

Optimization of the Operating Parameters of a Reverse Flow Reactor for Methane Thermal Oxidation

LIU RUIXIANG, LIU YONGQI, GAO ZHENQIANG

School of Transportation and Vehicle Engineering

Shandong University of Technology

12 Zhangzhou Road, Zibo, Shandong 255049

CHINA

lrxdlut@sina.com <http://jtxy.sdut.edu.cn>

Abstract: - The potential of the thermal reverse flow reactor for reducing the methane emission has been demonstrated, but the further optimization of the structure parameters and operation parameters of the reactor is necessary because the intrinsic feedback of heat gives rise to a complex dynamic behavior. The effects of switch time and velocity on the stability of the reactor were studied experimentally using the presented pilot scale reactor. Emphasis was the effect of switch time from a perspective of minimizing the thermal energy loss to the outlet stream. Based on detailed analyses, it was found that for the lower velocity, the longer switch time should be used, on the other hand, for the higher velocity, the shorter switch time should be used. Further, for a given reacting section length, the production of velocity and switch time is a constant. The switch time determined by the method can ensure to accumulate more energy within the reactor and sustain itself under the lower methane concentration conditions.

Key-Words: - Reverse flow reactor; Methane; Thermal oxidation; Switch time; Optimization

1 Introduction

Methane is believed to be a main contributor to climate warming today. Finding efficient methods of reducing methane emissions is stringent. Because most of these methane emissions is typically lean methane [1], the lean methane disposal is extra important. The feasibility of the lean methane thermal oxidation in a reverse flow reactor has been demonstrated by the results of our previous study. The lean methane thermal oxidation can be run and sustained in the reverse flow reactor, and the methane conversion efficiency is over 95%. Furthermore, the thermal oxidation has the advantage of considerably lower cost and is preferred where catalyst poisoning could be a problem. Therefore, the lean methane thermal oxidation is a promising technology for the efficient mitigation of methane.

However, because the intrinsic accumulation and transfer of heat lead to a complex dynamic behavior, the further investigation is necessary.

So far, many investigations on a catalytic reverse flow reactor have been carried out [2,3,4,5], the experimental results show that the behavior of the catalytic reverse flow reactor changes with many parameters, which include the structure parameters, such as properties of the ceramic monolith, properties of the insulation

layer and length of the reactor, and the operating parameters, such as methane concentration, feed velocity and switch time. Similarly, the behavior of the thermal reverse flow reactor should change with these parameters, too. In order for the thermal reverse flow reactor to be applied to practice, the effect of these parameters must be studied, too. To reduce the complicity, the focus of this investigation will be on the effect of velocity and switch time on the thermal reverse flow reactor. In addition, the reactor used by this investigation only reduces the greenhouse impact of the methane by oxidizing it without extraction of thermal energy [6,7]. Thus, the objective of this investigation is to develop an efficient method, which can be used to determine switch time, which can match with the different velocities.

2 Experimental reactor

Tests of the methane thermal oxidation were carried out in a reverse flow reactor. The entire configuration of the pilot scale reactor and test apparatus is shown in Fig.1.

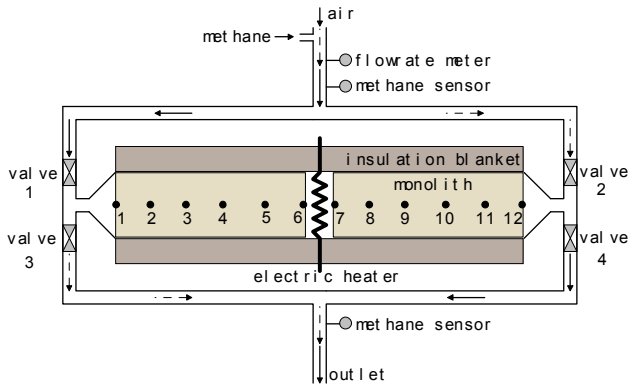


Fig.1 Configuration of the reactor and test apparatus

In a reverse flow reactor the feed is periodically switched between the two reactor ends using control valves. When control valves 1 and 4 are open, the feed flows to the reactor from left to right (forward flow). When control valves 2 and 3 are open, the feed flows to the reactor from right to left (reverse flow). The total cycle consists of these two operations, and the term switch time denotes the time at which the flow is changed from forward to reverse flow or from reverse to forward flow. The sum of the times for forward and reverse flow is the cycle duration.

Thermal energy generated in an exothermic methane oxidation reaction can be captured with the solid heat storage medium. Then, with switching the flow direction, the captured thermal energy within the heat storage medium can be used to preheat the feed. This allows the reactor core to remain at high reaction temperatures, even if the inlet feed is at lower temperatures. With reverse flow, the lean methane oxidation reactions that are not normally auto-thermal may be run and sustained at lower inlet temperatures and higher conversions.

It is clear that the thermal mass of the solid heat storage medium is very important to the reverse flow reactor, the higher thermal mass will give the higher stability [8,9]. In the experiments presented in this paper, the ceramic honeycomb monoliths were used as heat storage medium. The monolith properties were given in previous paper.

To prevent heat losing from the reactor to the surroundings, the reactor was surrounding with a layer of insulation ceramic fibre blanket. The ceramic fibre blanket properties were given in previous paper.

Air was supplied to the reactor using an air compressor. The inlet air was at ambient temperature. The methane gas was supplied from the gas cylinder. The flow rate of inlet methane air mixture was measured using a flow rate meter. The methane concentrations in the feed and outlet

stream were measured using two methane concentration sensors. In the experiments, the methane concentration was 0.4% over time. Thermal profiles from the reactor were obtained using twelve thermocouples (denoted from 1 to 12 in Fig.1). The data acquisition system recorded all sensor values at a specified interval, generally 1 second.

The four switching valves controlled the flow direction. The valves were controlled by the computer control system on the given switch time.

3 Testing procedure and results

3.1 Effect of switch time

The switch time is a key factor that can affect the stability of the reactor [9,10]. To explore the best switch time, experiments were carried out while changing the switch time alone at a constant velocity. The inlet velocity at inlet temperature (25°C) was 0.4 m/s. The experiments run for 45 cycles with an inlet methane concentration of 0.4 %. The axial temperature profiles recorded at the end of flow from left to right are shown in Fig.2 and Fig.3.

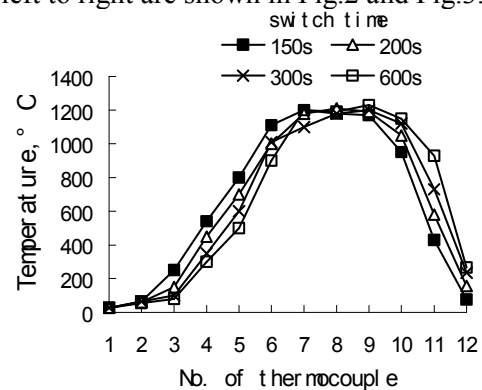


Fig.2 Axial temperature profiles at the end of the forward flow at longer switch time

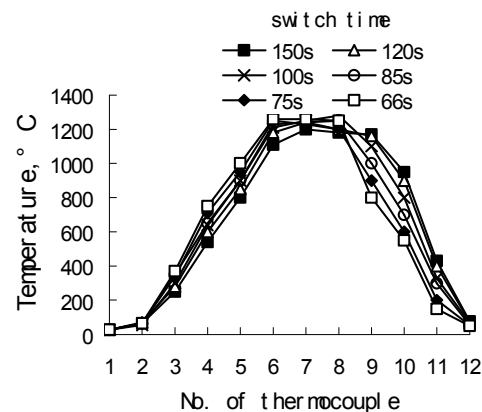


Fig.3 Axial temperature profiles at the end of the forward flow at shorter switch time

The results show that longer switch time will lead to more movement of the temperature profile, higher outlet temperature, and thus more energy loss from the reactor. Ultimately, too longer switch time will lead to extinction of the methane oxidation. On the other hand, shorter switch time will lead to more energy accumulation and higher temperature in the central section of the reactor, and thus less energy loss from the reactor. It is clear that shorter switch time can improve the reactor stability. However, too shorter switch time may result in overheating of the reactor.

3.2 Effect of velocity

Velocity affects the residence time in the reactor, and hence affects the stability. To investigate the effect of velocity, experiments were carried out while changing the velocity alone at a fixed switch time of 120s, and the same inlet methane concentration as before. The axial temperature profiles recorded after 45 cycles at the end of flow from left to right are shown in Fig.4 and Fig.5.

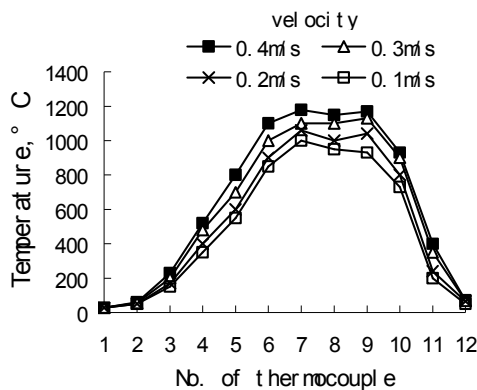


Fig.4 Axial temperature profiles at the end of the forward flow at lower velocity

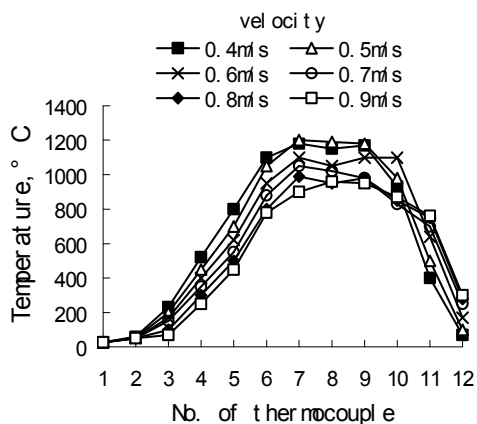


Fig.5 Axial temperature profiles at the end of the forward flow at higher velocity

Fig.4 shows that with decreasing velocity, the energy accumulated in the reactor decreases.

Ultimately, too lower velocity, 0.2 m/s and 0.1 m/s, will lead to extinction of the methane oxidation. This is because the gas flow rate decreases and thus the energy output of the methane oxidation decreases.

Fig.5 shows that with increasing velocity to 0.5 m/s, although the outlet temperature become higher and thus energy loss increases, because the gas flow rate increases and thus the energy output of the methane oxidation increases, the energy accumulated in the reactor increases. However, after 0.5 m/s, further velocity increase will result in shorter residence time and incomplete oxidation.

3.3 Combining effect of switch time and velocity

The switch time should be related to the gas velocity. The switch time and velocity will determine the position of the axial temperature profile together. Fig.6 and Fig.7 show that the effects of changing both the switch time and velocity such that the product of switch time and velocity is a constant.

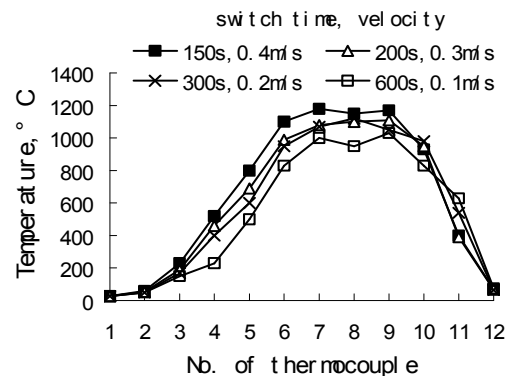


Fig.6 Axial temperature profiles at the end of the forward flow at longer switch time and lower velocity

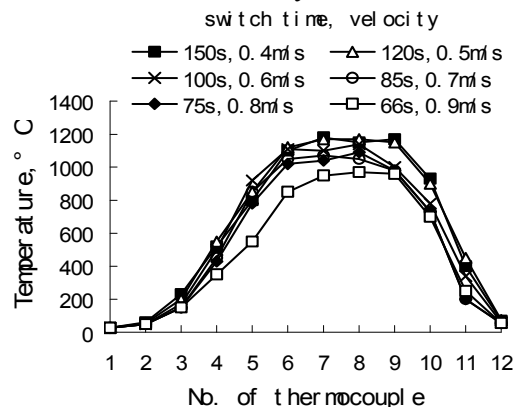


Fig.7 Axial temperature profiles at the end of the forward flow at shorter switch time and higher velocity

Fig.6 shows the axial temperature profiles after 45 cycles for lower velocity and longer switch time combinations. Comparing with Fig.4, the performance of the reactor operated at the velocity under 0.3 m/s is not improved markedly because the gas flow rate and energy output decreasing play a more important role than longer switch time.

Fig.7 shows the axial temperature profiles after 45 cycles for combinations of higher velocity and shorter switch time. Comparing with Fig.5, with increasing velocity, decreasing switch time will lead to the similar outlet temperature and thus similar energy loss. This effect is beneficial to oxidating completely and operating stably. As a result, the highest velocity at which the oxidation is complete and operation is stable is extended to 0.8 m/s from 0.5 m/s. With the highest velocity extended, the methane mixture processing capability of the same reactor is proportionally extended.

Overall, with the product of switch time and velocity being a constant, the velocity extent of stable operation is from 0.3 m/s to 0.8 m/s, which is much wider than that with the fixed switch time of 150s. Depending on the relationship between the switch time and velocity, the switch time matching with the different velocity can be exactly determined.

4 Conclusion

The relationship between the switch time and velocity is found. For a given length of reacting section, the production of the velocity and switch time is a constant. With switch time matching with the different velocity, the velocity extent of stable operation widens significantly, the methane mixture processing capability of the same reactor increases markedly and the performance of the reactor is improved. The results indicate that the thermal reverse flow reactor is an effective technology for reducing methane emissions.

Acknowledgments

This work was financially supported by the Science Foundation of Shandong University of Technology (No. 4040-306005).

References:

[1] Crutzen P J, On the role of CH₄ in atmospheric chemistry: sources, sinks and possible reductions

in anthropogenic sources, *Roy Swedish Academy of Sciences*, Vol.24, No.1, 1995, pp. 52-55.

- [2] Hayes R E, Kolaczowski, Awdry S, The palladium catalyzed oxidation of methane: reaction kinetics and the effect of diffusion barriers, *Chemical Engineering Science*, Vol.56, No.6, 2001, pp. 4815-4835.
- [3] Matros Y S, Bunimovich G A, Reverse flow operation in fixed bed catalytic reactors, *Catalytic Reviews: Science and Engineering*, Vol.38, No.1, 1996, pp. 20-68.
- [4] Williams J L, Monolith structures, materials, properties, and uses, *Catalysis Today*, Vol.69, No.1, 2001, pp. 3-9.
- [5] Pablo M, Miguel A G H, Salvador O, et al., Combustion of methane lean mixtures in reverse flow reactors: Comparison between packed and structured catalyst beds, *Catalysis Today*, Vol.105, No.3, 2005, pp. 701-708.
- [6] Shi Su, Andrew Beath, Hua Guo, et al., An assessment of mine methane mitigation and utilization technologies, *Progress in Energy and Combustion Science*, Vol.31, No. 2, 2005, pp. 123-170.
- [7] Krzysztof G, Krzysztof W, Effect of the mode of heat withdrawal on the asymmetry of temperature profiles in reverse-flow reactors: Catalytic combustion of methane as a test case, *Chemical Engineering Science*, Vol.62, No.10, 2007, pp. 2679-2689.
- [8] Moshe Sheintuch, Analysis of design sensitivity of flow-reversal reactors: Simulations, approximations and oxidation experiments, *Chemical Engineering Science*, Vol.60, No.11, 2005, pp. 2991-2998.
- [9] G. Kolios, J. Frauhammer, G. Eigenberger, Auto-thermal fixed-bed reactor concepts, *Chemical Engineering Science*, Vol.55, No.4, 2000, pp. 5945-5967.
- [10] Ulrich Nieken, Grigorios Kolios and Gerhart Eigenberger, Fixed-bed reactors with periodic flow reversal: experimental results for catalytic combustion, *Catalysis Today*, Vol.20, No.3, 1994, pp. 335-350.