the oil when the carbonization of the coconut shell is carried out in the same process.

The activation of the coconut charcoal is yielded in the reductive gas environment, and the reductive gas environment is formed when the water gas reaction occurs to generate hydrogen. Though the reaction temperature is between 850 °C and 1,100 °C [2], carbonization occurs at much lower temperature. Unlike most chemical processes handling liquid or gas, the production of the activated carbon involves solid material processing. The carbonization process of this study has a similar process of material handling. Two main works in the activation are the heating of the solid material and maintaining the reductive gas environment, but the carbonization process has heating only.

Coal pyrolysis in a rotary kiln is similar to the carbonization process of coconut shell except using different heating methods. In coal pyrolysis an internal burner of oil or gas provides the necessary heat

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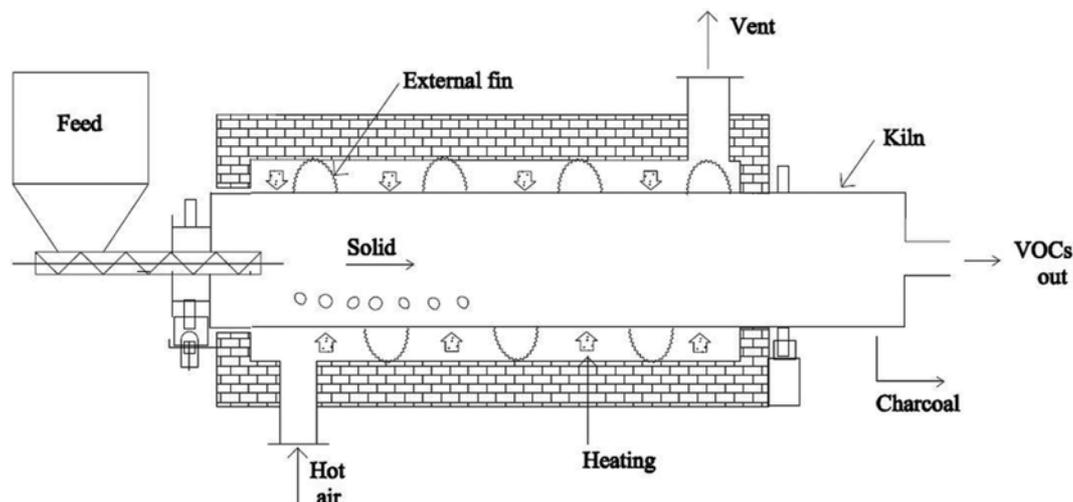


Fig. 1. A schematic diagram of the rotary kiln for the carbonization of coconut shell.

to raise the gas temperature. Instead, the carbonization process of coconut shell uses external heating [3-5]. When the internal burner used in coal pyrolysis is used in the VOC recovery process, the recovered VOC burns and the VOC yield drops sharply. The rotary kiln for the coal pyrolysis has been modeled to examine the role of operating parameters, such as the kiln rotation speed for the process optimization [6,7]. A review of the carbonization technology was also published to explain the production technique and its relation with the charcoal properties [8].

Though most industrial processes of activated carbon production use a rotary kiln and coconut charcoal is their feed material, several processes of different raw materials have been developed recently. Cangialosi et al. [9] investigated the oxidation of coal in a bench-scale rotary kiln. In the modeling of the process, an advective-dispersion model was utilized to determine the axial dispersion coefficient. The reaction rate constants were obtained by thermal analyses, and the residence time distribution of ashes was used to calculate combustion efficiencies in the rotary kiln. The energy recovery from municipal solid waste using a rotary kiln has been introduced by Baggio et al. [10]. The proposed process produces char, tar and syngas for the energy sources (19.2 MJ/kg) of almost the same heating value of wood (19.3 MJ/kg). A simple equilibrium model and the equation of the solid waste pyrolysis were developed to predict the syngas composition and tar and char fractions. The production of activated charcoal and syngas from sewage sludge using a rotary kiln has also been proposed by Chun et al. [11]. The optimal operating conditions of the process have been experimentally determined for the high adsorption performance of the yielded activated charcoal and the production of syngas having high heating value. Instead of the thermal activation a chemical activation of paper mill sludge has been studied by Kang et al. [12], and the kinetics of water gas reaction was explored in a hydrogen production process [13]. The surface modification and adsorption application were also investigated with a wood-based activated carbon [14]. Though the carbonization and activation processes have been examined in many studies [2,6-8,15], the recovery of the VOCs is not handled as a significant topic.

In this study, the process model of carbonization of coconut shell

utilizing a rotary kiln is developed, and the role of parameters in the heat transfer at the kiln is examined to show the difference between the process models of carbonization and activation. The parameters include the convective heat transfer coefficients of wall to solid and wall to gas heat transfers. The optimal operating conditions of the rotary kiln are searched for the design and control of the kiln. In addition, the production and handling of the VOCs are explained as well.

#### DESCRIPTION OF CARBONIZATION KILN

The rotary kiln is a cylindrical tube having screw-type fins attached outside of it for the enhanced heat transfer as demonstrated in Fig. 1. The cylinder rotates and is slightly inclined for the mixing and movement forward of the processing material. The structural difference between the carbonization kiln and the activation kiln comes from the heating system. While the activation kiln utilizes an internal heating and the insulation inside the kiln wall, the carbonization kiln does an external heating. Therefore, a brick chamber is

Table 1. Equipment characteristics and operating conditions

Length (m)	1.2
Internal diameter (m)	0.2
Raw material	Coconut shell
Solid flow rate ( $\text{kg h}^{-1}$ )	12
Moisture content (%)	0
Yield (%)	33
Inclination	5/1000
Insulation thickness (m)	0.05
Rotary kiln pressure	Atmospheric
Rotation speed (rpm)	4
Residence time (h)	1
Solid input temperature ( $^{\circ}\text{C}$ )	Room temperature
Solid output temperature ( $^{\circ}\text{C}$ )	200
Inlet gas temperature ( $^{\circ}\text{C}$ )	Room temperature
Outlet gas temperature ( $^{\circ}\text{C}$ )	310

necessary for the insulation as shown in Fig. 1. The carbonization process is an operation of relatively low temperature, and the air supply is limited to prevent the burning of processing material. For the simulation of the kiln the process data from a pilot kiln is utilized here. The dimension of the kiln and the operating data of pilot test are shown in Table 1. The values listed in Table 1 and the schematic in Fig. 1 were adopted from a pilot rotary kiln. It is horizontally placed and rotated at the speed of 4 rpm. The processing time of the feed stock is one hour. At the inlet of feed, a screw feeder is installed to prevent air supply. Two rollers support the kiln as shown in Fig. 1. The front rollers hold the kiln from moving horizontally, while the rear rollers control the horizontal sliding of the kiln due to thermal expansion. Two pieces of rollers are placed each location for stable support. The product, charcoal, is evacuated from the rear end of the rotary kiln. When the hot charcoal contacts air directly, an explosion occurs. Therefore, a large chamber of excessive space is equipped at the rear section of the rotary kiln for the cooling of the charcoal. However, the chamber was not considered in the modeling of this study. The VOCs produced from the raw material are evacuated from the rear of the rotary kiln with the processed charcoal. When a large chamber is installed for the charcoal cooling, the chamber temperature has to be controlled to prevent the liquefaction of the VOCs. The VOCs in vapor phase are easier to transport, and the heat recovery is more efficient due to its high temperature. While typical flow rates of liquid are between 0.15 m/s and 0.3 m/s, those of gas are between 9 m/s and 30 m/s [16]. The practical moisture content in raw coconut shell is 10%, but the moisture content is eliminated for the simple calculation and matching with the TGA data shown in Fig. 2.

### MATHEMATICAL MODEL

Material balances are used in the simulation of temperature distributions of solid and environmental gas in the rotary kiln. Though various kinds of heat transfers among solid, gas and kiln wall occur in the processing of the solid, the heat transfer between the kiln wall and solid is dominant for monitoring the carbonization process. The

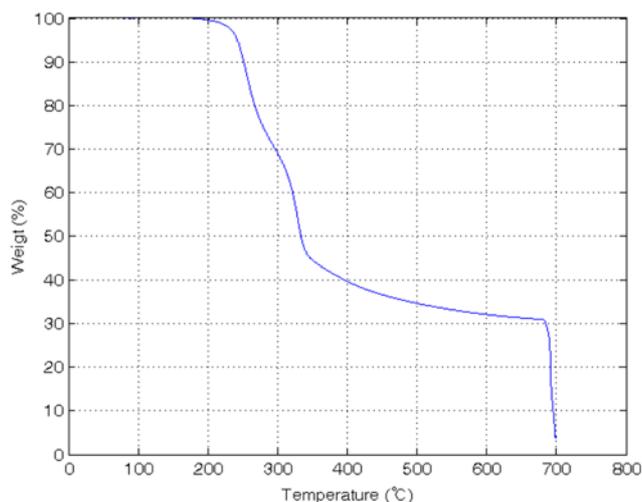


Fig. 2. Weight variation with temperature increase in the thermal gravimetric analysis of dry coconut shell.

main target of the carbonization process is raising the solid temperature to a desired level. An external heat supply is provided through the kiln wall, and the mechanism of the heat transfer is convection. Though the heat transfer is between solid kiln wall to solid material, the mechanism of heat transfer is convection because the solid material is particles and is constantly mixed to move forward like a fluid. The carbonization process is simpler than the activation process. The processing temperature is also about 300 °C lower than the activation, and there is no significant chemical reaction except the vaporization of VOCs [15]. The two processes use a rotary kiln, and the main process is heat transfer. Therefore, the system equations for the charcoal activation [17] can be used in this study. The assumptions used here are as follows:

- There is no axial mixing of solid and vapor.
- There is no radial variation of process variables.
- Variation of kiln wall temperature in angular direction is negligible.
- Pressure in the kiln is constant.
- Axial heat transfer of conduction is neglected.

When the heat transfer among the gas, solid and kiln wall is considered, the following energy balances for gas, solid and kiln wall can be formulated from Ortiz et al. [2] and Kim [17]:

$$\frac{1}{V_g} \frac{\partial(Q_g C_g T_g)}{\partial t} = \frac{\partial(Q_g C_g T_g)}{\partial z} - C_1(T_g - T_s) - C_2(T_g^4 e_g - T_s^4 A_v) - C_3(T_g - T_w) - C_4(T_g^4 e_g - T_w^4 A_v) \quad (1)$$

where the left hand side is the energy accumulation term, and the first of the right hand side denotes axial convection of gas flow. The next terms are the convection and radiation between gas and solid and the convection and radiation between gas and kiln wall, respectively.

In the same manner, the energy balance for the solid material is

$$\frac{1}{V_s} \frac{\partial(Q_s C_s T_s)}{\partial t} = \frac{\partial(Q_s C_s T_s)}{\partial z} - C_1(T_g - T_s) - C_2(T_g^4 e_g - T_s^4 A_v) + C_5(T_w - T_s) + C_6(T_w^4 e_w - T_s^4 A_v) - \frac{\partial Q_v}{\partial z} H_v \quad (2)$$

where the terms from the second of the right hand side are the convection and radiation between gas and solid, the convection and radiation between wall and solid, and heat consumption for the vaporization of VOCs, respectively. The energy balance for the kiln wall is found from the same procedure as follows:

$$\frac{\partial(M_w C_w T_w)}{\partial t} = \frac{\partial(M_w C_w T_w)}{\partial z} + C_3(T_g - T_w) + C_4(T_g^4 e_g - T_s^4 A_v) - C_5(T_w - T_s) - C_6(T_w^4 e_w - T_s^4 A_v) \quad (3)$$

Material balance for the solid is also given as

$$\frac{1}{V_s} \frac{\partial Q_s}{\partial t} = - \frac{\partial Q_s}{\partial z} - \frac{h_v A (T_g - T_s)}{H_v} \quad (4)$$

where the terms of the right hand side are the axial mass flow and loss by the vaporization of VOCs. Similarly, the balance for the gas is given as

$$\frac{1}{V_g} \frac{\partial Q_g}{\partial t} = \frac{\partial Q_g}{\partial z} + \frac{h_v A (T_g - T_s)}{H_v} \quad (5)$$

For the steady state simulation of this study the unsteady state

**Table 2. Model parameters**

$$C_1 = h_{gs} L_{cu}$$

$$C_2 = \frac{\sigma L_{cu} e_s}{1 - (1 - e_s)(1 - A_v)}$$

$$C_3 = h_{gw} L_{li}$$

$$C_4 = \frac{\sigma L_{li} e_w}{1 - (1 - e_w)(1 - A_v)}$$

$$C_5 = h_{ws} L_{es}$$

$$C_6 = \sigma L_{cu} \phi_{sw} e_w e_s$$

$$\phi_{sw} = \left\{ \frac{1}{1 - e_g} - (1 - e_w) \left[ \frac{L_{cu}}{L_{li}} (1 - e_s) + \left( 1 - \frac{L_{cu}}{L_{li}} \right) \right] \right\}^{-1}$$

$$A_v = 0.07$$

$$C_g = 1.2$$

$$C_s = 0.2$$

$$C_w = 0.7$$

$$e_s = 0.1$$

$$e_w = 0.9$$

$$h_{gs} = 5$$

$$h_l = 4.3$$

$$h_{ws} = 35$$

$$h_{gw} = 9$$

$$L_{cu} = 0.1848$$

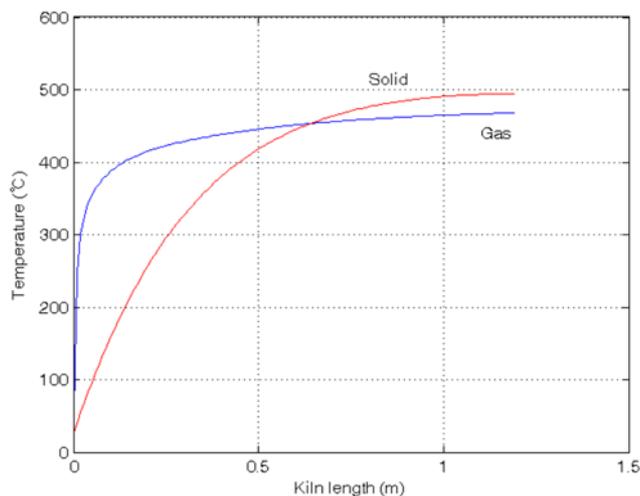
$$L_{es} = 0.2356$$

$$L_{li} = 0.3927$$

terms in the left hand side of the balance equations are eliminated, and a system of ordinary differential equations is formulated. The Euler integration was used in the numerical computation. The values of model parameters are listed in Table 2. The emissivities of solid and kiln wall are adopted from the coal pyrolysis model [7], and the data of the processing material are for the carbonization of coconut shell in a pilot test.

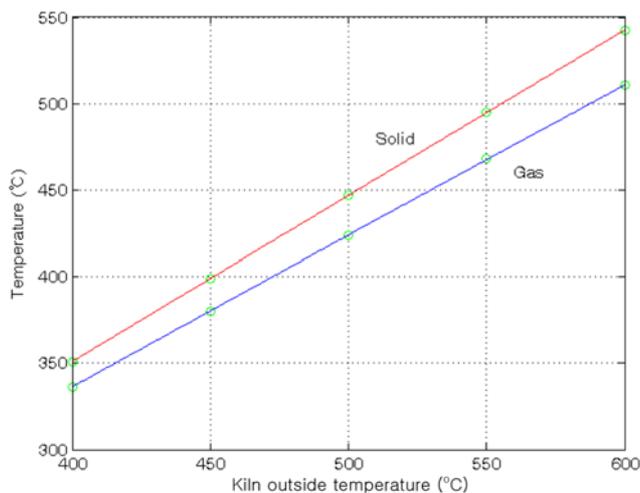
## RESULTS AND DISCUSSION

The composition of volatile organic compounds (VOCs) in the coconut shell charcoal is not important, when it is only used for the raw material of the activated carbon production. However, if VOC recovery is accompanied by the carbonization for an energy source, the VOC composition in the charcoal has to be minimized for the full VOC recovery. The processing temperature of the coconut shell determines the composition of the remaining VOCs in the coconut charcoal. The thermal gravimetric analysis (TGA) gives the information of the VOC content in the charcoal. Fig. 2 shows the result of the TGA of the dry coconut shell. There are three different regions: the first is up to 330 °C, the next is between 330 °C and 690 °C and the last is over 690 °C. In the first region the vaporization of the VOCs is very fast as seen in the figure. The sudden drop of weight indicates the fast vaporization of the VOCs. In the second the drop is slow and steady. Though more recovery of the VOC is desirable, raising the processing temperature too much for the recovery of the VOC in the second region is not economical. In this study the cut-off temperature was set at 500 °C after considering the economics of the temperature increase and VOC recovery. In the third region over 690 °C burning occurs, and therefore there is a sudden weight loss and the final weight becomes zero.

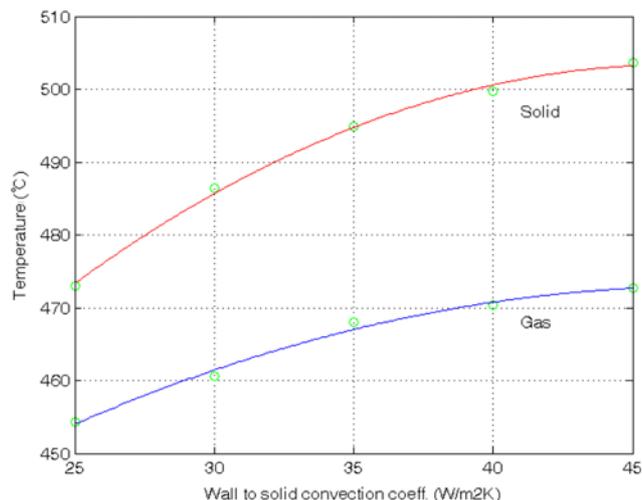


**Fig. 3. Temperature distribution of solid material and gas in the rotary kiln for the carbonization of coconut shell.**

The temperatures of solid and gas are calculated along the rotary kiln length, and they are plotted in Fig. 3. The external temperature of the kiln wall is maintained at a temperature of 550 °C. Near the inlet of the feed the gas temperature increases fast, while the solid temperature rises slowly. The main heat transfer in the kiln is the convection from the kiln wall to solid and gas. The heat capacity of solid is much larger than that of gas, and therefore the temperature elevation of solid near the feed entrance is slower than that of gas. But the kiln length is enough to raise the temperature to around 500 °C, which is the target temperature for the successful removal of the VOCs, some 65% of the raw coconut shell. The measured gas temperature at the middle of the pilot kiln was 430 °C, and it was comparable to the calculated temperature in Fig. 3. The calculated gas temperature at the middle of the kiln is 451 °C, which is close to the measured temperature. In another experiment [11], the measured gas temperatures in a rotary kiln were between 385 °C and 500 °C. The measured temperatures are comparable to the computed temperature indicated in Fig. 3. When paper sludge and olive pits were carbonized, the kiln was operated at 400 °C [12,18,19].



**Fig. 4. Effects of different kiln wall temperatures on solid and gas temperatures.**

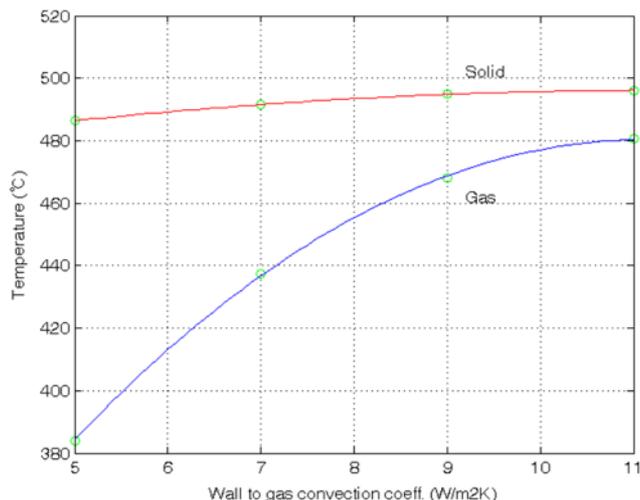


**Fig. 5. Effects of different heat transfer coefficients in kiln wall to solid convection on solid and gas temperatures.**

Though the measured temperature is a little lower than the computed temperature of this study, the predicted temperature from the model is reasonable to use in the design of the carbonization rotary kiln. The effect of kiln wall temperature on the solid and gas temperatures is shown in Fig. 4. The relation between the kiln temperature and the material temperature is almost linear. The plotted temperatures are the maximum over the whole kiln length for the given wall temperature. The solid and gas temperatures are simply displayed with the kiln outside temperature.

Because the convection from the kiln wall to solid and gas is the main heat transfer in the carbonization process, the value of convective heat-transfer coefficient is critical to calculate the temperature distribution in the kiln. The effect of the coefficient is demonstrated in Fig. 5. As the coefficient increases, both temperatures of solid and gas are elevated, and the increase of the solid temperature is twice as much as that of the gas temperature. Though the coefficient represents the convection between the kiln wall and solid, there is also the convection between the solid and gas as given in Eq. (1). In other words, the elevation of the convective heat-transfer coefficient raises heat transfer rate from the wall to solid first, and then the elevated solid temperature increases the gas temperature. But the increase is smaller than the solid temperature elevation because it is secondary. After considering the target temperature of 500 °C, we used 35 Wm<sup>-2</sup> K<sup>-1</sup> in this study. The calculation of the coefficient is explained in Patisson et al. [7]. In the study the values between 3 Wm<sup>-2</sup> K<sup>-1</sup> and 30 Wm<sup>-2</sup> K<sup>-1</sup> are suggested for the convective heat transfer, but the value used in this study is not far from the suggested values for the coal pyrolysis application in the previous study [7].

For the convective heat-transfer coefficient of wall to gas, the temperature variation is shown in Fig. 6. While the solid temperature is not affected by the coefficient variation, a large increase of gas temperature is observed as expected from the heat transfer between the kiln wall and gas. The value of 9 Wm<sup>-2</sup> K<sup>-1</sup> is employed in the simulation of this study after the result of a pilot test is compared. The role of gas emissivity in the radiation heat transfer was also examined, and no significant variation was found, unlike the



**Fig. 6. Effects of different heat transfer coefficients in kiln wall to gas convection on solid and gas temperatures.**

activation process. The adopted value of 0.065 from the other study [20] is used here. In the activation process of coconut charcoal, the gas temperature is much higher at about 950 °C [17], and therefore radiation heat transfer is dominant and emissivity widely changes the gas and solid temperatures.

The difference between the carbonization process of coconut shell and the activation process of coconut charcoal is that the carbonization process does not have a significant chemical reaction in its processing. Therefore, the control of the process operation is much simpler than that of the activation process. Maintaining the temperature of the external kiln wall is the key objective of the carbonization process operation. In the recovery of the VOCs from the process the wall temperature determines not only the amount of VOCs but also the condition of the VOCs. For the easy transportation of the VOCs, they have to be in vapor phase. Otherwise, the liquefied oily VOCs mixed with fine carbon particles clog up the pipe connecting the carbonization and activation kilns when the two processes are connected for the waste energy recovery from the VOCs. The control of the carbonization kiln temperature is more important to prevent from shutting down the connection in the combined processes.

The model parameters presented in this study are adjusted with the data obtained from the pilot test carried out at a local company producing the activated carbon using the coconut charcoal. The energy source of the activation process is currently bunker-C oil, and therefore the reduction of the energy cost has been pursued. One of the options is utilizing the VOCs from the carbonization process, which is also necessary process for the raw material preparation in the charcoal activation plant. It is the reason that the recovery of the VOCs is important in the production of the activated carbon.

## CONCLUSIONS

A model of the VOCs recovery process from coconut shell is proposed and simulated for the understanding of the process, which provides the basic knowledge of the kiln design and operation. From the simulation results, the convection from the kiln wall to the solid

and gas is the dominant heat transfer in the kiln, and the convective heat-transfer coefficient between the kiln wall and solid has the most significant role in the control of solid temperature determining product quality. The analysis of the model parameters indicates that the kiln wall temperature and solid and gas temperatures are almost linearly dependent. When the external wall of the rotary kiln is heated at 550 °C, some 65% of the raw coconut shell is recovered as the VOCs. For the refinement of the proposed model, more experiments have to be conducted and the operation and control of the kiln need to be investigated in the future.

#### ACKNOWLEDGEMENTS

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#### NOMENCLATURE

A	: gas-solid surface per unit of length [m <sup>2</sup> /m]
A <sub>v</sub>	: absorptivity [-]
C	: parameters listed in Table 2 [-]
C <sub>g</sub>	: average specific heat of gas [kJ/(kg K)]
C <sub>s</sub>	: average specific heat of solid [kJ/(kg K)]
C <sub>w</sub>	: average specific heat of kiln wall [kJ/(kg K)]
e <sub>g</sub>	: gas emissivity [-]
e <sub>s</sub>	: solid emissivity [-]
e <sub>w</sub>	: emissivity of internal surface of wall [-]
h <sub>gs</sub>	: heat transfer coefficient between the solid and gas [W/(m <sup>2</sup> K)]
h <sub>gw</sub>	: heat transfer coefficient between the wall and gas [W/(m <sup>2</sup> K)]
h <sub>i</sub>	: convection coefficient between the gas and solid [W/(m <sup>2</sup> K)]
h <sub>w</sub>	: heat transfer coefficient between the wall and solid [W/(m <sup>2</sup> K)]
H <sub>v</sub>	: latent heat of vaporization of VOC [kJ/kg]
L <sub>cut</sub>	: cord length [m <sup>2</sup> ]
L <sub>es</sub>	: covered wall surface [m <sup>2</sup> ]
L <sub>fi</sub>	: cord length [m <sup>2</sup> ]
M <sub>w</sub>	: wall mass per unit kiln length [kg/m]
Q <sub>g</sub>	: gas flow rate [kg/s]
Q <sub>s</sub>	: solid flow rate [kg/s]
T <sub>g</sub>	: gas temperature [K]
T <sub>s</sub>	: solid temperature [K]
T <sub>w</sub>	: temperature of internal surface of the wall [K]
t	: time [s]
V <sub>g</sub>	: gas velocity [m/s]
V <sub>s</sub>	: solid velocity [m/s]

z : kiln length [m]

#### Greek Letters

φ<sub>sw</sub> : radiation number listed in Table 2 [-]  
 σ : Boltzmann constant 5.57 × 10<sup>-8</sup> [W/(m<sup>2</sup> K<sup>4</sup>)]

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