

High-Temperature Liquid-Fluoride-Salt Closed-Brayton-Cycle Solar Power Towers

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Liquid-fluoride-salt heat transfer fluids are proposed to raise the heat-to-electricity efficiencies of solar power towers to about 50%. The liquid salt would deliver heat from the solar furnace at temperatures between 700°C and 850°C to a closed multireheat Brayton power cycle using nitrogen or helium as the working fluid. During the daytime, hot salt may also be used to heat graphite, which would then be used as a heat storage medium to make night-time operations possible. Graphite is a low-cost high-heat-capacity solid that is chemically compatible with liquid fluoride salts at high temperatures. About half the cost of a solar power tower is associated with the mirrors that focus light on the receiver, and less than one-third is associated with the power cycle and heat storage. Consequently, increasing the efficiency by 20–30% has the potential for major reductions in the cost of electricity. Peak temperatures and efficiencies of current designs of power towers are restricted by (1) the use of liquid nitrate salts that decompose at high temperatures and (2) steam cycles in which corrosion limits peak temperature. The liquid-fluoride-salt technology and closed Brayton power cycles are being developed for high-temperature nuclear reactors. These developments may provide the technology and industrial basis for an advanced solar power tower. [DOI: 10.1115/1.2710245]

Keywords: solar power tower, liquid fluoride salt, Brayton cycle, graphite heat storage

1 Introduction

The central challenge for solar power towers is economics. About half the cost [1] of a solar power tower system is associated with the sunlight collection system, which focuses sunlight on a high-temperature furnace. The remainder of the cost is associated with absorbing that heat and converting the heat into electricity. The sunlight collection system is almost independent of the power conversion system; thus, improvements in power-conversion-system efficiency can have a major impact on total power plant costs per unit of electricity that is generated.

It is proposed to replace molten-nitrate-salt coolant systems with liquid-fluoride-salt coolant systems and thus make it possible to increase peak salt coolant temperatures from ~565°C to between 700°C and 850°C. Increasing the peak coolant temperatures and using a higher-temperature closed-Brayton-power cycle have the potential to increase heat-to-electricity efficiency by 20–30% with an equivalent reduction in capital costs. The fluoride salts are thermodynamically stable (i.e., they do not decompose upon heating) and have atmospheric boiling points above 1000°C. The basis for using liquid fluoride salts and closed-Brayton-power cycles in solar power towers is outlined, including: (1) a general system description of a solar power tower; (2) development of liquid-fluoride-salt coolants and Brayton-power cycles for high-temperature nuclear reactors that are applicable to solar power towers; (3) the status of liquid-fluoride-salt technology; (4) a discussion of the graphite storage media; and (5) a description of closed-Brayton-power cycles.

2 Power Tower System

The solar power tower system (Fig. 1) consists of several major subsystems as follows:

1. *Heliostat field*: This focuses the sun's rays on the tower;

2. *Tower and solar receiver*: The tower contains the solar receiver, which absorbs the sun's rays and heats a fluid;
3. *Heat storage*: To continuously provide heat to the power system at night or under cloudy conditions, a heat storage device is required. For a system using fluoride salts, the preferred heat-storage medium is graphite. Graphite is a relatively low-cost high-temperature heat sink that is chemically compatible with the salt. While the salt could be used to store heat, it is more expensive than graphite. When the solar power tower is operating with thermal output that exceeds the requirements for the power conversion unit, hot salt flows downward through channels in the graphite, heats the graphite, and returns to the power tower. When the solar power tower is not operating and heat is required, salt flows upward through coolant channels in the graphite, is heated, and then flows to the power conversion unit;
4. *Power conversion*: Hot salt from the power tower or graphite heat storage provides the heat to a closed multireheat Brayton-power cycle that converts the heat to electricity. The working fluid may be nitrogen or helium; and
5. *Salt storage tank*: The salt storage tank provides space to store the salt when the power tower is shut down for an extended time. Typical salts freeze between 350°C and 500°C. The high volumetric heat capacity of the salt and the thermal storage provide a very large thermal inertia that prevents freezing; however, for protracted outages the system must be drained before cooling to low temperatures.

The same technologies can be used for solar power towers and parabolic trough systems. However, the higher-temperature fluoride coolant salts also have higher melting points relative to traditional nitrate salts. The complications of avoiding salt freezing in equipment are significantly less in a solar power tower than in a parabolic trough system; thus, solar power towers would be the initial application of the technology.

Contributed by the Solar Energy Engineering Division of ASME for publication in the JOURNAL OF SOLAR ENERGY ENGINEERING. Manuscript received May 3, 2005; final manuscript received July 8, 2006. Review conducted by Robert Pitz-Paál.

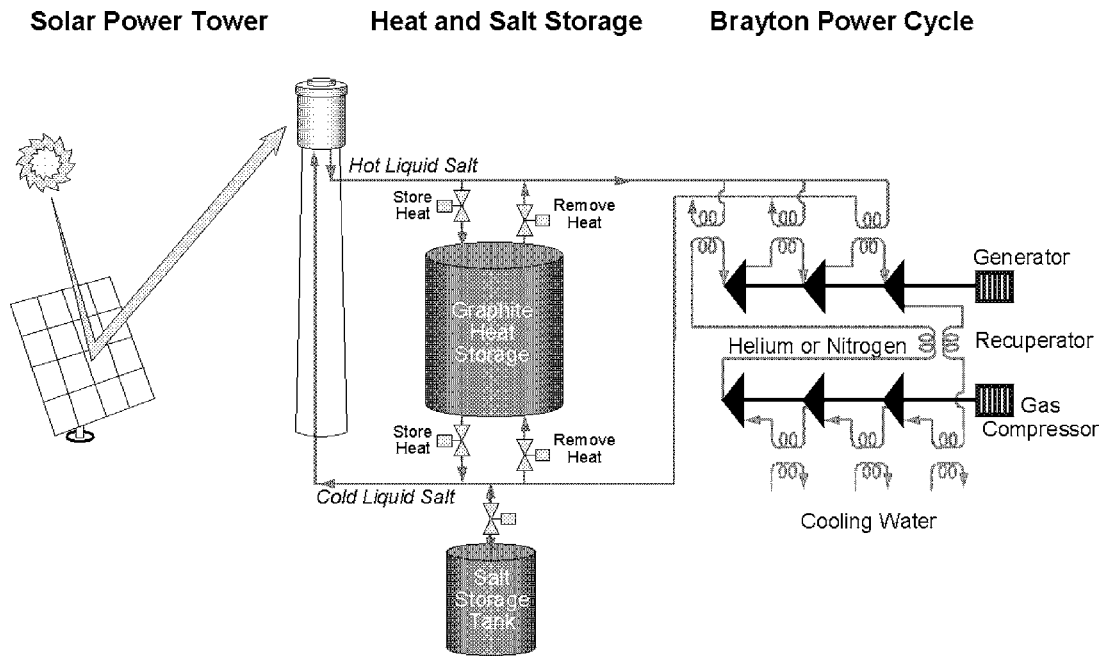


Fig. 1 Solar power tower with liquid-salt heat transport system, graphite heat storage, and Brayton power cycle

3 Technology Basis

The nuclear power industry is developing fluoride-liquid-salt heat-transport systems and closed-Brayton-power cycles for use in high-temperature reactors. The temperatures are in the same range of temperatures as required for solar power towers. These developments provide the basis for consideration of an advanced solar power tower.

Liquid fluoride salts have been used on a large industrial scale for a century. Since the 1890s, essentially all aluminum has been produced by the Hall process. In the Hall process, aluminum oxide is dissolved in cryolite (a sodium-aluminum-fluoride-salt) at $\sim 1000^{\circ}\text{C}$ in a graphite-lined bath. Electric current is passed through the salt solution to produce aluminum metal.

In the 1950s, at the beginning of the Cold War, the United States launched a large program to develop a nuclear-powered aircraft. Molten salt reactors (MSRs) were to provide the very-high-temperature heat source, with the heat transferred to a jet engine via an intermediate heat-transport loop. In a MSR, uranium fuel is dissolved in a liquid fluoride salt. In the 1960s and 1970s, the MSR was investigated as a thermal-neutron breeder reactor [2]. Fluoride salts were chosen for these applications because of their high-temperature heat transfer and nuclear characteristics. At operating temperatures, these salts have heat transfer properties similar to those of water. However, the boiling points are above 1000°C , which allows low-pressure operations.

Two experimental reactors were built and successfully operated. The Aircraft Reactor Experiment, the first MSR, was a 2.5 MW (t) reactor that was operated for 1 month in 1954 at a peak temperature of 860°C , used a sodium-zirconium-uranium fluoride salt, and proved the principle of a 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. In addition, test loops with liquid fluoride salts were operated for hundreds of thousands of hours and materials of construction were code qualified to 750°C and expected to exhibit a 30 year lifetime. Over 1000 technical reports were produced. These billion dollar programs were ultimately ended but developed the technology base from materials to operations for use of liquid fluoride salts as coolants.

In the last decade, there has been renewed interest in high-temperature reactors cooled with helium or liquid salts. This renewed interest is a consequence of two factors.

3.1 Brayton Power Cycles. For almost a century, the only practical method to convert heat to electricity on a large scale was the steam turbine which historically operated with peak temperatures of $\sim 550^{\circ}\text{C}$ (recent advances are increasing this temperature). There was little incentive to develop high-temperature power reactors because there was no efficient practical method to convert the high-temperature heat to electricity. The last decade has seen the development and deployment of large, efficient, low-cost high-temperature Brayton power systems that can operate at temperatures in excess of 1000°C . While these utility Brayton cycles burn natural gas or oil, the base technology and design methodologies now enable the design of Brayton power cycles to efficiently convert energy from other high-temperature heat sources to electricity [3].

The advances in Brayton power cycles have resulted in new reactor development programs in the United States, France, Japan, South Korea, and South Africa to develop high-temperature reactors using helium cooling [4,5] or salt cooling [6,7] with closed Brayton-power cycles using helium or nitrogen. This new interest is partly driven by the potential for lower costs because of higher plant efficiency and the potentially lower capital costs of Brayton power cycles compared to traditional steam cycles [8]. In closed Brayton power cycles, the entire power cycle operates at relatively high pressure that implies smaller equipment than for steam cycles with their large low-pressure low-power-density turbines. Japan has recently begun operation of its new 30 MW(t) high-temperature test reactor and is developing a closed-helium-Brayton power plant [9] with a 300 MW(e) output. The first such demonstration reactor with a closed-Brayton helium cycle (165 MW(e)) is projected to begin operation in South Africa [10] in 2011. Power systems are being considered with peak temperatures between 750°C and 950°C .

3.2 Hydrogen Production. The U.S. Department of Energy Nuclear Hydrogen Initiative, a component of a presidential hydrogen initiative, has as its goal the production of hydrogen using methods that require high-temperature heat and water as inputs.

Table 1 Characteristics of coolants and graphite^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)	$\nu \cdot 10^6$ (m ² /s)
LiF–NaF–KF (46.5–11.5–42)	454	1610 (est)	2020	1.89	3820	~0.6–1	1.4
NaF–NaBF ₄ (8–92)	385	700 ^b	1750	1.51	2640	0.5	0.5
NaNO ₃ –KNO ₃ ^c (66.3–33.7)	221	<i>c</i>	1772	1.53	2710	0.53	
Sodium	97.8	883	790	1.27	1000	62	0.25
Lead	328	1750	10,540	0.16	1700	16	0.13
Helium (7.5 MPa)			3.8	5.2	20	0.29	11.0
Water (7.5 MPa)	0	290	732	5.5	4040	0.56	0.13
Graphite			1700	1.9	3230	200	

^aApproximate physical properties at 700°C and 1 atm, except for water and nitrate salts. Pressurized water data are shown at 290°C for comparison. Nitrate salt data are shown at 500°C. Salt compositions are in mole %. Nomenclature: ρ is density; C_p is specific heat; k is thermal conductivity; and ν is viscosity. Additional references: Refs. [12–14].

^bAbove 700°C the system must be pressurized; however, the salt components do not decompose.

^cThis mixture has a composition by weight of 60% NaNO₃ and 40% KNO₃. It begins to decompose somewhat above 600°C.

These systems require that large quantities of heat be transported from high-temperature nuclear reactors to hydrogen production plants. Liquid fluoride salt heat transport loops are the leading technology to meet this requirement [11]. The requirements for transferring heat from a high-temperature nuclear reactor to a hydrogen plant are similar to those for transferring high-temperature heat from a solar receiver to an electric generator on the ground.

This work has major implications for closed Brayton cycle, salt-cooled solar power towers. Solar power towers were developed by combining heliostats and solar receiver designs with existing nitrate salt coolant and steam-turbine technologies. The high-temperature nuclear reactor programs over the next decade will create a technological and industrial basis that may make possible a relatively low-cost development program for a high-temperature liquid fluoride salt closed Brayton cycle solar power tower. Manufacturing and economic assessments of these technologies for high-temperature reactors will provide within the next several years the basis for a credible economic assessment of such an advanced solar power tower.

4 Liquid-Fluoride-Salt Coolants

4.1 Characteristics. Liquid fluoride salts are excellent high-temperature coolants. Table 1 shows the properties of several liquid fluoride salts and other coolants, including the typical nitrate salt used in current power tower designs. 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. Many other properties, such as vapor pressure, density, thermal expansion coefficient, and viscosity can also be modified with the appropriate salt mixture to produce more desirable qualities. For example, the lithium-sodium-

potassium fluoride salt (LiF: 46.5%, NaF: 11.5%, KF: 42%) has a melting point of 454°C, whereas LiF has a melting point of 845°C, NaF has a melting point of 992°C, and KF has a melting point of 856°C. With some three- and four-component mixtures, it is possible to reduce melting points to ~300°C. At operating conditions, the thermophysical properties of liquid salts are similar to those of room temperature water except for the very low vapor pressure of many liquid fluoride salts.

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. Liquid fluoride salts have been injected into water with no violent reactions (steam explosion or chemical reaction). There are several candidate salts. The selection of a specific fluoride-salt depends upon the particular application and requirements [12]. Solar power towers do not have nuclear requirements (such as low nuclear cross sections for capture of neutrons); thus, a wider choice of fluoride-salts exists for such applications. Table 2 lists some of the candidate salts. Additional studies will be required to define the preferred salt for solar power towers.

To avoid corrosion, liquid-fluoride-salt coolants must be thermodynamically stable relative to the materials of construction; that is, the materials of construction must be chemically noble relative to the salts. This requirement is necessary because fluoride salts are fluxing agents that rapidly dissolve protective layers of oxides associated with many metals, thus limiting the choice to highly thermodynamically stable salts if metal alloys of construction are to be used. Table 3 shows candidate fluorides suitable as components of a liquid salt coolant and their thermodynamic free energies of formation. The more negative the free energy of formation, the more stable the chemical compound. For example, if

Table 2 Physical properties of selected liquid salts

Salt (mol%)	Form. wt. (g/mol)	Melting pt. (°C)	Density	700°C heat capacity (J/g°C)	Viscosity	Thermal cond. (W/cm°C)
			(g/cm ³)		(cP)	
LiF–NaF–KF (46.5–11.5–42)	41.2	454	Alkali–Fluorides (IA) 2.53–0.00073 × T	1.89	0.04 exp(4170/T)	0.006–0.01
NaF–ZrF ₄ (50–50)	104.6	510	Alkali + ZrF ₄ 3.79–0.00093 × T	1.17	0.071 exp(4168/T)	~0.01 (est)
NaF–KF–ZrF ₄ (10–48–42)	102.3	385	3.45–0.00089 × T (est.)	1.09 (est.)	0.061 exp(3171/T) (est.)	~0.01
Li–NaF–ZrF ₄ (42–29–29)	71.56	460	3.37–0.00083 × T Fluoroborates	1.46	0.0585 exp(4647/T)	~0.01
NaF–NaBF ₄ (8–92)	104.4	385	2.252–0.000711 × T	1.51	0.0877 exp(2240/T)	~0.005

Table 3 Thermodynamic stability of components of liquid salts and structural materials

Constituent	Free energy of formation at 1000 K (kcal/mol°F)
LiF	-125
MgF ₂	-113
NaF	-112
RbF	-112
KF	-109
BeF ₂	-104
ZrF ₄	-94
¹¹ BF ₃	~-95
AlF ₃	-90
CrF ₂	-75.2
FeF ₂	-66.5
NiF ₂	-55.3
MoF ₆	-50.9

lithium metal was mixed with any other fluoride salt in the table, the lithium would preferentially form the fluoride salt and convert the other fluoride salt to a metal.

The general rule to ensure that the materials of construction (chromium, nickel, iron, etc.) are compatible 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). The corrosion strategy is the same as that used in sodium and other liquid-metal systems where the materials of construction are noble relative to metallic sodium.

In high-temperature systems, various temperature-induced mechanisms for corrosion exist 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 with adverse consequences on the strength of the metals. Parallel mechanisms have been seen in liquid-salt test loops. In salt systems, the primary concern has been the selective chemical transport of chromium (a constituent of many alloys) from hot to cold locations in heat transfer systems by multivalent cations due to a redox equilibrium that is temperature dependent. For most high-temperature alloys of interest, chromium is the least thermodynamically stable element among the materials of construction. Because these salts were originally developed for nuclear applications, most of the corrosion studies examined this corrosion mechanism with uranium ($2\text{UF}_4 + \text{Cr} = 2\text{UF}_3 + \text{CrF}_2$) as the multivalent cation.

This and other corrosion mechanisms were studied and resulted in the development of an optimized alloy, Hastelloy-N, as a material of construction. This alloy offers very good corrosion resistance and is code qualified to 750°C. Static corrosion tests have identified traditional high-temperature alloys for operation to higher temperatures; however, these higher temperature materials have not been qualified for use in salt systems [13].

These classes of corrosion mechanisms can be reduced or eliminated by: (1) using alloys or materials that do not have constituents that corrode; or (2) operating under chemically reducing conditions. As in nuclear systems, thick-wall tanks and piping would use lower-cost alloys for strength with high-nickel-alloy cladding for corrosion protection. Under chemically reducing conditions, chromium fluoride has an extremely low solubility, which limits chromium transport. Higher-temperature (815°C) test loops [13] indicate low corrosion rates with liquid fluoride salts when high-purity salts are prepared and maintained in a pure state.

Three classes of salts are candidates for a solar power tower. They are as follows:

1. *Alkali-fluorides*: The leading candidate is LiF-NaF-KF

(46.5–11.5–42). This is a highly stable, well-understood nontoxic fluoride salt;

2. *Fluoroborates*: The fluoroborates are the lowest-cost salts. They have many desirable characteristics; however, the cover gas does contain BF₃; and
3. *Alkali+zirconium fluorides*: The leading candidate is NaF-KF-ZrF₄, which has a melting point below 400°C.

4.2 Alternative Salts. The other potential liquid coolants at these temperatures are chloride salts. Selected chloride salts have somewhat lower melting points (355°C for LiCl-KCl) than some of the fluoride candidates and potentially lower costs. However, there are added challenges [14].

1. *Knowledge base*: The knowledge base for the use of liquid chlorides as coolants is much less extensive than that for fluorides;
2. *Thermodynamic stability*: It is believed that the thermodynamic driving force for corrosion in chloride systems is somewhat higher than that in fluoride systems. The difference in free energies between salt constituents and metallic corrosion products is less for chlorides than fluorides. The experience with fluoride systems suggests that it may be possible to develop chloride systems with low corrosion rates; however, there is little information on these systems in the context of high-temperature heat transport systems; and
3. *Purity*: Low corrosion rates in liquid salt systems are generally obtained with low impurity levels (similar to experience in water systems). While practical methods have been developed to purify fluorides, appropriate methods for chloride salts are not yet demonstrated [15].

5 Graphite Heat Storage

Solar towers require storage of heat to provide power at night. Heat can be stored in a wide variety of media with an indirect system where heat in the fluoride salt is transferred to a secondary heat transfer fluid via a heat exchanger and the heat storage media is in the secondary system. Alternatively, the salt can be used to directly heat graphite, a material that has relatively low costs and is compatible with liquid fluoride salts. The high mass and volumetric heat capacity of graphite (Table 1) provide an efficient storage medium. For a century, the aluminum industry has used very large graphite baths at ~1000°C to contain fluoride salts that were used to dissolve and electrolyze bauxite into aluminum metal. Laboratory tests have demonstrated compatibility of fluoride salts and graphite to 1400°C [16].

In the nuclear industry, many types of graphite reactors have been built consisting of stacked graphite blocks with cooling channels and fuel. Many of these reactors have diameters exceeding 16 m. This experience provides the starting point for the design of large graphite heat-storage systems for solar power towers.

The cost of high-quality machined electrolytic graphite is less than the cost goals (\$40/kWh) for heat storage in power towers [1]. Because the proposed 700 to 850°C temperatures are significantly lower than the traditional temperatures for other graphite applications, the potential exists for the use of significantly lower-cost graphites for this application.

6 Brayton Power Cycles

Brayton power cycles are the enabling technology for high-temperature solar power towers with peak coolant temperatures between 700°C and 850°C. The ongoing development of closed Brayton power cycles [3] for high-temperature nuclear reactors may provide the base technology for use in solar power towers. Several factors constrain the peak salt temperature as follows:

1. *Turbine efficiency*: For efficient Brayton power cycles, the helium or nitrogen temperatures should be above 650°C. This implies salt temperatures near 700°C;
2. *Power tower*: As power tower temperatures increase, in-

Table 4 Helium-Brayton power system design parameters for 600 MW(t) solar power tower

	Case 1	Case 2	Case 3	Case 4
Primary max./min. temperature (°C)	700/450	800/550	750/500	850/500
Turbine inlet/outlet temperature (°C)	670/430	770/530	720/480	820/480
Compressor inlet/outlet temperature (°C)	35/97	35/90	35/94	35/116
System pressure (MPa)	10	10	10	10
Numbers of turbines and compressors	3/6	3/6	3/6	2/4
Helium mass flow rate (kg/s)	157	156	156	165
Cycle pressure ratio	11.0	8.42	9.55	7.65
Pressure loss fraction	0.06	0.06	0.06	0.04
Overall cycle efficiency	0.463	0.517	0.492	0.504
Electrical power (MW)	278	310	295	302
The ratio of recuperator power to electrical power	0.93	1.1	1.0	0.98
Specific helium inventory (kg/MW(e))	10.7	10.2	10.4	9.5

creased radiation out of the solar furnace occurs. This begins to become important above 700°C [17] and a major consideration as temperatures approach 1000°C. Peak coolant temperatures should be significantly below this level to maximize efficient furnace design; and

3. **Materials:** Practical industrial high-temperature metals have temperature limits near 850°C. More exotic materials are available; however, major development requirements are associated with their use. For turbines, actively cooled turbine blades with gas cooling inside the blades can be used to higher temperatures. This enables the turbine blade to be at a much lower temperature than the gas and is standard industrial practice for turbine-gas temperatures above 850°C. However, there are significant heat losses to the turbine-blade cooling systems that degrade efficiency. Furthermore, in closed Brayton power cycles there are significant incentives to use helium and helium-nitrogen gas mixtures because the improved heat transfer characteristics of these gases reduces the sizes of heat exchangers. However, the same working gases are more efficient at removal of heat from actively cooled turbine blades. This implies additional heat losses and efficiency penalties with activity-cooled turbine blades. The practical implication is that the added complexity and efficiency penalties produce little incentive to go to higher temperatures unless the peak fluid temperatures are 100–200°C higher than these metallurgical limits and the gains in efficiency exceed the additional costs.

Planned and midterm (2008) solar-power-tower steam cycles [1] have efficiencies between 38% and 41.4%. Advanced solar-power-tower plants using *new* technologies (high-temperature (650°C) supercritical steam cycles systems and nitrate salts with oxygen overpressure to avoid decomposition) have been proposed with efficiencies up to 45.6%. Efficiencies of ~50% are expected for higher-temperature multireheat helium Brayton cycles. Efficiency is defined as the fraction of the heat collected by the solar furnace that is converted into electricity. Table 4 provides the details of four possible closed helium Brayton cycles for different combinations of inlet and outlet salt temperatures. For each of these cases, it is assumed that the thermal input is 600 MW. This size matches that of a large solar power tower and is equal in size to the closed Brayton power systems using helium and nitrogen that are being developed for high-temperature nuclear reactors. For nuclear reactor applications, both helium and nitrogen are being considered as the working fluid for closed Brayton power cycles. Either is applicable to a solar power tower. The preferred option will depend upon the results of ongoing tests and studies by turbine manufacturers [10].

The high efficiencies are a consequence of the high temperatures and multireheat Brayton power cycles that approximate ideal Carnot cycles. In these cycles (Fig. 1), the high-pressure helium is heated to its peak temperature and passes through a turbine, pro-

ducing work. This process is repeated several times, with the gas pressure being further reduced as it passed through each subsequent turbine. The lower-pressure gas goes through a recuperator and is cooled. The gas is then passed through a series of compressors and is cooled before each stage and then reenters the recuperator and is reheated and sent back to the turbines. One important operational characteristic of these systems is that they can be used for electrical load followed by changing the gas inventory (pressure). This approach enables the Brayton cycle efficiency to remain approximately constant over a wide range of power outputs.

The high-temperature Brayton cycles create economical options for dry cooling with their higher rejection temperatures, an important advantage in arid locations. Dry cooling will result in some loss of efficiency. Initial assessments indicate that the capital costs of the closed Brayton power systems per unit power output may be less than for steam cycles [8]. This is a consequence of the much higher power density (smaller equipment) in closed Brayton power cycles because of the higher minimum operating pressures.

7 Observations and Conclusions

Economics is the central challenge for solar power towers. Because the largest fraction of the capital cost is associated with the focusing solar mirrors, strong incentives exist to raise peak coolant temperatures and thus increase heat-to-electricity efficiencies. Major improvements in solar-power-tower performance are potentially possible by using high-temperature efficient Brayton cycles, low-pressure high-temperature high-heat-capacity liquid-fluoride-salt coolants, and graphite heat storage.

The nuclear power industry has expanding programs underway to develop high-temperature reactors with closed Brayton power cycles and liquid-salt heat-transport systems. Within the next decade, these development activities are expected to create: (1) much of the technology and industrial infrastructure required for a liquid-salt-cooled closed Brayton cycle solar power tower; and (2) sufficient information to enable credible cost estimates for many of the systems. For a liquid-salt-cooled closed Brayton cycle solar power tower, there are significant technical challenges including many interface issues that have not been addressed. However, the potential economic gains combined with these advancing technologies warrant serious consideration of such an advanced solar power tower.

Acknowledgment

This paper has been authored by a contractor of the U.S. Government Oak Ridge National Laboratory, UT-Battelle, LLC, for the U.S. Department of Energy under Contract No. 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.

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