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Research Paper

Combustion of coal mine ventilation air methane in a regenerative combustor with integrated adsorption: Reactor design and optimization

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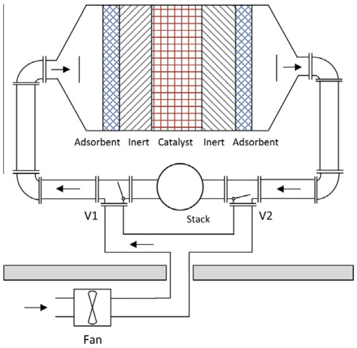
HIGHLIGHTS

- Coal mine ventilation air methane catalytically oxidised in reverse flow reactor.
- Water adsorption before the catalytic bed prevents catalyst inhibition.
- Model-based reverse flow reactor design.
- Simulations with a real coal mine ventilation air predicts stable operation.
- Economical optimization of reactor design.

GRAPHICAL ABSTRACT



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ABSTRACT

Coal mine ventilation air methane is an important environmental concern due to its contribution to global warming. Catalytic combustion in reverse flow reactors is an efficient treatment technique, but high emission moistures lead to catalyst inhibition. To overcome this issue a novel reverse flow reactor with integrated water adsorption has been proposed.

In this work, the design of a reverse flow reactor adequate to treat a typical real coal ventilation stream, 45 m^3/s with 0.30% (mol) methane and 5% (mol) water, has been studied. The performance of the reactor design has been simulated using a 1D heterogeneous dynamic model, previously validated with experimental results. Particular attention has been paid to reactor stability when water and methane feed concentration change upon time. Real coal mine ventilation air data have been used to produce realistic simulations. The optimization of the operating conditions (surface velocity and switching time) has been carried out based on the total cost of the reactor (considering fixed capital and 10-year variable cost).

1. Introduction

In the last decades, environmental issues related to global warming have gained importance. Methane is considered a major contributor to global warming (second in importance, after carbon dioxide), with global atmospheric concentrations raising from a pre-industrial value of about 0.7 up to 1.8 ppm in 2014 [1]. The main anthropogenic sources of methane are coal mines, landfills, natural gas distribution systems, sewage systems, etc. [2]. It is well known that the global warming potential of methane is 25 times greater than that of carbon dioxide, and for this reason the combustion of methane to carbon dioxide before releasing to the atmosphere is environmentally beneficial, as it reduces greatly the net warming potential of the emissions [3].

Coal mining has an important negative impact in relation to greenhouse emissions, because important amounts of methane (concentration 0.1–1%) are released to the atmosphere during coal

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Nomenclature

a	surface to bed volume ratio (m^2/m^3_{bed})
c_i	gas molar concentration (mol/m^3)
C_P	heat capacity (J/kg k)
D_{im}	molecular diffusion coefficient (m^2/s)
D_{ax}	axial dispersion coefficient (m^2/s)
D_{ep}	effective pore diffusion coefficient (m^2/s)
D_h	hydraulic diameter of the monolith (m)
d_P	particle diameter (m)
f_w	washcoating fraction (%)
h	gas to solid heat transfer coefficient (W/m^2 K)
k	thermal conductivity (W/m K)
K_{eqi}	water adsorption equilibrium constant (mol/kg_{ads}) $(m^3/mol)^{0.5}$
K_{inh}	water inhibition constant (1/Pa)
L_w	washcoating size (m)
K_C	gas–solid mass transfer coefficient (m/s)
k_w	kinetic constant (mol/kg_{cat} s Pa)
L_R	reactor length (m)
m	Freundlich exponent
n_i	concentration in the solid (mol/kg)
Nu	Nusselt number ($Nu = hD_h/k_G$)
p	Pressure (Pa)
Pr	Prandtl number ($Pr = C_{PG}\mu_G/k_G$)
Re	Reynolds number ($Re = D_h v \rho_G/\mu_G$)
r_i	reaction rate (mol/kg s)
R	ideal gas constant (8.314 J/mol K)
Sc	Schmidt number ($Sc = \mu_G/(\rho_G/D_{im})$)
Sh	Sherwood number ($Sh = K_C D_h/D_{im}$)
t	time (s)
T	temperature (K)
T_{ph}	pre-heating temperature (K)
t_{sw}	switching time (s)
v	gas (interstitial) velocity (m/s)
y	mole fraction (–)
z	spatial coordinate (m)

Greek symbols

ϵ_b	bed porosity (–)
ΔH_i	reaction enthalpy (−802.5 kJ/mol)
ϕ	Thiele modulus (–)
κ_{Gax}	axial dispersion coefficient for the gas phase (W/m K)
η	internal effectiveness factor (–)
ρ	density (kg/m^3)

Subscripts

0	inlet
cat	catalyst
G	gas
S	solid

extraction [4]. Thus, ventilation air methane represents the main contribution (approximately 78%) to the carbon footprint of the coal mining activity [5].

Catalytic combustion is a very interesting alternative for the treatment of these emissions [6], because of its low energy consumption (especially if compared with thermal oxidation) and the negligible formation of noxious by-products, such as thermal NOx [7]. An appropriate option for the treatment of coal mine ventilation air methane is the use of regenerative catalytic oxidation (RCO) in a reverse flow reactor [8].

Reverse flow reactors (RFR) consist of a catalytic fixed bed reactor in which the feed flow direction is periodically reversed [9]. By selecting the appropriate switching time (t_{sw}), defined as the time elapsed between two consecutive flow reversals, a great amount of the heat released in the reaction can be stored inside the reactor in consecutive cycles, and therefore autothermal operation (e.g. without the addition of external heat or fuel) can be possible even for very low concentration of hydrocarbons (1500 ppm methane) [10]. For higher hydrocarbon concentration, the energy released by the reaction can be also recovered [11].

The RFR potential advantages are a consequence of its forced unsteady state operation [12]. However, this can also be a drawback from the stability point of view, i.e. maintaining ignited operation under disturbances in flow rate or concentration. For example, if the feed is too lean, there is a risk of extinction, because the amount of heat released by the reaction is very low; in these situations the RFR heat regeneration capacity is crucial to maintain autothermal operation [13]. Under rich feed conditions, the heat released and accumulated in the reactor can result in too high bed temperatures, with the risk of causing catalyst thermal deactivation [14]. This issue has limited the industrial use of this type of reactors, and encouraged research in the development of suitable control systems [15].

The performance of the catalyst can be affected by the presence of other compounds, different from methane, in the ventilation air [16]. Water is usually present in high amounts (nearly at the saturation point), and it is well-known that water has a negative effect on the activity of supported precious metal catalysts [17]. Palladium catalysts on various supports, the most active for methane oxidation, have been studied by different authors with a general agreement in the existence of a reversible inhibitory effect of water [18]. This inhibition reduces the relative stability of the reverse flow reactor, and to overcome this, the reactor must be oversized, with the corresponding increase in purchase and operation costs.

One solution to avoid the decrease in catalyst activity caused by water is the use of a reverse flow reactor with integrated adsorption. Such a device, proposed by Bos et al. [19], consists of a reverse flow reactor in which the inert beds are replaced, at least partially, by an appropriate adsorbent material [20]. Thus, the water of the feed is retained in these adsorbent beds before reaching the central catalytic bed [21]. The adsorbent is regenerated in situ, because of the parabolic temperature profile that develops in reverse flow reactors, with high temperature in the middle and low temperature at the sides. The temperature profile is not static, but moves in the direction of the flow, and produces a displacement of heat from the centre to the sides of the reactor. Thus, while feed water is being adsorbed at low temperature in the first adsorbent bed (in the corresponding direction of the flow), the water adsorbed in the previous cycle desorbs at a higher temperature from the adsorbent bed placed in the opposite side of the reactor.

The working principle of the reverse flow reactor with integrated adsorption was experimentally demonstrated in a previous work for methane oxidation in a bench-scale device [22]. In the same work, a mathematical model to simulate the behaviour of this reactor was proposed. This model accurately predicted the experimental results.

The aim of the present work is to apply all this knowledge on reverse flow reactors with integrated adsorption to the design and optimization of commercial scale devices for the treatment of methane emissions from coal mines. The scope of the work is