

# **The Advanced High-Temperature Reactor (AHTR) for Producing Hydrogen to Manufacture Liquid Fuels**

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## ABSTRACT

Conventional world oil production is expected to peak within a decade. Shortfalls in production of liquid fuels (gasoline, diesel, and jet fuel) from conventional oil sources are expected to be offset by increased production of fuels from heavy oils and tar sands that are primarily located in the Western Hemisphere (Canada, Venezuela, the United States, and Mexico). Simultaneously, there is a renewed interest in liquid fuels from biomass, such as alcohol; but, biomass production requires fertilizer. Massive quantities of hydrogen ( $H_2$ ) are required (1) to convert heavy oils and tar sands to liquid fuels and (2) to produce fertilizer for production of biomass that can be converted to liquid fuels. If these liquid fuels are to be used while simultaneously minimizing greenhouse emissions, nonfossil methods for the production of  $H_2$  are required.

Nuclear energy can be used to produce  $H_2$ . The most efficient methods to produce  $H_2$  from nuclear energy involve thermochemical cycles in which high-temperature heat (700 to 850°C) and water are converted to  $H_2$  and oxygen. The peak nuclear reactor fuel and coolant temperatures must be significantly higher than the chemical process temperatures to transport heat from the reactor core to an intermediate heat transfer loop and from the intermediate heat transfer loop to the chemical plant.

The reactor temperatures required for  $H_2$  production are at the limits of practical engineering materials. A new high-temperature reactor concept is being developed for  $H_2$  and electricity production: the Advanced High-Temperature Reactor (AHTR). The fuel is a graphite-matrix, coated-particle fuel, the same type that is used in modular high-temperature gas-cooled reactors (MHTGRs). The coolant is a clean molten fluoride salt with a boiling point near 1400°C. *The use of a liquid coolant, rather than helium, reduces peak reactor fuel and coolant temperatures 100 to 200°C relative to those of a MHTGR.* Liquids are better heat transfer fluids than gases and thus reduce three temperature losses in the system associated with (1) heat transfer from the fuel to the reactor coolant, (2) temperature rise across the reactor core, and (3) heat transfer across the heat exchangers between the reactor and  $H_2$  production plant. Lowering the peak reactor temperatures and thus reducing the high-temperature materials requirements may make the AHTR the enabling technology for low-cost nuclear hydrogen production.

## 1. HYDROGEN NEEDS FOR LIQUID FUELS

Conventional world oil production is expected to peak within a decade [Giles 2004]. In the next several decades, shortfalls in production of liquid fuels (gasoline, diesel, and jet fuel) from conventional oil production are expected to be offset by increased production of fuels from heavy oils and tar sands [Williams 2003]. These hydrocarbon resources far exceed worldwide conventional oil resources. The Western Hemisphere (Canada, Venezuela, the United States, and Mexico) contains the world's largest deposits of these resources. These hydrocarbon resources have  $H_2$ -to-carbon ratios as low as one. The  $H_2$ -

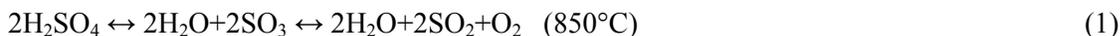
to-carbon ratio in liquid fuels is about two. Therefore, to produce liquid fuels from heavy oils and tar sands, this ratio must be raised to two. The H<sub>2</sub>-to-carbon ratio of heavy oils and tar sands can be changed to make liquid fuels (1) by thermal cracking—extracting “excess” carbon that is ultimately released as carbon dioxide or (2) by hydrocracking—the addition of massive quantities of H<sub>2</sub>. Today, H<sub>2</sub> is made by steam reforming of fossil fuels with the release of large quantities of carbon dioxide. If these hydrocarbon resources are to be used to produce liquid fuels while simultaneously minimizing greenhouse emissions, hydrocracking with the use of nonfossil methods for the production of H<sub>2</sub> is required.

In parts of Brazil, the United States, and other countries, ethanol and several other liquid fuels are made from biomass. One of the major energy inputs into producing biomass is fertilizer—primarily nitrogen in the form of ammonia. The production of ammonia fertilizer requires massive quantities of H<sub>2</sub> and currently is half the market for H<sub>2</sub>. If biomass is to be a major source of liquid fuels, additional quantities of H<sub>2</sub> are required for fertilizer production.

In the long term, H<sub>2</sub> is being considered as a transport fuel. However, it is unclear whether methods for on-board vehicle storage of H<sub>2</sub> will be successfully developed. If they are not successfully developed, the likely liquid fuels are methanol (H<sub>2</sub> plus carbon dioxide from the air) or ammonia (H<sub>2</sub> plus nitrogen). Transport fuel production will require increasing amounts of H<sub>2</sub>, independent of the choice of fuel type.

## 2. THERMOCHEMICAL HYDROGEN PRODUCTION

The leading candidates for low-cost, large-scale H<sub>2</sub> production using nuclear energy are two thermochemical processes [DOE 2003]. A thermochemical process consists of a set of chemical reactions in which the input is high-temperature heat plus water and the output is H<sub>2</sub> and oxygen (O<sub>2</sub>). Two [NEA 2003] of the highest-rated processes have the same high-temperature highly endothermic (heat-absorbing) steps that require heat input between 800 and 850°C.



The two thermochemical processes have different lower-temperature chemical reactions. The sulfur-iodine process has two other chemical reactions (Equations 2 and 3), which, when combined with the reaction in Equation 1, (1) yield H<sub>2</sub> and O<sub>2</sub> from water and heat and (2) recycle all other chemical reagents.



The hybrid sulfur process (also known as Westinghouse, GA-22, and Ispra Mark 11) has a single low-voltage electrochemical step (Equation 4) that completes the cycle.



In each of these cycles, the high-temperature sulfur trioxide (SO<sub>3</sub>) dissociation reaction (Equation 1) is an equilibrium chemical reaction that requires a catalyst. High temperatures and low pressures drive the reaction towards completion. Recently, the use of inorganic membranes has been proposed to reduce the peak temperature of the SO<sub>3</sub> dissociation step to ~700°C. This may be accomplished by the separation of SO<sub>2</sub>, H<sub>2</sub>O, and O<sub>2</sub> from the SO<sub>3</sub> at temperatures near 700°C. If these reaction product gases are removed, the remaining SO<sub>3</sub> (with a catalyst and heat) will dissociate into its equilibrium concentrations.

Significant experimental work is required before it will be known whether this approach will be technically and economically viable and will enable lower-temperature sulfur thermochemical cycles.

There are strong economic incentives to use thermochemical processes for hydrogen production. Their efficiencies are estimated to be near 50%. In contrast, the efficiency of electrolysis is much lower. If a high-temperature reactor has 50% efficiency (heat to electricity) and electrolysis has 70% efficiency (electricity to hydrogen), the overall efficiency is only 35%.

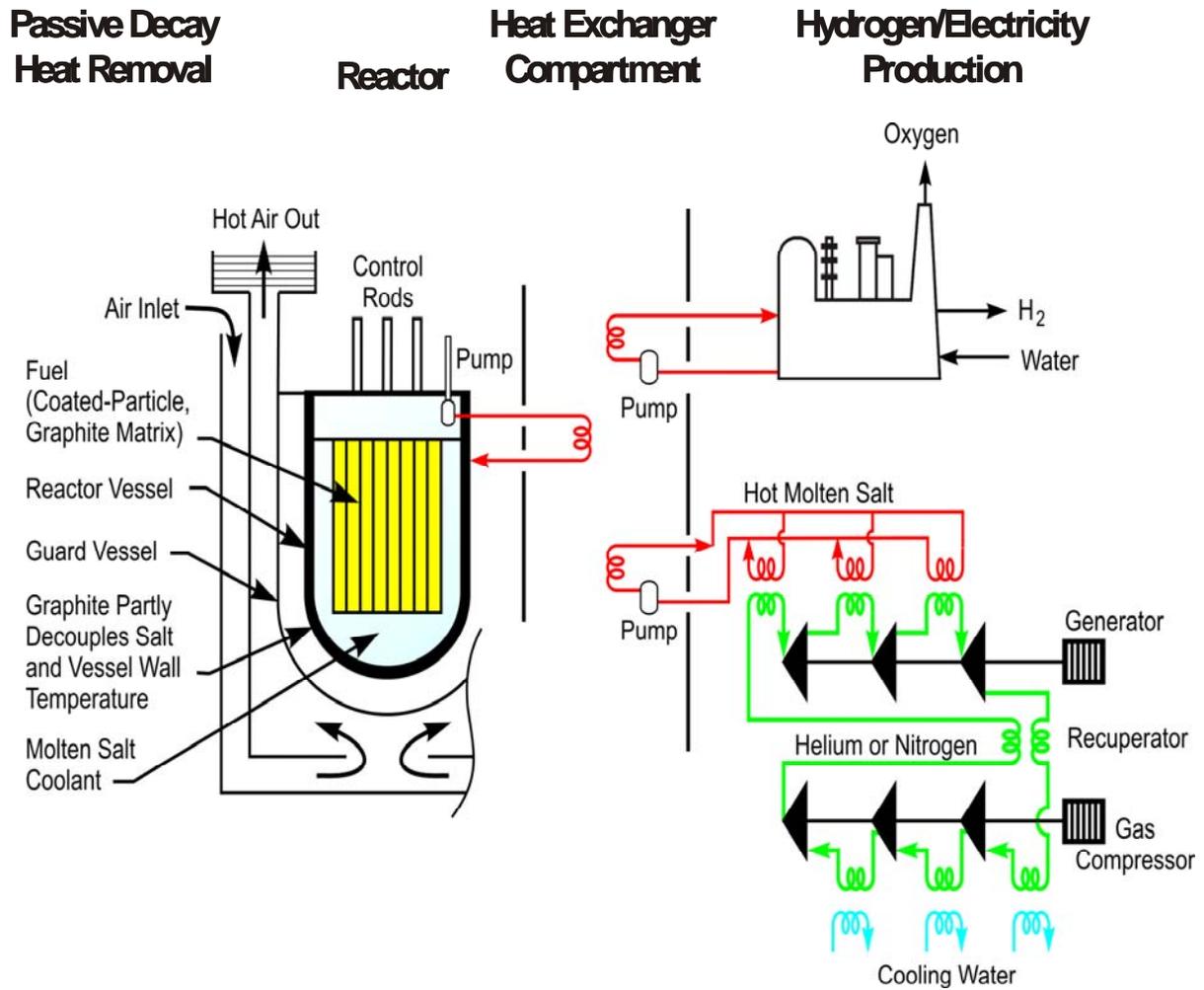
### 3. THE ADVANCED HIGH-TEMPERATURE REACTOR

The primary challenge for nuclear H<sub>2</sub> production is the requirement to deliver heat to the thermochemical plant at very high temperatures. These temperatures are at the very limits of practical engineering materials. A new reactor concept, the Advanced High-Temperature Reactor (AHTR), is being developed to meet these specific requirements. The strategy is to limit the peak reactor temperature and still deliver the process heat at the required temperatures to the H<sub>2</sub> plant. This section briefly describes the AHTR. The following section describes the specific characteristics to minimize peak reactor and heat exchanger temperatures; thus, minimize the technical and economic challenges to develop practical materials for such high temperatures. The development is a joint effort of Oak Ridge National Laboratory, Sandia National Laboratories, and the University of California at Berkeley.

The AHTR (Fig. 1, Table 1) uses coated-particle, graphite-matrix fuels and a molten fluoride-salt coolant [Forsberg et al. 2003; Ingersoll et al. 2004; Forsberg 2004]. The fuel is the same type that is used in modular high-temperature gas-cooled reactors (MHTGRs), with fuel-failure temperatures in excess of 1600°C. The optically transparent molten salt coolant is a mixture of fluoride salts with freezing points near 400°C and atmospheric boiling points of ~1400°C. Several different salts are being evaluated as the primary coolant, including lithium-beryllium and sodium-zirconium fluoride salts. The reactor operates at near-atmospheric pressure. At operating conditions, the molten salt heat-transfer properties are similar to those of water. Heat is transferred from the reactor core by the primary molten-salt coolant to an intermediate heat-transfer loop. The intermediate heat-transfer loop uses a secondary molten-salt coolant to move the heat to a thermochemical H<sub>2</sub> production facility to produce H<sub>2</sub> or to a turbine hall to produce electricity. If electricity is produced, a multi-reheat nitrogen or helium Brayton power cycle is used.

The baseline AHTR facility layout (Fig. 2) that was developed is similar to the S-PRISM sodium-cooled fast reactor designed by General Electric. Both reactors operate at low pressure and high temperature; thus, they have similar design constraints. The 9.2-m-diam. vessel is the same size as that used by the S-PRISM. The vessel size determines the core size that, in turn, determines the power output. In the initial baseline studies, it was assumed that the fuel and power density (8.3 W/cm<sup>3</sup>) were essentially identical to those of the MHTGR. This is a conservative assumption because higher power densities are possible with liquid coolants.

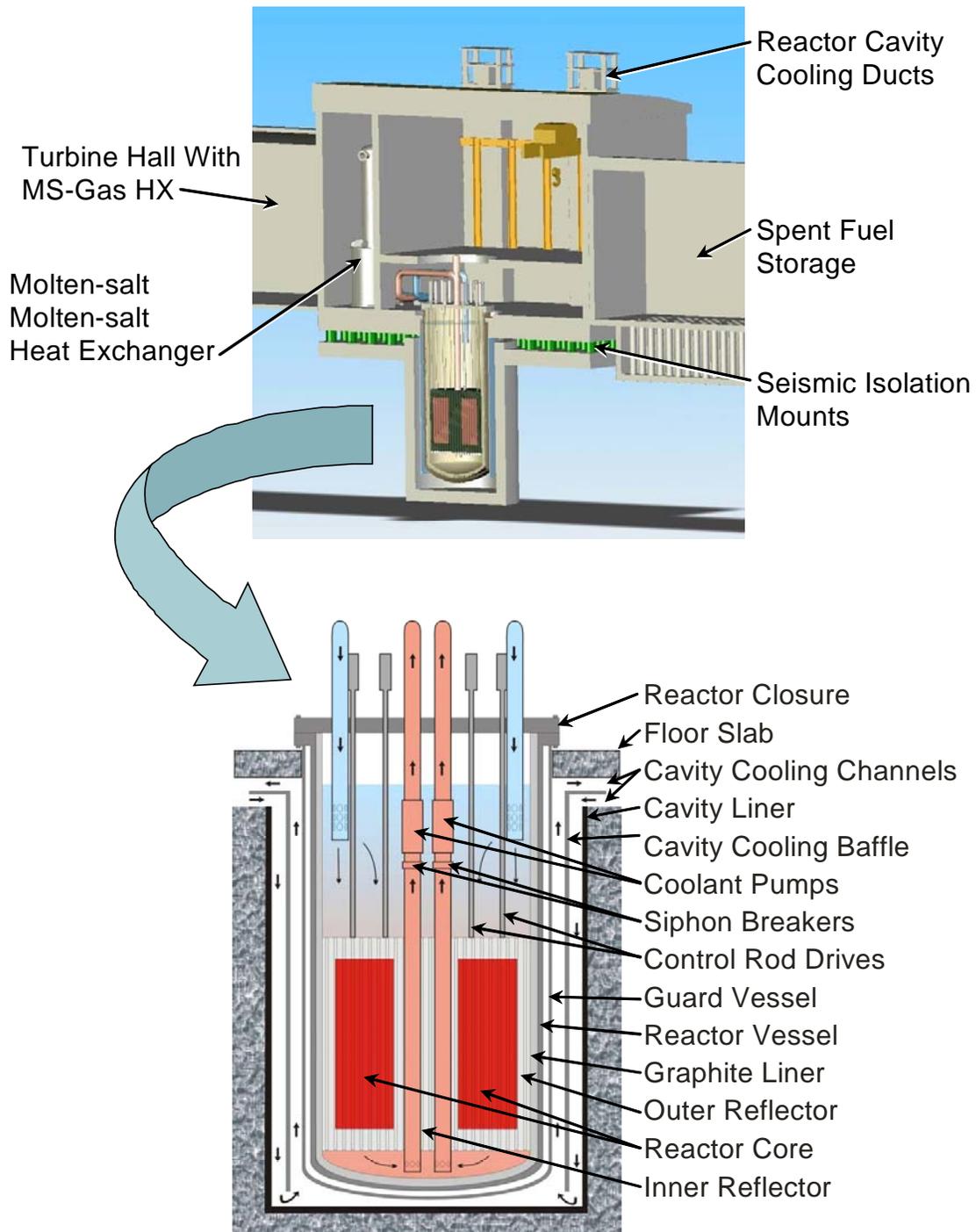
Three peak coolant temperatures were evaluated: 705, 800, and 1000°C for the AHTR—Low Temperature (AHTR-LT), the AHTR—Intermediate Temperature (AHTR-IT), and the AHTR—High Temperature (AHTR-HT), respectively. If the reactor power output is 2400 MW(t), the respective electricity production capacities are 1151 MW(e), 1235 MW(e), and 1357 MW(e). The AHTR-LT uses existing materials, the AHTR-IT uses existing materials that have not been fully tested, and the AHTR-HT uses advanced materials. The AHTR-HT and AHTR-IT include a graphite blanket system, while the AHTR-LT has a metallic blanket system that separates and insulates the reactor vessel from the reactor core so that the fuel and coolant can operate at higher temperatures than the vessel. This insulation ensures long vessel life (minimizing long-term creep) and minimizes heat losses during normal operations.



**Fig. 1. Schematic of the AHTR for electricity production.**

**Table 1. AHTR preconceptual design parameters**

Power level	2400 MW(t)	Power density	8.3 W/cm <sup>3</sup>
Core inlet/outlet temperature (options)	900°C/1000°C 700°C/800°C 670°C/705°C	Fuel element	Form: prismatic Diam.: 0.36 m Height: 0.79 m
Coolant (several options)	<sup>2</sup> LiF-BeF <sub>2</sub> (NaF-ZrF <sub>4</sub> )	Intermediate heat transport loop	Molten salt (Several options)
Fuel		Vessel	
Kernel	U carbide/oxide	Diameter	9.2 m
Enrichment	10.36 wt % <sup>235</sup> U	Height	19.5
Reactor core		Reactor fuel columns	
Shape	Annular	Fuel	324
Diameter	7.8 m	Reflector (outer)	138
Height	7.9 m	Reflector (inner)	55
Fuel annulus	2.3 m		
Volumetric flow rate	5.5 m <sup>3</sup> /s	Coolant velocity	2.3 m/s



**Fig. 2. Schematic of the AHTR nuclear island and vessel.**

In the current preconceptual design, the AHTR has an annular core through which the coolant flows downward. The molten salt coolant flows upward through the nonfuel graphite section in the middle of the reactor. The molten-salt-coolant pumps and their intakes are located above the reactor core with appropriate siphon breakers; thus, the reactor cannot lose its coolant except by failure of the primary vessel. The guard vessel is sized so that even if the primary vessel fails, the core remains covered with salt.

When a reactor shuts down, radioactive decay heat continues to be generated in the reactor core at a rate that decreases over time. If this heat is not removed, the reactor will ultimately overheat and the core will be damaged, such as occurred during the Three Mile Island accident. The reference AHTR design uses passive reactor vessel auxiliary cooling (RVAC) systems similar to that developed for decay heat removal in the General Electric sodium-cooled S-PRISM. The reactor and decay-heat-cooling system are located in a below-grade silo. In this pool reactor, RVAC system decay heat is (1) transferred from the reactor core to the reactor vessel graphite reflector by natural circulation of the molten salts, (2) conducted through the graphite reflector and reactor vessel wall, (3) transferred across an argon gap by radiation to a guard vessel, (4) conducted through the guard vessel, and then (5) removed from outside of the guard vessel by natural circulation of ambient air.

The rate of heat removal is controlled primarily by the radiative heat transfer through the argon gas from the reactor vessel to the guard vessel. Radiative heat transfer increases by the temperature to the fourth power ( $T^4$ ); thus, a small rise in the reactor vessel temperature (as would occur upon the loss of normal decay-heat-removal systems) greatly increases heat transfer out of the system. Under accident conditions such as a loss-of-forced-cooling accident, natural circulation flow of molten salt up the hot fuel channels in the core and down the edge of the core rapidly results in a nearly isothermal core with about a 50°C temperature difference between the top and bottom plenums. For a typical simulation of the reactor with a nominal coolant exit temperature of 1000°C, the calculated peak fuel temperature in such an accident is ~1160°C, which will occur at ~30 hours with a peak reactor vessel temperature of ~750°C at ~45 hours. The average core temperature in this accident rises to approximately the same temperature as the hottest fuel during normal operations.

In terms of passive decay-heat-removal systems, a major difference is noted between the liquid-cooled AHTR and gas-cooled reactors. The AHTR can be built in very large sizes [ $>2400$  MW(t)], while the maximum size of a gas-cooled reactor with passive-decay-heat removal systems is limited to ~600 MW(t). The controlling factor in decay heat removal is the ability to transport this heat from the center of the reactor core to the vessel wall or to a heat exchanger in the reactor vessel. The AHTR uses a liquid coolant where natural circulation can move very large quantities of decay heat to the vessel wall with a small coolant temperature difference (~50°C). Unfortunately, in a gas-cooled reactor under accident conditions when the reactor is depressurized, the natural circulation of gases is not very efficient to transport heat from the fuel in the center of the reactor to the reactor vessel. The heat must be conducted through the reactor fuel to the vessel wall. This inefficient heat transport process limits the size of the reactor to ~600 MW(t) to ensure that the fuel in the hottest location in the reactor core does not overheat and fail.

Because the AHTR uses the same basic fuel type and the molten salt coolant has a low neutron-absorption cross section, the reactor core physics and fuel cycle options are generally similar to those for helium-cooled high-temperature reactors. Reactor power is limited by a negative temperature coefficient, control rods, and other emergency shutdown systems.

Several molten fluoride salts with generally similar properties are being evaluated to determine the optimum coolant salt [Ingersoll et al. 2004]. Such evaluations involve trade-offs in neutronics, cost,

operations, and other parameters. The initial baseline AHTR design used the same salt ( $2^7\text{LiF-BeF}_2$ ) that was used in the Molten Salt Reactor Experiment (MSRE). The MSRE (*Nucl. Appl. Technology* 1970) was a molten salt test reactor in which the fuel was dissolved in the molten salt—in contrast to the AHTR in which there is solid fuel and clean molten salt coolant.

This salt is well understood and has a negative coolant void coefficient. Several other salts, such as the sodium-zirconium fluoride salts, have operational and cost advantages. However, with traditional gas-cooled-reactor coated-particle-fuel and core designs, these other salts have small positive void coefficients. While undesirable, a small positive void coefficient may be acceptable because of the small predicted consequences of a voiding accident. The preliminary transient analysis indicates that the very large negative temperature reactivity coefficient limits the temperature rise to a few 10s of degrees C in the core upon large-scale coolant voiding. The long neutron lifetime results in transients of several tens of seconds. This type of mild behavior under accident conditions is very different than the behavior of other reactors. Conceptual core design studies with more heterogeneous core designs are under way to determine whether negative coolant void coefficients can be obtained for a wide variety of fluoride salts. Earlier versions of CANDU (heavy-water moderated) and Hanford-type (water-cooled, graphite-moderated) reactors had positive void coefficients. The Advanced CANDU reactor and Hanford-N reactor have negative void coefficients. The same core design strategies used in these reactors may allow a wide variety of salts in the AHTR with negative void coefficients.

#### **4. MINIMIZING PEAK HYDROGEN TEMPERATURES**

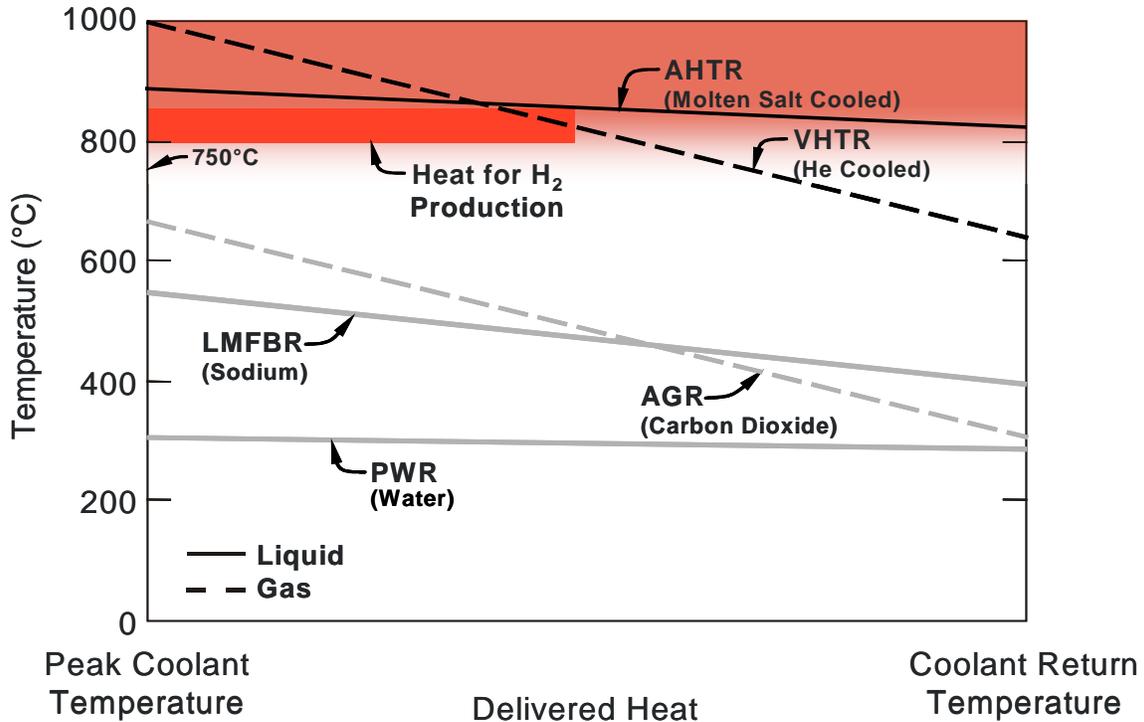
The primary technical challenge of nuclear thermochemical  $\text{H}_2$  production is the requirement for high temperature heat, 700 to 850°C, depending upon the process used to produce the  $\text{H}_2$ . The use of molten salts as the reactor coolant and the coolant in the heat transport system can reduce peak reactor temperatures by several hundred degrees compared to reactors with helium coolant. This significantly reduces the technical challenges for development of a reactor for hydrogen production. The lower temperatures are a consequence of three factors.

##### **4.1 Heat Delivery at Required Temperatures**

Gas-cooled reactor systems have high pumping costs relative to liquid-cooled systems (Fig. 3). Because gas cooling has high pressure losses, practical designs of gas-cooled reactors [such as the General Atomics helium-cooled GT-MHR and the British carbon-dioxide-cooled Advanced Gas Reactor (AGR)] have large temperature increases across the reactor core and deliver their heat to the power cycle over a large temperature range. Typical temperature increases across the core are 350°C. In contrast, liquid-cooled reactors such as the French sodium-cooled Super Phoenix liquid-metal fast breeder reactor (LMFBR) and pressurized-water reactors (PWRs) have low pumping costs and are designed to deliver their heat from the reactor core to the power cycle over a small temperature range. The same will be true of the molten-salt-cooled AHTR.

The production of  $\text{H}_2$  requires the delivery of a significant fraction of the heat at a relatively-uniform high temperature for the near-constant-temperature dissociation of  $\text{SO}_3$ . If half the heat has to be delivered at a given temperature, a gas-cooled reactor will deliver that heat over a temperature range that exceeds  $\sim 150^\circ\text{C}$ —equivalent to about half the temperature rise across the reactor core. If half the heat has to be delivered at a given temperature, a liquid-cooled reactor such as the AHTR will deliver that heat over a temperature range of  $\sim 50^\circ\text{C}$ —equivalent to about half the temperature rise across the reactor core. A gas-cooled reactor may have to have peak reactor exit temperatures a 100°C or higher to deliver heat to a thermochemical  $\text{H}_2$  plant at the same temperatures as a molten-salt-cooled reactor. The temperature range

over which heat is required for the thermochemical cycle will depend upon the specific cycle; however, in all cases, the reactor peak temperature will be less for the AHTR than for a helium-cooled reactor.



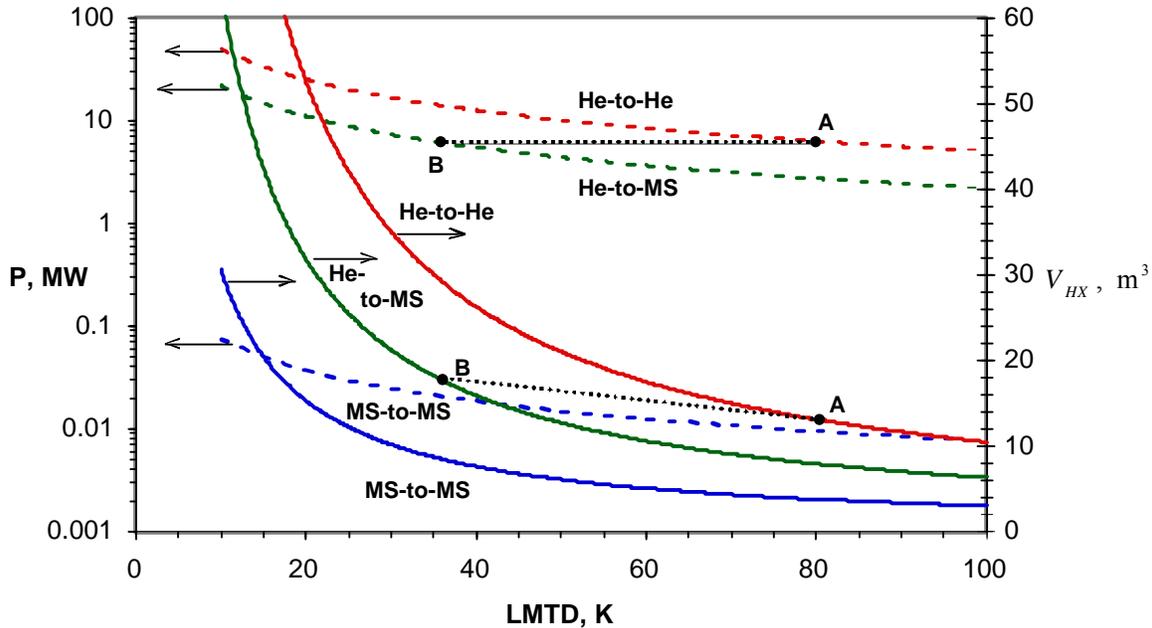
**Fig. 3. Temperature of delivered heat from different reactors.**

#### 4.2 Temperature Drops Across Heat Exchangers

An intermediate heat transport loop is required to transport heat from the reactor to the thermochemical  $H_2$  plant. This heat transport system will be, at a minimum, several hundred meters in length because of (1) the large size of the chemical plant and (2) the need to separate the two facilities for safety. The performance of liquid heat-transport systems is superior to gas heat-transport systems in terms of minimizing temperature drops across heat exchangers and minimizing pumping costs. The power required to overcome heat-exchanger pressure drops in a molten-salt system is typically about one-twentieth that of a helium system.

A recent analysis [Peterson et al. 2003] compared the use of helium with that of molten salts (Fig. 4) in heat transport systems using advanced plate-type compact high-temperature, composite heat exchangers. However, the basic trends apply to any heat exchanger. The performances of 600-MW(t) helium-to-helium, helium-to-molten salt, and molten salt-to-molten salt heat exchangers are shown. For each combination, the power required to force the fluids through the heat exchanger and the volume of the heat exchanger are shown as a function of the log mean temperature difference (LMTD) across the heat exchangers. These three parameters describe the trade-offs in heat-exchanger design. For example, consider the differences between a heat exchanger designed for helium-to-helium heat transfer versus a

heat exchanger designed for helium-to-molten salt heat transfer. If both heat exchangers have the same pumping power requirements (dashed lines) and the helium-to-helium heat exchanger has an 80°C LMDT (point A), the helium-to-molten salt heat exchanger LMDT (point B) will be less than 40°C. If a helium-cooled reactor is used, converting from a helium intermediate heat-transport loop to a molten salt intermediate heat-transport loop reduces the temperature drop across this one heat exchanger by 40°C. A similar reduction in the temperature drop would occur in the second heat exchanger inside the thermochemical H<sub>2</sub> plant. Given that heat must be delivered at a specific temperature to the thermochemical cycle, the reduced temperature drops across these two heat exchangers would allow the peak reactor temperatures to be reduced by 80°C.

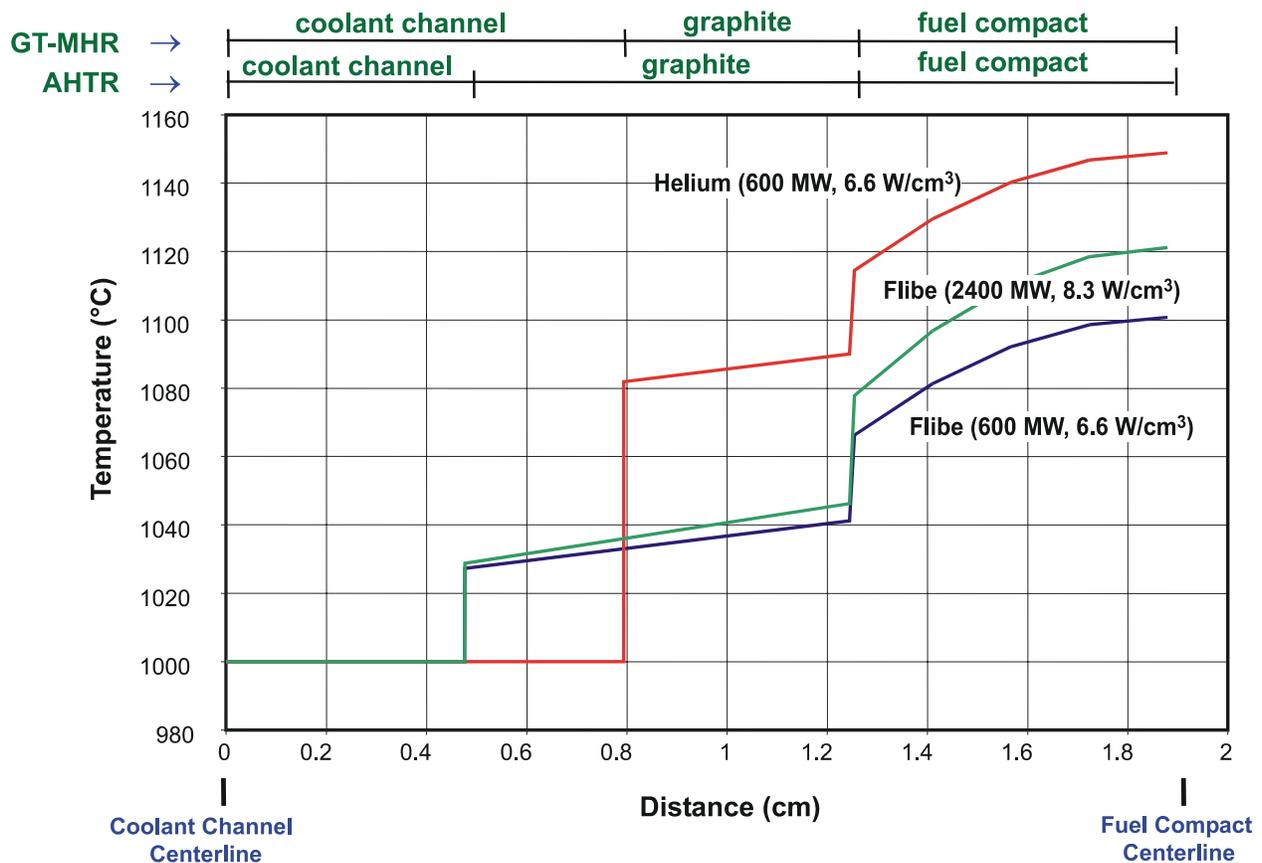


**Fig. 4.** Total pumping power  $P$  (dashed lines) and total volume  $V_{HX}$  (solid lines) for a 600-MW(t) compact heat exchanger as a function of LMTD for three combinations of helium (7 MPa) and/or molten salt.

#### 4.3 Temperature Distributions in Reactor Cores

The heat transfer between a flowing fluid and a fuel element is significantly better for liquids than gases. Figure 5 shows the temperature profile from the centerline of the coolant channel to the centerline of the fuel compact for the average channel at the outlet of the core for different designs of AHTRs and a comparable MHTGR. For this configuration, the coolant temperature was fixed at 1000°C. In all cases, the narrow gap between the fuel compact and the graphite matrix is assumed to be filled with helium. These curves show that for the average channel, the 2400-MW(t) AHTR fuel temperature will be 50°C cooler than that of a MHTGR with the same core power density.

The temperature peaking between coolant channels across the reactor core will be less in an AHTR than in a gas-cooled reactor. The viscosity of liquids decreases with temperature whereas the viscosity of gases increases with temperature. The hottest coolant channel in an AHTR will have greater flow than the coldest channel. The hottest coolant channel in a gas-cooled reactor will have less flow than the coldest channel. This can be partially overcome with appropriate coolant flow control for individual coolant channels. However, the fundamental physical properties of liquids versus gases favors liquids for reducing peak core versus average temperatures.



**Fig. 5. Radial temperature profiles from coolant channel centerline to fuel compact centerline for average temperature channel.**

## 5. ECONOMICS

Preliminary overnight capital costs of the AHTR for several exit temperatures were determined relative to MHTGRs (Ingersoll 2004). Depending upon the assumptions, the AHTR capital costs per kW(e) were estimated between 53 and 61% of that for modular gas-cooled reactors. There are significant uncertainties in these estimates. The lower capital costs are primarily a consequence of two factors: economics of scale [2400-MW(t) vs a 600-MW(t) reactor] and the capability to deliver heat over a small temperature range that translates into higher plant efficiencies for the same reactor coolant exit temperatures.

## 6. CONCLUSIONS

The future production of liquid fuels requires H<sub>2</sub>. Thermochemical production of H<sub>2</sub> using nuclear energy is potentially attractive. However, efficient thermochemical cycles require heat delivered at temperatures between 700 and 850°C—depending upon the specific process. The reactor temperatures will be significantly higher than the peak thermochemical process temperatures and near the limits of materials of construction. The AHTR is a liquid-cooled high-temperature reactor that uses the same basic fuel as gas-cooled reactors. *However, the use of liquid cooling versus gas cooling can reduce peak reactor temperatures by 100 to 200°C for the same temperature of delivered heat.* Although the AHTR is a new

reactor concept with significantly larger technical uncertainties associated with design compared to gas-cooled reactors, the lower AHTR peak reactor temperatures may significantly reduce the challenges in developing very high-temperature materials. The materials challenges may be greater than the design challenges. The larger size of the AHTR may offer significant economic advantages relative to modular reactors.

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