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Thermodynamic Analysis of an Electrochemically Driven Chemical Looping Heat Pump

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ABSTRACT

Electrochemical cells have been widely explored for their use in high efficiency energy systems. In this paper a novel heat pump cycle is proposed that utilizes chemical looping driven by electrochemical cells. Chemical looping is a method that has been applied to various applications such as combustion and air separation. It consists of cycling a substance between different chemical compositions in order to produce a desired effect. When the chemical composition of a fluid changes, various properties such as its saturation pressure will also change. In the proposed concept, the chemical looping of electrochemically active fluids is leveraged to generate a heat pumping effect. A number of electrochemically active liquid organic hydrogen carriers including alcohols such as ethanol and isopropanol have been theoretically investigated for use in the proposed cycle. These organic fluids were integrated into a thermodynamic model of the proposed cycle. When operating as an air conditioner the model indicates that an increase in cooling COP of over 20% could be achieved in comparison to a conventional vapor compression system using R410A.

1. INTRODUCTION

Buildings currently account for nearly 40% of primary energy consumption within the United States. They consume around 74% of the electricity generated and 34% of the natural gas produced. Around 40% of all CO₂ emissions can be attributed to the building sector (U.S. Department of Energy, 2011). A significant portion of this energy is used to maintain thermal comfort via heating and cooling. In addition, the demand for air conditioning is continually rising. Some estimates predict that the global energy demand for air conditioning will grow 33-fold over the next century (Birmingham Energy Institute, 2015). Efficiency improvements of air conditioning and heat pumping equipment represent an opportunity to simultaneously reduce the costly energy consumption and harmful greenhouse gas emissions associated with heat pumping systems.

The majority of air conditioners and heat pumps on the market utilize vapor compression (VC) technology. These systems utilize a mechanical compressor to pressurize a refrigerant that then cycles through condensation and evaporation stages to move heat from one temperature to another. The efficiency of these systems is strongly dependent on the performance of the compressor. When compared to Carnot heat pumping limits, these conventional systems still have room for improvement, which can lead to a reduction in primary energy consumption. A myriad of alternatives to vapor compressions systems have been investigated including absorption, magneto-caloric, thermoelectric, and chemical systems.

Chemical reactions have been studied for a wide range of heat pumping applications with hopes of being more environmentally friendly and achieving higher performance than vapor compression systems. They can be driven by either thermal energy or work input and can be classified as direct or indirect systems. Direct chemical heat pumps utilize the endothermic nature of a reaction to absorb heat and the exothermic behavior of a separate reaction to reject heat (Wongsuwan et al. 2001, Cai et al. 2011, Karaca F. et al. 2002, Kawasaki et al. 1999). Indirect systems utilize chemical reactions to promote a secondary effect (Gerlach, 2004). This secondary effect is then used to drive a heat pumping cycle. Thermal energy can be used as an input to drive chemical heat pump systems by promoting

desorption from a substrate or chemical dissociations. Work driven chemical heat pumps typically employ mechanical compressors or electrochemical devices to promote chemical transformations.

Kreysa et al. (1990) proposed a direct electrochemical heat pump consisting of two electrochemical cells together forming galvanic and electrolysis cells using various metal based reactions. Newell (1997) proposed a similar direct electrochemical heat pump system with the use of water electrolysis and a hydrogen/oxygen fuel cell. Gerlach (2004) presented an indirect electrochemical refrigeration cycle which relied on the evolution of a condensable gas and the throttling and evaporation of the fluid to generate a heat pumping effect.

2. CHEMICAL LOOPING HEAT PUMP

2.1 System Description

The chemical looping heat pump (CLHP) presented here is an indirect chemical heat pump wherein a chemical reaction will be used to promote a secondary effect to drive a heat pumping cycle. The concept of chemical looping has been investigated for a variety of applications such as combustion and air separations (Fan et al., 2010). In these applications a substance is cycled between various chemical compositions in order to facilitate a process such as separation. For the heat pump concept presented here, the chemical cycling of a working fluid will be used to change the saturation pressure of a working fluid which can in turn be leveraged to generate a heat pumping effect.

In typical vapor compression systems, the vapor coming out of the evaporator is compressed to high pressure in order to be condensed to a liquid. If, however, a chemical reaction was used to transform the vapor into a liquid, a pump as opposed to a compressor could be used to recompress the fluid. The working principle of the cycle is shown in Figure 1. Starting at state ① a substance in the liquid state denoted as 'A' is pumped to high pressure. A chemical reaction is then utilized to convert fluid A into fluid B at state ②. An energy input such as heat or work is required to drive the reaction. The reaction should be chosen such the fluid B is more volatile than fluid A. Fluid B is then throttled to low pressure, state ③, and then evaporated, state ④. The process from ② - ④ is similar to that of vapor compression cycles. A reverse reaction is then used to convert fluid B back into fluid A. Due to the selection of fluid B being more volatile than fluid A, the output of this reaction can be in the liquid state depending on the temperature and pressure of the reaction.

There are a number of ways in which the chemical transformations could be accomplished. For this study we will investigate the use of electrochemical cells similar to those used in the fuel cell industry. These electrochemical cells work through interactions between an anode, cathode, and electrolyte. At the anode a chemical undergoes oxidation generating electrons. The electrons are transferred through an external circuit while ions generated either at the anode or cathode travel through the electrolyte. At the cathode the electrons combine with a separate substance in a reduction reaction. There are a variety of electrolytes that are used in fuel cell systems. Examples of various fuel cell types are shown in Figure 2.

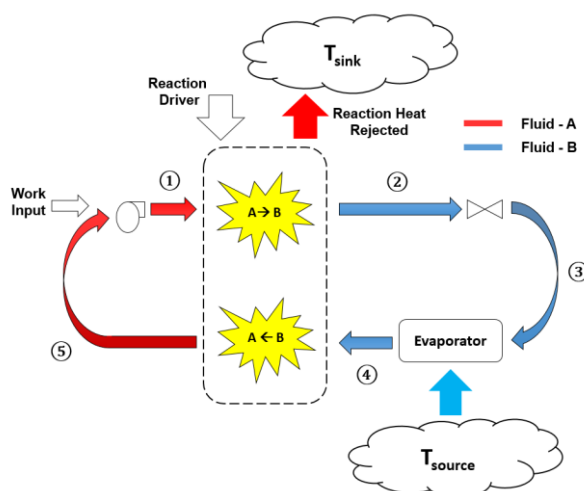


Figure 1: Schematic of Chemical Looping Heat Pump Concept.

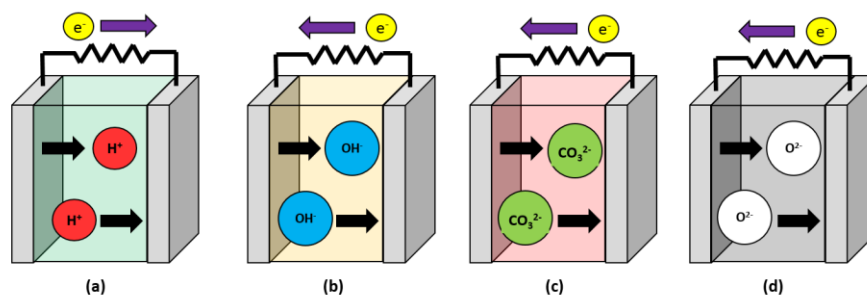


Figure 2: Electrolyte and ion transport for various fuel cell types. (a) proton exchange membrane fuel cell (b) alkaline fuel cell (c) molten carbonate fuel cell (d) solid oxide fuel cell.

Using electrochemical cells such as these, an embodiment of the CLHP is shown in Figure 3. Here the forward and reverse chemical transformations occur within the anode and cathode of the cell and the work input of an electrical current is used as the reaction driver.

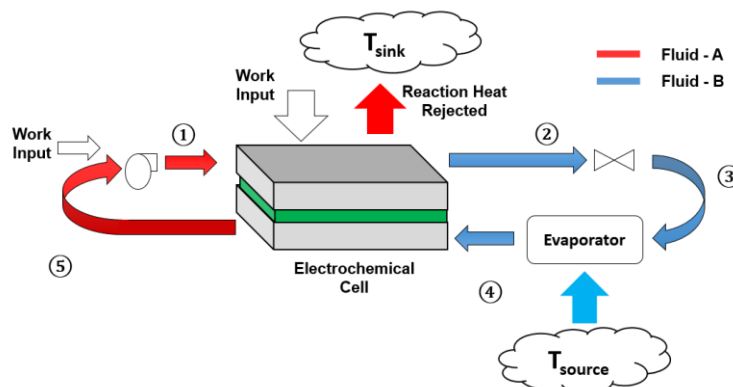


Figure 3: Schematic of CLHP using electrochemical cell as reaction driver.

2.2 Working Fluid Selection

When using electrochemical cells, the choice of the electrolyte influences the type of reactants that can be used within the cell. For the duration of this investigation the use of proton exchange membrane (PEM) cells will be assumed. PEM fuel cells have been widely studied and are the most popular choice for low temperature reactions in the range of interest to commercial and residential heat pumping.

Being that PEM cells operate through the transfer of hydrogen ions (protons) the working fluids used for the CLHP employing PEM cells need to be capable of undergoing hydrogen transfer reactions where hydrogen ions are lost (dehydrogenation) and hydrogen ions are accepted (hydrogenation). The fluids must also be condensable at temperatures of interest to heat pumping applications. One of the challenges of the fuel cell industry has been the selection of high density hydrogen carriers. To this end different liquid organic substances have been investigated for use in conjunction with PEM cells (Araujo et al. 2012, Soloveichik, Kariya 2014). Many of these substances are capable of undergoing hydrogenation/dehydrogenation reactions and as such are potential candidates for working fluids for the CLHP. The use of PEM cells have also been explored for electro-organic synthesis where the desired outcome is the generation of a particular chemical substance (Yuan et al, 2005, Wiyaratn, 2010). These studies have also lead to the identification of potential working fluids for the CLHP.

The CLHP utilizes the change in saturation temperature of a fluid in order to re-pressurize the fluid in the liquid phase after the evaporation stage. As such one of the most important criteria for a potential CLHP working fluid is

that there be a significant change in the saturation temperature of the fluid after it undergoes a transformation in a PEM cell. Imposing the criteria that the difference in normal boiling points of the chemicals before and after the reaction be greater than 15°C, possible working fluid pairs for the CLHP were selected and are shown in Table 1. For each pair the chemical transformation can be achieved via hydrogenation and dehydrogenation reactions. These fluids were selected from work done investigating liquid organic hydrogen carriers and in electro-organic synthesis. The substances in Table 1 should not be considered an exhaustive list of all the possible working fluids that can be used in a CLHP using PEM cells.

Table 1: List of potential working fluids for CLHP

Pair Abbreviation	Compound	Reaction Formula	$\Delta T_{\text{boiling}} [^{\circ}\text{C}]$
MTH	Methanol	$\text{CH}_3\text{OH} \leftrightarrow \text{CH}_2\text{O} + \text{H}_2$	83.59
	Formaldehyde		
ETH	Ethanol	$\text{C}_2\text{H}_6\text{O} \leftrightarrow \text{C}_2\text{H}_4\text{O} + \text{H}_2$	57.28
	Acetaldehyde		
IPA	Isopropanol	$\text{C}_3\text{H}_8\text{O} \leftrightarrow \text{C}_3\text{H}_6\text{O} + \text{H}_2$	26.38
	Acetone		
BUT	Butanol	$\text{C}_4\text{H}_{10}\text{O} \leftrightarrow \text{C}_4\text{H}_8\text{O} + \text{H}_2$	20.06
	Butanone		
PENT	2-Pentanol	$\text{C}_5\text{H}_{12}\text{O} \leftrightarrow \text{C}_5\text{H}_{10}\text{O} + \text{H}_2$	16.79
	2-Pentanone		
PGLY	Propylene Glycol	$\text{C}_3\text{H}_8\text{O}_2 \leftrightarrow \text{C}_3\text{H}_6\text{O}_2 + \text{H}_2$	31.3
	Hydroxyacetone		
OXYL	o-xylene	$\text{C}_8\text{H}_{10} + 3\text{H}_2 \leftrightarrow \text{C}_8\text{H}_{16}$	118
	1,2-dimethylcyclohexane		
TETRA	tetrahydrofuran	$\text{C}_4\text{H}_8\text{O} \leftrightarrow \text{C}_4\text{H}_4\text{O} + 2\text{H}_2$	34.25
	furan		
ANIL	aniline	$\text{C}_6\text{H}_7\text{N} + 3\text{H}_2 \leftrightarrow \text{C}_6\text{H}_{13}\text{N}$	49.9
	cyclohexylamine		

Since polymer membranes ideally only transmit protons, it may be possible to concurrently utilize different reacting pairs in the CLHP. In this manner, one chemical transformation could be occurring on one side of the cell and a reaction involving a dissimilar chemical pair could occur on the other side of the cell. An embodiment of the CLHP using two reacting pairs in two different circuits is shown in Figure 3. Fluids 1A and 1B make up one circuit and fluids 2A and 2B make up a second circuit.

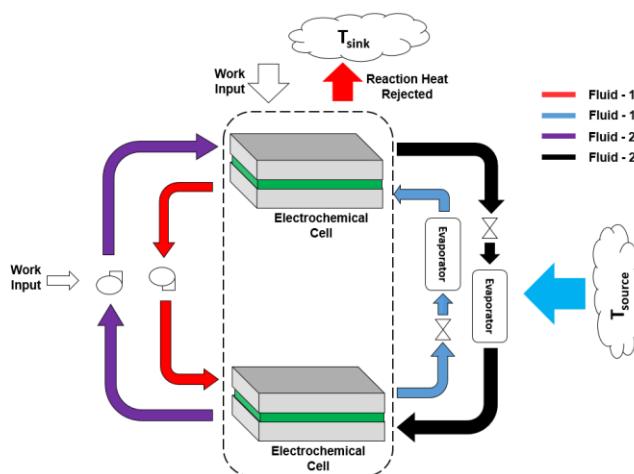


Figure 3: Diagram of CLHP using two circuits of reacting pairs

3. SYSTEM MODELING

A thermodynamic model was developed in order to assess the performance of the CLHP using PEM cells. There are many similarities between the modeling of the single circuit CLHP shown in Figure 2 and the dual circuit embodiment shown in Figure 3. For both systems the fluids going through a circuit undergo the same steps of pumping, hydrogenation/dehydrogenation, throttling, and evaporation. The modeling approach for the single circuit CLHP will be described and can be easily extended to a dual circuit CLHP by adjusting the inputs and outputs of the electrochemical cells. The following assumptions were made to simplify the modeling:

- Chemical reactions go to completion
- No crossover of chemicals through cell membrane
- Negligible pressure drops in lines

The electrochemical cells are cooled by rejecting heat to the ambient. Using the approach temperature the temperatures of the evaporator and of the electrochemical cell can be determined from Equations 1 & 2.

$$T_{evap} = T_{source} - T_{approach} \quad (1)$$

$$T_{cell} = T_{sink} + T_{approach} \quad (2)$$

Once the working pair for a circuit has been identified the low and high side pressures can be calculated using the saturation pressures of the fluids. From the state points in Figure 3, states ① & ② are at high pressure and states ③ - ⑤ are at low pressure. The low and high side pressures are given by Equations 3 & 4.

$$P_{low} = P_{sat}(fluidB, T_{evap}) \quad (3)$$

$$P_{high} = P_{sat}(fluidB, T_{cell}) \quad (4)$$

To ensure that the fluid at state ⑤ will be in the liquid phase after the chemical reaction the saturation temperature of the fluid needs to be evaluated at P_{low} . If T_{cell} is greater than the saturation temperature at ⑤ then the temperature lift is outside of the range of the particular working fluid pair. A different pair should be used or the temperature lift has to be reduced.

For each circuit the flow rate of one fluid making up the reacting pair needs to be specified. From here the molar ratios from the stoichiometric reaction equations are used to determine the molar flow rates of the other fluid in the pair as well as the hydrogen transferred during the reaction. Using Equation 5 as an example illustrating the use of aniline and cyclohexylamine, specification of the aniline molar flow rate dictates that the flow of cyclohexylamine be equal to that of aniline and the molar flow rate of hydrogen is 3 times higher than that of aniline.



The pump work can be calculated using Equation 6. The electrochemical devices are assumed to be isothermal components. The ideal work required to drive the chemical reactions is equal to the change in Gibbs free energy between the product and reactant streams and is shown in Equation 7 (Revankar et al. 2014). For the single circuit CLHP the reactants are taken as the input streams to the cell at states ① & ④ and the products are the output streams at states ② & ⑤. For the CLHP using dual circuits, the reactant and product streams need to be adjusted accordingly. Using this formulation, if the work comes out as positive then the electrochemical process produces work, whereas if the work is negative then the process consumes work.

$$\dot{W}_{pump} = \frac{\dot{m}_{fluid,①} v_{fluid,①} (P_{high} - P_{low})}{\eta_{pump}} \quad (6)$$

$$w_{cell}^o = -\Delta\bar{g} = -(\sum \bar{g}_{products} - \sum \bar{g}_{reactants}) \quad (7)$$

The Gibbs free energy is calculated according to Equation 8. Using the enthalpy and entropy of formation, the enthalpy and entropy of each stream can be calculated as functions of temperature and pressure according to Equations 9 - 12. The formation properties should be selected so as to represent the phase of the fluid at the specified state. The state entering the cell from the evaporator is gaseous while all other inputs and outputs are assumed to be in the liquid phase.

$$g = h - Ts \quad (8)$$

$$h = h_f^o + h(T) \quad (9)$$

$$h(T) = \int_{T^o}^T c_p dT \quad (10)$$

$$s = s_f^o + s(T) \quad (11)$$

$$s(T) = \int_{T^o}^T \frac{c_p}{T} dT - R \ln \left(\left(\frac{P}{P^o} \right)^n \right) \quad (12)$$

Using the change in Gibbs free energy the voltage required to drive the reaction can be found from Equation 13. Here F is Faraday's Constant and n_e is the number of electrons transferred during the reaction. This is related to the amount of H_2 transferred during the stoichiometric reaction via Equation 14.

$$V_{cell}^o = \frac{-\Delta g}{n_e F} \quad (13)$$

$$n_e = 2 \cdot n_{H_2} \quad (14)$$

Equation 13 represents the ideal voltage required to drive the electrochemical reaction. In reality irreversibilities will necessitate greater voltage input to drive the reaction. For a work producing reaction the actual voltage will be given by Equation 15 and for a work consuming reaction the required voltage is given by Equation 16.

$$V_{cell} = V_{cell}^o \cdot \eta_{cell} \quad (15)$$

$$V_{cell} = \frac{V_{cell}^o}{\eta_{cell}} \quad (16)$$

The electrical current through the cell can be calculated from the molar flow rates of the fluids and the number of electrons transferred via each reaction n_e as shown in Equation 17.

$$I = \dot{n}_{fluid} \cdot n_e \cdot F \quad (17)$$

Using the actual cell voltage and the calculated current, the power consumed or produced during the chemical transformations is given by Equation 18.

$$\dot{W}_{cell} = V_{cell} \cdot I \quad (18)$$

After leaving the electrochemical cell at (2), the fluid is throttled to low pressure in an isenthalpic expansion process and Equation 19 applies.

$$h_{(3)} = h_{(2)} \quad (19)$$

After the expansion process the fluid is evaporated to a saturated vapor. Saturation temperatures and pressures are used in conjunction with the Clausius–Clapeyron equation to determine the enthalpy of vaporization of the fluid, Equation 20. In order to account for the quality of the fluid after the expansion stage the difference in the enthalpies of the liquid state evaluated before and after the expansion stage are used to adjust the total enthalpy of vaporization according to Equation 21. The total rate of cooling can be determined from Equation 22.

$$h_{vap} = \frac{\ln \left(\frac{P_2}{P_1} \right) \cdot R}{\frac{1}{T_2} - \frac{1}{T_1}} \quad (20)$$

$$h_{evap} = h_{vap} - (h_{Liq,(2)} - h_{Liq,(3)}) \quad (21)$$

$$\dot{Q}_{cool} = \dot{n}_{fluidB} h_{evap} \quad (22)$$

The coefficient of performance of the entire system can then be calculated as the ratio of the cooling capacity to the sum of the work inputs as shown in Equation 23. Because the electrochemical work input will be negative, its absolute value is taken. For the dual circuit arrangement the COP_C is determined using the sum of the cooling capacity of both evaporators and the sum of the work consumed by both electrochemical cells and pumps as shown in Equation 24.

$$COP_C = \frac{\dot{Q}_{cool}}{\dot{W}_{pump} + abs(\dot{W}_{cell})} \quad (23)$$

$$COP_C = \frac{\dot{Q}_{cool,1} + \dot{Q}_{cool,2}}{\dot{W}_{pump,1} + \dot{W}_{pump,2} + abs(\dot{W}_{cell,1} + \dot{W}_{cell,2})} \quad (24)$$

4. SYSTEM COMPARISONS

4.1 CLHP Working Fluids

Using the aforementioned CLHP models the relative performance of various working fluid pairs were evaluated. The following inputs were provided to the model:

- $T_{source} = 20\text{ }^{\circ}\text{C}$
- $T_{sink} = 35\text{ }^{\circ}\text{C}$
- $T_{approach} = 5\text{ }^{\circ}\text{C}$
- $\eta_{pump} = 0.7$
- $\eta_{cell} = 0.6$

The efficiency of the electrochemical cell will depend on the current density through the device. The value of 60% represents a level of performance that can be achieved by various fuel cells. Following the nomenclature from Table 1 the calculated COP_C of the various fluid combinations using the dual circuit embodiment are shown in Figure 4. For the instances where the same fluid pairs are in both circuits, the single circuit CLHP can be used to produce the same results. Overall the performance of the CLHP degrades when different fluid pairs are used in combination with each other. This is due to an increase in the required voltage to drive the reactions. For the dual circuit CLHP the total voltage is taken as the sum of the voltages of the two cells. As shown by Figure 5 the overall voltage requirement increases when multiple fluid pairs are used consecutively. This is due in part to the differences in the oxidation and reduction potentials of the chemical species.

Of the working fluids investigated isopropanol, ethanol, and methanol produce the highest COPs when paired with themselves. As such a single circuit CLHP employing these fluids would be the most promising CLHP system due to performance and reduced complexity.

	MTH																		
MTH	6.93	ETH																	
ETH	1.298	7.59	IPA																
IPA	0.969	2.587	8.104	BUT															
BUT	0.932	2.188	6.136	5.374	PENT														
PENT	0.921	1.988	4.53	5.34	5.31	PGLY													
PGLY	1.0428	1.759	1.728	1.593	1.521	1.177	OXYL												
OXYL	0.172	0.2048	0.292	0.316	0.348	0.287	4.374	TETRA											
TETRA	1.09	5.43	1.81	1.586	1.48	1.483	0.157	6.235	ANIL										
ANIL	0.167	0.1917	0.274	0.295	0.323	0.272	1.368	0.153	0.946										


low COP  high COP

Figure 4: Cooling Coefficient of Performance for dual circuit CLHPs operating between 20°C and 35°C

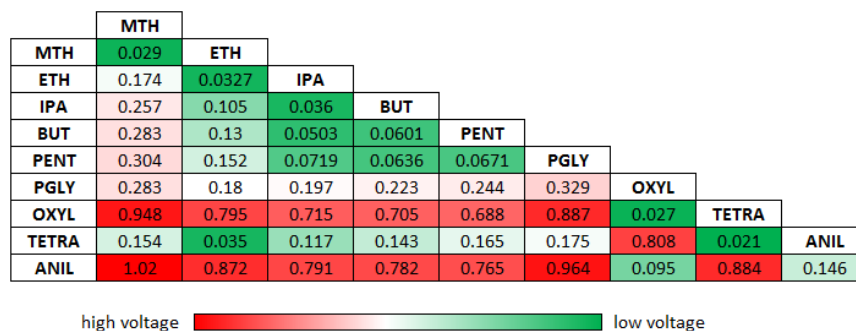


Figure 5: Voltage requirements of dual circuit CLHP operating between 20°C and 35°C

4.2 Electrochemical Systems

As previously mentioned, a number of electrochemically driven heat pump systems have been previously investigated. Being that these systems share some similarities with the CLHP, the performance of the CLHP relative to these systems was assessed. Electrochemical heat pumps are typically highly sensitive to component efficiencies. This is due to the fact that the electrical power cycled between cells in these system is typically orders of magnitude higher than the heat pumped through the system (Gerlach, 2004).

Using the same source, sink, and approach temperatures as the previous section, Figure 6 shows the response of COP_c to the efficiency of the electrochemical cells for the direct electrochemical heat pump of Newell (1997), the indirect electrochemical system using the evolution of a condensable gas of Gerlach (2004), and a single circuit CLHP using ethanol/acetaldehyde. Here the cell efficiencies are defined as in Equations 15 & 16.

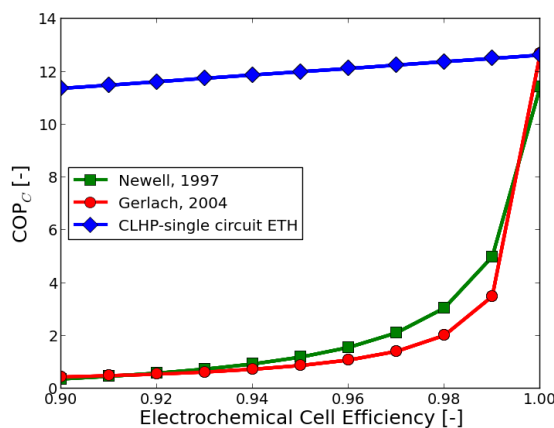


Figure 6: Performance of various electrochemical systems in response to electrochemical cell efficiency

A notable difference between the PEM CLHP and the other electrochemical heat pumps is that the performance of the PEM CLHP is not highly sensitive to component efficiency. This is due to the fact that in the CLHP the product of one side of the electrochemical cell is feed in as the reactant to the other side. While such a flow configuration would be counterproductive in a fuel cell power generation or electrochemical synthesis application, this greatly reduces the voltage requirements of the cells for the CLHP. By bringing the magnitude of the electrical power requirements closer to that of the heat transferred through the system, the sensitivity to the component efficiencies decreases.

4.3 Conventional Systems

Vapor compression (VC) systems account for the majority of heat pumping systems used worldwide. In order to see if the CLHP can serve as a viable alternative to VC the performance of the CLHP was compared to that of a VC system. For the VC heat pump the compressor was modeled using a constant isentropic efficiency of 75%. The temperatures at the outlets of the evaporator and condenser were determined according to equations 25 and 26

assuming 5 °C approach temperatures. The superheat out of the evaporator was assumed to be 1.0 °C and no sub-cooling was assumed after the condenser. For the CLHP the electrochemical cells were assumed to operate at 60% efficiency.

$$T_{evap} = T_{source} - T_{approach} \quad (25)$$

$$T_{cond} = T_{sink} + T_{approach} \quad (26)$$

The sink temperature was fixed at 35°C. The COP_C of the VC and CLHP systems are shown in Figure 7 over a range of source temperatures. For the working fluids investigated the CLHP can provide COP_C improvement of over 20% in comparison to the VC heat pumps for the assumed parameters and conditions.

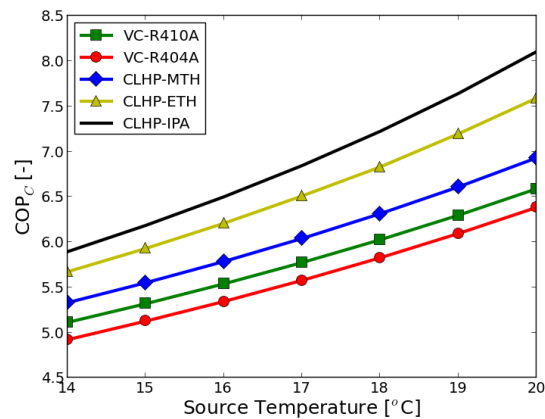


Figure 7: COP_C of vapor compression and CLHP systems vs source temperature for sink temperature of 35°C

5. CONCLUSIONS

A concept for a chemical looping heat pump has been presented. The operation of the system using a PEM electrochemical cell was modeled for single and dual circuit arrangements. A variety of working fluid pairs have been evaluated for use in the CLHP with Methanol/Formaldehyde, Ethanol/Acetaldehyde, and Isopropanol/Acetone showing the highest performance. Due to the voltage reduction caused by of the product stream returning as a reactant to the other side of the cell the CLHP is more tolerant to device inefficiencies than previously investigated electrochemical heat pumping devices. The CLHP has the potential to surpass the performance of typical vapor compression heat pumps by over 20%. Further experimental work will need to be done to prove the viability of using the CLHP for high efficiency heat pumping.

NOMENCLATURE

c_p	specific heat capacity	(J/mol-K)
F	Faraday's constant	(Col/mol)
g	Gibbs free energy	(J/mol)
h	enthalpy	(J/mol)
h_f^o	enthalpy of formation	(J/mol)
I	current	(A)
\dot{m}	mass flow rate	(kg/s)
n	number of moles	(-)
\dot{n}	molar flow rate	(mol/s)
η	efficiency	(-)
P	pressure	(Pa)
P^o	reference pressure	(Pa)
\dot{Q}	heat flow rate	(W)
R	universal gas constant	(J/mol-K)

s	entropy	(J/mol-K)
s_f^0	entropy of formation	(J/mol-K)
T	temperature	(K)
T^0	reference temperature	(K)
v	specific volume	(m ³ /kg)
V	voltage	(V)
V^0	reversible voltage	(V)
w^0	molar work	(J/mol)
\dot{W}	power	(W)

Subscript

cell	electrochemical cell	cond	condenser
cool	cooling load	e	electrons
evap	evaporator	H ₂	hydrogen
Liq	liquid	sat	saturation
vap	vaporization		

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