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Thermodynamic assessment and techno-economic analysis of a liquid indium-based chemical looping system for biomass gasification

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A R T I C L E   I N F O

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A B S T R A C T

A detailed thermochemical analysis is carried out to assess the energetic performance of a proposed process based on liquid metal slurry in a chemical looping gasification process. The system is designed to produce synthetic gas and generate electricity from low-grade (waste) solid carbon black collected from a thermal plasma plant. Indium oxide-indium slurry mixture was used as an oxygen carrier. The thermodynamic models showed that oxygen availability in the fuel reactor is the determining parameter that controls the operating mode of the system. The molar ratio of liquid metal to feedstock (LMO/C) and the steam to feedstock (S/C) are identified the key factors that regulate the level of exergy partitioned in the gas products. Generating steam by heat-recovery from the vitiated air (exhausted from the air reactor), is a proof that the process is partially self-sustained – capable of generating electricity to drive the pumps and the air compressors in the process. At LMO/C = 0.1 and S/C = 1.5, the largest exergy is partitioned in the synthetic gas and a syngas quality (molar ratio of H_2: CO) of ~1.55 is achieved. The highest syngas quality was achievable, however, at the cost of unreacted steam, which increased the exergy destruction of the plant. The peak performance of the system is achieved when the (fuel and air) reactors operated at near-isothermal conditions. At these conditions, the exergy destruction between reactors is minimised and the power production in the power block is maximised. Based on indicative available price indexes, a techno-economic analysis evaluated the economic viability and the levelised cost of energy for a different price for various scenarios. It showed that the proposed system offers a competitive LCOE against several existing energy and hydrogen production systems.

1. Introduction

As most of the human activities and industrial processing plants are partially or fully shut down during to the coronavirus (COVID-19) pandemic, a notable reduction in global emissions of CO_2 and other GHGs were reported [1]. This situation provided the scientific community with a unique insight and undeniable proof of the adverse impact of human-made environmental emissions and their carbon footprints on our planet. This outcome is consistent with the earlier recommendations of the Paris agreement in 2016. The census reached then was that burning fossil fuels using conventional combustion technology is not sustainable and hence, the focus should be redirected towards the development of new technologies and cleaner energy production systems to reduce the emission of GHGs [2] and the consumption of fossil fuels [3]. Despite promising results and successful demonstrations of several renewable energy plants, however, fossil fuel will remain the main source of energy for several decades [4]. Therefore, it is essential to continue the development of low-emission technologies that can be retrofitted into existing power systems and explore new chemical pathways to efficiently harness the potential of low-grade carbon-containing feedstock for energy production [5] and sustainable systems [6].

The main processes for converting a carbonaceous feedstock into value-added products and cleaner fuels are gasification and reforming of hydrocarbons, such as coal [7], biomass [8] or natural gas [9]. The product of these processes is a synthetic gas (referred to as syngas, which is a mixture of H_2, CO, CH_4 and a small amount of CO_2) that has relatively high heating values (LHV 18–21 MJ/kg) [10]. The conventional air gasification processes, however, inherently introduce nitrogen into the synthetic gas. Generally, the molar ratio of H_2: CO (syngas quality) determines the end-use application of the synthetic gas. Therefore, the presence of nitrogen in the mixture reduces the syngas quality and limits its end-use application. Similarly, in conventional gasification systems, a fraction of the feedstock is burnt to provide the required enthalpy to maintain specific reactor temperature for the gasification reactions to proceed. This in turn reduces the chemical and thermal efficiency of the...
achieved at 500 to 750 \degree C, followed by a series of solid reactions at T \degree C, which indicates that the percent purity of the gasified material is highly dependent on the temperature. In the gasification reactor, the carbonaceous fuel and/or feedstock reacts with a solid oxygen carrier (SOC) and absorbs the oxygen from the SOC stream. The reduced SOC is then fed into the air reactor where it regenerates through an oxidation reaction with air. The regenerated SOC is then fed back into the gasification reactor and the cycle is repeated. This process provides a suitable environment that separates the air and the solid carbon feedstock, hence preventing the nitrogen from diluting the composition of the syngas. Also, a large fraction of the thermal energy released in the air reactor (from the exothermic oxidation reaction, ΔH < 0) is transferred to the gasification reactor. That heat provides the energy required to support the (endothermic, ΔH > 0) gasification reactions, and therefore promoting fuel economy and improve the thermal efficiency of the system [12].

Chemical looping gasification is a flexible process that can be adapted to the type of feedstock. It can be employed for gasification of high-grade feedstock such as natural gas, or low-grade feedstock, such as pyrolysis oil, biomass, municipal waste and coal. As an example, Chein et al. [13] carried out thermodynamic modelling showing that gasification of biomass feedstock, such as Miscanthus or peach stone, can be achieved at 500 to 750 \degree C using CLG systems. However, the performance of the process was limited by the reactivity of CaO, which is reduced by increasing the temperature of the gasification reactor [13]. A thermogravimetric analysis was conducted by Yan et al. [14] to investigate the chemical looping gasification process for corn straw using iron-based oxygen carrier. They noticed that the process includes multiple stages of pyrolysis. The first occurs at T < 500 \degree C, followed by a series of solid–gas and solid–solid reactions at T < 1100 \degree C, which indicates that the performance of the CLG operating with iron particles is controlled by the kinetics of the reactions in different stages. As solid–solid reactions are relatively slow, the chemical performance of the system is limited by the reactivity and the kinetics of the reaction between solid particles. Wu et al. [15] showed that bimetallic solid oxygen carriers are potential candidates for lignin gasification. They considered CaFe_2O_4, CaMn_2O_4, BaFe_2O_4 and BaMn_2O_4 and demonstrated that these solid oxygen carriers have good regeneration capability and are selective to produce CO and H_2. However, these oxygen carriers required complex material processes to be developed and used in the CLG systems. Similar experiments were conducted by He et al. [16] using NiFe_2O_4 as an oxygen carrier for biomass gasification and reported carbon conversion of 92.5%.

In other studies, calcium and calcium oxides were identified as a potential oxygen carrier for CLG systems. These materials can be cost-effective if used for large-scale production of H_2-enriched syngas. Due to its catalytic effect, CaO can suppress the chemical activation barrier in the reduction–oxidation reactions [17–19]. However, the deactivation of the CaO particles over a continuous operation remains a critical limitation and challenge that requires further research [20].

Carbon particles deposition on the oxygen carrier is another challenge associated with the use of solid oxygen carriers in CLG processes [21]. Carbon particles can cover the reaction sites on the external layer of the SOC and can also be carried over into the air reactor, hence producing CO_2 and CO gases that are released to the environment. Similar findings have been reported for other solid oxygen carriers such as iron and copper oxide. Consequently, the performance of a CLG process is limited by the reactivity of the oxygen carrier. Apart from the chemical structure of the oxygen carrier, controlling the operating temperature of CLG systems is also technically challenging. For example, the oxidation reaction in the air reactor generates a large amount of heat, which in turn increases the temperature of the reactor. Despite great thermal properties and heat transfer characteristics of micron-sized solid particles [22] and nano-particles [23], particularly in convective heat transfer applications [24], at high reactor temperatures, the structure and morphology of the solid particles changes resulting in the reduction of the reactivity of the oxygen carrier. Similarly, the temperature of the gasification reactor should be high enough to drive the hydrogasification reaction [25]. This can potentially introduce other challenges such as agglomeration, sintering and breakage of the solid oxygen carriers [26,27] and reduced the thermal conductivity of the solid particle which is the key parameter affecting the thermal performance of the system [28].

To address the challenges of CLG associated with utilising SOC, it is proposed to replace the SOC with oxygen carriers in liquid, eutectic or slurry phases with low vapour pressure (high boiling points). Liquid metals are potential candidates that offer a reasonably high thermal conductivity and the capacity for cyclic reduction–oxidation, which are aligned with the nature of CLG operations. Liquid metals can successively be reduced and oxidised without any changes in their metallic structure once impurities (such as induced heavy metals, sulphur or carbon) are separated from the bulk of the metal. Liquid metal oxides with high density (e.g. 6500 kg.m^-3 for copper oxide or 5900 kg.m^-3 for gallium [29]) can be separated from ash and any impurity using conventional filters and separators such as those used in blast furnaces.
Any carry-over particles due to the entrainment of the gas in the liquid metal can also be avoided using sparging and degassing systems for liquid metals [32].

In a CLG process with liquid metal oxides (LMO) carriers, the melting temperature of the liquid metal is the dominant factor. This explains why some metal oxides are impractical for such a process due to their relatively high melting temperatures, for example, Cr₂O₃ has a melting temperature of 2500 °C [33]. Considering this limiting criterion, the list of potential metal oxides candidates is narrowed down to a few metals. One of these candidates is indium oxide. It has a negative Gibbs free energy (ΔG < 0), a relatively low melting temperature of 1910 °C, and can be used in a slurry phase (in an oxygen lean environment). Once all oxygen in the LMO carrier is consumed in the gasification reactor, the slurry phase is converted to a pure liquid phase of indium, which has a melting temperature ~157 °C. The pure liquid indium forms the base fluid and also the medium for the dispersion of indium oxide particles.

In a thermodynamic system consisting of carbon, indium and indium oxide, the Gibbs free energy of the reaction between carbon and indium oxides and that between pure indium and oxygen, is negative. This implies that the reduction and oxidation reactions are feasible and spontaneous. Carbon feedstock is the main by-product of thermal plasma systems and pigment industry. It is usually dumped or mixed with soil to maintain the moisture in the soil or to preserve its structure to reduce environmental pollution [34]. Carbon can also be activated using steam for use in water treatment and filtration systems. However, that carbon ultimately ends up in a combustion process through waste management systems, hence adding to the GHG emissions [35].

The objective of this study is to develop and analyse a thermodynamic model for a chemical looping gasification process using liquid indium slurry, and waste carbon-black feedstock to produce high-quality synthetic gas. The thermodynamic performance of the proposed system is evaluated, and a techno-economic assessment is conducted to calculate the levelised cost of energy (LCOE) of the system. Harnessing the potentials of the proposed system can create a new business opportunity for biomass and landfill waste gasification, hence contributing to the mitigation of global warming and environmental protection measures.

### 2. Conceptual design

A conceptual design of the proposed chemical looping gasification unit based on liquid metal oxides carriers is shown in Fig. 1. The system includes also a heat recovery unit for steam production and a power block unit for power generation. The chemical looping unit consists of a fuel reactor (FR) and an air reactor (AR). The indium oxide/indium liquid metal slurry acts as the oxygen carrier and also provides the medium for heat and mass transfer between the reactants and across the reactors. In the fuel reactor, the carbon-containing feedstock and steam react to reduce the liquid metal oxide slurry of the indium oxide (In₂O₃) to a pure liquid Indium (In). This reaction is endothermic requiring thermal energy to proceed to completion. Depending on the amount of oxygen available in the fuel reactor, either a partial or a complete oxidation reaction can be achieved, which effectively controls the operating mode of the system. If sufficient oxygen is available in the fuel reactor, the oxidation reaction proceeds to completion and the final product of the system is carbon dioxide (CO₂), water, and heat (combustion mode). However, if the quantity of oxygen is sub-stoichiometric, a partial oxidation reaction occurs resulting in the production of synthetic fuel (gasification mode). The former operating mode is suitable for on-site electrical power generation. The gasification mode is suitable for producing transportable combustible gaseous fuels that can be used elsewhere for high-temperature applications (1500–2000 °C [36]) or in a Fischer-Tropsch process [37].

The focus of the present analysis is the development of CLG-LMOC process, and therefore, the oxygen concentration in the slurry (in the fuel reactor) should be sub-stoichiometric. The reduced pure liquid indium in the fuel reactor is then fed back into the air reactor where it reacts with air to form indium oxide slurry via an exothermic reaction. A fraction of the generated heat is transferred to the fuel reactor through the sensible heat transfer of the liquid metal slurry, which in turn reduces the thermal load requirement of the fuel reactor. The exhaust gas of the air reactor is predominantly “vitiated air”; a mixture of oxygen-depleted, nitrogen-enriched gas with relatively large specific heat capacity ratio. The emitted vitiated air provides suitable thermodynamic conditions for use in gas-turbine power plants directly, or for steam production in heat recovery steam generators. However, since the

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**Fig. 1.** Schematic illustration of the proposed process for co-production of electricity and syngas using chemical looping gasification with liquid metal slurry.
operating temperature of the fuel and air reactors are \(<1000\, ^\circ\text{C}\), a steam turbine application is more suitable than a gas turbine to avoid installing a burner to increase the vitiated air temperature to \(~1200\, ^\circ\text{C}\) [38].

The generated electricity from power block can be used to drive the pumps and compressors, hence increasing the overall thermal efficiency and sustainability of the process. There also is an option to use an electric furnace or an electrical inductive heater for melting the slurry in the fuel reactor. The generated electricity can be also dedicated to

Fig. 2. Detailed schematic diagram of the proposed CLG-LMOC process for co-production of syngas and electricity, a) detailed schematic process flow diagram of the system, b) schematic representation of the model developed using Aspen Plus software package.
domestic use or fed into the grid network. However, a detailed assessment of the power cycle and end-use of electricity is beyond the scope of the present study. It is worth noting that steam bleeding from the power cycle cannot be used for preheating the feedstock or providing the steam for the fuel reactor. This is because the amount of steam generated in the power block unit does not match the required steam for preheating and/or gasification. Also, the quality of steam is low (<1) creating a potential hazard of freezing the liquid metals in the injection points.

3. Process description and modelling

Fig. 2 represents the schematic process flow diagram that was developed and modelled for the proposed CLG-LMOC system. Carbon is selected as a generic representative of feedstock because it has simple structure, relatively moderate heating value (LHV of 32.08 MJ/kg versus 50 MJ/kg for CH₄) and contains less impurity than complex feedstock structure. Worth noting that using full biomass composition, rather than a pure carbon, would require adjusting the ratios of the various fluids in the system to compensate for the change in oxygen concentration. In previous studies [39,40], it was demonstrated that the gasification of biomass using liquid metals is feasible. For example, in the HydroMax project (HyMelt) [41], molten metals such as iron were used to produce hydrogen-enriched synthetic fuel. It was also found that depending on the level of moisture, hydrogen, carbon and oxygen, the molar ratio of steam to carbon and liquid metal oxide to carbon must be tuned to compensate for the lack of oxygen. Also, the moisture in the biomass was found to be released at 300–400 °C, which in turn can contribute to the reduction of steam consumption of the system and associated cost with steam production. However, the technology readiness level of the proposed system is low and there is room for further investigation on the effect of biomass impurity on the performance of the system.

Pure carbon is an abundant by-product (waste) of several processes, such as thermal plasma, methane cracking and reforming. Hence, the proposed system can contribute to a reduction in the carbon footprint of these processes by eliminating carbon black waste and converting it to value-added products such as synthetic gas. In this process (Fig. 2a and b), carbon feedstock, air, water, and liquid indium metal oxide streams are preheated to 300 °C using external heat exchangers. The inlet temperature of the carbon should be kept below 400 °C to avoid partial/complete combustion. The preheating stage reduces the heating load in the units and adds thermal penalty (Q_{HEX,a}) that should be included in the analysis. The outlet gases from the fuel reactor are quenched and compressed before being fed into storage units. In the present work, the gases are cooled to 25 °C. The outlet gases from the air reactor are fed into a steam generator unit to produce dry steam at 470 °C and 60 bars to drive the steam turbine. To produce the high-pressure steam, a pump with a driver efficiency of 0.75 and mechanical efficiency of 0.8 is used in the analysis. The energy required for the pumps and compressors is provided by the electricity generated from the steam turbine, hence the power block unit is self-sustained. Notably, the red arrows show the potential operating units that can consume electricity produced with the power block.

The enthalpy of reaction (ΔH_{rea}) and the change in the Gibbs free energy (ΔG_{rea}) of the reactions in the process are calculated using the following equation:

$$\Delta H_{rea} = \sum_{prod} \Delta H_f^{\text{prod}}(T) - \sum_{react} \Delta H_f^{\text{react}}(T).$$  \hspace{1cm} (1)

where ΔH_f represents either the enthalpy of reaction (ΔH) or the change in the Gibbs free energy of the reaction (ΔG) for specie i. The subscripts “rea”, “react” and “prod” refer to reaction, reactants and products of each reactor, respectively. To evaluate the exergy partitioning in synthetic gas, an exergy efficiency (\(\chi\)) is defined:

$$\chi = \frac{n_{\text{H}_2} \times \Delta H_{\text{H}_2} + n_{\text{CO}} \times \Delta H_{\text{CO}}}{n_{\text{feed}} \times \Delta H_{\text{feed}}} \hspace{1cm} (2)$$

In this equation, \(n\) is the molar flow rate of each stream, \(\Delta H\) is the lower heating value of the reactive species (10.1 MJ/kg for CO and 119.9 MJ/kg for H₂). The total thermodynamic efficiency of the process is defined as follows:

$$\eta_{\text{th}} = \frac{\sum_{i=1}^{n} n_i \times \Delta H_{\text{prod}}}{\sum Q_{\text{net}}, + \sum_{i=1}^{n} n_i \times \Delta H_{\text{rea}}}_{\text{in}}$$ \hspace{1cm} (3)

Here, \(W_{\text{power,block}}\) is the thermodynamic work produced by the steam turbine, and \(W_{\text{pump}}\) is the energy consumed by the pump to produce the high-pressure water stream for the power block system. \(Q_{\text{loss}}\) is the heat loss to the environment, which based on literature is assumed to be ~3% of total heat input [42]. The partitioning in the power block, defined in Eq. (4), represents the fraction of the energy converted into electricity in the power block unit:

$$\eta_{\text{elec.}} = \frac{W_{\text{elec.}}}{n \times \Delta H_{\text{H}_2}} \hspace{1cm} (4)$$

In this equation, \(W\) is energy, and “FR” and “elec.” stand for fuel reactor and electricity, respectively. The total energy in the syngas is calculated as:

$$E_{\chi} = n_{\text{H}_2} \times \text{LHV}_{\text{H}_2} + n_{\text{CO}} \times \text{LHV}_{\text{CO}}$$ \hspace{1cm} (5)

Here, \(n\) is the molar flow rate of each stream.

To evaluate the economic viability of the process, a techno-economic assessment is also carried out using the methodology proposed in "Australian Liquid Fuel Techno-Economic Assessment (ALFTA)" [43]. In the present study, two competitive reference cases were considered; a conventional reforming plant for hydrogen production, and the CSIRO membrane system [44]. The techno-economic assessment accounts for the capital cost, operating and maintenance cost (O&M) – assumed to be 2.5% of total capital cost, and insurance at 1% of total capital cost. The levelised cost of energy (LCOE), is calculated using the following equation:

$$\text{LCOE} = \frac{\sum_{i=1}^{n} \frac{l_i M_i P_i}{(1+r)^{t}}}{(1+r)^{t}/(1+r)^{t}} \hspace{1cm} (6)$$

In this equation, \(l_i\) is the capital cost of all plant equipment in a year, \(t\) ($), \(M_i\) is the maintenance and operation cost in a year, \(t\) ($), \(P_i\) is the cost of fuel spent in year \(t\) ($), \(E_i\) is the energy produced by the system in year \(t\) (GJ); \(r\) is the discount rate (%), \(n\) is the lifetime of the system plus the construction period (Amortisation period of 23 or 30 years) [45]. To calculate the indicative price of the system units and capital costs associated with the plant, the following equation is used:

$$C_i = C_{0i} \left( \frac{S_i}{S_o} \right)^r \hspace{1cm} (7)$$

In Eq. (7), \(C_{0i}\) is the available cost for capacity \(S_o\), while \(C_i\) is the calculated cost for capacity \(S_i\). \(R\) is a scaling exponent, which is different for each operating unit. The values associated with natural gas consumption, the operating cost of the CSIRO membrane system, and a conventional reforming plant can be found in Ref. [44]. The values for insurance, operating cost were estimated using the values suggested by Hinkley et al. [45,46] and [43]. Table 1 shows the assumptions made for technoeconomic modelling of the process.

4. Thermodynamic stability and phase diagram

Fig. 3 shows the calculated stability diagram for the indium-based slurry mixture. The figure identifies the most stable phase(s) at various partial pressures of oxygen and operating temperatures. Within
the proposed CLG operating temperature range of 300 to 1000 °C, the stable components of the slurry are indium oxide (In$_2$O$_3$) and pure indium (In). However, in the fuel reactor, by depleting the oxygen due to the reaction with carbon, the In$_2$O$_3$ is converted back into a pure In. Once all the oxygen in the reactor is consumed, the indium oxide–to-indium reaction proceeds to equilibrium such that indium becomes the dominant product in the reactor. This demonstrates the feasibility of using In$_2$O$_3$/In slurry as a liquid metal oxide carrier in the CLG process.

Table 2 lists the reactions between liquid metal oxide slurry and the carbon feedstock in the fuel reactor (R$_1$ and R$_2$). It also shows the reaction between pure liquid indium and oxygen in the air reactor (R$_3$). In this table, $T_{\text{rxn, min}}$ is the minimum operating temperature for the reactions to occur. At $T < T_{\text{rxn, min}}$, the reactions are terminated as the Gibbs free energy of the reactions become positive. The phase diagram of indium oxide-indium-oxygen system (Fig. 4) identifies the thermodynamic state and phase state of the system. In the fuel reactor where the temperature range is between 200 °C and 1000 °C, the initial state is a solid–liquid phase (slurry) with indium oxide as the dominant component. By decreasing the oxygen content, the composition of the slurry changes such that the quantity of indium oxide decreases as it is converted into indium, reaching a pure indium state once oxygen in the slurry is fully released. This released oxygen is fully absorbed by carbon feedstock. At the endpoint (air reactor), the dominant product is pure indium, which can react with air. By oxidising the indium, the quantity of oxygen increases such that the phase point is shifted towards the re-formation of the slurry (pure indium/oxide indium). Hence, from the thermodynamic aspect, the process is feasible within the selected region on the phase diagram. This outcome is consistent and aligns well with the stability diagram shown in Fig. 3.

5. Results and discussion

A series of sensitivity analysis is conducted to evaluate the thermodynamic performance of the proposed system against key operating

Table 2

| No. | Reaction | $\Delta G_{\text{rxn}}$ (kJ/mol) | $\Delta H_{\text{rxn}}$ (kJ/mol) | $T_{\text{rxn, min}}$ (K) |
|-----|----------|-------------------------------|-------------------------------|--------------------------|
| R1  | In$_2$O$_3$(l) + 2C(s) + H$_2$O(g) → 2In(l) + 2CO$_2$(g) + H$_2$(g) | -3.8 | 306.1 | 550 |
| R2  | In$_2$O$_3$(l) + 4C(s) + H$_2$O(g) → 2In(l) + 4CO(g) + H$_2$(g) | 38.9 | 642.8 | 550 |
| R3  | 2In(l) + 1.5O$_2$(g) → In$_2$O$_3$(Slurry) | -671 | -923 | 250 |

Reactions (1) and (2) occur in the fuel reactor and they are endothermic, reaction (3) is exothermic and occurs in the air reactor. To prevent solidification of liquid metals, a $\Delta T_{\text{buffer}} = 50$ °C was applied to all reactors. A pinch point of 20 °C was also considered for heat exchangers.

Fig. 4. The proposed CLG-LMOC process shown in the phase diagram of the In-O system at various concentrations of oxygen and temperature (phase diagram). Adapted from [47,48]
parameters, such as the reactor temperature, the molar ratios of liquid metal oxide slurry to carbon, and steam to carbon.

5.1. Gibbs free energy and enthalpy of reaction

Although the focus in this study is mainly on gasification mode, the inclusion of combustion mode analysis provides a reference point for comparison and also for the completeness of the presentation. Fig. 5 represents the variation of the calculated Gibbs free energy and enthalpy of the reaction for complete oxidation of feedstock in the fuel reactor. As shown, with increasing the operating temperature of the fuel reactor, the magnitude of the Gibbs free energy of the reaction ($\Delta G_{rxn}$) increases linearly indicating that the reaction is spontaneous. For example, at $T = 550^\circ$C, $\Delta G_{rxn} = -3$ kJ/mol, but reaches $-141$ kJ/mol at $1000^\circ$C. Notably, for positive values of the Gibbs free energy, the reaction does not occur in conventional reactors, thereby requiring active techniques such as a thermal plasma or photocatalytic reactors to shift the equilibrium point and the $\Delta G_{rxn}$ value from the positive to the negative region. Hence, for the fuel reactor, the minimum operating temperature is $550^\circ$C, including a $50^\circ$C temperature buffer.

Interestingly, the enthalpy of reaction ($\Delta H_{rxn}$) is $>0$ within the temperature range of $500$–$1000$ °C showing the need for an external energy source for the reaction(s) to proceed to completion. The magnitude of $\Delta H_{rxn}$ decreases with the increase in the operating temperature of the reactor, such that at high temperatures, the energy dependence of the reactor decreases. For example, by increasing the reactor temperature from $500^\circ$C to $1000^\circ$C, the required energy decreases by $\sim 14\%$ from $330$ kJ/mol to $285$ kJ/mol, respectively. Fig. 6, shows that $\Delta G_{rxn}$ for the partial oxidation of feedstock is negative at $T > \sim 650^\circ$C indicating that partial oxidation of carbon feedstock is feasible over an endothermic reaction with $\Delta H_{rxn} = +645$ kJ/mol. Again, by increasing the operating temperature of the reactor, $\Delta G_{rxn}$ is intensified and the thermal energy demand of the reactor is reduced. For example, a $4\%$ decrease in $\Delta H_{rxn}$ is observed by increasing the temperature from $650^\circ$C to $1000^\circ$C. For the air reactor, the variation in $\Delta G_{rxn}$ and $\Delta H_{rxn}$ with the temperature is shown in Fig. 7. In this case, for reactor temperatures $>200^\circ$C, the oxidation of indium with air is feasible and it is exothermic, as indicated by the negative values of $\Delta G_{rxn}$ and $\Delta H_{rxn}$, respectively.

The reactor temperature also controls the level of exergy transported to the power block unit and the exergy loss due to the temperature difference between the air and fuel reactors. Therefore, the temperature difference between the reactors should be minimised to decrease the exergy loss, and hence, a near-isothermal operation is favourable. However, from a practical view, achieving isothermal conditions is technically challenging and needs further research.

5.2. Equilibrium constant

So far it has been shown that the reactions are sensitive to the operating temperature of the reactors. A change in the temperature can shift the equilibrium state from a feasible to unfeasible operating region. Therefore, the equilibrium constant is another parameter that can be used to further evaluate the performance of the system at different temperatures. A lower equilibrium constant ($K_{eq}$) shifts the operating point into the unfeasible operating region. In the fuel reactor, Fig. 8, at $T > 500^\circ$C, the equilibrium constant for both partial ($K_{eq, CO}$) and complete oxidation ($K_{eq, CO_{2}}$) is $>1$, which thermodynamically indicates the plausibility for the reactions to proceed towards an equilibrium state.

It is worth pointing out (Fig. 8) that the equilibrium constant for complete oxidation is always larger than that for partial oxidation (at any temperature $>500^\circ$C). This is probably due to the higher heat released by the exothermic reaction in the air reactor, which accelerates the rate of reactions.

5.3. Syngas quality ($H_2$: $CO$)

The variation of the syngas quality (molar ratio of $H_2$: $CO$) against the reactor temperature is shown in Fig. 9. For a syngas quality $>2$, the
Energy conversion and management is suitable for cleaner applications such as alcohol production. However, for syngas quality $\leq 1$, the syngas can be used only as fuel for combustion systems and propulsion applications. The advantage of using syngas in combustion systems is that a higher combustion temperature increases the thermal efficiency for many systems such as IC engines, rockets and burners. It is also worth mentioning that the composition of syngas includes a small amount of CO$_2$. However, if biomass is used as a feedstock, the CO$_2$ is regarded as a green (recycled) emission and does not impact the sustainability and environmental friendliness of the process.

The molar ratio of CO: CO$_2$ is another index for identifying the operating mode of the process. If the ratio is small, for example, 2 at 1000 °C, the system is operating in gasification mode, however, for the same temperature, and ratio $> 6$, the system is a combustion unit. This parameter is strongly controlled by the amount of oxygen in the fuel reactor. Thus, to assess the thermal performance of the system, both molar ratios of H$_2$: CO and CO: CO$_2$ should be evaluated. As shown in Fig. 9a, by increasing the operating temperature of the fuel reactor, the syngas quality decreases nonlinearly. For example, at $T = 550$ °C and molar ratio of steam to carbon (S/C) = 1.0, the H$_2$: CO ratio is 5.8, reducing to 0.9 at 1000 °C. This can be attributed to the suppression of the Boudouard reaction, which leads to an increase in the CO concentration (also increases the CO: CO$_2$ ratio) and a considerable decrease in the syngas quality. As represented in Fig. 9b, by increasing the S/C ratio, the molar ratio of H$_2$: CO increases. For example, at $T = 800$ °C and S/C = 1.0, the syngas quality is ~0.9, increasing to 1.55 at S/C = 2. However, this comes at the expense of producing wet syngas and unreacted steam at the outlet, which requires a robust condenser or an efficient cooling system to recover the heat and water. Also, increasing the S/C ratio requires more steam to be generated, hence adding to the thermal penalty in the system. As the quality of syngas increases, the LHV of the syngas is increased due to the presence of more hydrogen in the product gas.

The molar ratio of liquid metal oxide to carbon (LMO/C) is another key parameter (Fig. 10) influences the performance of the system. By increasing the LMO/C ratio from 0.1 to 1, for example, the operating mode of the system changes from gasification to a combustion unit. In the gasification regime, the available oxygen in the fuel reactor is relatively low and is quickly consumed by a large amount of carbon-containing feedstock. Hence, the products are hydrogen and carbon monoxide. As an example, at 1000 °C, for LMO/C ratios between 0.1 and 0.5, the syngas quality is ~0.9. However, by increasing the LMO/C ratio to 1.0, the syngas quality drops to ~0.5, hence limiting it to combustion applications. At this condition, the LHV of the syngas is suppressed. Likewise, as shown in Fig. 10b, the molar ratio of LMO/C controls the operating mode of the system. Therefore, depending on the requirements of the end-user, the process can be adjusted to operate as combustion, gasification or a combination of both operating modes.

5.4. Exergy transported in syngas

Fig. 11 shows the variation of the total exergy partitioned in the syngas with temperature and molar ratio of steam to feedstock (S/C). The largest portion of exergy is transported in the syngas at a temperature range 500 °C < T < 600 °C, and gradually decreases as the temperature, or the S/C molar ratio increases. A rise in the reactor temperature increases the production of CO and suppresses the
The variation of the exergy transported in the syngas with operating temperature and molar ratio of LMO/C is depicted in Fig. 12. In combination with the results from Figs. 9 to 11, shows that the largest exergy can be transported in the syngas occurs at 500 °C < T < 600 °C and LMO/C < 0.2. For example, at T = 600 °C and LMO/C = 0.1, the LHV is 8900 kJ/mol, decreasing to 2300 kJ/mol for the same temperature and LMO/C = 1.0. That is, reducing the temperature of the fuel reactor can decrease the thermal efficiency of the system. It can also increase the risk of solidification and freezing of the liquid metal oxide slurry.

Besides, the temperature of the air reactor controls the level of exergy transported to the power block. This is because the steam generation unit is connected to the vitiated air originating from the air reactor. Hence, there is a complex trade-off between the quality of syngas, operating temperature and electricity production in the power block. A lower temperature of the fuel reactor leads to a higher quality of the syngas. However, due to the oxidation reaction, the temperature of the air reactor is relatively high. As a result, the temperature of the fuel reactor must also be kept close to that of the air reactor to minimise the temperature difference between reactors, hence resulting in the reduction in the quality of syngas.

5.5. Energetic performance

Fig. 13 represents the variation of the exergy partitioning in the proposed system with the temperature of the air reactor. The figure shows that an increase in the temperature of the air reactor leads to an increase in the exergy fraction dedicated to the power block unit. For fuel reactor temperature of 500 °C, the fraction of exergy transported in the syngas is 0.74, which is the highest value, but it decreases to 0.58 at 1000 °C. However, increasing the temperature of the fuel reactor, the exergy fraction in the syngas decreases. Hence, depending on the end-use application, the temperature of the fuel and air reactor should be regulated accordingly.

The highest exergy partitioned in the syngas is 0.74, which is achieved for reactors temperatures (T_A and T_FR) of ~500 °C, corresponding to a minimum fraction of exergy destruction of 0.18. The rest of the exergy is transported to the power block. At 500 °C, the exergy fraction partitioned in the power block is 0.07, increasing to 0.12 at 1000 °C. This is a clear indication that the temperature of the air reactor is the controlling parameter for the exergy partitioning in the power block system.

5.6. Indicative techno-economic analysis

For the techno-economic assessment, the system is evaluated for processing 110 tonnes per day of carbon feedstock. The operating conditions of the process were regulated for quality of synthetic gas of around 0.5. This corresponds to an S/C = 2 at T_FR = 600 °C, and T_AR = 800 °C, generating 1.67 MW via the power block and releasing 500 kg/h of green CO₂ through syngas line at an LMO/C = 0.1. Table 3 represents the flow rates, temperatures and composition of the main streams of the process for the nominated operating conditions. Several models were developed to assess the potential effect of the equation of state (EoS) and “activity models” on the outcomes. It was found that the process model is reliable and only a deviation of ±5.1% is observed between the results obtained for each EoS simulation. This deviation is attributed to the Vapour-Liquid Equilibrium modelling (VLE), and also the vapour pressure of the indium oxide. Since, indium oxide and indium do not evaporate at T < 1000 °C, the Antoine partial pressure constants for the liquid metals were set to zero, thereby enabling faster convergence of the models.

In Table 4, the calculated indicative costs associated with the capital expenditure of the operating units are presented. The largest calculated cost is associated with the containment, storage and handling of liquid metals. However, it is expected that developing new technologies and
evaluation to justify the feasibility, competitiveness and economic viability of the process. LCOE estimation. Nonetheless, this assessment provides a reasonable water for steam production in the process and also the power block plant based on the data provided in the literature [50].

In systems compatible with high-temperatures and liquid metals, the total capital cost will decrease over time. It should be emphasised that this estimation of the capital cost is based on existing technologies and prices available in the literature for other systems, considering the proposed scenario based on biomass feedstock were defined to assess the economic sensitivity to the price of the feedstock. Therefore, different price scenarios were performed in the techno-economic analysis. For the assessed case, the higher cost of the biomass is considered as 0.6% of the total capital cost, which becomes 1.02% of the total cost of the process. Insurance is considered as 1% of the total capital cost. Air compressor is the least expensive unit employed and are cost-effective in terms of overhaul and maintenance. Air compressors are assumed to be gas to gas radiators which are cheap to be employed and are cost-effective in terms of overhaul and maintenance.

The air and fuel reactors, including design and installation, constitute 49% of the total cost. Apart from that, trace heating of liquid metal might be required in cold locations, which is ignored in this assessment. The air and fuel reactors, including design and installation, are the second most expensive items costing 18% of total capital cost, followed by scrubber and syngas cleaning system that costs 6% of total capital cost. Air compressor is the least expensive unit <1% and heat exchangers are assumed to be gas to gas radiators which are cheap to be employed and are cost-effective in terms of overhaul and maintenance.

To perform a back-to-back comparison, natural gas was considered as a carbon-containing feedstock with a projected cost of 6.5 $/GJ. As shown in Fig. 15, annually, about 57.51% of total expenditure is spent on fuel, while ~40% of the total expenditure is dedicated to the equipment. The O&M cost is normally considered as 2.5% of the capital cost, which becomes 1.02% of the total cost of the process. Insurance is also considered as 1% of the total capital cost which is 0.06% of total capital and O&M cost. Hence, the final price of the synthetic gas is sensitive to the price of the feedstock. Therefore, different price scenarios based on biomass feedstock were defined to assess the economic viability of the process. Fig. 14 represents the price breakdown of the proposed CLG-LMOC system for producing synthetic gas from carbon as a surrogate for more realistic fuel.

Fig. 14. A price breakdown of the proposed CLG-LMOC system for producing synthetic gas from carbon as a surrogate for more realistic fuel.

### Table 3

| Stream Name | Units | AIR | COLD-SYN | CARB-P | FEED | HOT-SYN | INDIUM | LM-RED | LM-OX | STEAM | TO-TP | VITI-AIR |
|-------------|-------|-----|---------|--------|------|---------|--------|--------|-------|-------|-------|---------|
| Temperature | °C    | 25  | 25      | 600    | 25   | 25      | 600    | 25     | 600   | 800   | 25    | 25     |
| Pressure    | bar   | 1   | 1       | 1      | 1    | 1       | 1      | 1      | 1     | 1     | 1     | 1      |
| Mole Flows  | kmol/h| 3.4 | 2.31    | 68.6   | 100  | 231     | 10     | 20.8   | 0.4   | 200   | 68.6  | 22.8   |
| In          | kmol/h| 0   | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 0     | 0     | 0      |
| InO₂        | kmol/h| 0   | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 0     | 0     | 0      |
| C           | kmol/h| 0   | 0       | 0      | 68.6 | 100     | 0      | 0      | 0     | 0     | 68.6  | 0      |
| H₂O         | kmol/h| 0   | 0       | 0      | 0    | 189     | 0      | 0      | 0     | 0     | 200   | 0      |
| CO          | kmol/h| 0   | 0       | 0      | 0    | 20.8    | 0      | 0      | 0     | 0     | 0     | 0      |
| CO₂         | kmol/h| 0   | 0       | 0      | 0    | 10.4    | 0      | 0      | 0     | 0     | 0     | 0      |
| Water       | kmol/h| 0   | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 0     | 0     | 0      |
| N₂          | kmol/h| 2.8 | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 0     | 0     | 2.8    |
| O₂          | kmol/h| 0.6 | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 0     | 0     | 0      |
| H₂          | kmol/h| 0   | 0       | 0      | 0    | 10.4    | 0      | 0      | 0     | 0     | 0     | 0      |
| Mass Flows  | kg/h  | 100 | 4480    | 825    | 1201 | 4480    | 2776   | 2396   | 121   | 3603  | 825   | 2375   |
| In          | kg/h  | 0   | 0       | 0      | 0    | 0       | 0      | 2296   | 0     | 0     | 0     | 0      |
| FeO          | kg/h  | 0   | 0       | 0      | 0    | 0       | 0      | 2296   | 0     | 0     | 0     | 0      |
| Fe₂O₃        | kg/h  | 0   | 0       | 0      | 0    | 0       | 0      | 2296   | 0     | 0     | 0     | 0      |
| Fe₃O₄        | kg/h  | 0   | 0       | 0      | 0    | 0       | 0      | 2296   | 0     | 0     | 0     | 0      |
| C           | kg/h  | 0   | 0       | 825    | 1201 | 0       | 0      | 0      | 0     | 825   | 0     | 0      |
| H₂O         | kg/h  | 0   | 3415    | 0      | 0    | 3415    | 0      | 0      | 0     | 3603  | 0     | 0      |
| CO          | kg/h  | 0   | 384.7   | 0      | 0    | 584     | 0      | 0      | 0     | 0     | 0     | 0      |
| CO₂         | kg/h  | 0   | 459.3   | 0      | 0    | 459     | 0      | 0      | 0     | 0     | 0     | 0      |
| Water       | kg/h  | 0   | 0       | 0      | 0    | 2776    | 0      | 121    | 0     | 0     | 0     | 0      |
| N₂          | kg/h  | 79  | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 79    | 0     | 0      |
| O₂          | kg/h  | 21  | 0       | 0      | 0    | 0       | 0      | 0      | 0     | 0     | 0     | 0      |
| H₂          | kg/h  | 0   | 21      | 0      | 0    | 21      | 0      | 0      | 0     | 0     | 0     | 0      |

*Including filtration system and separators, †Small-scale blower, ‡Including Sulphur removal, §Price extracted from suppliers in www.Alibaba.com.

**Table 4**
The calculated indicative cost associated with the capital cost of the process plant based on the data provided in the literature [50].

| Unit                | Calculated cost, (M$) | Base cost, (M$) | Base capacity | Scaling exponent |
|---------------------|-----------------------|-----------------|---------------|-----------------|
| LM handling*        | 18.5                  | 173             | 214 t/h       | 1               |
| Reactor FR          | 7.05                  | 217             | 1618 MW       | 0.66            |
| Reactor AR          | 7.05                  | 217             | 1618 MW       | 0.66            |
| Air compressing unit³| 0.001                | 4               | 10 MWₑ        | 1               |
| Syngas scrubbing*   | 2.16                  | 122             | 453 t/h       | 0.67            |
| Heat recovery        | 0.3                   | 6               | 156 MWₑ      | 1               |
| Heat Exchangers      | 0.35                  | –               | –              | 1               |
| Oxygen carrier cost† | 0.69                  | –               | –              | –               |
| Steam Generator      | 0.3                   | 6               | 156 MWₑ      | 1               |
| Steam Turbine        | 1.5                   | 14              | 54 MWₑ       | 0.67            |
| Process water cost   | 0.01                  | 2.2 $/m³      | –             | –               |
| Water pump           | 0.1                   | –               | –              | 1               |

*Including filtration system and separators, †Small-scale blower, ‡Including Sulphur removal, §Price extracted from suppliers in www.Alibaba.com.
2.36 MW of exergy (LHV-basis) is purged through carbon waste. This carbon can be fed into the agricultural sector for soil conditioning or can be burnt as a feedstock.

Depending on the geographical locations and type of the biomass, different pricing scenarios can be considered for the price of the feedstock: Scenario 1: negative price of biomass: this is associated with the cost of collection, package and transport from the source to the location of the plant that is covered by the owner of the biomass e.g. landlord or government subsidy; Scenario 2: Free biomass, which can be considered for the condition in which the process is localised and is installed on the land with available biomass resource; Scenario 3: purchase biomass at current price of 2$/GJ according to the Australian roadmap for renewable and solar thermal energy [49], for collecting and transport of biomass to be paid by the owner of the plant; Scenario 4: natural gas as a feedstock, which provides a condition to conduct a back-to-back comparison with the other similar systems; and Scenario 5: natural gas as a feedstock while the projected cost is considered to be increased to 20 and 30 $/GJ in the future.

Fig. 17 shows that the lowest levelised cost of energy for the system is observed for the first scenario (the price of biomass is negative), which means that the final price of the syngas is 11.46 $/GJ to 20.38 $/GJ for fuel prices of ~10 $/GJ to ~2 $/GJ, respectively. For scenario 2 (free biomass), the LCOE is 22.61 $/GJ increasing to 29.3 $/GJ for biomass feedstock with a price of 6 $/GJ, and further increased to 39.4 $/GJ for natural gas as a feed with a price of 15 $/GJ (scenario 3). The highest LCOE is 56$/GJ for the case of natural gas with a projected cost of 30 $/GJ (three times higher than the current price of natural gas).

A first-order comparison is made between the estimated LCOE versus several process plants reported in the literature [45] and presented in Fig. 18. The figure shows that despite the low technology readiness level (TRL) of the proposed CLG-LMOC process, it is still competitive with other technologies of higher TRLs. For example, the case of biomass as a feedstock offers a price of 2 $/GJ and final LCOE of 24.5 $/GJ, which is calculated using Eq. (6) and the data in Table 1. This is still lower than the projected LCOE calculated for CLC with recuperating reactors (LCOE ~ 40 $/GJ), fluidised bed CLC water splitting system for hydrogen production with LCOE ~ 46 $/GJ, natural gas cracking and chemical looping combustion with solid oxygen carrier and LCOE ~ 28 $/GJ and natural gas cracking with LCOE ~ 28 $/GJ.

The estimated LCOE for the proposed system against other technologies is likely to become more competitive, once carbon-tax (or similar) schemes are introduced. Also, it is estimated that the energy penalty for preheating the inlet streams adds 5% to 15% to the predicted LCOE value. Despite this uncertainty, the final LCOE for the CLG-LMOC system still competitive against many of the existing processes.

5.7. Advantages and technical challenges

The proposed CLG-LMOC system has been successfully demonstrated as a thermodynamically feasible process with a potential to produce high-quality syngas from a low-grade carbon-containing feedstock. By assessing the energetic performance of the proposed system, the following advantages of the proposed process are identified:

1) The exergy partitioning in the syngas is ~80% of the total exergy, which is reasonably good chemical exergy efficiency for a plant
operating at \( T < 1000 ^\circ C \). This also shows the potential of integrating this system with solar thermal energy units that at the current state-of-the-art operates at temperatures \(<1000 ^\circ C \).

2) Considering the low vapour pressure of the selected liquid indium metal (boiling temperature \( \sim 2300 ^\circ C \)) the indium does not evaporate, hence, corrosion and erosion effects due to the vapour of liquid metal do not pose a serious problem.

3) The system operates at a near-isothermal condition that provides a thermodynamic advantage to minimise the exergy destruction due to the temperature difference between reactors. For other metal oxides, such as copper or iron, the temperature difference between AR and FR can exceed 400 \(^\circ C\), which causes massive exergy loss.

4) Although this study focused on carbon as the feedstock, the proposed system offers flexibility in the type of the feedstock and can be utilised for a wider range of feedstock from municipal waste to waste tyres (providing that a robust scrubbing system, impurity separator, and syngas washing unit are used to remove the impurities in the feedstock to avoid any reactions with liquid metals).

Alongside these advantages of the system, the development of CLG-LMO processes inevitably will face technical challenges and engineering limitations, which require further investigation. Some of these challenges, for example:

1) **Gas distribution system:** The proposed system uses steam to enrich the hydrogen content in the syngas product. Since the oxygen carrier is a slurry liquid metal oxide, injecting steam below the freezing point of the slurry mixture result in freezing of the slurry that could generate blockage in the reactor and the piping systems. Hence, the temperature of the steam line should be monitored and controlled particularly in the expansion and injection points.

2) **Reliable trace heating:** Trace heating is another challenge associated with liquid metals in the process. The system might require a constant temperature controller in the reactors, pipes, valves and the dead zones inside the reactor. In the present work, such a system was not considered, however, in a practical design, this can add a penalty to the energy consumption of the system and the cost. There are also penalties for the external heat exchangers for the preheating systems before and after the reactor, which were not considered in this paper.

3) **Containment of liquid metal:** Although the slurry mixture (indium oxide/indium) is used at low and moderate temperatures, nonetheless, some level of corrosion is expected to occur over an extended period of operation. Hence, a robust suitable material might be required for containment of the slurry and construction of the reactors. Likewise, materials for piping and pumping systems need to be retrofitted with corrosion-resistant materials.

4) **Assessment of other liquid metals:** Although indium and indium oxide showed a thermodynamic potential, the performance of the proposed system using other liquid metal oxides warrant an evaluation in future studies.

6. Conclusions

In this work, we presented the thermodynamic performance and techno-economic analysis of a proposed CLG-LMO concept to convert low-grade carbonaceous feedstock, such as solid carbon black, into high-quality (>1) syngas. The system, together with an electricity generation unit, proved to be partially self-sustained. The results showed that the proposed concept is thermodynamically feasible and capable of producing high-quality syngas. Overall, the following concluding remarks are drawn:

1) The thermochemical analysis showed that for \( T > 500 ^\circ C \), the Gibbs free energy of the reactions is negative in both reactors, thus demonstrating the feasibility of using indium oxide/indium slurry mixture as the working fluid.

2) The operating temperature of the reactors is kept \(<1000 ^\circ C \), which offers the potential to hybridise the system with concentrated solar thermal energy systems. Also, considering the nature of the slurry metal oxide, it can be used as a thermal energy (sensible, latent and chemical) storage system.

3) The most influential thermodynamic parameters on the performance are identified to be the steam to feedstock (S/C) and the liquid metal oxide to feedstock (LMO/C) ratios. Higher S/C ratios induce unreacted steam to the system, while high LMO/C ratio adds more oxygen to the system, which in turn changes the operating mode of the process from gasification to a combustion process. Hence, the system is flexible and can be adjusted to produce heat, syngas, electricity, or a combination.

4) The results showed that the temperature of the air reactor is the controlling factor, which regulates the level of exergy to be partitioned in the power block. Also, the operating temperature of the fuel reactor should be kept close to that of the air reactor to minimise the energy destruction.

5) It was also shown that at \( T_{AR} = 500 ^\circ C \) and \( T_{FR} = 500 ^\circ C \), approximately 0.74 of the total exergy is partitioned in the syngas, while only 0.077 of exergy is partitioned in power block, and 0.183 is wasted as exergy destruction due to the temperature difference between reactors.

CRediT authorship contribution statement

M.M. Sarafraz: Conceptualization, Methodology, Writing - review & editing. F.C. Christo: Conceptualization, Methodology, Supervision, Software.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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