

Article

Experimental Study on Ultra-Low Concentration Methane Regenerative Thermal Oxidation

Junhui Yang ¹, Liguang Yang ^{2,*}, Jida Zhang ¹, Xiaoxu Fan ², Sheng Li ², Luyao Zhang ² and Weijie Zhang ²

¹ China Coal (Tianjin) Underground Engineering Intelligent Research Institute Co., Ltd., Tianjin 300000, China; yangjunhui@chinacoal.com (J.Y.); zhangjida@chinacoal.com (J.Z.)

² Energy Research Institute, Qilu University of Technology (Shandong Academy of Sciences), Jinan 250014, China; fanxx@sderi.cn (X.F.); lis@sderi.cn (S.L.); zly4824@163.com (L.Z.); zwj0531@126.com (W.Z.)

* Correspondence: yangliguo@sdkhjzyyxs.wecom.work; Tel.: +86-13256100771

Abstract: As a major coal country, China faces the issue of significant gas emissions during the coal mining process. This study aims to improve the utilization efficiency of mine gas, reduce greenhouse gas emissions, and promote the low-carbon and green transformation of the coal industry. A 10 kW gas regenerative thermal oxidizer (RTO) experimental system was constructed. The effects of initial methane concentration, low-temperature flue gas proportion, and operating load on combustion temperature, methane oxidation rate, high-temperature flue gas energy, and system thermal efficiency were studied. The results show that when the combustion temperature is below 600 °C, the CH₄ combustion reaction cannot proceed effectively, and the system temperature continuously decreases and cannot be maintained stably. The experiment determines the stable operating methane concentration range of the RTO. In this experimental system, the lower limit of the initial methane concentration is 0.28%, corresponding to an 86% methane oxidation rate. As the initial methane concentration decreases, the combustion temperature also decreases, and the methane oxidation rate follows suit. The higher the low-temperature flue gas proportion, the higher the combustion temperature, and the system's thermal efficiency and output heat decrease with the increase in the low-temperature flue gas proportion. This experiment explores multiple factors affecting regenerative thermal oxidation, providing a basis for ensuring the safe and stable operation of the system and its optimization. Improving the thermal insulation and heat exchange performance of the storage body can expand the lower limit of the initial methane concentration, thereby increasing the stability and thermal efficiency of the system.

Keywords: ultra-low-concentration methane; regenerative thermal oxidation; carbon emission reduction



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1. Introduction

Methane gas emitted from coal mines worldwide accounts for 17% of the total anthropogenic greenhouse gas emissions [1], and the amount of methane emitted into the atmosphere as diluted air constitutes approximately 70% of the total methane emitted from coal mines [2]. In China, the methane content in diluted gas accounts for about 90% of the total methane in coal mine gas [3], resulting in an annual greenhouse gas effect of approximately 200 million tons of CO₂ equivalent [4]. The greenhouse effect coefficient of CH₄ is more than 20 times that of CO₂, and the conversion of CH₄ into CO₂ emissions after combustion can achieve a reduction in the greenhouse gas effect by more than 80% of CO₂ equivalent. As the largest source of anthropogenic methane emissions, the coal mining area has tremendous potential for reducing methane emissions from coalbed gas [5], and researching the utilization technology of ultra-low concentration methane is of great significance from both an energy and environmental perspective.

The regenerative thermal oxidizer (RTO), used for processing low-concentration methane, is currently the only technology that can effectively utilize extremely low-concentration coalbed methane. Maintaining a relatively high ambient temperature inside

the reactor promotes the oxidation of low-concentration methane in the reactor, with it reacting to produce carbon dioxide and water and simultaneously releasing heat. This reaction heat is used to maintain the required temperature inside the reactor, and excess heat is recovered and utilized through water or air as a medium. Its development can be traced back to Weinberg's concept of super enthalpy combustion proposed in 1971, which involves recovering and reusing the heat generated by combustion through heat exchange media or other means to preheat fresh reactants. Many scholars at home and abroad have carried out a series of research work on this topic [6,7]. Jones found that by utilizing the characteristics of heat exchange media and using external thermal recirculation between the products and reactants, stable combustion of the mixed fuel can be achieved within the flow range, and full combustion of low calorific value fuel and other low calorific value mixtures can be achieved [8]. Roney made a one-dimensional assumption for a thermal recirculation reactor and conducted comprehensive theoretical research on this basis [9]. Kotani found that porous media super enthalpy combustion was able to broaden the flow rate range of the fuel and air mixtures used. Shinoda further studied the structural characteristics of a thermal cycling burner composed of ceramic materials and the thermal characteristics during the stable combustion of methane/air and lean fuel/air. Hoffman et al. also conducted a comprehensive theoretical and experimental study on the super enthalpy combustion of rarefied fuel gases in porous media [10–12]. The research results all indicate that porous media super enthalpy combustion can broaden the combustion range and has certain applicability for the combustion of low calorific value or dilute gas fuels. A VOCSIDIZER (Counterflow Reactor) is a typical RTO device developed by MEGTEC in the United States. In many industrial fields, the VOCSIDIZER oxidizes low-concentration methane and uses inserted steam pipes to recover energy, and it has been proven to be a mature technology. The practice has shown that if the methane concentration in the exhaust air is as low as 0.1%, the VOCSIDIZER can still operate without the need for additional energy supplementation. In 1994, a coal company in the UK installed a VOCSIDIZER testing device, which used exhaust air with a methane content of 0.3–0.6% and a flow rate of 8000 m³/h. This project confirms that the VOCSIDIZER can be used to treat low-concentration exhaust gas. In 2001, MEGTEC utilized the VOCSIDIZER device to recover 90% of the energy from mine exhaust in Australia for the production of hot water. In China, companies such as Shengli Power Machinery Group and Shandong University of Technology have adopted this technology to treat coal mine exhaust. In 2008, Shengli Power Machinery Group selected the Wangying Mine of the Fuxin Mining Group for industrial testing of a coal mine exhaust gas heat countercurrent oxidation device. The concentration of exhaust gas in the mine was 0.25–0.6%, and the oxidation device operated continuously and stably with a processing capacity of 12,500 m³ for exhaust gas³/about h. Shandong University of Technology independently developed a vertical exhaust air thermal oxidation device with a capacity of 10,000 m³ to handle exhaust air³/h. The methane conversion rate reaches 98%, and its minimum oxidation exhaust concentration is 0.2%. Since the “Twelfth Five-Year Plan”, China has invested over USD tens of hundreds of millions in scientific and technological innovation in the field of oxidative utilization technology for mine gas with low methane concentrations [13–17]. With the implementation of China's “Thirteenth Five-Year Plan for Controlling Greenhouse Gas Emissions” and the “National Climate Change Response Plan (2014–2020)”, mining districts such as Lu'an and Yangquan in Shanxi and Huainan in Anhui have successively constructed and operated more than 20 methane regenerative thermal oxidation utilization projects. These projects annually process a total of over 300 million cubic meters of coal mine diluted gas and perform low-concentration gas extraction, with the heat produced from oxidation being used for steam turbine power generation, heating, combined cooling and power supply, and coal slime drying, effectively reducing the direct emission of low-grade coalbed gas while alleviating the tight situation of clean energy supply in coal mines. Foreign companies with oxidative utilization projects in China include American MEGTEC, German EISENMENN, and DURR companies, among others. Current research on the oxidation technology of diluted gas in China is still in

the preliminary stage [18–24]. The regenerative thermal oxidation technology used in the coal industry has issues such as the poor temperature resistance of the heat storage body, low thermal efficiency, the high failure rate of high-temperature control valves, increased exhaust gas temperature, and high self-consumption of energy. Moreover, challenges such as low methane concentration, unstable gas sources, low concentration, and poor economy in coal mining areas cannot be fundamentally solved in the short term. It is necessary to conduct in-depth research on the related technologies of methane utilization in coal mines and further break through the bottleneck of existing gas regenerative thermal oxidation utilization to provide a basis for industrialization.

2. Materials and Methods

2.1. Experimental Device

The process flow of the gas regenerative thermal oxidation experimental system used in this experiment is shown in Figure 1.

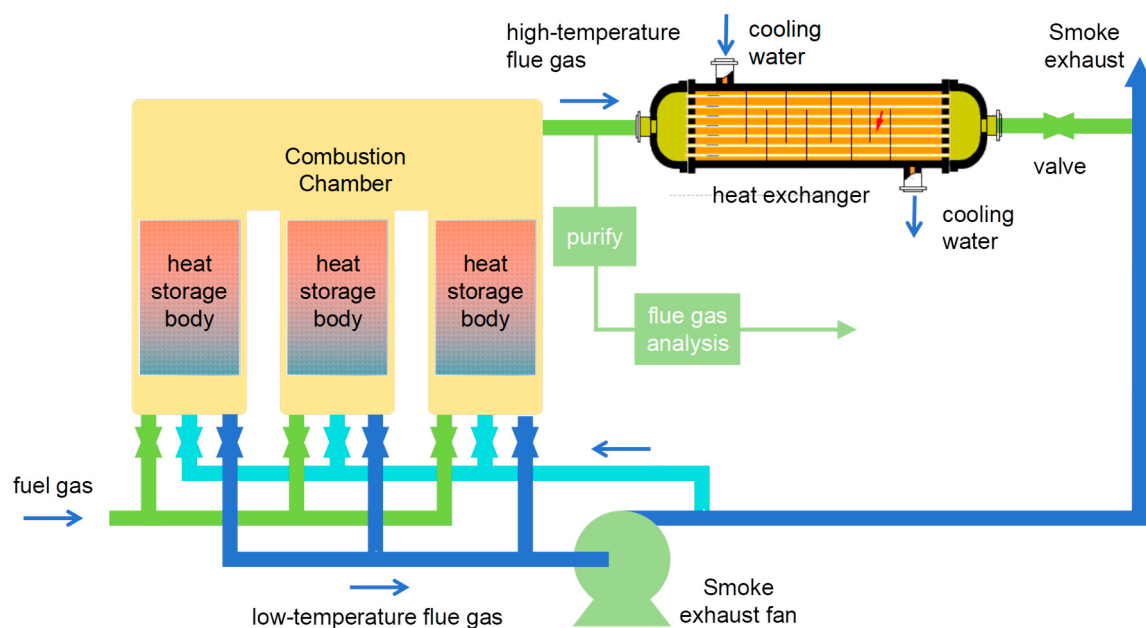


Figure 1. Diagram of the ultra-low-concentration methane regenerative thermal oxidation process.

The RTO unit employs a three-bed regenerative heat exchange system. The mixture of methane and air is used as fuel gas to simulate low concentration gas, and the specific operating conditions are shown in Table 1. The combustion products form smoke exhaust, mainly composed of N_2 , CO_2 and H_2O . After heat storage in the regenerative chamber, the fuel undergoes oxidation reactions in the combustion chamber, with some of the combustion products forming high-temperature flue gas. The high-temperature flue gas pipeline is equipped with a heat exchanger, and a flue gas sampling point is added before the heat exchanger, with a valve added after the heat exchanger to adjust the flow rate. Another portion of the flue gas from the combustion chamber exchanges heat with the regenerative material to form low-temperature flue gas. A photograph of the RTO system can be seen in Figure 2.

Table 1. Working condition data.

Name	Unit	Value	Remark
Initial Methane Concentration	%	0.28~2.5	The remaining components are air
Low-Temperature Flue Gas Proportion	%	0~100	/
Operational Load	kW	5~15	100% load is 10 kW

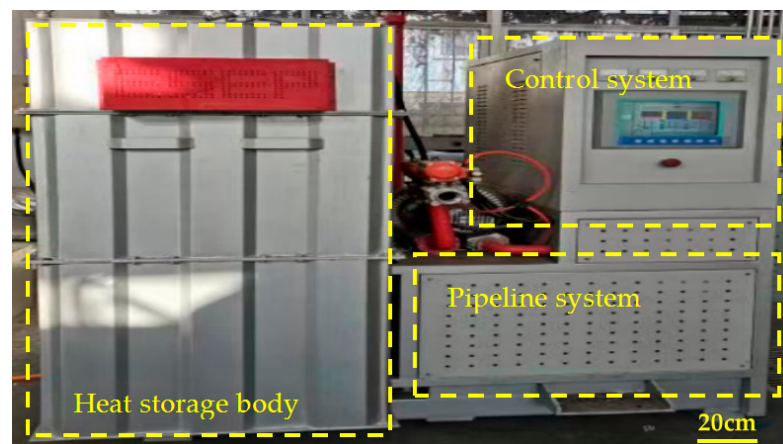


Figure 2. Photograph of the RTO test bench.

2.2. General Experiment Information

The experiment uses the initial methane concentration, fuel gas flow rate, and low-temperature flue gas proportion as the main parameters for adjusting the regenerative thermal oxidation system, exploring their influence on factors such as combustion temperature, high-temperature output energy, and thermal efficiency of the system. The working condition data can be found in Table 1.

The definitions of the concepts used in the Results and Discussion sections are as follows:

$$\text{Low-temperature flue gas proportion} = \frac{\text{Low-temperature flue gas volume}}{\text{Total flue gas volume}}$$

$$\text{High-temperature output energy} = \text{High-temperature flue gas volume} \times (\text{High-temperature flue gas enthalpy} - \text{Ambient-temperature flue gas enthalpy})$$

$$\text{System thermal efficiency} = \frac{\text{High-temperature output energy}}{\text{Fuel input energy}}$$

$$\text{Methane oxidation rate} = \left(1 - \frac{\text{Total flue gas volume} \times (\text{Flue gas CO concentration} + \text{Flue gas CH}_4 \text{ concentration})}{\text{Fuel input energy}}\right) \times 100\%$$

3. Results and Discussion

3.1. Experiment on Concentration Range

Studying the lower limit concentration for the stable operation of regenerative combustion is of great significance. According to China's Coal Mine Safety Regulations, in order to prevent gas accidents, the concentration of exhaust gas in actual mine production must generally be lower than 7%. There is a difference in the concentration of exhaust gas between low-gas mines and high-gas mines in China. The concentration of exhaust gas in low-gas mines is generally between 0.05% and 0.4%; the concentration of exhaust gas in high-gas mines is generally between 0.2% and 0.6%. If the methane concentration is too low, due to the presence of heat loss and low-temperature flue gas loss, the system cannot maintain self-sustaining stable combustion, according to the law of energy conservation. There is a correlation between the initial methane concentration, the proportion of low-temperature flue gas, and the combustion temperature. The combustion temperature increases with the increase in the initial methane concentration. There is a lower limit for the initial methane concentration, and the system cannot sustain combustion when the initial methane concentration is below this lower limit. Figure 3 illustrates the variation in combustion temperature with the initial methane concentration.

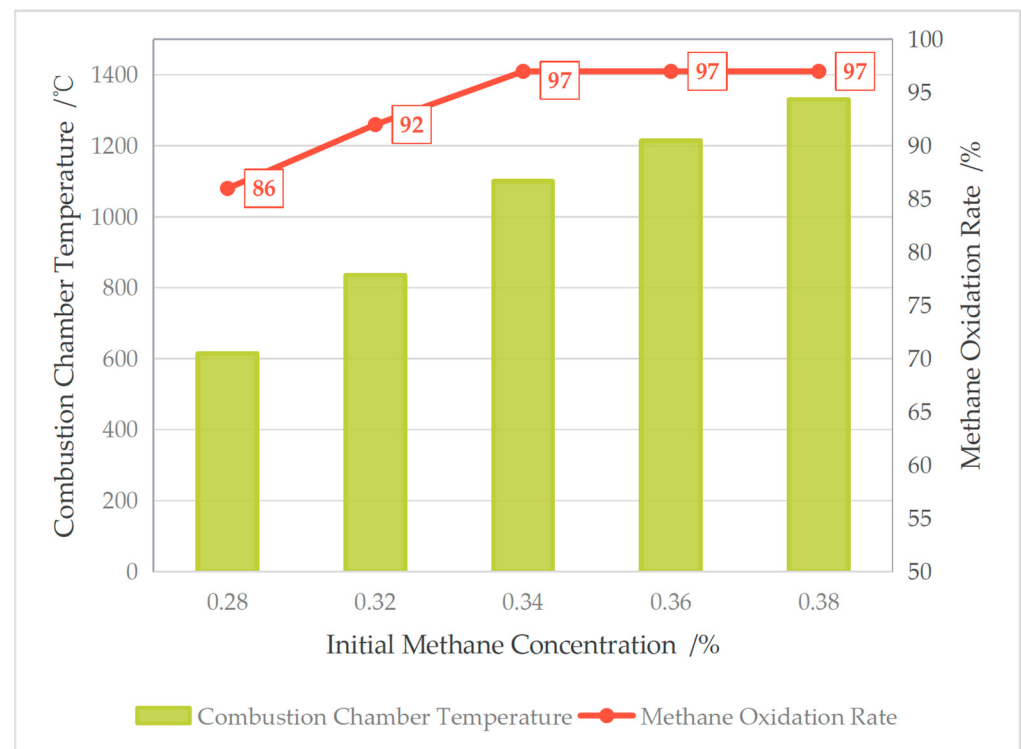


Figure 3. Variation curve of combustion temperature and methane oxidation rate with initial methane concentration.

As can be seen from Figure 3, the combustion temperature gradually decreases as the initial methane concentration decreases. When the initial methane concentration is less than 0.28%, the combustion chamber cannot maintain stable combustion due to the low temperature, and at the same time, the methane oxidation rate will also drop to 86%. This indicates that methane cannot react sufficiently under these furnace temperature conditions. Under any initial methane concentration, the higher the proportion of low-temperature flue gas, the higher the combustion temperature. When the proportion of low-temperature flue gas reaches 100%, the highest furnace temperature achieved with this initial methane concentration is reached. Figure 3 shows that as the initial methane concentration gradually decreases, the maximum combustion temperature also gradually decreases. The decrease in combustion temperature further leads to a decrease in the CH_4 oxidation rate, and the combustion reaction cannot proceed effectively, resulting in the system being unable to maintain stability due to the continuous decrease in temperature. At this point, the initial methane concentration is the lower limit concentration, which is 0.28% in this experimental system. The corresponding combustion temperature is 600 °C, and the CH_4 oxidation rate is 86%. It should be noted that the lower limit concentration changes with the heat dissipation performance of the system and the performance of the heat storage body. Enhancing insulation or improving the heat storage capacity and heat transfer performance of the heat storage body can expand the lower limit of the initial methane concentration. This conclusion has been confirmed by the experimental results of Wang Pengfei et al.; the amount of heat loss can be controlled by adding an electric heating device to the outer surface of the RTO device. When the set temperature is increased, heat dissipation on the external surface of the device can be reduced. The other conditions remain unchanged, and the lower limit concentration of methane in the intake air that maintains the self-sustaining operation of the system can be reduced from 0.45% to 0.4%, expanding the methane concentration range in the intake air. Under other unchanged conditions, as the set temperature of the heating strip on the outer wall surface of the device continuously decreases, i.e., the heat loss increases, the highest temperature of the oxidation bed and the outlet temperature both decrease [25]. The lower limit of methane

concentration required to maintain the self-sustaining combustion of the system obtained by Lv Yuan in their experiment is 0.5% [20]. Salomons et al. conducted a catalytic combustion reaction of low-concentration CH_4 on a pilot flow reversal reactor, indicating that the reaction can still operate on self-sustaining combustion even when the volume fraction of CH_4 in the feed is as low as 0.19%, and CH_4 maintains a high conversion rate. It can be seen that the lower limit of methane concentration is not a fixed value. Improving the insulation performance of the exhaust heat countercurrent oxidation system and reducing heat loss is very beneficial for increasing the oxidation bed temperature and methane conversion rate. Figure 3 also reveals that the methane oxidation rate increases with the rise of combustion temperature. Similarly, the methane oxidation rate is not only related to the reaction temperature but also to the flue gas residence time and the properties of the heat storage body.

3.2. The Impact of Initial Methane Concentration

Four different low-temperature flue gas proportions, 0.60, 0.48, 0.36, and 0.12, were selected for an experimental study on the variation in combustion temperature with low-temperature flue gas proportion, and the results are shown in Figure 4. It can be observed that the combustion temperature gradually increases with the increase in the initial methane concentration. Under the same initial methane concentration, the combustion temperature is positively correlated with the low-temperature flue gas proportion. Conversely, at the same combustion temperature, the low-temperature flue gas proportion is negatively correlated with the initial methane concentration.

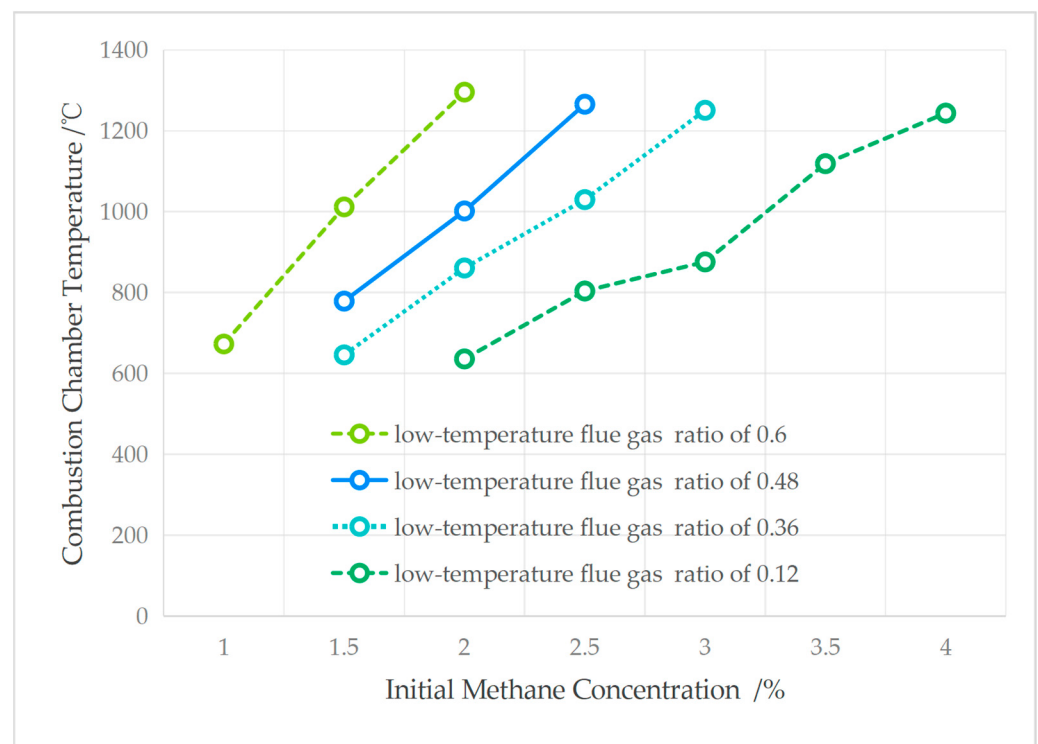


Figure 4. Variation curve of combustion temperature with initial methane concentration.

The research conclusion obtained by Qi et al. [26] on the experimental setup is that increasing methane concentration will increase the temperature in the setup and expand the range of high temperature zones; As the flow rate increases, the temperature inside the device increases, and the exhaust temperature also increases. The research results of Lü et al. [20]. believe that prolonging the switching period will increase the temperature field inside the device, while increasing the airflow speed will increase the temperature inside the device. The research results on the influence of methane concentration are the same

as other scholars, and the temperature inside the device will increase with the increase of methane concentration.

3.3. The Impact of Low-Temperature Flue Gas Proportion

The experimental study explores the variation of combustion temperature and system thermal efficiency with low-temperature flue gas proportion, and the results are shown in Figure 5.

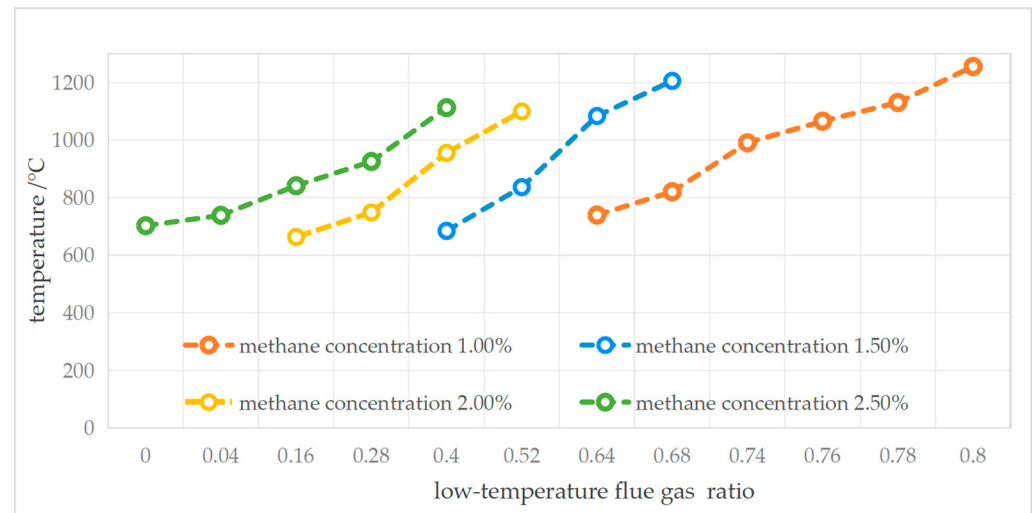


Figure 5. Variation curve of combustion temperature with low-temperature flue gas ratio.

As shown in Figure 5, for any initial methane concentration, the higher the low-temperature flue gas proportion, the higher the combustion temperature. A higher low-temperature flue gas proportion means less high-temperature flue gas is produced, which requires a greater high-temperature flue gas enthalpy to maintain the energy balance, thus resulting in a higher operating temperature. The higher the initial methane concentration, the broader the range of adjustment for the low-temperature flue gas proportion, and the smaller the low-temperature flue gas proportion at the same combustion temperature. When the initial methane concentration is less than 2.5%, there is always a lower limit value for the low-temperature flue gas proportion. The combustion temperature corresponding to this lower limit value is around 600 °C. Below this lower limit value, the reaction cannot proceed sufficiently due to the too-low combustion temperature, making it impossible for the system to maintain stability. When the initial methane concentration is higher than 2.5%, any low-temperature flue gas proportion will maintain a stable combustion temperature (above 600 °C, thus preventing the furnace temperature from being too low and slowing down the CH₄ reaction).

The variation curve of system thermal efficiency with low-temperature flue gas proportion is shown in Figure 6. Under any initial methane concentration, the system's thermal efficiency decreases with the increase in low-temperature flue gas proportion. The higher the initial methane concentration, the smaller the change in system thermal efficiency with the low-temperature flue gas proportion. Taking the case of an initial methane concentration of 1.5% as an example, when the low-temperature flue gas proportion reaches 0.7, the system thermal efficiency is 71%. If the low-temperature flue gas proportion is further increased, the combustion temperature will become too high. When the low-temperature flue gas proportion is 0.36, the system thermal efficiency is 73.5%. If the low-temperature flue gas proportion is further decreased, the combustion temperature would become too low, making stable combustion impossible. The experimental results show that to improve the thermal efficiency of the RTO system and enhance its heating capacity, it is necessary to increase the initial methane concentration or reduce the low-temperature flue gas pro-

portion. However, too low a proportion of low-temperature flue gas may lead to unstable operation due to excessively low combustion temperatures.

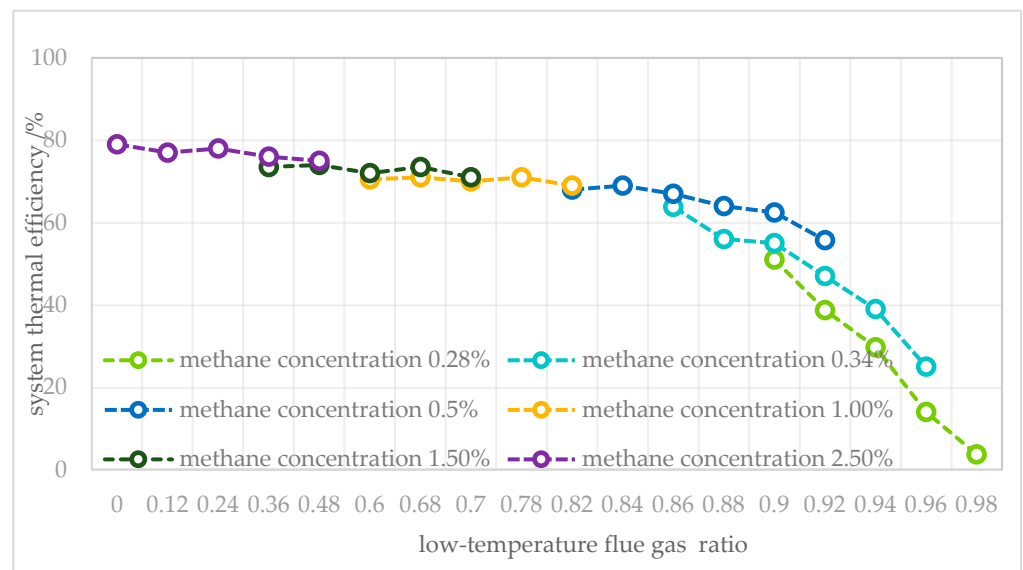


Figure 6. The variation curve of system thermal efficiency with low-temperature flue gas proportion.

The curve showing the change in system output heat with the low-temperature flue gas proportion is presented in Figure 7. Under any initial methane concentration, the system output heat decreases as the low-temperature flue gas proportion increases. The pattern of change in system output heat is consistent with that of system thermal efficiency. It is without question that the higher the initial methane concentration, the stronger the heating energy of the system.

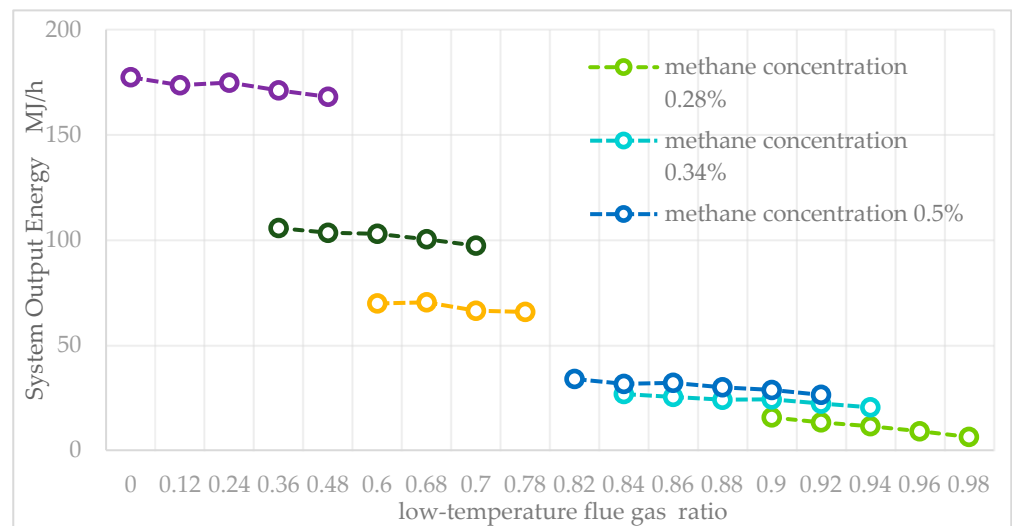


Figure 7. Curve showing the change in system output energy with the low-temperature flue gas proportion.

3.4. The Impact of Operational Load

The influence of the operational load rate on the temperature of the combustion furnace and the oxidation rate of methane is shown in Figure 8.

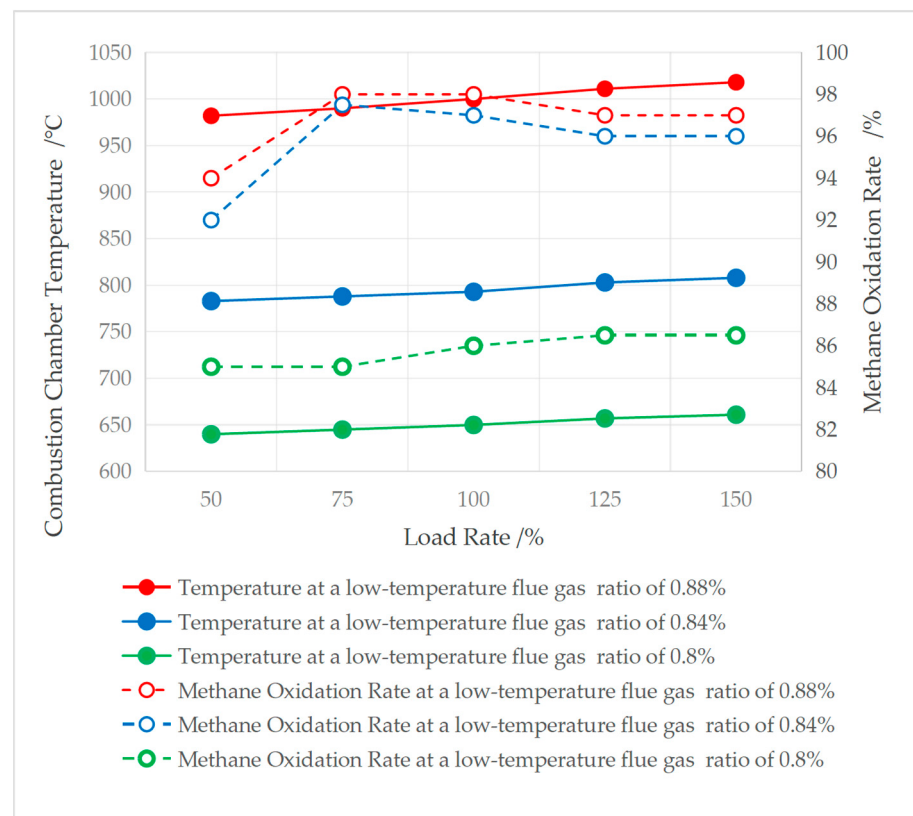


Figure 8. The curve showing the influence of load rate on the temperature of the combustion furnace and methane oxidation rate.

The system is designed to handle raw gas with a methane concentration of 0.5% at a flow rate of 200 m³/h (operating at 100% load). The experiment selected three Low-temperature flue gas proportions, which were 0.88, 0.84, and 0.8. As the operating load rate increased from 0.5 to 1.5, the combustion temperature showed a consistent upward trend, although the increase was not significant; the methane oxidation rate increased with the load rate from 0.5 to 1, but decreased from 1 to 1.5. At a load rate of 1.5, the combustion temperature reached its peak, yet the methane oxidation rate slightly decreased, which may be related to the reduced residence time of the fuel in the combustion chamber as the load rate increased. The results of this experiment are also consistent with the research of other researchers such as Wang Pengfei; under the same simulated exhaust flow rate, the conversion rate of low-concentration methane increases with the increase in the preheating temperature at the center of the oxidation bed, and the CO content in the reaction tail gas also significantly decreases with the increase in temperature. At the same preheating temperature, the methane conversion rate and flow rate in the simulated exhaust air are closely related. As the simulated exhaust air flow rate increases, the methane conversion rate shows a downward trend. This is because the increase in flow rate increases the speed of the simulated exhaust air passing through the oxidation bed, thereby shortening the gas preheating time and making the low-concentration methane reaction in the simulated exhaust air insufficient. In their experiment, Lv Yuan studied the operating characteristics of the actual ventilation rate under a fluctuation of 60% above or below the design processing capacity. He found that as the ventilation rate decreased, the high-temperature zone gradually shortened, and the main impact on the operation of the device was to reduce the total energy flow of the device. The decrease in the ventilation rate shortens the length of the high-temperature zone required for methane oxidation while ensuring the same residence time in the high-temperature zone. The ability of the heat storage bodies on both sides to preheat and intake air is enhanced, which has a positive effect on the methane oxidation reaction and is beneficial for improving the methane conversion rate. Increasing

the ventilation rate will reduce the methane conversion rate, which is consistent with the research results in this paper. In the experiment conducted in this article, to ensure the full oxidation of methane in the simulated exhaust air, the preheating temperature at the center of the oxidation bed needed to be above 800 °C and the feed gas flow rate needed to be between 150–250 m³/h.

4. Conclusions

At any initial methane concentration, the higher the low-temperature flue gas proportion, the higher the combustion temperature. When the low-temperature flue gas proportion is 100%, the highest combustion temperature is achieved. The experiment found that when the combustion temperature is below 600 °C, the CH₄ combustion reaction cannot proceed effectively, and the system temperature continues to decline and cannot be maintained stably. In the present experimental system, the lower limit of the initial methane concentration is 0.28%, corresponding to a methane oxidation rate of 86%. Below this concentration, the methane cannot react effectively due to the too-low combustion temperature, resulting in the inability to maintain a stable temperature in the combustion chamber.

The initial methane concentration has a significant impact on the stability and temperature of combustion. When the methane concentration is too low, heat loss and low-temperature flue gas loss lead to the system's inability to maintain self-sustained stable combustion. Under the same initial methane concentration, the combustion temperature is positively correlated with the low-temperature flue gas proportion. At the same combustion temperature, the low-temperature flue gas proportion is negatively correlated with the initial methane concentration.

The low-temperature flue gas proportion is positively correlated with the combustion temperature, which means that a higher combustion temperature is required to maintain energy balance with a larger low-temperature flue gas proportion. There is a lower limit for the low-temperature flue gas proportion when the initial methane concentration is low; below this limit, the combustion temperature is too low, and the reaction cannot proceed sufficiently. Moreover, both the system thermal efficiency and the high-temperature output energy decrease with the increase in the low-temperature flue gas proportion.

The operating load rate has a certain impact on the combustion temperature and methane oxidation rate. With the increase in the operating load rate, the combustion temperature tends to rise, but the increase is not significant. The methane oxidation rate decreases after the load rate reaches a certain value, which may be related to the reduced residence time of the fuel in the combustion chamber.

To maintain the stable operation of regenerative thermal oxidation, it is necessary to consider the initial methane concentration, low-temperature flue gas proportion, and operating load rate comprehensively. By adjusting these parameters, it is possible to ensure the stability of combustion while improving the system's thermal efficiency and output heat. Furthermore, the system's heat dissipation performance and the performance of the regenerative material are also important factors affecting the stable operation of combustion. By optimizing these properties, it is possible to further expand the lower limit of the initial methane concentration.

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Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Conflicts of Interest: Authors Junhui Yang and Jida Zhang were employed by the company China Coal (Tianjin) Underground Engineering Intelligent Research Institute Co., Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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