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Experimental study on thermal oxidation of ultra-low concentration methane in a non-catalytic reverse-flow reactor

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A series of experiment were carried out on a 60000 m^3/h non-catalytic reverse-flow reactor for thermal oxidation of ultra-low concentration methane. The start-up performance, methane conversion rate, lowest methane concentration of stable operation, reactor temperature distribution and the yield of super-heated steam were studied. The results show that the temperature distribution on the cross section of the reactor is quite uniform during the whole process including the start-up period, while on the section along the direction of flow, a peak temperature appears at the middle, and temperature falls as it gets farther away from the axis. With the methane concentration at inlet increases from 0.3% to 1.0%, the peak temperature enhances form 990˚C to 1066˚C, while methane conversion rate almost keeps constant (about 98.6%). As the flow of the reactor is 60000 m^3/h, the lowest inlet methane concentration of stable operation is 0.28%.

Keyword: ultra-low concentration methane; thermal oxidation; non-catalytic reverse-flow reactor.

INTRODUCTION

Methane is a kind of conventional energy, and also a kind of greenhouse gas. The huge emission of ventilation air methane (VAM) from coal mine causes serious environmental problem and results in a waste of energy. The ultra-low concentration VAM combustion is difficult to achieve with traditional methods. Nowadays the oxidation technologies of ultra-low concentration VAM are TFRR (thermal reverse-flow reactors) and CFRR (catalytic reverse-flow reactors) [1]. A series of theoretical and experimental study have been done on CFRR [2-8], CFRR achieves relatively low react temperature, however, the reaction activity of the catalyst is greatly affected by temperature, the catalyst is expensive and oxidation process is complex, all of this limit the application of CFRR. TFRR is less expensive and complex; however it needs relatively high reaction temperature. Also, more detailed researches on temperature distribution and methane conversion rate are required for stable and reliable operation of TFRR [9]. While researches on TFRR reported are not enough. Zheng Bin, etc. carried out a series of experimental researches on ultra-low concentration VAM oxidation based on a self-developed pilot regenerative reverse-flow oxidation reactor [10]. Wang Pengfei, etc. established a one dimensional single-temperature model of ultra-low concentration VAM thermal reverse-flow oxidation reactor as well as fractal systems [11, 12, 13]. However, up to now, there is still not any applying plant of TFRR is reported.

A 60000m^3/h non-catalytic reverse-flow reactor for thermal oxidation of ultra-low concentration VAM was proposed, the goal of current paper is to study its performance, including the start-up performance, methane conversion rate, lowest methane concentration of stable operation, and temperature distribution in the reactor.

EXPERIMENT EQUIPMENT AND CONDITIONS

The experiments were carried out on a 60000m3/h non-catalytic reverse-flow reactor for thermal oxidation of ultra-low concentration methane, the plant mainly consists of oxidation reactor, heat extraction system, conveying and mixing system, start-up system, deducting and dehumidifying system, monitoring and control system.

Three vertical columns of K-type thermocouples (which were labeled as left, middle and right) were fixed on the flow direction to obtain temperature distribution of oxidation reactor. There were 15 offset temperature measuring point in each column. The schematic diagram of temperature points arrangement show in figure 1.

The fuel gas used for start-up is liquefied petroleum gas. The concentration of ultra-low concentration methane is between 0.3% and 1.0%, which is obtained by mixing coal mine draining methane and VAM from air return ventilation mine shaft. The water used in the experiments is softened water produced by a LMZF-12 automatic water

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soften system in Julong coal mine methane pumping plant. The soften water is in line with national water supply standards for low pressure boiler. Concentration of methane was measured with a Madur maMos100 methane analyzer, of which the regulation error is 0.025%. Reversing valve actuators (DA-149) were used to achieve the reversing operation. The interval time between two adjacent reversing was 90~150, and the average consuming time of each reversing operation is 2.78s. A plug-in V-cone flow meter was set on the pipeline (DN1200) after the blower to obtain the process capacity. Start-up system consisted of hot air generation, regulation and distribution system, so that the demanding hot air may achieved by adjusting the power of burners and the opening of flow control valve. As core temperature of the reactor reached 800 °C, flow rate was increased, cooperating with which hot air valve was switched off gradually, until the normal operation of the reactor was achieved.

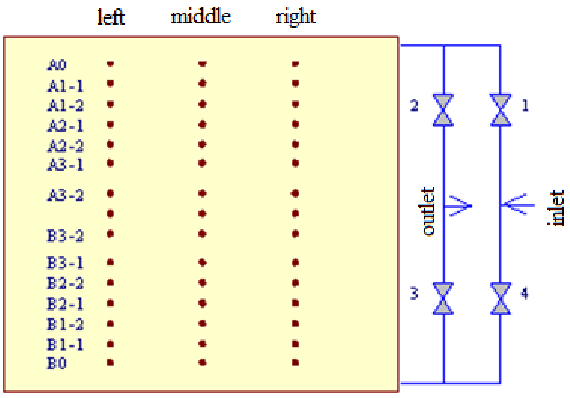


Fig. 1. The schematic diagram of temperature points arrangement.

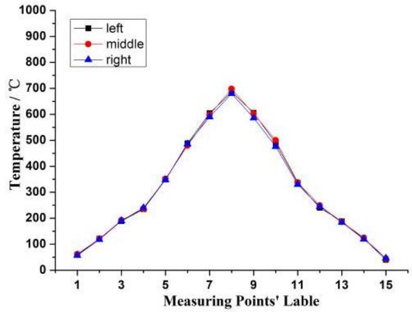
RESULTS AND DISCUSSION

Start up

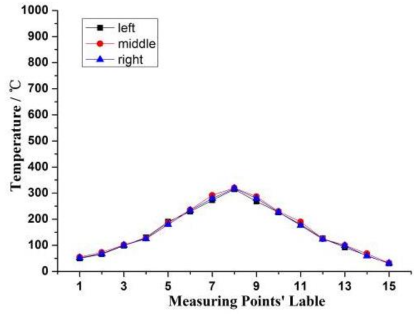
To achieve the normal operation of the plant, core temperature of 900~1000 °C and appropriate reactor temperature distribution must be established at first. Therefore, start-up performance experiments were carried out, and the results are shown in Figure 2.

Figure 2 shows that the temperature at the same level are similar, which indicates that during the start-up period temperature at nearly all the points on the same cross section increases synchronously.

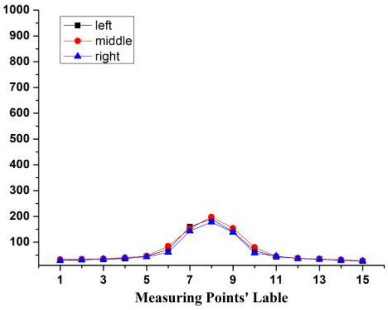
However, on the section along the direction of flow, a peak temperature appears at the middle, and temperature falls as it gets farther away from the axis. It is because gas flow direction reverses periodically, and heat is concentrated at the core of the ceramic regenerator.



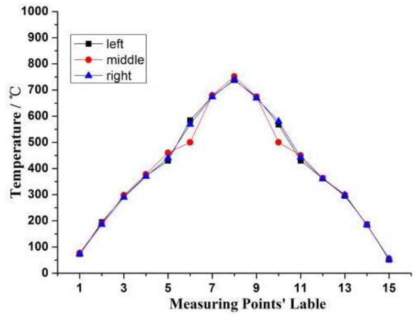
(a) after 3 hours



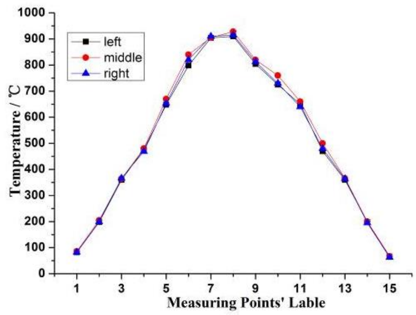
(b) after 12 hours



(c) after 24 hours



(d) after 36 hours



(e) after 48 hours

Fig. 2. The temperature distributions of start-up perior.

As more and more heat is sent to the reactor, core temperature increases, and results in the temperature distribution as mentioned above. This temperature distribution is quite similar to that under normal operation conditions, so that the time cost from start up condition to normal operation condition can be controlled.

Methane conversion rate

The experiments were carried out in order to obtain the peak temperature and methane conversion rate under different inlet methane concentration. During these experiments flow rate was 60625±385m^3/h, concentration of methane was adjusted from 0.3% to 1.0%, results are shown in Figure 3.

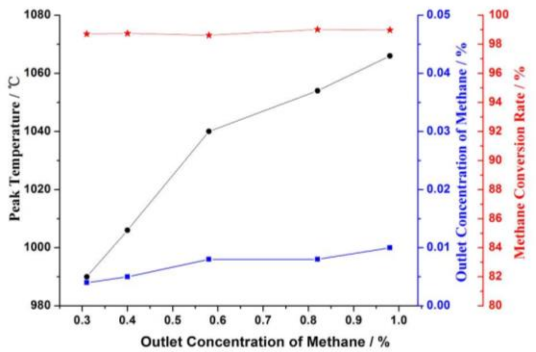


Fig. 3. The effects of methane on peak temperature and methane conversion

Figure 3 shows that as the increase of methane more and more heat is released, so that the peak temperature enhances apparently from 990 °C~1066 °C, meanwhile the outlet methane concentration shows slightly increase (from 0.004% to 0.01%), however methane conversion rates under different inlet methane concentration almost keeps constant, which are all larger than 98.6%, and shows an average value of 98.82%. This indicates that high efficient conversion can be achieved under relative wide range of inlet methane concentration.

The lowest methane concentration of stable operation

Heat released by methane oxidation reaction is partially kept in the reactor to maintain the reaction, and the rest heat may be extracted by the inner heat exchanger. As methane concentration gets too low to maintain the reaction, the temperature may decrease gradually until reaction gets its end. Therefore design of reactor and heat extraction system plays a significant role on the stable operation of the plant. In this experiment, the stable operation condition as following: the flow is about 60000m^3/h, under a certain inlet methane concentration, after 2 hours continuous operation average temperature of the reactor never falls, the peak temperature is larger than 950˚C, and methane conversion rate is no less than 97%. Table 1 shows several record of the lowest methane concentration of stable operation. The results show that as air flow rate keeps about 60000m^3/h, the lowest average methane concentration of stable operation is 0.28%.

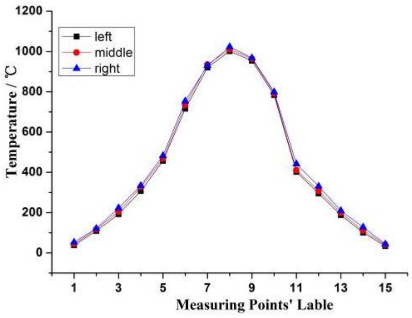
Uniformity of temperature distribution

Experiments on uniformity of temperature distribution in the reactor were carried out under different methane concentration, which was achieved by adjusting the amount of drainage methane for mixing.

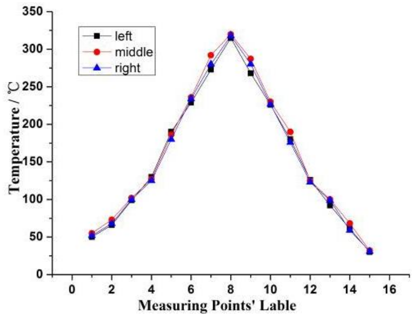
Temperature distribution in Figure 4 under different methane concentration shows that the temperature profiles at different location (left, middle and right) coincide well which reveals the perfect temperature uniformity.

Table1. The lowest methane concentration of the plant stable operation.

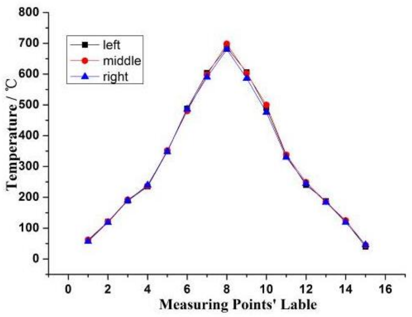
Number	Air flow (m^3/h)	Peak temperature (˚C)	Lowest methane concentration (%)
1	60258	965	0.27
2	61212	980	0.29
3	60480	976	0.28
4	60650	970	0.28
Average value	/	/	0.28



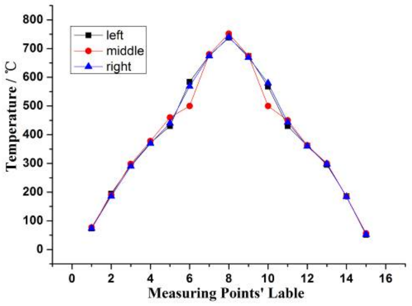
(a) 0.3%



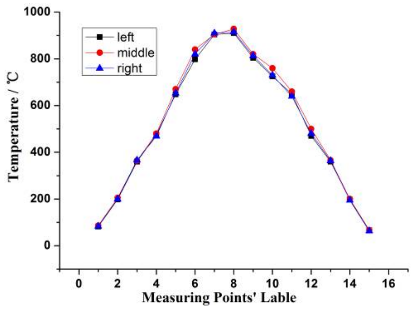
(b) 0.4%



(c) 0.6%



(d) 0.8%



(e) 1.0%

Fig. 4. The effects of methane concentration on the temperature of reactor.

As methane concentration increases, core temperature of the reactor is enhanced, which enlarges the extent of high temperature zone and oxidation zone, and then promotes methane conversion. However, the increase of outlet temperature with concentration can be ignored, which says that the affect of methane concentration variation on heat loss of exhausted gas is little.

CONCLUSION

A series of experiment were carried out on a 60000m^3/h non-catalytic reverse-flow reactor for thermal oxidation of ultra-low concentration methane. With results, following conclusions can be obtained:

(1) Temperature distribution on the cross section of the reactor is quite uniform during start-up period, while on the section along the direction of flow, a peak temperature appears at the middle, and temperature falls as it gets farther away from the axis, which is quite similar to that under normal operation conditions, and helps to achieve normal operation as soon as possible.

(2) The peak temperature enhances apparently from 990~1066 °C, meanwhile the outlet methane concentration shows slightly increase (from 0.004% to 0.01%), however the methane conversion rates under different inlet methane concentration almost keeps constant (98.82%), which indicates that high efficient conversion may achieved under relative wide range of inlet methane concentration.

(3) As the flow is about 60000m^3/h, the lowest methane concentration of stable operation is 0.28%, under which average temperature keeps stable, peak temperature is larger than 950 °C, and methane conversion rate is no less than 97%

(4) Under normal operation conditions, temperature profiles at different location (left, middle and right) coincides well and with the increase of methane concentration high temperature zone and oxidation zone is enlarged, while heat loss of exhausted gas increases little.

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ЕКСПЕРИМЕНТАЛНО ИЗСЛЕДВАНЕ НА ТЕРМИЧНОТО ОКИСЛЕНИЕ НА СВРЪХ-НИСКИ КОНЦЕНТРАЦИИ НА МЕТАН В НЕ-КАТАЛИТИЧЕН РЕАКТОР С ОБРАТЕН ПОТОК

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(Резюме)

Проведена е серия от експерименти в не-каталитичен реактор с обратен поток за термичното окисление на свръх-ниски концентрации на метан при дебит 60000 m^3/h. В пусковия период са изследвани степента на превръщане, най-ниската допустима концентрация на метана за устойчива работа, температурния профил и добива на прегрята пара. Резултатите показват, че температурното разпределение по напречното сечение на реактора е почти равномерно по време на целия процес, включвайки и пусковия период, докато по дължината на реактора се наблюдава максимална температура, която спада по-нататък. При повишаване водната концентрация на метана от 0.3% до 1.0% максималната температура се покачва от 990˚C до 1066˚C при постоянна степен на превръщане (около 98.6%). При избрания дебит на газа най-ниската концентрация на метан за стабилна работа е 0.28%.