Advancing Chemical Looping Combustion Technology for Victorian Brown Coals

Final Report

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EXECUTIVE SUMMARY

This is an industry-academia collaborative project on Chemical Looping Combustion (CLC) of Victorian brown coal. The project partners included Monash University, CSIRO Energy, Huazhong University of Science and Technology (HUST) -China, University of Alberta - Canada, Alstom –Germany, Victorian power generator Energy Australia, oxygen carrier manufacturer VITO-Belgium and Engineering Consultancy Lycopodium-Australia, drawing on relevant and complementary strengths from the respective participants.

Chemical looping combustion of fossil fuels such as brown coal is considered to be an emerging alternative technology to facilitate capture of CO_2 at a lower energy and cost penalty. The technological approach uses metal oxides (OC) rather than concentrated gaseous oxygen from air separation plants which are known to be expensive and very energy intensive for other technologies such as oxy-combustion and gasification. Chemical looping concept has been widely studied worldwide for combustion of natural gas; however, its application to solid fuel, such as coal, is being studied relatively recently.

In CLC, a reducible metal oxide supplies the oxygen to the fuel, and the spent oxygen carrier is separately re-oxidised in an air stream, unlike using pure oxygen gas obtained from energy intensive air separation units in oxy-fuel combustion. The products of CLC are CO_2 and water vapour. By condensing the water vapour from the mixture, CO2 is produced in a highly concentrated form which does not require further costly separation or purification. In addition to easier CO_2 capture, the technology also offers high fuel combustion efficiencies with the introduction of reforming gases like CO_2 and steam which promote the gasification of char.

The overall technical feasibility of the process is largely dependent on several factors - oxygen carrier particles, their interaction with brown coal's mineral matter, their stability, attrition resistance and uniform distribution of coal in the fuel reactor. The future commercial prospect and viability of the process would depend on:

- Identification of optimum condition for high conversion of brown coal in a fully looping CLC environment
- Availability of effective, stable and affordable metal oxide as an oxygen carrier
- Optimum reactor system that is scalable with efficient integrated energy management
- Low overall operating and capital requirements.
- Most importantly, acceptable techno-economics of the process, *i.e*. meeting the USDOE target of the levelized cost of electricity (LCOE) increase of no more than 35% over the SCPC without CCS and efficiency decrease no more than 10% over the SCPC without CCS.

However, such technical and cost analysis has not been carried out for the Victorian brown coals due to limited relevant performance data. To better understand these uncertainties and opportunities, it was necessary to take the previous BCIA funded work to the next level, through further testing of the CLC of brown coal in a continuous fully looping facility while also performing fundamental scientific work on coal-OC interactions. This project addressed this shortfall in knowledge through experimental and analytical work that included the following:

- Experiments at Monash using a non-looping CL reactor and thermo-gravimetry apparatus
- Experiments at CSIRO using a newly-designed and commissioned fully-looping CL reactor
- X-ray diffraction – at Monash University and the Australian Synchrotron, Temperature Programmed Reduction (TPR) and Scanning Electron Microscopy with Energy Dispersive Spectroscopy (EDS) at Monash University and the CSIRO

- Process simulation of a 550MWe net CLC plant complete with drying and all necessary units following the DOE report for 550MWe “Commercialization of an atmospheric iron-based CDCL process for power production. Phase 1: Technoeconomic analysis”. The costs were converted to AUD following the guidelines of a report prepared for “Australian Government Department of Resources, Energy and Tourism” by “Electric Power Research Institute (EPRI)”. A standard power rule of 0.6 was used to adjust the cost of equipment for the increase in capacity. The costs were also adjusted for the year 2017 using Chemical Engineering Plant Cost Index (CEPCI) from the base cost year of 2011.

Three Victorian brown coals - Loy Yang, Morwell and Yallourn -, a Canadian lignite and a Chinese bituminous coal was tested in this project. Three different oxygen carriers identified from the previous project (Hematite, Ilmenite and an Mn/Fe -based synthetic oxygen carrier prepared and supplied by project partner VITO) were used as oxygen carriers. The key information generated are summarised below:

<u>Carbon conversion, agglomeration propensity, coal type, the performance of oxygen carrier</u>

- High oxygen release from oxygen carrier can be achieved between 800- 900^{0}C.
- From the unoptimised bench scale non-looping reactor operation between 800-1000°C, carbon –conversion is >90%, while the CO_2 yield was >80%. These figures will be higher under optimised condition.
- There were no visible signs of agglomeration for the Victorian brown coal ash-OC under the 800-1000°C range tested for up to 40 hours in the bench scale non-looping reactor
- In thermo-gravimetry apparatus based tests, the high ash Canadian and Chinese coals performed poorly compared to the Victorian brown coal regarding OC activity and carbon conversion due to deposition of coal ash on the OC surface
- Based on the 50 alternating reduction-oxidation cycles in a bench scale using Yallourn brown coal and three oxygen-carriers – hematite, ilmenite and Mn-Fe synthetic oxide – the order of activity (oxygen transfer ability) was found as hematite > ilmenite > synthetic carrier.

  It is important to note that synthetic carriers will be generally more expensive compared to the natural minerals hematite and ilmenite.
- Based on the observation above, further tests to assess the interaction of volatiles and OC, volatile-OC tests using hematite were undertaken in the bench-scale reactor; the hematite activity dropped after 120 hours of operation. This is an important information for the techno-economic study, in particular, the operating cost involving the cost of the oxygen carrier and determining it’s the make-up rate.
- The volatiles released from coal pyrolysis are CO, H_2, CH_4 and CO_2. The most reactive gas in converting metal oxide is H_2 with lower reactivity from CO and CH_4.which is mostly unreactive. The (very well controlled) temperature programmed reduction experiments with the three oxygen carriers suggested that initial reduction or oxygen transfer starts around 400°C and continue through to 1000°C. These are broadly in line with what was observed in much larger bench scale operation mentioned earlier. However, for CLC the presence and strong reactivity of hydrogen may result in a lack of available oxygen for CO reduction in the oxygen carrier resulting in the lower conversion of CO to CO_2. This is an important information in designing the internals for the fuel reactor to ensure effective contact between all components of the volatiles and the OC.

<u>Fully looping reactor operation at CSIRO</u>

The test campaign is designed to obtain key scientific information about CLC process using brown coal such as:

- CLC behaviour under a continuous fully looping environment
- Effect of process parameters on CLC conversion
- Overall effectiveness and stability of oxygen carrier
- The possible impact of coal ash-oxygen carrier interaction on CLC performance

The following summarizes the key findings and conclusions from the test campaign, relevant to addressing some of the scientific and engineering questions associated with CLC technology for brown coal.

- Stable continuous CLC operation for up to 35 hours using brown coal together with ilmenite has been demonstrated using the versatile prototype scale CLC reactor. The reactor was able to deliver the required circulation of oxygen carrier (metal oxide) to continuously sustain the CLC redox reaction.
- The 35-hour CLC operation has been accumulated over a number of consecutive test runs which involve separate start-ups and shut-downs. From these independent test runs, it was possible to demonstrate the robustness of the process, in term of process reproducibility and repeatability of the CLC operation with good control.
- The CLC is shown to be very responsive to process dynamic changes. Steady state operation can be achieved within minutes, thus providing a very important operational knowledge and understanding of the process. Specifically, the tests have been able to demonstrate the consistent operational behaviour of the CLC especially in response to the parameter changes, e.g. inter-relationship between combustion rate of fuel and oxidation of metal oxide.
- The reactivity of the oxygen carrier (ilmenite) is found to be improving over the course of the current test campaign (35 hrs). The CO_2-to-CO ratio of the Fuel Reactor exit gas increases progressively from a factor of just over 2 to close to 9 after 35 hrs of CLC operation.
- Detailed characterisation and analysis of the oxygen carrier (ilmenite) using XRF, XRD and SEM-EDX have provided valuable insights and a possible explanation for the increased reactivity of the oxygen carrier.
- No notable adverse effect of ash on the reactivity of the oxygen carrier or the CLC performance is found from the current study, equivalent to over 500 redox cycles based the oxygen carrier particle residence time.
- The preliminary carbon balance analysis suggests that the current CLC experimental test unit could achieve about 50% carbon conversion efficiency. The other losses are due to breakthrough of the volatiles, tar and fine char due to either short gas residence time in the bed or elutriation of very fine char from the Fuel Reactor because there is no mechanism for separating and recycling the fine char back to the reactor in order to further improve the overall carbon conversion efficiency.

In summary, the study has shown that the Victorian brown coal can be used effectively with low cost Australian naturally occurring mineral (ilmenite) for CLC process. Useful operational knowledge and insights have been gained from the experimental program. The prospect of using a low cost Australian natural mineral (ilmenite) as CLC oxygen carrier is key in offering an economically and commercially viable solution for conversion of brown coal with low emission potential using the CLC process.

Process simulation and techno-economics study

Process simulation was carried out using Aspen Plus for a Fe-based CLC flowsheet for a 550MWe plant developed by the USDOE. We integrated an RWE-WTA type dryer at the front end but with a very conservative integration avoiding “deep integration” for heat recovery which is often seen in academic studies. Our methodology included broadly reproducing the DOE report’s flowsheet before starting ours.

The economic analysis of the CDCL process was conducted following DOE/NETL's Quality Guidelines. The cost for the CDCL plant was developed at the Total Plant Cost (TPC) level, which includes equipment, material, indirect labour costs, engineering and contingencies. The cost of balance of plant equipment was developed following the 550MWe net CDCL process from DOE report, "Commercialization of an atmospheric iron-based CDCL process for power production. Phase 1: Techno-economic analysis", submitted by "The Babcock & Wilcox Generation Group, Inc". The costs were converted to AUD following the guidelines of a report prepared for "Australian Government Department of Resources, Energy and Tourism" by "Electric Power Research Institute (EPRI)". A standard power rule of 0.6 was used to adjust the cost of equipment for the increase in capacity. The costs were also adjusted for the year 2017 using Chemical Engineering Plant Cost Index (CEPCI) as the base cost year was 2011.

CDCL process equipment include the reducers, combustors, risers, distributers, coal injection system, particle makeup, steam generating surface in the combustor and heat transfer surface at the exit of the combustor, air and CO_2 heaters, burners, and CO_2 compressor. For the CDCL equipment, 15% project contingency and 20% process contingency were applied. The levelized Cost of electricity (CoE) was developed following the guidelines of a report prepared for "Australian Government Department of Resources, Energy and Tourism" by "Electric Power Research Institute (EPRI)"Key conclusions are:

- The HHV, net efficiency is 35% *meaning that the DOE efficiency target is met*
- The total plant cost is approximately A$4200 per net kW
- With a capacity factor of 85%, cost of electricity is estimated at approximately $150/MWh

The COE compares with the COE for supercritical brown coal without CCS of $125/MWh (BREE, 2013), therefore the CLC COE from this study is at an acceptable point. Another comparison point is Alstom's figure for their 2015 study for bituminous coal where the COE is US$97/MWh which is roughly A$130/MWh; the brown coal data from this study involved moist (62% moisture and hence includes a dryer and considers associated larger coal feed rate.

- A ±20% sensitivity to the major cost items - coal dryer cost, CLC equipment cost and CO_2 removal and compression cost – suggest that there will be a ±12% effect on the total plant cost and ±3% effect on the COE.

We emphasise here that CLC is still an emerging technology, and therefore, some of the cost figures may be uncertain. There will be unknown fuel costs during start-up even though we have considered natural gas start-up and one can argue that this will be no different to the practices used in commercial fluidized beds currently operating at comparable scale – 600MWe. However, the availability of plant involving the operation of two reactors – fuel reactor and the OC combustor – in tandem is still uncertain. At the same time, one can argue that this is no different from what happens in a large CFB plant with the main riser resembling the fuel reactor and the external heat exchanger resembling the OC combustor to a large extent.

<u>Skills development</u>

The build-up of critical expertise and skills locally in this emerging area was one objective of this project. As a means of capacity building, the project has so far trained two PhD students, one Research Fellow, and four undergraduate students. Apart from the fundamentals of chemical looping combustion and gasification, these researchers gained knowledge in the design, construction, commissioning and operation of high temperature chemical looping reactor, advanced chemical analysis of fuel samples, and thermodynamic modelling. The reactors built for this project and the previous project also funded by the BCIA will be useful for ongoing work in this area by postgraduate and undergraduate students.

<u>Recommendations</u>

While the project has generated substantial scientific and technical information on chemical looping combustion behaviour of Victorian brown coals and other coals from around the world, the following additional investigations are recommended:

- Extend the test to fully evaluate the stability of the oxygen carrier by conducting longer test campaign in the continuous looping reactor at the CSIRO using Victorian brown coal, more than 100 hrs; this could not be done due to resource and time constraints.
- Evaluate the efficacy of the CLC process for other Victorian brown coals as well as examining other ilmenite types and natural minerals such as manganese-rich ore (or mixed oxides) to exploit chemical looping oxygen uncoupling (CLOU) effect.
- Evaluate the impact of the high CO_2 concentration on the combustibility behaviour has not been fully evaluated here. It is proposed to conduct a more detailed study where a majority of the fluidizing gas in the Fuel Reactor is fluidised with CO_2 and in combination with steam to enhance the in-situ gasification.
- Modify the reactor system may also be considered to separate and reinject the fine char escaping from the Fuel Reactor back again to improve the overall carbon conversion.
- Modify the current CLC test system to allow evaluation of the efficacy of the CLC-based hydrogen production process. This is given the fact that a separate desktop study has shown the prospect of using CLC to enable low cost hydrogen production using Victorian brown coal.

<u>Final thoughts</u>

It is clear from the process simulation and the techno-economics analysis that the DOE efficiency target is met while the COE is at an acceptable point and is sufficiently favourable to warrant further development of the CLC for Victorian brown coals.

It is also clear from the two BCIA-ANLEC R&D projects and works from around the world at up to 1 MWth on CLC that this is a prospective technology and will have a niche application for highly reactive low-ash coals such as the Victorian brown coals relative to the high ash coals from around the world. Although these two projects' focus was on CLC based power generation, we would like to emphasise that the CLC process can also be adjusted towards alternative ways of improved usage of the brown coal resource, such as coal-to-liquids and chemicals production.