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Autothermal reverse-flow reactors: Design and comparison of valve-operated and rotary systems

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HIGHLIGHTS

- Valve-operated and rotary reverse-flow reactors are simulated and compared.
- Five different monolithic structures are considered for performance comparison.
- A design strategy is proposed considering a range of VOC content to be treated.
- The rotary scheme avoids the emission of unreacted VOCs after flow reversal.
- The rotary scheme results substantially more compact than the conventional.

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ABSTRACT

The valve-operated reverse-flow catalytic reactor is an efficient system for the treatment of air streams contaminated with small amounts of volatile organic compounds (VOCs). Nonetheless, it has two drawbacks when operated at low cycle periods: the emission of the volume without treatment present in the region close to the reactor input and an unavoidable reduction of the valve lifetime. Upon consideration that the use of shorter cycle periods would enable the design of more compact units, we investigated the option of operating with reverse flow by means of a rotary catalytic reactor. This alternative eliminates the necessity of valves for the flow reversal and enables to allot a small fraction of the total cross section to be fed with clean air and prevent the VOCs that remain close to the reactor input from being discharged to the atmosphere.

The aim of this work is to analyse the treatment of an air stream contaminated with ethanol and ethyl acetate by means of mathematical simulation of reverse-flow operations in both valve-operated and rotary catalytic reactors. To that end, monolithic structures with square channels are assumed for both types of reverse-flow reactors, a design strategy is proposed and the results for both types of reactors are compared. It is concluded that the rotary reverse-flow reactor arises as a better option.

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1. Introduction

As the result of increasingly demanding environmental regulations, an interest in technology for the control of air contamination has grown considerably in recent decades. Volatile organic compounds (VOCs) are among the most common air contaminants, with catalytic combustion being the usual technology for their elimination when the levels of VOCs are low – *i.e.*, $\leq 1\%$ – (Kolaczkowski, 2006).

The industrial air streams to be purified are usually at near room temperature and, in practice, the flow rates of the air to be processed are quite high. Furthermore, depending on the nature of the substances to be treated and the catalyst used, the temperature necessary for the catalytic combustion ranges between 200 and 400 °C. The necessity therefore arises to utilize combustion schemes that enable an efficient recovery of the energy of the processed gases to raise the temperature of the input air stream.

Among the possible schemes to realize the energy recovery, the reverse-flow system has been widely studied (*e.g.*, Chen et al., 2011; Marín et al., 2010; Matros and Bunimovich, 1996). The system consists in the operation of a catalytic bed (with either a structured or a granular packing) with periodically inversion of the flow direction of the air stream. By this means, the operation

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remains transient though with periodically repeated cycles so as to maintain the central zone of the bed at the temperature required for the VOCs combustion, while the ends operate as preheating sectors (Matros and Bunimovich, 1996). This type of operation becomes particularly attractive when the adiabatic temperature rise is low, a condition that occurs in the treatment of VOCs owing to their low concentration.

In practice, the reversal in the direction of the flow is achieved by a valve (or a set of valves) in the conventional scheme. Though this means is the simplest, the use of valves has two disadvantages when the cycle period is short: first, each time the valves change the direction of the flow, they emit the small fraction of the untreated VOCs that remain in the entrance of the unit into the atmosphere; second, the lifetime of a valve is proportional to the cycle period. Both of these features set a lower limit on the cycle period. The use of short cycle periods, however, would enable the design of more compact units. Indeed, the shorter the cycle period, the higher will be the average temperature of the bed. Thus, the operation with flow reversal by means of a rotary scheme in which the bed turns on an axis, while the input air enters and the product is extracted through the use of segmented fixed heads, arises as an interesting option. This alternative eliminates the necessity of valves for the flow reversal and allows avoiding the emission of VOCs close to the reactor inputs, as will be explained in Section 2.1.

The rotary scheme has been postulated by Kolios et al. (2000), who indicated: "An elegant possibility of operating a regenerative fixed-bed reactor in a continuous mode with integrated purge steps without using valves is to use a Ljungström-type rotating-regenerator design". Such a scheme has been proposed for the process of Chemical Looping Combustion (CLC). In this system, the alternative of employing a monolithic-type structure (Lecomte et al., 2010) or a packed bed of particles has been suggested (Noorman et al., 2010). A model of this type of system has been presented by Zhao et al. (2013). To the best of our knowledge in the literature to date no report exists dealing with a modelling and design procedure of a rotary scheme for the treatment of VOCs in order to determine in a concrete manner the advantages of that scheme over the conventional reverse-flow reactor (CRFR).

Within this context, the objective proposed in this contribution is to apply a model of the rotary reverse-flow reactor (RRFR) to the treatment of VOCs to compare the performance of such scheme against the CRFR. For both systems, we propose a design procedure taking into account the fluctuations that could occur in the input air stream. The elimination of a mixture of ethanol and ethyl acetate in an air stream is employed as a case study. A monolithic structure with square channels is assumed for both types of reverse-flow reactors. Five configurations of the monolith cell have been considered in order to select the most suitable one for each type of reactor.

2. Case-study

Ethyl acetate (EA) and ethanol (Et), employed as solvents in printing processes, are typical VOCs released by the manufacture of packaging. The catalytic incineration over Mn and Cu oxides (Morales et al., 2008) of an air stream containing those compounds has been chosen as the case-study. Table 1 presents the specific characteristics of the stream to be treated. In terms of the maximum adiabatic temperature rise, ΔT_{ad}, the minimum and nominal VOC contents shown in Table 1 correspond to 26.6 and 53.2 °C, respectively. The need to consider a minimum content of VOCs will be discussed in Section 5.

The reaction system has been studied by Campesi et al. (2012b). They proved that ethanol combustion goes through the production of acetaldehyde. The reactions and kinetic expressions involved – identified by indices 1, 2, and 3 for ethyl acetate, ethanol, and acetaldehyde, respectively – are:

Table 1
Characteristics of the VOC-laden air stream.

Mass flow rate [kg s^{-1}]	Pressure [bar]	Temperature [°C]	Minimum composition [ppm V]		Nominal composition [ppm V]	
12.7	1.06	50	EA 307	Et 125	EA 615	Et 250

Table 2
Kinetic parameters.

Preexponential factor [s^{-1}]		Activation energy [J mol^{-1}]		Reference temperature [K] and adsorption constant [m^3 mol^{-1}]	
$k_{1,ref}$	6.62×10^1	E_1	1.48×10^5	T_{ref}	448
$k_{2,ref}$	1.81×10^3	E_2	1.10×10^5	K_1	4.19×10^1
$k_{3,ref}$	1.22×10^1	E_3	1.69×10^5	K_3	6.75×10^1

$$C_4H_8O_2 + 5O_2 \rightarrow 4CO_2 + 4H_2O$$

$$C_2H_6O + 1/2O_2 \rightarrow C_2H_4O + H_2O$$

$$C_2H_4O + 5/2O_2 \rightarrow 2CO_2 + 2H_2O$$

$$r_i = \frac{k_i C_i}{1 + K_1 C_1 + K_3 C_3} \qquad \text{for} \quad i = 1..3 \tag{1}$$

$$k_i = k_{i,ref} \exp\left[-\frac{E_i}{R}\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right] \qquad \text{for} \quad i = 1..3 \tag{2}$$

Table 2 summarizes the kinetic parameters (Campesi et al., 2012b), which correspond to intrinsic values, as intra-particles diffusional resistances were accounted for in the analysis of the experimental data.

The catalytic reactors are monolithic structures comprising an inert matrix coated with the catalyst. A 10 μm thick uniform catalytic coating, δ_{cat}, was assumed. For the sizing of the units and their comparison, a set of 5 types of square cells, whose characteristics are shown in Table 3, were considered. The dimensions in Table 3 include the catalytic coating. The first three of them correspond to commercial standards of Corning Incorporated (Boger et al., 2004) with 200, 400, and 600 cells per square inch and wall thicknesses of 12.5, 7.5, and 4.0 thousandths of an inch, respectively. Configurations 4 and 5 are alternative designs to numbers 2 and 3, respectively, with the wall thickness having been modified in such a way that the void fraction is the same as that of configuration number 1. The properties of solid structure are evaluated as those of nonporous cordierite (Gulati, 2006).

Table 3
Cell configurations and relevant geometrical properties.

Nº	$10^{-5}N_{cell}$ [m^{-2}]	$10^4\delta_T$ [m]	ε	a_v [m^{-1}]	$10^3 d_h$ [m]
1	3.1	3.18	0.678	1833	1.48
2	6.2	1.91	0.722	2677	1.08
3	9.3	1.02	0.814	3480	0.94
4	6.2	2.25	0.678	2593	1.05
5	9.3	1.83	0.678	3176	0.85

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