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Thermochemical Heat Transformer for Waste Heat Recovery: consideration of dynamic aspects for design

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ABSTRACT

Waste Heat Recovery in the industrial sector often requires to upgrade the temperature of the heat fluxes in order to make them useful for the process. Thermochemical Heat Transformer can play a major role as they can provide high temperature lift when compared to alternative technologies.

The aim of the presented work is twofold:

- to identify suitable working pairs using water as working fluid taking into account three criteria: COP, temperature lift and equilibrium drop (difference of the operating temperature and pressure to their value at equilibrium)
- to run a parametric study, thanks to a dynamic 2-salt/2-reactor model, to identify optimal specific heat power as a function of cycle time, working pairs and reactor heat transfer coefficient.

For waste heat temperature at 90 °C and a cold source at 30 °C, the CaCl₂/H₂O and Ca(NO₃)₂/H₂O working pairs were identified as promising. The achieved temperature lift is as high as 90 K for a theoretical COP of 0.42. In dynamic operating, a specific heat power of 325 W/kg was found.

Keywords: Thermochemical Heat Transformer, Waste Heat, Salt screening

1. INTRODUCTION

As reported by Ademe, waste heat valorization is a promising solution to dramatically increase the energy efficiency of the industrial sector (ADEME, 2015). According to this report, heat to electricity conversion (e.g. ORC) becomes competitive for temperatures of recovered heat above 150 °C whereas a huge amount of waste heat is available at lower temperature. While sorption systems are industrially available for heat to cold conversion, there is today a great challenge for heat to heat conversion, i.e. to upgrade waste heat temperature from 90-100°C to 150°C and above.

Compared to Electrical Heat Pumps (EHP) and Absorption Heat Transformers (AHT), Thermochemical Heat Transformer (THT) is a far less mature technology (Li et al., 2013; Wu et al., 2017). However, THT could potentially tackle the issues of EHP and AHT by achieving high temperature lifts with 1-stage heat driven system without any crystallization or corrosion issues (Li et al., 2013). Furthermore, 2-salt technology (Figure 1a), also called resorption heat pump, allows a reduction of the high-pressure lift occurring in AHT when high temperature lifts are sought (Yu et al., 2008). In this case, the evaporator and the condenser are replaced by two reactors filled with a so-called low temperature salt (LT salt). Salt/ammonia pairs have been extensively studied for THT applications while there have been some evaluations of salt/methanol, salt/H₂O and salt/CO₂ pairs with encouraging results for the latter two (Yu et al., 2008). In such systems, a main issue is to select the appropriate salt pair for given operating temperatures (T_L , T_m & T_H), to achieve high performance.

Hence, the first objective of this paper is to perform a salt screening for industrial waste heat upgrade thanks to a static thermodynamic approach. The second objective is to study the dynamic behavior of the THT for pre-selected pairs. The model developed and validated for this purpose is presented as well. Furthermore, it was chosen to focus on water as working fluid.

2. STATIC THERMODYNAMIC APPROACH FOR SALT SCREENING

In this section, a static thermodynamic approach is used to perform a rapid screening of available salt-H₂O pairs (N'Tsoukpoe et al., 2014; Visscher et al., 2004). It is based on the following set of equations. Eq. 1 states the hydration equation of a salt (S) by a vapour (G). The reaction equilibrium is given by Eq. (2), where p_{eq} is the equilibrium pressure at T_{eq} as shown in Eq. (3).

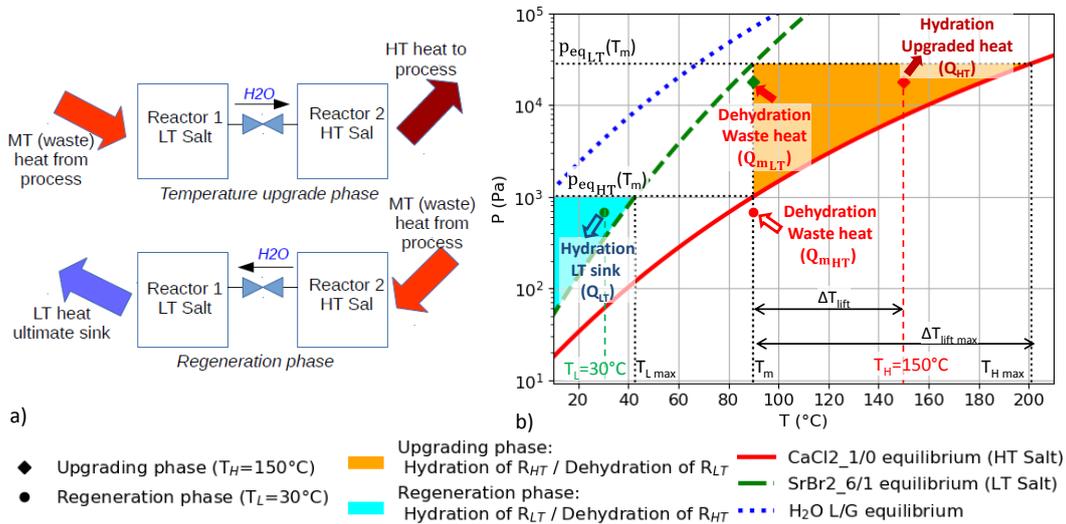


Figure 1: a) Working principle of a two-salt THT. b) Clapeyron diagram representation of the operating areas during hydration and dehydration for the CaCl₂/SrBr₂ pairs.

$$S + vG = S' + v\Delta h_r^0 \quad \text{Eq. (1)}$$

$$\Delta g_r = \Delta g_r^0 + RT_{eq} \ln\left(\frac{p_{eq}}{p^0}\right)^v = v\Delta h_r^0 - vT_{eq}\Delta s_r^0 + RT_{eq} \ln\left(\frac{p_{eq}}{p^0}\right)^v = 0 \quad \text{Eq. (2)}$$

$$p_{eq} = p^0 e^{-\frac{\Delta h_r^0}{RT_{eq}} + \frac{\Delta s_r^0}{R}} \quad \text{Eq. (3)}$$

For the sake of clarity, the work presented in this paper is focused on given heat source temperatures representative of operating conditions suitable for heat upgrading in the industry. The cold source temperature is chosen at $T_L = 30^\circ\text{C}$, the waste heat temperature at $T_m = 90^\circ\text{C}$ and a high temperature source at $T_H = 150^\circ\text{C}$.

As shown on Fig. 1a, during the regeneration phase (phase 1), waste heat at T_m is supplied to the HT reactor and the LT salt is connected to a heat sink at T_L . In order to allow simultaneously the hydration of the LT salt and the dehydration of the HT salt, the operating conditions (vapor pressure and temperature) of the LT reactor must be in the blue zone presented on Fig. 1b. Therefore, this reactor has to be connected to a heat sink at $T_L < T_{L,max}$ (i.e. the equilibrium pressure of the LT salt at T_L must be inferior to the equilibrium pressure of the HT salt at T_m). In the case of T_L is superior to $T_{L,max}$, the operating conditions do not allow the hydration of the LT salt and the dehydration of the HT salt. At the end of this phase, the reactive LT salt is fully hydrated and the HT salt is fully dehydrated.

During the temperature upgrade phase (phase 2), the LT reactor is connected to the waste heat at T_m . In order to achieve the dehydration of the LT reactor and the hydration of the HT reactor, the operating condition of the HT reactor must be in the orange zone of the Fig. 1b, thus the HT reactor temperature must be inferior to $T_{H,max}$ (i.e. the equilibrium pressure of the LT salt at T_m have to be superior the equilibrium pressure of the HT salt at T_H). Therefore, this phase allows to supply heat at high temperature (T_H). Finally, at the end of this phase, the reactive LT salt is fully dehydrated and the HT salt is fully hydrated, requiring the beginning of a new regeneration phase.

From these considerations, a simple numerical tool was developed to perform a first selection for LT and HT salts by checking whether the temperature conditions are satisfied ($T_L < T_{L,max}$ and $T_H < T_{H,max}$). 77 pairs were identified from 66 initial reactions.

A second step to discriminate between possible pairs is to consider the achievable performance in terms of COP and maximum temperature lift.

Using the thermodynamic considerations previously presented, neglecting heat accumulation by the salts (heat capacities are low compared to reaction enthalpies) and assuming that there are no heat losses, the theoretical maximal COP of the 2-salt THT is given by:

$$COP_{max} = \frac{Q_{HT}}{Q_{m_{LT}} + Q_{m_{HT}}} = \frac{\Delta h_r^0 S_{HT}}{\Delta h_r^0 S_{LT} + \Delta h_r^0 S_{HT}} \quad \text{Eq. (4)}$$

With $\Delta h_r^0 S_n$ the reaction enthalpy of the salt n (in J/mol_v).

The maximal temperature lift is achieved for adiabatic operation and therefore corresponds to the theoretical maximum lift attainable by the HT salt during the temperature upgrade phase: $T_{Hmax} - T_m$. For the targeted high and waste heat temperatures (150 & 90 °C), a minimum temperature lift of 60 K is then required. To take into account the temperature pinches required in the HT reactor for the heat transfer to occur in finite time, only pairs achieving a minimum temperature lift of 80 K are considered on Fig. 2.

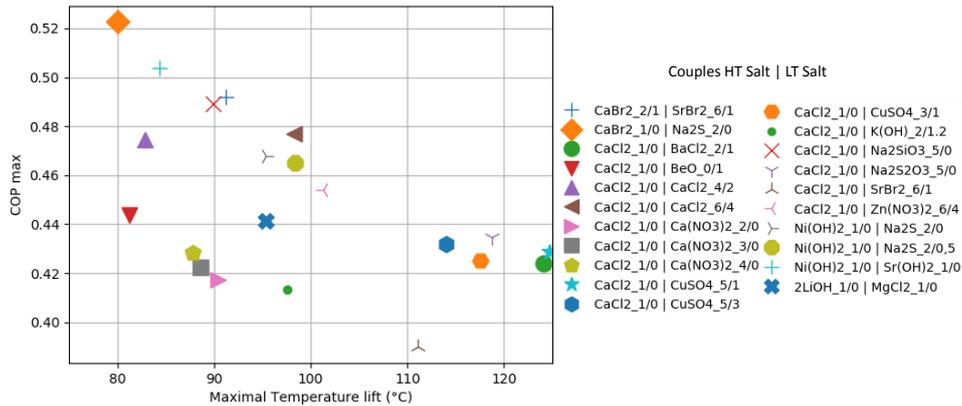


Figure 2: Maximal COP versus theoretical maximal temperature lift, for $T_{LT}=30^\circ\text{C}$ and $T_m=90^\circ\text{C}$.

From these results, 21 of the 77 promising reaction couples are kept. A large discrepancy between ΔT_{max} values is observed, from 80 K to 125 K (for the $\text{CaCl}_2/\text{CuSO}_4/\text{H}_2\text{O}$ pair) with a COP_{max} ranging from 0.39 to 0.52. Besides the static thermodynamic considerations, dynamic aspects have also to be taken into account in order to estimate specific heating power.

3. DYNAMIC MODEL

3.1 Governing equation

Several modeling of solid/gas reactor, with different approaches (local, global) and complexity levels (1/2D, stationary/dynamic, ...) have been presented in the literature (Castaing-Lasvignottes and Neveu, 1997; Marias et al., 2014; Nagel et al., 2016). Most of the reported works refer to cooling or heat storage applications and only few models deal with THT applications, especially for those based on 2-salt cycles.

Thus, from modeling approaches of a solid/gas reactor developed by Lebrun and Spinner (1990), a simple and original model of 2-salt THT reactor has been developed in the frame of this work. It should allow to study the dynamic behavior to estimate its performances, mainly heating capacity, and temperature lift. The following assumptions are made:

- Uniform temperature (T_{SLT} and T_{SHT}) and local thermal equilibrium between the gas and solid phases is assumed in the reactors.
- Heat accumulation in the gas phase and heat losses are neglected.
- The heat capacities are considered constant during the reaction.
- In closed thermochemical systems (working with pure gas), the mass transfer usually does not limit reaction kinetic. Thus, the vapor pressure is assumed uniform inside the reactor.
- The assumptions (a) and (b) imply that the salt reaction advancement (X_s) is uniform in each reactor. X_s equals 0 when the salt is hydrated and 1 when is dehydrated
- The kinetics of the solid/gas reactions depends only on the reaction advancement and on the equilibrium drop. This drop corresponds to the difference of the operating temperature and pressure to their value at equilibrium.

3.1.1 Energy balance

According to assumptions (a), (b) and (c), the energy balances of the HT and LT salt reactors are respectively written as:

$$\left[m_{S_n} \left(1 - (1 - X_{S_n}) v_{S_n} \frac{M_v}{M_{S_n}} \right) c_{S_n} + m_{exch_n} c_{exch_n} \right] \frac{dT_{S_n}}{dt} = \dot{q}_{S_n} + \dot{m}_{htfn} c_{htfn} (T_{ihtfn} - T_{otfn}) \quad \text{Eq. (5)}$$

with:
$$\dot{q}_{S_n} = -\dot{m}_{v_{S_n}} \Delta h_{r_{S_n}}^0 = v_{S_n} \frac{dX_{S_n}}{dt} \frac{m_{S_n}}{M_{S_n}} \Delta h_{r_{S_n}}^0 \quad \text{Eq. (6)}$$

With $\dot{m}_{v_{S_n}}$ the source ($\dot{m}_{v_{S_n}} > 0$ in dehydration phase) or sink ($\dot{m}_{v_{S_n}} < 0$ in hydration phase) of vapor due the reaction of the salt n .

The outlet temperature of the heat transfer fluid is given by:

$$T_{otfn} = \frac{(T_{ihtfn} - T_{S_n})}{\frac{U A_{S_n}}{e \dot{m}_{htfn} c_{htfn}}} + T_{S_n} \quad \text{Eq. (7)}$$

3.1.2 Kinetics of the solid/gas reaction

According to the assumption (f), the reaction kinetic depends on the reaction advancement and on the equilibrium drop. Mazet et al., (1991) have shown that it can write as a 1st order kinetic law:

$$\frac{dX_{S_n}}{dt} = k_{cin_{S_n}} Y_{S_n} \left(1 - \frac{p_{eq}(T_{S_n})}{p_v} \right) \quad \text{Eq. (8)}$$

where: $Y_{S_n} = (1 - X_{S_n})$ in hydration phase and $Y_{S_n} = X_{S_n}$ in dehydration phase.

3.1.3 Operating vapor pressure

The vapor operating pressure of the interconnected HT and LT salt reactors, p_v , is defined such as the vapor mass flow rate caught/released by the HT salt reactor is equal to the vapor mass flow rate respectively released/caught by the LT salt reactor. Thus, using Eq. (6) and Eq. (8), the vapor pressure writes:

$$p_v = \frac{A p_{eq_{S_{HT}}}(T_{S_{HT}}) - p_{eq_{S_{LT}}}(T_{S_{LT}})}{A - 1}, \text{ with: } A = - \frac{m_{S_{HT}} M_{S_{LT}} v_{S_{HT}} k_{cin_{HT}} (1 - X_{S_{HT}})}{m_{S_{LT}} v_{S_{LT}} k_{cin_{LT}} X_{S_{LT}} M_{S_{HT}}} \quad \text{Eq. (9)}$$

Note that in case of the kinetic coefficients of both reactive salts are equals and if the salts masses are chosen in a balanced way (i.e. in a way that the overall of both reactive salts could react, cf. § 3.2), we have $A = -1$. Therefore, the operating vapor pressure is equal to the average of the equilibrium pressure of each salt:

$$p_v = \frac{p_{eq_{S_{HT}}}(T_{S_{HT}}) + p_{eq_{S_{LT}}}(T_{S_{LT}})}{2} \quad \text{Eq. (10)}$$

3.2 Model parameters

3.2.1 Reactor heat transfer coefficient (UA)

The global heat transfer coefficient of the reactor has been identified from experimental results of Esaki et al., (2017) obtained with a prototype of THT using $\text{CaCl}_2/\text{H}_2\text{O}$ as working pair. Although UA depends on the reactor geometry and on the reactive material used, we will suppose in this paper, that the change of the reactor scale and reactive material does not affect the heat transfer characteristics. The following specific heat transfer coefficient has been evaluated:

$$\frac{U A_{S_n}}{m_{S_n}} = 131.36 e^{-3.35 X_{S_n}} \quad \text{Eq. (11)}$$

3.2.2 Kinetic parameter

The kinetic constant, k_{cin} , lumps the heat and mass transfers in the solid pores and the intrinsic kinetics of the reaction. A value of $68.10^{-4} \text{ s}^{-1}$ has been identified from an experimental hydration of a THT using CaCl_2 as reactive salt (Esaki et al., 2017).

As shown by Michel et al. (2014), this value indicates that the kinetic is not the main limitation of the thermochemical reaction. In the following, this constant value of k_{cin} has been used for all the studied working pairs.

3.2.3 Mass of salts

In order to avoid that one of the two reactive salts limits the reaction of the other one, the mass of each salts should be chosen in a way that both reactive salts could totally react. Thus, each salt must allow to catch (respectively release) the water vapor released (respectively caught) by the other salt:

$$m_{v_{HT}} = m_{v_{LT}} \Leftrightarrow v_{S_{HT}} \frac{m_{S_{HT}}}{M_{S_{HT}}} = v_{S_{LT}} \frac{m_{S_{LT}}}{M_{S_{LT}}} \Rightarrow m_{S_{LT}} = m_{S_{HT}} \frac{v_{S_{HT}} m_{S_{HT}}}{v_{S_{LT}} M_{S_{HT}}} \quad \text{Eq. (12)}$$

3.3 Model validation

The model has been validated using experimental results of a $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ THT prototype (Esaki et al., 2017). Fig. 3 presents simulations that fit adequately to experimental results. The difference between experimental and simulated reaction rates and outlet heat transfer fluid temperature is rather low: at $X=0.5$, it is respectively of 9% and 0.5%. These results demonstrate that this simple model predicts in a satisfactory way the thermal behavior of a reactor.

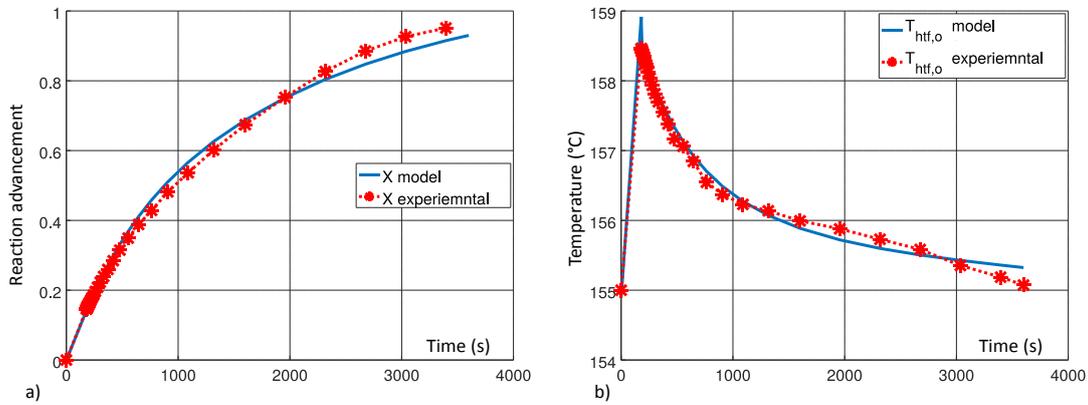


Figure 3: Experimental and simulated results of the hydration of the THT (A type reactor, Esaki et al., 2017)) a) Reaction advancement vs. time during. b) Outlet heat transfer fluid temperature vs. time.

4. SALT SELECTION CONSIDERING DYNAMIC ASPECTS

In a real cycle an equilibrium drop must be considered in order to realize the thermochemical reaction. This parameter influences the heat and mass transfer rates and therefore the reaction kinetics (Mazet and Lu, 1998; Obermeier et al., 2015). Thus, any change in the operating temperature (T_L , T_m or T_H) results in a change in the equilibrium drop and then in the reaction kinetic. However, as indicated by Obermeier et al. (2015), the equilibrium drop is not an independent parameter and equal values do not produce the same heat and mass transfer rates for different reaction systems.

This parameter can be related to the chemical potential difference which represents the thermodynamic driving force of the reaction. Usually, the greater the driving force, the greater the reaction kinetic is. Under the assumptions of pure substances and ideal mixtures, the driving force can be described by the Gibbs free energy of the reaction (Obermeier et al., 2015). Thus according to the eq. (2) and (3) this parameter can be writes as:

$$F_m = \frac{\Delta G_r}{vRT} = \ln\left(\frac{p_v}{p_{eq}}\right) \quad \text{Eq. (13)}$$

In order to take into account the thermal resistances inside the reactors of a 2-salts THT, the reactor temperature must be evaluated for each reactor. Thus, assuming that the heat accumulation is negligible compared to the reaction enthalpy, the eq. (6), (7) and (8) lead:

$$T_{S_n} \left(1 - e^{\frac{-UA_{S_n}(T_{ohf_n} - T_{ihtf_n})}{k_{cin_{S_n}} Y_{S_n} \left(1 - \frac{p_{eq}(T_{S_n})}{p_v} \right) \Delta h_{r_{S_n}}^0}} \right) - T_{ohf_n} + T_{ihtf_n} e^{\frac{-UA_{S_n}(T_{ohf_n} - T_{ihtf_n})}{k_{cin_{S_n}} Y_{S_n} \left(1 - \frac{p_{eq}(T_{S_n})}{p_v} \right) \Delta h_{r_{S_n}}^0}} = 0 \quad \text{Eq. (14)}$$

Thus, assuming a reaction advancement of 0.5, determining the vapor pressure using eq. (10) and imposing a heat transfer fluid pinch (5°C for the HT reactor and 1°C for LT reactor), the temperature of each reactor has been determined and then the reaction driving forces have been evaluated.

Fig. 4 presents a comparison of the driving force of the hydration of the LT and HT reactors for different reactive couples (i.e. a comparison of the driving force during the regeneration and the upgrading temperature phases). A large discrepancy between F_m values is observed, from 0.001 to 0.35. However, promising reactive pairs are identified, as $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$, $\text{CaCl}_2/\text{SrBr}_2$ and $\text{CaCl}_2/\text{K}(\text{OH})$. They present high driving forces whose values are close between each cycle phase. This allows to avoid large reaction times gap between these phases.

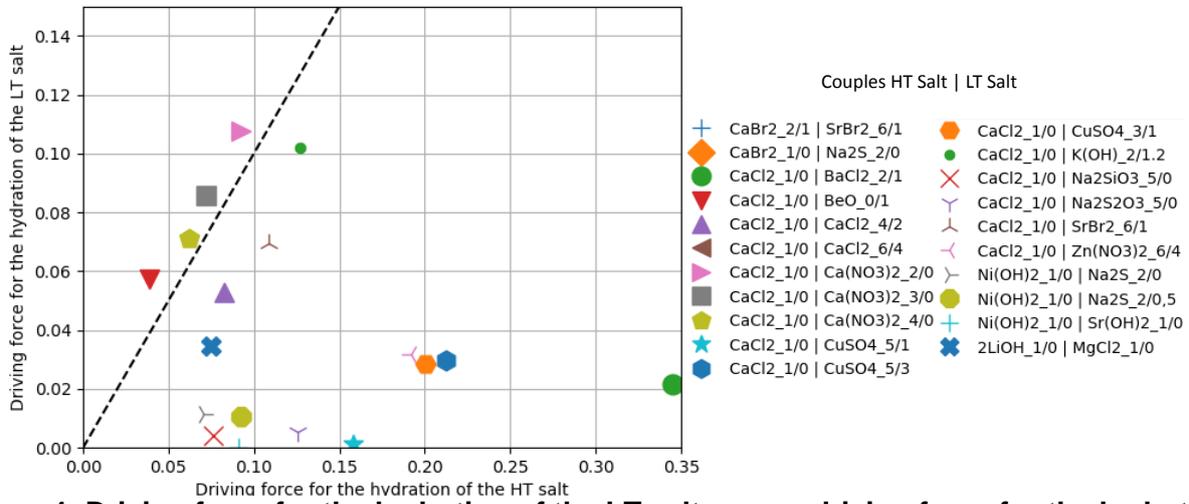


Figure 4: Driving force for the hydration of the LT salt versus driving force for the hydration of the HT salt (for $X_{S_n}=0.5$, $T_L=30^\circ\text{C}$, $T_m=90^\circ\text{C}$, $T_H=150^\circ\text{C}$, $\Delta T_{htf,HT}=5^\circ\text{C}$ and $\Delta T_{htf,LT}=1^\circ\text{C}$).

5. CYCLE PERFORMANCES

This part aims at analyzing the dynamic behavior and performances of a 2-salt THT. Three key parameters allowing to define the performance of the system are studied: the system heating power (during the upgrading phase), the COP and the temperature lift. The cyclic behavior of the two LT and HT reactors is simulated with successive heating and cooling. The results are given at cyclic steady state and optimal cycle time (maximum heating power).

5.1 Dynamic behavior

The dynamic behavior for $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$ and $\text{CaCl}_2/\text{BaCl}_2$ reactive couples is presented Fig. 5a and b. Maximal specific heat power respectively of 325 W/kg and 300 W/kg have been reached during the temperature upgrade phase, corresponding to average specific heat power of 49 W/kg and 26.9 W/kg. Furthermore, due to the system regulation, the temperature and specific heat power are not stable, especially during the temperature upgrade phase.

In agreement with the driving forces presented in the Fig. 4, the reaction advancement evolution shows that the reaction kinetic of each phase is quite equilibrated for the $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$ THT, while the temperature upgrade phase is much faster than the other phase for the system using the $\text{CaCl}_2/\text{BaCl}_2$ couple (30 min instead of 1.5h). In this last case, the system performances are therefore limited by the regeneration phase, especially due to the adverse operating conditions.

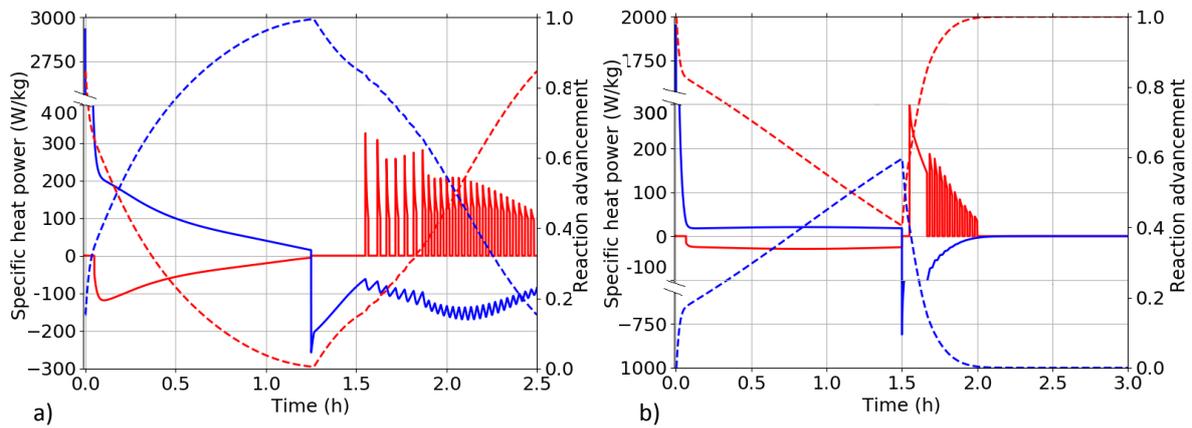


Figure 5: Specific heat power and reaction advancement versus time of 2-salt THT operating with: a) $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$ working pairs. b) $\text{CaCl}_2/\text{BaCl}_2$ working pairs.

5.2 Sensitivity to operating temperatures

Simulations have been performed to study the sensitivity of the operating temperatures. Their influence to the average specific heat power and the reaction driving forces is presented on Fig. 7. The impact of the operating temperatures clearly depends to the driving force values. Thus, although the diminution of T_L increases the driving force associated to the generation phase, no significant change of the specific heat power is observed. As contrary, the increase of T_H (and then of the driving force associated to the temperature upgrade phase) allows in a first time to significantly increase the system performances. This is because in the reference case, the temperature upgrade phase corresponds to the lower driving force value. This phase is therefore the limiting phase, which is subject to the most unfavourable conditions on which it is necessary to play in priority.

Other limitations, as the heat transfer or the kinetic reaction can impact the system performances. However, this is out the scope of this paper. Note that in the studied cases, these parameters are weakly limiting.

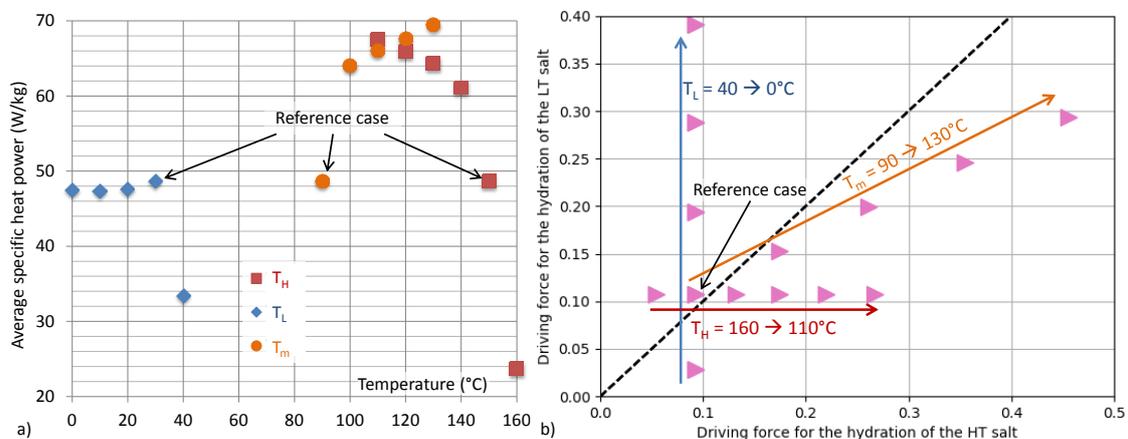


Figure 6: Impact of the operating temperatures. a) Evolution of the driving forces. b) Specific heat power release during the upgrading temperature phase. Case of the $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$ reactive couples.

6. CONCLUSION

A screening of salt couples was performed to design a 2-salt Thermochemical Heat Transformer for waste heat recovery. For the sake of clarity a representative case was considered with a low temperature source at 30 °C, an intermediate at 90 °C and a high temperature source at 150 °C. A first tool was developed to discriminate pairs using a static thermodynamic approach insuring cycle feasibility. From the selected 77 pairs, 21 were kept considering a minimum requirement in temperature lift. The last step was based on dynamic consideration (driving force) and allows to identify three candidates: $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$, $\text{CaCl}_2/\text{SrBr}_2$ and $\text{CaCl}_2/\text{K}(\text{OH})$.

Cycle operation was simulated for the most promising pair $\text{CaCl}_2/\text{Ca}(\text{NO}_3)_2$. An average specific heating power as high as 49 W/kg is achieved.

ACKNOWLEDGEMENT

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NOMENCLATURE

c	heat capacity ($\text{J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$)	Δs_r^0	standard entropy of reaction ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)
COP	coefficient of performance (-)	ν	stoichiometric coefficient (mol/mol_s)
F_m	Driving force (-)	<i>Indices</i>	
k_{cin}	kinetic constant (s^{-1})	<i>eq</i>	solid/gas equilibrium
M	molar weight ($\text{kg}\cdot\text{mol}^{-1}$)	<i>exch</i>	exchanger
m	mass (kg)	<i>i</i>	inlet
\dot{m}	mass flow rate ($\text{kg}\cdot\text{s}^{-1}$)	<i>htf</i>	heat transfer fluid
\dot{m}_v	sink or source of gas ($\text{kg}\cdot\text{s}^{-1}$)	<i>H</i>	high temperature
p	pressure (Pa)	<i>HT</i>	high temperature salt
P_m	specific power ($\text{W}\cdot\text{kg}^{-1}$)	<i>L</i>	low temperature
\dot{q}	sink or source of heat ($\text{J}\cdot\text{s}^{-1}$)	<i>LT</i>	low temperature salt
Q	heat exchanged (J)	<i>m</i>	medium
R	ideal gas constant ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)	<i>max</i>	maximum
T	temperature (K or $^\circ\text{C}$)	<i>n</i>	HT or LT salt
UA	heat exchange coefficient ($\text{W}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$)	<i>v</i>	vapor
X	reaction advancement (-)	<i>s</i>	salt
<i>Greek symbols</i>		<i>o</i>	outlet
Δg_r	free Gibbs energy ($\text{J}\cdot\text{mol}^{-1}_s$)	<i>Exponents</i>	
Δh_r^0	standard enthalpy of reaction ($\text{J}\cdot\text{mol}^{-1}_v$)	<i>0</i>	reference

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