HYDROGEN PRODUCTION PROCESS REQUIREMENTS AND NUCLEAR REACTOR OPTIONS

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INTRODUCTION

Nuclear energy has the potential to become the primary method for the production of hydrogen (H_2). However, production of H_2 on a large scale imposes a set of technical, system, and economic requirements. An evaluation was made to define the requirements for feasibility, identify the nuclear reactor concepts that could be used for H_2 production, and assess the capability of each concept. The evaluation establishes which reactors are most likely to be candidates for H_2 production and the major barriers each must overcome to perform this mission.

REQUIREMENTS FOR HYDROGEN PRODUCTION

The choice of method to produce H_2 using a nuclear reactor depends upon multiple factors [1]: (1) scale of operation, (2) H_2 plant requirements, (3) nuclear reactor development status, and (4) nuclear fuel cycle requirements.

Scale of operation. Nuclear plants are economical for industrial and utility applications only when they are built in large sizes. If H_2 production from nuclear energy is to be viable, the scale of H_2 production must match the economic scale of nuclear energy production. Changes within the last decade have eliminated the historic mismatch between H_2 demand and the scale of nuclear energy production. These changes include (1) growth in worldwide H_2 consumption to 50 million t/year, (2) expected future H_2 growth rates of 4 to 10%—primary because of the decreased quality of crude oil and the need for more hydrogen to upgrade the crude oil to gasoline—and, (3) development of pipelines that allow for very large H_2 production units. The world-class H_2 plants that are under construction have production capacities of 200 million standard cubic feet of H_2 per day (scfh/d). New plants have been announced with capacities of 300 million scfh/d [1200 MW(t) of H_2 energy, based on the higher heating value). The next generation of ammonia plants (large H_2 consumers) are expected to produce 3000 t/day, equivalent to 200 million scfh/d. Most of these plants use steam reforming of natural gas to produce H_2.

Several processes are being developed to produce H_2 from water and high-temperature heat from nuclear reactors (see below). If such a process is 50% efficient, a 2400-MW(t) reactor would be required to produce 300 million scfh/d. In terms of energy flows, the size of today's H_2 production plant is now equivalent to the size of a nuclear power plant. The nuclear reactor output should match the energy requirements for the H_2 production facility to minimize costs—all other factors being equal. This defines the preferred size for the nuclear reactor.

Hydrogen Plant Requirements. Three approaches [1] have been identified for the efficient production of H_2 using nuclear energy. The first approach, nuclear-assisted steam reforming of natural gas, uses nuclear heat to reduce the amount of natural gas needed to produce a given quantity of H_2. The second approach, hot electrolysis, involves electrolysis of water at high temperatures to produce H_2 and oxygen. At high temperatures (1) some of the energy input is heat rather than electricity and (2) internal loses in the electrolyzer are reduced. Finally, thermochemical cycles use a series of chemical reactions and high-temperature heat to convert water to H_2 and oxygen. The thermal-to-H_2 efficiency of these processes is ~50%. All of the processes impose similar requirements on the high-temperature heat source.

Today, H_2 is produced primarily from the steam reforming of natural gas (net reaction: $CH_4 + 2H_2O$ Y $CO_2 + 4H_2$). Steam reforming is an energy-intensive endothermic low-pressure process requiring high-temperature heat input. The natural gas is (1) used as the reduced chemical source of H_2 and (2) burned to produce heat to drive the process at temperatures of ~800EC. The amount of natural gas required for steam reforming can be significantly reduced when heat is provided by a nuclear reactor. The Japan Atomic Energy Research Institute [2] is currently preparing to demonstrate the production of H_2 by steam reforming of natural gas with the heat input provided by its recently completed High-Temperature Engineering Test Reactor (HTTR). The nuclear reactor provides the energy to replace heat from a gas flame and thus reduces the amount of natural gas required to produce a unit of H_2. Because this system uses standard H_2 production technology, it represents the near-term nuclear H_2 technology. Only nuclear reactor issues must be addressed. For Japan and other countries with high-cost natural gas, economic analysis indicates that H_2 from nuclear-assisted steam reforming of natural gas will have lower costs than H_2 from natural gas alone using near-term technologies.

About a 100 thermochemical processes have been identified to produce H_2 from heat and water. The leading candidate is the sulfur-iodine process which consists of three chemical reactions:

$2H_2SO_4$: $2SO_2 + 2H_2O + O_2$ (Heat input at 800EC)

$2HI$] $I_2 + H_2$ (Heat input at 450EC)

$I_2 + SO_2 + 2H_2O$] $2HI + H_2SO_4$ (Heat rejection at 120EEC)

Overall, heat and water are added to the process to produce H_2 and oxygen. Many of the papers presented at this conference describe ongoing research on this process. All other chemical reagents are fully recycled. The first step, the catalytic decomposition of sulfuric acid, is the high-temperature, energy-intensive step. It is an equilibrium chemical reaction that can proceed in either direction. High temperatures and low pressures drive the reaction to the right toward completion. At 10 bars, the reaction is estimated to go to 31% completion at 625EC, 79% completion at 725EC, and 99% completion at 925EC.

Preliminary economic analysis indicates that the thermochemical cycles will ultimately be the low-cost option. Although traditional low-temperature electrolysis is a proven technology, it is less efficient than the high-temperature processes. This finding parallels that from experience with production of H_2 from natural gas. Steam reforming of natural gas, a thermochemical cycle, has proven much more economic for H_2 production than electrolysis of water with the electricity produced from natural gas. With a light-water reactor (LWR), the thermal-to-hydrogen energy conversion efficiency is ~24%—half that expected for an efficient thermochemical cycle. This value represents the product obtained by multiplying the typical LWR efficiency of converting thermal energy to electricity (33.3%) by a typical efficiency of electrolysis in an industrial system (~72%). Japanese researchers estimate that the cost of nuclear thermochemical hydrogen production may be as low as 60% of that for nuclear H_2 production by the electrolysis of water; consequently, thermochemical processes have received the most attention.

While there are a variety of nonelectrolyzer processes to produce H_2, all of them (nuclear-assisted steam reforming of natural gas, hot electrolysis, and thermochemical) impose somewhat similar technical requirements on the reactor.

Temperature. All the potentially low-cost H_2 production methods require high temperatures (750 to 900EC).

Temperature range of delivered heat. All of the H_2 production methods involve an endothermic high-temperature chemical dissociation reaction that operates over a relatively small temperature range. Delivery of the heat under such conditions maximizes process efficiency.

Pressure. The high-temperature, energy-consuming chemical reactions for H_2 production go to completion at low pressures while high pressures reverse the desired reactions. The H_2-nuclear interface should be at low pressure to (1) minimize the risk of pressurization of the chemical plant with release of large inventories of toxic chemicals and (2) minimize the need for high-strength, high-temperature materials.

Isolation. To ensure that potential accidents in one facility do not impact the other, the nuclear and chemical facilities should be separated by a significant distance. In the 1970s, researchers in Germany examined the use of various reactors to provide high-temperature heat to the chemical industry. They concluded that the last two requirements could be best met by using an intermediate molten-salt or other high-heat-capacity (low heat losses and small piping), low-viscosity (low pumping cost) liquid for the heat transfer loop between the reactor and the chemical plants.

Nuclear Reactor Development Status. No commercially available reactor capable of meeting the requirements of H_2 production presently exists. The difficulty of developing a reactor for H_2 production will strongly depend upon the choice of reactor.

Nuclear Fuel-Cycle Requirements. The fuel cycles associated with nuclear reactors are significantly more complex than those associated with natural-gas plants. The fuel cycle requirements can, under many scenarios, control the choice of reactor system.

Technology status. Only limited research and development (R&D) is required for some fuel cycles. In other cases, the R&D requirements are very large. This impacts the practical choices for future reactors.

Scale of deployment. If nuclear-generated H_2 is to have a significant impact on the market, it must be deployed on a large scale. Although rapid large-scale deployment of some reactors and associated fuel cycles is possible, others (even if the technology is available) require many decades for large-scale deployment.

REACTOR OPTIONS

Five reactors meet the minimum requirement for production of H_2: operation at the necessary temperatures. A brief description of each is provided herein with appropriate references that provide more detailed information. These advanced reactors are designed with passive safety systems that do not require diesel generators, and other such active components to assure safety.

Very-High-Temperature Reactor (VHTR). The VHTR [3, 4] is a higher-temperature version of the high-temperature gas-cooled reactor (HTGR). The solid fuel consists of microspheres of uranium oxide or carbide with multiple refractory coatings that retain fission products. The microspheres are embedded in a graphite matrix with cooling channels. High-pressure helium, the reactor coolant, is used to transfers heat from the reactor core to the H_2 production facility. The energy output is limited to ~600 MW(t)—the largest size compatible with its passive safety systems. There has been renewed utility interest in HTGRs and VHTRs for electric generation. The high-pressure helium can be directly coupled to a direct-cycle gas turbine to produce electricity—potentially a major simplification and improvement in electric power plant design. The reactor size matches the capabilities of large gas turbines.

Several demonstration HTGRs have been built for electricity production. Japan recently started the HTTR [2], a small VHTR [30 MW(t)] to develop the technology for efficient production of H_2 and electricity. The helium reactor exit temperature is 950EC. One goal of this test reactor is to demonstrate nuclear-assisted steam reforming of natural gas. The reactor will later be used to demonstrate thermochemical H_2 production.

Molten Salt Reactor (MSR). The MSR [3] uses a liquid molten-fluoride salt as fuel and coolant. The uranium or plutonium fuel is dissolved in the molten salt. Two test reactors were built. In the 1950s, the Aircraft Reactor Experiment operated normally with molten salt exit temperatures of 815EC with peak operating temperatures up to 860EC and very low primary system pressures. Work continued on MSR technology for power applications until 1976. The reactor can be built in large sizes with passive safety systems.

Advanced High-Temperature Reactor (AHTR). The AHTR [1] uses a solid coated-particle graphite-matrix fuel and a clean molten-fluoride salt coolant. It combines the fuel from the VHTR with the coolant (minus dissolved fuel and fission products) of the MSR. Graphite fuels are compatible with fluoride salts. There is a century of industrial experience with the compatibility of graphite and molten fluoride salt; aluminum is electrolytically produced from cryolite ($3NaF$-AlF_3) in very large graphite baths at ~1000EC. The Ni-based high temperature alloys used in the VHTR (at the HTTR) are similar to those developed for molten salts. The AHTR is a new reactor concept and the first such reactor designed explicitly to match the requirements for H_2 production. The reactor can be built in large sizes with passive safety systems.

Lead-Cooled Fast Reactor (LFR). The LFR [3] uses a solid metal or nitride fuel with metal cladding and molten lead (or a lead alloy) as the reactor coolant to transfer heat from the reactor core to the H_2 production facility. The technology was originally developed by Russia for nuclear submarine propulsion. Several submarines with this power system were built and operated for limited times. The operating temperatures of these reactors were near 500EC. Lead has a very high boiling point; thus, this reactor could be designed to operate at very high temperatures. However, serious corrosion problems have occurred at lower temperatures and new materials are required for higher-temperature operation. The LFR requires a closed fuel cycle. The reactor can be built in large sizes with passive safety systems.

Gas-Cooled Fast Reactor (GFR). The GFR [3] uses an advanced fuel (several options being investigated) and high-pressure helium as the coolant. It couples the helium coolant technology of the HTGR and VHTR with the fast-neutron reactor technology originally developed for sodium-cooled fast reactors and LFRs. The GFR requires a closed fuel cycle.

COMPARISONS OF REQUIREMENTS VERSUS REACTOR OPTIONS

The requirements for H_2 production were compared with the characteristics of the different reactor systems. The relative rankings are shown in Table 1, where "high" indicates a good match between the requirement and the reactor characteristics. Significant uncertainties exist in many of these comparisons because of the limited work that has been done on H_2 production using nuclear energy. Consequently, a more detailed ranking is not justified at this time. The basis for each conclusion is described below.

Table 1. Relative Ranking of Different Reactors for Large-Scale Hydrogen Production

Parameter	AHTR	VHTR	MSR	LFR	GFR
Scale of operation	high	low	high	high	low
Hydrogen plant requirements					
Temperature	high	high	high	low	low
Temperature range of delivered heat	high	low	high	high	low
Pressure	high	low	high	high	low
Isolation	high	low	middle	high	low
Nuclear reactor development status	high	high	low	low	low*
Nuclear fuel-cycle requirements					
Technology status	high	high	low	low	low
Scale of deployment	high	high	low	low	low*

*Ranking significantly below other concepts.

Scale of operation. The newest H_2 production facilities on order have a capacity of ~300 million scfh/d. A 2400-MW(t) reactor would be required to produce this quantity of H_2. Because of economics of scale and simplicity of plant design, the optimum plant configuration is to match the size of the reactor to that of the chemical plant (i.e., a single reactor with a single-train H_2 plant). The AHTR, MSR, and LFR (all low-pressure reactors) would normally be built in this size range with passive safety systems; consequently, they are given a high rating. Current designs of the VHTR and GFR are for smaller reactors. These two reactors use high-pressure helium coolant; thus, vessel pressure limitations place constraints on the reactor design. If passive nuclear safety systems for the VHTR and GFR are to be used (a highly desirable feature in terms of economics and public acceptance), the size limit is typically near 600 MW(t).

Hydrogen Plant Requirements. There are four technical requirements that any reactor used to make H_2 must meet.

Temperature. All the potentially low-cost H_2 production methods require high temperatures (750 to 900EC). All of the reactor concepts described herein have the potential to meet this requirement. However, operation at in this temperature range has been demonstrated only fuels and coolants used by the AHTR, VHTR, and MSR—not for the LFR or GFR fuels.

Temperature range of delivered heat. All of the H_2 production methods involve endothermic high-temperature chemical dissociation reactions that operate over a small temperature range. The temperatures are near the limits of current materials; thus, peak temperatures should be minimized to reduce materials requirements. This can be accomplished by using a reactor with a liquid coolant.

Liquid coolants have good heat transfer capabilities and low pumping power costs in comparison with gas coolants. Liquid coolants minimize within the reactor core the fuel clad-coolant temperature drop, and thus keep the fuel cooler for the same reactor coolant exit temperatures. Liquid-cooled reactors can deliver most of their heat at near-constant temperatures while gas-cooled reactors generally deliver their heat over a wide range of temperatures to reduce pumping power costs. This implies the reactor exit temperatures and peak heat exchanger temperatures in a liquid-cooled reactor will be significantly less than a gas-cooled reactor for heat delivered to the H_2 plant at a fixed temperature.

Some industrial examples (Table 2) demonstrate these differences between gas-cooled and liquid-cooled nuclear reactors. The General Atomics gas-cooled HTGR (the GT-MHR) has a ΔT across the reactor core of 359EC, while the British Advanced Gas-Cooled Reactor (Hinkley Point B) has a ΔT of 355EC. Liquid-cooled reactors typically have much-smaller temperature increases across the reactor core. The Point Beach pressurized-water reactor (PWR) has a ΔT across the reactor core of 20EC, while the French liquid-metal fast reactor (Super Phenix) has a ΔT of 150EC. A liquid-cooled reactor can deliver all of its heat with small temperature differences (20 to 150EC) between (1) the hottest temperatures in the reactor coolant, piping, and heat exchangers and (2) the maximum temperature of the chemical reagents in the H_2 production facility.

If heat is needed at 750EC, the maximum temperature of the gas coolant in a gas-cooled reactor and heat exchangers may exceed 1000EC whereas that of the liquid coolant in a liquid-cooled reactor will only be 20 to 150EC higher—depending upon the design. This can significantly reduce the high-temperature demands on materials. Liquid-cooled reactors include the AHTR, MSR, and LFR

Table 2. Temperature Increases Across Reactor Cores for Different Reactor Coolants

System	Delta T. Inlet to Outlet (EC)	Inlet T (EC)	Outlet T (EC)	Coolant
GT-MHR	359	491	850	Gas (Helium)
Advanced Gas Reactor (Hinkley Point B)	355	310	665	Gas (CO_2)
PWR (Point Beach)	20	299	319	Liquid (Water)
Liquid Metal Reactor (Super Phenix)	150	395	545	Liquid (Sodium)

Pressure. The chemical reactions for H_2 production go to completion at low pressures, while high pressures reverse the desired reactions. Low-pressure reactor coolants are preferred that (1) minimize material strength requirements for heat exchangers between the reactor and chemical plant and (2) avoid the potential for overpressurization of the chemical plant in the event of heat exchanger failure. These consideration favor the use of low-pressure liquid cooled reactors: the AHTR, VHTR, and MSR.

Isolation. To ensure that potential accidents in one facility do not impact the other, the nuclear and chemical facilities may be separated by a significant distance. The most efficient methods to transfer heat from the reactor to a chemical plant over some distance involve the use of high-heat-capacity liquid heat transfer agents. In the 1970s, researchers in Germany examined the use of various reactors to provide high-temperature heat to the chemical industry. They concluded that this requirement could be best met by using an intermediate heat transfer loop with a high heat capacity liquid with low pumping costs. Molten salts are currently used in the chemical industry for this purpose. A reactor with molten salt best couples with such a heat transfer system to minimize temperature drops and heat loses. Liquid-cooled reactors such as the AHTR, MSR, and LFR best meet this requirement.

Another factor, unique to MSRs, "down rates" this reactor in comparison with other liquid-cooled reactors in terms of isolation. In an MSR, the fuel is dissolved in the molten salt. The fission process produces tritium, the radioactive form of H_2. This places an additional requirement on the intermediate heat transfer loop to ensure that tritium does not reach the H_2 production facility. Significant work has been conducted to develop methods to ensure tritium does not cross the heat exchanger. Most of this work is associated with development of fusion reactors that have very large tritium inventories. It is unclear how serious this issue is.

Nuclear Reactor Development Status. Only two reactors are potential near-term candidates for production of H_2: the AHTR and VHTR. Both reactors use graphite-matrix coated-particle fuel, the only nuclear fuel that has been demonstrated on a significant scale at the required operating temperatures for H_2 production. Fuel development is usually the most complicated and time consuming activity in development of a reactor.

Several HTGR demonstration plants have been built for production of electricity and a small VHTR is operating in Japan. Consequently, the VHTR is a leading candidate for near-term deployment for H_2 production. The AHTR has not been demonstrated. However, because it is the first reactor concept designed for H_2 production, its characteristics are significantly better matched to this mission. The reactor operates at lower temperatures and pressures than the VHTR for heat delivered at the same temperatures to the H_2 plant. Its larger size better matches the expected size of H_2 plants. While the major technologies for the AHTR have been demonstrated in other systems, significant uncertainties are associated with any new reactor concept.

The MSR is a serious candidate for H_2 production because of its history. MSRs were originally developed as part of the U.S. Aircraft Nuclear Propulsion Program, a very large development program with the goal of building a military aircraft with unlimited range. The requirements for aircraft propulsion (high temperature, low pressure, etc.) are almost identical to those required for H_2 production; thus, the MSR offers relatively good coupling with H_2 production. (The AHTR was not considered for aircraft propulsion because the high-temperature coated-particle fuel had not yet been invented.) The complication with an MSR is that it uses liquid fuels, whereas all other reactors use solid fuels. Because the MSR represents a very different reactor technology, major resources would be required to address its own specific development and regulatory issues.

The LFR and GFR are in the very early stages of development as commercial reactors. While LFRs have been built, the operating temperatures are much lower than required for H_2 production. New fuel clad materials would be required for high-temperature operation. No fuel has yet been chosen for the GFR—an advanced concept with many uncertainties.

Nuclear Fuel-Cycle Requirements. There are two types of nuclear fuel cycles: once through (open) and recycle (closed). The AHTR, VHTR, and MSR can be operated in either mode while the LFR and GFR require a closed fuel cycle. With a once-through fuel cycle, the fuel is made with enriched uranium and the spent nuclear fuel (SNF) is a waste. With a recycle fuel cycle, the SNF is chemically processed to recover fissile materials that are used to produce new fuel.

Fast-neutron reactors (the LFR and GFR) require large inventories of fissile material per unit of energy output. The high cost of fissile material in these reactors requires that the SNF be processed, the fissile material recovered, and the fissile material converted into new fuel. At current uranium prices, once-through fuel cycles are less expensive than recycle fuel cycles for those reactors (VHTR, AHTR, MSR) that need only small inventories of fissile materials to operate. Fast reactors (the LFR and GFR) and the MSR, with the appropriate closed fuel cycle, can be designed to produce more fuel than they consume; i.e., convert fertile ^{238}U or ^{232}Th into fissile fuels. This is a major long-term advantage in reducing the cost impact of uranium on the fuel cycle by a factor of 50 or more. It is the basis for the long-term interests in such reactors.

Technology Status. The AHTR and VHTR use coated-particle fuels in a once-through fuel cycle. Current power reactors primarily use once-through fuel cycles. Uranium mines, chemical conversion facilities, and uranium enrichment plants already exist. The only commercial-scale component of the fuel cycle that does not exist for these reactors is fuel fabrication of the coated-particle fuel. The basic fuel fabrication technology, however, exists. Thus primary requirement is to develop and demonstrate the fabrication technology on a commercial scale. The SNF from these reactors can be directly disposed of. Consequently, the fuel cycle status of these two reactors is given a high rating.

Much, but not all of the fuel technology for the MSR exists. Most of the LFR fuel cycle exists because it uses the same basic technology that was developed for the sodium-cooled fast reactor. (The sodium-cooled fast reactor itself is not a candidate for H_2 production because of the low boiling point of sodium.) However, the LFR technology has not been deployed on a commercial scale. The GFR fuel cycle technology is in a very early state of development (fuel form not yet defined).

Scale of deployment. Reactors with once-through fuel cycles (AHTR and VHTR) can be rapidly deployed on a large scale. Creation of the fuel cycle requires only construction of a relatively low-cost fuel fabrication plant (at most a few tens of millions of dollars). Reactors that require SNF recycle [5] will require many decades to deploy on a large scale and involve very large expenditures of resources because: (1) economics demands construction of large-scale facilities to recover fissile material from SNF and (2) the current inventories of fissile material in SNF are limited. The limited fissile inventories imply that many decades will be required to obtain the necessary materials for deployment of sufficient reactors on a scale that makes a major impact on the world's H_2 production. Such fuel cycles require very-large-scale commitments over very long time periods; but, require very little uranium or thorium to operate.

CONCLUSIONS

Economic large-scale production of H_2 is challenging. An examination of the requirements and reactor options indicates that two reactors are potential candidates for the large-scale production of H_2: the AHTR and the VHTR. Both reactors use the same type of fuel. The AHTR, which is designed for H_2 production, is potentially superior for this role because of the better coupling of a low-pressure liquid coolant with the H_2 production facility and a size that matches expected H_2 plant sizes. However, there is experience in building small VHTRs. In terms of R&D, perhaps ~80% commonality between the two concepts exists.

The other reactors represent much longer term options. The LFR and GFR would require many decades and very large expenditures of resources, partly because these reactors require a closed fuel cycle. In the very long term (beyond 2050), these reactors have the advantage of being able to can create their own fuel.

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