DEVELOPMENT OF CONSTRUCTION OF A CATALYTIC REACTOR
FOR METHANE OXIDISING IN VENTILATION AIR IN COAL MINES
AND THE RESEARCH ON INTEGRATED “HEAT PIPE” RECUPERATOR

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ABSTRACT

The article presents the analysis whose result is the selection of appropriate design and construction of a monolithic CMR reactor intended for oxidising methane from ventilation air in coal mines. The description of “heat-pipe” recuperator cooperating with the reactor was also presented. The research was mainly aimed at verifying the compliance with the work of autothermity premise obtained by the return of part of heat from catalytic reaction. The result of research was to define the range volumetric fume expense ensuring autothermity and the definition of maximum recuperator efficiency. The range of volumetric expense was 18–25 m³/h and maximum value of efficiency coefficient was \( \eta = 0.50 \) for the volumetric expense of 18 m³/h.

Key words: “heat pipe” recuperator, catalytic reactor.

INTRODUCTION

One of anthropogenic sources of methane emission to atmosphere is the ventilation air from coal mines, containing 0.1–1% of methane. It is estimated that c.a. 15 Mtons of \( \text{CH}_4 \) is released to atmosphere, what makes c.a. 70% of methane emitted from coal mines [7]. Apart from ecological aspects, the stream of methane is the source of energy that has not been used so far. Annual emission of methane in ventilation air is c.a. 600 mln Nm³, and its calorific value reaches 2,1\cdot10^8 \text{ GJ}, what constitutes the economic value of 1.2 bln PLN [4].

Due to relatively low concentration of methane in ventilation air the oxidation of homogeneous air is impossible. The process can be conducted exclusively in catalytic reactors working in autothermic conditions. The technical and technological problems related mainly to highly reactive and stable catalysts, autothermic reactors of specialised construction and cooperating effective heat exchangers ensuring the capture, return and distribution of heat energy in the system require further research and development of the issue [5]. The works were undertaken by the Consortium of Methane Utilisation from Coal Seams, established by AGH University of science and Technology, Wrocław University of Technology and Maria Curie-Skłodowska University. One of the consortium tasks was to construct an experimental large-size installation for catalytic oxidation of methane from ventilation air in coal mines and the tests on both the catalysts and the whole installation. The key issue for the quality of the planned research tasks, aimed at magnifying the scale, was to select the type and form of catalytic reactor construction and heat exchanger.

The work presents the analysis of technique which is the basis for the selection of catalytic reactor type and the description of research on recuperator using “heat pipe” technique in modern version cooperating with the catalytic reactor. The research was primarily conducted for compliance with the work of autothermity premise obtained by the return of the part of heat from the catalytic reaction.
CHOOSING THE CONSTRUCTION OF CATALYTIC REACTION

In order to oxidise the methane included in ventilation air from coal mines three types of reactors were used:
- TFFR (thermal flow reversal reactor),
- CFRR (catalytic flow reversal reactor),
- CMR (Catalytic monolith reaktor).

Figure 1 and Figure 2 presents the characteristics of monolith and reversal reactors.

The analysis of the presented courses of temperatures and the data available in thematic literature [1, 2, 7] showed that:
- the process realised in CFRR reactors is fully controllable; the profile of temperature is a stationary course which is easier for modelling;
- fitting the CFRR reactor with opening and closing dampers generates disturbances in flows and mixing masses of fuel and fumes, what leads to disturbances in heat and mass exchange, manifested in the course of output temperature,
- minimum CH₄ in ventilation air ensuring stable reactor work for CFRR and CMR reactors is 0.1% and 0.4% respectively,
- CMR reactor has a simple, compact construction,
- the control of process in CMR is easier,
- CMR reactor must be equipped with additional source of heat in case of concentration under 0.4%,
- the costs of construction and exploitation of CMR reactor with the throughput of 34 m³/s is by 12% and 5% lower than the costs of construction and exploitation of CFRR reactor.

![Diagram 1](image1.png)

**Fig. 1.** The schemes of construction and typical temperature courses of monolith reactors (CMR):
- classical and reverse flow; Tᵯ – input temperature, Tᵰ – initiation temperature,
  - T_max – maximum temperature, T₁ – output temperature, ΔT_ad = T₁ – T₀ [2]

![Diagram 2](image2.png)

**Fig. 2.** The scheme and profile of temperature in catalytic flow reversal reactor CFRR, marks as in Figure 1 [2]
The main criterion for the selection of experimental catalytic reactor to realise the basic research tasks, especially the evaluation of the proposed catalysts, was the capability to obtain parameter stability and the elimination of disruption and interference of heat and mass streams. Moreover, according to the author, due to the research on heat effects of methane oxidation processes and exploitation reasons, it is more reasonable to couple the heat exchanger with the reactor in the external manner.

Consequently, was chosen a monolithic CMR reactor which was integrated with external heat exchangers. It was designed and constructed.

**CHARACTERISTICS OF HEAT EXCHANGER CONSTRUCTION**

The construction premises of heat exchangers cooperating with CMR reactor were imposed by the specification of the work of catalytic components. They include the temperature of initiation of the catalytic reaction, which is 320 °C for the used palladium catalysts, and high heat exchange efficiency conditioning the autothermity of reactor work.

Selecting the construction of capture and return of heat to the reactor, the author used the recuperator with a modern “heat pipe” technique, as the most efficient solution. Figure 3 presents the construction of a single section of the “heat pipe” exchanger.

“Heat pipe” exchanger consists of the case with collars at its ends, divided with a horizontal adiabatic panel dividing it into two canals. In the canals there are hexagonal rows of vertical heat pipes fitted with radiators. In the lower canal there is the medium releasing heat and in the upper canal there is the medium absorbing heat. The transportation of heat is realised inside the heat pipe, where the medium mediating in the exchange evaporates in the lower part and condenses in the upper part (condensation), what intensifies the heat exchange process.

Considering the compliance with the postulate of maximum heat reception from the fumes, three-section construction of heat exchanger was adopted with the following temperature ranges: section I: 300–750 °C, section II: 120–300 °C, section III: 20–150 °C. Each section was equipped with heat pipes located in hexagonal layout with 1.5 D gap and an adiabatic septum, which ensures the sealing of the sample pressure equal to 1,5×10⁵ Pa.

**TEST BENCH AND THE CHARACTERISTICS OF THE OBTAINED RESULTS**

The aim of the research was to define the volumetric flow of agents in which the amount of the returned heat to the catalytic residue should ensure the autothermity of reactor’s work. Moreover, it was necessary to define the characteristics of recuperator efficiency and the amount of usable heat in the assumed range of fume’s flow.

The constructed exchanger was subjected to tests in the test bench developed for this purpose (Fig. 4).

The test bench is composed of the systems of hot air preparation consisting of a ventilator (1), air flow meter (2), humidifier (3), heater (4), three-section heat exchanger (5) and reception system, i.e. ventilator (6), air flow meter (7) and computer control system for data collection and analysis (8). The test bench was fitted with temperature (t), humidity (W), flow (Prz) and pressure (p) sensors. The recuperator was thermally insulated.

The test bench allows the tests of heat recuperation in the flow and counter flow conditions for the following ranges of parameters:

- **volumetric agent expense**: 5–30 Nm³/h,
- **sensor temperature**:
  - releasing heat: 100–700 °C,
  - absorbing heat: 10–250 °C,
- **relative humidity of heat releasing agent** – up to 95%.

Sensory system ensured the measurement, recording and reading of the parameters in real time.
The properties of recuperator define its efficiency and usable heat stream. Recuperator efficiency is defined for both the releasing and absorbing agents as the ratio of the amount of absorbed or liberated heat, to the amount of maximum, theoretic capacity (for the surface of exchange approaching infinity) [6]:

\[
\eta_o = \frac{Q^r_o / Q^r_{o, \text{max}}}{(T^0_{we} - T^0_{wy}) / (T^0_{we} - T^0_{we})}, \\
\eta_p = \frac{Q^r_p / Q^r_{p, \text{max}}}{(T^0_{wy} - T^0_{we}) / (T^0_{we} - T^0_{we})},
\]

where: \(\eta_o\) – recuperator efficiency on the side of heat releasing agent (evaporator efficiency), \(\eta_p\) – recuperator efficiency on the side of heat absorbing agent (condenser efficiency), \(Q^r_o\) – current heat flux of the heat releasing agent, \(Q^r_{o, \text{max}}\) – maximum heat flux of the heat releasing agent, \(Q^r_p\) – current heat flux of the heat absorbing agent, \(Q^r_{p, \text{max}}\) – maximum heat flux of the heat absorbing agent, \(T^0_{we}\) – initial temperature of the heat releasing agent, \(T^0_{we}\) – output temperature of the heat releasing agent, \(T^0_{wy}\) – initial temperature of the heat absorbing agent, \(T^0_{wy}\) – output temperature of the heat absorbing agent.

It was assumed that the total heat absorbed by the recuperator will be returned to the catalytic residue. Whereas the heat surplus, with relation to the conditions of the surrounding in the heat releasing agent released from the recuperator, what constitutes the usable heat, which is then received in a separate heat pipe exchanger in fumes-water system. The flux of usable heat calculated from the relation:

\[
Q^u = m' \cdot \Delta I [J/s]
\]

where: \(Q^u\) – the flux of usable heat [W]; \(m'\) – the flux of heat releasing agent mass [kg/s]; \(\Delta I\) – the difference in enthalpy between the flux of releasing agent and surrounding [J/kg].

The tests were conducted in the temperature range of heat releasing agent between 520–600 °C, what corresponds to the range of fume temperatures in the output of catalytic residue and the volumetric consumption of agents ranging 10–30 m³/h. The tests were conducted in five series of parameter measurements for different volumetric values of the parameter and different volumetric expenses but the same amounts of releasing and absorbing agents in the conditions of heat balance obtained after c.a. 3 hours of test bench work.

Figure 5 presents the relation between initial temperature of heat absorbing agent and the obtained usable power \(Q^u\) to the volumetric expense of the agents.

In the examined range of expenses the temperature of heat absorbing agent oscillated in the range 275–340 °C, whereas for the flows from 18 m³/h to 25 m³/h the temperature exceeded the value of 320 °C. This is a minimum temperature initiating the work of catalytic residue. In the above range of expenses the obtained usable power was from 330 W to 360 W with its maximum for the expense of 25 m³/h.

Figure 6 presents the characteristics of efficiency of evaporator and condenser of the examined recuperator.

In the examined range of agents’ volumetric expenses the efficiency for heat release was 88–94% and for heat absorption – 50–58%. Maximum total efficiency of the heat exchanger was 50% for the expense of 18 m³/h.

**SUMMARY**

1. As a result of the analyses experimental CMR reactor was chosen, constructed and integrated with external heat exchangers. The basic crite-
The relation between initial temperature of heat absorbing agent $T_{wy}$ and the obtained usable power $Q_{uz}$ to the volumetric expense of the agents.

The characteristics of efficiency of evaporator and condenser.

Criteria of selection were: the possibility to obtain stable work parameters, elimination of heat and mass interferences that is characteristic for CMR reactors.

2. The exploitation tests showed that the construction solution of the exchanger with a hexagonal layout of heat pipes with 1.5D gap and the division of condensation zones, zone scaling methods and the use of radiators meets the requirements of functionality.

3. The conducted tests showed that the temperature of 320 °C in the exchanger output, which ensures autothermity of reactor work is obtained for the expenses (the flux of fumes and heated reagents’ mixture) from 18 m$^3$/h to 25 m$^3$/h. In the above range of expenses the obtained usable power was from 330 W to 360 W with its maximum for the expense of 25 m$^3$/h.

4. Maximum total efficiency of the heat exchanger was 50% for the expense of 18 m$^3$/h for the range of agents’ volumetric expenses, the efficiency for heat release was 88–94% and for heat absorption – 50–58%.

REFERENCES