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# The Optimization Model of Effective Thermal Conductivity for Metal Hydride Heat Pump of Refrigeration Cycle

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## ABSTRACT

The main challenge for the practical application of metal hydride heat pump (MHHP) is the relatively poor system performance, which is mainly caused by the undesirable heat transfer performance of reaction bed. However, there is no significant improvement of system performance when enhancing the effective thermal conductivity (E.T.C) of reaction bed. In order to identify the relationship between the E.T.C and system performance, a numerical model has been carried out first in this research. The effect of E.T.C on cooling temperature, cycle time, the coefficient of performance (C.O.P) and cooling power is investigated according to the numerical results, which indicate that the variation trend of C.O.P and cooling power is opposite when E.T.C increasing. There therefore exists an optimum value of (E.T.C), which is 0.7-0.8 W/mK in this research. The experimental confirmation is conducted eventually, and the comparison of results shows a reasonable agreement with an acceptable error range.

## 1. INTRODUCTION

Answering the call for the eco-friendly energy development, the thermally driven metal hydride heat pump (MHHP) is one of feasible approaches for its great potential in the recovery of low-grade heat without greenhouse-gas emissions, and that makes it become a leading contender among different applications of metal hydride.

The main challenge for the practical application of MHHP is the relatively poor system performance due to the low effective thermal conductivity of metal hydride reaction bed (Takeda 1998). Therefore, aiming for improving heat transfer performance, previous researchers had proved that inserting high- conductivity sheet into the reactor is one of the most practical methods. Bae (2012) tried to enhance the effective thermal conductivity by inserting the MH sheets, which were contained MH powder, carbon fiber, and pulp into reactor, and the results showed that it was increased 1.32 times. Besides, Yasuda (2013) also used MH sheets on MHHP system, and pointed out the rate of reaction were accelerated, especially on low temperature side. Moreover, Aluminium Foam Sheets (AF sheets) also was used on MHHP systems on ETC enhancing, and it succeed to increase the ETC values much greatly than using MH sheets on (Lin 2105); however, even the effective thermal conductivity has been increased, the efficiency of the system is still relatively low for that the enhancement of performance (C.O.P) is not statistically significant.

In addition, a number of studies had developed analysis about the effect of operating conditions as well as design parameters on the system performance. Satheesh (2010a) showed that for a given reactor geometry and overall heat transfer coefficient, there was an optimum value of thermal conductivity to cycle time. Kang (1996) indicated that the optimum value of a cycle period could be selected on the basis of the (C.O.P) and the heating output for different geometric configurations. In addition, it had been found that the average heat storage rate per unit mass of the metal

hydride reactors could be maximized by the optimal design of parameters involving the number of heat exchanger tubes, the heat exchanger tube diameter and the heat exchanger tube pitch (Bao, 2012). Although Førde (2009) discovered that the effective thermal conductivity was sensitive to the system, there have been no studies focusing on the relationship between effective thermal conductivity with system performance. To solve this problem, an optimization model of E.T.C is proposed in this paper.

## 2. OPERATING PRINCIPLE

### 2.1 Mechanism

In the metal hydride heat pump (MHHP), the reversible reaction of metal hydrides includes the hydriding (exothermic) process and dehydriding (endothermic) process, which can be indicated as below (Muthukumar,2010):

$$M + \frac{x}{2} H_2 \xrightarrow{hydriding} MH_x + \Delta H_f \tag{1}$$

$$MH_x + \Delta H_f \xrightarrow{dehydriding} M + \frac{x}{2} H_2 \tag{2}$$

Where $M$ stands for a metal (or metal alloy); x is a non-stoichiometric constant and $\Delta H_f$ is the hydride formation heat. By controlling the equilibrium pressure in the reaction, the direction of the reaction can be controlled. Figure1 (a) P-C-T curve is employed to demonstrate the relation between the pressure, hydrogen capacity and isotherm properties of metal hydrides. Under each isotherm condition, P-C-T curve denotes the plateau process in the main stage of this reaction, which is described in direct relationship to Van't Hoff equation:

$$\ln(P) = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \tag{3}$$

Where $\Delta H$ and $\Delta S$ are the enthalpy and entropy variation of metal hydrides and R is the gas constant. Thus building MHHP system requires two different alloys possessing different properties of P-C-T curves, which is translated into the Van't Hoff plot in Figure 2 for a refrigeration cycle.

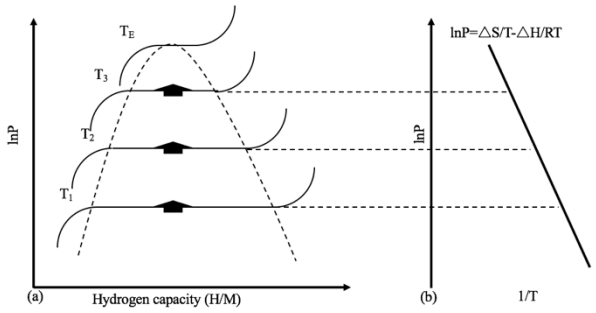



**Figure 1 :** PCT curve of metal hydride

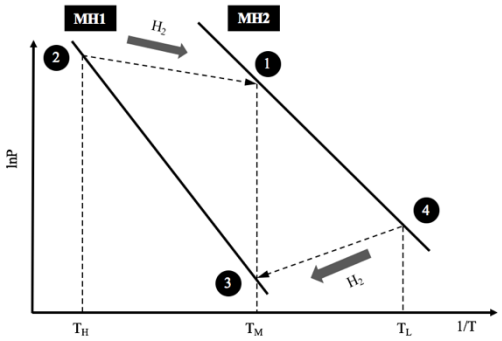



**Figure 2 :** MHHP of refrigeration cycle

### 2.2 The Valve-Controlled Metal Hydride Refrigeration System

Based on the theory of Van't Hoff, only relying on temperature changes enables hydrogen pressure to increase or decrease to change the hydrogen flow, so that the operation of the system could be achieved by the regulation of temperature and switching the valve. A valve-controlled metal hydride refrigeration system then is established, including a high-temperature reactor A, a low-temperature reactor B and the hydrogen tube and valve, which is shown as the Figure 3. In this system, the heat source and heat sink provide the regulation of temperature for high-temperature side and low-temperature side, which is selected as 80℃ and 20℃ respectively based on the consideration of experimental environment. In addition, the variation of hydrogen flow and cycle time could be achieved by controlling the valve.

(1) Preheating process: In the beginning, both of reactor A and reactor B are placed under low-temperature side, but MH1 is kept as metal hydride (spot 3) meanwhile the MH2 is in the state of alloy (spot1), and the valve is

closed. For increasing the hydrogen pressure, the temperature of reactor A could be increased to heat source temperature by using the heat from heat source. In the meantime, reactor B is still maintained at low-temperature side. Therefore, the purpose of this process is to increase the equilibrium hydrogen pressure of MH1 (spot2) above that of MH2 (spot1), preparing for the next process of regeneration.

(2) Regeneration process: In this process, opening the valve, due to the the pressure difference, the hydrogen would flow from reactor A to reactor B. Therefore, the desorption of hydrogen is carried out with endothermic reaction in reactor A, and the heat is supplied from heat source. Meanwhile, the absorption of hydrogen is running with exothermic reaction in reactor B, and the reaction heat is moved by heat sink.

(3) Precooling process: After the regeneration process, MH1 is in the state of desorption (spot2) and MH2 is in the state of absorption (spot1). In order to cycling the hydrogen, closing the valve and using the middle temperature (heat sink) for cooling the reactor A, the hydrogen pressure of MH1 (spot3) would be decreased lower than MH2 (spot1).

(4) Cooling process: The process of precooling results in a pressure difference of hydrogen, so that the hydrogen would flow from reactor B to reactor A once the valve is opened. In this process, MH1 would absorb hydrogen with endothermic reaction and MH2 desorb hydrogen with exothermic reaction. This exothermic reaction enables the temperature of MH2 to decrease to cooling load temperature (spot4), achieving the cooling purpose.

## 3.MATHEMATICAL MODELLING

### 3.1 System Model

The numerical model describes a pair of cylindrical reactor filled with metal hydride alloy. In practical application, however, a continuous power output requires at least two pairs of reactors. The physical model is showed in Figure3. Each reactor is placed at the thermostatic bath and is connected with a tube for transferring hydrogen by controlling the valve. In order to simplify this analysis model, some assumptions are made as following:

(1) Each part of the system is under the adiabatic condition.
(2) The convective heat transfer in hydride materials is negligibly small comparing with conductive heat transfer.
(3) The wall temperature of the reactor is constant with the temperature of thermostatic bath without considering of the heat convection, and the heat loss from the system is neglected.

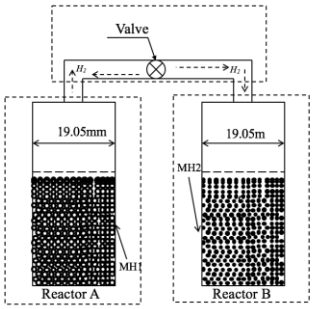



**Figure 3 :** The calculation model schematic diagram of MHHP

### 3.2 Numerical Equations

Considering the desorption or absorption reacting inside the reactor as an internal heat source, it is essential for mass transfer analysis to connect the hydrogen flow rate with to the physical-thermodynamic parameters of the hydrogen gas and hydride. Thus, the equation governing the hydrogen concentration is employed (Gambini,1994a), which has been experimentally proved suitable for the engineering application with reasonable accuracy in earlier literatures and is expressed as following:

In the process of desorption:

$$\frac{d\theta_d}{dt} = \varepsilon_d e^{-E_d/RT_d} ln\left(\frac{P_{eq}}{P_1}\right)\theta_d \quad (4)$$

where $\theta_d$ is the hydrogen concentration, thus $d\theta_d/dt$ stands for the hydrogen concentration variation in reactor during desorption process, and it is proportional to the hydrogen flow rate:

$$G = \frac{dm}{dt} = \alpha \frac{d\theta}{dt} \quad (5)$$

where $G$ is hydrogen flow rate and the constant α could be calculated by:

$$\alpha = \frac{N_{MH} W_{MH} M_{H_2}}{2M_{MH}} \quad (6)$$

By contrast, hydrogen concentration variation during absorption process is expressed as:

$$\frac{d\theta_a}{dt} = \varepsilon_a e^{-E_a/RT_a} ln\left(\frac{P}{P_{eq}}\right)(\theta_\infty - \theta_a) \quad (7)$$

where $\theta_\infty$ is the the maximum achievable value of hydrogen concentration when $d\theta_a/dt=0$.
From above equations, it shows that the key dynamic of reaction is the variation of hydrogen pressure and temperature that causes the hydrogen flow to change, affecting the reaction heat of system. As it is coupled and influenced each other between mass transfer and heat transfer in the reactor (Choi, 1990), to build a complete analysis model equations governing heat transfer should be decided appropriately according to assumptions. Without considering the convective heat transfer, the unsteady heat conduction of a cylinder with an inner heat source can be normally represented as (Çengel, 1998):

$$\frac{\rho c}{k_{eff}}\frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) + \frac{1}{r^2}\frac{\partial}{\partial \varphi}\left(\frac{\partial T}{\partial \varphi}\right) + \frac{\partial}{\partial z}\left(\frac{\partial T}{\partial z}\right) + \frac{\phi}{k_{eff}} \quad (8)$$

where $\phi$ is the internal heat source from reaction heat and is the product of hydrogen flow rate $G$ and the enthalpy $\Delta H$ of hydrides. In addition, $k_{eff}$ stands for the effective thermal conductivity (E.T.C) of the reaction bed. In this case, the heat transfer mainly occurs in the radial direction due to the symmetry of reactor and negligible hydrogen pressure drop in the axial direction. The conductive heat transfer equation therefore would be simplified as:

$$\frac{\rho c}{k_{eff}}\frac{\partial T}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) + \frac{\phi}{k_{eff}} \quad (9)$$

In the process of regeneration and cooling, the tube of hydrogen is connected since the valve is opened. The transient analysis to the hydrogen properties in the tube should therefore be considered, determining whether the reaction process of whole system is finished, which is solved coupling with energy conservation equation. Considering this process is insulated without mechanic work, the internal energy variation would be:

$$dU = dm_d H_d - dm_a H_a = d\left(m_g H_g\right) = dm_g U_g + m_g dU_g \quad (10)$$

For hydrogen, assuming a linear relationship between the enthalpy and internal energy and temperature as $H=c_pT+H_0$ and $U=c_vT+U_0$, the above equation would be expressed as:

$$\frac{dm_d}{dt}c_P T_d - \frac{dm_a}{dt}c_P T_a + H_0\left(\frac{dm_d}{dt} - \frac{dm_a}{dt}\right) = \frac{dm_g}{dt}c_v T_g + \frac{dm_g}{dt}U_0 + m_g c_v \frac{dT_g}{dt} \quad (11)$$

According to mass conservation:

$$\frac{dm_g}{dt} = \frac{dm_d}{dt} - \frac{dm_a}{dt} \tag{12}$$

and assuming $H_0=U_0$, the temperature of hydrogen would be:

$$\frac{dT_g}{dt} = \frac{1}{m_g}\left[\frac{d\theta_d}{dt}\left(\frac{c_p}{c_v}T_d - T_g\right) - \frac{d\theta_a}{dt}\left(\frac{c_p}{c_v}T_a - T_g\right)\right] \tag{13}$$

As ideal gas requires that $PV=nRT$, the pressure of hydrogen in the tube is represented as:

$$\frac{dP_g}{dt} = P_g\left[\frac{1}{T_g}\frac{dT_g}{dt} + \frac{1}{m_g}\frac{dm_g}{dt}\right] \tag{14}$$

### 3.3 Performance of a Metal Hydride Heat Pump

For the purpose of studying the relationship between heat transfer properties and system performance, cycle time, the coefficient of performance (C.O.P) and cooling power are selected as the indicators of system performance.
For numerical results,

$$COP = \frac{Q_{out}}{Q_{in}} \tag{15}$$

where $Q_{out}$ and $Q_{in}$ is the heat generated during cooling process and regeneration process and can be calculated as:

$$Q = \sum_{i=0}^{n} \frac{(\phi_i + \phi_{i+1})\cdot\Delta t}{2} \tag{16}$$

For experimental verification, the equation can be derived following the general thermodynamic theory (Yang,2010):

$$COP = \frac{Q_{out}}{Q_{in}} = \frac{\Delta H_{MH2}\int_0^{t_c} G_c dt - C_L(T_M - T_L)}{\Delta H_{MH1}\int_0^{t_r} G_r dt + C_H(T_H - T_M)} \tag{17}$$

The cooling power is a calculation determining the amount of heat that needs to be removed from a system by a cooling mechanism in a unit time. It is one of the things considered during the design phase for a cooling system, and the goal is to build a structure with better efficiency to remove increasingly amount of heat during certain time.

$$P_{cooling} = \frac{Heat\ for\ cooling}{Cooling\ time \cdot Mass\ of\ MH} = \frac{\Delta H_{MH2}\int_0^{t_c} G_c dt}{t_c \cdot m_{MH2}} \tag{18}$$

## 4.RESULTS AND DISCUSSION

### 4.1 Numerical Results

The results of calculation computing by MATLAB identify the performance of metal hydride heat pump for a variety of effective thermal conductivity. Based on the consideration of available data about parameters of metal hydride, $LaNi_{4.8}Al_{0.2}$ and $LaNi_5$ are selected as high temperature metal hydride (MH1) and low temperature metal hydride (MH2) respectively. The properties are taken from (Gambini,1994b) and shows in Table1. The effective thermal conductivity of hydride bed is varied from 0.5to 2.0 W/mK by keeping other parameters as constant.

**Table 1:** Parameters used in calculation

| **Parameters** | | | **MH1** | **MH2** |
|---|---|---|---|---|
| Reaction Heat | $\Delta H$ | kJ/mol | 33.89 | 30.8 |
| Entropy Change | $\Delta S$ | kJ/mol.K | 0.107 | 0.108 |
| Kinetic Coefficient of Desorption | $\varepsilon_d$ | | 1420000 | 1320 |
| Kinetic Coefficient of Absorption | $\varepsilon_d$ | | 15200 | 35.8 |
| Activation Energy of Desorption | $E_d$ | kJ/kg | 20330 | 12400 |
| Activation Energy of Absorption | $E_a$ | kJ/kg | 13890 | 8430 |
| Specific Heat | c | kJ/kg.K | 0.426 | 0.359 |
| Density | ρ | kg/m$^3$ | 8110 | 8300 |
| Gas Constant | $R$ | kJ/mol.K | 0.008314 | 0.008314 |

Figure 4 shows the relation between E.T.C and cooling temperature and cycle time. By definition, the cooling temperature is the attainable lowest temperature during cooling process, which is proportional hydrogen desorption capacity, reflecting refrigeration effect. In Figure 4 when ETC increasing, the cooling temperature rises slightly at first and then is increasing rapidly once E.T.C is up to 1.3 W/mK. In contrast, the cycle time is found to an opposite trend. It is observed that due to the improvement of heat transfer the cycle time is decreasing significantly with the growth of E.T.C up to 0.9 W/mK and then shows a slowing trend, which is matching with the result from past research conducted by Satheesh (2010b). It indicates that the shortening cycle time leads to a higher cooling temperature, weakening refrigeration effect for the reason that available hydrogen capacity will be less provided. As refrigeration capability is tending towards stability once hydrogen is all reacted, cooling temperature increases slowly at first. Because it is found that heat transfer determines the rate at which hydrogen is transferred (BJURSTROM 1989).

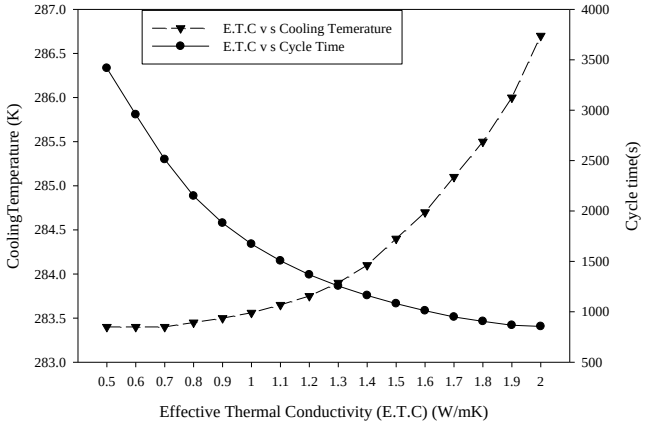



**Figure 4:** The effect of E.T.C on cooling temperature and cycle time by calculation

The effect of E.T.C on the cooling power and C.O.P is investigated in Figure5 and Figure6. It is showed that cooling power is gradually increased while C.O.P is slightly declining with an increase in the E.T.C. This phenomenon is explained by the relation between E.T.C and cooling temperature and cycle time. According to the data from Figure4, it is showed that when E.T.C changes for certain range, a wider range of cycle time is represented comparing with the change of cooling temperature, which means the variation rate of cycle time is more dramatically than refrigeration capacity. When it comes to cooling power, it would gradually increase with the growth of E.T.C. As when cycle time increasing, the available hydrogen capacity increases, and thus the heat output increases, resulting in a better C.O.P. In addition, once a certain cycle time has been reached, in this scenario when E.T.C decreases to 0.7 W/mK, E.T.C has less effect on the hydrogen capacity, so that C.O.P begins to change slowly with a decreasing of E.T.C. By comparing the variation trend, it is noticed that there exists an optimal cycle time, which could achieve a high COP value while a good cooling power. For this stimulation condition, the optimal range would be 0.7-0.8 W/mK, which is presented in Figure7 for temperature distribution of each process.

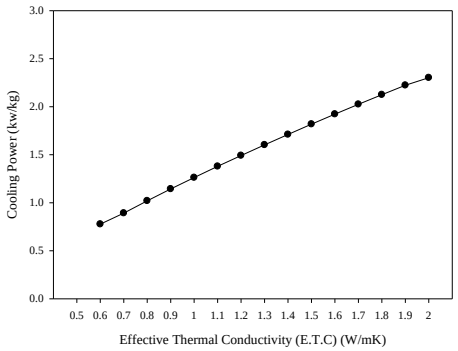



**Figure 5:** The effect of E.T.C on cooling power

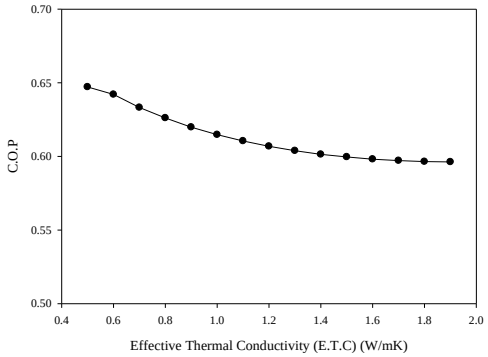
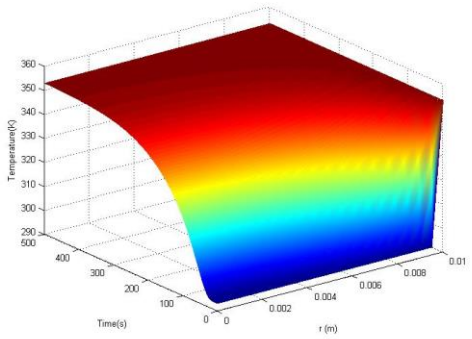
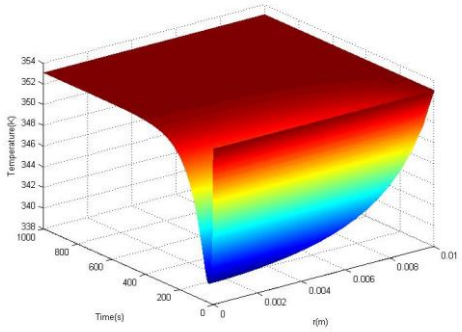


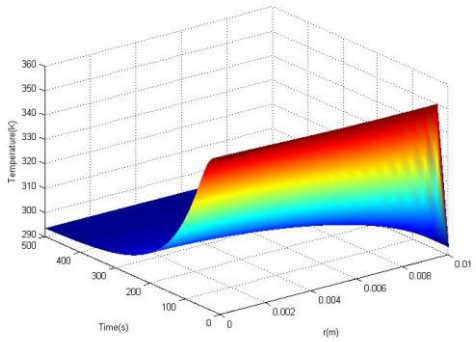

**Figure 6:** The effect of E.T.C on C.O.P



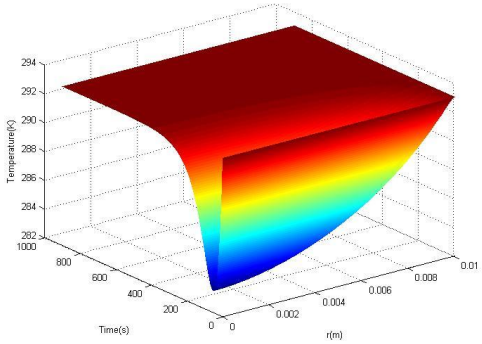
(a) Preheating process



(b) Regeneration Process



(c) Precooling process



(d) Cooling Process

**Figure 7:** The temperature distribution of the MHHP system

## 4.2 Experiment Set Up and Results

There are lots of researches discussing about the cycle time and reaction temperature on MH reaction period, besides, the values of cooling power and C.O.P of heat pump system are also mentioned frequently to evaluate how the system is. Our research team have also done lots of researches about enhancing the C.O.P values through increasing the effective thermal conductivities. However, until now, there is no any research talking about the relationship between C.O.P and effective thermal conductivity of MH particles. Originally, the experiment was set to show this relationship, but it's difficult to control the values of effective thermal conductivities on all cases in the experiment. Instead of that, considering the relationship between ETC and reaction time for linear situation part, the total reaction time was controlled as 25, 28, 31, 34, 37, 40,43, 46 minutes for k=0.5-2.0 to observe the values of C.O.P and cooling power on every cases. Before the experiment, the MHs ($LaNi_5$ and $LaNi_{4.8}Al_{0.2}$) have been activated and stabilized already.

The experiment equipment is shown as figure 8. There are two reactors (filled into 50g LaNi5 and 75g LaNi4.8Al0.2 alloy powder), hydrogen supply, vacuumed pump, thermostatic bath, and controlling valves on the device. The thermostatic bath is set as 20 degrees for lower temperature bath and 80 degrees for higher temperature bath. The reactors, shown as figure 9, are made by steel circular pipe with 19.05 mm external diameter and 15.75 mm internal diameter, and thermocouples are inserted into the center of the reactors from the tube bottom.

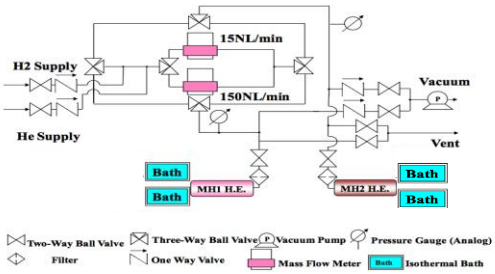



**Figure 8:** The schematic diagram of MHHP experiment

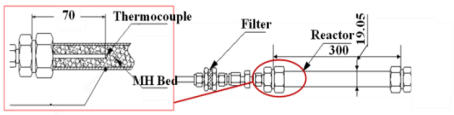



**Figure 9:** Specification of reactor

According to the MHHP cycle written in theorem, experiment is set for four process, preheating, regeneration, pre-cooling, and cooling process. These process are converted by controlling valves and moving the reactor from higher (lower) temperature bath to the other. Parameters (pressure, temperature, and flow rate) are recorded during all of the process, and the values of C.O.P and cooling are calculated by experimental equations.

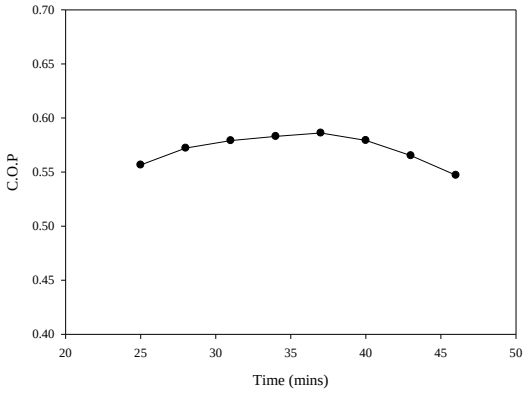



**Figure 10:** COP vs controlled cycle time by Exp.

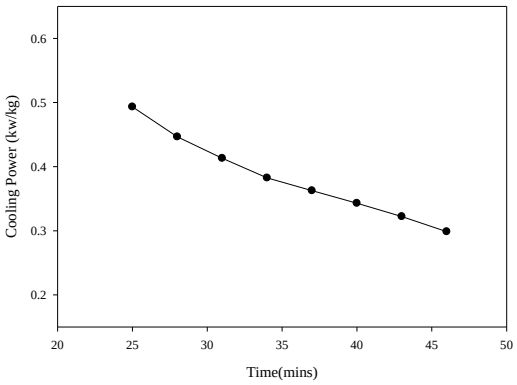



**Figure 11:** Cooling power vs controlled cycle time by Exp.

Figure 10 shows that C.O.P values changing while adjusted reaction time changing. At first, because the reaction between hydrogen and MH happened on the reactor, considering the generation heat, when increasing the reaction time, C.O.P was also increased; however, considering heat loss of the system, when increasing the reaction time of the system, it produces much more total heat loss during the reaction period. Combined these two reasons, the peak value of C.O.P happened between 35-40 minutes for k = 0.7-0.8W/mK.

Figure 11 shows the relationship between reaction time and cooling power. The result indicates that the value of cooling power become smaller when the reaction time is increased. According to the formula (18), the value of cooling power is the ratio of heat for cooling and cycle time multiple MH mass. It can be explained as the rate of cooling heat reduced when the reaction time extended due to the reaction amount of MH reduced. Instead of that, reaction time become a leading factor on the value of cooling power when the reaction time increasing.

### 4.3 Result Verification

Figure 12 left shows that the comparison between experimental data and simulation data on C.O.P values. Both of them are increased when the reaction time increasing. On simulation case, C.O.P values are remained the same as before while controlled reaction time over 50 minutes due to the same total generation heat of the system. Under the adiabatic condition of system, C.O.P values is related to total generation heat during the reaction period. However, on the experimental case, heat loss should be take into account on C.O.P calculation. It can be observed that the values of C.O.P are decreased when the reaction time is over 40 minutes due to the affection of heat loss. Besides, it

also be found that neglecting over 40 minutes' data, the average error differences between the simulation values and the experimental value were less than 5%

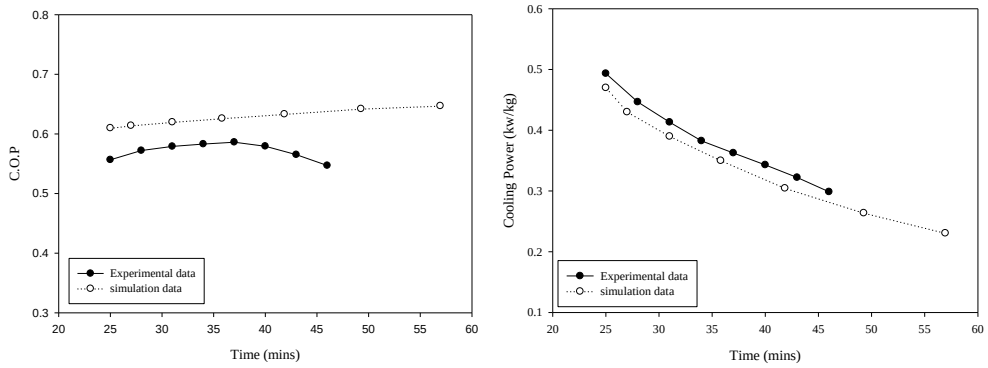



**Figure 12:** The comparison between experimental results and numerical results on C.O.P and cooling power

Figure 12 right shows that the comparison between experimental data and simulation data on cooling power. It can be observed that the tendency of cooling power changing with different reaction time on experimental values is the same as simulation data; moreover, the error band between experimental data and simulation data is less than 3%. It can strongly prove the accuracy of this calculation model

## 5.CONCLUSIONS

As the conclusion, a numerical model identifying the relation between E.T.C and system performance of MHHP is carried out to obtain the optimal condition. Based on this model, the effect of E.T.C on cooling temperature, cycle time, C.O.P and cooling power could be investigated. The numerical results indicate that for a given reactor geometry and operation condition, there exists an optimum value of effective thermal conductivity which could achieve a high value of C.O.P as well as a good cooling power. On this ideal situation, the optimal value range of E.T.C is between 0.7-0.8W/mK. To verify this numerical model, the performance of system is evaluated experimentally, and the comparison between experimental data and numerical results shows a reasonable agreement, for that the accuracy of this calculation model is within acceptable level, especially for cooling power which error band less than 3%.

## NOMENCLATURE

| | | |
|---|---|---|
| E | activation energy | (kJ/kg) |
| G | hydrogen flow rate | (kg/s) |
| H | enthalpy | (kJ/mol) |
| M | molecular weight | |
| N | number of atoms in hydride molecule | |
| P | pressure | (atm) |
| Q | heat | (kJ) |
| R | gas constant | (kJ/mol.K) |
| S | entropy | (kJ/mol.K) |
| T | temperature | (K) |
| U | internal energy | (kJ/kg) |
| W | hydride and reactor mass | (kg) |
| c | specific heat | (kJ/kg.K) |
| $c_p$ | specific heat at constant pressure | |
| $c_v$ | specific heat at constant volume | |
| $k_{eff}$ | effective thermal conductivity | (W/mK) |
| m | mass | (kg) |
| r | radius | (m) |
| t | time | (s) |

**Greek Letters**

| | | |
|---|---|---|
| $\theta$ | hydrogen concentration | ([H]/[MH]) |
| $\varepsilon$ | kinetic coefficient | |
| $\alpha$ | constant | |
| $\rho$ | density | ($kg/m^3$) |
| $\phi$ | internal heat source | (kW) |

**Subscript**

| | |
|---|---|
| MH | metal hydride |
| $H_2$ | hydrogen |
| a/d | absorption or desorption |
| eq | equilibrium |
| g | gas |
| c/r | cooling process or regeneration process |
| out/in | output or input |
| 0 | referred to initial condition |
| $\infty$ | referred to infinite time |
| 1,2 | referred to MH1 and MH2 |

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