A THERMODYNAMIC ANALYSIS OF CHEMICAL LOOPING COMBUSTION

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ABSTRACT

Recently, interest has grown in chemical looping combustion (CLC) because it is seen as a technique that may allow cost-effective carbon capture and storage (CCS). In CLC the overall reaction by which chemical energy is released is between a hydrocarbon and air as in conventional combustors. However, the reaction is completed in two separate oxidation and reduction steps occurring in different reaction vessels. In the oxidizer (or air reactor) an oxygen carrier, usually a metal, is exothermically oxidized in air resulting in an oxide and a hot air stream (oxygen depleted). The exhaust gasses may be expanded through a turbine to produce work, while the oxide passes to the reduction vessel (or fuel reactor). Here, it reacts with the fuel, is reduced and the metal regenerated. The metal then returns to the oxidizer to complete the loop. The exhaust gasses from the reducer contain only carbon dioxide and water so that, after expansion and work extraction, the water may be condensed leaving a stream of pure CO₂ ready for storage.

Hydrocarbon fuels will continue to be used for decades, so, in the face of ambitious emission reduction targets, CCS is an important technology and methods, such as CLC, that offer automatic CO_2 separation (so-called inherent carbon capture) are particularly attractive. Despite this obvious advantage CLC was not originally conceived for the purposes of CCS, but rather as a means to produce pure carbon dioxide free from contamination by inert gases such as nitrogen. In the context of power generation it was then proposed as a means to improve the exergetic efficiency of energy conversion processes using hydrocarbons. Combustion is usually a highly irreversible process and necessitates the rejection of large quantities of heat from power cycles leading to the low thermal efficiency of gas turbines and the like. The two-stage reaction approach of CLC can reduce the irreversibility and the extent of heat rejection and hence provide improved cycle efficiency.

Ideally, both goals would be simultaneously achieved thereby offsetting both the cost of carbon capture and of compression, transportation and storage. In the paper we present a thermodynamic analysis of CLC to illustrate its potential for improving efficiency. We will then develop a methodology for selecting oxygen carriers based on their thermodynamic properties and review several candidate materials. In particular, we will compare, from a thermodynamic perspective, solid phase oxygen carriers as used in fluidised bed based reaction systems and the liquid/vapour phase carriers previously suggested by the authors. Finally, comments on practical implementations of CLC in power plant will be presented.

NOMENCLATURE

Roman

CCS	carbon capture and storage	
CLC	chemical looping combustion	
G	stream or reaction Gibbs function	MJ/kmol
Η	stream or reaction enthalpy	MJ/kmol
Ι	irreversibility or lost work	MJ/kmol
S	stream or reaction entropy	kJ/kmol.K
Т	Temperature	K
TET	turbine entry temperature	Κ

Greek

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\eta_{II} second law efficiency
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Subscripts

с	cold sink
EQ	equilibrium condition
(g)	gaseous state
h	hot source
(1)	liquid state
0	sink condition: - 25°C, 1.01bara
OX	oxidation
R	reaction
RED	reduction

(s) solid state

INTRODUCTION

A move to low carbon sources of energy is underway with the rapid growth in the world's installed wind turbine capacity (a more than six fold increase between 2001 and 2009 WWEA 2010) and the expected renaissance in nuclear fission based electricity generation being clear examples. Nevertheless, fossil fuel based thermal power plant still provide the vast majority of the electricity generated today (68%) and are likely to continue to do so for decades to come (65% predicted in 2035, EIA 2010) . Indeed, given the extent of known resources, it may well be the case that coal and gas fired powered stations will play a significant part in electricity generation for another century or more. Carbon Capture and Storage (CCS), therefore has a critical part to play in managing CO_2 emission from power plant and in managing/stabilizing atmospheric levels of this greenhouse gas.

Development of CCS is gathering pace and there are a number of pilot plant in operation or planned - including one at Imperial College. In 2008, at the Hokkaido Toyako Summit, G8 leaders called for 20 large-scale CCS demonstration projects to be launched by 2010. Whilst progress is undoubtedly being made, this challenging target seems unlikely to be met. One of the main drawbacks of CCS is the energy penalty associated with it, which the IPCC (2005) puts at between 10% and 40% with most of this going to capture and compression. That is to say, a CCS equipped power station that can reduce CO₂ emissions to the atmosphere by 80-90% will consume between 10% and 40% more fuel than a non-CCS station delivering the same amount of electricity. This penalty will hasten the exhaustion of reserves and increase the cost of electricity – neither attractive propositions for individuals or nations alike

It is for these reasons that Chemical Looping Combustion (CLC) has been attracting increased attention in recent years. The method provides '*inherent carbon capture*' in that there is very little energy penalty associated with separating the CO_2 from the exhaust gases. Indeed, it was first proposed by Lewis and Gilliland (1954) as a means for producing pure carbon dioxide free from contamination with inert gases such as nitrogen. Another potential benefit of CLC, however, is that, in the context of power generation, the method can lead to

improved cycle efficiency as proposed by Richter and Knoche (1983). Hence, if the cycle is well optimized, CLC has the potential to offset the energy penalties associated with both capture and compression of CO_2 and so could provide CCS without excess fuel consumption or the cost associated with it. In this paper, we review the thermodynamics of CLC as compared to conventional thermal power cycles to show how improved efficiency can be achieved and to derive a methodology for CLC cycle optimization. We then compare on a thermodynamic basis a commonly cited solid phase CLC cycle with an alternative derived by the authors and which shows the potential for greater efficiency. We conclude by providing some comments on practical aspects of the implementation of the technique.

THE CHEMICAL LOOPING CONCEPT

In CLC the energy source remains a hydrocarbon but in a sense the fuel changes to a metal that is referred to as the oxygen carrier. A schematic of the resulting system is provided in figure 1. In a conventional thermal power system, a single exothermic air/fuel reaction takes place in a combustion chamber with sensible heat deposited either directly into the working fluid as in the Brayton cycle or subsequently transferred to it as in the Rankine cycle. CLC by contrast involves two separate reactions, one oxidation and one reduction. The oxidizer is analagous to the standard combustion chamber and in it the 'fuel' (oxygen carrier) is oxidized in air via an exothermic reaction. The chemical energy liberated can, in principle, be converted to work in a heat engine in the usual way via a Brayton, Rankine or indeed other conventional cycle. The difference in CLC lies in the addition of a second reaction chamber, the reducer. Here, the metal oxide is reduced in a reaction with the hydrocarbon and hence the metal 'fuel' regenerated. From here it returns to the oxidizer to burn again and the loop is complete.

At this stage the principle advantage of CLC becomes clear. The exhaust stream from the oxidizer consists of oxygendepleted air whilst that from the reducer comprises carbon



Figure 1 Schematic diagram of an ideal chemical looping combustion system for power production

dioxide and water vapour. A control volume drawn around this two-reactor system would show a hydrocarbon and air entering and nitrogen, carbon dioxide, water, useful work and rejected heat leaving. Hence, the CLC system is analogous to a conventional thermal power system with one main difference, namely, that the carbon dioxide and nitrogen leave in separate streams. Carbon capture therefore requires only that the water vapour be condensed.

To this point we have made no mention of the thermodynamics of this process, i.e. of the likely cycle efficiency. It transpires that this depends not only on operating temperatures and pressures but also on the choice of oxygen carrier. The oxidation reaction is exothermic and by an appropriate choice of oxygen carrier the reduction reaction can be made endothermic. As shown in figure 1, it can then be arranged so that the heat necessary to drive this reaction is that rejected by the heat engine. To establish if the CLC approach conveys any thermodynamic advantage, it is instructive to compare the thermodynamics of CLC with those of conventional fuel/air combustion cycles.

THE THERMODYNAMICS OF COMBUSTION AND CHEMICAL LOOPING

Conventional combustion is a highly irreversible process that results in a great deal of lost work (almost a $1/3^{rd}$ of the energy content of the fuel typically) and it is this that limits the cycle efficiency of current power plants. The problem stems from the fact that the reaction takes place at a temperature far below that required for reversibility, resulting in significant entropy generation. Cengel (2007) ascribes the irreversibility of combustion to an "uncontrolled electron exchange between reacting components" but goes on to explain that we are not constrained to use combustion to convert chemical energy and not restricted by the Carnot heat engine efficiency. A fuel cell provides the means for a controlled exchange of electrons and can achieve a greater efficiency than a practical heat engine burning hydrogen. Chemical looping can achieve a similar effect, even though it still employs a heat engine, by using a staged reaction approach. To illustrate we can compare CLC with conventional combustion as follows.

It is well known that the turbine entry temperature (TET) limits the efficiency of gas turbines and that what limits TET is the material from which the turbine blades are made. For decades engineers have steadily increased TET with improved materials, manufacturing techniques and thermal management. The development path through forging, casting, directional solidification, single crystals, film cooling and ceramic thermal barrier coatings is well known and today TET for land based gas turbines can exceed 1500 K and for aerospace turbines it can approach 2000 K for short periods. An obvious question to pose, however, is does thermodynamics suggest an optimum or limiting temperature, what is it and can we realistically increase TET to match it?

If we consider the combustion of methane, assuming reactants enter and products leave the system at the dead state (taken to be 1bar and 298.15 K) the maximum work output is given by the fall in Gibbs function, $-\Delta G_0$, whilst the energy released is given by the fall in enthalpy, $-\Delta H_0$. To take full advantage of the available work we require a heat engine with an efficiency of:

$$\eta = \frac{-\Delta G_0}{-\Delta H_0} = \frac{-(\Delta H_0 - T_0 \Delta S_0)}{-\Delta H_0} = 1 - T_0 \frac{\Delta S_0}{\Delta H_0} \quad \dots (1)$$

The best heat engine available, albeit theoretically, is the Carnot engine which has an efficiency of:

$$\eta_{Carnot} = 1 - \frac{T_c}{T_h} \dots (2)$$

Equating the two and assuming that $T_0=T_c$, we can deduce the necessary hot source temperature:

$$T_h = \frac{\Delta H_0}{\Delta S_0} \dots (3)$$

In the case of methane the required temperature is found to be approximately 160000 K. This is an indication of the sort of TET value that would be required to make full use of the available energy from methane. The extreme value illustrates the futility of the task. A similar result is found for the majority of hydrocarbons and stems from the fact ΔG_0 and ΔH_0 are approximately equal or that, equivalently, $\Delta S_0 \approx 0$. Another way to interpret this result is as the equilibrium temperature of the reaction i.e. the temperature at which the reaction would proceed without generating entropy, or, the temperature at which it would proceed reversibly. In the case of methane, this presents another problem, since the reactants would have to be heated to this value before the reaction starts. Nevertheless, since reversibility is the engineering ideal, the expression for T_h provides us with a means to assess different fuels (oxygen carriers) and their prospects for achieving a high second law efficiency.

As has already been mentioned, in CLC, metals are the preferred oxygen carriers. By way of an example, we consider cadmium, chosen since it was the thermodynamically preferred metal identified by Richter and Knoche in their 1983 paper. Oxidation of cadmium proceeds according to the following reaction:

$$Cd + 0.5O_2 + 1.88N_2 \rightarrow CdO + 1.88N_2$$

Equation 3 suggests that the equilibrium temperature of this reaction to be approximately 1700 K. At this temperature and 1 bar pressure, cadmium is gaseous and its oxide liquid, both of which, have been accounted for in the reaction temperature calculation. So, with cadmium as the fuel and a TET of around 1700 K, all of the available energy could theoretically be accessed – a second law efficiency of 100%. However, the world is not blessed with a geologic endowment of pure metals

for use as fuels but rather with a substantial quantity of hydrocarbons. In CLC then, hydrocarbons are used to regenerate/reduce the oxygen carrier completing the cycle and allowing continuous energy conversion with a limited inventory of metal.

Another way of viewing the challenge associated with fuel/air combustion of methane is to say that, at practical temperatures it is not possible to transfer the availability of the methane to its combustion products without entropy generation and consequent lost work. However, in other reactions of methane, its availability can theoretically be completely transferred to the reaction products without loss and one such reaction is the reduction of cadmium oxide. The reaction proceeds as follows:

$$4CdO + CH_4 \rightarrow 4Cd + CO_2 + 2H_2O$$

The reaction is endothermic and, if it proceeds isothermally, the products are of greater entropy than the reactants. Nevertheless, the reaction can proceed without entropy generation if the heat transferred in to maintain the temperature causes a drop in entropy of the heat source that matches the increase due to the reaction. That is, there will be no entropy generation if:

$$\frac{\Delta H_R}{T_R} = \Delta S_R$$

Here, ΔH_R is the enthalpy of the reaction, ΔS_R is the entropy increase of the reactants relative to the products and also the entropy decrease of the heat source and T_R is the temperature of the reaction. If the values in this expression are evaluated based on those of formation at standard conditions, that is to say ignoring the effect of sensible enthalpy, then the above expression, when rearranged for temperature, is the same as equation 3. In the case of cadmium the required temperature is approximately 800 K.

The obvious source of heat to feed the reduction reaction is that rejected from the heat engine. Now the rationale for the layout of the CLC system shown in figure 1 is complete. In the *combustor* metal *fuel* is burnt in an isothermal process with the heat generated transferred to the working fluid of a Carnot engine at a temperature such that the entropy decrease of the reaction products exactly matches the entropy rise of the working fluid. The engine does work and rejects heat to a reducer wherein the metal *fuel* is regenerated in an isothermal endothermic reaction that occurs at a temperature such that the entropy drop in the working fluid exactly matches the entropy rise in the reaction products.

With hot and cold source temperatures of 1700 K and 800 K respectively the cadmium fueled Carnot engine has a first law efficiency of approximately 53% but the CLC system is reversible and has a second law or exergetic efficiency of 100%.

At 800 K the rejected heat retains substantial availability but this is not lost as it combines with the availability *top-up* received from the methane and returns to the *combustor*. In a sense then, the reducer can be thought of as an availability recuperator. In figure 2 a Sankey diagram shows the fluxes of energy for the reversible cadmium based CLC system. It is important to note that the only energy input to the system is from the methane and that it is not all available. Hence as shown in the diagram, the unavailable portion is rejected to the surroundings, in this case, via a second heat engine.



Figure 2 Sankey diagram for energy fluxes in CLC using cadmium as the energy carrier

The above analysis, although highly idealized, serves to demonstrate that the staged reaction approach of CLC has the potential to make better use of the available energy in hydrocarbons and thereby has the potential to offset the energy penalty associated with CCS. It is also helps define the desirable properties of oxygen carriers. These should oxidize exothermically and reversibly at a temperature in keeping with the limits on current TET's. They should then be reduced endothermically and reversibly, ideally at a low temperature to maintain a reasonable specific power output. In both cases, the entropy change is key in setting the desirable reaction temperature. During oxidation, ideally there should be a drop in entropy that can be matched by heat rejection to the working fluid. Cadmium allows this to happen because in the oxidation reaction 1.5 gaseous moles are replaced by a single liquid mole. During reduction, ideally there should be a rise in entropy that can be matched by the heat absorbed from the working fluid. Cadmium, again, fits the bill in that during reduction 1 gaseous mole becomes 7 gaseous moles.

Although thermodynamically well suited to power generation, cadmium is impractical as an oxygen carrier due to its toxicity. In the sections that follow we therefore consider oxygen carriers that have been proposed for practical CLC cycles and compare them on a thermodynamic basis.

SUGGESTED EMBODIMENTS OF CLC

Almost every CLC system considered to date has been based on solid phase oxygen carriers, indeed that the metal and its oxide remain solid at the operating temperatures of the cycle was identified by Adenez et al (2004) as one of a number of selection criteria for oxygen carriers. In such systems interconnected fluidized bed reactors are used as shown schematically in figure 3. The oxygen carriers are encapsulated in an inert carrier, which is typically a ceramic (Fang et al, 2009, Lyngfeldt et al, 2008, Hossain and de Lasa, 2008) and provides improved chemical and mechanical properties for the particles.

In contrast, the authors, in pursuit of thermodynamic efficiency, have considered oxygen carriers in solid, liquid and vapour phases with the sodium based cycle of McGlashan et al (2011) an example. Herein, as in Botsis (2010), we compare, on a thermodynamic basis, one of the commonly suggested solid phase oxygen carriers, iron, with a multi-phase carrier, zinc.



Figure 3. Chemical looping combustion based on interconnected fluidised beds

IRON VS ZINC OXYGEN CARRIERS

In the discussion so far it has been assumed that the reactions take place at their respective equilibrium temperatures. As such, there has been no entropy generation or irreversibility. In this section we will compare iron and zinc oxygen carriers, using methane as the energy source, at temperatures thought to be achievable in real power systems. These may differ from the equilibrium temperatures and we therefore need a means to account for this. If we can retain the assumption of isothermal reactions then the irreversibility generated in a non-equilibrium reaction is given by:

$$I = -T_0 \left(\frac{1}{T_R} - \frac{1}{T_{EQ}} \right) \Delta H_0 \dots (4)$$

which is the same equation that results from a consideration of heat transfer across a finite temperature difference.

Considering, again, a control volume around the CLC system with only methane and air entering and combustion products leaving, it should be apparent that the availability of the system is that of methane. This is given by the fall in Gibbs energy between methane and its combustion products and hence can define the second law efficiency as:

$$\eta_{II} = \frac{-\Delta G_0 - \left(I_{OX} + I_{RED}\right)}{-\Delta G_0}$$

In the analysis that follows, we have assumed the maximum cycle temperature based on practical limitations but set the temperature of the reduction reaction to its equilibrium value. This makes I_{RED} equal to zero. Substituting for I_{OX} from equation (4) and for T_{EQ} from equation (3) the expression for the second law efficiency becomes:

$$\eta_{II} = 1 - \frac{T_0 \left(\Delta S_0 - \frac{\Delta H_0}{T_R}\right)}{-\Delta G_0}$$

and we can now use this equation to compare the selected oxygen carriers. The analysis will be conducted for a maximum cycle temperature of 1300 K since this corresponds to a realistic TET for a modern gas turbine. Results for single step methane combustion are also included for completeness. The reactions for methane, iron and zinc (with nitrogen omitted for clarity) proceed as follows:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$

$$\frac{8}{3}Fe + 2O_2 \rightarrow \frac{4}{3}Fe_2O_3$$

$$\frac{4}{3}Fe_2O_3 + CH_4 \rightarrow \frac{8}{3}Fe + CO_2 + 2H_2O$$

$$4Zn_{(g)} + 2O_2 \rightarrow 4ZnO_{(s)}$$

$$4ZnO_{(s)} + CH_4 \rightarrow 4Zn_{(g)} + CO_2 + 2H_2O$$

where the moles of the respective species have been modified so that in each case a single mole of methane is consumed. The results of the analysis are shown in table 1.

Table	1.	Equilibrium	temperature	and	second	law
efficiencies for iron and zinc oxygen carriers						

	T _h	T _{EQ_OX}	T_{EX_RED}	η_{II}
Methane	1300 K	160000 K	NA	77%
Methane/iron	1300 K	2995 K	819 K	82%
Methane/zinc	1300 K	1840 K	1080 K	83%

PRACTICAL ASPECTS OF CLC

The analysis presented in the previous section is instructive in showing what might be possible with CLC but is highly idealized, relying, as it does, on the use of a hypothetical Carnot engine exchanging heat with the two reaction vessels. It is therefore worthwhile to consider how CLC might be embodied in real power systems and how close we might be expected to get to the theoretical ideal that the values in table 1 represent. The first question is what type of heat engine should be employed.

One possibility is to use a Rankine cycle with a working fluid exchanging heat indirectly with the oxidation and reduction reactions, boiling and condensing respectively as it does so. For efficiency the hot and cold temperatures of the cycle should match those of the reactions as closely as possible. Although modest relative to the equilibrium values, the temperatures in table 1 are too high for the use of steam as the working fluid. A possible alternative for high temperature Rankine cylces is the use of metal vapour as the working fluid, something that has been actively considered for space power systems. Using refractory metals for hot section components allows the operating temperatures to be increased and it is theorized that cycles with working fluid temperatures as high as those in table 1 might eventually be possible, Bevard and Yoder (2003). McGlashan (2008), considered a Rankine CLC system using sodium as the working fluid that captured the thermodynamic advantages of CLC but was ultimately rejected due to the necessity for indirect heat exchange at unrealistically high temperatures.

An alternative approach would be to use a gas turbine to extract the useful work. Here, indirect heat transfer may be replaced by direct heat transfer to the oxygen-depleted air exiting the oxidation reactor that can subsequently be expanded through a turbine. This is the approach suggested by Ishida and Jin (1993) who outlined a system using nickel as the oxygen carrier, a methane energy supply and work extracted from the oxidizer exhaust using a humid air turbine at an inlet temperature of 1473 K. Heat energy for the endothermic reduction reaction was then provided by transfer of the hot nickel oxide to the reducer with further work extracted from the expansion of the reducer exhaust (CO_2 and H_2O) through a second turbine with an inlet temperature of 1373K. This approach is much more in keeping with existing engineering capabilities and Ishida estimated an overall LHV plant efficiency of 55.1%.

Another gas turbine based CLC concept, more in keeping with the thermodynamic elegance of the approach outlined in this paper, involves using the oxygen carrier as the working fluid and expanding it through a gas turbine with the turbine exhaust directed to the reducer where its residual enthalpy can be used to drive the endothermic reduction. However, there is a problem with this approach in that the oxidizer exhaust contains nitrogen and cannot therefore be transferred to the reducer if the key feature of CLC, namely inherent CO₂ separation, is to be realised. To address this issue. McGlashan et al (2011) proposed a cycle in which sodium is used as the working fluid and the oxygen carrier and is lean burnt in oxygen provided via an external air separation unit. Energy for the reduction reaction is provided partly by liquid Na₂O transferred from the oxidizer but mostly, as originally intended, as heat rejected from the turbine exhaust. The cycle has a turbine inlet temperature in the range 1750-1850 K and so significant engineering challenges would need to be addressed to implement it. Nevertheless, accounting for the work required to drive the air separation until the potential cycle efficiency was estimated to be in excess of 75%.

DISCUSSION

The results presented in table 1 show that at a relatively modest peak temperature of 1300 K both CLC systems show improved second law efficiency relative to single step combustion of methane and hence both could help offset he cost of CCS beyond the initial separation of the CO_2 which is inherent.

Although the advantage of zinc over iron is modest, the equilibrium oxidation temperature of zinc is over 1000 K lower than that for iron so that 100% second law efficiency might conceivably be obtained within the material limits of realizable turbo-machinery.

The methodology for assessing oxygen carriers set out in this paper is designed to enable selection of carriers on the basis of their potential for high second law efficiency in the context of a CLC system as represented in figure 1. This approach leads one to consider a wider range of options than would be the case if one were constrained to the solid phase oxygen carriers required for the dual fluidized bed embodiment of CLC. Not surprisingly, however, a theoretical thermodynamic approach throws up candidate carriers that prove to be quite impractical in the field - cadmium being a good example. If efficiency is the primary goal then one is drawn to consider oxygen carriers that may exist in multiple phases at different points in the cycle. The comparison between iron and zinc as oxygen carriers is just one example of this.

Although the efficiencies of the zinc and iron based CLC systems are similar, the multi-phase behaviour of zinc means that the respective power plant needed to exploit them would be very different. Since iron and its oxide exist in the solid phase throughout, the interconnected dual fluidized bed system discussed earlier represents the best implementation route.

For zinc and other multi-phase carriers the authors have considered metal vapour Rankine cycles although, here, the problems associated with high temperature indirect heat exchange seem almost insurmountable. A more intriguing option would be to use the oxygen carrier as the working fluid which, again, would require a metal vapour turbine. In previous work, the authors have considered a sodium based cycle with an estimated efficiency in excess of 75%.

CONCLUSIONS

Chemical looping combustion offers the potential for power systems with inherent carbon capture and with improved thermal efficiency to further offset the costs of CCS, namely compression, transportation and storage. Although, energy is still derived from a hydrocarbon, in CLC a metal oxygen carrier is burnt in the work producing reaction. This process produces far less irreversibility than standard one-stage combustion of methane as the reaction can be conducted at or near equilibrium. Cadmium is a thermodynamically favorable oxygen carrier but is impractical due to its toxicity. Two other carrier, iron and zinc, have been considered. Iron and its oxide remain solid throughout whilst zinc may be present in solid, liquid and gas phases. For a peak temperature of 1300 K, the second law efficiencies of iron and zinc based CLC were similar (82% and 83% respectively). However, the lower equilibrium temperature of zinc oxidation makes it a better candidate for achieving a high (close to 100%) exergetic efficiency. Iron may be used in a CLC system based on interconnected fluidized beds. Zinc requires a different implementation and, as has been reported elsewhere, might even be used as the working fluid.

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