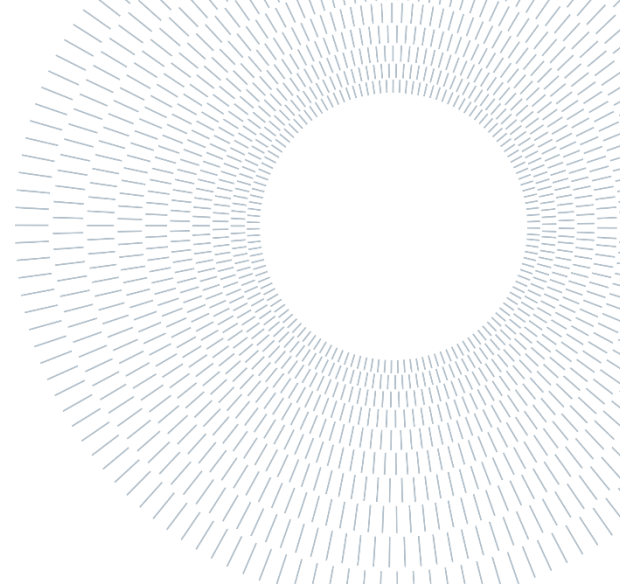




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EXECUTIVE SUMMARY OF THE THESIS

Investigation and Design of Novel Peak Shaving District Heating Systems based on Thermochemical Materials

TESI MAGISTRALE IN ENERGY ENGINEERING – INGEGNERIA ENERGETICA

AUTHOR: POUYA JAVANMARD

ADVISOR: PROF. ANDREA GIOSTRI

EXTERNAL ADVISORS: DR.IR. AMIR MAHMOUDI, DR. MINA SHAHI

ACADEMIC YEAR: 2021-2022

1. Introduction

The Energy sector is the source of around three-quarters of greenhouse gas emissions today. The IEA (International Energy Agency) has set a global pathway to net-zero emissions by 2050 which requires all the governments to strengthen and then successfully implement their energy and climate policies. Switching to cleaner renewable energy technologies has a key role in this roadmap [1].

Currently, domestic heating for the households is one of the major end uses of fossil fuel (fig1) [2]. In fact, heat sources like gas boilers or co-generation plants which are largely based on natural gas are widely used for this application. There is a need to derive sustainable heat from alternative sources.

The Netherlands established a National Climate Agreement (Klimaatakkoord) in 2019, targeting for removing natural gas as the main heat source for over 7 million households by 2050. A new sustainable district heating network will be developed and initially, 1.5 million households

must connect to the new heating network by 2030[3].

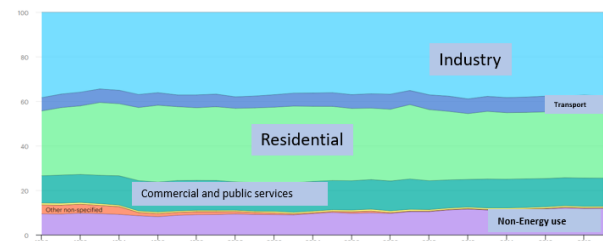


Fig1. World final Natural Gas consumption by sector (in percentage), 1990-2019 [2]

Seasonal heat storage using a thermochemical material (TCM) is a novel heat storage technology which can transform the conventional district heating network to a newer, cleaner and more sustainable network. This system can be charged during the summer period with renewable sources (e.g. solar) and then discharged in the winter to provide the required heat to the households for domestic use.

The aim of this study is to investigate the TCM based heat storage system, design and optimize a novel reactor for this application using the COMSOL Multiphysics® software.

2. Methodology

First, a literature review was done to understand the different configuration types and components for thermochemical heat storage technologies and their comparison. Then, a numerical investigation of the model was performed to explain the kinetic modelling and governing equations.

Next, a basic 2D model was designed and sensitivity analysis was made to figure out the main design parameters controlling this model.

Later, based on the studies and the basic modelling, two possible designs were proposed for the reactor. The results were processed for each model, together with the related analyzes and explanations. Also, improvement procedure of these models was discussed and finally, some relevant comparisons were done.

Finally, all the main conclusions obtained during the study were explained and some relevant recommendations on the designing were discussed to have a better engineering view for future studies.

3. Theory

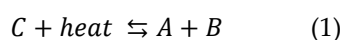
Thermal energy storage is divided in three main types including Sensible, Latent and Thermochemical heat storage [4].

In Sensible heat storage system, energy (or heat) is stored/released by cooling/heating a liquid or solid storage material through a heat transfer interaction.

Latent heat storage is done thanks to the phase change of a material (e.g., evaporation, melting and crystallization) and the high enthalpy change during this process.

Thermochemical heat storages actually use a reversible chemical reaction to store thermal energy. This method has high energy density and allows long-term thermal energy storage [4].

The main principle of thermochemical heat storage is based on a reversible reaction as equation 1:



Where C is the TCM and can absorb energy and convert chemically into A and B. This is the charging process which is endothermic and

absorbs energy. After the charging, A and B are stored separately with little or no energy losses. Thermochemical heat storage has advantages like high storage density, low heat losses and possibility of charging in summer.

3.1. Open and Closed Systems

In an open system concept (figure 2), there is one vessel which is the reactor. This reactor contains the solid materials and the moist air at atmospheric pressure flows through it. The pressure is set to atmosphere pressure and there is no internal heat exchanger and no evaporator/condenser [5].

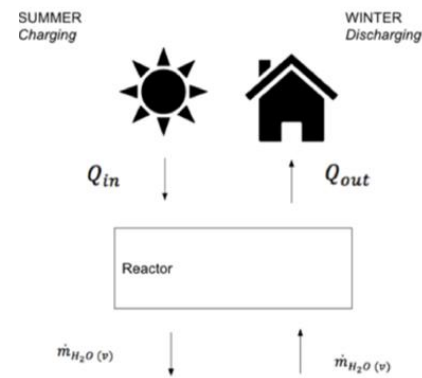


Fig 2: An integrated configuration for an open system

In a closed system, there are two vessels, a reactor which contains the material and a condenser/evaporator holding the reactive gas. Unlike open system, in closed system the heat energy is transferred via a heat exchanger. In closed system, material reacts with a pure gas. Fig 3 demonstrates a closed system configuration [5].

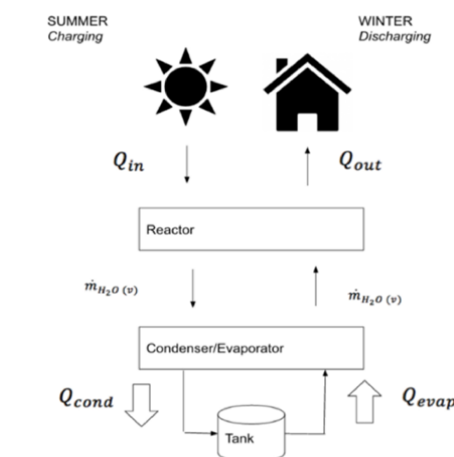


Fig 3: An integrated configuration for a closed system

3.2. Reactor Types

Reactors could be divided into fixed (packed) bed, moving bed or fluidized bed [6].

In Fixed bed reactors, the material is stationary inside a vertical vessel (Figure 4a). Their simple design and easier modelling have made them widely usable and so suitable for hydration/dehydration reactions due to the solid-gas states.

In Moving Bed Reactors, the bed which also contains the TCM salt moves with respects to the vessel (Figure 4b). These reactors have uniform fluid flow and high heat transfer coefficient whereas their design is more complex due to having moving parts.

In Fluidized bed reactors (figure 4c), the solid is in fine particles which are suspended by the upwards flow of the fluid(gas). When this upward velocity of the fluid goes higher than a minimum fluidization velocity, the solid particles start moving randomly [6].

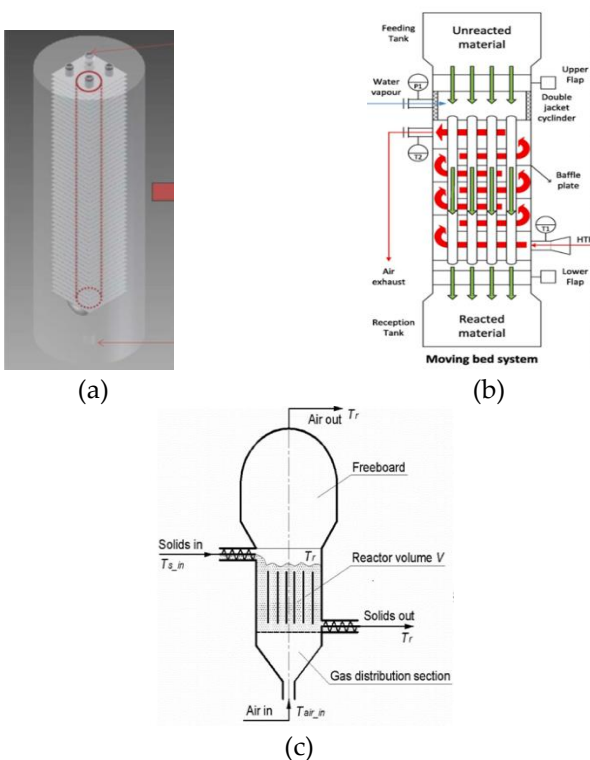


Fig4: Examples of fixed bed (a), moving bed(b) and fluidized bed reactors (c)

This study will focus on designing a closed system fixed bed reactor.

3.3. Heat Exchangers

There are quite a lot of heat exchanger designs and configurations, each with its own benefits and drawbacks. Several heat exchangers have been investigated in this study. Among them, shell and tube and helical coil have been chosen for designs. The first one consists of several tubes which are inside a cylindrical shell. One of the fluids can move inside the tubes and one of them inside the shell and heat transfer takes places between them. They are easy for maintenance, have flexible design and can also withstand some severe conditions. However, they may have a large footprint and low performance at low temperatures [7]. The latter has a shell called annulus and inside it, there is a helical coil. It occupies less space and provides more surface area for effective heat transfer as compared to shell and tube heat exchanger [8].

3.4. Heat Transfer Fluid

There are several deciding criteria to choose a suitable heat transfer fluid including high heat conductivity, low viscosity, large heat capacity, high working temperature range, nontoxicity, non-explosivity, easy replacement and cleaning and price [9]. Thermal Oils are too expensive for widespread use. Liquid metals and molten salts have high melting point problem.

Air and water have been concluded as two better options as they are both cheap and easy to model. Between them, water is preferred due to higher thermal conductivity [9].

3.5. TCM Material

The TCM is the material which involves in hydration/dehydration reactions in the reactor and gives heat to the HTF which can be finally used for space heating and hot tap water application. Donkers et al. collected thermodynamic data for 262 salts, investigated 563 reactions, introduced several filters to come up with the best possible salts for the application[10]. A summary of the salts analyzed by them has been reported in Table 1.

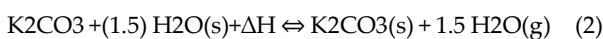
	Energy density open system (GJ/m ³)	Energy density closed system (GJ/m ³)	Price (euro/kg)	Point of Concern
GdCl ₃	2.70	1.56	R	Rare Earth
LiCl	2.08	1.36	37	Price
Ca(ClO ₄) ₂	1.75	1.17		Explosive
RbF	1.57	1.10	>10	Price
CaCl ₂	1.54	1.06	0.29	Deliquescence and higher hydrates
Mg(NO ₃) ₂	1.53	1.04		Instable
LaCl ₃	1.48	1.03	R	Rare Earth
K ₂ CO ₃	1.30	0.96	1	
MgCl ₂	1.93	1.24	0.18	HCl formation - Instable
Na ₂ S	1.60	1.14	0.65	H ₂ S formation-Safety/Instable

Table 1: characteristics of some possible TCM salts

The TCM selected for this study is Potassium Carbonate (K₂CO₃) as it is highly available, chemically robust, safe, cheap and not so much corrosive [10].

3.6. Reaction Kinetics

In this application, a reversible reaction of hydration of potassium carbonate (exothermic) and its dehydration (endothermic) takes place as equation 2 [11]:



A dependent variable called α (alpha) is defined as the status of conversion. This variable shows how much the conversion process of the solid material has been completed and it can range from 0 to 1. The reaction rate is generally formulated as equation 3:

$$\frac{d\alpha}{dt} = K(T) \cdot f(\alpha) \cdot h(P, P_{eq}) \quad (3)$$

where K defines the rate coefficient, $h(P, P_{eq})$ defines the pressure dependence and the function $f(\alpha)$ describes the reaction mechanism.

Mahmoudi et al. [11] performed TGA (Thermogravimetric Analysis) measurements for a thermochemical heat storage system based on potassium carbonate to evaluate the kinetics of K₂CO₃. Finally, they came up with equation 4 and the corresponding values of table 2:

$$\frac{d\alpha}{dt} = A_f \cdot e^{-\frac{E_a}{RT}} \cdot (1 - \alpha)^q \cdot \left(1 - \frac{P_{eq}}{P}\right) \quad (4)$$

A_f is the pre-exponential factor in 1/s,

E_a is the activation energy in J/mol

R is the universal gas constant in J/kg/K

T is the temperature in K

α is the status of conversion.

P_{eq} is the equilibrium water vapor pressure in Pa

P is the water vapor pressure in Pa

	A_f (1/s)	E_a (J/mol)	q
Hydration	2.7×10^{-9}	-34828	0.7
Dehydration	225	43382	0.8

Table 2: Experimental Values for parameters of reaction kinetics

The phase diagram of K₂CO₃ (Figure 5) is constructed based on the Clausius-Clapeyron equation (equation 5):

$$\frac{d \ln(P_{eq})}{d\left(\frac{1}{T}\right)} = -\frac{\Delta h}{R} \quad (5)$$

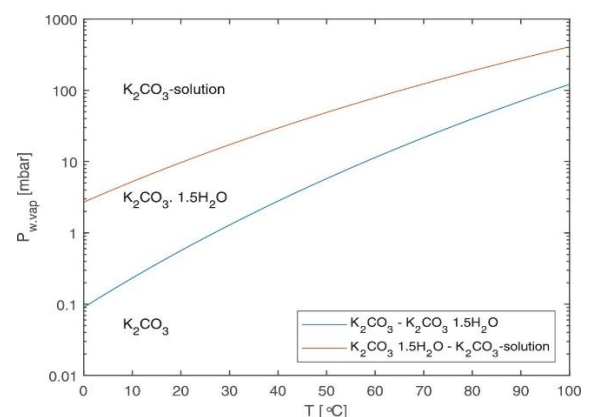


Fig 5: Phase diagram of K₂CO₃

The equilibrium pressure from this phase diagram can be fitted by the equation 6:

$$P_{eq} = 4.228 \times 10^{12} e^{-\frac{7337}{T}} \quad (6)$$

Porosity of a porous medium (equation 7) is defined as the void volume (in m³) divided by the total bulk volume of the medium (in m³).

$$\varepsilon_a = \frac{V_{pore}}{V_{bulk}} \quad (7)$$

3.7. Flow and Permeability Model

Reynolds Number for a porous media can be derived as equation 8 [12]:

$$Re = \frac{v_\varepsilon \sqrt{\kappa} \rho}{1750 \mu \varepsilon^{1.5}} \quad (8)$$

Where:

- v_ε is the real flow velocity in cm/s (and is obtained from v/ε)
- κ is the permeability in μm^2
- ρ is the density of the water vapor in g/cm³
- μ is the viscosity of the water vapor in mPa.s

In case $Re < 10$, the flow is darcian, meaning that it follows Darcy's law (equation 9) where the pressure gradient is a linear function of the flow velocity [12].

$$u = -\frac{\kappa}{\mu} \nabla p \quad (9)$$

Where u is the Darcy's velocity in m/s and κ is the permeability of the porous medium in m².

For this study, the values will be selected such that the flow becomes darcian.

Permeability is another fundamental parameter in the porous media which correlates to the porosity. Higher porosity which means higher void fractions, leads to higher permeability. For darcian flow, the Kozeny-Carman equation (equation 10) is used [12]:

$$\kappa = \frac{d_p^2}{180} \frac{\varepsilon_p^3}{(1-\varepsilon_p)^2} \quad (10)$$

Where d_p is the average particle diameter and ε_p is the porosity.

3.8. Energy Balances

Equations 11 and 12 show energy balance in heat transfer fluid and porous media respectively [12]:

$$\text{Heat Accumulation} = \text{Heat Conduction} + \text{Heat Convection} \\ C_{p,htf} \rho_{htf} \frac{\partial T}{\partial t} = \nabla \cdot (\lambda_{htf} \nabla T) - \rho_{htf} C_{p,htf} (u_{fd,htf} \cdot \nabla T) \quad (11)$$

Heat Accumulation = Heat Conduction + Heat Convection + Heat Source Term

$$(C_{p,s} (1-\varepsilon) \rho_s + C_{p,v} \varepsilon \rho_v) \frac{\partial T}{\partial t} = \nabla \cdot (K_{eff} \nabla T) - \\ \rho_v C_{p,v} (u_{fd} \cdot \nabla T) + (1-\varepsilon) \frac{\rho_{s0}}{M_{s0}} \frac{\partial \alpha}{\partial t} \Delta H_r \quad (12)$$

Where:

- $C_{p,htf}$ is the specific heat capacity of the HTF in J/(kg.K)
- ρ_{htf} is the density of the HTF in kg/m³
- λ_{htf} is the thermal conductivity of the plate in W/(m.K)
- $u_{fd,htf}$ is the velocity field of the HTF in m/s
- ε is the bed porosity
- $C_{p,s}$ is the specific heat capacity at constant pressure of the TCM in J/kg.K
- $C_{p,v}$ is the specific heat capacity at constant pressure of the water vapor in J/kg.K
- ρ_s is the density of the TCM in kg/m³
- ρ_v is the density of the water vapour in kg/m³
- u_{fd} is the velocity field of water vapor through the TCM in m/s
- M_s is the molar mass of TCM in kg/kmol
- ΔH_r is the reaction enthalpy of the TCM in J/mol
- K_{eff} is the effective thermal conductivity of the porous bed in W/(m.K)

3.9. Evaluation Indicators

Every reactor design should be evaluated according to some important results:

Average Heat Transfer Fluid Outlet Temperature:

is the main result of the study which shows the effectiveness of the reactor for the space heating and/or hot water usage. Due to using a fixed bed reactor, a very high temperature may not be expected, but values above 45°C can be desired for space heating.

Energy Density: is the amount of energy that can be stored in a given system given by equation 13:

$$\text{Energy Density} = \frac{\text{TCM Mass} \cdot \text{Hr}}{\text{Total Volume}} \quad (13)$$

Where:

• Hr is the heat of reaction in kJ/kg
TCM mass which can be stored in porous bed is derived by:

$$\text{TCM Mass} = \rho_s \cdot V \cdot (1 - \varepsilon) \quad (14)$$

Where:

- ρ_s is the density of the TCM in kg/m³
- V is the bed volume in m³
- ε is the porosity

Average Status of Conversion(α): which can show the speed of the reaction in the bed and cycle time.

4. Basic Modelling

As already mentioned, before moving to the final design proposals, a simple 2D model was created in COMSOL Multiphysics® using the heat transfer fluid and reactor bed parts not only to understand the physics of the problem and get familiar with modelling, but also to perform some sensitivity analysis to figure out some of the main design parameters which can be also used in the final reactors for improvement. Figure 6 shows the schematic of the created basic model.

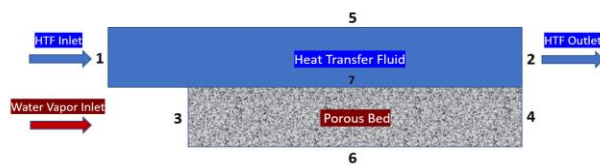


Fig 6: Schematic of the basic 2D Model

The main boundary conditions are as Table 3:

Boundary	Type	Boundary Conditions
1	Mass Flow Rate Inlet	$\dot{m} = 0.01 \text{ kg/s}$ $T = \text{const} = 30^\circ\text{C}$ $\frac{\partial c}{\partial x} = 0$
2	Pressure Outlet	$P = 0 \text{ Pa}$
3	Pressure Inlet	$P = 1200 \text{ Pa}$ $T = 10^\circ\text{C}$ $c = \text{const} = 0.5 \text{ [mol/m}^3 \text{]}$
4	No Flow	$q \cdot n = 0$ $\rho u \cdot n = 0$

	Thermal Insulation	
5	Wall	No slip wall $u = 0$ $q \cdot n = 0$
6	Symmetry	$q \cdot n = 0$ $\rho u \cdot n = 0$
7	Wall	No slip wall $u = 0$

Table 3: Boundary Conditions for model in Fig 6

Initial Heat Transfer Fluid temperature, initial TCM temperature, initial α , initial TCM pressure have been set to 30°C, 30°C, 0.1 and 130 Pa.

4.1. Assumptions

1. The Material bed is a homogenous porous medium and has constant properties such as heat capacity and thermal conductivity.
2. The TCM particles are stationary.
3. Permeability and Bed Porosity are constant over time.
4. Local Temperature Equilibrium ($T_s \approx T_v \equiv T$) is considered in the porous media, meaning that one single equation is solved for two phases (solid and gas) using effective material properties.
5. Heat transfer by radiation in the porous media is neglected.
6. Inlet Mass Flow Rate of heat transfer fluid is constant.

4.2. Constant Parameters

The main constant parameters are as Table4:

A_f : Pre-exponential Factor (1/s)	2.7E-9
E_a : Activation Energy (J/mol)	-34828
Hr: Heat of Reaction (kJ/mol)	-91.32
D_p : particle diameter (mm)	1
C_{p,K_2CO_3} : K_2CO_3 Specific Heat Capacity (J/(g.K))	0.86527
$\rho_{K_2CO_3}$: K_2CO_3 density (kg/m ³)	2290
$K_{K_2CO_3}$: K_2CO_3 Thermal Conductivity (W/m.K)	0.8
$C_{p,wv}$: Water vapor Specific Heat Capacity (J/(g.K))	1.878
ρ_{wv} : Water Vapor density (kg/m ³)	0.0094
K_{wv} : Water Vapor Thermal Conductivity (W/m.K)	0.02

Table 4: Constant Parameters of the model

4.3. Sensitivity Analysis – Design Parameters

Based on the knowledge about the physics and equations, Potential design variables can be divided into geometry variables like the bed length and thicknesses of TCM and HTF layers, and operation variables like Porosity, HTF mass flow rate and the evaporator pressure.

Sensitivity Analysis was performed for these parameters on the basic 2D Model to grasp the design parameters which can better be used for improvements. Results were compared considering both heat transfer fluid outlet temperature and the status of conversion.

It has been concluded that, increasing bed length and TCM layer thickness can increase the heat exchange surface and therefore the outlet temperature but also decrease the status of conversion a bit due to having more salt. So, geometric variable should increase considering a trade-off.

Furthermore, a porosity equal to 0.7 has been selected as a good value for the porous bed. Higher evaporator pressure and conversion status due to more pressure difference (reaction potential) and lower inlet mass flow rate increased the outlet temperature noticeably because of lower HTF velocity and more heat exchange time.

5. Final Designs

Two different reactors have been proposed, one based on multi-tube (shell and tube) design and the other with helical coil design. Based on the experience from previous sensitivity analysis, improvement

5.1. First Multi-Tube Design

Figure 7 illustrates the first multi-tube design. the TCM particles are inside the tubes and water vapor enters this TCM tubes from an inlet section. The liquid water as the heat transfer fluid enters the shell and moves with 90 degrees beside the tubes and through the shell, taking the heat of hydration reaction of the salt, increasing its temperature and flows to the HTF outlet inlet. Note that from now on, all B.C.s and I.C.s are the same as basic 2D model except where is mentioned.

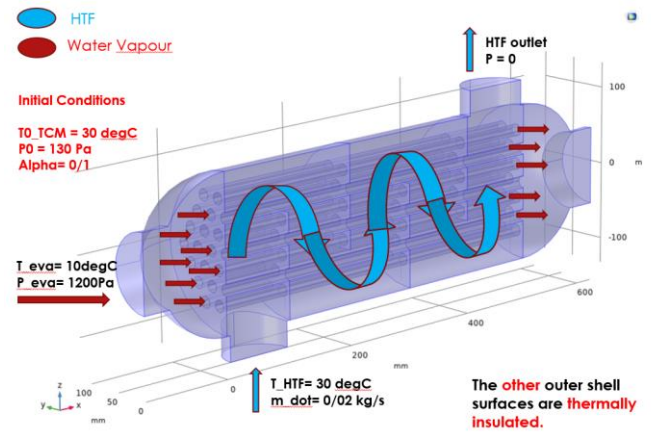


Fig 7: The Initial Multi-Tube Design

5.2. Improved Multi-Tube Design

To improve the initial design, total reactor length has been increased, tube diameters have been doubled (to increase TCM mass) and HTF mass flow rate has been halved (Figure8).

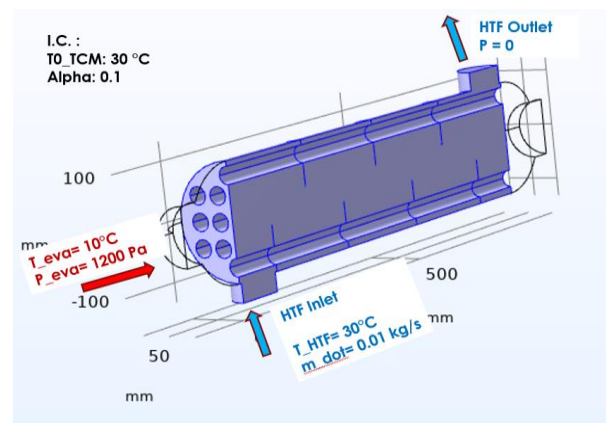


Fig 8: The improved Multi-Tube Design

5.3. Results: Multi-Tube

Figure 9 and Table 5 are the main comparisons.

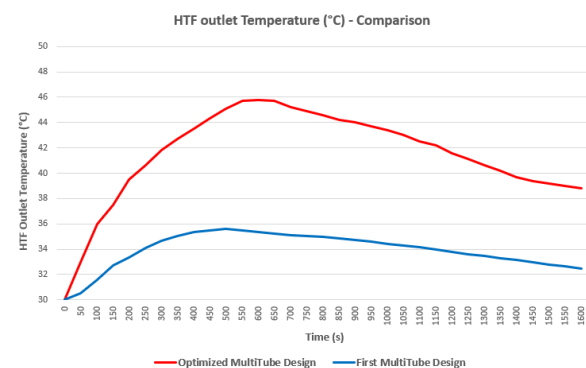


Fig 9: The Average HTF outlet temperature for the initial(blue) and improved(red) designs

	Initial Multi-Tube	Improved Multi-Tube
Total Reactor Volume (m ³)	0.01	0.02
Bed Volume (m ³)	0.0039	0.012
$\frac{Bed\ Volume}{Total\ Volume}$	40 %	60 %
TCM Mass (kg)	2.67	8.24
Energy Density (kJ/m ³)	169698	272464
Ideal Energy density (kJ/m ³)	445651	454107

Table 5: Volume, TCM Mass and Energy density for two multi-tube models

It can be concluded that the improved reactor has higher average HTF outlet temperature (peak value of 46°C) due to increasing the heat transfer surface by geometric improvements. This is a good value, considering that a fixed bed is being used.

Table 8 shows that in the bigger reactor, the portion of the volume related to the TCM bed has been increased from 40% in the initial design to 60%. Bigger bed has higher TCM and higher energy density.

5.4. First Helical Coil Design

The helical coil design (Figure 10) consists of a helical coil inserted inside a cylinder shell.

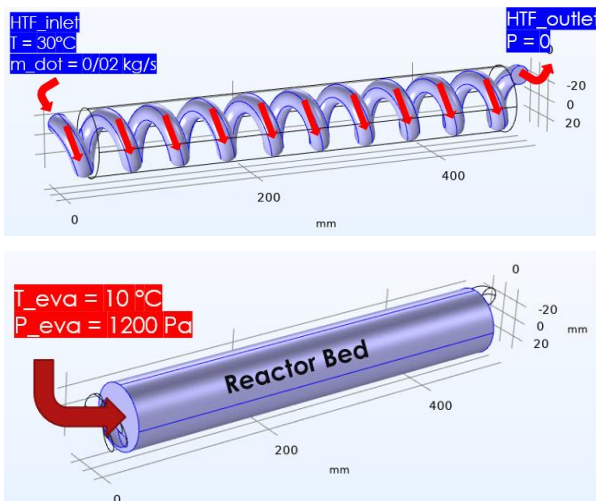


Fig 10: The initial helical coil design

The cylinder is the reactor bed, containing the TCM material. Water vapor coming from the evaporator enters this bed at the evaporation pressure and temperature and hydrates the TCM. The heat

released from this reaction is transferred to the heat transfer fluid which is flowing through the helical coils.

5.5. Improved Helical Coil Design

To improve the initial design, axial pitch was decreased and number of turns increased to improve the heat transfer and reaction, diameter of helix increased to have more HTF, mass flow rate was halved and bed length was increased (Fig11).

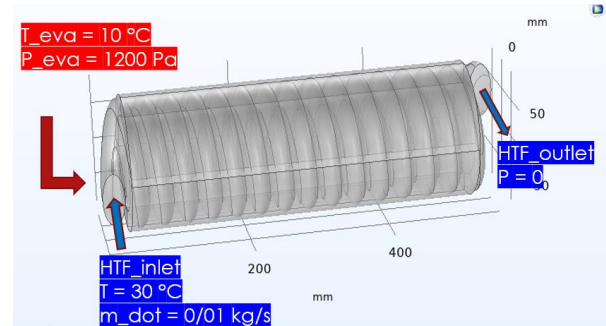


Fig 11. The improved Helical Coil Design

5.6. Results: Helical Design

Figure 12 and Table 6 are the main comparisons.

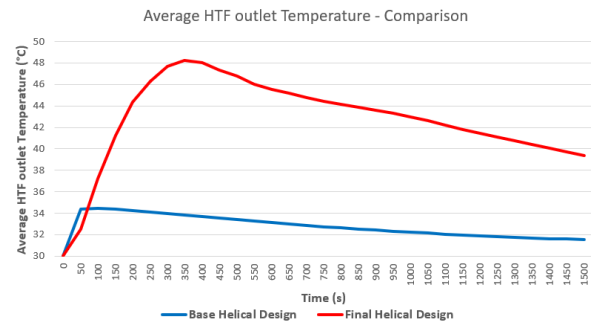


Fig 12: Average HTF outlet temperature for first(blue) and final(red) helical coil design

	Initial Helical Design	Improved Helical Design
Total Reactor Volume (m ³)	0.0022	0.020
Bed Volume (m ³)	0.0017	0.012
$\frac{Bed\ Volume}{Total\ Volume}$	82 %	60 %
TCM Mass (kg)	1.17	8.245
Energy Density (kJ/m ³)	350900	272497
Ideal Energy density (kJ/m ³)	445651	454107

Table 6: Other Comparisons of two helical designs

As expected, the improved reactor has higher average HTF outlet temperature (peak value of 48.5°C) due to increasing the heat transfer surface by geometric improvements. Although decreasing TCM bed volume portion has decreased the energy density of the improved case, but the resulting HTF portion increase has helped the helical reactor to achieve better conversion, heat transfer and high Average HTF outlet temperature.

5.7. Results: Helical vs. Multi-Tube

The Helical model was improved such that it also has the same total volume, bed volume, TCM mass and thus energy density with the improved multi-tube design. Therefore, the average HTF outlet temperature can be used for a fair comparison as figure 13. Equal parameters for two designs are:

- Total Reactor Volume = 0.02 m³
- Bed Volume = 0.012 m³ ($\frac{\text{Bed Volume}}{\text{Total volume}} = 60\%$)
- TCM Mass: 8.245 kg
- Energy Density = 272497 kJ/m³ (which is 60% of the ideal energy density)

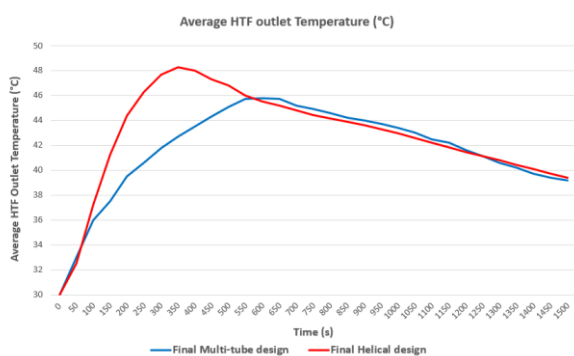


Fig 13: Average HTF Outlet Temperature for the final multi-tube(blue) and helical(red) designs

6. Conclusions

- 1) A suitable reactor design requires a trade-off optimization among the HTF outlet temperature, average status of conversion and energy density.
- 2) Both reactors have good potential to provide acceptable HTF outlet temperature and power output for space heating purposes, but for hot water usage, they need more improvement or should be upgraded to moving bed reactors.
- 3) The Helical Coil Heat Exchanger has had a slightly better performance mainly because of

larger heat transfer surface compared to the shell and tube designs.

4) From literature review, it was found out that both these designs may have difficulty while cleaning, and clogging can be an issue. However, Helical heat exchangers need cleaning less often. Also, for the helical design of this study, the cleaning may be easier because of the simpler and more accessible tubes.

7. Recommendations

- 1) In both designs, status of conversion remained as the main improvement target because some parts of the design had lower α due to less heat transfer. So, in any future design, the geometry, HTF and TCM sections should be such that there is sufficient heat transfer and thus good material conversion in all the domain.
- 2) One may be able to reach to temperatures above 50°C with further improvements of the designs of this study, but it is recommended also to put effort on simulating moving bed and open systems.
- 3) Cost and Economic evaluations were not in the scope of this study. For final designs for industry, cost must also be considered as another indicator.
- 4) The reactors in this study were small models for research purpose. Definitely, TCM mass and energy density can grow in bigger reactors. However, energy density should always be compared with the ideal energy density.

8. Acknowledgements

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