

## Review Article

# Lead-Cooled Fast Reactor Systems and the Fuels and Materials Challenges

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Anticipated developments in the consumer energy market have led developers of nuclear energy concepts to consider how innovations in energy technology can be adapted to meet consumer needs. Properties of molten lead or lead-bismuth alloy coolants in lead-cooled fast reactor (LFR) systems offer potential advantages for reactors with passive safety characteristics, modular deployment, and fuel cycle flexibility. In addition to realizing those engineering objectives, the feasibility of such systems will rest on development or selection of fuels and materials suitable for use with corrosive lead or lead-bismuth. Three proposed LFR systems, with varying levels of concept maturity, are described to illustrate their associated fuels and materials challenges. Nitride fuels are generally favored for LFR use over metal or oxide fuels due to their compatibility with molten lead and lead-bismuth, in addition to their high atomic density and thermal conductivity. Ferritic/martensitic stainless steels, perhaps with silicon and/or oxide-dispersion additions for enhanced coolant compatibility and improved high-temperature strength, might prove sufficient for low-to-moderate-temperature LFRs, but it appears that ceramics or refractory metal alloys will be necessary for higher-temperature LFR systems intended for production of hydrogen energy carriers.

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## 1. ENERGY SOURCES IN THE 21ST CENTURY

The demand for affordable energy will continue to grow based on both population growth and increases in per capita energy usage. Conservation will slow but not obviate the need for additional production. The following trends are likely to drive the manner in which energy is used in the 21st century.

- (i) Technological innovation and market deregulation will increase the options for both producing and transmitting energy.
- (ii) Consumers will continue to demand energy sources that are increasingly environmentally benign.

Currently, the energy production and distribution system is quite simple. For transportation, the primary method to supply consumers with energy is to move oil from the ground through refineries to distribution sites near the consumer. For heating, oil and natural gas are moved directly to the consumer for use, or electricity is generated at remote sites and transmitted to the user. Other major power needs are met by electricity that is generated remotely and distributed to the user via power lines. For most users, production and con-

sumption are separate functions. Households and businesses rarely create and supply energy; they just use energy.

Three innovations are likely to change this dynamic. The first is the addition of hydrogen as an energy storage medium and energy carrier, operating in synergy with electricity. The addition of hydrogen as an energy carrier provides additional flexibility to the energy distribution system. Electricity generated during periods of low fuel cost will be usable in manufacturing hydrogen. The hydrogen can be used directly in transportation-based fuel cells or can be converted back to electricity via fuel cells at a later time when it is economically viable. Excess electricity or hydrogen can be used to produce potable water. Either fuel can be transported to the near user or energy can be transported via electricity or hydrogen.

The second innovation is digital technology. As each person on the energy grid obtains the ability to monitor, in real time, the use or transmission of energy from their site, they can decide if it is more economical to use energy or sell it [1].

The third innovation is the increased use of distributed production methods. Through the use of hydrogen-driven fuel cells, solar panels, wind generation, or other technologies, users will have the ability to manufacture their own energy and to sell excess energy. A user's decision on obtaining

production technology and when to distribute rather than use energy will be based on cost relative to traditional central station distribution methods.

In an age where terrorism poses a potential threat to energy production and distribution, energy sources may increasingly be hardened against attack or distributed to minimize the repercussions of a single attack.

Current nuclear energy plants are large base load units for electricity production. The nuclear plants are usually sited remotely from population centers, and the electricity generated is distributed via high-voltage transmission lines. This technology was developed during a time when electricity production was part of a regulated market and nuclear plants were essentially designed in a manner similar to coal plants. The nuclear reactor replaced the boiler in a Rankine cycle coal-driven steam system. Fuel is used once through the system that throws away a significant portion of the energy content of the fuel and does not attempt to recycle waste products. Compared to other energy production technologies, nuclear reactors produce far less waste per unit of energy production, but the waste is more toxic per unit mass of waste. The cost of operating a nuclear plant is low compared to other sources, but the capital cost to build the plant is larger than other technologies.

While these central station plants are likely to continue to be a significant portion of the 21st century energy distribution systems, the changes in the manner in which energy is produced and distributed allow for the possibility that nuclear systems can also be used in a different manner. Designers of nuclear systems now need to consider

- (i) the ability to produce hydrogen,
- (ii) smaller distributed plants,
- (iii) making the plants increasingly environmentally benign,
- (iv) the ability to load-follow to match production with need.

Lead-cooled fast reactor (LFR) concepts address some or all of these objectives, with advantages for innovative use of nuclear technology. The last three considerations (small plants, reduced environmental footprint, and load following) can be also achieved using other fast reactor concepts (sodium, salt, or gas cooling). Because of the higher boiling temperature of lead and lead-bismuth, lead-alloy systems are better positioned to link to high-temperature hydrogen production than the sodium-cooled systems.

In this paper, the authors (who have each worked for a short time with the US Department of Energy's LFR effort) attempt to provide a dispassionate status of LFR technology which is proposed or under consideration in recent years. Emphasis is placed on materials of construction, as issues with materials constitute the biggest technological barrier to realizing these systems. This paper will briefly describe the key features of three different LFR concepts, each designed for a separate function: a small modular system (STAR), a medium-sized sustainable electricity production system (BREST), and an actinide burner (INEEL/MIT design). These are only examples of potential LFR technology and not an exclusive list. As an example, recently, the

European Union has proposed the ELSY concept, which is a 600 MWe lead-cooled pool-type system that lowers the operating temperature to allow a faster deployment using mainly commercially available technology [2]. The only industrial experience with lead-alloy cooling comes from the Soviet Union submarine program. Following the description of each plant, a section on critical fuels and materials R&D needs is presented.

## 2. LFR CONCEPTS

LFR systems are Pb or Pb-Bi alloy-cooled reactors with a fast neutron spectrum, with fuel flexibility supporting options for use within a closed fuel cycle. Technology options include a range of plant ratings, including a long refueling interval transportable system ranging 50–150 MWe, a modular system ranging 300–400 MWe, and a large monolithic plant at 1200 MWe. These options also provide a range of energy products.

In designing fast reactor systems, lead coolant provides two advantages over sodium as a coolant. First, the boiling point of lead or lead-bismuth eutectic (LBE) is much higher providing greater safety margins and the ability to operate at higher temperature. Second, lead alloys do not react exothermically with water and air as sodium does. At the same time, lead has certain drawbacks relative to sodium. Lead alloys are more corrosive to structural steels than sodium. The high density and low heat conductivity, as well as the erosion of protective oxide layers, reduce possible lead flow rates, thus limiting the total allowed power density and the doubling time to breed plutonium. Finally, the high melting temperature of lead requires special efforts to keep the lead from freezing in the primary circuit.

Lead-bismuth has a lower melting point than lead, and using lead-bismuth simplifies prevention of primary coolant freezing. For this reason, LBE was chosen as the coolant in the Soviet submarine program. For a large-scale deployment of nuclear energy, the Russians desire to move away from LBE towards lead because of the higher cost of LBE (primarily due to the cost of Bi) and due to the higher radioactivity levels associated with polonium production from bismuth [3].

Lead-cooled systems, like all fast spectrum systems, can be used as net breeders or net burners of fissile material. The BREST system was designed to operate in a sustainable manner with no net production of minor actinides. This system is briefly described in the next section.

### 2.1. BREST

As a means of developing a nuclear power system that took full advantage of the capability of nuclear energy while addressing the issues of safety, economics, proliferation resistance, and waste disposal, the Russians designed a series of intermediate spectrum lead-cooled plants ranging in size from 300 to 1200 MW. The 300 MWe version was described in the *White Book of Nuclear Power* [3]. The description below follows the White Book.

The 300 MWe BREST reactor was designed such that each core loading lasts roughly five years. The coolant inlet

temperature is 417°C and the core outlet temperature is 537°C. Although supplied with pumps, the system relied heavily on natural circulation flow with steam generators placed six meters above the core. The feedwater temperature of the supercritical steam-driven secondary system was maintained at 337°C to prevent primary system freezing. The secondary system is designed to produce power at 43% efficiency.

Mononitride fuel with stainless steel cladding and a lead-filled gap was chosen for the BREST system. A 900 mm gap was placed at the top of the fuel rod to collect released fission gasses. To prevent production of weapon-grade plutonium, BREST designers avoided the use of traditional uranium blankets and instead used a lead reflector that surrounded the entire core. The core used three separate radial zones, each with its own fuel loading, but with similar Pu content over the whole core. Within each zone, the diameter of the fuel rods differed to match fuel assembly power with flow. The three-zone system flattened the power distribution and maximized the temperature margins on the cladding. Fuel lifetime was limited by cladding performance rather than burnup.

The BREST design calls for the nitride fuel to be enriched in N-15 to prevent the buildup of C-14, which is a radiation hazard. The cost of enriching the nitrogen for the fuel was expected to be offset by 2.5% increase in  $k_{\text{eff}}$ , reducing the fuel inventory by roughly 10%.

To optimize fuel utilization and minimize waste, all minor actinides are assumed to be recycled in the BREST system. To maintain an equilibrium fuel loading, after each reprocessing step U-238 is added in equal amounts by mass of the fission products separated. The core was designed with a conversion ratio near 1 such that burnup of fissile isotopes was replaced by bred plutonium.

An advantage of lead coolant is that it provides shielding of reactor components against damage. In the BREST design, the shroud that separates the coolant inlet and outlet was only subjected to roughly 0.2 dpa/year, while the reactor vessel was limited to roughly 0.01 dpa/year.

The BREST system was designed to prevent inadvertent reactivity accidents from becoming significant by establishing a system such that the reactivity margin stays well below  $\beta_{\text{eff}}$ . Additional safety features to enhance stability include stabilizers to keep fuel assembly pitch unchanged in case the lead coolant level drops below its nominal level, and thermal expansion boosters that widen the gaps between fuel assemblies if the temperature exceeds its permissible level. To prevent gas bubbles from entering the core in a steam generator leak, possibly leading to reactivity addition accidents, the coolant leaving the primary pump first entered a “free level” above the core before traveling down through the steam generator. At this free level, any bubbles would be released into a large plenum above the core rather than traveling to the core itself.

Lead-cooled systems can also be used as actinide burners. The INEEL/MIT actinide burning design is described in the next section.

## 2.2. INEEL/MIT actinide burner

The Idaho National Engineering and Environmental Laboratory (INEEL) (which is now known as the Idaho National Laboratory) and the Massachusetts Institute of Technology (MIT) proposed three 300 MWe actinide burning lead-cooled reactor systems, each tailored for a specific design mission [4–6]. A major goal of these designs is to optimize the destruction of minor actinides from LWR fuel, and the concepts are designed to operate within a multirecycle fuel cycle. The three concepts are a fertile-free actinide burner for incineration of all transuranics from light water reactor (LWR) spent fuel (actinide burning reactor (ABR)), a fertile-free minor actinide burner for preferential burning of minor actinides working in tandem with LWRs or gas-cooled thermal reactors (minor actinide-burning reactor (MABR)), and an actinide burner with thorium fuel (actinide-burning reactor with thorium (ABRT)) aimed also at reducing the electricity generation costs through longer-cycle operation.

These three concepts operate using a lead-bismuth eutectic coolant with identical power ratings, 461°C inlet temperature, and 553°C outlet temperature. The secondary system is a supercritical carbon dioxide-cooled Brayton cycle. The reactor vessel, heat exchangers, pumps, decay heat removal strategy, and heat removal path for normal operation are identical for all three systems with differences only occurring in core design and number of control rods. Similar to the design philosophy of the integral fast reactor (IFR) concept [7], inherent safety features such as Doppler coefficient, fuel expansion, coolant density change, and radial expansion are designed into the INEEL/MIT designs. The INEEL/MIT designs improve the values for these parameters to manage the reduction caused by adding minor actinides to the fuel.

The ABRT uses reprocessed spent LWR fuel with the addition of U and Th as fertile nuclides. Sufficient U-238 is added to denature bred-in U-233 for proliferation resistance reasons. The ABR is fertile-free and uses reprocessed spent LWR fuel in a zirconium matrix. The MABR is fertile-free in a zirconium matrix with the actinide concentration being optimized for burning. Fuel design was based heavily on the experience of the IFR fuel development results. Metallic fuel was chosen, partly for safety reasons, under the belief that in an accident, the cladding would yield and allow fuel to be dispersed through the coolant, taking it away from higher-worth regions of the core and thereby preventing excess reactivity accidents. With a nitride fuel, the belief was that the cladding would break away during severe accidents before the nitride fuel would melt.

HT9 cladding was chosen based on the extensive radiation effects database. The cladding steady-state temperature is assumed to be limited by creep strength to 600°C and the transient temperature to 725°C by fuel cladding chemical interaction. The fuel-side constraints (e.g., fuel-cladding interdiffusion) were considered to be more restrictive than corrosion constraints on the coolant side of the cladding. A corrosion temperature limit of 650°C was based on Russian design assertions.

The reactor vessel in the concept is constructed of 316 stainless steel and has a maximum temperature of 430°C.

The size was set to ensure transportability by rail. The ABR uses an RVACS system for decay heat removal, similar to the IFR concept, with the gap between the pressure vessel and guard vessel being filled with lead-bismuth to improve heat transfer. Reactor power is ultimately limited by the decay heat removal ability of the RVACS. The pressure vessel is coupled to a guard vessel by a lead-filled gap, with the guard vessel being cooled by natural circulation atmospheric air. A modular design was chosen to reduce construction time and limit manufacturing variability.

During normal operation, forced circulation of the primary coolant is used. Forced circulation allowed for a smaller core with smaller temperature changes along the core. Coolant circulation uses the Russian dual free level approach to prevent reactivity insertions from gas entry into the core in a heat exchanger failure.

The recent focus of the US program is on sustainable (conversion ratio of one) transportable modular concepts. These modular systems will be described in the next section.

### 2.3. Modular systems

The secure transportable autonomous reactor (STAR), as well as other systems such as the encapsulated nuclear heat source (ENHS), is designed as a modular, sustainable energy system. The description below on the STAR reactor follows the description provided in [8]. Modular systems are targeted for use in countries or regions with a need for smaller power stations, delivering a variety of energy products including hydrogen, electricity, and potable water. In small countries where large additions of power to the grid are not needed, modular plants may be more appropriate. The plant design assumes that the generating company running the plant has no capability or desire for its own fuel cycle infrastructure. Cores are designed to operate for 20- to 30-year lifetime and then be replaced with a new “cartridge or battery” core.

Deployment of modular plants is envisioned as a means to shift investment decisions from the traditional economy-of-scale approach to an economy-of-mass-production approach. In the traditional economy-of-scale approach, a regulated utility could afford the risk in building a large capital cost plant because the rates at which electricity could be sold were assured, thus minimizing the economic risk. Under those conditions, a larger plant would usually produce a higher output per unit of capital cost, due to the economy of scale. The STAR vision considers increasingly deregulated electricity markets in which the financial risk is shifted from the electrical generating company to the nuclear system supplier. For this environment, a modular construction is attractive because it ensures that a revenue stream is made available to the plant owner as soon as possible.

The system is designed to use full actinide recycle, sending only fission products to the waste stream, using all minor actinides and keeping the minor actinides in a highly radioactive “self-protecting” fuel form. To obtain high coolant outlet temperature, uranium-plutonium-nitride fuel is selected allowing for higher fuel temperatures. Expanded cores are envisioned to be transported to a regional fuel cycle center where fuel is recycled. The recycle process would use elec-

trometallurgical recycle and remote vibrocompaction refabrication of the nitride fuel, similar to that deployed at engineering scale for oxide fuels at the Russian Institute of Reactors [9]. The large size of the core cassettes makes the fissile material diversion without detection difficult. To minimize the production of C-14 which, as a radioactive volatile species, would be difficult to control during recycle operations, the fuel will need to be enriched in nitrogen 15, adding cost to the fuel cycle.

The most ambitious STAR design is the STAR-H<sub>2</sub>, a 400 MWe natural-circulation-cooled system. This reactor is designed with a coolant outlet temperature of roughly 800°C and would drive a Ca-Br thermochemical process for hydrogen separation, a Brayton cycle for electricity production, and finally a bottoming cycle for potable water production. These cascading secondary systems are designed to optimize the use of the heat transferred out of the core. Lower-temperature STAR systems, along with the ENHS, are designed for electricity production and not hydrogen production. The STAR plant is designed to follow load on the grid.

The STAR systems are designed such that the secondary balance of the plant has no safety function. All safety functions are part of the core and hence the responsibility of the supplier company. This centralization of the safety function will improve plant operation across multiple platforms. The safety basis of the plant relies on natural feedback mechanisms rather than engineered control. The plants are located underground in an earthen berm to help isolate them from the colocated hydrogen plant. A final unique safety aspect of the modular systems is the desire to perform licensing of the design by test of a prototype reactor.

The plant size, with a power density being approximately the same as that of an LWR, was chosen such that the reactor vessel would be rail shippable. The plant size was also kept smaller than a typical LWR to allow energy suppliers with limited capital to finance the plant. Shipping occurs in large 200-ton casks with the fuel being encapsulated in frozen lead. This provides a measure of safety during transportation.

## 3. PROGRESS IN R&D

From a design perspective, the LFR provides a wide range of possibilities for delivering energy products in a sustainable manner. The modular systems, such as the STAR, hold promise to revolutionize the manner in which energy is distributed. Works in [10–12] provide good descriptions of the reactor design goals of the LFR. The success of the LFR depends on critical developments in fuels and materials technology. The following sections describe the state of fuels and materials development, with emphasis on materials development, as compatibility of materials with high-temperature Pb or Pb-Bi remains a key viability issue for the LFR concepts.

### 3.1. Fuels

Functionally, for the majority of reactor types and concepts, fuel rods and assemblies serve three major purposes: they configure the fissile material into a desired location in the

reactor core to allow for a controlled chain reaction and adequate heat removal; they provide a first safety barrier to the release of hazardous radionuclides; they provide a convenient means for handling or exchanging fuel to replenish or adjust reactivity in the core. Reactor designers and developers consider fuel performance relative to each of these three purposes when selecting or developing a fuel for a particular application. In general, properties of fast reactor fuel rods that have impact on performance include melting temperature (or other phase transition temperatures), chemical compatibility and interdiffusivity among fuel and cladding constituents, fission gas release and fission gas-driven fuel swelling, cladding strength and creep resistance, and geometric stability under irradiation. Specific requirements for fuel performance and characteristics often evolve from those considerations, particularly as safety-related equipment failures and malfunctions are addressed in safety analysis.

Selection of fuel type for LFR application begins with consideration of fuel types investigated for sodium-cooled fast reactor use. Fast reactor fuel development during the 1960s through the 1990s addressed several fuel types, including ceramic fuels selected for their high melting temperatures and high-temperature strength and metallic fuels selected for their high atom density, high thermal conductivity, and ease of fabrication. Of these, the most extensive work has been performed on mixed oxide, (U,Pu)O<sub>2</sub> fuel, and U-Pu-Zr metal alloy fuel.

Unfortunately, initial consideration of those fuel types suggests that each may not be well suited for LFR application. Published data indicate that U, Pu, and Zr each have some solubility in Pb and/or Bi [13], suggesting potential fuel dissolution into coolant if cladding were to breach. Actual dissolution kinetics and realized rates of dissolution in actual breached-cladding geometry might prove to be sufficiently benign so that metal alloy fuel can be considered for moderate-temperature core conditions (e.g., at temperatures similar to those used in sodium-cooled fast reactors). For example, UO<sub>2</sub> reacts with water at high temperature to form UO<sub>3</sub> in LWRs [14], and (U,Pu)O<sub>2</sub> reacts with sodium coolant to form less dense compounds that can further breach cladding [15]. However, these phenomena have been shown to be of little or no consequence, either because operating practices avoid conditions where the phenomena matter, or because the actual phenomena involved have little impact on radionuclide release into the coolant under normal operating conditions (e.g., [16]). For the case of oxide fuel, the most relevant properties to the current stages of concept development are that designers prefer higher atom densities, higher thermal conductivities, and lower heat capacities than those typical of oxide fuels. The high temperatures that would be associated with oxide fuels lead to a requirement to use more engineered safety features and less passive safety features [17]. For those reasons, LFR designers have preferred nitride fuels for various LFR concepts.

Nitride fuels have properties that are attractive for reactor designers, with thermal conductivities and atom densities closer to those for metal alloy fuels than for oxide fuels, and melting temperatures considerably higher than those for metal fuels [18, 19]. The single valence state for actinide ni-

trides provides good miscibility for the actinide nitride compounds, which facilitates fabricating pellets of uniform composition. UN fuel appears to have excellent irradiation stability in a fast spectrum [20] although the irradiation performance database for such fuels is not substantial [21]. An additional advantage of nitride fuels is the compatibility with lead, allowing lead bonding to be used in the fuel rod design. A potential disadvantage regarding mixed nitride fuels is the dissociation of the compounds at temperatures substantially lower than their congruent melting point if a nitrogen overpressure is not maintained [20]. This would be an issue for an overpower or loss-of-coolant event that would substantially raise fuel temperatures—events which designers hope to obviate with passive safety features in these lead or lead-bismuth systems.

### 3.2. Materials

#### 3.2.1. Material candidates

Materials for the LFR must meet three criteria:

- (1) adequate mechanical properties (strength, ductility, and toughness) over the range of operating temperatures, stresses, and doses;
- (2) adequate dimensional stability (resistance to void swelling and thermal and irradiation creeps) over the range of operating temperatures, stresses, and doses;
- (3) adequate corrosion, stress corrosion, and liquid metal embrittlement resistance under all operating conditions.

Materials of construction for a lower-temperature LFR like the BREST or INEEL burner design are likely to be metallic, specifically austenitic stainless steels or ferritic/martensitic steels.

Ceramic or refractory metals will be required for higher-temperature LFR designs like STAR-H<sub>2</sub>. The technological readiness for steels is much higher than for ceramic materials or refractory metals. A lower-temperature LFR could be ready for deployment much earlier than a high-temperature LFR, but with a much more limited set of capabilities. In high-dose components of lower-temperature LFRs, where void swelling limits materials performance, ferritic/martensitic steels will be used.

HT9 is a ferritic/martensitic steel that was developed for the power-generation industry in the 1960s and introduced into the US fast reactor and fusion materials programs in the 1970s. However, since that time, several improved ferritic/martensitic steels have been developed for the power-generation industry, offering significant improvements on HT9. Table 1 outlines the evolution of five generations of elevated-temperature steels over the last 60 years, beginning with 21/4Cr-1Mo steel (T22) (labeled as a zeroth-generation steel) in the 1940s [22].

HT9 is a first-generation steel with limited temperature capability relative to subsequent generations. The table also indicates that EM12, a duplex ferritic/martensitic steel, was investigated in detail in France for cladding and duct applications in fast reactors. Other first-generation steels were

TABLE 1: Evolution of ferritic/martensitic steels for power-generation industry.

Generation	Years	Steel modification	10 <sup>5</sup> h rupture strength (MPa)	Steels	Maximum use temperature (°C)
0	1940–60		40	T22, T9	520–538
1	1960–70	Addition of Mo, Nb, and V to simple Cr-Mo steels	60	EM12, HCM9M, HT9, HT91	565
2	1970–85	Optimization of C, Nb, and V	100	HCM12, T91, HCM2S	593
3	1985–95	Partial substitution of W for Mo and addition of Cu	140	NF616, E911, HCM12A	620
4	Future	Increase W and add Co	180	NF12, SAVE12	650

investigated for fast reactor applications as well in Germany (W. Nr. 1.4914) and the United Kingdom (FV448). All of these steels showed promise for the intended nuclear applications. However, since the 1970s, the steel and power-generation industries have improved on these steels, and for future nuclear applications like the LFR, the improved steels that replaced first-generation steels should allow the reactor designer a greater opportunity to achieve the elevated temperatures being desired in the new reactor designs. Because the fourth-generation steels contain cobalt, they are not suitable for nuclear use. Nonetheless, the use of third-generation ferritic/martensitic steels would allow an increase in operating temperature of roughly 50°C.

The Russian programs developed the steel EP823 specifically for use in lead-bismuth [23]. EP823 is a 12 Cr ferritic/martensitic steel with elevated silicon levels (1–1.3 wt%). No comparable US steel is currently being produced. Development of steels for the LFR may require the development of a similar alloy.

To obtain improved high-temperature creep strength from a ferritic steel, oxide dispersion-strengthened (ODS) steels have been produced and evaluated. These steels derive their elevated-temperature strength from a fine distribution of Y-Ti-O particles, as opposed to carbide precipitation in conventional elevated-temperature steels. Effective strengthening by carbides, which is the typical approach with application of many steels, is not possible above  $\approx 650^\circ\text{C}$ , as the carbides are not stable. Different oxide dispersion-strengthened steels have been developed in different alloy development programs [22]. The Japanese Atomic Energy Agency, for example, has developed advanced ODS steels with improved high-temperature strength [24] and they are currently being tested as fuel cladding in test assemblies being irradiated in Russia. In addition, they are manufactured by mechanical alloying/power metallurgy techniques that introduce further difficulties and expense. Joining of the steels being a challenge, due to segregation of the oxide particles to the surface of a molten weld zone, has yet to be adequately addressed. Therefore, although ODS steels offer the promise of a steel that can operate at 800°C, the steels are still in the early development stage and the performance database is limited.

An additional R&D path to improve creep strength of structural materials is grain boundary engineering [25]. Grain boundaries are altered to minimize the fraction of high-energy grain boundaries, thus improving creep strength. For this approach, the long-term stability of special boundaries must be proven.

Austenitic stainless steels are appropriately considered for lower-temperature and lower radiation dose LFR components. The austenitic stainless steels AISI 304 and AISI 316 have been used extensively in the light water reactor industry as well as the fast reactor development programs. The performance of 316 is superior and would be the leading candidate for austenitic components in an LFR.

Because austenitic and FM stainless steels are not adequate for the higher-temperature LFRs, ceramics, superalloys, or refractory metals are being considered. The existing data in these high-temperature materials are summarized in [26]. Silicon carbide composites are the leading candidate materials for high-temperature LFR components, based on development in the fusion materials programs. The technological readiness of SiC composites is much lower than austenitic or ferritic/martensitic steels, so high-temperature LFR development is a much longer-term prospect than for lower-temperature versions with high-dose radiation resistance, cost, and fabricability still being major open issues. Because of radiation embrittlement typical in Ni-base alloys and because of poor fracture toughness and poor oxidation resistance in refractory metals, the superalloys and refractory metals are not likely to be acceptable in LFRs without further breakthroughs.

### 3.2.2. Radiation stability

Significant radiation stability data under LFR conditions exist only for a limited number of metallic alloys and one ceramic composite. Extensive radiation stability databases exist for the austenitic stainless steels 304 and 316. Additionally, a Ti-stabilized, 316-like alloy named D9 was developed in the fast reactor programs to optimize the composition for maximizing swelling resistance. These alloys are expected

to have adequate radiation resistance for lower-temperature LFR components.

For high-dose metallic components where swelling resistance is important, ferritic/martensitic steels are the primary candidates in lower-temperature versions of the LFR. The US fast reactor cladding and duct development program created an extensive database on mechanical properties and dimensional stability under radiation for HT9, a 12 Cr steel with adequate swelling performance up to doses of roughly 200 dpa at temperatures of roughly 600°C. To use HT9 in an LFR, some additional research would still be required including verifying the ability to control corrosion in lead alloys, proving adequate resistance to fuel-cladding chemical interaction with minor actinide bearing fuels, establishing high-temperature properties under hypothetical accident scenarios, and ensuring adequate low-temperature (<400°C) fracture toughness. Additionally, there is a need to verify uniformity of composition and thermomechanical properties of cladding material from batch to batch and to establish quality assurance of material supply and fabrication and welding techniques. There is currently no supplier of HT9 in the United States.

T91, a 9Cr FM steel, has significant radiation test data and shows improved fracture toughness properties over HT9, but it does not have as extensive radiation performance database as HT9. The T91 alloy showed improved irradiation resistance compared to that of HT9, primarily because of the lower carbon concentration in T91 [22]. In particular, under irradiation conditions where HT9 develops an increase in the ductile-brittle transition temperature of 120–150°C, the modified 9Cr-1Mo develops a shift of only 52–54°C. Because of the lower chromium content, the corrosion resistance of T91 in lead alloys is expected to be inferior to HT9. Recent work has shown that the embrittlement of T91 correlates strongly with the available He produced in the material during irradiation, with DBTT reaching values above 100°C [27].

Only one irradiation experiment on the third-generation steels has been carried out [28]. In that experiment, two of the new commercial steels, NF616 and HCM12A, along with T91 were irradiated at 300°C in the mixed neutron spectrum of the high-flux reactor (HFR) in Petten, the Netherlands. For these experiments, the Charpy impact properties of T91 were superior to the new steels both before and after irradiation [28]. For this test, the modified 9Cr-1Mo developed a larger shift than normally observed. No ready explanation is available for these large increases in DBTT in T91 seen in this experiment. The authors suggested that temperature variations may have played a role, as the temperature on the specimens was said to vary from 265°C to 312°C.

Tensile properties indicated that the NF616 showed the least hardening followed by modified 9Cr-1Mo, and the HCM12A hardened the most. The presence of boron in the two new steels might be a factor, since in the mixed neutron spectrum of HFR, the thermal neutrons can transform the  $^{10}\text{B}$  in the steel to helium. Recently, ion irradiation studies on HCM12A at 400°C have shown HCM12A to have similar hardening to T91 [29]. Additionally, the grain boundary chromium concentration increases during radiation, leading

to potential corrosion susceptible regions adjacent to, but just off, the boundaries.

SiC-SiC composites have two critical issues relative to the radiation response [26]. The thermal conductivity decreases under radiation, and subsequent work is needed to determine whether a minimal acceptable thermal conductivity can be maintained under irradiation. Additionally, the material undergoes swelling at low temperature (~120°C) due to amorphization and at high temperature (~1000°C) due to void swelling. The swelling limit at high temperature is based on a limited dataset and will need to be further refined.

In summary, HT9 and 316 have the most complete radiation effects database of all LFR candidate materials. T91, JNC ODS, and SiC-SiC composites have some data, but they require additional radiation testing before being qualified for LFR use. Advanced FM steels have little radiation data. The priority radiation data needs for LFR application are for the following materials: ODS, SiC-SiC, and advanced FM steels like NF616 and HCM12A.

### 3.2.3. Corrosion, stress corrosion, and liquid metal embrittlement

As part of the Russian lead reactor design programs, significant development was performed to understand and deploy materials and coolant chemistry control schemes for lead-bismuth eutectic cooled systems [30–45]. Outside Russia, the technological readiness level of lead-alloy nuclear coolant technology is at an earlier development stage, but the partial knowledge of the Russian experience is available to the Western Technical Community, and it has been a major driver in corrosion research. Over the past ten years, a significant amount of corrosion R&D has occurred in Europe, Asia, and the US.

Russian LBE coolant technology relies on active control of the oxygen thermodynamic activity in LBE to control corrosion and coolant contamination. Within this framework, a series of structural materials was developed and tested in Russia for enhanced corrosion resistance and acceptable lifetime. The operating temperature is below 550°C, with fuel cladding temperature below 650°C. The LBE flow velocity design limit is about 2 m/s to prevent the onset of erosion.

The oxygen control technique, when properly applied, leads to the formation of “self-healing” protective oxide films on the surfaces of the materials in contact with lead-alloys. This is effective because the base element (typically Fe) and the alloying elements (Cr, Ni) of many structural materials have higher chemical affinity to oxygen than to the coolant alloy constituents, and such films can then provide a barrier to attack on the base metal by the lead or lead-bismuth coolant. Without such protective measures, Fe, Cr, and especially Ni all have nonnegligible solubility in lead-alloys which causes severe dissolution attacks. Because of the higher solubilities of Fe, Cr, and Ni in LBE than in lead, any dissolution problems are more severe in LBE at similar temperatures [11]. Oxygen sensors and control systems are thus important components of the reference coolant technology.

The most advanced Russian alloys, although similar to some western alloys, have no direct counterpart. Austenitic

alloys similar to AISI 304 and 316, as well as 12Cr ferritic/martensitic steels similar to HT9, were the primary materials of construction, but the Russian alloys had significant modifications. One important difference was the addition of silicon to improve corrosion resistance.

In the past five years, many international programs [46–52] have expended resources towards understanding corrosion in lead-bismuth eutectic (LBE). These programs have just started to generate data with somewhat scattered results. A recent summary by Zhang and Li [53] provides an excellent overview of LBE properties, corrosion mechanisms, corrosion mitigation including oxygen control, and corrosion models. Zhang and Li summarize the knowledge of LBE corrosion as follows.

- (i) At very low oxygen, both austenitic and ferritic/martensitic steels are subject to dissolution, even at low temperature.
- (ii) From 300°C to 470°C, with sufficient oxygen ( $>0^{-4}$ -ppm), protective oxide films can be formed on both austenitic and ferritic/martensitic steels.
- (iii) For temperatures above 550°C, austenitic stainless steels undergo heavy dissolution and ferritic/martensitic steels form a very thick and potentially unstable oxide. This thick oxide may be susceptible to erosion at high flow rates. Between 470°C and 550°C, the corrosion behavior in structural steels appears to make a transition from oxidation to dissolution and it may be acceptable using oxygen control. Furukawa et al. determined that at these higher temperatures, the iron oxide form changed from magnetite to wustite, which was a less adhesive form [21].

Gómez Briceño et al. [49] showed that for high oxygen concentration in the coolant, alloys with increased chromium concentration had better corrosion resistance as the chromium promoted the formation of a stable spinel layer. Alternately, at low coolant oxygen concentration, lower bulk chromium (e.g., carbon steels) minimizes Cr dissolution. Additionally, changes in oxygen concentration during a test can significantly alter material response. This may pose strict operational concerns on coolant chemistry.

As pointed out by Zhang and Li [53], although much is being learned about corrosion in LBE, the technical basis to connect corrosion to the multiple influential factors is still immature. Significant advances are required in the areas of precipitation and deposition, oxygen transport, oxide formation and kinetics, hydrodynamic effects, composition, microstructure, surface effects, and corrosion modeling.

A few studies have pointed out methods towards improving corrosion control. Kurata and Futakawa [46] recently showed that the high silicon-containing alloy SX (Fe-18Cr-20Ni-5Si) formed a protective oxide at 550°C in tests where JPCA and 316 did not form stable oxides. The result is consistent with the Russian development of alloy EP 823 in that increased silicon allows for the formation of a stable SiO<sub>2</sub> layer that prevents dissolution. Because of concerns on radiation performance and weldability, the silicon level in alloy SX is likely to be too large, but optimizing silicon content is likely to be a critical portion of corrosion control.

Limited data exist for refractory metals. Fazio et al. [47] studied the corrosion of AISI 316L, T91, W, and Mo in very low oxygen ( $3.1 \times 10^{-10}$  to  $7.3 \times 10^{-8}$  wt%) for times up to 1500 hours. At these low-oxygen potentials, W and Mo had a corrosion rate two orders of magnitude less than the steels although, based on previous testing, a protective oxide is not expected at higher-oxygen potentials for the refractory alloys. Runge et al. [54] tested the corrosion response of V-4Cr-4Ti exposed to lead at 650–800°C for 1000 hours. This alloy, which has undergone significant development for the fusion materials programs, was severely attacked.

The corrosion resistance of HT9 or other ferritic/martensitic steels still needs to be proven before it is chosen as the cladding. Both Russian experience and preliminary US corrosion studies indicate that elevated silicon levels may be required to provide adequate corrosion resistance when using oxygen control as the method for cladding corrosion protection. If alloys with higher silicon are required, the irradiation test base must be established for the new higher silicon.

Initial corrosion studies on  $\beta$ -SiC [55] indicated excellent corrosion resistance in lead at 800°C with no interaction between the SiC and the lead. A SiC-SiC Nicalon composite material was also tested in 800°C lead for 1000 hours [55]. For the composite, lead intruded and wet the composite surface and structure forming a reaction phase attributed to residual silica within the Nicalon fibers.

Although there is no report of liquid metal embrittlement (LME) in the oxygen-controlled regime in Russian LBE technology, there is emerging experimental evidence that LME may occur when oxygen is depleted (such as in abnormal operating conditions and during accidents) or when a hydrogen concentration influence is present.

In summary, the initial corrosion studies indicate that oxygen control may be tenable as a corrosion control solution for lower-temperature ( $\sim 470^\circ\text{C}$ ) systems. Above this temperature, dissolution mechanisms become more significant. For both metal and ceramic candidates, significant testing and development are required before a specific candidate can be selected and an engineering database can be established.

### 3.2.4. Surface treatment

Coating or surface modification is an R&D solution being evaluated as a method to establish corrosion control for materials in contact with lead or LBE. Successfully developed, the surface modification would provide corrosion protection, but it would maintain the acceptable mechanical and dimensional stabilities of the bulk metal performance under irradiation.

Alloying materials with elements promoting tenacious and protective oxides (e.g., Si and Al) or treating/coating the surface with appropriate materials for enhanced corrosion resistance have been developed and tested with oxygen control. The surface treatment procedures include procedures to aluminize (FeCrAlY base treatment) steel surfaces via heat treatment and electron beam processing (the GepulsteElektronenStrahlAnlage (GESA) process) [51]. Initial testing (both static and dynamic) shows exceptional resistance to

LBE. However, it is not known how the performance of such materials will change if the film is accidentally damaged, and how it behaves in radiation environments. Tests will need to be performed over a range of operating and accident conditions, with emphasis on coating stability and response to damage.

#### 4. CONCLUSION

The emerging global energy market is motivating the consideration of innovative nuclear energy technologies. The properties of lead and lead-bismuth eutectic coolants allow for the design of lead-cooled fast reactor (LFR) systems with innovation features. However, realization of such systems requires further development of technology to address issues with fuels and materials in these systems. Fuels and core materials used for sodium-cooled fast reactor application could be similarly attractive for LFR application; however, issues or uncertainties with the compatibility of those fuels and materials with lead and lead-bismuth eutectics must be addressed. Fuels based on U-Pu-Zr fuel alloys and (U,Pu) $O_2$  have significant performance histories, but metal fuels have potentially poor compatibility with lead and lead-bismuth coolants, and the higher operating temperatures associated with oxide fuels diverge from the passive safety goals of LFR designers. Nitride fuels are reputed to have good compatibility with lead and lead-bismuth, but the application maturity of those fuels is low. Similarly, structural stainless steels developed for sodium-cooled fast reactor application, such as D9 or HT9 in the US, have reasonable development maturity but inferior lead and lead-bismuth compatibilities at temperatures of interest for LFR systems. Later-generation alloys with improved high-temperature radiation performance have been developed (such as ODS, NF616, and HCM12A), but they are also unproven relative to corrosion performance and as such they are sufficiently immature to preclude their application to LFR systems for at least a decade. No metallic or ceramic material has currently proven corrosion and radiation resistance under LFR relevant conditions. The LFR systems are attractive, but they will have limited deployability until suitable fuel designs and materials of construction are identified.

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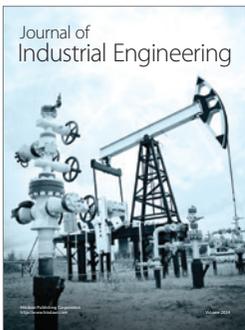
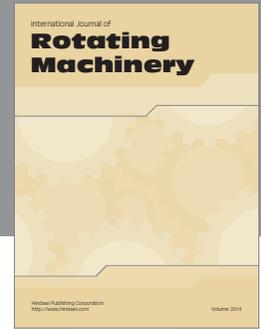
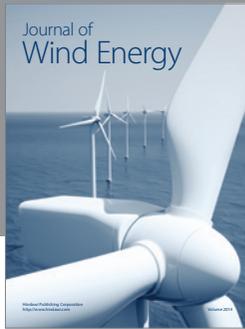
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