Chemical Engineering Science 134 (2015) 423–432

Contents lists available at ScienceDirect

Chemical Engineering Science

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

An efficient computational scheme for building operating maps for a flow reversal reactor

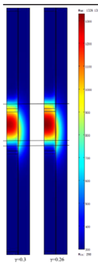
Z. Jia [a], R.E. Hayes [b,*]

[a] State Key Laboratory of Chemical Resources, Beijing 100029, China
[b] Department of Chemical and Materials Engineering, University of Alberta, Edmonton, AB, Canada T6G 2G6

HIGHLIGHTS

- A detailed model of a full scale CFRR is developed for lean methane combustion.
- A simple empirical model is developed to predict maximum and average reactor temperatures.
- An acceptable operating zone is defined in terms of these temperatures.
- Stability maps for acceptable operation are generated.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 3 February 2015
Received in revised form
7 May 2015
Accepted 8 May 2015
Available online 22 May 2015

Keywords:
CFRR
Catalytic combustion
Stability map
Simulation
Process

ABSTRACT

This paper describes the use of a two dimensional heterogeneous computational model for the catalytic reverse flow reactor for the combustion of lean methane mixtures. A uniform design method is used to generate a table of 41 operating points. The results from these points were used to generate a simple empirical model for the reactor at a single specified value of the superficial gas velocity. Stability maps that show the envelope for operation that satisfy a minimum level of conversion and a maximum operating temperature are generated using the empirical model. These maps delineate all possible combinations of methane concentration, switch time and mass extraction rate that are capable of giving stable operating conditions that meet these two criteria. The model is potentially useful in model predictive control of the reactor.

1. Introduction

Methane emissions from a variety of sources contribute strongly to the total greenhouse gases emitted worldwide. Methane is widely considered to be the second most important greenhouse gas, after carbon dioxide, with a global warming potential about 23 times that of carbon dioxide. Large amounts of methane are discharged into the atmosphere with the ventilation air from underground coal mines. A single ventilation shaft can emit over 500,000 m^3 (STP)/h of ventilation air in which the methane content can be as high as 1 vol%. The abatement of this emission source is therefore important from both economic and environmental points of view, as the gaseous fuel is inadvertently lost, simultaneously contributing to the greenhouse effect and wasting a source of potentially useful energy (Gosiewski et al., 2008).

The most convenient method to eliminate the methane emissions is by combustion, which reduces the global warming potential by about 88% and also may recover useful energy. The low methane content of the mine ventilation air places it outside of the flammability limits for standard homogeneous combustion, but it may be burned

---

* Corresponding author.
E-mail address: bob.hayes@ualberta.ca (R.E. Hayes).

http://dx.doi.org/10.1016/j.ces.2015.05.023

using catalytic combustion (Hayes and Kolaczkowski, 1997). The key requirement for a good catalytic combustion solution is that of achieving auto-thermal reactor operation (Hayes, 2004). Although this operating mode can often be achieved using feed pre-heat, sometimes this approach is not desirable or is not possible.

One method for achieving auto-thermal reactor operation for lean methane feed is the use of reverse flow operation (RFO). RFO has been used for many years (Cottrell, 1938; Gilbert and Daniels, 1948; Frank-Kamenetski, 1955). When used for reactors, the catalytic flow reversal reactor (CFRR) consists of a fixed catalytic bed through which the feed flow directions are periodically reversed, which causes heat to be trapped within the rector, thus increasing its temperature beyond the adiabatic temperature rise. Inert sections are usually included on either side of the catalyst bed to increase the heat trapping potential. See Matros and Bunimovich (1996) for a review of the CFRR. The catalytic combustion of methane in a CFRR has been investigated by many workers for the reduction of fugitive emissions from different sources (Liu et al., 2000a, 2000b, 2001a, 2001b, 2007; Eigenberger and Nieken 1988; Cittadini et al., 2001).

Over the past three decades, many experimental and numerical studies based on relatively small-diameter CFRR have been made (Salomons 2003; Salomons et al., 2003, 2004; Fissore et al., 2005). These systems can be classed as lab or small pilot scale. The drawback of these smaller diameter systems is that the higher ratio of external reactor surface to volume, which is characteristic of small reactors (low diameters), causes the heat losses to be relatively more important with regard to the overall thermal balance of the system (Li and Jia 2011). It is particularly difficult to obtain the near adiabatic conditions that can be approached in an industrial scale apparatus. This poses a number of problems in the scale-down of industrial size reactors, and may produce unreliable results in pilot-scale demonstration units. As a consequence, results obtained in a lab-scale reactor cannot be used to design an industrial apparatus with any degree of accuracy (Fissore et al., 2005; Hevia et al., 2006). Fissore et al. (2005) designed an experimental reactor with a special temperature-control system based on a dynamic compensation of the thermal losses to reproduce as closely as possible the behavior of large-scale industrial reactors. However, it is important to realize that the heat transferred to the reactor through the wall may change the radial temperature distribution in the reactor, and the results from this test-rig should be carefully considered. This approach is also somewhat complicated experimentally. Therefore, a numerical simulation on a reactor of a larger diameter is still desirable to study the reactor performance and to determine its operating window, and that study was one purpose of this work.

Computer aided design offers many advantages for the development of industrial scale processes. There has been much progress in recent years in this area. Models of increasing complexity have been developed as the power of the hardware and the sophistication of the software increase. In spite of these improvements, the use of detailed computer models may still require significant amounts of time. For full size reactors, the number of mesh points that must be used can be considerable, especially if two or three dimensional solutions are required, which is generally the case for the reverse flow reactor. For the CFRR, the problem is exacerbated by the need to run many cycles after startup before the stationary state is attained, thus finding the limit points of extinction and reactor over heating can take time. Cittadini et al. (2001) discussed some of the drawbacks of using a detailed model for the simulation of the CFRR, and presented in that work a simplified model. They also give a good review of some of the other simplified models that have been proposed. A compromise solution is to use a detailed model to generate sufficient information to establish a simpler model that is totally empirically in nature, but yet captures the desired operating features and can be used to evaluate the limit points.

Another important issue that relates to the desire for a computationally simple, yet accurate, model is the need to control the reactor during its operation. There are several works that deal with the control of a CFRR, with different methods proposed. For example, the switch time was adjusted to deal with the fluctuation of the inlet concentration in a lab-scale reactor (Hevia et al., 2005; Gao and Jia, 2014). It is not clear if this method would be effective for a large-scale reactor or a rich methane concentration. Marín et al. (2010) used the temperature in the middle point of the reactor to control the flow reversal and to overcome disturbances in the feed concentration for some conditions. However, the temperature in the middle of the reactor is not necessarily, or indeed even usually, the maximum temperature and thereby not a typical point for a controller. Others have used the rate of mass extraction to control the performance (Balaji et al., 2007; Fuxman et al., 2007, 2008; Balaji and Lakshminarayanan, 2005). Implementation of an appropriate control strategy requires a complete understanding of the operating window plus a predictive model that is computationally efficient.

The generation of some simpler criteria for the design and operation of a CFRR is a desirable objective. In previous work in this area, Cittadini et al. (2001) used a simplified model to study the limiting values of the length of the catalyst section and switch time for auto-thermal operation, and presented the results in terms of stability maps. Hevia et al. (2005) also used stability maps of the reactor to delimit regions where the maximum temperature (or conversion) is higher or lower than a given value. The full map is a function of three parameters: the switch time, the inlet concentration and the fluid velocity. The stability map is a useful representation that has been used and extended in this paper.

In this paper, we show how a detailed model can be used to generate data which can in turn be used to develop a simple empirical model that captures all of the data necessary for the description of the reactor. The model is used to develop complete stability maps that delineate the operating window over a wide range if inlet methane concentrations. Stability maps are generated for a large pilot scale unit, which is quite different in behaviour to the lab scale reactors often reported. Because of the difficulty in measuring conversion in an operating unit, we show the appropriate relationships between temperature and conversion. It is shown that for a given velocity, the temperature can be used to predict the conversion accurately. Such a model should have application in the robust control of the CFRR.

The objectives of this work were: (1) to develop a numerical model for a relatively large diameter CFRR and use it to study the operating conditions; (2) to develop a simple, yet accurate empirical model, and (3) to use the simple model to determine the acceptable operating regions for the reactor. It is our intent to demonstrate a complete workflow that can be used for other, similar systems, albeit with different operating limits.

2. Description of the CFRR used in the investigation

As noted in the introduction, there is significant literature on the CFRR for small small-diameter reactors, but the conclusions from these small reactors are difficult to apply in industrial applications directly. Therefore, a larger diameter CFRR was selected for this study. A schematic of the reactor concept is shown in Fig. 1. The central catalyst section was a packed bed of Rashigs rings 1.2 m in length, whilst inert monoliths 3 m long were used on each side. Mass extraction was done from either end of the catalyst section, depending on the flow direction. The dimensions of the reactor are summarized in Table 1. Values for the physical properties are based on the small pilot scale reactor reported by (Salomons et al., 2004), with the catalyst of (Aubé and Sapoundjiev, 2000).