

Hydrogen Production Using the Advanced High-Temperature Reactor

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ABSTRACT

The demand for hydrogen (H₂) is growing rapidly because of increased use of H₂ to convert heavy crude oils into clean transport fuels. If low-cost H₂ were available, the energy content of liquid fuels could be increased by up to 15% per barrel of oil by the addition of more H₂ to those fuels—which represents a massive new market. In addition to this option is the potential to use H₂ directly as a transport fuel. The size of the H₂ market and the scale of single H₂ production facilities are now sufficient large to justify the development and potential deployment of nuclear reactors for H₂ production under either the business-as-usual or the H₂-economy futures.

Nuclear energy can be used to produce H₂ by multiple methods: nuclear-heat-assisted steam reforming of natural gas, high-temperature electrolysis, and high-temperature thermochemical processes. The requirements for efficient H₂ production are demanding. Consequently, it may be necessary to develop nuclear reactors explicitly for H₂ production. A new reactor concept is being developed for this specialized application: the Advanced High-Temperature Reactor (AHTR). With the AHTR, the fuel and reactor coolant are chosen to match H₂ production requirements. The fuel is a coated-particle fuel with a demonstrated operating temperature limit of - 1200EC. Heat is transferred using molten salt coolants from the reactor core to an intermediate heat exchanger and from the intermediate heat exchanger and then to the H₂ production plant.

I. INTRODUCTION

The growing demand for hydrogen (H₂), the potential for a H₂ economy, the incentives to avoid imports of foreign oil, and concerns about greenhouse gas emissions have created strong incentives to develop new methods of H₂ production. One potentially attractive option is the use of nuclear energy. However, the requirements for H₂ production are demanding. To meet these requirements, a nuclear reactor specifically designed for H₂ may be required. One such candidate, the Advanced High-Temperature Reactor (AHTR), is described herein, including the basis for selection of specific features of the reactor.

II. HYDROGEN MARKETS

Nuclear plants are economical only when built in large sizes (hundreds to thousands of megawatts). If H₂ production from nuclear energy is to be viable, the demand for H₂ must match the economic scale of nuclear energy production. Changes (Forsberg 2003a) within the last decade have eliminated the historic mismatch between H₂ demand and the scale of nuclear energy production. These changes include (1) growth in worldwide H₂ consumption to 50 million tons/year, (2) expected future H₂ growth rates of 4 to 10%—primarily because of the decreased quality of crude oil and the need for more H₂ to upgrade the crude oil to gasoline, and (3) development of pipelines that allow for very large H₂ production units. The world-class H₂ plants that are under construction have production capacities of 200 million standard cubic feet of H₂ per day (scf/d). New plants have been announced with capacities of 300 million scf/d [1200 MW(t) of H₂ energy based on the higher heating value]. The next generation of ammonia plants (large H₂ consumers) are expected to produce 3000 t/day of ammonia, requiring 200 million scf/d of H₂. Most of these plants use steam reforming of natural gas to produce H₂.

Several processes are being developed to produce H₂ 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 scf/d. In terms of energy flows, the size of today's H₂ production plant is now equivalent to the size of a nuclear power plant. There is no longer a size mismatch between the scale of conventional H₂ production facilities and nuclear reactors.

If H₂ can be generated economically using nuclear energy, the demand for H₂ could ultimately exceed 5% of the total U.S. energy market in a modified business-as-usual scenario. The world is exhausting its supplies of light crude oil, which requires little processing to produce liquid fuels. The rapid growth market for H₂ is the upgrading of more abundant low-cost heavy crude oils to clean gasoline and diesel fuels. These crude oils have H₂-to-carbon ratios as low as 0.8. The H₂-to-carbon content of the crude oil must be raised to - 1.5 to produce usable liquid fuels. However, the ratio of H₂ to carbon in liquid fuels is a variable. If low-cost H₂ were available, the yield of gasoline, jet, and diesel fuel per barrel of oil could be increased by up to 15% by increasing the H₂-to-carbon ratio in the final product. This is accomplished primarily by breaking one ring of double- and triple-ring compounds in liquid fuels and saturating the carbon chain with H₂.

Refineries can add more H₂ to liquid fuels. The H₂-to-carbon ratio is dictated by economics. The economics are near breakeven but not profitable. If the price of oil increased or the price of H₂ decreased, further hydrogenation of fuel would be adopted. However, there is a problem with this approach. Most H₂ is made from natural gas. Natural gas and crude oil prices are coupled because (1) both can be used as boiler fuel and (2) natural gas can be converted to liquid fuels. As a consequence, if oil prices rise, natural gas and H₂ prices also rise. If economic H₂ from nuclear energy can be developed, the links between oil, gas, and H₂ prices are broken. If the links are broken, more H₂ can be added to the liquid fuel supply as oil prices rise.

There are other benefits from additional hydrogenation of liquid fuels. Oil imports are then reduced, with the production of more fuel per barrel of oil. The CO₂ emissions from burning liquid fuels are also decreased. The sulfur content of liquid fuels is reduced because the

processes that add H₂ to liquid fuels also tend to remove sulfur. Smaller quantities of nitrogen oxides are formed in internal combustion engines because H₂-rich fuels lower the peak burn temperature in the combustion chamber. The particulate emissions from diesel engines are reduced because the H₂ addition reduces the quantities of multiring carbon compounds in diesel fuel—the compounds primarily responsible for soot from diesel engines. Last, the adoption of H₂-enriched liquid fuels creates a H₂ infrastructure that greatly simplifies any future conversion to a H₂ economy.

Ultimately, the world will likely adopt some type of H₂ economy. Such an economy implies a world market for H₂ that exceeds that for electricity. The near-term and potential long-term markets for H₂ are sufficient to justify the development of methods to produce H₂ using nuclear energy.

III. HYDROGEN PRODUCTION USING NUCLEAR ENERGY

Three approaches (Forsberg 2003a, OECD-NEA 2000) have been identified for production of H₂ using nuclear energy. In each application, energy in the form of high temperature heat is used to manufacture H₂.

Nuclear-assisted steam reforming of natural gas uses nuclear heat to reduce the amount of natural gas needed to produce a given quantity of H₂. Steam reforming of natural gas is the primary method used for H₂ production today. It involves the conversion of methane and water into H₂ and carbon dioxide. The process is highly endothermic; that is, high-temperature heat is required. The natural gas is (1) used as a source of chemically reduced H₂ and (2) burned to produce high-temperature heat for the process. Nuclear-assisted steam reforming of natural gas uses a nuclear reactor to provide the high-temperature heat and thus reduce the quantity of natural gas required. Because the chemical process is fully developed, this is the near-term process option for H₂ production using nuclear energy.

Hot electrolysis involves electrolysis of water at high temperatures to produce H₂ and oxygen. At high temperatures, some of the energy input for electrolysis can be heat rather than the more expensive electricity. Major research and development is required to make this a viable process.

Thermochemical cycles use a series of chemical reactions and high-temperature heat to convert water to H₂ and oxygen. Over a 100 such cycles have been identified. The estimated thermal-to-hydrogen efficiency for the best of these processes is greater than 50%. Japanese researchers estimate (OCED-NEA 2000) that the cost of nuclear thermochemical H₂ production may be as low as 60% of that for H₂ production by the electrolysis of water; consequently, thermochemical processes are the leading long-term options for production of H₂ using nuclear reactors. The projected cost is lower than that for electrolysis because a thermochemical process converts heat directly to H₂ whereas electrolysis involves conversion of heat to electricity and the electricity to H₂. Added conversion steps increase inefficiencies and capital costs. Significant work is required before these processes become commercial. The leading candidate is the sulfur-iodine process which consists of three chemical reactions:



Overall, heat and water are added to the process to produce H₂ and oxygen. 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.

The nuclear-assisted steam reforming of natural gas reduces greenhouse gas emissions compared to current H₂ processes whereas the other two H₂ production techniques totally avoid greenhouse gas emissions. Each of the three approaches to H₂ production imposes similar requirements on the reactor.

Temperature. Low-cost H₂ production methods require high temperatures (750 to 900EC).

Temperature range of delivered heat. All of the H₂ production methods involve an endothermic high-temperature chemical dissociation reaction that operates at nearly constant temperatures. Heat is best delivered over a small temperature range.

Reactor power. The most recently announced plants to produce H₂ from natural gas will yield about 300 million scf/d—equivalent to 1200 MW(t). Assuming a 50% efficient thermochemical cycle, the required reactor output is ~2400 MW(t).

Pressure. The chemical reactions for H₂ production go to completion at low pressures, while high pressures reverse the desired reactions. The H₂–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 in the high-temperature materials.

Isolation. To ensure that potential accidents in one facility do not impact the other, the nuclear and chemical facilities must be separated.

IV. THE ADVANCED HIGH-TEMPERATURE REACTOR (AHTR)

To meet the requirements for H₂ production, an advanced reactor concept is proposed: the AHTR (Forsberg 2003b). The AHTR design is based on two considerations: (1) meeting the functional requirements for production of H₂—as defined above—and (2) using existing technologies where possible. The use of existing technologies implies that such a reactor can be developed and deployed on a significant scale in less than 20 years.

Figure 1 shows a simplified schematic of the AHTR. The reactor uses a graphite-matrix coated-particle fuel capable of operating at steady-state temperatures up to 1200EC. Heat is generated within the nuclear fuel and transferred to the coolant. The heat is then transported from the reactor core to the H₂ production facility by (1) pumping a hot molten fluoride salt coolant from the reactor core to the H₂ production facility, (2) transferring the heat to the process facility through heat exchangers, and (3) returning the molten-salt coolant to the reactor. The reactor operates near atmospheric pressure with a power output of up to 2400 MW(t).

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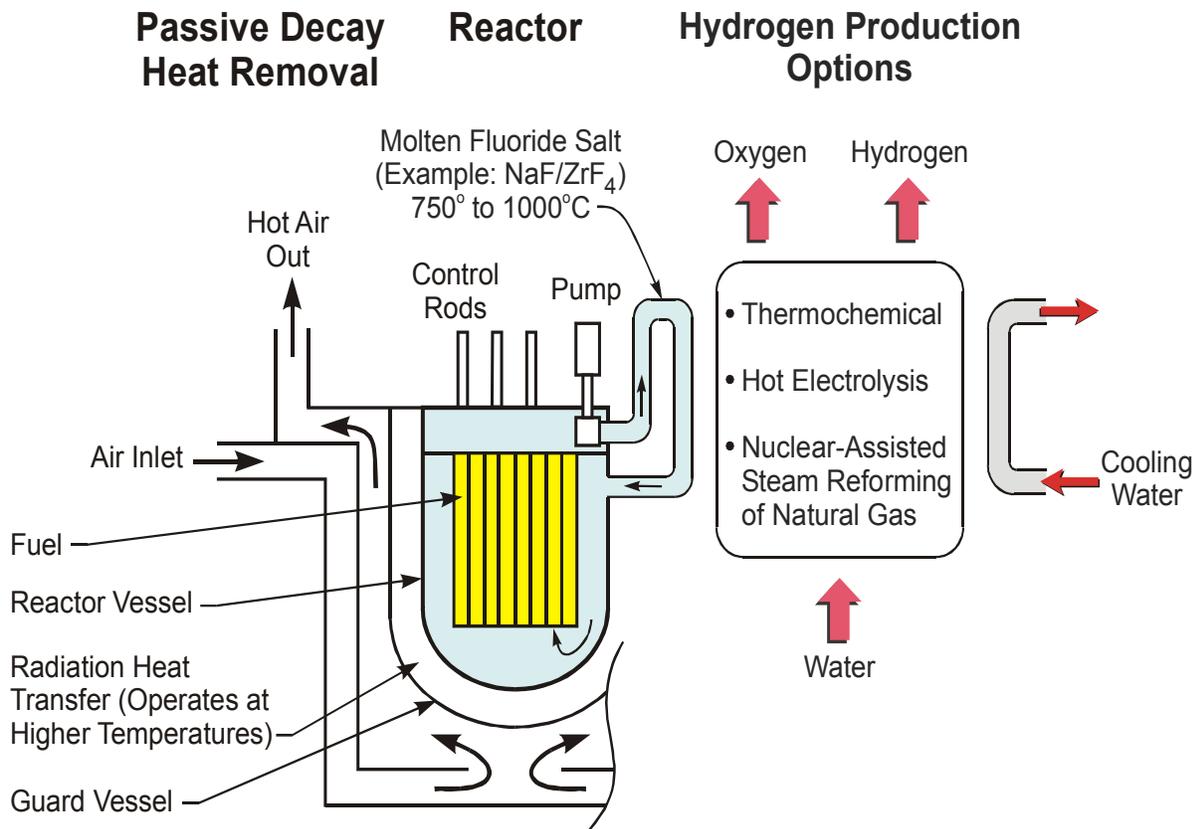


Fig. 1. The Advanced High-Temperature Reactor.

Fuel Design

Because graphite-matrix coated-particle fuel is the only high-temperature nuclear fuel that has been demonstrated on a significant scale to operate at the required temperatures, it is the nuclear fuel that will be used if H_2 is to be manufactured within the next several decades. This type of fuel was originally developed for the high-temperature gas-cooled reactor (HTGR). Figure 2 shows the specific fuel design for the Japanese High-Temperature Test Reactor, which uses helium cooling with a helium reactor exit temperature of 950EC. The nominal peak fuel operating temperature is 1200EC. Under accident conditions, the fuel temperature can go as high as 1600EC for limited periods of time before fuel failure occurs.

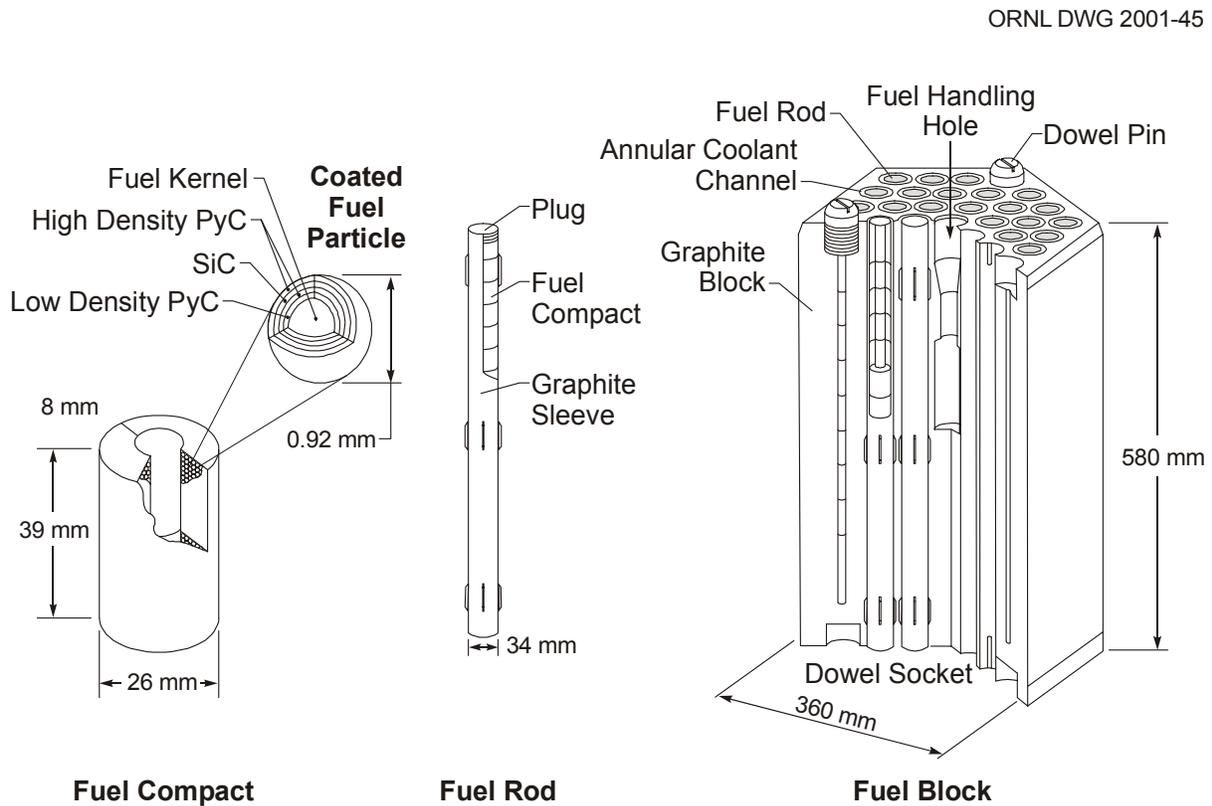


Fig. 2. Graphite-Matrix Coated-Particle Fuel Used in the High-Temperature Test Reactor.

The fuel consists of microspheres of uranium oxide or carbide that are coated with layers of carbon and silicon carbide. The multilayer structure contains the radioactivity from fission of the uranium. The microspheres are incorporated into a carbon matrix that forms a fuel compact. Compacts are inserted into large graphite fuel element blocks. Heat is removed by the flow of coolant through coolant channels in the graphite block. The reactor core is made of three very high-temperature materials: uranium dioxide, silicon carbide, and various forms of carbon.

Molten-Salt Coolants

The type of fuel used defines the options for reactor coolant. The graphite-matrix coated-particle fuel has been demonstrated to be compatible with only two coolants: high-pressure helium and low-pressure molten fluoride salts. Industry has shown interest in the use of this fuel in helium-cooled reactors to produce electricity. The high-pressure, high-temperature helium that leaves the reactor can be sent directly to a helium gas turbine for the production of electricity. This technology is an attractive method to produce electricity.

However, the requirements are different from those for H₂ production. The AHTR uses a molten-salt coolant because liquid coolants are more efficient than gas coolants in transferring heat from the reactor fuel to the H₂ plant. This reduces the peak temperatures in the reactor, pumps and heat exchangers.

Figure 3 compares the performance of liquid and gaseous coolants in typical reactor designs. The temperature difference between the coolant entering a reactor and leaving a reactor is typically a few tens of degrees for liquid coolants versus several hundred degrees for gaseous coolants. This difference has been demonstrated in operating reactors. Over half the world's power reactors are pressurized-water reactors. The temperature rise of the liquid coolant (water) through the reactor core is ~20°C. In contrast, the temperature rise in British advanced gas reactors with carbon dioxide coolant is ~350°C. Similarly, for a liquid coolant the temperature drop from the hottest fuel microsphere to the reactor coolant is half that for a gas-cooled reactor (Forsberg 2003b). Careful design can reduce these temperature losses for both types of coolants but at considerable costs. If the heat must be delivered at 800°C, peak reactor coolant and fuel temperatures in a liquid-cooled reactor will be significantly less than those in a comparable gas-cooled reactor. The temperatures are near the limits of standard (metallic) engineering materials; thus, there are strong incentives to lower peak reactor temperatures. Furthermore, unlike high-pressure coolants, low-pressure liquid coolants reduce the stresses on components.

There is a century of industrial experience with graphite and fluoride salt compatibility; aluminum is electrolytically produced from molten cryolite (3NaF·AlF₃) in very large graphite baths at ~1000°C. There is also reactor experience with molten salts. The Aircraft Reactor Experiment (ARE), a 2.5-MW(t) reactor, operated in the 1950s with a NaF-ZrF₄ molten salt at peak temperatures of 860°C. The ARE was part of the large billion-dollar Aircraft Nuclear Propulsion Program, which had as its goal a nuclear-powered military jet aircraft with unlimited range. The reactor was designed to deliver heat to a heat exchanger, which then heated the air going through the jet engine. The ARE was followed by the Molten Salt Reactor Experiment (MSRE), an 8-MW(t) reactor, operated in the 1960s with a ⁷LiF/BeF₂ molten salt.

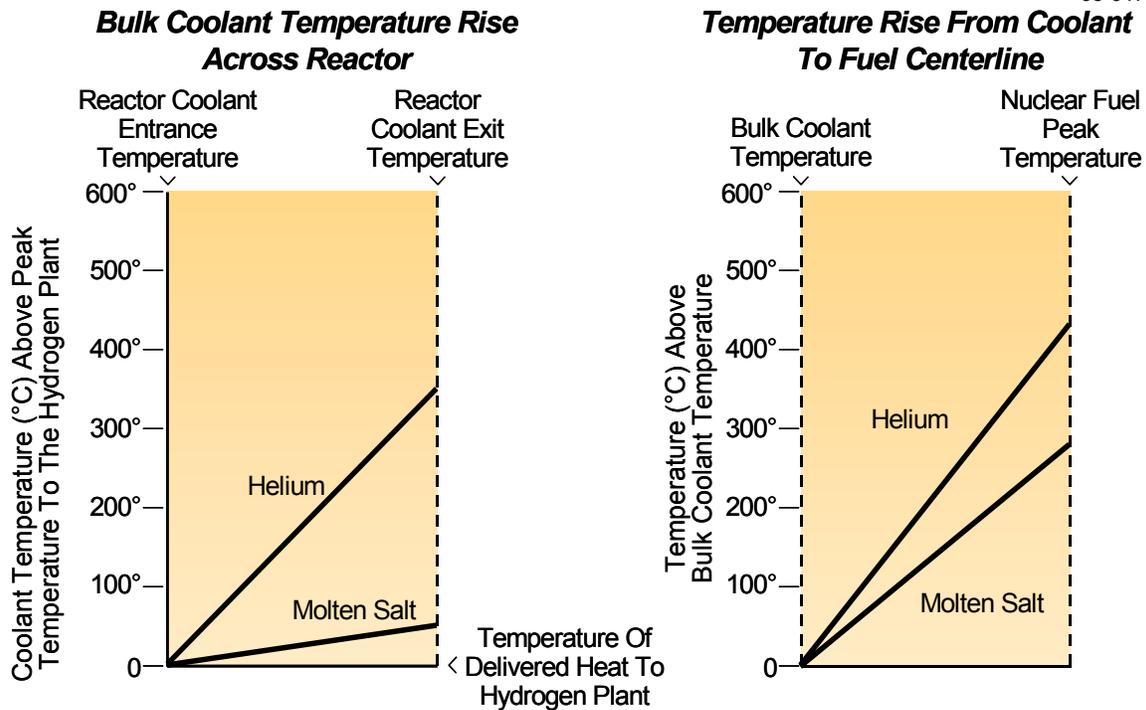


Fig. 3. Typical Temperature Drops Used to Move Heat in Reactors with Different Coolants.

The ARE and MSRE were molten-salt-fueled reactors (uranium fuel dissolved in the coolant) whereas the AHTR is a molten-salt-cooled reactor. The AHTR uses solid fuel with a clean molten-salt coolant (no fuel or fission products in the salt). The use of clean molten salts as coolants avoids some of the issues associated with liquid-fuel reactors. When the original work on molten-salt-fueled reactors was underway, the solid fuel had not yet been invented. Thus, molten-salt-cooled reactors were not developed at that time. After the particulate fuel was developed, interest was focused on helium-cooled reactors, in which the high-temperature, high-pressure helium could be efficiently coupled to a gas turbine for electricity production. It is the requirement to transfer high-temperature heat to a H_2 production plant that creates the strong incentive to use molten-salt coolants. The technical requirements for a reactor to produce H_2 (very high temperatures and low pressures) are similar to those for a reactor to provide heat to a jet engine; consequently, similar technologies apply.

All of the candidate fluoride salts have somewhat similar properties. Neutron absorptions in these salts are low, and atmospheric boiling points are near 1400EC. Molten salts are transparent. At operating conditions, the heat transfer properties of molten salts are similar to those of water. Although molten salts do not react with air or carbon dioxide, they will slowly react with water. For H₂ production, the candidate molten salts include the ARE salt (NaF-ZrF₄) and various salts with a third component. Molten fluoride salts are also leading candidates for cooling future fusion energy machines. Other molten salts are used in the chemical industry for heat transfer.

Reactor Safety Systems

Nuclear reactors contain large quantities of radioactivity. Consequently, safety systems are required to minimize the risk of accidents. For advanced reactors, passive safety systems are proposed that are not dependent on human actions or operating mechanical equipment (diesel generators, pumps, etc.). In this context, the HTGR and AHTR both have unique safety potential because of the unique high-temperature capability of the graphite-matrix coated-particle fuel.

In terms of plant design and economics, the most important safety requirement is to avoid overheating the reactor core, which can result in subsequent fuel failure. When a reactor is shut down, the fuel continues to produce heat from decay of radioactive fission products but at a rate that continually decreases with time. Failure to provide adequate cooling, such as happened at the Three Mile Island reactor, will cause the fuel to overheat and fail.

The graphite-matrix coated-particle fuel can withstand temperatures as high as 1600EC for limited periods of time without releasing significant quantities of radionuclides. If the fuel temperature can be kept below this peak temperature, a major radionuclide release is avoided. Several methods are used to limit temperatures.

Normal decay-heat cooling. Reactors are designed with decay-heat cooling systems to prevent damage to the reactor core upon shutdown. There are several options for the design of this system. One option for the AHTR is the decay-heat-removal system that was originally developed for the General Electric S-PRISM sodium-cooled reactor. This system requires a low-pressure coolant. The decay heat (Fig. 1) is conducted through the reactor vessel wall, transferred across an argon gap by radiation to a guard vessel, conducted through the guard vessel, and removed from the outside of the guard vessel wall by natural circulation of air. The radiation heat transfer rate from the reactor vessel to the guard vessel increases by the temperature to the fourth power (T^4); thus, a small rise in the reactor vessel temperature greatly increases the heat transfer out of the system. The argon gap acts as a thermal switch to limit heat losses during normal operation but allows radiation heat transfer to increase heat losses if the reactor overheats. With this type of passive safety system, the reactor size can be as large as 2400 MW(t).

Beyond-design-basis accident. In a severe accident, safety is ensured by systems that protect public health; however, in such scenarios the plant might be destroyed. There are two interconnected ultimate decay-heat removal systems in molten-salt-cooled reactors. Both systems are based on the fact that the fuel remains intact at temperatures far beyond that of the metallic components in the reactor—a unique capability of this fuel.

- Salt boiloff. The molten salt boils at $\sim 1400^{\circ}\text{C}$, below the failure temperature of the fuel. The boiling (and condensing) of the salt removes heat from the reactor core.
- Decay heat to ground. If the temperatures are sufficiently high, vessel failure will occur. The silo in which the reactor is located will fill with molten salt. The inventory of molten salt in the vessel assures that the molten-salt level is above the reactor core. Because molten salts freeze between 300 and 400°C , the salt seals the silo. Heat is then transferred by convective currents to the silo wall and conducted into the surrounding soil. The fuel temperature is kept below 1600°C , the temperature at which it may begin to fail.

While the details of operation for these systems are complicated, there is an important economic implication. The liquid coolant allows heat to be removed from the hottest locations in the reactor core to the outside of the reactor core with small temperature drops by natural circulation of the liquid and conduction. Gas-cooled reactors can only remove heat from the hottest locations in the reactor core by conduction through the fuel. The cooling effect of a gas by natural circulation and conduction is low—particularly if the reactor is depressurized. As a consequence, there is a large temperature drop between the center of the reactor core and outside the reactor core that limits the reactor size if the peak fuel temperature in an accident is to be kept below 1600°C . The practical implications are that the liquid-cooled AHTR may be built in sizes up to 2400 MW(t) with passive safety systems whereas the gas-cooled HTGR with passive safety systems may be limited to 600 MW(t) under accident conditions when the reactor depressurizes.

Isolation of the Nuclear Reactor from the Hydrogen Production Facility

Nuclear facilities and H_2 production facilities incorporate different safety philosophies, which are a consequence of the different properties of radioactive materials and H_2 . In a nuclear facility, the safety systems are designed to contain radioactivity and prevent release to the atmosphere. Critical components are bunkered inside containment buildings with walls more than 1 meter thick. In contrast, H_2 production facilities are open-air facilities. Therefore, (1) leaks of H_2 can rapidly disperse to below 4% H_2 —the explosion limit for H_2 in air and (2) the energy from any fire or explosion can be dispersed with minimum damage. This difference in safety philosophy requires the physical separation of the reactor from the H_2 production plant. This, in turn, requires efficient methods to transfer high-temperature heat over significant distances.

Traditionally, molten salts have been used in the chemical industry to transfer heat at high temperatures and low pressures. Molten salts have very high heat capacities and low pumping costs. The AHTR, as a molten-salt-cooled reactor, is fully compatible with a heat transfer loop using a molten salt.

V. ECONOMICS, DEPLOYMENT, AND STATUS

The AHTR is a new reactor concept—approximately 2 years old. Consequently, significant uncertainties remain. HTGRs have already been built. Thus, they are the near-term option for H₂ production, particularly for nuclear-assisted steam reforming of natural gas, which represents the near-term H₂ production technology.

In the longer term, the AHTR has the potential for significantly improved performance and economics. This is a consequence of designing a reactor for a specific mission rather than using an existing reactor design that was originally developed for another purpose. In this specific case, it is the adoption of a low-pressure liquid coolant that potentially improves the economics by two sets of mechanisms.

- *Liquid coolant properties.* The excellent heat transfer properties of liquids versus gases reduces (1) peak temperatures in the reactor, piping, and heat exchangers and (2) the size of heat exchangers. The high heat capacity liquid reduces piping and pump size, including the system to transfer heat to the H₂ plant.
- *Economics of scale.* The larger plant size [2400 MW(t) for the AHTR versus 600 MW(t) for the HTGR] offers the potential for significant economics of scale. Industrial experience indicates that the relative costs for plants of different size can be estimated by:

$$\text{Cost of large plant} = \text{cost of small plant} \times [\text{capacity of large plant}/\text{capacity of small plant}]^x$$

The traditional scaling exponent (x) in the chemical industry is 0.6. If the plant is four times larger, the cost per unit capacity in the larger plant will be 58% of the smaller plant. If a more conservative scaling exponent of 0.8 is used, the cost per unit of capacity of the larger plant will be 75%. It is expected that the same economic drivers that have increased the size of natural-gas-fired H₂ plants will also apply to nuclear H₂ facilities.

The current estimates (EPRI 2003) are that in the United States, nuclear-assisted steam reforming of natural gas using an HTGR is roughly competitive with H₂ production using steam reforming with only natural gas. With modest increases in natural gas prices, thermochemical production of H₂ would be economic. If these estimates are correct, the AHTR creates the potential for H₂ production costs less than today's H₂ production costs.

VI. CONCLUSIONS

The economics of nuclear energy generally favor large plants. The rapid growth in demand and the scale of new H₂ production plants are now sufficient to support the use of nuclear energy for H₂ production. Three technological approaches exist to use high-temperature heat from a nuclear reactor to produce H₂. Only graphite-matrix coated-particle nuclear fuels are capable of operating at the temperatures required for efficient H₂ production. Two coolants are compatible with this nuclear fuel: high-pressure helium gas (HTGR) and low-pressure molten-salt liquids (AHTR). Molten-salt cooling has potential advantages for H₂ production because (1) the molten-salt coolant better couples with the requirements of H₂ production, (2) the reactor

size matches the preferred scale for H₂ production, and (3) the long-term economics are potentially superior for this specific application. The commonalities between the HTGR and AHTR suggest that most of the research and development activities for nuclear-generated H₂ using the two reactors will be identical. The AHTR is a new reactor concept with significant technical and economic uncertainties. However, it has the potential for economic, non-greenhouse-gas-producing H₂ production.

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