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HEAT EXTRACTION FROM A FLOW REVERSAL REACTOR IN LEAN METHANE COMBUSTION

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This paper describes the use of a flow reversal reactor for the destruction of lean emissions of methane with ambient temperature feed. The reactor was comprised of two packed bed reactors and inert sections to act as heat traps. Inert sections were one of ceramic monolith, metal monolith or packed beds of ceramic spheres. In this paper, the effect on reactor performance of the extraction of a portion of the process stream between the two reactor sections is illustrated. The effect of the inert type on the axial and radial temperature profiles, the rate of heat accumulation and the reactor stability is presented.

It is observed that the packing type has a significant influence on the temperature profiles, especially the radial profiles. For a metal monolith, the magnitude of the radial gradients was reduced, in addition to an overall reduction in temperature being achieved. For the metal monolith, the decrease in width of the axial temperature profile was most pronounced.

Keywords: catalytic combustion; heat trap; reverse flow; energy generation; methane.

INTRODUCTION

Catalytic combustion is increasingly being used to produce usable thermal energy, often for environmental reasons (Forzatti and Groppi, 1999). Catalytic combustion is a flameless combustion process that can be used to oxidize fuels such as hydrocarbons and carbon monoxide at lower temperatures than conventional combustion, and also to combust mixtures with fuel to air ratios that cannot sustain a conventional flame, either because the energy content of the fuel is too low, or the mixture composition lies outside of the flammability limits. The relatively low temperature found in catalytic combustion also produces lower emissions of NO_X. The combustion unit is usually smaller than a conventional firebox and can be located in areas where conventional fired units would not be allowed (Hayes and Kolaczkowski, 1997). Catalytic combustion is also used in emissions control, for example, the automotive catalytic converter.

Recent attention has focused on the use of catalytic combustion to provide useful energy from streams of fugitive emissions (Aubé and Sapoundjiev, 2000) from such sources as coal mine air. Air extracted from underground coal mines contains up to 1.0% by volume methane. Potentially a useful source of energy, such streams of dilute methane have one drawback, that is, they are usually available at low temperatures. To achieve auto-thermal reactor operation in standard operating configuration, a fixed bed reactor would require a significant amount of preheating. To avoid preheating, a reverse flow reactor can be used. The concept of the catalytic flow reversal reactor (CFRR) was first discussed by Frank-Kamenetski (1955) and is reviewed by Matros and Bunimovich (1996). In a reversing flow reactor the feed is periodically switched between the two reactor ends using control valves; Figure 1 illustrates the concept. In Figure 1(a) control valves 1 and 4 are open and the feed flows to the reactor from left to right (forward flow). In Figure 1(b) control valves 2 and 3 are open and the feed flows to the reactor from right to left (reverse flow). The total cycle consists of these two operations, and the term switch time denotes the time at which the flow is changed from forward flow to reverse flow. Liu *et al.* (2001) demonstrated the effectiveness of a CFRR for lean methane combustion in an automobile application, the combustion of methane emissions from a natural gas/diesel dual fuel engine. The use of periodic flow reversal in a packed bed reactor for catalytic decontamination of waste gases was reported by Grozev and Sapundzhiev (1997) to achieve conversions of 99.5%. They placed a heat exchanger at the centre of the reactor to remove energy to ensure that the reactor did not overheat and deactivate the catalyst or damage the reactor. Heat removed from the reactor may be used for building heat or to drive a turbine. Heat removal from a reverse flow reactor is also discussed in Eigenburger and Nieken (1988) and Matros and Bunimovich (1996).

The purpose of this work is to present some experimental results obtained using a pilot scale CFRR for the combustion of lean methane with the extraction of energy from

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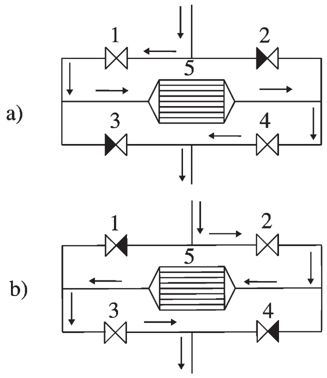

Figure 1. Illustration of the reverse flow reactor concept.

the midsection. The extraction of energy was accomplished by the removal of a fraction of the flowing gas, rather than the insertion of a heat exchanger, as discussed above. The effects of using different materials for inert sections on either end of the reaction sections is demonstrated. For application of this reactor, we are primarily interested in sources of lean methane that are available at ambient pressure, and therefore some compression is required to ensure flow through the reactor. Compression costs can be a significant fraction of total operating costs, thus there is interest in ensuring a minimum compression requirement. Structured packing, such as monoliths, can give a lower pressure drop than packed beds of particles. Typically, however, these structured packings have different thermal properties than packed beds. Therefore, we examined the effects of using two different types of monoliths and compared them to the results for packed beds. This comparison was performed for the inert sections of the reactor.

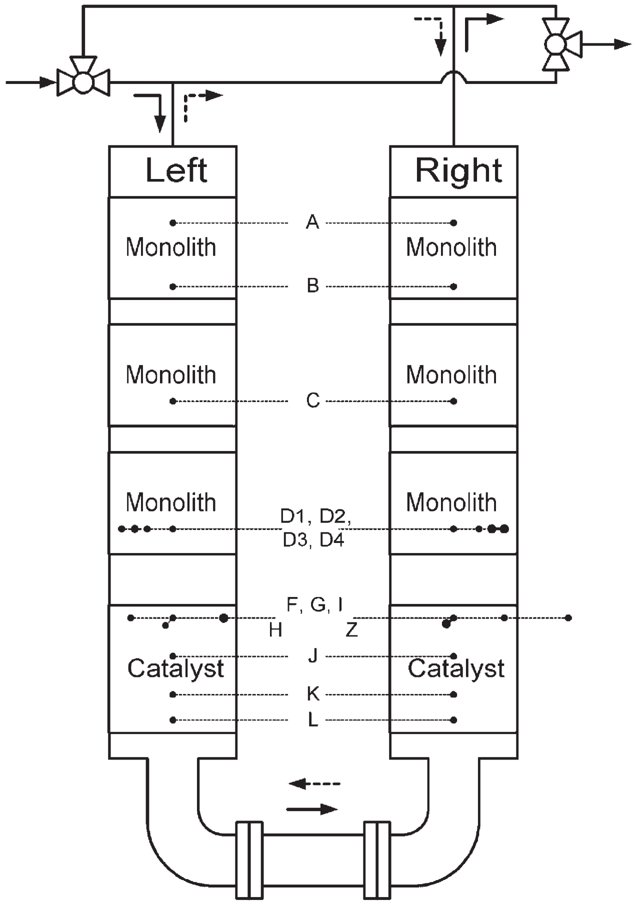

Figure 2. Schematic of the reactor used for the study with ceramic monolith inert sections shown. The letters denote thermocouple locations. Forward flow is from left to right.

EQUIPMENT

The reactor used in this investigation was located at the CANMET Energy Technology Centre, Varennes, Quebec, Canada. This reactor was used in previous investigations without heat extraction, which reported its operating characteristics (Salomons, 2003; Salomons *et al.*, 2003; Salomons *et al.*, 2004; Kushwaha *et al.*, 2004). Figure 2 gives a schematic of the reactor with ceramic inert sections fitted. The reactor had two parallel sections with an internal diameter of 0.2 m mounted side by side and connected by a U-bend at the bottom. Two three-way valves and associated transfer piping allowed for either forward or reverse flow operating modes. For reference purposes, forward flow occurs when the gas flow was from the left section to the right section (see Figure 2), whilst flow from right to left is reverse flow. The reactor was made from Hastelloy to allow operating temperatures up to 1000°C. The reactor walls were 3.2 mm thick and were surrounded by an insulation blanket 0.28 m thick. Physical properties of the wall and insulation are given in Table 1.

The reactor internals consisted of a combination of both inert and catalyst sections. The role of the inert sections was to trap energy in the reactor, giving a higher maximum temperature (Eigenberger and Nieken, 1988). Sapundzhiev *et al.* (1990) used an inert layer at the entrance and exit of the reactor to retain heat and minimize the amount of catalyst.

The porosity, bulk density, solid thermal conductivity, heat capacity and surface area to volume ratio for the catalyst particles and inert materials are given in Table 1. Other properties are given in the following. The catalyst sections were packed beds of Raschig rings with a characteristic length of 7.5 mm and containing a non-noble metal catalyst. Properties of the catalyst packing were given by Aubé and Sapoundjiev (2000). The inert sections were one of ceramic monoliths, metal monoliths or Denstone balls. The ceramic monoliths, supplied by Corning Inc., were composed of Celcor 9475 (EX-20) Cordierite with a density of 1683 kg/m^3. The monoliths had 100 cells per square inch (CPSI). The hydraulic diameter of each

Table 1. Summary of physical properties of the reactor internals.

	ρ_{bulk} (kg/m^3)	ε	φ (m^2/m^3)	k W/(m·K)	C_P J/(kg·K)
Reactor wall	7800	NA	NA	14.3	460
Insulation	128	NA	NA	0.144	1340
Ceramic monolith	423	0.75	1340	1.46	1040
Metal monolith	532	0.93	2000	26.0	461
Denstone balls	1440	0.40	360	1.46	1040
Catalyst rings	632	0.51		0.5	1020