



## Full Length Article

# Performance of a Victorian brown coal and iron ore during chemical looping combustion in a 10 kW<sub>th</sub> alternating fluidized bed



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## HIGHLIGHTS

- Chemical looping combustion of Victorian brown coal and char studied with iron ore.
- The optimum temperature for CO<sub>2</sub> yield and carbon conversion was 900 °C.
- Oxygen carrier diameter decreases with increased reaction cycles.
- Coal is preferable to char, with less time required and higher carbon conversion.
- No ash deposition was observed on the oxygen carrier particles.

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## ABSTRACT

This paper presents the performance of Victorian Yallourn coal during chemical looping combustion with iron ore in a 10 kW<sub>th</sub> alternating fluidized bed reactor. CO<sub>2</sub> was used as the gasification agent and fluidization gas. The effect of reaction temperature, fuel type and number of redox cycles were investigated. The NO<sub>x</sub> emission from the system was determined. The used oxygen carrier particles were characterized using SEM, EDS, XRD, and surface area analyser. It was found that the optimum condition for CO<sub>2</sub> yield and carbon conversion was obtained at a temperature of 900 °C. Under this condition with the present reactor configuration, the average carbon conversion and CO<sub>2</sub> yield over six cycles were found to be 92.2% and 82.9% respectively. Although Yallourn ash has high amounts of iron and silica, no ash deposition was detected on the oxygen carrier particles. The NO<sub>x</sub> concentration was found to be 25 ppm. Results from the carbon balance showed that about 1% was present in the solid phase; about 6.8% was unaccounted for and possibly elutriated out of the reactor. While favourable results were obtained for this configuration, the use of circulating fluidized bed reactors and an improved cyclone separation system would increase the carbon conversion and CO<sub>2</sub> yield.

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## 1. Introduction

Combustion of fossil fuels is a major contributor to CO<sub>2</sub> emissions from power generation [1], with the CO<sub>2</sub> concentration in the atmosphere exceeding 400 ppm in 2015 [2]. According to the International Energy Agency (IEA) current estimates show carbon-based fuels meet over 80% of the world's electricity demands [3]. With such a large reliance on fossil fuels, CO<sub>2</sub> capture and storage (CCS) technology can be an important option to reduce CO<sub>2</sub> emissions from fossil fuel combustion. However, most CO<sub>2</sub> capture processes remain highly energy intensive [4]. Chemical Looping Combustion (CLC) has emerged as a promising CO<sub>2</sub> capture process with the benefit of inherent CO<sub>2</sub> separation,

eliminating the need for energy-intensive and costly external separation processes. CLC employs the use of oxygen-containing solid materials, called oxygen carriers (OCs), to transfer oxygen from air for the combustion of fuel, avoiding direct contact between the fuel and air. Subsequently, the reduced OC are re-oxidized in the presence of air. The cycling process can be repeated and upon condensation, will yield a flue gas stream consisting primarily of CO<sub>2</sub> ready for capture.

Recent investigations have shown that low-rank coals have potential as a fuel source for CLC [5–15]. Victoria in Australia has large reserves of low-rank coals classified as brown coal. Low-rank coals are characterized by higher moisture content, volatile matter, and reactivity compared to bituminous coal and anthracite. However, they have a lower energy density than coals of higher rank. Compared to bituminous coals, the gasification of low-rank coal occurs faster [16].

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Promising oxygen carriers (OC) can be evaluated by three criteria: oxygen capacity, availability or economics and non-toxicity [17]. Recent investigations using lignites and brown coals have focused on employing Fe-based oxygen carriers for CLC. Fe-based materials have been attractive as oxygen carriers due to their low cost, mechanical and thermal stability, lack of environmental impact and non-toxicity [18], and have been widely utilized as singular or combined oxygen carriers with other metal oxides. Larring et al. investigated a range of Fe-Mn-based minerals, comparing their redox capacity and oxygen capacity to Ilmenite [17]. Larring et al. found Kryvbas and Sinai-A oxygen carriers give higher reaction rates and oxygen capacities, and have speculated the mixed phases may result in a longer lifespan and a reduced need for oxygen carrier pre-treatment [17]. Zhang et al. utilized a combined Fe-Mn oxygen carrier for CLC with Victorian brown coal in a TGA and fluidized bed reactor [15]. In a similar manner, Rajendran et al. combined a Fe-based oxygen carrier with Mn, Ni, and Cu through impregnation [7]. It was concluded that the addition of the Mn, Ni, and Cu oxide particles improves the conversion of the coal volatile and gasification products, as well as enhancing the carbon conversion. However, there is little information in the literature regarding the effect of long-term usage of Victorian brown coal with a Fe-based ( $\text{Fe}_2\text{O}_3$ ) OC in a CLC system. Furthermore, continuous operation over an extended period has not been investigated when using Victorian brown coal. Therefore, this investigation will use a continuous fuel feeding 10 kW<sub>th</sub> alternating fluidized bed reactor to generate information crucial for the determination of Fe-based OCs viability with Victorian brown coals in a CLC system.

## 2. Materials and methods

### 2.1. Materials

The fuel used in this study was Yallourn coal, Victorian brown coal mined from the Latrobe Valley in Australia and is currently used for power generation in Victoria. The proximate, ultimate analysis and ash analysis of the coal are found in Table 1. This coal is known to be very friable under fluidized bed conditions and at higher temperatures [16]. Char was used as a comparative fuel type and was prepared by pyrolysis at 1000 °C under  $\text{N}_2$  for 8 h. Both coal and char were sieved to a size range of 600–1000  $\mu\text{m}$ . The oxygen carrier (OC) used in this study was iron ore from Western Australia, with its composition after calcination in Table 2. The OC contains predominantly  $\text{Fe}_2\text{O}_3$  (~95%) with small quantities of  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ , among other minerals. The oxygen carrier was sieved to a size range of 150–350  $\mu\text{m}$  for all experiments.

### 2.2. Experimental procedure

An important aspect of the 10 kW<sub>th</sub> reactor is its comparison with laboratory scale reactors. The larger size ensures that phenomena observed in laboratory experiments, such as bed

**Table 1**  
Proximate, Ultimate and Ash composition of Yallourn coal.

Proximate analysis		Ash composition (%)	
Ash, % db	2.4	$\text{SiO}_2$	10.4
Volatile matter, % db	50.4	$\text{Al}_2\text{O}_3$	4.0
Fixed carbon, % db	47.2	$\text{Fe}_2\text{O}_3$	40.2
Ultimate analysis		$\text{TiO}_2$	0.2
C, % daf	70.6	$\text{K}_2\text{O}$	0.6
H, % daf	4.6	MgO	15.7
N, % daf	0.4	$\text{Na}_2\text{O}$	5.6
S, % daf	0.2	CaO	7.8
O, % daf	24.2	$\text{SO}_3$	15.5

(% db = % dry basis, % daf = % dry ash free basis).

**Table 2**  
Calculated oxygen carrier (OC) composition.

Composition	Mass fraction (%)
$\text{Fe}_2\text{O}_3$	95.15
$\text{SiO}_2$	2.21
$\text{Na}_2\text{O}$	1.20
$\text{Al}_2\text{O}_3$	1.11
MgO	0.20
$\text{P}_2\text{O}_5$	0.07
$\text{SO}_3$	0.02
CuO	0.02
$\text{As}_2\text{O}_3$	0.01
ZnO	0.01

agglomeration due to wall effects, become less noticeable. The 10 kW<sub>th</sub> alternating fluidized bed reactor is shown in Fig. 1 with a 100 mm inner diameter and 1.5 m high reaction zone with an expanded 150 mm 2.5 m freeboard section. The fuel is fed continuously over a 3 min period using a screw feeder, with the outlet of the feeding tube terminating at the base of the oxygen carrier bed to ensure contact of the volatiles evolved during feeding with the oxygen carrier as the coal enters the reactor. 1 kg of sand was added to the reactor as the inert bed material followed by 5 kg of OC. The fluidization gas flow rate during reduction was 25 L/min and the gas was composed of 20 v/v%  $\text{CO}_2$  with  $\text{N}_2$  balance. During the oxidation cycle, the same flow rate of 25 L/min was used composed of 80 v/v% air with an  $\text{N}_2$  balance. Fifty grams of coal was added during each reduction reaction to achieve an OC to fuel ratio of 100:1. Cyclones at the reactor outlet were used to collect fly ash and coal fines elutriated from the combustion zone. Most experiments were conducted over 5–6 reduction cycles, as operational experience has shown that typically 5–6 reduction cycles were required to obtain a stable gas composition. The effect of temperature was investigated by performing experiments at 100 °C intervals between 700 and 1000 °C. Experiments were also conducted with both Yallourn coal and char to study the effect of the fuel type. Finally, a 36-cycle experiment was completed to study the performance of the OC over an extended period.

The outlet gas composition was analysed for  $\text{CO}_2$ , CO,  $\text{CH}_4$ ,  $\text{H}_2$ , and  $\text{O}_2$  using a Fuji Electric Systems Co., Ltd. online gas analyser. For trace  $\text{NO}_x$  gases, NO and  $\text{NO}_2$  concentrations were measured using a combination of Dräger tubes and Gas Chromatography.

### 2.3. Sample characterization

The oxygen carrier samples, which were collected at the end of every run, were characterized using a range of techniques to study the changes in composition, morphology, and structure. To investigate the morphological and chemical composition changes of the OC a JOEL 7001F Scanning Electron Microscope (SEM) together with an Oxford Instruments X-Max 80 Silicon Drift Energy Dispersive Spectroscopy (EDS) were used. To detect the presence of surface crystalline compounds in the oxygen carrier, a Rigaku Miniflex 600 X-ray Diffraction (XRD) unit, Cu targeted with  $\text{K}\alpha$  radiation was used at 40 kV and 15 mA. The analysis was conducted in a step-scan manner with a step size of 0.02° over a 2 $\theta$  range of 3–80°. The Brunauer, Emmett and Teller (BET) surface area, cumulative pore volume and average pore size was obtained using a Micromeritics Accelerated Surface Area and Porosimetry (ASAP) 2010 instrument.

### 2.4. Data processing

To obtain the instantaneous gas fraction during the reduction reaction, the following equation was used:

$$f_i = \frac{x_i}{(x_{\text{CO}} + x_{\text{CO}_2} + x_{\text{CH}_4} + x_{\text{H}_2})}$$

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