**ISMS 2016** 

# Multi-bed type oxidation reactor applied to the coal mine ventilation air methane

Weifeng Zou<sup>a,b,\*</sup>, Bo Lan<sup>a,b</sup>, Jiandong Kang<sup>a,b</sup>

<sup>a</sup> Key Laboratory of Low-grade Energy Utilization Technologies & Systems of Ministry of Education, College of Power Engineering, Chongqing University, Chongqing, China, 400044

<sup>b</sup> Chongqing Research Institute Co.,Ltd. of China Coal Technology & Engineering Group, Chongqing, China, 400037

### ABSTRACT

The utilization of coal mine ventilation air methane plays an important role in saving energy, reducing pollution, improving the safety in coal mine production, and adjusting the energy structure. The thermal flow-reversal reactor is one of the main technologies of ventilation air methane utilization which is at a critical stage of industrial application. The thermal flow-reversal reactor of MEGTEC company and those designed by some Chinese scholars generally use a two-bed type. The gas that is detained in the oxidation chamber and reversal valves will not enter the combustion chamber, but through the chimney directly into the atmosphere when switching the flow direction, the average oxidation rate of the reactor will be reduced due to the residual gas.

In order to solve the problems including low oxidation rate of methane, pressure build-up of the main fan, and high failure rate of valves in existing two-bed type oxidation reactor of coal mine ventilation air methane, an innovative structure with multi-bed type oxidation reactor has been designed. The paper analyzes the unique advantages of the multi-bed type oxidation reactor by elaborating on the working principles of three-bed and five-bed type device. In the design of the oxidation device for industrial demonstration projects, the device whose processing capacity is 100,000 m<sup>3</sup>/h adopts the five-bed type structure from the aspects of methane oxidation rate, waste heat utilization effect, cost, etc. The thermodynamic calculation process of regenerative chambers as the core unit of the oxidation device is described in this paper.

The technology has an industrial demonstration project whose reactor uses a five-bed type design located in Chongqing Songzao Datong No.1 mine. This project can produce 64800 t of superheated steam and reduce emissions of  $CO_2$  equivalent to 107000 t annually, and therefore has excellent energy-saving and emission reduction benefits.

KEYWORDS: multi-bed type; coal mine ventilation air methane; thermal flow-reversal oxidation; industrial application

### 1. INTRODUCTION

Coal bed methane is a kind of clean and efficient energy which is one of the rising unconventional natural gases in recent years. The ventilation air methane (VAM) accounts for about 60% of total methane emissions from coal mines, and its utilization plays an important role in saving energy, reducing pollution, improving the safety in coal mine production, and adjusting the energy structure (Gosiewski et al., 2009). However, the development of the utilization technology is restricted by factors such as low concentration, large air volume and instability of ventilation air methane. The thermal flow-reversal reactor (TFRR) is one of the main technologies of VAM utilization which is at a critical stage of industrial application. The small thermal oxidation device has a processing capacity of 1000 m<sup>3</sup>/h and was designed by Zheng. It uses a two-bed horizontal type (Zheng et al., 2009). The methane

oxidation rate is slightly higher than 95% of this device. The thermal oxidation device with a maximum processing capacity of 1000 m<sup>3</sup>/h was designed by Lv. It also uses a two-bed horizontal type (Lv et al., 2011). The device should be operated steadily when the flow is in the range of 400-800  $m^{3}/h$  and the concentration of methane is in the range of 0.5%-0.8%. The thermal oxidation device has a maximum processing capacity of 500 m<sup>3</sup>/h and was designed by Wang. It uses a two-bed vertical type and some experimental studies are carried out based on this device (Wang et al., 2012). The MEGTEC Company whose oxidation device uses a two-bed vertical type also is the most representative of this technology abroad. It has been found that the current oxidation device of VAM on the regenerative chamber structure is generally a two-bed type. However, the gas that was detained in the oxidation chamber and reversal valves will not enter the

combustion chamber but goes through the chimney into the atmosphere directly when switching the flow direction, so the average oxidation rate of the reactor will be reduced due to the residual gas (Kang et al., 2015). To solve this problem, this paper puts forward the design idea of the multi-bed type for the structure of thermal flow-reversal reactors.

### 2. THE NECESSITY OF DEVELOPING THE MULTI-BED REGENERATIVE OXIDATION REACTOR

# 2.1 Working principle of two-bed type regenerative oxidation reactor

For a two-bed type oxidation reactor, there are two combustion chambers, one as the inlet and the other as the outlet of mixed gas. After the reactor is heat-actuated, the VAM flows at ambient temperature from the low temperature side (e.g., chamber 1) into the regenerative chamber, then is heated, oxidized and released as thermal energy. The released energy during methane oxidation is recovered by ceramic as the gas moves to the outlet side of the chamber. This process is often referred to as the upper half cycle of the circulating operation. At this time, chamber 1 is in the thermal release stage and chamber 2 is in the thermal storage stage. The direction of VAM flow should be switched and the reversed process carried out, and the system should be auto-thermal operational with continuous transformation between two chambers at high temperature and low temperature.



Figure 1: Schematic of two-bed type TFRR.

### 2.2 Disadvantages of two-bed type TFRR

Chongqing Research Institute of China Coal Technology & Engineering Group independently developed a small test device of TFRR with a processing capacity of 1000 m<sup>3</sup>/h during the 11th Five-Year Plan period of China, named Low Concentration Coal Bed Methane Utilization Technology and Equipment which belongs to the National Science and Technology Major Project. A lot of studies have been carried out based on the utilization system of coal mine ventilation air methane. The experimental system is shown in Figure 2.



Figure 2: Diagram of two-bed type TFRR experimental system.

The two-bed type thermal flow-reversal reactor has some inevitable disadvantages due to structure, as follows.

a. There will be a large fluctuation of the gas flow rate from the total intake to zero when switching the flow direction, so that the main fan will be suppressed and its life will be affected.

b. The flue gas flow rate of the waste heat boiler will periodically fluctuate, which may affect the quality of the superheated steam in the use of waste heat.

c. The large size of valves results in a large force being requires for action and failure rate will be high.

d. The gas distribution in the regenerative chamber and the combustion chamber is not uniform and it is not conducive to the oxidation of methane in VAM because of the large cross section of the regenerative chamber.

e. The gas who was detained in the oxidation chamber and reversal valves will not enter the combustion chamber but go through the chimney into the atmosphere directly when switching the flow direction, so the average oxidation rate of reactor can only reach 95% due to the residual gas.

# 3. WORKING PRINCIPLE OF MULTI-BED TYPE TFRR

The core idea of multi-bed regenerative oxidation reactors is to add some regenerative chambers for oxidizing the methane in VAM, and also to add a regenerative chamber whose main function is sweeping. For a three-bed type oxidation reactor, as shown in Figure 3, there are three regenerative chambers, one as the inlet, one as the outlet of mixed gas, and another as the inlet of sweeping gas. For a five-bed type oxidation reactor, as shown in Figure 4, there are five regenerative chambers, two as the inlet, two as the outlet of mixed gas, and another as the inlet of sweeping gas. The data in Table 1 uses the five-bed type oxidation reactor as an example. A complete working cycle consists of five cycles, where each regenerative chamber periodically switches between the three stages of thermal storage, sweeping, and thermal release.



Figure 3: Structure schematic diagram of three-bed type TFRR.



Figure 4: Structure schematic diagram of five-bed type TFRR.

Tuble 1. Work process of five bed type 11 KK.								
No.	Bed 1	Bed 2	Bed 3	Bed 4	Bed 5			
Cycle 1	а	а	b	b	с			
Cycle 2	с	а	а	b	b			
Cycle 3	b	с	а	а	b			
Cycle 4	b	b	с	а	а			
Cycle 5	а	b	b	с	a			
Cycle 5	а	b	b	с	а			

Table 1: Work process of five-bed type TFRR

Note: a. thermal storage, b. thermal release, c. sweeping

The present study also considered the situation of increasing the number of bed on the basis of the fivebed type oxidation reactor. The numerical simulation and theoretical calculation of the seven-bed and ninebed type oxidation reactor were carried out. It was found that there would need to be a large number of valves due to the increase of the oxidation beds. The control of oxidation reactors is very complex and not conducive to its stable operation. In addition, the cost will increase and the economical profits will be reduced. Therefore, the so-called multi-bed type of TFRR in industrial applications usually refers to the three-bed or five-bed type devices.

## 4. ADVANTAGES OF MULTI-BED TYPE TFRR

Compared with the two-bed type oxidation reactor, the multi-bed type oxidation reactor has the following advantages:

a. There are more than two chambers in the multi-bed type oxidation reactor and one of them is designed to sweep the detained gas. The methane in VAM is not oxidized and distributed between the reversal valves and regenerative chamber to the combustion chamber in the traditional structure of the oxidation reactor when the flow direction is switched, so it avoids discharging into the atmosphere directly.

b. The cross-sectional area of the oxidation chamber should be decreased, and its gas distribution is more uniform due to the smaller processing capacity of a single chamber of the multi-bed type oxidation reactor. This avoids the non-uniform distribution of gas in traditional oxidation reactors which should oxidize the larger flow rate of mixed gas.

c. The working state should be changed from intake to sweep in only one chamber of the inlet chambers, and the rest of the inlet chambers remain unchanged when switching the flow direction. The gas flow rate is less, and the effect of waste heat utilization is better.

# 5. INDUSTRIAL APPLICATION OF MULTI-BED TYPE TFRR

#### 5.1 Selection of industrial device

An industrial demonstration project will be established in the 12th Five-Year Major Project (Direct use technology of low concentration coal bed methane) as a continuation of the research tasks of 11th Five-Year Major Project of China. Based on the summary of the application, the industrial device whose processing capacity is 100000  $\text{m}^3$ /h will be intended to adopt a multi-bed type structure design.

The results obtained by calculating and analyzing the parameters of two-bed, three-bed, and five-bed type structures according to the design and calculation method of the regenerative heat exchanger are shown in Table 2. The industrial device whose processing capacity is 100000 m<sup>3</sup>/h adopted the five-bed type structure from the aspects of methane oxidation rate, waste heat utilization effect, cost, etc.

	two-bed type	Three-bed type	Five-bed type
Single cross-sectional area of regenerative chamber $(m^2)$	4×5.5=22	4×5.5=22	4×2.95=11.8
Single volume of regenerative chamber (m <sup>3</sup> )	46	46	23
Total volume of regenerative chamber (m <sup>3</sup> )	92	138	115
Volume of heat preservation material (m <sup>3</sup> )	107	145	177
Device size(m <sup>3</sup> )	14.4×4.7×8	18.6×4.7×8	18.45×4.7×8
Reversing valves	DN1300	DN1300	DN1000
Number of reversing valves	4	6	10
Number of sweeping valves	0	3	5
Cost of regenerative chamber (RMB, Million Yuan)	110.4	165.6	138
Cost of valves (RMB, Million Yuan)	60	99	95
Cost of heat preservation material (RMB, Million Yuan )	53.5	72.5	88.5
Total cost (RMB, Million Yuan)	223.9	337.1	321.5
oxidation rate of methane	95%	98%	98%

Table 2: Comparison of different structure types.

#### 5.2 Design of industrial device

The thermal flow-reversal reactor of coal mine ventilation air methane is a precise and complex system. The whole device is composed of the regenerative chambers, combustion chambers, some valves, and so on. As the core part of the device, the regenerative chamber is the cornerstone in the course of whole device design. The heat exchange area, the volume, and the size of the regenerative chamber are calculated according to the design method of the regenerative heat exchanger. The thermodynamic calculation process is as follows:

a. The heat (Q) required for VAM in a cycle of flow reversal

 $Q = V_f \left( c_f^{\dagger} t_f^{\dagger} - c_f^{\dagger} t_f^{\dagger} \right) \tau_h \quad (1)$ 

Where:  $V_f$  is the flow rate of VAM that be heated,  $m^3/s$ .

 $c'_{f}, c'_{f}$  are the inlet and outlet specific heat of VAM, J/(kg·K).

 $t_{f}, t_{f}$  are the inlet and outlet temperature of VAM. °C

- $\tau_h$  is the duration of VAM for heating, s.
- b. The flue gas outlet temperature( $t_y$ ) of regenerative chamber

$$t_{y}^{"} = \frac{1}{c_{y}^{"}} \left( c_{y}^{'} t_{y}^{'} - \frac{Q}{V_{y} \tau_{y} \eta} \right)$$
(2)

Where:  $V_{y}$  is the flow rate of flue gas, m<sup>3</sup>/s.

 $c'_{y}, c'_{y}$  are the inlet and outlet specific heat of flue gas, J/(kg·K);

 $t'_{y}$  is the inlet temperature of flue gas, °C

 $\tau_{\rm y}$  is the duration of flue gas for cooling, s.

 $\eta$  is the heat efficiency of regenerative chamber,  $\eta = 0.9 \sim 0.95$ .

c. The logarithmic mean temperature difference  $(\Delta t)$ 

$$\Delta t = \frac{\Delta t_{\max} - \Delta t_{\min}}{\ln \Delta t_{\max} / \Delta t_{\min}}$$
(3)

Where:  $\Delta t_{\max} = t_{y} - t_{f}$ ,  $\Delta t_{\min} = t_{y} - t_{f}$ .

d. The convection heat transfer coefficient ( $\alpha$ )

$$\alpha = 7.5\phi \frac{W^{0.3}}{de^{0.33}}$$
(4)

Where:  $\phi = 0.184T^{0.25}$ , the temperature correction factor, *T* is temperature, K.

*de* is the equivalent diameter of honeycomb ceramic, m.

W is the velocity of flow at the minimum cross section, m/s.

e. The integrated heat exchange coefficient ( K )

$$K = \frac{\left[\frac{1}{\alpha_{y}\tau_{y}} + \frac{1}{\alpha_{f}\tau_{f}} + \frac{S}{6\lambda}(1/\tau_{y} + 1/\tau_{f})\right]^{-1}}{(\tau_{y} + \tau_{f})}$$
(5)

Where: *S* is the thickness of honeycomb ceramic, m.

 $\alpha_f, \alpha_y$  are the convection heat transfer coefficient of VAM-side and flue gas-side, W/(m<sup>2</sup>·K).

 $\lambda$  is the thermal conductivity of honeycomb ceramic, W/(m·K).

f. The heat exchange area of a regenerative chamber ( *A* )

$$A = \frac{Q}{K\Delta t\eta \left(\tau_{y} + \tau_{f}\right)}$$
(6)

g. The volume of a regenerative chamber (V)

$$V = \frac{A}{a_v} (7)$$

Where:  $a_v$  is the specific surface area of honeycomb ceramic, m<sup>2</sup>/m<sup>3</sup>.

h. The horizontal cross-sectional area of a regenerative chamber (F)

$$F = \frac{V_f}{W_f \sigma}$$
(8)

Where:  $W_f$  is the velocity of VAM, m/s.

Table 3: Parameters and thermodynamic calculation results.

i. The height of regenerative chamber (H)

$$H = \frac{V}{F} (9)$$

According to the calculation formulas above, the basic parameters of the industrial device (processing capacity of VAM is 100000  $m^3/h$ ) are given and the FORTRAN software are used for thermodynamic calculations. The related parameters and calculation results are shown in Table 3.

No.	Items	Units	Numerical values
1	VAM Processing capacity of single oxidation bed	m³/h	50000
2	Inlet temperature of VAM	C°	20
3	Outlet temperature of VAM	°C	787
4	Flow rate of flue gas	m <sup>3</sup> /h	50000
5	Inlet temperature of flue gas	C°	900
6	Outlet temperature of flue gas	C°	60
7	Heat loss coefficient	%	5.0
8	Logarithmic mean temperature difference	C°	70
9	Duration of the cycle of flow reversal	min	3
10	Volume of honeycomb ceramic	m <sup>3</sup>	23.35
11	Circulation cross-sectional area	m <sup>2</sup>	11.12
12	Height of honeycomb ceramic	m	2.1
13	Total volume of honeycomb ceramic	m <sup>3</sup>	116.75
14	Size of regenerative chambers	m×m×m	18.45m×4.7m×2.6m
15	Velocity of VAM	m/s	1.86

In this point, it is necessary to point out that the parameters obtained such as the height of the honeycomb ceramic and the circulation crosssectional area are based on the determined parameters and structure of the honeycomb ceramic. In this paper, the selection of honeycomb ceramic and its layout principles are omitted, see Figure 5.



Figure 5: Schematic diagram of ceramic's layout.

#### 5.3 Industrial demonstration project

The industrialization of this utilization technology is promoted through the establishment of the industrial demonstration project of the VAM. Some industrial tests were carried out about the industrial demonstration system in which the reactor uses a five-bed type design in Chongqing Songzao Datong No.1 mine, to investigate the effect of collecting cover, test the methane oxidation rate of



the reactor, test the technology of steam produced by waste heat, and to improve and perfect the industrial

device according to the test results.

 Diffusing tower. 2. VAM collecting cover. 3. Mixer.
4. TFRR. 5. Waste heat boiler. 6. Chimney.
Figure 6: Schematic diagram of industrial demonstration system for VAM utilization.

The whole system is composed of the VAM collecting and mixing system, regenerative oxidation reactor, the waste heat utilization system, and the monitoring system, as shown in Figure 6. The ventilation air methane is exhausted from the ventilating shaft and sent into the pipeline after collection by the collecting cover, then brought into

the oxidation reactor by induced-draft fan. The methane in VAM is heated and oxidized and the oxidation products such as  $CO_2$  and  $H_2O$  (water vapor) are discharged into the atmosphere. The burner in the combustion chamber will burn diesel oil to preheat the regenerative ceramic for starting the reactor. In the test of waste heat utilization the drainage gas is mixed with VAM in order to make the volume fraction of methane reach 1%, and take part of the high temperature flue gas to the boiler, to produce superheated steam.



Figure 7: Scene diagram of industrial demonstration project.

The construction of the demonstration project was completed in October 2015 at which point the system was debugged. It is estimated that when it is put into operation, it will produce 64800 t of superheated steam and reduce emissions of  $CO_2$  equivalent to 107000 t annually, and therefore has excellent energy-saving and emission reduction benefits.

### 6. CONCLUSIONS

a. Through the introduction of working principles of the traditional two-bed type oxidation reactor, it is found that this type of reactor has some disadvantages including low oxidation rate of methane, pressure build-up of the main fan, and the high failure rate of valves.

b. In view of the design idea for a multi-bed type oxidation reactor, their unique advantages are pointed out by comparing with the two-bed type oxidation reactor.

c. By calculating and analyzing the parameters of two-bed, three-bed, and five-bed type structures, the industrial oxidation reactor whose processing capacity is 100000 m<sup>3</sup>/h adopted the five-bed type structure design, and according to the design and calculation method of the regenerative heat exchanger, the thermodynamic calculation process and results of regenerative chamber were introduced.

d. When the demonstration project located in Chongqing Songzao Datong No.1 mine is put into operation, it will produce 64800 t of superheated steam and reduce emissions of CO<sub>2</sub> equivalent to

107000 t annually, and therefore has excellent energy-saving and emission reduction benefits.

### 7. ACKNOWLEDGEMENT

This study was financially supported by National Science and Technology Major Project of China (2011ZX05041-005).

### 8. References

Deng, H.X., Xiao, Q. and Xiao, Y.H. (2014). Design method of thermal flow-reversal reactor for ventilation air methane based on regenerative heat exchange model. Journal of China Coal Society, Volume 39, No. 7, pp. 1302–1308.

Feng, T., Wang, P.F. and Hao, X.L. (2012). Experimental study on thermal flow-reversal oxidation of coal mine ventilation air low concentration methane. China Safety Science Journal, Volume 22, No. 10, pp. 88–93.

Gosiewski, K. and Warmuzinski, K. (2007). 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, Volume 62, pp. 2679–2689.

Gosiewski, K., Pawlaczyk, A. and Warmuzinski, K. (2009). A study on thermal combustion of lean methane-air mixtures: simplified reaction mechanism and kinetic equations. Chemical Engineering Journal, Volume 154, pp. 9–16.

Kang, J.D., Lan, B. and Zou, W.F. (2015). Design and application on five-bed type thermal accumulation oxidized device of mine ventilation air methane. Coal Science and Technology, Volume 43, No. 2, pp. 136–139.

Lv, Y., Jiang, F. and Xiao, Y.H. (2011). Experimental study of coal mine ventilation methane oxidization. Journal of China Coal Society, Volume 36, No. 6, pp. 973–977.

Wang, P.F. (2012). Study on theory and experiment of thermal flow-reversal oxidation of coal mine ventilation air methane. Central South University, Hunan, China.

Xiao, Q., Deng, H.X. and Lv, Y. (2012). Lean methane-air premixed-gas heating process research. Journal of Mining & Safety Engineering, Volume 29, No. 2, pp. 295–300.

Zheng, B., Liu, Y.Q. and Liu, R.X. (2009). Oxidation of coal mine ventilation methane of thermal flow-reversal reactor. Journal of China Coal Society, Volume 34, No. 11, pp. 1475–1478.

Zhou, X. (2009). Experiment study of coal mine ventilation air methane oxidation. Institute of Engineering Thermophysics Chinese Academy of Science, Beijing, China.