ACCELERATORS FOR SUBCRITICAL MOLTEN SALT REACTORS*

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Abstract

Reactors built using solid fissile materials sealed in fuel rods have an inherent safety problem in that volatile radioactive materials in the rods are accumulated and can be released in dangerous amounts. Accelerator parameters for subcritical reactors that have been considered in recent studies have primarily been based on using solid nuclear fuel much like that used in all operating critical reactors. An attractive alternative reactor design that used molten salts was experimentally studied at ORNL in the 1960s, where a critical molten salt reactor was successfully operated using enriched U235 or U233 tetrafluoride fuels. These experiments give confidence that accelerator-driven subcritical molten salt reactors will work as well or better than conventional reactors since they will have better efficiency due to their higher operating temperature, the inherent safety of subcritical operation, and constant purging of volatile radioactive elements to eliminate their accumulation and potential accidental release in dangerous amounts.

INTRODUCTION

Energy, Safety, Waste

The US nuclear power industry provides about 20% of the nation's electricity, in spite of the fact that no new reactors have been built in the US in several decades. Public perceptions regarding possible nuclear accidents, weapons proliferation, terrorist activities, and the mounting stockpile of waste from conventional reactors have made the growth of this industry difficult. However, even the general public is coming to realize that nuclear power could be an effective mitigation to the greenhouse gas emissions related to climate change. Accelerator Driven Subcritical Reactors (ADSR) have the potential to address the concerns of the public in all respects, from intrinsic safety from subcritical operation, discouraging proliferation and terrorists, to burning the waste from conventional reactors.

Lessons from Fukushima

The multiple disasters at Fukushima, starting from the earthquake and tsunami, have uncovered problems with solid-fuel rod designs that can be overcome with molten-salt fuels. Figure 1, taken from the NY Times, shows the radiation levels at the plant gates plotted by date after the March 11, 2011 earthquake and tsunami. Radiation level increases can be seen that can be attributed to the nature of the solid fuel rods used in light water reactors. In these reactors, long tubes of strong metal (zirconium alloy) enclose small cylinders of solid uranium or plutonium oxide ceramic. Moderators to slow the neutrons can be inserted between the tubes or rods to control the reaction rates while water circulates between the cylinders to transfer the heat to steam turbines.

Only the first three of six reactors were operating at the time of the earthquake. These three correctly "scrammed" when the earthquake happened, stopping fission reactions. However, due to the decays of fission products, the power output of the reactor fuel rods continued at something like 5% of the rate of normal operation. Once the tsunami hit, the emergency cooling systems failed, the rods became very hot, and the first vulnerability of the zircaloy-enclosed fuel rods manifested itself in hydrogen explosions in each of the first three reactors as seen in Fig. 1. Steam and hot zircaloy react exothermically to produce large amounts of hydrogen ($Zr + 2\ H_2O \rightarrow ZrO_2 + 2\ H_2$) at a rate that increases exponentially with temperature. This is surely the source of the explosions.

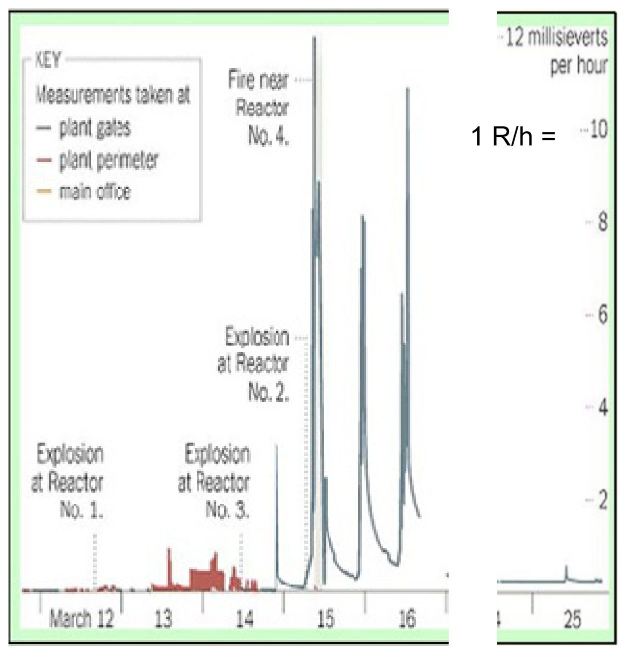

Figure 1: Radiation levels at the gates of the Fukushima power plant following the March 11 earthquake and tsunami. Twenty minutes at the peak on March 15 would correspond to the average yearly dose from natural radiation in the USA.

Once the zircaloy melts or burns, volatile radioactive fission products (Xe, Kr, I, Sr, Cs, Ru,..) that have been accumulated in the fuel rods during months or years of operation can be released. Increases in radiation levels are seen at the time of each explosion. The large spike in radiation that correlates with the fire near the non-

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operational reactor #4 is presumably due to loss of water in the spent fuel storage pond, leading to failure of the zircaloy enclosing the spent fuel.

The large spikes of radiation on March 15 and 16 are suspected to be due to the accumulation of fuel pellets from failed fuel rods at the bottom of the pressure vessels in reactors #2 and #3. These became hot enough to melt through the steel vessel walls.

In molten-salt fuel designs, there is no zircaloy to cause explosions and little accumulation of volatile fission products to create dangerous and frightening fallout.

Extra Neutrons

The only materials that exist in the world in significant quantity that are capable of a sustained nuclear reaction are U235, found as 0.7% of natural uranium, and Pu239, which has been created in U235-based reactors. A critical mass of either of these fissile isotopes will create enough neutrons so that a chain reaction will continue. Other fertile materials such as naturally occurring thorium, nuclear waste from conventional reactors, or natural uranium can also be converted to fissile elements to produce energy if additional neutrons can be added, e.g.:

$$n + \underset{\text{fertile}}{{}^{238}U_{92}} \rightarrow {}^{239}U_{92} \xrightarrow[\text{24 m}]{\beta^-} {}^{239}Np_{93} \xrightarrow[\text{2.4 d}]{\beta^-} \underset{\text{fissile}}{{}^{239}Pu_{94}}$$

$$n + {}^{232}\mathbf{Th}_{90} \rightarrow {}^{233}Th_{90} \xrightarrow[\text{22 m}]{} {}^{233}Pa_{91} \xrightarrow[\text{27 d}]{} {}^{233}U_{92}$$

The two known methods to add neutrons use 1) fast breeder reactors or 2) particle accelerators similar to those developed for basic physics research. While the development of fast breeder reactors has been on hold in the USA because of nuclear weapon proliferation concerns, particle accelerator technology has reached the point where spallation neutrons can be produced in sufficient quantity for practical ADSR uses. In particular, with an accelerator, neutrons can be added to overcome the build-up of neutron-absorbing fission products, whereas for long-term breeder operation the fuel must be reprocessed.

ACCELERATORS FOR ADSR

Molten-Salt Fuel ADSR

Some features of a molten-salt reactor are displayed in Fig. 2, which shows the ADNA GEM*STAR conceptual design [1]. The molten-salt fuel mixture (e.g. UF_4, ThF_4, LiF) is held in a graphite-reflector, Hastelloy-N container, which also contains the heat exchanger (non-radioactive) liquid salt. Beams of energetic protons hit a uranium target to cause spallation neutrons to enter the fuel mixture. Volatile radioactive fission products are constantly purged by a flow of helium gas.

This helium purging feature practically eliminates the possibility of accidental releases of radioactivity that is a well-known problem of technologies that employ fuel rods that must contain years of built-up radioactive volatile elements. In the molten-salt case, the usual containment vessel is not required.

The heat exchanger molten-salt can include a large-volume reservoir to reduce sensitivity to accelerator beam interruptions to the point that power output from the turbine/generator can continue even for accelerator down times of several hours.

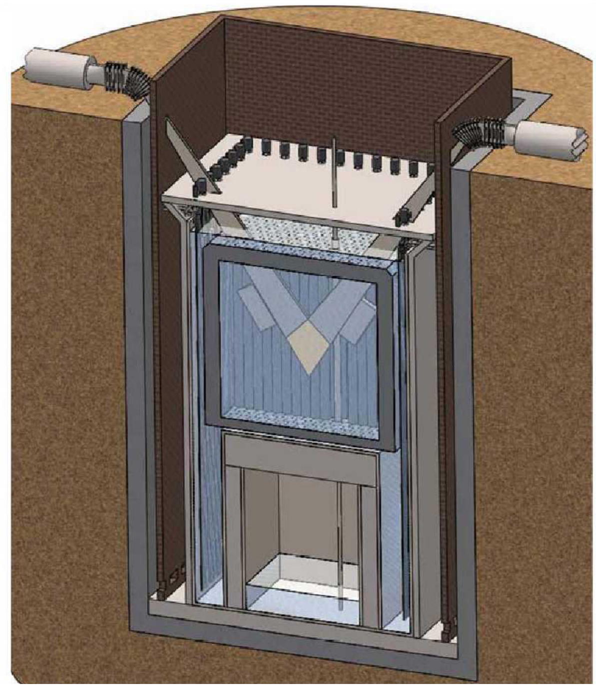

Figure 2: Conceptual design of the ADNA Green Energy Multiplier - Subcritical-technology, Thermal-spectrum, Accelerator-driven, Recycling molten salt reactor (GEM*STAR) in its underground placement. The vertical dimension is about 9 m. The gray box is the graphite reflector for the core. Here, horizontal beams from two accelerators are shown at the top being bent by magnets into the core where they strike a uranium metal target shown schematically in the center of the core.

MOLTEN-SALT REACTOR EXPERIMENT

The practicality of the molten-salt concept was demonstrated in an experiment at ORNL in the 1960s. The following is from the 1969 Report Abstract [2].

"The MSRE is an 8-MW(th) reactor in which molten fluoride salt at 1200°F circulates through a core of graphite bars. Its purpose was to demonstrate the practicality of the key features of molten-salt power reactors.

Operation with ^{235}U (33% enrichment) in the fuel salt began in June 1965, and by March 1968 nuclear operation amounted to 9,000 equivalent full-power hours. The goal of demonstrating reliability had been attained - over the last 15 months of ^{235}U operation the reactor had been critical 80% of the time. At the end of a 6-month run which climaxed this demonstration, the reactor was shut down and the 0.9 mole% uranium in the fuel was stripped very efficiently in an on-site fluorination facility. Uranium-233 was then added to the carrier salt, making *the MSRE the world's first reactor to be fueled with this fissile material. Nuclear operation was resumed in October 1968, and over 2,500 equivalent full-power hours have now been produced with ^{233}U.*

The MSRE has shown that salt handling in an operating reactor is quite practical, the salt chemistry is well behaved, there is practically no corrosion, the nuclear characteristics are very close to predictions, and the system is dynamically stable. Containment of fission products has been excellent and maintenance of radioactive components has been accomplished without unreasonable delay and with very little radiation exposure.

The successful operation of the MSRE is an achievement that should strengthen confidence in the practicality of the molten-salt reactor concept."

It remains to recapture the expertise that was demonstrated more than 40 years ago and to extend it based on technology that has been developed since then.

SRF-BASED ACCELERATOR

Superconducting RF Linac

The most popular accelerator contenders for ADSR applications that have the potential to provide 10 MW, 1 GeV continuous proton beams include cyclotrons and fixed-field alternating gradient synchrotrons as well as SRF linacs. While SRF linacs are likely to be the most expensive due to their complexity and component costs, they are also the most likely to achieve the required energy and power parameters. Beam from sources and RFQs that generate hundreds of mA of proton current can be accelerated to energies of several GeV for extrapolated beam power of hundreds of MW.

One uncertainty in this extrapolation is the requirement of loss rates in the linac, which should be kept below 1 W/m to avoid levels of induced residual radioactivity in the linac components that would make hands-on maintenance difficult.

Economies of scale will eventually be important for ADSR, since in many markets, the issue will be cost per kw-h in comparison with other power sources, including conventional reactors and fossil fuel power stations. The natural maximum size of a molten-salt reactor such as GEM*STAR as shown in Fig. 2 (500 MWt or 220 MWe) implies that a multi-GW power station may have one or two dozen reactors that need 10 MW each for operation. The present design of the first phase of the (~1$B) Fermilab Project-X proton driver [3] is for a 5 to 10 ma H-minus source (limited to 1 ma average) that will operate CW at 3 GeV. One can imagine that it could eventually be upgraded to handle a 100 ma proton source for 300 MW to feed 30 molten-salt reactors.

The upgrade would require only slightly more cryogenic infrastructure but would need higher-power RF sources and couplers, added redundancy, efficient beam splitting and distribution schemes based on the same transverse-kicking RF used at CEBAF, and highly efficient and fast trip recovery and beam loss protection techniques. These additional requirements could be met with an enthusiastic R&D program that could be an additional goal of Project X to augment its HEP importance. Such an ambitious goal would complement the development of a national power grid with sufficient capability to handle very large power stations.

However, to get started and to develop confidence that there are no surprises, it is possible to use a smaller energy and beam power, perhaps as little as 500 MeV at 2.5 MW. Very little development effort for reliability may be needed for the first stage because of the intrinsic insensitivity of the molten-salt fuel to short-term interruptions and because of the buffering provided by the large volume non-radioactive molten-salt heat exchanger to cover longer interruptions.

Spallation Target for ADSR

The SNS spallation neutron target and other high-power targets under development are expensive and complex in part because the heat produced is large and difficult to remove from the relatively small interaction region where the beam is absorbed, and also because the beam is pulsed, which adds shock to the problem. For ADSR, the beam is CW, relatively diffuse, and the target and molten-salt fuel can share the same volume inside the reactor. In this latter case, the heat from the target can be handled naturally by the heat exchange salt, and some of the power used by the accelerator can be recovered.

CONCLUSIONS AND OUTLOOK

ADSR using molten-salt fuel has impressive advantages: 1) ability to burn any number of materials including conventional reactor waste, excess plutonium from weapons, and very abundant thorium; 2) exceptional safety advantages including subcriticality to eliminate Chernobyl-like disasters, 3) no build-up of volatile radioactive elements to eliminate Fukushima or 3-Mile Island problems; 4) no storage of solid nuclear waste that can catch fire. Because of its insensitivity to beam interruptions, ADSR with molten-salt fuel also relaxes the accelerator requirements.

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[3] http://projectx.fnal.gov/index.shtml