Optimum Conditions for Effective Decomposition of Toluene as VOC Gas by Pilot-Scale Regenerative Thermal Oxidizer

S. Iijima, K. Nakayama, D. Kuchar, M. Kubota, and H. Matsuda

Abstract—Regenerative Thermal Oxidizer (RTO) is one of the best solutions for removal of Volatile Organic Compounds (VOC) from industrial processes. In the RTO, VOC in a raw gas are usually decomposed at 950-1300 K and the combustion heat of VOC is recovered by regenerative heat exchangers charged with ceramic honeycombs. The optimization of the treatment of VOC leads to the reduction of fuel addition to VOC decomposition, the minimization of CO_2 emission and operating cost as well.

In the present work, the thermal efficiency of the RTO was investigated experimentally in a pilot-scale RTO unit using toluene as a typical representative of VOC. As a result, it was recognized that the radiative heat transfer was dominant in the preheating process of a raw gas when the gas flow rate was relatively low. Further, it was found that a minimum heat exchanger volume to achieve self combustion of toluene without additional heating of the RTO by fuel combustion was dependent on both the flow rate of a raw gas and the concentration of toluene. The thermal efficiency calculated from fuel consumption and the decomposed toluene ratio, was found to have a maximum value of 0.95 at a raw gas mass flow rate of 1810 kg·h^{-1} and honeycombs height of 1.5m.

Keywords—Regenerative Heat Exchange, Self Combustion, Toluene, Volatile Organic Compounds.

I. INTRODUCTION

VOC are often used as solvents in many industrial processes such as painting, printing and laminating. However, as a consequence of their utilization, aromatic and chlorinated hydrocarbons are widely discharged to the environment. For example, an amount of 101,736 tons of toluene was discharged in FY 2006 in Japan and this amount represented the largest amount among the target chemicals [1]. In general, VOC are considered to be responsible for various health problems due to the generation of photochemical oxidants and SPM (Suspended Particulate Matter) from VOC [2].

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As a countermeasure of VOC emission, Japanese air pollutant control law had been revised in May 2004 and more rigorous measures were taken to prevent the release of VOC to the atmosphere [3]. Concentrations of VOC in exhaust gases of driers, paint booth, industrial cleaning as well as VOC storage tank, were targeted by the revised regulation which is in accordance with regulations in other countries such as Council Directive 1999/13/EC and Clean Air Act Amendments of 1990 [4]. For example, total hydrocarbons concentration permitted in exhaust gases of a drier for a gravure printing was set to 700 ppmC. Regarding the treatment of VOC from printing dryer and laminator, etc., various methods on the recovery or thermal decomposition of VOC have been in practical use. For VOC recovery, the methods on PSA (Pressure Swing Adsorption) [5] and membrane separation [6] have been used to recover mainly a single species of VOC from exhaust gases. However, in general, the exhaust gas discharged from industrial process contains different kinds of VOC, which may cause a lot of energy consumption for the separation and recovery of the desired VOC. Meanwhile, among the decomposition methods for VOC treatment, there are several methods in use; combustion, chemical agents, plasma [7] and biochemical decomposition [8]. In particular, the combustion method is widely used for the treatment of VOC such as toluene, ethyl acetate, methyl ethyl ketone, etc. In the combustion method, a RTO is known as an effective device for the oxidative decomposition of the majority of VOC, in terms of high thermal efficiency, robustness and economical merit. Further, the RTO covers a wide range of VOC concentration

Generally it is considered that the efficiency of VOC treatment in the RTO is affected by the concentration of VOC, the residence time and the temperature of a raw gas to be treated, etc. Among these operation factors, flow rates and VOC concentration of a raw gas are of primary significance. However, so far, there have been only few reports which gave the guideline for the optimum operation of an RTO to accomplish a higher thermal efficiency as well as high decomposition efficiency for VOC treatment. Therefore, in the present work, the thermal efficiency and the thermal performance of an RTO during the oxidative decomposition of VOC was studied in a pilot-scale RTO unit using toluene as VOC, and the optimum conditions for a higher thermal efficiency as well as a cost-effective design of the RTO were

discussed.

II. PROCEDURE FOR EXPERIMENTS

A. Operation Principle of Regenerative Thermal Oxidizer

An illustrative diagram of a typical RTO is shown in Fig. 1. The RTO consists of a set of two heat exchange columns where ceramic honeycombs are charged as heat storage/regeneration media. These two heat exchangers are connected to a combustion chamber on the top of the unit.
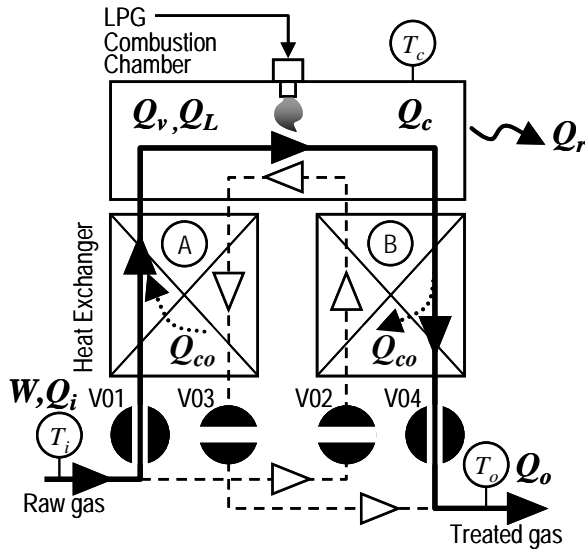

Fig. 1 Schematic diagram of RTO operation

The raw gas containing VOC is made flow into heat exchanger "A" which has stored the remainder of combustion heat for VOC decomposition, by operating V01 and V04 valves open, while V02 and V03 valves are closed. In the first step of operation, the raw gas receives the heat stored in heat exchanger "A" during preheating period and enters the combustion chamber. When, the preheated raw gas enters the combustion chamber, VOC undergo oxidative decomposition and the treated gas of a higher temperature leaves the combustion chamber. Thereafter, the treated gas free from VOC flows into heat exchanger "B" where the honeycombs are heated up storing the exhaust heat from the combustor, and leaves the RTO.

In the second step, a direction of the raw gas flow in the RTO is reversed by opening V02 and V03 valves, with V01 and V04 valves closed. When the raw gas enters heat exchanger "B" where the combustion heat of VOC has been stored in the honeycombs in the former step, the raw gas recovers the heat from the honeycombs and is preheated before entering the combustion chamber. After VOC in the raw gas had been decomposed in the combustion chamber, the treated gas enters heat exchanger "A". Thus, one cycle of cyclic operation of VOC treatment in the RTO is completed.

B. Experiments

As shown in Fig. 1, the raw gas was sucked into the RTO by an exhaust fan operated by a frequency controller. The mass flow rate W (kg·h^{-1}) was measured by a mass flow meter. A LPG burner was used to keep the gas temperature in the combustion chamber at a given value (973-1173 K). The LPG consumption was measured by a gas flow meter, and the combustion heat calorific value of LPG Q_L (kJ·h^{-1}) was calculated from the consumption and heating value of LPG.

As a typical representative of VOC, toluene was used in this study and was supplied to the RTO by a spray nozzle. The mass flow rate of the supplied toluene was monitored, and the concentration of toluene in the raw gas and the treated gas was measured by an FID gas chromatograph. The combustion heat calorific value of toluene Q_v (kJ·h^{-1}) was calculated from the supplied toluene flow rate and the heating value of toluene ΔH (kJ·mol^{-1}).

Table I shows the specification of the ceramic honeycombs used in the present study. A cross section of each heat exchanger was 0.36 m^2, and ceramic honeycombs were charged up to a given height H (0.9 - 1.5 m) in both heat exchangers.

TABLE I
SPECIFICATION OF HONEYCOMB

Geometric Surface	830	m^2·m^{-3}
Open Space	0.625	
Weight	5.7	kg
Raw Density	2.3x10^{-3}	kg·m^{-3}
Specific Heat Capacity	900	J·kg^{-1}·K^{-1}
Thermal Conductivity	1.7	W·m^{-1}·K^{-1}
Material	Cordierite	

The combustion chamber and the heat exchangers were insulated around the inside of the outer walls, so that the heat loss from the combustion chamber and the heat exchangers by heat conduction, heat convection and heat radiation, Q_r (kJ·h^{-1}) from the RTO could be lowered.

The gas temperatures at measuring points indicated in Fig. 1 were measured by K-type thermocouple. In Fig. 1, T_i (K) is the temperature of the raw gas in the inlet duct of the RTO and T_c (K) is the gas temperature in the combustion chamber, and T_o (K) is the temperature of the treated gas in exhaust dust. The calorific values of Q_i, Q_c and Q_o (kJ·h^{-1}) are those of the gas at each temperature of T_i, T_c and T_o. The calorific value, Q_{co} (kJ·h^{-1}) shown in Fig.1 is that of charged or discharged heat in the heat exchanger. The calorific value of Q_{co} charged in the heat exchanger is equal to the calorific value regenerated for preheating the raw gas, in the steady-state condition.

In the experiment, the ambient air was introduced into the RTO for about 2 hours to start up the RTO and the main valves (V01 - V04) were switched continuously at a given interval, t_V. During this operation, the gas temperature in the combustion chamber was slowly heated up to a given temperature by a LPG burner. When a stable state condition of the RTO was achieved, the raw gas containing toluene was introduced into the RTO and the treatment of the raw gas was started.

The experimental conditions were as follows; the gas

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temperature in the combustion chamber was set to a value from the a temperature range of 973 to 1273 K and the mass flow rate of the raw gas was in the range of 1090 to 2550 kg·h^{-1}. The time interval of switching the main valves to change the direction of gas flow was set to a value from a range of 60 to 180 s.

C. Temperature Efficiency η_T

The temperature efficiency η_T is practically used to evaluate the heat exchange performance of various heat exchangers, since it can be calculated only from the temperatures of a high temperature fluid and a low temperature fluid at the inlet and the outlet. The η_T of typical heat exchangers is calculated by (1) [10]-[11].

$$\eta_T = \frac{T_{hi} - T_{ho}}{T_{hi} - T_{ci}} \tag{1}$$

where, T_{hi} is the temperature of a high temperature fluid in the inlet of heat exchanger, T_{ho} is the temperature of a high temperature fluid in the outlet of heat exchanger and T_{ci} is the temperature of a low temperature fluid in the inlet of heat exchanger.

As shown in Fig.1, the sensible heat of the treated gas leaving the combustion chamber at T_c was stored in the honeycombs while the treated gas was flowing downward heat exchanger "B", and the treated gas was discharged as exhaust gas at T_o. After heat exchanger "B" was heated up and kept at a prescribed temperature, the raw gas at the temperature of T_i was allowed to flow into heat exchanger "B", so that the raw gas could be preheated in heat exchanger "B". When the temperature efficiency is adapted to the RTO system, (1) is replaced by (2).

$$\eta_T = \frac{T_c - T_o}{T_c - T_i} \tag{2}$$

In the operation of RTO, the temperature of the exhaust gas, T_o changed with time, which meant that the η_T value calculated by (2) was dependent on time. For example, variation of T_i, T_o and T_c at $t_V = 120$ s is shown in Fig.2. In the present work, the overall η_T value of RTO was calculated using average gas temperatures of T_c, T_i and T_o, as expressed in (3). As shown in Fig.2, T_c and T_i can be assumed to be constant, and then (3) is simplified as (4).

$$\eta_T = \frac{\dfrac{1}{t_V}\int_0^{t_V} T_c(t)dt - \dfrac{1}{t_V}\int_0^{t_V} T_o(t)dt}{\dfrac{1}{t_V}\int_0^{t_V} T_c(t)dt - \dfrac{1}{t_V}\int_0^{t_V} T_i(t)dt} \tag{3}$$

$$\eta_T = \frac{T_c - \dfrac{1}{t_V}\int_0^{t_V} T_o(t)dt}{T_c - T_i} \tag{4}$$

Heat exchange characteristic of the honeycombs was

evaluated using the η_T calculated by (4).
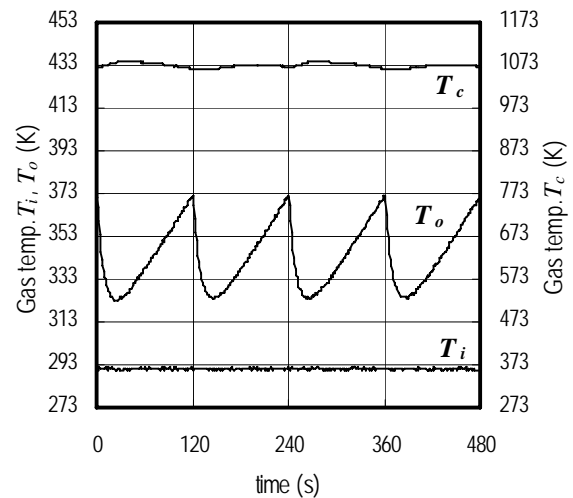
$(W = 2180$ kg·h^{-1}, $H = 1.2$m, $t_V = 120$ s$)$
Fig. 2 Gas temperatures variation

D. Thermal Efficiency η_H

As expressed in (4), a decrease in the temperature of the exhaust gas, T_o led to an increase of temperature efficiency η_T. However, at a higher temperature level, the heat loss such as the radiation heat loss, Q_r should not be neglected, since as a radiation heat loss from heat exchanger surface increases, η_T increases due to a decrease in T_o. To estimate the radiation heat loss, Q_r, the enthalpy balance in the RTO during the steady-state condition was considered. In accordance with the energy conservation law, the enthalpy values of the raw gas Q_i and the exhaust gas from the RTO Q_o, and the calorific values of VOC, Q_v and LPG, Q_L are related as expressed in (5).

$$Q_v + Q_L = Q_o - Q_i + Q_r \tag{5}$$

In the case that the concentration of VOC is low, the combustion heat of LPG is required to provide heat to raise the temperature of the raw gas from T_i to T_c. On the other hand, when VOC concentration is high, the heat supply to the RTO by LPG combustion is not necessary. Furthermore, when the concentration of VOC is high enough providing an excess amount of combustion heat to the RTO, the surplus heat should be removed from the RTO, so that T_c may be kept within a safe temperature level for a commercial operation of RTO. In the present work, such an operation condition will not be discussed, since the surplus heat can not be removed from the combustion chamber of the present RTO unit.

The thermal efficiency η_H of the RTO can be expressed by (6), in accordance with its definition.

$$\eta_H = \frac{Q_{co}}{Q_c - Q_i} \tag{6}$$

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As shown in Fig. 1, the raw gas at the temperature of T_i is heated by the regenerative heat stored in the honeycombs and attains T_c by the combustion heats of toluene and LPG. These enthalpy values are related to (7).

$$Q_c = Q_i + Q_{co} + Q_v + Q_L \qquad (7)$$

When (8) is replaced by (7) and substituted for (6), η_H is calculated by (9) using Q_v and Q_L.

$$Q_{co} = Q_c - Q_i - (Q_v + Q_L) \qquad (8)$$

$$\eta_H = 1 - \frac{Q_v + Q_L}{Q_c - Q_i} = 1 - \frac{Q_o - Q_i + Q_r}{Q_c - Q_i} \qquad (9)$$

III. RESULTS AND DISCUSSION

A. Effect of Gas Temperature on VOC Decomposition Efficiency

Fig. 3 shows the experimental results on the decomposition efficiency of toluene and the yield of by-products obtained in the temperature range of T_c =973 to 1173 K, at the mass flow rate W =2180 kg·h^{-1} and the time interval t_V =120 s. The concentration of toluene in the raw gas was kept in the range of 100 to 400 ppm.

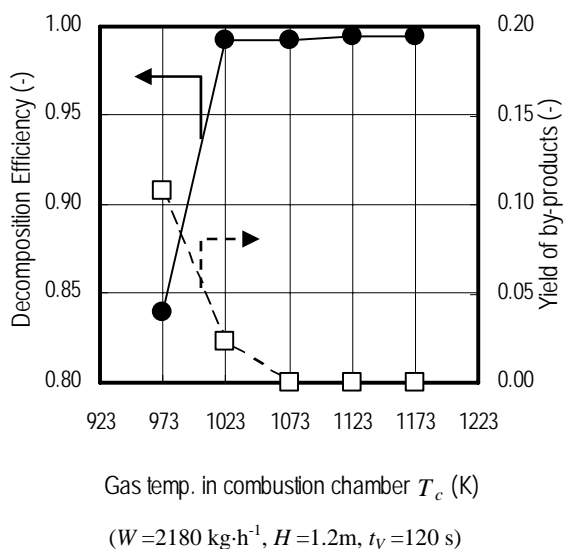$(W$ =2180 kg·h^{-1}, H =1.2m, t_V =120 s)
Fig. 3 Effect of gas temperature on toluene decomposition efficiency

It is seen in Fig. 3 that the decomposition efficiency of toluene was approximately 1.0 when T_c is raised above 1023 K. Further, benzene was detected as the main by-product produced in toluene decomposition, when the temperature was below 1073 K. However, when the temperature raised up to 1073 K, no residual by-products were detected and toluene was completely decomposed at 1073 K.

B. Effect of Height of Honeycombs and Mass Flow Rate on η_T and η_H

As shown in Fig. 3, toluene was completely decomposed above 1073 K. Then, at a constant gas temperature in the combustion chamber of T_c =1073 K, the effect of height of ceramic honeycombs H and the mass flow rate W on the temperature efficiency η_T and the thermal efficiency η_H was studied and the results are shown in Fig. 4.
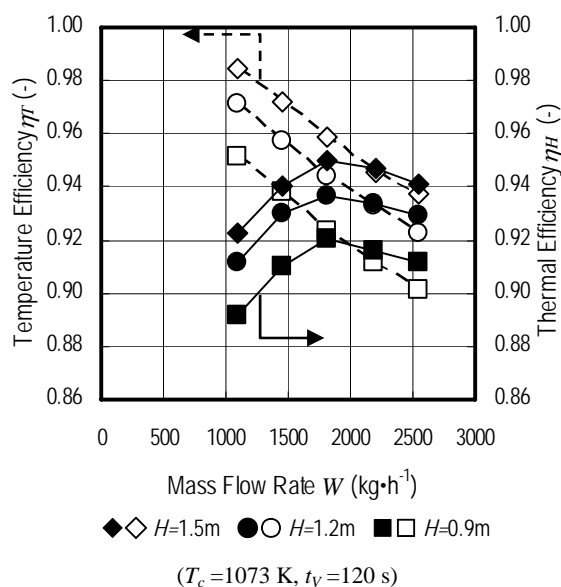
$\blacklozenge\lozenge$ H=1.5m $\bullet\circ$ H=1.2m $\blacksquare\square$ H=0.9m

$(T_c$ =1073 K, t_V =120 s)
Fig. 4 Effect of the mass flow rate W on η_T and η_H

As shown in Fig. 4, η_T increased from 0.92 to 0.96 and η_H increased from 0.92 to 0.95 when H was increased from 0.9 to 1.5 m at W =1810 kg·h^{-1}. Moreover, η_T increased from 0.90 to 0.95 when W was decreased from 2550 to 1090 kg·h^{-1} at H =0.9 m. On the other hand, η_H increased from 0.91 to 0.92 when W was decreased to 1810 kg·h^{-1} at H =0.9 m. But, when W was further decreased, the value of η_H decreased.

The heat transfer performance of honeycombs was improved, so that an increase in H or a decrease in W led to increase in the residence time of the gas in the honeycombs. Thus, an increase in η_H with a decrease in W at a constant height of honeycombs was also predicted. But η_H had a maximum value at W =1810 kg·h^{-1}.

This phenomenon can be explained by the effect of the heat radiation loss, Q_r. The total enthalpy for heating the raw gas from T_i to T_c decreases proportionally to a decrease in W. Further, a ratio of enthalpies of both toluene and LPG to the total enthalpy for heating the raw gas decreased. On the other hand, the enthalpy of both toluene and LPG in order to make up the radiation heat loss, Q_r was a constant. This is because Q_r was almost independent on W at a constant temperature of T_c. Therefore, when the mass flow rate W was low, the combustion heats of toluene and LPG were mostly lost in the form of Q_r, and η_H decreases.

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C. Effect of Residence Time on η_T and η_H

As seen in Fig. 4, the temperature efficiency η_T and the thermal efficiency η_H were affected by the mass flow rate of treated gas W and the height of ceramic honeycombs H. From this result, it was considered that η_T and η_H were dependent on the space velocity or the residence time of the gas in the honeycombs. Hence, the residence time of the treated gas, t_R was defined by (10).

$$t_R = \frac{3600 \times V}{F} \tag{10}$$

where, V is the channel volume of honeycombs in the heat exchanger, F is the volumetric flow rate of the raw gas at the average gas temperature T_{ave} in the heat exchanger. The values of V and F are expressed by (11) and (12), respectively.

$$V = A \times H \times O_s \tag{11}$$

$$F = \frac{W}{\gamma} \times \frac{T_{ave}}{273} \tag{12}$$

where, A is a cross section of the honeycombs 0.36 m², O_s is an open space of the honeycombs 0.625, γ is the specific gravity of the raw gas 1.293 kg·m⁻³ at 273 K.

Fig. 5 shows the effect of the residence time of the treated gas, t_R on the efficiencies of η_T and η_H.
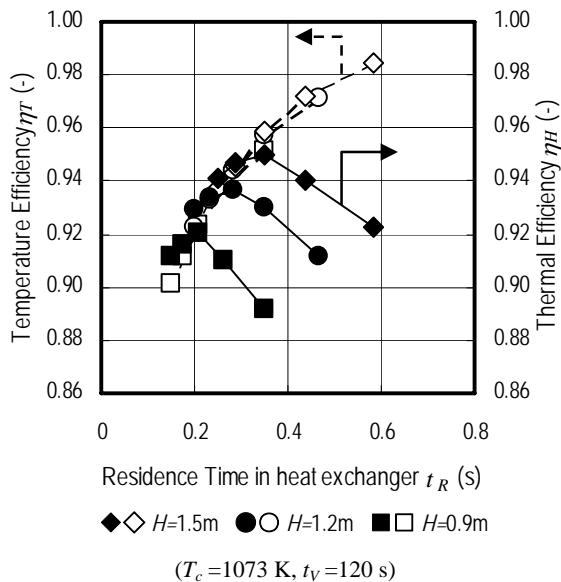

$(T_c = 1073$ K, $t_V = 120$ s$)$
Fig. 5 Effect of the residence time t_R on η_T and η_H

As seen in this figure, η_T increased almost linearly from 0.90 to 0.98 when t_R increased from 0.15 to 0.58 s at each honeycombs height of $H = 0.9$, 1.2 and 1.5 m. It was found that η_T is almost independent on H at a constant t_R. On the other hand, η_H took the maximum value a constant height of H when t_R was increased. The value of t_R which gave the maximum value of η_H was increased from 0.21 to 0.35 s when H was

increased from 0.9 to 1.5 m. From these results, it can be said that the amount of heat recovered from honeycombs was increased with an increase in the residence time of the raw gas. However, as already explained on the reason of the decrease in η_H value in Fig.4, the radiation heat loss from the RTO became relatively large when the mass flow rate was increased.

D. Effect of Time Interval on Thermal Efficiency

As was shown in Fig. 2, the treated gas temperature at the outlet of the duct T_o changed from 323 to 372 K with time at each interval of $t_V = 120$ s. Based on this, it is considered that T_o and thermal efficiency η_H should vary with the change in t_V. Fig. 6 shows the effect of t_V on η_H and T_o.

$\diamond$ T_o max. $\square$ T_o ave. $\triangle$ T_o min.

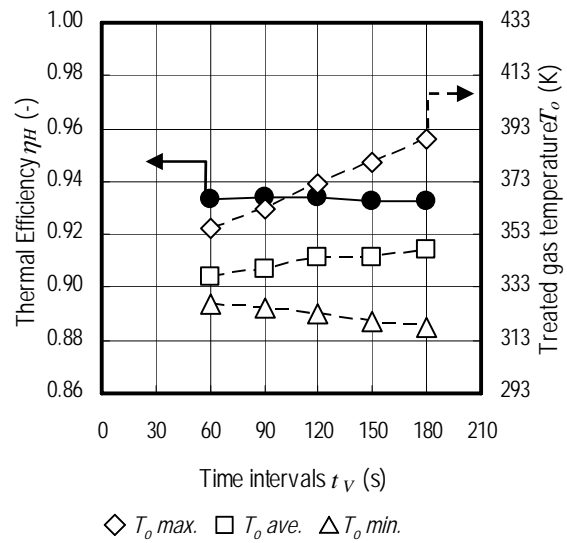$(W = 1810$ kg·h⁻¹, $T_c = 1073$ K, $H = 1.2$ m$)$
Fig.6 Effect of time interval t_V on thermal efficiency

As seen in this figure, when t_V was changed from 60 to 180 s, the minimum value of the temperature of T_o decreased from 327 to 318 K, and the maximum value of the temperature of T_o increased from 355 to 389 K. In addition, the thermal efficiency η_H decreased only slightly, although the time-averaged value of T_o increased from 337 to 347 K. When t_V was increased, the temperature of the upper part of honeycombs approaches the gas temperature in combustion chamber T_c and then the heat transfer from the treated gas to honeycombs is reduced.

E. Optimization of Honeycombs for Self Combustion

As was shown in the previous section, the maximum value of the thermal efficiency η_H was obtained at $W = 1810$ kg·h⁻¹ and η_H increased with an increase in H. In an optimum RTO operation, the shortage of calorific value for a stable combustion of toluene in the raw gas was provided by heat exchanger when LPG combustion was not used ($Q_L = 0$). Hence, the minimum concentration C of toluene in the raw gas to achieve self combustion can be calculated from (13), in terms of calorific value of combustion Q_V, heat value of toluene ΔH and mass flow rate of the gas W at $H = 0.9$, 1.2 and 1.5 m.

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$$C = \frac{Q_V}{\Delta H} \times \frac{22.4 \times \gamma}{W} \times 10^3 \qquad (13)$$

$$Q_V = (1 - \eta_H) \times (Q_c - Q_i) \qquad (14)$$

where, Q_V is calculated from (14), which was replaced by (9) on the assumption that Q_L was equal to 0.

Fig. 7 shows the relation between the calculated value of C and the height of honeycombs at $W = 1810$ kg·h^{-1}.
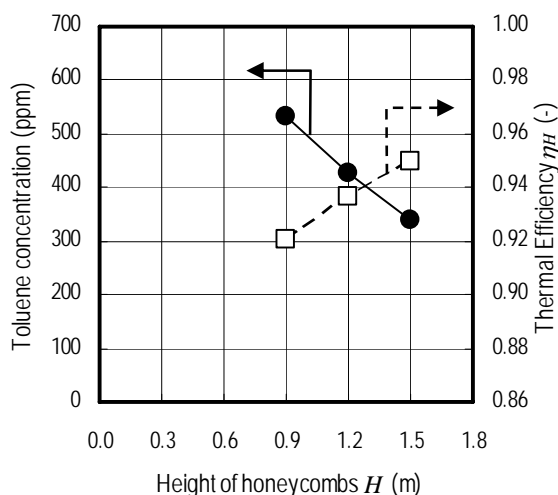
$(W = 1810$ kg·h^{-1}, $T_c = 1073$ K, $t_V = 120$ s)
Fig. 7 Toluene concentration in the raw gas necessary for self combustion

It was found that toluene concentration of 530 ppm in the raw gas was necessary for self combustion at $H = 0.9$ m and this concentration decreased to 340 ppm due to the increase in η_H from 0.92 to 0.95 when H was increased up to 1.5 m. However, if an excess amount of honeycombs is charged in the heat exchangers, the pressure drop in heat exchangers increases, which leads to a deterioration in cost effectiveness of RTO. Thus, based on these findings, it is possible to determine an optimum height of honeycombs for self combustion of toluene in the raw gas.

IV. CONCLUSION

In the present work, the thermal efficiency of the RTO was investigated using a pilot-scale RTO unit for effective decomposition of toluene. As a result, it was recognized that the heat transfer of honeycombs depended only on the residence time of the gas in the honeycombs. In addition, the optimum value of the raw gas mass flow rate was determined to be 1810 kg·h^{-1} in this RTO unit. As for the gas mass flow rate lower than 1810 kg·h^{-1}, it was considered that the majority of combustion heat of toluene and LPG was used to compensate the radiation heat loss. Finally, it was found that when the height of honeycombs decreased from 1.5 to 0.9 m, a minimum concentration of toluene to achieve self combustion without additional heating increased from 340 to 530 ppm at an optimum mass flow rate of 1810 kg·h^{-1}.

NOMENCLATURE

Symbol	Quantity	unit
C	Toluene concentration in the raw gas	ppm
C_c	Specific heat capacity at T_c	kJ·kg^{-1}·K^{-1}
C_i	Specific heat capacity at T_i	kJ·kg^{-1}·K^{-1}
C_o	Specific heat capacity at T_o	kJ·kg^{-1}·K^{-1}
F	Volumetric flow rate of the gas	m3·h^{-1}
H	Height of ceramic honeycombs	m
ΔH	Heat value of toluene	kJ·mol^{-1}
Q_r	Radiation heat loss from surface of the combustion chamber and the heat exchangers	kJ·h^{-1}
Q_c	Calorific value of the gas at T_c in combustion chamber	kJ·h^{-1}
Q_{co}	Calorific value of transferred heat from treated gas to honeycombs	kJ·h^{-1}
Q_i	Calorific value of raw gas at T_i	kJ·h^{-1}
Q_L	Combustion heat calorific value of LPG	kJ·h^{-1}
Q_o	Calorific value of treated gas at T_o	kJ·h^{-1}
Q_v	Combustion heat calorific value of VOC	kJ·h^{-1}
T_c	Gas temperature in combustion chamber	K
T_i	Raw gas temperature in inlet duct of RTO	K
T_o	Treated gas temperature in exhaust duct of RTO	K
t_R	Residence time of raw gas in heat exchanger	s
t_V	Time interval for reversing gas flow	s
V	Total channel volume of honeycombs	m^3
W	Mass flow rate of the gas	kg·h^{-1}
η_H	Thermal efficiency	
η_T	Temperature efficiency	

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