Solar Fuels from Concentrated Sunlight

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Executive Summary

Sunlight is by far the most abundant carbon-neutral energy resource on earth. However, solar energy is intermittent and does not necessarily match the variations in demand. If it is to become a major contributor to our energy supply, some form of storage is necessary.

Conversion of solar energy into chemical fuels is an attractive method of solar energy storage. Solar fuels, such as hydrogen, can be used for upgrading fossil fuels, burned to generate heat, further processed into electrical or mechanical work by turbines and generators or internal combustion engines, or used directly to generate electricity in fuel cells and batteries to meet energy demands whenever and wherever required by the customers. The challenge is to produce large amounts of chemical fuels directly from sunlight in robust, cost-effective ways while minimizing the adverse effects on the environment.

The success of solar thermal power generation – known as 'concentrating solar power' (CSP) – is already moving towards sustainable, large scale fuel production: concentrating solar radiation with reflecting mirrors provides high temperature process heat for driving efficient thermochemical processes. Although the technical feasibility of various technologies has been demonstrated, commercialization of these processes has been hindered by the economics. Nevertheless, solar fuels are among the most promising technologies to curb the growing demand for fossil fuels and to mitigate the effects of climate change. To achieve this it is recommended that commercial implementation steadily evolves, starting from the current state-of-the-art fossil fuel production technologies. To facilitate the introduction of new solar fuels production processes, the existing know-how from both the fuel production industry and the CSP research institutes should be merged. At a later stage, the emerging solar fuel technologies will be based on processes that are completely independent of any fossil fuel resources.

The main vector for this transformation will be the production of hydrogen, a potentially clean alternative to fossil fuels, especially for use in transport. Today, however, more than 90% of hydrogen is produced by using high temperature processes from fossil resources, mainly natural gas. If hydrogen is generated from solar energy, it is a completely clean technology; no hazardous wastes or climate changing byproducts are formed and only sunshine and water are required as inputs to the process. This is the vision outlined in the European Commission's 'European hydrogen and fuel cell roadmap', which runs up to 2050.

Solar fuels production

There are basically three routes, alone or in combination, for producing storable and transportable fuels from solar energy. The *electrochemical* route uses solar electricity made from photovoltaic or concentrating solar thermal systems followed by an electrolytic process; the *photochemical/photobiological* route makes direct use of solar photon energy for photochemical and photobiological processes; the *thermochemical* route uses solar heat at high temperatures followed by an endothermic thermochemical process.

The thermochemical route offers some intriguing thermodynamic advantages with direct economic implications.

Concentrating solar technologies

The state-of-the-art CSP technology capable of achieving high process temperatures is the 'solar tower' configuration, where a field of

heliostats (tracking mirrors) focuses the sun-rays onto a solar receiver mounted on top of a centrally located tower. Such solar concentrating systems already operate in large-scale pilot and commercial plants, such as the 11 MW$_{el}$ PS10 plant near Seville in Spain, in operation since 2007 to deliver solar generated electricity to the grid. They make use of a heat transfer fluid (typically air, water, synthetic oil, helium, sodium, or molten salt) that is heated by solar energy and then used in traditional steam or gas turbines. The typical land area required for a 50 MW$_{th}$ plant is about 300,000 m^2.

Solar thermochemical applications, although not as far advanced as solar thermal electricity generation, employ similar solar concentrating technologies. However, high-temperature thermochemical processes require higher solar concentration than CSP plants, which has an impact on heliostat field layout and plant operation.

For efficient development of solar fuels production technologies, in particular solar hydrogen, dedicated concentrating research facilities – solar furnaces and solar simulators – are employed. The largest solar furnaces currently operating in France and Uzbekistan have a thermal power of 1 MW.

Solar hydrogen

Clean hydrogen production will be based on water and energy from renewable sources. Replacing fossil fuels with renewable energy will shift the balance between electricity and fuels. For many applications, electricity will be used instead of fuels, but two major applications will require a massive production of solar hydrogen. Firstly, renewable energy must be stored for balanced use, and secondly, mobility will probably be based on fuels rather than electricity.

The European Union's World Energy Technology Outlook scenario predicts a hydrogen demand equivalent to about 1 billion tons of oil in 2050.

Solar electricity generated by CSP technology, and followed by electrolysis of water, is a viable technical route for producing hydrogen. It can be considered as a benchmark for other routes, such as solar-driven water-splitting thermochemical cycles that offer the potential of energy efficient large-scale production of hydrogen. The projected costs of hydrogen produced by CSP and electrolysis, assuming solar thermal electricity costs of 0.08 $/kWh$_{el}$, range from 0.15-0.20 $/kWh, or 6-8 $/kg H$_2$.

There are also a range of thermochemical routes to solar production of hydrogen. All of these involve energy consuming (endothermic) reactions that make use of concentrated solar radiation as the source of high temperature process heat.

Economic solar hydrogen

The economical competitiveness of solar fuel production is closely related to two factors: the cost of fossil fuels and the necessity to control the world climate by drastically reducing CO$_2$ emissions.

Both the US Department of Energy and the European Commission have a clear vision of the hydrogen economy, with firm targets for hydrogen production costs. The US target for 2017 is 3 $/gge (gasoline gallon equivalent; 1 gge is about 1 kg H$_2$), and the EU target for 2020 is 3.50 €/kg H$_2$.

The economics of large scale solar hydrogen production has been assessed in numerous studies, which indicate that the solar thermochemical production of hydrogen can be competitive compared with the electrolysis of water using solar-generated electricity. It can even become competitive with conventional fossil-fuel-based processes at current fuel prices, especially if credits for CO$_2$ mitigation and pollution avoidance are applied.

Further R&D and large-scale demonstrations are therefore justified. This would have positive effects on achievable efficiencies and investment cost reduction for materials and components. An important factor will be the massive installation of commercial solar thermal power plants, in particular power towers, since heliostats will be one of the most expensive components of a solar thermal hydrogen production plant.

Recommended strategy

A range of research activities are already under way, with the ultimate goal of developing technically and economically viable technologies for solar thermochemical processes that can produce solar fuels, particularly hydrogen. Implementation should start immediately to accelerate the transition from today's fossil-fuel-based economy to tomorrow's solar driven hydrogen economy.

The EU-FP6 project INNOHYP-CA (2004-2006) has already developed a roadmap, which shows the pathway to implementing thermochemical processes for massive hydrogen production.

The future for solar fuels

Solar energy is free, abundant and inexhaustible, but at least two crucial steps are necessary for a successful market introduction of solar fuels. Firstly, solar chemical production technologies must be further developed and proven to be technically feasible and economical. Secondly, a worldwide consensus on the most promising future energy carriers – both renewable electricity and hydrogen – needs to be reached. The arguments in favor of a future hydrogen economy are excellent, and the political commitment to move in this direction has been manifested in many initiatives. What is urgently needed now is a clear decision to start the transition from fossil to renewable energies and from gasoline to hydrogen.

We encourage politicians and policy-makers, energy officials and regulators, utility companies, development banks and private investors to firmly support the massive production of solar fuels – primarily hydrogen – by taking concrete steps to enable future infrastructure and market development without delay.

We have only a short time window of opportunity to tackle and solve the critical problems of greenhouse gas emission and climate change. Solar fuels are part of the solution – they have the capacity to help satisfy the energy needs of the world without destroying it.

Intermittent Solar Energy – Need for Storage

CSP
Concentrated Solar Power plants produce electric power by converting the sun's radiative energy into high-temperature heat using various mirror configurations.

Sunlight is by far the most abundant carbon-neutral energy resource on earth. More energy from the sun strikes the earth's surface in one hour than is consumed annually by all of the fossil fuels. However, solar energy is intermittent and does not necessarily match the variable daily and seasonable demands for energy. If solar energy is to become a major contributor to our energy supply, some form of energy storage is necessary for use during times when there is little or no sunlight. The growth in worldwide usage of solar energy will be constrained until reliable and low-cost technologies for storing solar energy become readily available. A viable solar energy conversion scheme must result in a 10–50 fold decrease in the cost-to-efficiency ratio for the production of stored fuels and must be stable and robust for a 20–30 year period.

Why solar fuels?
Conversion of solar energy into chemical fuels is an attractive method of solar energy storage (figure 1). Solar fuels, such as hydrogen, can be used for upgrading fossil fuels, burned to generate heat, further processed into electrical or mechanical work by turbines and generators or internal combustion engines, or used directly to generate electricity in fuel cells and batteries to meet energy demands whenever and wherever required by the customers. The challenge is to produce large amounts of chemical fuels directly from sunlight in robust, cost-effective ways to deal with growing energy demands while minimizing the adverse effects on the economy and environment.

Sustainable, large scale fuel production relies on the success of solar thermal power generation (→CSP)[1]: concentrating the incident solar radiation (figure 2) with reflecting mirrors provides high temperature process heat for driving efficient thermochemical processes in compact centralized plants. Although the technical feasi-
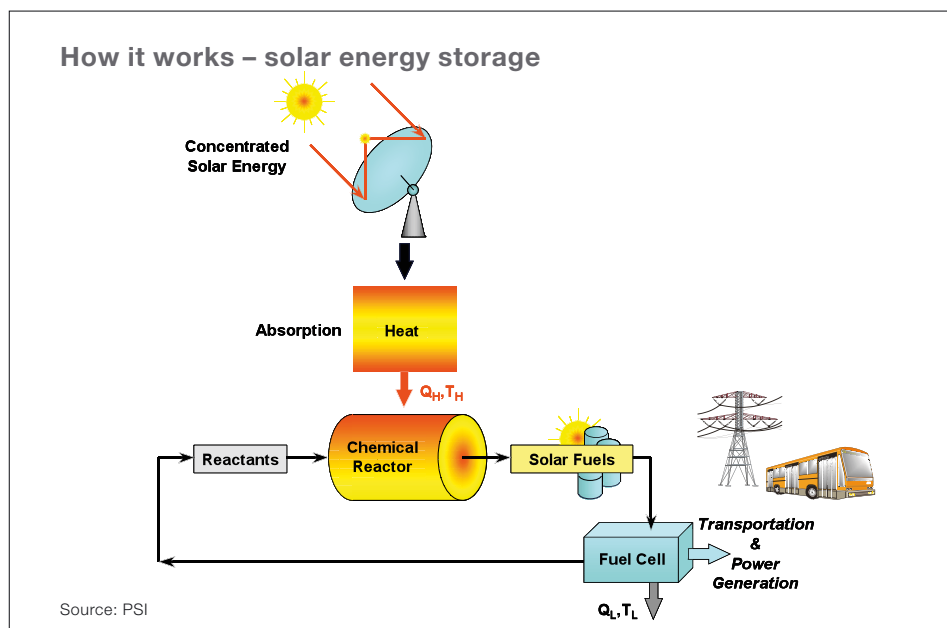
How it works – solar energy storage

Source: PSI

Figure 1: Energy conversion into solar fuels – **Concentrated solar radiation is used as the energy source of high-temperature process heat for driving thermochemical reactions towards the production of storable and transportable fuels.**

[1] Concentrating Solar Thermal Power – Now! Brochure published by Greenpeace, ESTIA, IEA SolarPACES, 2005.

GHG
Greenhouse gas emissions result in global warming, climate change and connected extreme weather events. CO_2 is the most prominent GHG but others like methane have much higher impact.

Solar thermochemical process
Any endothermic chemical process that uses concentrated solar energy as the source of high-temperature process heat.

Solar cavity receiver
A well-insulated enclosure – with a small opening to let in concentrated solar energy – approaching a blackbody absorber in its ability to capture solar energy.

Exergy efficiency
Efficiency for converting solar energy into chemical energy, given by the ratio of the maximum work that may be extracted from a solar fuel to the solar energy input for producing such a fuel by a solar thermochemical process.

Carnot efficiency
Maximum efficiency of a cyclic process for converting heat from a high-temperature thermal reservoir at T_H into work and rejecting heat to a low-temperature thermal reservoir at T_L, given by $\eta_{Carnot} = 1 - T_L/T_H$.

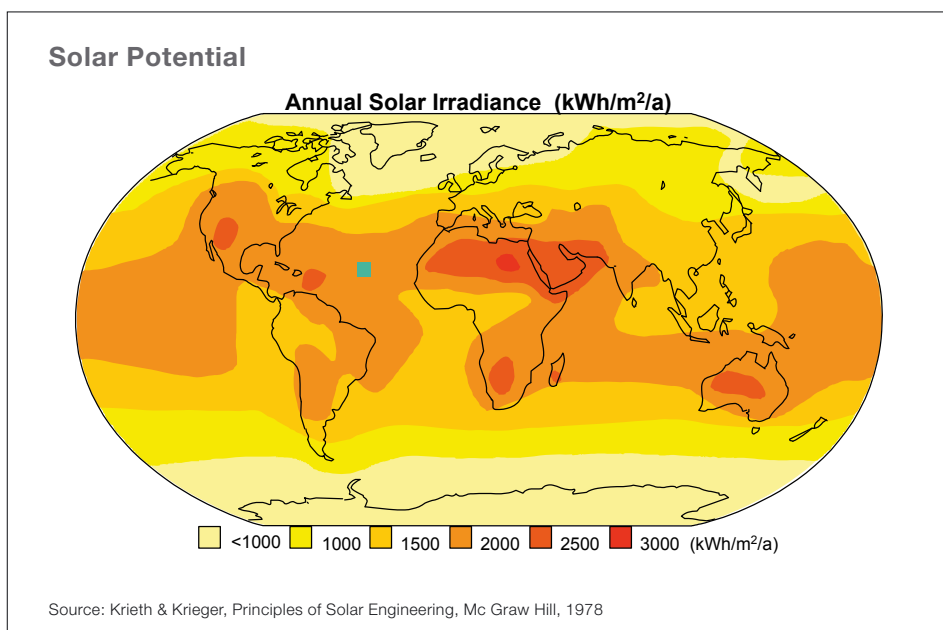Source: Krieth & Krieger, Principles of Solar Engineering, Mc Graw Hill, 1978

Figure 2: Annual solar irradiance on Earth – **The square of 1000 x 1000 km^2 plotted in the middle of the Atlantic Ocean stands for the land area required for supplying the world's energy needs with concentrated solar energy.**

bility of various technologies has been demonstrated in the past, commercialization of these processes had been hindered – predominantly due to economical reasons.

Nevertheless, solar fuels are among the most promising technologies to curb the growing demand for fossil fuels – associated with soaring prices for diminishing fossil fuel resources – and to mitigate the effects of climate change. Their rapid implementation will create new markets for developing countries and increase the energy security, due to greater independence in fuel production and a larger number of countries supplying solar fuels.

It is recommended that commercial implementation steadily evolves, starting from the current state-of-the-art fossil fuel production technologies. To facilitate the introduction of new solar fuels production processes, the existing know-how from both the fuel production industry and the CSP research institutes should be merged. At a later stage, the emerging solar fuel technologies will be based on processes that are completely independent of any fossil fuel resources.

Why solar hydrogen?
Many believe that over the next several decades, there will be a shift away from today's fossil fuel economy toward a much cleaner hydrogen future. The enormous problems accompanied with a fossil fuel economy and the significant environmental advantages of the Hydrogen Economy (see box) are strong drivers toward clean hydrogen (H_2) production, supply and uti-

> **Hydrogen Economy – political initiatives**
>
> - International Partnership for the Hydrogen Economy (IPHE)
> - Hydrogen Implementing Agreement of the International Energy Agency (IEA-HIA)
> - Hydrogen program of the US DOE
> - European Hydrogen and fuel Cell Platform (HFP)
> - European Hydrogen and Fuel Cell Joint Technology Initiative (JTI).

lization, as manifested in a number of political initiatives. One striking vision is outlined in the European hydrogen and fuel cell roadmap (figure 3), which runs up to 2050.[2]

Although H_2 is widely accepted as the energy carrier of the future, it must still fulfill the requirements of a sustainable energy economy, i.e. it must be produced from unlimited energy sources, without →GHG emission, at an affordable price.

Today, more than 90% of the H_2 is produced from fossil resources, mainly natural gas (NG). Half of the annual production of more than 100 million tons is used for producing fertilizers and about 45% for petro-chemical processes. The latter application alone will drastically rise in the next years because an increasing amount of H_2 is needed for refining heavy oils.

[2] European Commission, EUR 20719 EN – Hydrogen Energy and Fuel Cells – A vision of our future, Luxembourg: Office for Official Publications of the European Communities, 2003 – 36 pp., ISBN 92-894-5589-6.
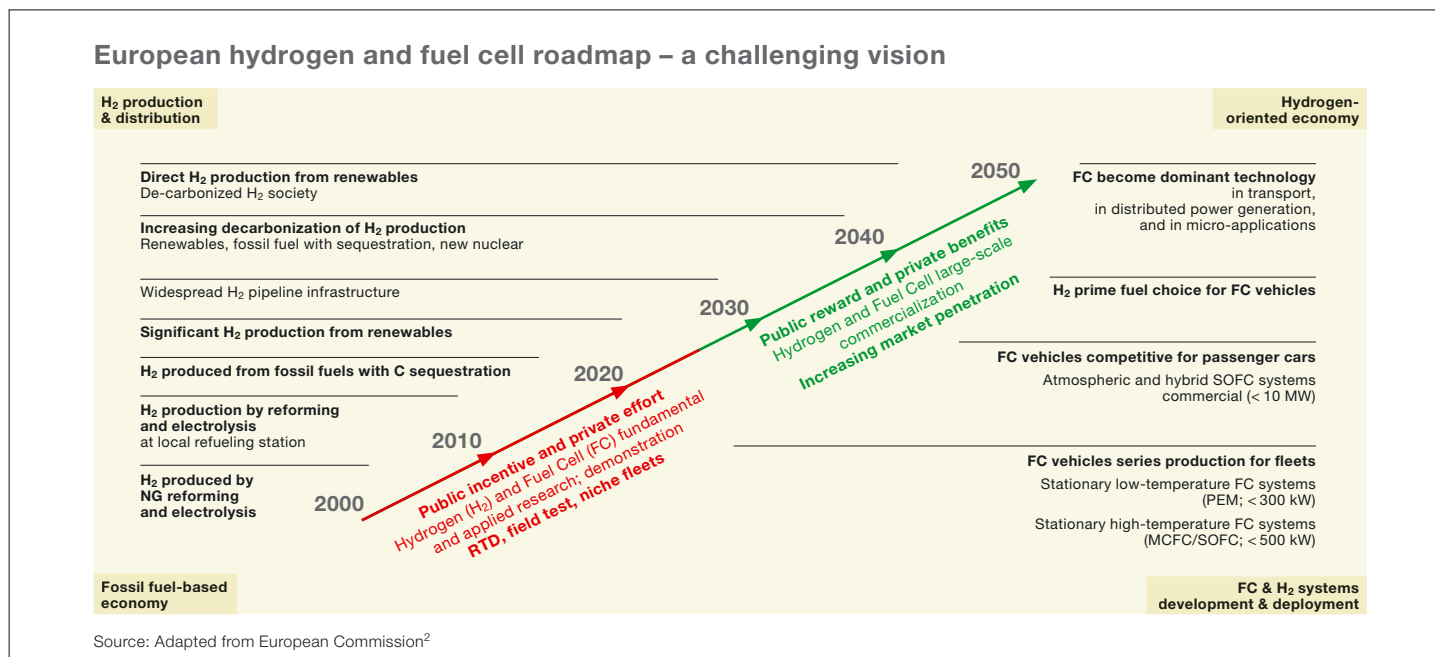
European hydrogen and fuel cell roadmap – a challenging vision

Source: Adapted from European Commission[2]

Figure 3: From fossil fuel-based to hydrogen-oriented economy – **Today, H$_2$ is produced by reforming of natural gas and electrolysis. In 2020, a significant amount of H$_2$ will be produced from renewables, and increasing decarbonization of H$_2$ production is expected for 2040. In a decarbonized H$_2$ society around 2050, H$_2$ will be directly produced from renewables.**

These demands can potentially be fulfilled by high temperature processes, which are able to provide large amounts of H$_2$ in centralized plants. Many think that H$_2$ produced using solar energy will provide the long-term solution for solar energy storage. In fact, if H$_2$ is generated from solar energy, it is a completely clean technology; no hazardous wastes or climate changing byproducts are formed and only sunshine and water are required.

A lot of research is currently being undertaken around the world, because solar H$_2$ seems to have the highest technical and economical potential for successful market introduction. However, since other technologies for energy storage are available – such as carbon dioxide (CO$_2$) or metal oxides – the pros and cons of all of those alternatives should be carefully evaluated.

Solar fuels production

The conversion of solar energy into solar fuels opens up numerous possibilities. There are basically three routes that can be used alone or in combination for producing storable and transportable fuels from solar energy:

- **Electrochemical:** solar electricity made from photovoltaic or concentrating solar thermal systems followed by an electrolytic process;
- **Photochemical/Photobiological:** direct use of solar photon energy for photochemical and photobiological processes;
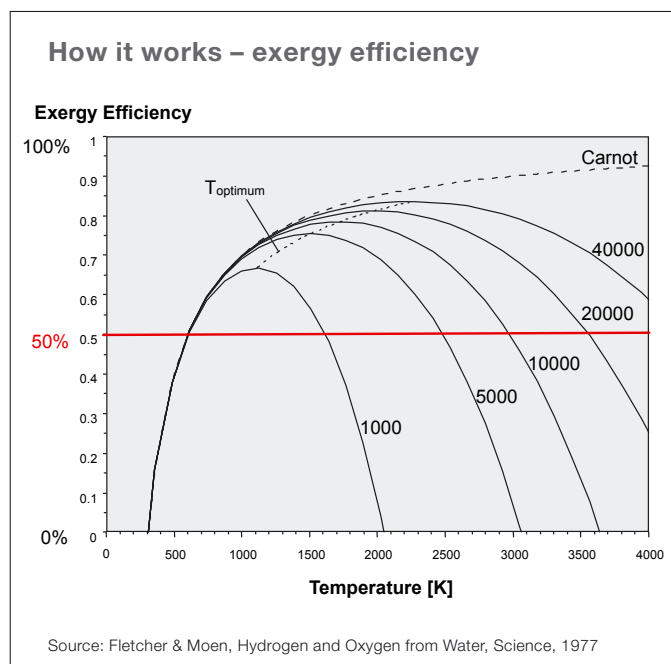- **Thermochemical:** solar heat at high temperatures followed by an endothermic →thermochemical process.

How it works – exergy efficiency

Source: Fletcher & Moen, Hydrogen and Oxygen from Water, Science, 1977

Figure 4: Exergy efficiency – **Variation of the ideal exergy efficiency as a function of the process operating temperature for a blackbody cavity-receiver converting concentrated solar energy into chemical energy. The mean solar flux concentration is the parameter (given in units of 1 sun = 1 kW/m^2): 1000; 5000; …; 40,000 suns. Also plotted are the →Carnot efficiency and the locus of the optimum cavity temperature T$_{optimum}$.**

The thermochemical route offers some intriguing thermodynamic advantages with direct economic implications. Irrespective of the type of fuel produced, higher reaction temperatures yield higher energy conversion efficiencies. However, higher temperatures also lead to greater losses by re-radiation from the →solar cavity-receiver.

Exergy efficiency – The measure of how well solar energy is converted into chemical energy stored in solar fuels is called →exergy efficiency (figure 4). The thermochemical route offers the potential of reaching exergy efficiencies exceeding 50%, higher than those obtained by all alternative routes. The higher the exergy efficiency, the lower the required power (solar collection area) for producing a given amount of solar fuel and, consequently, lower costs for the solar concentrating system, which usually correspond to half of the total investments of the entire solar chemical plant. Thus, high exergy efficiencies imply favorable economic competitiveness.

In Focus – Solar Concentrating Technologies

Solar concentration ratio
Non-dimensional ratio of the solar flux intensity – e.g., given in "suns" (1 sun = 1 kW/m^2) – achieved after concentration to the normal insolation of incident beam.

Solar tower facilities
The state-of-the-art solar concentrating technology for large-scale solar energy collection at high →solar concentration ratio is based on the solar tower optical configuration using a field of heliostats (two-axis tracking parabolic or flat mirrors) that focus the sun rays onto a solar receiver mounted on top of a centrally located tower. Solar tower systems (figure 5) – also referred to as central receiver systems – may have either a circular field of heliostats with a cantered receiver on top of the tower, as in the case of (a) the Solar Two power plant at Barstow, USA, or an asymmetric field, like the south facing test facilites at (b) the Plataforma Solar de Almería (PSA), Spain, and (c) Targassonne,

Solar tower facilities – state of the art
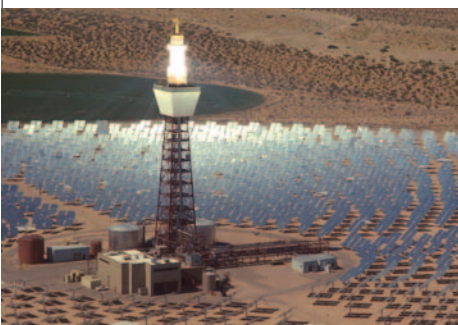
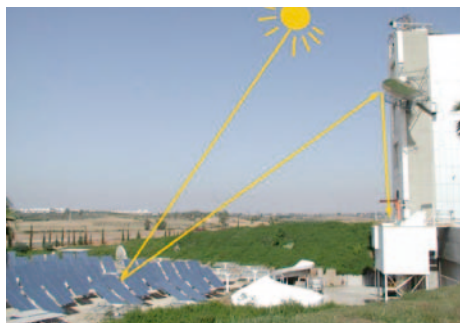(a) 60 MW$_{th}$ solar tower, Barstow, USA (Solar Two)

(b) 7 MW$_{th}$ solar tower, PSA, Almería, Spain (CESA 1)

(c) 4.5 MW$_{th}$ solar tower, Targassonne, France (THEMIS)

(d) 500 kW$_{th}$ solar tower, CSIRO, Newcastle, Australia

(e) 3 MW$_{th}$ solar tower with "beam down" optics, WIS, Rehovot, Israel

Figure 5: Solar towers – **Examples of concentrating solar research facilities based on the tower concept.**

CPC
Compound Parabolic Concentrator, a secondary optical element intended to augment the concentration of incoming solar radiation, e.g. on top of a solar tower facility.

Solar furnace
Two main configurations are used:
(a) On-axis: heliostat, parabolic concentrator, and experimental platform are positioned in one line.
(b) Off-axis: facetted mirrors with varying focal lengths reflect the solar radiation to the side, where the solar receiver-reactor is mounted in the focal plane.

HFSS
High-Flux Solar Simulators (HFSS) in operation:
(a) 150 kW_{el} HFSS consisting of 10 Xenon arc lamps with common focus at Paul Scherrer Institute (PSI), Villigen, Switzerland.
(b) 60 kW_{el} HFSS using 10 IR radiators at German Aerospace Center (DLR), Cologne, Germany.

France, or (d) CSIRO's north facing test facility in Newcastle, Australia. A novel optical configuration – called "beam-down" – developed at (e) WIS in Israel for the tower system makes use of a hyperboloidal reflector at the top of the tower to re-direct sunlight to a secondary concentrator and a receiver-reactor located at ground level.

Secondary concentrators – The solar flux concentration ratio typically obtained by solar tower systems is between 1000–1500 suns (1 sun = 1 kW/m^2). In principle, such concentrating solar devices can reach temperatures exceeding 1700°C, with optimal cavity temperatures of around 1000°C (figure 4). To some extent, the flux concentration can be further augmented with the help of a secondary concentrator (figure 6), such as a compound parabolic concentrator (→CPC), which is positioned in tandem with the primary parabolic concentrating system. Higher flux concentrations have the advantage of lower heat losses from smaller aperture areas and higher attainable temperatures in the cavity receiver. The disadvantage of such solar concentrating op-

Secondary concentrator (CPC)

a — Source: DLR

b — Source: WIS

Figure 6: CPC – **a) Multiple CPC mounted in front of a solar receiver on the CESA 1 solar tower at PSA, Spain; b) CPC for "beam down" application at WIS, Israel.**

First commercial solar tower power plants

Source: Abengoa Solar

Figure 7: Commercial electricity production using solar tower concept – **The 11 MW_{el} PS10 plant (back) is in operation since 2007, and the 20 MW_{el} PS20 plant (front) is in operation since 2009, both at Sanlúcar La Mayor near Seville, Spain.**

tics is that their smaller acceptance angle – which requires a longer and less efficient heliostat field – results in optimum temperatures closer to 1000°C rather than to 2000°C for efficientl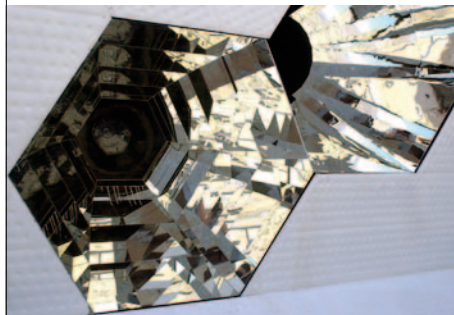y operating a solar tower system on an annual basis. The solar concentrating systems have proven to be technically feasible in large-scale pilot and commercial plants aimed at producing electricity (figure 7). They make use of a working fluid (typically air, water, synthetic oil, helium, sodium, or molten salt) that is heated by solar energy and then used in traditional steam or gas turbines. The typical land area required for a 50 MW_{th} solar tower plant is about 300,000 m^2, with a land use factor of 0.25 (fraction of reflective mirror area).

Solar thermochemical applications, although not as far advanced as solar thermal electricity generation, employ the same solar concentrating technologies. However, the heliostat field layout may look different since high-temperature thermochemical processes require a higher performance in solar concentration than solar thermal power plants. This also has an impact on investment costs for the concentrating solar facilities and their operational strategies.

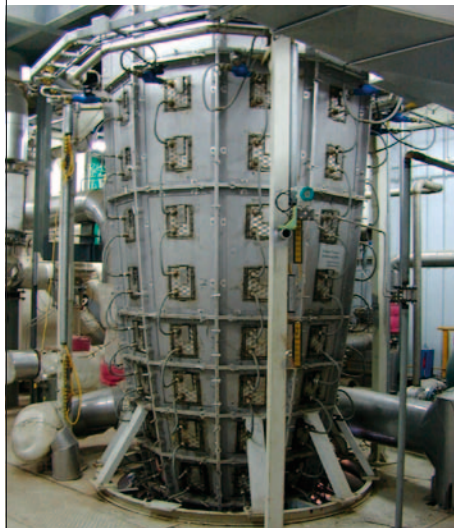**Concentrating solar research facilities**
All over the world, research groups are developing the scientific and engineering know-how for large scale production of solar fuels, in particular solar hydrogen. For these state-of-the-art R&D efforts, dedicated concentrating research facilities – solar furnaces and solar simulators – are employed (figure 8).

Solar furnace – Solar furnaces receive the solar radiation by one or more heliostats like solar towers. But instead of a tower with a central re-

Solar research infrastructure worldwide – state of the art

20 kW$_{th}$ HFSS at DLR, Cologne (D)

50 kW$_{th}$ HFSS at PSI, Villigen (CH)

25 kW$_{th}$ off-axis solar furnace
at DLR, Cologne (D)

40 kW$_{th}$ on-axis solar furnace
at PSI, Villigen (CH)

1 MW$_{th}$ solar furnace
at CNRS-PROMES, Odeillo (F)

40 kW$_{th}$ on-axis solar furnace
at KIER, Daejeon (KOR)

60 kW$_{th}$ on-axis solar furnace
at Plataforma Solar de Almería (E)

500 kW$_{th}$ solar tower
at CSIRO, Newcastle (AUS)

25 kW$_{th}$ off-axis solar furnace
at NREL, Golden, CO (USA)

3 MW$_{th}$ solar tower with "beam down"
optics at WIS, Rehovot (IL)

1 MW$_{th}$ solar furnace
at Parkent (Uzbekistan)

16 kW$_{th}$ on-axis solar furnace
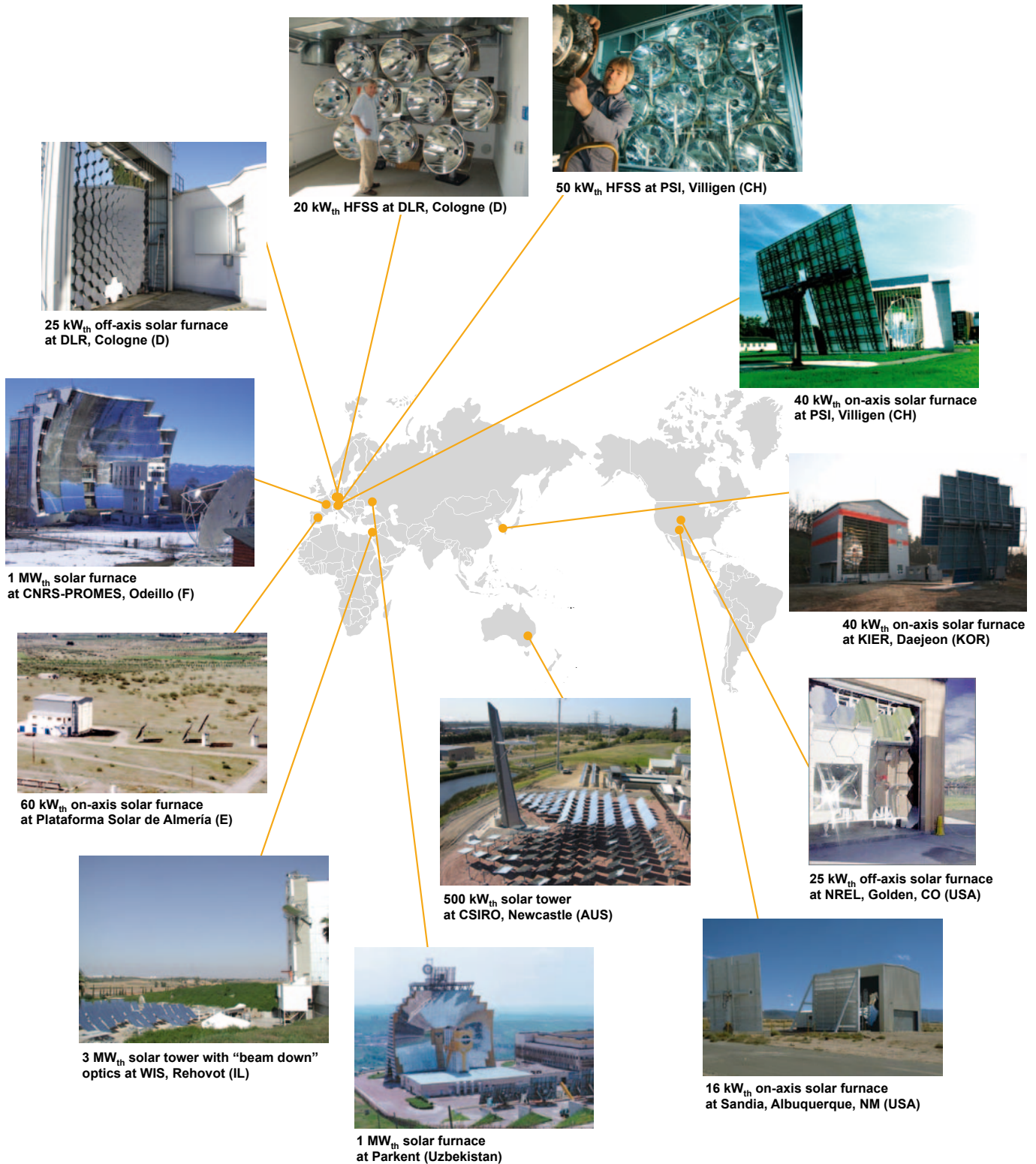at Sandia, Albuquerque, NM (USA)

Figure 8: Concentrating solar research facilities – **High-flux solar furnaces (HFSF) and simulators (HFSS), and solar towers.**

ceiver, a parabolic mirror is used as a secondary optical component to concentrate the sunlight up to 20,000 times. The parabolic mirror may be facetted because single mirrors are restricted in their size. The largest solar furnaces in Odeillo, France, and Parkent, Uzbekistan, have a thermal power of 1 MW and a size equivalent to a 12-story building. The standard geometrical configuration of a →solar furnace consists of a heliostat, parabolic concentrator, and experimental platform in one line and, therefore, is called 'on-axis' (figure 9). Usually, the reactor is mounted on a platform and then raised up to the focus of the parabolic mirror, thereby, shadowing part of the concentrator. To avoid shadowing of the concentrator, facetted mirrors with varying focal lengths can be used to reflect the solar radiation to the side. This so-called 'off-axis' concept has the advantage of easier operation but is more complex and, therefore, more expensive.

Solar simulator – In contrast to solar furnaces, which depend on varying weather conditions and deal with problems of intermittent solar irradiation and a moving radiation source, high-flux solar simulators (→HFSS) offer more stable and reproducible operating conditions (figure 10). Experiments requiring long durations can be performed easily without the problem of daytime-only operation. For efficient development of solar fuel production processes, however, both concentrating research facilities are extremely important and, because of their different characteristics, neither one can be substituted for the other.
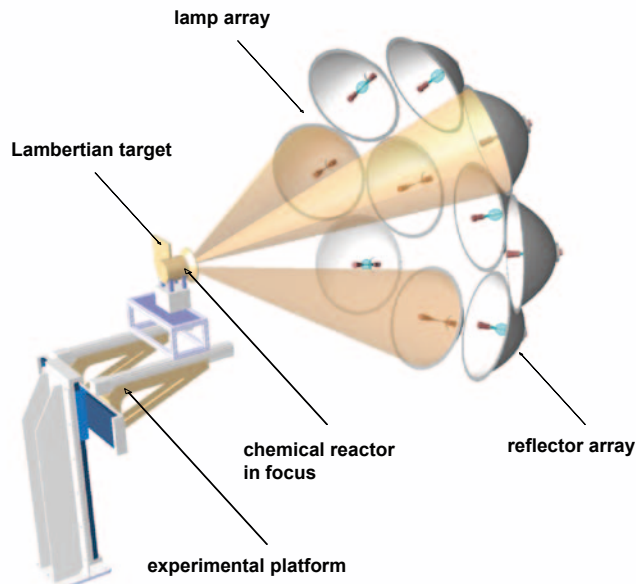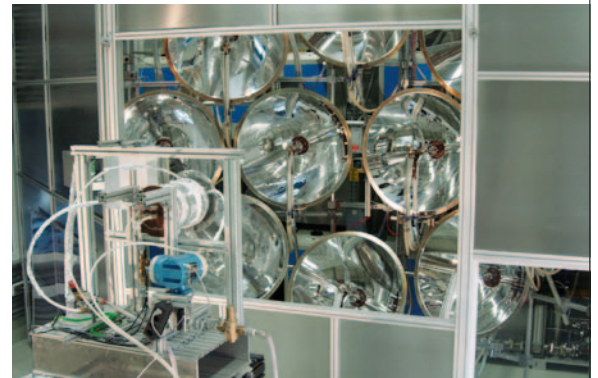
How it works – solar furnace
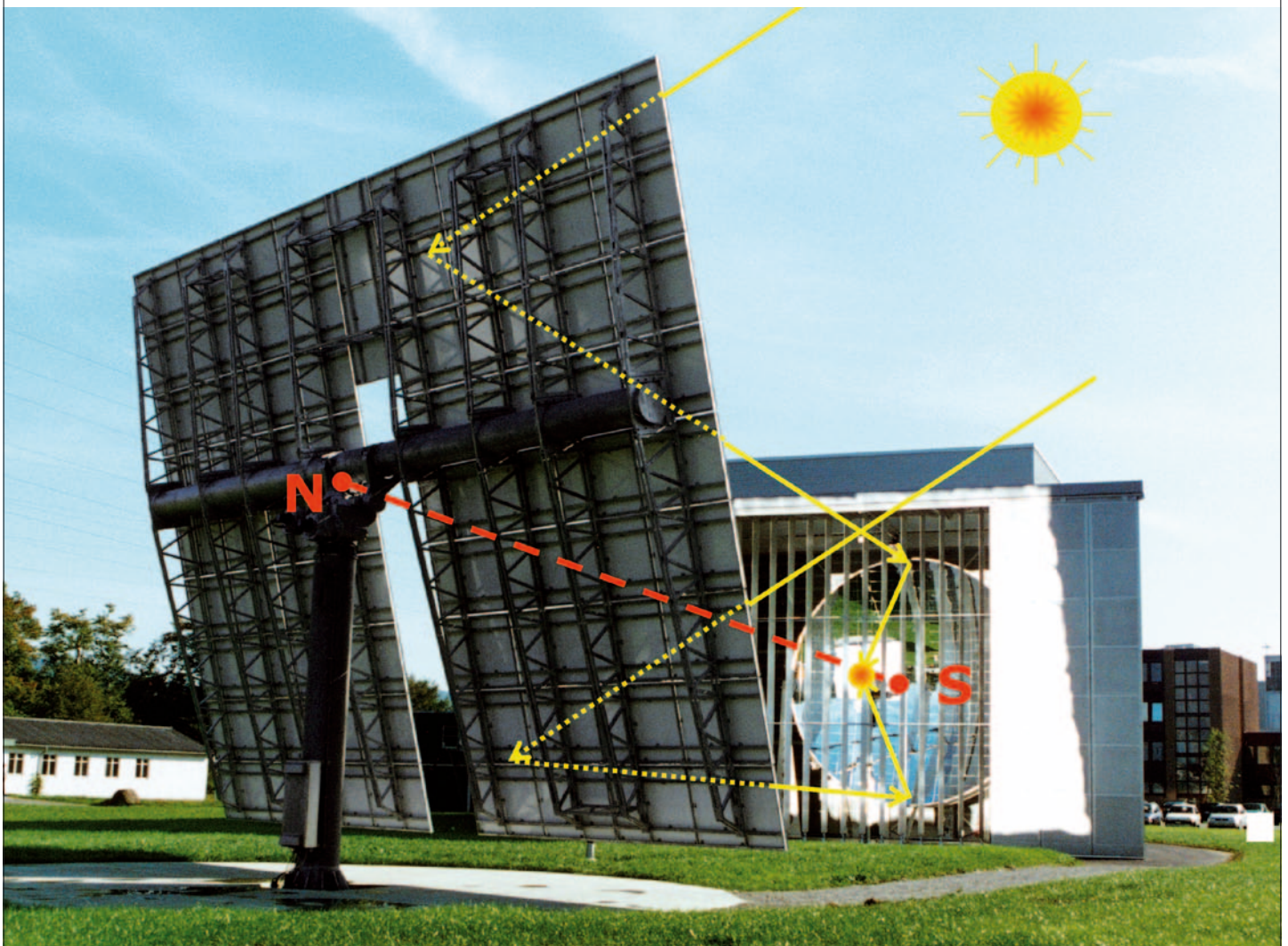
Source: PSI

Figure 9: Solar furnace – **PSI's High-Flux Solar Furnace (HFSF) consists of a 120 m² sun-tracking flat heliostat on-axis with an 8.5 m-diameter paraboloidal concentrator. It delivers up to 40 kW at peak concentration ratios exceeding 5000 suns. The solar flux intensity can be further augmented to up to 10,000 suns by using CPC secondary concentrators. A Venetian blind type shutter located between the heliostat and the concentrator controls the power input to the reactor.**

How it works – solar simulator

lamp array

Lambertian target

chemical reactor
in focus

reflector array

experimental platform

Source: PSI

Radiation flux measurement

(4) data processing

(3) CCD camera,
optical filter

image
acquisition

(5) intensity
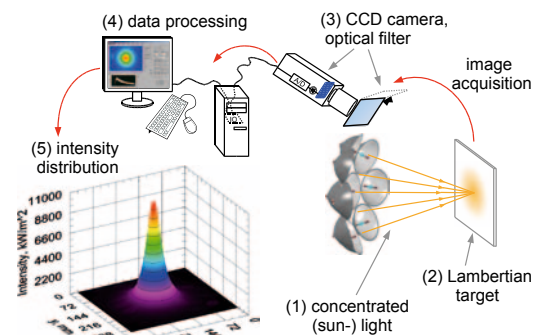distribution

(2) Lambertian
target

(1) concentrated
(sun-) light

Figure 10: Solar simulator – **PSI's High-Flux Solar Simulator (HFSS) comprises an array of ten 15 kW$_{el}$ high-pressure Xenon arcs, each closed-coupled with truncated ellipsoidal specular reflectors of common focus. Each Xe-arc can be switched on and off individually, allowing for adjustment of the radiative power input into the chemical reactor. The radiation flux distribution in the focal plane is measured optically by recording the image on a Lambertian target, acquired by a fast CCD camera equipped with optical filters. This facility is able to deliver a total radiative power of 50 kW with a peak radiative flux exceeding 10,000 suns. The total radiative power intercepted by a 6-cm diameter circular target – representing the reactor's aperture – is 20 kW, and the average radiative power flux over this aperture is more than 7,000 suns. Such high radiation fluxes correspond to stagnation blackbody temperatures exceeding 3300 K. Further coupling with a tandem 3D-CPC may augment the power flux concentration by approximately 90%. Thus, PSI's HFSS features the world's highest performance level of combined radiative power and power flux.**

Solar Hydrogen – Future Energy Carrier

Electrolysis of water
Decomposition of water (H_2O) into oxygen (O_2) and hydrogen gas (H2) due to an electric current being passed through the water.

Electrolyzer
An apparatus to separate chemically bonded elements and compounds by passing an electric current through them.

LHV
The lower heating value of a fuel is defined as the amount of heat released by combusting a specified quantity (initially at 25°C) and returning the temperature of the combustion products to 150°C, which assumes the latent heat of vaporization of water in the reaction products is not recovered.

Hydrogen is envisioned as a future energy carrier. The production routes require chemical and energy sources that are truly sustainable only if they offer closed material cycles that do not produce CO_2. Clean H_2 production from water (H_2O) and energy from renewable sources will become the paradigm of the hydrogen economy concept. Replacing fossil fuels with renewable energy will shift the balance between electricity and fuels. For many applications, electricity will be used instead of fuels, but two major applications will require a massive production of solar H_2. Firstly, renewable energy must be stored for balanced use, and, secondly, mobility will probably be based on fuels rather than electricity. Therefore even in rather conservative studies, the drastic increase in demand for electricity will be coupled with a rising demand for H_2. This development is starting slowly now, but various scenarios indicate that H_2 will become more important after 2030. The WETO-H_2 scenario predicts a H_2 demand equivalent to about 1 billion tons of oil in 2050 (figure 11).

Solar production of hydrogen
Solar electricity – generated via photovoltaics (PV) or concentrating thermal power (CSP) – followed by →electrolysis of water, is a viable technical route for producing H_2. Today, it can be considered as a benchmark for other routes such as solar-driven water-splitting thermochemical cycles that offer the potential of energy efficient large-scale production of H_2 (figure 12). With solar electricity from PV and solar thermal at efficiencies of around 15%
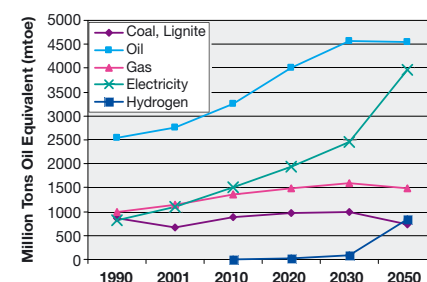
Source: WETO-H_2, 2006

Figure 11: Global energy carrier demand – **The WETO (World Energy Technology Outlook) study[1] by the European Union is a projection of the world energy system in 2050 with emphasis on the hydrogen economy. After 2030, the increasing demand for electricity will be coupled with a rising demand for H_2.**

and 20%, respectively, and →electrolyzers at 80% efficiency, the overall solar-to-hydrogen energy conversion efficiency ranges between 12% and 16%. Projected costs of H_2, assuming solar thermal electricity costs of 0.08 $/kWh$_{el}$, range from 0.15–0.20 $/kWh (→LHV of H_2), i.e. from 6–8 $/kg H_2. For PV electricity, costs are expected to be twice as high. The electricity demand for electrolysis can be significantly reduced if the electrolysis of water proceeds at

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[1] World Energy Technology Outlook – 2050, Luxembourg: Office for Official Publications of the European Communities, 2006, ISBN 92-79-01636-9, online: http://ec.europa.eu/research/energy/pdf/weto-h2_en.pdf

SOEC
A Solid Oxide Electrolyzer Cell is able to split water at high temperatures up to 900°C with a higher efficiency than conventional steam (alkaline) electrolysis near room temperature. This technology currently under development is one promising candidate for efficient high temperature hydrogen production.

Effusion
Flow of gas through an orifice whose diameter is small as compared with the distance between molecules of the gas.

Quench
Rapid cooling – for example used as gas separation technique to prevent recombination of product gases.

VHTR
The Very High Temperature Reactor is one of six designs under discussion for the next generation of nuclear reactors. Recently, a pebble bed nuclear reactor was demonstrated at the research center Jülich, Germany.

Sulfur-iodine cycle
3-step cycle based on the thermal decomposition of sulfuric acid (H_2SO_4) at 850°C. Mainly developed by General Atomics (GA) and demonstrated as an electrically heated laboratory plant at Japan Atomic Energy Agency in 2004.

UT-3 cycle
4-step cycle based on the hydrolysis of calcium and iron bromide ($CaBr_2$ and $FeBr_2$) at 750°C and 600°C, respectively. University of Tokyo (UT), Japan.

Redox reaction
Reduction/oxidation reactions describe all chemical reactions in which atoms have their oxidation number (oxidation state) changed.

Aerosol-flow reactor
The aerosol flow reactor method is a simple and efficient one-step process that can directly produce particles within a desirable particle size range with consistent and controlled properties.
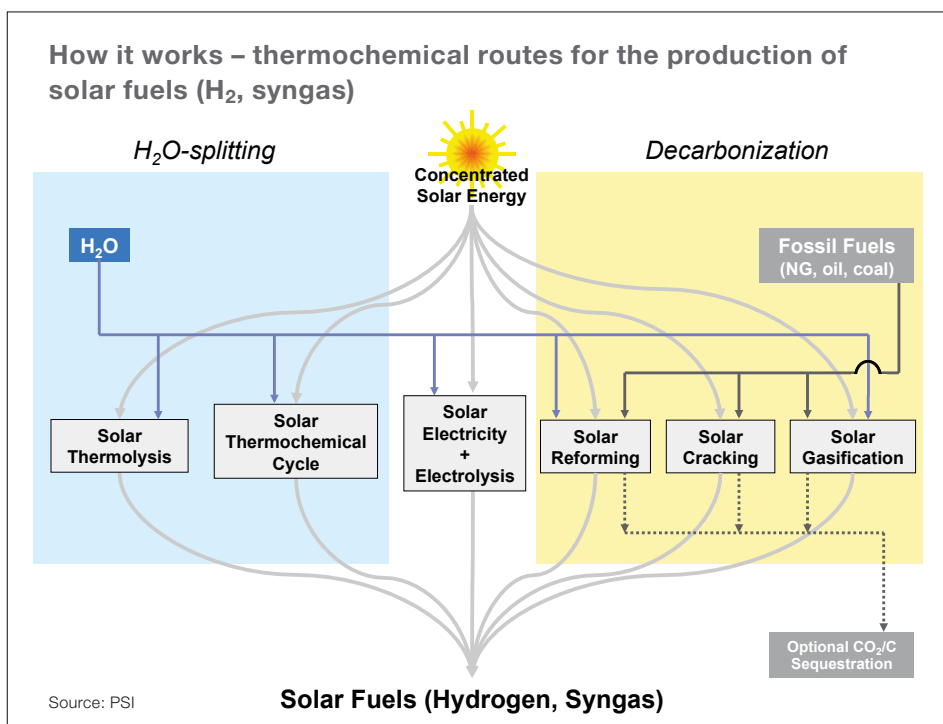

Figure 12: Thermochemical routes for solar hydrogen production – **Indicated is the chemical source of H_2: H_2O for the solar thermolysis and the solar thermochemical cycles; fossil fuels for the solar cracking, and a combination of fossil fuels and H_2O for the solar reforming and solar gasification. For the solar decarbonization processes, optional CO_2/C sequestration is considered. All of those routes involve energy consuming (endothermic) reactions that make use of concentrated solar radiation as the energy source of high-temperature process heat.**

higher temperatures (800–1000°C) via Solid Oxide Electrolyzer Cells (→SOEC). Concentrated solar energy can be applied to provide the high-temperature process heat.

H_2 from H_2O by solar thermolysis
The single-step thermal dissociation of H_2O to H_2 and O_2 is known as water thermolysis. Although conceptually simple, its realization is very challenging since it needs a high-temperature heat source above 2200°C for achieving a reasonable degree of dissociation, and an effective technique for separating H_2 and O_2 to avoid an explosive mixture. Among the ideas proposed for separating H_2 from the products are →effusion separation and electrolytic separation. Membranes based on ceramics like zirconia and other high-temperature materials have been tested above this temperature, but they usually fail to withstand the severe thermal shocks that often occur when working under high-flux solar irradiation.

Rapid →quench by injecting a cold gas, expansion in a nozzle, or submerging a solar-irradiated target in liquid water, is simple and workable, but the quench introduces a significant drop in the exergy efficiency and produces an explosive gas mixture. Furthermore, the very high temper-

atures required by the thermodynamics of the process (e.g. 2725°C for 64% dissociation at atmospheric pressure) pose severe material limitations and can lead to significant re-radiation from the reactor, thereby lowering the efficiency.

H_2 from H_2O by solar thermochemical cycles
Water-splitting thermochemical cycles bypass the separation problem and further allow operation at moderately high temperatures. Previous studies performed on thermochemical cycles were mostly characterized by using process heat at temperatures below 950°C, which are expected to be available in the future from very high temperature nuclear reactors (→VHTR). These cycles required three or more chemical reaction steps (two steps in the case of the hybrid sulfuric acid cycle incorporating one electrolysis step) and are challenging because of material problems and inherent inefficiencies associated with heat transfer and product separation at each step. The leading candidates for multi-step thermochemical cycles include mainly a three-step →sulfur iodine cycle based on the thermal decomposition of sulfuric acid at 850°C and a four-step →UT-3 cycle based on the hydrolysis of calcium and iron bromide at 750°C and 600°C, respectively.

In recent years, significant progress has been made in the development of optical systems for large-scale solar concentrators capable of achieving mean solar concentration ratios that exceed 5,000 suns. Such high radiation fluxes allow temperatures above 1200°C, which are needed for the more efficient two-step thermochemical cycles using metal oxide →redox reactions (figure 13).
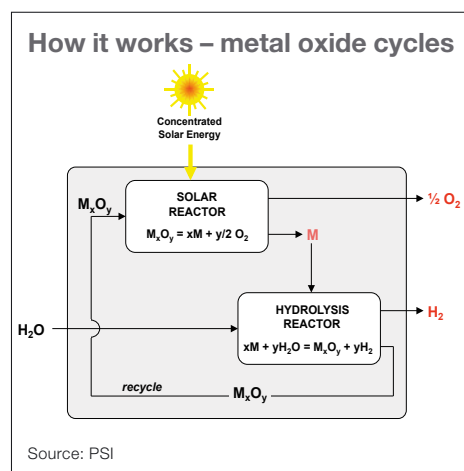

How it works – metal oxide cycles

Source: PSI

Figure 13: Thermochemical route based on metal oxide redox reactions – **The first step of the cycle is the solar thermal release of O_2 from the metal oxide (M_xO_y). This step requires very high temperatures. The second step is the reaction of the metal (M) with H_2O to form H_2 and the corresponding M_xO_y. This step proceeds at lower temperatures and does not require additional heating in some cases. Since H_2 and O_2 are formed in different steps, the need for high-temperature gas separation is thereby eliminated. This cycle was originally proposed for an iron oxide redox system.**
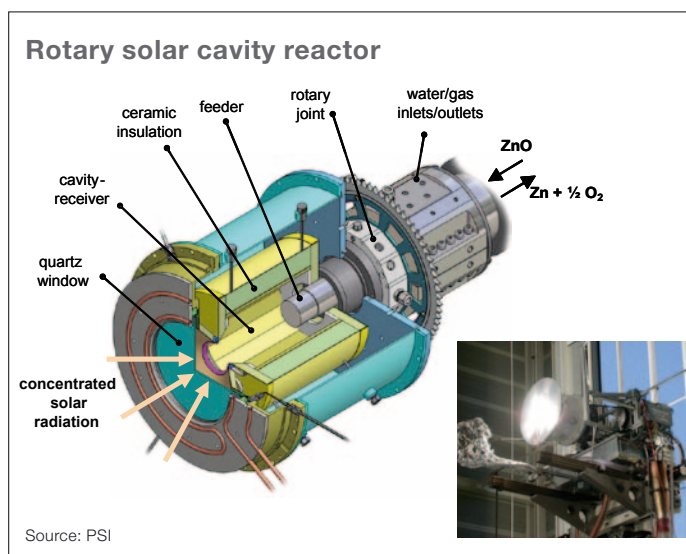
Rotary solar cavity reactor

Source: PSI

Figure 14: Rotary solar reactor for the thermal dissociation of zinc oxide to zinc and oxygen at above 1700°C – **The concept features a windowed rotating cavity-receiver lined with ZnO particles that are held by centrifugal force. With this arrangement, ZnO is directly exposed to high-flux solar irradiation and serves simultaneously the functions of radiant absorber, thermal insulator, and chemical reactant.**
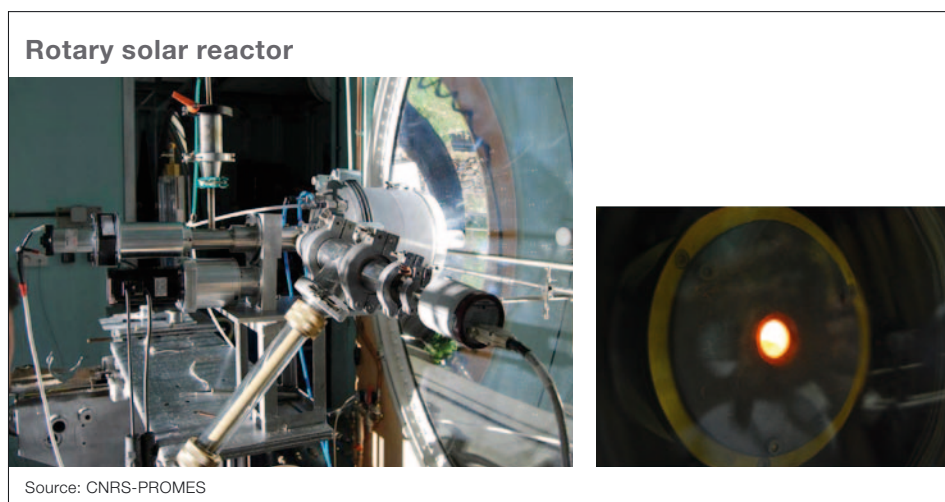
Rotary solar reactor

Source: CNRS-PROMES

Figure 15: Lab-scale solar reactor for the reduction of tin oxide (SnO_2) at 1500°C – **The rotary reactor can operate continuously by particle injection and at reduced nitrogen pressure in order to decrease the decomposition temperature.**

Zn/ZnO cycle – One of the most researched metal oxide redox pairs for the two-step cycle is based on zinc (Zn) and zinc oxide (ZnO). Since the products of the solar high temperature ZnO decomposition – gaseous Zn and oxygen (O_2) – rapidly recombine, a quenching process is necessary. The estimated exergy efficiency of this cycle is 35% without heat recovery from the quench process. Alternatively, electro-thermal methods for in-situ separation of gaseous Zn and O_2 at high temperatures have been experimentally demonstrated in small-scale reactors. High-temperature separation further enables recovery of the sensible and latent heats of the products to enhance the efficiency. A high-temperature solar chemical reactor concept was developed for this process (figure 14). Solar tests carried out at PSI's solar furnace, Switzerland, with a 10 kW reactor prototype subjected to a peak solar concentration of 4000 suns revealed the low thermal inertia of the reactor system – the surface temperature reached 1700°C in two

seconds and was resistant to thermal shocks. In 2010, the solar chemical reactor concept for the thermal dissociation of ZnO will be demonstrated in a 100 kW$_{th}$ pilot plant at a large solar research facility.

For the H_2 production step (Zn + H_2O → ZnO + H_2) laboratory studies and preliminary tests with a novel concept of a hydrolyser (hydrolysis reactor) indicate that the water-splitting reaction works at reasonable rates above 425°C. This was experimentally demonstrated using a so-called →aerosol-flow reactor for in-situ formation and hydrolysis of Zn nano-particles. In principle, the heat liberated by the exothermic reaction could be used to melt Zn and produce steam. Alternatively, if the H_2 production plant was located next to the solar plant, molten Zn from the quencher unit at 425°C (or higher) could be fed directly to the hydrolyser. On the other hand, transportation of solid Zn to the site where H_2 is finally used eliminates the need for storage and transportation of H_2. Another pos-

sible application of this cycle is to use the energy carrier Zn directly in Zn-air batteries. This technology is already commercially available, and some companies are pursuing a fuel-cell analogue with 'mechanically-rechargeable' Zn-air batteries for stationary or mobile applications.

SnO/SnO$_2$ cycle – Another promising metal oxide cycle is based on tin oxide (redox pair SnO/SnO$_2$). Exergy and energy efficiencies of this redox pair are 30% and 36% respectively. Atmospheric and reduced pressure experiments showed that the SnO_2 reduction can be performed efficiently at 1500°C and the SnO hydrolysis at 550°C. A 1 kW solar reactor prototype was successfully tested for continuous operation at Odeillo, France (figure 15).

Mixed iron oxide cycle – Other metal oxides such as manganese oxide or cobalt oxide, as well as mixed oxides redox pairs – mainly based on iron – have also been considered. The

Nano-material
Consists of nano-sized particles having diameters of less than 1 micrometer.

Reducing agent
Donates electrons to a chemical compound, e.g. de-oxidizes a metal.

Syngas
Synthesis gas is a mixture of H_2 and CO produced by reacting natural gas with steam at 850°C and high pressure. The name comes from its role as an important starting substance for chemical synthesis.

Sequestration
Term mainly used for permanent storage of CO_2, either in gaseous form in deep geological formations, in liquid form in the ocean, or in solid form in mineral carbonates.

Fischer-Tropsch
A process developed in 1925 by Franz Fischer and Hans Tropsch to convert syngas into liquid hydrocarbons, e.g. fuels like gasoline or diesel.

Watergas shift reaction
Chemical reaction in which CO reacts with water to form CO_2 and H_2.

Pressure swing adsorption
Technology used to separate some gas species from a mixture of gases under pressure at near-ambient temperatures.

Monolithic solar reactor

four-way valve · to mass spectrometer · ducts for feed gases · inlet for feed gases · N_2 · steam · N_2 (O_2) · H_2 · steam · four-way valve · double tube for feed gas preheating · cylindrical SiC housing · coated honeycomb structure · quartz windows · sealing
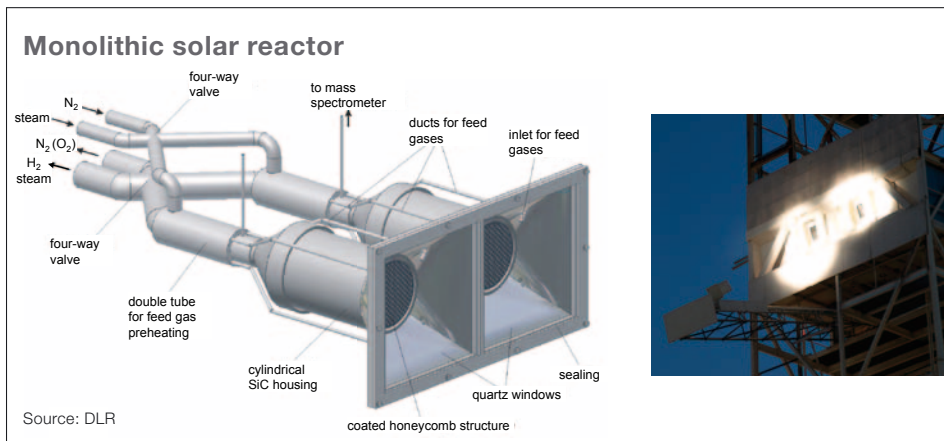
Source: DLR

Figure 16: Monolithic dual chamber solar receiver-reactor for continuous H_2 production – **The concept features a closed receiver-reactor constructed from ceramic multi-channeled monoliths (left). Cyclic operation of the water-splitting and regeneration steps is established in two reaction chambers. Their individual temperature levels are controlled by focusing and defocusing heliostats during thermal tests at PSA, Spain (right).**

mixed iron oxide cycle was demonstrated at the 10 kW$_{th}$ level within the EU's R&D project **HYDROSOL** (2002–2005). The model for the monolithic solar thermochemical reactor (figure 16) was the catalytic converter used for exhaust treatment in automobiles. The reactor contains no moving parts and is constructed from ceramic multi-channeled monoliths that absorb the solar radiation. The monolith channels are coated with mixed iron oxide →nano-materials, which are activated by heating to 1250°C. After releasing the O_2, they are capable of splitting water vapor passing through the reactor by trapping the O_2 and leaving H_2 as the product in the effluent gas stream at 800°C. Thus, no explosive gas mixtures are produced. Cyclic operation is established on a single, closed receiver-reactor system. A quasi-continuous H_2 flow is produced by alternate operation of two or more reaction chambers.

Continuation within the EU's award-winning P&D project **HYDROSOL 2** (2005-2009) is directed at testing a 100 kW$_{th}$ dual chamber pilot reactor at the Plataforma Solar de Almería (PSA), Spain.

Carbothermal reduction of metal oxides – The carbothermal reduction of metal oxides using coke, natural gas (NG), and other carbonaceous materials as →reducing agents brings about reduction of the oxides at even lower temperatures. Carbothermal reductions of metal oxides like iron oxide, manganese oxide, and zinc oxide with carbon and natural gas to produce the metals and →syngas have been demonstrated in solar furnaces. Such a solar chemical reactor concept – PSI's 'two-cavity' solar reactor (figure 17) based on the indirect irradiation of ZnO and carbon (C) for producing Zn and carbon monoxide (CO) – was scaled up

Two-cavity solar reactor

300 kW concentrated solar power · upper cavity (absorber) · carrier gas inlet · quartz window · lower cavity (reaction chamber) · ZnO/C packed-bed · gaseous products
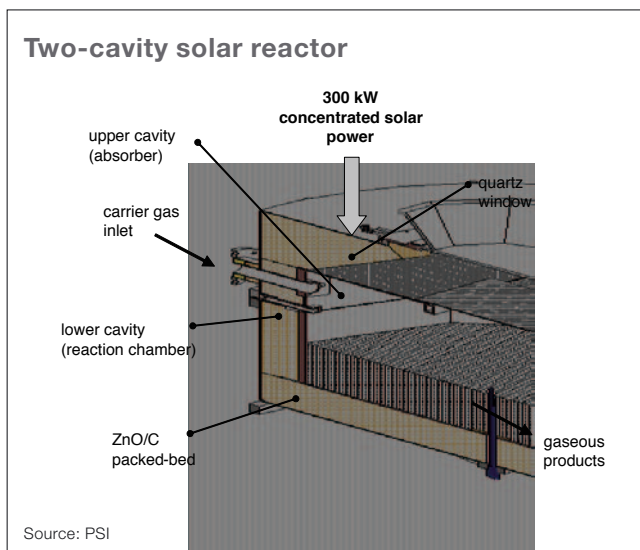
Source: PSI

Figure 17: Two-cavity solar reactor concept for the carbothermal reduction of ZnO – **The upper cavity (absorber) serves as the solar receiver and radiant emitter. The lower cavity is a well-insulated enclosure containing a ZnO/C packed bed. It serves as the reaction chamber and is subjected to thermal radiation from the upper cavity. With this arrangement, the upper cavity protects the quartz window against particles and condensable gases coming from the reaction chamber.**

within the EU's R&D project **SOLZINC** (2001–2005). Testing of the 300 kW$_{th}$ solar chemical reactor (figure 18) at the solar tower research facility of WIS, in Israel, at temperatures ranging from 1000–1200°C yielded up to 50 kg/h of 95%-purity Zn and an energy conversion efficiency of around 30%. The **SOLZINC** process provides an efficient thermochemical route for storing and transporting solar energy. The

Source: PSI

Figure 18: 300 kW$_{th}$ solar chemical pilot plant developed and tested in the EU's R&D project SOLZINC – **The two-cavity reactor (1.4 m inner diameter) for the carbothermal reduction of ZnO is specifically designed for beam-down incident radiation, as obtained at the solar tower research facility at WIS, Israel.**

How it works – decarbonization processes
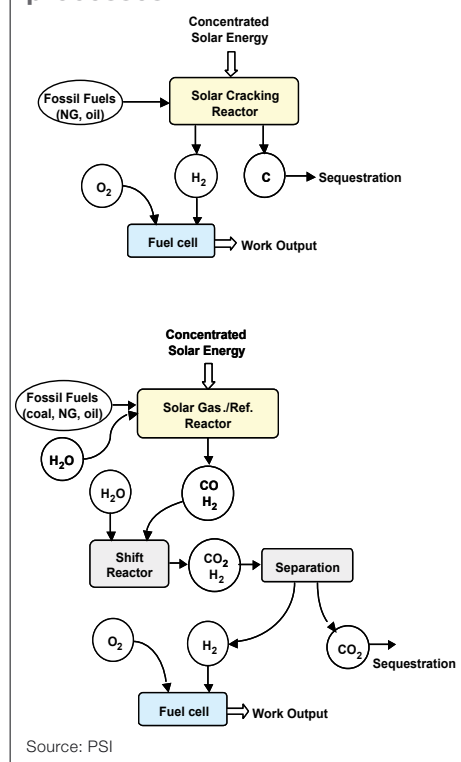
Source: PSI

Figure 19: Solar thermochemical routes for H$_2$ production using fossil fuels and H$_2$O as the chemical source – **Solar cracking (top), and solar reforming and gasification (bottom).**
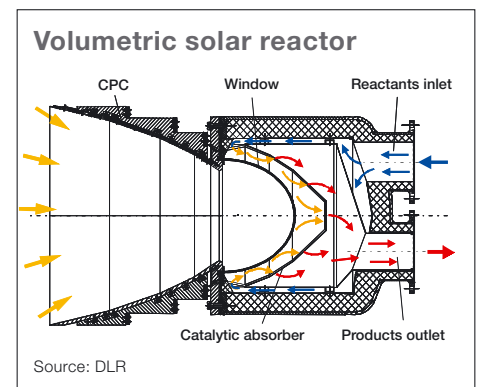
process can be CO$_2$-neutral if charcoal is used for reducing ZnO.

H$_2$ by decarbonization of fossil fuels

Three solar thermochemical processes for H$_2$ production using fossil fuels as the chemical source are considered: cracking, reforming, and gasification (figure 19). The solar cracking route refers to the thermal decomposition of NG, oil, and other hydrocarbon into H$_2$ and carbon. The carbonaceous solid product can either be →sequestered (see box) without CO$_2$ release or used as material commodity or reducing agent under less severe CO$_2$ restraints. Steam-reforming of NG, oil, and other hydrocarbons, and steam-gasification of coal and other solid carbonaceous materials yield syngas, the building block for a wide variety of synthetic fuels including →Fischer-Tropsch type chemicals, H$_2$, ammonia, and methanol. The quality is determined mainly by the ratios between H$_2$, CO, and CO$_2$. The CO content in the syngas can be shifted to H$_2$ via a catalytic reaction with steam – the so called →watergas shift reaction. The product, CO$_2$, can be separated from H$_2$, e.g. by using the →pressure swing adsorption technique.

Solar reforming – Solar reforming of NG, using either steam or CO$_2$, has been extensively studied in solar concentrating facilities with small-scale solar reactor prototypes. The solar re-

How it works – carbon sequestration

From the point of view of carbon sequestration, it is easier to separate, handle, transport, and store solid carbon than gaseous CO$_2$. Furthermore, while the steam reforming/gasification method requires additional steps for shifting CO and for separating CO$_2$, the thermal cracking accomplishes the removal and separation of carbon in a single step.

In contrast, the major drawback of the thermal decomposition method is the energy loss associated with the sequestration of carbon. Thus, the solar cracking may be the preferred option for NG and other hydrocarbons with high H$_2$/C ratio. For coal and other solid carbonaceous materials, the solar gasification has the additional benefit of converting a solid fuel traditionally used for combustion to generate electricity into a cleaner fluid fuel – cleaner only when using solar process heat – that can be used in highly efficient fuel cells.

For the solar NG cracking and the solar coal gasification without CO$_2$ sequestration, specific CO$_2$ emissions amount to 0.53-0.56 kg CO$_2$/kWh$_{el}$, about half as much as the specific emissions discharged by conventional coal-fired power plants.

forming process developed within the EU's R&D project **SOLREF** (2004–2009) has been scaled-up to power levels of 300–500 kW$_{th}$ and tested at 850°C and 8–10 bars in a solar tower using two solar reforming reactor concepts: indirect-irradiation and direct-irradiation (see box). The indirect-irradiated solar reforming reactor (not shown here) consists of a cavity receiver insulated with ceramic fibers that contains a set of vertical tubes externally exposed to the concentrated solar radiation. A matching CPC is installed at the windowless cavity →aperture for capturing radiation spillage, augmenting the average solar flux concentration, and providing uniform heating of the tubes. Similar to the indirect-irradiated reactor, a CPC is installed at the aperture of the direct-irradiated solar reactor – also referred to as the volumetric solar reactor (figure 20).

Volumetric solar reactor

Source: DLR

Figure 20: Volumetric solar reactor concept for the reforming of NG – **The main component is the porous ceramic absorber, coated with Rhodium catalyst, which is directly exposed to the concentrated solar radiation. A concave quartz window, mounted at the aperture, minimizes reflection losses and permits operation at elevated pressures.**

Solar reactor – direct vs. indirect irradiation

Direct-irradiation reactor – Provides efficient radiation heat transfer to the reaction site where the energy is needed, by-passing the limitations imposed by indirect heat transport via heat exchangers. The major drawback when working with reducing or inert atmospheres is the requirement for a transparent window, which is a critical and troublesome component in high-pressure and severe gas environments.

Indirect-irradiation reactor – Eliminates the need for a transparent window. The disadvantages are linked to the limitations imposed by the materials of construction of the reactor walls: constraints in maximum operating temperature, thermal conductivity, radiant absorbance, inertness, resistance to thermal shocks, and suitability for transient operation.

Aperture
Opening of a solar cavity-receiver for accepting incoming solar radiation.

Catalyst
Catalysis is the process in which the rate of a chemical reaction is increased by means of a chemical substance known as a catalyst.

Rankine cycle
Thermodynamic cycle that converts heat into work. The heat is supplied externally to a closed loop, which usually uses water as the working fluid.

Parabolic dish
A 3-dimensional parabola-shaped reflective device (e.g. faceted mirror) used to collect and focus solar energy.

Petcoke
Petroleum coke (often abbreviated petcoke) is a carbonaceous solid by-product derived from oil refinery coker units or other cracking processes.

Vortex flow
Motion of a spinning, often turbulent flow of fluid swirling rapidly around a center.

Claus process
Chemical process originally developed to produce sulfur from H_2 sulfide, which is being formed as by-product in the formation of coke from coal or the desulfurization of crude oil.

Dry reforming of methane (CH_4) with CO_2 was performed in an aerosol solar reactor similar to the one used for NG cracking (see figure 24 below). Operating with residence times around 10 milliseconds and temperatures of approximately 1700°C, methane and CO_2 conversions of 70% and 65%, respectively, were achieved in the absence of →catalysts.

The solar dry reforming of CH_4 with CO_2 can also be used in a closed-loop chemical heat pipe (figure 21). The product of this reversible reaction is syngas which can be stored at ambient temperatures and transported to sites requiring energy. Stored solar energy is released by the reverse exothermic reaction in the form of heat, which can be used for generating electricity via a →Rankine cycle. The products of this reverse reaction are again recycled to the solar reactor to complete the cycle.

Similarly, the dissociation/synthesis of ammonia (NH_3) can be applied in a chemical heat pipe for storage and transportation of solar energy. In this system developed by the Australian National University (ANU), ammonia is dissociated in an energy storing (endothermic) chemical reactor as it absorbs solar thermal energy. At a later time and place, the reaction products of H_2 and nitrogen (N_2) react in an energy releasing (exothermic) reaction to re-synthesize ammonia. An industrial demonstration plant has been announced using four of ANU's 400 m^2 →parabolic dishes.

Solar steam gasification – The steam-gasification of carbonaceous materials and related reactions has been performed using concentrated solar energy in early exploratory studies with coal and oil shale. A conceptual design of a solar reactor for the gasification of carbonaceous materials was performed using optical fibers to direct the solar radiation into the reaction chamber. In the framework of the industrial project **SYNPET** (2003–2009), the solar chemical technology for the steam-gasification of →petcoke particles was developed. For this process, a 10 kW$_{th}$ solar reactor was designed that featured a continuous gas-particle →vortex flow confined to a cavity receiver and directly exposed to concentrated solar radiation. A scale-up of the vortex solar reactor (figure 22) to 500 kW$_{th}$ and testing at the Plataforma Solar de Almería is progressing within the timeframe of the project.
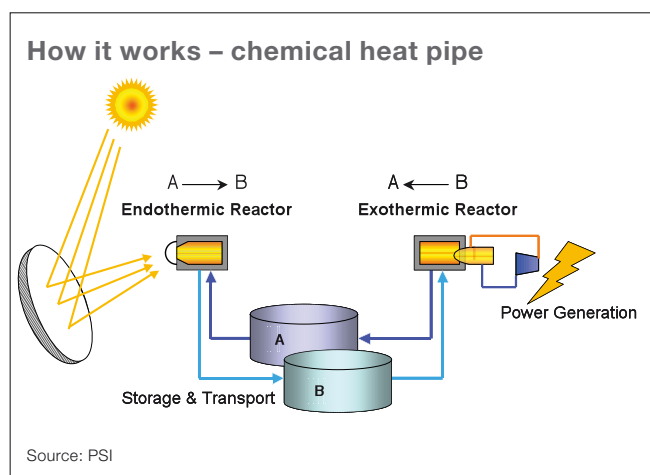

How it works – chemical heat pipe

Source: PSI

Figure 21: Solar chemical heat pipe for the storage and transportation of solar energy – **High-temperature solar process heat is used to drive the endothermic reversible reaction A–B. The product B may be stored and transported to the site where the energy is needed. At that site, the exothermic reverse reaction B–A is effected and yields high temperature process heat in an amount equal to the stored solar energy. The chemical product A of the reverse reaction is returned to the solar reactor for reuse.**

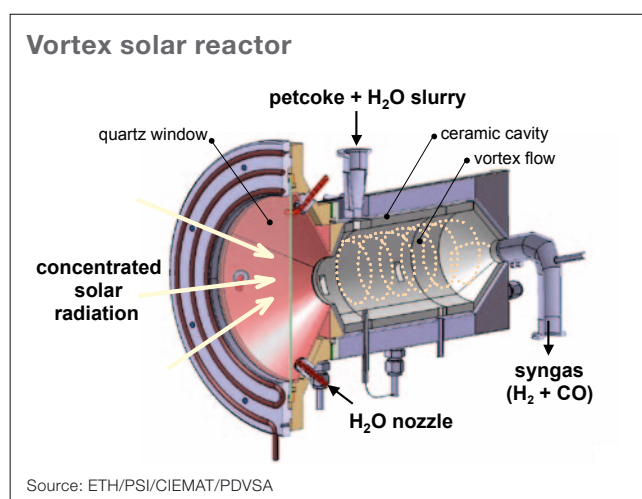
Vortex solar reactor

Source: ETH/PSI/CIEMAT/PDVSA

Figure 22: Vortex solar reactor for the steam-gasification of petcoke – **This configuration features a continuous gas-particle vortex flow confined to a cavity receiver and directly exposed to concentrated solar radiation. The reactants are injected as liquid slurry of petcoke particles and water.**
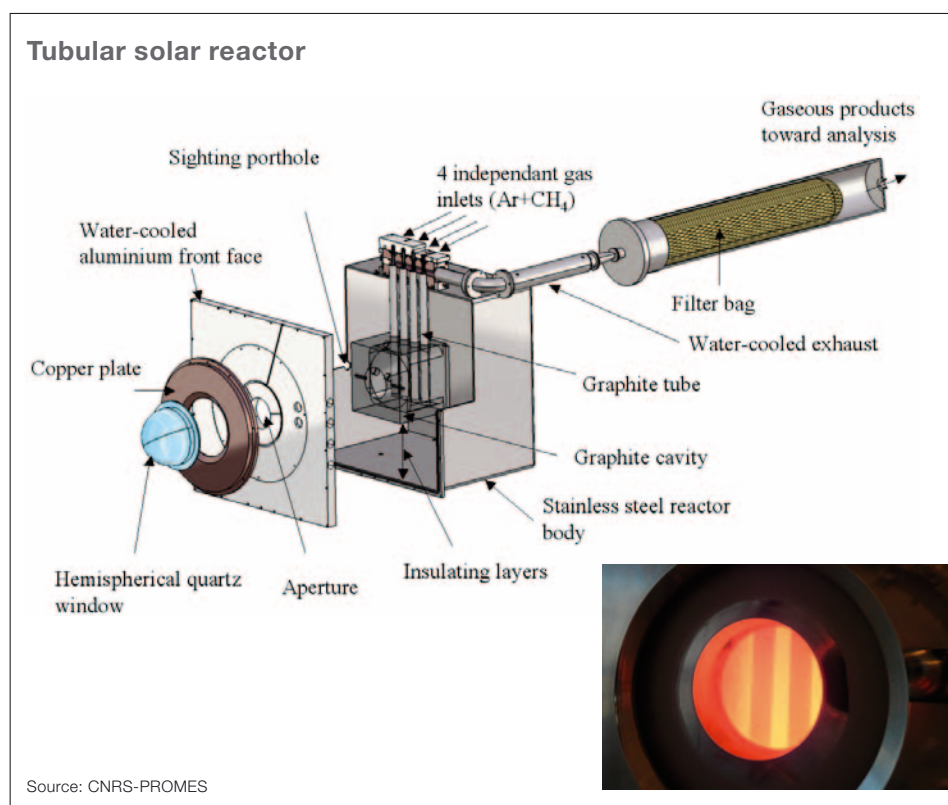
Figure 23: Tubular solar reactor for NG cracking – **Methane (CH₄) and argon (Ar), are indirectly heated inside graphite tubes (top). Test campaign with a 10 kW$_{th}$ solar reactor prototype for NG cracking at the focus of the 1 MW solar furnace, Odeillo, France (bottom).**
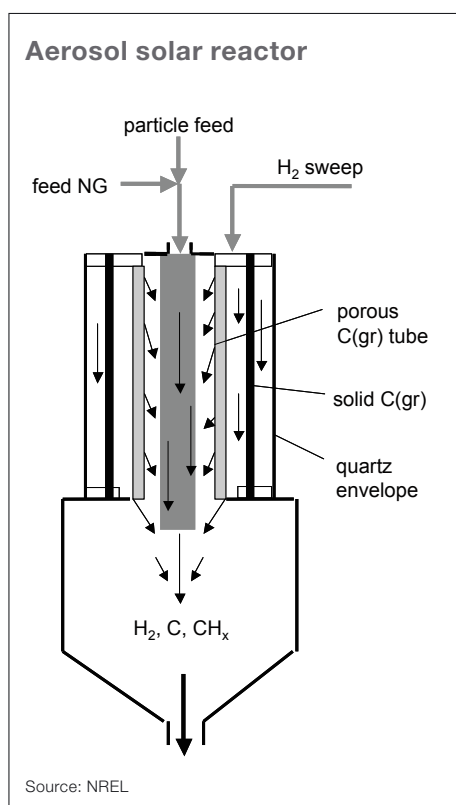

Figure 24: Aerosol solar reactor for thermal cracking of NG – **This configuration features two concentric graphite tubular reactors: the outer solid tube serves as the solar absorber and the inner porous tube contains the reacting flow consisting of fine carbon black particles suspended in a CH₄ feed gas stream.**

Solar cracking – Cracking of CH_4 or NG is a nonconventional route for potentially cost-effective H_2 production and carbon nano-material synthesis with concentrated solar energy. The process thermally decomposes NG in a high temperature solar chemical reactor, yielding two valuable products: a H_2-rich gas and a marketable high-value nano-material called 'Carbon Black' (CB). Thus, both H_2 and CB are produced by renewable energy, resulting in potential CO_2 emission reduction of 14 kg of CO_2 and energy saving of 277 MJ per kg H_2 compared to conventional NG steam reforming and standard CB processing.

A novel 10 kW$_{th}$ tubular solar reactor (figure 23) was successfully tested at a temperature range of 1430–1800°C at the solar furnace of CNRS-PROMES in Odeillo, France. A high dissociation of 98% was achieved with a 90% H_2 yield. An advanced 50 kW$_{th}$ solar reactor prototype is being developed in the framework of the EU's R&D project **SOLHYCARB** (2006–2010). Also, a tubular aerosol reactor (figure 24) was tested at NREL in Golden, Colorado, where methane conversion of up to 90% was obtained. Further studies are being performed at ETH Zurich, Switzerland, in a so-called vortex solar reactor configuration (figure 22), which was tested in a methane flow laden with carbon particles that serve simultaneously as radiant absorbers and nucleation sites for the heterogeneous decomposition reaction.

H_2 from H_2S by solar thermolysis

An optional source of H_2 is hydrogen sulfide (H_2S), a toxic industrial byproduct derived from NG, petroleum, and coal processing. Current industrial practice uses the →Claus process to recover sulfur from H_2S, but H_2 is wasted by oxidizing it to H_2O. Alternatively, H_2S can be thermally decomposed above 1500°C to produce H_2 and sulfur (S), which, after quenching, naturally separate due to phase change. Solar experiments indicate high degrees of chemical conversion.

Solar Hydrogen –
At Competitive Costs!

The economical competitiveness of solar fuel production is closely related to two factors: the cost of fossil fuels and the necessity to control the world climate by drastically reducing CO_2 emissions. Transition processes like the solar steam reforming of NG will be economically competitive at a NG price of 11 $/MMBtu or 0.42 $/Nm^3; for comparison, the NG price for long-term contracts was 8-9 $/MMBtu, and the spot price was 14-16 $/MMBtu (DOE, 2008).[1]

Both the US Department of Energy and the European Commission have a clear vision of the hydrogen economy with firm targets for H_2 production costs: the US target (figure 25) for 2017 is 3 $/gge (gasoline gallon equivalent; 1 gge is about 1 kg H_2), and the EU target for 2020 is 3.50 €/kg H_2. DOE's →H2A Analysis Group has developed the tools necessary to conduct rigorous and consistent analyses of a wide range of H_2 technologies and the transition to a H_2 economy. Predicted solar thermal hydrogen production cost are 2–4 $/kg for efficient solar thermochemical cycles.

The economics of massive solar H_2 production (figure 26) has been assessed in numerous studies, which indicate that the solar thermo-chemical production of H_2 can be competitive compared with the electrolysis of H_2O using solar-generated electricity. As described above, it can even become competitive with conventional fossil-fuel-based processes at current fuel prices, especially if credits for CO_2 mitigation and pollution avoidance are applied.

The weaknesses of these economic assessments are primarily related to the uncertainties in the viable efficiencies and investment costs of the various solar components due to their early stage of development and their economy of scale. Therefore, further development and large-scale demonstrations are warranted. This would have positive effects on achievable efficiencies and investment cost reduction for materials and components. In this respect, an important factor will be the massive installation of commercial solar thermal power plants, in particular power towers, since heliostats will be one of the most expensive components of a solar thermal H_2 production plant.

Massive hydrogen production – economics

Characteristics	Units	2006 Target	2012 Target	2017 Target
Solar thermo-chemical H_2 cost	$/gge H_2	10	6	3
Installed helio-stat capital cost	$/m^2	180	140	80
Process energy efficiency	%	25	30	>35

Source: STCH

Figure 25: Technical and economic targets of the US DOE hydrogen program – **Cost improvements by technical innovations (process energy efficiency), scaling up the solar chemical plant, and increasing the production volume are expected for the STCH project (Solar-driven high temperature Thermo Chemical Hydrogen production) endorsed by the IPHE.**
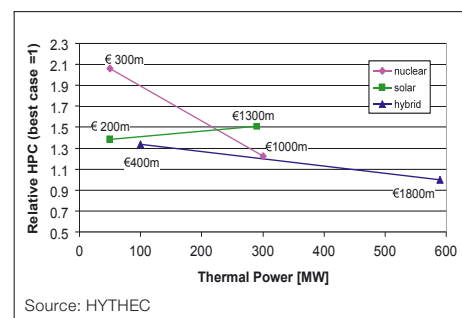

Source: HYTHEC

Figure 26: Comparison of H_2 production costs (HPC) for solar, nuclear, and hybrid powered H_2 production plants based on the hybrid sulfuric acid cycle – **The EU-FP6 project HYTHEC (2004–2007) has evaluated 26 scenarios for large-scale H_2 production based on the hybrid sulfuric acid thermochemical cycle using either solar, nuclear, or hybrid (combined solar and nuclear) sources of high temperature process heat. Interestingly enough, H_2 production cost would be lower for solar thermal plants than for nuclear plants in a power range up to 250 MW$_{th}$, whereas hybrid operation would be the cheapest solution at power levels above 300 MW$_{th}$. These findings are in good agreement with conceptual studies presented by General Atomics (GA) in the early 1980s where the high temperature step of sulfur-based thermochemical cycles was assumed being carried out in a solar reactor.**

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[1]   US DOE, February 2008,
     http://tonto.eia.doe.gov/oog/info/ngw/ngupdate.asp
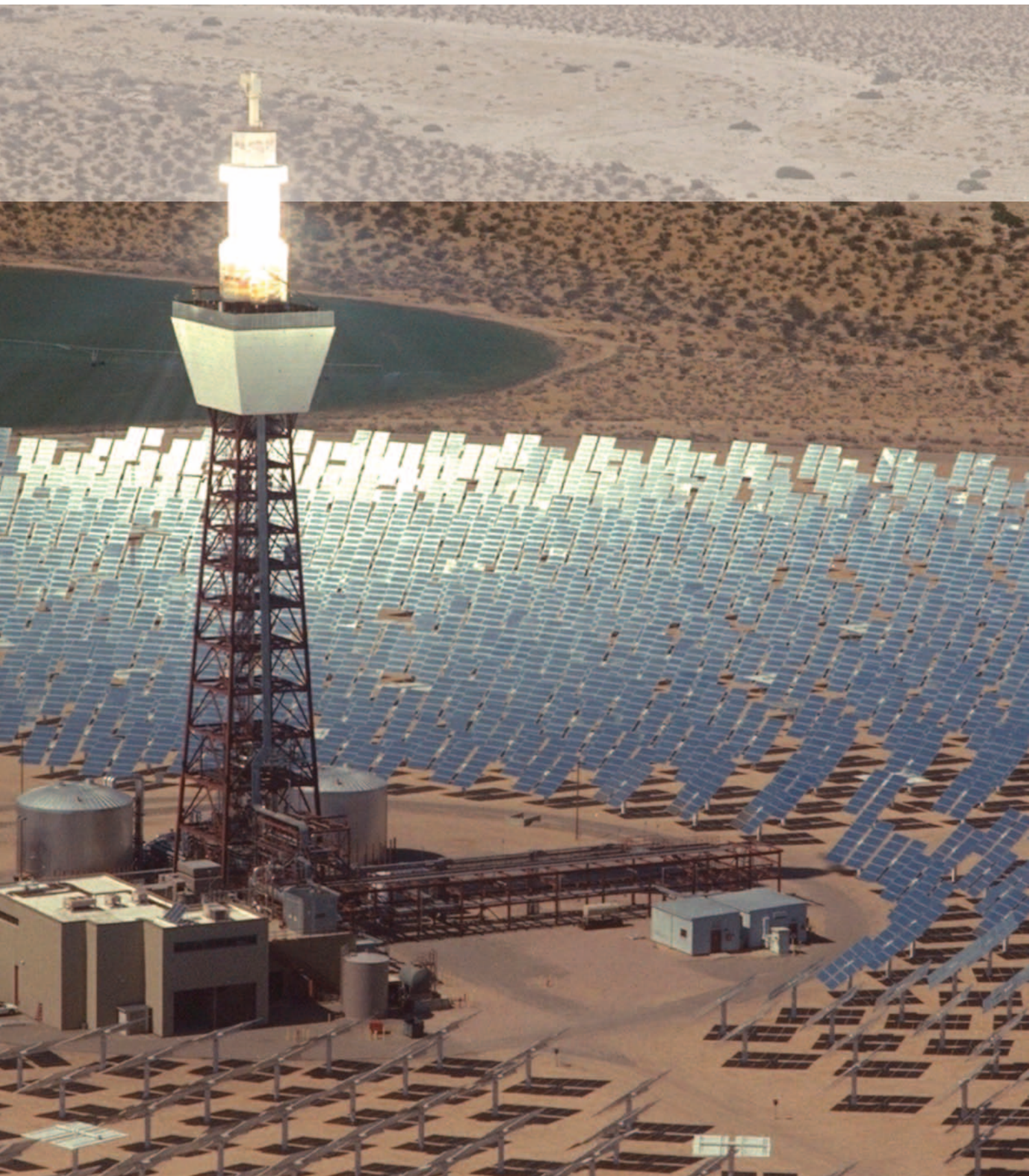
Boosting Solar Fuels – Need for Worldwide R&D

H2A
US Department of Energy (DOE) standard format and list of parameters for reporting realistic economic and technology-based analysis results for central production, distributed production, and delivery of H_2.

Most activities on solar fuel production are funded by national or international research programs like the Framework Programs of the European Union[1], the Hydrogen and Fuel Cell Joint Technol-ogy Initiative (JTI)[2] and the hydrogen program of the US DOE[3] (figure 27). The projects are generally carried out by international consortia from R&D, industry and academia (see figure 28).

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[1] EU FP7: http://cordis.europa.eu/fp7/

[2] JTI: https://www.hfpeurope.org/hfp/jti
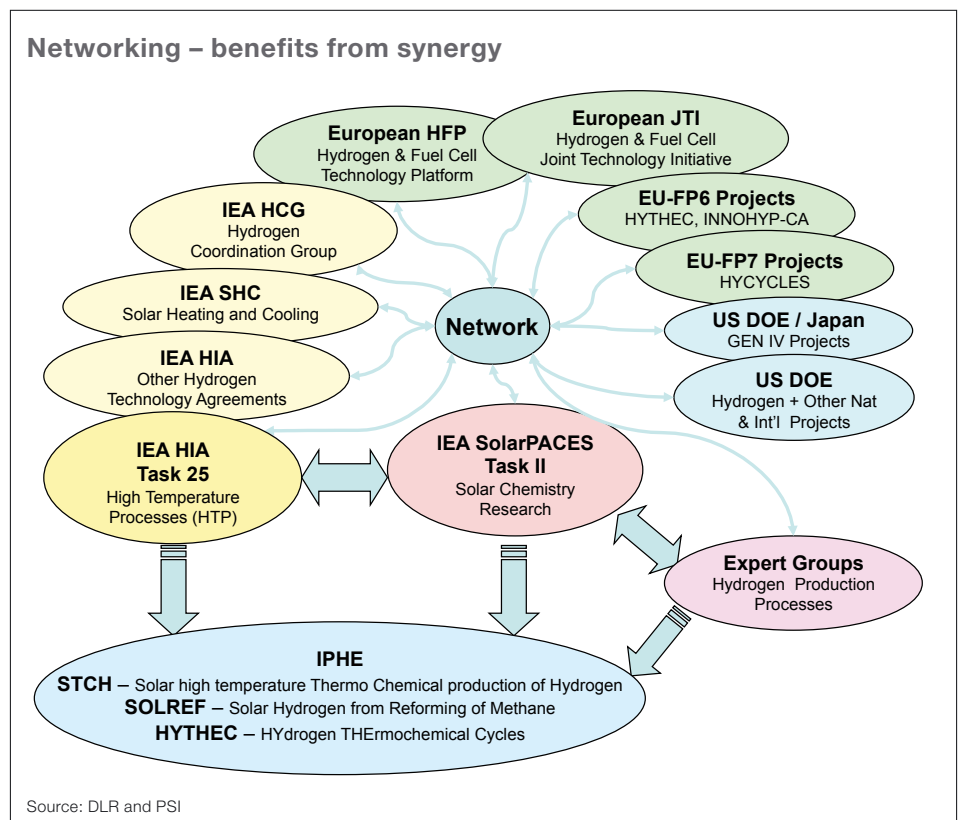[3] US DOE: www.hydrogen.energy.gov/

Figure 27: Network for solar fuels – **Linking pertinent research activities for the production of solar hydrogen and other fuels under the umbrella of international organizations such as IEA or IPHE.**

International networking – The global relevance of solar fuels makes it necessary to link the pertinent research activities under the umbrella of organizations such as the International Energy Agency **(IEA)**[4] and the International Partnership for the Hydrogen Economy **(IPHE)**[5].

International Energy Agency – The main network for solar fuels (figure 27) is Task II on Solar Chemistry Research of the **IEA Solar-PACES** Implementing Agreement[6]. Task 25 on High Temperature Hydrogen Production of the IEA Hydrogen Implementing Agreement **(IEA-HIA)**[7] creates a link between solar, nuclear, and conventional H_2 production technologies.

International Partnership for the Hydrogen Economy – The IPHE was established in 2003 as an international institution to accelerate the transition to a global hydrogen economy. This key organization is supported by the governments of the member states. The purpose of the IPHE is to provide a mechanism for partners to organize, coordinate and implement effective, efficient and focused international research, development, demonstration and commercial utilization activities related to H_2 and fuel cell technologies. A number of high-quality solar thermochemical projects for the massive production of H_2 are endorsed by the IPHE (see box).

Solar projects – endorsement by IPHE

STCH – Solar Thermo Chemical production of H_2
- Within this project, the most promising thermochemical cycles for H_2 production will be identified, and one or two cycles will be down-selected for demonstration.
- Lower cost solar concentrating technology will be developed, as well as solar receiver and thermochemical reactor technology to demonstrate a fully integrated thermochemical process on-sun.

SOLREF – Solar H_2 from Reforming of Methane
- The project aims at designing, testing, and demonstrating a unique, low temperature, steam reforming reactor using concentrated solar energy.
- A world-class solar facility for international collaboration in H_2 production from solar sources will be built to integrate the system.

HYTHEC – HYdrogen THErmochemical Cycles
- Within the former EU-FP6 project HYTHEC (2004–2007), the potential for massive H_2 production by the sulfur-iodine (S-I) cycle was investigated and compared to the hybrid sulfuric acid cycle. The work is being continued in the EU-FP7 project HyCycleS (2008–2010).
- The emphasis is on the suitability of materials and the reliability of components of a scalable reactor. Of particular interest is the potential use of solar primary energy sources for the decomposition of sulfuric acid.

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[4] IEA: http://www.iea.org/index.asp
[5] IPHE: http://www.iphe.net/
[6] IEA-SolarPACES Task II: http://www.solarpaces.org/Tasks/Task2/task_ii.htm
[7] IEA-HIA Task 25: http://www.ieahia.org/page.php?s=static&p=task25

Solar fuels production – worldwide activities (national and international research programs)

Topic of R&D	Participating Entity	Remarks
Solar Decarbonization	• CIEMAT (E) + ETH/PSI (CH) + PDVSA (Venezuela)	Petcoke gasification; Industrial project SYNPET
	• ETH/PSI (CH)	Gasification of carbonaceous material

	• CNRS-PROMES (F) + ETH/PSI (CH)
	  + TIMCAL (B) + ABENGOA (E)
	  + N-GHY (F) + CREED (F)
	  + APTL (GR) + WIS (IL) + DLR (D)
	• Florida Solar Energy Center (USA)
	• CIEMAT (E)
		NG cracking;
		EU-FP6 project SOLHYCARB

	• DLR (D) + WIS (IL) + ETH (CH) • CSIRO (Australia) • Tokyo Institute/Niigata U. (Japan)	NG reforming; EU-FP6 project SOLREF

Solar Thermochemical Cycles	• APTL (GR) + DLR (D)
	  + Johnson Matthey (UK)
	  + StobbeTech (DK) + CIEMAT (E)
		Fe-based cycle;
		EU-FP5 project HYDROSOL,
		EU-FP6 project HYDROSOL 2

	• PSI/ETH (CH) + U. of Minnesota (USA)	ZnO/Zn cycle
	• PSI/ETH (CH) + WIS (IL) + CNRS-PROMES (F) + ScanArc (S)	ZnO+C reaction; EU-FP5 project SOLZINC
	• CNRS-PROMES (F)	SnO_2/SnO cycle and comparison with ZnO/Zn cycle; Ce-based mixed oxide cycles; Fe-based cycles
	• Sandia + NREL + U. of Colorado + Pinnacle West (USA)	ZnO cycle; Fe-based cycle; SI cycle; DOE project
	• Tokyo Institute / Niigata U. (Japan)	Fe-based cycle
	• ENEA (I)	UT3-cycle

	• CEA (F) + DLR (D) + ProSim (F)
	  + U. of Sheffield (UK) + DIMI (I)
	  + Empresarios Agrupados (E)
		SI cycle; hybrid sulfuric acid cycle;
		EU-FP6 project HYTHEC

	• DLR (D) + CEA (France) + ETH (CH)
	  + U. of Sheffield (UK) + BoosTec (F)
	  + Empresarios Agrupados (E)
	  + APTL (GR) + JRC (EU) + ENEA (I)
	  + General Atomics (USA) + Westinghouse (USA)
	  + JAEA (Japan) + CSIRO (Australia)
		SI cycle, materials, components;
		EU-FP7 project HyCycleS

Figure 28: Solar fuels production – **Overview on research institutes active in national and international research programs for producing solar hydrogen and other fuels.**

Transition to Solar Fuels – Recommended Strategy

The ultimate goal of all research activities described above is to develop technically and economically viable technologies for solar thermochemical processes that can produce solar fuels, particularly H_2. The implementation should start immediately to accelerate the transition from today's fossil-fuel-based economy to tomorrow's solar driven hydrogen economy (figure 29).

R&D strategy for solar H_2 production

The EU-FP6 project **INNOHYP-CA (**2004–2006) has developed a roadmap to implement thermochemical processes for massive H_2 production (figure 30). Within this project, processes with both solar and nuclear sources of process heat were thought to benefit from synergies in their development. Some of the proposed processes can be operated by both energy sources separately or in hybrid installations. However, the nuclear option is impeded in many countries due to political opposition, and the processes driven by future VHTR are limited to 950°C. In particular, the processes with highest exergy efficiency can only be attained by concentrating solar systems.

Processes and timeframe – Since the various solar fuel production technologies under consideration are at diverse stages of maturity, the **INNOHYP-CA** project proposes three phases and a staggered timeframe for their individual development and commercial demonstration.
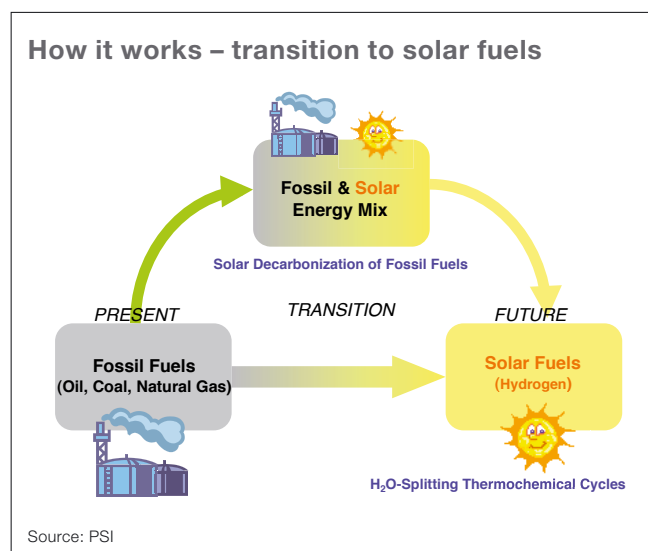

How it works – transition to solar fuels

Fossil & Solar Energy Mix

Solar Decarbonization of Fossil Fuels

PRESENT   TRANSITION   FUTURE

Fossil Fuels
(Oil, Coal, Natural Gas)

Solar Fuels
(Hydrogen)

H_2O-Splitting Thermochemical Cycles

Source: PSI

Figure 29: Strategy for substitution of fossil fuels with solar H_2 involving research on two paths – **The long-term approach via H_2O-splitting thermochemical cycles brings us to the complete substitution of fossil fuels with solar H_2. The short-to-mid term approach via decarbonization of fossil fuels creates a link between today's fossil-fuel-based technology and tomorrow's solar chemical technology.**

First phase – The first phase lasts until 2015 and is devoted to the pilot demonstration of transition processes like solar steam reforming of NG **(SOLREF)**, solar gasification of petcoke **(SYNPET)**, or solar carbothermal reduction of ZnO **(SOLZINC)**. The further development of the zero CO_2 emission technologies will need more time for market introduction. Additional work should be dedicated to studying alternative processes, which in the long term offer a high potential but also a high risk of failure.

Second phase – The second phase lasts until 2020 and investigates the most promising nominally CO_2-free thermochemical processes such as the iodine sulfur (IS) cycle, the hybrid sulfuric acid cycle (WH), the mixed iron oxide cycle (Ferrites), as well as the high temperature steam electrolysis (HTSE) and will be demonstrated at pilot scale. Their chances for successful commercialization will then be assessed.
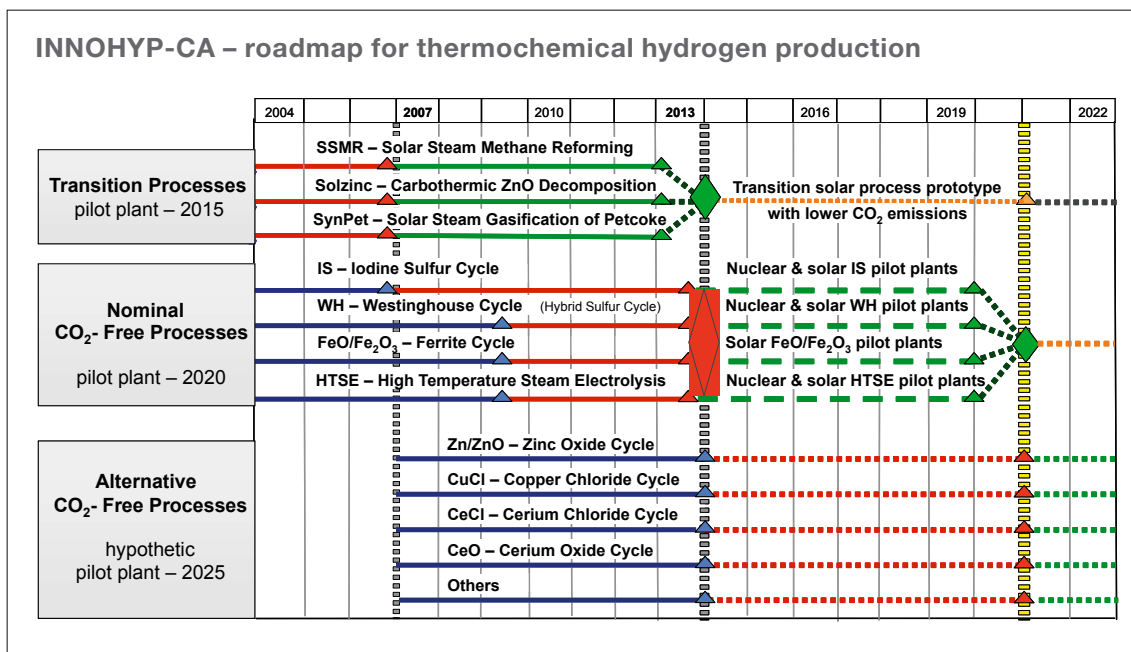

Figure 30: INNOHYP-CA road-map – **An important aspect is the continuous evaluation and benchmarking of the processes (marked by triangles) to avoid time consuming dead ends. Since the various solar fuel production technologies under consideration are at diverse stages of maturity, three phases and a staggered timeframe are proposed for their individual development.**
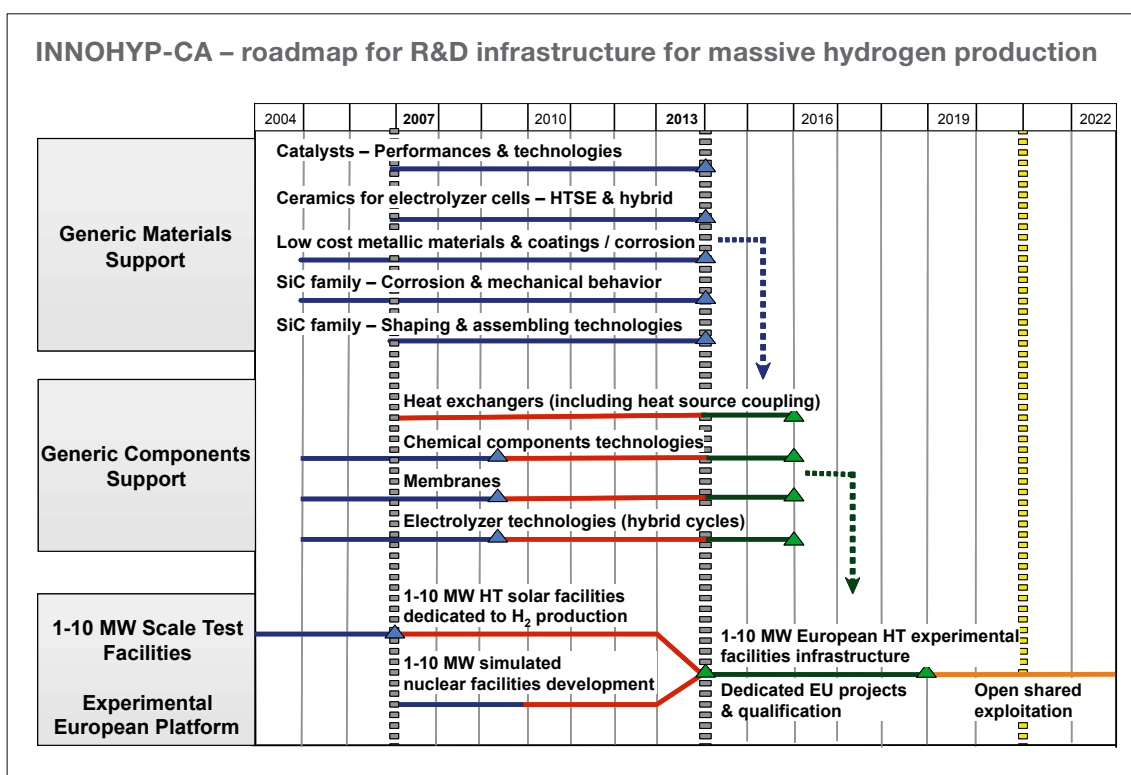

Figure 31: INNOHYP-CA R&D roadmap – **Cross-cutting actions are proposed to develop high-temperature materials and infrastructure components. The foundation of a worldwide alliance of research facilities is envisaged to exploit synergies and to provide access to experimental platforms. The realization of these far-reaching initiatives can be fostered by IEA's SolarPACES, while organizations like the IPHE or the European JTI also work in this direction.**

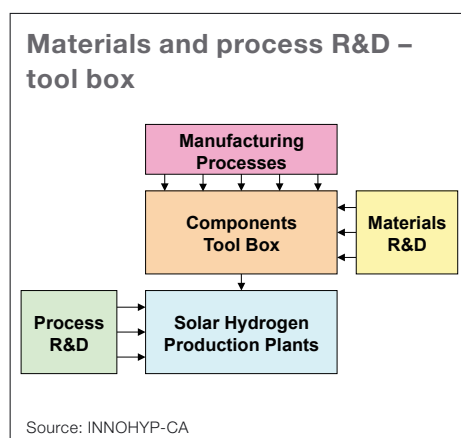**Materials and process R&D – tool box**

Source: INNOHYP-CA

Figure 32: Integration of components tool box and process R&D – **The components tool box combines materials and components developments required for safely and efficiently running thermochemical processes for massive H_2 production.**

Third phase – The third phase lasts until 2025 and will be committed to improving the nominally CO_2-free technologies (see second phase), as well as the demonstration of alternative CO_2-free technologies, provided their technical and economical feasibility can be shown. Emphasis is given to potentially highly efficient two-step thermochemical cycles (in particular the Zn/ZnO cycle) for solar application, and low temperature thermochemical cycles (like the hybrid copper chloride cycle) for nuclear application.

R&D needs for solar H_2 production

In parallel, a development of the high temperature technologies for massive H_2 production, the **INNOHYP-CA** project, proposes a roadmap for further improving the existing R&D infrastructure, mainly for the efficient use of concentrating solar facilities (figure 31). The roadmap also addresses material and component developments required for safely and efficiently running thermochemical processes for the production of solar fuels, in particular solar H_2.

Future national and international research programs will need to focus on further evaluating and improving the proposed thermochemical processes, since adequate technical data and information on production cost and GHG emission are still missing. In addition, important cross-cutting actions are required to develop high-temperature materials and infrastructure components.

At present, hardly any reliable reactor wall materials are available that can withstand the severe operating conditions of the various solar fuel production processes. For many lower temperature processes, e.g. the sulfur based thermochemical cycles, the major problem is corrosion. For very high temperature metal oxide cycles, the challenge is the thermal shock resistance of the materials. As soon as thermally and chemically stable ceramic materials will be available, the next challenge is to manufacture complete components that can be safely integrated into the solar chemical plants. An example is the gas-tight connection of ceramic materials to metals at temperatures above 900°C. For this purpose, the **INNOHYP-CA** project suggests the creation of a components tool box

(figure 32) that combines R&D of appropriate materials for the proposed chemical reactor components with adequate manufacturing processes, e.g. shaping, brazing and welding.

It is imperative to know the limitations of materials and manufacturing technologies before designing and constructing industrial solar chemical plants (figure 33). For example, the size of a single chemical reactor may be limited to less than 50 MW$_{th}$ solar power input, meaning that a cyclic process coupled to a 500 MW$_{th}$ solar tower plant would be realized in at least ten modular units.

A vital factor is the availability of adequate research and test infrastructure for large-scale solar testing of thermochemical cycles. It is indispensable to build such demonstration plants in an international context by pooling R&D budgets to achieve a maximum impact of the huge investments. This development process can be facilitated by founding strategic alliances of research institutes such as **SOLLAB** (Alliance of European Laboratories on Solar Thermal Concentrating Systems)[1] or virtual institutes like **vICERP** (virtual Institute of Central Receiver Power Plants).[2]

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[1] SOLLAB: www.sollab.eu
[2] vICERP http://vicerp.com/

Industrial solar chemical plant for massive hydrogen production – design concepts
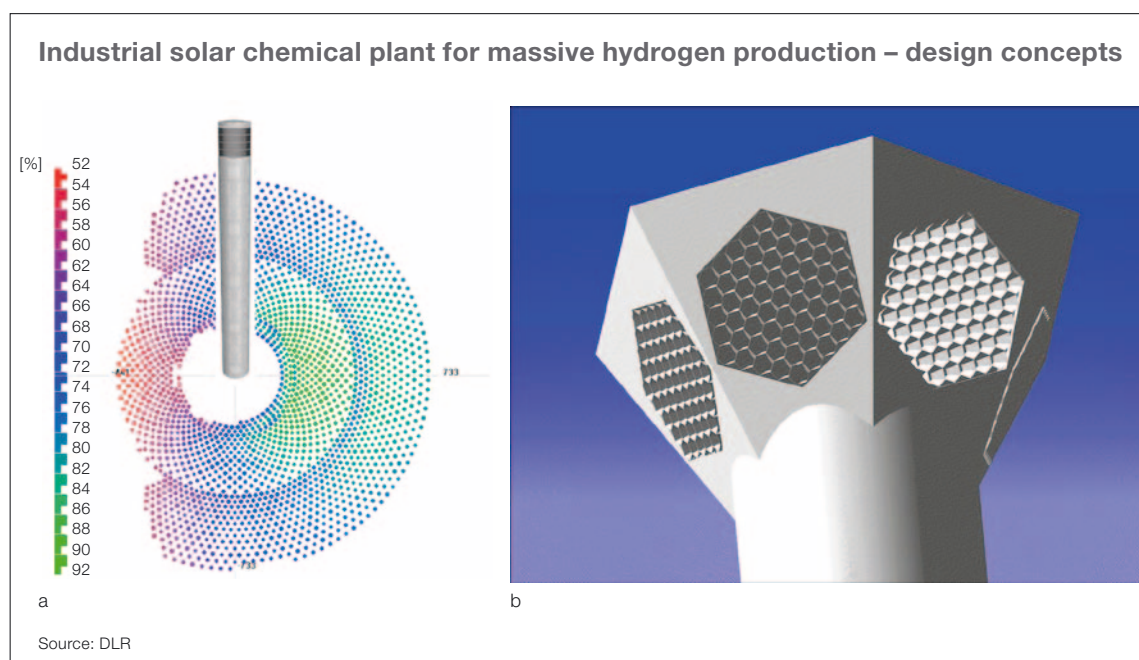
[%]

a

b

Source: DLR

Figure 33: Design concepts for large-scale solar tower plants – **a) Example of a circular heliostat field layout showing largest distances of 733 m from central tower and annual efficiencies of each heliostat (color code). b) Conceptual design of multi-eye tower for multiple solar thermochemical reactor operation.**

Bright Future for Solar Fuels – Outlook

Suggestions for Further Reading

Fletcher, E. A. (2001)
Solarthermal Processing: A review.
J. Solar Energy Engineering 123, 63-74.

Abanades S., Charvin P., Flamant G., Neveu P. (2006)
Screening of water-splitting thermochemical cycles potentially attractive for hydrogen production by concentrated solar energy, Energy 31, 2469-2486.

Solar thermal power plants have paved the way for solar fuels production plants. Similar to concentrating solar power (CSP) plants emerging as an alternative for large scale power generation, concentrating solar chemical plants will presumably become the salient option for sustainable renewable fuels production.

Solar energy is free, abundant and inexhaustible, but at least two crucial steps are necessary for a successful market introduction of solar fuels. Firstly, the solar chemical production technologies must be further developed and proven to be technically feasible and economical. Secondly, a worldwide consensus on the most promising future energy carriers – both renewable electricity and H_2 – needs to be reached.

The arguments in favor of a future hydrogen economy (figure 34) are excellent, and the political commitment to move in this direction has been manifested in many initiatives. What is urgently needed now is a clear decision to start the transition from fossil to renewable energies and from gasoline to H_2.

We encourage politicians and policy-makers, energy officials and regulators, utility companies, development banks and private investors to firmly support the massive production of solar fuels – primarily H_2 – by taking concrete steps to enable future infrastructure and market development without delay.

We have only a short time window of opportunity to tackle and solve the critical problems of greenhouse gas emission and climate change. Solar fuels are part of the solution – they have the capacity to contribute to the energy supply and security and to help satisfy the energy needs of the world without destroying it.
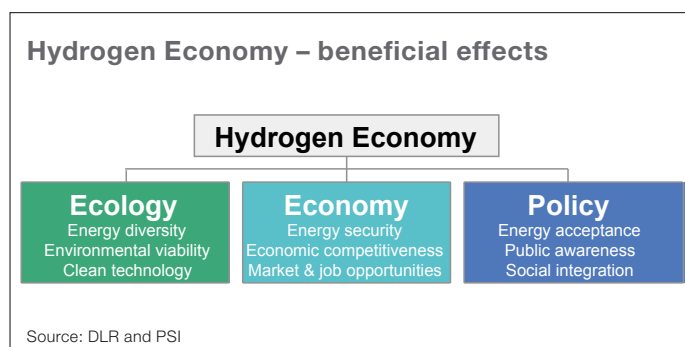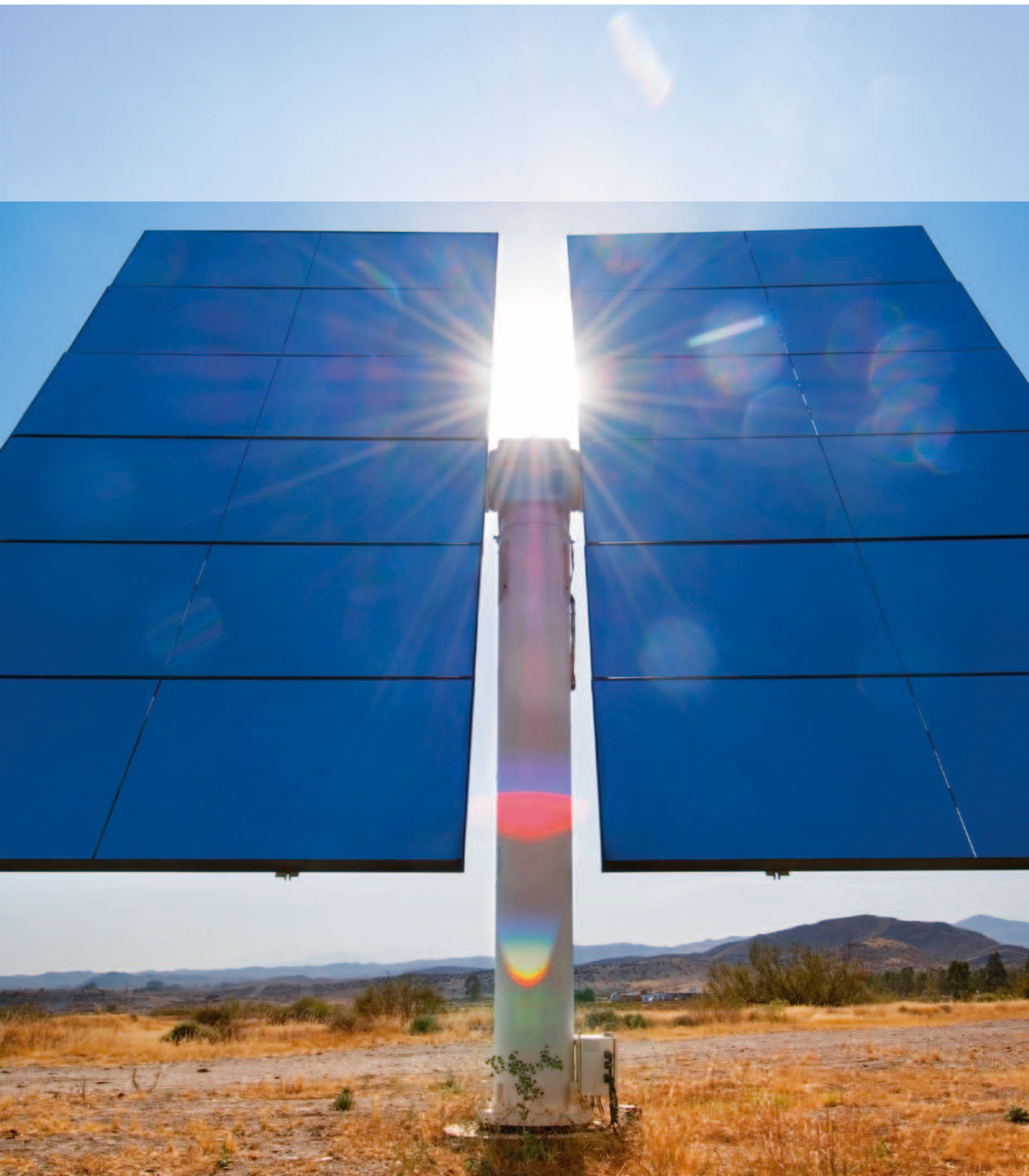

Hydrogen Economy – beneficial effects

Hydrogen Economy

Ecology	Economy	Policy
Energy diversity	Energy security	Energy acceptance
Environmental viability	Economic competitiveness	Public awareness
Clean technology	Market & job opportunities	Social integration

Source: DLR and PSI

Figure 34: Benefits of the hydrogen economy – **H_2 is a superb fuel for road transport, distributed heat and power generation, and for energy storage. It is widely considered to have a strong potential for use in future energy systems, meeting climate change, air quality, resource use goals, and securing energy supply.**

**Solar Power and Chemical Energy Systems
Implementing Agreement of the
International Energy Agency**