Materials Research Directions Towards a Green Hydrogen Economy: A Review

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Abstract

A constellation of technologies have been researched with an eye towards enabling a hydrogen economy. Within the research fields of hydrogen production, storage, and utilization in fuel cells, various classes of materials have been developed targeting higher efficiencies and utility. This Review examines recent progress in these research fields from the years 2011-2021, exploring the concepts and materials directions important to each. Particular attention has been given to catalyst materials enabling the green production of hydrogen from water, chemical and physical storage systems, and materials used in technical capacities within fuel cells. Quantification of publication and materials trends provides a picture of the current state of development within each node of the hydrogen economy.

Introduction

Green hydrogen economy

Despite decades of research towards alternatives, fossil fuels account for more than 80% of global energy consumption today.¹ Facing dwindling natural resources and burgeoning ecological consequences, we are challenged with charting a sustainable course for modern life using renewable energy sources. This will require safe and reliable methods of converting, storing, and using energy that can compete with hydrocarbon fuels extracted from the Earth. While the optimal solutions may vary depending on geographical location and availability of alternative energy-enabling materials, one major proposed avenue is the use of hydrogen as an energy carrier and hydrogen fuel cells as a primary method of converting energy into electricity. The integrated system of hydrogen production, storage, and utilization on a societal scale is aspirationally referred to as the *hydrogen economy*.

Hydrogen has the potential to act as a superior energy carrier when compared to fossil fuels, as it has approximately twice the gravimetric energy density and could have none of the carbon emissions.² However, the primary mode of producing hydrogen today is from reforming of fossil fuels (natural gas, oil, and coal), which together account for 96% of production.³ This is hardly a solution for enabling sustainable energy. Additionally, as of 2020 most (>98%) hydrogen is produced for chemical, nonenergetic purposes such as ammonia production and oil refinement.⁴ It is well known that industrial ammonia production is central to sustaining the Earth's human carrying capacity; this underscores the importance of developing renewable hydrogen production avenues. In this paper, we refer to the integration of *renewable* hydrogen into the global energy system as the *green hydrogen economy* (GHE) and explore research trends in each of the three facets of the green hydrogen economy: green hydrogen production, hydrogen storage, and hydrogen-based fuel cells. Using data from the CAS Content Collection[™], we analyze the academic and patent literature from 2011-2021 to understand the general progress of each field as well as the classes of materials and concepts driving their innovation. As an expert-curated resource, CAS content is utilized here for quantitative analysis of publications against variables including time, country/region, research area, and substance details. We hope that this manuscript serves as a broad overview of the materials research directions driving the potentially transformative set of GHE technologies.

Production by water electrolysis

Efforts towards renewable hydrogen production center on water electrolysis, which proceeds via oxidation at the electrolyzer anode to produce O_2 and reduction at the cathode to produce H_2 . These two reactions are respectively referred to as the oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) and proceed according to the following equations in acidic media (Equations 1-3).

$HER: 2 H^+ + 2 e^- \rightarrow H_2$	(1)
OER: $H_2O \rightarrow \frac{1}{2}O_2 + 2 H^+ + 2 e^-$	(2)
Overall: $H_2O \rightarrow H_2 + \frac{1}{2}O_2$	(3)

These chemical principles can be applied in various electrolyzer configurations to produce H₂ from water. The three primary technologies of interest for industrial application are alkaline electrolyzers, polymer electrolyte membrane (PEM) electrolyzers, and solid oxide electrolyzers (SOEs), all shown schematically below in Figure 1:



Figure 1. Electrolyzer configurations of interest for application

Materials challenges motivate research in each of these electrolyzer categories, although alkaline electrolyzers are a mature and globally commercialized technology. In an alkaline electrolyzer, a tank of water containing KOH as a base and electrolyte interacts with two electrodes immersed in the tank. Water is continuously added to the tank as hydrogen and oxygen are recovered from the splitting reaction.

For example, minimizing internal resistances, optimization of the membrane-electrode assembly, and selection of separator material all play into the device efficiency.³ With these in mind, research on the catalyst components of electrolyzers is more active and will be considered in this manuscript from a critical materials perspective.

Just as with energy stored in battery electric vehicles, water electrolysis-produced hydrogen is only as green as the electricity used in its generation. Moreover, technology that decreases the amount of electricity needed to split water is beneficial. These concepts are well-explored in life cycle analyses comparing energy-normalized global warming potential of hydrogen production methods by alternative

energy sources.⁵ Energy is a significant factor in particular when assessing SOEs, which can operate at high temperatures (~1000°C).

New directions in hydrogen production

Water electrolysis is an energy-intensive process that benefits from the use of catalysts. Because the canonical hydrogen evolution catalyst is Pt and oxygen evolution catalyst is RuO₂, an important research focus for green hydrogen production has been the development of catalysts that rival scarce metals in performance but with reduced or eliminated metal loading.^{6, 7} The recent materials research landscape in this area can be visualized in many ways; we begin by presenting the most commonly co-occurring concepts found to be important within each respective study in a clustered network diagram (Figure 2).



Figure 2. Top 125 pairs of co-occurring concepts in the green hydrogen production literature from 2011-2021.

From this high-level conceptual analysis, we see that "Hydrogen evolution reaction" and "Water splitting" concepts have approximately equal primacy in the discussion, while "Oxygen evolution reaction" also frequently co-occurs within documents. This finding underscores that studies of overall water splitting must consider both OER and HER to reach a complete understanding.^{8, 9} In addition, "Photocatalysts" are a common research theme from 2011-2021 and have significant overlap with several nanomaterial-related concepts; the fact that "Electrochemical reaction catalysts" co-occur at approximately the same rate as "Photocatalysts" demonstrates how important photocatalysts have become in the field. Finally, the inclusion of a cluster of surface-oriented concepts such as "Surface structure", "Surface area", and "Pore size" shows the relevance of surface phenomena in catalyst design.



Figure 3. Publication trends and distinct substances used for catalysts by year in green hydrogen production research from 2011-2021

Between 2011 and 2021, an over five-fold increase of publication volume is observed in green hydrogen production (Figure 3). This is driven by a concomitant increase in both journal articles and patents. After experiencing rapid growth throughout the decade, the publication volume appears to be leveling off.

Concerning catalytic materials in green hydrogen production journal articles and patents, the scope of several substance classes expanded appreciably in the 2010s and culminated in a peak of substance diversity in 2019. The slight dip in distinct substances from 2019-2021 contrasts with the continued overall increase in publication count (Figure 3). In addition to the general increase in distinct substances, green hydrogen production had a primarily academic research focus in the first half of the decade but then had a gradual increase in the share of patents to reach 15% of the total publication volume in 2021. Based on these observations, green hydrogen production catalysis appears to be reaching maturity as a research field with commercial potential.

Substance information found within publications provides additional insights. While publication volume alone provides a picture of relative research interest over time, it does not detail the diversity of

approaches being taken to a given topic. Analysis of relevant substance classes within this progression over time reveals several research trends. For example, the increase in compounds of classes Alloy and Element show the exploration of chemical space for alternative electrocatalysts to Pt¹⁰ and as components in composite materials as electro- and/or photocatalysts. Coordination compounds, on the other hand, saw an increase in material diversity in the first half of the decade followed by sustained research interest. In this time period, metal-organic framework (MOF)-based and MOF-derived materials saw increased interest in heterogeneous catalysis.¹¹ The application of semiconductor engineering to photocatalysis saw compounds of classes General Inorganic and Oxide increasingly applied to green hydrogen production throughout the decade alongside their use as catalyst supports.^{12, 13} Finally, polymers began to be studied as components in heterojunction catalysts¹⁴, as tunable stand-alone porous photocatalysts (in the case of covalent organic frameworks),¹⁵ and as precursors to engineered carbonaceous catalyst materials.^{16, 17} The top-studied materials from these classes in 2021 are shown below in Table 2 alongside their respective important research focuses.

As detailed in Table 2, control and exploitation of nanoscale morphology is currently a heavy focus of green hydrogen production catalysts. Several factors play into this research focus. First, it is desirable in electrocatalysis to maximize the electrochemically active surface area of the catalyst to increase the number of active sites available, and in photocatalysis, to maximize light absorption. In addition, the dispersal of precious metals onto a nanostructured surface can improve the catalytic properties on an atomic metals basis. Finally, in heterostructured semiconductor photocatalysts as well as Schottky junction materials, managing variables such as exciton separation, carrier diffusion, and mass transfer becomes quite complicated.¹⁸

The relative prevalence of the most common nanomaterial types in GHE research are shown in Table 1, normalized to the number of publications in each respective research area. For green hydrogen production, the "Nanoparticles" concept is the most common, followed by "Nanosheets" and "Nanocomposites". The popularity of nanoparticles is well known, with Pt nanoparticles being considered among the top-performing HER electrocatalysts. Concerning the almost equally popular nanosheets, materials chemistry has been enamored with this 2-dimensional materials for the last 15 years, and a diverse set of products can be convinced to form into atomically thin dimensions to give rise to novel and useful phenomena for catalysis.¹⁹ When combined with other materials (into nanocomposites), high surface area catalysts can be prepared which take advantage of nanoscale effects such as quantum confinement²⁰ and surface plasmon resonance,²¹ as well as interfacial effects including the aforementioned semiconductor heterojunctions and Schottky junctions.²² In addition to the transition metal dichalcogenides²³ (e.g. MOS_2) and graphitic carbon nitride²⁴ (C_3N_4) mentioned in Table 3, nanosheets used for green hydrogen production have thus far included layered double hydroxides,²⁵ graphene,²⁶ MXenes,²⁷ bismuth oxyhalides,²⁸ halide perovskites,²⁹ and 2D MOFs³⁰ and covalent-organic frameworks.³¹ The vast selection of electronic materials available and the toolbox of synthetic methods impart control over particle size, shape, doping and defect levels, crystallinity, and material interfaces, thus motivating the large number of observed studies on nanoscale morphology in green hydrogen production.

We also consider here the relative prevalence of elements in catalysts used for green hydrogen production on a document-level basis, shown below in Figure 4. This provides a rough landscape for assessing the overall elemental distribution of research interest and may point to future resource requirements for adoption on a societal scale. Overall, emphases on carbonaceous materials as well as transition metal oxides and sulfides are evident. There has been strong interest in critical metals including cobalt, nickel, and platinum, with the peak publication volume centered at the expected d⁸ transition metals typical of HER catalysts.

# Publications	Concept	Green Production	Storage	Fuel Cells	Proportion of
14,810	Nanoparticles		•	•	Publications
6,314	Nanosheets	•	•	•	• 0.0016
4,990	Nanocomposites	•	•	•	0.0500
4,505	Nanostructures	•	•	•	0.1000
2,455	Nanorods	•		•	0.1465
2,285	Nanowires	•		•	
1,965	Nanotubes	•	•	•	
1,732	Nanocrystals	•	•	•	
1,731	Nanostructured materials	•	•	•	
1,666	Nanocatalysts	•	•	•	
1,379	Nanofibers	•	•	•	
758	Core-shell nanoparticles	•		•	
652	Nanospheres	•		•	
608	Nanoporous materials	•	•		
584	Nanoclusters				

Table 1.	op nanotechnology-related o	concepts in each area d	of GHE research	from 2011-2021
	1		1	,

H 6,869	Number of Documents 0 17,382													He 0			
Li	Be	Be											C	N	0	F	Ne
152	10	10											11,639	6,996	17,382	725	0
Na	Mg											AI	Si	P	S	CI	Ar
889	537											1,539	1,214	3,621	7,249	1,369	7
К	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
615	464	100	5,227	1,187	686	1,085	4,553	6,251	8,116	3,301	2,685	472	138	74	1,348	438	O
Rb	Sr	Y	Zr	Nb	Mo	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	ا	Xe
41	727	229	798	606	4,383	27	2,231	849	1,524	1,213	2,121	917	877	148	266	224	1
Cs	Ba		Hf	Та	W	Re	Os	lr	Pt	Au	Hg	TI	Pb	Bi	Po	At	Rn
108	284		112	702	1,860	186	95	1,360	5,493	1,436	32	14	217	1,136	0	0	0
Fr 0	Ra 0																
			La 734	Ce 918	Pr 104	Nd 73	Pm 4	Sm 74	Eu 40	Gd 85	Тb 25	Dy 21	Ho 12	Er 46	Tm 16	Yb 37	Lu 10
			Ac 0	Th 6	Pa 1	U 15	Np 1	Pu 1	Am 0	Cm 0	Bk 0	Cf 0	Es 0	Fm 0	Md 0	No 0	Lr 0

Figure 4. Occurrence of elements in materials used as green hydrogen production catalysts by number of documents from 2011-2021

Catalyst Substance	Substance	REG #	2021	Feature(s)	Exemplary Publications
Class			Pubs		
Oxide	RuO ₂	9002-89-5	185	Standard for comparison for OER, but also	32-34
				frequently used in nanocomposite	
				electrocatalysts	
Oxide	TiO ₂	13463-67-7	421	Catalyst support; frequently doped and/or	35-37
				heterostructured nanocomposites for both	
				photocatalysis and electrocatalysis	
General Inorganic	C ₃ N ₄	143334-20-7	477	Facile synthesis into nanostructures, amenable to	38-40
				vacancy engineering for photocatalysis	
General Inorganic	MoS ₂	1317-33-5	308	Exfoliatable semiconductor nanosheets for	41-43
				photocatalysis	
Element	C	7440-44-0	917	Prepared via various sources to control	44-46
				morphology and doping level of	
				(photo)electrocatalyst component	15 17 10
Element	Pt	7440-06-4	899	Nanostructured or "single-atom" catalysts for	45, 47, 48
				decreased Pt loading in HER	16 10 50
Element	Ni	7440-02-0	681	Ni foam as electrocatalyst component; in-situ	46, 49, 50
				transformations into active nanocatalyst	
				components; single-atom catalyst studies	51_52
Coord. Compd.	UIO-66(NH ₂)	1260119-00-3	11	Visible light-responsive porous photocatalyst	21-22
	715 67	46204 07 4	27	component	54-56
Coord. Compd.	ZIF-67	46201-07-4	27	Doped, surface-engineered, and/or calcined to	54 50
All		11110 22 6	<u> </u>	produce novel Co-based (photo)electrocatalysts	57-59
Alloy	Fe-NI	11148-32-6	64	Electrodepositable nanocomponent in overall	
		11101 12 0	42	Nanagementation algorithm and algorithm with a their term	60-62
Alloy	CO-INI	11101-13-0	43	manocomposite electrocatalysis with other top	
			45		63-65
Polymer	Polyaniline	25233-30-1	15	Conductive polymers in nanocomposite	05 05
Polymer	Polypyrrole	30604-81-0	13	(pnoto)electrocatalysts	64, 66, 67

Table 2. Key substances in green hydrogen production catalyst research

Hydrogen storage

Renewable clean hydrogen which does not contain any environmental issues is a desirable energy storage carrier and an efficient fuel; it has the highest energy per mass (142kJg⁻¹) of any fuel.⁶⁸ For multiple decades, the major hurdle to development of the GHE has been that hydrogen is difficult to store. Its low ambient temperature density results in a low energy per unit volume, therefore requiring the development of advanced, safe, and efficient storage methods with potential for high energy density.⁶⁹ Hydrogen can be stored in several ways including physical-based and chemical-based storage. Conventional hydrogen storage methods include compression, liquefaction, and cryo-compression.

Compressed hydrogen. The compression of hydrogen into tanks provides the benefits of quick charge and discharge times. However, the volumetric density of hydrogen is much lower than that of other energy sources, for example four times lower than natural gas. Therefore, hydrogen needs to be compressed to very high pressures (700-800 bar or higher) to achieve reasonable volumetric density.⁷⁰ Thick walled steel tanks with steel or aluminum (Type-3) or polymer linings (Type-4) therefore become necessary. The tanks are accompanied by inherent safety risks including explosion and fire, and troubled by the permeability of hydrogen and embrittlement of the tank walls. A recent study of 350-bar and 700-bar polymer-lined H₂ storage tanks has shown that the carbon fiber-reinforced composite needed to provide the structural strength for these fuel tanks. For 700-bar H₂ storage tanks, the high-density polyethylene (HDPE) liner fully wrapped with carbon fiber composites is required.^{71, 72}

Liquified hydrogen. Liquefaction is an option if an application requires hydrogen volume to be reduced further than compression can achieve.⁷⁰ MHI Group and the space industry have used liquefied hydrogen to fuel rockets for many years, however liquid hydrogen storage is energy intensive, technically complex, and very costly. Hydrogen must be cooled to -253°C (near to absolute zero) and stored in insulated tanks to maintain this extremely low temperature. To minimize the losses due to vaporization, transfer of liquid hydrogen should be performed in a vacuum-insulated system. In addition, this transfer should be conducted in a closed system with a proper safety relief device to avoid a flammable atmosphere or explosive mixture of air and liquid hydrogen.⁷³

Cryo-Compressed hydrogen. The two techniques – compression and liquefaction – can also be combined. A volumetric density of 70.8 kg/m3 can be achieved using 15.2 kWh/kg of energy as work through a process of compression and cooling in cryogenic tanks while the gravimetric density is influenced by the tank size. In Type 3 (metal-lined) cryogenic tanks, a 2-mm stainless steel liner meets the 15,000-fatigue cycle life requirement for storage pressures up to 700 bar.⁷⁴

Because these physical methods of H_2 storage do not typically meet applicable safety and/or density requirements, especially for transport applications, chemical hydrogen storage methods have received the widespread attention of scientists.



Figure 5. Physical vs. chemical hydrogen storage.

Hydrogen storage materials can be divided into two categories based on the relative strength of the material interaction with hydrogen: "physisorption materials" and "chemisorption materials." In physisorption materials, H₂ molecules are adsorbed via a weak van der Waals interaction on the surface of the pores. The physisorption process is reversible since the interaction energy is incredibly low. A dominant material in this class are carbon sorbents where physisorption is proportional to their specific surface area. ^{75, 76} Storage pressures for physisorption based sorbent systems can be much lower than physical storage methods without a significant reduction in capacity. Storage temperatures can also be higher, thus reducing the cost for insulation and the energy consumption for cooling. Lastly, sorption is a completely reversible process and does not require off-board regeneration as is needed for chemical storage options. The main challenge for physisorption materials is low binding energy for H₂. An approach is to use cryogenic temperatures to enhance gravimetric and volumetric storage capacities of physisorbents, although this comes at the cost of some of the benefits.⁷¹

In chemisorption-based materials, hydrogen chemically interacts with the storage medium. Chemisorption process may not be fully reversible due to relatively high activation energy in the adsorption and desorption process. On-board hydride materials belong to this class.^{77, 78}



Figure 6. Publication trends and distinct substances used for material by year in hydrogen storage research from 2011-2021

In contrast to green hydrogen production, the number of annual publications on H₂ storage is steady at about 1500 with yearly fluctuations between 2011-2021 (Figure 6). However, this total hides the declining number of journal publications. The surge of the 2012-2013 publications coincides in time with the first commercially produced hydrogen fuel cell vehicle, Hyundai ix35 FCEV, introduced by Hyundai in 2013.⁷⁹ A decrease in journal publications occurred up to 2017. Since, journal increases occur through 2021. At the same time, the number of patents shows more steady growth with fewer fluctuations. The growth of patents may indicate a special interest of manufacturers such as Toyota, Honda, Hyundai, and Panasonic in developing new on-board H₂ storage technologies (see Table 6).

The progress in the H₂ storage research is illustrated by the concept cluster map shown in Fig.7. "Hydrogen storage materials" is the key concept of the map. The concept is commonly used together with "Dehydrogenation", "Hydrogenation", "Dehydrogenation catalysts", and "Hydrides" indicating H₂ storage and release via chemisorption. Also "Hydrogen storage materials" frequently co-occurs with "Absorption", "Desorption", "Metal-organic frameworks", and "Carbon nanotubes" which indicate physisorption and common physisorbents. "Microstructure", "Ball Milling", "Nanoparticles",

and "Nanostructures" demonstrate current trends in material modification. They often co-occur with "Activation energy", "Surface area", "Pore size", "Pore size distribution", and "Binding energy". Finally, "Fuel cells" and "Secondary batteries" show the way for on-board H₂ storage.



Figure 7. Top 125 pairs of co-occurring concepts in the hydrogen storage literature from 2011-2021

The most frequently reported substance classes for hydrogen storage selected from 2011-2021 publications are summarized in Figure 6. The development of hydrogen storage materials can be divided in two periods, steady 2011-2015 with a slight increase in 2013, ups and downs during 2016-2021 with a visible decrease in 2020. Three main substance classes such as alloys, general inorganic, and small organic/inorganic molecules continued to be a focus of hydrogen storage research. During the 2011-2015 period, publications on alloys dominated since alloys were used for both physical storage (tanks/pipelines materials) and chemical storage (sorbents). It is interesting that research on polymers also increased at

that time due to polymer reinforcement of metal storage tanks. New interest in polymers appeared in 2021 when porous polymers able reversibly stored and released hydrogen were reported. Coordination compounds, which were of great interest in 2011-2015, show a decline in 2016-2021. There is continuous attention to hydrides with a surge in 2021. Elements and oxides are always in the spotlight. Those substances represent carbonaceous sorbents, dehydrogenation catalyst and modifiers.

Elements. The elements class includes carbon-based materials such as activated carbon, carbon nanotubes, graphitic nanofibers, graphene, graphite and fullerene. Carbonaceous sorbents are promising materials for hydrogen storage due to their low densities, good chemical stability, high surface area, and porosity.⁸⁰ Carbonaceous sorbents research focuses on increasing effective adsorption temperature through increase of the hydrogen binding energies and improving volumetric and gravimetric storage capacities through optimizing the material's pore size, increasing pore volume and surface area, and investigating effects of material densification. Hydrogen can be physically adsorbed on activated carbon and be "packed" on the surface and inside the carbon structure more densely than if it has just been compressed. The best results were achieved with carbon nanotubes.⁸¹ Carbon nanotubes, when decorated with metal or metal oxide nanoparticles, show a significantly improved hydrogen storage capacity. For instance, multi-walled carbon nanotubes (MWCNTs) decorated by Dy₃Fe₅O₁₂ nanoparticles can store H_2 at temperatures as low as 196.15°C and pressures as low as 6 MPa, providing a gravimetric density of 10.8 wt% and volumetric density of 41 kg/m^{3.82} Fullerenes are potential hydrogen storage material based on the ability to react with hydrogen via hydrogenation of carbon-carbon double bonds. The theory predicts that a maximum of 60 hydrogen atoms can be attached to both the inside and outside of the fullerene spherical surface. A stable $C_{60}H_{60}$ isomer can be formed with the theoretical hydrogen content of \sim 7.7 wt %. It seems that the fullerene hydride reaction is reversible at high temperatures. The 100% conversion of C₆₀H₆₀ indicates that 30 moles of H₂ gas will be released from each mole of fullerene hydride compound. However, this reaction is not possible because it requires a high temperature, about 823–873 K.⁸³ A new trend in carbonaceous sorbents is preparing porous carbon materials from biomass pyrolysis. It has been shown that the pyrolysis temperature, pyrolysis heating rate and carbon-containing precursor strongly affect the yield and structure of the resulting porous carbons.⁸⁴ Another attempt is to fabricate highly porous carbon sorbents by carbonizing highly crystalline metal-organic frameworks (MOFs) without any carbon precursors.⁸⁵

Other widely used **elements** are transition metals which can be applied alone or together with noble metals (Pt, Pd) as dehydrogenation catalysts for metal hydrides and liquid organic hydrogen carriers. It was shown that using nickel nanoparticles as ammonia borane dehydrogenation catalysts is a promising step toward a feasible hydrogen storage medium for fuel cells. ⁸⁶

Coordination compounds. In coordination compounds such as metal–organic frameworks, H₂ is physiosorbed on the surface of the pores. In general, the interactions of the type "H₂-MOF" are very weak. Therefore, MOFs' high storage capacities can be achieved at liquid nitrogen temperature and high pressures. Still, MOFs have been extensively studied as promising hydrogen storage materials. Various metal ions such as Zn(II), Cu(II), Mn(II), Cr(III), and La(III), and ligands such as carboxylates, imidazolates, triazolates, tetrazolates have been employed for the construction of the MOFs. Promising H₂ storage data were reported for MOF-5 (4.5 wt % at 77 K and 1 atm), prepared from benzene-1,4-dicarboxylate (BDC) and Zn(II) salt.⁸⁵ Significant research is focused on densified hybrid MOF@CNT and MOFs impregnated with fullerene composites for hydrogen storage. Various MOFs such as MOF-5, MIL-101, Zr-MOFs and HKUST-171 have been compacted and their hydrogen adsorption capacities evaluated.⁸⁷ After compaction

at ~700 MPa, the densified UiO-66 achieved a total H_2 uptake of 5.1 wt% at 100 bar and 77 K compared to 5.0 wt% for the UiO-66 powder.⁸⁸

Metal Hydrides. Light and complex metal hydrides are prominent hydrogen storage materials as they require comparatively feasible working temperatures and have good hydrogen storage capacity. To form light metal hydrides, hydrogen interacts with metals through different bonds. Ionic or covalent hydrides, such as LiH and MgH₂ are quite suitable for hydrogen storage due to their high hydrogen storage capacity but require a very high operating temperature. Research on hydrides focuses on improving the volumetric and gravimetric capacities, hydrogen adsorption/desorption kinetics, cycle life, and reaction thermodynamics of potential material candidates. Pristine MgH₂ can store ~7.6 wt.% hydrogen.⁸⁹ However, both MgH₂ hydrogenation and dehydrogenation reactions are very slow. Magnesium hydride forms ternary and quaternary hydride structures by reacting with various transition metals (Fe, Co, Ni, etc.) and thus improved kinetics. Moreover, the nanoscale version of these transition metal particles offers an additional hydrogen sorption mechanism via its active surface sites.⁹⁰ Noble metals (Pt, Ag, Au, Pd, and Ru) are beneficial to improve dynamical stability and dehydrogenation properties of LiH.⁹¹

Doping nanoparticles of Fe and Ti with a few mol% of carbon nanotubes improves the sorption behavior of MgH₂.⁹² Ball milling is widely used for modification of the hydrogenation/dehydrogenation process in magnesium-based hydrogen storage materials. The phenomenon of mechanical milling helps to pulverize the particles of MgH₂ into micro- or nano-crystalline phases and thus leads to lowering the activation energy of desorption.⁹³ The activation energies of the H₂ sorption and dehydrogenation temperatures for the bulk MgH₂, mechanically milled MgH₂ and nanocatalyst-doped MgH₂, are shown.

(a) 162 kJ/mol H₂; 415 C (b) 144 kJ/mol H₂; 340 C (c) 71 kJ/mol H₂; 245 C



The activation energies of H₂ sorption and dehydrogenation temperatures for MgH₂ (a) bulk, (b) ball-milled nanocrystalline, (c) nanocrystalline, transition metal nanoparticle-doped

Figure 8. Activation energies of H₂ sorption and dehydrogenation temperatures for MgH₂ (a) bulk, (b) ball-milled nanocrystalline, and (c) nanocrystalline, transition metal nanoparticle-doped

Activation barriers are drastically lowered by nanocatalyst doping which suggests that the collision frequency between the H_2 molecules and transition metal nanoparticles increases with decreasing crystallite size (Figure 8). In addition, mechanochemical milling of MgH₂ introduces defects and particle size reduction. In an example, commercial MgH₂ exhibits weight loss due to hydrogen decomposition at 415°C, the ball milled nanocrystalline MgH₂ shows endothermic hydrogen decomposition at 340°C.⁹⁴

Both alkali metals and alkaline-earth metals borohydrides have a significant hydrogen storage capacity. For example, LiBH₄ demonstrates the highest gravimetric (18.5 wt. % H₂) and volumetric (121 kg H₂ m⁻³) hydrogen storage capacity. However, pristine LiBH₄ undergoes different changes with the effect of temperature. It was shown that hexagonal boron nitride (h-BN) additive greatly improves the dehydrogenation of LiBH₄.⁹⁵

Sodium borohydride (NaBH₄) is an excellent hydrogen storage material, but its application is restricted by irreversibility of hydrolysis and high cost of regeneration. To regenerate NaBH₄ with high yield and low costs, the hydrolytic product NaBO₂ reacts with CO₂, forming Na₂B₄O₇·10H₂O and Na₂CO₃, both of which are ball-milled with Mg under ambient conditions to form NaBH₄ in high yield (close to 80%). This method is expected to effectively close the loop of NaBH₄ regeneration and hydrolysis, enabling a wide deployment of NaBH₄ for hydrogen storage.⁹⁶

Hydrogen storage alloys. Divided into magnesium, rare earth and titanium hydrides, hydrogen storage alloys use metals or alloys to store hydrogen by reacting with it to form hydrides, a much safer way of storing hydrogen than using conventional physical storage methods.⁹⁷The stored hydrogen can then be released for use when the hydrides are heated. Some examples of these storage alloy hydrides are LaNi₅, Mg₂Ni, FeTi.⁹⁸ Magnesium-based alloy hydrides have the most promising hydrogen storage properties. Intermetallic compounds such as TiFe and LaNi₅ have good hydrogen storage reversibility. Recently, a new class of alloys, TiZrNbFeNi high-entropy alloys (HEA) were investigated for H₂ storage.⁹⁹

¹⁰⁰LaNi₅-based alloys have been used in practical applications because of their stable reversible hydrogen absorption and desorption reactions under moderate conditions (i.e., room temperature and hydrogen gas pressure less than 1 MPa).

Recent studies of rare-earth/magnesium/nickel alloys showed that REMgNi₄-based alloys, $RE_{(2-x)}Mg_xNi_4$ (RE: rare-earth metals; 0 < x < 2), with a AuBe₅-type crystal structure, exhibit reversible hydrogen absorption and desorption reactions, which are known as hydrogen storage.¹⁰¹

Hydrogen storage alloys are designed to absorb and release hydrogen without compromising their own structure. Hydrogen storage alloys represent an excellent solution for fuel cell storage.

Small inorganic/organic molecules

Ammonia is a kind of material that offers a path to turning hydrogen into a liquid fuel. Ammonia's energy density by volume is nearly double that of liquefied hydrogen, making it far easier to store and transport.¹⁰²

Formic acid with its high volumetric concentration of H_2 (53 g L⁻¹) is a potential liquid organic hydrogen carrier (LOHC). ¹⁰³ Formic acid is a renewable hydrogen carier, it can be produced from carbon dioxide via direct catalytic hydrogenation. The most promising catalysts for FA selective dehydrogenation are based on nobel metals such as ruthenium and iridium.¹⁰⁴

Ammonia borane (AB), BH₃NH₃, is a stable solid at room temperature, melts at a temperature 110 - 114 C. It is a promising chemical hydrogen storage material for use in fuel cells for automotive industry because of its high H₂ intrinsic material capacity and the exothermicity of the dehydrogenation reaction. Storing H_2 on-board in the form of solid AB, however, has a technical barrier that is typical to any solid fuel that needs to be regenerated.⁷⁷

N-ethylcarbazole are liquid organic H₂ storage materials which require dehydrogenation catalysts. At the present time, the bottleneck of N-ethylcarbazole dehydrogenation technology is how to develop a catalyst with higher conversion, better selectivity, and stability. Among those catalysts are the binuclear ruthenium and rhodium catalysts,¹⁰⁵ noble metal catalysts,¹⁰⁶ ruthenium-nickel TiO2 supported catalysts¹⁰⁷. Recent trend, using nickel or other non-precious metals in bimetallic composites with Pd or Ru, is reported.¹⁰⁸

General inorganic: Carbides, Nitrides, Phosphides

Two-dimensional MXenes, such as Ti_3C_2 and other transitional metal carbides (Ni₃C, Mo₂C, Cr₃C₂ and NbC) were used to enhance the H₂ storage behavior of MgH₂.¹⁰⁹ The carbides were introduced into MgH₂ by mechanical ball milling without changing their phase. Ti_3C_2 shows the best catalytic effect on dehydrogenation kinetic properties of MgH₂, followed by Ni₃C, NbC, Mo₂C and Cr₃C₂.¹⁰⁹ Ti₃C₂ and Ni/Ti₃C₂ serve as dehydrogenation catalysts for metal hydrides.¹¹⁰ A recent study on hydrogen physisorption on AM-2DSnC (AM = Li, Na, and K) complexes reported that the K-2D SnC monolayer has the highest hydrogen-storage capacity with one potassium atom adsorbing up to 6 hydrogen molecules, followed by Na-2D SnC with 5 hydrogen molecules and Li-2D SnC with 3 hydrogen molecules; demonstrating the possibility of these complexes overcoming the US-DOE recommended target of 5.5 wt% for onboard automotive systems.¹¹¹

Bilayer hexagonal boron nitride (h-BN) shows a trend of decreasing binding energies and desorption temperatures that is useful for potential H2 storage. The theoretically calculated overall storage capacity of the bilayer is 6.7 wt % with Eads of 0.223 eV/H2.¹¹² TiO₂-coated boron nitride nanofibers showed improved morphology, surface area and hydrogen storage.¹¹³

In a recent study, alkali-earth metal (Be, Mg and Ca) encapsulated $Al_{12}N_{12}$ nanoclusters have been considered for hydrogen storage via adsorption and demonstrated: an increase in adsorption energy when compared to untreated $Al_{12}N_{12}$ and H_{2} - $Al_{12}N_{12}$ and a decrease in the HOMO- LUMO energy gap.¹¹⁴ Another example of nanomaterials for hydrogen storage is g-C₃N, a graphitic carbon nitride whose structure permits the storage of a significant amount of hydrogen.¹¹⁵.

A study on the thermodynamic stabilities of Li- and Na-decorated 2D boron phosphide (BP) systems found that the dispersed Li and Na atoms on the monolayer surface significantly increase both the H_2 binding energies and the H_2 storage capacities.¹¹⁶ An improvement in hydrogen-storage capacity due to the process of intracell Kubas-enhanced hydrogen adsorption in Co₂P nanoparticles obtained by ball-milling has also been discovered.¹¹⁷

Metal oxides are promising candidates for long-term hydrogen storage. They participate in a reversible redox cycle with H_2 as a reducing agent and H_2O as an oxidizing agent, where the oxidation reaction results in a high-purity hydrogen production (Eq 4).¹¹⁸

 $Me_xO_y + zH_2 \iff Me_xO_{y-z} + z H_2O(g)$ (4)

The best H₂ storage oxides such as Fe₃O₄, GeO₂, MoO₂, SnO₂, ZnO, and WO₃ are supported with Al₂O₃, TiO₂, Cr₂O₃, MnO, and MgO. ¹¹⁸ Ti₄M₂O_y mixed oxides (Ti₄Fe₂O, Ti₄Ni₂O) have demonstrated good hydrogen storage properties at room-temperature.¹¹⁹ Also hybrid ceramics, such as NiCo₂O₄/TiO₂, are an efficient and novel hydrogen storage material.¹²⁰

Metal oxides, especially transition metal oxides, are universally used as hydrogenation and/or dehydrogenation catalysts. For example, TiO_2 supported on multi-walled carbon nanotubes improves both hydrogenation and dehydrogenation of a Mg–Ni alloy (absorbs 5.60 wt% H₂ at 373 K and releases 6.08 wt% H₂ at 553 K).¹²¹ The addition of TiO_2 to a Mg₈₀Ni₁₀La₇Ce₃ alloy significantly improves its dehydrogenation.¹²²

Metal oxide nanoparticles produced by ball milling have a superior effect on the hydrogenation and/or dehydrogenation of MgH₂. Thus, Ni@TiO₂ core-shell nanoparticles significantly improve hydrogen desorption from MgH₂.¹²³ Nb₂O₅ milled with MgH₂ catalyzes the hydrogenation of MgH₂.¹⁴⁷ The catalytic effect of a VNbO₅ ternary oxide on MgH₂ dehydrogenation is also remarkable.¹²⁴

Recently solid oxides with ABO₃ perovskite structures became key substances for enhancing hydrogen storage properties.¹²⁶ It was shown that porous NiTiO₃ and CoTiO₃ nanorods can decrease the dehydrogenation temperature of MgH2 and provide faster hydrogen desorption (Tdes = 261.5°C and Tdes = 298°C for NiTiO₃ and CoTiO₃, respectively).¹²⁷ Other mixed oxides such as NiMoO₄ and CoMoO₄ nanorods similarly enhance the non-isothermal and the isothermal desorption performance of magnesium hydride.¹²⁸

Polymers. Porous organic polymers, hyper-crosslinked polymers, and polymers with intrinsic microporosity reversibly store and release hydrogen through hydrogen physisorption on their highly porous structures and in some cases use combinations of physisorption and chemisorption to store H₂. The introduction of functional groups in polymer networks can also provide an enhancement of the hydrogen uptake. Porous polymers containing highly electron-deficient carborane-components were successfully used as adsorbents for hydrogen.¹²⁹ Ketone and N-heterocycle containing polymers fixed and stored hydrogen at atmospheric pressure through the formation of chemical bonds to form the corresponding alcohol and hydrogenated N-heterocycle polymers, respectively. The hydrogenated polymers released hydrogen in the presence of catalysts at mild conditions. Quinaldine-substituted polyacrylic acid and its hydrogenated 1,2,3,4-tetrahydroquinaldine derivative reversibly release hydrogen by heating (80°C) in the presence of an aqueous iridium complex catalyst.¹³⁰ Conducting polymers consisting of a polyaniline matrix, which can be functionalized by catalytic doping or by the introduction of chemical groups into a polymer molecule, is a potential promise for hydrogen storage. It was reported that polyaniline could store up to 6–8 wt% of hydrogen.¹³¹ Porous polyaniline P-PANI facilitates hydrogen diffusion and reaction kinetics of the hydrogen storage alloys.^{132, 133} Polyacrylamide blending with ammonia borane enables the dehydrogenation of the polymeric composite to occur at lower temperature with enhanced hydrogen purity.¹³⁴

Although hydrogen storage by means of physisorption has some limitations, polymers seem very promising materials, due to their high potential of structural and functional tuning, as well as good thermal and chemical stability.

Nanomaterials for H₂ storage.

Nanomaterials for hydrogen storage have attracted great interest in recent years. As shown in Table 1, "Nanoparticles" is the most popular concept followed by "Nanocomposites" and "Nanostructures". Metal hydride nanoparticles, polymer and metal-organic frameworks nanocomposites are advantageous for storing substantial amounts of hydrogen. ¹³⁵ Carbon "nanotubes" are efficient H₂ storage materials. "Nanocrystals" and "Nanocatalysts" concepts reflect the synergistic effects of nanocrystallinity and nanocatalyst doping on improving the thermodynamics and hydrogen reaction kinetics in metal hydrides. The development of new "nanocatalysts" maximizes hydrogenation/dehydrogenation efficiency while minimizing the use of precious noble metals. ¹³⁶ "Nanoporous materials" confirms that nanoconfinement of hydrides and borohydrides in carbon nanopores significantly improves their hydrogen sorption properties.¹³⁷ The nano-concepts extracted from 2011-2021 publications confirm the widespread use of nanostructured materials in hydrogen storage.

The most cited key substances for hydrogen storage are summarized in Table 3.

Substance Class	Substance	CAS REG #	Storage Type/Feature	Publications/ Citations 2021	Exemplary Publication
All	LaNi₅	12196-72-4	Chemical/Hydrogenation	13/7	138-142
Alloys	Mg ₂ Ni	12057-65-7	Chemical/Hydrogenation	11/20	143, 144
	FeTi	1223-04-0	Chemical/Hydrogenation	13/44	145-150
	Stainless steel	12597-68-1	Physical/Tank material	16/8	74, 151
	MgH ₂	7693-27-8	Chemical/Dehydrogenati	86/195	94, 152-155
Hydrides	LiH	7580-67-8	Chemical/Dehydrogenati on	17/32	91, 156
	NaBH ₄	16940-66-2	Chemical/Dehydrogenati on	23/20	157-162
	AlH ₃	7784-21-6	Chemical/Dehydrogenati on	12/45	163, 164
	LiAlH ₄	16853-85-3	Chemical/Dehydrogenati on	14/14	165, 166
	Mg(BH ₄) ₂	16903-37-0	Chemical/Dehydrogenati on	13/25	167, 168
Elements	Carbon	7440-44-0	Physical/Sorbent	100/134	169-172
	Graphene	1034343-98-0	Physical/Sorbent	60/76	173-177
	Graphite	7782-42-5	Physical/Sorbent	16/7	178, 179
	Nickel	7440-02-0	Chemical/Catalyst	54/55	180-182
Small organics	9-ethylcarbazole	86-28-2	Chemical/Dehydrogenati on	16/10	183, 184
	Methylcyclohexane	108-87-2	Chemical/Dehydrogenati on	15/14	185-188

Table 3. Key substances in hydrogen storage research

	Ammonia	7664-41-7	Chemical/Dehydrogenati	37/34	189-193
			on		
	Ammonia borane	13774-81-7	Chemical/Dehydrogenati	13/73	194-198
			on		
Small inorganics	UiO-66(Zr)	1072413-89-8	Chemical/Sorbent	4/4	199-201
	Triaqua[µ-[1,3,5-	222404-02-6	Chemical/Sorbent	4/4	202-204
	benzene				
	tricarboxylato(3-)				
	кО1:кО′1]][µ3-[1,3,5-				
	benzenetricarboxylat				
	o(3-				
)kO1:kO3:kO'1]]trico				
	pper				
	Zinc Tris[µ-[1,4	255367-66-9	Chemical/Sorbent	5/9	204-206
Coordination	benzenedicarboxylat				
compounds	o(2-)-kO1:kO'1]]-m4-				
	oxotetra				
Oxides	MgO	1309-48-4	Chemical/Catalyst	14/44	207, 208
	Nb ₂ O ₅	1313-96-8	Chemical/Catalyst	5/7	209-211
Polymers	Polyethylene glycol	25322-68-3	Chemical/Dispersant for	14/1	212, 213
			Dehydrogenation		
			catalysts		
	Nylon 6	25038-54-4	Physical/Storage tank	10/1	214, 215
			reinforcement		



Figure 9. Occurrence of elements in materials used for hydrogen storage research by number of documents from 2011-2021.

As discussed above, carbon is the most prevalent element of H₂ storage (Fig.9). Carbon is a major part of various carbonaceous sorbents such as activated carbon, graphene, MOFs, as well as liquid organic hydrogen carriers and polymers. Magnesium is another key element found in hydrides, borohydrides and hydrogen storage alloys (MgH₂, Mg (BH₄)₂, Mg ₂Ni). Transition metals, namely Ni, La, Ti, and Fe, are important components of H₂ storage alloys (LaNi₅, FeTi) and metal oxides (Ti₄Fe₂O, Ti₄Ni₂O). In addition, the growing interest in transition metal nanoparticles as dehydrogenation catalysts is since they can successfully replace expensive noble metals.

Utilization of Hydrogen to Generate Energy

With hydrogen now generated and stored for future use, we now concentrate on methods of converting the hydrogen into energy. The primary method of using hydrogen in the GHE is a fuel cell, an electrochemical device that converts chemical energy into electrical energy. Though similar to batteries because fuel cells and batteries both convert energy released during a chemical reaction into electric energy, fuel (H₂) can be continuously fed into fuel cells for continuous electrical power output. ²¹⁶ They are compatible with renewable sources and offer quiet operation due to their static nature. A diverse range of applications have already been commercialized, for example: in the transportation sector, in stationary power generation for residential, commercial, and industrial applications, and in the portable sector for military equipment and even toys²¹⁷.

A fuel cell that uses hydrogen fuel works through a process that is the reverse of the before mentioned water electrolysis. Instead of using water and electricity to produce hydrogen and oxygen, fuel cells use hydrogen fuel and oxygen from the air to produce water, usually as steam. In general, a typical fuel cell consists of a thin electrolyte material, typically a semi-permeable membrane, in between two porous electrodes, the cathode and anode.²¹⁸ Molecular hydrogen is delivered to the anode via a gas flow, where the anode catalyst oxidizes the hydrogen, producing hydrogen cations and electrons. Hydrogen cations pass through the electrolyte/membrane from the anode to the cathode. The electrons in the system cannot transfer from the anode to the cathode through the layers of the electrolyte, but only through an external electrical circuit.²¹⁶ It is this movement of electrons that produces the electric current. At the cathode, molecular oxygen combines with the hydrogen protons and electrons to form water. Figure 10 shows this process in a single unit cell.



Figure 10. General structure and operation of a hydrogen fuel cell.

Hydrogen fuel cells work on the principle of three partial reactions:

Anode:	$2H_2 \rightarrow 4H^+ + 4e^-$	(5)
Cathode:	$O_2 + 4e^- \rightarrow 20^{-2}$	(6)
	$20^{-2} + 4H^+ \rightarrow 2H_2O$	(7)
Overall equation:	$2H_2 + O_2 \rightarrow 2H_2O$	(8)

Fuel cells in general can be categorized by various criteria such as type of fuel or operating temperature, but since this paper will be focused on hydrogen fuel cells, we will classify them according to the type of electrolyte they employ. Though other types of hydrogen fuel cells exist, the four types of fuel cells that tend to use hydrogen as their primary source of fuel (even if it is extracted from natural gas using a reformer) are: Alkaline fuel cells (AFC), Proton exchange or polymer electrolyte membrane fuel cells (PEMFC), Phosphoric acid fuel cells and Solid oxide fuel cells (SOFCs).

Alkaline fuel cells (AFC) were first widely used in the U.S. space program to produce electrical energy and water on-board a spacecraft, specifically in the Apollo missions to the Moon and later in the space shuttle program, specifically in the orbiter.^{219, 220} AFC have also been used on vehicles like forklifts, as stationary

power applications, backup power and in military applications.^{220, 221} The electrolyte is a concentrated alkaline solution, usually potassium hydroxide due to its high conductivity, and operates on average bellow 100°C, though temperatures can range from below zero to 230°C.^{217, 220, 221} Anion exchange membranes have also been used as the electrolyte.²²⁰ A wide range of electrocatalysts can be used at the anode and cathode due to the flexibility of the AFC design, although nickel, silver and platinum tend to be the most commonly used ²¹⁹. AFCs have a slightly different operating principle than other fuel cells because the electrolyte/membrane conducts hydroxyl anions (OH⁻) instead of H⁺ cations²²⁰. The reactions that take place at each electrode are the following:

Anode:	$2H_2 + 4OH^- \rightarrow 4H_2O + 4e^-$	(9)
Cathode:	$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$	(10)
Overall equation:	$2H_2 + O_2 \rightarrow 2H_2O$	(11)

The biggest disadvantage of AFCs is their high sensitivity to contaminants, especially CO_2 which reacts with KOH to form K_2CO_3 , degrading the cell performance and durability. This poisoning has required pure hydrogen and oxygen to be used instead of air, for the CO_2 to be removed via absorbers (soda lime, molecular sieves) or by electrolyte recirculation where the electrolyte is passed through a cleaning system to remove carbonates.²²² Using an alkaline membrane as a portion of the electrolyte helps, allowing a lower susceptibility to CO_2 poisoning. However, carbon dioxide still affects the performance of the Alkaline Membrane Fuel Cells (AMFC) ²²³.

Proton exchange membrane fuel cells (PEMFC) were invented by General Electric for use in NASA's 'Gemini' manned space vehicles . ²²⁴ Also known as solid polymer electrolyte membrane fuel cells, They use a solid, acidic polymer membrane that conducts hydrogen cations through its structure when saturated with water. Most commercial cells use a perfluorosulfonic acid ionomer membrane from the Nafion family developed by DuPont.²²⁵ A primary advantage of PEM cells is low weight in comparison to liquid electrolytes, making it the main fuel cell candidates to power electric vehicles as well as more portable power applications, although stationary applications are also possible.^{217, 223-225}

The disadvantage is that the membrane must be hydrated to conduct protons, which means the membrane must be kept at around 80°C below the boiling point of water (though high temperature versions above 200°C have been studied) and water management in general can be an issue ^{217, 224, 225}. PEM fuel cells also require very pure hydrogen with minimal or no CO, which poisons the expensive platinum catalysts at low temperatures, but this becomes mostly a non-issue if the hydrogen is produced via greener methods like water electrolysis instead of steam reforming. PEMFC fuel cells have the same operating principles shown in Figure 10 and Eq. 5-8.

Phosphoric acid fuel cells (PAFC) use phosphoric acid (H₃PO₄) in silicon carbide as the electrolyte. This acid is a solid at room temperature but melts at 42°C and is stable at 200°C, allowing this cell to operate at higher temperatures and reducing the sensitivity to carbon monoxide poisoning ²²⁶. The advantage of this high operational temperature is that these fuel cells can not only be used in stationary power applications, but the waste heat from operation can be captured and used for space heating and hot water ²²⁶. The disadvantages of the cell are that it must be heated first for it to be able to operate (long start up), it needs pure platinum as the catalyst for the cathode and a platinum-ruthenium alloy as catalyst for the anode, and it is very sensitive to sulfur contaminants.^{221, 226} Its operating principle are the same as PEMFC.

Solid oxide fuel cells (SOFCs) use an insulating ceramic solid oxide, most commonly ZrO_2 doped with Y_2O_3 , as the electrolyte that conducts oxygen ions.²²⁷ Due to the solid structure of the electrolyte, it has a very high operating temperature between 600°C to 1,000°C to achieve sufficient ionic conductivity.²²⁸ The high

temperature of operation is a disadvantage due to slow device startup and thermal shielding requirements, but it also removes the need for precious-metal catalyst, thereby reducing cost.²²³ SOFCs are also the most sulfur-resistant fuel cell type and are not poisoned by carbon monoxide.²²¹ High operating temperatures also cause durability issues and strict material requirements. Therefore, the development of high temperature stable device materials or lowering the temperature of the cells to more intermediate levels are the main challenges facing this technology.²²⁹ These types of fuel cells are mostly used for stationary applications (auxiliary power, electric utility, distributed generation) and are highly used for production of electrical and useful thermal energy known as combined heat and power (CHP).^{221, 227}



Figure 11. Publication trends and distinct substances used by year in hydrogen fuel cell research from 2011-2021

In the area of hydrogen fuel cells, publication volume was flat with an almost equal distribution of documents between patents and journals up to 2018 (Figure 11). It is in 2018 that one begins to see a rise in patents as well as documents in general with 2021 being the year of most overall interest and patent publications.



Figure 12. Top 125 pairs of co-occurring concepts in the hydrogen fuel cell literature from 2011-2021

Similar to water splitting, the fuel cell field seeks to improve performance and durability while lowering the costs of fuel cell components to make it more viable for market applications.^{217, 230} This is reflected in the most commonly co-occurring concepts clustered network diagram (Figure 12). The primary concept in the literature is as expected "Fuel Cells", but it is important to note that it is followed by "Solid Oxide Fuel Cells". SOFCs garnered more attention in the 2011-2021 decade than "Proton exchange membrane fuel cells" or "Polymer electrolyte fuel cells". One can also see that there is no mention of "Alkaline Fuel Cells" or "Phosphoric acid Fuel cells" on the top 125 concepts, demonstrating that these are losing popularity in modern times and that the future is moving towards SOFCs and PEMFCs.

Another popular concept is "electric current-potential relationship", a standard concept used when referring to the voltammetry measurements of the cell or half-cell performance utilized in performance evaluation, as can be seen to its link with the "Impedance" cluster that includes impedance, current density, conductivity, etc. Out of the individual fuel cell components, the data shows that although "Fuel Cell electrodes" is a relatively popular topic, higher interest in "Fuel Cell Cathodes" exists. We suspect this is due to research efforts directed to the Oxygen Reduction Reaction (ORR), which occurs in the cathode of most fuel cells, being one of the major challenges when it comes to reducing cost.²³¹⁻²³⁹ One can also see the interest in tackling this challenge with other concepts, particularly "electrochemical reaction catalysts" and "electrochemical reduction" cluster. Similar to water splitting, the inclusion of concepts such as "nanoparticles" (also reflected on Table 1) and "Surface area" in this blue cluster confirms morphology and surface research as important parameters when optimizing catalysts. This is also supported by the key role of carbon and graphene in hydrogen fuel cell research (Table 4.)

The diversity in materials used in Hydrogen Fuel Cell Devices in GHE research has slowly declined (See Figure 11), with 2013 being the year with the highest number of distinct subtances being researched. The major interest throughout the decade has been in Oxides due to the growing interest in materials for SOFCs. The constant appearance of Organic/Inorganic Small molecules is mostly due to electrolyte

components like water, acids as alternative electrolytes (ex: phosphoric acid and sulfuric acid fuel cells), alternative oxidants like H_2O_2 ,²⁴⁰ and association with chemical hydrogen storage like ammonia and methanol.^{191, 192, 241, 242} Polymer interest is due to research for membranes for the PEMFC and AEMFC electrolytes.²⁴³⁻²⁴⁶

There has been some continued interest in substances under the Alloys and Elements designation due to catalyst research. As before mentioned, the never ending pursuit of more affordable yet effective catalysts is also reflected in the interest of nanoparticles (Table 1). Just like in the case of Green Hydrogen Production and Hydrogen Storage, nanoparticles and nanomaterials are playing a big role in hydrogen fuel cells. The application of these materials is diverse, but the literature shows that the main interests are for the increase of surface area, porosity or just general improvements in morphology of Fuel Cell catalysts, especially for ORR. ^{232, 234, 236, 238, 239} This is further explored and demonstrated in the features of the most popular substances in hydrogen fuel cell research and their exemplary publications shown in Table 6.

Table 4. Key substances in hydrogen fuel cell research

Substance Class	Substance	CAS #	# Publicati ons/Citati on	Feature/Areas of interest	Exemplary Publication
Alloy	Cobalt platinum alloy	11134-15-9	42/61	Catalyst; focus on reducing the cost of ORR catalysts by reducing Pt via nanostructuring, catalyst ink formulations, alloying with non-noble metals, etc.	231, 232, 236, 239, 247
	platinum alloys	1273120- 99-2	53/10	Catalyst; reducing the cost of ORR catalysts by reducing Pt via high surface area nanoalloys/Pt-M nanoparticles	234, 238
Element	Carbon	7440-44-0	1562/208 4	Catalyst support; alternatives to noble metal catalysts for oxygen reduction reactions (ORR) via non noble metal-N-C catalysts, high surface area micro/nanostructures of Pt/C and non Pt catalysts, increase of surface defects and porous structures	233, 235, 237, 248, 249
	Graphene	1034343- 98-0	461/624	Catalyst support; filler material for PEMFC; alternatives to noble metal catalysts for oxygen reduction reactions (ORR) via doping of graphene structure with Silicon, Sulphur and/or nitrogen usually on non-noble metal graphene electrocatalysts	235, 245, 250-252
	Nickel	7440-02-0	694/734	Electrode/electrolyte component SOFC; ORR and/or HOR catalyst with focus on nanostructures, porosity, single atom alloys and nano/microstructures; metal foam as flow distributor in PEMFC; attempts to reduce Pt via nanoalloys of Pt as catalysts	238, 253-259
	Platinum	7440-06-4	1239/156 1	Most used and versatile catalyst component, expensive, focus on reducing loading amount Pt via nanoalloys, micro/nanostructures and Pt-M catalysts	248, 260-262
Oxide	Ceria (CeO ₂)	1306-38-3	250/276	interlayer material between electrode and electrolizer in ceramic fuel cells; doped ceria catalyst/electrolyte for SOFC; used in interfaces of membrane and catalysts in PEMFC to better contact area	258, 263-266

	Silica (SiO ₂)	7631-86-9	221/193	used as a template for catalyst synthesis; component in Proton exchange composite membranes; hybrid nanofluid coolant for PEMFC	267-270
	Titania (TiO ₂)	13463-67-7	315/417	ORR catalyst nanocomposite component; catalyst support; organic-inorganic composite membranes for AEMFC	271-274
	Nickel monoxide (NiO)	1313-99-1	225/258	Part of ceramic anode or cathode composition for SOFC, usually reduced to Ni	256, 275, 276
	Yttrium sesquioxide (Y ₂ O ₃)	1314-36-9	127/89	SOFC solid electrolyte dopant or electrode component, focus on formation techniques (printing, plasma spray); degradation studies and microstructures	277-280
	Yttrium zirconium oxide	64417-98-7	125/127	SOFC electrolyte or electrode component, focus on perovskite structure modification, electrode-electrolyte interface, and degradation	264, 279, 281
	Zirconium dioxide (ZrO ₂)	1314-23-4	210/139	SOFC electrolyte; focus on replacing with materials that lower operating temp, obtaining electrolyte thin layer structures, degradation studies and microstructures	279, 282, 283
Polymer	Ethene, homopolymer	9002-88-4	83/8	proton-conducting membranes, bettering performance and durability	246, 284, 285
	Poly(vinyliden e fluoride)	24937-79-9	103/31	proton-conducting membranes; functionalization for selective proton conducting, polymer-ceramic composites for SOFC	286, 287
	Polypropylene	9003-07-0	75/36	proton-conducting membranes, functionalization for better conductivity; Conductive polymer composites for bipolar plates	243, 288, 289
	Polytetrafluor oethylene	9002-84-0	304/137	Enhanced proton-conducting membranes; support for composite membranes; interest in porosity and better PTFE loading	244, 290-292

H 33,647	Number of Documents 47 0 41,784													He 84			
Li	Be	Be											C	N	0	F	Ne
2,064	72	72											34,006	13,255	41,784	7,227	15
Na	Mg	Mg											Si	P	S	CI	Ar
3,318	2,946	2,946											6,862	5,175	6,993	5,022	281
К	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
2,444	1,889	1,186	6,456	1,438	4,648	6,717	11,444	10,281	13,631	5,759	2,690	1,337	279	110	282	970	14
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	ا	Xe
86	7,080	5,368	8,419	1,811	2,858	31	3,253	1,375	4,461	3,007	279	920	2,055	343	160	457	19
Cs	Ba		Hf	Ta	W	Re	Os	lr	Pt	Au	Hg	TI	Pb	Bi	Po	At	Rn
297	2,868		371	883	1,955	302	587	1,749	15,037	3,178	149	41	514	928	7	1	5
Fr 3	Ra 5																
			La 7,470	Ce 7,600	Pr 1,007	Nd 681	Pm 33	Sm 2,132	Eu 130	Gd 2,936	Tb 111	Dy 164	Но 95	Er 179	Tm 63	Yb 458	Lu 69
			Ac 3	Th 39	Pa 4	U 51	Np 5	Pu 14	Am 4	Cm 2	Bk 0	Cf 2	Es 0	Fm 0	Md 0	No 1	Lr 0

Figure 13. Occurrence of elements in materials used for hydrogen fuel cell device research by number of documents from 2011-2021

Many of the substances in Table 6 are comprised of the highlighted elements on Figure 13. Some of the highlighted elements are part of basic components of fuel cells, for example: hydrogen as fuel; oxygen for ORR or oxides in electrodes; Pt, Ni,N, Fe and C for catalysts, etc. Still table 6 did not show all substances associated with solid oxide fuel cell electrolytes and electrodes, especially the important perovskite like ceramics. These are reflected in Figure 13 by the high occurrence of Co, La and Sr.

Cobalt, Lanthanum and Strontium are very common components in perovskite, a type of crystalline material. The compositions of perovskites are very varied, and modifications of the structure to better efficiency and durability are a major research topic in solid oxide fuel cells, especially since perovskites can be used as both electrodes and electrolytes in SOFC.^{228, 264, 293} A great example that includes all three is the perovskite lanthanum strontium cobalt ferrite (LSCF), one of the leading materials for Intermediate temperature SOFCs that possesses mixed ionic and electronic conductivity (MIEC). ^{294, 295} Though cobalt containing cathodes are known to have good performance, there is also a lot of interest in cobalt-free cathodes due to high cost and compatibility issues with electrolytes.²⁹⁶ Many of these cobalt free perovskites still contain La and Sr as well as other elements like Fe, Ba, Cu, Ti, Cr, Sm among others that are reflected in Figure 13.

Global Publication Trends in GHE Technologies by Geography

Our search of the GHE literature from 2011-2021 retrieved a total of 107,293 journal articles and 79,193 patents. Leading the way in number of publications throughout the decade are China, Japan, the US, the Republic of Korea and Germany with China leading the way (Figure 14).

China has the largest number of journal articles, but it's publication numbers in general across the decade have an almost exponential growth and dwarf all other countries (Figure 15). Interest in green hydrogen has surged thanks to China's drive to achieve carbon neutrality by 2060. Some recent real-world examples of China's rising interest in GHE are: the city of Zhangjiakou inaugurating the world's largest electrolyzer for green hydrogen production to provide fuel for hydrogen fuel cell vehicles during the 2022 Winter Olympics;²⁹⁷ Sinopec, China's largest oil refiner, starting to build the world's largest green hydrogen plant, to be entirely powered by solar;²⁹⁸ and top Chinese solar manufacturer, Longi Green Energy Technology Co., investing in the production of electrolyzers for green hydrogen production.²⁹⁹ Though China has yet to introduce a national hydrogen plan, the Chinese Hydrogen Alliance was launched in 2018 by China Energy Corporation and currently has 87 members including universities, research institutions and large companies in the energy production and manufacturing sectors.³⁰⁰

Japan has had the most patents published throughout the decade (Figure 14). Though its patent publications are decreasing yearly (Figure 15), the country is investing significantly. The government provided 370 billion yen (\$3.4 billion) in funds to research, develop, support and promote hydrogen with 70 billion yen allocated toward hydrogen production via water electrolysis.³⁰¹ The interest in hydrogen is also promoted by the Japan Hydrogen Association which includes 274 members between companies, municipalities and universities.³⁰² Japan promoted this technology thoroughly in the Tokyo 2020 Olympics, where the Olympic cauldron was powered by hydrogen, Toyota provided 500 Mirai fuel cell vehicles and 100 fuel cell buses for transportation, and hydrogen based electricity was used in the Olympic village.³⁰³ Another example is the New Energy and Industrial Technology Development Organization's (NEDO) green hydrogen plant, where 45 acres in Namei are now a solar farm where excess solar power is used for water electrolysis.³⁰⁴

The United States produced more journal articles than patents, but interest in in GHE decreased slightly throughout the decade (Figure 15). Still, we see that GHE publications will increase again because clean hydrogen is crucial to the US Department of Energy's strategy for achieving a 100% clean electrical grid by 2035 and net-zero carbon emissions by 2050, and the recent Bipartisan Infrastructure Law includes: \$8 billion for Regional Clean Hydrogen Hubs to expand use of clean hydrogen, \$1 billion for a Clean Hydrogen Electrolysis Program to reduce costs of green hydrogen production, and \$500 million for Clean Hydrogen Manufacturing and Recycling Initiatives to support equipment manufacturing and strong domestic supply chains. ³⁰⁵



Figure 12. Journal and patent publications on GHE by top organization countries/regions



Figure 13. Journal articles and patents over time on GHE for selected countries/regions.

There seems to be a larger academic interest in green hydrogen production due to the large number of journal publications compared to patents (Figure 3 and Table 5). Leading the way in number of publications is China, with its top patent assignee being the Dalian Institute of Chemical Physics, a council member of the Chinese Hydrogen Alliance. This aligns with the recent surge in interest of the solar and oil refining industries of China to build large electrolyzers and green hydrogen plants. The United States places second in number of publications, mostly in journals, followed by Korea and Japan. Table 6 also demonstrates that no US based company in the top assignees have patents, though Japan's Toyota and JX Nippon Oil & Energy Corporation are tied for the second highest number of patents.

The biggest producer of publications on hydrogen storage is China, followed by Japan, the US and Korea. The leading countries are shown to pay attention to both academic research and practical development. Toyota leads the way in patents, supporting it's push in Japan for FCVs. While China has had the most publications overall in the GHE space, Japan leads the way in fuel cell publications, followed by China, the US, south Korea and Germany. Japan has the most patents, which aligns with 9 of the top 15 patent assignees being Japanese based multinational companies (Table 5). China's publications are split almost evenly between patents and journals. The US comes in third with GM being its highest patent assignee, followed by South Korea with Hyundai taking the lead followed by Kia, and then Germany with Bosch and Daimler.

The automotive industry is leading the way in hydrogen fuel cell and storage patent publication, transportation in general being one of the principal applications where new technology and better efficiency is being developed. Some examples of these patents in real life are Toyota's Mirai, Honda's Clarity and Hyundai's NEXO commercial FCVs available in market, as well as FC trucks and buses like Hyundai's XCient and Toyota's Sora.³⁰⁶⁻³⁰⁹ Other manufacturing companies are joining in, with Panasonic just launching a 5kW Hydrogen Fuel Cell Generator³¹⁰ and its plan to build a large facility that uses pure hydrogen fuel cell generators (500 kW) as part of its in-house power for its fuel cell factory department at its Kusatsu site in Shiga Prefecture.³¹¹

Country/Region	Green Hydrogen Production		Hydrogen Storage		Hydrogen Fuel Cells	
	Journal	Patent	Journal	Patent	Journal	Patent
China	24,528	4,829	4,041	3,190	13,747	14,311
Japan	2,188	405	709	970	4,193	28,134
United States	3,785	356	842	391	6,093	5,492
Korea, Republic of	2,475	218	420	377	3,635	7,707
Germany	1,616	222	326	226	2,087	5,278
India	2,553	75	634	25	2,269	252
France	553	159	253	160	1,249	1,272
United Kingdom	786	42	222	44	1,279	806
Taiwan	762	28	123	60	1,080	621
Canada	582	32	179	22	1,336	338
Iran, Islamic Republic of	772	0	230	0	1,115	4
Italy	626	42	198	4	1,043	170

Table 5. Journal articles and patents on GHE by top-producing countries/regions from 2011-2021

Russian Federation	494	54	202	32	950	185
Spain	661	20	123	6	817	82
Australia	701	22	206	20	389	66
Turkey	472	3	241	5	650	29
Switzerland	389	26	55	26	397	161

Table 6. Top patent assignees on GHE in each research area from 2011-2021. Multinational companies are combined under individual names.

Assignee	Number of Patents			
	Green Hydrogen	Hydrogen	Hydrogen	
	Production	Storage	Fuel Cells	
Toyota	37	205	6,768	
Honda	22	28	2,893	
Hyundai	7	41	1,964	
Panasonic	21	47	1,651	
Nissan	2	11	1,629	
Bosch	24	14	1,171	
Daimler	2	14	972	
Kyocera Corp.	2	0	790	
Dalian Institute of Chemical Physics, Chinese				
Academy of Sciences	61	37	626	
Кіа	3	19	670	
NGK Insulators, Ltd.	0	2	566	
JX Nippon Oil & Energy Corporation	37	27	483	
Aisin Seiki Co., Ltd.	0	0	476	
GM Global Technology Operations, Inc.	0	25	452	
Toto Ltd.	6	0	462	

Conclusions and Outlook

Analysis of the literature from 2011-2021 reveals that of the three areas in GHE research, green hydrogen production had the largest upward momentum throughout the decade in terms of total publication volume, whereas hydrogen storage and hydrogen fuel cells experienced a dip in publication followed by an increase in the second half of the decade largely fueled by increasing patent activity. These increasing patent volumes suggest that hydrogen storage and fuel cells are more technologically mature than green hydrogen production, whose proportion of patents has yet to reach 20%. Throughout the decade, the diversity of catalytic materials used in green hydrogen production experiments expanded, while hydrogen storage and fuel cells narrowed in the scope of materials used as a whole. When combined with the increases in patent volume, it appears that these two fields have been focusing in on potentially commercially viable materials, whereas green hydrogen production is still at an exploratory stage where many different avenues are being explored.

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