Chemical Engineering Journal 238 (2014) 78–85

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Catalytic or thermal reversed flow combustion of coal mine ventilation air methane: What is better choice and when?

Krzysztof Gosiewski *, Anna Pawlaczyk

Institute of Chemical Engineering, Polish Academy of Sciences, Gliwice, Poland

HIGHLIGHTS

- Methane concentration range is divided into low (below 0.4%) and high (above 0.4%).
- Catalytic reactor is competitive to thermal in low concentrations while in high is not suitable.
- In low concentrations heat recovery is unprofitable – only greenhouse gas mitigation is effective.
- Heat recovery is attractive in high CH_4 concentrations achieving environmental and energy effect.

GRAPHICAL ABSTRACT

ARTICLE INFO

Article history:
Available online 20 July 2013

Keywords:
Reversed flow reactors
Catalytic combustion
Thermal combustion
Methane
VAM

ABSTRACT

The paper presents a comparison of the two options of reverse flow reactors destined for the utilization of coal mine ventilation air methane by catalytic (CFRR) or thermal (TFRR) combustion. It has been shown that both solutions have advantages and drawbacks. The use of the catalyst significantly decreases reactor temperature and makes the operation becomes to be autothermal for methane concentrations lower than in TFRR (even as low as over 0.06 vol.%). On the other hand when methane is combusted, particularly if average concentration is higher than ca. 0.4 vol.% the maximum temperature in the reactor appears to be too high for available cheap catalysts, while the use of the noble metals as active components (e.g. Pd) is not economically viable. Moreover lifetime of the catalysts is much lower than of the inactive heat exchange packing. For TFRR autothermicity threshold is higher (ca. 0.2 vol.%) but it enables cost-effective heat recovery if CH_4 concentration is higher than approx. 0.4 vol.%. In conclusion, the paper states that for lower VAM concentrations, when only greenhouse gas mitigation is an aim of combustion CFRR can have some advantages over TFRR. Should the heat recovery be seriously taken into account the TFRR is economically and technically the most advantageous solution, however.

1. Introduction

Methane released during coal mine operation is not only greenhouse gas but also a valuable energy carrier. There are three streams of gas containing coal mine methane: gas drained from a seam before mining (60–95 vol.% CH_4), gas drained from work areas of the mine (30–95 vol.% CH_4) and carried with mine ventilation air (0.1–1.0 vol.% CH_4) [1], the so-called Ventilation Air Methane (VAM). In Poland due to safety regulations CH_4 concentration of VAM is lower and usually does not exceed 0.7 vol.%. In other countries e.g. in the US [1] it can reach or even slightly exceed 1 vol.%. In Poland nearly 600 millions m^3/year of CH_4 acc. to [2] are released to the atmosphere, while worldwide VAM emission in 2009 was estimated at 28.7 billion m^3. As the concentration of VAM is not very high, utilization of this source via the direct conventional combustion processes is only possible with supplemental fuel. However, such a lean mixture can be an alternative fuel for energy production if modern combustion technologies, i.e. Flow Reversal Reactors (FRRs): either catalytic – CFRRs or

* Corresponding author. Tel.: +48 32 2310811.
E-mail address: k.gosiewski@iich.gliwice.pl (K. Gosiewski).

1385-8947/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.cej.2013.07.039

Nomenclature

Q_{rec}	flux of heat recovered, MW_t
Q_{gen}	flux of heat generated by methane combustion, MW_t
T^{ign}	combustion ignition temperature
T^{max}	maximum temperature in the reactor, °C
ΔT_{ad}	adiabatic temperature increase of the reaction, °C
z	axial coordinate, m

Acronyms

CFRR	catalytic flow reversal reactor
CFRR(MnO_2)	catalytic flow reversal reactor with manganese catalyst 12% MnO_2/γ-Al_2O_3
CFRR(Pd)	catalytic flow reversal reactor with palladium catalyst 0.5% Pd/γ-Al_2O_3
CMR	catalytic monolith combustor
CSS	cyclic steady state
FRR	flow reversal reactor
GWP	global warming potential
HC	high VAM concentrations (above 0.4 vol.%)
LC	low VAM concentrations (below 0.4 vol.%)
TFRR	thermal flow reversal reactor
TFRR 10000	TFRR with flow throughput 10,000 m^3_{STP}/h
RCO	regenerative catalytic oxidizer
RTO	regenerative thermal oxidizer
STP	standard temperature and pressure: 273.15 K (0 °C), 100 kPa
VAM	ventilation air methane
VOC	volatile organic compounds

Note: Units of power abbreviations

MW_t	thermal power
MW_e	electrical power

thermal – TFRRs are applied. These methods have specific advantages but also drawbacks. Su et al. [3] present and compare various methods of VAM utilization and divide them into two basic categories: *ancillary uses* and *principal uses*. For the *ancillary uses*, ventilation air is used tosubstitute ambient air in combustion processes, including gas turbines, internal combustion engines and coal-fired power stations. For the *principal uses*, methane in ventilation air is a primary fuel. Methods of *ancillary uses* seem to be the easiest and the cheapest, but they are hardly feasible in practice because of large flow rates of ventilation air, accounting for at least hundreds of thousands of cubic meters of air per hour. It is difficult to find or to build in vicinity of a ventilation shaft a power station requiring such large amount of air for combustion. A power plant ready to consume such huge volumes of air must have been rated for several hundreds MW.

As the *principal uses* in [3] the following are discussed:

- Catalytic Flow Reverse Reactor (CFRR),
- Thermal Flow Reverse Reactor (TFRR),
- Catalytic Monolith Combustor (CMR),
- Catalytic lean burn gas turbine,
- Recuperative gas turbine,
- Methane concentrator, etc.

Nowadays, however, only CFRR and TFRR are seriously taken into account for industrial usage, so only these two are discussed in this paper.

The interesting discussion of the two types as applied to the volatile organic compounds (VOC) combustion was given by Matros et al. in [4] (FRRs were named regenerative oxidizers, either catalytic – RCO or thermal – RTO). The problem of VAM mitigation could be slightly different as in some cases large amount of heat can be recovered, which in VOC combustion seldom takes place. Thus economy and technical requirements for the heat recovery equipment should also be taken into account. Concentration of VAM varies significantly and depends on particular coal mine. There are many mines of average concentration of ca. 0.3 vol.% or even lower [1]. The objective of the VAM utilization, is either environmental mitigation of this strongly greenhouse gas (GHG) emission or energy recovery, or both. It will be shown in the paper how these two goals affectthe choice of the combustion method and technical solutions applied.

2. FRRs for greenhouse gas mitigation, for heat recovery or for both goals?

Methane as the GHG has a global warming potential (GWP) at least over 20 times higher than CO_2. Therefore combustion even without the heat recovery could be ecologically or economically attractive. Matros and Bunimovich in [5] claim that CFRR (RCO) requires adiabatic temperature increase ΔT_{ad} > ca. 15 °C to be autothermal. The similar threshold for TFRR (RTO) is 50–90 °C acc. to [5] or 45–70 °C acc. to [4]. Estimated adiabatic temperature increase for methane combustion is 265 °C per 1 CH_4 vol.%. It means that VAM combustion in the CFRR should be autothermal for CH_4 - concentrations above ~0.06 vol.% while TFRR requires at least ~0.19 vol.%, which generally agrees with the conclusions given in [3]. Thus advantage of CFRR over TFRR for very low concentrations seems to be obvious. On the other hand when significant heat recovery is taken into account, i.e. for concentrations above 0.4 vol.%, maximum temperature in CFRR seems to be too high for any cheaper catalyst cf. [6]. For concentrations reaching 1 vol.% even if Pd catalyst is used its temperature will be close to the permissible limit.

Maximum temperature, at least by 300 °C higher in TFRR than in CFRR, can be considered as disadvantage, due to possible too high NO_x emission [4,7] and more expensive construction material. Otherwise, as experiments carried out at TFRR research and demonstration plant [8] revealed a maximum temperature actually did not exceed 1100 °C therefore these drawbacks occurred to be insignificant.

The analysis of influence of the reactor temperature on the amount of the heat recovered (cf. [9]) has shown that the higher the temperature the higher is heat recovery. No matter that, the analysis in [9] was conducted for CFRRs only, the general finding that higher reactor temperature favors heat recovery is also valid for TFRRs. In the next sections of this paper a comparison of CFRR and TFRR with the respect of VAM mitigation purposes is given. The two aspects are taken into account:

- greenhouse gas mitigation,
- heat recovery efficiency.

For these purposes the scope of VAM concentrations was divided into two ranges: