

Article

Study on the Hydrogen Storage Properties of LaNi₅ Framework Materials

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Abstract

The hydrogen storage properties of LaNi₅ framework materials prepared by filling porous substrates with LaNi₅ powders were studied. Aluminum silicate fiber blanket, 110 pores per inch (PPI) polyester foam, and 70 PPI and 110 PPI nickel foams were used as porous substrates, and 45-, 80-, and 200-mesh LaNi₅ powders were sequentially poured onto the substrates. To reduce particle detachment, LaNi₅ framework materials with 70 and 110 PPI nickel foam substrates were compressed at 0.5 MPa for 20 min. Microstructural analysis showed that LaNi₅ powders were uniformly distributed within the 70 PPI and 110 PPI nickel foam substrates. At 303 K and 4 MPa, all LaNi₅ framework materials could be activated after two hydrogen absorption cycles. PCT test results show that the hydrogen storage capacity values of the LaNi₅ framework materials with the aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam, 110 PPI nickel foam, compressed 70 PPI nickel foam and compressed 110 PPI nickel foam substrates at 303 K are 1.31, 1.27, 1.37 and 1.63, 1.29 wt% and 1.57 wt%, respectively. The corresponding ΔH values are −30.0, −28.4, −27.8, −26.3, −28.4 and −29.2 kJ·mol^{−1}, respectively. After 10 cycles, the capacity retention rates of the LaNi₅ framework materials ranged from 95.7% to 98.8%, all higher than those of the pure 45-, 80-, and 200-mesh LaNi₅ powders (91.7–94.1%). The LaNi₅ framework material with the 110 PPI nickel foam substrate exhibits the best comprehensive hydrogen storage properties.

Keywords: LaNi₅ framework materials; Hydrogen storage properties; LaNi₅ powders; porous substrates; heat and mass transfer

1. Introduction

Hydrogen has become a promising, new clean energy carrier due to its high energy conversion efficiency, zero carbon emissions, and wide availability [1–3]. Hydrogen storage remains a major bottleneck for the large-scale utilization of hydrogen energy [4,5]. The low density of hydrogen gas results in low volumetric storage density for conventional gaseous storage. Although high-pressure storage can increase the gaseous hydrogen storage density (at 70 MPa, the volumetric storage density is approximately 40 g·L^{−1}), it also brings safety risks [6,7]. Although low-temperature liquid-hydrogen storage has higher volumetric storage density (at 20 K and 0.1 MPa, the volumetric storage density is approximately 70 g·L^{−1}), it requires maintaining an ultra-low temperature of 20 K, which involves high liquefaction energy consumption and strict requirements for storage containers [8]. In recent years, solid hydrogen storage has attracted widespread attention

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owing to its high volumetric storage density, moderate hydrogen storage pressure, and safety [9,10].

Metal hydrides are the most widely investigated and applied solid hydrogen storage materials [11,12]: $2M + xH_2 \rightleftharpoons 2MH_x$. Among various metal hydrides, LaNi₅ alloy has a hydrogen storage capacity of approximately 1.5 wt%. Despite its moderate hydrogen storage capacity, its mild hydrogen absorption/desorption conditions, easy activation, and rapid kinetics have attracted extensive attention [13,14], making LaNi₅ alloy one of the most widely used commercial hydrogen storage alloys. It has been widely applied in vehicle-mounted solid-state hydrogen storage tanks, stationary hydrogen storage systems, and negative electrodes of nickel–metal hydride batteries. Despite these advantages, the large-scale practical application of LaNi₅ alloy still faces challenges. In practical applications, LaNi₅ alloy is used in powder form. During hydrogen absorption and desorption cycles, the volume expansion and contraction of LaNi₅ powder particles generate internal stresses, which cause particle cracking and pulverization [15,16]. Pulverization reduces hydrogen storage capacity, and the accumulation of fine particles creates a dense powder bed with poor hydrogen gas permeability. Moreover, the hydrogen absorption reaction of LaNi₅ is an exothermic reaction. The dense powder bed hinders the timely dissipation of reaction heat, resulting in local heat accumulation and preventing the complete progress of the hydrogen absorption reaction. As a result, the capacity retention of LaNi₅ powder decreases rapidly.

To address the pulverization of LaNi₅ powder particles, recent research has mainly focused on improving the structural stability of LaNi₅ alloy through elemental substitution [17,18]. Chen et al. [14] reported that Mn substitution in LaNi_{5-x}Mn_x ($x = 0, 0.25, 0.5, 0.75, 1$) alloys stabilized the crystal structure during hydrogen cycling, with LaNi_{4.25}Mn_{0.75} achieving 94.0% capacity retention after 1000 cycles. However, the introduction of larger Mn atoms reduces interstitial sites for hydrogen storage, resulting in decreased hydrogen capacity. Liu et al. [19] found that Al substitution in LaNi_{5-x}Al_x ($x = 0, 0.25, 0.5$) alloys reduced lattice expansion during hydrogenation and enhanced cycling stability, while excessive Al addition reduced hydrogen storage capacity due to the occupation of hydrogen storage sites. Zhu et al. [20] demonstrated that Co substitution enhanced the cycling stability of LaNi_{5-x}Co_x ($x = 0, 0.25, 0.50, 0.75$) alloys. After 1000 cycles, LaNi_{4.5}Co_{0.5} maintained a more complete crystal structure and larger particle size, and its cycle stability was significantly better than that of pure LaNi₅.

Elemental substitution can improve the structural stability of LaNi₅ alloys to a certain extent, but this method often compromises hydrogen storage capacity and fails to eliminate ~25% volume expansion during the hydrogen absorption process of LaNi₅ [21]. The expansion generates internal stresses that lead to cracking and eventual pulverization. Therefore, completely preventing pulverization during hydrogen absorption/desorption cycles remains challenging, and the key issue is how to improve heat and mass transfer. In recent years, the polymer encapsulation and porous framework strategies have attracted much attention. Zadorozhnyy et al. [22] prepared three-dimensional porous LaNi₅ bulk materials by laser beam powder bed fusion, which exhibited significantly higher thermal conductivity than LaNi₅ powders. Ugaddan et al. [23] embedded AB₅-type metal hydride powders in polyethylene (PE) and polymethyl methacrylate (PMMA) matrices to prepare polymer-based composite pellets by cold pressing. The polymer matrices effectively suppressed alloy pulverization, with the PE-based composite maintaining stable hydrogen storage performance and less than 3% powder loss after 20 hydrogen absorption/desorption cycles.

Based on the above, this study proposes a novel structural design strategy for LaNi₅ hydrogen storage alloys. LaNi₅ framework materials with aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam, and 110 PPI nickel foam substrates were

prepared by filling the porous substrates with LaNi₅ powder of the different particle sizes. This design utilizes the interconnected pore network in the porous substrates to facilitate hydrogen gas flow and rapid heat dissipation. This strategy provides a feasible approach to developing solid hydrogen storage materials with excellent cycling stability.

2. Experimental Methods

2.1. Preparation and Characterization

To improve the heat and mass transfer of LaNi₅ powders, LaNi₅ framework materials were prepared by filling porous substrates with LaNi₅ powders. Commercial LaNi₅ powder with a purity of 99.9% was purchased from Baotou Lanxi New Materials Technology Co., Ltd (Baotou, China). Three types of LaNi₅ powder with different particle sizes (45, 80, and 200 mesh) and four types of porous substrates (aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam and 110 PPI nickel foam) were used in this study. The macroscopic photographs of the porous substrates are provided in Figure S1 to illustrate their morphologies and dimensions. Among these substrates, the 70 PPI and 110 PPI nickel foam substrates were used to investigate the effects of pore size, whereas the aluminum silicate fiber blanket and 110 PPI polyester foam substrates were used to investigate the effects of thermal conductivity and physicochemical properties on the hydrogen storage capacity of LaNi₅ alloy powders.

The porous substrate was first fully immersed in polyethylene glycol (PEG) solution. Subsequently, 45-, 80-, and 200-mesh LaNi₅ powders were poured onto the porous substrates in turn. Afterward, the porous substrates filled with LaNi₅ powders were vacuum-dried at 0.1 Pa to remove the solvent and then heated to 300 °C at a heating rate of 5 °C·min⁻¹ under the same vacuum condition and maintained for 30 min to complete the solidification of PEG.

To prevent the detachment of powder during hydrogen absorption and desorption cycling, the LaNi₅ framework materials with 70 PPI and 110 PPI nickel foam substrates were compressed at 0.5 MPa for 20 min. The pore sizes of the porous substrates and the particle sizes of the LaNi₅ powder used in this study are summarized in Table 1. The loading rates of the LaNi₅ framework materials, defined as the mass ratio of LaNi₅ powder to the total mass of LaNi₅ framework material, are listed in Table 2.

Table 1. Pore sizes of porous substrates and average particle sizes of LaNi₅ powder used in this study.

Porous Substrate	Pore Size (μm)	LaNi ₅ Powder	Average Particle Size (μm)
70 PPI	~350–450	45 mesh	~355
110 PPI	~200–300	80 mesh	~180
		200 mesh	~75

Note: The conversion relationship between PPI and m⁻¹ is 1 PPI ≈ 39.37 m⁻¹.

Table 2. LaNi₅ loading rate of LaNi₅ framework materials.

Substrate	Substrate Mass (g)	LaNi ₅ Mass (g)	LaNi ₅ Loading Rate (wt%)
Aluminum silicate fiber blanket	6.63	29.47	81.63
110 PPI polyester foam	3.27	9.83	75.04
70 PPI nickel foam	2.30	18.32	88.85
110 PPI nickel foam	1.22	6.94	85.05
70 PPI nickel foam (compressed)	2.42	14.38	85.60
110 PPI nickel foam (compressed)	1.12	9.73	89.68

A PHENOMWORLD scanning electron microscope (Phenom-World B.V., Eindhoven, The Netherlands) was used to observe the microstructures of the LaNi₅ framework materials in secondary electron mode. The accelerating voltage was 15 kV, and the chamber vacuum was 1 Pa. Mercury intrusion porosimetry (MicroActive AutoPore V 9600) (Micromeritics Instrument Corporation, Norcross, GA, USA) was used to measure the pore sizes and the porosity of the LaNi₅ framework materials.

2.2. Hydrogen Storage Property Measurements

Activation, pressure–composition–temperature (PCT) and cycling performance measurements were performed using a Sieverts apparatus (Zhejiang University, Hangzhou, China). For the Sieverts measurements, samples in the shape of small rectangular pieces were cut from the prepared framework materials and then placed in the sample chamber. The mass of LaNi₅ powder contained in each sample was calculated by the total mass of the sample multiplied by the corresponding LaNi₅ loading rate listed in Table 2.

The Sieverts apparatus used in this study has a pressure measurement range of 0.1 Pa to 10 MPa, a temperature control range from room temperature to 800 °C, a hydrogen purity of 99.99%, and a sample chamber volume of 79 mL. Before testing, the samples were evacuated under vacuum at 423 K for 30 min to remove surface oxides. The temperature was then cooled to 303 K, and hydrogen absorption activation tests were conducted under a hydrogen pressure of 4 MPa.

The PCT curves were measured at 293 K, 303 K, and 313 K. The enthalpy change (ΔH) and entropy change (ΔS) during hydrogen absorption were calculated using the Van 't Hoff equation:

$$\ln\left(\frac{P_{\text{eq}}}{P^0}\right) = \frac{\Delta H}{RT} - \frac{\Delta S}{R} \quad (1)$$

where P_{eq} is the equilibrium plateau pressure, P^0 is the standard pressure (0.1 MPa) and R is the universal gas constant.

The cycling performance tests were conducted at 303 K under a hydrogen pressure of 4 MPa. The hydrogen capacity retention after 10 cycles was calculated according to the ratio between the hydrogen storage capacity after cycling and the initial hydrogen storage capacity.

3. Results

3.1. Microstructure

The microstructures of LaNi₅ framework materials with the different substrates are shown in Figure 1. The differently sized LaNi₅ powder is uniformly distributed on the surface of the aluminum silicate fiber substrate (Figure 1a); however, due to the relatively large interval of aluminum silicate fiber blanket substrate, it does not fully fill the interior of the substrate (Figure 1b). The loading rate of the aluminum silicate fiber blanket substrate is 81.63 wt%. For the 110 PPI polyester foam substrate, the differently sized LaNi₅ powder fully fill the surface (Figure 1c); however, due to the poor connectivity of the 110 PPI polyester foam substrate, the LaNi₅ powder does not fill the internal pores, especially the large particles (Figure 1d). The loading rate of the 110 PPI polyester foam substrate is 75.04 wt%. For the 70 PPI nickel foam substrate, due to its good connectivity and large pore size, the differently sized LaNi₅ powder fully fills the surface and the pores, with a uniform distribution throughout the framework material (Figure 1e,f). The loading rate of the 70 PPI nickel foam substrate reaches 88.85 wt%, which is the highest among these framework materials. For the 110 PPI nickel foam substrate, due to its small pores (200–300 µm), which are smaller than the particle size of 45-mesh powder (~355 µm), only the smaller powder particles filled the interior of the substrate, leaving large particles on

the surface (Figure 1g,h). The loading rate of the 110 PPI nickel foam substrate is 85.05 wt%.

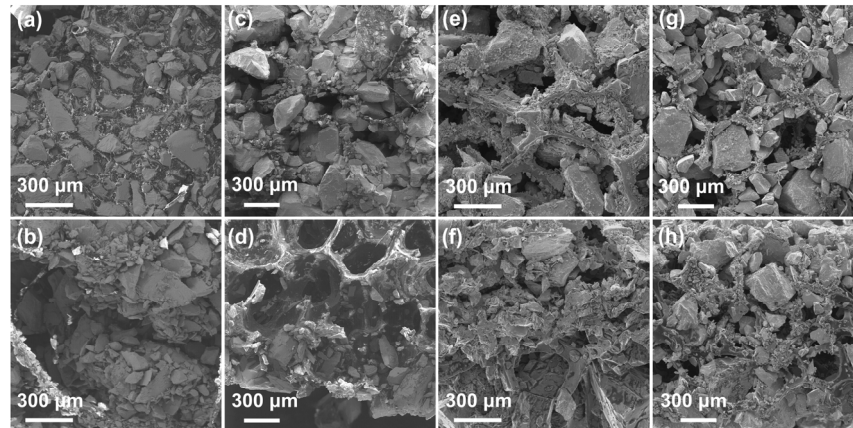


Figure 1. SEM images of LaNi₅ framework materials: (a) surface and (b) side view of aluminum silicate fiber blanket substrate; (c) surface and (d) side view of 110 PPI polyester foam substrate; (e) surface and (f) side view of 70 PPI nickel foam substrate; (g) surface and (h) side view of 110 PPI nickel foam substrate.

To prevent particle detachment, both LaNi₅ framework materials with 70 PPI and 110 PPI nickel foam substrates were compressed at 0.5 MPa for 20 min. Their microstructures are shown in Figure 2. After compression, the microstructure of the LaNi₅ framework material with the 70 PPI nickel foam substrate becomes more packed while still retaining a large number of gaps and pores (Figure 2a,b). Meanwhile, some large LaNi₅ particles on the surface are pressed into the porous interior during compression. For the LaNi₅ framework material with the 110 PPI nickel foam substrate, after compression, the microstructure becomes denser and more compact, with few gaps and pores left (Figure 2c,d). Compression similarly results in the large LaNi₅ particles on the surface being pressed into the porous interior.

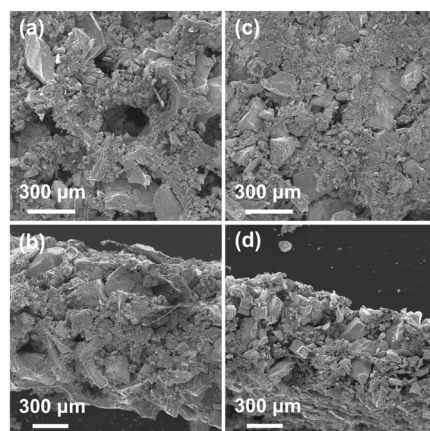


Figure 2. SEM images of compressed LaNi₅ framework materials: (a) surface and (b) side view of 70 PPI nickel foam substrate; (c) surface and (d) side view of compressed 110 PPI nickel foam substrate.

The pore size distribution curves and porosity of the LaNi₅ framework materials with 70 PPI and 110 PPI nickel foam substrates are shown in Figures 3 and 4. The pore sizes of the LaNi₅ framework materials with 70 PPI and 110 PPI nickel foam substrates are determined to be 250 μm and 200 μm (Figure 3), respectively. After compression, the pore

sizes of both framework materials show a significant decrease. The porosity of the LaNi₅ framework material with the 70 PPI nickel foam substrate is determined to be 64.58% (Figure 4), and after compression, it decreases to 58.90%, while the porosity of the LaNi₅ framework material with the 110 PPI nickel foam substrate is 71.42%, and after compression, it decreases to 42.18%, nearly half of its initial value.

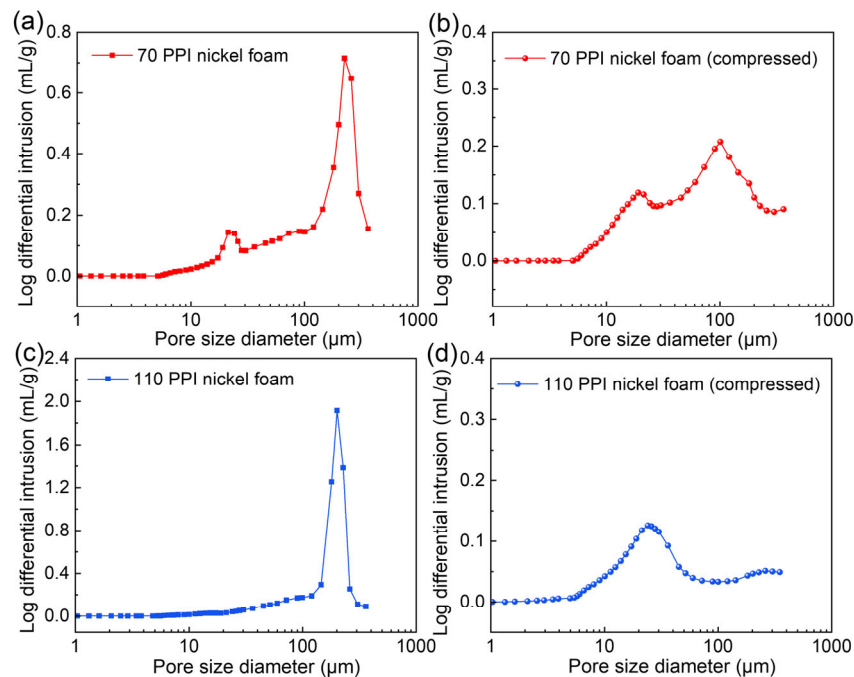


Figure 3. Pore size distribution curves of LaNi₅ framework materials with: (a) 70 PPI nickel foam substrate, (b) compressed 70 PPI nickel foam substrate, (c) 110 PPI nickel foam substrate, and (d) compressed 110 PPI nickel foam substrate.

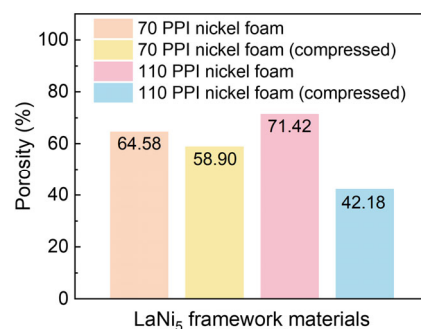


Figure 4. Porosity of LaNi₅ framework materials.

3.2. Activation Curves

The activation curves of the LaNi₅ framework materials for the first and second hydrogen absorption cycles at 303 K and 4 MPa are shown in Figure 5. In the first hydrogen absorption cycle (Figure 5a), the LaNi₅ framework material with the 110 PPI nickel foam substrate has the shortest hydrogen absorption induction period, while that with the 110 PPI polyester foam substrate has the longest induction period. Among all samples, the LaNi₅ framework material with the 110 PPI nickel foam substrate has the maximum hydrogen absorption capacity, while the other three samples have nearly the same, relatively low hydrogen storage capacity. After compression, the hydrogen storage

capacity of both the LaNi₅ framework material with the 70 PPI and 110 PPI nickel foam substrate decreases.

In the second hydrogen absorption cycle (Figure 5b), the hydrogen absorption induction periods of the LaNi₅ framework materials are significantly decreased, implying that they exhibit good activation properties at 303 K and 4 MPa, except for the LaNi₅ framework material with the aluminum silicate fiber blanket substrate, which still has a low hydrogen absorption rate.

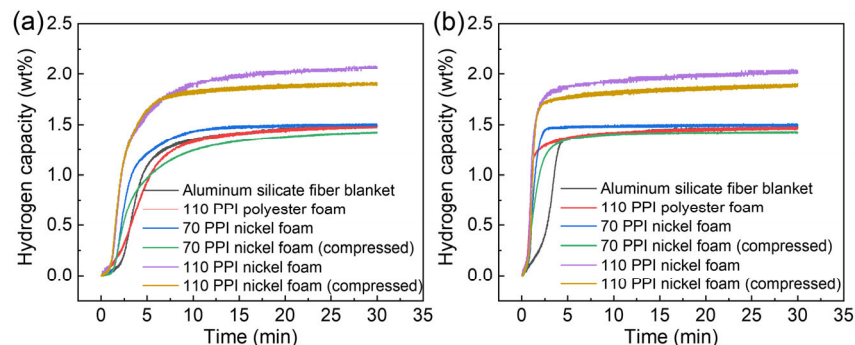


Figure 5. Hydrogen absorption activation curves of LaNi₅ framework materials at 303 K and 4 MPa: (a) First cycle. (b) Second cycle.

3.3. PCT Curves

The PCT curves of the LaNi₅ framework materials at 293 K, 303 K, and 313 K are shown in Figure 6, and the hydrogen storage properties are summarized in Table 3. All the samples exhibit a single hydrogen absorption and desorption plateau, and the platform pressure increases with the increase in temperature. The hydrogen storage capacity values at 303 K are 1.31, 1.27, 1.37 and 1.63 wt% for the LaNi₅ framework materials with the aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam and 110 PPI nickel foam substrates, respectively. The LaNi₅ framework material with the 110 PPI nickel foam substrate has the maximum hydrogen storage capacity. After compression, the hydrogen storage capacity of the LaNi₅ framework materials decreases, from 1.37 wt% to 1.29 wt% for the 70 PPI nickel foam substrate and from 1.63 wt% to 1.57 wt% for the 110 PPI nickel foam substrate.

Based on the Van 't Hoff equation and the hydrogen absorption plateau pressure at different temperatures, the ΔH and ΔS of hydrogen absorption of the LaNi₅ framework materials are obtained (as shown in Figure S2) and are listed in Table 3. The ΔH values of the LaNi₅ framework materials with aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam and 110 PPI nickel foam are determined to be -30.0 , -28.4 , -27.8 and -26.3 kJ·mol⁻¹, respectively. Among them, the LaNi₅ framework material with the 110 PPI nickel foam substrate has the smallest absolute ΔH value, implying that it has the lowest thermodynamic stability of the hydride phase. After compression, the ΔH of the LaNi₅ framework materials with the 70 PPI and 110 PPI nickel foam substrates decreases to -28.4 and -29.2 kJ·mol⁻¹, respectively.

The ΔS values of the LaNi₅ framework materials range from -98.5 to -111.0 J·mol⁻¹·K⁻¹, exhibiting the same variation trend as ΔH .

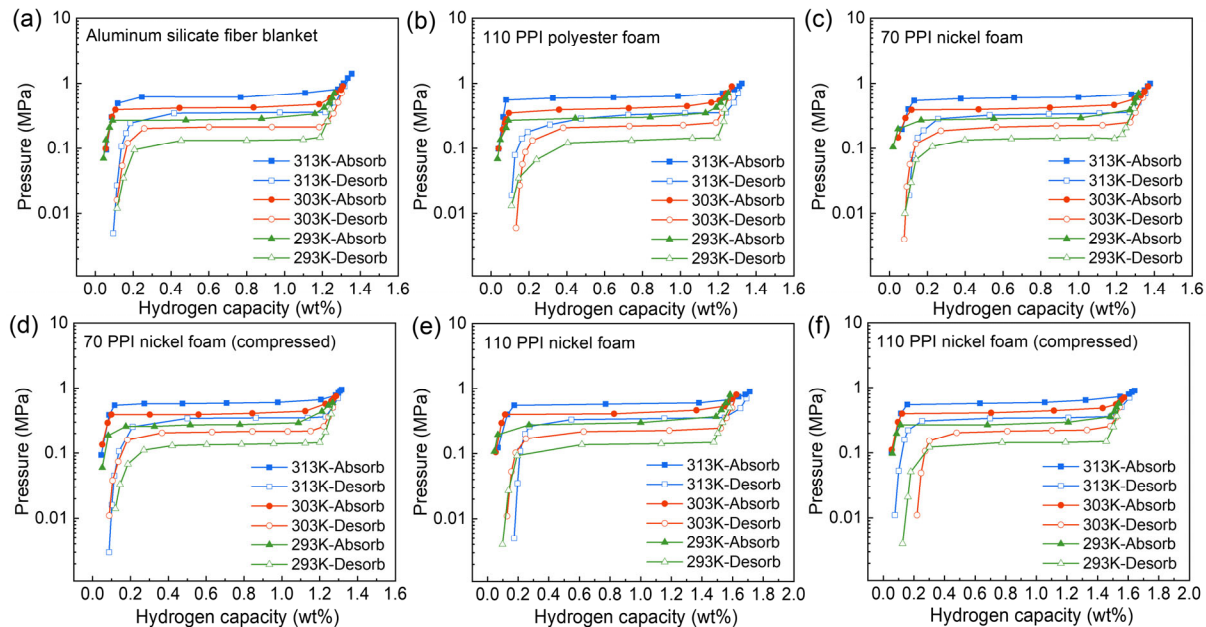


Figure 6. PCT curves of LaNi₅ framework materials with (a) aluminum silicate fiber blanket, (b) 110 PPI polyester foam, (c) 70 PPI nickel foam, (d) compressed 70 PPI nickel foam, (e) 110 PPI nickel foam and (f) compressed 110 PPI nickel foam substrates at 293 K, 303 K and 313 K.

Table 3. Hydrogen storage property parameters of LaNi₅ framework materials at 293, 303, and 313 K.

Sample	Temperature (K)	Capacity (wt%)	P_{ab} (MPa)	P_{de} (MPa)	ΔH (kJ·mol ⁻¹)	ΔS (J·mol ⁻¹ ·K ⁻¹)
Aluminum silicate fiber blanket	293	1.26	0.277	0.132	−30.0	−111.0
	303	1.31	0.434	0.212		
	313	1.36	0.608	0.351		
110 PPI polyester foam	293	1.25	0.291	0.133	−28.4	−105.5
	303	1.27	0.406	0.219		
	313	1.32	0.613	0.331		
70 PPI nickel foam	293	1.32	0.288	0.138	−27.8	−103.7
	303	1.37	0.411	0.217		
	313	1.38	0.598	0.322		
110 PPI nickel foam	293	1.58	0.289	0.143	−26.3	−98.5
	303	1.63	0.408	0.222		
	313	1.71	0.576	0.347		
Compressed 70 PPI nickel foam	293	1.27	0.276	0.136	−28.4	−105.2
	303	1.29	0.399	0.212		
	313	1.32	0.581	0.346		
Compressed 110 PPI nickel foam	293	1.54	0.272	0.144	−29.2	−107.9
	303	1.57	0.417	0.218		
	313	1.64	0.584	0.343		

For comparison, the PCT curves of 45-mesh, 80-mesh, and 200-mesh LaNi₅ powders at 293 K, 303 K, and 313 K are shown in Figure 7, and the hydrogen storage properties are summarized in Table 4. ΔH and ΔS are obtained using the Van 't Hoff equation shown in Figure S3. The ΔH values of the 45-, 80-, and 200-mesh LaNi₅ powders are −30.8, −31.3, and −31.4 kJ·mol⁻¹, respectively. The relatively small variation in ΔH indicates that particle size has little influence on the thermodynamic properties of LaNi₅ powder. Therefore, the

differences in ΔH among the LaNi_5 framework materials may be related to framework structure, instead of particle size.

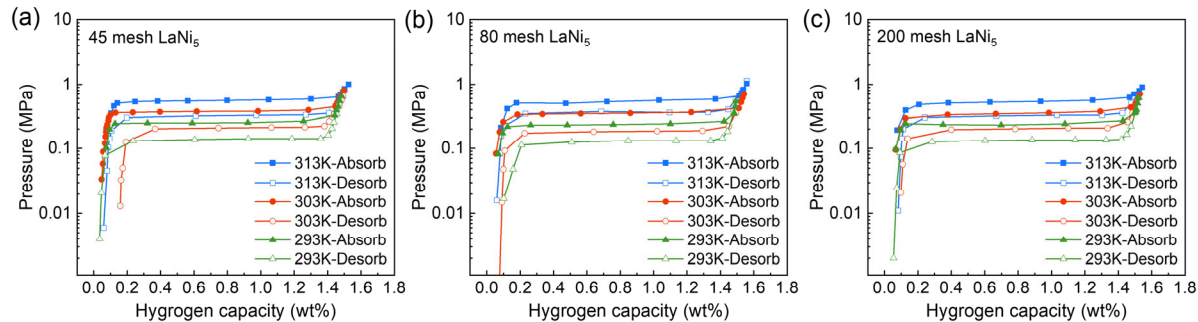


Figure 7. PCT curves of (a) 45-mesh, (b) 80-mesh and (c) 200-mesh LaNi_5 powders at 293 K, 303 K and 313 K.

Table 4. Hydrogen storage property parameters of 45-mesh, 80-mesh and 200-mesh LaNi_5 powders at 293, 303, and 313 K.

Sample	Temperature (K)	Capacity (wt%)	P_{ab} (MPa)	P_{de} (MPa)	ΔH (kJ·mol ⁻¹)	ΔS (J·mol ⁻¹ ·K ⁻¹)
45 mesh	293	1.48	0.249	0.140	−30.8	−112.6
	303	1.50	0.382	0.209		
	313	1.53	0.558	0.322		
80 mesh	293	1.49	0.236	0.131	−31.3	−114.0
	303	1.54	0.354	0.184		
	313	1.56	0.537	0.337		
200 mesh	293	1.52	0.233	0.132	−31.4	−114.1
	303	1.53	0.348	0.201		
	313	1.54	0.531	0.324		

3.4. Capacity Retention

The hydrogen absorption curves and capacity retention rates of the LaNi_5 framework materials and pure LaNi_5 powder after 10 cycles are shown in Figure 8 and 9. It can be seen that the capacity retention rates of the LaNi_5 framework materials with the aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam and 110 PPI nickel foam substrates are 98.5, 95.7, 97.8 and 98.8%, respectively. Among them, the LaNi_5 framework material with the 110 PPI nickel foam substrate has the highest capacity retention rate. After compression, the capacity retention rate of the LaNi_5 framework material with the 70 PPI nickel foam substrate increases from 97.8% to 98.5%, while that of the 110 PPI nickel foam substrate decreases from 98.8% to 96.6%. After 10 cycles, the capacity retention rates of the 45-mesh, 80-mesh, and 200-mesh LaNi_5 powders are 92.6%, 91.7%, and 94.1%, respectively. The microstructures of the LaNi_5 framework materials after 10 cycles are shown in Figure S4. The SEM images show that the LaNi_5 particles retained relatively large particle sizes without obvious pulverization.

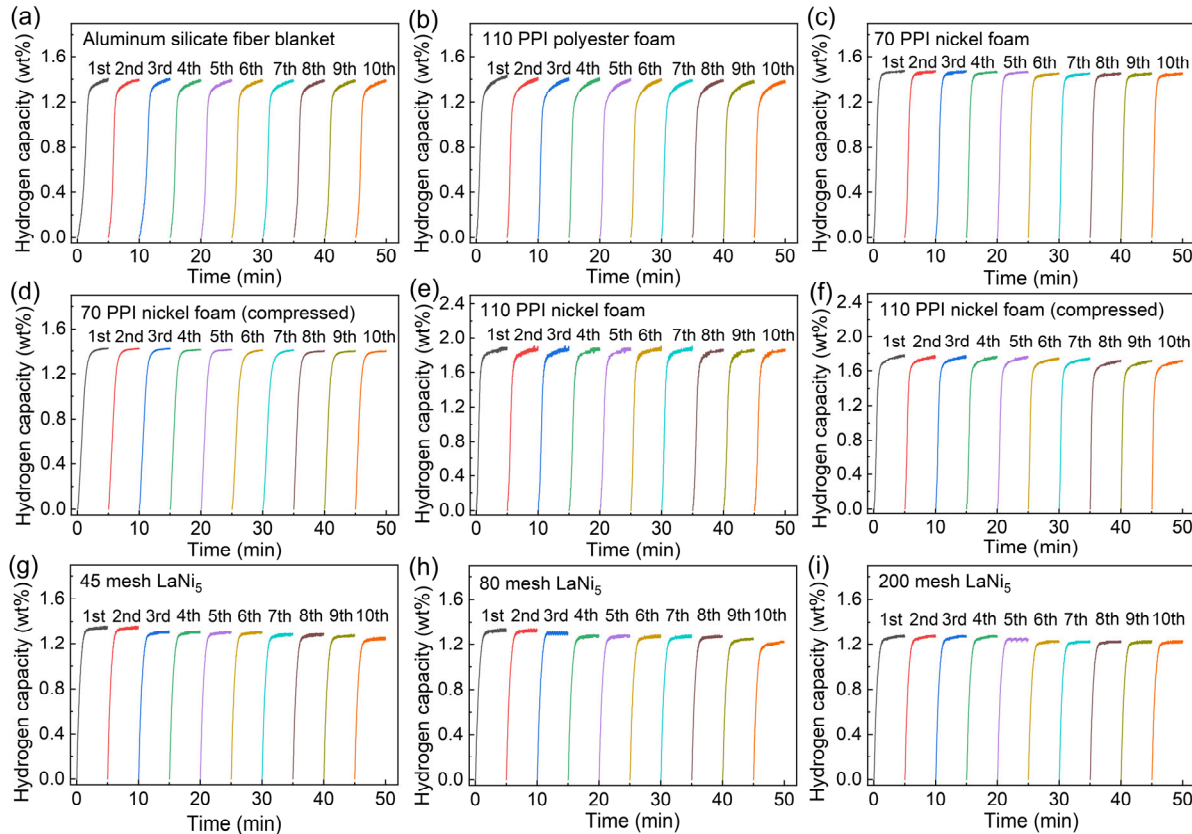


Figure 8. Hydrogen absorption curves of the LaNi₅ framework materials with (a) aluminum silicate fiber blanket, (b) 110 PPI polyester foam, (c) 70 PPI nickel foam, (d) compressed 70 PPI nickel foam, (e) 110 PPI nickel foam, (f) compressed 110 PPI nickel foam substrates and (g) 45-mesh, (h) 80-mesh and (i) 200-mesh LaNi₅ powders during 10 cycles.

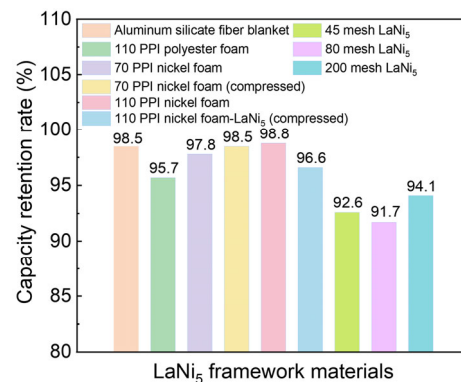


Figure 9. Capacity retention rates of the LaNi₅ framework materials and LaNi₅ powders after 10 cycles.

4. Discussion

The capacity retention rates of all LaNi₅ framework materials (95.7–98.8%) are higher than those of pure LaNi₅ powder with different particle sizes (91.7–94.1%) after 10 cycles of hydrogen absorption and desorption. This improvement may be attributed to the porous substrates, which enhance heat and mass transfer and reduce the pulverization of LaNi₅ powder during cycling.

In practical applications, LaNi₅ alloy is used in powder form. The repeated volume expansion and contraction during hydrogen absorption and desorption cycles generate internal stresses within the alloy particles, ultimately leading to particle cracking and pulverization. The pulverization of powder into fine particles inhibits hydrogen transport. As a result, the hydrogen storage capacity of LaNi₅ powder is limited. In addition, hydrogen absorption is an exothermic reaction that generates a large amount of reaction heat. The poor heat transfer performance of the dense powder bed inhibits the timely release of reaction heat, resulting in local heat accumulation, which inhibits the hydrogen absorption reaction process, making the hydrogen absorption reaction insufficient. As a result, the hydrogen storage capacity of LaNi₅ powder decreases.

In this study, porous substrates are filled with LaNi₅ powder, where the powder is uniformly distributed within the porous substrates, forming a large number of pores supported by the porous substrates. These pores provide sufficient extra space for LaNi₅ powder, facilitating the timely transfer of reaction heat. As a result, the reaction can proceed more thoroughly, thereby maintaining high hydrogen absorption capacity. At the same time, the sufficient extra space is also beneficial to the rapid flow of hydrogen gas, enabling almost all individual LaNi₅ powder particles to fully participate in the hydrogen absorption and desorption reactions and thereby maintain high hydrogen absorption capacity during cycling.

In addition, the LaNi₅ framework materials can also reduce the pulverization of LaNi₅ powder. Due to sufficient extra space within the frameworks, the microstress caused by the expansion and contraction of LaNi₅ powder, as well as temperature fluctuations during hydrogen absorption and desorption, can be reduced. As a result, the pulverization of LaNi₅ powder can be inhibited, resulting in higher capacity retention rates of LaNi₅ framework materials.

The heat and mass transfer properties are related to the pore structure of the materials. Laurencelle et al. [24] confirmed through simulations that the effective thermal conductivity of pure LaNi₅ powder is approximately 0.15 W·m⁻¹·K⁻¹, while the effective thermal conductivity of aluminum foam filled with LaNi₅ powder increases to 10.0 W·m⁻¹·K⁻¹, demonstrating that the porous structure can significantly enhance the heat transfer performance of LaNi₅ powder. Regarding mass transfer, the Bruggeman equation is employed to calculate the effective diffusion coefficient of hydrogen in the porous structures, as follows [25]:

$$D_{\text{eff}} = D_0 \varepsilon^{1.5} \quad (2)$$

where D_{eff} is the effective diffusion coefficient; D_0 is the diffusion coefficient of hydrogen in gas phase, which is a constant at the same temperature and pressure; and ε is the porosity. Higher porosity results in a larger D_{eff} . Therefore, porous substrates can enhance the heat and mass transfer of LaNi₅ powder, which is consistent with our analysis.

The hydrogen storage capacity of LaNi₅ framework materials is also influenced by their porosity. As shown in Figure 4, the porosity values of the LaNi₅ framework materials with the 70 PPI and 110 PPI nickel foam substrates are 64.58% and 71.42%, respectively. According to Table 3, the hydrogen storage capacity values of the LaNi₅ framework materials with the 70 PPI and 110 PPI nickel foam substrates are 1.37 wt% and 1.63 wt% at 303 K, respectively. These results indicate that the porosity of LaNi₅ framework materials indeed influences their hydrogen storage capacity. After compression, the porosity of the LaNi₅ framework materials with the 70 and 110 PPI nickel foam substrates decreases, resulting in corresponding decreases in their hydrogen storage capacity.

The hydrogen capacity values of the LaNi₅ framework materials with the 110 PPI and compressed 110 PPI nickel foam substrates, as shown in Figure 5, reach approximately 1.7–2.0 wt%. We speculate that the relatively higher hydrogen storage capacity may be

related to the structural characteristic of the porous substrate. More research work will be needed in future studies to verify this assumption.

Porous substrates also affect the thermodynamic properties of LaNi₅ framework materials. As summarized in Tables 3 and 4, the absolute values of ΔH for the LaNi₅ framework materials (26.3–30.0 kJ·mol^{−1}) are generally lower than that reported for LaNi₅ alloy (30.4 kJ·mol^{−1} [26]). Similarly, the absolute values of ΔS for the LaNi₅ framework materials (98.5–111.0 J·mol^{−1}·K^{−1}) are generally lower than that reported for LaNi₅ alloy (111.2 J·mol^{−1}·K^{−1} [26]). However, the reasons and their underlying mechanism remain unclear and require further investigation.

5. Conclusions

This work systematically investigated the microstructures, activation, PCT curves, thermodynamic properties and cycling stability of LaNi₅ framework materials and compared their cycling stability with pure 45-, 80- and 200-mesh LaNi₅ powder. The main conclusions are summarized as follows.

(1) LaNi₅ framework materials were prepared by filling porous substrates with LaNi₅ powder. LaNi₅ powder was uniformly distributed within the LaNi₅ framework materials with the 70 PPI and 110 PPI nickel foam substrates. After compression, the LaNi₅ powder was pressed into the porous structures, forming a more compact structure.

(2) All LaNi₅ framework materials were activated after two hydrogen absorption cycles at 303 K and 4 MPa, and the LaNi₅ framework material with the 110 PPI nickel foam substrate had the shortest hydrogen absorption induction period.

(3) The porosity of the LaNi₅ framework materials influences their hydrogen storage capacity. The porosity values of the LaNi₅ framework materials with 70 PPI and 110 PPI nickel foam substrates are 64.58% and 71.42%, and their hydrogen storage capacity values at 303 K are 1.37 wt% and 1.63 wt%, respectively.

(4) The ΔH values of the LaNi₅ framework materials with the aluminum silicate fiber blanket, 110 PPI polyester foam, 70 PPI nickel foam, 110 PPI nickel foam, compressed 70 PPI nickel foam, and compressed 110 PPI nickel foam substrates are −30.0, −28.4, −27.8, −26.3, −28.4, and −29.2 kJ·mol^{−1}, respectively.

(5) The capacity retention rates of the LaNi₅ framework materials after 10 cycles (95.7%–98.8%) are all higher than those of pure 45-, 80-, and 200-mesh LaNi₅ powder (91.7%–94.1%).

(6) The LaNi₅ framework material with the 110 PPI nickel foam substrate exhibits the best comprehensive hydrogen storage properties, including high hydrogen capacity (1.63 wt%, at 303 K), the shortest hydrogen absorption induction period, and excellent cycling stability (98.8%, after 10 cycles).

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/met16101095/s1>, Figure S1: Digital photographs showing the macroscopic morphology of porous substrates: (a) aluminum silicate fiber blanket, (b) 110 PPI polyester foam, (c) 70 PPI nickel foam, and (d) 110 PPI nickel foam; Figure S2: Van't Hoff plots log P_{H_2} vs. $1000/T$ of the LaNi₅ framework materials with: (a) Aluminum silicate fiber blanket, (b) 110 PPI polyester foam, (c) 70 PPI nickel foam, (d) 110 PPI nickel foam, (e) compressed 70 PPI nickel foam, and (f) compressed 110 PPI nickel foam substrates; Figure S3: Van't Hoff plots log P_{H_2} vs. $1000/T$ of the (a) 45 mesh, (b) 80 mesh and (c) 200 mesh LaNi₅ powders; Figure S4: SEM images of the LaNi₅ framework materials with: (a) Aluminum silicate fiber blanket; (b) 110 PPI polyester foam; (c) 70 PPI nickel foam; (d) compressed 70 PPI nickel foam; (e) 110 PPI nickel foam and (f) compressed 110 PPI nickel foam substrates after 10 cycles.

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Abbreviations

The following abbreviations are used in this manuscript:

PPI Pores Per Inch

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