

The production of hydrogen by nuclear and solar heat

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Abstract

Both nuclear and solar energy represent significant carbon-free sources, which may contribute robust elements to a cleaner energy economy, to develop domestic energy sources for the purpose of energy security and stability, and to reduce national dependencies on imports of fossil fuels. Hydrogen, on the other hand, represents a fuel which is clean, powerful and an environmentally benign source of energy to the end-user. The current production of hydrogen is mainly based on hydrocarbons as feedstock, e.g. steam reforming of natural gas. Over the long term, however, water-splitting processes such as thermochemical cycles or high temperature electrolysis will become the principal basis of H₂ production. In Germany, R&D activities started in the past to investigate the possibility of utilising nuclear and solar primary energy input to provide process heat in the temperature range of 800-1 000°C. Such “allothermal” production processes promise significant savings of the fossil resources and a reduction in specific CO₂. In the long run, coupling nuclear or solar heat sources to thermochemical cycles enables bringing down CO₂ emissions to zero, since the only raw material is water. The paper will give an overview on the potential of nuclear and solar primary energy input to the hydrogen production processes of steam-methane reforming and thermochemical cycles of the sulphur family and describe respective research activities of the past and present in Germany.

Introduction

In 2004, the world's consumption of primary energy amounted to 10 224 million TOE, of which a dominant fraction of 88% originates from fossil fuels. Satisfying the need of future generations for a more sustainable development, the objective of climate change policies must be to enhance the trend towards renewable energies and other non-fossil alternatives. Both nuclear and solar energy represent significant carbon-free sources, which may contribute robust elements to a cleaner energy economy, to develop domestic energy sources for the purpose of energy security and stability and to reduce national dependencies on imports of fossil fuels. Hydrogen, on the other hand, represents a fuel which is clean, powerful and an environmentally benign source of energy to the end-user.

Currently hydrocarbons are the most applied feedstock for mass production of hydrogen and will continue to be so for the next decades. Steam reforming of natural gas covering world wide about half of the hydrogen demand, is one of the essential processes in the petrochemical and refining industries. In conventional plants, the process heat required is provided by burning a part of the natural gas (autothermal). An alternative option would be to provide an external process heat source (allothermal) and, thus, saving resources and reducing specific CO₂ emissions. In the long term, the clean production methods will most certainly be based on water as feedstock. High temperature electrolysis reduces the electricity needs up to about 30% and could make use of high temperature heat and steam from an HTGR. In thermochemical (hybrid) cycles, a series of thermally-driven chemical reactions takes place where water is decomposed into hydrogen and oxygen at moderate process temperatures.

Nuclear and solar primary energy may serve different purposes, if used for the clean production of hydrogen, and they complete each other. While nuclear energy could play a major role in centralised H₂ production on a large scale at a constant rate, solar and other renewable energy sources with their low-density energy and typically intermittent operation mode will be preferable for dispersed systems of H₂ generation. In addition, both types can be applied to generate electricity and provide it to the grid at any place. Also natural gas could be used as feedstock for centralised and decentralised H₂ production. Both grid-bound forms of energy are consumer-friendly and widely available.

Nuclear and solar primary heat source

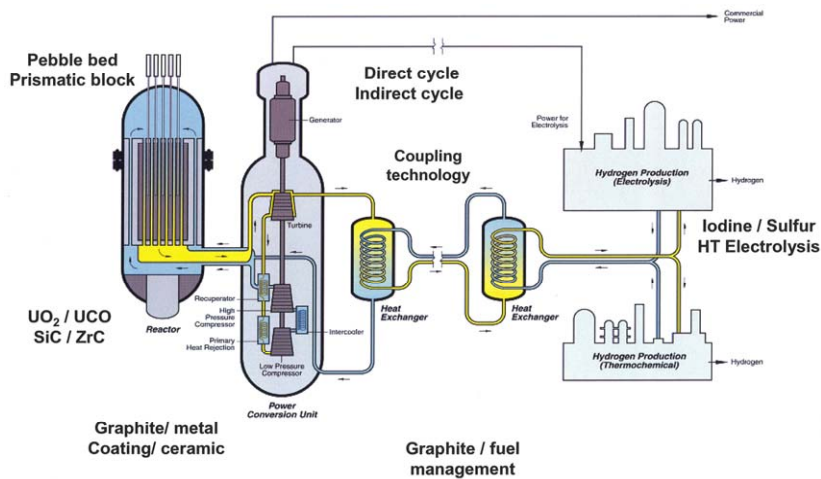
Before a CO₂-free hydrogen production will be established on the future energy market, it is desirable for a transition phase to replace autothermal with allothermal processes, where an adequate heat carrier system is taken to transfer heat from outside into the hydrocarbon splitting process. The output of hydrogen per unit feed stock may be increased by 30-40%. This strategy includes the production not only of hydrogen, but also of liquid energy alcohols in a subsequent step, e.g. methanol synthesis.

Nuclear process heat reactor

Future nuclear reactors are expected to be further progressed in terms of safety and reliability, proliferation resistance and physical protection, economics, sustainability (GIF, 2002). One of the most promising nuclear reactor concepts of the next generation (Gen-IV) is the VHTR. Characteristic features are a helium-cooled, graphite-moderated thermal neutron spectrum reactor core with a reference thermal power production of 400-600 MW. Coolant outlet temperatures of 900-1 000°C or higher are ideally suited for a wide spectrum of high temperature process heat applications.

The co-generation of hydrogen is done by connecting the VHTR to a H₂ production plant via an intermediate heat exchanger (Figure 1). Top candidate production methods, considered presently by various countries, are the sulphur-iodine (S-I) or alternative thermochemical (hybrid) cycles and high temperature electrolysis (HTE). Assuming a plant availability of 90% and an overall conversion efficiency of (aimed at) 50%, the system would have a capacity of 27.4 t/d of hydrogen (higher heating value) per 100 MW of nuclear thermal power.

The technology of the VHTR benefits from the broad experience gained from respective research projects in the past such as the German Prototype Nuclear Process Heat (PNP) project. But also HTGR operation like the currently operated HTTR in Japan and the HTR-10 in China, as well as comprehensive R&D efforts which were recently initiated in many countries to investigate HTGR systems in connection with nuclear hydrogen production provide valuable knowledge.

Figure 1: Very high temperature reactor with hydrogen production system

Heat from concentrating solar systems

To achieve process temperatures in the range beyond 800°C, in principle, two mature solar technologies are available: solar dishes and central receiver systems. The former technology is limited in size (< 100 kW per unit) and suitable for decentralised applications. For a commercial scale-up, solar-thermal central receiver systems (CRS), also called solar tower systems, can achieve the required temperature level and multi-megawatt power levels. CRS have been under investigation since the 1970s, when various installations were planned and built world wide. One example is the research plant CESA-1. Several hundred tracked and typically slightly shaped mirrors, so-called heliostats, are arranged around a tower. They redirect and concentrate solar radiation to one spot on top of the tower, where the radiation is collected and converted to process heat to power a steam or gas turbine. Alternatively the heat can be used for high temperature (600-2 000°C) chemical reactions either directly at the tower platform or by transferring the heat via a heat transfer fluid to a chemical plant.

In 2007, the first commercial CRS was brought into operation and connected to the power grid, the 10 MW(e) plant PS10 near Seville in Spain (Figure 2). It has a 75 000 m² heliostat field of 624 heliostats with 120 m² each, a saturated steam receiver and a 30 minute thermal storage. It generates 20 GWh/yr of solar electricity. The construction of a second power tower, the 20 MW PS20, was completed nearly at the same site. The Spanish Abengoa group is building at their Plataforma Solar of Sanlucar la Mayor (PSSM) the world's largest solar platform with a total CSP capacity of 300 MW, including the 11 MW PS10, two 20 MW PS20 power towers, and five 50 MW parabolic trough plants. A high temperature prototype plant has recently been inaugurated in Jülich, Germany. It features a DLR development, an open volumetric receiver made of ceramic honeycomb structures.

Figure 2: View of Abengoa's Plataforma Solar Sanlucar La Mayor near Seville, Spain, with the 11 MW PS10 and construction of the 20 MW PS20 (Abengoa Solar)



Heat decoupling

The combination of an external heat source with chemical processes will need a device to decouple the heat from its origin to the heat utilisation system. In the case of a nuclear plant, the intermediate heat exchanger (IHX) must provide a clear separation between nuclear plant and heat application. Under normal operating conditions, the IHX prevents the primary coolant from accessing the process plant and, on the other side, process gases from being routed through the reactor containment, thus limiting or excluding a radioactive contamination of the product (*e.g.* by tritium). Furthermore the physical separation allows for the heat application facility to be conventionally designed meaning easy maintenance and repair works under non-nuclear conditions. The VHTR will have three heat exchanging levels (see Figure 1), from the primary side to an intermediate circuit, then to a heat delivery system of the chemical plant before transferred to various chemical processes.

Different technologies for heat exchanging components designated for nuclear applications have been developed in the past. Two IHX components, one with a helical tube bundle and the other one with U-tubes, designed for a power level representative for large and medium-sized plants were constructed by German companies and tested under conditions of nuclear process heat applications in a 10 MWt component test loop (KVK) with 950°C helium on the primary and 900°C helium on the secondary side (Harth, 1990). In the Japanese HTTR, a 10 MW helically coiled IHX component is presently being operated under the same conditions. New IHX designs currently under investigation for nuclear applications are the so-called printed circuit heat exchangers (PCHE), composed of metal plate layers containing alternately coolant channels for the primary and secondary fluid and stacked together to a solid, all-metal core. PCHE are highly compact, robust and thermally efficient.

In the case of solar powered systems, decoupling of heat source and chemical plant facilitates the compensation of fluctuating and intermittent available power input. This is particularly important if units of the chemical system require steady-state conditions over long periods. Beyond that decoupled systems allow for an easier integration of thermal and chemical storage units to compensate daily or seasonal variation of solar supply. The same applies for hybrid operation, *i.e.* the combination with burner firing or with a nuclear heat source.

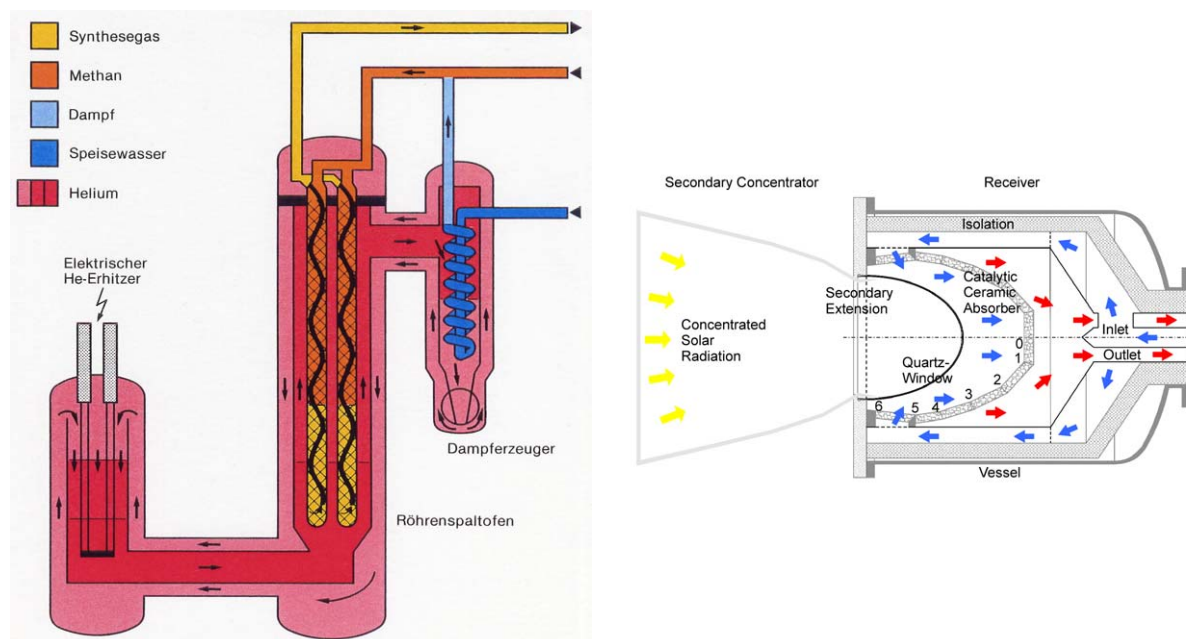
For both nuclear and solar systems, appropriate material selection will be essential. A qualification programme for high temperature metallic materials must demonstrate their good long-term performance. In the nuclear case, candidate materials will be exposed to helium of 1 000°C with impurities such as CO, CO₂, H₂, H₂O, CH₄ and to neutron irradiation. The experience gained so far has disclosed that the technical solution of material problems requires further efforts in the future.

Allothermal steam reforming of methane

Nuclear steam reforming

Within the German projects on the application of nuclear process heat for steam reforming of methane, the nuclear high-temperature reactor design, the development of major components in the 125 MW scale was confirmed with the operation of the EVA/ADAM test facilities. The follow-up test facility, EVA-II, represented a complete helium circuit containing a bundle of reformer tubes (Nießen, 1988). The nuclear heat source was simulated by an electrical heater with a power of 10 MW to heat up helium gas to a temperature of 950°C at 4.0 MPa. As can be seen from the schematic on the left-hand side of Figure 3, the principal components of heater, reformer bundle and steam generator were housed in separate steel pressure vessels in a side-by-side arrangement. The vessels of helium heater and steam reformer were connected by a co-axial helium duct of 5 m length.

Two reformer bundles both with convective helium heating were investigated in the EVA-II facility. They differed in the way of channelling the helium flow. The first bundle tested was a baffle design tube bundle for a power of 6 MW consisting of 30 tubes. Inside each splitting tube, an internal helical tube with an outer diameter of 20 mm was used to recirculate the product gas. The second steam reformer bundle tested was of annulus design for a power of 5 MW. It consisted of 18 tubes with each splitting tube placed inside a guiding tube channel, where the hot helium was flowing upwards through the annular gap. The specific data of both tubes and catalytic systems were very similar compared to components planned for nuclear applications. Also the loads imposed on the supporting

Figure 3: Schematic of the EVA nuclear steam reformer (left) and the SOLASYS solar reformer (right)

structures were characteristic to the nuclear case. The tests have demonstrated the industrial feasibility of the components for a large-scale hydrogen production and have been providing results of high importance for any allothermal chemical process (Nießen, 1988).

Solar steam reforming

At the Weizmann Institute of Science (WIS) in Rehovot, Israel, a 480 kW solar steam/ CO_2 methane reformer was designed, constructed and operated intermittently during two years as a complete solar heat pipe system (Epstein, 1996). The insulated pentagon-shaped reformer box contained eight tubes (length: 6.4 m, diameter: 51 mm) in two rows made of Inconel 617. The 4.5 m long heated section contained the catalyst. In the case of CO_2 reforming, which is thermodynamically more efficient and easier to operate under changing conditions (as in solar), the catalyst was 1% Ru on alumina. The tubes were directly heated by the solar radiation entering through a 600 mm aperture and reflected from the walls of the enclosure. The heat take-up was found to be quite uniform with no tendency of local overheating. The maximum average surface temperature was $\sim 930^\circ\text{C}$ and the heat flux in the range of 80-84 kW/m^2 . The flow rate of the process gas, which could be varied over a wide range of $\text{CO}_2/\text{H}_2\text{O}/\text{CH}_4$ feed mixtures, was limited to ~ 300 kg/h due to higher than expected pressure drop. It entered the reformer tubes at $\sim 500^\circ\text{C}$ and 1.6-1.8 MPa and left at ~ 800 - 830°C .

CSIRO in Australia has successfully demonstrated steam reforming of natural gas using solar energy. The test rig used a commercially-available solar dish concentrator (Edwards, 1996). The test rig is equipped with a tube-type receiver-reactor containing spiral-shaped metallic absorber tubes filled with a catalyst bed. Investigations into using a mix of steam/ CO_2 for reforming the high CO_2 natural gas sources in Australia are being considered.

The project ASTERIX (Langnickel, 1993) has demonstrated that solar energy can be used for steam reforming of methane and be coupled to conventional production lines for mass production of synthesis gas. The ASTERIX plant was a small but complete chemical production plant installed at a solar tower (CESA-1) of the Plataforma Solar de Almería. In co-operation with research centres and industrial partners from Germany and Spain, DLR developed process units for steam generation, pre-heating of feed, reactor, cooling and water separation and adapted it to the solar tower. The central unit, the reformer, included registers of tubes convectively heated by hot air. The heat transfer

medium air was provided by the so-called GAST loop (Becker, 1989) at temperatures of up to 980°C and a pressure of 0.8 MPa. It made use of the GAST receiver equipped with ceramic tubes, which was located on a platform of the CESA-1 tower and heated by concentrated radiation from the adjacent heliostat field. The tests were carried out at a power level of 150-200 kW (power absorbed by the reformer) over several hundreds of hours.

An alternative concept is the so-called direct absorption concept, which applies solar-receiver reactors. According to this concept a solar reformer was developed by DLR in the SOLASYS project (Tamme, 2003) based on earlier experiences in the projects SCR and CAESAR (Bauer, 1994). It allows the concentrated radiation to penetrate through a transparent aperture into the reformer, where it is absorbed directly by the irradiated absorber. The reaction gases pass through the absorber which serves simultaneously as a heat transfer unit and as support for the catalyst. The quartz window, used as aperture closure, enables the reformer to be operated under pressure. A schematic of the solar receiver is shown in Figure 3 (right). The reformer was operated up to 0.9 MPa and 780°C. The pilot reformer was tested at a power level of about 300 kW(th) at the solar tower of the Weizmann Institute of Science in Rehovot, Israel. In a follow-up project SOLREF (Möller, 2006), the operation conditions will be about 1.5 MPa and 950°C.

EVA-ADAM energy transportation system

Steam reforming reactions in combination with their reverse reaction, the methanation, can form the basis of a long-distance energy transportation system using the energy carrier hydrogen. Such a system in connection with an HTGR was first suggested in the 1970s by FZJ and became known under the acronym “EVA-ADAM”. Its principle is the storage of energy in form of latent heat by an endothermic chemical reaction in a gas, the transportation of the gas to the consumer’s site, where in the exothermic reverse reaction, the stored energy is liberated in the form of heat for further utilisation. The transportation system is a closed cycle.

Within the project NFE, German acronym for “nuclear long-distance energy”, comprehensive R&D activities were conducted to utilise process heat from an HTGR for the production of synthesis gas by steam methane reforming. The reforming part in the EVA experimental facility was already previously described. The counterpart of EVA is ADAM, acronym for “plant with three adiabatic methanation reactors”, where in the reverse reaction the original educts methane and water plus heat can be regained. After separation the methane can be piped back to the EVA site to close the cycle, while the steam could be used in other process applications.

The WIS solar reformer described above was completed by a methanation system to demonstrate the EVA-ADAM energy transportation providing valuable information for system improvements in the future.

Thermochemical water-splitting

In water-splitting processes, the heat input is directly converted into the chemical energy of the hydrogen. Thermochemical cycles are composed of several reaction steps which, in the sum, lead to a decomposition of water into hydrogen and oxygen. The supporting chemical substances are regenerated and recycled, and remain – ideally – completely inside the system. The only inputs are water and heat. The mainly welcome by-product is oxygen that can be further used as a commodity.

Numerous cycles have been proposed in the past and investigated in terms of their characteristics like reaction kinetics, thermodynamics, separation of substances, stability, processing flow scheme and cost analysis. Only a few, however, were deemed sufficiently promising and thus worth further investigation. Among the cycles whose partial reactions are being investigated in more detail also with respect to their coupling to an HTGR or a solar heat source, are those of the sulphur family originally developed in the USA (Norman, 1981), later pursued and modified by various research groups.

Other processes considered worth of further investigation are the so-called Westinghouse process, a sulphuric acid hybrid (HyS) cycle where the low-temperature step runs in an electrolysis cell to produce the hydrogen, the CuCl cycle, a lower temperature (< 500°C) hybrid cycle investigated at ANL, or the so-called UT-3 process based on a four-step cycle with calcium and bromine.

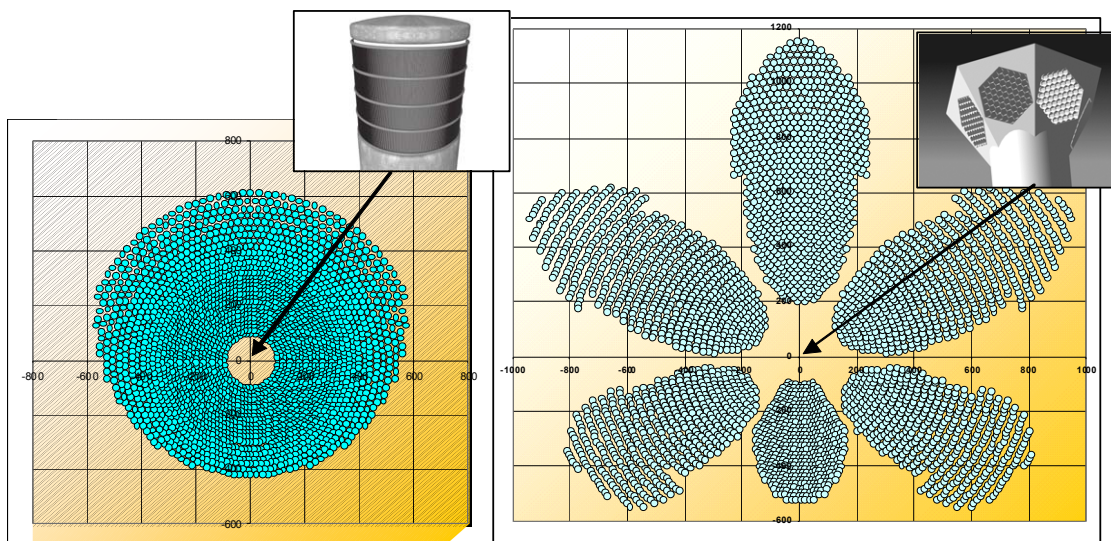
Solar-driven HyS cycle

A special focus of the development regarding solar thermochemical processes is the high temperature endothermic decomposition of sulphuric acid. The solarisation of this step was part of the R&D activities of the European project HYTHEC (HYdrogen THERmochemical Cycles) with an emphasis on components development and process improvement (Nogliki, 2009).

A prototype receiver-reactor for the solar decomposition of sulphuric acid has been developed and qualified. The concept of a volumetric receiver-reactor has been proven feasible in practice. High conversion almost up to the maximum achievable can be achieved with a platinum catalyst. Further investigations with a less expensive catalyst, iron oxide, provided evidence of the potential of such material.

The potential of solar-powered processes was investigated for the HyS hybrid cycle including flow sheeting, component sizing and techno-economic analyses. The studies were based on an annual average thermal power of 50 MW(th) for a region close to Lake Nasser in Egypt (Figure 5). A step-wise iteration led to a basic flow sheet for this specific process version, which formed the basis for the assessment of heat balance and component sizing. The determining factors for the economics of the process were evaluated and alternatives were developed. A first optimisation of the process indicated that for the electric power generation, the application of a separate receiver is by far advantageous compared to an integrated version.

Figure 5: Results of a design study – potential heliostat field set-ups for an annual average receiver power of 50 MW(th)



Solar-nuclear hybrid systems

Solar-nuclear hybrid variants of the HyS cycle process have also been investigated to explore potential synergies. Two hybrid plant designs were analysed:

- In the first, trivial case of hybridisation, all thermal energy required for the process steps, mainly vaporisation and splitting of sulphuric acid, is delivered by solar, whereas all electric consumers, mainly electrolysis are driven by nuclear. This way, the electrolysis can be run continuously, while the sun provides the high-temperature heat for the splitting process.
- In the second approach, an HTGR and a solar reactor are assumed to provide all thermal energy needed. In addition, nuclear energy drives the electric power generation for the electrolysis of all SO_2 produced. It is thus a constantly-running nuclear-powered HyS cycle which consumes additional solar produced SO_2 . This way, each technology is well used, but there is a high degree of redundancy regarding the plant components.

A preliminary evaluation of the hydrogen production costs based on solar, nuclear and hybrid operation lead to the following results: Small plants are powered most favourably by solar energy, while nuclear plants are most economic at high power levels > 300 MW(th); hybrid systems may have their niche in the mid-range of 100 to 300 MW(th).

The recently started EU FP7 project HycycleS (Roeb, 2009) builds up upon the results of HYTHEC and aims at the qualification and enhancement of materials and components for key steps of solar- and nuclear-powered thermochemical cycles. Emphasis is placed on materials and components for sulphuric acid evaporation, decomposition and sulphur dioxide separation. The suitability of materials and the reliability of the components will be shown in practice by decomposing sulphuric acid and separating its decomposition products in scalable prototypes.

Conclusions

For future hydrogen production based on allothermal reforming processes, it will be important and necessary to retrieve and re-evaluate all data and manufacturing experiences obtained in the frame of the above-mentioned large-scale experiments in terms of a technological and economical appraisal. The experience and knowledge on nuclear steam methane reforming, which were bundled in the Research Centre Jülich, was also significantly influencing the development of solar steam reformer systems. Both nuclear and solar can be adjusted to the typical thermal power sizes of present modular power plant designs of up to 600 MW. It will thus support the design of novel components up to an industrial scale, which are required in allothermal reforming processes as well as in high temperature electrolysis, or in thermochemical processes. The decomposition of sulphuric acid which is the central step of the sulphur-based family of those processes, especially the hybrid sulphur cycle and the sulphur-iodine cycle will be further investigated in the recently started EU project HycycleS, which is co-ordinated by the German Aerospace Centre.

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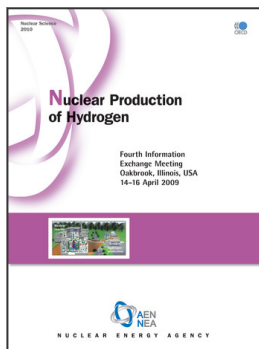
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