Production of Hydrogen Using Nuclear Energy

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Background Information

- The views expressed herein are those of Dr. Charles Forsberg and do not necessarily reflect the views of Oak Ridge National Laboratory, UT-Battelle, or the U.S. Department of Energy
- Charles Forsberg
  - Experience
    - Oak Ridge National Laboratory
    - Bechtel
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    - Sc.D.: MIT: Nuclear Engineering
    - B.Ch.Eng.: U. Minnesota: Chemical Engineering
  - Professional Engineer (Tennessee)
  - Other
    - American Nuclear Society (ANS) Fellow
    - ANS Special Award 2002: Innovative Reactors
    - AIChE Nuclear Energy Division Hydrogen Subpanel Chair
    - 200+ Publications; 9 patents
- Presentation odd pages; backup information even pages

Hydrogen Demand and Nuclear Energy

The size of individual hydrogen production plants and the total market demand for hydrogen are sufficient to justify development and use of nuclear reactors for hydrogen production

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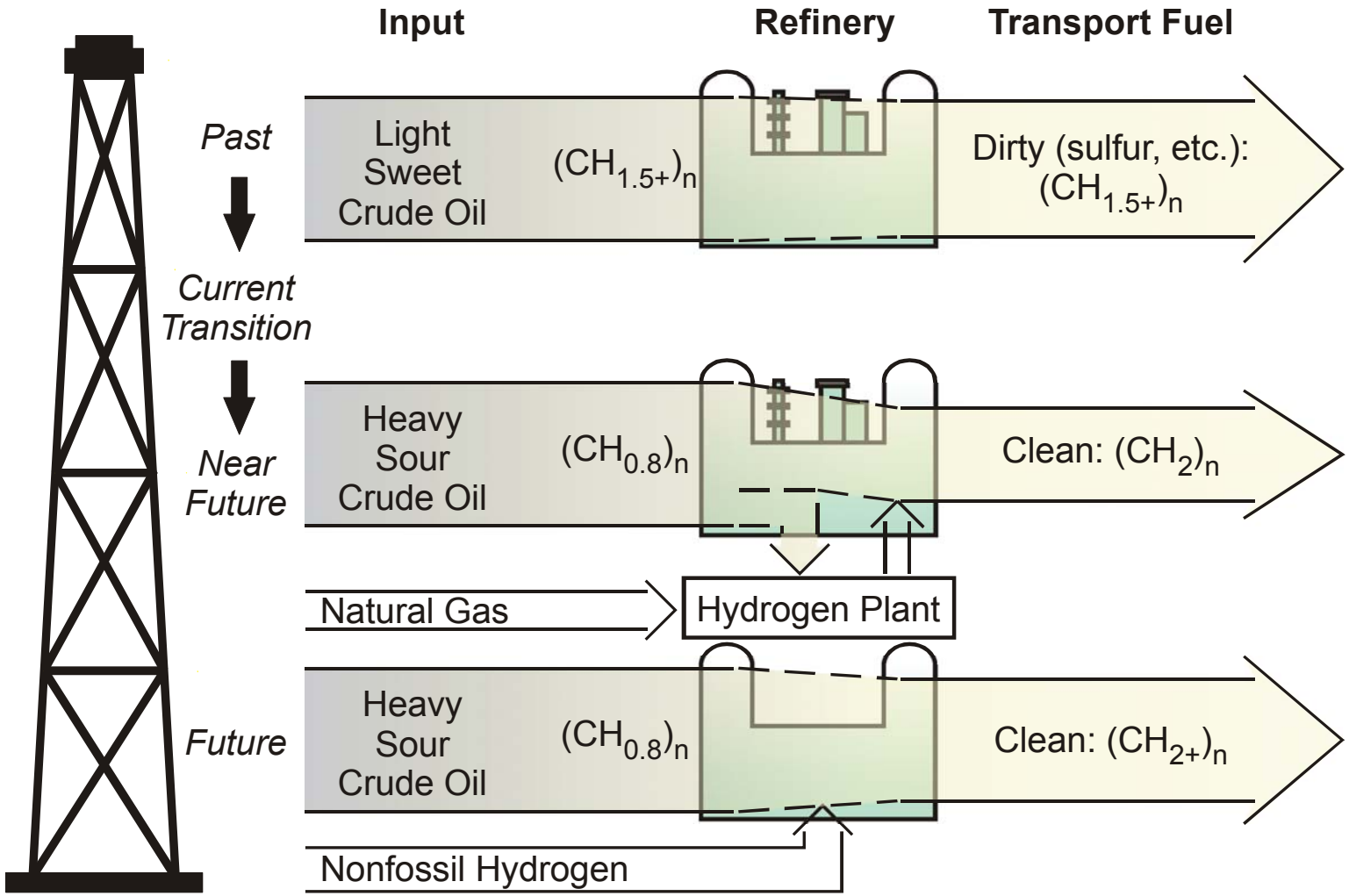
Liquid Fuels Production Is Rapidly Becoming the Major Market for Hydrogen



Hydrogen Demand Is Large and Growing Rapidly

- World consumes 50 million tons of H_2/year
  - ~80% intentionally produced; remainder, by-product H_2
  - 200 GW(t) if the H_2 is burned
  - 4 to 10% growth per year
  - Applications: fertilizer, chemical industry, liquid fuels
- Within decades, the energy to produce H_2 in the United Sates may exceed current energy production from nuclear power
- Increased use of heavy crude oils is anticipated as sources of light crude oils become depleted
  - More H_2 is required to produce equivalent liquid fuels
  - Trend has existed for decades with no expectation for a change in the trend

Nuclear Energy Is Now Compatible with Hydrogen Production Because of the Growth in the Size of Hydrogen Production Facilities

Plant Size

H_2 Plants	Energy to Produce H_2 (50% Efficient)
200×10^6 scf/d (Existing)	1600 MWT
300×10^6 scf/d (Future)	2400 MWT

Time

Nuclear Reactor

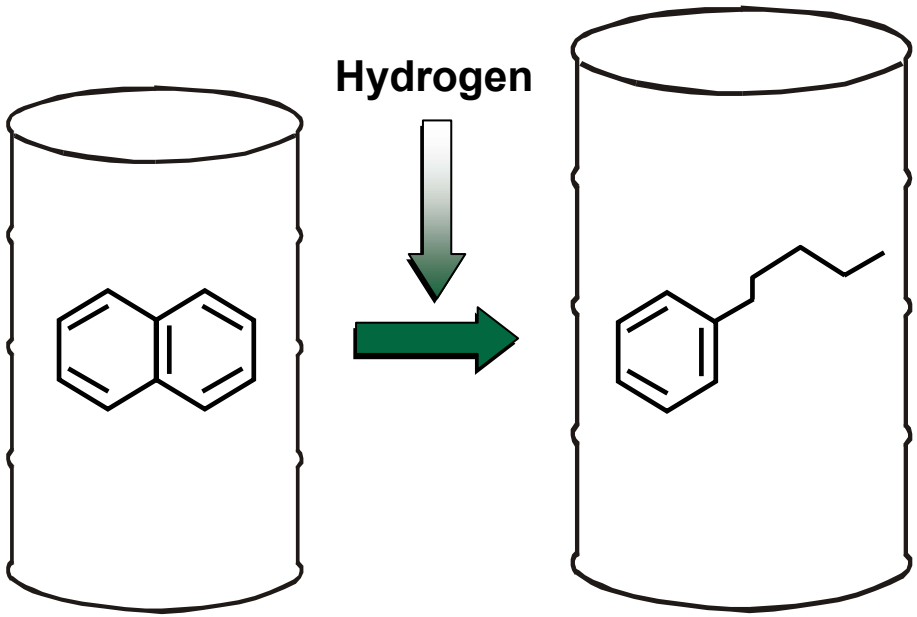
600–4000 MWT

Hydrogen Plant Size Is Now Compatible with Nuclear Energy

- Nuclear reactors are economic only in large sizes
- Large growth in demand for H_2 by refineries
- In many parts of the world, refineries are connected to hydrogen pipelines
  - Avoid downtime
  - Trade H_2
  - Merchant plants
- Scale of hydrogen production matches nuclear energy
  - Total demand is sufficient to justify development of nuclear hydrogen production systems
  - Single-train hydrogen plant size matches nuclear reactor size
- System is suited to hydrogen production using nuclear reactors with a business-as-usual future

Addition of Hydrogen Can Increase Liquid Fuel Yields per Barrel of Oil by Up to 15%

(Potential Nuclear-Hydrogen Midterm Market)



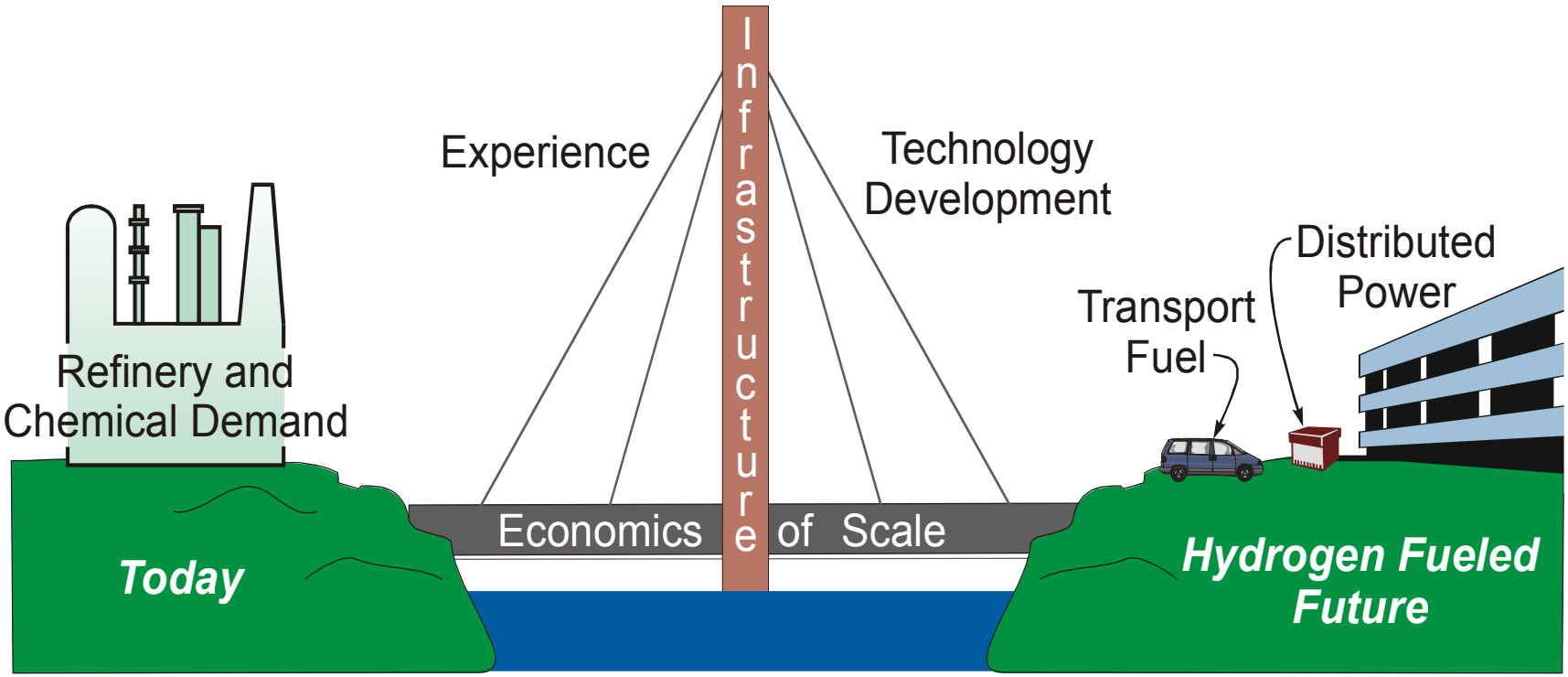
Benefits

- **Lower imports**
- **Reduced CO_2**
- **Reduced NO_X**
- **Reduced sulfur**
- **Reduced diesel particulate**

There Are Large Incentives to Develop Lower-Cost H_2 Production Methods for H_2-Boosted Liquid Fuels—Independent of Any "H_2 Economy"

- **Benefits**
  - 15% more fuel per barrel of oil (best case; decreased dependence on imports)
  - Reduced CO_2 emissions from the vehicle
  - Reduced NOX (Lower flame temperature)
  - Reduced sulfur in fuel (processes remove sulfur)
  - Reduced particulate with fewer multi-ring compounds (diesel smoke)
- **Economics near break-even point**
  - Dependent upon H_2 costs and costs of crude oil
  - Hydrogen production not tied to natural gas or oil enables option as oil prices increase (natural gas for hydrogen production tied to oil prices, thus hydrogen from this route may follow cost curve for oil)
- **Potential hydrogen market >5% of the total U.S. energy demand**
- **Technology (primary chemical mechanism)**
  - Break polyaromatic rings
  - Open one ring and saturate with hydrogen
  - Ring with tail
- **Technical contacts (ORNL): W. Steele and R. Uhrig**

The Growing Hydrogen Demand Creates a Bridge to the Hydrogen Economy—With a Future Hydrogen Energy Demand That May Exceed That for Electricity



Hydrogen Could Exceed Electricity as an Energy Carrier by 2050

- Auto companies and Presidential initiative to develop fuel cells for cars within 10 years
- Technological transitions typically take several decades
  - 40 years from whale-oil lamps to electricity
  - 30 years from horses to cars
- If H_2 replaces liquid fuels, H_2 demand may exceed electricity demand
  - Y in energy projections
  - One future, electricity; the other future, H_2 and electricity
  - No solid basis to predict which future will occur

Methods to Make Hydrogen Using Nuclear Energy
(Reactor Provides High-Temperature Heat)

Nuclear-assisted steam reforming of natural gas

Hot electrolysis

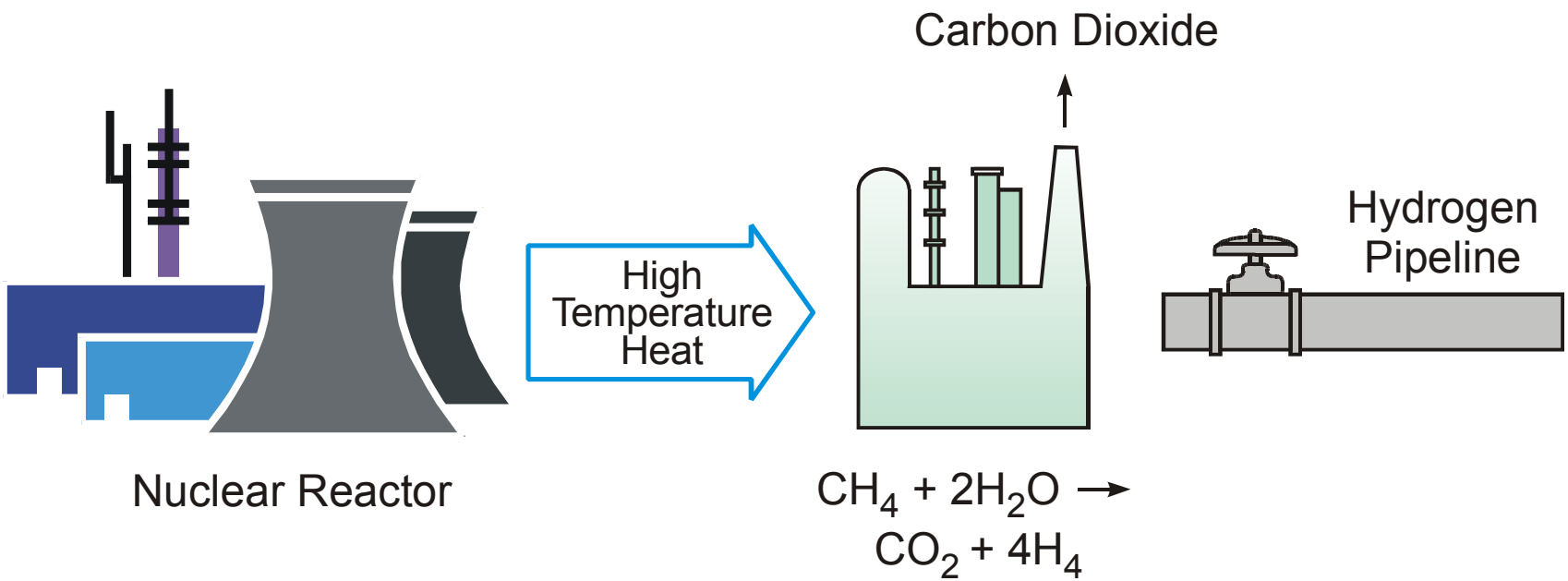
Thermochemical cycles

Characteristics of Current Hydrogen Production Techniques

- Most H_2 is made from natural gas
  - Heat + methane (CH_4) + water (H_2O) $\Rightarrow$ hydrogen (H_2) + carbon dioxide (CO_2)
  - Endothermic process with heat input at ~800°C
- Water electrolysis is used to produce small quantities of H_2
  - Inefficient: heat to electricity to H_2
  - Viable when electricity is cheap

Nuclear-Assisted Hydrogen Production Uses High-Temperature Heat (~800°C) to Reduce Energy Requirements for Steam Reforming of Natural Gas

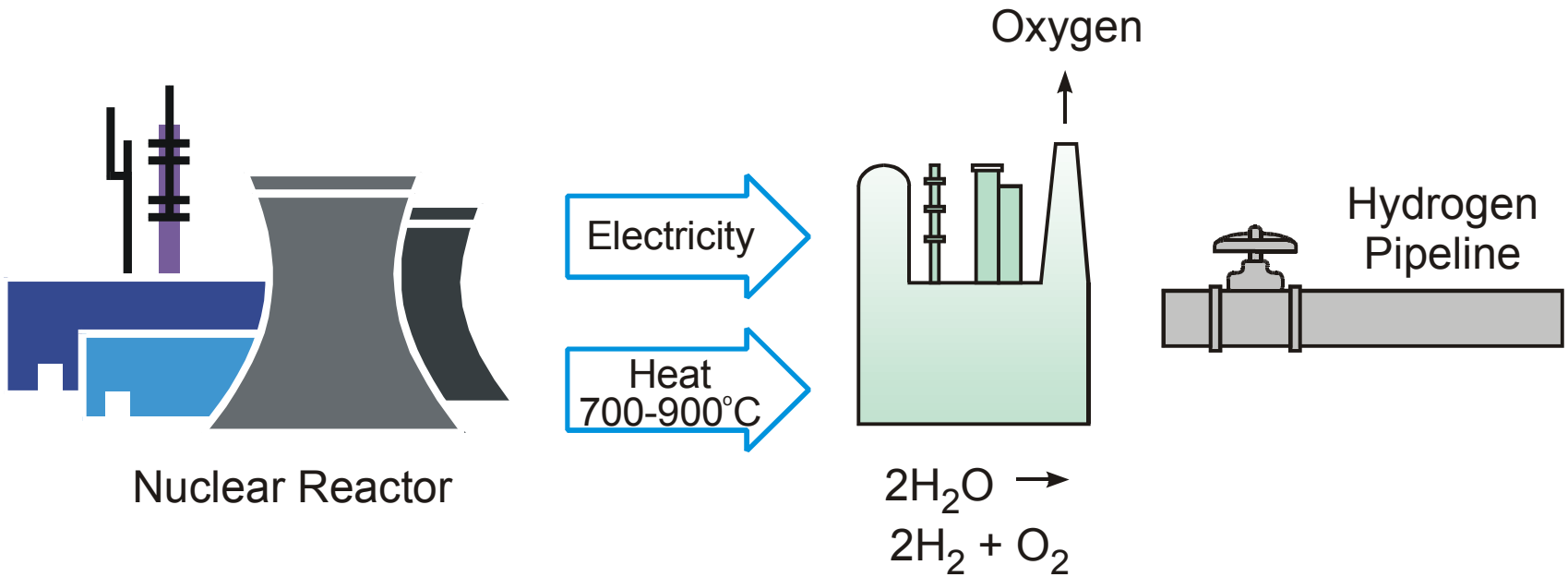
(Development Program in Japan)



Nuclear-Assisted Steam Reforming of Natural Gas Is a Near-Term Option

- Chemical reactions in steam reforming
  - $CH_4 + H_2O \Leftrightarrow CO + 3H_2$
  - $CO + H_2O \Leftrightarrow CO_2 + H_2$
- Steam reforming is an endothermic process
  - Natural gas used for two purposes
    - Reduced form of hydrogen
    - Source of heat
  - High-temperature heat from nuclear reactor replaces use of natural gas as a heat source in the process
- Japan has a major R&D program
  - Japanese estimates indicate that the technology is economically viable today (Japan imports liquified natural gas)
  - Cold test bed (140 m^3/h) operational
  - High-Temperature Test Reactor (HTTR) to supply heat
    - Started operation in 2001
    - Peak temperature: 950°C
    - 30 MW(t)

Hydrogen Production Using Hot Electrolysis Requires High-Temperature Heat (700-900°C) and Electricity

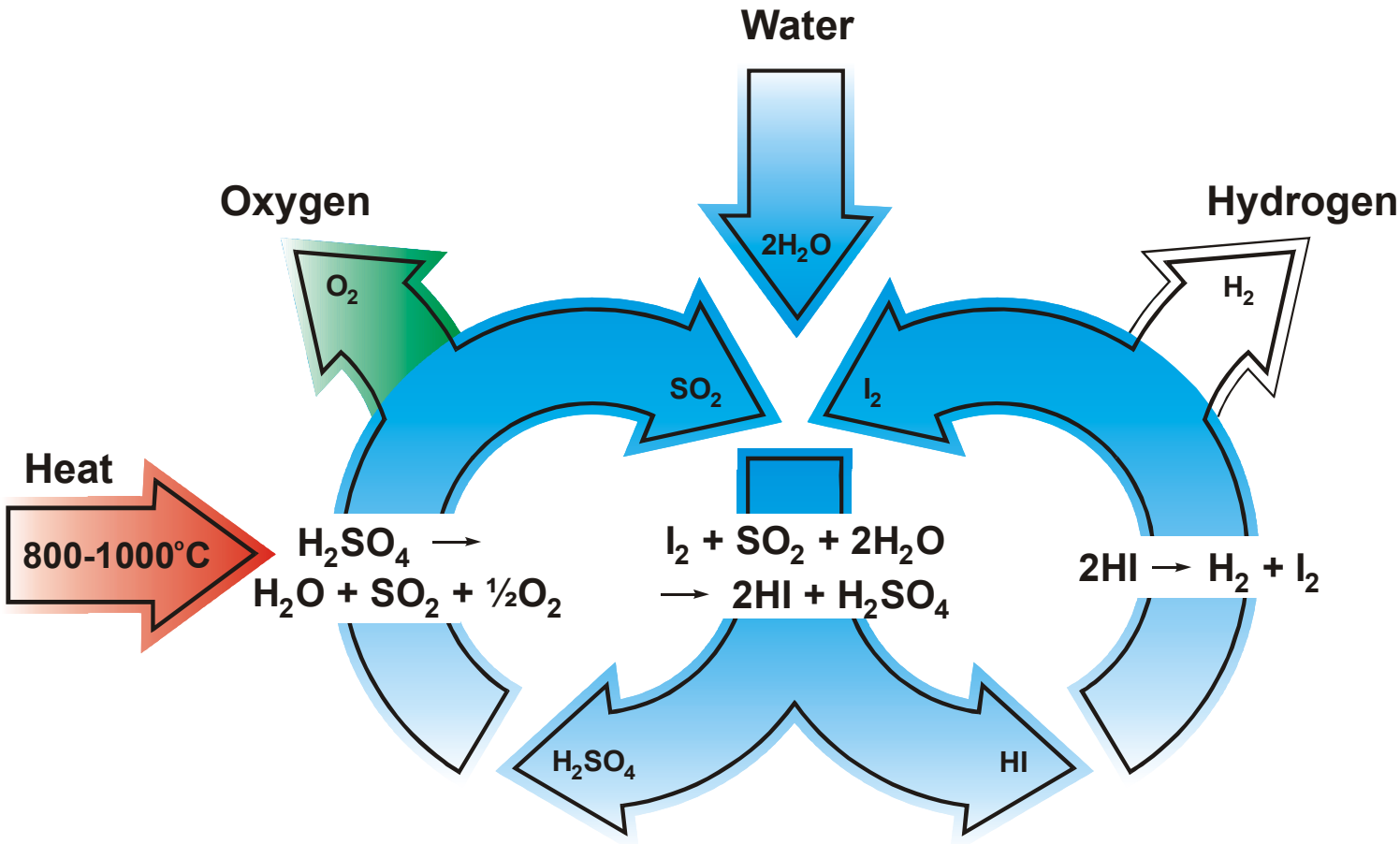


Hydrogen from Hot Electrolysis

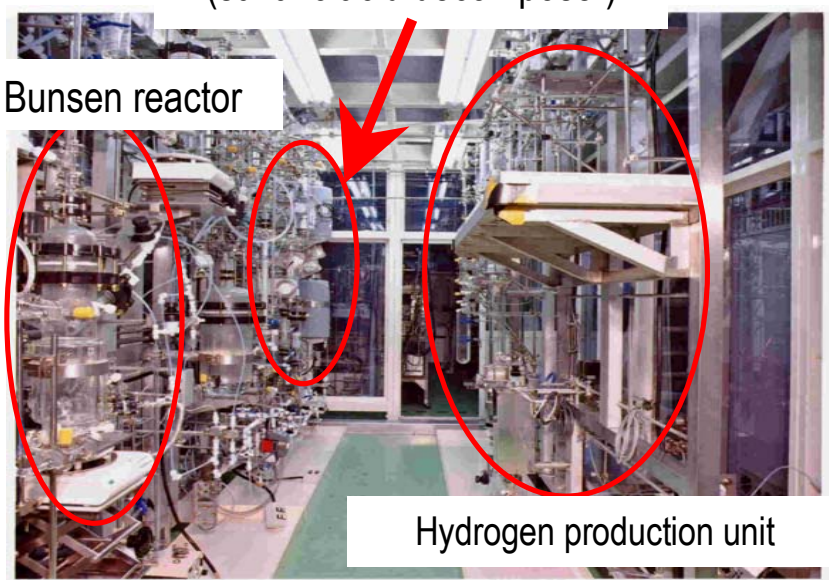
- Heat + water (H_2O) + electricity $\Rightarrow$ hydrogen (H_2) + oxygen (O_2)
- Heat replaces some of the electric demand
- Heat input at 700 to 900°C
- High-temperature operation may also reduce other losses in the electrolysis process
- Based on high-temperature fuel cell technology

Thermochemical Processes Convert High-Temperature Heat and Water to H_2 and Oxygen

(Leading Candidate: Iodine-Sulfur Process: Potential for Lower Peak Temperatures)



Thermochemical Processes



- **General characteristics**
  - Heat + water ⇒ hydrogen (H_2) + oxygen (O_2)
  - Heat input >750°C
  - ~100 such cycles identified
  - Sulfur-Iodine is the leading candidate (new technologies may significantly lower peak temperatures)
- **Low pressure**
  - Chemical reactions favored at low pressures
  - Materials requirements minimized
- **Lowest potential costs**
  - Projected efficiencies of >50%
  - Projected best long-term economics (60% cost of cold electrolysis)
- **Program initiated in the United States**
- **Larger program in Japan on iodine-sulfur (IS) process**
  - Laboratory IS loop (left): 50 l/h
  - Cold pilot scale: 30 m^3/h (2010)
  - Plan to connect to reactor (after nuclear-assisted steam-reforming tests)
    - High-Temperature Test Reactor (Operating)
    - 1000 m^3/h
  - Ref: K. Onuki et. al, "R&D on Iodine-Sulfur Thermochemical Water Splitting Cycle at JAERI", 2ed Topical Conference on Fuel Cell Technology (Embedded Topical), 2003 American Institute of Chemical Engineers , New Orleans, March 30-April 3, 2003

Efficient Hydrogen Production Imposes Tough Requirements on the Reactor

- Economics: match expected H_2 plant size
  - Largest H_2 plants on order: 300 million ft^3/d
  - Nuclear heat (50% efficiency): 2400 MW(t)
- High temperatures (750 to 1000°C)
- Heat delivered at constant temperature
- Low pressure to hydrogen facility
  - Match process requirements
  - Minimize potential accidental releases of toxic chemicals (off-site public safety)
- Separation of hydrogen chemical plant from the reactor (plant safety)
  - Corrosive and hazardous chemicals

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Reactor Options for Hydrogen Production

Hydrogen Production Requirements

Define Reactor Requirements, Which, in Turn, Define Viable Reactor Options

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Five Reactor Concepts Have the Potential for Hydrogen Production

(However Existing Nuclear Fuels Technology Implies Only Two Viable Candidates for Deployment Before 2020)

Reactor Option	Demonstrated Temperature	Developed Fuel
Advanced High-Temperature Reactor	Yes	Yes
Very-High-Temperature Reactor	Yes	Yes
Molten Salt Reactor	Yes	No
Lead-Cooled Fast Reactor	No	No
Gas-Cooled Fast Reactor	No	No

Details on Reactor Options

- **VERY-HIGH-TEMPERATURE REACTOR (VHTR).** The VHTR 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 transfer 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 is 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. Several demonstration HTGRs have been built for electricity production. Japan recently began operating the High-Temperature Test Reactor (HTTR), a small VHTR [30 MW(t)] to develop the technology for efficient production of H_2 and electricity. The helium reactor exit temperature is 950°C. 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 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. The Aircraft Reactor Experiment (1950s) operated normally with molten salt exit temperatures of 815°C, with peak operating temperatures up to 860°C 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 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 ($3NaF\text{-}AlF_3$) in very large graphite baths at ~1000°C. 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 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 periods of time. The operating temperatures of these reactors were near 500°C. 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 uses an advanced fuel (several options are 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. It is at a very early stage of development, and no decisions have been made on what type of fuel to use.

Two Nuclear Power Plans Options For Hydrogen Production

Very High Temperature Reactor

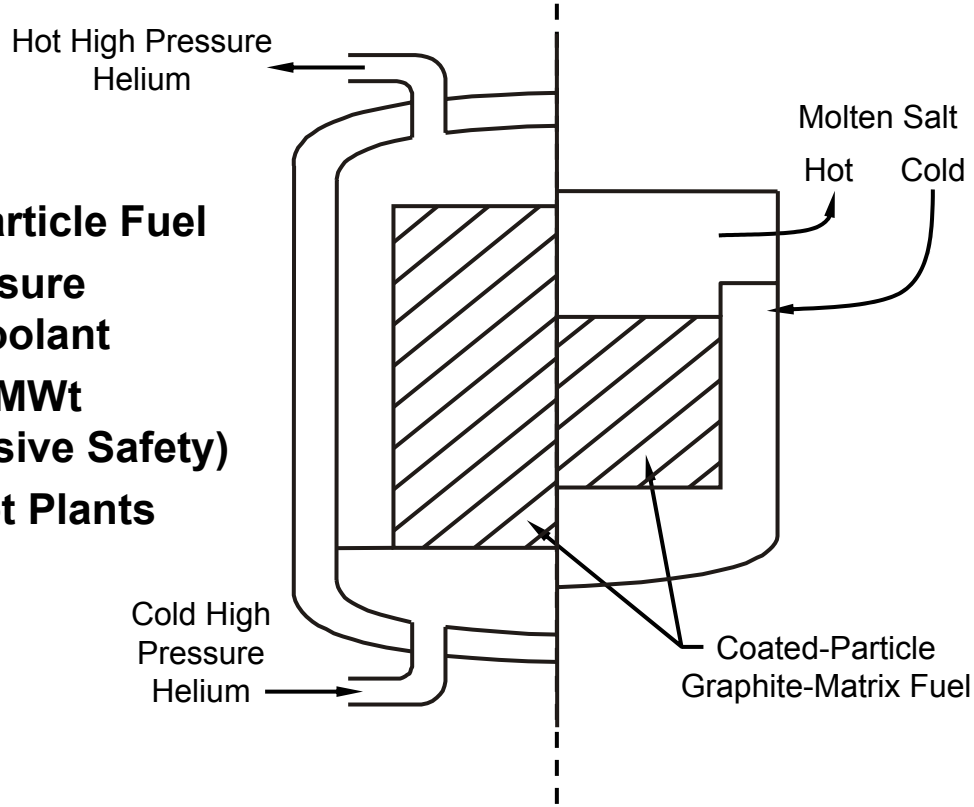
(Developed Originally for Electric Production)

- **Coated Particle Fuel**
- **High Pressure Helium Coolant**
- **Size: 600 MWt (With Passive Safety)**
- **Small Pilot Plants**

Advanced High Temperature Reactor

(Designed for Hydrogen Production)

- **Coated Particle Fuel**
- **Atmospheric Pressure Molten Salt Coolant**
- **Size: To 2400 MWt (With Passive Safety)**
- **New Concept**



Viable Reactor Options Within 20 Years
(Both Use the Same High-Temperature Fuel: The Limiting Factor in the Choice of Reactor Options)

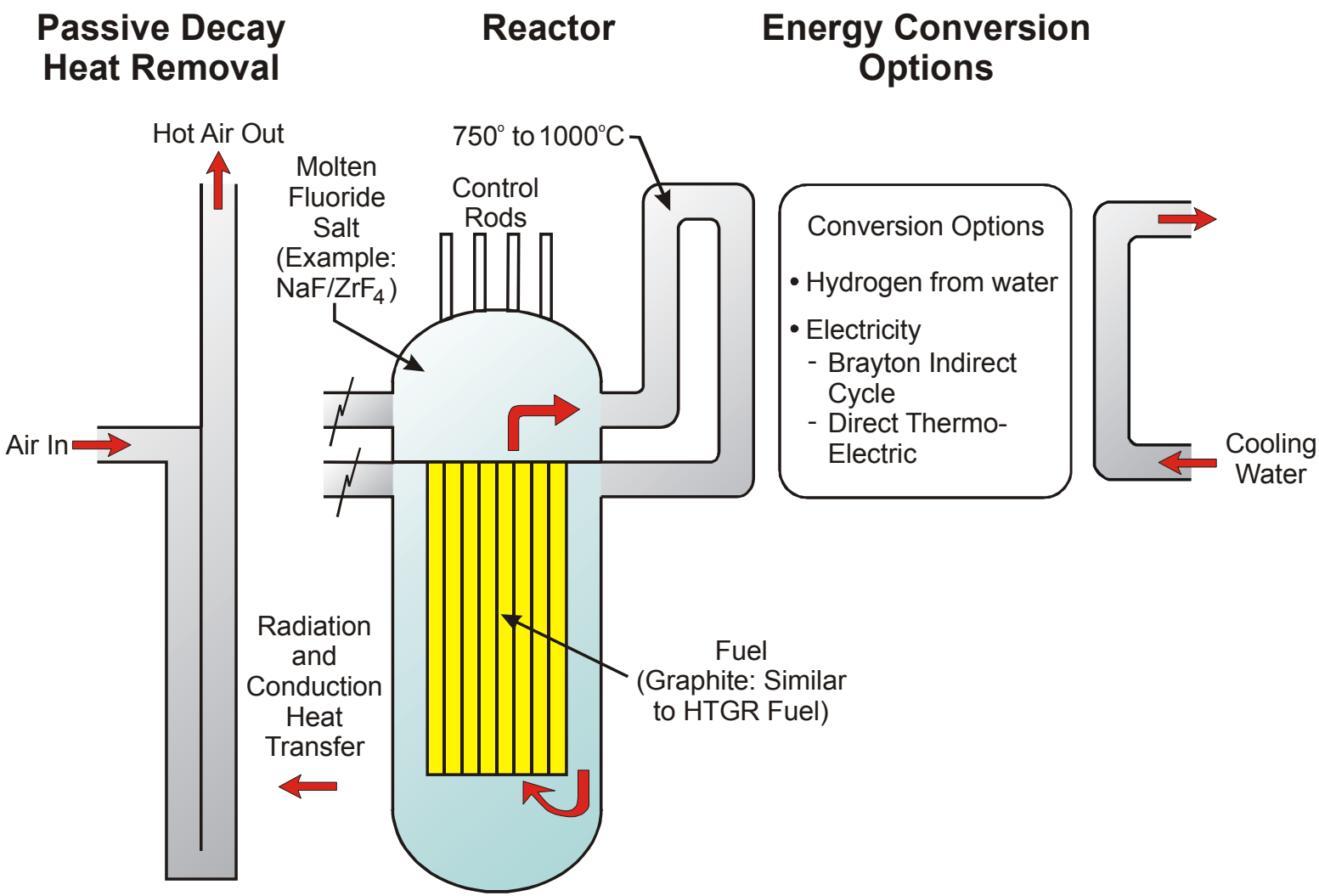
AHTR

- New concept
  - Designed to match hydrogen production requirements
  - Base technologies demonstrated
- Graphite-matrix coated-particle fuel
- Molten-fluoride-salt coolant
  - Atmospheric pressure
- Size: 2400 MW(t)
  - Size defined by the goal of passive nuclear safety system
- Technology considerations
  - Developing integrated design
  - Testing basic reactor concept
  - Maximum reactor/heat-exchanger temperatures less than those of the VHTR. Key components operate at significantly lower temperatures

VHTR

- Several demonstration reactors for electricity production
  - Good match for electricity
  - Direct-cycle helium gas turbine
  - Not demonstrated for hydrogen production
- Graphite-matrix coated-particle fuel
- Helium coolant (high pressure)
- Size: 600 MW(t)
  - Size defined by the goal of passive nuclear safety system
- Technology considerations
  - Boosting temperatures to match requirements

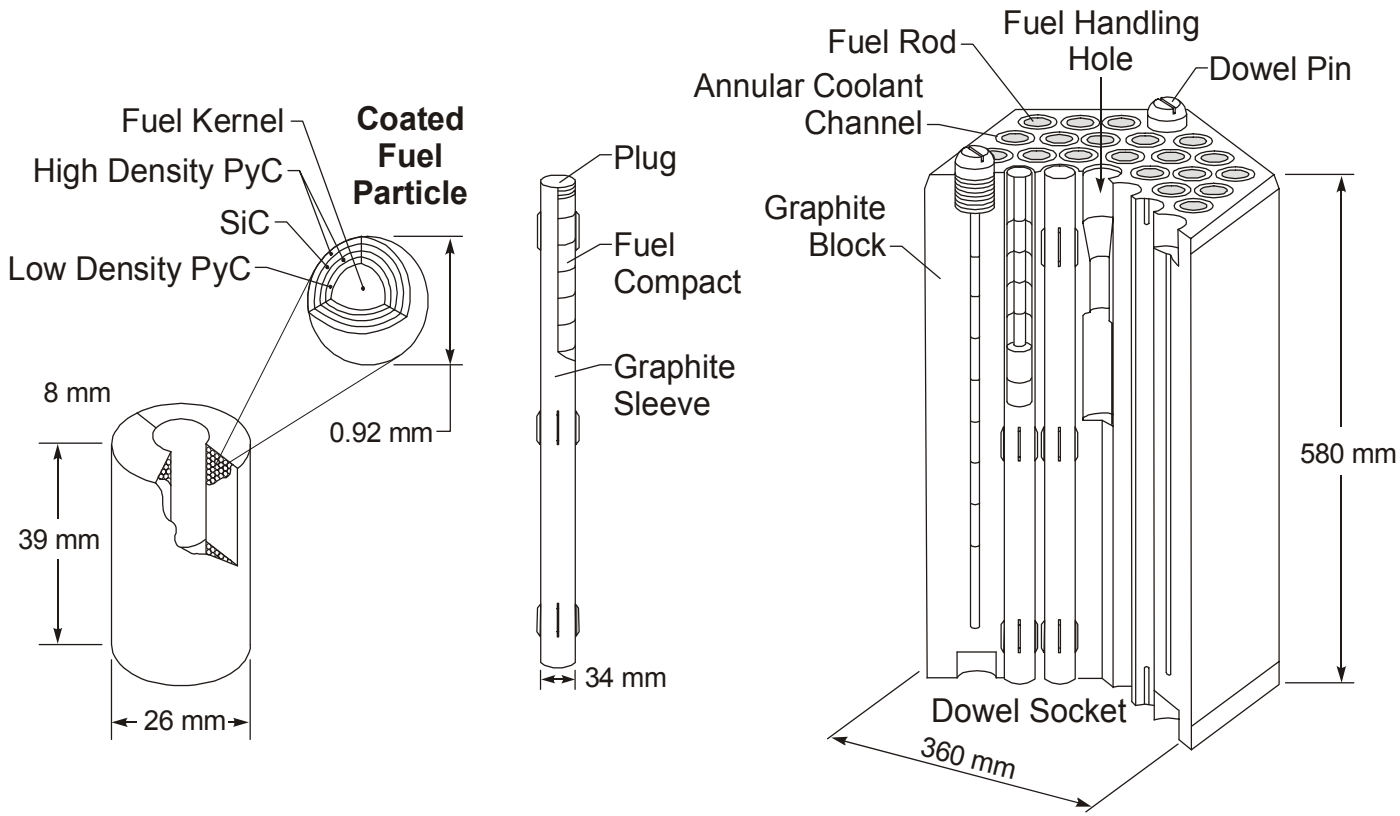
Advanced High-Temperature Reactor



Advanced High Temperature Reactor
(Very Early Pre-conceptual Design)

- Power level: 2400 MW(t)
- Nuclear safety systems
  - Reactor kinetics somewhat similar to HTGR
    - Preliminary concept with annular core
  - Passive decay heat removal (multiple options)
    - Air-cooled (similar to GE PRISM Fast Reactor)
    - Water jacket (similar to Modular HTGR (S. Africa))
- Fuel: graphite-matrix coated particle
  - Same basic fuel as VHTR and HTGR
  - Nominal peak temperature 1200°C
  - Maximum accident temperature without major fission product release: 1600°C
- Coolant: molten salt
  - Leading candidate (same as aircraft propulsion program): $NaF\text{-}ZrF_4$
  - Several other salt options with similar properties such as $NaF\text{-}RbF\text{-}ZrF_4$ (8%/50%/42%)
  - Boiling point ~1400°C
- Other
  - May use down-flow core

The AHTR and VHTR Use Graphite-Matrix Coated-Particle Fuels: Only Proven Fuel That Operates at Required Temperatures for Hydrogen Production



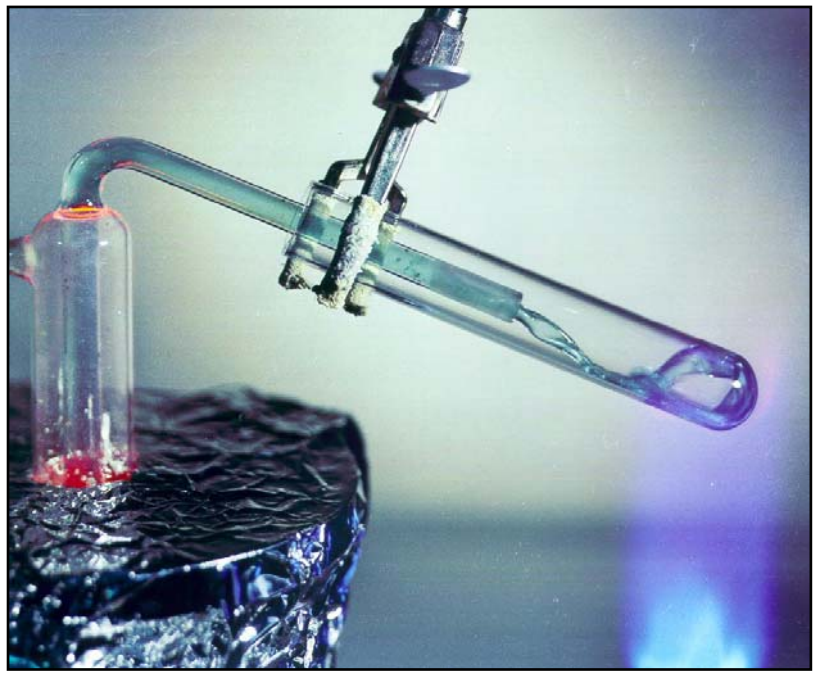
Fuel Compact **Fuel Rod** **Fuel Block**

The AHTR and VHTR Use Graphite-Matrix Coated-Particle Fuels

- Only high-temperature fuel with which there is any significant experience
  - Used in multiple test and demonstration reactors
  - Used in High-Temperature Test Reactor (HTTR)
    - Operated in Japan since 2001
    - 950°C helium exit temperature
- Coated-particle graphite-matrix fuel
  - Normal temperature limit of ~1200ºC
  - Accident limit of ~1600°C
    - Limited time with no significant radionuclide release

AHTR Molten Salt Reactor Technology Was Developed in the Aircraft Nuclear Propulsion Program with Test Reactor Operations at 860°C

(Aircraft and Hydrogen Reactor Requirements Similar : High Temperature and Low Pressure; Current Molten Salt coolant Interests: AHTR and Fusion Reactor Cooling)



Molten Salt Coolant Properties and History

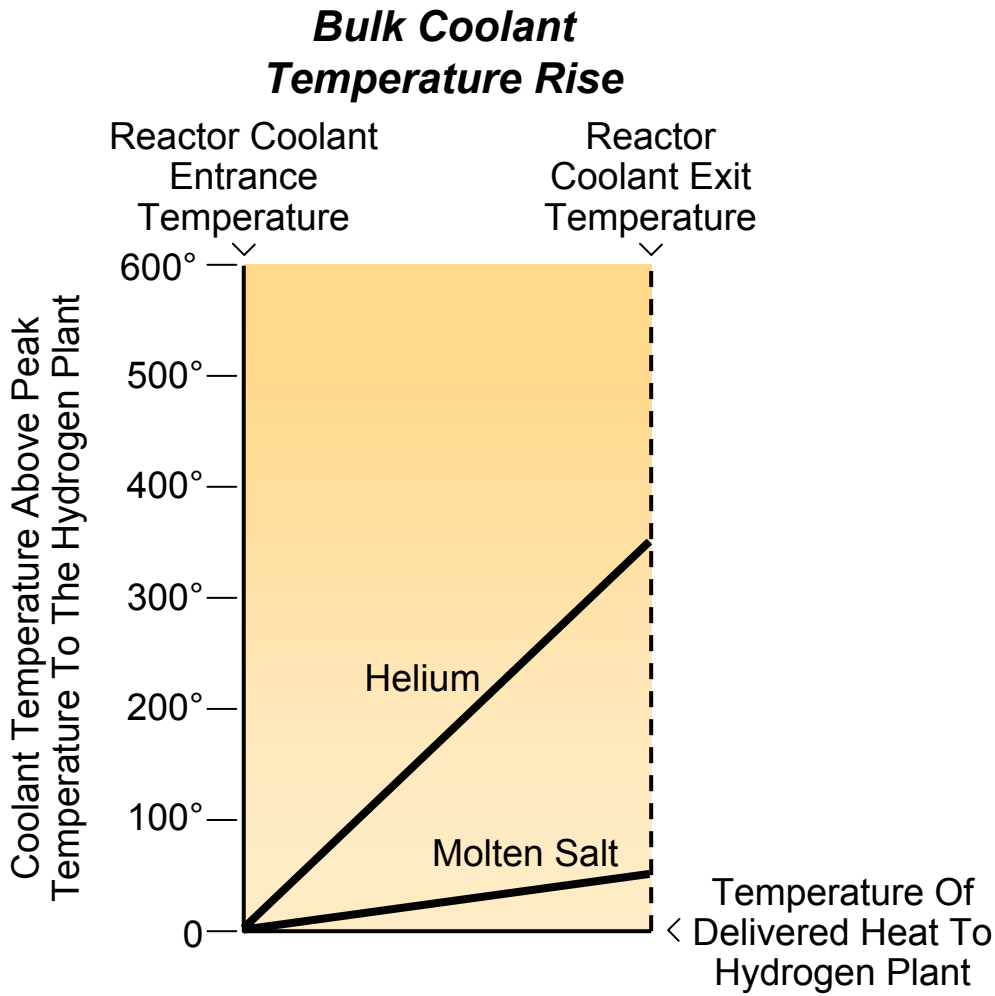
Properties

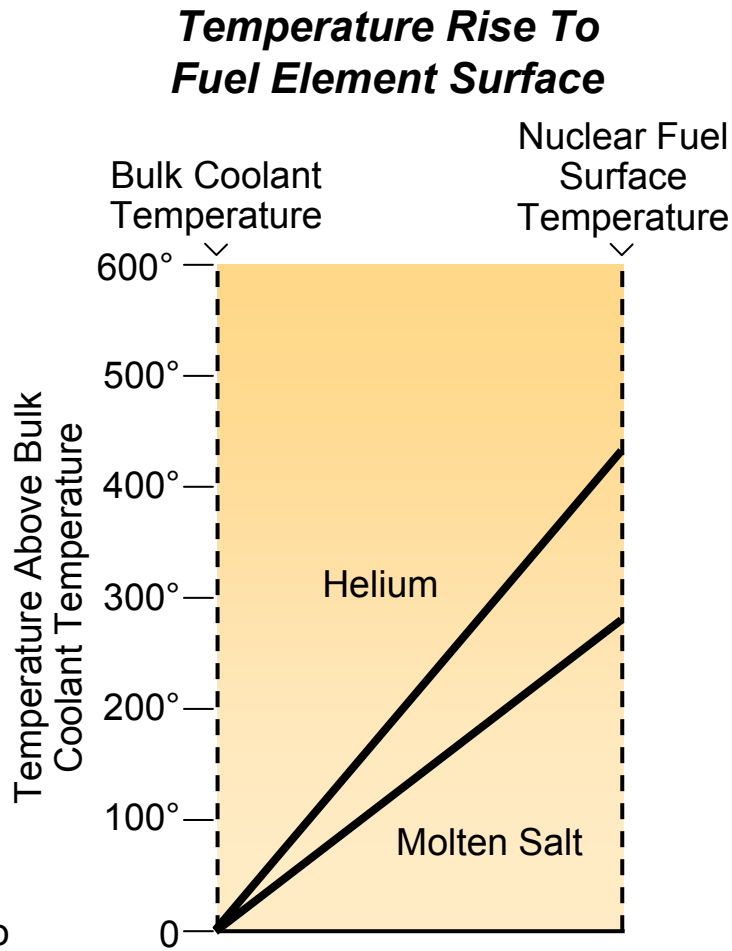
- Only fluoride molten salts have appropriate nuclear properties (low nuclear absorption cross sections)
- Very large experience base
  - Aluminum industry
    - Hall process for aluminum production (used since 1890s)
    - Electrolysis in cryolite ($3NaF\text{-}AlF_3$) bath at about 1000°C
  - Nuclear industry (see history on right)
- Molten salts compatible with fuel
  - Graphite-based fuel
  - Graphite used in aluminum industry to hold salt
  - Reactor experience (see right)
- Molten salt compatibility with metals
  - Maintain salts under reducing conditions
  - Noble with respect to materials of construction
- Transparent
- High heat capacity
  - 4X better than sodium
- No rapid chemical reactions
- Low pressure
- Fluoride salt options
  - Salt mixtures
  - Candidate fluorides (Na, Zr, Rb, Be, etc.)

History

- Historical progression
  - Early programs (Aircraft Nuclear Propulsion Program and Molten Salt Breeder Reactor Program examined molten-salt-fueled reactors (fuel dissolved in salt)
  - AHTR has solid fuel and clean coolant
    - Coated-particle fuel developed after termination of these programs
    - All other nuclear reactors worldwide use solid fuels
  - Current molten salt R&D
    - Leading candidate for cooling fusion reactors
    - Test loops operating for several applications of molten salts
- Aircraft Nuclear Propulsion Program
  - Goal: Unlimited-range military aircraft
  - $ 1B Investment (1946-1961)
  - Operated the Aircraft Reactor Experiment—a small test reactor with a $NaF\text{-}ZrF_4$ molten salt
  - Project cancelled because of excessive aircraft weight
  - Aircraft and hydrogen reactor requirements similar: high temperature and low pressure
- Molten Salt Reactor
  - Goal: Breeder reactor
  - Graphite moderator (not fuel)
  - Operated Molten Salt Reactor Experiment—an 8 MW(t) test reactor
  - Project cancelled because of U.S. decision to develop only one breeder reactor concept

Liquid Coolants Are Better Heat Transfer Agents Than Gases



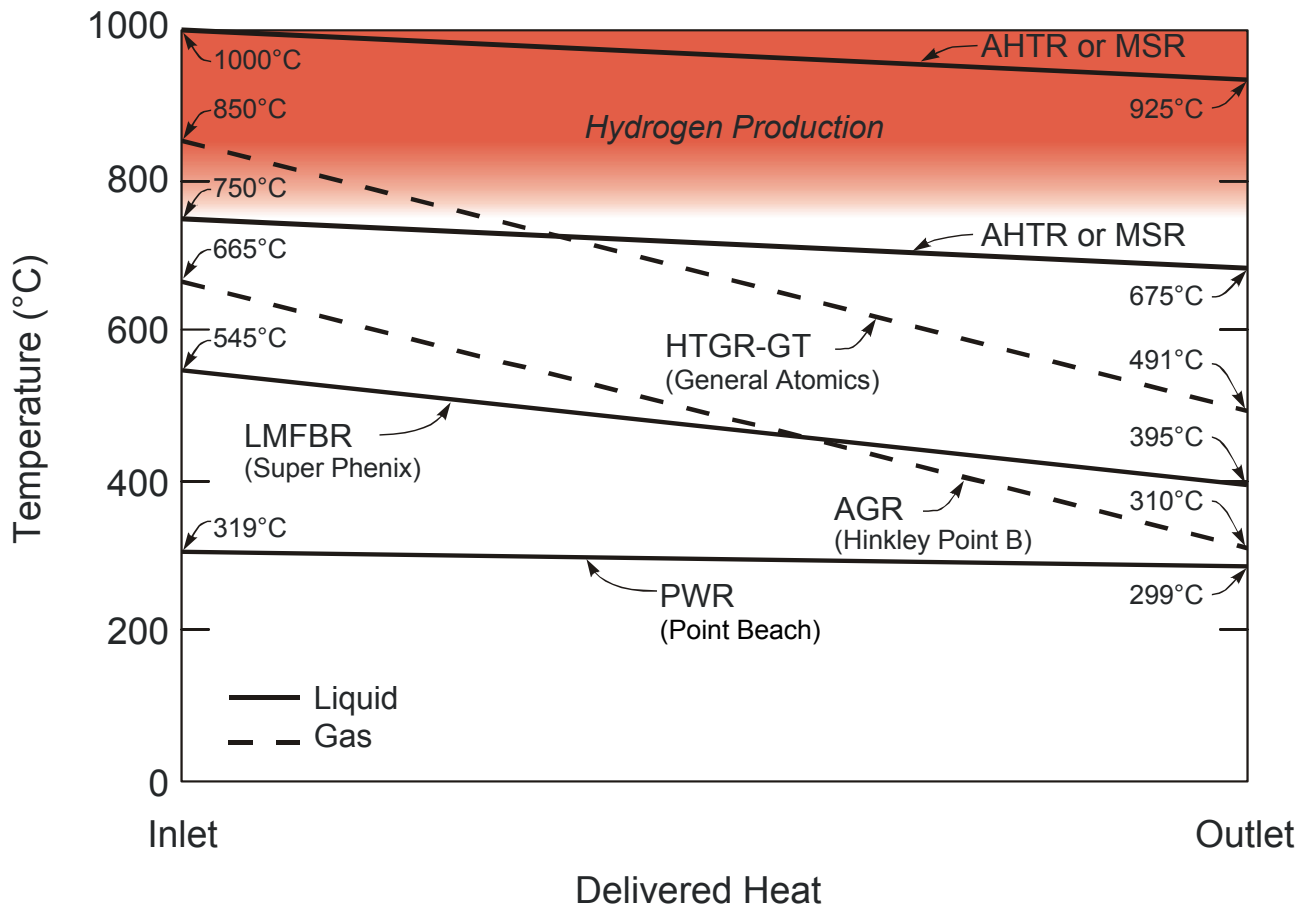


Heat-Transfer Considerations

- Temperature drops are required to transfer heat from the reactor fuel to the hydrogen production plant
- Comparison of molten-salt liquid coolant (AHTR) and helium gas coolant (VHTR)
  - Example uses historical demonstrated practice—the new HTTR uses a new fuel design that may lower temperature drops, but at a cost of more expensive fuel designs and greater pumping costs
  - Temperature rise across the reactor core (temperature drop across hydrogen plant)
    - Liquid cooled: 50°C
    - Gas cooled: 350°C
  - Temperature drop from surface of standard fuel block to fluid in cooling channel
    - Liquid cooled: 280°C
    - Gas cooled: 415°C
  - Calculations exclude:
    - Probable need for an intermediate heat-exchanger loop with temperature losses
    - Enhanced molten-salt heat transfer (not well understood in terms of engineering) at very high temperatures from infrared heat transfer in a transparent high-heat-capacity salt. Mechanism for heat transfer from fuel to coolant
- Relatively few reactor heat-transfer studies at these temperatures

Liquid Reactor Coolants Can Deliver the Heat Over a Small Temperature Range

(Minimizing the Reactor Core and Heat-Exchanger Temperatures)



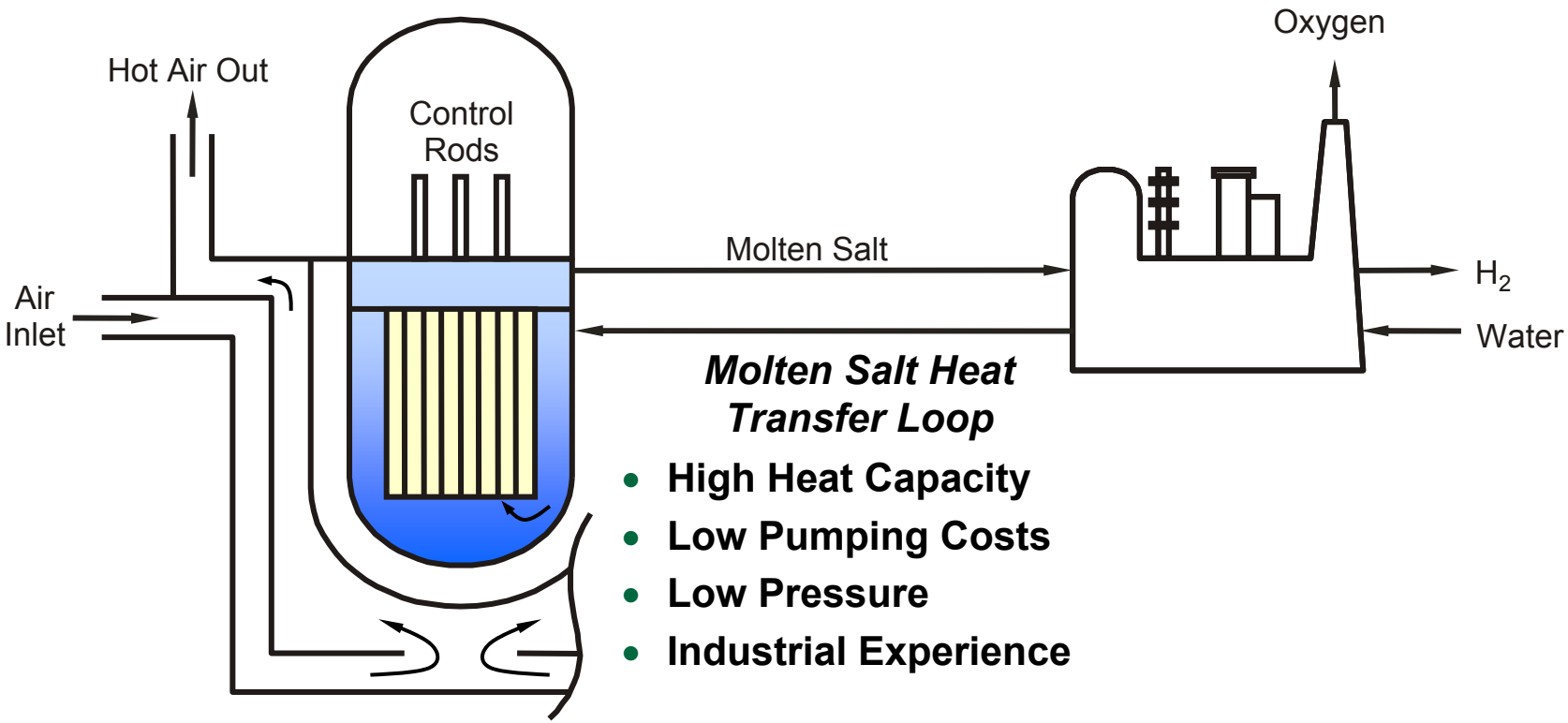
Factors Controlling Transfer of Heat From a Reactor to the Application

- The temperature increase across a reactor core and the corresponding temperature drop to the application for the heat involve economic tradeoffs
- Liquids are better than gases for moving heat
- Experience with real reactors shows the very large difference between liquid-cooled and gas-cooled reactors
  - PWR (liquid coolant). The example is the operating Point Beach pressurized-water reactor in Wisconsin. The heat produces steam for production of electricity. There is an 11°C difference from the hottest to coldest temperatures in the primary coolant circuit.
  - LMFBR (liquid coolant). The example is the now-shutdown French Super-Phenix liquid metal (sodium) fast breeder reactor. The heat was used to produce steam for production of electricity. There was a 150°C difference from the hottest to coldest temperatures in the primary coolant circuit.
  - AGR (gas coolant). The example is the operating British Hinkley Point B advance gas-cooled (carbon dioxide) reactor. The heat produces steam for production of electricity. There is a 355°C difference from the hottest to coldest temperatures in the primary coolant circuit.
  - HTGR (gas coolant). The example is the proposed General Atomics high-temperature gas (helium) reactor, which drives a helium gas turbine for electricity production. There will be a 359°C difference from the hottest to coldest temperatures in the primary coolant loop.

The Reactor and Hydrogen Production Facility Will Be Physically Separated to Ensure Safety
(Molten Salt Provides for Efficient Heat Transfer)

Nuclear Safety by Isolation

Hydrogen Safety by Dilution



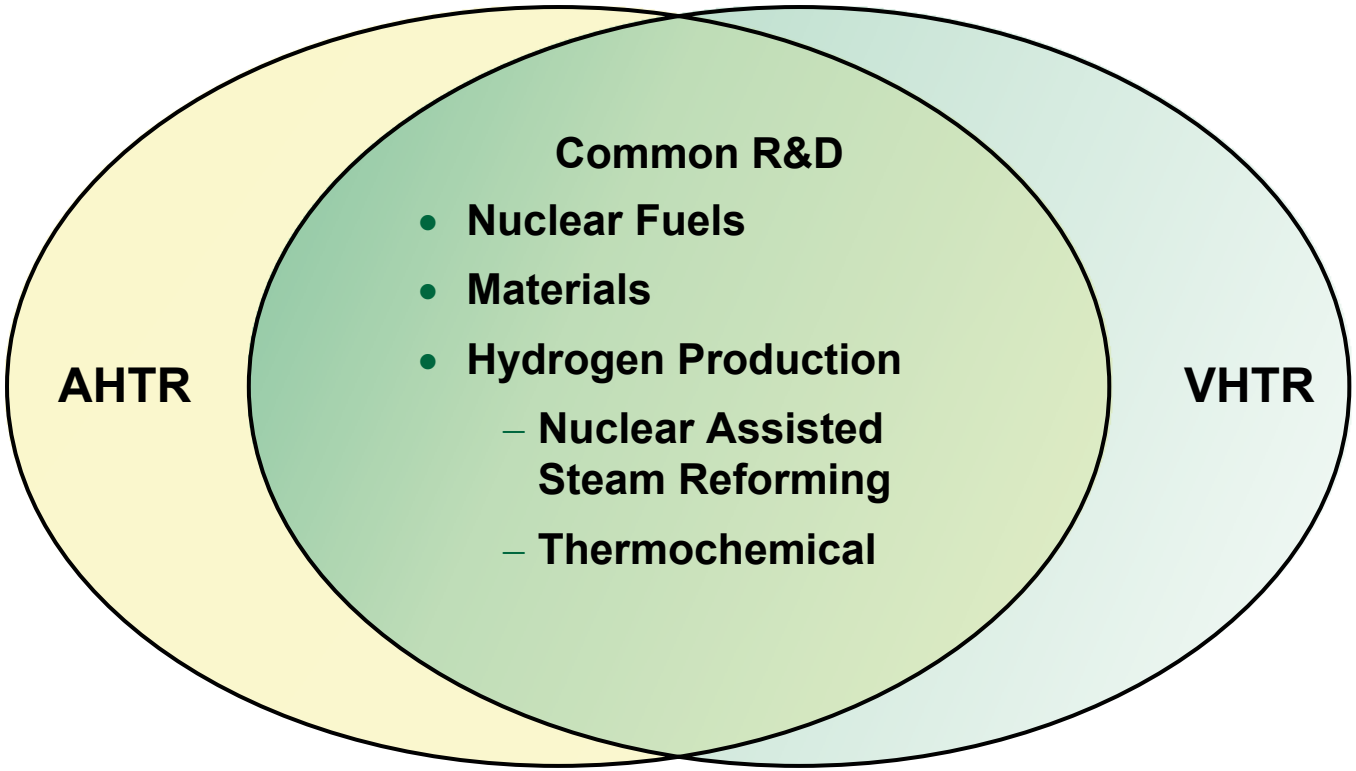
Molten Salt Heat Transfer Loop

- **High Heat Capacity**
- **Low Pumping Costs**
- **Low Pressure**
- **Industrial Experience**

The Nuclear Reactor and the Hydrogen Plant Will Adopt Different Safety Philosophies

- Nuclear plant safety philosophy
  - Contain radioactivity
  - All key components in massive buildings with partly buried main structure
- Hydrogen plant safety philosophy
  - Dilution provides protection against leaks and explosions
  - Open plant without buildings
- Different safety philosophies require physical separation of facilities
  - Heat may be transported over significant distances
  - Method of heat transfer important
- Molten salts offer major advantages for heat transport
  - Technical considerations
    - Low pumping costs
    - Efficient heat transfer
    - High heat capacity (4X of sodium)
    - Low energy release during an accident (atmospheric pressure, no compressed gas)
  - Industrial experience: chemical industry traditionally uses low-pressure molten salts for high-temperature heat transport
  - German nuclear-hydrogen studies concluded that molten salts were the preferred heat-transfer fluid
- AHTR interfaces efficiently with molten-salt heat-transport systems

The R&D Requirements For Different Systems Have Much In Common



Research and Development

- Early in the R&D cycle
- Thermochemical cycles
  - Major R&D remains
  - Potential to improve Iodine Sulfur cycle
    - Significantly lower peak temperatures
    - Better separations
    - Similar to status of steam reforming in the 1950s
    - Possibility of new cycles
- Reactors
  - 80% commonality between VHTR and AHTR
- Institutional
  - Business model
  - Regulation

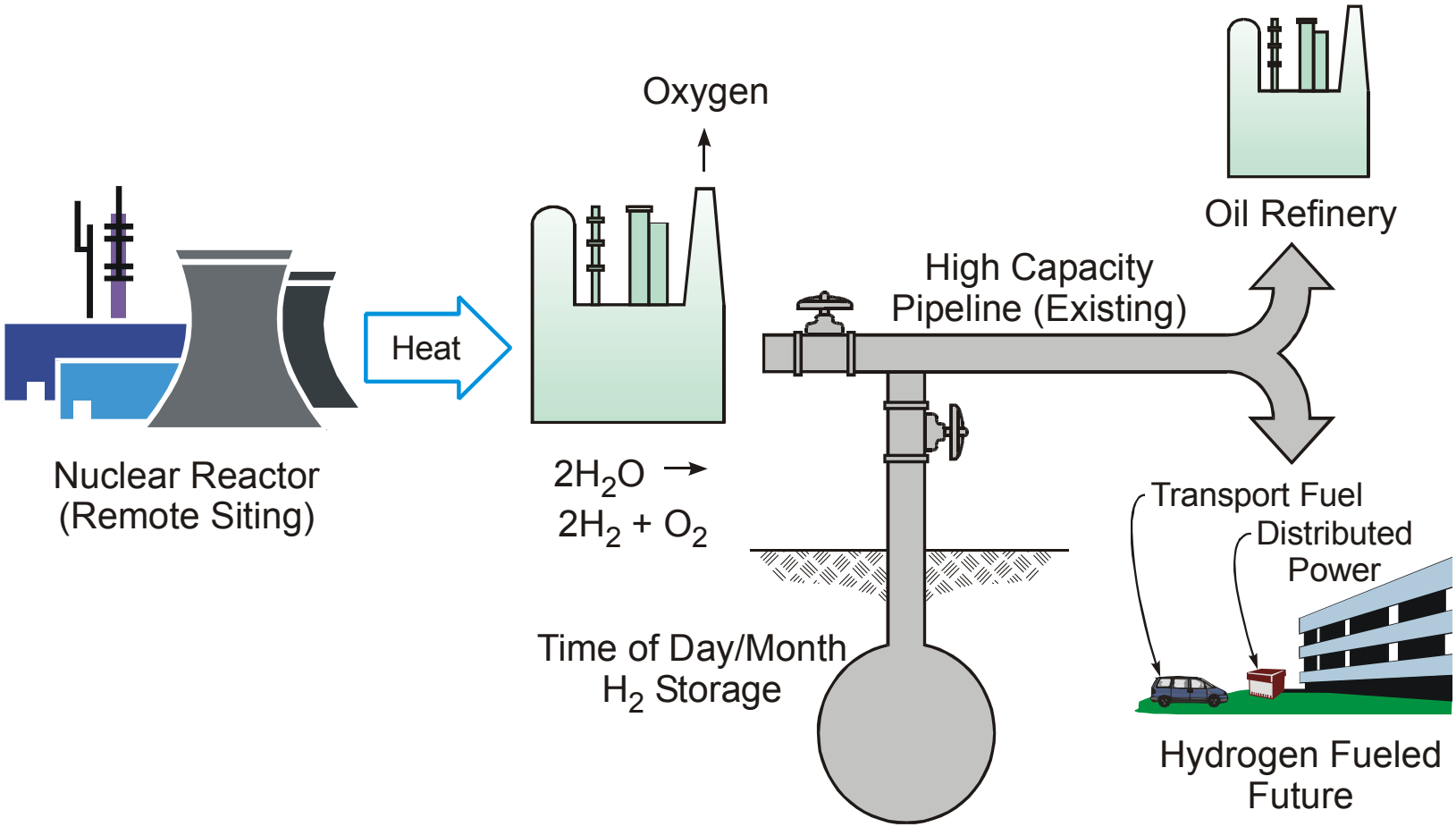
Conclusions

- Economic production of hydrogen is difficult
- Growth in hydrogen demand and plant size makes nuclear hydrogen a realistic possibility
- Several options exist for hydrogen production techniques (heat plus water yields hydrogen and oxygen)
  - Steam reforming of natural gas (closest option)
  - Thermochemical cycles (midterm option)
  - Hot electrolysis
- Two reactor options could be deployed before 2020
  - AHTR
  - VHTR

Backup Information

The Intrinsic Characteristics of Nuclear Power Are Compatible with Hydrogen Production

(Remote Siting, Scale of Operations, and Full-Load Operations)

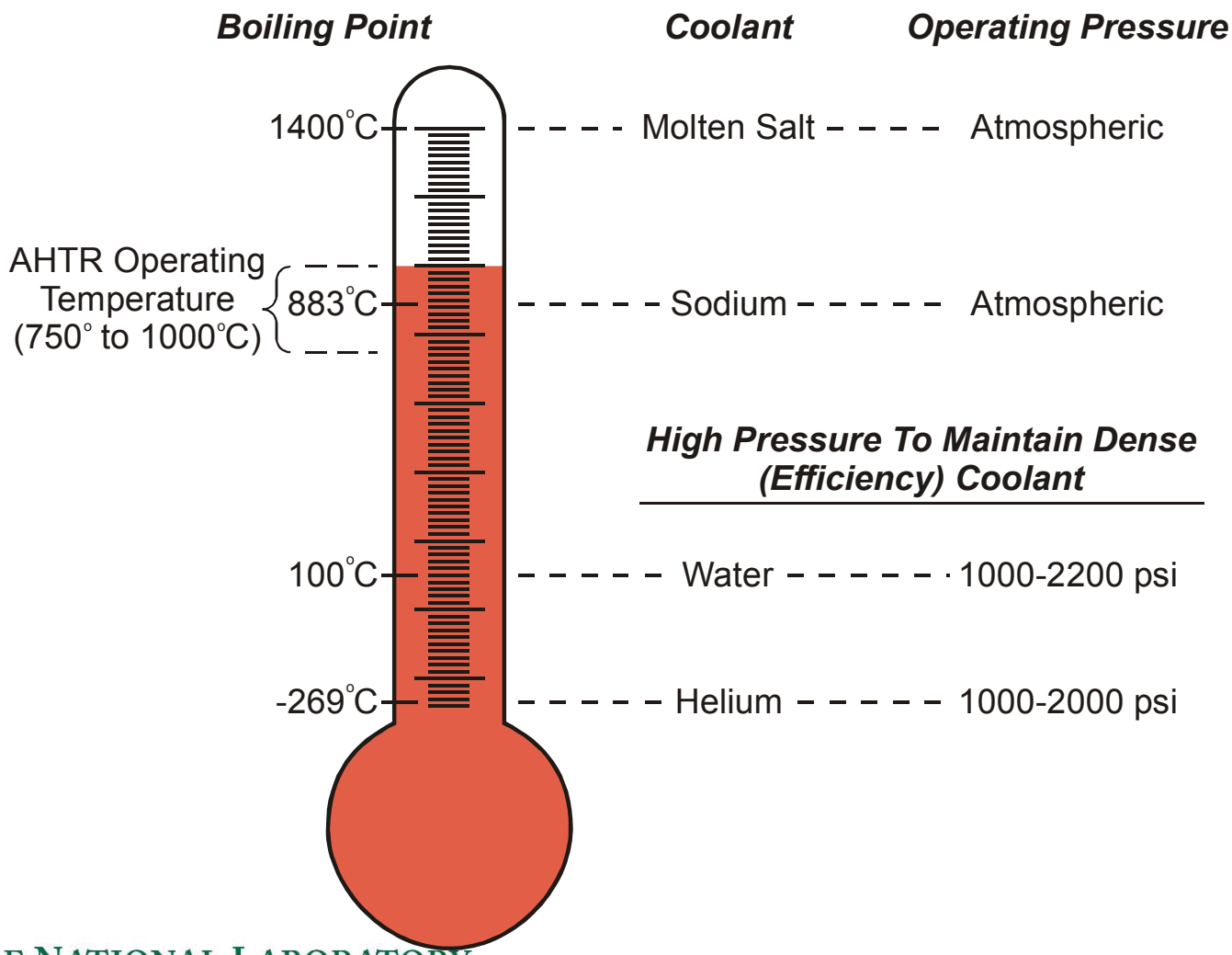


Scale of Hydrogen Production Matches Nuclear Energy

- The economics of nuclear reactors implies energy outputs of 600 to 4500 MW(t)
- Newest world-class H_2 plants (natural gas fuel) that have been announced will produce 300 million ft^3/day
- Equivalent to 2400-MW(t) reactor (Assuming 50% thermal-to-H_2 conversion efficiency)
- No size mismatch

Molten Salts Are a Low-Pressure Reactor Coolant

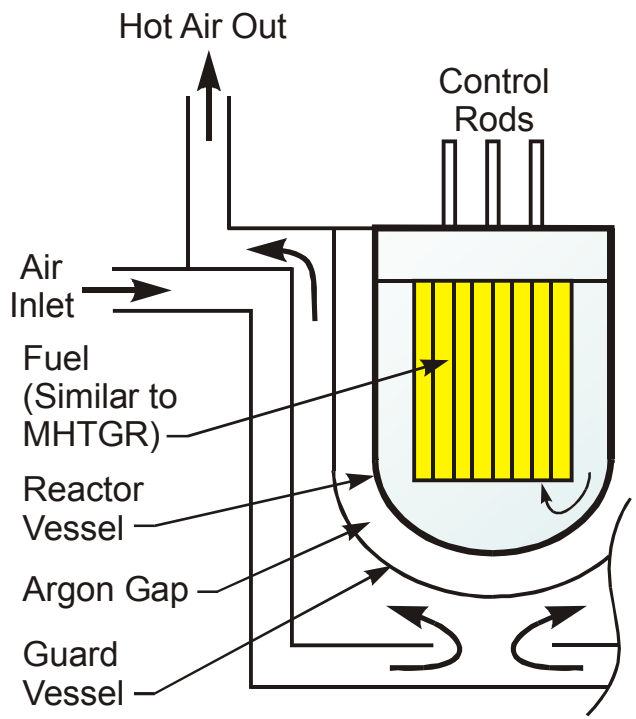
(Avoid Pressurization Risk at Chemical Plant; Minimum Stress on Materials)



The High-Temperature, Low-Pressure Liquid Coolant Enables Passive Decay Heat Removal in Large Reactors

(Example: AHTR Using Modular Liquid-Metal Reactor Decay-Heat System)

Passive Decay Heat Removal **Reactor**

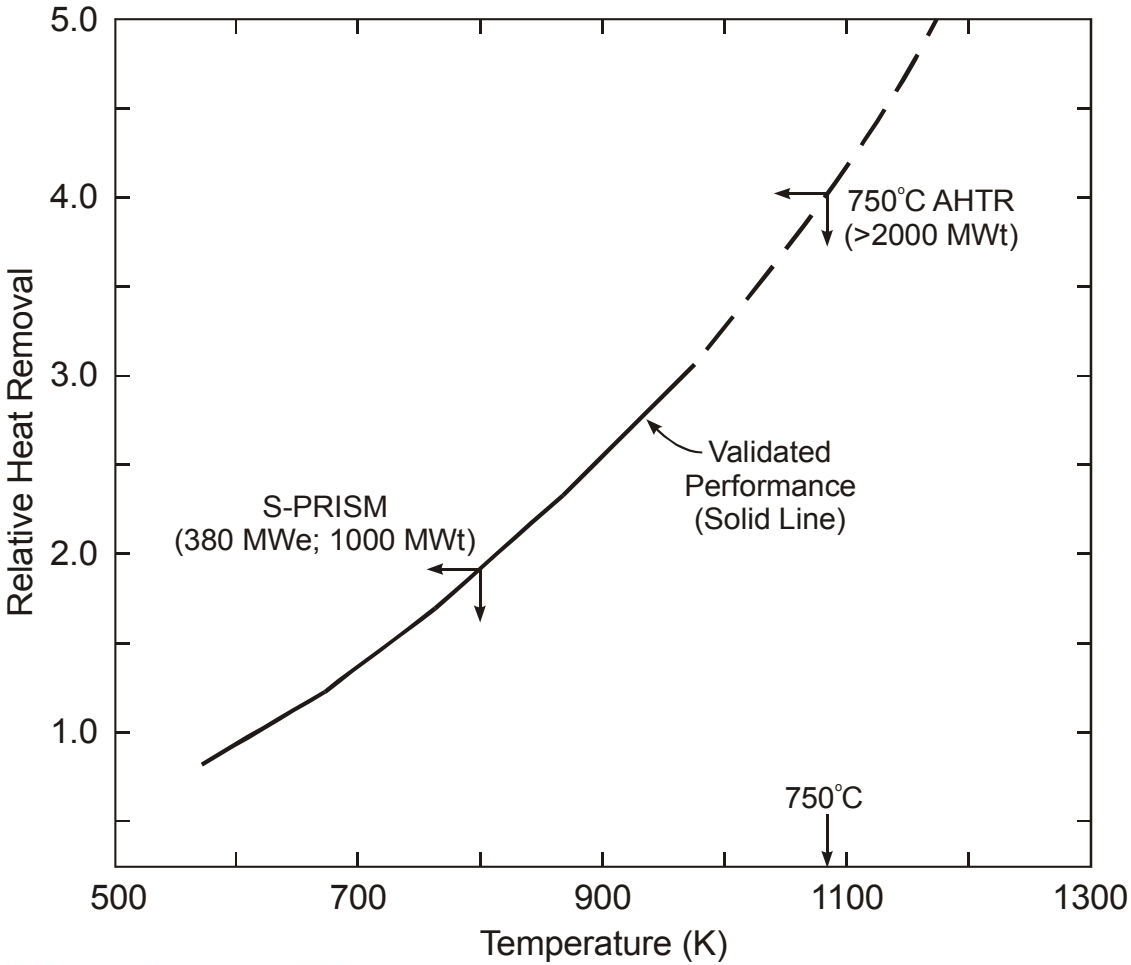


Decay-Heat Cooling System Characteristics

- Similar to GE S-PRISM (LMR)
- Argon Gap
  - Heat Transfer ~T^4
  - Thermal Switch Mechanism
- Heat Rejection Temperature Dependent
  - LMR: 500-550°C (~1000 Mw(th))
  - AHTR: 750-1000°C (>2000 Mw(th))

Higher Temperatures of the AHTR Allow Much Larger Reactors with the Same Size Systems

(Economic Implication: S-PRISM and AHTR With Identical 9-m Vessels)



Molten Salt Options

- Choice of molten fluoride salt is dependent upon multiple factors
  - Neutronics
  - Economics
  - Melting points (mixtures lower melting points)
- Different fluoride salts have somewhat similar properties
- Leading candidates
  - $NaF\text{-}ZrF_4$ (50%/50%): used in Aircraft Reactor Experiment—experimental test reactor
  - $NaF\text{-}RbF\text{-}ZrF_4$ (8%/50%/42%)
  - $^{7}Li_2BeF_4$