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1 Pathways to Electrochemical Solar-Hydrogen Technologies

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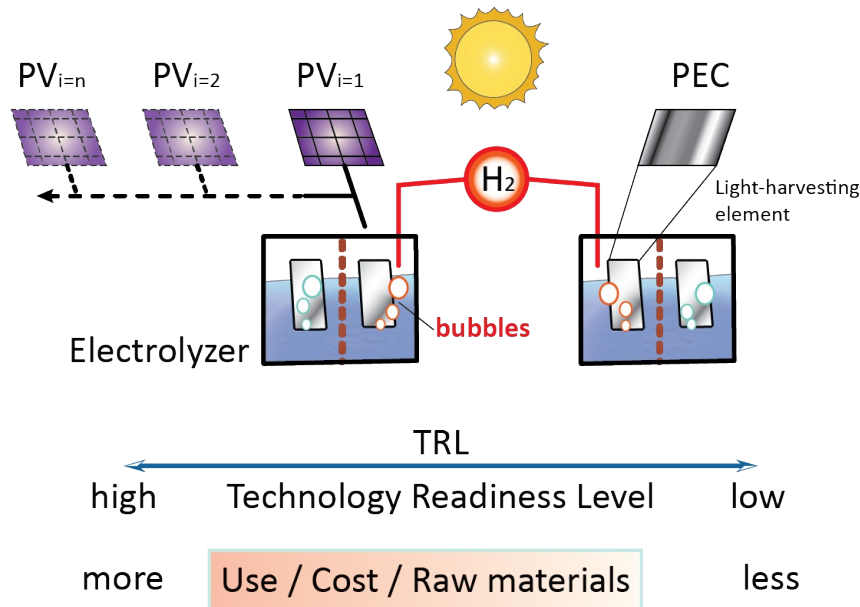
78

791. **Introduction**

80 Solar-powered technologies for the electrochemical production of hydrogen through water
81electrolysis are of significant immediate interest. These so-called “solar hydrogen” technologies are able
82to capture solar energy and efficiently store it as hydrogen for widespread use when demand is high,
83uniquely for stationary applications, as a mobile transportation fuel, and as a reductant for various
84chemical transformations. This application space complements others covered by alternative technologies
85that capture solar energy and generate electricity (e.g. photovoltaics) or heat (e.g. solar-thermal systems).
86Over the past decade, several large research programs around the globe have been implemented with the
87aim of accelerating the development of the science and technology of solar-hydrogen devices: The
88Swedish Consortium for Artificial Photosynthesis, NSF Center for Chemical Innovation in Solar Fuels,
89the Joint Center for Artificial Photosynthesis, The Korean Center for Artificial Photosynthesis, the
90Institute for Solar Fuels at the Helmholtz Center in Berlin, the Japan Technological Research Association
91of Artificial Photosynthetic Chemical Process, The VILLUM Center for the Science of Sustainable Fuels
92and Chemicals in Denmark, the Center for Multiscale Catalytic Energy Conversion and the Towards
93BioSolar Cells program in The Netherlands, the PEC House and Solar Hydrogen Integrated
94Nanoelectrolysis Project (SHINE) in Switzerland, the UK Solar Fuels Network, among others. These
95large-scale programs, in conjunction with the efforts of small teams of researchers worldwide, have
96contributed to a clearer understanding of the requirements and challenges of solar-hydrogen
97technologies,¹⁻¹⁰ placing us in an appropriate position to perform an informed assessment on the feasibility
98of their future deployment. On June 13–17, 2016, fifty-two participants from 10 countries and 32 different
99organizations with expertise in multiple areas of solar hydrogen gathered at the Lorentz Center in Leiden,
100The Netherlands (<http://www.lorentzcenter.nl/>). Participants represented leading research institutions, the
101industrial sector, social scientists evaluating the societal impact and perception of solar-hydrogen
102technologies, and delegates from several governments. Attendees with this breadth in expertise and
103experience in solar hydrogen and broad topic discussions made this workshop unique. Over the five days

104of the workshop multiple topics were discussed and debated, including the state-of-the-art and limitations
105of materials, device architectures, early-stage market opportunities, and a roadmap for the implementation
106of solar-hydrogen technologies into large-scale energy markets. Several coupled considerations were
107examined for successful implementation of solar-hydrogen devices: (1) technical constraints for the
108robust and stable long-term operation of the system, (2) economic viability and environmental
109sustainability, and (3) societal impacts and political drivers. The most important outcome from the
110workshop was a specific technology roadmap for solar hydrogen devices, which had not previously
111existed.

112 The minimum requirement for a practical solar-hydrogen system is that it uses sunlight to convert
113water to a hydrogen stream that contains oxygen at a concentration below the flammability limit.^{11, 12}
114Here, we only consider devices and systems that generate H₂ via proton/electron-transfer redox reactions
115driven by gradients in electrochemical potential formed by non-thermal photovoltaic action resulting from
116sunlight absorption. While this includes processes such as solar photovoltaic plus electrolyzers,
117photoelectrochemistry, photocatalysis, and molecular approaches, we recognize that other processes are
118possible as well (e.g., using light to drive thermochemical hydrogen generation). For clarity and
119simplicity, we classify device architectures into two broad categories as described in Figure 1 and Table 1:
120photovoltaic-driven electrolysis (PV–Electrolysis) and photo-electrochemistry (PEC).¹³⁻¹⁵



121

122**Figure 1.** Scheme representing PEC and PV–Electrolysis device concepts, including current use, projected costs,
 123amount of raw materials, and current relative level of technology readiness. For more details, see Table 1.

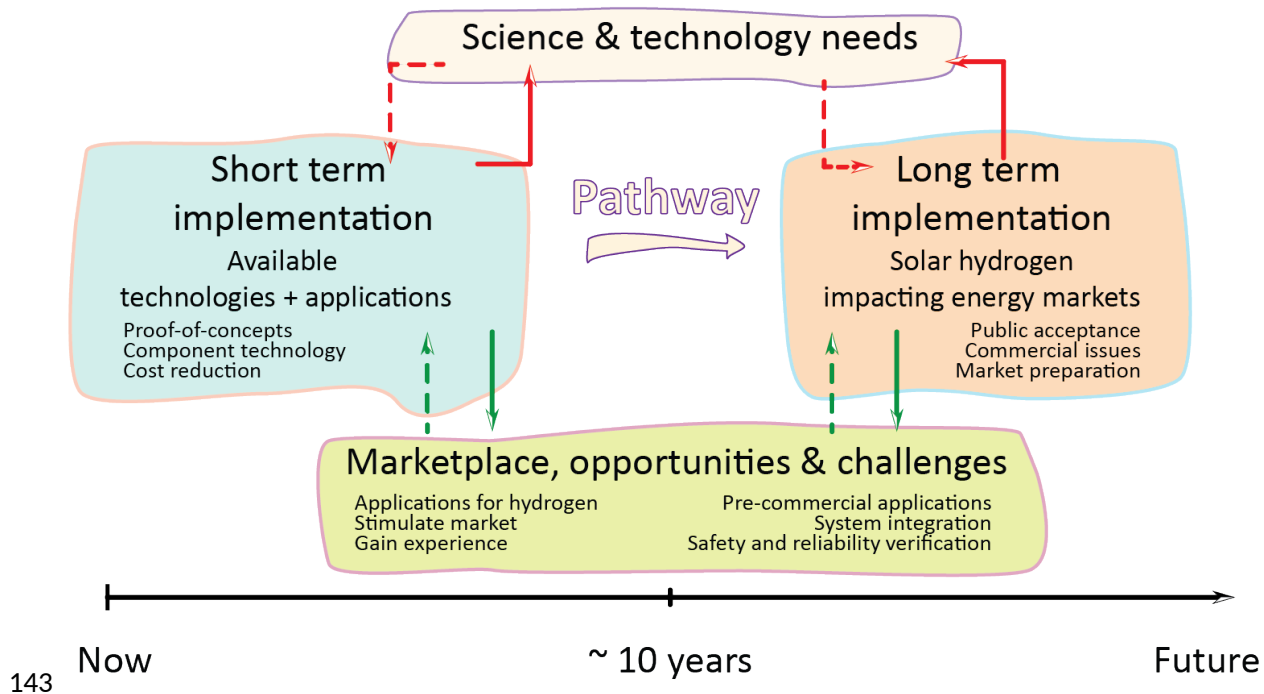
124**Table 1. PV–Electrolysis versus PEC systems.** Overview of general concepts, and comparison of unique
 125characteristics, technological considerations, economic challenges and political factors for each device type.

	PV–Electrolysis systems		PEC systems
General concept	Over large areas, sunlight is used to convert water to a stream of hydrogen that contains an oxygen concentration below the flammability limit		
Terminology	<i>Components:</i> Light absorbers, Electrocatalysts, Ion-exchange membranes, Electrolytes, etc. <i>Devices:</i> PV, PEC, Light absorber in electrolyte with co-catalyst, etc.		
Unique aspects	Light absorption component (PV) physically separated macroscopically from water splitting (electrolysis) component		Light absorption and water splitting components are integrated in one region
Technological options	<i>Distributed</i> Hydrogen production is independent of energy generation (different sources, electricity grid)	<i>Centralized</i> Hydrogen production occurs at the site of energy generation; requires hydrogen transport	<i>Centralized</i> Design concept exclusively allows centralized operation
Technology readiness	Advanced stage		Early stage, and exploratory for: nano-/micro-structured, and particulate/molecular components
Maximum demonstrated Solar-to-Hydrogen	30% for > 48 h ¹⁶		10% for > 40 h ¹⁷

efficiency[†]	
Economic challenges	Competition with conventional sources of non-renewable energy (fossil fuel, nuclear), battery-backed renewable energy, and hydrogen generated by other means (methane reforming) in terms of cost, availability, and accessibility
Socio-political factors	Investments are not always stable (e.g. elections, political agendas, influential special interest groups); events affect public and political perception, perceived relevance and public acceptance (e.g., oil spill, nuclear disaster, hydrogen explosion, decreasing energy prices, environmental benefits, societal push for renewable or more sustainable energy solutions)

126 [†]Based on laboratory-scale device demonstrations capable of producing nearly pure H₂.

127 The first family comprises at least two devices where the light absorption component (PV) is
128physically separated from the water-splitting/electrolysis component (Electrolyzer). These types of
129devices are the most mature and benefit from modularity, allowing individual devices to be optimized for
130the integrated operation. However, this modularity also often necessitates use of two encapsulation and
131support structures. For the other category of PEC devices, the light absorption and water splitting
132components are co-located or assembled into a single component and the light absorber is directly
133influenced by the properties of the electrolyte potentially simplifying the device architecture. In this
134context, PEC devices include those based on photoelectrodes where two half reactions can be spatially
135separated by a membrane and particles suspended in an electrolyte where they cannot be separated.^{13, 18}
136PEC devices are less mature, and therefore less technology readied, than PV–Electrolysis devices, yet we
137do not define a quantitative technology readiness level for either technology because of differing global
138metrics. In its place, we refer to “Low technology readiness” for technologies that are far from
139commercialization, and “High technology readiness” for technologies that are already commercialized or
140beyond the large prototype stage, and evaluated in their intended environment. A technology may be
141assigned a high technology readiness at the device or system level, while advanced components for
142improved performance may still be at a low technology readiness level.



144 **Figure 2.** Schematic representation of a pathway and timeline for solar H₂ technologies and interrelated aspects
 145 discussed in this article.

146

147 In this perspective paper, we discuss potential pathways for solar-hydrogen technologies, as depicted
 148 in Figure 2. The first section describes general considerations for solar-hydrogen technologies, including
 149 technical approaches for device and system architectures, economic challenges, and societal and political
 150 impacts. The second section describes pathways for implementation of solar-hydrogen technologies,
 151 including specifically, markets for short-term implementation (≤ 10 years) of combined PV–Electrolysis
 152 devices and systems together with technological challenges and research opportunities. For long-term
 153 implementation, potential pathways for both combined PV–Electrolysis devices and systems, as well as
 154 PEC devices, are considered together with other important societal, economic, political drivers and
 155 technological requirements.

156

157 **2. General considerations**

158 **2.1. Technical options**

159 When evaluating the device categories (PV–Electrolysis or PEC), it is instructive to classify the
160 design strategy. One classification is whether a technology is considered distributed or centralized. Within
161 this article, *Distributed* approaches are defined as those that rely on the collection of sunlight by discrete
162 solar-module installations followed by transport of energy to electrolyzer units at a different and possibly
163 distant location. *Centralized* approaches are defined as solar installations that directly drive the water-
164 splitting processes. Based on this technology classification, for a given hydrogen production goal, both
165 *Centralized* and *Distributed* approaches could be implemented as either large-scale production facilities
166 placed in one single location or as a collection of small-scale facilities dispersed geographically. PV–
167 Electrolysis designs can be classified as either distributed or centralized while the inherent integrated
168 nature of PEC designs necessitates that they are only centralized. Agnostic to the classification of the PEC
169 or PV–Electrolysis designs is the requirement that they must operate with fluctuating energy inputs,
170 because of the intermittency of solar irradiation. This challenge is significant and will also affect the
171 implementation of centralized solar-hydrogen technologies. Section 3.1 presents a more detailed
172 discussion on possible solutions for the PV–Electrolysis approach using alternative energy sources
173 present in traditional electricity grids and the research opportunities that may provide solutions to
174 overcome it in the short-term.

175 The distributed PV–Electrolysis design strategy can take advantage of electricity grids for the
176 required electronic transport, and by doing so the electrolyzer can also utilize energy from various sources
177 (e.g. from wind, fossil fuels), therefore avoiding fluctuations in its operation due to the intermittency of
178 solar irradiation.¹⁹ By having the option to transport charge instead of hydrogen over large distances,
179 hydrogen transportation from centralized sunny locations to consumer centers is not necessary.
180 Distributed approaches require implementation of power electronics to enable electricity transmission
181 from PV installations to the electricity grid (e.g. DC–DC converters, AC–DC inverters) and subsequently
182 to the electrolyzers.²⁰ Power electronics add to the cost of the system and decrease system efficiency,
183 while transmitting electricity through the grid results in additional costs which are defined by the

184electricity markets. A specific option for distributed approaches is the implementation of alternative
185electricity grids that are exclusively used for PV–Electrolysis (possibly operated under direct current, like
186those envisioned in Europe and China and only requiring DC–DC converters).^{21, 22} If new infrastructure is
187needed for these DC grids, this approach requires a large upfront capital investment but saves operational
188expenses related with electricity grid transmission costs and management.

189 In contrast to the distributed PV–Electrolysis design strategy, an advantage of centralized PV–
190Electrolysis implementation is the ability to optimize the PV array operation for the electrolysis needs.
191This also enables the option to operate with minimal power conversion, which can result in cost
192reductions and efficiency improvements. Moreover, because larger sizes result in greater economic
193benefits, both the PV component and the electrolysis component can be implemented on very large scales;
194a similar situation can be seen for the centralized case. The main disadvantage of centralized solar-
195hydrogen facilities is the need to cover large land mass areas and then transport the generated fuel to its
196point of use. In the case of PEC approaches, by definition the light absorption and water splitting
197components operate at the same centralized location, and thus PEC has similar benefits and deficiencies
198as centralized PV–Electrolysis.

199 PV–Electrolysis devices have a higher technology readiness level than PEC devices.^{23, 24} PV panels
200and electrolyzers are already established in the market and are continually optimized (as independent
201installations). PEC devices are still in the early stage of development and could enter the market in the
202medium-to-long-term (> 10 years) (Figure 2). In the medium-term, the technologies most likely to
203succeed are those that leverage semiconductor manufacturing techniques to fabricate planar
204photoelectrodes. In the long-term, advanced structural designs may be cost-effective where the PEC units
205are micro-/nano-structured, inexpensive flexible substrates are used, or particles or molecules are
206suspended or dissolved in liquid electrolytes. Complex PEC structures may ultimately enhance
207performance of solar-hydrogen devices, including light absorption, catalysis, and mass transport.^{25, 26}
208Suspensions could benefit from economic advantages associated with low-cost plastic reactors that do not

209require electrical wiring or framing required to physically support heavy electrically conductive
210substrates.²⁷

211 **2.2. Economic challenges**

212 In comparison to the technical options, the economic feasibility requirements are broader and depend
213on the ultimate application of the technology. Applications in the energy sector provide opportunity for
214the largest and most impactful implementations of solar-hydrogen technologies. The scale of these
215markets is massive (> 28,000 Terawatt-hours (TWh) per year in the US alone). In the energy share, solar-
216hydrogen technologies can be used for direct energy generation, as a fuel for transportation, or for
217temporary storage and ultimate electricity production. To date, hydrogen's direct contribution to energy
218markets is almost negligible, with hydrogen being almost exclusively produced from non-renewable
219energy sources, and small-scale uses of hydrogen that include demonstrations of grid-level energy
220storage, hydrogen fuel cell vehicles, and crude oil refining.^{28, 29} The multiple orders-of-magnitude
221difference between the current scale of the energy markets and the hydrogen market represents a clear
222opportunity for solar-hydrogen technologies. For solar-hydrogen devices to be deployable at the energy-
223market scale, however, the conditions of cost competitiveness and availability must be satisfied;
224specifically, solar-hydrogen technologies must be scalable so that collectively they have the potential to
225supply a significant fraction of the future global hydrogen needs (likely hundreds of GW) at a competitive
226price point on a "per kWh" basis. In terms of the active components of the technology, the scalability
227requirement is related to the current and projected ease of accessibility and processability of the
228materials.^{30, 31} While noble-metal catalysts that are currently implemented in state-of-the-art electrolyzers
229allow production of systems at a scale approaching GW/year, research on the development of improved
230utilization of precious metals and use of non-precious-metal electrocatalysts and low-cost light absorbers
231and ancillaries, such as transparent-conductive oxides and protective coatings, could enable production at
232larger scales.³² This is a classic trade-off between cost and efficiency; the challenge is to optimize these
233aspects to improve the desired metric (\$/kWh or \$/kg H₂). This cost metric needs to account for not only

234the cost of the device and its balance-of-system costs, but also the costs associated with the operation and
235maintenance (O&M) of the technology. O&M costs may include, for example, energy costs associated
236with feeding water to reaction sites, cleaning of the system, gas collection, compression and
237transportation to distribution centers, each which are likely to cost more in integrated systems that operate
238at low current densities and therefore occupy large areas.

239 The bottom line for cost-competitiveness in the hydrogen market (where hydrogen is used not only
240for energy purposes, but also for chemical processing such as petroleum refining and ammonia and
241methanol production) is that solar hydrogen will need to compete ultimately with hydrogen from fossil
242fuels (i.e. usually produced from methane reforming and coal gasification routes, which tend to be
243situated in close proximity to points of utilization, such as ammonia production plants, thus reducing
244transportation costs). In the broader energy markets scale, the cost of energy produced via solar-hydrogen
245routes will need to compete with energy produced from other sources, e.g. fossil, nuclear, hydroelectric,
246wind. These non-solar energy sources define the baseline cost that determines the viability of solar-
247hydrogen technologies. At early stages of technological development, smaller-scale applications may
248benefit from use of solar hydrogen when the characteristics of the technology pose an advantage over
249other technologies. Below, a series of potentially viable market opportunities where solar hydrogen could
250be impactful in the short-term (i.e. within the next 10 years) are presented, and a critical assessment of the
251requirements for inclusion in large-scale energy markets in the long-term is made. For completeness,
252“cost” includes not only the monetary value of energy, but also any other value that society assigns to the
253externalities associated with different energy production mechanisms (e.g. CO₂ emissions, nuclear
254disasters, ecological damage).³³ In anticipation of the future global energy markets, the costs of
255externalities are incompletely internalized by either energy producers or energy consumers, and instead
256the monetary value of their impact is shared over many entities that may not have been involved in the
257energy-generation process or have not derived any benefit from the energy use. Although new successful
258applications of solar-hydrogen technologies will need to stand alone without heavily relying on

259regulation, advanced energy policies could incorporate the costs of externalities via various market
260mechanisms (e.g. carbon taxes, emission limits, incentives).³⁴ In practice, this could render polluting or
261risky technologies costlier on a monetary basis than safe renewable energy technologies, such as solar
262hydrogen.

263 **2.3. Societal and political impacts**

264 In addition to technical and economic challenges, other unknown or emerging societal and political
265events will influence the deployment of solar-hydrogen technologies. Building an adequate physical
266infrastructure (e.g., pipelines, fuel stations, two-way electricity grids) could favor the deployment of
267particular new technologies, including solar hydrogen. On the other hand, events such as oil spills,
268nuclear disasters, or hydrogen explosions can change public perception and the political agenda of
269specific governments, and therefore the funding scheme. The Fukushima nuclear accident in 2011, for
270example, received intense media coverage and led to demonstrations against nuclear power in Germany.³⁵
271The growing public concern and resistance resulted in requests for more transparency and into a drastic
272change of the German national policy toward more renewable energy.^{36, 37} The awareness and perception
273of risks and advantages of a new technology can thus influence the acceptance of the public for new
274technological or infrastructural changes which are crucial for its deployment. As social studies show,
275safety and price are the main concerns for public acceptance of hydrogen technologies.³⁸ However, the
276general attitude of people towards technology and the types of information they are given also greatly
277influences their opinion about hydrogen technology.^{39, 40} In addition to public acceptance, political
278decisions can have an impact on technological development. In 1990 for example, the California Air
279Resources Board obliged major car manufacturers to bring zero emission vehicles to the market by 2003,
280which led to an increase in funding for research and development activities and pushed the development
281of new technologies in this field.⁴¹ The political agenda in several countries support emerging
282technologies via funding schemes, e.g. in large programs on renewable energy. For example, Norway will
283ban the sale of fossil fuel cars by 2025.⁴² Political and public attention around a particular topic thus help

284to mobilize research funding and relevant actors, while unfulfilled research promises can lead to a shift to
285other technological options. Hydrogen-based technologies for example have already seen major ups and
286downs in political and public attention in the past.^{43, 44} Specific to solar-hydrogen technologies is that they
287must also compete with other research activities not only in the field of renewable energy but also with
288technologies that promise to reduce energy consumption or net CO₂ emissions. The scientific community
289will likely have more influence on the opinion of policy-makers if applied research goals are focused on
290realistic research targets that can be delivered in a timely fashion and that satisfy society's evolving
291expectations. Of course, realistic research targets are mostly based on pre-existing long-term fundamental
292research products.⁴⁵ Understanding how to continue to fund fundamental research, while yielding tangible
293deliverables that have social impact, constitutes a challenge for all stakeholders in the hydrogen
294technology sector.

295

296**3. Identifying pathways for implementation of solar-hydrogen technologies**

297 A pathway for inclusion of solar-hydrogen technologies in energy markets likely requires successful
298incorporation in early-stage markets. In this section, we describe and critically assess short-term
299opportunities (≤ 10 years) for solar-hydrogen technologies and identify criteria for penetration of solar-
300hydrogen systems into large-scale energy markets in the long term, where it becomes critical for the
301technology to be socio-economically, politically, and technically beneficial.

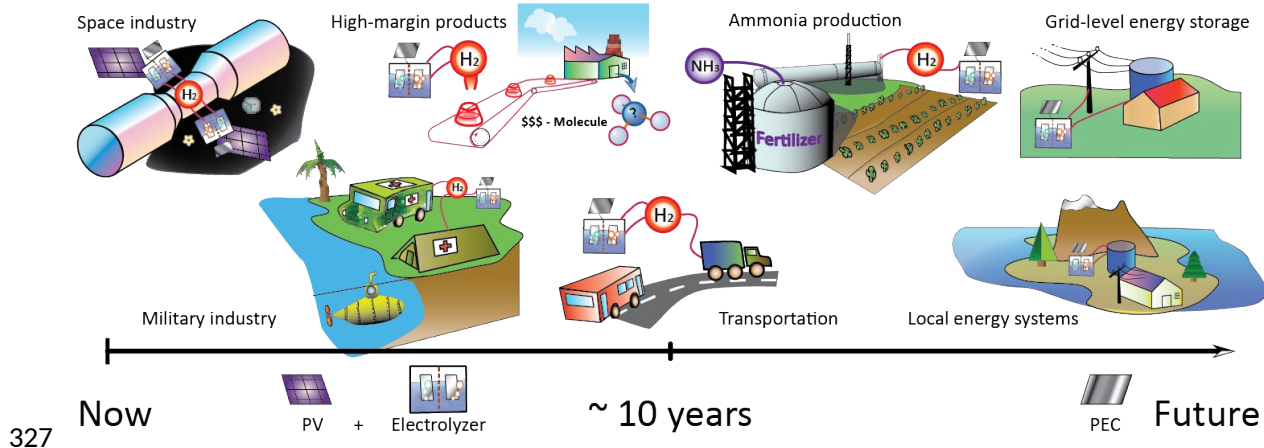
302 ***3.1. Short-term implementation (10-year timeframe)***

303 This subsection describes short-term markets and technological opportunities that could lead to
304favorable economic conditions for entry-scale implementation of solar-hydrogen technologies,
305specifically focusing on the more mature PV–Electrolysis devices.

306 *3.1.1. Market opportunities*

307 Although solar-hydrogen technologies use sunlight and water to generate hydrogen directly, under
308 current market conditions they must compete with hydrogen generated from methane reforming or from
309 grid-powered electrolysis. As long as fossil fuels remain as the predominant source of grid-level
310 electricity, hydrogen produced by either of these non-solar routes has a substantial CO₂ footprint, and
311 therefore, has clear environmental costs. Moreover, while hydrogen can be obtained inexpensively from
312 methane reforming at large-scale plants, its use in the transportation sector could be hampered by the
313 additional costs and added emissions from delivery to consumer locations. In addition, reformer-produced
314 H₂ must have carbon species (e.g., CO, CO₂, CH₄), as well as trace sulfur in natural gas, removed from the
315 reaction products at an additional cost.

316 While generating H₂ from a pure water feedstock does not require removal of carbonaceous reaction
317 products, residual water must be removed in both cases. Given these product differentiators, application
318 areas where solar-hydrogen technologies could potentially succeed in the near-term should aim at
319 exploiting *a) environmental aspects of the production processes, b) generation of hydrogen close to the*
320 *point of utilization, and c) purity of the produced hydrogen.* This would aid in the competitiveness of the
321 technology in cost-inelastic markets that require high-purity hydrogen, production (decentralized) near
322 the point of application, and with low environmental impacts that solar-based technologies can provide.
323 Broadly speaking, plausible early-stage application fields can be divided in to seven distinct areas that are
324 depicted in Figure 3: (i) grid-level energy storage, (ii) local or isolated permanent energy systems, (iii)
325 transportation, (iv) as a precursor for the production of high-margin products, (v) the military industry,
326 (vi) the space industry, and (vii) the agricultural sector.



328 **Figure 3.** Short-term (10-year timeframe) application fields that are likely to provide the most promising utilization
 329 routes. The chronological ordering of these application fields is based on projected timelines for practical
 330 implementation.

331

332 i. Grid-level energy storage: While more challenging to break into, large markets are also of interest
 333 for solar-hydrogen technologies because even small impacts would result in large installations.
 334 Grid-level energy storage applications are advantageous because distributed solar-hydrogen
 335 technologies benefit from backing by the electricity grid. Therefore, challenges due to
 336 intermittency can be mitigated, at the expense of requiring some level of AC-DC and DC-AC
 337 conversion. For this proposed application field, both photovoltaic installations and electrolyzers
 338 that are coupled to a fuel cell or are regenerative (i.e. they serve the dual role of electrolyzer and
 339 fuel cell) would be connected to the electricity grid. The most cost-effective use strategy would
 340 be to generate hydrogen during periods of high solar insolation, when electricity prices are low
 341 due to a large supply of electricity generated from sunlight, and in certain locations with very
 342 high penetration of photovoltaics or other renewables, so low that the electricity is nearly free.
 343 The hydrogen would then be temporarily stored until solar insolation is poor and other sources of
 344 renewable electricity are scarce. The low supply of clean electricity would mean that electricity
 345 prices would be dictated by baseload power and would be high. Solar hydrogen could capitalize
 346 on these electricity prices by generating electricity through reacting hydrogen and oxygen (from
 347 the air) electrochemically in a fuel cell or by combustion in a turbine. Given the current relative

348 high prices of electrolysis units and large energy losses incurred during both generation of
349 hydrogen from water and recombination of hydrogen and oxygen, grid-level energy storage
350 would be a difficult market to access and build a profitable business case.^{46, 47} Under current
351 market conditions, batteries are economically more viable for short-term energy storage due to
352 their high round-trip efficiencies. Despite their own challenges, batteries would serve in the same
353 role as hydrogen in grid-level energy storage, where, in general, most storage requirements are on
354 the scale of days.⁴⁸⁻⁵⁰ Additionally, gas peaker plants that operate on methane combustion are able
355 to rapidly adapt to different electricity production levels, and can be used to smooth intermittent
356 energy produced by solar or wind power installations both for short- and long-term energy
357 storage needs.⁵¹ In summary, the current alternatives (i.e. battery energy storage and natural gas
358 fired power generation) tend to be more cost effective than solar-hydrogen technologies and
359 therefore, it is unlikely that grid-level energy-storage solutions based on solar-hydrogen
360 technologies will be economically viable in the short-term, although even small impacts represent
361 large opportunities.

362 **ii. Local or isolated permanent energy systems:** Communities without grid access, including those
363 on small islands, could benefit from localized, independent energy systems where the
364 implementation of renewable energy sources may be advantageous. As such, solar-hydrogen
365 technologies could play a key role in these energy solutions, especially when these communities
366 or military bases receive high solar insolation. These implementations would also likely benefit
367 from a local electricity microgrid that contains photovoltaics and energy-storage systems. As
368 described above, battery economics favor short-term energy storage while electrolyzers coupled
369 to use as a fuel cell compare favorably to batteries for larger periods of storage.⁵² Unlike grid-
370 level energy storage, which is backed by enormous baseload power that can adjust to seasonal
371 variability, isolated permanent electrolysis units would serve the purpose of buffering long-term
372 fluctuations in photovoltaic output (i.e. weeks to seasons). This time frame and scale are not

373 practical for battery energy storage due to slow self-discharge that becomes significant over long
374 timescales and unit size because battery mass scales proportionally with energy needs.⁵³ The
375 distribution of batteries and hydrogen storage units would depend on seasonal fluctuations in
376 local-specific resources. For example, desert locations would require fewer electrolysis units due
377 to small seasonal fluctuations in solar insolation, while temperate regions would require larger
378 and/or more electrolysis units due to more seasonal variability in the solar resource.

379 **iii.** Transportation: In the short-term, solar-hydrogen technologies can directly impact the
380 transportation sector. Hydrogen can be mixed into natural gas pipelines to provide some of the
381 available energy during combustion, even in internal combustion engines.⁵⁴ In addition, small
382 fleets of hydrogen fuel-cell vehicles (HFCVs) recently entered the market, and they have been
383 allocated in local communities with hydrogen fueling capabilities. Early adopters of HFCVs are
384 predominantly environmentally conscious and technologically knowledgeable individuals with
385 the appropriate economical means. Currently, the vast majority of hydrogen available for fueling
386 is produced via CO₂-emitting methane reforming. This method is implemented because the cost
387 of hydrogen production from a centralized methane reforming plant, while variable, is lower than
388 via electrolysis methods. Also, large capital investments are required for compression, storage,
389 and dispensing in hydrogen fueling stations which deters the additional investment required to
390 produce renewable hydrogen locally. Nonetheless, given the low supply of hydrogen fuel, the
391 price charged at hydrogen fueling stations must be significantly higher than the cost to produce
392 and distribute hydrogen. A non-negligible subset of the population would be willing to pay a
393 premium for hydrogen from clean sources, just as a subset of the population is willing to pay for a
394 HFCV.

395 Public transportation represents a logical opportunity for implementation of HFCVs and use
396 of solar-hydrogen technologies to generate hydrogen fuel. Already some example demonstration
397 projects have been implemented in the US, Germany, Switzerland, Japan among others.⁵⁵⁻⁵⁹ These

398 projects are easier to implement than infrastructure changes required for personal HFCVs,
399 because vehicles for public transportation have predetermined and limited routes, and require
400 access to fueling stations in close proximity to their service route. Depots for public
401 transportation vehicles can even be co-located with solar-hydrogen technologies so that the solar-
402 hydrogen light absorbers can shade the vehicles from sunlight, thus keeping the vehicles cooler
403 when not in use and ultimately saving on air conditioning needs. Furthermore, public
404 transportation is often government regulated, and therefore a direct and rapid pathway to
405 implementation may exist due to pressures from clean-energy policy. For similar reasons, long-
406 distance shipping and transportation may benefit from HFCVs and solar-hydrogen technologies.

407 Nations in the process of developing their energy infrastructure represent opportunities for
408 implementation of solar-hydrogen technologies, notably for HFCV car rentals in cities of the
409 future. In these planned cities, it may make sense to locate fueling stations along the outer edge of
410 each city, where there is more space available for large area photovoltaic installations and
411 electrolyzers. In this scenario, people could use predominantly public transportation or battery-
412 electric vehicles within the confines of the city, and rental cars for longer-distance travel to places
413 outside the city, including for transportation to other cities. Car rental agencies would be located
414 on the outer edge of the city and near the fueling stations. The ability to design a city with
415 collocation of solar-hydrogen technologies (e.g. photovoltaic farms and electrolyzer plants),
416 hydrogen fueling stations, and HFCV car rental agencies at the nexus of the city and open land,
417 provides a unique opportunity for the design of synergistic infrastructure that optimizes the
418 benefits of each technology. This is common practice in chemical plant design, where collocation
419 of multiple plants that utilize equipment and use products from one plant in another process is
420 often economical. Moreover, as in the case of personal HFCVs, consumers could drive this
421 opportunity of synergistic infrastructures for solar-hydrogen technologies if tourism is a big
422 market.

423 **iv.** High-margin products: Hydrogen is a chemical feedstock widely used in the electronics, food,
424 pharmaceutical, cosmetics, lubricants, and chemical industries. For example, hydrogen is used to
425 change the rheological and sensory properties of foods through hydrogenation of unsaturated
426 fatty acids and many lipids. For many of these applications high purity hydrogen is required, with
427 no trace of the typical contaminants found in hydrogen produced by methane reforming, which is
428 a niche filled by solar hydrogen generated by electrolysis. Additionally, the cost of hydrogen in
429 the final product is often negligible, in part due to the small volumes that are required, and small
430 differences in the price of hydrogen do not affect the cost structure of these industries. Because
431 purity is the dominant factor, these high-margin products are produced most economically via
432 electrolysis. Moreover, implementing solar-hydrogen technologies in these industries will allow
433 them to market their products to environmentally conscious consumers, especially for food and
434 cosmetics. All of these characteristics of high-margin products make the short-term
435 implementation of solar-hydrogen technologies potentially viable. Other high-margin chemicals
436 include those produced on large scales in chemical plants, many of which can be made
437 electrochemically, and several of which constitute rather large markets. If instead of electrolyzing
438 water, solar-hydrogen generation could be coupled to another oxidation reaction, such as chloride
439 oxidation to chlorine gas or perchlorate salts that would increase the economic incentive to
440 produce solar hydrogen.⁶¹

441 **v.** Military industry: Military applications provide another specialized market entry point for solar-
442 hydrogen technologies. Small-scale, easily deployable, portable, and robust microgrid energy
443 systems are of interest to deployed troops in isolated locations. Larger installations could supply
444 power for grid-independent bases, which are therefore less vulnerable to cybersecurity hacks or
445 attacks on the electrical grid. Again, for remote and isolated applications, reliability, mass, and
446 volume are often more important than the cost of the technology. In addition, remote generation
447 of hydrogen is useful for powering fuel cells for aeromedical evacuations, which enable longer

448 flight times compared to those powered by batteries. Similar to use for respiration during space
449 exploration, the generation of medical grade oxygen from water splitting is also of importance for
450 military hospital installations and any people who are involved in remote projects and
451 expeditions.

452 **vi.** Space industry: Specialized applications in the space industry might also be a viable entry point
453 for solar-hydrogen technologies. The cost of devices to generate hydrogen and oxygen are of
454 minor importance, while the most important factors are reliability and the mass and volume of the
455 systems, including feedstocks. For space applications, this is because enormous amounts of fuel
456 are required to transport payloads and therefore the mass of the fuel, and oxidant for return
457 missions, dominate the cost of space missions. Onboard generation of fuel (by reaction of H₂ with
458 CO₂) and for prolonged and distant space missions (e.g. between Earth and Mars), generation of
459 an oxidant (O₂) to release the energy stored in the fuel in space and create thrust, would result in a
460 much lighter payload and therefore, a lower mission cost. For this reason, lightweight and flexible
461 designs for on-demand energy production and storage are extremely beneficial strategies.
462 Moreover, recycling water and electrolyzing it for direct onboard oxygen generation for
463 respiration is a common approach used in space applications, and driving the process with
464 sunlight affords a reliable, low-mass option for energy generation and storage. Lightweight solar
465 panels consisting of thin films of III-V materials deposited on Kapton supports are already used
466 in space applications, and lightweight designs for solar-hydrogen technologies have also recently
467 been proposed.^{16, 60} For these applications, it is even more critical that devices operate at the
468 highest possible efficiency, and that is why the highest-performing photovoltaics are preferred
469 over low-cost alternatives. In addition, the solar spectrum differs between space and earth, and
470 terrestrial size constraints for deployed devices are often relaxed for implementations in space
471 where vast regions are unoccupied, as long as the devices can be effectively bundled for delivery.

472 **vii.** Agriculture sector: More than half of the 50 million tons of hydrogen produced annually is used
473 for the production of ammonia using the Haber-Bosch process, and more than half of the
474 ammonia is used for the production of nitrogen-based fertilizers. Without these, we would not be
475 able to grow enough food to sustain a population of 7 billion people. While the massive scale of
476 the Haber-Bosch and fertilizer production processes make early-stage implementation of solar-
477 hydrogen technologies unlikely, the sheer size of this market means that even small contributions
478 from solar-hydrogen technologies will constitute substantial implementations that will further aid
479 near-term deployment.

480 While the seven sectors mentioned above represent possible entry points for implementation of solar-
481hydrogen technologies, advances in the component technologies themselves could impact other industries
482involved in the electrochemical production of alternative commodity chemicals to hydrogen (e.g.
483chloralkali, zinc production, aluminum production)⁶¹ or on electrochemical wastewater treatment.⁶² These
484industries enjoy higher margins than the energy industry and already use electrochemical methods for
485large-scale production,⁶³ which could facilitate early-stage implementation of solar-hydrogen
486technologies.

487 3.1.2. *Technological implementation*

488 The technology readiness of solar-hydrogen technologies is low; the readiness of the specific subset
489of PEC solar-hydrogen technologies is even lower. Generally, for applications where cost is a significant
490market driver, the cost of the PV–Electrolysis device would be the most important factor. Because > 90%
491of the PV market consists of solar cells made from silicon (either mono-crystalline or multi-crystalline),⁶⁴
492they are likely to be the most appropriate light absorbers to implement, although other commercially
493available light absorbers could compete with silicon based on the application. CdTe and CIGS
494photovoltaics represent a viable option which is likely to result in solar-hydrogen costs in a similar range
495to those achievable using silicon photovoltaics.⁶⁵ In most cases, PV modules based on III-V

496semiconductors are currently not economically viable for terrestrial applications, but are predominant in
497space applications where their efficiency and thin lightweight designs offset their capital cost. There are
498also active research programs aimed at lowering the cost of III-V solar cells and PEC devices while
499maintaining their conversion efficiency, thus enabling their use in conventional flat-plate and low-
500concentration applications.⁶⁶⁻⁶⁸

501 In terms of electrolysis technologies likely to be implemented in the short-term there are two
502prominent commercial options: alkaline electrolyzers and proton-exchange membrane (PEM)
503electrolyzers. Despite the fact that solid oxide electrolyzers are not discussed in this article, the
504conclusions and discussion also generally apply to this class of water-splitting devices.

505 *Liquid electrolyte alkaline electrolyzers* have been deployed commercially for more than 100 years.⁶⁹
506⁷⁰ Because of this, they have already been developed and implemented on larger scales than PEM
507electrolyzers, but they require additional attention and safety considerations due to the use of a strongly
508corrosive *liquid* alkaline electrolyte and the need for tightly balanced pressures of H₂ and O₂. Alkaline
509electrolyzers also tend to be less efficient than the acidic PEM electrolyzers at a given current density.
510This is due to the larger overpotential required for the alkaline-stable Ni-based electrocatalysts for
511hydrogen evolution and the larger ohmic losses caused by the lower conductivity of the electrolyte and
512the larger inter-electrode gap. Alkaline electrolyzers are also less amenable to changes in their operation
513conditions, because they usually implement porous separators between the electrodes with higher gas
514permeability and hence high crossover rates. Contrarily, PEM electrolyzers implement highly selective
515gas-separating ion-exchange membranes.

516 *PEM electrolyzers* are the state-of-the-art for most small-scale hydrogen generation applications.
517They implement ion-conducting polymer membranes as *solid* acid electrolytes that are selective for
518cations, allowing proton transport from the site of water oxidation to the site of hydrogen generation. Use
519of a solid electrolyte and liquid deionized water as a feedstock is much less of a safety concern than the

520corrosive liquid electrolytes needed in alkaline electrolyzers. Yet, because PEM electrocatalysts are in
521direct contact with the solid electrolyte membrane, which is acidic and corrosive, the only efficient
522catalyst materials that remain bound and stable are those based on noble metals (e.g. Pt and IrO_x are the
523state-of-the-art). While the terrestrial scarcity of noble metals could preclude the implementation of PEM
524electrolyzers on large TW scales, their implementation at early stages on GW scales is not expected to be
525limited by the availability of specific raw materials. In comparison to alkaline electrolyzers, PEM
526electrolyzers are in many ways more amenable to PV–Electrolysis devices. The use of state-of-the-art
527electrocatalysts in PEM electrolyzers allow for more efficient operation. Moreover, PEM electrolyzers
528operate more effectively under conditions of fluctuating power input, particularly when intermittent solar
529insolation drives electrolysis consistently outputting a pressurized hydrogen product (up to 30 bar).⁷¹
530While PEM electrolyzers do have significant technical advantages over alkaline electrolyzers, they still
531tend to be more costly (currently costing ~1.2 USD/W)⁷² partly because of lower production volumes and
532limited system sizes, with the largest planned systems being on the order of several MW.^{73, 74} As their
533production volumes increase, it is likely that their costs will continue to decrease due to economies of
534scale and technological advances.

535 3.1.3. *Science and technology opportunities*

536 There are significant challenges for the implementation of PV–Electrolysis devices, mainly arising
537from complications caused by the PV-driven intermittent use of electrolyzers. These challenges can at
538least in part be mitigated using today’s electrolyzer technologies if electronic buffering mechanisms are in
539place to maintain operation above a threshold and therefore avoid large amounts of gas crossover and
540formation of explosive gas mixtures.¹² Buffering approaches include incorporation of an array of batteries
541or capacitors, or utilization of grid electricity, where available. An alternative to buffering is removal of
542the hydrogen and oxygen reaction products from the reaction chambers during periods of slow operation,
543for example, by flushing the system with water, or to implement other engineering approaches to avoid
544the formation of explosive gas mixtures.⁷⁵ Additionally, electrical circuits of photovoltaic arrays and AC-

545 driven peripheral components (e.g. pumps, fans and control systems) could be re-designed to directly
546 drive water electrolyzers without the need for power electronics (i.e. maximum power trackers or DC–DC
547 converters).^{16, 76} If electricity buffers, product removal, and power electronics could be avoided, a scenario
548 that seems reasonable within the next decade, solar-hydrogen technologies will be simplified, therefore
549 ensuring smooth operation and ultimately driving down their cost.

550 **3.2. Long-term deployment in energy markets**

551 The opportunities identified in the short term could help solar-hydrogen technologies enter energy
552 markets and build the foundation for more widespread implementation in the long term. This subsection
553 first describes societal and policy changes, as well as technological opportunities that could lead to
554 favorable economic conditions for larger-scale implementation of solar-hydrogen technologies. Long-
555 term pathways for both PV–Electrolysis and PEC devices are discussed.

556 *3.2.1. Societal, economic, and policy changes and drivers*

557 Environmental challenges associated with burning large quantities of fossil fuels to generate energy
558 have triggered a strong interest in implementation of renewable-energy systems.^{77, 78} As a testimony to
559 this, the number of energy-conversion installations driven by sunlight or wind has experienced
560 exponential growth over the past decade. In the case of solar energy, this growth is directly apparent from
561 the enormous increase in the production capacity of photovoltaics, which has resulted in significant
562 reductions in their cost.⁷⁹ On the production side, government incentives facilitated this market increase
563 by providing strong investment that led to the rapid increase in production. An increase in demand was
564 propelled by policy drivers that aimed to curtail use of non-renewable energy sources. For example,
565 China, India, and even smaller size countries all have policies to promote renewable energy technologies.
566 Further policy drivers such as controls on CO₂ emission as well as incentives for clean-energy
567 technologies will help increase penetration of renewables into the energy markets and raise awareness for
568 the need to realize accessible, reliable and affordable supply of energy. The Paris Climate Agreement

569helped set the stage for this development.⁸⁰ The Dutch government, for example, targets 40% renewable
570energy by 2030 and a > 80% reduction in CO₂ emissions by 2050.⁸¹ Societal aspects can also trigger the
571large-scale adoption of clean energy technologies. Changes to the environment, violent and more frequent
572natural disasters, and local pollution can favor the adoption of clean technologies on the basis of world
573energy and global transportation scenarios created by the World Energy Council.⁸² Additionally,
574investment in education and in accessible and accurate information regarding environmental effects of
575various energy sources can help shape society's perceptions of the energy markets. Ultimately, these
576changes in public perception can decisively lead to the enactment of long-lasting clean energy policies.⁸³
577⁸⁴

578 Changes in energy markets can also favor clean technologies. Market failures in the gas and oil sector
579(e.g. drop in demand, decrease in production, curtailments) can lead to spikes in energy prices, therefore
580indirectly improving the economic viability of alternative renewable-energy sources. Additionally, market
581and ecological factors could lead to the collapse of large-scale fossil fuel suppliers, therefore necessitating
582the development and broad deployment of clean-energy technologies.^{85, 86} To date, the growth of the
583photovoltaic sector has been facilitated by the ability to integrate solar-energy-conversion devices into our
584current electricity transmission and distribution infrastructure. A larger penetration of photovoltaics into
585the energy markets will result in changes in the operation of the electricity grid. Energy storage
586mechanisms will have to be implemented to bridge the time gap between production periods and
587consumer demands. Under conditions of direct storage and use, an electricity grid may not even be
588required. This will further motivate the decoupling of photovoltaic installations from the grid, favoring
589options like centralized solar-hydrogen facilities for the production of transportation fuels and for long-
590term energy storage needs. Similarly, as outdated and unreliable grid structures continue to age, new
591energy-efficient systems such as microgrids emerge, which are in general more compatible with
592renewable technologies over traditional large-scale power plants.^{87, 88} Moreover, as government incentives

593for PV phase out, soft costs must continue to decrease to keep PV competitive with fossil sources of
594electricity.

595 3.2.2. *Science and technology opportunities*

596 In the long-term, solar hydrogen generated by both PV–Electrolysis and PEC routes could play a
597significant role in the energy market. The socio-economic and policy drivers mentioned above would
598facilitate the use of solar-hydrogen technologies as a competitive energy-storage option. At the same time,
599significant scientific and technological barriers need to be overcome in order for the technologies to
600succeed in a highly competitive market. Despite some demonstrations of functioning devices, the long-
601term stable operation of efficient and cost-effective devices has not yet been proven for PEC routes.
602Possible technology development pathways are presented below for the two families of devices that, if
603successful, could lead to viable solar-hydrogen systems.

604 3.3. *Pathways for PV–Electrolysis*

605 To a large extent, PV–Electrolysis advances can be commercialized by independently optimizing each
606of the constituent components,¹⁰ i.e. the PV module, the cell stack materials, and the electrolyzer design.
607However, the ultimate goal of a practical system coupling the two components must be kept in mind
608while performing this independent optimization. Although at a first glance this statement might seem
609obvious and non-constraining, there is a significant number of peripheral components (mainly power
610electronics) that are incorporated into PV installations and electrolysis units to couple their operation with
611the electrical grid. These components account for a non-trivial fraction of the overall capital costs of the
612equipment, and furthermore poor integration will result in efficiency decreases on the order of at least
61310%, with ~5% losses on each of the two AC/DC conversion steps, and even larger losses at low power
614output. While under some circumstances PV–Electrolysis will operate in conjunction with the grid to
615maximize the utilization of the electrolyzer unit, lean alternatives with fewer peripheral components and a
616more integrated operation will likely be preferred as the technology progresses and electrolyzers become

617more capable of operating with fluctuating loads. This integrated PV–Electrolysis approach would not
618require that power electronics be incorporated in current electrolyzers systems, as PV arrays may be
619designed to directly power electrolyzers units with the appropriate DC characteristics. The reduced
620balance-of-system costs of integrated PV–Electrolysis devices and the higher efficiencies achievable due
621to short transmission distances could favor their implementation in the long term, assuming that no new
622durability challenges emerge during intermittent or fluctuating operation.^{5, 7, 8} In the short term the value
623proposition of on-site or wastewater-derived solar-hydrogen generation can be realized in niche markets.
624Those gains would need to compensate for the economic losses from the low utilization of the
625electrolyzer units if powered exclusively with solar energy.

626 In the photovoltaic space, it is likely that silicon will continue to be the most promising technology in
627the short to medium term (< 30 years). Laboratory-based examples of silicon PVs directly coupled to
628electrolyzers have demonstrated efficiencies for hydrogen production in excess of 14%.⁷⁶ Following a
629pathway of reasonable improvements, silicon PVs could be implemented in solar-hydrogen devices to
630attain efficiencies of up to 18%. These advances involve improvements in surface passivation of Si,
631introduction of back contacting techniques in the cell fabrication, and small improvements in the quality
632of the crystalline silicon solar cells. Achieving even higher efficiencies using single silicon PVs would be
633difficult. On the cost side, only small reductions are expected from silicon manufacturing, as the prices
634have already decreased significantly (currently at < USD 0.5/W) and gains from economies of scale will
635saturate. Alternative materials for PVs including cadmium telluride, copper indium gallium selenide
636(CIGS), hybrid organic–inorganic halide perovskites, III-V semiconductors, or tandem architectures could
637be disruptive to the PV space.^{16, 89} However, currently they are significantly disadvantaged with respect to
638Si PVs.^{65, 90} There are many factors that limit the practicality of each alternative PV material, such as
639stability, toxicity, efficiency, durability, but ultimately each of these technologies suffers from the same
640limiting factor for large-scale viability: economic competitiveness. Advances that improve PV scalability,
641cost, stability, and performance for these materials classes will be needed before they have a significant

642impact on solar-hydrogen devices. Lastly, inexpensive optical concentration or light management
643schemes and heat and mass transfer optimizations that enhance efficiency and materials utilization of PV–
644Electrolysis over PV or electrolyzers alone, could improve the viability of PV–Electrolysis.

645 Although the contribution of the electrolyzer to the projected costs of a PV–Electrolysis system is
646minor, an improved efficiency of this component means that less PV cells are needed to produce the same
647amount of hydrogen, so that the hydrogen can become significantly cheaper. While the PV industry has
648grown aggressively in the recent past, and current yearly installation levels approach a 85 GW capacity,⁹¹
649the electrolyzer industry lags behind in terms of installations by more than two orders-of-magnitude. The
650production scale of the electrolysis industry will need to approach levels comparable to the PV sector, and
651as this happens, significant cost gains for both technologies are expected. Porous transport layers and
652bipolar plates are important from cost, stability, and efficiency perspectives. Their optimization enables
653higher current densities and lower catalyst loadings. Active component improvements in performance and
654stability (catalysts layers and membranes) are also needed. In particular, as the scale of production
655increases, it will be important to develop earth-abundant electrocatalysts with comparable performance to
656the noble-metal electrocatalysts used in current PEM electrolyzers. In addition to standard cation-
657exchange-membrane-based electrolyzers, membrane-free systems have seen significant advances due to
658their tolerance for impurities in water feedstock and potentially lower upfront capital costs.⁹²⁻⁹⁵ Moreover,
659the development of anion-exchange membranes can enable implementation of alkaline polymer–
660electrolyte–membrane electrolyzers that use high-performing and earth-abundant Ni-based catalysts.^{96, 97}
661These membranes must exhibit long-term stability and avoid excessive gas crossover even at lower
662sunlight-driven rates.

663 In addition to economies of scale, cost reductions in electrolyzers may arise from lowering the capital
664cost requirements of the system (currently at ~1/3 of the total cost), or by reducing costs associated with
665the electricity feedstock required for their operation. Solar-to-hydrogen efficiency improvements will
666directly affect electricity feedstock expenses, as less electricity will be needed for a given rate of solar-

667hydrogen production. Important sources of efficiency improvements in current PEM electrolyzers may
668come from reduction of ionic resistances in membranes, improvement in electrocatalyst activity, and
669mitigation of mass transport limitations in catalyst and porous transport layers.⁹⁸ If the efficiency
670improvements lead to larger operating current densities, electrolyzer units could be designed with smaller
671footprints for a given production level, thus reducing their capital costs. Additionally, the feedstock costs
672could be reduced if the electrical grid is circumvented in a direct PV–Electrolysis configuration. In this
673configuration, the costs associated with electricity transmission and distribution through the grid would be
674eliminated. Opportunities exist for defining application-specific guidelines for membranes used for direct
675PV–Electrolysis. Research and development of membranes for direct PV–Electrolysis configurations
676include identifying those with lower gas permeability and optimal ion-transport and mechanical
677properties, information on the molecular and morphological characteristics of membranes during mass
678transport processes, and ion-conducting membranes that can operate under intermittent electrolysis
679conditions. These fundamental science developments can lead to advances in the long term that ultimately
680may brighten the economic prospects of PV–Electrolysis technologies.

681 **3.4. Pathways for PEC**

682 Even if all the advancements in component performance and cost of coupled PV–Electrolysis systems
683are achieved, the nature of their design will require significant cost reduction of the auxiliary components
684in order for them to be cost-competitive with other hydrogen production pathways. This is similar to the
685case of current PV plants where the cost of the PV does not dominate system cost. Such cost reductions
686might not even be possible given the inherent system architecture of coupled PV–Electrolysis systems.
687For this reason, PEC systems could provide an opportunity for this necessary cost reduction, given that
688their design can be completely different than PV–Electrolysis systems and therefore could lead to
689disruptive and significant cost reduction. Opening up the design space to a broader set of architectures can
690only have a positive impact on the potential to identify a cost-optimal option. One example is systems
691based on photocatalyst particles.^{4, 99, 100} However, to date, large-scale deployment of PEC-based solar-

692hydrogen technologies appears to be disadvantaged with respect to PV–Electrolysis approaches. PEC
693devices are significantly less developed, and their efficiencies are generally worse than for coupled PV–
694Electrolysis devices.¹⁰¹ Moreover, they suffer from poor stability due to the requirement of light absorbing
695materials to be in contact or close proximity with often caustic electrolytes. Despite great efforts to
696develop protection strategies, this challenge remains largely unsolved and precludes deployment of PEC
697technologies.¹⁰¹ One important development challenge is the scale: for PEC devices to reach the same rate
698of H₂ output as PV–Electrolysis technologies the projected electrochemically active H₂ production area
699would have to be at least ~50 times larger.^{5, 14} These large electrochemical areas would lead to significant
700challenges in product handling due to the low current density at the photoelectrode surface, but could
701result in higher operating efficiencies and less stringent catalytic requirements. Enabling large-scale
702efficient PEC devices requires advances in materials durability and the ability to control at the atomic-
703level reproducible material engineering across macroscopic areas.¹⁰² From a topological viewpoint, PEC
704devices are a subset of PV–Electrolysis devices where the electrocatalytic components are collocated with
705the light absorbers, and in fact can then be the same material. However, viable implementation pathways
706for PEC architectures will require the discovery of a PEC system that can perform solar water-splitting at
707a cost per kg of H₂ that is equal to or lower than available PV–Electrolysis systems, and as a consequence,
708PEC devices cannot be based on components that could also be used to fabricate a PV–Electrolysis device
709with equivalent or higher economic benefits. If this goal is not achieved, long-term solar-hydrogen
710technologies will tend toward PV–Electrolysis architectures. In a PV–Electrolysis configuration, each of
711the device components (e.g. light-absorber and water-splitting units) can be independently engineered so
712that the overall device is optimized, often with the aid of power electronics. Furthermore, there are
713significant fundamental advantages of decoupling the light-absorption and water-splitting functions in
714solar-hydrogen devices, which arise from increased flexibility in device design, optimization, and
715operation. For example, in a PEC configuration, the light absorbers will require innovative electrode
716designs to minimize shading due to optical absorption and scattering by the catalysts and to facilitate gas

717 evolution and mitigate occlusion of electrocatalytic sites, for example, due to evolved bubbles that can
718 block mass transfer and light incidence.¹⁰³

719 It has been argued that economic benefits for PEC devices arise from the component integration
720 aspects of light absorbers with electrolysis technologies, no peripheral electronics, the possibility of
721 achieving higher efficiencies when the reactions take place at semiconductor–liquid junctions due to
722 fewer ohmic losses, and the ease of forming a high-quality junction.¹⁰¹ While the first two potential
723 advantages have not been demonstrated, there are several additional advances that could facilitate
724 realizing them. Understanding at a fundamental level the interfacial interactions between light absorbers,
725 electrocatalysts, and electrolytes might lead to improved solar-to-hydrogen efficiencies and better
726 stability. Also, continuing to use chemical engineering principles to develop design rules and
727 demonstrations of integrated devices and solar-hydrogen production plants would provide realistic
728 prospects on the economic and environmental viability of PEC approaches.^{8, 26, 104-112} Furthermore,
729 developing engineering solutions for the mass-production of promising PEC materials will be needed to
730 achieve large-scale hydrogen production.¹¹³ Specifically, to the case of so-called photocatalyst particle-
731 based PEC devices, selective catalysis approaches will need to be developed to preferentially drive the
732 water-splitting reaction,^{114, 115} while avoiding undesirable recombination reactions of the products.^{100, 116} In
733 addition, avoiding the formation of explosive hydrogen streams will require development of new
734 separation materials and engineering schemes, including flow-cell designs that introduce improved
735 mechanisms of gas separation and collection,^{104, 117} especially over large areas.

736

737 **4. Conclusions and perspectives**

738 This article presented a broad perspective on pathways for the implementation of solar-hydrogen
739 technologies. Several niche market opportunities were identified for solar hydrogen implementation on
740 the short-term (≤ 10 years). In this time frame, it is anticipated that PV–Electrolysis systems will be the

741only approach that could be implemented for such applications and still be economical. In the long term,
742solar-hydrogen technologies could be deployed more broadly in the energy markets. For that to happen,
743hydrogen produced via solar routes might need to be competitive against other energy carriers, such as
744fossil fuels. This is a daunting challenge, as the cost of energy from fossil sources has been historically
745low, even though extremely volatile, and it suggests that hydrogen production costs today would need to
746sum to less than \$2/kg hydrogen.¹¹⁸ Despite the scale of the challenge, solar-hydrogen technologies
747provide a promising path to clean alternative fuels, and if externalities from fossil fuel utilization were
748internalized, the prospects for hydrogen fuel implementation would be greatly enhanced. Implementing
749PV–Electrolysis units manufactured using currently available commercial devices would lead to costs of
750hydrogen that exceed this value by at least a factor of three.⁷ Therefore, achieving that cost target with
751PV–electrolysis devices would require significant technology advances, cost reductions, and possibly also
752political/policy measures, such as a CO₂ tax. Currently, one high-impact research focus is to advance
753electrolysis that is directly driven by PV installations. Under this mode of operation, electrolyzers will
754need to accommodate the natural intermittency of solar irradiation, in a stable way over lifetimes
755comparable to current PV technologies. This approach would result in significant capital cost reductions
756due to elimination of power electronics required in existing systems, and would increase overall
757efficiency; at the expense of a reduced capacity factor of the electrolyzer. Important long-term goals
758include the ability to operate PV–Electrolysis devices using inexpensive and efficient electrocatalysts.
759This will require the development of new catalytic materials that are stable under acidic electrolytes or
760anion-exchange membranes with significantly improved stability. PEC routes present even more
761significant challenges but have a significantly more disruptive potential. For a PEC system to be
762implemented, it would have to perform at least equally as well as available PV–Electrolysis alternative
763systems on economic grounds. Additionally, if the components used for the fabrication of such a PEC
764device could be utilized in a PV–Electrolysis arrangement, the integrated PEC architecture would need to
765be economically preferable to an alternative PV–Electrolysis arrangement and also show advantages in
766terms of sustainability even while it is less flexible in design, optimization, and operation. Understanding

767fundamental science aspects and developing reactor engineering design guidelines can help to achieve
768these goals.

769 Even if the scientific community achieves all of the advances in PV–Electrolysis or PEC devices
770outlined in this report, it is uncertain whether solar-hydrogen technologies will be competitive in large-
771scale energy markets in the long term. This will depend on a variety of factors that include, but are not
772limited to, system efficiencies, materials cost, balance-of-system costs, lifetime, externalities, social
773acceptance, and price of energy or hydrogen from alternative sources. The possible impact of some of
774these factors have been described in more detail in recent DOE reports.¹¹⁹ Economic policy mechanisms
775to account for the environmental effects of CO₂ emissions can help facilitate this prospect. As a
776worldwide community, we should emphasize the development of CO₂-free, sustainable energy
777technologies at comparable cost than today’s CO₂-heavy alternatives. While scientific curiosity should
778never be hindered by economic considerations, cost can and should be considered at a stage when more
779applied research programs or policy decisions need to be designed. There has been tremendous progress
780in the fundamental understanding of solar-hydrogen systems in the past decades and the interdisciplinary
781knowledge accumulated can be implemented in new electrochemical processes, wastewater treatment, or
782applications for which the purity or sustainability of the hydrogen is more important than the price, with
783greater prospects for profitability, sustainability, and societal impact. The creativity of the scientific
784community and its ability to pivot into new promising application areas will have a decisive effect on the
785future societal and environmental impacts of solar-hydrogen technologies.

786

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