

Carbon-Carbon-Composite Salt-Cooled Electric Space Reactors

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

Timothy D. Burchell
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, TN
Tel: (865) 576-8595
E-mail: burchelltd@ornl.gov

David F. Williams
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, TN
Tel: (865) 574-5769
E-mail: williamsdf2@ornl.gov

David E. Holcomb
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, TN
Tel: (865) 576-7889
E-mail: holcombde@ornl.gov

Ray F. Holdaway
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, TN
Tel: (865) 576-2509
E-mail: holdawayrf@ornl.gov

A. L. Qualls
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, TN
Tel: (865) 574-0259
E-mail: quallsal@ornl.gov

File Name: ANS05.SpaceReactorPaper
Manuscript date: March 3, 2005
Final paper due date: April 15, 2005

Space Nuclear Conference 2005 (SNC'05)
Manuscript Number: 1148; Session 1.03
Embedded Topical Meeting at the 2005 American Nuclear Society Annual Meeting
American Nuclear Society
San Diego, California
June 5-9, 2005

The submitted manuscript has been authored by a contractor of the U.S. Government
under contract DE-AC05-00OR22725. Accordingly, the U.S. Government retains a
nonexclusive, royalty-free license to publish or reproduce the published form of this contribution,
or allow others to do so, for U.S. Government purposes.

*Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy
under contract DE-AC05-00OR22725.

Carbon-Carbon-Composite Salt-Cooled Electric Space Reactors

C. W. Forsberg, T. D. Burchell, D. F. Williams, D. E. Holcomb, R. F. Holdaway, and A. L. Qualls
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, TN 37831-6165
Tel: (865) 574-6783; E-mail: forsbergcw@ornl.gov

Abstract — The primary requirements for a deep space or planetary nuclear reactor for production of electricity are reliability, long life, and a high power-to-mass ratio. Advanced reactors (core, structure, power-conversion systems, radiator, etc.) are proposed that are built entirely from carbon-based materials that use salts (liquid or gas) as the heat transfer medium between the reactor and power-generation equipment and/or heat rejection systems to create reactor systems with very high power-to-mass ratios. While currently proposed space nuclear reactors have peak temperatures between 900 and 1400K with potential efficiencies as high as 30% for advanced Stirling engines, the new reactors would have peak operating temperatures between 1800 and 2300K with potential efficiencies twice that of other concepts. Because there are no other classes of materials that can potentially operate at such temperatures and have very low masses, most of the components of such a space electric nuclear reactor must be built of carbon-based materials. Based on theoretical considerations and the developments in carbon-carbon technologies over the last 20 years, such machines appear to be potentially viable. However, significant research is required to demonstrate feasibility and a major long-term development program would be required to build such machines.

I. INTRODUCTION

The primary requirements for a deep space or planetary reactor are reliability, long life, and a high power-to-mass ratio. Proposed space nuclear reactors have peak temperatures between 900 and 1400K with efficiencies as high as 30% for advanced Stirling power-conversion cycles. Advanced reactors (core, structure, power systems, radiator, etc.) are proposed that are built entirely from carbon-based materials that use salts (liquid or gas) as the heat transfer medium between the reactor and power-generation equipment and/or heat rejection systems to create reactor systems with very high power-to-mass ratios.

- Carbon-carbon composites are among the strongest and lightest high-temperature engineered materials. Moreover, carbon-carbon composites today can be fabricated into complex forms. Unlike metals, the strength of carbon-based materials increases up to temperatures of $\sim 2300\text{K}$. *This technology has been developed primarily over the last 20 years, after the last major effort to develop nuclear space systems in the United States. Carbon-carbon composites are the only new class of materials with the potential to dramatically improve performance.*
- Coated-particle nuclear fuels with zirconium carbide and other coatings have operating temperatures that approach 2300K. This coated-particle fuel can be incorporated into carbon-based matrixes.

- Carbon foams are a new material with extraordinary low densities and high thermal conductivities—a class of materials that may enhance heat transfer in radiators.

Only two classes of fluids are chemically compatible with carbon-based materials: inert gases (e.g., helium-xenon mixtures) and fluoride-based salts. Liquid metals are incompatible with carbon-based materials. Fluoride-based liquid salts (molten salts) were used in the Nuclear Aircraft Propulsion program and have demonstrated compatibility with carbonaceous materials. The choice of constituent salts in a salt mixture can be adjusted to modify the physical properties (freeze point, volume change upon melting, vapor pressure, viscosity, boiling point, etc.) to meet mission requirements.

These innovative materials are logical candidates for the construction of advanced space reactors operating at very high peak temperatures. At such extreme temperatures, the entire system would be built of carbon-carbon composites because no other potential materials of construction exist that can withstand the temperature and have low masses. Higher-temperature operation fundamentally leads to higher cycle efficiency (smaller reactors, power-conversion units, heat rejection systems) and thereby to higher power-to-mass ratios. The first applications of such a technology would be for space missions. Planetary missions provide the additional challenge of protectively coating the exterior, high-temperature carbon-based materials against oxidizing environments (such as that on Mars).

The characteristics of carbon composites, coated-particle fuels, and carbon foams, as well as their compatibility with salts and the constraints and characteristics of these salts, are described. The challenges, some potential reactor configurations, and the implications for a family of advanced space reactors are described.

II. CARBON-BASED MATERIALS

II.A Carbon Material Properties

The ultimate efficiency of a nuclear power system depends upon peak temperatures which, in turn, depend upon the materials of construction. Carbon-carbon composites¹ can operate at higher temperatures than other materials (Fig. 1), retain their room-temperature strength at temperatures up to $>2500\text{K}$ ($>2225^\circ\text{C}$), and have much higher strength-to-weight ratios than other candidate materials. The theoretical peak temperatures of carbon-based materials are limited by the carbon sublimation temperature of $\sim 3350^\circ\text{C}$, which is close to the 3400°C melting point of tungsten. Carbon-carbon composites have been demonstrated to (1) maintain reproducible strength at 1650°C , (2) withstand large thermal gradients, (3) have low coefficients of thermal expansion and thus the potential to minimize thermal stresses, (4) have tolerance to impact damage, and (6) be manufacturable.

Carbon-based materials have two major restrictions. First, carbon atoms must be protected against oxidizing environments (planetary environments such as Mars). The current temperature limit for protective coatings is about 1875K ($\sim 1600^\circ\text{C}$). Second, high-performance carbon materials are more difficult to repair than many metals. Although these are major limitations for

terrestrial applications, these restrictions do not apply to typical space applications. For most space or lunar applications, no oxidizing environment is present and there are no requirements for repair. Planetary surface applications would require protection against oxidizing and reactive atmospheres.

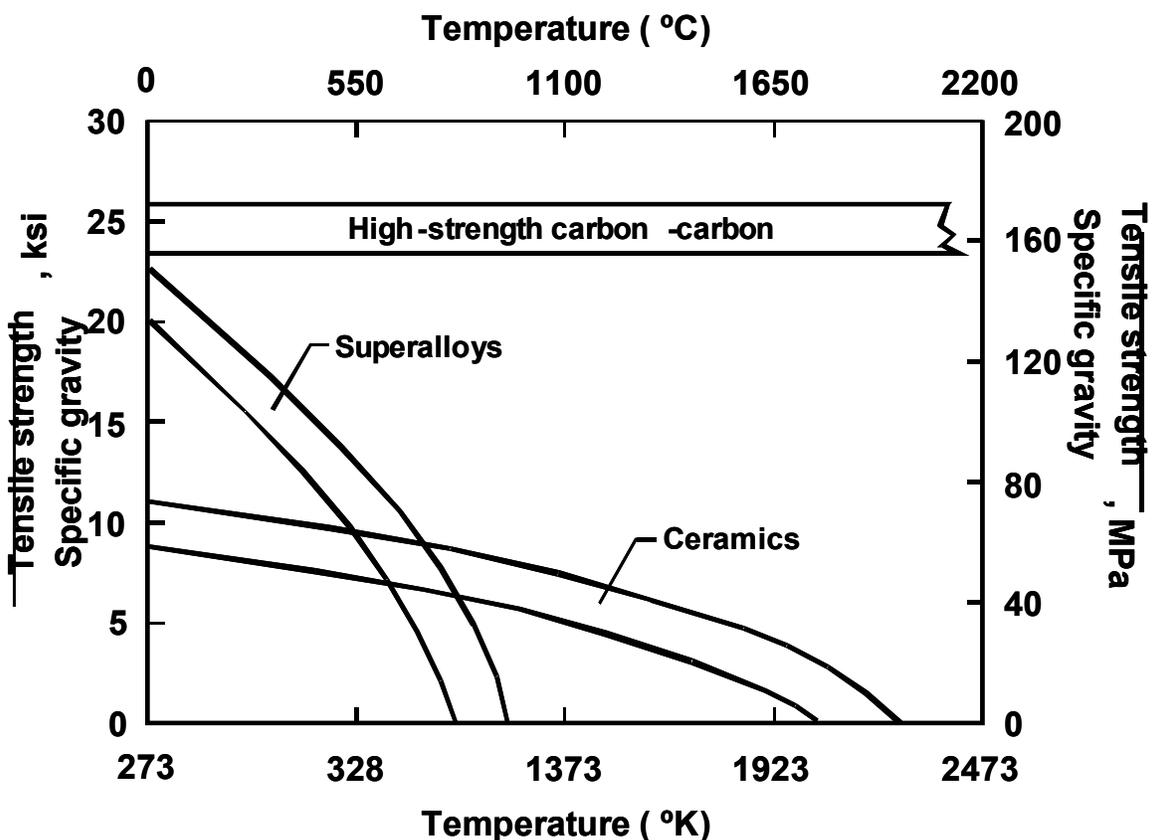


Fig. 1. Strength of High-Temperature Materials Versus Temperature

Carbon-carbon composites have been developed for fusion² and fission³ applications with short-term operating temperatures up to 1600°C. Graphite and carbon-carbon composites are used in a variety of high-temperature nuclear reactors. The characteristics of these materials under neutron radiation have been extensively studied and are dependent upon the specific material, the type of neutron damage, and the temperatures. In most cases, radiation damage is reduced as temperatures increase.

II.B. Carbon Foams

Carbon can be produced in many forms. The past decade has seen the development of carbon foams.^{4,5} While the thermal conductivity of these foams is similar to that of aluminum, their densities are 15 to 25% of those of aluminum and they can operate at extreme temperatures. Figure 2 shows the low-density structure of these materials. Such materials have three major applications relevant to a very high temperature space reactor.

- *Space radiator.* Carbon foams have been flight qualified as a low-temperature radiator for spacecraft applications. They can be operated at high temperatures.
- *Boiling heat transfer.* The foams can be placed inside heat transfer surfaces to conduct heat from tubes through the foam to the fluid. In effect, the internal surface area for heat transfer can be greatly expanded.
- *Condensing heat transfer.* The foams can be placed inside heat transfer surfaces to conduct heat from the fluid to tubes.

The surfaces of the foam can be coated with various materials including high-temperature metals to change their surface properties (wetting characteristics, etc.)

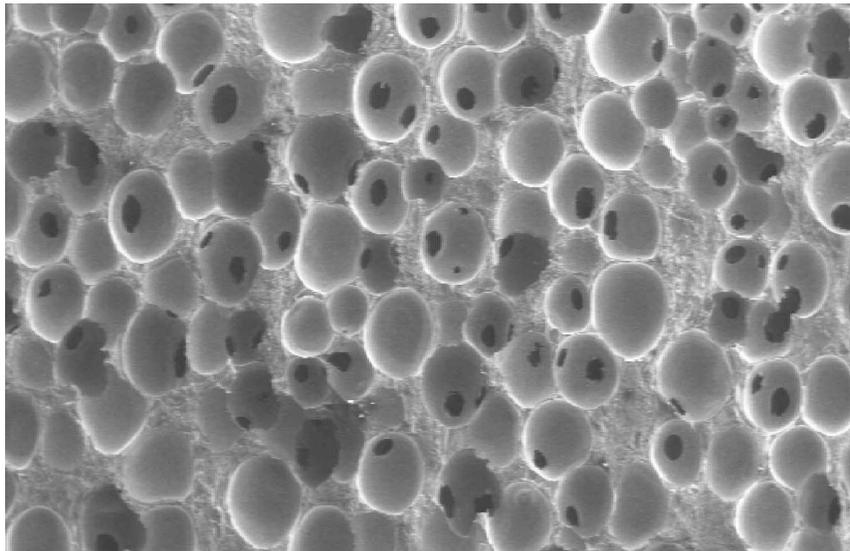


Fig. 2. Carbon Foam

II.C. Carbon Challenges

Two major challenges exist for very high temperature reactor applications of carbon-based materials.

- *Radiation damage.* Carbon-carbon composites and graphites are used in many nuclear reactors; however, these materials degrade when subjected to in-core radiation levels. At the same time, it is known that most types of radiation damage are reversed by treatment at temperatures near 2000°C. Theoretical considerations and indirect experimental evidence suggest that at very high temperatures there may be sufficient self-healing of the carbon materials to enable extreme fuel burnups and long-term operations at temperatures between 1500 and 2000°C
- *Permeability.* Unlike metals, the permeability of composites is not necessarily zero. Development of very low permeability carbon-carbon composites for operation at extreme temperatures for very long time periods may be a major challenge. Current successful techniques include infusion of carbon and sometimes other materials into the matrix by a variety of techniques. However, none of these methods have been tested for very high temperatures and very long time periods.

III. NUCLEAR FUELS

High-temperature reactors require high-temperature fuels. Multiple options are available. Coated particles consisting of uranium oxide or carbide microspheres covered with multiple layers of different types of carbon and other materials to retain fission products are the traditional fuel for high-temperature graphite reactors. For very high temperature operation, the likely preferred coating material is zirconium carbide (ZrC).⁶⁻⁹ The ultimate temperature limit for ZrC is dictated by its formation of a eutectic with carbon at 2850°C. If the ZrC-coated particles can be isolated from the carbon, the melting point of the ZrC is approximately 3540°C. Only limited operating data exists above 1600°C, but a few short-term tests have been conducted to 2500°C. Work is ongoing to develop this fuel form. Alternative higher-temperature coated-particle fuels based on NbC and TaC have been proposed, but very little work has been done on these materials.

IV. LIQUID SALTS

Only two families of coolants are compatible with carbon-based materials: noble gases (e.g., helium) and fluoride salts. Liquid metals are incompatible with carbon-based materials. Liquid fluoride salts were originally developed as a coolant in the 1950s and 1960s for the Aircraft Nuclear Propulsion Program and the Molten Salt Breeder Reactor Program.¹⁰ This particular reactor concept used fuel dissolved in the salt coolant. Two experimental reactors were built and successfully operated. The Aircraft Reactor Experiment, the first molten salt reactor (MSR), was a 2.5-MW(t) reactor that was operated in 1954 at a peak temperature of 860°C, used a sodium-zirconium-uranium fluoride salt, and proved the principle of an MSR. This was followed in 1965 by the Molten Salt Reactor Experiment, an 8-MW(t) reactor that used a lithium-beryllium fluoride salt, demonstrated most of the key technologies for a power reactor, and operated over a

4-year period of time. These programs operated various salt test loops for several hundred-thousand hours and produced over 1000 technical reports. More recent programs are examining the use of liquid salts with graphite-matrix fuels. While experiments to demonstrate the compatibility of graphite with salts were conducted only to 1400°C, these experiments and theory provide a basis for consideration of these salts as liquids or vapors to much higher temperatures. (The chemical industry uses a variety of liquid salts in heat transport systems but at somewhat lower temperatures.)

Fluoride salts are thermodynamically stable at high temperatures with very high boiling points. Liquid fluoride salts do not react with helium or nitrogen but will react slowly with water. 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 and the physical properties can be modified to match mission requirements. The compatibility of liquid salts with graphite has been demonstrated in chemical tests, loop tests, and reactors. Most fluoride salts do not wet graphite; however, surfaces of carbon components can be modified to alter wetting behavior. Table 1 shows the properties of some salts relative to those of other coolants. The very high heat capacities imply low coolant inventories relative to those for other coolants.

Fluoride salts are fluxing agents that rapidly dissolve protective layers of oxides. After carbon, borides and nitrides appear to be the most compatible nonmetallic materials. To avoid corrosion in systems that contain metallic components, liquid-fluoride-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 2 shows some candidate fluorides suitable for a liquid-salt coolant 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/(mol °C). Nuclear reactor considerations impose requirements that the salt constituents have low neutron absorption cross sections, which represents a second independent constraint on the selection of salt constituents. Several potential salt constituents have low cross sections. In the cases of lithium and boron, there are very low cross-section isotopes that are potential salt constituent candidates—provided that the high-cross-section isotopes are removed.

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 · 10 ⁶ (m ² /s)	N _p (mol/cm ³)
Li ₂ BeF ₄ (Flibe)	459	1,430	1,940	2.34	4,540	1.0	2.9	0.137
0.58NaF-0.42ZrF ₄	500	1,290	3,140	1.17	3,670	~1	0.53	0.108
Sodium	97.8	883	790	1.27	1,000	62	0.25	0.034
Lead	328	1,750	10,540	0.16	1,700	16	0.13	0.051
Helium (7.5 MPa)			3.8	5.2	20	0.29	11.0	0.001
Water (7.5 MPa)	0	100	732	5.5	4,040	0.56	0.13	0.122

^aNomenclature: ρ is density; C_p is specific heat; k is thermal conductivity; v is viscosity; and N_p is number density.

Table 2. Thermodynamic stability of components of liquid salts and structural materials

Constituent	Free energy of formation at 1000K (kcal/mol-F)	Cation thermal capture cross section (barns)
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 B ^a F ₃	~-95	0.05 (¹¹ B)
AlF ₃	-90	0.23
F	N	0.01
CrF ₂ ^a	-75.2	3.0
FeF ₂ ^a	-66.5	2.5
NiF ₂ ^a	-55.3	4.5
MoF ₆ ^a	-50.9	2.5

^a Components of metal alloys for fluoride salt systems

V. REACTOR APPLICATIONS AND CONCEPTS

V.A. Reactor Core

No work has been undertaken to develop an ultra-high-temperature space-reactor core using carbon-based materials, coated-particle fuels, and liquid salt coolants. However, work is underway in the United States and elsewhere to develop the Advanced High-Temperature Reactor,¹¹⁻¹³ which uses similar technologies at lower temperatures. The AHTR goal is a commercial 2400-MW(t) high-temperature reactor designed to produce electricity and hydrogen with peak temperatures between 700 and 1000°C with a plant lifetime in excess of 60 years. Liquid-salt-cooled heat transport systems are also being developed for (1) transport of heat from high-temperature nuclear reactors to hydrogen production plants, (2) fast breeder reactors,¹⁴ and (3) fusion reactors.¹⁵ While the temperatures of all of these machines are much lower, they provide a technological base for a space reactor.

One option for a high-power core is to design the core as a printed-circuit heat exchanger. Printed-circuit heat exchangers (Fig. 3) are currently manufactured from various metals and

carbon-carbon composites.¹⁶ Such heat exchangers have been built for high-pressure and moderate-temperature (900°C) operations. Plates are machined with coolant channels, stacked in layers, and bonded together. For a reactor core, the same approach can be used with separate channels milled for the fuel microspheres and coolant. For larger heat exchangers (kilowatt and megawatt), the power densities of these heat exchangers exceed those for other designs. Similar high-power densities should be viable for space reactors.

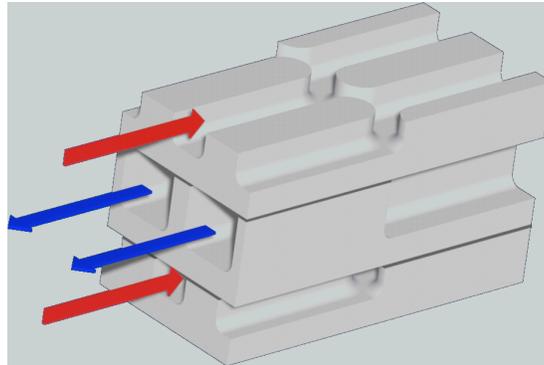


Fig. 3. Graphite-Composite Printed-Circuit Heat Exchangers

V.B. Power Cycles

Spacecraft power-conversion systems can be divided into two classes: static (thermoelectric and thermophotovoltaic) and dynamic (Stirling, Rankine, and Brayton). For the static power options, fundamentally new systems would be required because of the extreme temperatures. For dynamic power systems, such as Stirling engines,¹⁷ there is the option to develop carbon-carbon composite versions of these systems that operate at much higher temperatures. For example, Stirling engines consist of pistons and heat exchangers. Programs are currently under way for development of ultra-high-performance internal-combustion engines with carbon-carbon composite pistons and cylinders. This technology base^{18,19} provides a starting point for development of very high temperature Stirling engines. Similarly, many turbine components have been built of carbon-based materials.¹ The higher-temperature options should allow major increases in efficiencies.

VI. CONCLUSIONS

If major advances in space nuclear electric power systems are to be achieved, new approaches are required. Based on presently understood material properties, significantly improved performance should be possible using space reactors built of carbon-carbon composites, coated-particle fuels, and liquid-salt coolants. The developments in carbon materials over the past two decades provide the basis for consideration of such an advanced reactor. However, major

research will be required to determine the real potential and major development will be needed to build a practical system. The primary limitation is the lack of experimental data at extreme temperatures.

REFERENCES

1. J. D. BUCKLEY, "Carbon-Carbon Overview," *Carbon-Carbon Materials and Composites*, ed. J. D. Buckley and D. D. Edie, Noyes Publications, Park Ridge, New Jersey (1993).
2. L. L. SNEAD, "Fusion Energy Applications" pp. 389-427 in *Carbon Materials for Advanced Technologies*, ed. T. D. Burchell, Pergamon, New York, (1999).
3. T. D. BURCHELL, "Fission Reactor Applications of Fusion," pg. 429-484 in *Carbon Materials for Advanced Technologies*, ed. T. D. Burchell, Pergamon, New York (1999).
4. J. KLETT, R. HARDY, E. ROMINE, C. WALLS, and T. BURCHELL, "High-Thermal-Conductivity, Mesophase-Pitch-Derived Carbon Foams: Effect of Precursor and Structure and Properties," *Carbon*, **38**, 953-973 (2000).
5. J. W. KLETT, A. D. McMILLAN, N. C. GALLEGO, and C. A. WALLS, "The Role of Structure on the Thermal Properties of Graphitic Foams," *J. Mater. Sci.*, **39**, 3659-3676 (2004).
6. K. MINATO, T. OGAWA, K. FUKUDA, H. NABIELEK, H. SEKINO, Y. NOZAWA, and I. TAKAHASHI, "Fission Product Releases from ZrC-Coated Fuel Particles During Post Irradiation Heating to 1600°C," *J. Nucl. Mater.*, **224**, 85-92 (1995).
7. K. MINATO, et al., "Irradiation Experiment on ZrC-Coated Fuel Particles for High-Temperature Gas-Cooled Reactors," *Nucl. Technol.*, **130**, 272-281 (2000).
8. T. OGAWA, K. MINATO, and K. SAWA, "Advanced Coated Particle Fuels: Experience of ZrC-TRISO Fuel Development and Beyond," *11th International Conference on Nuclear Engineering (ICONE-11)*, Tokyo, Japan April 20-23 (2003).
9. K. Verfondern, J. Sumita, S. Ueta, and K. Sawa, *Modeling of Fuel Performance and Fission Product Release Behavior During HTTR Normal Operation*, JAERI-Research 2000-067, Japanese Atomic Energy Research Institution, Ibaraki, Japan, (March 2001).
10. *Nucl. Appl. Technol.*, **8**(2) (Entire issue) (1970).
11. C. W. FORSBERG, P. S. PICKARD, and P. F. PETERSON, "Molten-Salt-Cooled Advanced High-Temperature Reactor for Production of Hydrogen and Electricity," *Nucl. Technol.* **144**, 289-302 (2003).

12. D. T. INGERSOLL, et al. *Status of Preconceptual Design of the Advanced High-Temperature Reactor*, ORNL/TM-2004/104, Oak Ridge National Laboratory, Oak Ridge, Tennessee (2004).
13. C. W. FORSBERG, “Reactors with Molten Salts: Options and Missions,” *Proc. Frederic Joliot & Otto Han Summer School on Nuclear Reactors: Physics, Fuels, and Systems, Cadarache, France, August 25–September 3, 2004* (2004).
14. C. W. FORSBERG, P. F. PETERSON, and D. F. WILLIAMS, “Practical Aspects of Liquid-Salt-Cooled Fast-Neutron Reactors,” *2005 International Conference on Advances in Nuclear Power Plants, Seoul, Korea, May 15–19, 2005*, American Nuclear Society, La Grange Park, Illinois (2005).
15. R. W. Moir, et al., “HYLIFE-II: A Molten-Salt Inertial Fusion Energy Power Plant Design—Final Report,” *Fusion Energy* **25**, 5–25 (1994).
16. P. F. PETERSON, C. W. FORSBERG, and P. S. PICKARD, “Advanced CSiC Composites for High-Temperature Nuclear Heat Transport with Helium, Molten Salts, and Sulfur-Iodine Thermochemical Hydrogen Process Fluids,” *Proc. OECD/NEA Second Information Exchange Meeting on Nuclear Production of Hydrogen*, October 2–3, 2003, Argonne National Laboratory, Argonne, Illinois, Nuclear Energy Agency, Paris (2003).
17. P. C. SCHMITZ and L. S. MASON, *Space Reactor/Stirling Cycle Systems for High-Power Lunar Applications*, NASA-TM-103698, NASA Glenn Research Center, Cleveland, Ohio (1991).
18. H. K. RIVERS, P. O. RANSONE, G. B. NORTHAM, and F. A. SCHWIND, *Pistons and Cylinders Made of Carbon-Carbon Composite Materials*, U.S. Patent 6, 044,819. United States Patent and Trademark Office, Washington, D.C. April 4 (2000).
19. H. K. RIVERS, P. O. RANSONE, G. B. NORTHAM, and F. A. SCHWIND, *Carbon-Carbon Piston Architectures*, U.S. Patent 6, 116,202. United States Patent and Trademark Office, Washington, D.C., September 12 (2000).