Chemical Looping Air Separation for Oxy-fuel Power Plants

by

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Discipline of Chemical Engineering

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Declaration

I hereby certify that this thesis contains no material which has been accepted for the award of any other degree or diploma in any university or other tertiary institution and, to the best of my knowledge and belief, contains no material previously published or written by another person, except where due reference has been made in the text. I give consent to the final version of my thesis being made available worldwide when deposited in the University’s Digital Repository, subject to the provisions of the Copyright Act 1968.

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August, 2014
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List of Publications

The following publications have arisen from the study presented in current thesis.


Song H., Shah K., Doroodchi E. and Moghtaderi B., *Development of a Cu-Mg-based oxygen carrier with SiO₂ as a support for chemical looping air separation*. Energy & Fuels, 2013. 28(1) P. 163-172


Song H., Shah K., Doroodchi E. and Moghtaderi B., *A spray dried CuO/MgAl₂O₄ oxygen carrier for chemical looping air separation*. (To be submitted to Energy & Fuels)

Song H., Shah K., Doroodchi E. and Moghtaderi B., *Reactivity of Cu-M/SiO₂ (M = Fe, Ni, Mg, Co and Mn) bimetallic oxygen carrier for chemical looping air separation* (Writing in Progress)

Song H., Shah K., Doroodchi E. and Moghtaderi B., *Demonstration of chemical looping air separation using an impregnated Cu-Mg/SiO₂ bimetallic oxygen carrier* (Writing in Progress)

The following papers, which are not related to this thesis, have also been published during my PhD study.

Song H., Doroodchi E. and Moghtaderi B., *Redox characteristics of Fe–Ni/SiO₂ bimetallic oxygen carriers in CO under conditions pertinent to chemical looping combustion*. Energy & Fuels, 2011. 26(1) P. 75-84

Executive Summary

Oxy-fuel combustion, which refers to the combustion of coal in the presence of oxygen rather than air, is one of the key technology options among the portfolio of carbon capture and storage (CCS) technologies. Oxy-fuel combustion captures carbon dioxide in-situ, producing a CO₂-enriched flue gas stream (95 vol% CO₂) ready for storage. The main shortcoming of the oxy-fuel combustion, however, is the high cost and energy intensity of its oxygen plant. At present, the main technology option for the oxygen plant is cryogenic air separation which typically consumes about 20% of the total electricity produced in oxy-firing mode and, thus significantly reduces the thermal efficiency of the oxy-fuel power plant. The oxygen plant also accounts for 30% of the total capital investment. In view of the above, a comprehensive program of study on alternative air separation technology - chemical looping air separation (CLAS), which offers a cost effective method for large-scale oxygen production, has been systematically conducted in the current study.

The present study aims to identify and characterise the suitable metal oxide oxygen carriers for CLAS applications. The broad objectives of the project have been achieved using a combined theoretical and experimental approach. A comprehensive review of the current state of oxygen carrier developments and utilisation was carried out. Other emerging air separation methods have also been reviewed in detail as part of this literature review. A thermodynamic method of identifying the feasible metal oxide oxygen carriers for high temperature air separation was developed in the present study. The energy cost associated with oxygen production using CLAS and its comparison with an advanced cryogenic air separation unit has been investigated in detail. The reaction mechanisms, underpinning the oxidation and oxygen release processes under conditions pertinent to CLAS, were also determined as one of the main objectives for the current study. The relevant experiments have been carried out using a variety of experimental setups, including thermogravimetric analysis (TGA), packed-bed, and interconnected circulating fluidised beds (ICFB) system.

Numerous oxides of metal elements from the periodic table were systematically investigated as potential oxygen carrier candidates for CLAS based on a thermodynamic
approach in the current study. The majority of the metal oxides exhibit the capability of releasing oxygen. However, most of them cannot be used in CLAS considering the oxygen equilibrium partial pressure (EPP) lower than 0.015, which may increase the use of sweep gas during the oxygen releasing process and correspondingly the energy consumption. Moreover, the likely carbonation and formation of hydrides on exposure to CO2/steam enriched conditions for metal oxides, such as the Ca and Na-based oxides, limit their application in the preparation of oxygen carriers. Only Mn3O4/Mn2O3, CoO/Co3O4, and Cu2O/CuO are found to be the most suitable oxidation pairs to transport oxygen in CLAS process. Furthermore, a special case for the integration of CLAS with oxy-fuel power plant, i.e., the direct use of clean CO2 flue gas as the sweep media during oxygen release for CLAS, was used for the operation optimisation based on these three systems. Results have shown CLAS has significant lower energy consumption for oxygen production, compared to the advanced cryogenic air separation.

Furthermore, oxygen carriers of Mn-, Co-, and Cu-based metal oxides with Al2O3 or SiO2 as a support were prepared by the dry impregnation method with an exception of the spray dried CuO/MgAl2O4. The reactivity of these prepared oxygen carriers was determined through TGA in the temperature range of 800-950°C. Among these studies, CuO/SiO2 and Co3O4/Al2O3 exhibit a very fast reaction rate in both oxidation and reduction processes. Co3O4/Al2O3 has shown a higher rate of oxygen transport (ROT) for the oxygen release than CuO/SiO2 at the same temperature, but a lower ROT of oxidation. It was also noted that the Mn and Co-based oxygen carriers on both Al2O3 and SiO2 have thermodynamic limitations in high temperature oxidation under air. The CuO/MgAl2O4 carrier was found to have a very stable performance and the highest oxygen transport capacity (OTC), however, very low mechanical strength after test, less than 0.8 N. Oxygen content attaining the equilibrium partial pressure was achieved from CuO/SiO2 and CuO/MgAl2O4 in a packed bed reactor at the temperature of 800, 850, 900, and 950°C.

The reduction and oxidation kinetics were determined for CuO/SiO2 and CuO/MgAl2O4. Avrami-Erofe'ev random nucleation and subsequence growth (A2) and phase boundary reaction (R2) mechanisms were found to fit the experimental results very well for both of these two oxygen carriers.
In response to the sinter issue of the mono-metallic CuO/SiO$_2$ oxygen carriers, attempts were made to add promoters to CuO/SiO$_2$ including MgO, Fe$_2$O$_3$, NiO, Mn$_2$O$_3$, and Co$_3$O$_4$. The results indicated that the agglomeration of CuO can be suppressed via the proper addition of secondary metal oxides, such as MgO, Fe$_2$O$_3$, NiO, and Co$_3$O$_4$. The best improvement was achieved by MgO with the added weight no less than 20.4 wt%.

In addition, an interconnected circulating fluidised beds (ICFB) system was designed, built, and operated for CLAS. The CLAS process has been successfully demonstrated using a Cu-Mg bimetallic oxygen carrier. Oxygen product at a level very close to the equilibrium concentration was continuously obtained in this rig.
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<th>Description</th>
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<tbody>
<tr>
<td>APP</td>
<td>Actual partial pressure</td>
</tr>
<tr>
<td>ASU</td>
<td>Air separation unit</td>
</tr>
<tr>
<td>BET</td>
<td>Brunauer-Emmett-Teller</td>
</tr>
<tr>
<td>CAR</td>
<td>Ceramic auto-thermal recovery</td>
</tr>
<tr>
<td>CAS</td>
<td>Cryogenic air separation</td>
</tr>
<tr>
<td>CCS</td>
<td>Carbon capture and storage</td>
</tr>
<tr>
<td>CLAS</td>
<td>Chemical looping air separation</td>
</tr>
<tr>
<td>CLC</td>
<td>Chemical looping combustion</td>
</tr>
<tr>
<td>CLOU</td>
<td>Chemical looping with oxygen uncoupling</td>
</tr>
<tr>
<td>DCFB</td>
<td>Dual circulating fluidised beds</td>
</tr>
<tr>
<td>EPP</td>
<td>Equilibrium partial pressure</td>
</tr>
<tr>
<td>HR</td>
<td>Heat recovery</td>
</tr>
<tr>
<td>ICFB</td>
<td>Interconnected circulating fluidised beds</td>
</tr>
<tr>
<td>ICP-OES</td>
<td>Inductively coupled plasma-optical emission spectrometer</td>
</tr>
<tr>
<td>ITM or OTM</td>
<td>Ion transport membrane</td>
</tr>
<tr>
<td>LNG</td>
<td>Liquid nature gas</td>
</tr>
<tr>
<td>Loss of OTC</td>
<td>Loss of oxygen transport capacity for oxygen carrier</td>
</tr>
<tr>
<td>OR</td>
<td>Oxidation reactor</td>
</tr>
<tr>
<td>OTC</td>
<td>Oxygen transport capacity for oxygen carrier</td>
</tr>
<tr>
<td>PSA</td>
<td>Pressure swing adsorption</td>
</tr>
<tr>
<td>ROT</td>
<td>Rate of oxygen transport for oxygen carrier</td>
</tr>
<tr>
<td>RR</td>
<td>Reduction reactor</td>
</tr>
<tr>
<td>SCM</td>
<td>Shrinking core model</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning electron microscopy</td>
</tr>
<tr>
<td>Abbreviations</td>
<td>Description</td>
</tr>
<tr>
<td>---------------</td>
<td>------------------------------</td>
</tr>
<tr>
<td>SLMs</td>
<td>Supported liquid membrane</td>
</tr>
<tr>
<td>sTPD</td>
<td>Standard ton per day</td>
</tr>
<tr>
<td>TGA</td>
<td>Thermogravimetric analyser</td>
</tr>
<tr>
<td>VSA</td>
<td>Vacuum swing adsorption</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
</tr>
</tbody>
</table>
Nomenclature

$A$ Pre-exponential factor, min$^{-1}$ (reduction), m$^{3/2}$ mol$^{-1/2}$ min$^{-1}$ (oxidation)

$A_1$ Coefficient for calculation of $C_p$, kJ mol$^{-1}$ K$^{-1}$

$Ar$ Archimedes number

$Au_{oxy-boiler}$ Auxiliary energy consumption for oxy-boiler

$B_1$ Coefficient for calculation of $C_p$, kJ mol$^{-1}$ K$^{-2}$

$C$ Oxygen mole concentration, mol m$^{-3}$

$C_1$ Coefficient for calculation of $C_p$, kJ mol$^{-1}$ K

$C_{eq}$ Oxygen equilibrium mole concentration, mol m$^{-3}$

$C_p$ Heat capacity at atmospheric pressure, kJ mol$^{-1}$ K$^{-1}$

$C_{p,air}$ Air heat capacity at atmospheric pressure, kJ mol$^{-1}$ K$^{-1}$

$C_{p,CO_2}$ CO$_2$ heat capacity at atmospheric pressure, kJ mol$^{-1}$ K$^{-1}$

$d_p$ Particle size, m

$d_p^*$ Dimensionless particle size

$D_1$ Coefficient for calculation of $C_p$, kJ mol$^{-1}$ K$^{-3}$

$E$ Activation energy, kJ mol$^{-1}$

$F_{blower}$ Power consumption for air blower, kW h

$g$ Gravitational acceleration, m s$^{-2}$

$G$ Gibbs free energy of chemical specie, kJ mol$^{-1}$

$G_p$ Gibbs free energy for product, kJ mol$^{-1}$

$G_r$ Gibbs free energy for reactant, kJ mol$^{-1}$

$G_s$ Solid circulation rate, kg min$^{-1}$

$H$ Enthalpy of chemical specie, kJ mol$^{-1}$

$H_{coal}$ Heat vale for coal, kJ kg$^{-1}$

$H_f(298.15)$ Standard enthalpy of formation at 298.15K, kJ mol$^{-1}$

$H_{tr}$ Enthalpy of transformation, kJ mol$^{-1}$
## Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td>Reaction rate constant, min$^{-1}$ (reduction) and m$^{3/2}$mol$^{-1/2}$min$^{-1}$ (oxidation)</td>
</tr>
<tr>
<td>$m_1$</td>
<td>Mass for density bottle loaded with water, kg</td>
</tr>
<tr>
<td>$m_2$</td>
<td>Mass for density bottle loaded with oxygen carrier and water, kg</td>
</tr>
<tr>
<td>$m_{air}$</td>
<td>Mass of air fed into CLAS unit, kg</td>
</tr>
<tr>
<td>$m_{CO_2}$</td>
<td>Mass of CO$_2$ required by CLAS unit, kg</td>
</tr>
<tr>
<td>$m_d$</td>
<td>Mass of density bottle, kg</td>
</tr>
<tr>
<td>$m_{O_2}$</td>
<td>Mass of oxygen recovered in CLAS unit, kg</td>
</tr>
<tr>
<td>$m_o$</td>
<td>Mass for oxygen carrier used in density bottle, kg</td>
</tr>
<tr>
<td>$m_{ox}$</td>
<td>Oxygen carrier mass at fully oxidised state, kg</td>
</tr>
<tr>
<td>$m_{red}$</td>
<td>Oxygen carrier mass at fully reduced state, kg</td>
</tr>
<tr>
<td>$m_t$</td>
<td>Oxygen carrier instantaneous mass during TGA test, kg</td>
</tr>
<tr>
<td>$m'_{O_2}$</td>
<td>Mass of oxygen consumed by coal, kg</td>
</tr>
<tr>
<td>$n_*$</td>
<td>Reaction order</td>
</tr>
<tr>
<td>$P$</td>
<td>Applied pressure for N$_2$ adsorption and desorption, Pa</td>
</tr>
<tr>
<td>$P_o$</td>
<td>Atmospheric pressure, Pa</td>
</tr>
<tr>
<td>$P_{O_2}$</td>
<td>Oxygen actual partial pressure</td>
</tr>
<tr>
<td>$P_{O_2,e}$</td>
<td>Oxygen equilibrium partial pressure</td>
</tr>
<tr>
<td>$Pr_{coal}$</td>
<td>Market price for coal, $\text{$ kg}^{-1}$</td>
</tr>
<tr>
<td>$Pr_e$</td>
<td>Market price for electricity, $\text{$ kg}^{-1}$</td>
</tr>
<tr>
<td>$R$</td>
<td>Constant of the ideal gases, kJ mol$^{-1}$ K$^{-1}$</td>
</tr>
<tr>
<td>$R_{air}$</td>
<td>Excessed air ratio for combustion</td>
</tr>
<tr>
<td>$Re$</td>
<td>Reynolds number</td>
</tr>
<tr>
<td>$Re_{mf}$</td>
<td>Reynolds number corresponding to minimum fluidisation</td>
</tr>
<tr>
<td>$S$</td>
<td>Entropy of chemical specie, kJ mol$^{-1}$ K$^{-1}$</td>
</tr>
<tr>
<td>$S(298.15)$</td>
<td>Standard entropy of chemical specie, kJ mol$^{-1}$ K$^{-1}$</td>
</tr>
<tr>
<td>$S_{BET}$</td>
<td>BET surface area, m$^2$ g$^{-1}$</td>
</tr>
<tr>
<td>$t$</td>
<td>Time, s</td>
</tr>
<tr>
<td>$T$</td>
<td>Absolute temperature, K</td>
</tr>
</tbody>
</table>
Nomenclature

\( T_{\text{air}} \) \hspace{1cm} Air inlet temperature, K
\( T_{\text{CO}_2} \) \hspace{1cm} Temperature for recycled \( \text{CO}_2 \), K
\( T_{tr} \) \hspace{1cm} Temperature for phase transformation, K
\( u \) \hspace{1cm} Superficial velocity, m s\(^{-1}\)
\( u_c \) \hspace{1cm} Superficial velocity limit toward turbulent fluidisation, m s\(^{-1}\)
\( u_{mf} \) \hspace{1cm} Minimum fluidisation velocity, m s\(^{-1}\)
\( u_t \) \hspace{1cm} Terminal velocity, m s\(^{-1}\)
\( U^* \) \hspace{1cm} Dimensionless velocity
\( U_t^* \) \hspace{1cm} Dimensionless terminal velocity
\( u_{se} \) \hspace{1cm} Superficial velocity limit toward fast fluidisation, m s\(^{-1}\)
\( V_a \) \hspace{1cm} Volume for the adsorbed \( \text{N}_2 \) at pressure of \( P \), m\(^3\)
\( V_d \) \hspace{1cm} Volume of density bottle, m\(^3\)
\( V_m \) \hspace{1cm} Volume for oxygen carries used in density bottle, m\(^3\)
\( V_s \) \hspace{1cm} Volume for the \( \text{N}_2 \) absorbed after the solid surface is covered by a monomolecular layer, m\(^3\)
\( \dot{V} \) \hspace{1cm} Volume flow rate of air, m\(^3\) s\(^{-1}\)
\( X \) \hspace{1cm} Oxygen carrier conversion

Greek letters

\( \alpha_{\text{ox}} \) \hspace{1cm} Oxygen carrier conversion during oxidation
\( \alpha_{\text{red}} \) \hspace{1cm} Oxygen carrier conversion during reduction
\( \Delta G \) \hspace{1cm} Variation of Gibbs free energy for chemical reaction, kJ mol\(^{-1}\)
\( \Delta G_{\text{oxidation}} \) \hspace{1cm} Variation of Gibbs free energy for oxidation reaction, kJ mol\(^{-1}\)
\( \Delta H \) \hspace{1cm} Variation of heat, kJ mol\(^{-1}\)
\( \Delta P_{\text{blower}} \) \hspace{1cm} Increase of air pressure via air blower, Pa
\( \Delta P_{or} \) \hspace{1cm} Oxygen partial pressure difference between oxygen EPP and APP in oxidation reactor
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta P_{rr}$</td>
<td>Oxygen partial pressure difference between oxygen EPP and APP in reduction reactor</td>
</tr>
<tr>
<td>$\Delta T$</td>
<td>Variation of temperature, K</td>
</tr>
<tr>
<td>$\eta_{\text{fan}}$</td>
<td>Fan efficiency</td>
</tr>
<tr>
<td>$\eta_{\text{motor}}$</td>
<td>Motor efficiency for air blower</td>
</tr>
<tr>
<td>$\eta_{\text{oxy-boiler}}$</td>
<td>Efficiency of oxy-boiler</td>
</tr>
<tr>
<td>$\eta_{\text{tr}}$</td>
<td>Electric transmission efficiency for air blower</td>
</tr>
<tr>
<td>$\mu_G$</td>
<td>Gas viscosity, kg m$^{-1}$s$^{-1}$</td>
</tr>
<tr>
<td>$\rho_m$</td>
<td>Real density for oxygen carrier, kg m$^{-3}$</td>
</tr>
<tr>
<td>$\rho_G$</td>
<td>Density of fluidising gas, kg m$^{-3}$</td>
</tr>
<tr>
<td>$\rho_l$</td>
<td>Density for water, kg m$^{-3}$</td>
</tr>
<tr>
<td>$\Phi$</td>
<td>Particle sphericity</td>
</tr>
</tbody>
</table>