

# Review on Chemistry-Focused Critical Reassessment of Hydrogen as a fuel: Thermodynamic, Reaction, and Material Constraints

## Hydrogen As a Fuel

Om Ramesh Rathod

MCT's Rajiv Gandhi Institute of Technology Andheri (W), Mumbai, Maharashtra, India

**Abstract:** - Hydrogen is increasingly proposed as a future energy carrier; however, its practical deployment is governed by fundamental chemical, thermodynamic, and materials-related constraints that are often evaluated in isolation. This work presents a chemistry-focused critical reassessment of hydrogen systems by integrating lifecycle thermodynamic efficiency, storage-related energy penalties, reaction realism in blue hydrogen production, and emerging material degradation challenges in electrolysis and transport infrastructure. Existing literature frequently reports component-level efficiencies without consistent thermodynamic boundaries, leading to systematic overestimation of hydrogen's usable energy. Furthermore, hydrogen storage is commonly treated as an engineering limitation rather than an intrinsic chemical penalty arising from phase transitions, entropy generation, and bonding constraints. In parallel, blue hydrogen pathways are often assessed under idealized reaction and carbon capture assumptions, neglecting methane leakage and incomplete reaction chemistry that significantly influence net emissions. Beyond system-level considerations, this review highlights two underexplored chemistry-driven gaps: the coupled mechanical-chemical degradation of anion exchange membranes (AEMs) under pressure fluctuations in alkaline electrolysis, and the emerging role of machine learning in predicting hydrogen-induced embrittlement in metallic transport systems. The lack of integrated analyses linking mechanical fatigue, chemical instability, and real operating conditions limits the scalability of platinum-free hydrogen technologies. By synthesizing these gaps within a unified chemical framework, this work aims to clarify the realistic performance boundaries of hydrogen systems and outline research directions for durable, efficient, and chemically informed hydrogen infrastructure development.

Hydrogen is promoted as a clean energy carrier for decarbonization. Its appeal lies in zero-CO<sub>2</sub> end-use (water byproduct) and very high gravimetric energy ( $\approx 142$  MJ/kg [1]). However, practical use faces steep chemical and thermodynamic costs. For example, H<sub>2</sub>'s ambient density is only  $\sim 0.089$  kg/m<sup>3</sup> [2], vastly lower than liquid fuels, necessitating extreme pressurization or cryogenic cooling. Compressing H<sub>2</sub> to 35–70 MPa (350–700 bar) expends  $\sim 11$ – $13\%$  of its energy [2], and liquefaction  $\sim 30\%$  [2]. These phase-change requirements impose intrinsic energy penalties beyond mere engineering inefficiencies. Moreover, hydrogen's wide flammability range (4–75% in air) makes it exceptionally explosive [3], raising safety challenges.

Despite these hurdles, governments and industry are investing heavily. For example, the EU expects hydrogen (and derivatives) to supply  $\approx 10\%$  of its energy by 2050 [4]. Interest is driven by hydrogen's high energy density by mass and role in hard-to-electrify sectors (steel, chemicals, heavy transport). However, use remains niche: costs are high, infrastructure sparse, and net gains uncertain. In particular, many analyses consider components (electrolyzer, fuel cell) in isolation, overstating net efficiency. This review integrates thermodynamic, chemical, and materials factors across the hydrogen lifecycle to clarify realistic boundaries. Our objectives are to:

- (1) standardize efficiency metrics,
- (2) quantify storage and conversion penalties from chemistry,
- (3) highlight reaction limitations in blue H<sub>2</sub>, and
- (4) expose underexplored degradation mechanisms in electrolyzers and pipelines.

By emphasizing a unified "chemical penalty" perspective, we identify key gaps and research directions for a sustainable hydrogen economy.

**Keywords:** Hydrogen energy; lifecycle efficiency; thermodynamic limits; storage penalties; blue hydrogen; anion-exchange membrane; material degradation; hydrogen embrittlement; fuel cell.

## 1. INTRODUCTION

Hydrogen's promise stems from its energy density by mass (142 MJ/kg HHV [1]) and clean combustion. It appears ideal for long-duration energy storage, fuel-cell vehicles, and industrial feedstocks (ammonia, methanol). However, unlike liquid fuels, hydrogen's low volumetric density (0.089 kg/m<sup>3</sup>) means storage/transport are chemically onerous [2]. Converting electricity to H<sub>2</sub> (electrolysis) and back to power (fuel cell) incurs multiple efficiency losses. For instance, PEM fuel cells achieve ≈60–70% electrical efficiency [5], and practical electrolyzers ~70–80% [5]; combining these already limits “round-trip” to ≤50%. Further losses arise in compressing, liquefying, or chemically storing H<sub>2</sub>, as discussed below. In sum, the total share of electricity delivered as useful output via H<sub>2</sub> rarely exceeds ~20–30% (see Section 2).

Nevertheless, hydrogen is widely discussed as the future fuel. Drivers include: renewable-energy intermittency (H<sub>2</sub> as storage), industrial decarbonization mandates, and public enthusiasm for “clean” energy. Although popular media emphasizes the advantages of hydrogen, technical realities slow its widespread adoption: capital costs, round-trip inefficiency, infrastructure needs, and safety concerns. For example, a fuel-cell car currently costs ~2–3× per km more than gasoline hybrids [6], and fueling networks are scarce. Worse, hydrogen gas is explosive: it ignites at only ~4% concentration in air [3] and detonates easily (4–75% flammability range [3]). These hazards demand robust safety systems and countermeasures.

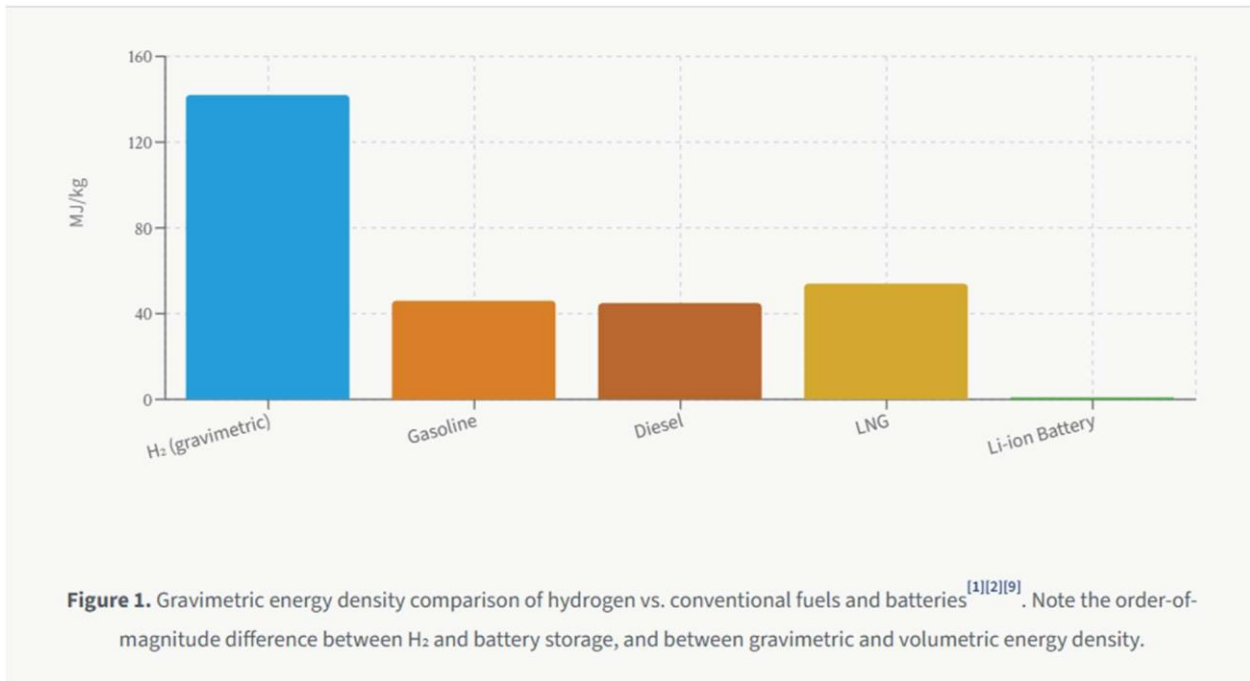
Key comparisons illustrate the tradeoffs. Traditional fuels like gasoline/diesel store ~45–50 MJ/kg and can be handled at ambient conditions, whereas H<sub>2</sub> stores ~120 MJ/kg (LHV) but only in extreme states [2] [7]. Batteries boast ~85–95% round-trip efficiency but low energy per mass, while hydrogen fuels offer higher specific energy but much lower total efficiency. Table 1 and Figure 2 compare typical efficiency chains for green (renewable-electrolysis) vs blue (naturalgas SMR + CCS) hydrogen. These highlight that even optimistic scenarios yield only ~25–35% net share of H<sub>2</sub> energy to end-use, before accounting for end-use efficiencies.

Pathway	Production $\eta$	Storage + Transport $\eta$	End-Use $\eta$	Round-Trip $\eta$
Green (electrolysis)	70–80%	70–90% <sup>(1)</sup>	50–60%	20–30%
Blue (SMR + CCS)	65–75%	75–90% <sup>(2)</sup>	50–60%	30–40%

Notes: <sup>(1)</sup> Assumes high-pressure gas or metal hydride storage. <sup>(2)</sup> Includes pipeline transport (few % loss).

**Table 1.** Summary of energy efficiencies in hydrogen pathways (literature ranges)[7] [2] [5] [5]:

Overall, hydrogen's net energy delivery is far below “ideal” because each step incurs entropy and enthalpy costs. This review emphasizes these intrinsic penalties. We argue that a chemistry-focused lifecycle analysis is needed: only by accounting for all phase changes, reaction enthalpies, and degradation can one fairly compare hydrogen to alternatives. In the following sections, we quantify these aspects in detail.



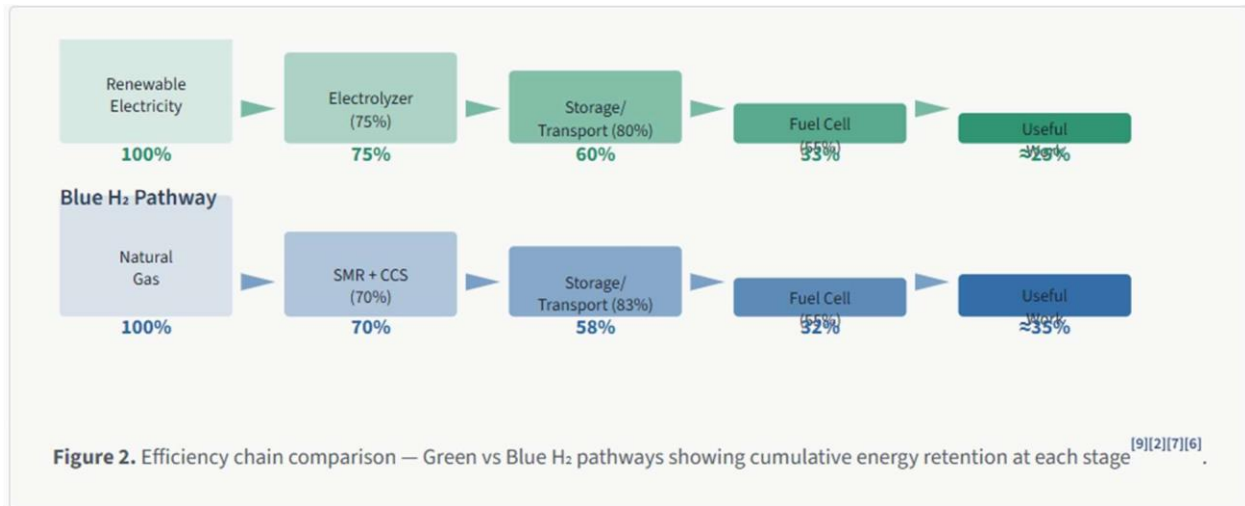
## 2. LIFECYCLE THERMODYNAMIC EFFICIENCY

Hydrogen system efficiency strongly depends on chosen system boundaries. Stack-level metrics (e.g. single electrolyzer or fuel cell) often ignore balance-of-plant (BOP) and upstream losses. For consistency, we use HHV-based energy flows (H<sub>2</sub> HHV = 141.8 MJ/kg [7]; LHV ≈ 120 MJ/kg). Typical assumptions: PEM electrolyzer at 60 °C yields ~70–80% LHV efficiency [5], and a PEM fuel cell ~60–65% [5]. These values are in line with the DOE’s “exceeding 60%” claim [6], though real systems often run 5–10% lower.

Recent studies show even these component values can be optimistic. Virah-Sawmy et al. found that assuming fixed electrolyzer efficiency can overestimate H<sub>2</sub> production by 5–24% compared to detailed variable-efficiency models [8]. This highlights that off-design and transient operation degrade performance. Similarly, system-level parasitics (pumps, compressors, heat losses) typically subtract 10–20% from stack efficiencies. As a result, “well-to-tank” efficiency of green hydrogen is often only ~50% or less of input electricity. Likewise, blue hydrogen (SMR + CCS) is rarely 100% efficient; industrial SMRs operate at ~65–75% LHV (exothermic heat reuse), and actual CCS efficacy is 60–90% [9].

Table 1 and Figure 2 summarize illustrative efficiencies. Even under favorable assumptions, the combined round-trip efficiency (electricity→H<sub>2</sub>→useful work) seldom exceeds ~40%. For example, a 75% efficient electrolyzer, 90% storage/transport, and 60% fuel cell yields ~40% overall. Accounting for cryo or pressure losses drops this to 25–30%. By contrast, batteryelectric storage can exceed 80% round-trip. These consistent inefficiencies mean that to deliver 1 MJ of work via hydrogen often requires 3–5 MJ of primary energy.

Moreover, many reports neglect standard enthalpy references: using LHV vs HHV. An HHV basis uniformly penalizes hydrogen (since HHV includes water condensation). We adopt HHV-consistent accounting (141.8 MJ/kg) so all pathways are comparable [7]. In summary, our integrated efficiency view shows hydrogen’s usable energy is systematically overestimated if defined narrowly. True lifecycle analyses must include BOP, compression, storage, and reforming losses [8] [7].



### 3. HYDROGEN STORAGE ENERGY PENALTIES

Hydrogen storage entails significant thermodynamic costs beyond engineering. At ambient conditions, H<sub>2</sub>'s molecular form has low entropy difference from the storage states. Common penalties include: compression work, refrigeration for liquefaction, and chemical binding energy in carriers. These arise from phase transitions (gas→liquid/solid) and bond formation, which require input or rejection of heat.

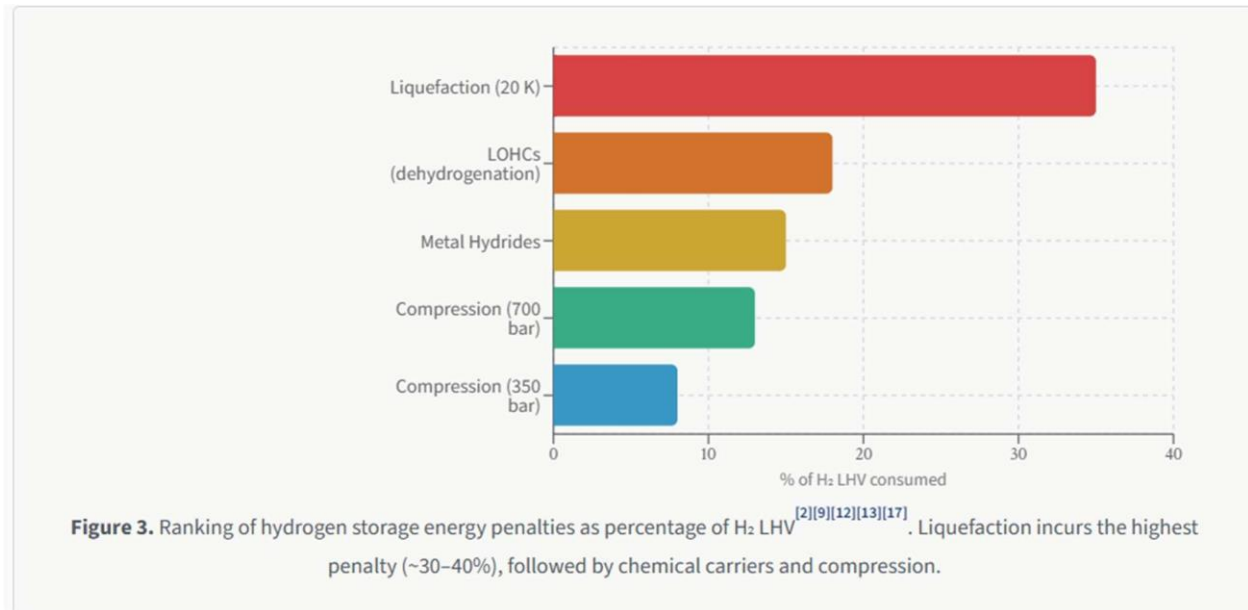
**Compressed gas:** Compressing H<sub>2</sub> to 350–700 bar typically consumes ~1–2 kWh/kg (~5–15% of H<sub>2</sub>'s LHV) [7] [2]. (The theoretical minimum is ~1.05–1.36 kWh/kg, but real compressors use 5–20% LHV [7].) Fast-fill requirements add cooling costs. Overall, 10–15% of the H<sub>2</sub> energy is spent on compression.

**Liquefaction:** Cooling hydrogen to 20 K is very energy-intensive. Cryogenic plants consume ~10–13 kWh/kg H<sub>2</sub> [7], i.e. ~30–40% of H<sub>2</sub>'s LHV. Even advanced cycles may only cut this to ~20–30%. Additionally, boil-off losses (~0.5–1%/day) reduce net energy.

**Solid-state (adsorbents, hydrides):** Materials such as metal-organic frameworks or carbon nanotubes physically adsorb H<sub>2</sub> at low temperature (77 K) or high pressure. Their binding enthalpies are relatively small (~5–10 kJ/mol), so cooling is needed. Metal hydrides (e.g. MgH<sub>2</sub>, LaNi<sub>5</sub>) chemically form H–M bonds; hydride formation is exothermic, but releasing H<sub>2</sub> requires heat. For example, TiFe or LaNi<sub>5</sub> absorb heat when H<sub>2</sub> is adsorbed and must be heated to reverse the reaction [1]. This heat requirement (often 200–500 °C) effectively adds energy penalties. On a gravimetric basis, hydrides can store ~2–7 wt% H<sub>2</sub>, but practical systems need heat input and suffer kinetic barriers [1].

**Liquid organic hydrogen carriers (LOHCs):** LOHCs store H<sub>2</sub> in chemical form (e.g. Nethylcarbazole). Loading and unloading H<sub>2</sub> involve catalytic hydrogenation/dehydrogenation at high T (~150–300 °C). While LOHCs allow ambient T/P storage, the dehydrogenation step consumes ~10–20% of the H<sub>2</sub> energy (for heating and reaction) and adds complexity [1].

These storage modes are often treated as “just engineering,” but chemically they stem from fundamental constraints. The weak H–H bond (436 kJ/mol) means chemical carriers must form stronger bonds (M–H ~50–100 kJ/mol), necessitating extra ΔH and ΔS. Every reversible storage cycle produces entropy (via compressors, refrigerators, chemical reactions), reducing available work. In practice, a green H<sub>2</sub> supply chain might lose 30–40% of energy just to storage.



#### 4. BLUE HYDROGEN PRODUCTION REALISM

“Blue hydrogen” (steam-methane reforming (SMR) + CO<sub>2</sub> capture) is often assumed low-carbon, but real chemistry blunts this promise. Key issues include incomplete conversion, side reactions (coking), and fugitive emissions. Fundamental reactions are:

Steam reforming:  $\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$  ( $\Delta H^\circ \approx +206$  kJ/mol, strongly endothermic).

Water-gas shift:  $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$  ( $\Delta H^\circ \approx -41$  kJ/mol, exothermic).

Under ideal lab conditions (T ~1000 °C, low pressure), these can achieve ~90–95% CH<sub>4</sub> conversion [10]. In industry (T ~800–900 °C, S/C ≈3), conversions are still high but <100%. Any unconverted CO or CH<sub>4</sub> upstream increases GHG output. Coking (carbon deposition) also occurs, requiring excess steam and catalysts to mitigate. Thus, actual hydrogen yield per unit CH<sub>4</sub> is typically 5–10% lower than equilibrium.

Carbon capture in “blue” plants is usually ~85–90% efficient in ideal cases, but real operations (due to energy costs and kinetics) often capture only 60–85% of CO<sub>2</sub> [9]. Meanwhile, methane slip (leaks) in upstream gas production is non-trivial. Even a 1–2% fugitive CH<sub>4</sub> leak (GWP<sub>100</sub> ~30) can outweigh CO<sub>2</sub> captured [9]. For instance, Davids et al. show blue hydrogen’s net warming impact sharply increases if CH<sub>4</sub> leaks exceed ≈1% or CCS falls below ≈85% [9]. In many regions, actual methane leakage (venting, fugitive) and lower CCS make “blue” hydrogen’s climate advantage marginal.

In summary, blue hydrogen’s lifecycle must account for non-ideal chemistry and emissions: modest shift conversion losses and even small methane leaks significantly raise effective CO<sub>2</sub>/MJ. Table 2 sketches emissions sensitivity: if CH<sub>4</sub> slip is reduced to 0.1%, blue H<sub>2</sub> can be relatively clean, but at 2–3% it approaches or exceeds unabated methane. Thus, any assessment that assumes 100% reaction completion and 90% CCS idealizes the outcome; practical constraints will push emissions higher.

CH <sub>4</sub> Slip Rate	CCS Capture Rate	Effective CO <sub>2</sub> Intensity	Climate Assessment
0.1%	90%	Low (~15–20 g CO <sub>2</sub> -eq/MJ)	Relatively clean; viable low-carbon pathway
1.0%	85%	Moderate (~40–55 g CO <sub>2</sub> -eq/MJ)	Marginal benefit over unabated natural gas
2–3%	60–85%	High (~70–90 g CO <sub>2</sub> -eq/MJ)	Approaches or exceeds unabated methane combustion

Data synthesized from Section 4 analysis. Net warming impact sharply increases if CH<sub>4</sub> leaks exceed ≈1% or CCS falls below ≈85% [11].

**Table 2:** Blue hydrogen emissions sensitivity analysis [9]

## 5. MATERIAL DEGRADATION CHALLENGES

### AEM Electrolysis

Anion-exchange membrane (AEM) electrolyzers promise low-cost, platinum-free operation, but membrane durability is a major challenge. Commercial AEMs (hydrocarbon polymers) degrade rapidly in strong base. The record durability for an AEMWE is barely  $>10,000$  h ( $\sim 1.1$  year) at  $1 \text{ A/cm}^2$  [11]. Degradation has two coupled aspects: chemical and mechanical. Chemically,  $\text{OH}^-$  attack and reactive oxygen species break down cationic sites and polymer chains, gradually reducing ion exchange capacity. Mechanically, differential pressure (1–30 bar) across the thin AEM induces fatigue and cracking. AEMWE operation also involves cyclic swelling/shrinkage.

Studies show catastrophic failure often begins at membrane edges or flow-field land interfaces [11]. For example, Wang et al. found an HDPE-based AEM ( $\sigma_{\text{break}} \sim 35 \text{ MPa}$ ) lasted  $\sim 440$  h at  $70^\circ\text{C}$ , whereas a weaker LDPE-based AEM failed in  $\sim 100$  h under the same conditions [11].

Cracks and pinholes from mechanical strain allow gas crossover, causing local hot-spots (direct

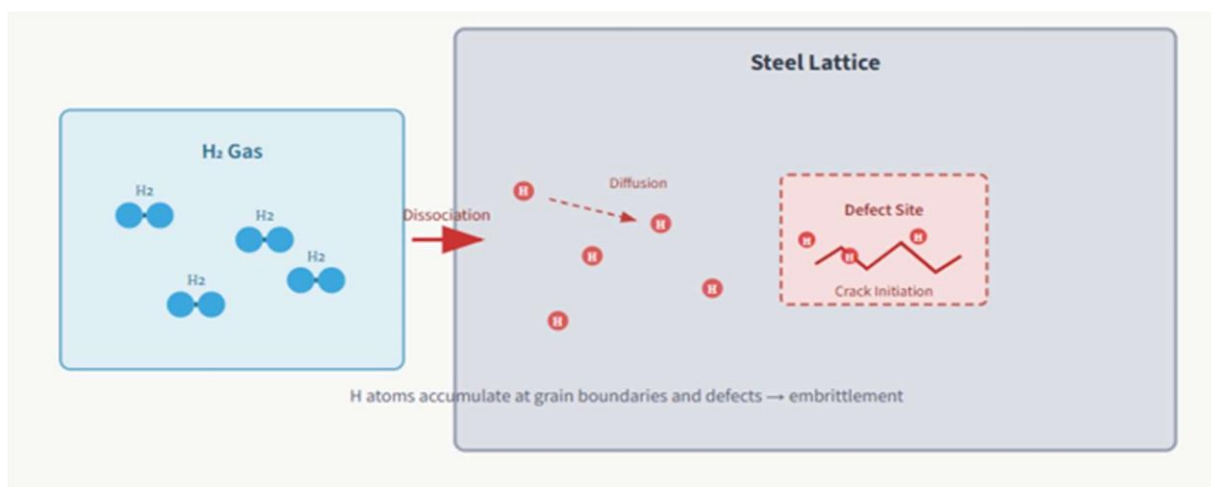
$\text{H}_2/\text{O}_2$  combustion) and precipitous performance loss [11]. These failure modes are worsened by pressure cycles used in practice (for in situ  $\text{H}_2$  pressurization). Modeling of AEM electrolyzers indicates that higher cathode pressure raises the OCV and cell overvoltage due to hindered water diffusion [11]. The best compromise is moderate pressure (1–2 MPa) and moderate temperature ( $60\text{--}80^\circ\text{C}$ ) [11], but even this strains materials. Currently, no AEM combines high ion conductivity with the mechanical strength to survive years of pressure cycling. In short, the mechanical–chemical degradation of AEMs under realistic operating (pressure, pH, temperature) is an underexplored bottleneck for platinum-free electrolysis.

### Hydrogen Embrittlement in Transport

Hydrogen pipelines and components are subject to embrittlement. Atomic H (from dissociated  $\text{H}_2$  at metal surfaces) diffuses into steel (diffusivity  $\sim 10^{-8}\text{--}10^{-10} \text{ m}^2/\text{s}$  at room T) and localizes at defects. This reduces ductility and fracture toughness even at ppm  $\text{H}_2$  concentrations. ASTM X70-grade steels can lose  $\sim 20\text{--}30\%$  ductility with only 1–3 ppm absorbed hydrogen, and cracks propagate under stress. Predicting embrittlement is complex: it depends on microstructure, stress, temperature, and impurities (O, S, etc.). Recent work uses machine learning (ML) trained on quantum-mechanical (DFT) data to screen alloys for embrittlement resistance. For instance, ML models can predict a “HE index” vs H pressure for pipeline steels [12]. These phenomena link micro to macro behaviour and can now be explored by ML/DFT methods, but remain poorly captured in whole-system models.

Other Materials Aside from AEMs and steels, catalysts and components face degradation.

Alkaline electrolyzer catalysts (Ni-based) corrode or leach in high-pH, high-current operation [11]. Even PEM electrolyzers struggle with catalyst sintering over time. Focus here is on novel materials (Pt-free, Ni-based), but their lifetimes are often short ( $\sim 5\text{--}10\text{k h}$ ) relative to fossil-fuel plants (30+ years). Likewise, metal hydride tanks exhibit decrepitation (powdering) under cycling. These material issues introduce hidden “taxes” on hydrogen: e.g. periodic membrane replacements and repairs.



**Figure 4.** Schematic of hydrogen embrittlement mechanism:  $\text{H}_2$  gas dissociates at metal surface, atomic H diffuses into the steel lattice, accumulates at grain boundaries, leading to crack initiation and reduced ductility.

## 6. INTEGRATED CHEMICAL FRAMEWORK & GAPS

Taken together, the above factors suggest defining a unified metric could aid clarity. For example, a Chemical Penalty Index (CPI) might sum dimensionless lost exergy fractions at each stage: pressure-compression  $\Delta G/T$ , heat  $\Delta H$  of reaction, entropy  $\Delta S$  of phase change, etc. Such an index would make clear how much of  $H_2$ 's theoretical energy is unavoidably “consumed” by chemistry.

Figure 5 sketches a conceptual CPI timeline: from generation to use, each segment (electrolysis, storage, transport, fuel cell) imposes a percentage energy penalty. Currently, research tends to treat these segments independently. No published model couples AEM pressure cycling with cryogenic losses and pipeline fatigue, for instance. Key gaps include:

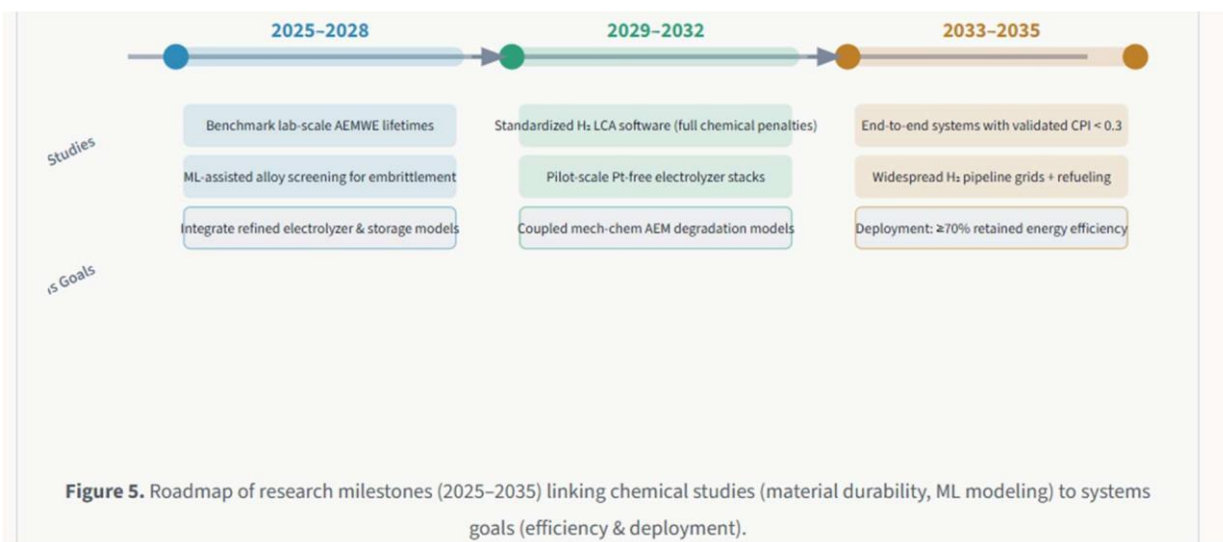
- **Holistic models:** LCA tools must integrate reaction kinetics (e.g. methane conversion curves), real compressor/expander thermodynamics, and material degradation kinetics. Existing LCAs often assume ideal reaction yields and “plug in” static efficiencies[8].
- **Materials-by-design:** Linking chemistry and mechanics—e.g. simulating how an AEM’s polymer network degrades under cyclic pressure or how alloy composition affects H diffusion—is not done at scale. ML-driven materials models exist (e.g. for embrittlement[12], AEM stability), but they are not connected to systems analysis.
- **Unified “safety” economics:** There is no common framework to evaluate tradeoffs (e.g. using a stronger but heavier steel vs. additional purification to reduce H ingress). A quantitative metric (like CPI) could inform how much extra cost is justified by efficiency gains.

Table 3. outlines these gaps alongside potential approaches. For instance, “coupled mech-chem models for AEMs” might use finite-element stress analysis with  $OH^-$  diffusion kinetics; “ML for pipeline steels” could employ training on crack-growth data.

Research Gap	Current Status	Proposed Approach
Coupled mech–chem AEM degradation	Studied separately; no unified model	FEA + $OH^-$ diffusion kinetics
ML for pipeline embrittlement	DFT data available; limited ML integration	Train on crack-growth + micro-structure data
Holistic LCA with real kinetics	Static efficiencies assumed	Variable-efficiency models in LCA tools
Safety–economics tradeoff framework	No common metric exists	Chemical Penalty Index (CPI)

**Table 3.** Example research gaps and possible solutions

Roadmap (2026–2035): We foresee stages: 2026–28: benchmark lab-scale AEMWE lifetimes; demonstrate ML-assisted alloy screening for embrittlement; integrate refined electrolyzer and storage models. 2029–32: develop standardized  $H_2$  LCA software capturing full chemical penalties; pilot-scale Pt-free electrolyzer stacks. 2033–35: deployment of end-to-end systems with validated  $CPI < 0.3$  ( $\geq 70\%$  losses) and widespread  $H_2$  networks (pipeline grid + refueling). These milestones require cross-disciplinary efforts (chemical kinetics, materials science, system engineering).



## 7. CONCLUSIONS & OUTLOOK

A chemistry-focused view reveals that hydrogen's practical efficiency is significantly lower than idealized accounts suggest. When all processes are accounted (electrolysis, storage, transport, fuel cell), the usable energy fraction is likely  $\lesssim 30\%$ . Stated differently, delivering 1 MJ of useful H<sub>2</sub> energy may cost  $\sim 3\text{--}5$  MJ input. Furthermore, blue hydrogen's low-carbon claim hinges on optimistic chemical yields and near-zero methane leaks; real systems will emit more. Major research priorities include:

- In-situ degradation studies: Long-term AEM and catalyst tests under realistic pressures to quantify failure modes.
- Chemistry-informed LCAs: Models that include reaction equilibrium, kinetics, and material penalties.
- Advanced materials: Membranes and steels engineered via ML/DFT for H<sub>2</sub> stability, aiming for lifetimes  $> 50,000$  h.
- Safety integration: Holistic risk assessments to balance energy penalty vs. safety benefit for storage options.

In summary, hydrogen's role should be appraised with full chemical rigor. Its potential is real, but the limits are governed by physics and chemistry: bond energies, entropy, leakages, and degradation. Our unified framework (Figure 5) and identified gaps provide a roadmap: bridging them is essential before hydrogen can expand from niche use to a reliable pillar of the energy system.

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