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Conception and experimental investigation of metal hydride hydrogen storage system

Mohamed Houcine Dhaou

[a] *Laboratoire d'Etudes des Systèmes Thermiques et Energétiques LESTE, Ecole Nationale d'Ingénieurs de Monastir ENIM, Université de Monastir, Rue Ibn Eljazzar, Monastir 5019, Tunisie.*
[b]*Department of physics, College of Science, Qassim University, KSA.*

Abstract. Metal hydrides offer the potential to store hydrogen at modest pressures and temperatures with high volumetric efficiencies. The process of charging hydrogen into a metal powder to form the hydride is exothermic. The heat released by the reaction must be removed quickly in order to maintain a rapid charging rate. An effective method for heat removal is to embed a heat exchanger within the metal hydride bed. If the heat released is not removed from the system, the resulting temperature rise of the hydride will reduce the hydrogen absorption rate. Hence, hydrogen storage systems based on hydride materials must include a way to remove the heat generated during the absorption process. The present work aim to size, design and fabricate a metal-hydrogen reactor to study the hydrogen storage in a closed metal-hydrogen reactor. The effects of water cooling temperature (T_F), hydrogen injection pressure (P_{H2}) and cooling fluid flow rate ($\dot{m}$) on the mass absorbed of hydrogen (H_2) are experimentally investigated and analyzed. Results show that the mass absorbed of H_2 by the metal ($LaNi_5$) raise when T_F decreasesand/or when P_{H2} increases.

Keywords*: Hydrogen, Conception, Metal-hydrogen reactor, Storage hydrogen.*

Nomenclature

D	Diameter of cooling water collectors (*m*)
e	Spacing between the water collectors (*m*)
H	Convective heat transfer coefficient ($W/m^2.°C$)
L	Length of the core reactor (*m*)
$\dot{m}$	Cooling fluid flow rate (*kg/s*)
P_{H2}	Hydrogen injection pressure (*Pa*)
R	Radius (*m*)
R_1	Inner radius of the core reactor (*m*)
R_2	Outer radius of the core reactor (*m*)
R_C	Critical radius (*m*)
T	Temperature (°*C*)
T_1	Temperature of the core reactor (°*C*)
T_2	Temperature of the interface stainless steel / copper (°*C*)
T_F	Temperature of the cooling water (°*C*)
Greek symbols	
λ_C	Thermal conductivity of the copper (*W/m.°C*)
λ_S	Thermal conductivity of the stainless steel (*W/m.°C*)
Φ	Radial heat flux (*W*)

1. INTRODUCTION

The present transition from a fossil fuel-based towards a renewable energy-based society has triggered enormous efforts in research and development for energy storage technologies. In particular, hydrogen becomes increasingly important as a chemical energy vector because it can be produced readily by water electrolysis using surplus renewable energy [1,2]. Hydrogen storage and transport are both key technologies in a hydrogen-based energy cycle [3,4]. In this regard, metal hydrides can be employed for safe, compact and low-pressure hydrogen storage solutions [5,6]. In addition, metal hydrides can be used for thermochemical heat storage [7-9], for thermochemical heat pumps [10], for hydrogen compressors [10-12], for hydrogen purification [13] or for pneumatic actuators [14]. However, for all these applications a thorough design of the entire metal hydride tank system including the hydrogen storage material, the tank compartment and the periphery (heat exchangers, valves, controllers, etc.) is mandatory to reach an optimal performance under given specific operation conditions. Hydride tank design is non-trivial because it relies on numerous physical, chemical and engineering principles which have to be considered, including the gas-solid reaction, the gas transport through a porous reaction bed and the thermal management. The latter issue has become increasingly important because most of the aforementioned applications require a high-dynamic tank operation for which the heat transfer inside the hydride bed needs to be fast. Since most hydrides show low intrinsic heat conductivities [15,16], auxiliary materials and/or structures with high heat conductivity are employed inside the reaction bed, e.g. metal fins [17], metal foams [18] or graphite [19,20]. Kais Harbrig et Al.[21] A comprehensive computer model to simulate the hydrogenation and dehydrogenation dynamics of hydrogen storage tanks based on hydride graphite composites was presented. The model assumes an axisymmetric 2D heat and mass transfer including reaction heat, pressure volume work, anisotropic thermal conductivity, convective heat transport, Darcy flow, mass conservation in pores and matrix including local shrinkage/swelling, heat effect of inflowing hydrogen and tempering by a thermal fluid. Recently, Eustathios et al. [22], developed effective heat management strategies and novel cooling design options for hydrogen storage systems in $LaNi_5$metal-hydride reactors by performing systematic simulation and optimization studies. Askri et al. [23] established a model to study the dynamic behavior inside various designs of MHVs and optimization results indicate that almost 80% improvement of the storage time can be achieved when the design includes a concentric heat exchanger tube equipped with fins and filled with flowing cooling fluid. DHAOU et al.[24] studied the improvement of thermal performance of spiral heat exchanger on hydrogen. The experimental results show that the absorption/desorption times of the MHV are considerably reduced due to the integration of the finned heat exchanger system. Bhouri et al. [25] developed scoping and numerical models describing phenomena occurring during the loading process in analanate storage system having the configuration of a cylindrical shell, tube and fin heat exchanger. The numerical tool is used to evaluate the influence of varying the fin thickness and the number of heat exchanger tubes on both the loading and discharging processes. However, for the discharge case, a quicker release rate is obtained since the increase of the fin thickness results in a more uniform distribution of hydride bed temperature and, thus, a more uniform distribution of consumed and formed species. Serge Nyallang et al. [26] concluded that the optimum dimensions of the annular fins have been obtained by the minimization of the overall thermal resistance under the constraint of constant fin volume. Under such a constraint there are an infinite number of solutions, but it was found that longer fins with slender thickness may reduce the thermal resistance. In the second part of this study, a 2-D numerical model has been developed to predict the performance of finned tube heat exchanger. The effects of fin size on the hydrogen charging time have been discussed. The hydrogen charging time maybe reduced by more than 600 s when the optimized design is used. In addition, increasing the cooling tube diameter can bring about 25% of hydrogen charging time reduction under the same heat transfer conditions, which is quite significant. Paggiaro et al. [27] developed an unsteady lumped model and analyzed the influence of the thermal effects during vessel operation. Zewei et al.[28] developed a mathematical model of heat and mass transfer in high-temperature magnesium hydride reactors. The absorption kinetic is accelerating for the increasing pressure supply and for the inlet temperature. The optimal operating parameters, the gravimetric capacity and volumetric capacity at the filling time of 12 min are increased greatly compared with the baseline set. Jinsheng et al. [29] simulated the air cooling hydrogen tank and the water cooling hydrogen tank with the Comsol and siumlink. The simulation model calculated by Simulink and Comsol are fit well with the experimental results during the four stages. The pressure increases quickly and reaches its maximum at 951 s. The thermal average temperature data is computed by the finite element model based on Comsol. The agreement between two simulation curves tests and verifies the reliability of the simulation results. Heat transfer is modeled under conditions of air cooling and water cooling, wall temperature is improved by varying heat transfer coefficient, a more realistic geometry with insert tube improves near inlet temperature. The water cooling condition is better than air cooling condition in decreasing the temperature of the storage tank and improving the storage capacity.

2. CONCEPTION OF THE HYDROGEN STORAGE SYSTEM

In order to study the sorption kinetic of hydrogen (H_2) by the metal hydride ($LaNi_5$), we propose to size and design a concentric metal-hydrogen reactor equipped with a copper coaxial heat exchanger.

In order to improve the sorption kinetic of H_2, the rate of hydrogen absorbed by the metal hydride must be optimized. Consequently, the cooling of the reactor core must be effective.

In this part, the main objective is to determine the critical radius R_C for which we obtain the maximum of heat flux Φ transferred to the cooling water.

2.1 Determination of the critical radius R_C

The cross-section of the reactor considered in this study is shown in figure1. It consists of a copper coaxial heat exchanger and a stainless steel core reactor.

To simplify, we suppose a steady state regime. Furthermore the heat flux transfer by conduction is supposed one-dimensional and occurs along the radial direction.

The temperature of core reactor is assumed to be uniform and equal to T_1. T_2 is the temperature of interface stainless steel / copper and T_F is the temperature of the cooling water.

- Heat flux through the core reactor (stainless steel)

Here L denotes the length of the core reactor and λ_S is the thermal conductivity of stainless steel.

$\forall R_1 \leq r \leq R_2$, the radial heat flux expression is written:

$$\Phi = -2\pi\lambda_S rL\frac{dT}{dr} \tag{1}$$

Heat flux is conservative $\forall R_1 \leq r \leq R_2$, so we can write:

$$\int_{T_1}^{T_2} dT = -\frac{\Phi}{2\pi\lambda_S L}\int_{R_1}^{R_2}\frac{dr}{r} \tag{2}$$

$$T_1 - T_2 = -\frac{\Phi}{2\pi\lambda_S L}Log\left(\frac{R_2}{R_1}\right) \tag{3}$$

- Heat flux through the heat exchanger (copper)

We suppose that the spacing e between the water collectors is very small (Figure 1) such that the cooling fluid manifolds are assimilated to an annular space with interior diameter R_C. Furthermore, it is assumed that the diameter D is very less than the length L.

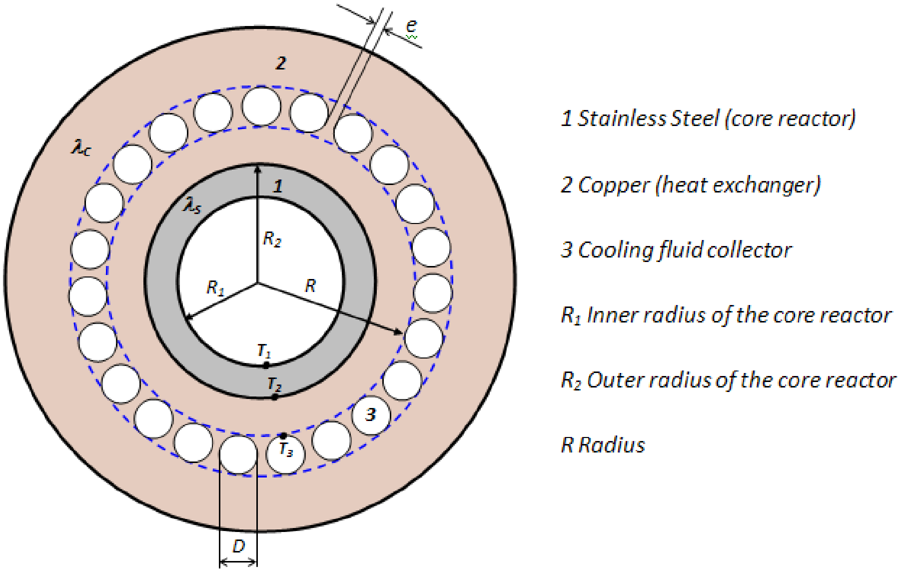

Figure 1:Cross-section view of the concentric metal-hydrogen reactor equipped with a coaxial heat exchanger made of copper.

Taking into the effect of convection, the expression of heat flux through the heat exchanger is written as follow:

$$\Phi = \frac{2\pi L(T_2 - T_F)}{\frac{1}{\lambda_C} Log\left(\frac{R}{R_2}\right) + \frac{1}{Rh}} \tag{4}$$

Where h denote the convective heat transfer coefficient.
Thus, we can write:

$$T_2 - T_F = \frac{1}{2\pi L}\left[\frac{1}{\lambda_C} Log\left(\frac{R}{R_2}\right) + \frac{1}{Rh}\right]\Phi \tag{5}$$

The sum of equations (3) and (5) gives:

$$T_1 - T_F = \frac{1}{2\pi L}\left[\frac{1}{\lambda_S} Log\left(\frac{R_2}{R_1}\right) + \frac{1}{\lambda_C} Log\left(\frac{R}{R_2}\right) + \frac{1}{Rh}\right]\Phi \tag{6}$$

We can deduce the expression of heat flux which can be written as follows:

$$\Phi(R) = \frac{T_1 - T_F}{\frac{1}{2\pi L}\left[\frac{1}{\lambda_S} Log\left(\frac{R_2}{R_1}\right) + \frac{1}{\lambda_C} Log\left(\frac{R}{R_2}\right) + \frac{1}{Rh}\right]} \tag{7}$$

The main objective is to determine the critical radius R_C for which the heat flux Φ, transferred to cooling fluid, is optimal (maximum flux). The derivative of Φ with respect to R is:

$$\frac{d\Phi}{dR} = \frac{\frac{1}{2\pi L}\left(\frac{T_1 - T_F}{R}\right)\left(\frac{1}{Rh} - \frac{1}{\lambda_C}\right)}{\left[\frac{1}{2\pi L}\left(\frac{1}{\lambda_S} Log\left(\frac{R_2}{R_1}\right) + \frac{1}{\lambda_C} Log\left(\frac{R}{R_2}\right) + \frac{1}{Rh}\right)\right]^2} \tag{8}$$

Hence, on can determine the critical radius R_C:

$$\frac{d\Phi}{dR} = 0 \Rightarrow \frac{1}{Rh} - \frac{1}{\lambda_C} = 0 \Rightarrow R = R_C = \frac{\lambda_C}{h} \tag{9}$$

In figure 2, we have plotted the heat flux evolution according to radius R. It is clear that the heat flux has a maximum in the vicinity of $R = R_C = \frac{\lambda_C}{h} = 4.10^{-2} m$. As a result, this critical radius R_C is retained for the design step.

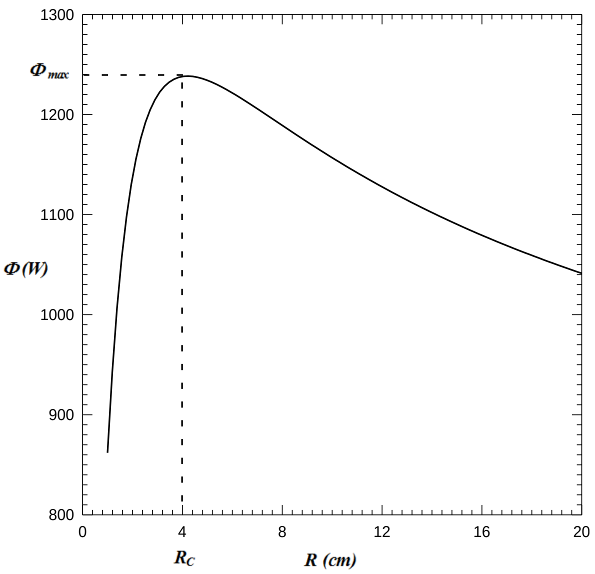

Figure 2: Heat flux Φ evolution according to radius R.

2.2 Design and experimental setup

Figure 3 shows the design scheme of hydrogen storage system (reactor). It is consists by two main parts: the reactor core made of stainless steel and the heat exchanger made of copper.

A cross-section view and a reactor photos are given by figures 4 and 5.

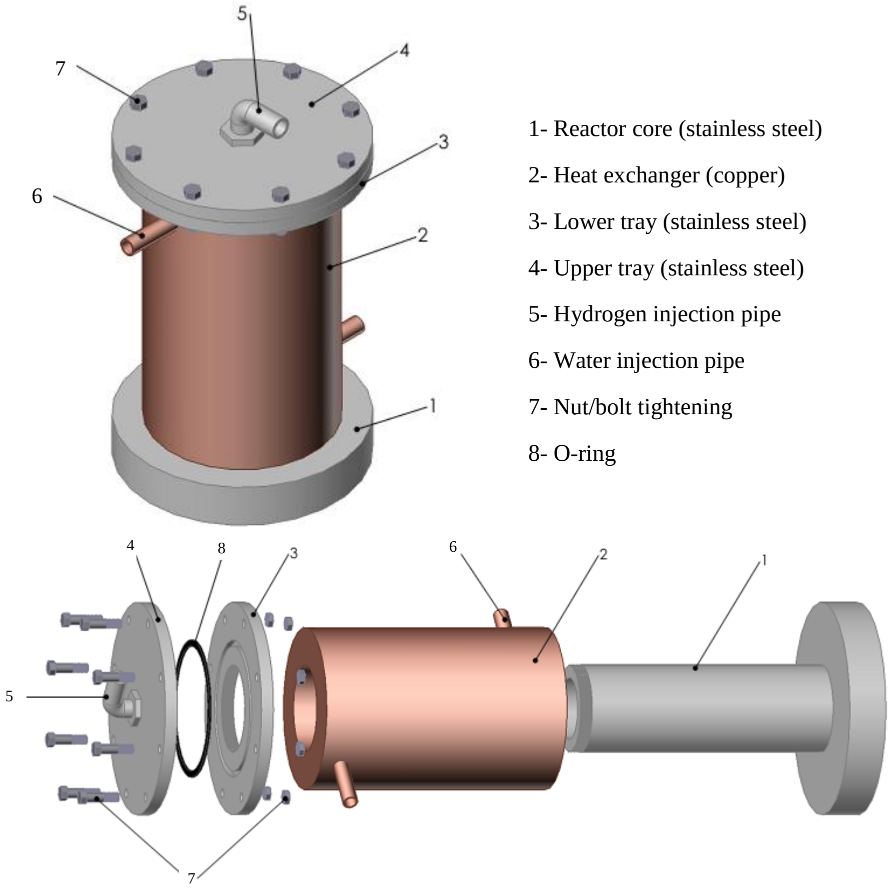

Figure 3: Design scheme of the hydrogen storage system.

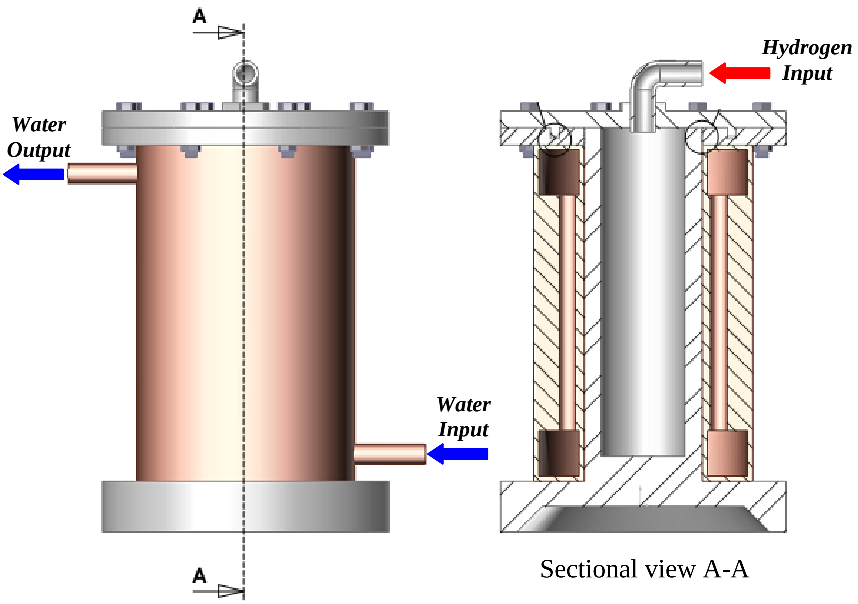

Figure 4: Cross-section view of the reactor.

Figure 5: Reactor photos after its fabrication.

The experimental set-up (figure 6) is a Sievert-type experimental apparatus mainly consists of a hydride reactor, two reference cylinders interconnected via valves, a vacuum pump, a thermostatic water bath (-10°C –>185 °C), a water pump, a data acquisition system and a 99.99% pure hydrogen gas cylinder. The temperature and pressure acquisition are recorded on a computer for further processing and interpretation. The experimental procedure for the absorption begins by starting the vacuum pump to maintain a low pressure in the reactor (0.01 bar) at high temperature (50 °C) for a long period of time (4 h) to ensure that the sample of $LaNi_5$ is emptied of hydrogen (or other gases). Then, thermostated water flow through the heat exchanger to bring the tank to the temperature value which will be the initial and final state of absorption test. The reference volume is fixed and then loaded with hydrogen at a desired pressure. The tank and the reference volume are then interconnected and the data acquisition (hydrogen dynamic pressure and temperatures of hydride bed and water outlet) is executed.

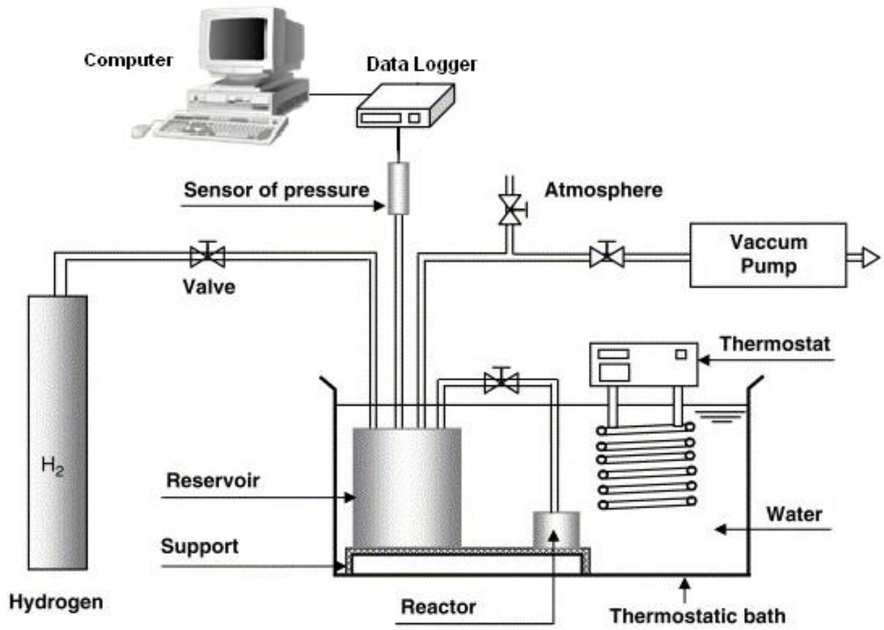

Figure 6: Experimental setup scheme.

3. RESULTS AND DISCUSSION

In this part we aim to study the effects of cooling fluid temperature (T_F), hydrogen injection pressure (P_{H2}) and cooling fluid flow rate ($\dot{m}$) on the hydrogen absorption rate.

3.1 Effect of cooling water temperature

To study the effect of cooling water temperature on the absorption rate, three values are considered (17, 27 and 37°C) and the other operating parameters are kept the same for the three cases. The obtained results shown in Figure 7 indicate that low temperature can accelerate the reaction but it has no effect on the total mass absorbed. Furthermore, it is important to note that decreasing the cooling temperature below certain value has no significant effect.

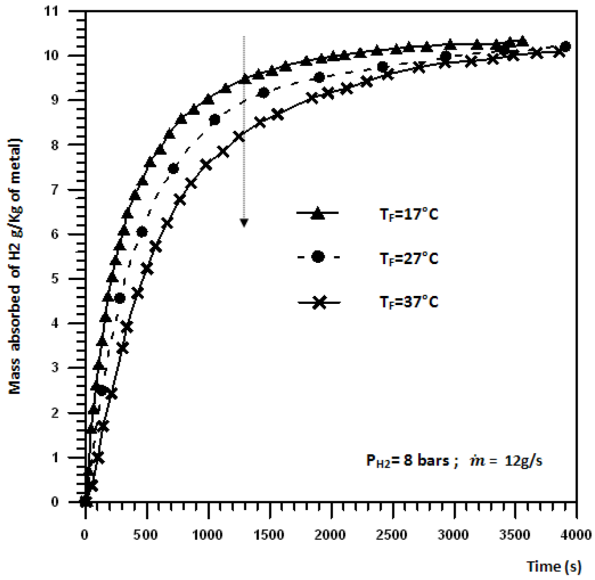

Figure 7: Effect of cooling water temperature on the time evolution of hydrogen absorption rate.

3.2 Effect of hydrogen injection pressure

The reactor absorbs hydrogen with various pressures by keeping a constant temperature of the coolant (17°C). The flow rate of cold fluid is maintained constant in order to fix the overall heat transfer coefficient. Figure 8 shows the variation of the rate of absorption and the temperature within the reactor. It is observed that for a given supply pressure, the rate of absorption reaches a peak at the beginning and decreases gradually towards zero at the end of the absorption process. This is due to the fact that, at the beginning of the absorption process, the potential of mass transfer (difference between the supply pressure and the equilibrium pressure) is high. While time progresses, the equilibrium pressure increases following the increase in the temperature of the bed under the effect of the absorption. This reduces the potential of the kinetics and thereafter the deceleration of the absorption rate. Because of the low thermal conductivity of the bed of hydride, the produced heat of absorption cannot be transferred starting from the bed with the cold fluid at the initial period from fast absorption and consequently excessive heat is stored in the bed of hydride itself, having for result a rise in the temperature of the bed.

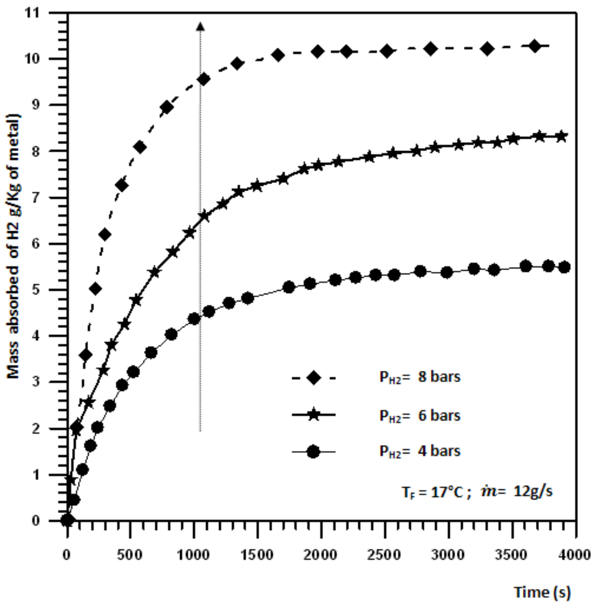

Figure 8: Effect of hydrogen injection pressure on the time evolution of hydrogen absorption rate.

3.3 Effect of cooling water flow rate

To study the effect of cooling water flow rate on the absorption rate, three values are considered (5, 7 and 12g/s) and the other operating parameters are kept the same for the three cases. The obtained results shown in Fig. 9 indicate that high flow can accelerate the reaction but it has no effect on the total mass absorbed. Besides, it is important to note that increasing the cooling water flow rate below certain value has no considerable effect.

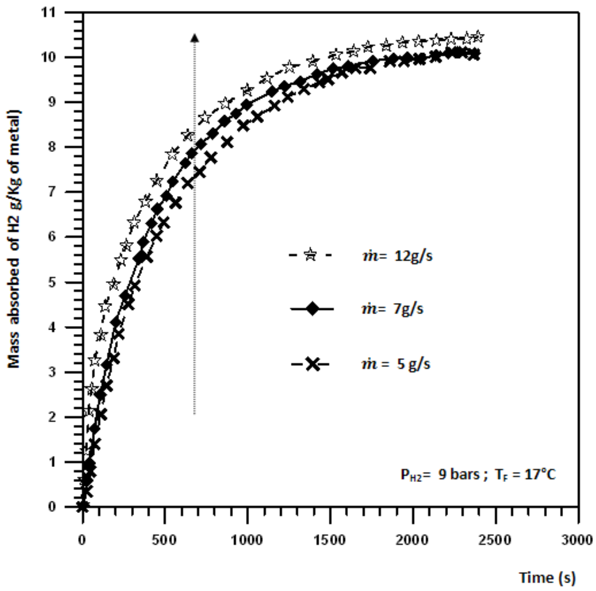

Figure 9: Effect of the mass flow of cooling water on the time evolution of hydrogen absorption rate.

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تصور وتحقيق تجريبي من نظام مفاعل الهيدروجين المعدني لتخزين الهيدروجين

محمد الحسين علي ضو[١،٢]

[١] مخبر دراسة انظمة الطاقة والحرارة، المدرسة الوطنية للمهندسين بالمنستير– تونس

[٢] كلية العلوم– قسم الفيزياء، جامعة القصيم، المملكة العربية السعودية.

ملخص البحث. الهدروجين من الغازات الصغيرة جدا، وقع اختراعه في عام ١٨٣٩ من طرف الباحث والعالم الانجليزي وليام روبرت. يستعمل غاز الهيدروجين في العديد من التطبيقات الصناعية، نذكر منها: تخزين الطاقة، خلايا توليد الكهرباء بالهيدروجين، مضخات الهيدروجين، الخ... لاستعماله في التطبيقات الصناعية يجب تخزين الهيدروجين بثلاثة طرق مختلفة: الهيدروجين المضغوط، الهيدروجين السائل والهيدروجين ذو الترابط الكيميائي (تفاعل الهيدروجين مع مواد صلبة. مثال: $LaNi_5$، Mg، ...). يستطيع الهيدروجين في الحالة الغازية نقل الطاقة كالكهرباء لمسافات بعيدة وعبر انابيب النقل وذلك بكفاءة عالية وبأقل تكلفة ممكنة وكما بإمكان الهيدروجين الاعتماد على تقنية وقود الطاقة او الآلات الاخرى المولدة للطاقة ليوفر لجمهور المستهلكين الكهرباء والماء النقي الصالح للشرب والهيدروجين اما بوصفه عنصرا كيميائيا فله استخدامات وتطبيقات متنوعة خلاف الطاقة الكهربائية ومن الممكن ان نصنف استخدامات الوقود الهيدروجيني بشكل رئيسي ضمن الحقول الاربعة الرئيسية التالية:

١– وقود لوسائط النقل كالسيارات والطائرات العاملة التي تعتمد على تقنية خلايا الوقود الهيدروجيني و تطبيقاتها الاوسع وصولا لاستخدامها مستقبلا في محطات توليد الطاقة.

٢– استخدام الوقود الهيدروجيني كبطارية بسعات تتدرج من الصغيرة المستخدمة في الحواسب الشخصية المحمولة و صولا الى بواخر نقل الهيدروجين التي تنقله من محطات الطاقة المتجددة الى اماكن توليد الكهرباء البعيدة لحل مشاكل و تكاليف الشبكات الطويلة و الضياعات الطاقية عبرها.

٣- يستخدم الوقود الهيدروجيني لتوليد للطاقة الحرارية باحتراقه المباشر في المراجل في محطات الطاقة اضافة لاستخدامه كوقود دفعي في الصواريخ.

٤- كما يستخدم الوقود الهيدروجيني عاملا في المفاعلات النووية ونخص بالذكر منها تقنية المفاعل الذي يعمل على مبدأ توليد الطاقة على سطح الشمس.

في هذا البحث قمنا بتصميم وتصنيع خزان لتخزين الهيدروجين (مفاعل الهيدروجين المعدني)، مع مبادل حراري خارجي، هذا الخزان وضع به ٤٠٠ غ من مادة صلبة ($LaNi_5$) لتخزين الهيدروجين. قمنا بدراسة العديد من العوامل المؤثرة على كمية الهيدروجين المخزنة بالمادة الصلبة مثال: تأثير ضغط الهيروجين، تأثير حرارة السائل المار بالمبادل الحراري، تأثير سرعة السائل المار بالمبادل الحراري.