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 accidentally 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 as well as the thorium-burning accelerator-driven energy amplifier proposed by Rubbia et al. 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 an accelerator-driven subcritical molten salt reactor will work as well or better than conventional reactors, having better efficiency due to their higher operating temperature, having the inherent safety of subcritical operation, and having constant purging of volatile radioactive elements to eliminate their accumulation and potential accidental release. Moreover, the requirements to drive a molten salt reactor can be considerably relaxed compared to a solid fuel reactor, especially regarding accelerator reliability, to the point that much of the required superconducting RF (SRF) technology exists today. It is proposed that a prototype commercial machine be built to produce energy for the world by, for example, burning thorium in India and nuclear waste from conventional reactors in the USA.

Work supported by ATI: http://acceltech.us
A few preliminary comments:

- **Muons, Inc.** is a small company founded 9 years ago to help the DOE solve its problems through SBIR-STTR projects and contracts with national labs and universities
  - Staff of 22 Ph.D. level accelerator physicists and engineers
  - The name comes from our obsession to have a muon collider be the next energy frontier machine, and with muon beam cooling
  - Developed **G4beamline**, a program to interface to GEANT4

- We have founded a new company to raise private capital to support R&D for ADSR using molten salt fuel:
  - Infomercial at [http://acceletech.us](http://acceletech.us)

- We believe ADSR needs close collaboration between accelerator and reactor people and have been very lucky to collaborate with Charlie Bowman of ADNA
  - 1992 patent: Apparatus for nuclear transmutation and power production using an intense ... Charles D. Bowman
Goal – US government pays industry to remove nuclear waste and produce energy from it

- Setting the stage – where we are – opportunities/problems
- Solid fuel nuclear reactor technology - what goes wrong
  - fuel rods – accidents waiting to happen?
- Molten-salt Reactor Experiment (MSRE) 1965-1969
  - continuous purging of volatile radioactive elements – no zircaloy
- Accelerator-Driven Subcritical Reactors (ADSR)
  - reactor concept uses molten salt fuel (e.g. UF₄ or ThF₄)
  - GEM*STAR example

- Avoids nuclear weapon proliferation concern of reprocessing for 200 years
- The next step is a prototype ADSR machine to inspire industry
  - basic design issues, safety systems, reliability, availability, residual radiation from beam losses, beam delivery, independent reactor control, economy of construction and operation, ...

- Rousing Conclusions
## Nuclear Power Capacity as of 02/2012

<table>
<thead>
<tr>
<th>Country</th>
<th># reactors</th>
<th>GW capacity</th>
<th>Nuclear share of electricity production</th>
</tr>
</thead>
<tbody>
<tr>
<td>Belgium</td>
<td>5.9</td>
<td>51.7%</td>
<td></td>
</tr>
<tr>
<td>Canada</td>
<td>12.7</td>
<td>14.8%</td>
<td></td>
</tr>
<tr>
<td>China (PRC)</td>
<td>10.2</td>
<td>1.9%</td>
<td></td>
</tr>
<tr>
<td>France</td>
<td>59</td>
<td>63.2%</td>
<td></td>
</tr>
<tr>
<td>Germany</td>
<td>20.3</td>
<td>26.1%</td>
<td></td>
</tr>
<tr>
<td>India</td>
<td>4.8</td>
<td>2.9%</td>
<td></td>
</tr>
<tr>
<td>Japan</td>
<td>54</td>
<td>47.3%</td>
<td></td>
</tr>
<tr>
<td>Korea, South</td>
<td>18.7</td>
<td>31.1%</td>
<td></td>
</tr>
<tr>
<td>Russia</td>
<td>23.0</td>
<td>17.8%</td>
<td></td>
</tr>
<tr>
<td>Spain</td>
<td>7.4</td>
<td>17.5%</td>
<td></td>
</tr>
<tr>
<td>Sweden</td>
<td>9.4</td>
<td>37.4%</td>
<td></td>
</tr>
<tr>
<td>Taiwan</td>
<td>4.9</td>
<td>20.7%</td>
<td></td>
</tr>
<tr>
<td>Ukraine</td>
<td>13.2</td>
<td>48.6%</td>
<td></td>
</tr>
<tr>
<td>United Kingdom</td>
<td>11.0</td>
<td>17.9%</td>
<td></td>
</tr>
<tr>
<td>United States</td>
<td>104</td>
<td>101.2%</td>
<td></td>
</tr>
<tr>
<td>Rest of World</td>
<td>25.4</td>
<td>20.2%</td>
<td></td>
</tr>
<tr>
<td>World</td>
<td>378.9</td>
<td>14%</td>
<td></td>
</tr>
</tbody>
</table>
Available US Nuclear Waste

- The United States Department of Energy alone has 470,000 tonnes of depleted uranium. About 95% of depleted uranium is stored as uranium hexafluoride.

- The US currently has more than 75,000 metric tons of spent nuclear fuel stacked up at 122 temporary sites in 39 states across the US, according to DOE reports. The nation’s 104 commercial nuclear reactors produce about 2,000 tons of spent nuclear fuel annually. Thousands more tons of high-level military waste also need a final home.

- Natural uranium U₃O₈ costs $114,000/tonne today, $17,600 in 2001
  - yellowcake is 70-90% U₃O₈

- If 1 tonne /GW-y, all of US electricity (500 GW-y) can be provided by:
  - Spent fuel 75,000/500 = 125 years
  - Depleted uranium = 470,000/500 = 940 years
To continuously generate a power output of 1GW for a year requires:

- **3,500,000 tonnes of coal**
  - Significant impact upon the Environment
  - especially CO₂ emissions

- **200 tonnes of Uranium**
  - Low CO₂ impact
  - but challenges with reprocessing
  - and very long-term storage of hazardous wastes
  - **Proliferation**

- **1 tonne of Thorium**
  - Low CO₂ impact
  - Can consume Plutonium and radioactive waste
  - Reduced quantity and much shorter duration for storage of hazardous wastes
  - **No proliferation**
What does Carlo’s slide mean?
It compares power according to how much
you dig up and how you use it.

- Only 0.7% of natural uranium is U-235, which is
  - capable of self-sustaining nuclear fission (fissile),
  - (the only element that exists in nature in sufficient quantity…)
  - So you need to dig up over 143 tonnes of U to get 1 of U-235
  - Then you enrich it (using centrifuges, which have proliferation concerns)

- the rest is U-238, which, like thorium-232, is fertile, not fissile.
  - i.e. you need to provide neutrons to convert it to a fissile isotope.
  - (Criticality is the point at which a nuclear reaction is self-sustaining;
    subcritical means additional neutrons are needed)

\[
\begin{align*}
\text{fertile} & \quad \text{fissile} \\
\text{n} + ^{238}\text{U}_{92} & \rightarrow ^{239}\text{U}_{92} \\
\beta^- & \quad \beta^- \\
24 \text{ m} & \quad 2.4 \text{ d} \\
\text{n} + ^{232}\text{Th}_{90} & \rightarrow ^{233}\text{Th}_{90} \\
\rightarrow ^{233}\text{Pa}_{91} & \quad \rightarrow ^{233}\text{U}_{92} \\
22 \text{ m} & \quad 27 \text{ d}
\end{align*}
\]

30% LWR

May limit k<.97
The extra neutrons needed to convert fertile elements can be provided by:

- A fast or Breeder reactor using fissile U-235 or Pu-239, above criticality
- A particle accelerator – very hot topic 20 years ago!

What is new:

- Superconducting RF Linacs can provide extraordinary neutron flux
  - Can easily outperform breeder reactors
- The advantages of continuous purging of radioactive elements from the nuclear fuel are apparent from Fukushima (and TMI and Chernobyl)
- Molten salt fuel can be continuously purged in new reactor designs without zircaloy, that can lead to hydrogen explosions
- Molten salt fuel eases accelerator requirements

Subcritical ADSR operation has always been appreciated
- Fission stops when the accelerator is switched off
Three Mile Island was a lesson unlearned; Fukushima has provided perhaps several more

- At Fukushima, perhaps 6 separate cases of things going wrong:
- 3 reactor explosions, (perhaps spreading radioactive uranium oxide fuel components over at least a mile),
- fuel in the bottom of 2 of these reactors then melted through the bottom of their pressure vessel.
- At least one storage pond went dry enough to expose used fuel rods so they got hot enough to release radioactivity.
  - After fission stops, heat from decays in rods is ~5% of operating level
  - (17,600 tons of spent fuel stored in ponds at Fukushima)

These events released enough radioactive material for class 7 status, with almost 10% of the fallout caused by Chernobyl, but without a criticality accident.
- Fukushima Dai-ichi reactors - 6 BWR-type Light Water Reactors –
  - #1, #2 and #3 turned off (scrammed), #4, #5 and #6 were off at the time of earthquake and tsunami. Radiation was released from 1, 2 and 3 and a storage pool.
  - Fuel melts through the bottom of pressure vessel in #2 and #3

Cited from NY Times
Fuel Rods of Conventional Reactors
are Fuel Rods an intrinsic problem?

Fuel rods are made of many small cylinders of enriched UO$_2$ or mixed oxide fuel (MOX) enclosed in a sheath of zirconium alloy.

- (a plant in France processes spent fuel rods to extract Pu$_{239}$, which is mixed with UO$_2$ to make MOX. Remains are returned to country of origin.)

- During operation, many radioactive elements are created that are contained by the zircaloy sheath

- If, during operation or storage, the zircaloy casing is damaged, these radioactive elements can be released and among other things scare the heck out of a lot of people. (fall-out near Fukushima may be 10% of Chernobyl).

- Radioactive Fission Products Partially Released from Damaged Fuel
  - Noble gases (Xe, Kr)
  - Volatile fission products (I, Sr, Cs, Ru, …)
  - Non-volatile fission products retained, but may be leached by water

- Hot zircaloy itself is a hazard – it can oxidize in steam to release hot H$_2$ in large quantities, which can explode when it rises to meet air.
  - Zr + 2 H$_2$O $\rightarrow$ ZrO$_2$ + 2 H$_2$
  - Exothermic
  - rate increases exponentially with temperature
Fuel Rods an intrinsic problem? (cont.)

- It will be more and more apparent that stored used fuel rods are not without risk. Losing coolant in these could cause zircaloy failures that could lead to released volatile radioactive elements.

- For reactors, there are lots of layers of protection that have been invented and used to mitigate the problems that follow from solid fuel rod technology.
  - See latest iteration on next slide.

- Is there an intrinsic safety solution?

- Like the manhole cover to protect workers below?
  - e. g. Trap door → safety chain → procedures → for safety
  - Or just making the hole round with a round cover of larger diameter?
Safety systems for conventional solid fuel reactors are still evolving
AREVA Evolutionary Power Reactor
http://en.wikipedia.org/wiki/European_Pressurized_Reactor

The EPR's main safety systems

- Double wall containment with ventilation and filtering systems
- Water tank inside containment
- Molten core spreading area
- Ultimate containment heat removal system
- 4-train redundancy of the main safety systems
• An intrinsic safety problem for conventional reactors is enclosed solid fuel.

• a natural solution is to use molten-salt fuel

• that is also well suited to accelerator-driven subcritical reactors.
  • A major difficulty is fatigue of UO$_2$ fuel in rods caused by accelerator trips – no such problem for molten salt fuel

• The technology of molten-salt fuel was developed in the 1960s in the Molten-Salt Reactor Experiment (MSRE) at ORNL.
  • Use of molten salt fuel was later abandoned
    • not enough Pu-239 for bombs?
    • President Nixon?
  (See MSRE on wikipedia for nice summary)
Molten-Salt Reactor Experiment

Glowing radiator
Molten-salt Reactor Experiment
"The MSRE is an 8-MW(th) reactor in which molten fluoride salt at 1200°F (650 C) 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 235U (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 235U 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 shutdown 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 233U.

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."

NOW FAST FORWARD 40 YEARS and add an accelerator
GEM*STAR concept without fuel reprocessing

How best to solve the dilemma of accumulated spent fuel depends on assumptions.

Because of nuclear weapon proliferation concerns, the USA decided not to reprocess spent fuel (i.e. separate into its components).

If this is to be the policy in the future, one possible approach to eliminate spent fuel is to consider iterations of fuel burning where neutron absorption by accumulated fission products (FP) is compensated by higher neutron flux. (not easy with a fast or breeder reactor)

This implies successive particle accelerator generations produce neutrons more efficiently.

First, spent UO₂ fuel is converted to UF₄ salt, then

- Gen 1 SRF
  - UF₄ salt -> GEM*STAR -> UF₄ outflow -> GEM*STAR -> UF₄ outflow
  - with more FP

- Gen 2 SRF
  - UF₄ outflow -> GEM*STAR -> UF₄ outflow
  - with more FP

- Gen 3 SRF
  - GEM*STAR -> etc.

After 5 years, the GEM*STAR has reached equilibrium, and its output can start a second unit.
GEM*STAR concept without fuel reprocessing

In all cases, the accelerator uses less than 15% of the generated power.

<table>
<thead>
<tr>
<th>Cycle</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_{eff}$</td>
<td>0.99</td>
<td>0.95</td>
<td>0.90</td>
<td>0.83</td>
<td>0.77</td>
</tr>
<tr>
<td>Start date</td>
<td>2020</td>
<td>2060</td>
<td>2100</td>
<td>2140</td>
<td>2180</td>
</tr>
<tr>
<td>Neutron source</td>
<td>Acc.1</td>
<td>Acc.2</td>
<td>Acc.3</td>
<td>Fusion 1</td>
<td>Fusion 2</td>
</tr>
<tr>
<td>End date</td>
<td>2060</td>
<td>2100</td>
<td>2140</td>
<td>2180</td>
<td>2220</td>
</tr>
<tr>
<td>Neutron multiplication</td>
<td>100</td>
<td>20</td>
<td>10</td>
<td>6</td>
<td>4</td>
</tr>
<tr>
<td>Relative neutron cost</td>
<td>1</td>
<td>5</td>
<td>5</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>Energy-weighted LWR waste remnant</td>
<td>0.5</td>
<td>0.324</td>
<td>0.183</td>
<td>0.114</td>
<td>0.068</td>
</tr>
</tbody>
</table>

How best to solve the dilemma of accumulated spent fuel depends on assumptions.

**Figure 11**
Five successive burn cycles for LWR spent fuel in GEM*STAR. The top line shows $k_{\text{eff}}$ for each of the five cycles. The bottom line shows the cumulative burn-up fraction for the five cycles. Note that each subsequent cycle consumes a larger fraction of the remaining $^{238}\text{U}$. The middle line shows the cumulative energy extracted from five successive recycles of LWR spent fuel. Note that if the total energy generated from the fresh fuel sent into the LWR is of interest, one must add 1.0 to the right ordinate. In each case, the power to produce the neutrons is assumed to never exceed 15% of the output power (see Table 9).
An Accelerator-Driven Subcritical Reactor Example with Molten Fuel (ThF$_4$ or UF$_4$)

Conceptual design of the GEM*STAR reactor in its underground placement. The vertical dimension is about 30 ft. The gray box is the graphite reflector for the core. Horizontal beams from two accelerators are shown at the top being bent by magnets about 45 degrees into the core where both strike a uranium metal target shown schematically in the center of the core.
GEM*STAR ADSR Molten-Salt Example

• GEM*STAR is shown schematically on the next slide.
  • Charles D. Bowman, et al. GEM*STAR: Handbook of Nuclear Engineering,
• The graphite core shown in gray surrounded by a reflector.
• The molten salt fuel takes up about 7% of the core volume and it is shown in red outside of the core. (less than critical mass!)
• The fuel flows upward to a free surface above the core and over to the sides where it is pumped down as shown on the left to the bottom of the unit. It turns upward and then horizontally and reenters the core through apertures in the bottom reflector.
• Heat is removed by a secondary (non-fissile) salt of lower melting point as shown on the right. (A reservoir can be added for reliability)
• The secondary salt flows downward on the inside of an array of pairs of concentric tubes, turns the corner at the bottom and flows upward through the outer tube with heat flow through the outer tube wall from the fuel salt to the secondary salt. A secondary salt reservoir is possible.
• The secondary salt then flows through a steam generator.
GEM*STAR Technology

- Electric motors
- He in
- He out
- UF₄ or fluorinated LWR spent fuel + ⁷LiF carrier
- Steam generator
- Molten salt pumps
- 750°C
- Molten salt
- Salt overflow
- Graphite/salt
- 650°C out
- Target
- Reflector
- Secondary salt loop with concentric piping
- 550°C in
- Turbine/generator
- Modified Hastelloy-N or graphite encloses all fuel salt
- Graphite reflector
- 650°C
- Steel base plate

500 MWt
220 MWe
• The maximum temperatures are 750 C for the fuel salt at the top of the core, 650 C for the secondary salt exiting the core and 550 C for the steam entering the turbine.

• The expected thermal-to-electric conversion efficiency exceeds 44%.

• Fuel is fed in liquid form at the rate of about 1 liter per hour for a power production of 220 MWe. The vertical pipe shown allows the fuel to overflow into an inner tank and then to an outer tank below the reactor.

• The tanks have storage capacity for forty years of fuel overflow. The overflow can be fed to another GEM*STAR unit.

• More than one internal target for neutron production will be normally present in the core instead of the external targets shown schematically.

• A flow of He across the salt surface above the core enables the prompt collection and removal of noble gases for storage away from the core so that the inventory of volatile fission products in the core is reduced by about 10 million from that of an LWR of the same power.
Status of Superconducting RF

• Discussed at SRF Workshop at SRF11.

• 20 years ago, the required power was not possible with any accelerator technology

• Several CW hadron Linacs can now be considered for ADSR
  • The International Fusion Materials Irradiation Facility (Japan)
  • note 125 ma (1 GeV gives 125 MW beam power!)
  • Like the LEDA RFQ proof-of-principle
  • 1 Linac is enough for 25 1st-Gen GEM*STARS or 5.5 GWe
  • MYRRHA (Belgium)
  • Japan ADS
  • Indian ADS
  • China ADS/SNS
  • Project-X?
Beam Power Frontier for ion beam accelerators

Adapted from Sang-Ho Kim, SRF11

12.5 mA CW
Conclusions: SRF Linacs with today’s technologies* can drive an ADSR with Molten-Salt-Fuel to simultaneously address
- elimination of dangerous stored nuclear waste
- production of safe, environmentally-friendly energy

ADSR nuclear power stations using molten salt fuel operate
• in an inherently safe region below criticality,
• without accidental releases of radioactive volatile elements,
• without generation of greenhouse gases,
• producing minimal nuclear waste,
• without byproducts useful to rogue nations or terrorists,
• fueled by and eliminating existing stockpiles of
  • LWR nuclear waste and depleted uranium
• and/or efficiently using abundant natural thorium or uranium,
  • which does not need enrichment.

*Molten-salt fuel allows an end-run around the solid fuel fatigue problem so that short-term accelerator trips are not important. Non-radioactive salt heat transfer reservoirs allow multi-hour interruptions.