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The relative performance of alternative oxygen carriers for liquid chemical looping combustion and gasification

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ABSTRACT

The relative performance of different potential liquid oxygen carriers within a novel system that can be configured for either chemical looping gasification or combustion is assessed. The parameters considered here are the melting temperature, the Gibbs free energy, reaction enthalpy, exergy and energy flows, syngas quality and temperature difference between the two reactors. Results show that lead, copper and antimony oxides are meritorious candidates for the proposed systems. Antimony oxide was found to offer strong potential for high quality syngas production because it has a reasonable oxygen mass ratio for gasification. A sufficiently low operating temperature to be compatible with concentrated solar thermal energy and a propensity to generate methane. In contrast, copper and lead oxides offer greater potential for liquid chemical looping combustion because they have higher oxygen mass ratio and a higher operating temperature, which enables better efficiency from a power plant. For all three metal oxides, the production of methane via the undesirable methanation reaction is less than 2% of the product gasses for all operating temperatures and an order of magnitude lower for lead.

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Introduction

Liquid Chemical Looping Gasification (LCLG) and liquid Chemical Looping Combustion (LCLC) are two recently proposed technologies to produce synthesis gas (syngas) and to provide integrated $CO₂$ capture from the combustion of a hydrocarbon fuel, respectively $[1-3]$ $[1-3]$ $[1-3]$. Chemical looping process using solid particles can also be used to produce hydrogen $[4-8]$ $[4-8]$. All chemical looping systems operate based via the indirect transfer of oxygen from the air to the fuel by means of a Oxygen Carrier (OC) that is cycled between two reactors, typically termed the air and fuel reactors [\[9\].](#page--1-0) However, for a LCLC system, the amount of oxygen transferred between the reduction and oxidation reactors with the LOC is significantly greater than the stoichiometric ratio, while for a LCLG system, it is sub-stoichiometric. Since the potential use of a liquid as the oxygen carrier has only recently begun to be explored, a wide range of potential LOC materials are yet to be assessed. The overall objective of the present investigation is therefore to assess the relative performance of different potential LOCs for applications in combustion or gasification.

Solid-phase CLG and CLC systems employ solid particles as the oxygen carrier. Although this has the advantage of enabling detailed control of the properties of the OC materials, it also introduces the challenges of attrition, agglomeration

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Liquid metal oxides have potential to be used as the OC for gasification process or hydrogen production $[18-20]$ $[18-20]$ $[18-20]$. For example, syngas is produced using liquid slag during the heat recovery process from high-temperature blast furnaces using liquid slag $[21-24]$ $[21-24]$. In this process, a carbonaceous fuel is oxidised (partially or totally) by injecting it into a high temperature molten slag. Extensive studies on this process $[18,25-29]$ $[18,25-29]$ $[18,25-29]$ reveal that it suffers from a low efficiency because of the low thermal conductivity of the slag (1 W/m.K for slag versus 120 W/m.K for molten copper oxide [\[30\]](#page--1-0), respectively), which is lowered by the presence of minerals. In addition, slag tends to form layers of fused materials of different composition (e.g. silica layer, minerals and non-metallic compounds) with relatively low thermal conductivity, which inhibit the thermal performance of the process [\[21,22\].](#page--1-0) However, pure liquid metals have a high thermal conductivity and can offer better chemical and thermal performance $[31-34]$ $[31-34]$ $[31-34]$ than molten slag. It has also been shown that the presence of molten metals can enhance the rate of reaction, the thermal performance $[32,35-40]$ $[32,35-40]$ $[32,35-40]$ and the chemical conversion of the reaction because they catalyse system $[41-43]$ $[41-43]$.

Recently, Jafarian et al. [\[17\]](#page--1-0) proposed one potential configuration in which to use a liquid metal oxides could be used as the oxygen carrier with the view to addressing the aforementioned challenges. They evaluated the thermodynamic potential of their cycle using molten iron oxide as a plausible oxygen carrier. This LOC offers the potential to achieve a high operating temperature of 1350 \degree C, although it suffers the disadvantage of a relatively high solidification temperature. They showed that the proposed system has significant potential to address major limitations of both solid-phase CLC systems and a liquid CLC system proposed by Lamond et al. $[44, 45]$ and McGlashan et al. $[46-48]$ $[46-48]$, which are temperature limited. Jafarian et al. [\[17\]](#page--1-0) also showed that it is also possible to configure the liquid chemical looping process to operate in either the CLC or CLG modes by controlling the stoichiometry of the reactor, which can be achieved by varying the relative molar flow rate of the LOC. However, no previous investigation has been reported of the potential for alternative types of liquid oxygen carrier for either CLC or CLG.

Although the influence of the type of solid oxygen carrier particles on the performance of chemical looping combustion has been assessed [\[49,50\]](#page--1-0), to the best of authors' knowledge, no previous assessment has been reported of the relative performance of alternative LOCs for combustion or gasification. Indeed, no criteria have been proposed with which to compare their performance. For this reason, the first aim of this work is to develop a set of thermodynamic criteria with which to assess the relative merit of alternative liquid oxygen carriers for chemical looping. The second is to assess the energetic performance of a particular configuration of reactors for liquid phase CLC and CLG.

Methodology

[Fig. 1](#page--1-0) presents a schematic representation of a potential system for liquid chemical looping combustion or gasification systems, as proposed by Jafarian et al. [\[17\]](#page--1-0). The main components of this system are two interconnected bubble column reactors, referred to as the fuel and air reactors. The former is used to reduce the LOC, by the oxidation of the fuel to produce syngas, while the latter is used to oxidize the LOC using oxygen from the air. During operation, the LOC is proposed to be circulated continuously between the reactors. The system is analysed here using graphite as a surrogate for a range of potential carbon containing feedstock together with steam as the gasifying agent. The assumed product of gasification is syngas whose dominant components are CO and H_2 . [Fig. 1](#page--1-0) also proposes an ash separator, which is reasonably assumed to be developable because of the significant difference in density between the molten metal oxides and fused ash [\[51,52\]](#page--1-0). The process of oxidation causes the reduced and vitiated air $(O₂ < 21%)$ to leave the oxidation reactor at an elevated temperature.

For the present analysis, it is assumed that the whole process is isothermal. This assumption is reasonable because the oxidation reaction, which is exothermic, can be used to supply the heat required for gasification, which is endothermic, via circulation of the LOC between the reactors. Furthermore, the circulation of heat between the reactors can be controlled via the flow rate of LOC and the partial pressure of oxygen. Isothermal operation also allows solidification to be avoided, which is necessary for reliable operation and is desirable to minimise the energy and exergy losses associated with heating and cooling of the reactants and products. Setting the circulation ratio between the reactors to be sub-stoichiometric will cause the system to operate in the gasification regime, while setting it to be at or above stoichiometric causes the system to operate in the combustion regime.

To assess the proposed system thermodynamically the Gibbs minimization method was employed to estimate the Gibbs free energy, enthalpy of reaction and oxygen content ratios. The total or partial oxidation of fuel and feedstock $(C_nH_{2m}O_p)$ with metal oxides shown as Me_aO_b was calculated as follows:

Combustion mode

In the fuel reactor:

$$
C_nH_{2m}O_p(g) + \left[\frac{m+2n-p}{b-ah/g}\right]Me_aO_b(l) \to \left[\frac{m+2n-p}{b-ah/g}\right] \frac{a}{g}Me_gO_h(l)
$$

+ $mH_2O(g) + nCO_2(g)$, (1)

In the air reactor:

$$
\left[\frac{m+2n-p}{b-ah/g}\right]Me_gO_h(l)+\frac{1}{2}[m+2n-p]O_2(g)\rightarrow \left[\frac{m+2n-p}{b-ah/g}\right]
$$

$$
\frac{a}{g}Me_aO_b(l),
$$

(2)

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