

# Experimental investigations of the influence of radial gas mixing in an inert ceramic foam bed on thermal combustion of lean methane–air mixtures

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## Abstract

The paper discusses the application possibilities of ceramic foam in a thermal combustion process of a lean methane–air mixture. The experiments were done in a ceramic foam bed. The foam (Vukopor<sup>®</sup> A) was made mainly of Al<sub>2</sub>O<sub>3</sub>. The foam samples were packed in a tubular reactor symmetrically placed in a laboratory furnace. It was assumed that the tested foam should have a surface close to the monolith surface area which was tested in a previous work (Pawlaczyk and Gosiewski, 2015). Pore density of the tested foam was 10 PPI. The tested air mixture contained 0.51–0.76 vol. % of methane. The results show that thermal methane oxidation in foam is possible in the acceptable range of temperatures. The combustion process in foam is characterized by similar ignition temperature to tests carried out in monolith, a more intense course, and better methane conversion at lower temperatures.

## Keywords

methane combustion, VAM, ceramic foam, thermal combustion, kinetic

## 1. INTRODUCTION

A lean methane–air mixture is produced by mining industry, wherever coal is extracted from methane mines. For many years the mixture, known as VAM (Ventilation Air Methane), was treated as a production waste. Nowadays, it is the unfriendly pollution to environment which should be destroyed. The issue of VAM utilization became physically tangible because of the announced inclusion of methane in the EU ETS system. However, it is difficult to use VAM as a fuel in industrial practice because of a very low methane concentration (often below 0.4 vol.%), a huge flux, a lot of pollution as dust (Baris, 2013; Su et al., 2008), a high humidity and other gases that might contain sulfur (Setiawan et al., 2014). The most advanced utilization technology, dedicated to VAM mitigation is the combustion in thermal reverse flow reactors (Pawlaczyk-Kurek and Suwak, 2021). Those reactors are filled by ceramic honeycomb monoliths. Due to a large number of parallel channels the monoliths are characterized by low flow resistance which is essential, especially when the reactor is in the final sections of the technological line, where the pressure is not too high. The biggest disadvantage of the monolith structure is the impossibility of mixing reactants in the reactor cross-section, which for an industrial scale reactor may contribute to uneven spreading of the stream in the bed (Gosiewski and Pawlaczyk-Kurek, 2019). In a high-temperature process the phenomenon directly reflects on the distribution of temperature in the cross-section, forming space without the reaction, and without heat generation. In such a space the accumulation of heat is also limited. The goal of the work is to show the impact of the implementation of ceramic foam in the reactor on the thermal

combustion process. Open solid foams are widely discussed in the literature. They are usually used in catalytic processes as a support of the catalyst (Cerri et al., 2000; Ciambelli et al., 2010; Maestri et al., 2005; Palma et al., 2013; Patcas et al., 2007; Twigg and Richardson, 2007). The foam structure ensures good reactant mixing with enhanced radial heat and mass transfer which is significant in highly endo- and exothermic reactions (Liu and Chen, 2014). The structure allows the medium to flow through the bed in all directions, which in high temperature processes also translates into a uniform temperature distribution in a cross section of the reactor and increased use of bed volume for storing the heat released during the reaction. In such a situation, the use of foam bed would reduce the size of the reactor and investment expenditure. Although, in comparison to the monoliths, the pressure drop in foams can be slightly higher (Gancarczyk et al., 2018; Patcas et al., 2007; Tsinoglou et al., 2009), they still would be an interesting alternative. The performed basic tests characterize the combustion process of lean methane–air mixtures, provide information about the conditions in which this process occurs, and allow us to determine the possibility of using ceramic foam in a non-catalytic high-temperature process. The collected data are discussed and compared with the results of similar experiments obtained for monolith packing.

## 2. EXPERIMENTAL SETUP

The experiments of thermal combustion of lean methane–air mixtures were done using the experimental setup described in (Pawlaczyk and Gosiewski, 2015). The difference is that another bed was previously studied, a monolith honeycomb,



in place of the foam currently being investigated. A tubular reactor filled with foam samples was symmetrically placed in a three-zone laboratory furnace with separate temperature control for each zone. The flowing gas mixture was heated to combustion temperatures. Due to the high temperatures in the reaction environment, the foam Vukopor® A was used. According to the manufacturer's data sheet, the foam consists of  $\text{Al}_2\text{O}_3$  and  $\text{SiO}_2$ . The detailed test of composition showed that  $\text{Al}_2\text{O}_3$  is the main component.

The crucial morphological foam parameters as specific surface area and porosity were determined with computer microtomography (micro-CT, SkyScan 1172). The obtained images were processed with the global thresholding method, using iMorph software. The method allowed to avoid errors in computing the total surface by eliminating closed surfaces inside the foam structure, not accessible to reagents. The method and used procedures are described in (Gancarczyk et al., 2019; Leszczyński et al., 2016).

The decision about choosing the right size of foams was made based on the size of the specific surface area of tested samples. It was assumed that this value in the case of ceramic foams should be similar to the monolith (870  $\text{m}^2/\text{m}^3$ ), tested previously and described in (Pawlaczyk and Gosiewski, 2015). Because the mentioned monolith was previously marked as monolith B (MB), the same designation was kept here. The pore density of investigated foam sample was 10 PPI, with a specific surface area of 860  $\text{m}^2/\text{m}^3$ .

The combustion experiments were carried out with stable flow of the gas mixture of 800  $\text{dm}^3/\text{h}$  through the foam bed. The methane concentrations were within a range of 0.51–0.76 vol.%. The basic parameters of the process and the tested foam bed as compared to the monolith B are presented in Table 1.

The change in methane conversion was obtained by changing the temperature set. The temperature in the combustion zone was measured with thermocouples, enabling the assign-

Table 1. Parameters of the combustion process and beds: monolith and set of parameters for different beds: monolith and foam.

Quantity	Unit	Kind of bed	
		Monolith B	Foam Vukopor® A -10 PPI
Ignition temperature	°C	675	680
Average temp. in combustion zone	°C	670–790	670–770
Methane concentration	vol.%	0.44–0.97	0.51–0.76
Flow	$\text{dm}^3/\text{h}$	800	800
Porosity	%	66	78
Specific surface area	$\text{m}^2/\text{m}^3$	870	860
Diameter	m	0.65	0.65
Length of the reactor	m	0.7	0.7

ment of a temperature profile along the bed (along the combustion zone). The composition of the gas mixture at the inlet and outlet of the reactor was measured with precision gas analyzers (IR).

The experimental setup was designed so that temperature in the combustion zone would be approximately constant. From the results based on the developed method of determination (Pawlaczyk and Gosiewski, 2013) the real volume of the combustion zone in structural bed and the average temperature in this zone, the kinetic parameters for the assumed reaction mechanisms and the form of kinetic equation were determined by constructing Arrhenius plots.

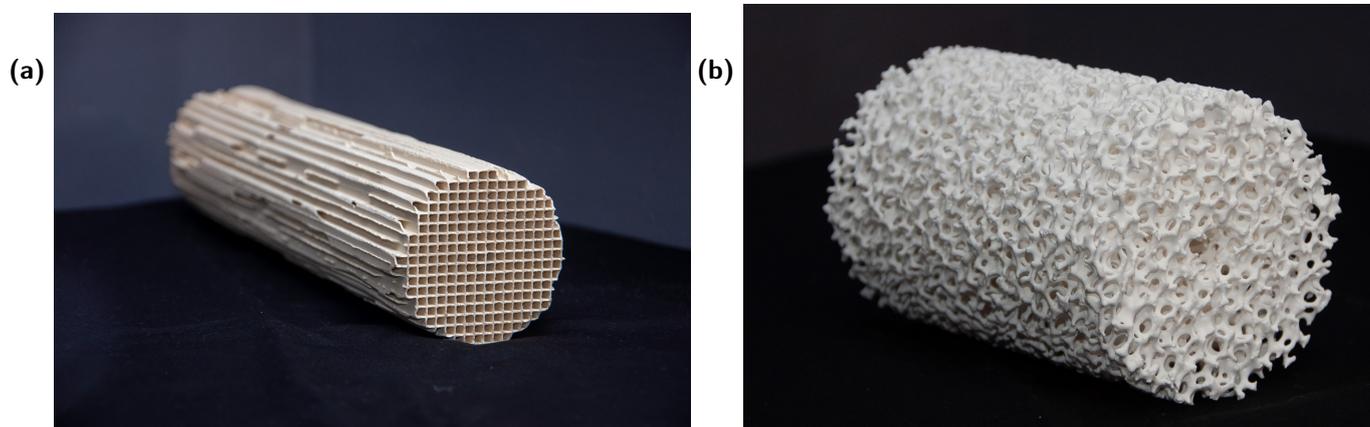


Figure 1. The comparison of reactor bed: a) monolith B tested in (Pawlaczyk and Gosiewski, 2015), b) foam 10 PPI.

### 3. ANALYSIS OF RESEARCH RESULTS

The experiments showed a dependence of the composition of the post-reaction mixture on the temperature in the reaction zone. Similar to other studies (Pawlaczyk and Gosiewski, 2015), the presence of CH<sub>4</sub>, CO, and CO<sub>2</sub> was observed in the stream flowing out of the reactor. The experimentally determined ignition temperature of diluted methane in the air in foam bed (680 °C) was close to the value obtained in the case of the monolith B (675 °C). It was noticed that in certain temperature ranges, methane conversion and the shape of the measured temperature profile was dependent on the concentration of the combustible component, which was not observed in the case of monolith tests. This dependence became visible when the average temperature in the combustion zone was higher than 720 °C and the total conversion of methane reached a value of about 30%. It was also noticeable that when methane conversion exceeded 70% with the average temperature in the reaction zone higher than 750 °C, the conversion was changeable in time. Then the achievement of stable conditions in the reactor was impossible. This is most likely due to the continuous increase in temperature in the reaction zone due to the release of reaction heat. The amount of this heat is large enough for the continuous increase in temperature in the reaction zone to be observed, which in turn translates into an increase in conversion. Moreover, a favourable effect of foam geometry on methane conversion was found, as shown in Fig. 2, which shows the dependence of methane conversion on the average temperature in the combustion zone for the foam bed and the monolith B. In the case of the monolith bed, methane conversion and the length of the combustion zone was prac-

tically independent of inlet methane concentration, thus the data presented in Figs. 2, 3 and 4 is the average value for sets of CH<sub>4</sub> concentration for the given temperature. Consequently, inlet methane concentration was not given in these figures.

The comparison of total methane conversion for foam and monolith shows that the difference between those values is lower at the low temperature and rises with the rising temperature in the combustion zone. In the case of the monolith the value is half of the conversion value obtained for the foam at 720 °C. Whereas at 760 °C the conversion for a reaction carried out in the foam is almost 3 times higher than that for the monolith bed.

Methane is combusted totally in foam beds at lower temperatures than in monolith beds. Complete methane conversion occurs at 770 °C for the tested foam. During the experiments the conditions for the formation of carbon monoxide and the complete utilization of methane were also determined. It can be seen in Fig. 2.

The analysis of the results revealed that it is possible to distinguish three temperature ranges in which the methane combustion reaction has a different course. In range I, the reaction proceeds mainly to carbon monoxide. The amount of CO in the outlet stream rises with the temperature rise. CO formation is more intensive in the ceramic foam than in the case of monolithic packing. There was no effect of the amount of combustible component in the mixture for methane conversion on individual products of the reaction. In range II, a stabilization is observed in relation to the conversion of methane to carbon monoxide, which is about 28% in the temperature range of 746 °C to 760 °C for foam. The combustion in foam

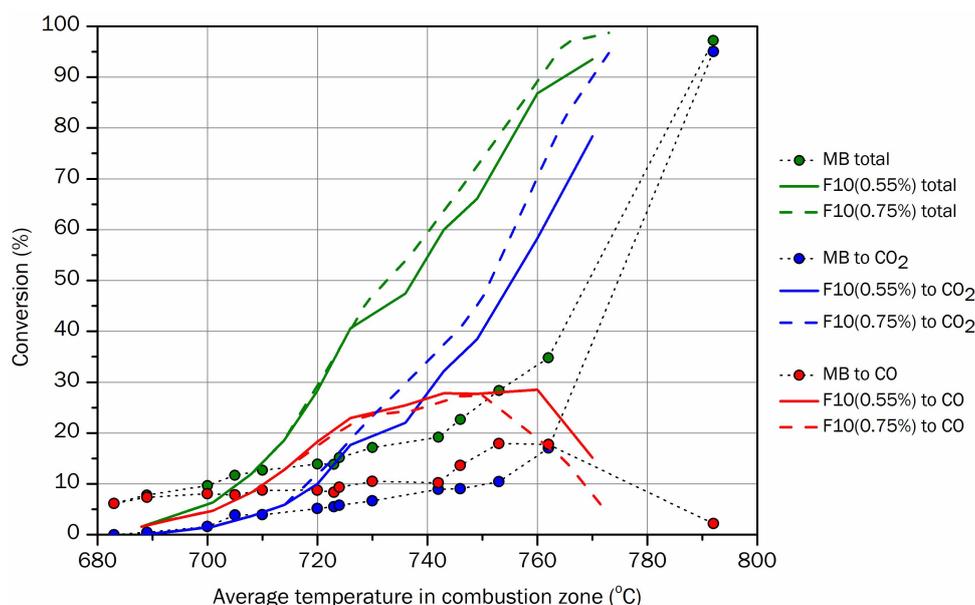


Figure 2. Dependence of methane conversion (total – green colour, to CO – red colour, to CO<sub>2</sub> – navy blue colour) on the average temperature in the combustion zone for MB and foam at the inlet CH<sub>4</sub> concentrations of 0.55% vol. – F10 (0.55%), and 0.75% vol. – F10 (0.75%).

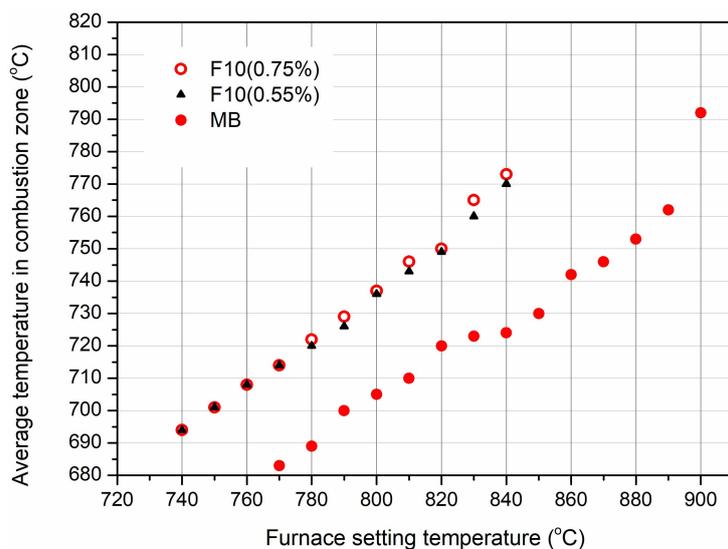


Figure 3. Dependence of the average temperature in the combustion zone on the furnace setting value for MB fillings and 10 PPI foam.

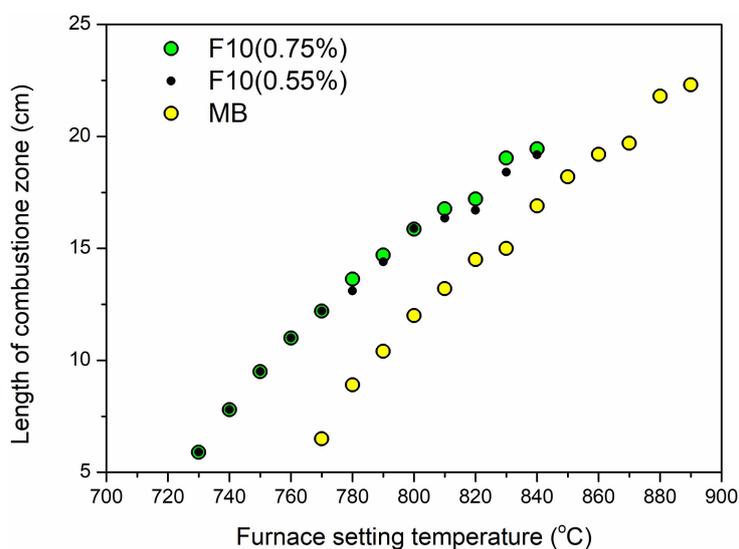


Figure 4. Comparison of the length of the combustion zone vs. the furnace setting temperature for foam and monolith.

promotes the formation of a higher amount of CO than in the case of the monolith. At the same time, an increase in the concentration of carbon dioxide in products is recorded with increasing temperature and the influence of methane concentration in the inlet mixture on the value of methane conversion. It is visible in Figure 2 that above 725 °C in the combustion zone, the mixtures with higher methane content allowed to achieve a little higher conversion of methane to CO<sub>2</sub> than comparable conversions to CO. In range III a problem with getting stable conditions in the reactor was revealed. The situation was observed when the temperature in the reaction zone was about 760 °C and higher. It was probably caused by the amount of heat released in the methane combustion reaction, which was large enough to cause a continuous increase of temperature in the reactor. It affected the composition of the post-reaction mixture directly. The

higher temperature, the higher the methane conversion was observed until the reaction was completed.

The investigations confirmed that the structure characteristic of the foam provides the conditions for good mixing of the mixture components, which influence the temperatures measured in the bed axis, and these in turn the average temperatures in the reaction zone and the length of the combustion zone. As shown in Fig. 3, the mean temperature differences can reach 30 °C for the lower furnace settings and about 45 °C for the higher ones.

Higher temperatures in the foam bed than in the monolithic bed confirm the fact that the use of the former may be advantageous and improve the uniformity of temperature distribution in the cross-section of the reactor. The results from the experiments carried out for the same temperature set point

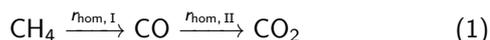
showed that the length of the combustion zone was longer for the foam bed than for the monolith bed. The differences in zone lengths are shown in Fig. 4.

While comparing data (see Table 2) for similar average temperatures in the combustion zones in the foam and the monolith it may be seen that the length of the combustion zone is shorter in the foam and the total conversion of methane determined in the combustion zone is much higher.

Table 2. Comparison of the length of the combustion zone and the total conversion of methane for selected similar average temperatures in the combustion zone.

Average temp. in combustion zone [°C]		Furnace setting temperature [°C]		The length of the combustion zone [cm]		Total conversion [%]	
MB	F10	MB	F10	MB	F10	MB	F10
689	694	780	740	9	7.8	7.8	3.8
700	701	800	750	12	9.5	11.7	6.3
720	720	820	780	14.5	13.1	13.9	28.3
730	729	850	790	18.0	14.7	17.1	45.8
746	746	870	810	19.8	16.8	22.7	68
762	760	890	830	22.4	18.4	34.8	86.8

The data collected during experiments allowed also to determine the kinetic parameters for assumed consecutive mechanisms in the tested reaction system (see Table 3), as in Eq. (1):



The method of determining the parameters of the kinetic equations was based on the method previously developed and described in (Pawlaczyk and Gosiewski, 2013). Due to the much higher concentration of oxygen than that of methane, the relative change in oxygen concentration can be negligible and the kinetic equation can be given by Eq. (2):

$$r_{\text{hom},j} = -\varepsilon \frac{dC_i}{dt} = k_{0,j} \exp\left(\frac{-E_j}{RT}\right) \cdot (C_i^{3\nu})^{a_j} \quad (2)$$

where:  $i = C_{\text{CH}_4}$  for reaction (I) or  $i = C_{\text{CO}}$  for reaction (II).

Table 3. Kinetic parameters for the follow-up mechanisms in the tested reaction systems.

No.	Reaction	$E_j$ [J/mol]	$k_{0,j}$ [mol <sup>(1-a)</sup> / (m <sup>3(1-a)</sup> s)]	$a_j$ [-]
I	$\text{CH}_4 \xrightarrow{r_{\text{hom,I}}} \text{CO}$	266 067	$6.08 \times 10^{12}$	0.1
II	$\text{CO} \xrightarrow{r_{\text{hom,II}}} \text{CO}_2$	251 637	$9.28 \times 10^{11}$	0.3

The calculated value of the reaction rate was determined in relation to the volume of the foam bed. Estimation of the kinetic data was done with the assumption of the isothermal conditions in the combustion zone.

Based on the measured temperature profiles for each set point of methane concentrations and temperature, the volume and length of the combustion zone, and the average temperature in the zone were calculated. This temperature was then used to determine subsequent points on the Arrhenius plots. The combustion zone was this part of the foam bed where the temperature was higher than the ignition temperature. The ignition temperature was taken as the highest measured temperature for which the first products of the reaction were observed.

## 4. CONCLUSIONS

The thermal combustion of lean-methane air mixtures can be possible in a reactor with the ceramic foam bed. The results were compared with the results of an experiment in the monolith bed with a similar size of surface area. The following conclusions were drawn:

- the specific structure of foam ensures a better mixture of reagents in the reactor and caused better heat transfer across and along the reactor;
- the ignition of the reaction in the foam bed starts at a similar temperature (680 °C) as in the monolith bed (675 °C);
- the foam bed allows intensification of the combustion process which is visible in the data obtained for similar average temperatures for the monolith and foam beds; in the case of foam the reaction occurs in shorter combustion zone with higher conversion;
- the total combustion of methane occurs in the foam bed at a lower temperature (average temperature in combustion zone of about 770 °C) than in the monolith (793 °C).

The obtained results will be the basis for further research on the application of ceramic foams in technologies for thermal utilization of VAM. The next step will include testing thermal properties of the ceramic foam and determining adsorption capacity of substrates and products of the reactions. The relevant issue will be measurements of gas flow resistance in a wide range of temperatures, especially in high temperatures. It is planned to develop a mathematical model of a reactor filled with a ceramic foam bed. The collected data will be used to verify the model.

## ACKNOWLEDGEMENTS

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## SYMBOLS

$a_j$	exponents at methane concentration in the kinetic equation
$C$	concentration, mol/m <sup>3</sup>
$E_j$	activation energy, J/mol
$k_{0,j}$	pre-exponential factor in kinetic equation, mol <sup>(1-a)</sup> /(m <sup>3(1-a)</sup> s)
MB	monolith B – bed of the reactor, tested in (Pawlaczyk and Gosiewski, 2015)
F10	foam – bed of the reactor, pore density: 10 PPI
$r_{hom,j}$	reaction rate, mol/(m <sup>3</sup> s)
$R$	gas constant, J/(mol K)
$t$	time, s
$T$	temperature, K
<i>Greek symbols</i>	
$\varepsilon$	void fraction, –
<i>Superscripts</i>	
av	average value
<i>Subscripts</i>	
$i$	$i$ -th component
$j$	$j$ -th reaction (I or II)

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