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ACCELERATORS FOR SUBCRITICAL MOLTEN-SALT REACTORS

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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 in dangerous amounts. 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 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

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. UF_4 or ThF_4)
  - 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

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Nuclear Power Capacity as of 02/2012

Country	# reactors	GW capacity	Nuclear share of electricity production
Belgium		5.9	51.7%
Canada		12.7	14.8%
China (PRC)		10,2	1.9% ←
France	59	63.2	75.2%
Germany		20.3	26.1%
India		4.8	2.9% ←
Japan	54	47.3	28.9%
Korea, South		18.7	31.1%
Russia		23.0	17.8%
Spain		7.4	17.5%
Sweden		9.4	37.4%
Taiwan		4.9	20.7%
Ukraine		13.2	48.6%
United Kingdom		11.0	17.9%
United States	104	101.2	20.2%
Rest of World		25.4	
World		378.9	14%

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 U_3O_8 costs \$114,000/tonne today, \$17,600 in 2001
  - yellowcake is 70-90% U_3O_8

- 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

Carlo Rubbia

Comparing alternatives

AkerSolutions

To continuously generate a power output of 1GW for a year requires:

3,500,000 tonnes of coal

Significant impact upon the Environment

especially CO_2 emissions

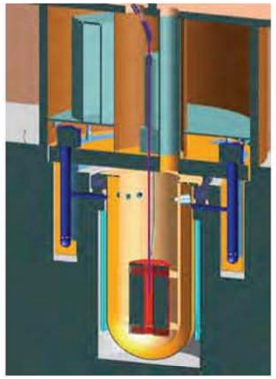
200 tonnes of Uranium

Low CO_2 impact

but challenges with reprocessing

and very long-term storage of hazardous wastes

Proliferation



1 tonne of Thorium

Low CO_2 impact

Can consume Plutonium and radioactive waste

Reduced quantity and much shorter duration for storage of hazardous wastes

No proliferation

C.Rubbia2, Energy 2050, Stockholm

Slide# : 16

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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)

fertile $\qquad \beta^-$ $\qquad \beta^-$ *fissile*

- $n + {}^{238}U_{92} \rightarrow {}^{239}U_{92} \xrightarrow[24\ m]{} {}^{239}Np_{93} \xrightarrow[2.4\ d]{} {}^{239}Pu_{94}$ ← 30% LWR
- $n + {}^{232}Th_{90} \rightarrow {}^{233}Th_{90} \xrightarrow[22\ m]{} {}^{233}Pa_{91} \xrightarrow[27\ d]{} {}^{233}U_{92}$ ← 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 or
  - A particle accelerator – very hot topic 20 years ago!

- What is new:
  - Superconducting RF 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

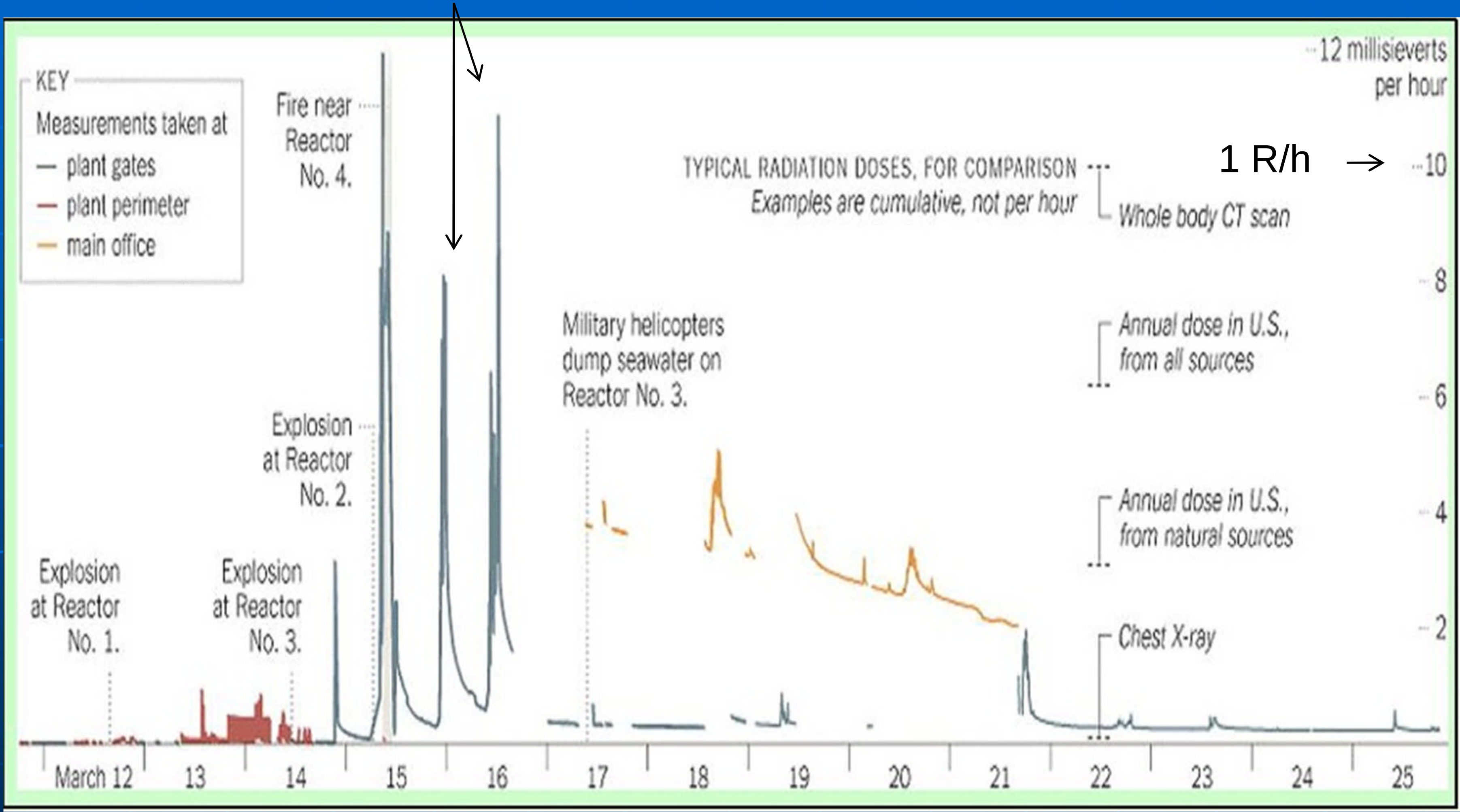
Three Mile Island was a lesson unlearned; Fukushima has provided perhaps several more

- 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

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Fuel Rods of Conventional Reactors

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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.
    - $Zr + 2\ H_2O \rightarrow ZrO_2 + 2\ H_2$
    - Exothermic
    - rate increases exponentially with temperature

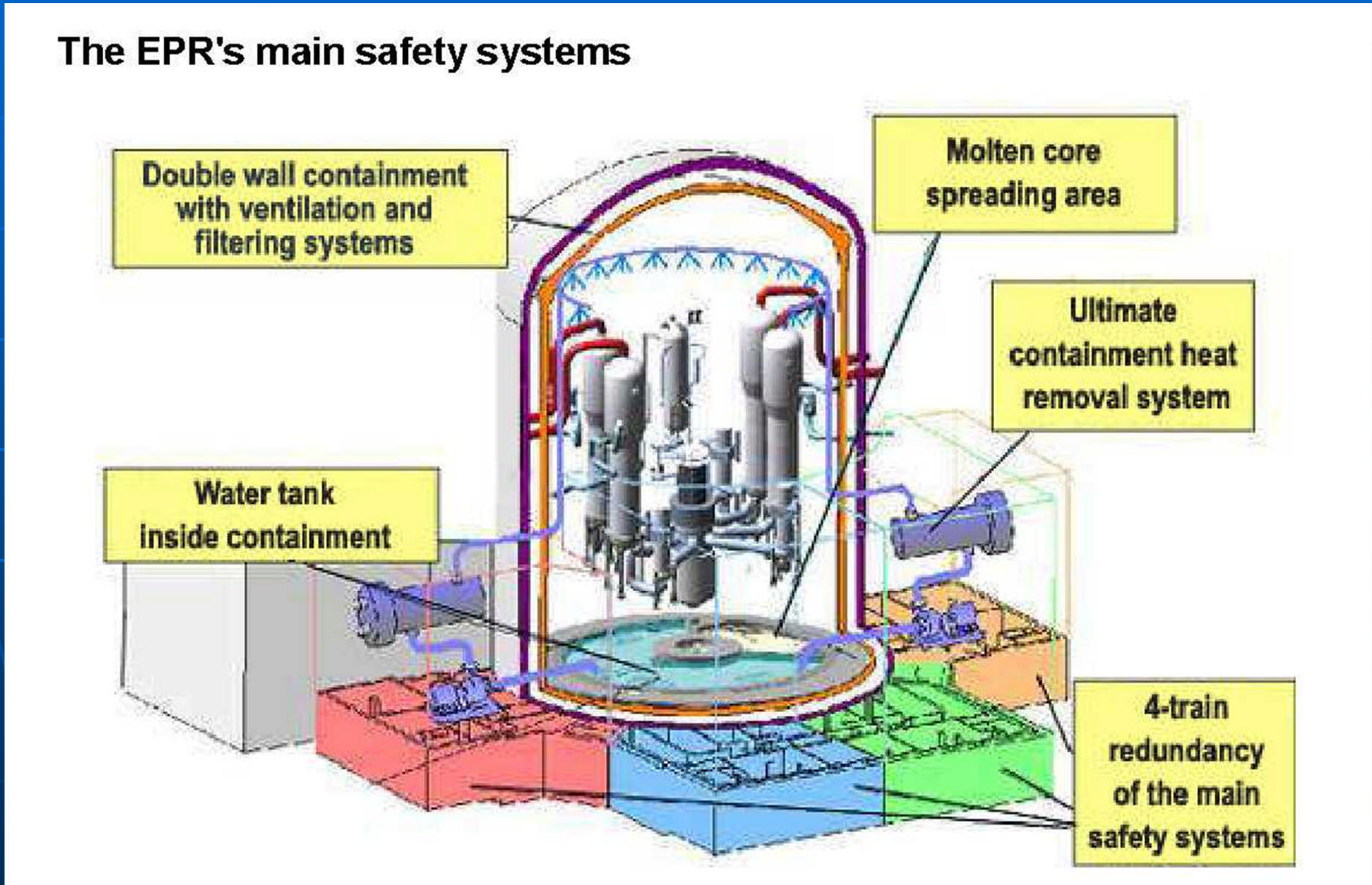
Fuel Rods an intrinsic problem (cont.)

- It will be more and more apparent that used stored 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



- An intrinsic safety problem for conventional reactors is enclosed solid fuel.
- a natural solution is to use <u>molten-salt fuel</u>
- 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 because the technique did not produce enough Pu-239 for bombs.

      (See MSRE on wikipedia for nice summary)

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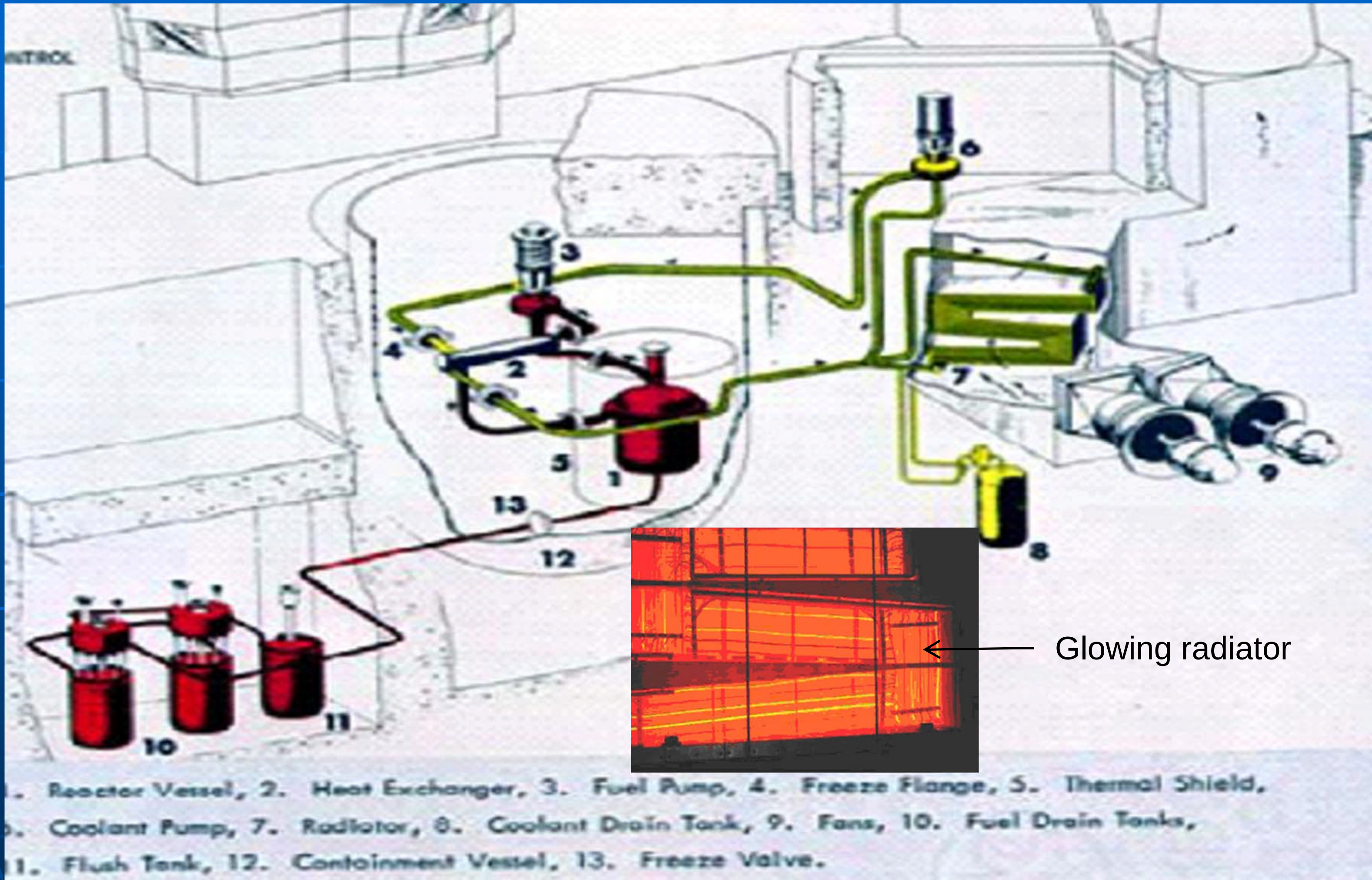
Molten-Salt Reactor Experiment



Molten-salt Reactor Experiment

From 1969 MSRE Report Abstract

“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 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 the accumulated spent fuel depends on assumptions.

Because of nuclear weapon proliferation, the USA decided not to reprocess spent fuel.

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 the build-up of fission products (FP) is compensated by higher neutron flux. (not possible 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   Gen 2 SRF   Gen 3 SRF

*F4 salt -> GEM*STAR -> UF4 outflow -> GEM*STAR -> UF4 outflow-> GEM*STAR-> etc.*

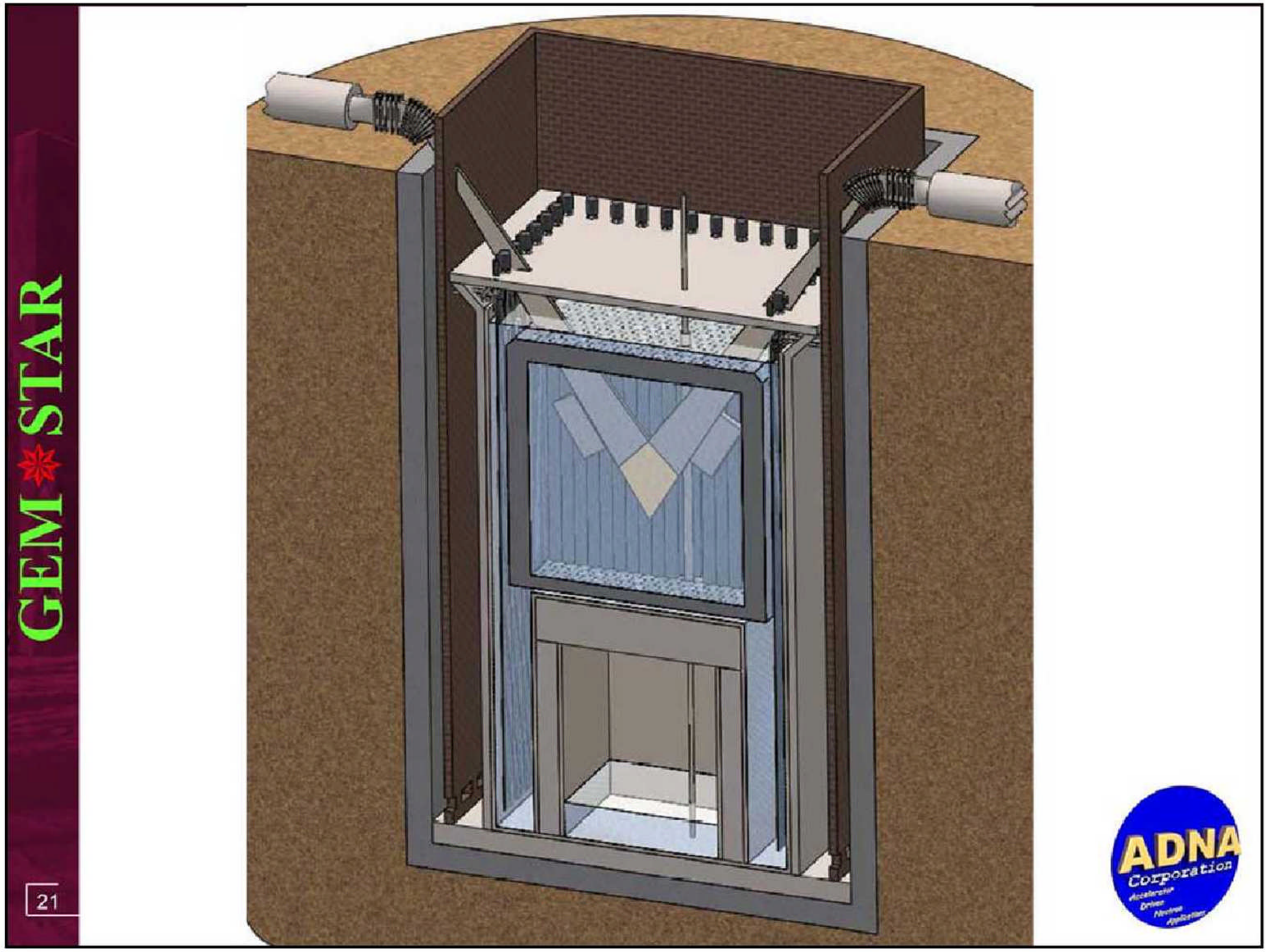
with more FP   with more FP

*After 5 years, the GEM*STAR has reached equilibrium, and its output can start a second unit.*

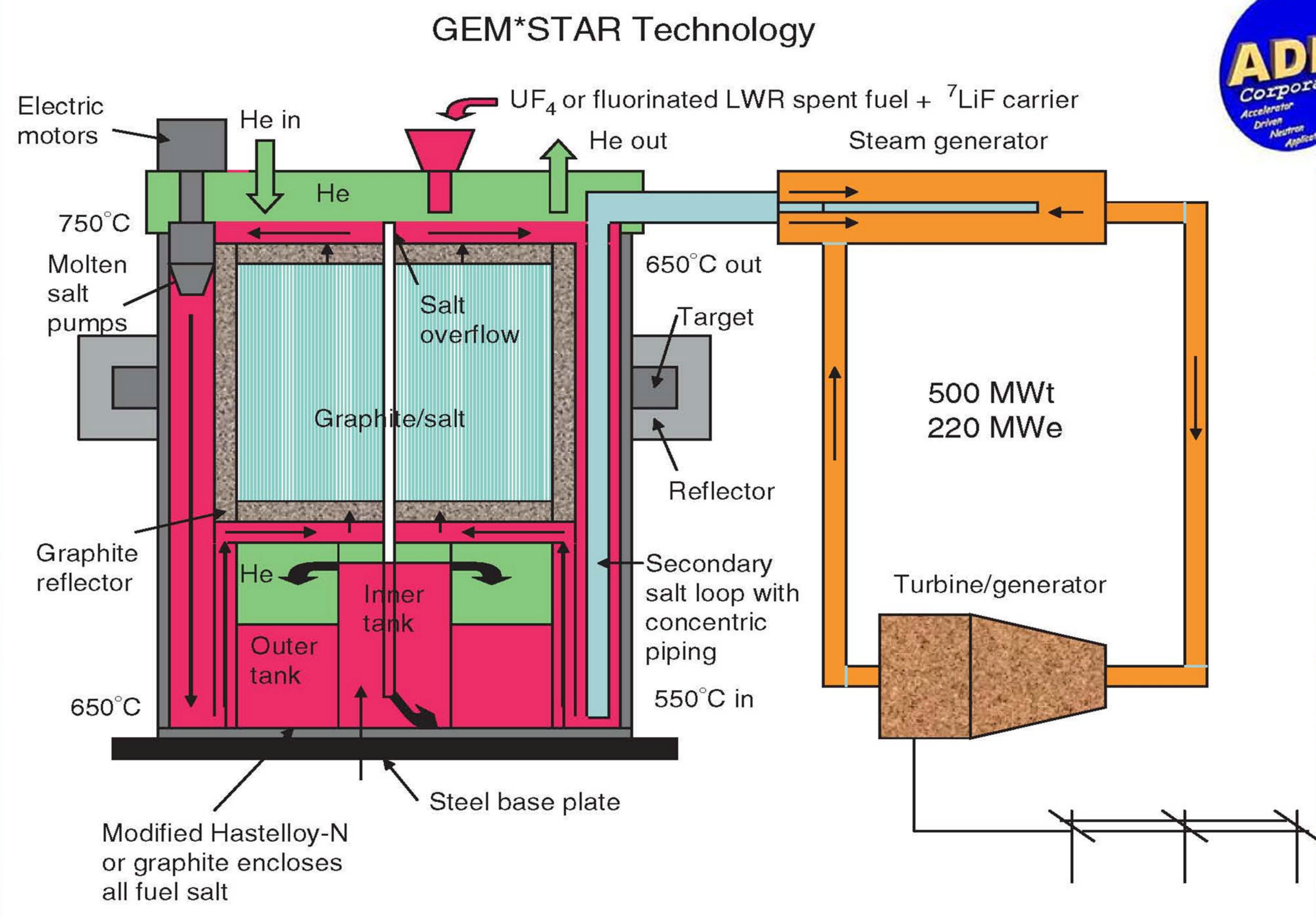
LWR fuel repeated recycle summary

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

An Accelerator-Driven Subcritical Reactor Example with Molten Fuel (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 Technology
Electric motors
He in
UF_4 or fluorinated LWR spent fuel + ^7LiF carrier
He out
Steam generator
He
750°C
Molten salt pumps
Salt overflow
650°C out
Target
Graphite/salt
500 MWt
220 MWe
Reflector
Graphite reflector
He
Inner tank
Outer tank
Secondary salt loop with concentric piping
Turbine/generator
650°C
550°C in
Steel base plate
Modified Hastelloy-N or graphite encloses all fuel salt

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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.
- 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 ADSR Molten-Salt Example (cont.)

- 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 <u>inventory of volatile fission products in the core is reduced by about 10 million from that of an LWR of the same power</u>.

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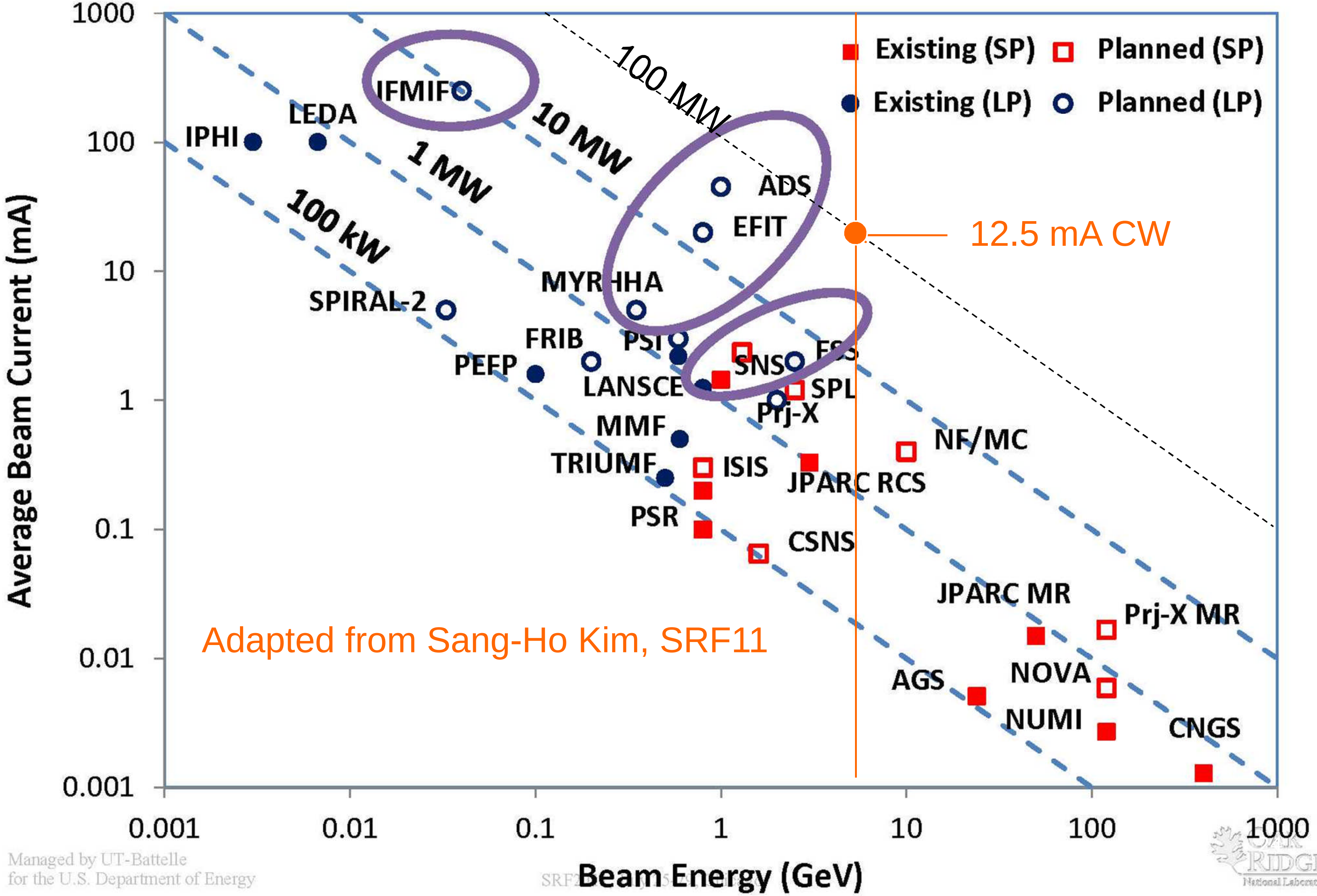
Status of Superconducting RF

- Discussed at SRF Workshop at SRF11.
    - http://conferences.fnal.gov/srf2011/
- 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 125ma
- MYRRHA (Belgium)
- Japan ADS
- Indian ADS
- China ADS/SNS
- Project-X?

Beam Power Frontier for ion beam accelerators



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.