Liquid Fuel Nuclear Reactors

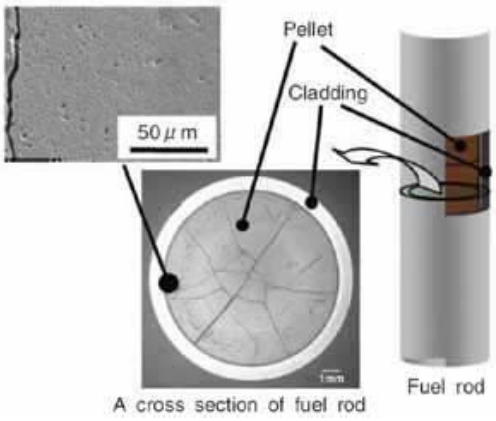
Robert Hargraves and Ralph Moir

Today's familiar pressurized water nuclear reactors use solid fuel -- pellets of uranium dioxide in zirconium fuel rods bundled into fuel assemblies. These assemblies are placed within the reactor vessel under water at 160 atmospheres pressure and a temperature of 330°C. This hot water transfers heat from the fissioning fuel to a steam turbine that spins a generator to make electricity. Alvin Weinberg invented the pressurized water reactor (PWR) in 1946 and such units are now used in over 100 commercial power-producing reactors in the US as well as in naval vessels.

Weinberg also pursued research on liquid fuel-reactors, which offer a number of advantages over their solid-fueled counterparts. In this article we review some of the history, potential advantages, potential drawbacks, and current research and development status of liquid-fueled reactors. Our particular emphasis is on the Liquid Fluoride Thorium Reactor (LFTR).

Before describing the characteristics of liquid-fuel reactors we review briefly in this paragraph the situation with PWRs. In a conventional PWR the fuel pellets contain UO_2 with fissile U-235 content expensively enriched to 3.5% or more, the remainder being U-238. After about 5 years the fuel must be removed because the fissile material is depleted and neutron-absorbing fission products build up. By that time the fuel has given up less than 1% of the potential energy of the mined uranium, and the fuel rods have become stressed by internal temperature differences, by radiation damage that breaks covalent UO_2 bonds, and by fission products that disturb the solid lattice structure (Figure 1). As the rods swell and distort, their zirconium cladding must continue to contain the fuel and fission products while in the reactor and for centuries thereafter in a waste storage repository.

In contrast, fluid fuels are not subjected to the structural stresses of solid fuels: liquid-fuel reactors can operate at atmospheric pressure, obviating the need for containment vessels able to withstand high-pressure steam explosions. Gaseous fission products like xenon bubble out while some fission products precipitate out and so do not absorb neutrons from the chain reaction. Like PWRs, liquid-fuel reactors can be configured to breed more fuel, but in ways that make them more proliferation resistant than the waste generated by conventional PWRs. Spent PWR fuel contains transuranic nuclides such as Pu-239, bred by neutron absorption in U-238, and it is such long-lived transuranics that are a core issue in waste storage concerns. In contrast, liquid-fuel reactors have the potential to reduce storage concerns to a few hundred years as they would produce far fewer transuranic nuclides than a PWR.



Courtesy of Japan Atomic Energy Agency R&D Review 2008

Figure 1. Solid fuel rods are stressed by fission products, radiation, and heat.

History of liquid fuel reactors

The world's first liquid fuel reactor used uranium sulfate fuel dissolved in water. Eugene Wigner conceived this technology in 1945, Alvin Weinberg built it at Oak Ridge, and Enrico Fermi started it up. The water carries the fuel, moderates neutrons (slows them to take advantage of the high fission cross-section of uranium for thermal-energy neutrons), transfers heat, and expands as the temperature increases, thus lowering moderation and stabilizing the fission rate. Because the hydrogen in ordinary water absorbs neutrons, an aqueous reactor, like a PWR, cannot reach criticality unless fueled with uranium enriched beyond the natural 0.7% isotopic abundance of U-235. Deuterium absorbs few neutrons, so, with heavy water, aqueous reactors can use unenriched uranium. Weinberg's aqueous reactor fed 140 kW of power into the electric grid for 1000 hours. The intrinsic reactivity control was so effective that shutdown was accomplished simply by turning off the steam turbine generator.

In 1943, Wigner and Weinberg also conceived a liquid fuel thorium-uranium breeder reactor, for which the aqueous reactor discussed above was but the first step. The fundamental premise in such a reactor is that a blanket of thorium Th-232 surrounding the fissile core will absorb neutrons, with some nuclei thus being converted

("transmuted") to Th-233. Th-233, in turn, beta decays to protactinium-233 and then to U-233, which is itself fissile and can be used to refuel the reactor. Later, as Director of Oak Ridge, Weinberg led the development of the liquid fluoride thorium reactor (LFTR), the subject of this article. Aware of the future effect of carbon dioxide emissions, Weinberg wrote "humankind's whole future depended on this." The Molten Salt Reactor Experiment, powered first with U-235 and then U-233, operated successfully over 4 years, through 1969. To facilitate engineering tests, the thorium blanket was not installed; the U-233 used in the core came from other reactors breeding Th-232. The MSRE was a proof-of-principle success. Fission-product xenon gas was continually removed to prevent unwanted neutron absorptions, online refueling was demonstrated, minor corrosion of the reactor vessel was addressed, and chemistry protocols for separation of thorium, uranium, and fission products in the fluid fluorine salts were developed. Unfortunately, the Oak Ridge work was stopped when the Nixon administration decided instead to fund only the solid fuel Liquid sodium Metal cooled Fast Breeder Reactor (LMFBR), which could breed plutonium-239 faster than the LFTR could breed uranium-233.

The Liquid Fluoride Thorium Reactor

A significant advantage of using thorium to breed U-233 is that relatively little plutonium is produced from the Th-232 because six more neutron absorptions are required than is the case with U-238. The U-233 that is bred is also proliferation-resistant in that the neutrons that produce it also produce 0.13% contaminating U-232 which decays eventually to thallium, which itself emits a 2.6 MeV penetrating gamma radiation that would be obvious to detection monitors and hazardous to weapons builders. For example, a year after U-233 separation, a weapons worker one meter from a subcritical 5 kg sphere of it would receive a radiation dose of 4,200 mrem/hr; death becomes probable after 72 hours exposure. Normally the reactor shielding protects workers, but modifying the reactor to separate U-233 would require somehow adding hot cells and remote handling equipment to the reactor and also to facilities for weapons fabrication, transport, and delivery. Attempting to build U-233-based nuclear weapons by modifying a LFTR would be more hazardous, technically challenging and expensive than creating a purpose-built weapons program using uranium enrichment (Pakistan) or plutonium breeding (India, North Korea).

Work on thorium-based reactors is currently being actively pursued in many countries including Germany, India, China, and Canada; India plans to produce 30% of its electricity from thorium by 2050. But all these investigations involve solid fuel forms. Our interest here is with the liquid-fueled form of a thorium-based U-233 breeder reactor.

The configuration of a LFTR is shown schematically in Figure 2. In a "two-fluid" LFTR a molten eutectic mixture of salts such as LiF and BeF_2 containing dissolved UF_4 forms the central fissile core. ("Eutectic" refers to a compound that solidifies at a lower temperature than any other compound of the same chemicals.) A separate annular region containing molten Li and Be fluoride salts with dissolved ThF4 forms the fertile blanket. Fission of U-233 (or some other "starter" fissile fuel) dissolved in the fluid core heats it. This heated fissile fluid attains a noncritical geometry as it is pumped through small passages inside a heat exchanger. Excess neutrons are absorbed by Th-232 in the molten salt blanket, breeding U-233 which is continuously removed with fluorine gas and used to refuel the core. Fission products are chemically removed in the waste separator, leaving uranium and transuranics in the molten salt fuel. From the heat exchanger a separate circuit of molten salt heats gases in the closed cycle helium gas turbine which generates power. All three molten salt circuits are at atmospheric pressure.

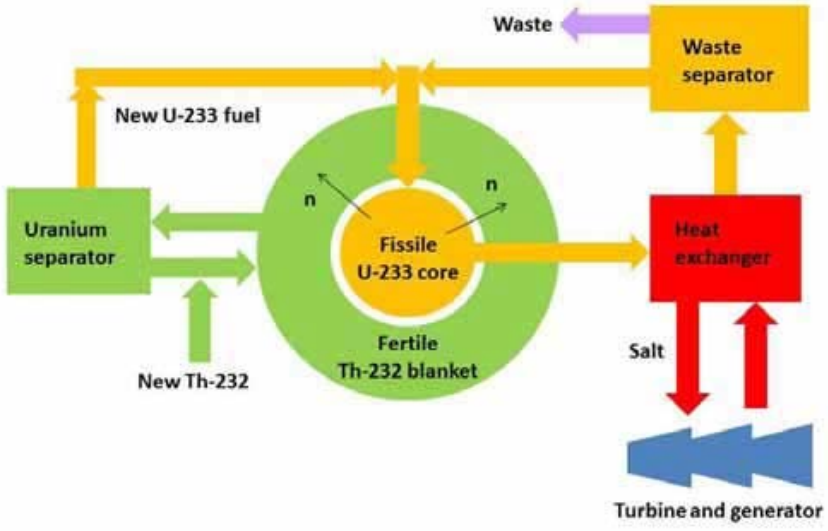


Figure 2. In a two-fluid liquid fluoride thorium reactor the fission of U-233 in the core heats molten carrier salt (yellow). It attains a noncritical geometry as it is pumped through small passages in a heat exchanger. A separate circuit of molten salt (red), with no radioactive materials, heats gases in the closed cycle helium gas turbine which spins to generate power. Excess neutrons are absorbed by Th-232 in the molten salt blanket (green), breeding U-233 which is removed with fluorine gas. Fission products are chemically removed in the waste separator, leaving uranium and transuranics in the molten salt fuel. All three molten salt circuits are at atmospheric pressure.

LFTRs would reduce waste storage issues from millions of years to a few hundred years. The radiotoxicity of nuclear waste arises from two sources: the highly radioactive fission products from fission and the long-lived actinides from neutron absorption. Thorium and uranium fueled reactors produce essentially the same fission products, whose radiotoxicity in 500 years drops below that of the original ore mined for uranium to power a PWR. A

products, whose radiotoxicity in 300 years drops below that of the original ore mined for uranium to power a PWR. A LFTR would create far fewer transuranic actinides than a PWR. After 300 years the LFTR waste radiation would be 10,000 times less than that from a PWR (Figure 3). In practice, some transuranics will leak through the chemical waste separator, but the waste radiotoxicity would be < 1% of that from PWRs. Geological repositories smaller than Yucca mountain would suffice to sequester the waste.

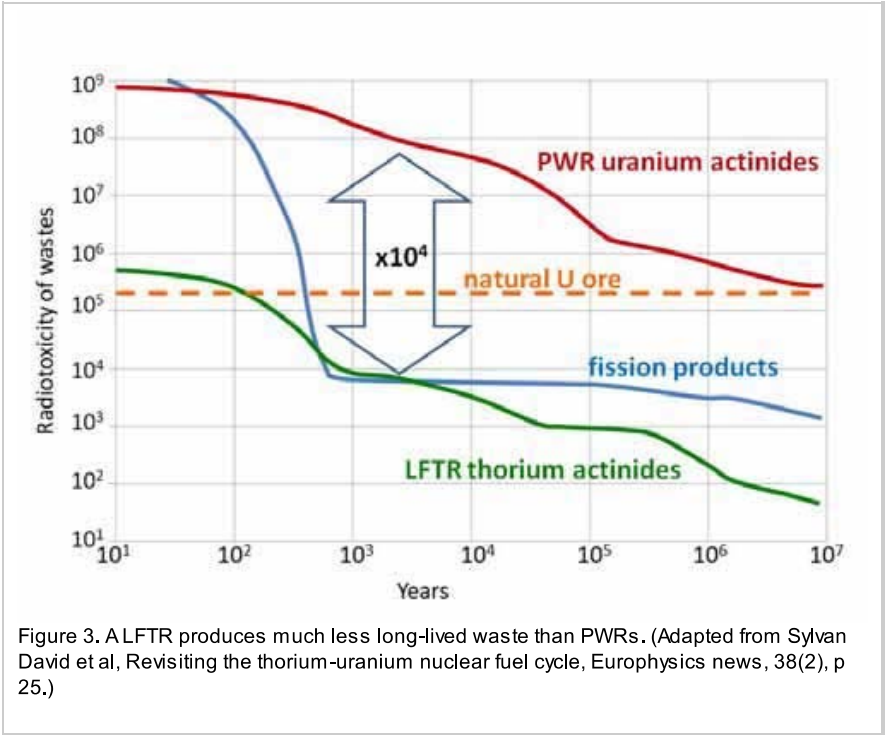


Figure 3. A LFTR produces much less long-lived waste than PWRs. (Adapted from Sylvan David et al, Revisiting the thorium-uranium nuclear fuel cycle, Europhysics news, 38(2), p 25.)

Existing PWR spent fuel can be an asset. A 100 MW LFTR requires 100 kg of fissile material (U-233, U-235, or Pu-239) to start the chain reaction. The world now has 340,000 tonnes of spent PWR fuel, of which 1% is fissile material that could start one 100 MW LFTR per day for 93 years.

A commercial LFTR will make just enough uranium to sustain power generation, so diverting uranium for weapons use would stop the reactor, alerting authorities. A LFTR will have little excess fissile material; U-233 is continuously generated to replace the fissioned U-233, and Th-232 is continuously introduced to replace the Th-232 converted to the U-233. Terrorists could not steal this uranium dissolved in a molten salt solution along with lethally radioactive fission products inside a sealed reactor, which would be subject to the usual IAEA safeguards of physical security, accounting and control of all nuclear materials, surveillance to detect tampering, and intrusive inspections.

It is also possible to configure a liquid-fuel reactor that would involve no U-233 separation. For example, the single fluid denatured molten salt reactor (DMSR) version of a LFTR with no U-233 separation is fed with both thorium and < 20% enriched uranium. It can operate up to 30 years before actinide and fission product buildup requires fuel salt replacement, while consuming only 25% of the uranium a PWR uses.

Starting up LFTRs with plutonium can consume stocks of this weapons-capable material. Thorium fuel would also reduce the need for U-235 enrichment plants, which can be used to make weapons material as easily as power reactor fuel. U-233, at the core of the reactor, is important to LFTR development and testing. With a half-life of only 160,000 years, it is not found in nature. The US has 1,000 kg of nearly irreplaceable U-233 at Oak Ridge. It is now slated to be destroyed by diluting it with U-238 and burying it forever, at a cost of $477 million. This money would be far better invested in LFTR development.

Can LFTR power be cheaper than coal power?

Burning coal for power is the largest source of atmospheric CO_2, which drives global warming. We seek alternatives such as burying CO_2 or substituting wind, solar, and nuclear power. A source of energy cheaper than coal would dissuade nations from burning coal while affording them a ready supply of electric power.

Can a LFTR produce energy cheaper than is currently achievable by burning coal? Our target cost for energy cheaper than from coal is $0.03/kWh at a capital cost of $2/watt of generating capacity. Coal costs $40 per ton, contributing $0.02/kWh to electrical energy costs. Thorium is plentiful and inexpensive; one ton worth $300,000 can power a 1,000 megawatt LFTR for a year. Fuel costs for thorium would be only $0.00004/kWh.

The 2009 update of MIT's *Future of Nuclear Power* shows that the capital cost of new coal plants is $2.30/watt, compared to LWRs at $4/watt. The median of five cost studies of large molten salt reactors from 1962 to 2002 is $1.98/watt, in 2009 dollars. Costs for scaled-down 100 MW reactors can be similarly low for a number of reasons, six of which we summarize briefly:

Pressure. The LFTR operates at atmospheric pressure, obviating the need for a large containment dome. At atmospheric pressure there is no danger of an explosion.

Safety. Rather than creating safety with multiple defense-in-depth systems, LFTR's intrinsic safety keeps such

Safety. Rather than creating safety with multiple defense-in-depth systems, LFTR's intrinsic safety keeps such costs low. A molten salt reactor cannot melt down because the normal operating state of the core is already molten. The salts are solid at room temperature, so if a reactor vessel, pump, or pipe ruptured they would spill out and solidify. If the temperature rises, stability is intrinsic due to salt expansion. In an emergency an actively cooled solid plug of salt in a drain pipe melts and the fuel flows to a critically safe dump tank. The Oak Ridge MSRE researchers turned the reactor off this way on weekends.

Heat. The high heat capacity of molten salt exceeds that of the water in PWRs or liquid sodium in fast reactors, allowing compact geometries and heat transfer loops utilizing high-nickel metals.

Energy conversion efficiency. High temperatures enable 45% efficient thermal/electrical power conversion using a closed-cycle turbine, compared to 33% typical of existing power plants using traditional Rankine steam cycles. Cooling requirements are nearly halved, reducing costs and making air-cooled LFTRs practical where water is scarce.

Mass production. Commercialization of technology lowers costs as the number of units produced increases due to improvements in labor efficiency, materials, manufacturing technology, and quality. Doubling the number of units produced reduces cost by a percentage termed the learning ratio, which is often about 20%. In *The Economic Future of Nuclear Power*, University of Chicago economists estimate it at 10% for nuclear power reactors. Reactors of 100 MW size could be factory-produced daily in the way that Boeing Aircraft produces one airplane per day. At a learning ratio of 10%, costs drop 65% in three years.

Ongoing research. New structural materials include silicon-impregnated carbon fiber with chemical vapor infiltrated carbon surfaces. Such compact thin-plate heat exchangers promise reduced size and cost. Operating at 950°C can increase thermal/electrical conversion efficiency beyond 50% and also improve water dissociation to create hydrogen for manufacture of synthetic fuels such that can substitute for gasoline or diesel oil, another use for LFTR technology.

In summary, LFTR capital cost targets of $2/watt are supported by simple fluid fuel handling, high thermal capacity heat exchange fluids, smaller components, low pressure core, high temperature power conversion, simple intrinsic safety, factory production, the learning curve, and technologies already under development. A $2/watt capital cost contributes $0.02/kWh to the power cost. With plentiful thorium fuel, LFTRs may indeed generate electricity at less than $0.03/kWh, underselling power generated by burning coal. Producing one LFTR of 100 MW size per day could phase out all coal burning power plants worldwide in 38 years, ending 10 billion tons per year of CO_2 emissions from coal plants.

Development Status of LFTRs

A number of LFTR initiatives are currently active around the world. France supports theoretical work by two dozen scientists at Grenoble and elsewhere. The Czech Republic supports laboratory research in fuel processing at Rez, near Prague. Design for the FUJI molten salt reactor continues in Japan. Russia is modeling and testing components of a molten salt reactor designed to consume plutonium and actinides from PWR spent fuel, and LFTR studies are underway in Canada and the Netherlands. US R&D funding has been relatively insignificant, except for related studies of solid fuel, molten salt cooled reactors at UC Berkeley and Oak Ridge, which hosted a conference to share information on fluoride reactors in September 2010.

Developing LFTRs will require advances in high temperature materials for the reactor vessel, heat exchangers, and piping; chemistry for uranium and fission product separation; and power conversion systems. The International Generation IV Forum budgeted $1 billion over 8 years for molten salt reactor development. We recommend a high priority, 5-year national program to complete prototypes for the LFTR and the simpler DMSR. It may take an additional 5 years of industry participation to achieve capabilities for mass production. Since LFTR development requires chemical engineering expertise and liquid fuel technology is unfamiliar to most nuclear engineers today, nuclear engineering curricula would have to be modified to include exposure to such material. The technical challenges and risks that must be addressed in a prototype development project include control of salt container corrosion, recovery of tritium from neutron irradiated lithium salt, management of structural graphite shrinking and swelling, closed cycle turbine power conversion, and maintainability of chemical processing units for U-233 separation and fission product removal. Energy Secretary Chu expressed historical criticism of the technology in a letter to Senator Jeanne Shaheen (D-NH) answering questions at his confirmation hearings, “One significant drawback of the MSR technology is the corrosive effect of the molten salts on the structural materials used in the reactor vessel and heat exchangers; this issue results in the need to develop advanced corrosion-resistant structural materials and enhanced reactor coolant chemistry control systems”, and “From a non-proliferation standpoint, thorium-fueled reactors present a unique set of challenges because they convert thorium-232 into uranium-233 which is nearly as efficient as plutonium-239 as a weapons material.” He also recognized, however, that “Some potential features of a MSR include smaller reactor size relative to light water reactors due to the higher heat removal capabilities of the molten salts and the ability to simplify the fuel manufacturing process, since the fuel would be dissolved in the molten salt.”

Other hurdles to LFTR development may be the regulatory environment and the prospect of disruption to current practices in the nuclear industry. The Nuclear Regulatory Commission will need funding to train staff qualified to work with this technology. The nuclear industry and utilities will be shaken by this disruptive technology that changes whole fuel cycle of mining, enrichment, fuel rod fabrication, and refueling. Ultimately, the environmental and human development benefits will be achieved only when the cost of LFTR power really proves to be cheaper than from coal.

References

Robert Hargraves and Ralph Moir, Liquid Fluoride Reactors, American Scientist, July/August 2010

Alvin Martin Weinberg, The first nuclear era: the life and times of a technological fixer. Springer, New York, 1997.

Oak Ridge National Laboratory document repository

S. David, E. Huffer, H. Nifenecker, Revisiting the thorium-uranium nuclear fuel cycle

David LeBlanc, Molten Salt Reactors: A New Beginning for an Old Idea

Ralph Moir, Edward Teller, Thorium fueled underground power plant based on molten salt technology, http://ralphmoir.com/moir_teller.pdf

Per Peterson, Pebble Bed Advanced High Temperature Reactor, http://www.nuc.berkeley.edu/pb-ahtr/

Oak Ridge National Laboratory, Fluoride Salt-Cooled High-Temperature Reactor Agenda, https://www.ornl.gov/fhr/agenda.html

A Technology Roadmap for Generation IV Nuclear Systems, http://gif.inel.gov/roadmap/pdfs/gen_iv_roadmap.pdf

Robert Hargraves teaches Energy Policy at the Institute for Lifelong Education at Dartmouth College. He received his Ph.D. in physics from Brown University.

Ralph Moir has published ten papers on molten salt reactors during his career at Lawrence Livermore National Laboratory. He received his Sc.D. in nuclear engineering from the Massachusetts Institute of Technology.

These contributions have not been peer-refereed. They represent solely the view(s) of the author(s) and not necessarily the view of APS.

APS Home | APS Units | Terms of Use © 2013 American Physical Society