Chemical Engineering Science 164 (2017) 122–132

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Experimental investigations on unmixed combustion for heat transfer applications

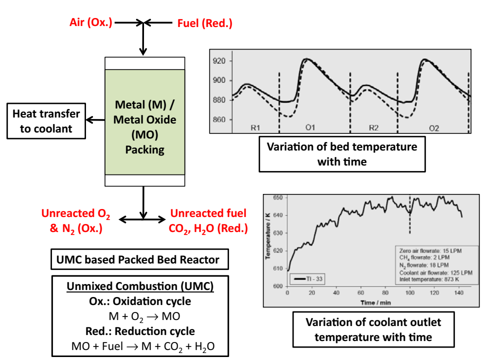
Amol Deshpande, Srinivas Krishnaswamy *, Krishnaswamy Ponnani

Department of Chemical Engineering, BITS - Pilani, K K Birla Goa campus, Zuarinagar, Goa 403726, India

HIGHLIGHTS

- Novel unmixed combustion process for heat transfer applications demonstrated.
- A test rig based on packed bed concept has been designed and successfully operated.
- Heat transfer under sustained combustion primarily by conduction has been achieved.
- Under cyclic operation, variations in bed and coolant outlet temperature was minimal.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 26 August 2016
Received in revised form 13 January 2017
Accepted 28 January 2017
Available online 8 February 2017

Keywords:
Unmixed Combustion (UMC)
Chemical Looping Combustion (CLC)
Heat transfer
Packed bed reactor
Oxygen Storage and Release Material (OSRM)

ABSTRACT

The potential of Unmixed Combustion (UMC), a novel variant of Chemical Looping Combustion (CLC), is demonstrated for heat transfer applications. A purpose-built test rig, based on a dynamically operated packed bed reactor concept, was designed, fabricated and commissioned. Experiments were conducted using representative Cu based Oxygen Storage and Release Material (OSRM) which was alternately reduced and oxidized in the bed using methane and zero air (21 mol% O_2) respectively. The energy generated in both reactions due to exothermicity was radially transferred by conduction and convection to coolant air under sustained combustion conditions. For a specific loading of 1.25 kg of OSRM and fixed reaction cycle times, the effect of varying zero air flowrate, methane concentration and coolant flowrate on the radial heat transfer was investigated. The radial heat transfer rate was maximized at 95 ± 2% of total energy in the bed at zero air, methane and coolant flowrates of 15 LPM, 2 LPM and 150 LPM respectively. Under "cyclic" steady state conditions, the variation of bed temperature and coolant outlet temperature was restricted to within ±30 K and ±3 K respectively and the combustion process was observed to be self-sustaining. The results obtained present a strong argument for using UMC based reactor systems as an alternative to "premixed" combustion for process heat transfer applications subject to suitable combination of operating conditions inclusive of cycle time, reactor material of construction and choice of OSRM.

1. Introduction

Over the years, combustion has been largely based on "premixing" fossil fuel with excess air in a closed firebox and heated to produce hot flue gas. The enthalpy of the flue gas is subsequently

* Corresponding author.
E-mail address: srinivas@goa.bits-pilani.ac.in (S. Krishnaswamy).

http://dx.doi.org/10.1016/j.ces.2017.01.072
0009-2509/

Nomenclature

Symbols

c_{pc}	coolant air specific heat (J kg^{-1} K^{-1})
ΔH	standard heat of reaction (J/mol)
$\dot{m}_c$	coolant mass flowrate (kg s^{-1})
n	number of moles (mol)
N	number of cycles
Q	total amount of energy generated or transferred (J)
Q_c	total energy transfer to coolant air (J)
$\dot{Q}_{ext}$	external power supply in N cycles (W)
Q_{ext}	total energy supplied externally (J)
t	cycle time (s)
T	thermodynamic temperature (K)
ΔT_c	coolant temperature rise across zone 2 of the furnace (K)
X	conversion

Subscripts

ox	oxidation
red	reduction

transferred primarily via convection and radiation and used for process applications like steam generation and power production. This process however has following limitations:

1. NO_x and carbon dioxide (CO_2) emissions in flue gas
2. Difficulties in CO_2 capture from flue gas
3. Energy loss from hot flue gas
4. Excess entropy generation due to high temperatures involved

These limitations pose a dual challenge of improving thermal efficiency and reducing the impact on environment simultaneously. Over the years, several researchers have attempted addressing the above issues, based on system improvements post combustion or investigating pre-combustion based technologies like gasification and oxy-fuel combustion (Kanniche et al., 2010; Leung et al., 2014; Cuellar-Franca and Azapagic, 2015). The emphasis of these investigations however has been on energy recovery and carbon dioxide (CO_2) capture for reuse or sequestration. Very few attempts have been made to change the fundamental nature of the combustion process itself which on the whole involves integrating or "premixing" fuel, air or O_2 and energy together to produce fire.

A novel form of combustion called "Unmixed Combustion (UMC)" was proposed for controlling puffing in rotary kiln incinerators (Lyon, 1993). This process referred to as an "alternative to fire" occurs when air and fuel in alternate cycles pass over an OSRM (Oxygen Storage and Release Material) that undergoes oxidation and reduction. These reactions if exothermic, can release energy in the form of heat that can potentially be transferred by conduction and convection to be subsequently utilized in process applications like providing heat for cold starting of engines, generation of inert gas for airplane fuel tanks, avoiding NO_x emissions and supplying heat for endothermic reactions like steam reforming (Lyon and Pittstown, 1998; Lyon and Cole, 2000). Nitrogen (N_2) and unreacted oxygen (O_2) exit during oxidation while CO_2 and steam are produced in the reduction cycle. Pure CO_2 becomes available for storage, sequestration or reuse purposes without any additional separation units, but merely by condensing water out. As this process operates at relatively lower temperatures than "premixed" combustion without generating flames, the amount of excess entropy generation can be reduced and formation of NO_x can also be avoided.

Lyon and Cole (2000) have discussed various potential applications of UMC including boiler systems. In this context, they passed CO and air alternately over 25 wt% Cu on alumina rings and the enthalpy of reaction was used for heating water, which resulted in a heat flux of 22.2 W/cm^2. The phenomenal increase in recovering energy is mainly due to heat transfer by conduction through the solid phase coupled with convection as against radiation and convection prevalent in an existing "premixed" combustion based boiler system. However, the practical possibility of effectively transferring energy from endothermic or exothermic reactions in alternate cycles by conduction and convection under sustained combustion conditions will pose a major challenge and has not been investigated till date. This formed the genesis of the present work.

UMC can be carried out in fluidized or packed bed reactors, the former being referred to as Chemical Looping Combustion (CLC). Hossain and de Lasa (2008) and Adanez et al. (2012) have presented comprehensive reviews on CLC reporting the main advances in this technology. More recently theoretical and experimental studies have been conducted on the use of packed bed reactor (PBR) technology for CLC and these investigations have concluded that this technology has a great potential as an alternative to the fluidized bed system (Noorman et al., 2007, 2010, 2011a, 2011b, 2011c). The emphasis of these studies was however on producing a very high temperature gas stream exiting the reactor axially to drive turbines in power plants and simultaneously obtaining pure CO_2 for sequestration purposes.

The choice of a suitable OSRM is vital to the design of UMC systems and its final selection is dictated by its reactivity, stability, resistance to coke formation and cost. Different metal oxides have been reported in literature as possible candidates namely: CuO, CdO, NiO, Mn_2O_3, Fe_2O_3, and CoO (Hossain and de Lasa, 2008; Chandel et al., 2009; Cho et al. 2004; Albert et al., 2007; Chuang et al., 2008).

The objective of this study was to demonstrate the potential of a dynamically operated UMC based packed bed reactor for radial heat transfer to a coolant using conduction and convection under sustained operating conditions. Cu/CuO was chosen as a representative OSRM in this study because of its high reactivity and stability and that both oxidation-reduction reactions involved are exothermic (Noorman et al., 2007, 2011c; Chuang et al. 2008). A proof-of-concept test rig was designed for this purpose and the fraction of radial heat transfer from the bed has been estimated. The effect of zero air, coolant and methane flowrates and reactive gas inlet temperatures on the radial heat transfer has been investigated. The reactions involved with their respective enthalpies are given in Table 1.

Table 1
Reaction details.

Reaction	Standard heat of reaction (ΔH)
$4Cu + 2O_2 \rightarrow 4CuO$ (Oxidation)	$\Delta H_{ox} = -312$ kJ/mol of O_2
$CH_4 + 4CuO \rightarrow 4Cu + CO_2 + 2H_2O$ (Reduction)	$\Delta H_{red} = -178$ kJ/mol of CH_4