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Research advances and trends of the dilute methane thermal reverse flow reactor technology

Pengfei Gao[1,2,3 a*]

[1]Power Engineering College, Chongqing University, Chongqing, 400044, China

[2]State Key Laboratory of The Gas Disaster Detecting, Preventing and Emergency Controlling, Chongqing, 400037, China

[3]China Coal Technology Engineering Group Chongqing Research Institute Chongqing, 400037, China)

[*]Corresponding author's e-mail: gpfmky@163.com

Abstract: Based on coal mine methane (CMM) emission policy in China and other main coal-mining countries in the world, a status quo of dilute methane (such as ventilation air methane (VAM) or similar concentration methane) reverse flow reactor (RFR) technology was looked back and the principal of the reverse flow reactor for dealing with dilute methane was introduced in detail. The development of the dilute methane RFR technology was carded and the main research directions and processes in this field were described, including reaction kinetics mechanism of dilute methane in the inert homogeneous channel, reaction heat front reverse movement rule in the RFR, numerical simulation of the RFR mathematical model, NOx emission of the RFR process, large-scale RFR experiments, and so on. The contradictions existed in the relative research of dilute methane RFR technology were discussed, and the evolution of the dilute methane RFR technology was predicted. And it would be necessary guidance for the scientific study, numerical simulation, optimization design and engineering application of the dilute methane RFR technology.

1.Introduction

Methane discharged through human activities, as a kind of non-CO_2 greenhouse gas, has caused significant effect to the global warming tendency. The total methane emission from coal mining in the world accounts for 8% of man-made methane emission, and 17% of anthropogenic greenhouse gas emission. Methane emission from VAM occupies 70% of the total coal mine methane discharged to the atmosphere [[1]]. VAM is a type of ventilation flow range from 100~300m3/s drained from the coal mine return air shaft with dilute methane, whose concentration is usually less than 1% for safety purpose [[2]]. Dealing with such a huge flow, and ultra-low concentration fuel gas of methane has caused a great challenge to the current energy utilization technology and engineering application.

Nowadays, research in the thermal or catalytic kinds of reverse flow reactor has made rapid progress, which brings light to the VAM or CMM destruction [[3]]. This kind of reverse flow reactor, with porous homogenous regenerative packing inside, works a state of pseudo-steady through the flow direction reverse switching periodically. Relying on the difference between fuel gas flow velocity and reaction heat front moving speed, the reactor would make a heat balance between heat released and heat losses through the continuous flow reverse switching periodically, and would expel redundant

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high-temperature flue for thermal utilization [[4]]. In general, the concentration of methane handled by the reverse flow reactor would be less than 1.2% volume concentration.

Some researches had been made to compare the thermal RFR and the catalytic RFR for dealing with the low concentration methane and VAM [[2],[5]]. The conclusion of the comparison believed that the type of catalytic RFR was suitable for the VAM with dilute methane concentration around 0.2%, even as low as 0.1%, and the type of thermal RFR would be fit to VAM with methane concentration higher than 0.4% for it had a better reward economically. And that's the reason why thermal RFR has wider application areas, such as heating, power generation, and so on [[6]]. The main researches of RFR were focused on mathematical modelling, methane reaction mechanisms, kinetics in the monolith ceramic channels, and lab-scale experiments, and some commercial engineering have been operating. The main idea of this paper was to present the theoretical progress and the technology status of the thermal RFR, evaluate the prospect of the thermal RFR potential market, and offer advice for methane mitigation policy making through technical vision.

2. Technical principle of thermal RFR

MEGTEC, taking lessons from RFR for dealing with volatile organic compounds (VOC), was the first company to develop a thermal RFR, named VOCSIDIZER^{TM} [[7]]. The principle of the VOCSIDIZER^{TM} was realizing the flameless oxidation of dilute methane by sustaining the combustion zone with temperature higher than the methane ignition point, relying on heat transfer between the dilute methane and the regenerative ceramics through reversing flow direction periodically, to keep the oxidation reaction going on spontaneously without extra energy. In this process, the reaction heat would be pulled out to produce superheated steam for turbine generation or other thermal utilization except holding heat balance of the RFR system. The diagram of the thermal RFR system could be seen from Fig.1.

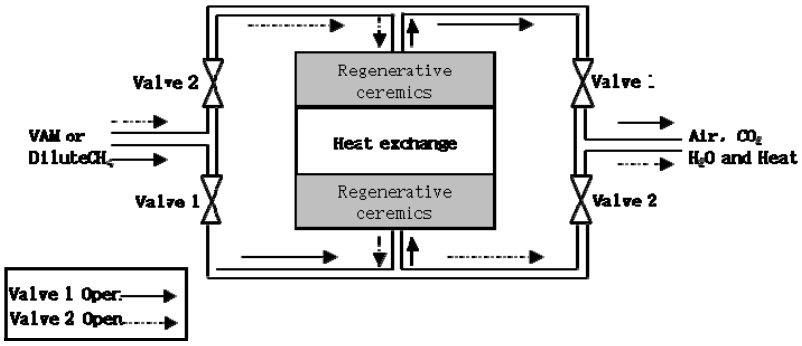


Fig.1 The diagram of the thermal RFR system

3. Main research direction and progress

The main research of the thermal RFR, including mathematical modeling and verification, experimental research, reaction mechanism reduction, reaction kinetics parameters, regenerative layer pressure, methane conversion, reactor operation stability, heat fatigue of material and design method of the reactor, and so on. We could get the main research roadmap of the thermal RFR from different research aspects during the time evolution from Fig.2.

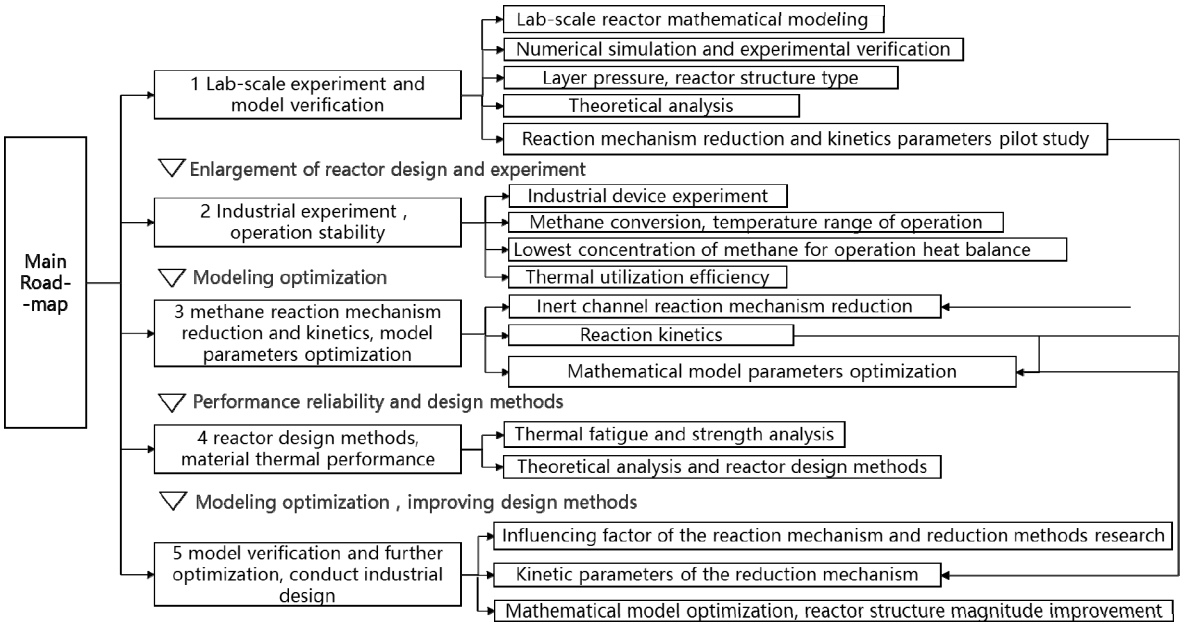


Fig.2 The main roadmap of the thermal RFR technology

3.1 Reaction mechanism and kinetics characteristics

Slepterev [[8]] gave research on dilute methane (lower than 2% v/v) uniform deep oxidation kinetics in ceramic tube and ceramic pellets packed beds, revealed that methane oxidation path was in a consecutive reaction style via CO formation and concluded that oxidation reaction velocity was relative to the equivalent diameters. In other words, it meant that inhibition effect of reaction in tubes and pellets were significant led by the influence of mass transfer to the free radical terminal in the ceramic surface. He also found that CO oxidation doesn't rely on quality exchange condition in fact, but is inhibited by methane obviously, and offered the recommended kinetic equation and parameters.

Slepterev [[9]] found that when the methane concentration was 0.5~2.0%, temperature was 700~1100℃, and contact time was 0.024 ~ 0.384s, methane oxidation occurred through CO, and the concentration of CO reaches its maximum with the rise of temperature, and the kinetic parameters were obtained through experiments. Still, his research discovered that methane is oxidized by two reaction pathways: directly generating CO_2 (at medium temperatures) and by intermediately producing CO (at high temperatures) with selection conversion rate reaching 70-90% as temperature increases. Under certain conditions, ethane, ethylene and formaldehyde are formed in the reaction mixture, and their content couldn't be negligible. As the characteristic size of the free volume (diameter of the tubular reactor) decreases, the role of chain fracture on the surface of the pipeline increases, and the methane conversion rate decreases significantly. The methane conversion rate depends on the packing type and diameter of the reactor.

Three simplified models of detailed reaction mechanism of methane were proposed by Gosiewski [[10]], and three reduction mechanisms were verified through comparative experiments between free space and regenerative packing channels. As shown in Fig.3, (a) was parallel reaction mechanism, (b) was consecutive reaction mechanism, and (c) was parallel-consecutive reaction mechanism. Gosiewski concluded that (a) and (c) would produce a kinetic equation with uncertainty form ($\infty \times 0$), and (b) would be more applicable in the numerical simulation of the RFR.

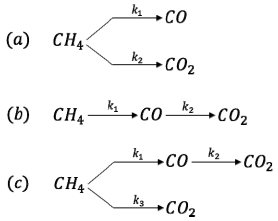


Fig.3 Three reduction reaction mechanisms of methane

And Gosiewski.et al [[11]] believed that packing size, surface type and temperature of the combustion zone would have an effect on reaction mechanism and kinetic characteristics, for bed surface type and specific surface area contacting with gas phase corresponds to unique radical activation intensities. The temperature-dependent reaction would be carried out according to different reaction mechanisms, the larger the specific surface area, the lower the ignition temperature, and the smaller the amount of CO generated in the flue gas. A calculation method of methane reaction rate during wide temperatures range was provided, based on kinetic parameters of the consecutive mechanism in the low temperatures zone and high temperatures zone.

Wang [[12]] studied the oxidation reaction mechanism of VAM, got a simplified two-step reaction mechanism based on the experiment data of VAM (simulated by a mixture of air and methane) homogeneous combustion in empty reactor and homogeneous packed reactor, and determined kinetic parameters of VAM homogeneous combustion reaction in the reactor. His study found that CO_2 concentration reaches local maximum, then decreases gradually, and finally shows an upward trend as the temperature rises. Experiments data showed that at low temperatures, methane may also be directly oxidized to CO_2, which reacts with methane and is reduced to CO at high temperatures. At high temperatures, CO_2 is mainly produced by CO. The existence of packing can reduce ignition temperature and promote methane combustion. The larger the contact area between packing and gas phase, the lower the ignition temperature. Increasing specific surface area or reducing pore diameter can promote combustion reaction, while an extremely small diameter may lead to complete combustion extinction.

3.2 Heat wave shift discipline of thermal RFR

Using single temperature approximation method, Dobrego [[13]] analyzed the seepage combustion wave of dilute methane-air mixture in semi-closed pipeline, built the analytical solution for the preheating zone, reaction zone and combustion product zone, and established the temperature of three different regions, the distribution regulation of methane component, the longitudinal extension of reaction zone, the ignition temperature of mixture and the moving velocity of system combustion wave.

In addition, combined methods of theoretical analysis and numerical simulation, Dobrego studied dilute methane-air mixture combustion in the lean combustibility limit (LCL) [[14]], by changing the parameters (heat loss coefficient, length of the reactor, pressure, particle size, porosity), established the corresponding curve of LCL, and obtained gas concentration rate function in LCL limit conditions. Numerical results showed that the particle size of porous media is the most important factor for LCL control, and higher pressure provides better concentration limit (LCL), which can be used to stabilize reactor operation.

From Gosiewski's opinion, compared to the method of laying out heat exchanger in the center of RFR, extracting flue gas from the RFR center is more likely to get the temperature distribution of symmetry, which would be also easy to appear in the cooled jacketed RFR structure, and the improvement control strategy to solve the thermal asymmetry was mentioned [[15]].

3.3 Theoretical analysis of thermal RFR

The study of theoretical analysis and analytical solution tries to obtain the solution result of the system mathematical model through no complex numerical simulation, and then guides industrial design or system performance optimization.

C.W.M. van der Geld [[16]] put forward an analytical method and two different numerical calculation methods for the one-dimensional mathematical model of RFR, and compared these three different solving methods with the experimental data, and regarded the analytic method as an easier and more advantageous way for dealing with practical problems.

Balaj [[17]] made use of dimensional analysis, gave mathematical derivation of highly nonlinear equations (conservation of energy and mass in flow phase and solid phase) in describing RFR, and obtained dimensionless numbers used for first order dimensionless analytic expression representing RFR operation characteristics, with the purpose of RFR design optimization. He also paid attention to sensitivity of parameters to the RFR, such as bed length, switch time, and so on.

Kaisare [[18]] carried out model validation and scale analysis of RFR, found that the optimal reversing time was close to the natural time scale of reaction heat release, and consider that the time scale analysis would provide guidance rules for the design and operation of RFR.

Through dimensionless analysis, derivation and reduction of the mathematical model of RFR, Haynes [[19]] used the correlation formula of dimensionless number and reactor parameters to guide the industrial design of RFR, and modified the calculation of RFR design.

3.4 Modeling solution and verification

In terms of mathematical model research, the polish academy of sciences [[20]] established two different versions of the one-dimensional model of the thermal RFR (heat storage on the inner wall and without heat storage on the inner wall), and verified the model through the experimental data on the large-scale RFR. It is concluded that the heat accumulation on the wall does not directly affect the temperature distribution of the packing in the reactor.

Deng et al. [[21],[22]] established a single-channel homogeneous reaction model of the thermal RFR, studied the effects of different flow rates and concentrations on the operating characteristics of the thermal RFR through numerical simulation, and analyzed the effects of different concentrations of methane exotherm on the preheating process. Based on the thermal RFR model, Deng et al. proposed a method to calculate the minimum stable running methane concentration [[23]]. Through the model, the calculation method of the ventilation range and the length of the RFR was raised.

Morrone et al. [[24]] built a one-dimensional unsteady calculation model for an RFR with particles and a fluidized bed RFR, and verified the model, which is applicable to actual production conditions. The process thermal efficiency and gas pressure drop were calculated according to the system geometry and operating parameters. Experimental data show good consistency with the model. The simulation results showed that although the gas-solid contact efficiency is high in fluidized bed, the thermal performance of fluidized bed RFR is poor due to its circulation characteristics.

Mario Amelio et al. [[25]] put forward a one-dimensional unsteady state model of RFR and a numerical calculation program for solution, which could be effectively applied to the analysis and design of the RFR system and could be used to select more appropriate structure types, such as structural type or packing bed (even considering various particle shapes). The energy efficiency of the random RFR is compared with that of the structure RFR. From the perspective of energy utilization, the performance of the random RFR is poor, and it should be used if the cost is low and the economic requirements are met.

3.5 Inert regenerative packing characteristics

Wang et al. [[26],[27]] put in use FLUENT software to study thermal structure characteristics, operation parameters and resistance of RFR. He considered that the temperature field in the RFR bed is basically in trapezoidal distribution, and the high temperature area moves periodically along RFR bed height. At the same time, he realized that the width of the high-temperature area of the RFR bed layer widens with the increase of methane concentration, and narrows with the extension of half cycle of switching period. The increase of wall heat loss will also decrease the reaction velocity, narrow the high temperature area, lower the temperature in the high temperature area and lower the methane conversion rate. The study of the resistance characteristics showed that, with the periodic change of

flow direction, the resistance of RFR presents the periodic change in the form of "V" wave, and the resistance of RFR decreases continuously with the increase of time in the first half cycle. Entering into the second half cycle, the resistance of RFR starts to rise, and at the end of the second half cycle returns to the resistance value at the beginning of the cycle. The pressure loss of RFR magnifies linearly with the increase of the volume of air, enlarges with the augment of methane concentration in the gas, and decreases with the rise of the porosity of honeycomb ceramics.

Lan et al. [[28]] conducted a numerical study on the thermodynamic characteristics of honeycomb ceramic channel using FLUENT software, and investigated the influence of mass flow rate, solid heat capacity and switch period on the temperature efficiency. The results showed that the temperature efficiency decreases linearly and the pressure drop increases linearly with each half cycle. With the increase of the switch period, the temperature efficiency decreases slowly in the heating process, while it is not affected in the cooling period of the RFR.

Liu et al. gave research on thermal fatigue properties of mullite ceramics commonly used in the thermal RFR [[29]], and predicted the working life of the reactor bed according to the intensity attenuation law and thermal fatigue test data. The fatigue test results showed that the bending strength of mullite ceramics generally decreases with the increase of thermal shock. The bending strength decreases faster at higher temperature differences. In 600~800 ℃ temperature difference, the bending strength began to decline, and then, after reaching a certain value, it remained the same, then fell again. The calculated results based on strength attenuation theory and thermal fatigue experimental data showed that the thermal fatigue life of the thermal RFR is about 1-8 months.

The effects of different mathematical models on thermodynamic process parameters, as well as the thermal asymmetry phenomenon of thermal RFR were analyzed by Gosiewski et al. [[30]], and then the technical conception of enhancing heat transfer such as increasing the hydraulic diameter of the pore channel and increasing the flow rate was put forwarded. Moreover, the mechanism and kinetics of the homogeneous reaction of methane were further studied, and the results were compared with those of Wang et al. [[12]].

3.6 Large-scale experimental studies

VOCSIDIZER^{TM}, A special kind of thermal RFR, the first system of which was tested on methane at the Thoresby coal mine in the UK in 1994. The results of experiments showed that VOCSIDIZER^{TM} had good stability when methane concentration fluctuated. MEGTEC ran its first commercial case of VOCSIDIZER^{TM} in 2001 at Applin coal mine in Australia, recycling heat and producing steam for 12 months. In 2007, the West-camp project of VOCSIDIZER^{TM} started operation in Australia, ran in the methane concentration of 0.9%v/v through mixing coal mine methane (CMM) with the volume flow of 70m3/s, actuated the steam turbine to generate electricity with the goal of 6MWe. MEGTEC also provided a VOCSIDIZER^{TM} system for China's Zheng Coal Group, which ran with a methane concentration of 0.3-0.7% and a flow rate of 18m3/s, with oxidizing heat used to produce hot water.

A series of experimental studies were carried out on a full-size (1.6m in diameter and 2.85m in height) columnar RFR (packing type is rectangular saddle ring) by C.W.M et al. [[31]], and the prediction results of one-dimensional mathematical model were compared and validated using the test results.

Liu et al. [[32]-[34]] analyzed the influence of operation parameters, such as methane concentration, flow rate, switching period and reaction region's temperature profile on the oxidation reaction, studied a mathematical model of flow resistance of RFR, and discussed flow uniformity in the RFR. Based on the porous media homogeneous model, control equations of the heat storage and oxidation process were established, and the influence of operating parameters of the RFR and the ceramic structure parameters on the flow resistance and outlet temperature of the RFR was studied by numerical simulation. It was concluded that the pressure loss and the outlet temperature both increase with the rise of the apparent flow velocity of intake gas, the pressure loss decreases significantly with the increase of the equivalent diameter, and the stable operation of the RFR increase slightly with the augment of the specific heat capacity.

Liu et al. [[35]] conducted a series of experiments on the dilute methane thermal RFR of 60000m^3/h, and studied its start-up performance, methane conversion rate, the minimum self-thermal equilibrium methane concentration, the temperature distribution profile in the reactor and the superheated steam productive rate. It was obtained that the lowest self-thermal balance concentration was 0.28%. In addition, when the methane concentration is 1.0%, the highest temperature in the RFR is 1066 ℃, and the methane conversion rate was 98.6%.

Lin [[6]] discussed the RFR operating characteristics in Dafosi coal mine, including 27 hours of preheating process (at the center of the bed temperature heated to 1000 ℃) and 9 hours of bed temperature distribution adjustment process (adjustment of bed temperature state to similar distribution of stable operation). The heat energy recovery rate for power generation in the stable operation period accounts for 31.61 ~ 46.82% of the methane reaction heat, the minimum methane concentration in self-maintaining equilibrium is less than 0.25%, and the system is recommended to operate at 1% of the concentration.

4. Conclusions

Through the discussion of research status and trend of dilute methane thermal RFR technology, it can be known that the dilute methane thermal RFR technology can completely destroy CMM and realize near-"zero" emission of CMM. The promotion and application of this technology have great ecological benefits, social benefits and certain economic benefits.

Many researches have been made in the understanding and analysis of the thermal RFR phenomenon, but the basic theory research of RFR is still very weak, such as the reaction mechanism and kinetics in the RFR homogeneous inert channels that used in the numerical calculation, and the rationality of the numerical computation of control equation, which would make it difficult to get accurate simulation results for guiding the reactor process design in engineering application.

Further theory researches, such as inert regenerative thermal shock resistance, thermal fatigue life of the packing material, different equivalent ratio methane reaction kinetics mechanism, the asymmetric temperature gradient produced by reverse flow of thermal wave, and the potential harmonic response of the RFR, will help to optimize the mathematical model of the RFR, improve the efficiency of industrial RFR design, and enhance the technical economy of the industry applications, to better serve the transformation and upgrading of green mining of coal mining enterprises strategy.

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