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TOPIC 2

THE REACTORS AND THEIR FUELS

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Reactors with Molten Salts: Options and Missions

Charles W. Forsberg
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831-6165
Tel: (865) 574-6783
Fax: (865) 574-0382
E-mail: **forsbergcw@ornl.gov**

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C. W. Forsberg
Oak Ridge National Laboratory
P.O. Box 2008, Oak Ridge, TN 37831-6165
Tel: (865) 574-6783
Email: **forsbergcw@ornl.gov**

Many molten fluoride salts have excellent heat transfer properties, have very low vapor pressures with boiling points in excess of 1400ºC, are highly stable in radiation fields, and can dissolve high concentrations of actinides and fission products. As a consequence, such salts have potentially unique roles as coolants in four high-temperature nuclear applications. These applications will be examined, with the primary emphasis on the Advanced High-Temperature Reactor (AHTR) and the Molten Salt Reactor (MSR).

The AHTR is a large passively safe reactor that uses solid graphite-matrix coated-particle fuel (similar to that used in gas-cooled reactors) and a clean molten salt coolant. The molten salt coolant transfers its heat to a secondary molten-salt heat-transport system that, in turn, transfers the heat to a Brayton power cycle or hydrogen production plant. The AHTR is a new concept with peak reactor coolant temperatures between 700 and 1000ºC, depending upon the application.

The MSR is a fluid reactor in which uranium and other actinides are dissolved in a molten fluoride fuel salt. The salt flows through a reactor core of graphite, where the graphite acts as a neutron moderator and fission occurs within the molten salt. The molten fuel salt transfers its heat to a secondary molten-salt heat-transport system that, in turn, transfers the heat to the Brayton power cycle. Because the MSR has a liquid fuel, the reactor has the unique safety advantage that in the event of an accident, the fuel salt is dumped to critically safe, passively cooled storage tanks. The fuel salt can be processed on-line, a capability that enables the MSR to be a thermal-neutron breeder reactor and have very different fuel cycles. Two experimental MSRs have been built.

Molten-salt heat-transport systems are being considered to move heat from the AHTR or gas-cooled reactors to thermochemical hydrogen production plants. Last, the fusion energy community is considering molten salts to cool fusion reactors and breed tritium fuel.

1. INTRODUCTION: THE ROLE OF MOLTEN SALTS IN NUCLEAR SYSTEMS

1.1 Rebirth of High-Temperature Reactors

There are strong incentives to develop high-temperature nuclear energy systems with coolant temperatures between 700 and 1000°C. Power plant efficiency increases with temperature. Consequently, the size and cost per kW(e) of energy conversion systems (heat to electricity), heat rejection systems (cooling towers, etc.), and nuclear safety systems (decay-heat removal) become smaller as the reactor temperature increases. The higher thermal efficiency implies lower nuclear fuel costs and less generation of waste per unit of energy.

While the above benefits of high-temperature reactors have been understood for decades, it is the new goals and enabling technologies that are creating a renewed interest in such reactors and the potential for practical economic high-temperature reactors.

- *Hydrogen production.* There is a growing interest in hydrogen production using thermochemical hydrogen cycles that require high-temperature heat to convert water to hydrogen and oxygen. Preliminary cost estimates indicate that hydrogen production costs using these cycles to be ~60% of the cost of hydrogen from electrolysis. Depending upon the system, heat must be delivered between 700 and 850°C.

- *Brayton power cycles.* Brayton power cycles using nitrogen or helium allow efficient conversion of high-temperature heat to electricity. It is only in the last decade that efficient Brayton power cycles, derived from aircraft engines, have been developed for utility applications. For over 80 years, the traditional utility power conversion cycle has been the Rankine steam cycle. Steam cycles have served the utility industry well; however, the upper limit of practical steam cycles is between 500 and 600°C. Earlier high-temperature reactors were coupled to steam cycles and thus were unable to efficiently utilize the high-temperature heat that was available from the reactor core for electricity generation.

- *High-temperature materials.* In the 1970s, graphite-based coated-particle high-temperature nuclear fuels were developed. Since then, a wide variety of carbon-carbon composite and other engineered materials have been developed. Practical materials for reactor cores, control rods, and other high-temperature components are becoming available.

- *Passive safety systems.* Passive safety systems have been developed for high-temperature reactors. Passive safety systems may improve economics (avoid the need for complex and expensive active safety systems with diesel generators, pumps, compressors, etc.); safety; and public acceptance.

- *Dry cooling.* In most parts of the world, nuclear reactor siting is limited by the availability of cooling water. Dry cooling towers can avoid this constraint but are expensive when coupled to light-water reactor (LWR) steam cycles. High-temperature reactors with high efficiency and Brayton cycles dramatically lower the cost of dry cooling and increase the number of options for nuclear-plant siting.

1.2 Liquid-Cooled High-Temperature Reactors

Nuclear reactor types can be classified by power output and the peak temperatures of their coolants (Fig. 1). LWRs are low-temperature, high-pressure reactors. Traditional fast reactors cooled with liquid sodium operate at medium temperatures and low pressures. Two options exist for high-temperature reactor coolants: (1) high-pressure gases and (2) low-pressure liquids with boiling points above the peak coolant temperatures. Helium is the traditional high-temperature, high-pressure gas coolant. Molten fluoride salts are the traditional high-temperature low-pressure liquid coolant. The only other potential candidates are liquid metals, particularly molten lead or lead alloys for fast-spectrum reactors. Because of their relatively low boiling points, traditional liquid metals such as sodium are not candidates for high-temperature operations. The characteristics of these different reactor coolants are summarized in Table 1.
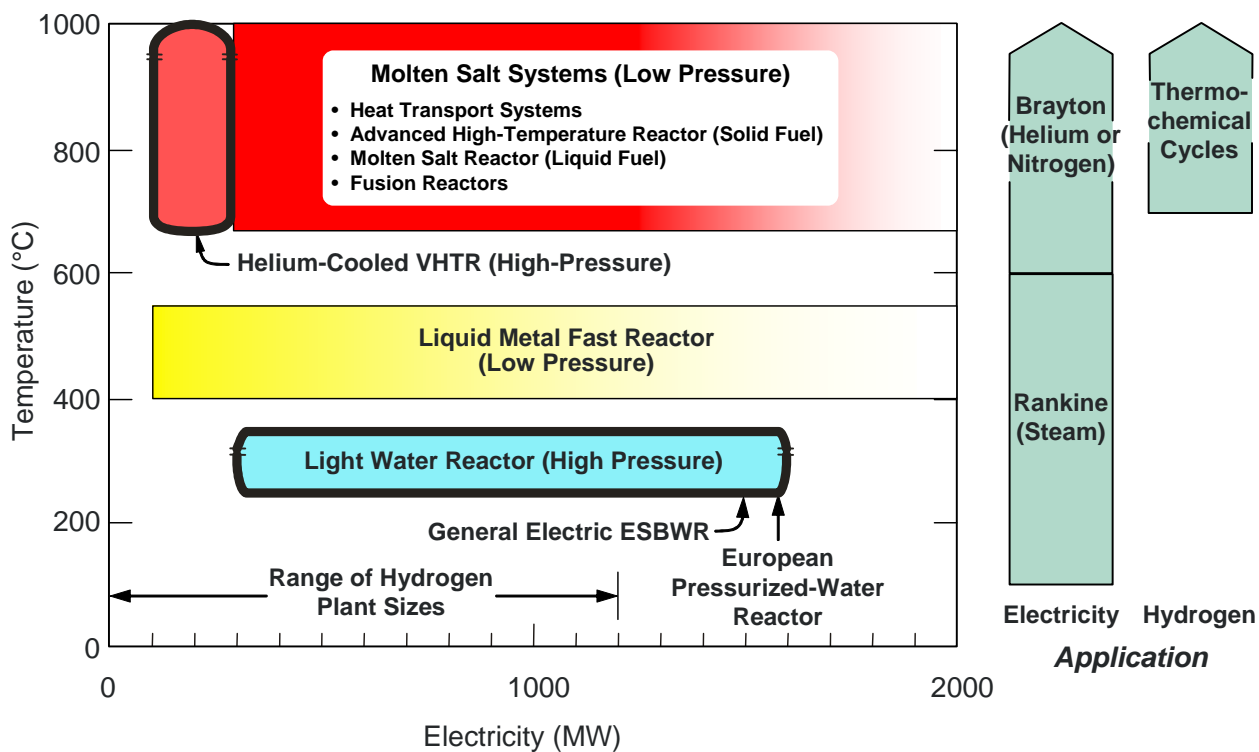
Fig. 1. Reactor type vs temperature and power output.

Table 1. Characteristics of reactor coolants[a]

Coolant	T_{melt} (°C)	T_{boil} (°C)	ρ (kg/m^3)	C_p (kJ/kg °C)	ρC_p (kJ/m^3 °C)	K (W/m °C)	$v \cdot 10^6$ (m^2/s)
Li$_2$BeF$_4$ (Flibe)	459	1,430	1,940	2.34	4,540	1.0	2.9
0.58NaF-0.42ZrF$_4$	500	1,290	3,140	1.17	3,670	~1	0.53
Sodium	97.8	883	790	1.27	1,000	62	0.25
Lead	328	1,750	10,540	0.16	1,700	16	0.13
Helium (7.5 MPa)			3.8	5.2	20	0.29	11.0
Water (7.5 MPa)	0	100	732	5.5	4,040	0.56	0.13

[a]ρ is density; C_p is specific heat; k is thermal conductivity; v is viscosity.

Current concepts of high-temperature reactors can be divided into two categories. The high-pressure helium-cooled reactors are modular in design and have relatively small power outputs. The various low-pressure reactors with transparent molten salt coolants have large power outputs. There are fundamental reasons for reactor size to be coupled to the choice of coolant.

- *Materials.* Most materials weaken as the temperature increases. Consequently, development of a large high-temperature, high-pressure machine is more difficult and expensive than design of a high-temperature, low-pressure machine.

- *Coolant volumetric heat capacity.* For any reactor, heat must be transported from the reactor core to the power conversion system. Pipe, valve, pump, and heat-exchanger size depend primarily on physical properties of the fluid. Table 2 shows the number of 1-m pipes required for different fluids to transport 1000-MW(t) of heat, assuming that the fluid temperature is changed by 100ºC. Molten salts have outstanding volumetric heat capacities compared with those of other fluids; thus, the fluid system size is smaller than in other reactors. Large reactors are viable with molten salt cooling. The large size of helium systems limits the ultimate energy output of a reactor with a helium coolant.

- *Passive safety systems.* As will be discussed, molten salts are the enabling technology for large reactors with passive safety systems.

Table 2. Relative capabilities of different coolants to transport 1000 MW(t) of heat with a 100ºC rise in coolant temperature

	Water	Sodium	Helium	Molten salt
Pressure, MPa	15.5	0.69	7.07	0.69
Outlet temp, ºC	320	545	1000	1000
Velocity, m/s (ft/s)	6 (20)	6 (20)	75 (250)	6 (20)
Number of 1-m-diam. pipes required to transport heat	0.6	2.0	12.3	0.5

1.3 Molten Salt Applications

Molten fluoride salts are being considered in four nuclear applications. These applications are listed in the likely order of deployment.

- *Molten-salt heat-transport systems.* Molten fluoride salts are being proposed to transport heat from all types of high-temperature reactors to hydrogen production systems.

- *AHTR.* The AHTR is a solid fuel reactor that uses a clean molten salt coolant to transfer heat from the solid reactor core to an intermediate heat exchanger. The intermediate heat-transport loop transfers the heat to a Brayton power cycle or a hydrogen production facility.

- *MSR.* The MSR is a liquid-fuel reactor in which uranium, fission products, and actinides are dissolved in the molten salt. The fuel salt flows through a graphite reactor core, which acts as a moderator. The heat is carried by the fuel salt to an intermediate heat exchanger. The intermediate heat-transport loop transfers the heat to a Brayton power cycle or a hydrogen production facility.

- *Fusion reactors.* Molten salts are major candidates for cooling inertial and magnetic fusion energy systems.

1.4 History

A significant experience base exists for only three high-temperature liquids: molten iron, molten glass, and molten fluoride salts. Since the 1890s, essentially all aluminum has been produced by the Hall electrolytic process. In the Hall process, aluminum oxide is dissolved in a mixture of sodium and aluminum molten fluoride salts (cryolite: $3NaF-AlF_3$) at ~1000ºC in a graphite-lined bath. Massive graphite electrodes provide the electricity that converts aluminum oxides to aluminum metal.

In the 1950s, at the beginning of the cold war, the United States launched a large program to develop a nuclear aircraft (Fraas and Savolainen 1956) for delivery of nuclear weapons. Nuclear submarines were being developed at that time and the U.S. Air Force wanted an equivalent aircraft with unlimited range. MSRs were initially developed to provide a heat source with the heat transferred via an intermediate loop to a jet engine. MSRs were chosen for this application to minimize aircraft weight. The high-temperature low-pressure fluid-fuel reactor avoided the need for heavy pressure vessels and the high temperatures maximized jet engine efficiency. Last, because there is no potential to burnout the fuel, liquid-fueled reactors can have much higher power densities than solid-fuel reactors, a factor that reduces reactor size. The nuclear aircraft program was ultimately cancelled because of the peacetime risks of aircraft crashes and the high shielding weight required to protect the crew.

In the 1960s and 1970s, the MSR was investigated as a thermal-neutron breeder reactor (*Nucl. Appl. Technol.* 1970). The liquid fuel offered a unique advantage: on-line processing of the fuel salt, which enabled the design of a thermal-neutron breeder reactor using a thorium-^{233}U fuel cycle. The program was ultimately cancelled when the United States decided to concentrate on development of a single breeder reactor concept.

These billion-dollar programs developed the technology base for use of molten salts in nuclear systems. Two experimental reactors were built and successfully operated. The Aircraft Reactor Experiment (ARE) was the first MSR. It was a 2.5-MW(t) reactor that was operated in 1954 at a peak temperature of 860ºC and used a sodium-zirconium fluoride salt. This was followed in 1965 by the Molten Salt Breeder Reactor (MSBR) Experiment, an 8-MW(t) reactor that used a lithium-beryllium fluoride salt and demonstrated most of the key technologies for a power reactor. In addition, test loops with molten salts were operated for hundreds of thousands of hours, materials of construction were code qualified to 750ºC, and a detailed conceptual design of a 1000-MW(e) MSBR was developed. Over a 1000 technical reports were produced.

Since the 1970s, smaller research programs examining the MSR, the use of molten salts for heat transport loops, and molten salts as coolants for fusion energy machines have been conducted in various countries. In the last several years, the AHTR has been developed as a new reactor concept.

1.5 Subjects for Study

Three subjects are to be addressed: descriptions of proposed molten salt applications, molten salt properties, and engineering characteristics of molten salt systems.

2. APPLICATIONS

2.1 Heat Transfer

Molten-salt-heat transport loops are being considered to transport heat from gas-cooled and molten-salt cooled reactors to thermochemical hydrogen production facilities. This requires the transport of hundreds to thousands of megawatts of heat over distances of hundreds of meters. Molten salts have been traditionally used in the chemical industry to transport heat to ~600ºC. However, hydrogen production may require heat transport at significantly higher temperatures. For several reasons, this type of heat transport system will likely require the use of high-temperature molten salts.

*   *Size*. The size of a molten-salt heat-transport loop is only a small fraction of those using helium and is small relative to those for other coolants (Table 2). The pipe diameter of a molten-salt system will typically be one-fifth of a high-pressure helium system.

*   *Design freedom*. While the vapor pressure of proposed molten salts is below 1-atmosphere, a molten-salt heat-transport system can be operated at any pressure by using a pressurizer with a small amount of inert gas. For example, the heat transport loop can be operated at a pressure level between that of the chemical plant and that of the reactor to minimize stresses on heat exchangers and minimize long-term creep.

*   *Safety*. Based on experiences such as the catastrophic chemical accident that occurred in Bhopal, India, in 1984, the primary hazards in chemical plants are associated with the release of toxic chemicals that drift off-site to populated areas. Thermochemical processes contain large inventories of hazardous chemicals. Molten salts are effectively incompressible; consequently, a failure of a heat exchanger in the chemical plant will not pressurize the plant. In contrast, if helium or other gases are used in the heat transport systems, a heat-exchanger failure can pressurize the chemical plant, with the potential for large releases of toxic materials. This consideration was the primary factor in the decision by the German chemical industry to recommend against helium heat-transport loops between reactors and chemical plants.

*   *Performance*. 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 of a helium system. A recent analysis (Peterson et al. 2003a) compared the use of helium with that of molten salts (Fig. 2) 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 80ºC LMDT (point A), the helium-to-molten salt heat exchanger LMDT (point B) will be under 40ºC. 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.
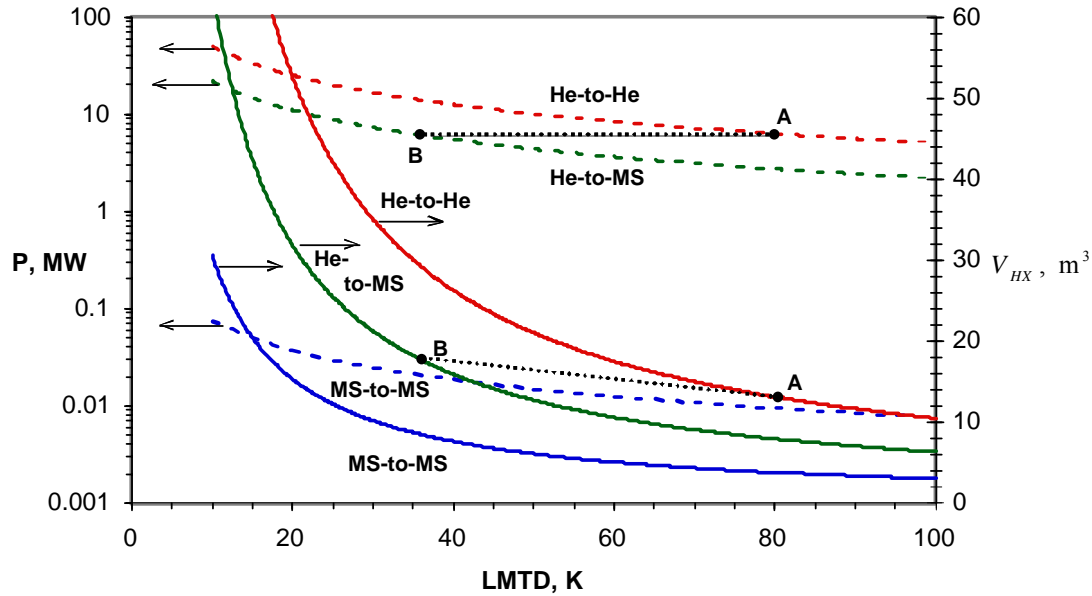
Fig. 2. Total pumping power P (dashed lines) and total volume V_{HX} (solid lines) for a 600 MW(t) compact heat exchanger as a function of LMTD for three combinations of helium (7 MPa) and/or molten salt.

A similar reduction in the temperature drop would occur in the second heat exchanger inside the thermochemical hydrogen 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.

2.2 The AHTR

2.2.1 General Design

The AHTR is a new reactor concept that has three technical characteristics: high temperature, passive safety, and a large power output. The high temperature is required to produce hydrogen and efficiently produce electricity. The passive safety features are required to reduce operating costs and improve public acceptance. The large power output, passive safety features, and high efficiency of electricity production (a consequence of high temperatures) are the enabling technologies to improve economics. The development is a joint effort of Oak Ridge National Laboratory, Sandia National Laboratories, and the University of California at Berkeley.

The AHTR (Fig. 3, Table 3) uses coated-particle graphite-matrix fuels and a molten-fluoride-salt coolant (Forsberg et al. 2003; Ingersoll et al 2004; Forsberg et al. 2004a). The fuel is the same type that is used in modular high-temperature gas-cooled reactors (MHTGRs), with fuel failure temperatures in excess of 1600EC. The optically transparent molten salt coolant is a mixture of fluoride salts with freezing points near 400EC and atmospheric boiling points of ~1400EC. Several different salts are being evaluated as the primary coolant, including lithium-beryllium and sodium-zirconium fluoride salts (Table 1). 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 the turbine hall. In the turbine hall, the heat is transferred to a multi-reheat nitrogen or helium Brayton cycle power conversion system. For hydrogen production, the heat is transferred to the thermochemical hydrogen production facility, which converts water and high-temperature heat to hydrogen and oxygen.
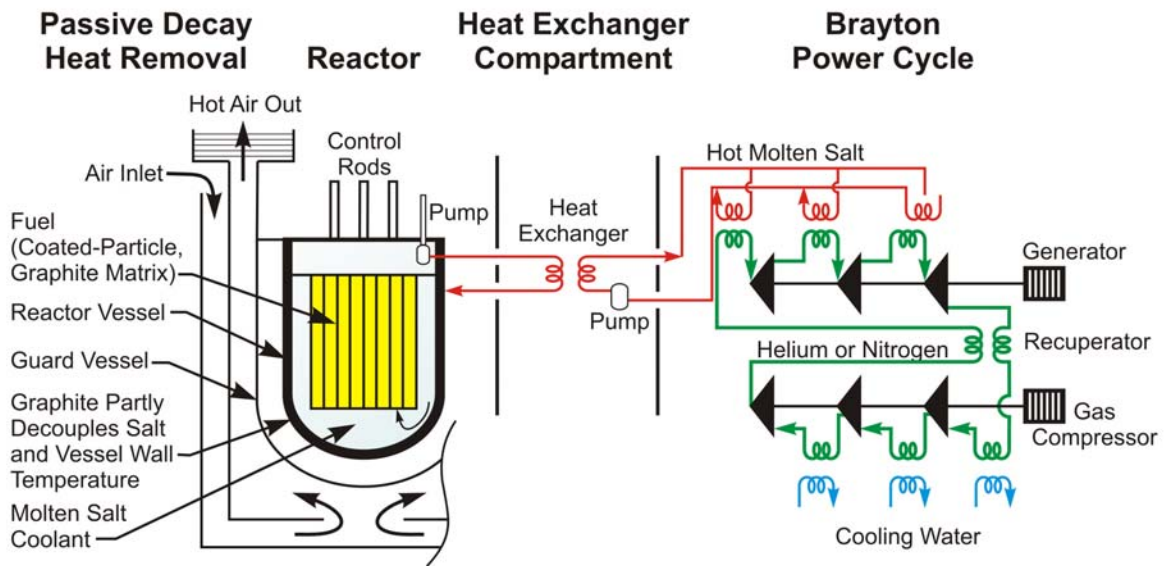

Fig. 3. Schematic of the AHTR for electricity production.

Table 3. AHTR preconceptual design parameters

Power level	2400 MW(t)	Power cycle	3-stage multi-reheat Brayton
Core inlet/outlet temperature (options)	900°C/1000°C 700°C/800°C 670°C/705°C	Electricity (output at different peak coolant temperatures)	1357 MW(e) at 1000°C 1235 MW(e) at 800°C 1151 MW(e) at 705°C
Coolant (several options)	$2^7LiF\text{-}BeF_2$ $(NaF\text{-}ZrF_4)$	Power cycle working fluid	Nitrogen (helium longer-term option)
Fuel		Vessel	
Kernel	Uranium carbide/oxide	Diameter	9.2 m
Enrichment	10.36 wt % ^{235}U	Height	19.5 m
Form	Prismatic	Reactor core	
		Shape	Annular
Block diam.	0.36 m (across flats)	Diameter	7.8 m
Block height	0.79 m	Height	7.9 m
Columns	324	Fuel annulus	2.3 m
Decay heat system	Air cooled	Power density	8.3 W/cm^3
Volumetric flow rate	5.54 m^3/s	Reflector (outer)	138 fuel columns
Coolant velocity	2.32 m/s	Reflector (inner)	55 fuel columns

The baseline AHTR facility layout (Fig. 4) 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. Earlier engineering studies indicated that this was the largest practical size of a low-pressure reactor vessel. 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^3) were essentially identical to those of the MHTGR. However, this is a conservative assumption. The better heat transfer of the molten salt relative to helium (similar to that seen in heat exchangers─see Fig. 2) implies (1) lower peak reactor temperatures if the power densities are similar or (2) a higher-power-density core that is smaller or has a higher total power output for a given reactor vessel size.

Three peak coolant temperatures are being considered: 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. 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.

In the current 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.

2.2.2 Decay Heat Cooling and Accident Management

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 AHTR (Forsberg et al. 2004a) uses passive reactor vessel auxiliary cooling (RVAC) systems (Fig. 3) 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. The effective thermal inertia, per unit volume of the reactor vessel, is much larger than that for gas-cooled reactors because of the high heat capacity of the molten salt and the low radial temperature gradient across the reactor core.

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 by the edge of the core rapidly results in a nearly isothermal core with about a 50℃ temperature difference between the top and bottom plenums. For a typical simulation of the reactor with a nominal coolant exit temperature of 1000℃, the calculated peak fuel temperature in such an accident is ~1160℃, which will occur at ~30 hours with a peak reactor vessel temperature of ~750℃ at ~45 hours. The average core temperature 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.
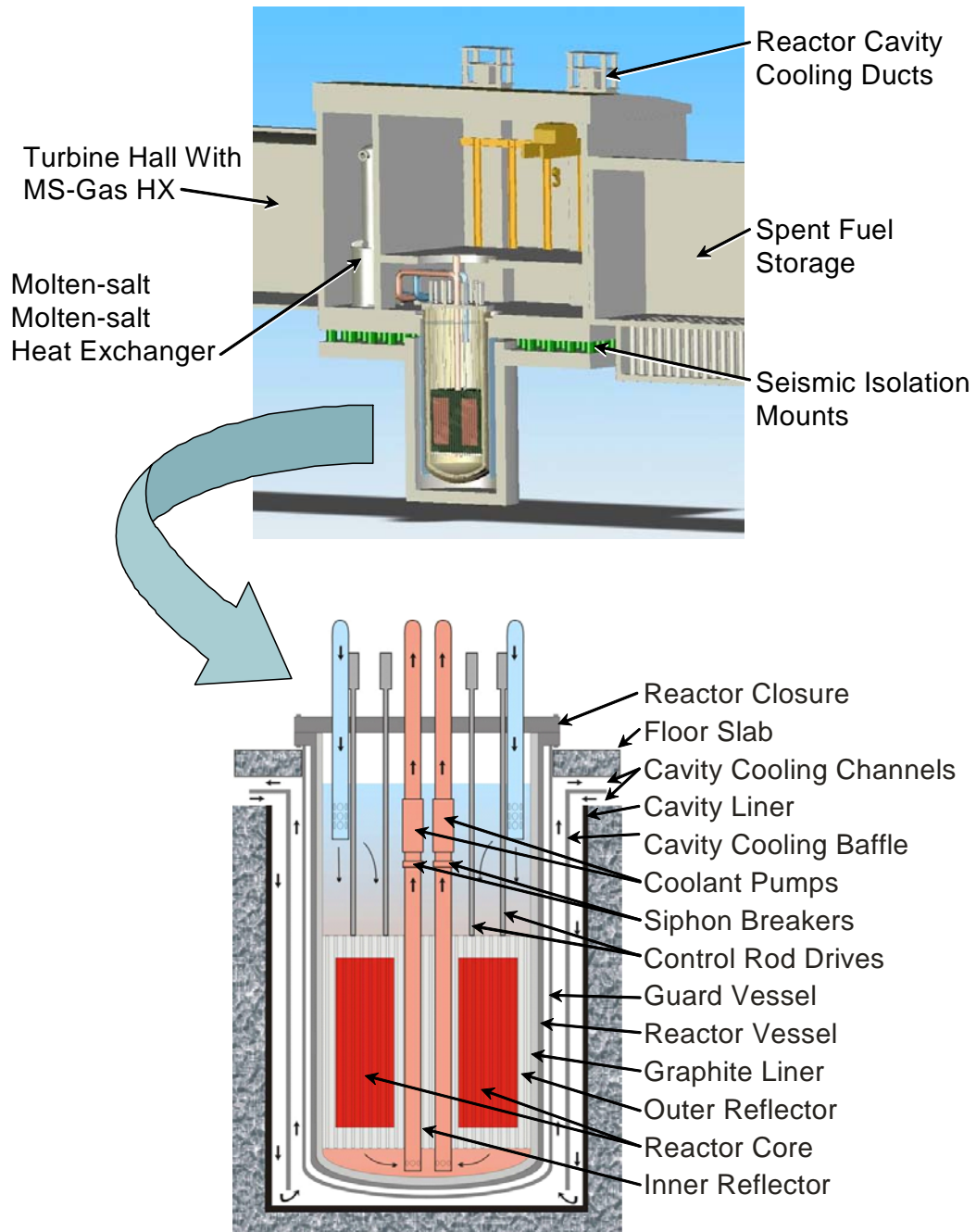
Fig. 4. Schematic of the AHTR nuclear island and vessel.

In terms of various accidents, there are no major stored energy sources within containment such as high-pressure fluids [helium and water] or reactive fluids [sodium]. This reduces requirements for the containment. The molten salt (Sect. 3) dissolves almost all actinides and fission products (except xenon and krypton). This provides a major barrier against the release of any radionuclide from fuel failure.

2.2.3 Reactor Physics and Fuel Cycle

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 ($2^7LiF\text{-}BeF_2$) that was used in the MSRE. 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 other reactors. Conceptual core design studies with more heterogeneous core designs are under way to determine if 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.

2.2.4 Economics

As shown in Table 4, preliminary overnight capital costs of the AHTR for several exit temperatures were determined relative to other higher-temperature reactor concepts [i.e., the S-PRISM and the gas turbine−modular helium reactor (GT-MHR)] based on the relative size of systems and quantities of materials. *This approach provides relative, but not absolute, costs.* Only the construction of multiple reactors provides reliable absolute costs. The lower capital costs are a consequence of several factors: economics of scale [2400-MW(t) reactor vs a 600-MW(t) or 1000-MW(t) reactor, passive safety in a large reactor system, and higher thermal efficiency (see Sect. 4.3). No cost comparisons with large LWRs have yet been completed.

2.2.5 Research and Development (R&D)

About 80% of the R&D required for the AHTR is shared with that for helium-cooled high-temperature reactors. This includes fuel development, materials development, and Brayton power cycles. Many other areas are in common with facility design for liquid-metal reactors. The required R&D is sensitive to the peak coolant temperatures. Near 700°C, existing materials and fuels may be used. At 1000°C, major material development programs are required. Other areas that require significant R&D include (1) the reactor vessel insulation system, (2) optimization of core design, and (3) refueling and maintenance operations in the reactor vessel at 400 to 500°C. As a new concept, the AHTR is early in its development. The largest ongoing effort is a joint program in the United States at Oak Ridge National Laboratory, Sandia National Laboratories, and the University of California at Berkeley. Several other efforts are beginning.

Table 4. Comparison of estimated overnight capital cost (2002$) of the AHTR-IT and AHTR-HT, as a percentage of the costs of the S-PRISM and GT-MHR [with multi-module output of 1145 MW(e)][a]

		S-PRISM $1681/kW(e)	GT-MHR $1528 /kW(e)
AHTR-IT	$930/kW(e)	55%	61%
AHTR-HT	$816/kW(e)	49%	53%

[a]The General Electric S-PRISM consists of four reactor modules, each producing 1000 MW(t) and 380 MW(e). The peak sodium temperature is 510ºC. The General Atomics GT-MHR consists of four reactor modules, each producing 600 MW(t) and 285 MW(e). The peak helium temperature is 850ºC.

2.3 MSRs

2.3.1 General Design

In an MSR (Fig. 5) the molten fluoride salt with dissolved fissile, fertile, and fission isotopes flows through a reactor core (Fig. 6) moderated by unclad graphite. In the core, fission occurs within the flowing fuel salt, which then flows into a primary heat exchanger, where the heat is transferred to a secondary molten salt coolant. The fuel salt then flows back to the reactor core. The graphite-to-fuel ratio is adjusted to provide the optimal neutron balance─an epithermal neutron spectrum. In the preconceptual designs developed in the early 1970s, the liquid fuel salt typically enters the reactor vessel at 565ºC and exits at 705ºC and ~1 atmosphere (coolant boiling point: ~1400ºC). The reactor and primary system are constructed of modified Hastelloy-N or a similar alloy for corrosion resistance to the molten salt. Volatile fission products (e.g., krypton and xenon) are continuously removed from the fuel salt.

The secondary coolant loop transfers the heat to the power cycle or hydrogen production facility. The secondary coolant (1) provides isolation between the low-pressure reactor and either the power cycle (if electricity is being produced) or a hydrogen production facility and (2) chemically reacts and traps tritium that escapes from the primary system. With a fluid-fuel reactor, the tritium is not trapped in the solid fuel and tends to migrate across hot heat exchangers. A small cleanup system removes the tritium from the secondary coolant.

The parameters developed for the 1000-MW(e) MSBR conceptual design (Robertson et al. 1971) developed in the late 1960s are shown in Table 5. These parameters are for a large (2250 MW(t)) [233]U-thorium, liquid-fuel breeder reactor designed for the production of electricity using a steam cycle.

The reactor characteristics minimize the potential for accident initiation. Unlike solid-fuel reactors, MSRs operate at steady-state conditions with no change in the nuclear reactivity of the fuel as a function of time. Fuel is added as needed; consequently, the reactor has low excess nuclear reactivity. No excess fuel is needed at reactor startup to compensate for fuel depletion, and no excess reactivity is required to override xenon poisoning. No significant buildup of xenon occurs over time. There is a strong negative temperature coefficient because increased temperatures lower the fuel-salt density and push fuel out of the reactor core. In normal operations, the control rods are fully removed from the reactor.
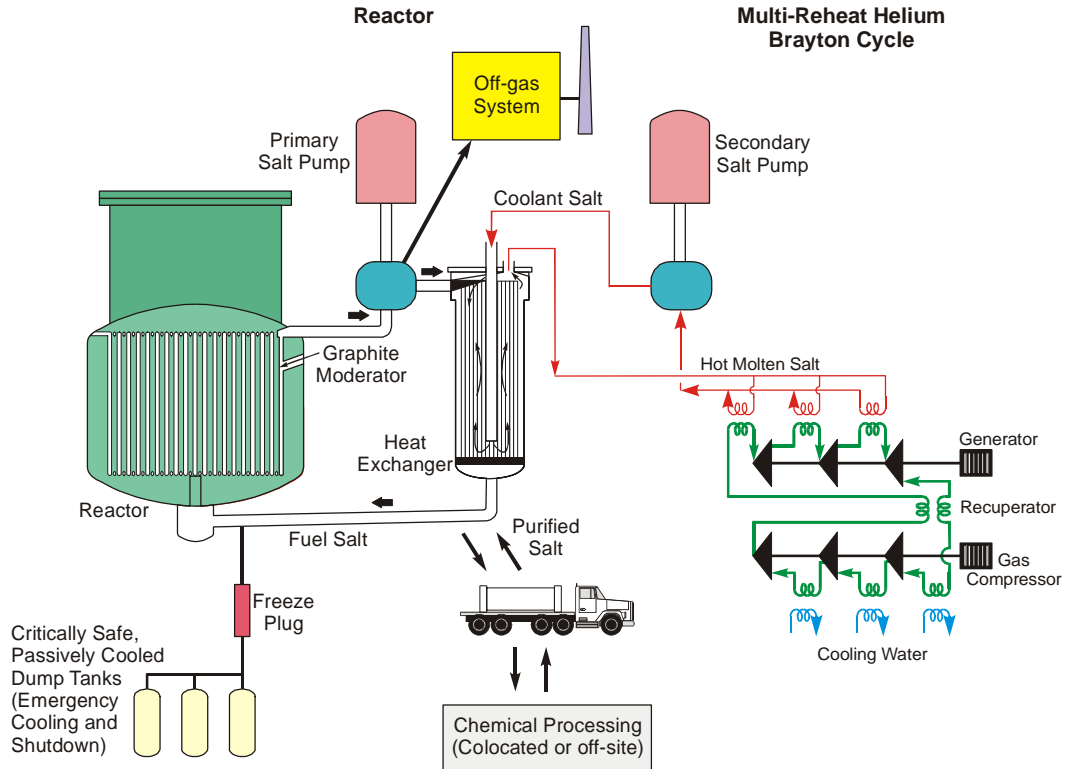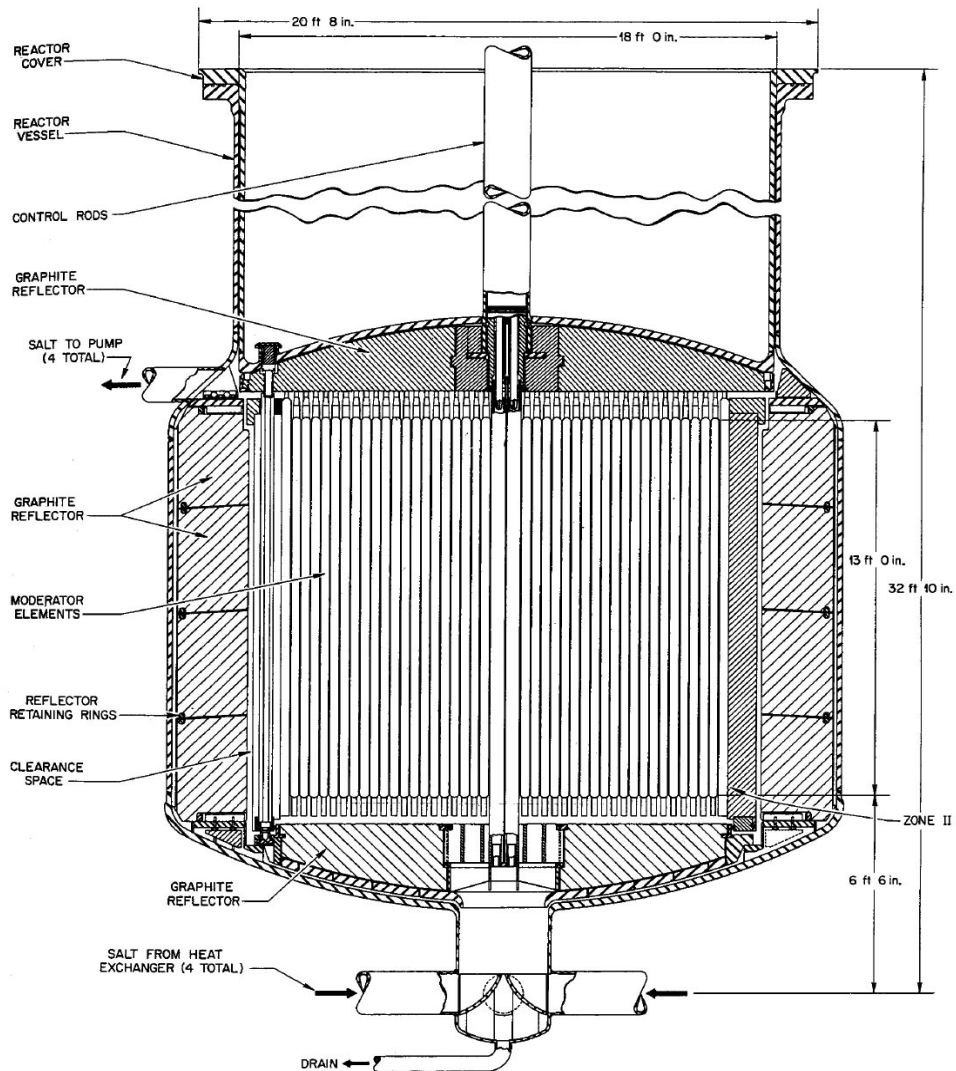
Fig. 5. MSR with multi-reheat helium Brayton cycle.

Fig. 6. Cross section of the 1970s 2250-MW(t) MSBR vessel (*Source*: Robertson et al. 1971).

Table 5. Design characteristics of the 1970s MSBR

Net electric generation	1000 MW	Maximum core flow velocity	2.6 m/s
Thermal efficiency	44.4% (steam cycle)	Total fuel salt	48.7 m^3
Core height	3.96 m	^{233}U	1,500 kg
Vessel design pressure	$5.2 \cdot 10^5$ N/m^2 (75 psi)	Thorium	68,100 kg
Average power density	22.2 kW/L	Salt components	^{7}LiF-BeF$_2$-ThF$_4$-UF$_4$
Graphite mass	304,000 kg	Salt composition (see entry above)	71.7-16-12-0.3 mol %

Early designs of the MSR proposed the use of a steam cycle for electricity production. Current proposals for an MSR use a multi-reheat helium or nitrogen Brayton cycle. The Brayton cycle has major advantages over the use of a steam Rankine cycle: simplified balance of plant with lower cost, improved efficiency, reduced potential for salt freezing in the heat exchangers, and simplified control of tritium within the reactor. The AHTR-LT, that was discussed earlier, has the same molten salt outlet temperature as the 1970s MSBR design with its steam cycle. However, the estimated helium Brayton power-cycle efficiency is 48% compared to 44% for the MSR with steam cycle. This improved efficiency (Sect. 4.4) is a consequence of adopting a Brayton power cycle that is better match to molten salt systems than steam power cycles. The helium or nitrogen Brayton cycle also minimizes difficulties in the control of tritium. In a liquid-fuel reactor, fission-product tritium is not trapped in solid fuel. It can migrate through hot heat exchangers to the power cycle. In a Brayton cycle, it is easy to remove any tritium that enters the power cycle from the dry gas. This is in contrast to a steam cycle where any tritium diffusing through hot heat exchangers with their very large surface area combines with the steam.

2.3.2 Decay Heat Cooling and Accident Management

Molten salt reactors use passive emergency core cooling systems that are radically different from those used in solid-fuel reactors. If the molten reactor fuel salt overheats, its thermal expansion causes it to overflow by gravity into an overflow weir. The fuel is then dumped to multiple critically safe storage tanks with passive decay-heat cooling systems. Freeze valves that open upon overheating of the salt can also be used to initiate core dump of fuel. Drains under the primary system also dump fuel salt to the storage tanks if a primary system leak occurs. This design approach allows very large reactors to be built with passive safety systems.

Many of the driving forces for an accident are reduced compared with those that exist for other reactors. Fission products (with the exception of xenon and krypton) and nuclear materials are highly soluble in the salt and will remain in the salt under both operating and expected accident conditions. The fission products that are not soluble (e.g. xenon and krypton) are continuously removed from the molten fuel salt, solidified, packaged, and stored in passively cooled storage vaults. There are no major stored energy sources within containment such as high-pressure fluids [helium and water] or reactive fluids [sodium]. This reduces requirements for the containment.

2.3.3 Reactor Physics and Fuel Cycle

MSRs are fluid-fuel reactors. Such reactors have several characteristics that are different from those of solid-fuel reactors.

- *Nuclear reactivity*. Negligible xenon effect occurs because xenon continuously escapes from the fuel salt into the off-gas system. There is no change in core reactivity with time, because fuel is continuously added as required. The fuel inventory in the reactor core is coupled to the reactor temperature. An increase in reactor temperature reduces the fuel inventory by expansion of the fuel salt with less mass of fuel salt in the reactor core.

- *Fissile inventory*. As a class, MSRs have very low fissile inventories compared with other reactors for several reasons (1) thermal neutron reactors require less fissile inventory than fast reactors; (2) a low fuel-cycle fissile inventory exists outside the reactor system [no conventional SNF]; (3) little excess reactivity is required to compensate for burnup [because fuel is added on-line]; (4) direct heat deposition in the fuel/coolant allows high power densities; and (5) the high-absorption fission products such as xenon are continuously removed. As a consequence, the MSR requires less than 2 kg of fissile material per MW(e) to reach criticality, compared with 3 to 5 kg/MW(e) for LWRs and over 25 kg/MW(e) for fast-spectrum reactors. This implies that the MSR has the potential to provide long-term, sustainable energy production while limiting the global inventory of plutonium and minor actinides to a total quantity over *an order of magnitude lower* than solid-fuel reactors.

- *Burnup and plutonium isotopics*. Relative to solid-fuel reactors, MSR fuel cycles (Rodriguez-Vieitex et al. 2002; Engel et al. 1978) have very high equivalent fuel burnups and unusual plutonium isotopics with high concentrations of ^{242}Pu.

  – In solid-fuel reactors, SNF burnup is limited by fuel-clad lifetime that, in turn, limits fuel burnup and the burnout of plutonium. In non-breeder reactors, SNF burnup is also economically limited—independent of the technology. Excess fissile material is in fresh fuel when it is initially placed in the reactor core. This is required to compensate for fuel burnup over time. To assure reactor control, burnable neutron absorbers are then added to the fresh fuel to avoid excess nuclear reactivity in new fuel assemblies. There is a significant economic cost (extra enrichment) in "storing" excess fissile fuel in the new fuel assembly until it is needed toward the end of the fuel assembly lifetime. These factors fundamentally limit solid-fuel burnup.

  – In an MSR, fuel is added incrementally to the liquid as required. No excess fuel and associated burnable absorbers are required. Selected fission products are removed from the molten salt and solidified as a waste form. As a consequence, the normal burnup limits that define solid fuels do not apply to a liquid-fuel reactor. The plutonium remains in the salt, with the lighter plutonium fissile isotopes burned out faster than ^{242}Pu. This has major implications in terms of proliferation resistance. The high ^{242}Pu content makes the plutonium from an MSR much less desirable than plutonium from any other reactor type for use in weapons because of its very high critical mass (Choi et al. 2004).

- *Delayed neutron fraction*. In all reactors, some fraction of the fission neutrons are delayed neutrons emitted from the decay of very-short-lived fission products. This fraction is used for reactor control. Unlike solid fuel reactors, the flowing fuel implies that some fraction of the delayed neutrons will occur in flowing fuel that has left the reactor core. This must be accounted for in all reactor physics calculations and evaluations.

Four major fuel cycle options exist to address different goals of reactor operation. The basic reactor remains unchanged except for the salt composition, salt-cleanup systems, and fuel cycle operations. Any of the MSRs can be started up using low-enriched uranium or other fissile materials. With the exception of the breeder reactor fuel cycle, the fuel salt processing for all the other fuel cycles can be performed off-site with removal of the fuel salt every few years.

Actinide burning. This fuel cycle (Rodriguez-Vieitex et al. 2002; Kurchatov 2001) burns multi-recycle Pu, Am, and Cm from LWR SNF or other sources to reduce the long-term hazard of wastes to the repository. It can also produce denatured ^{233}U as a by-product. The penalty for burning actinides in an epithermal neutron flux is partly offset by the greater fission neutron yield of the higher actinides. As an actinide-burner, the production of electricity from the MSRs will be up to 10% of the electricity that is produced by the other reactors that originally generated the actinides. This mode of operation requires a molten salt, such as a sodium-zirconium fluoride salt, that has a high solubility for actinides. In the process of burning actinides, the actinides with high fission cross sections are burnt out first. It requires substantially longer times to burnout low-nuclear-cross-section actinides. Consequently, there is a buildup of low-cross-section actinides in the reactor. This implies that any reactor burning actinides from LWRs will have a larger inventory of actinides in the reactor core than with other MSR fuel cycles.

Much of the current interest in MSRs is a result of the capabilities to burn actinides to reduce waste management burdens. Because they are liquid-fuel reactors, MSRs offer three advantages over solid-fuel reactors in this application:

- *No isotopic blending.* Different lots of SNF have very different Pu, Am, and Cm isotopics. The MSR has a homogeneous liquid fuel. Any fissile material can be fed to the reactor. The very different nuclear characteristics of different batches of higher actinides are addressed by the rate of addition to the homogeneous molten fuel salt. In contrast, in solid-fuel reactors, the quantity and isotopics of the fissile materials in every location of every fuel assembly must be controlled to avoid local overpower conditions that burn out the fuel. With complex mixtures of isotopics, this process is expensive and difficult to accomplish.

- *No fuel fabrication.* The higher actinides have small critical masses and high rates of decay heat, representing a serious technical and economic challenge for fuel fabrication. This is a non-issue for an MSR, because no fuel fabrication is required.

- *Minimal reprocessing.* In an MSR, fission products are removed from the molten salt, while actinides remain in the salt. This is the reverse of traditional processing, in which clean fissile materials are extracted from SNF. In an MSR, the cleaned fuel salt is to be mixed back with the salt in the reactor. Some fraction of the fission products must be removed, but there is no reason to fully clean the salt. Processing would be done as a batch process at a collocated or off-site location.

Once-through fuel cycle. The once-through fuel cycle (Engel et al. 1980) converts thorium to ^{233}U internally in the reactor and uses 20% enriched uranium as fresh fuel to the reactor. The annual fuel consumption is ~45 t/GW(e), or about one-fifth that of an LWR. No recovery of fissile material from the discharged salt (SNF) would be required.

Thorium-^{233}U breeder cycle. MSRs can operate as breeder reactors. After startup, only thorium is added as a fuel. A breeder reactor with efficient fuel production also requires on-line processing of the fuel salt because of the nuclear characteristics of breeding fuel with thermal neutrons using the ^{233}U-Th fuel cycle. In a thermal neutron breeder reactor, the breeding reaction is ^{232}Th + n → ^{233}Pa → ^{233}U. Unfortunately, ^{233}Pa has a moderately large absorption cross section and a half-life of 27 days. If it is left in the reactor, parasitic capture of neutrons by ^{233}Pa will occur, resulting in a significant reduction

in the breeding ratio. To avoid this scenario and to obtain high breeding ratios, on-line processing is required for removal of the ^{233}Pa and storage outside of the reactor until it decays to ^{233}U. With an efficient processing system, the breeding ratio is ~1.06, with an equilibrium ^{233}U inventory of about 1500 kg (Table 5). If the reactor is to be a breeder reactor, the fuel salt characteristics must be optimized and will almost certainly be a mixture of ^{7}LiF, BeF$_2$, ThF$_4$, and UF$_4$. This salt mixture provides better neutron economy. The use of a thorium-^{233}U breeder reactor cycle results in a high-level waste with a very low actinide content because─as neutrons are added to the thorium─the various fissile isotopes that are produced (^{235}U, ^{239}Pu, etc.) tend to fission.

Denatured thorium-^{233}U breeder cycle. This is a breeder reactor fuel cycle (Engel et al. 1978; Engel et al. 1979) designed to maximize proliferation resistance by minimal processing of the fuel salt and by addition of ^{238}U to isotopically dilute fissile uranium isotopes. This lowers the breeding ratio to slightly above 1 and results in a very low fissile plutonium (^{239}Pu and ^{241}Pu) inventory of ~0.16 kg/MW(e). The use of a thorium-^{233}U breeder reactor cycle results in a high-level waste with a low actinide content because─as neutrons are added to the thorium─the various fissile isotopes that are produced (^{235}U, ^{239}Pu, etc.) tend to fission.

2.3.4 R&D Requirements

In terms of R&D, the MSR represents an unusual situation. A major program existed in the 1960s and early 1970s that operated a test reactor, operated various test loops with several hundred thousand hours of operating time, code qualified materials of construction, developed a detailed conceptual design of a 1000-MW(e) reactor, and was on the path to building a medium-size demonstration plant when the program was cancelled. Research then stopped. In the last several years, because of changing goals and new technology, there has been a renewed interest in MSRs. This renewed interest in MSRs was initially driven by changes in goals for reactors.

- *Passive safety*. MSRs intrinsically have passive safety systems, a factor that was not considered important in the 1970s but is viewed as significant today.

- *Actinide-free wastes*. MSRs with thorium fuel cycles intrinsically have wastes with low actinide contents. This issue, which was not important in the 1970s, is considered extremely relevant today and has resulted in a general renewed interest in fluid-fuel reactors (Slessarev et al. 2004).

- *Breeding*. Less emphasis currently exists on breeder reactors with high breeding ratios. This has a major impact on the complexity of any on-line molten salt processing system. MSRs have breeding ratios only slightly greater than 1. High breeding ratios imply significant rates of salt processing to manage ^{233}Pa and remove fission products. If a breeding ratio of 1.01 is acceptable, major simplifications of the salt processing systems may be possible.

- *Fuel cycle fissile inventory*. There is an increasing interest in minimizing the total fissile inventory in the nuclear power system to reduce proliferation risks, reduce safeguards uncertainties, and reduce the time required to perform waste transmutation missions. The MSR system has a much lower fissile inventory than any other reactor system.

- *Actinide burning*. MSRs have some unique capabilities to burn actinides from other reactors. Because burning actinides from other reactors was not historically a development goal for the MSR, this change in goals has significant impacts on R&D needs. The needed R&D includes (1) obtaining a better understanding of the solubility of minor actinides and lanthanides in molten fluoride salt fuel for actinide burning missions with high actinide concentrations, (2) developing appropriate separations technologies, and (3) developing methods to produce appropriate waste forms.

The second factor in the rebirth of interest in the MSR has been the major advances in technology (Forsberg et al. June 2004b) such as Brayton cycles, compact heat exchangers, and carbon-carbon composites that (1) eliminate many of the technical difficulties associated with the earlier MSR designs, (2) significantly lower costs, and (3) improve performance. Much of the current R&D is designed to explore the implications of these technological advances on MSR economics and technical viability.

Other required R&D activities include (1) the consideration of impact of changing regulatory requirements developed over 30 years, (2) development of appropriate waste forms for repositories—an issue not addressed in the 1960s, (3) integration of nonproliferation and safeguards into plant design—an issue only partly addressed in the 1960s, (4) added materials testing, (5) improved molten salt processing technologies, (6) improved remote maintenance methods, and (7) better management of noble metal fission products. The fission process generates noble metal fission products. These do not dissolve as stable fluorides in the molten salt and can plate out on heat-exchanger walls as metal or lower-valence metal-metal bonded fluoride clusters. Molten salts do not wet carbon, so MSR primary systems built with advanced carbon composite materials may enable simple recovery and separation of noble metal fission products using metallic sponge filter materials. Better removal techniques for these fission produces is desirable and may be possible.

Many of the earlier technical challenges of the MSR have disappeared with advancing technology. However, it will be several more years before a clear understanding exists of the remaining challenges in developing a modern MSR. At present, the largest research program exists in France, with smaller programs in the United States and elsewhere.

2.4 Fusion

Molten salts (primarily lithium beryllium fluoride salts) are serious candidates for fusion energy machines to remove heat from the fusion reactor, breed tritium, and provide a renewable material for neutron shielding to reduce the mass of solid material exposed to damaging fusion neutron fluences. In a fusion reactor, tritium and deuterium fuse together to release energy, neutrons, and helium. The fusion machine must breed tritium to continue operation: $^6Li + n \rightarrow \,^4He + \,^3H$ (tritium). The lithium can be a solid in the blanket of the fusion energy machine or as part of the coolant. Consequently, low-pressure lithium-containing molten salts are being considered as coolants for fusion energy machines.

Molten salts are also being considered for one other application in inertial fusion machines where heavy ion beams, pulsed electrical power, or lasers are used to compress small pellets to extreme temperatures and cause a fusion explosion. This type of fusion machine produces an impulse on the fusion reactor wall that damages the wall over time. Molten salts (Moir 1994; Peterson 2001) may be used to form a liquid wall inside the fusion machine to shield structures from fusion neutrons and to absorb shock loads from targets. For heavy-ion and laser drivers, the vapor pressure of the salt must be extremely low to permit focusing of the beams on the target. As with other fusion applications, the molten salt is also used to breed tritium.

3. MOLTEN SALT PROPERTIES

Unlike water, sodium, or helium, fluoride molten salts are a family of coolants with similar general properties. The choice of a specific molten salt for a specific application is determined by functional requirements and costs. Many salts have been examined. Table 6 lists the leading candidates for various nuclear molten salt applications and key physical properties. The remainder of this section discusses the choices of salts and the constraints that limit the choice of salt.

3.1 Stability of Molten Salts and Materials of Construction

3.1.1 Molten Salts and Metals

A major challenge for all reactors is the development of materials of construction for components and heat exchangers that are compatible with the environment. For high-temperature service, there are four issues: strength over time (avoiding changes in metallic structure from various thermal mechanisms), long-term creep, corrosion resistance to air, and corrosion resistance to molten fluoride salts. While only coolant-metal compatibility will be discussed herein, this may not be the primary materials challenges for high-temperature reactors. The experience with high-temperature molten-salt-corrosion test loops is that air corrosion has often been more of a limitation than salt corrosion (Koger 1972)─particularly above 800°C. The other three materials issues are common challenges for all high-temperature reactors. In molten salt systems, Hastelloy-N has been code qualified for service to 750⁰C. Because of limitations from long-term high-temperature creep, this is near the upper long-term temperature limit for Hastelloy-N. Although many other candidate alloys exist, these have not been qualified for nuclear service.

Fluoride salts are fluxing agents that rapidly dissolve protective layers of oxides and other materials. To avoid corrosion, molten salt coolants must be chosen that are thermodynamically stable relative to the materials of construction of the reactor; that is, the materials of construction are chemically noble relative to the salts. This limits the choice to highly thermodynamically-stable salts. Table 7 shows the primary candidate fluorides suitable for a molten salt and their thermodynamic free energies of formation. The general rule to ensure that the materials of construction are compatible (noble) with respect to the salt is that the difference in the Gibbs free energy of formation between the salt and the container material should be >20 kcal/(mole °C). The corrosion strategy is the same as that used in sodium-cooled reactors where the materials of construction are noble relative to metallic sodium.

In high-temperature systems, there are also temperature-induced mechanisms for corrosion that are dependent upon large temperature differences in the heat transport system. The classic example is the transport of carbon in the form of various oxides in gas-cooled systems resulting in depletion of carbon in one part of the system and the deposition of carbon in another part. Parallel mechanisms have been seen in molten salt test loops. In this system, the primary concern has been the selective chemical transport of chromium (an alloy constituent of many alloys) from hot to cold locations in the reactor system by uranium where the equilibrium is temperature dependent:

$$2UF_4 + Cr = 2UF_3 + CrF_2$$
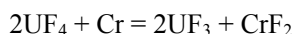
Chromium is the least thermodynamically stable element among the materials of construction. This and other corrosion mechanisms resulted in the development of Hastelloy-N as a material of construction, which offers very good corrosion resistance in MSR systems.

In this context, avoiding corrosion in an MSR with metallic components is significantly more difficult than avoiding corrosion in clean-salt-coolant applications (heat transport loops, AHTR, and fusion). In an MSR, the dissolved uranium in the fuel salt cannot be removed from the system so alloys such as Hastelloy-N that are resistant to such corrosion mechanisms must be developed. In clean-salt applications, these types of corrosion mechanisms can be reduced or eliminated by (1) using purified salts that do not contain chemical species that can transport chromium and other alloy constituents or (2) operating under chemically reducing conditions. Under chemically reducing conditions, chromium fluoride has an extremely low solubility which limits chromium transport. In the Molten Salt Reactor Experiment (MSRE), almost no corrosion was observed in the intermediate heat-transport loop with its clean salt. Higher-temperature test loops (Williams et al. 2003) indicate low corrosion rates with molten fluoride salts when these conditions are met. The practical engineering conclusion is that the development and qualification of metallic materials for higher-temperature clean-molten-salt systems (>750°C)─such as heat transport loops, the AHTR, and fusion reactors─will be significantly easier than for a higher-temperature MSR with its fuel salt. Molten salt systems are similar to water and helium coolant systems in that corrosion control depends upon control of coolant chemistry.

Table 6. Physical properties of selected molten salts[a]

Salt (mol %)	Form. wt. (g/mol)	Melt. pt. (°C)	Density (g/cc), T(°C)	700°C Heat capacity (cal/g-°C)	Viscosity (cP), T(K)	Thermal cond. (Watts/ cm-°C)
Alkali-Fluorides (IA) : Nontoxic						
LiF-NaF-KF (11.5-46.5-42)	41.2	454	2.53–7.3E-4*T	0.45	0.04exp(4170/T)	0.006-0.01
LiF-RbF (43-57)	70.7	475	3.30–6.9E-4*T	0.284 (est)	0.021exp(4678/T)	~ 0.06
Alkali + Alkaline Earth Fluorides (IA + IIA)						
LiF-BeF$_2$ (66-34)	33.1	458	2.28–4.884E-4*T	0.57	0.116exp(3755/T)	0.011
NaF-BeF$_2$ (57-43)	44.1	360	2.27–3.7E-4*T	0.52	0.034exp(5164/T)	~ 0.01
Alkali + ZrF$_4$: Nontoxic and Low Tritium Yield						
NaF-ZrF$_4$ (50-50)	104.6	510	3.79 –9.3E-4*T	0.28	0.071exp(4168/T)	~0.01 (est)
NaF-KF-ZrF$_4$ (10-48-42)	102.3	385	3.45–8.9E-4*T	0.26 (est.)	0.061exp(3171/T) (est.)	~0.01
Li-NaF-ZrF$_4$ (42-29-29)	71.56	460	3.37–8.3E-4*T	0.35	0.0585exp(4647/T)	~0.01
Fluoroborates: Secondary Salt Candidates						
NaF-NaBF$_4$ (8-92)	104.4	385	2.252–7.11E-4*T	0.36	0.0877exp(2240/T)	~0.005
KF- KBF$_4$ (25-75)	120.48	460	2.258–8.02E-4*T (est.)	> 0.32	Similar to KBF$_4$	~0.005
RbF - RbBF$_4$ (31-69)	151.25	442	2.494–8.7E-4*T (est.)	?	?	?
NaBF$_4$	109.8	408	2.263–7.51E-4*T	0.36	0.0832exp(2360/T) 0.0787exp(2406/T)	~0.005
KBF$_4$	125.9	570	2.228–8.15E-4*T	0.32	0.0946exp(2280/T)	~0.005
RbF$_4$	172.27	582	2.795–10.4E-4*T	?	?	~0.005

[a]Other mixtures of interest: NaF-RbF-ZrF$_4$ (8-50-42, mp = 400°C) and LiF-NaF-RbF (45-10-45, mp = 430°C), References: ORNL/TM-2316, ORNL/TM-4308, ORNL-4229; ORNL-4344, ORNL-4449, ORNL-4586, ORNL-4622, ORNL-4676; *Fluid Fueled Reactors* (1958); *Progress in Nuclear Energy* Series 4, Vol. 2, p. 140 (1960), *Nucl. Appl.Technol.* 8 (1970); *Nucl. Sci. Eng.* Vol. 71 p.200 (1963); R. DeWitt in *Phys and Chem.Liq.* Vol. 4 (no.2─3), pp.113-123 (1974).

Table 7. Thermodynamic stability of components of molten salts and structural materials

Constituent	Free energy of formation at 1000°K (kcal/mol-F)	Cation thermal capture cross section (barns)
Majority Constituents (>99.9 mol %)		
^{7}LiF	-125	0.033 (^{7}Li)
MgF_2	-113	0.063
NaF	-112	0.52
RbF	-112	0.70
KF	-109	2.1
BeF_2	-104	0.01
ZrF_4	-94	0.18
$^{11}BF_3$	~-95	0.05 (^{11}B)
AlF_3	-90	0.23
SiF_4	~XX	0.16
F	N	0.01
Structural Metal Constituents (Trace)		
CrF_2	-75.2	3.0
FeF_2	-66.5	2.5
NiF_2	-55.3	4.5
MoF_6	-50.9	2.5

3.1.2 Molten salts and graphite

A large experience base has demonstrated the compatibility of molten fluoride salts and graphite in radioactive and nonradioactive systems. The MSBR program investigated the compatibility of molten salts with graphite in chemical tests, loop tests, and reactors. In an MSR, the reactor core is made of bare graphite (the moderator) with the molten fuel salt flowing through channels in the graphite. In a classical molten salt reactor where the uranium and fission products are dissolved in the fuel salt, the fuel salt is dumped to storage tanks during shutdown. For safety and maintenance purposes, it is essential to know exactly where all the salt, fission products, and uranium are located. As a consequence, the interactions of salt and graphite were carefully investigated.

Post-irradiation examination from the MSRE showed no interactions (erosion or corrosion) between the salt and the graphite (McCoy and McNabb 1972). The original machining marks were still clearly visible. Out-of-reactor tests were conducted to 1400°C with no interactions between the salt and graphite (Rosenthal et al. 1969). Experiments show the nonwetting behavior (Fig. 7) of the fluoride salts of interest and that molten salts will not penetrate small cracks in the graphite (Briggs 1963; ORNL-3626, 1964; Fontana and Winard 1970). For the AHTR, this has the practical implication that the clean coolant will never contact the fuel micro spheres that are embedded in a carbon matrix.
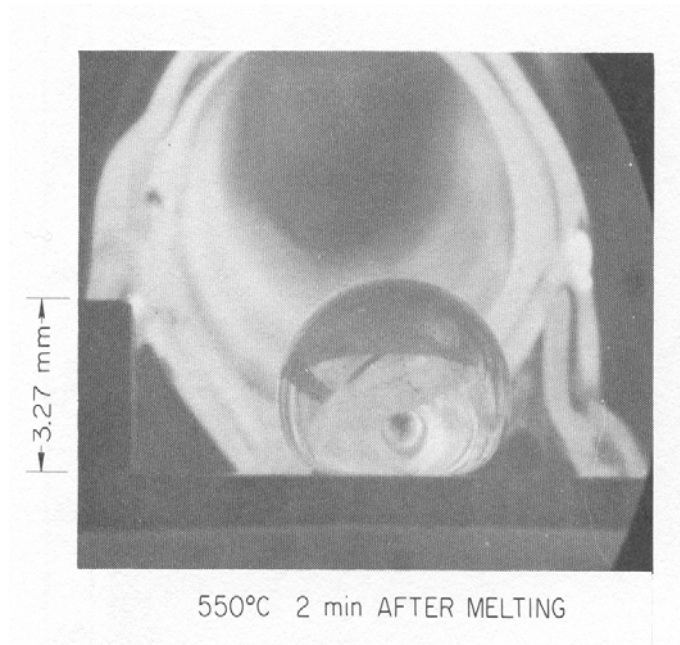
Fig. 7. Nonwetting characteristics of molten fluoride salts and graphite.

Because of the excellent compatibility of graphite with molten salts, there is ongoing research on the use of various carbon-carbon composites for all types of components in molten salt systems (Peterson et al. 2003b). In particular, the development of carbon-carbon composite heat exchangers may allow operations at temperatures significantly above 1000ºC and avoid the issues associated with metallic systems.

3.1.3 Reactions of molten salts with other fluids

Molten salts do not react with helium or nitrogen but will react slowly with water over time. Experiments have been conducted to determine the impact of injecting molten salt into water, such as might occur in some types of systems that have water. In the largest experiment, 230 kg of molten salt at 815°C was injected into a water bath over 45 to 50 seconds with the discharge pipe 1.5 m (5 ft) under the water level. No steam explosion occurred, only a negligible amount of steam reached the surface of the water, and no entrainment of water or steam into the air was observed (Mann 1955).

3.2 Nuclear Criteria for Salt Selection

Chemical criteria provide the first set of constraints for selection of usable salts. Nuclear criteria provide the second set of constraints.

3.2.1 Low-nuclear-absorption cross section

The neutron-absorption cross sections of any molten salt for reactor applications must be low to avoid excessive parasitic capture of neutrons. For thermal and intermediate neutron-spectrum reactors, this probably eliminates chloride salts with their higher nuclear cross sections, even if the higher the high-cross-section ^{35}Cl is removed. Only fluoride salts are candidates. A wide variety of atoms have low cross sections; however, the realistic candidates are also restricted by the requirements of thermodynamic stability to ensure viable materials of construction for the container. Table 7 shows the primary salt options and their cross sections. Isotopically separated lithium and boron are required to have a salt with a low absorption cross section. The exception is fusion reactors, where ^{6}Li with its high cross section is required to breed tritium.

3.2.2 Radiolysis of molten salts

Unlike water, fluoride salts do not undergo radiolysis in radiation fields when *molten,* and no fluorine will be generated. No radiolysis (ORNL-3708, 1964) was detected in flowing loops of molten salt operated in intense radiation fields of the Materials Testing Reactor. Molten fluoroborate coolant salts (containing BF_3 + alkali-fluorides) were also tested for their radiolysis response, and none was found (Compere et al. 1970). In fact, the reverse reaction—recombination—typically counteracts primary radiolysis events far below the melting point of the salt. For *solid* $2LiF–BeF_2$ salt in a radiation field, the temperature that inhibited a net radiolysis response was ~150°C (Toth and Felker 1990). For other solid fluoride salts (ZrF_4), no radiolysis response was found above room temperature, and radiolysis at room temperature was not observed for NaF-ZrF_4 salts in the ARE (ORNL-3626, 1964). This radiation stability is partly a consequence of the materials requirement that only very stable fluoride salts can be considered to ensure that the metals of construction are thermodynamically stable with respect to the salt.

3.2.3 Environmental and occupational considerations

Several environmental and occupational factors impact the choice of salt for specific applications.

*   *Waste management.* The choice of salt can have major impacts on waste management. In addition to their relatively high neutron absorption cross sections, chloride salts create significant waste management challenges. In a reactor the high-cross-section ^{35}Cl is activated to ^{36}Cl—a long-lived radionuclide with a half-life of 300,000 years. Most chlorides are soluble in water, which makes it more difficult to avoid long-term release of ^{36}Cl from a repository to the environment (Poinssot et al. 2002). In contrast, fluorides have low neutron cross sections, do not activate to radionuclides that create major challenges in waste management systems, and form many insoluble waste forms. If a chloride salt was used, isotopically-separated ^{37}Cl would probably be required.

*   *Chemical toxicity.* The toxicity of the molten fluoride coolant depends upon the specific salt and varies from the fluoride salts used in toothpaste for prevention of tooth decay to toxic materials.

*   *Neutron activation.* Some salts produce tritium under radiation (lithium and beryllium). Other salts such as sodium produce gamma-emitting radionuclides. The choice of coolant impacts the need for tritium control systems and radiation shielding in the primary system.

3.3 Physical Properties

The chemical and nuclear criteria define the allowable elements for a molten salt coolant. Physical property requirements are used to define the candidate molten salts for specific applications. The requirements include (1) a good coolant with coolant properties generally between those of water and sodium, (2) a coolant freezing point as low as possible, and (3) application-specific requirements.

In all cases, binary or more complex fluoride salt mixtures are preferred because the melting points of fluoride salt mixtures are much lower than those for single-component salts. For example, the molten salt Li_2BeF_4 has a melting point of 457°C, whereas pure LiF has a melting point of 847°C and pure BeF_2 has a melting point of 544°C. Other traditional candidate salts include $NaF-ZrF_4$ (50 mol % NaF, 50 mol % ZrF_4), with a melting point of 510°C, and $NaF-RbF-ZrF_4$ (8 mol % NaF, 50 mol % RbF, and 42 mol % ZrF_4), with a melting point of 400°C. With some three-component mixtures such as $^7LiF-BeF_2-NaF$, and potentially with four-component mixtures, it is possible to reduce melting points to ~350°C. At operating conditions, the thermophysical properties of molten salts are similar to those of water except for the very low vapor pressure.

The MSR imposes additional requirements on the salt that do not exist for molten-salt heat transport loops, the AHTR, or fusion energy machines. Actinides and fission products must have very high solubility in the salt. For operation as a converter reactor for electricity or hydrogen production or for burning of actinides, a NaF and ZrF_4 containing salt similar to the ARE salt could be used. These salts are inexpensive, relatively nontoxic, and have a high solubility for actinides and lanthanides. For breeder reactor applications, the nuclear constraints in minimizing neutron absorption will almost certainly require a mixture of ^7LiF, BeF_2, ThF_4, and UF_4.

4. ENGINEERED SYSTEMS

4.1 Status of Engineering

In the 1960s, a major effort was undertaken to develop a 1000-MW(e) MSR with an exit coolant temperature of 705°C. This initiative included developing code-qualified materials (Hastelloy-N) for long-term operating temperatures up to 750°C, pump test loops, and a wide variety of other developmental work. As a consequence, there is reasonable confidence that the base technology (pumps, materials, etc.) exists for these operating temperatures.

4.2 Heat Transfer

Molten salts have a unique set of properties in terms of heat transfer: they are transparent high-heat-capacity liquids. As a consequence, radiation heat transport can become an important mechanism at high temperatures (>700°C) where the rate of radiation heat transport varies as T^4. While the basic theory of heat transfer accounts for radiation heat transfer, very little practical engineering experience exists. Although traditional correlations that do not account for radiation heat transfer will provide conservative estimates of heat transfer, actual performance may be significantly different.

4.3 Reactor Heat Transport

Low-pressure liquid-cooled reactor systems can have substantially higher efficiencies than gas-cooled reactor systems for the same reactor exit temperatures (Fig. 8). Gas-cooled reactor systems have high pumping costs relative to liquid-cooled systems. Because direct 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 any reactor (AHTR, MSR, or fusion) with a molten salt coolant. If one compares helium-cooled and molten-salt-cooled high-temperature reactors, a helium-cooled reactor (the GT-MHR) with a peak temperature of 850°C

delivers its average heat at the same temperature as a molten-salt-cooled AHTR with a peak coolant temperature of 750EC. With multi-reheat Brayton cycles, the small temperature range of delivered heat with molten salts results an increase in thermodynamic efficiency of 4 to 8% above that of gas-cooled reactors with similar core outlet temperatures.
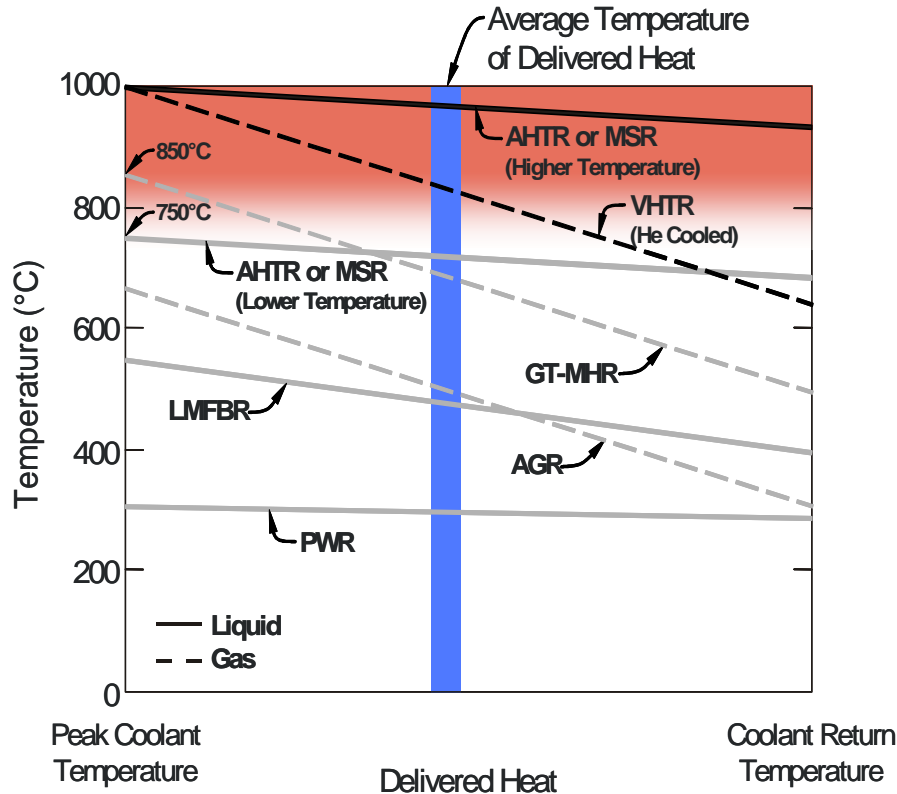

Fig. 8. Temperature of delivered heat from different reactors.

4.4 Brayton Power Cycles

For electricity production, recuperated gas (nitrogen or helium) Brayton cycles (Fig. 3) are proposed for use with molten salts with two to three stages of reheating and three to eight stages of intercooling (Peterson 2003). This is independent of whether the reactor is the AHTR, an MSR, or a fusion energy machine. Nitrogen and helium are inert relative to molten salts. Brayton power cycles are the only efficient power cycles that have been developed that match the temperatures of high-temperature reactors. The multi-reheat Brayton cycle is equivalent to the multi-reheat steam cycle used in many coal-fired power stations. The gas pressure is reduced through multiple turbines in series, with reheating of the gas to its maximum temperature with hot molten salt before it reaches each turbine. The gas then flows through a recuperator and is compressed in multiple stages with interstage cooling. Table 8 describes the characteristics of three Brayton high-efficiency Brayton cycles with different peak molten salt temperatures. Both nitrogen and helium Brayton cycles are being considered. The major advantage of nitrogen Brayton cycles is that the turbomachinery is similar to commercially available natural-gas, combined-cycle plants.

4.5 Dry Cooling

Historically, one of the major limitations of thermal energy systems is the need for cooling water to reject heat. Most power plants are located on rivers or the seashore. Siting options for nuclear power plants would be greatly expanded with the use of dry cooling towers. Power plant siting is further complicated by shortages of cooling water in many parts of the world. In the United States, cooling water for steam plants is the largest single use of water (Hutson et al. 2004). There are major legal and political battles over water rights. While dry cooling is used, it is uncommon because of the high cost: both the direct cost of the cooling towers and the hidden, but equally real cost of lower plant efficiency. Cost estimates (California Energy Commission 2002) for dry cooling are as high as several hundred dollars per kW(e) when considering both direct costs and the efficiency penalties. The economic penalties of dry cooling are drastically reduced with high-temperature Brayton-cycle power stations.

- *Less heat rejection*. Current LWRs have efficiencies of ~33%. For AHTRs, MSRs, and fusion reactors with peak coolant temperatures of 705, 800, and 1000ºC, the respective plant efficiencies are 48, 51.5, and 56.6%. While the LWR rejects 2 kW(t) of heat for every kW(e), the three high-temperature power cycles reject, respectively, 1.08, 0.94, and 0.77 kW(t) for every kW(e). The higher efficiency reduces the capacity requirements for heat rejection by about a factor of two.

- *Reduced penalty for higher heat-rejection temperatures.* The capital costs of dry cooling systems can be reduced by rejecting heat at a higher temperature. The efficiency penalty becomes smaller as the peak temperature of the power cycle increases. For the three Brayton cycles, the *losses of efficiency* for a 10ºC rise in the compressor inlet temperature are about 1.1, 1.3, and 1.5%, respectively, for the high-, intermediate-, and low-temperature systems.

- *Heat rejection over a temperature range*. Dry cooling involves heating air from a lower temperature to a higher temperature. If the power cycle rejects heat over a temperature range rather than at a single temperature, the appropriate design of countercurrent dry-cooling-tower heat exchangers maximizes the temperature drop across the heat exchangers, which reduces the required size of these heat exchangers. With dry cooling, Brayton cycle has a major advantage over the Rankine cycle. Rankine (steam) cycles deliver the rejected heat at a constant temperature—a characteristic is consistent with evaporative cooling, in which water is vaporized at a nearly constant temperature. In contrast, a Brayton cycle delivers rejected heat over a temperature range as the gas is compressed. In the designs herein, the heat is rejected over a 50ºC range.

5. CONCLUSIONS

High-temperature reactors offer major advantages relative to other reactors in terms of efficiency and hydrogen production. High-temperature, low-pressure molten salt coolants are an enabling technology for large passively safe reactors (AHTR, MSR, fusion reactors) that may be required to meet economic goals. The AHTR potentially represents the next generation nuclear power plant (after LWRs) that could be commercialized before 2020. While the MSR is an intermediate-term option, fusion energy is a long-term option. There is a natural sequence of technical development from the AHTR to an MSR or to a fusion energy machine.

6. ACKNOWLEDGEMENTS

Table 8. Comparison of parameters for different-temperature Brayton cycles[a]

	High Temperature	Intermediate temperature	Low temperature
Thermal power	2400 MW(t)	2400 MW(t)	2400 MW(t)
Electrical power	1357 MW(e)	1235 MW(e)	1151 MW(e)
Primary max. temperature	1000°C	800°C	705°C
Primary min. temperature	900ºC	700ºC	670ºC
Intermediate loop max. temperature	920°C	770°C	690°C
Intermediate loop min. temperature	860°C	690°C	620°C
Intermediate flow rate	10.3 m^3/s	7.4 m^3/s	8.3 m^3/s
Turbine inlet temperature	900°C	750°C	675°C
Compressor inlet temp	35°C	35°C	35ºC
Compressor outlet temp	86°C	76°C	80°C
IHX power density	120 MW(t)/m^3	30 MW(t)/m^3	50 MW(t)/m^3
Recuperator power density	40 MW(t)/m^3	50 MW(t)/m^3	40 MW(t)/m^3
Heater power density (from high pressure to low pressure)	120 to 80 MW(t)/m^3	80 to 60 MW(t)/m^3	70 to 50 MW(t)/m^3
Overall cycle efficiency	0.566	0.515	0.48
PCU power density	560 kW(e)/m^3	490 kW(e)/m^3	460 kW(e)/m^3

[a]IHX = intermediate heat exchanger; PCU = power conversion unit. The power conversion system (Peterson 2003) is a helium Brayton cycle with three stages of reheat and six stages of intercooling. The peak Brayton cycle pressure is 10 MPa.

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