Metamaterial Reinforced Reactor Design with Internal Heat Transfer Channels for Enhanced Metal Hydride as Hydrogen Storage Solution

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Effective operation of metal hydride (MH) hydrogen storage systems hinges on reactor configuration, which is crucial for large-capacity and stationary applications. However, when utilised for mobility usage, MH gravimetric density limits the range of efficient scenarios. One way to overcome this limitation is by designing a reactor that can also be suitable as a frame, pursuing to create a system where the MH weight becomes negligible. Employing a metamaterial of lattice octet design with inner channels for heat transfer fluid flow creates a design that is capable of withstanding load while maintaining MH hydrogen storing performance. This study focuses on developing a lightweight, compact, and efficient MH reactor capable of accommodating substantial hydrogen inventory. Numerical analysis evaluates three reactor configurations to visualise the impact of various design elements during storing operation. Compared to baseline reactors, the lattice octet design demonstrates significantly improved performance, with a 28 % reduction in hydrogen storage time and 48 % more storage capacity.

1. Introduction

Hydrogen has emerged as a promising energy alternative to replace fossil fuels, addressing both the need for sustainable energy to sustain human living standards and mitigate the environmental damage caused by non-renewable resources. Recent studies highlight hydrogen's potential to transition from fossil fuel-based energy production to more environmentally friendly methods (Juangsa et al., 2021), particularly in systems producing zero carbon emissions.

Various methods exist for hydrogen production, including biomass gasification (Widyawati et al., 2011), methane decomposition, electrolysis, and chemical looping (Holladay et al., 2009). Despite its advantages, the storage of hydrogen poses challenges. While liquefaction and compressed gas forms are commonly used, they suffer from high volume per energy storage ratios, safety concerns, and efficiency issues. Pressurized storage requires chambers designed to withstand pressures above 700 bar, raising safety considerations. Additionally, maintaining hydrogen in a cryogenic state for liquefaction incurs high energy costs. Promising solutions include catalytic or non-catalytic NH_3 synthesis and metal hydride (MH) based storage, known for its safety properties and volumetric energy density (Niaz et al., 2015).

The absorption and desorption of hydrogen are influenced by heat exchanger (HE) layouts, such as pipes (Elhamshri and Kayfeci, 2019), finned spiral pipes (Dhaou et al., 2010), and finned multi-tubular tanks (Ma et al., 2014). Research indicates that a higher surface area leading to uniform temperature distribution enhances sorption kinetics efficiency. Recent studies emphasize the importance of uniform temperature distribution across the metal hydride bed for improved HE performance.

MH forms when a metal compound absorbs hydrogen through sorption, which is illustrated by the pressure-composition-temperature (PCT) connection curve. Initially, a metal solution (α phase) forms just before an increase in hydrogen pressure and concentration. MH formation (β phase) begins under these conditions, reaching a plateau once α and β phases reach equilibrium. This process is represented by the equation:

$$H2 + Metal \rightleftharpoons MH + \Delta HR; \Delta HR > 0 \tag{1}$$
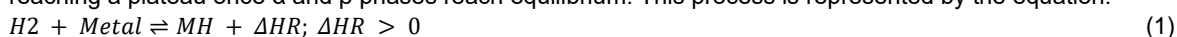
Due to its safe operation and high hydrogen density per unit volume, MH technology holds promise for hydrogen storage. However, its low hydrogen density per unit mass and slow charging and discharging rates limit its application in mobile contexts. One potential solution is integrating MH reactors into structural elements of mobile applications, similar to incorporating lithium batteries into electric cars, to make them smaller and lighter (Lesmana and Aziz, 2023). Previous research has explored utilizing metamaterial-based reactors to enhance charging efficiency and hydrogen capacity within vehicle structures. The metamaterial structural qualities, such as high compression capability and surface area density, enable this feature.

Cellular materials exhibit opposing mechanical characteristics, combining low density with high stiffness (Sinha and Mukhopadhyay, 2023). However, finding naturally occurring materials that possess all the necessary qualities in a single cohesive unit is challenging. There are limitations to extending the qualities of natural materials. The mechanical and physical properties of natural materials, like mass density, are interconnected. Typically, traditional materials with high strength and stiffness have high mass density and vice versa. Overcoming these connections to invent materials that are simultaneously stiff, strong, and lightweight is crucial in fields like mechanical engineering, robotics, and aerospace. Such materials are often classified as mechanical metamaterials (Contreras et al., 2024).
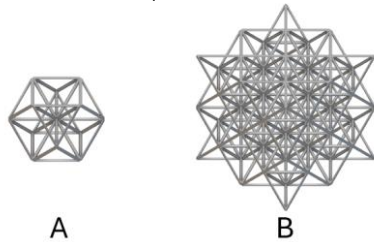

Figure 1: Visualisation of (a) single cell lattice octet and (b) 2x2x2 matrix of connected octet cells

Previous studies have explored the relationship between topology and increasing nodal connectivity, leading to enhanced macroscopic rigidity (Panesar et al., 2018). The octet lattice demonstrates unique characteristics, including high compressive strength and linear scaling strength with consistent and reliable density under various conditions. As a lattice metamaterial, the octet lattice is recognised for its ability to produce combinations of high stiffness and strength. Figure 1 illustrates the octet lattice and its visualisation from the strut junction to the matrix of interconnected cells.

The above study indicates a strong correlation between MH bed design and HE implementation to further achieve more effective sorption kinetics of hydrogen, and other studies show cellular materials characteristic of high compression strength capability (Kaur and Singh, 2021). However, a numerical study of hydrogen storage utilising metamaterial in a lattice structure was found to be limited. The object of this study is to analyse the opportunity for octet structure to be implemented as a solid hydrogen storage option that could replace structural parts for high mobility usage scenarios like hydrogen cars as car chassis. Previous studies utilising the gyroid structure show great usability of the same concept (Lesmana and Aziz, 2022). In this study, the effect of hydrogen charging rate by using the octet structure is investigated using CFD analysis.

2. Method

2.1 Reactor Design Preparation

This study uses a simple case of a canister supported by an octet structure. The octet structure, inherently self-supporting, is feasible for manufacturing through 3D printing. However, if certain portions of the strut are absent, it results in overhangs, making the structure challenging to handle during processing. All the octet struts are required to exist. This study uses a tube canister model with hollowed walls and piping channels inside the bed following the octet strut shapes. The size of the model is 60 mm in height and 40 mm in diameter. Octet struts with 3 mm thickness were then designed to have 1 mm channels inside them for the heat transfer fluid to flow. MH bed and the internal octet are visualised in Figure 2(a), while the octet structure itself is depicted in Figure 2(b). The fluid chamber can be viewed in Figure 2(c), which contains both the inner channel inside the octet strut and the chamber that envelops the MH bed. The MH bed 3D model was then used in CFD analysis for evaluating the performance of hydrogen absorption and compared with a simple pipe-in-pipe design that uses the same heat transfer fluid domain volume as depicted in Figure 2(d).
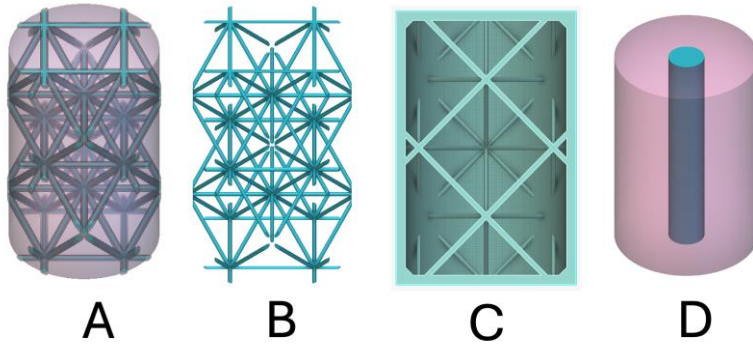
Figure 2: Visualisation of (a) MH bed, (b) octet reinforcement structure ,(c) heat transfer fluid domain and (d) simple baseline reactor design of pipe inside pipe with same fluid domain volume

2.2 Numerical Model

The governing equations for the sorption kinetics of hydrogen in LaNi$_5$ bed include volume averaged energy balance equation, volume averaged mass balance equation, and reaction kinetics of MH. To simplify the numerical model, the following assumptions are made: (1) hydrogen in the gas phase is treated as an ideal gas, (2) local thermal equilibrium is assumed between hydrogen and the metal hydride (MH), (3) the MH bed is regarded as a homogeneous, porous, and isotropic medium, (4) the volumetric expansion of the MH bed is ignored, and its properties, such as porosity, permeability, and thermal conductivity, are considered constant, and (5) the MH bed is assumed to be perfectly in contact and uniformly packed. The energy balance equation is expressed as,

$$(\rho c_p)_{eff} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(k_{eff} \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(k_{eff} \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(k_{eff} \frac{\partial T}{\partial z}\right) - (1-\varepsilon)\dot{m}\,\Delta H \tag{2}$$

$$(\rho c_p)_{eff} = \varepsilon(\rho c_p)_{gas} + (1-\varepsilon)(\rho c_p)_{mh} \tag{3}$$

$$k_{eff} = \varepsilon\, k_{gas} + (1-\varepsilon)\, k_{mh} \tag{4}$$

The mass balance equation is expressed as,

$$(\varepsilon)\frac{\partial \rho}{\partial t} = \dot{m} + (1-\varepsilon)\frac{\partial}{\partial x}\left(D\frac{\partial \rho}{\partial x}\right) + (1-\varepsilon)\frac{\partial}{\partial y}\left(D\frac{\partial \rho}{\partial y}\right) + (1-\varepsilon)\frac{\partial}{\partial z}\left(D\frac{\partial \rho}{\partial z}\right) \tag{5}$$

$$D = D_o\exp\left(\frac{-H_a}{k_B T}\right) \tag{6}$$

The rate at which hydrogen is absorbed per unit volume during absorption can be expressed as a function of several parameters, including density (ρ), mass diffusion coefficient (D), porosity of the material (ε), and the mass of hydrogen absorbed over time ($\dot{m}$). Additionally, there are other factors involved such as the pre-exponential factor (D$_o$), the Boltzmann constant (k$_B$), the activation enthalpy (H$_a$), and the temperature (T).

$$\dot{m}_a = C_a\exp\left(\frac{-E_a}{RT}\right)\ln\left(\frac{P}{P_{eq}}\right)\left(\rho_{sat} - \rho_{t,a}\right) \tag{7}$$

In this context, ρ_{sat} and $\rho_{t,a}$ denote the saturated density and the density of LaNi$_5$ MH when it's empty. P$_{eq}$ stands for pressure equilibrium, which can be mathematically described using the Van 't Hoff equation.

$$\ln(P_{eq}) = A - \frac{B}{T} \tag{8}$$

where A and B denote Van 't Hoff constants.
With HE is integrated into the system, the flow of the coolant at a constant rate is controlled by an energy balance equation.

$$(\rho c_p)_f \frac{\partial T_f}{\partial t} + (\rho c_p)_f \vec{u}\, grad T_f = div(k_f\, grad T_f) \tag{9}$$

Initially, the MH pressure, density, and temperature are considered constant.

$$T = T_0, \ \rho = \rho_0, \ P = P_0 \quad u_x = u_y = 0, u_z = u_{in}, T = T_{in} \tag{10}$$

$$u_x = u_y = 0, u_z = u_{in}, T = T_{in} \tag{11}$$

A case study was used to validate the numerical model created in COMSOL. By using material properties from Table 1, a case study canister was designed and used as geometry for MH bed analysis. A fine mesh setting was used for this study. Hydrogen inlet pressure is set to 15 bar. The comparison between the numerical model and experiment is presented in Figure 3. The comparison shows that the mathematical model represents the experiment result with the deviation that is assumed caused by the assumption that is being made.

Table 1: MH LaNi$_5$ properties

| Properties	Value
Density, ρ (kg m^{-3})	8,200
Specific heat, c_p (J kg^{-1} K^{-1})	419
Thermal conductivity, k (W m^{-1} K^{-1})	2.4
Porosity, ε $(-)$	0.5
Van 't Hoff constants used in Eq. (2.7), A $(-)$, B (K)	12.99; 3,704.59
Plateau slope, α $(-)$	0.038
Hysteresis factor, β $(-)$	0.137
Initial concentration of MH bed, c_0 (mol m^{-3})	18,981.6
Activation energy – Absorption, E_a (J mol^{-1})	21,170
Initial pressure P_0 (MPa)	2
Material constant used in Eq. (2.6), C_a (s^{-1})	59.187
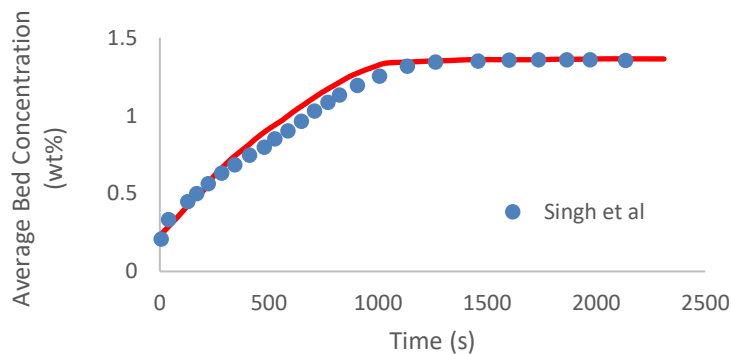
Figure 3: Comparison between past experiment and numerical model

3. Result and Discussion

The simulation used $300\frac{W}{m^2 K}$ heat transfer coefficient to simulate the heat transfer fluid flowing trough the channel inside the octet struts. Figure 4a show the hydrogen fraction over time alongside the concentration map. During the first charging state under 1,000 s, the high absorption state is only concentrated around the cooled area. The result shows direct corelation with heat transfer performance to improve the reaction rate of hydration. The trend also shows that the growing hydrogen fraction is increasing from the area where octet strut are located. Compared with baseline design, the octet insert shows faster charging rate on average (see Figure 5).
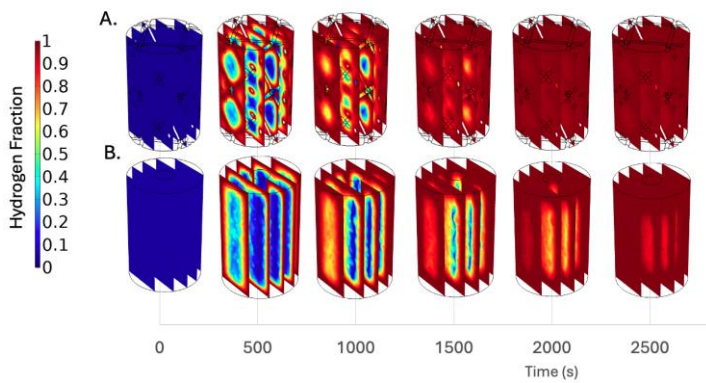
Figure 4: Concentration of hydrogen fraction over time of (a) octet insert structure canister and (b)simple pipe in pipe canister for baseline design
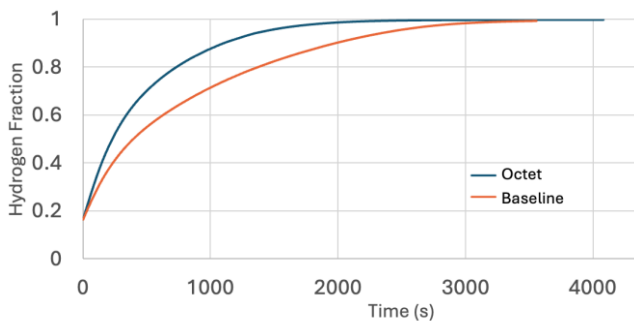

Figure 5. Average hydrogen fraction over time of (a) octet insert structure canister and (b)simple pipe in pipe canister for baseline design

During 1,000 s, the charging rate slope for octet design is at the highest rate, and the effect of cooling of the octet strut can be observed through the initial MH that reaches the full fraction located on the parameter of the strut. Full charge state of this MH bed reached at 2,000 s, however 90 % of charging state only requires 1,200 s. This shows a favourable outcome considering charging and discharging rate of MH bed can be scaled to a bigger volume by applying same heat transfer solution. Compared with octet-based reactor, the Baseline reactor shows a slower charging rate, requiring 2,000 s for 90 % charged and almost 3,500 s to be fully charged.

The temperature map during the same period also provided in Figure 6. The heat transfer effect from the fluid chamber to the MH bed also can be observed and the temperature returns back to room temperature once the hydrogen fraction becomes 1, resulted from the exothermic reaction is no longer occurred. Enhancing the performance of this canister could involve addressing the blue spotted region noticeable on the state of charge map depicted in Figure 5. This could be achieved by either incorporating additional struts or increasing the cell count. This adjustment would lead to more uniform structures that function as effective heat transfer surfaces, augmenting the reaction rate of the metal hydride (MH).
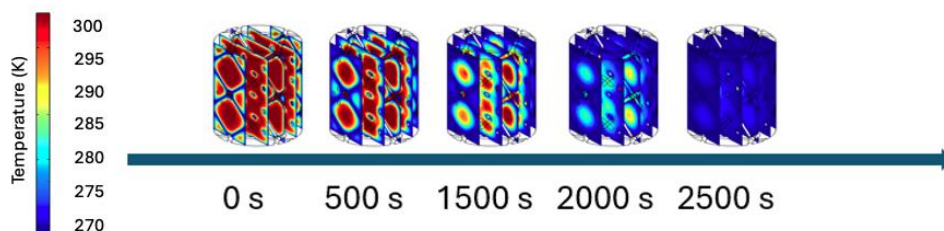

Figure 5: MH temperature distribution during charging

4. Conclusion

The effect of octet insert as canister support and heat transfer enhancement is investigated in this study. This study investigated the use of an octet lattice structure in MH reactor for hydrogen storage, targeting high mobility applications like hydrogen-powered vehicles. The octet structure, designed for 3D printing, integrated fluid channels to enhance heat transfer, crucial for efficient hydrogen absorption. Numerical model analysis showed a strong link between efficient heat transfer and increased hydrogen absorption rates, with full hydrogenation achieved in about 2,000 s and 90 % capacity in 1,200 s. Improved heat transfer was observed around the cooled octet struts compared to baseline reactor design with only pipe in pipe, indicating faster hydrogen absorption that connected with more surface area provided by octet insert when compared with simple pipe in pipe deseign. Further enhancements could be made by increasing the number of struts or reducing cell sizes for better heat distribution. The octet lattice structure shows great potential for mobile hydrogen storage due to its structural strength and efficient heat transfer. Integrating MH beds into structural components such as chassis, cases, and bodies can offset the low gravimetric density of MH. With advancements in hydrogen utilization efficiency, hydrogen-based vehan MH reactor for hydrogen storage, targeting high-mobilityhat electric cars currently incorporate lithium batteries for frame integrity, utilizing lattice insert with MH bed similarly could expand hydrogen use, providing a safe and clean energy solution without the hazardous waste issues associated with lithium batteries. Future research should optimize this design and explore larger-scale applications to fully leverage its advantages.

Acknowledgments

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