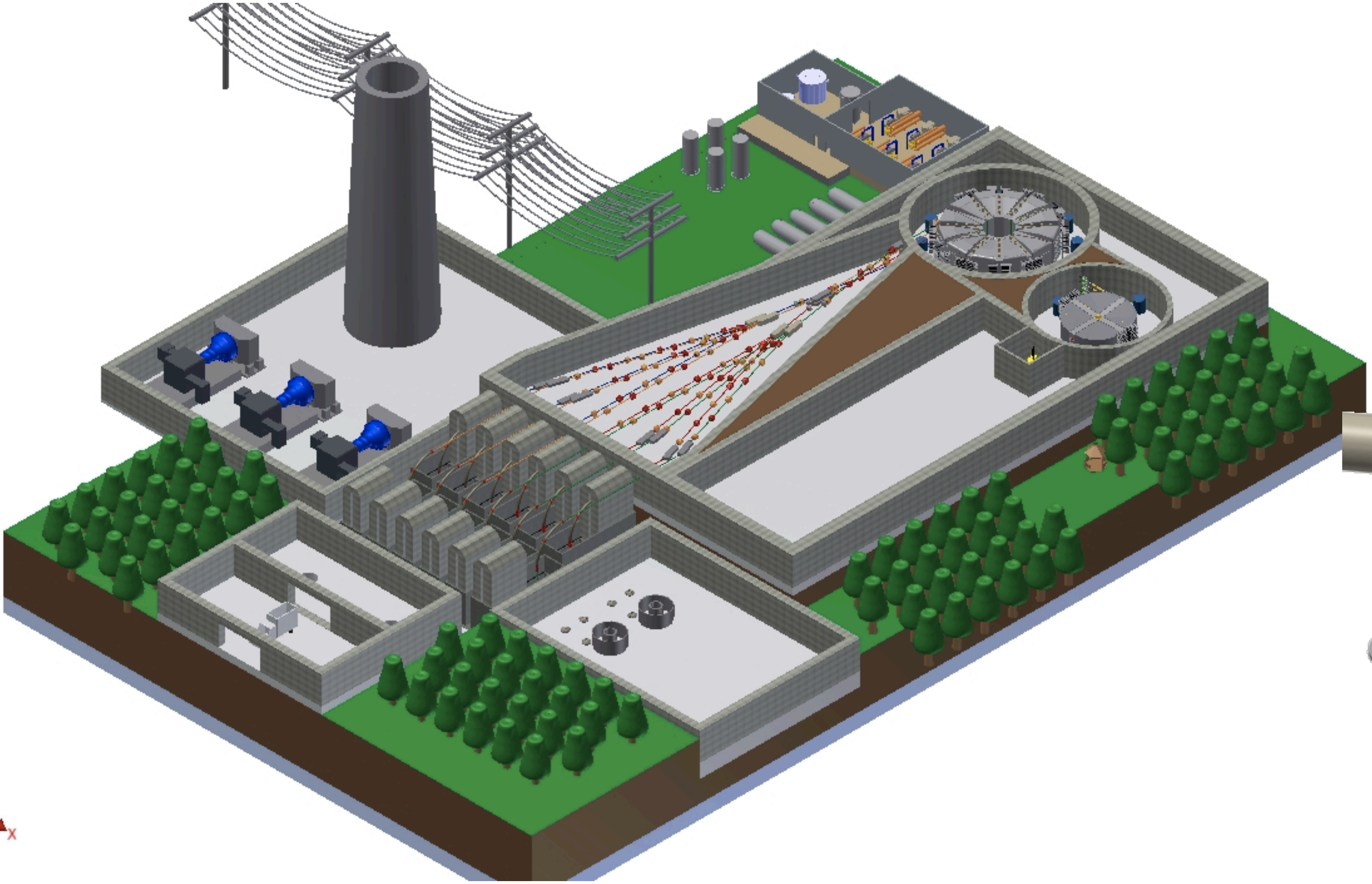
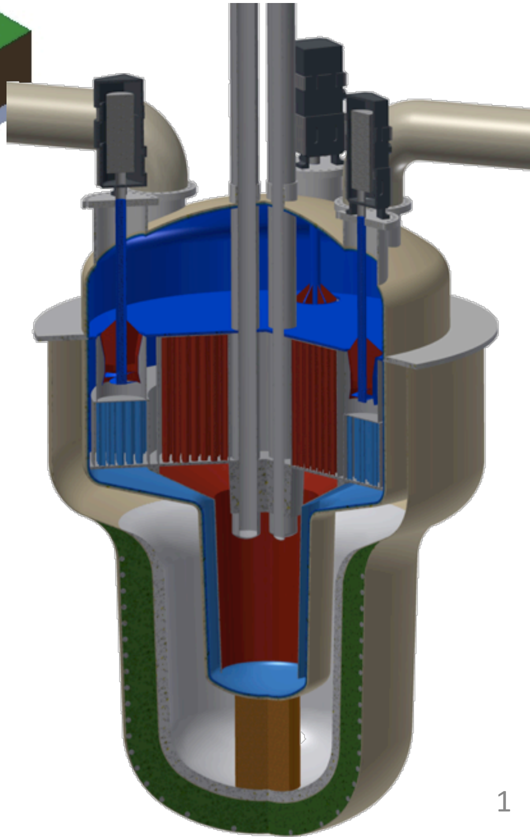
Accelerator-Driven subcritical fission in A Molten salt core: Closing the Nuclear Fuel Cycle for Green Nuclear Energy





Peter McIntyre, Texas A&M University

For the ADAM Collaboration

ADS Fission in a Molten Salt Core

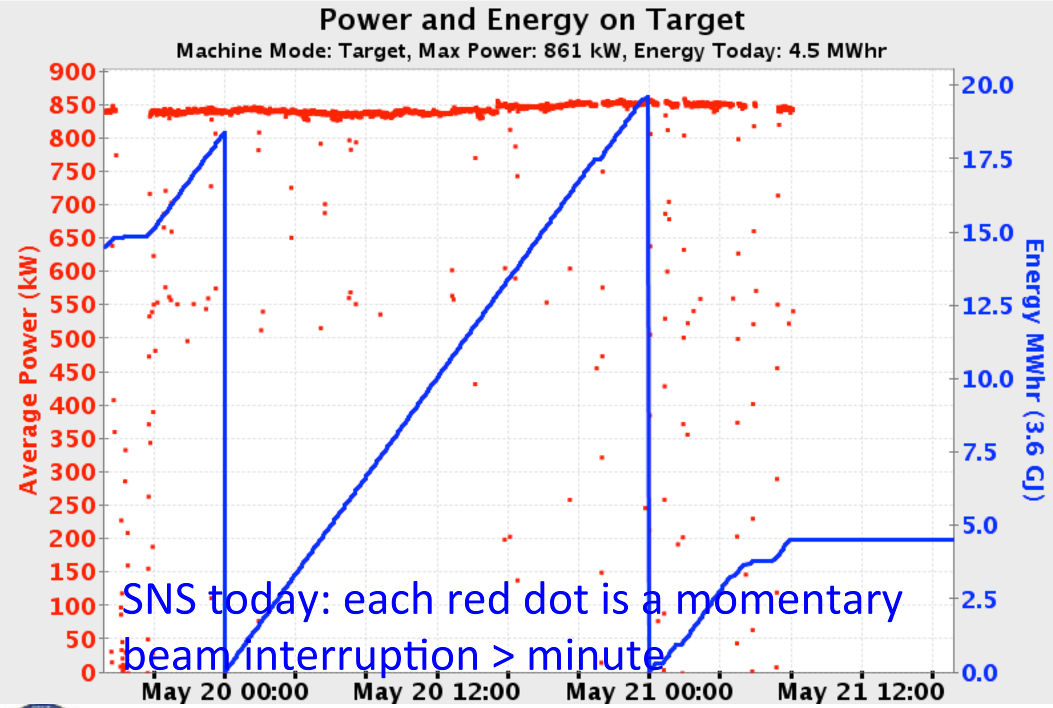
- Extract the minor actinides and long-lived fission products from spent fuel into molten salt
  - Pyroprocessing and electroseparation
  - Developed at ANL, INL, PRIDE
  - Never separate Pu from other TRU

- Fast neutronics in a subcritical moletn salt core
  - Fastest neutron spectrum ever designed $\langle E_n \rangle = 1$ MeV
  - Burns all the transuranics together at the same rate
  - No thermal shock when drive beam is interrupted
  - Cannot go critical, cannot overtemp even if power fails

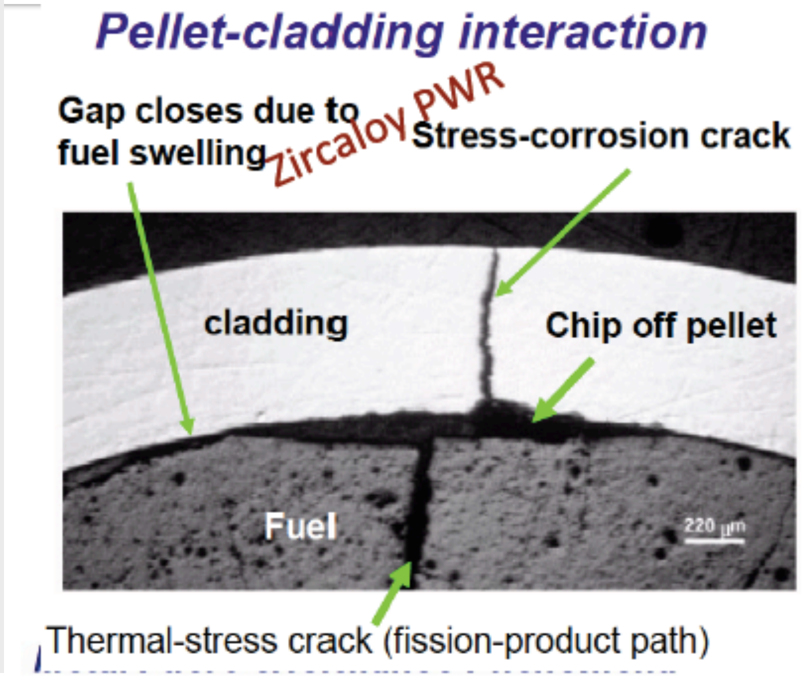
ADS has a rich history of ideas

- 1958 Ernest Lawrence invented the idea, and gave it as an argument in the proposal to AEC for funding the Alvarez Linac.
- 1980 Bob Wilson dubbed it electroproduction, and conceived using higher-energy protons to drive fission.
- 1995 Carlo Rubbia revived the idea and posed it as a basis for thorium-cycle fission power.
- Today MYRRHA is developing a first research system to explore the parameters for subcritical cores.
- We are adding two important elements:
  - A strong-focusing cyclotron that can deliver 40 MW of proton beam with high energy efficiency
  - A molten salt core that provides for high-power targetry and avoids issues of thermal shock from accelerator interruptions.

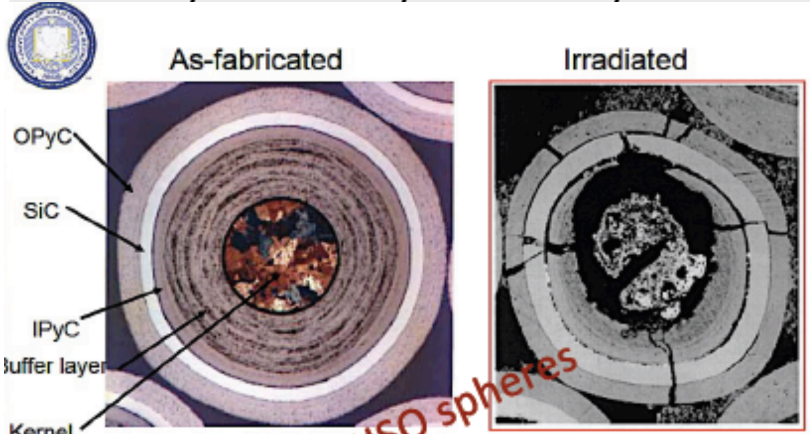
Molten salt fuel eliminates thermal shock



SNS today: each red dot is a momentary beam interruption > minute



Zircaloy PWR



TRISO spheres

- Xe and Kr released from fuel kernel
- oxygen liberated from $(U,Pu)O_{2-x}$
- $2C + O_2$ → 2CO [illegible] of carbon in buffer layer

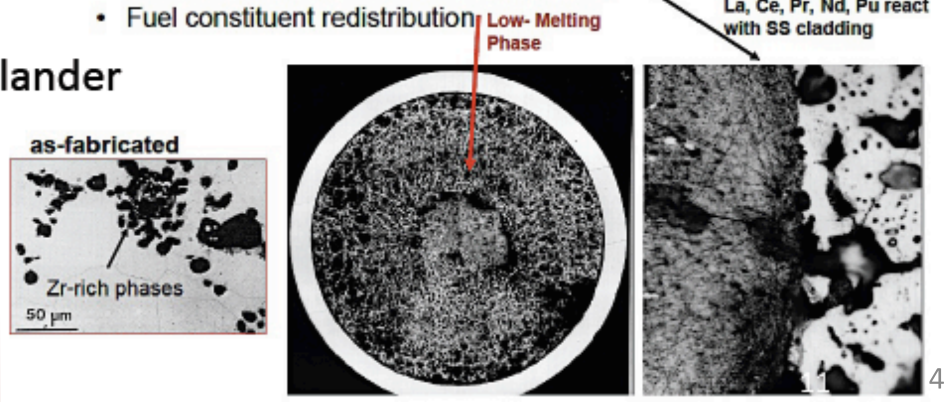
Probably failed by overpressure due to fission gases and CO

ADSMS

- Irradiation growth: ~ 3% at 14% burnup of metal atoms
- Fuel swelling and fuel-cladding mechanical interaction (FCMI)
- Gas release
- Fuel-cladding chemical interaction (FCCI)
- Fuel constituent redistribution

Metal fuel for HTGR

D. Olander

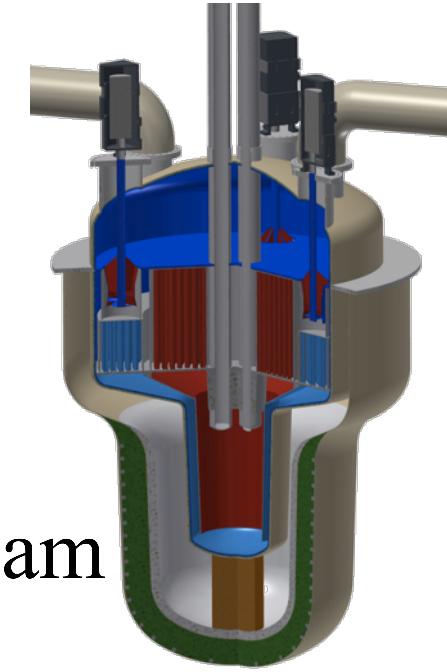


A molten salt core optimizes TRU-burning

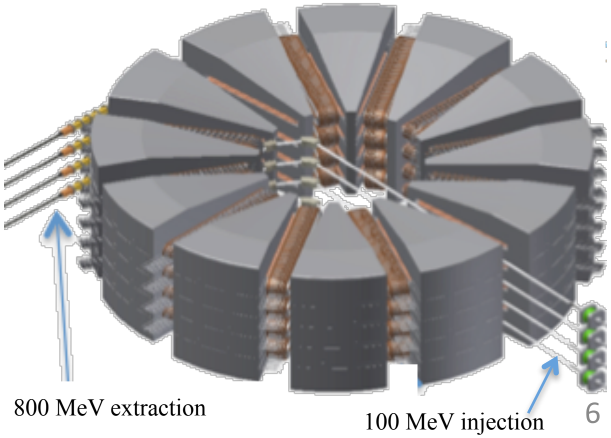
- The TRU contents can be extracted from UNF using pyroprocessing technology developed at ANL and INL.
- The molten salt serves as spallation target, moderator, and fissile inventory.

- The molten salt flow on the beam window makes delivery of a 2.7 MW proton beam realistic.
- The core is designed to provide passive cooling of decay heat in event that HX flow were lost.

- Molten salt core – simple to fuel, simple to recycle
  - Every 3 months add 90 kg of TRU to replace what was burned
  - Every 5 years, transfer fuel salt from core to remove fission products, then return to core
  - Fuel salt is 100% contained in 5 layers for 5 years of operation

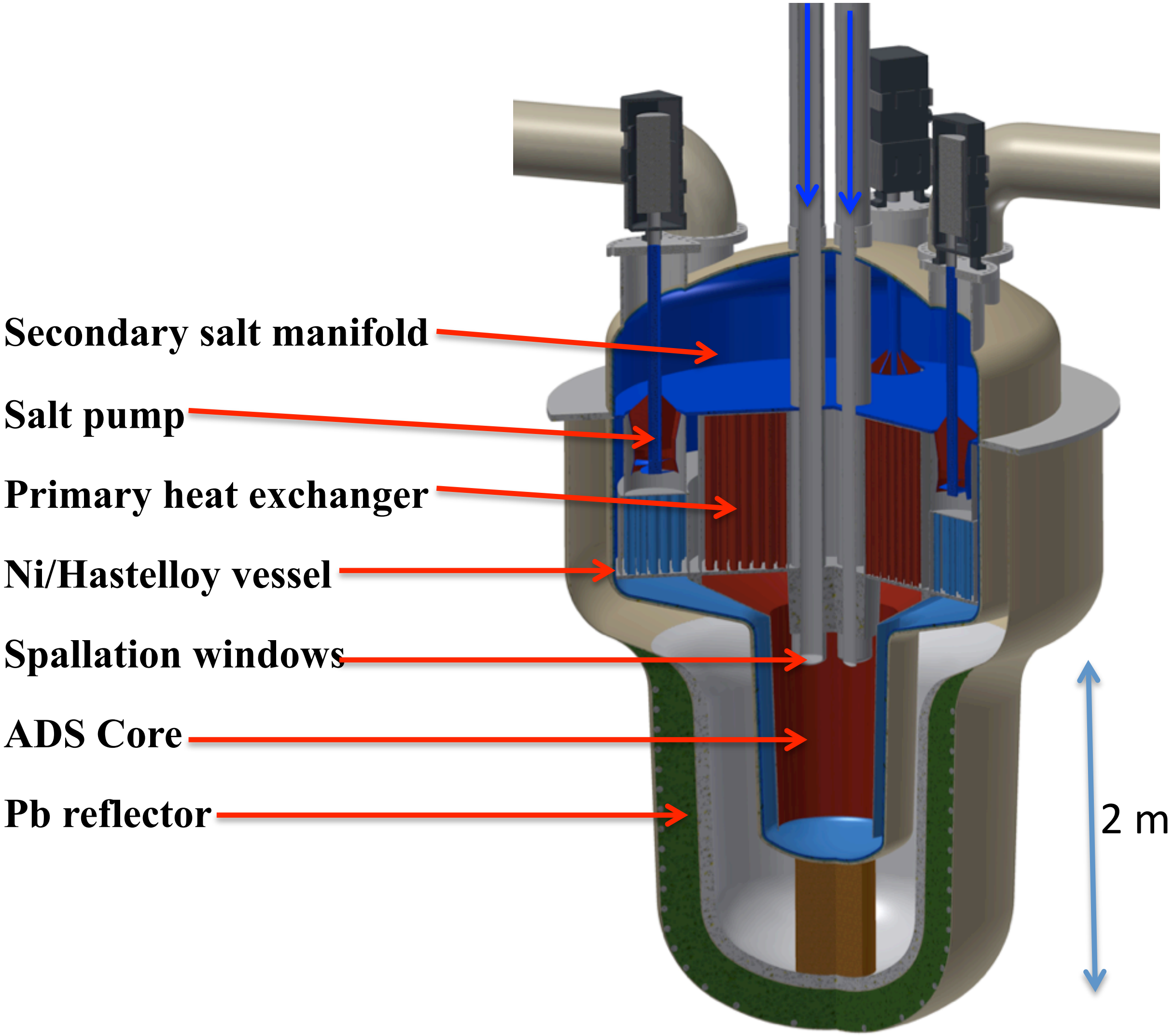


- Drive the subcritical core with proton beam
  - Stack of 3 cyclotrons
  - Drives 3 ADSMS cores
  - Modulate current 9➔12 mA for const P_t
  - **5:1 Energy Amplifier**



290 MW ADAM Core

three 2.8 MW proton drive beams



Molten salt fuel:

70 NaCl – 15 $TRUCl_3$ – 13 UCl_3

Fast fraction 20% E_n>1 MeV

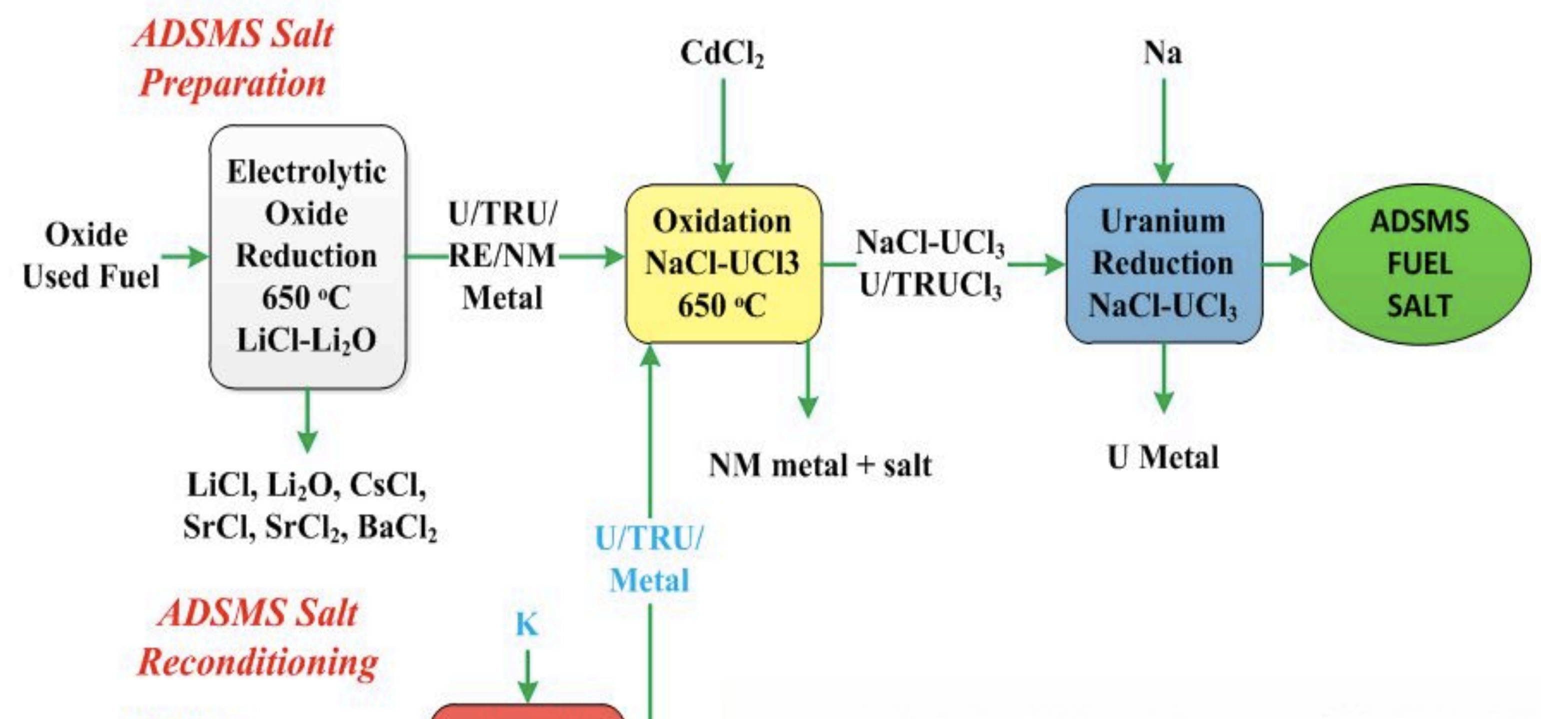
All fuel salt in one vessel

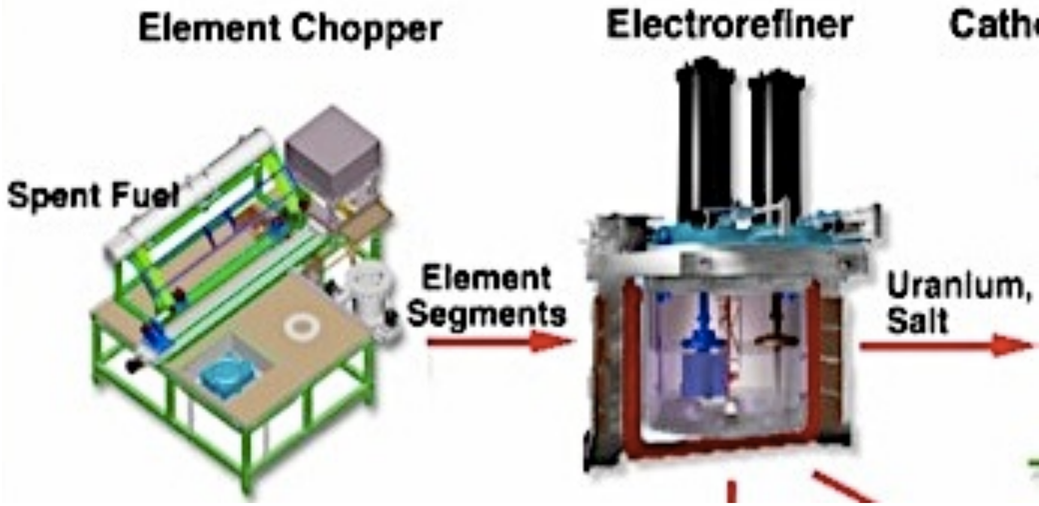
575 –675 C operating temp

The molten salt chemistry is important

- LiF-based salts were used in the original MSRE, and have been proposed for many designs of critical and subcritical molten salt cores.
- LiF has several problems for a TRU-burner:
  - The light elements moderate the neutron spectrum;
  - Multiple ionization states of TRU elements are metastable, including volatile species (analogs of UF_6).
  - LiF is corrosive, which presents a challenge for the lifetime of core vessel and HX components.
  - Loading the necessary mole% of TRU would push a F-based salt beyond the eutectic limit at reasonable operating temp – TRU salt could drop out of the mixture if the salt freezes.
- ***All of these issues are resolved by using $TRUCl_3$-NaCl.***

Extracting TRU from UNF fuel bundles

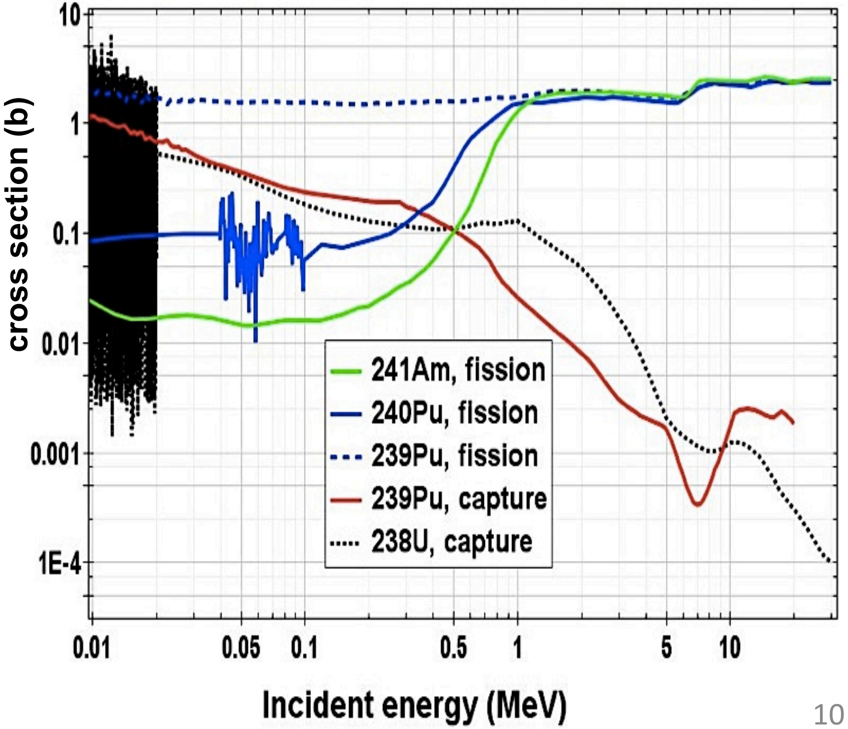




The PRIDE facility in ROK has developed to pilot-industrial scale

Neutronics for Isoburning

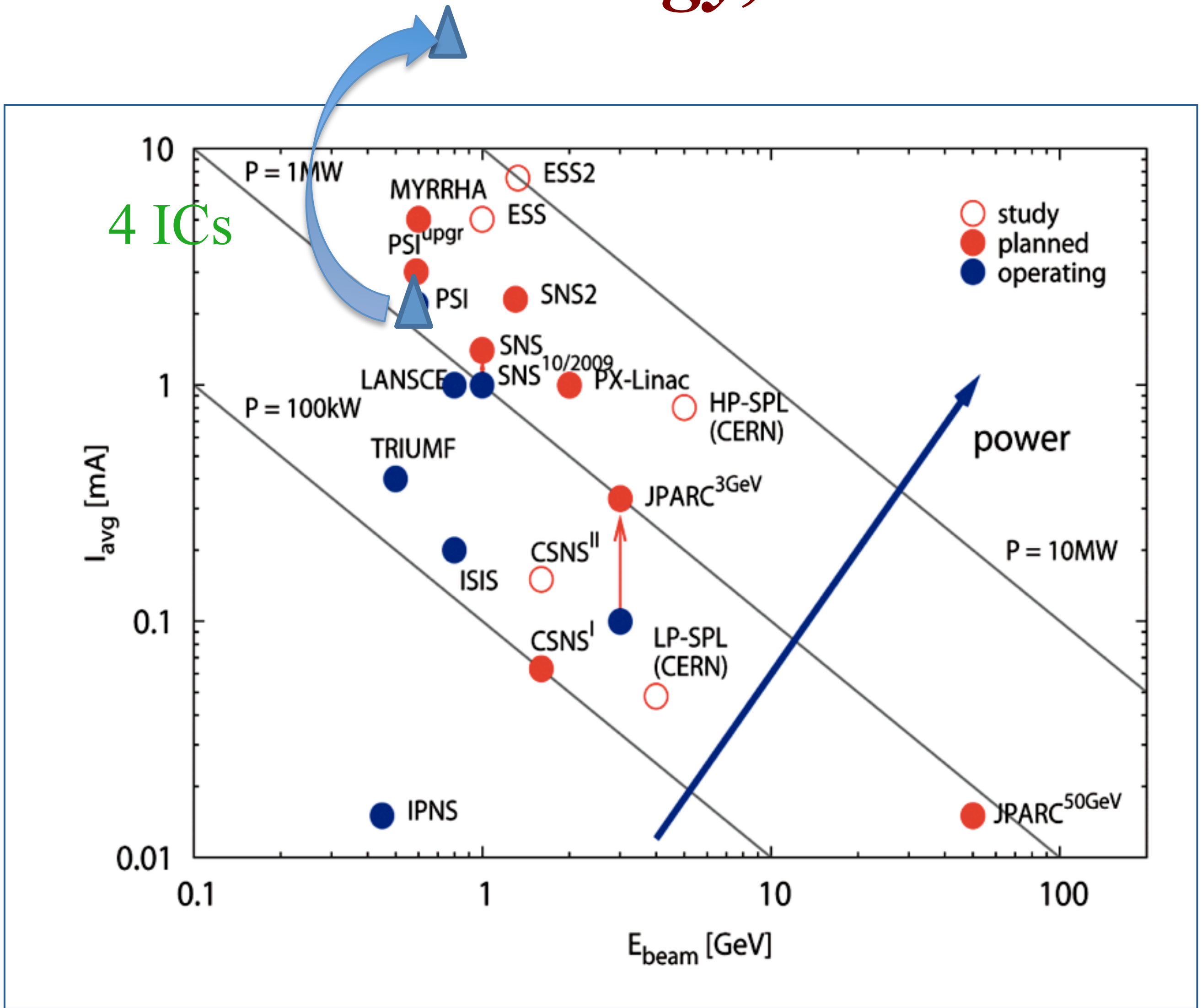
- One batch of UNF has a ton of ^{239}Pu
- Non-proliferation – keep Pu with intensely radioactive ingredients – TRU, FP
- Strategy – *we extract all the TRU elements together from UNF; we destroy them together*
- The fission cross-sections for Pu, TRU are equal for $E_n > 1$ MeV
- But for $E_n < 1$ MeV MA fissions 10 times less than ^{239}Pu



Choice of criticality k_{eff}

- We need to run the core subcritical
  - ^{239}Pu has 3x fewer delayed neutrons than ^{235}U
  - ^{241}Am has 5x fewer delayed neutrons than ^{235}U
  - ^{239}Pu fissions faster than ^{241}Am ➔ neutronics shifts
  - TRU-burning is a challenge for any critical core design.
- Suppose cooling is lost…
    - Passive heat pipes remove decay heat
    - The salt cannot freeze – k_{eff} has strong negative temp coeff.
- Design core to operate with $k_{eff} = 0.97$.
- Core cannot go critical under any of the many failure modes considered.
- But we need lots of proton drive…

Now for the proton driver: To destroy the TRU produced by a GW_e power plant, we need proton drive with 800 MeV energy, 30 MW CW!



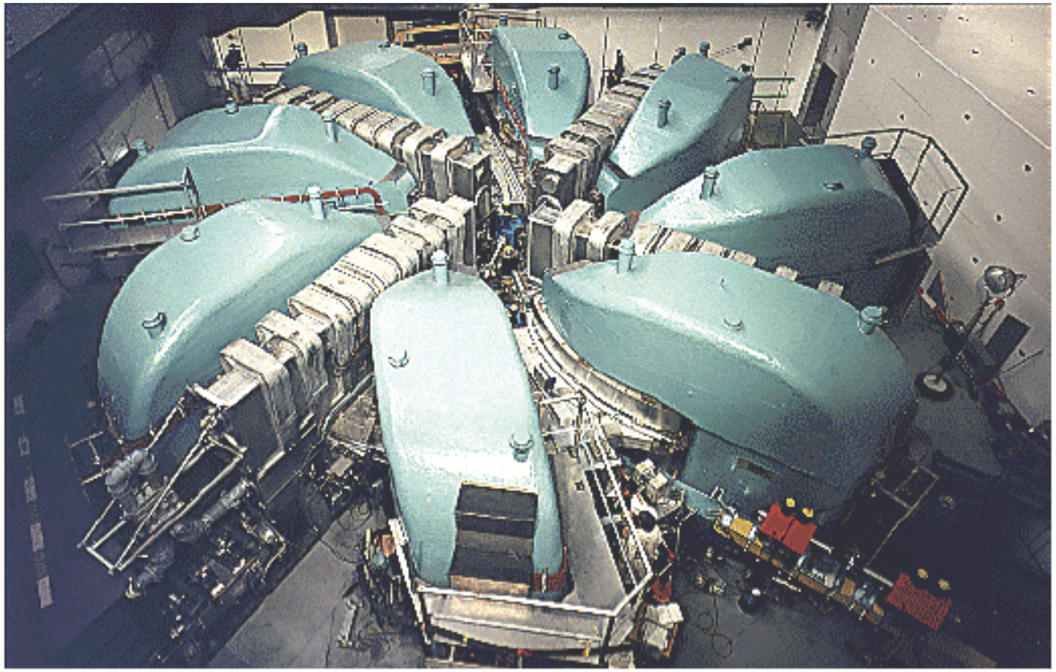
4 ICs

Each 290 MW_t ADAM core requires 3 x 4 mA of 800 MeV proton drive beams, and destroys 130 kg/year of TRU.

Each GW_e nuclear plant produces 390 kg/year of TRU.

So how do we make 9 x 4 mA of 800 MeV protons?

invented by Ernest Lawrence, 1930 at Berkeley



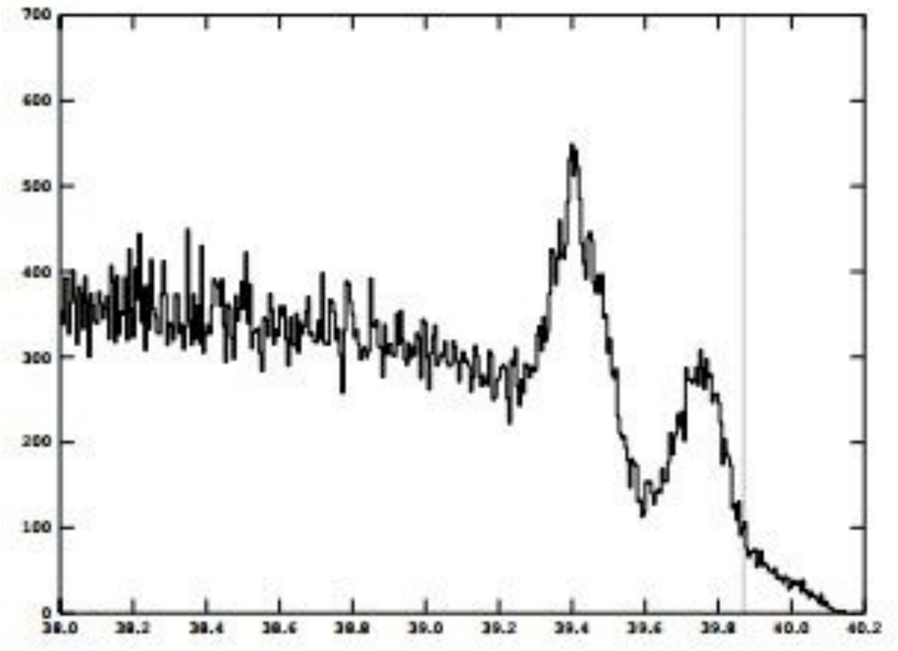
PSI operates the highest power accelerator in the world: 2.3 mA @ 590 MeV

The **cyclotron** is among the oldest of particle accelerators, and it still holds the world record for the highest beam power – 1.3 MW.

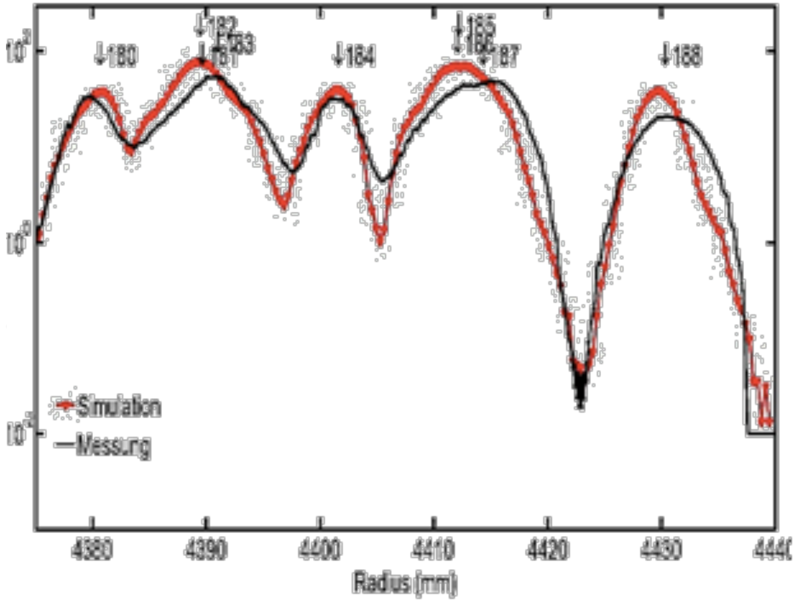
Even teenagers can build one:

http://www.youtube.com/watch?v=d7tKxqwfZoE&feature=autoplay&list=ULTLdlKDP76b4&index=1&playnext=1

Current limits in cyclotrons:
1) Overlapping bunches in successive orbits



http://www.nscl.msu.edu/~marti/publications/beamdynamics_ganil_98/beamdynamics_final.pdf



http://cas.web.cern.ch/cas/Bilbao-2011/Lectures/Seidel.pdf

Overlap of N bunches on successive orbits produces N x greater space charge tune shift,

non-linear effects at edges of overlap.

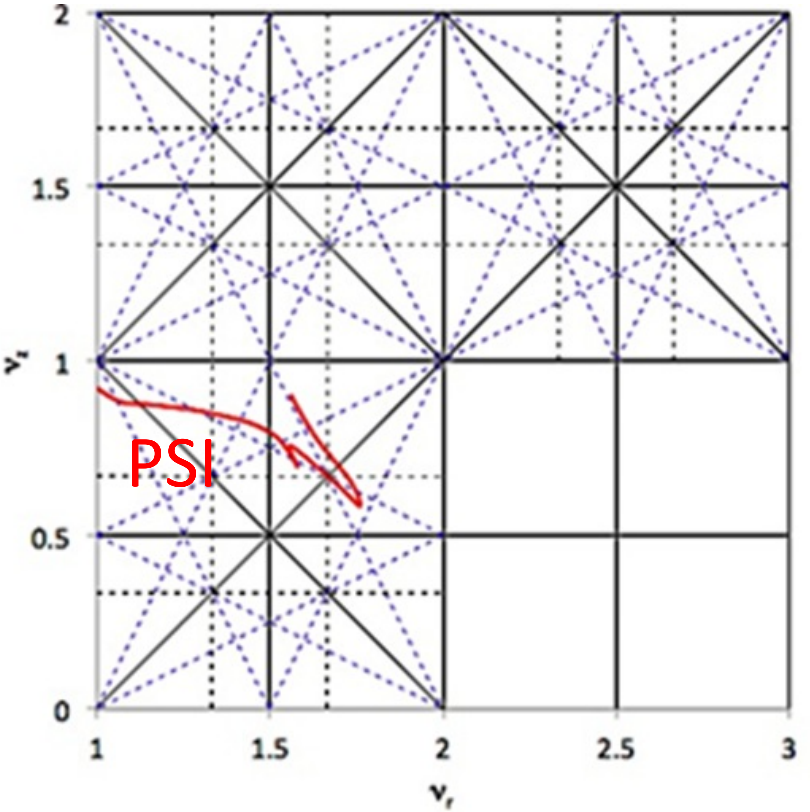
2) Weak focusing, Resonance crossing

Cyclotrons are intrinsically weak-focusing accelerators

- Rely upon fringe fields
- Low tune requires larger aperture
- Tune evolves during acceleration
- Crosses resonances

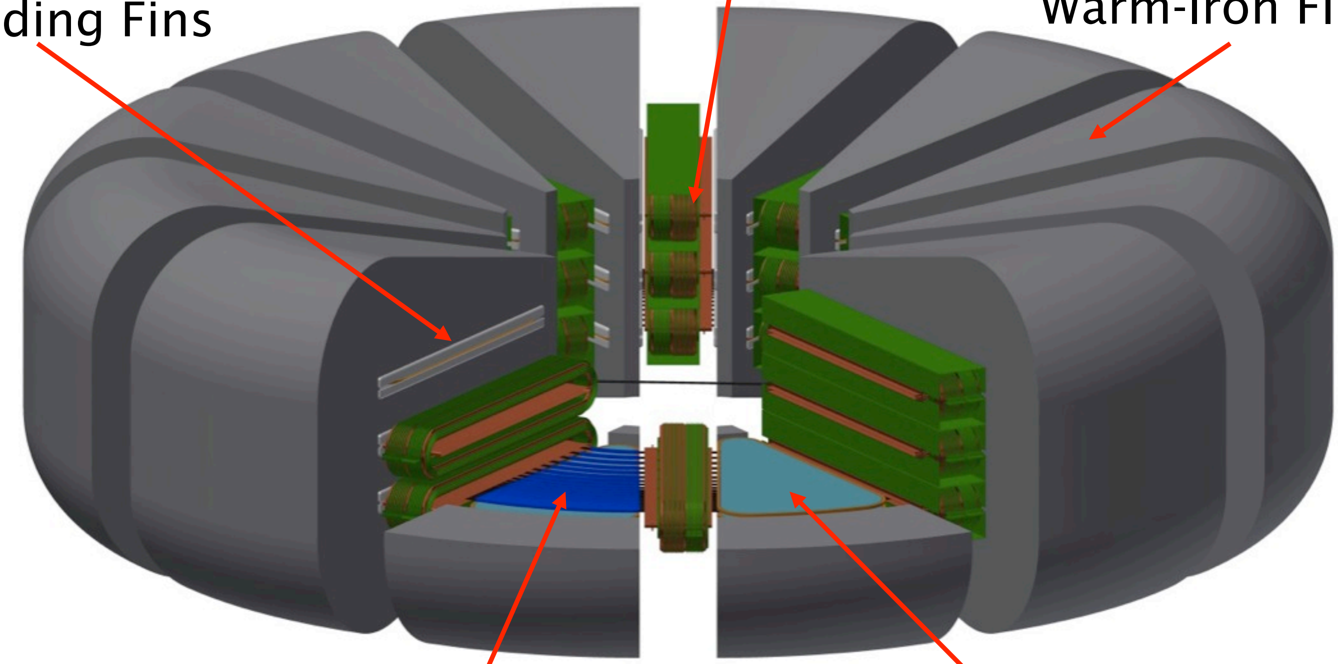
Scaling, Non-scaling FFAG utilize non-linear fields

- Rich spectrum of unstable fixed pts



Space charge shifts, broadens resonances, feeds synchro-betatron
Even if a low-charge bunch accelerates smoothly, a high-charge bunch may undergo breakup even during rapid acceleration

Hence the Strong-Focusing Cyclotron…



SRF Cavities

Warm Shielding Fins

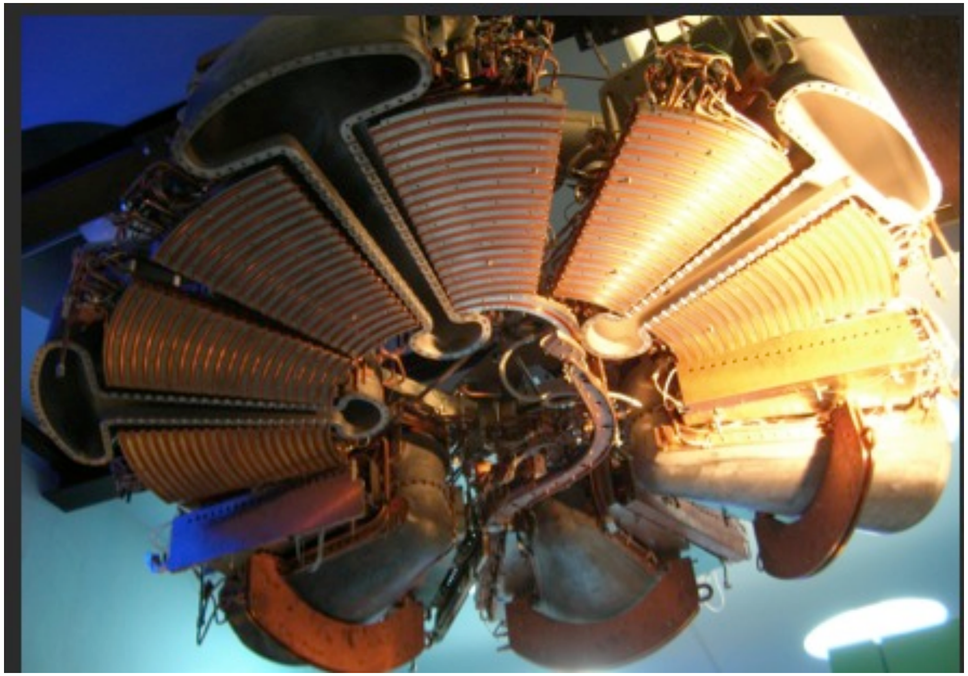
Warm-Iron Flux Return

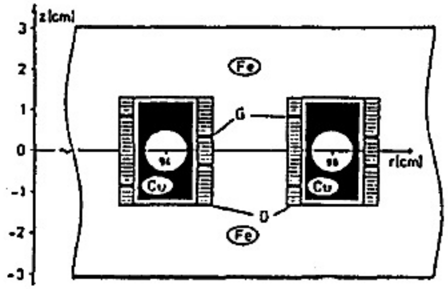
Beam Transport Channels

Cold-Iron Pole Piece

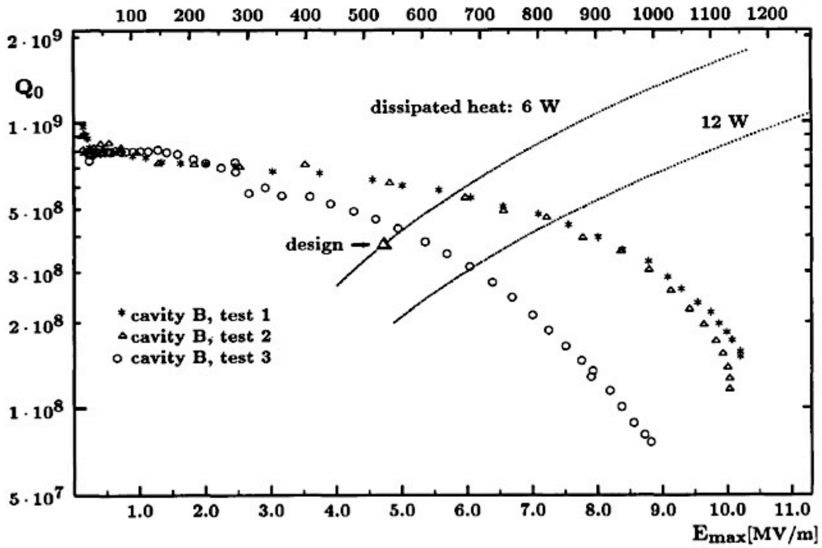
- SRF cavities provide 20 MeV/turn energy gain – fully separate orbits
- Sectors are simple radial wedges – optimum for integrating SRF
- Beam transport channels control betatron tunes, isochronicity

TRITRON was the first to attempt to make a separated orbit cyclotron





The good-field fraction of radial aperture was <50% for each orbit, so admittance was limited.



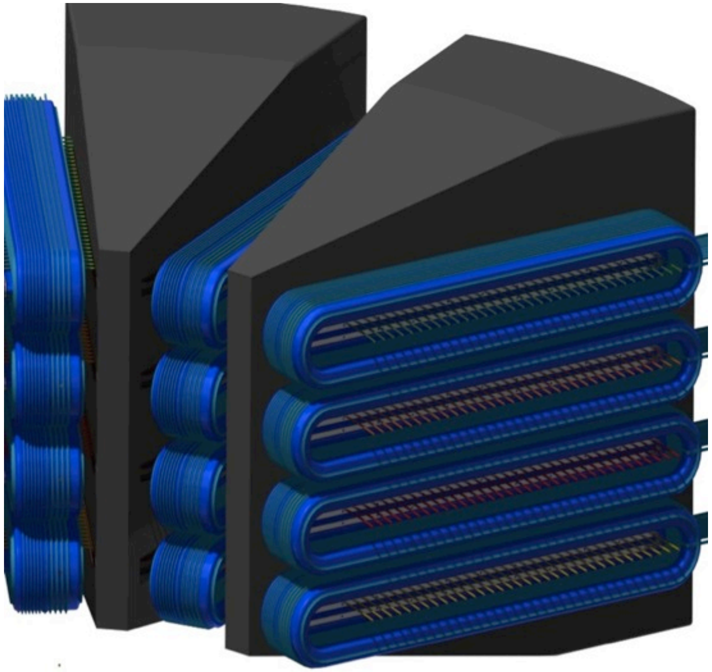
Energy gain in its superconducting Pb cavities was limited by multipacting.

The intervening years of superferric magnet technology (and now MgB_2) and Nb cavity technology make this a fertile time to make a strong-focusing cyclotron for high current.

Three key innovations enable us to make the drive beam power to drive 3 cores:

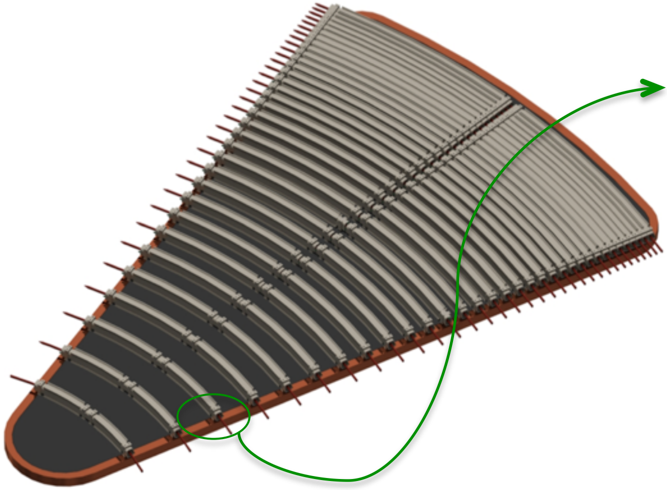
The Strong-Focusing Cyclotron Stack

Flux-coupled stack of cyclotrons



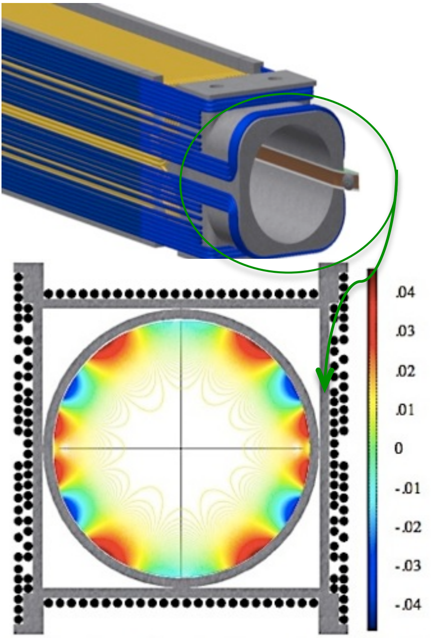
12 mA CW proton beam in each SFC
3 x 12 mA x 800 MeV = **30 MW power**

Superconducting RF cavities



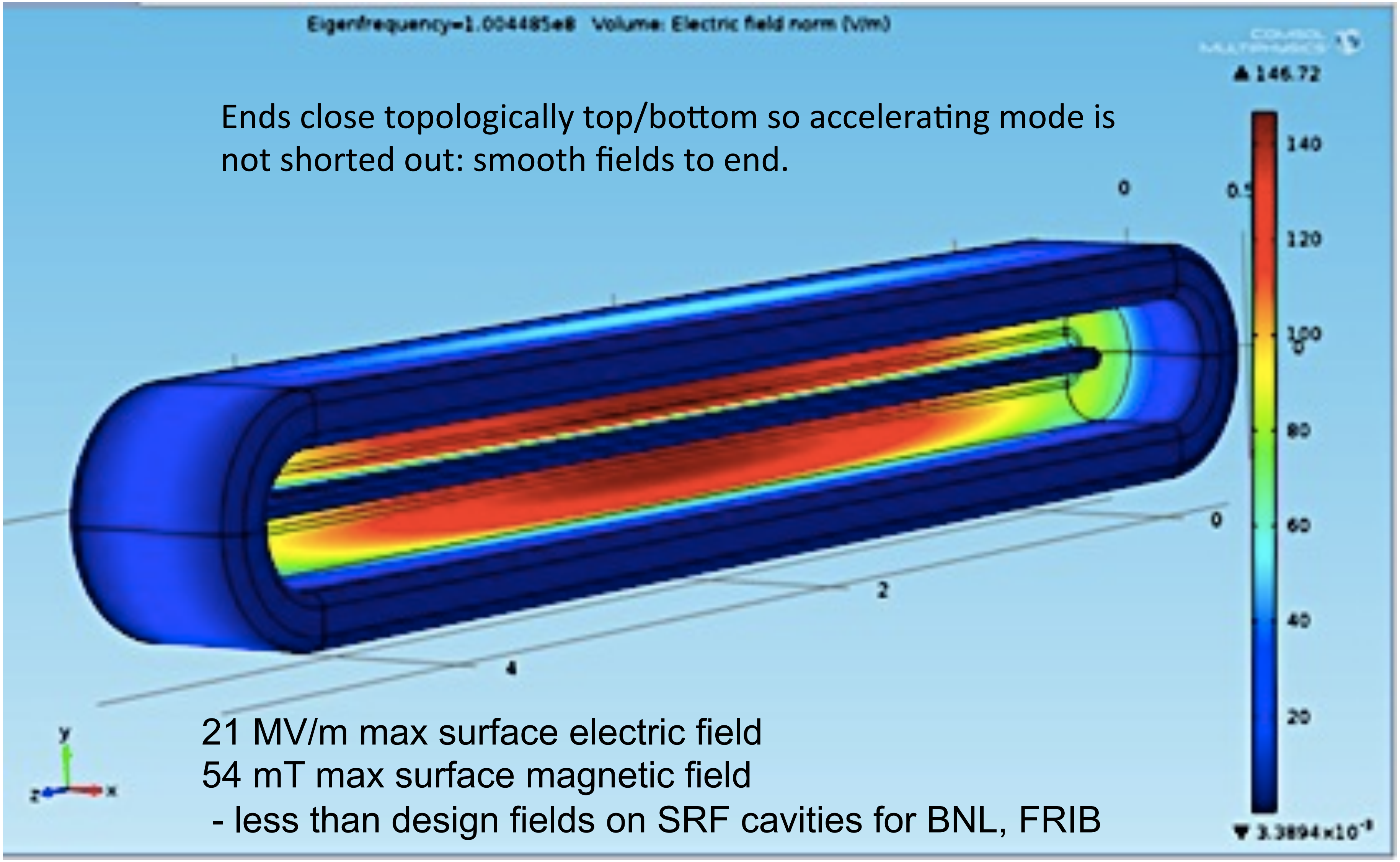
All orbits fully separated.
12 cavities x 1.8 MV/cavity
=20 MV/turn

Quadrupole focusing channels



Fix betatron tunes throughout acceleration.

SRF Cavity: slot-geometry ¼-wave structure



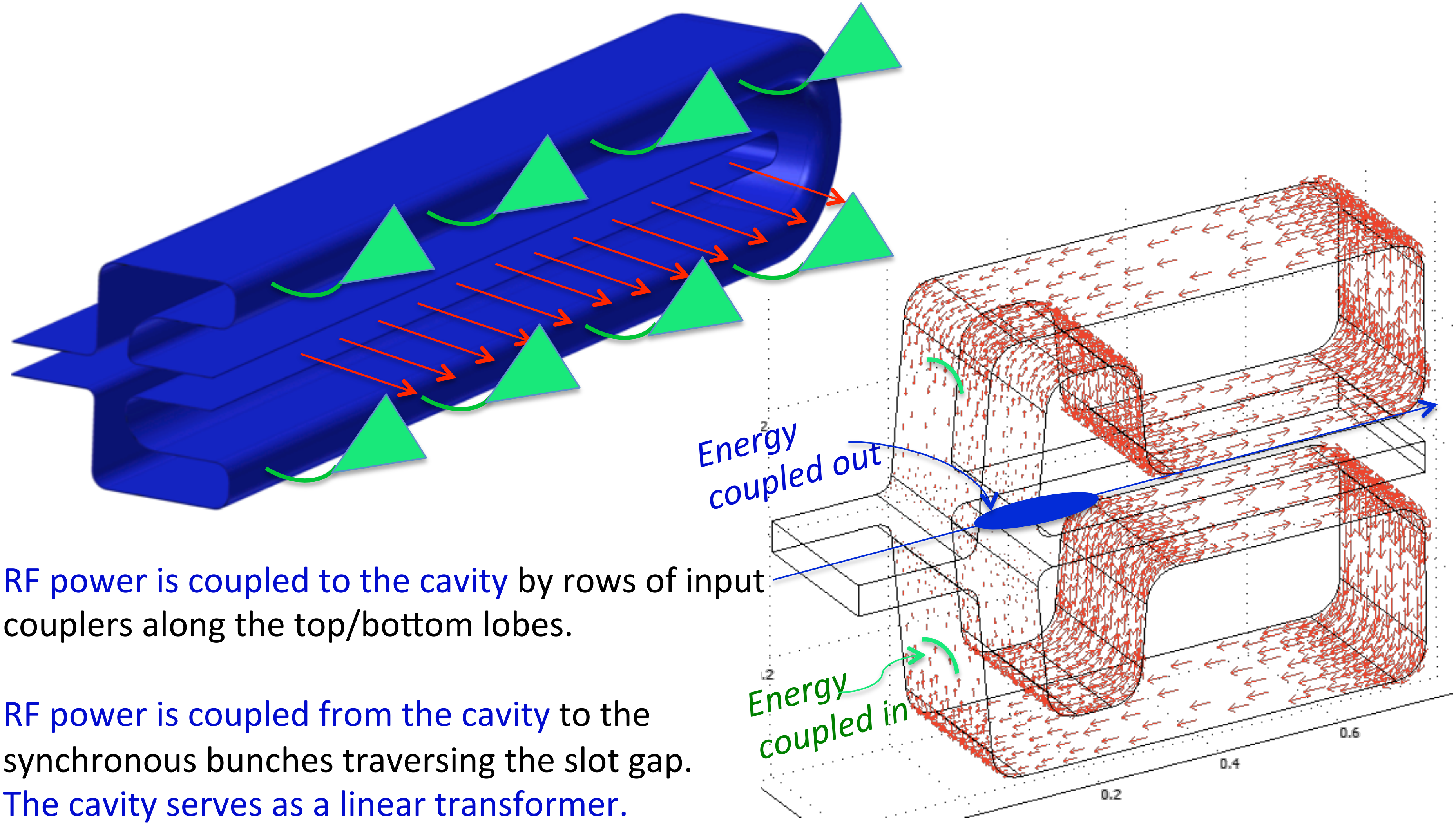
Ends close topologically top/bottom so accelerating mode is not shorted out: smooth fields to end.

21 MV/m max surface electric field

54 mT max surface magnetic field

- less than design fields on SRF cavities for BNL, FRIB

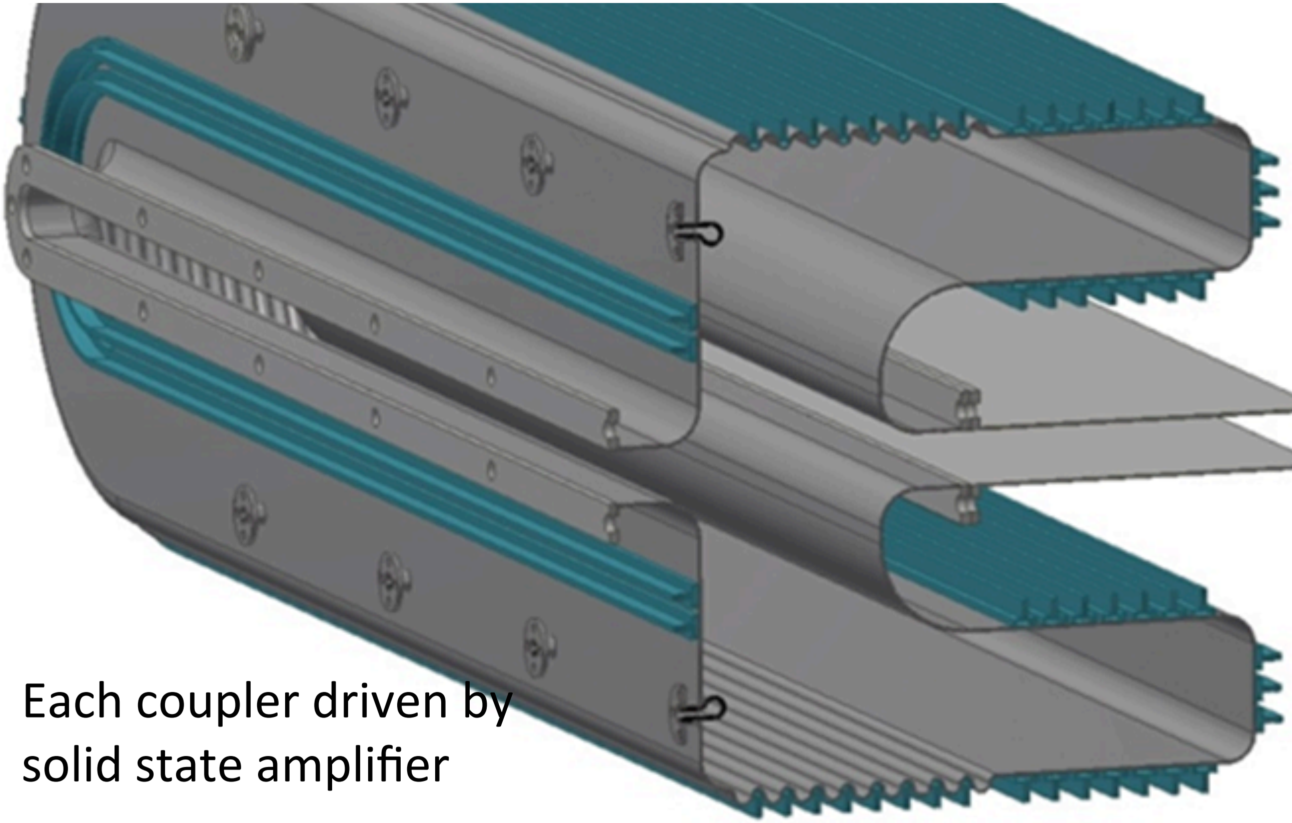
Slot-geometry ¼ wave cavity structure and distributed RF drive suppresses perturbations from wake fields



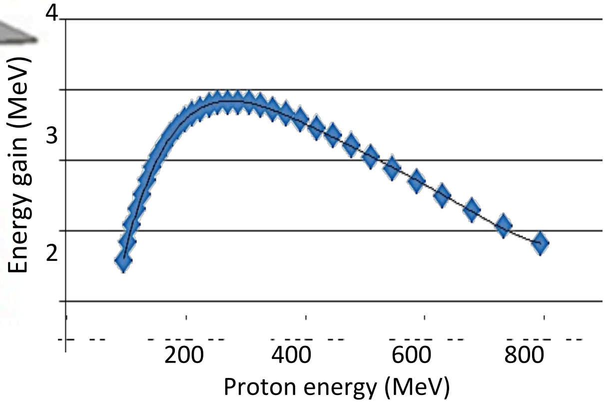
RF power is coupled to the cavity by rows of input couplers along the top/bottom lobes.

RF power is coupled from the cavity to the synchronous bunches traversing the slot gap.
The cavity serves as a linear transformer.
Its geometry accommodates transverse mode suppression

Linear coupler array to match drive to beam loading, convolutes to suppress multipacting



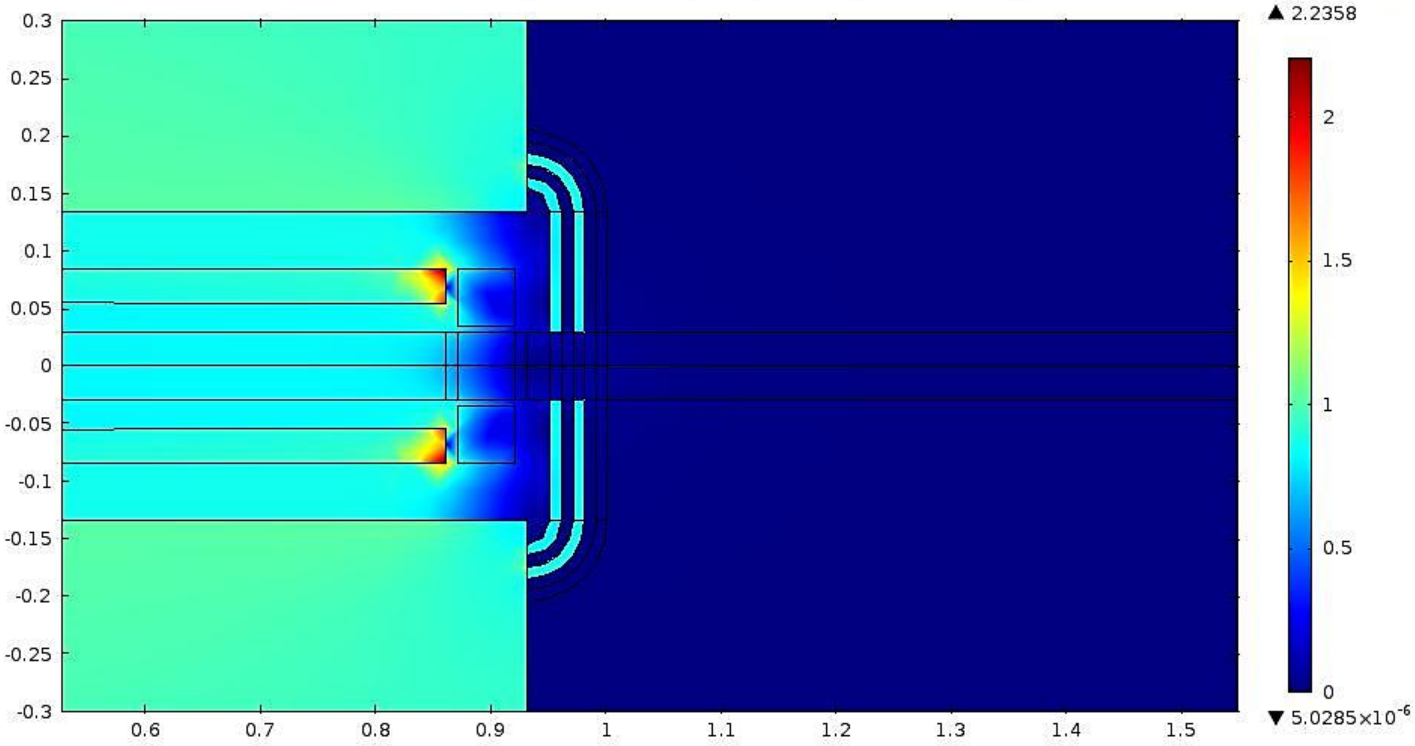
Each coupler driven by solid state amplifier



Distributed drive matches to distributed beam loading for stability under high beam loading.

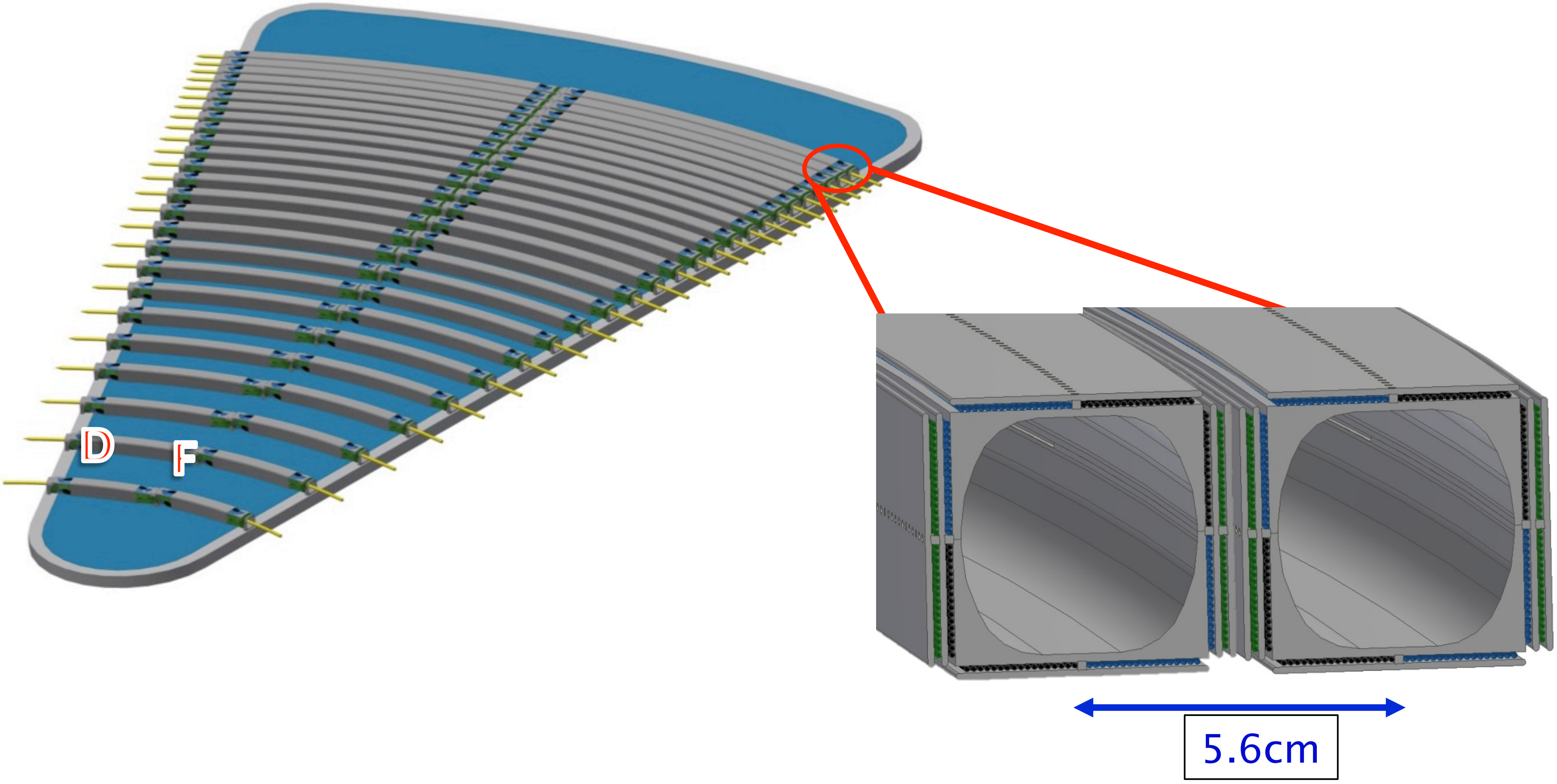
Note: this requires that all orbits are made very close to isochronicity…

Shielding the Sector dipole field from the neighboring Superconducting Cavities



Cross section of the magnet at the mid-point of the orbits. In the coils themselves the fields reach 2.24 T in TAMU800.
The fins reduce the maximum field to 40 mT @ 5 cm beyond the warm iron. Multi-layer mag insulating foils reduce that to 3 μT at closest SRF cavity surface.

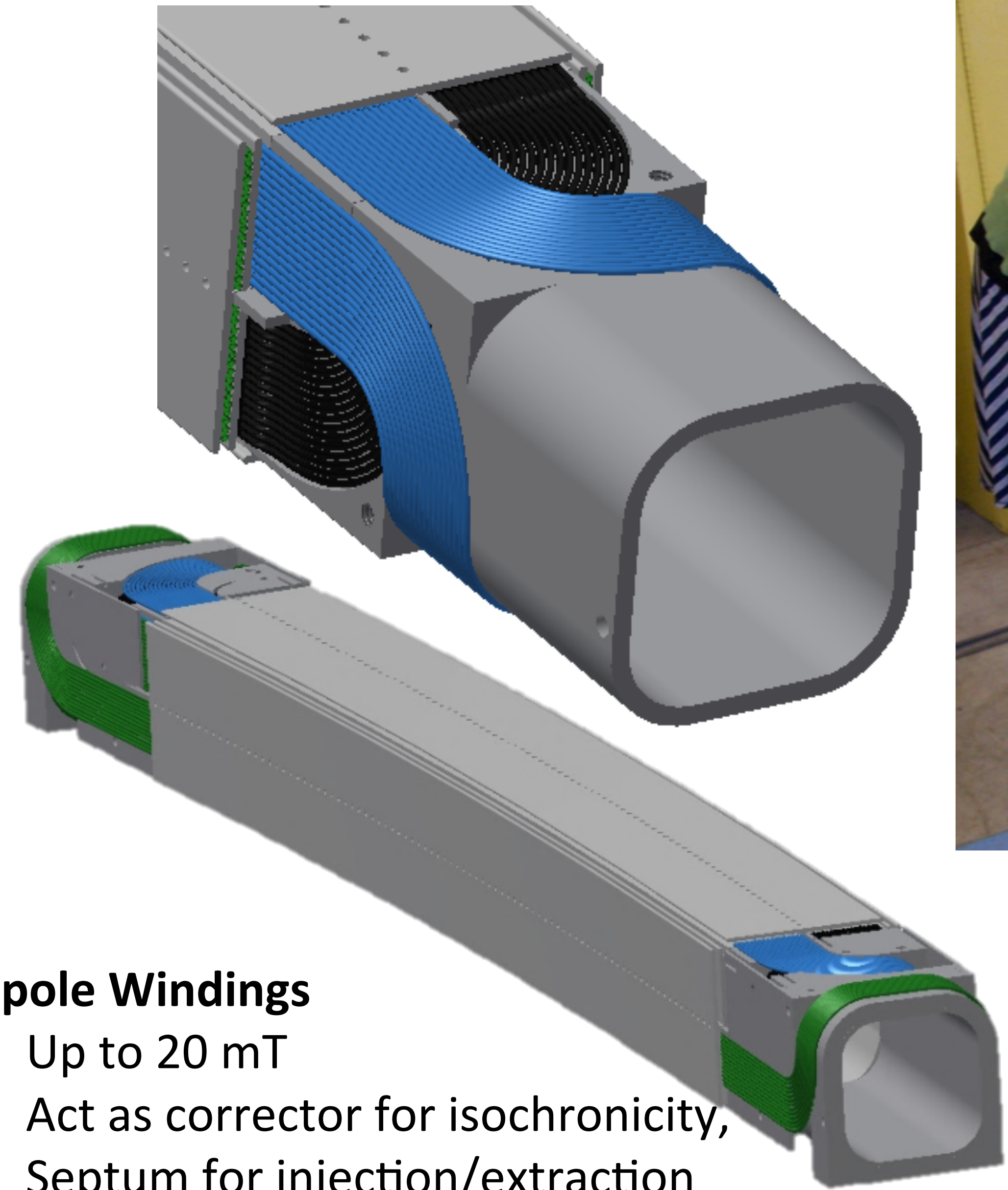
F-D doublet on each orbit, each sector



BTC dimensions are set by the requirements for beam separation at extraction.

>80% of horizontal aperture is useful for orbits.

MgB_2 windings on beam transport channels



Dipole Windings

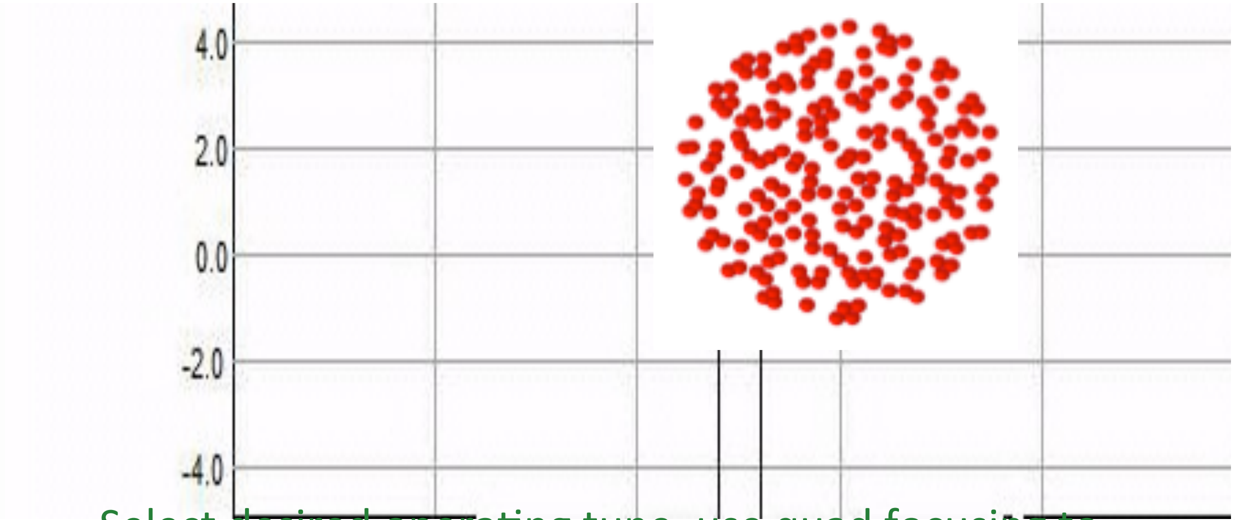
- Up to 20 mT
- Act as corrector for isochronicity,
- Septum for injection/extraction

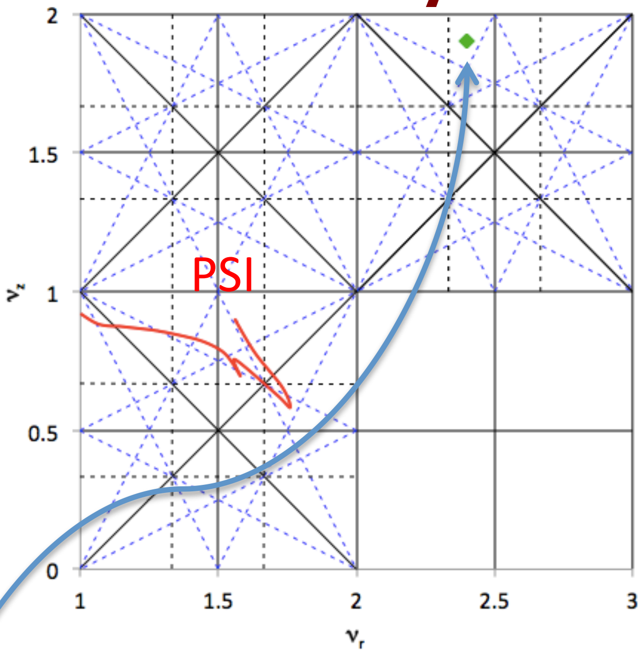
Quadrupole Windings

- Up to 6 T/m
- Panofsky style
- Alternating-gradient focusing
- 6 families provide tune control

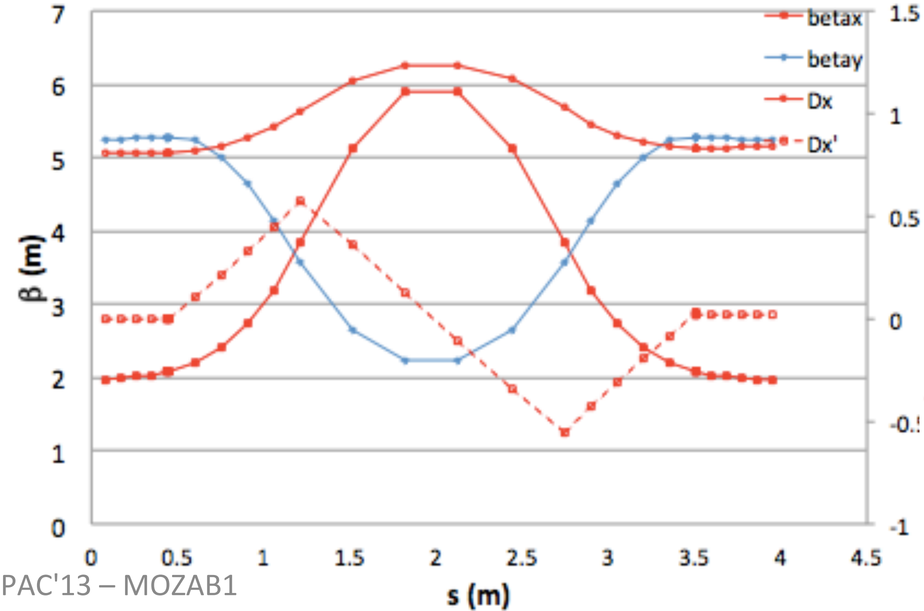
BTCs control tune, isochronicity

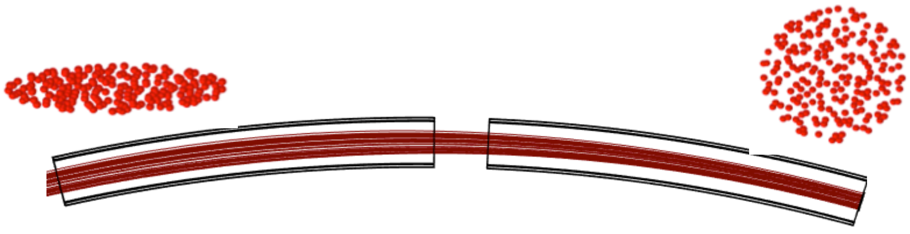
Uniform gradient in each channel: excellent linear dynamics





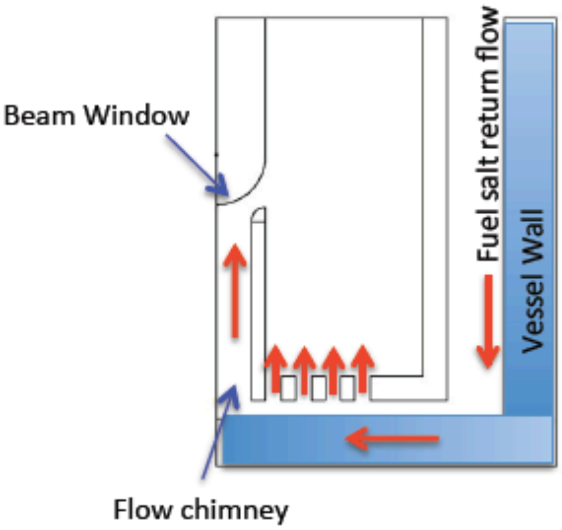
Select desired operating tune, use quad focusing to lock the tune for all energies

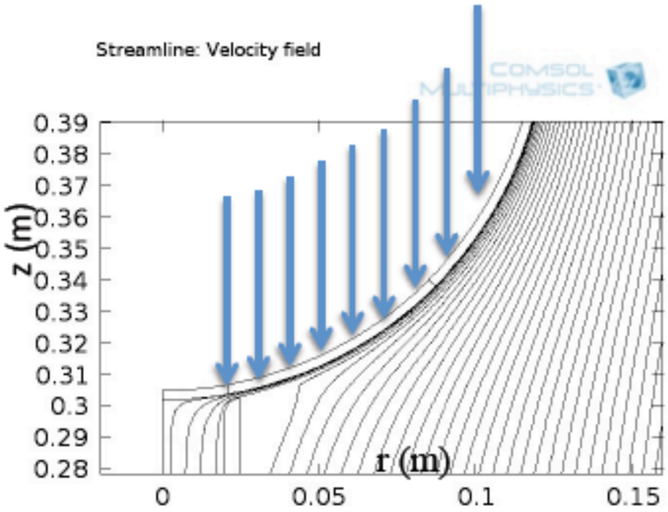


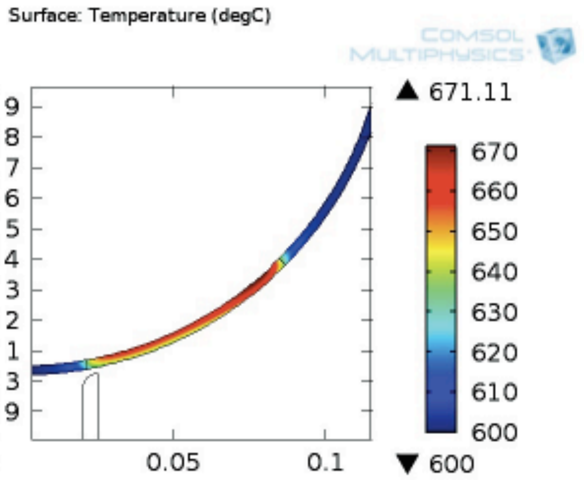


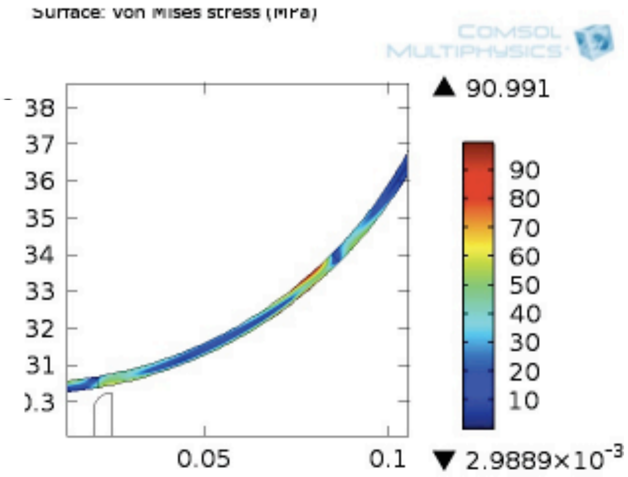
We inject 2.8 MW protons through a 3 mm-thick Hastelloy window

We direct a dedicated molten salt flow on the window in the HX circuit.









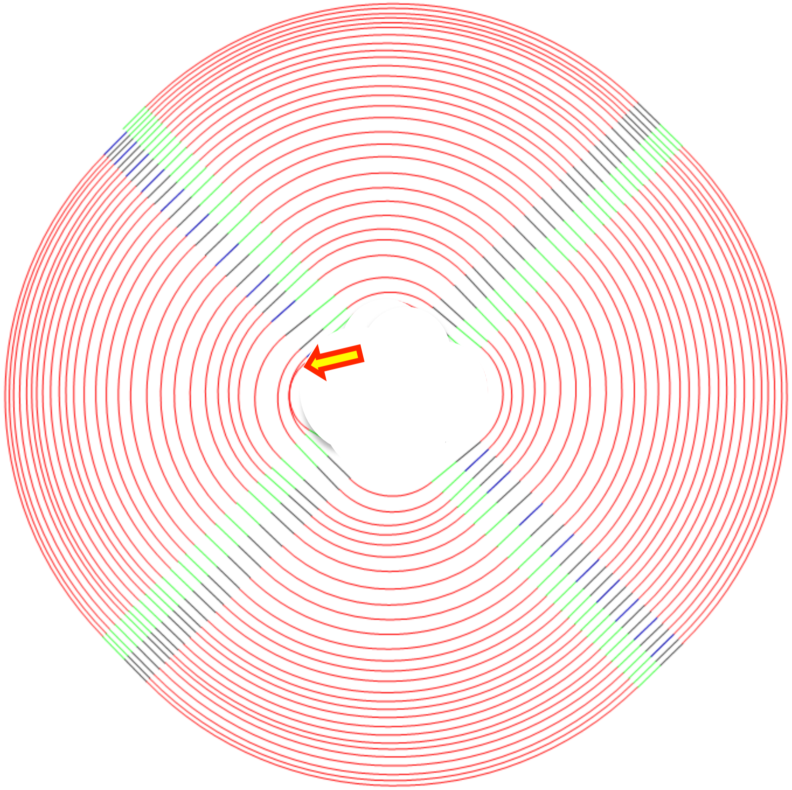
Protons pass through window, deposit most of their energy in molten salt.

~22 kW is deposited in the 3 mm Hastelloy window.

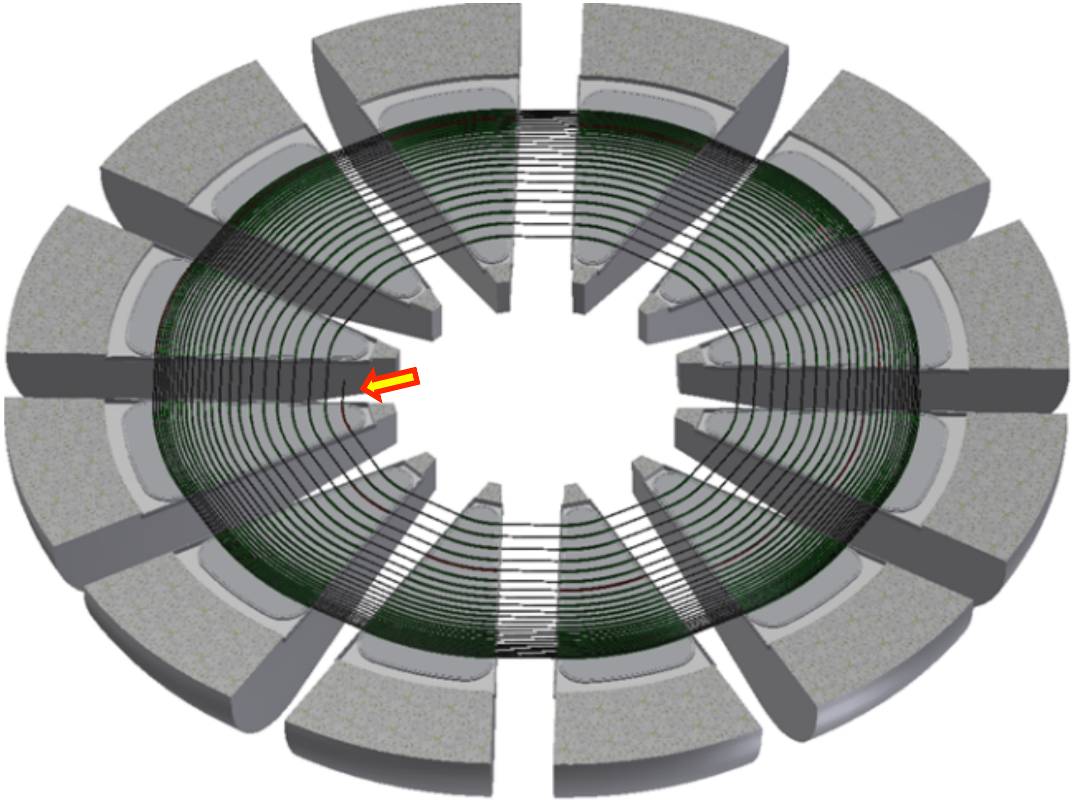
Max temp gradient ~60 C, max von Mises stress ~60 MPa.

Should be fine, we will do experiments to verify.

Control all orbits:
betatron tunes, isochronicity, position



TAMU100: 6.5→100 MeV



TAMU800: 100→800 MeV

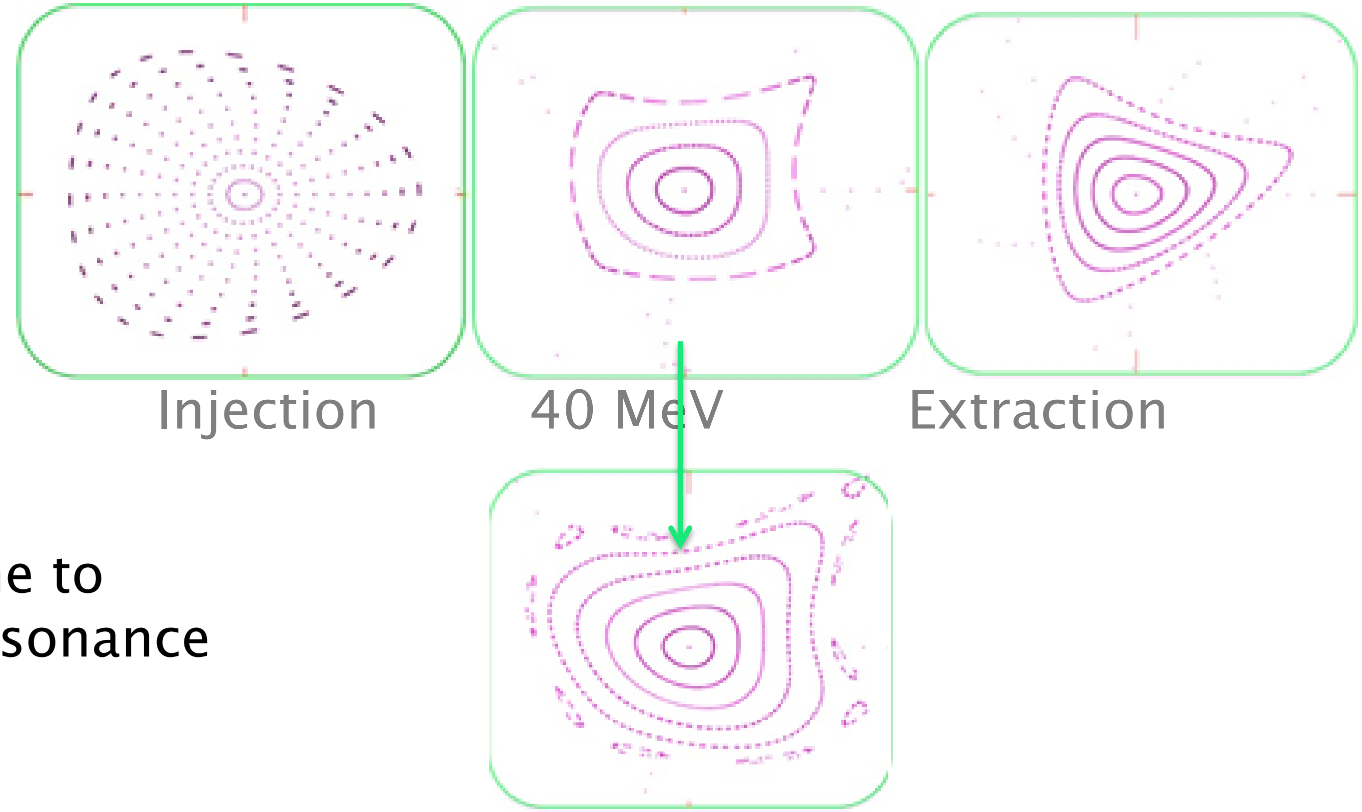
If any one of the 10 rf cavities malfunctions, increase gradient in the remaining 9 to maintain energy gain/turn, use trim dipoles in the beam transport channels to maintain equilibrium orbit unchanged. Works like a ‘spiral linac’.

We have simulated spiral transmission line, including x/y coupling, synchrobetatron, space charge

Poincare Plots of 1-5 σ contours in TAMU100

3.5 mA beam

First lock tune to favorable operating point:



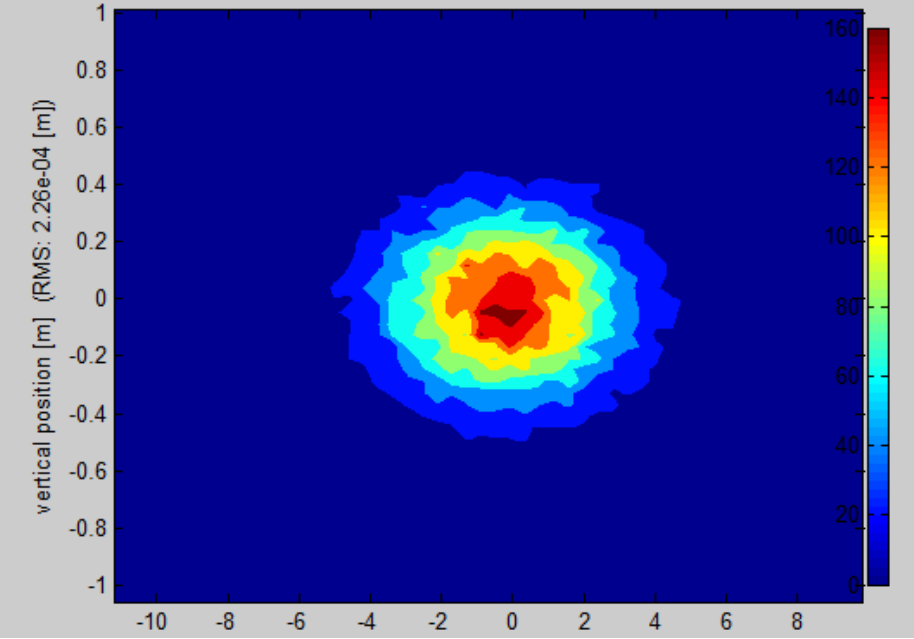
Now change the tune to excite a 7th order resonance

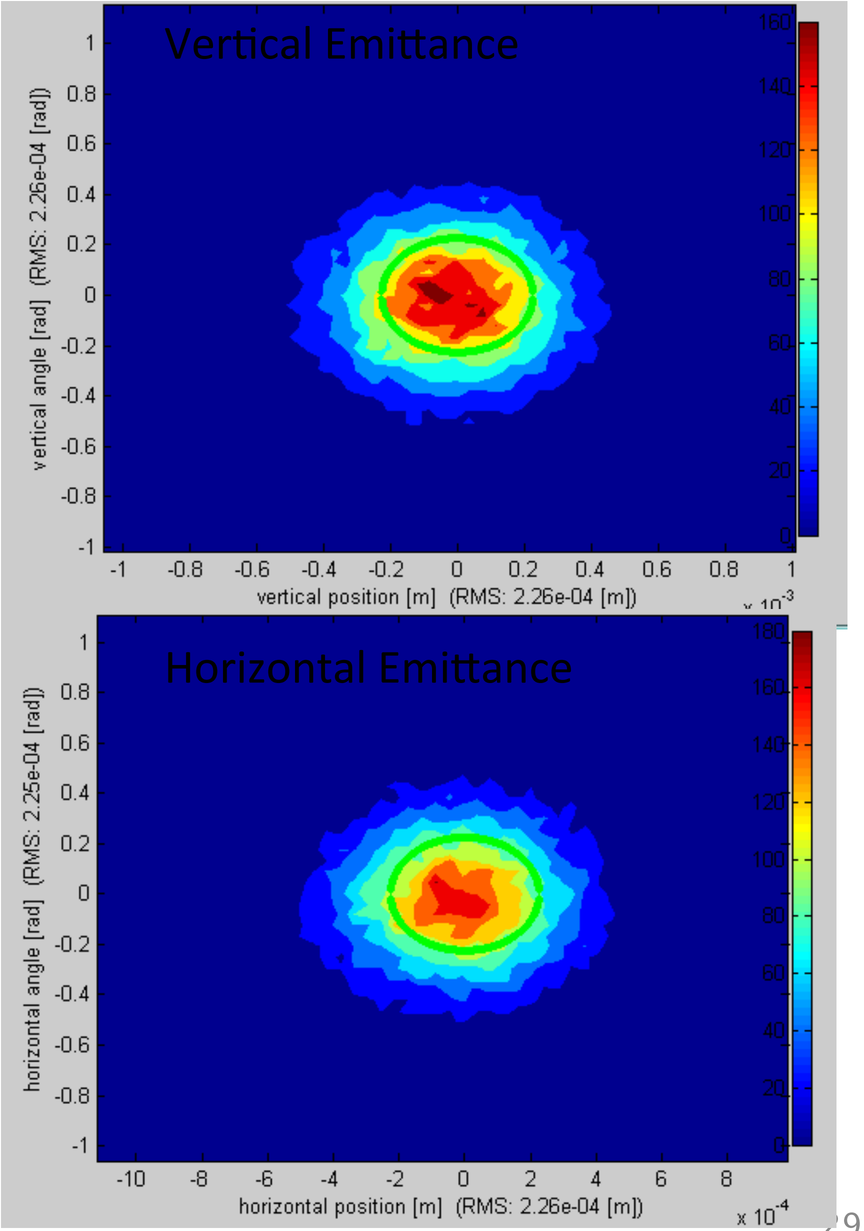
We are seeing the origins of the current limits in PSI from overlapping bunches, tune trajectory. Both are cured in the SFC.
Next studies: beam loading of cavities, wake fields…

Transverse phase space of 10 mA bunch

First at injection:

x/y profile

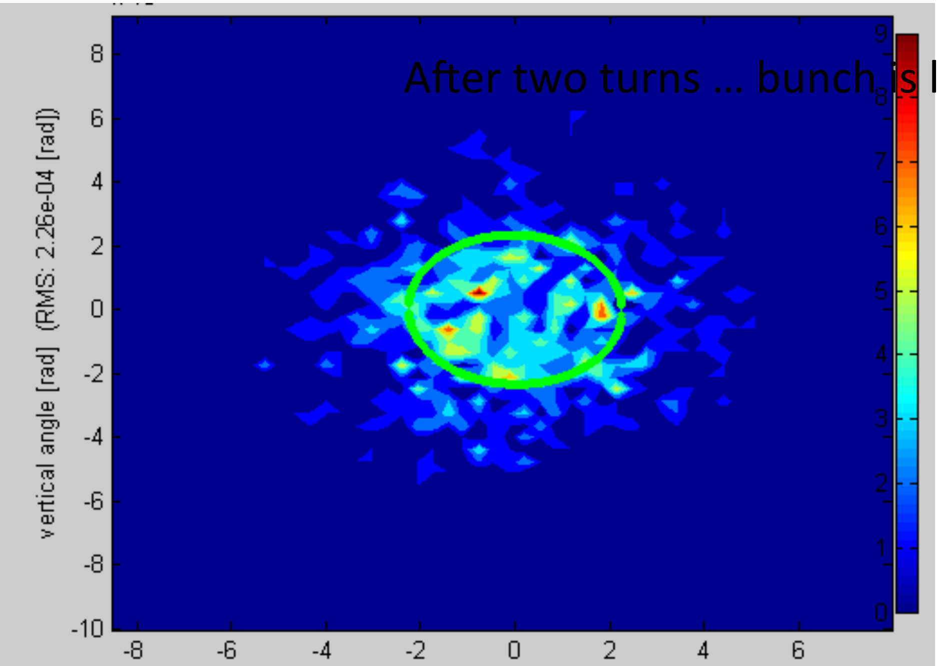




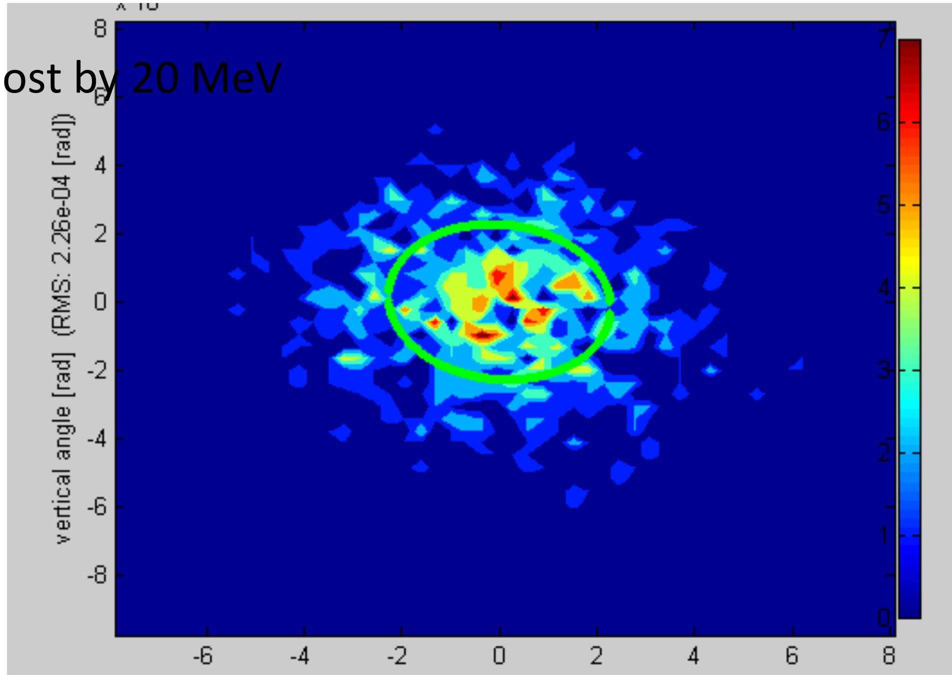
Now look at effects of synchrobetatron and space charge with 10 mA at extraction:

Move tunes near integer fraction resonances to observe growth of islands

After two turns … bunch is lost by 20 MeV



1/3 order integer effect

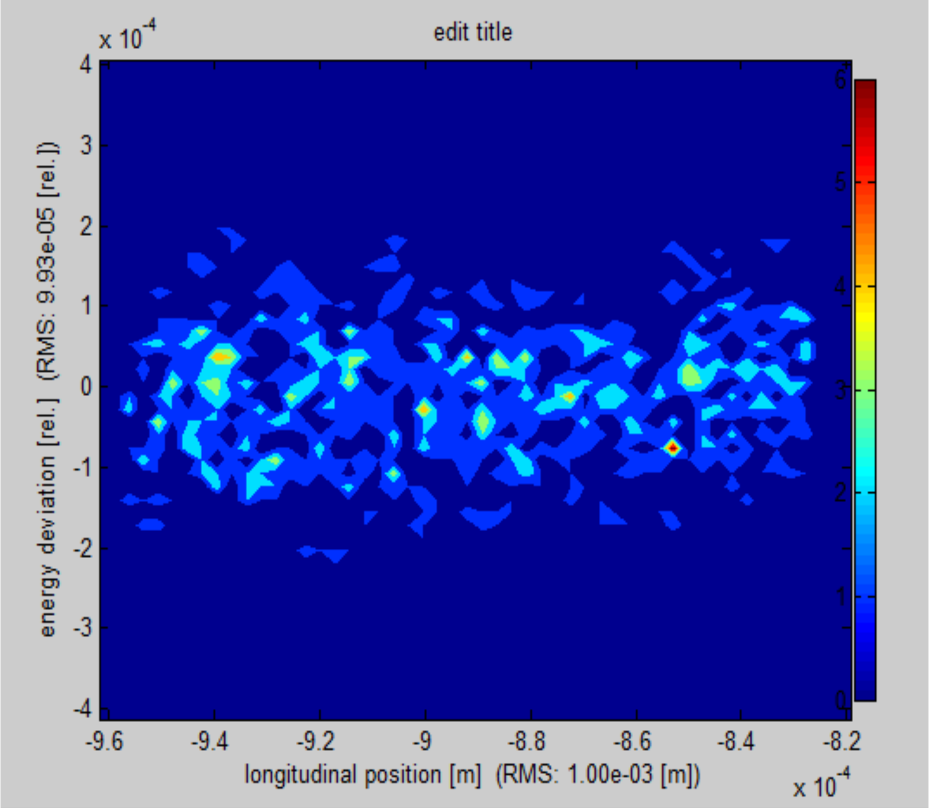


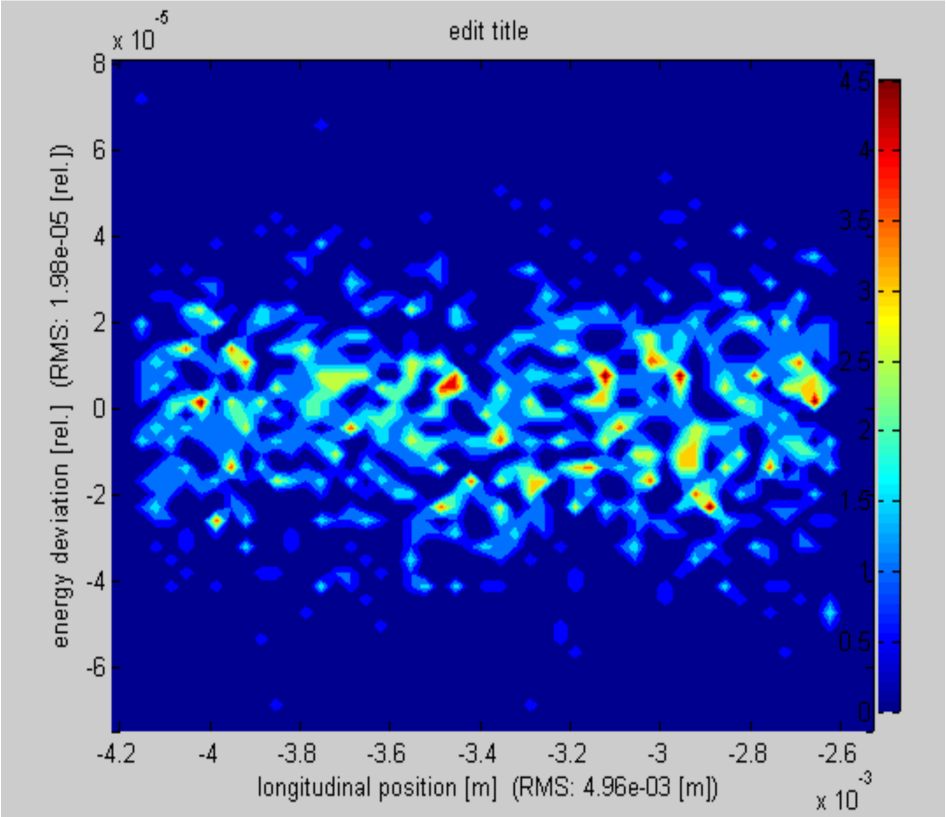
1/5 order integer effect

1/5-order islands stay clumped, 1/3-order islands are being driven. Likely driving term is edge fields of sectors (6-fold sector geometry). We are evaluating use of sextupoles at sector edges to suppress growth.

Synchrobetatron/space charge in longitudinal phase space:

Tunes again moved to approach resonances, but retaining transmission through lattice





Injection

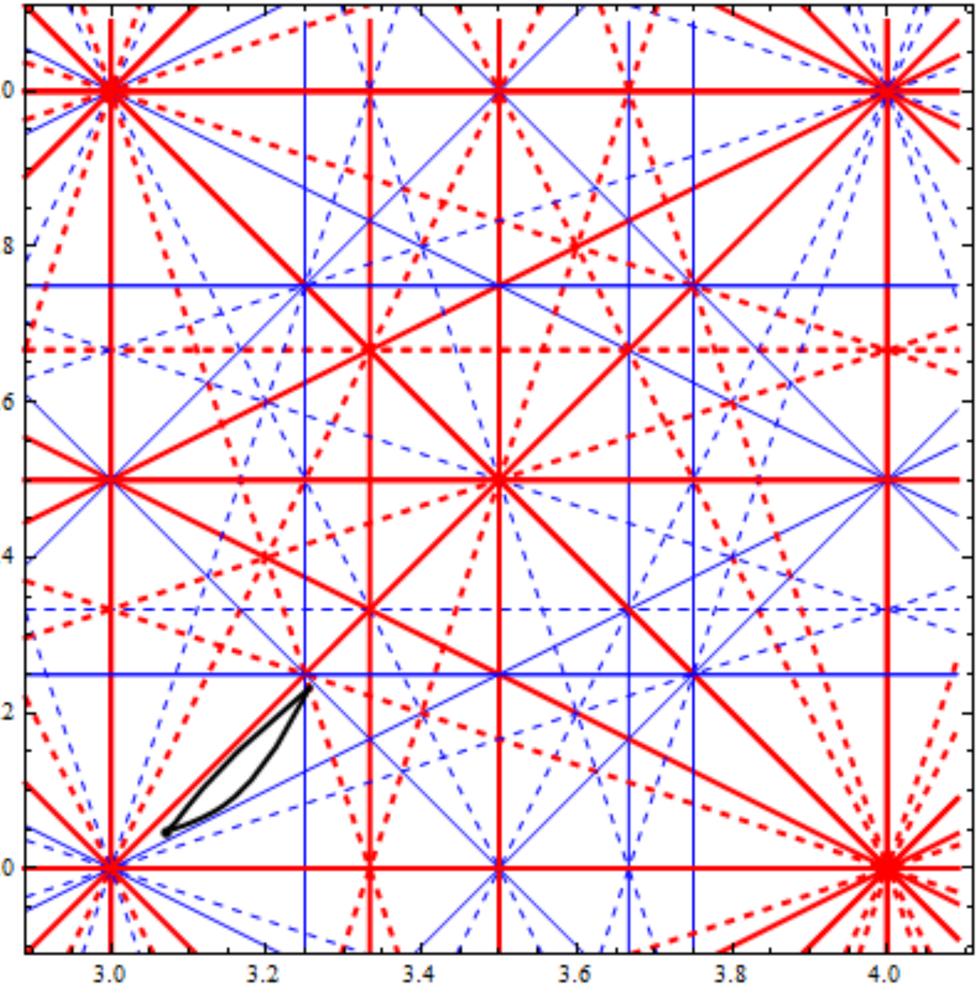
extraction

Phase width grows x5 at extraction

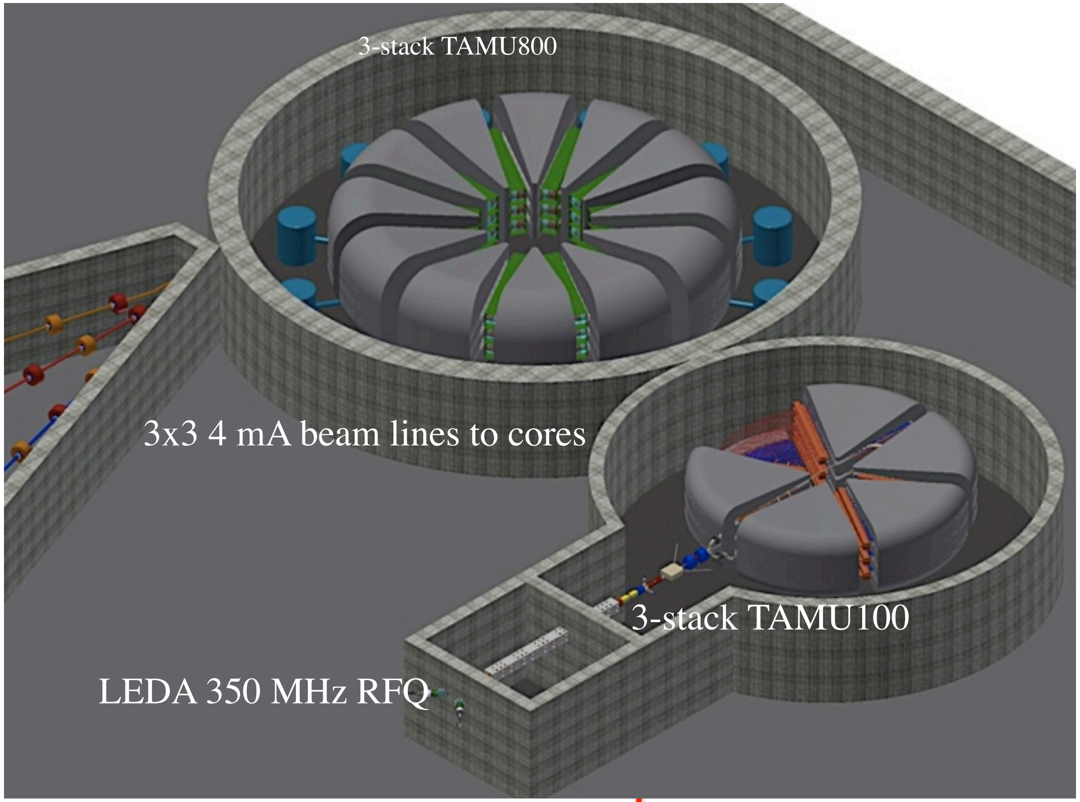
Now find tunes for all particles on the 5 s contour in a 10 mA beam accelerated to 800 MeV:

Since we can control tune using BTCs, we can place the operating point so that no significant resonance is crossed by any beam out to 5σ

We are exploring placement of 4 families of sextupole correctors after each sector;
We expect that to enable us to push further in current…



To destroy TRU generated from a GW$_e$ power plant:



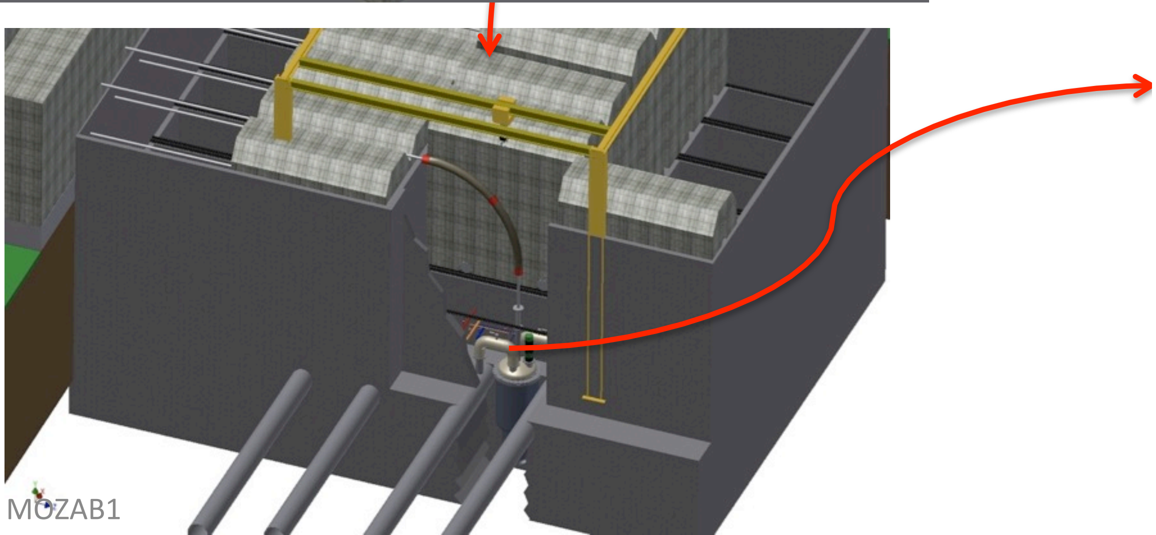
Each 800 MeV SFC

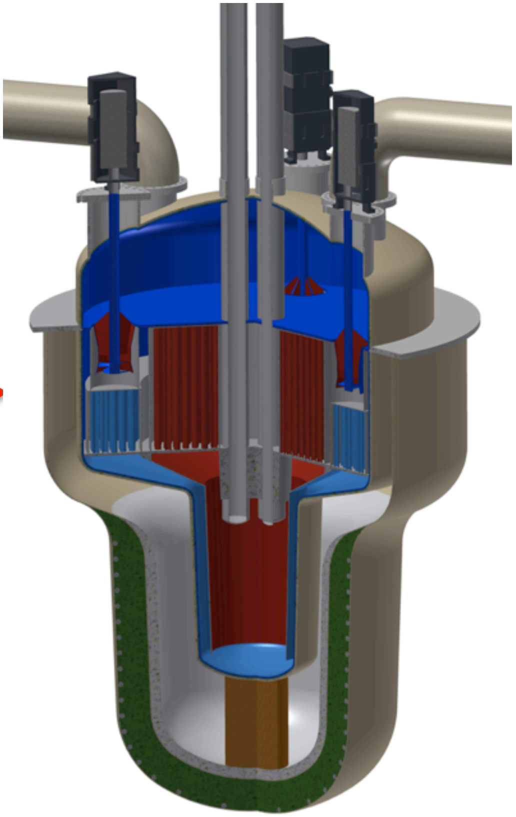
    12 mA current ➔3 beams

Total 30 MW CW:

    9 drive beams

    3 ADSMS cores

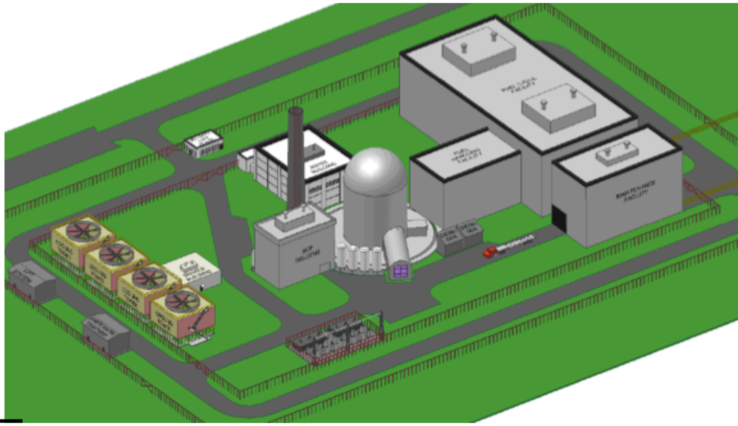




Compare performance for TRU-burning between ADAM and three flavors of critical fast reactors:

Critical reactors to burn TRU must operate with fast spectrum and non-H coolant/moderator:

- Sodium-cooled fast reactor SFR
- High-temperature gas fast reactor GFR
- Lead-cooled fast reactor LFR

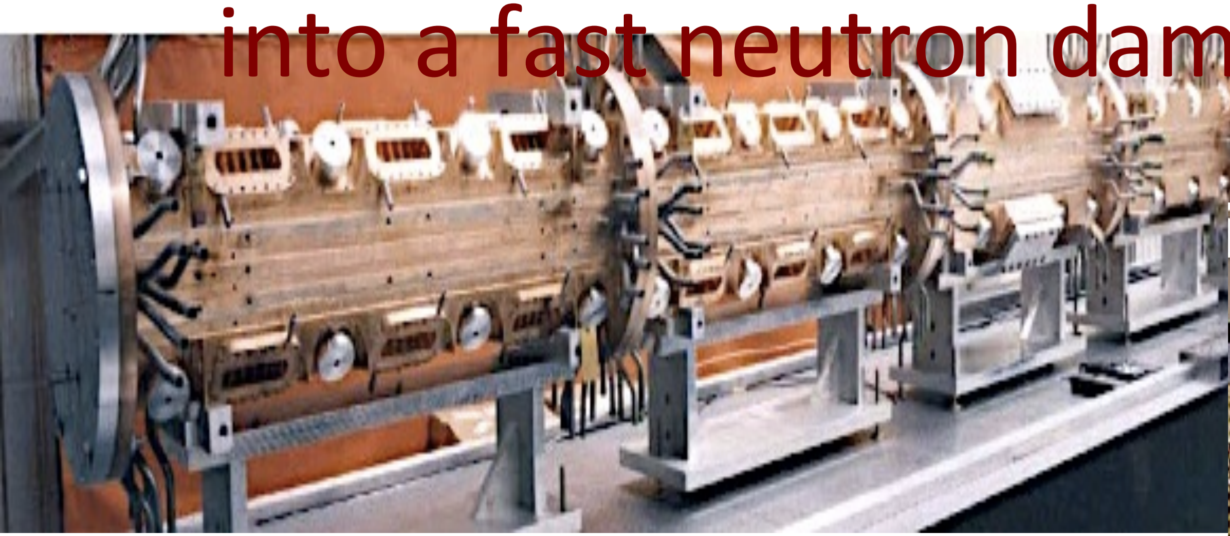


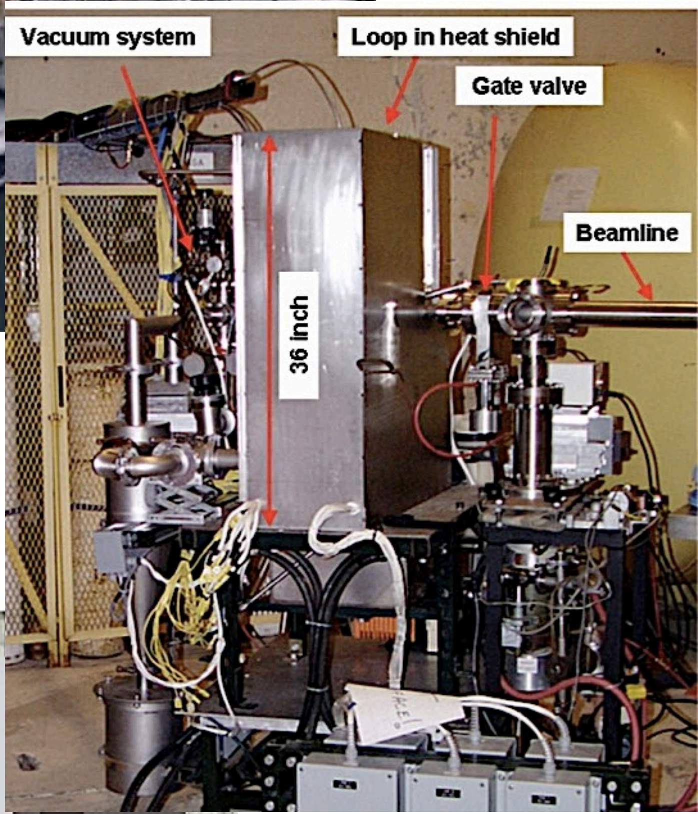
System	ADAM	SFR	GFR	LFR	
Net TRU Destruction	0.84	0.74	0.76	0.75	g/MW$_t$-day
System Power	**290**	**840**	**600**	**840**	**MW$_t$**
Outlet Temperature	665	510	850	560	C
Thermal Efficiency	44	38	45	43	%
Power Density	200	300	103	77	W/cc
TRU Inventory	**1733**	**2250**	**3420**	**4078**	**kg**
Fuel Volume Fraction		22	10	12	%
TRU Enrichment	53	44 - 56	57	46 - 59	% TRU/HM
Fuel Burnup	129.5	177	221	180	GWd/tHM
dTRU/TRU	**0.056**	**0.086**	**0.049**	**0.048**	**/year**

ADAM burns TRU as well as the best critical core yet designed, it operates with smallest TRU inventory, and it has no potentially disastrous failure modes.

RN Hill and TA Taiwo, PHYSOR-2006, ANS Topical Meeting, Vancouver, 9/2006.

We plan to move LEDA to TX, transform it into a fast neutron damage facility

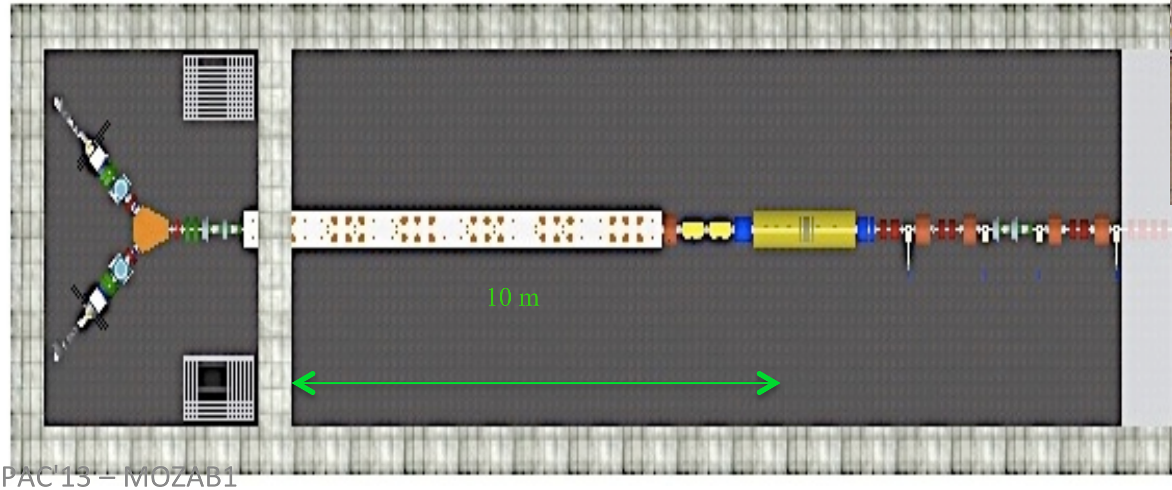




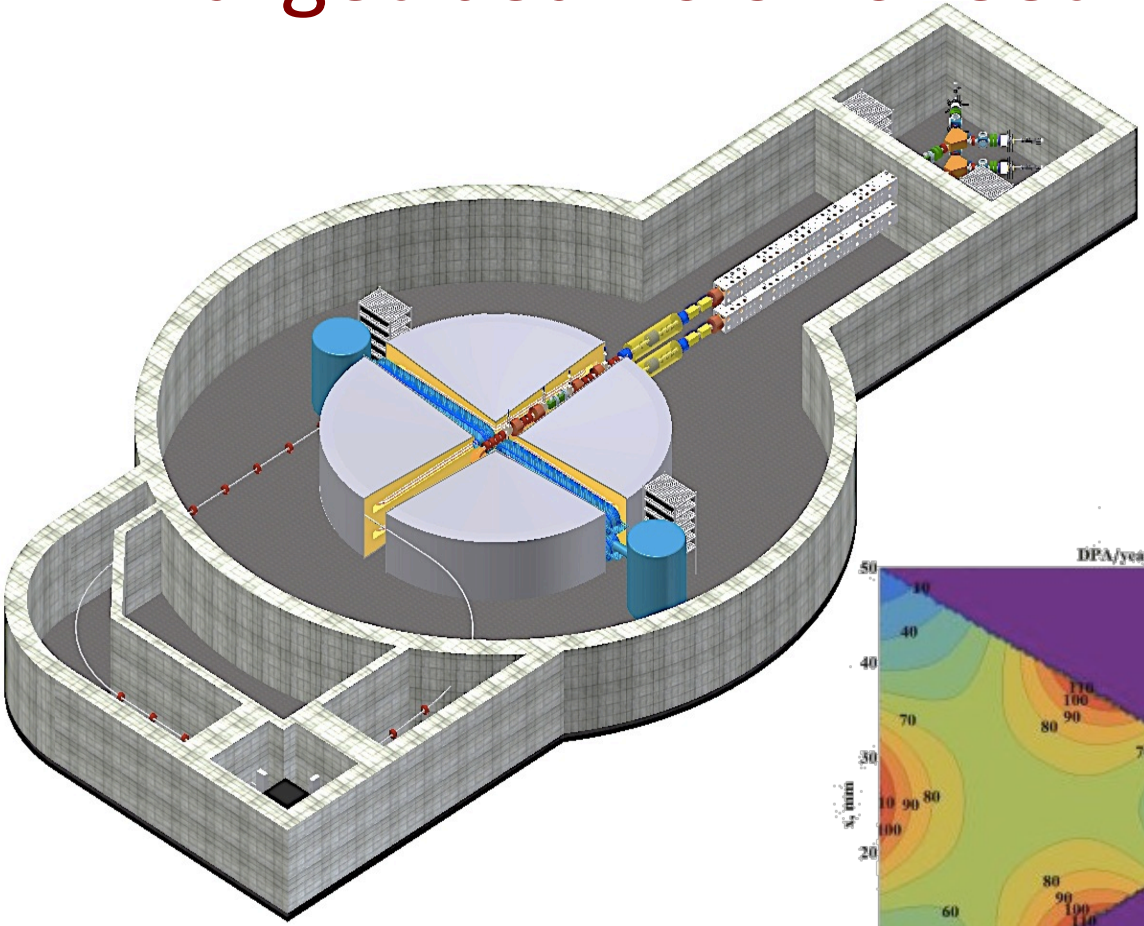
LEDA rfq: 6.5 MeV, 100 mA CW

ANL sheet-flow Li target: 6 m/s

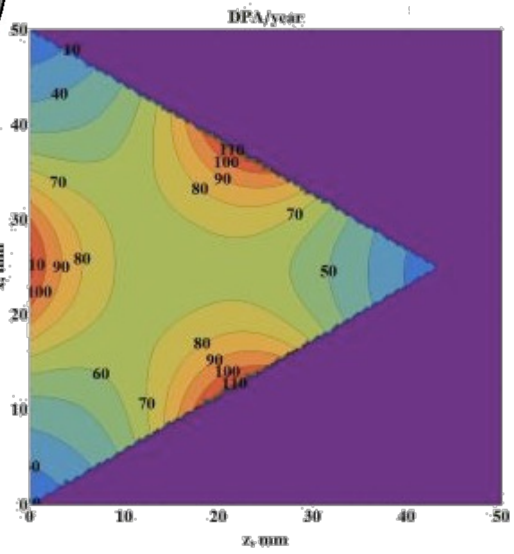
AND-1 at Texas A&M: 8 dpa/yr with fast neutrons

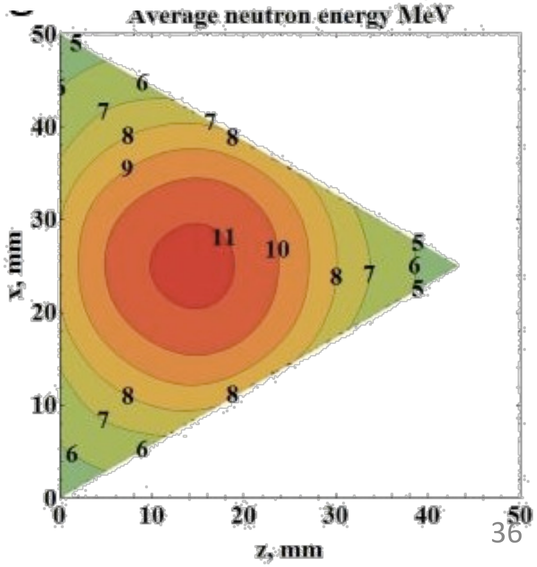


AND-2: Split the LEDA beam into 3 117 MHz bunch trains. Inject to a 3-stack TAMU100. Target beams on sheet-flow molten Pb.

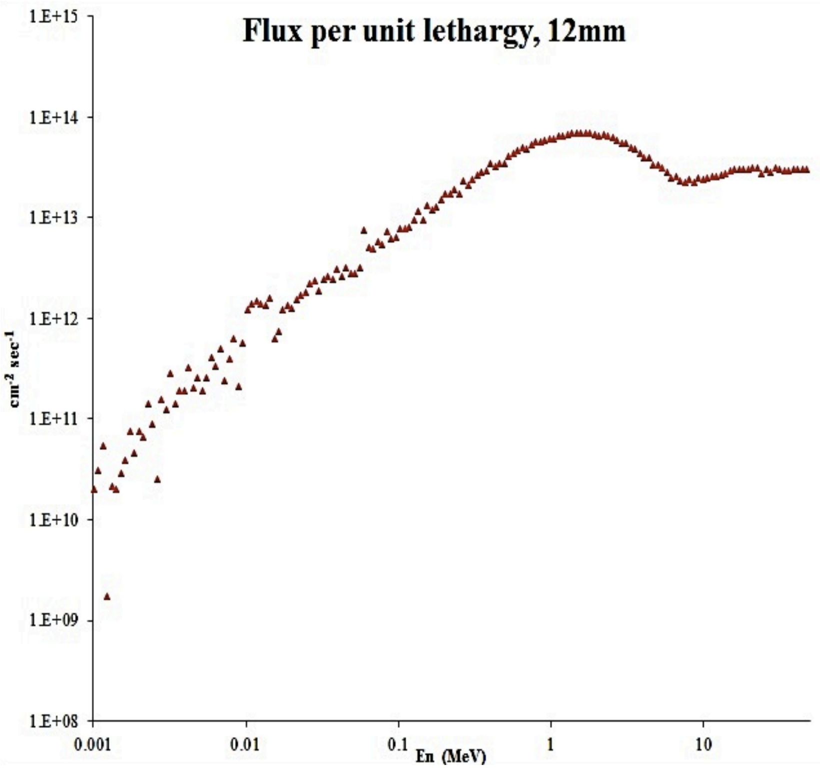


- 100 dpa/year of fast neutron damage to samples
- Fast spectrum - <E_n> can be adjusted by target angle 2-10 MeV
- Provides for all materials studies for fast reactorss and for fusion first wall

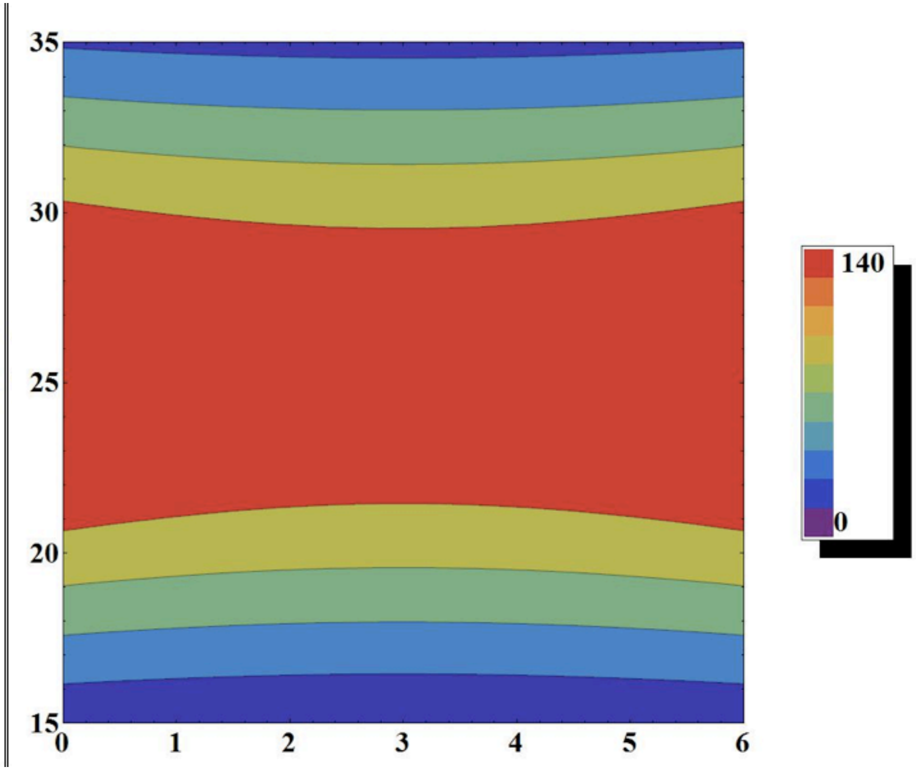




Neutron spectrum covers the needs for fast-spectrum fission and fusion



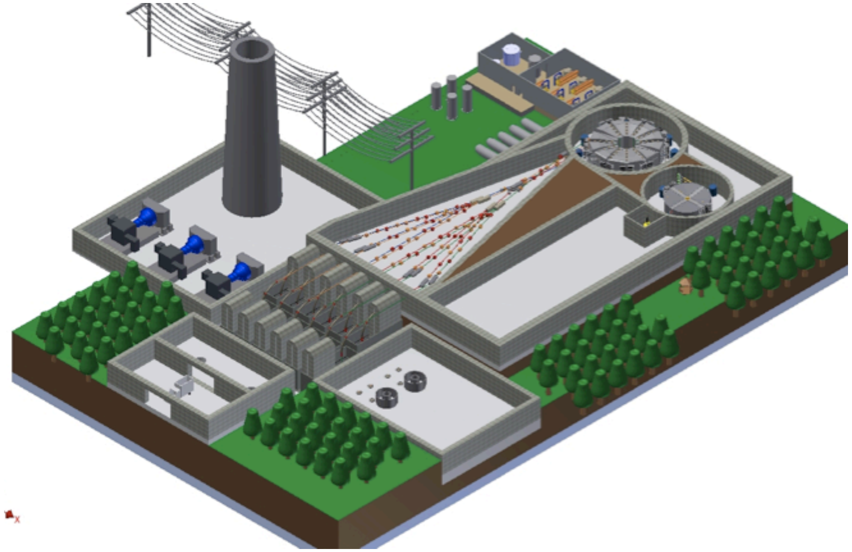
Mean neutron energy 10.6 MeV



140 dpa/year in 1x1x0.6 cm^3

- 140 dpa/year of fast neutron damage to samples
- Fast spectrum: $\langle E_n \rangle$ can be adjusted by target angle 2-10 MeV
- Provides for all materials studies for fast fission reactors and for fusion first wall

Destroying transuranics is the gift we can give our future generations…



Our plans to make it all happen:

- 2014-2016 Build LEDA-based n damage facility AND-1
- 2017-2019 Build 3-beam 100 MeV 140 dpa/year AND-2
- 2020-2022 Build ADSMS Isoburner core and TAMU800
- 2023-2024 Commission with La surrogate fuel
- 2025 Operate ADSMS with TRU/U fuel

The ADAM Collaboration

Texas A&M University:

Physics: Saeed Assadi
Karie Badgley
Austin Baty
Kyle Damborsky
James Gerity
Joshua Kellams
Tom Mann
Al McInturff
Peter McIntyre
Nate Pogue
Akhdiyor Sattarov
Elizabeth Sooby

Nuclear Engineering:
Pavel Tsvetkov

Chemistry: Abraham Clearfield

University of Utah:
Michael Simpson

VCU:
Supathorn Phongikaroon

Brookhaven National Lab
Bill Horak

Seoul National University
Il Soon Hwang

blue = students!

Please join us!

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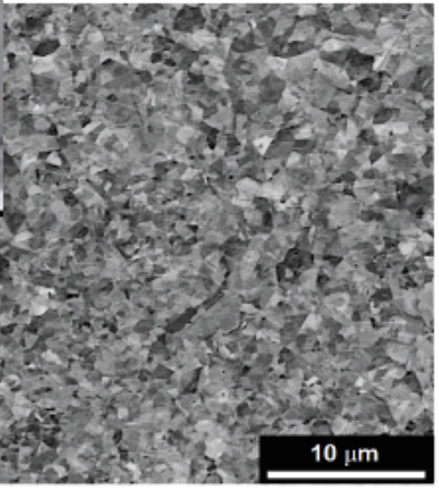
Material issues for the core vessel, beam window, HX

- Ni is the most robust metal against chemical corrosion in the molten salt eutectic at 550-650 C
- Ni exhibits 2 forms of radiation damage:
  - swelling from dislocations and voids caused by n scatters
  - He embrittlement by an (n,a) reaction of fast neutrons
- Swelling limits at <20% for Ni, provide for that in the mechanical design
- The dominant embrittlement actually arises from a pathway through ^{59}Ni which is mediated by thermal neutrons, and we have 100x less than any ‘fast’ reactor.

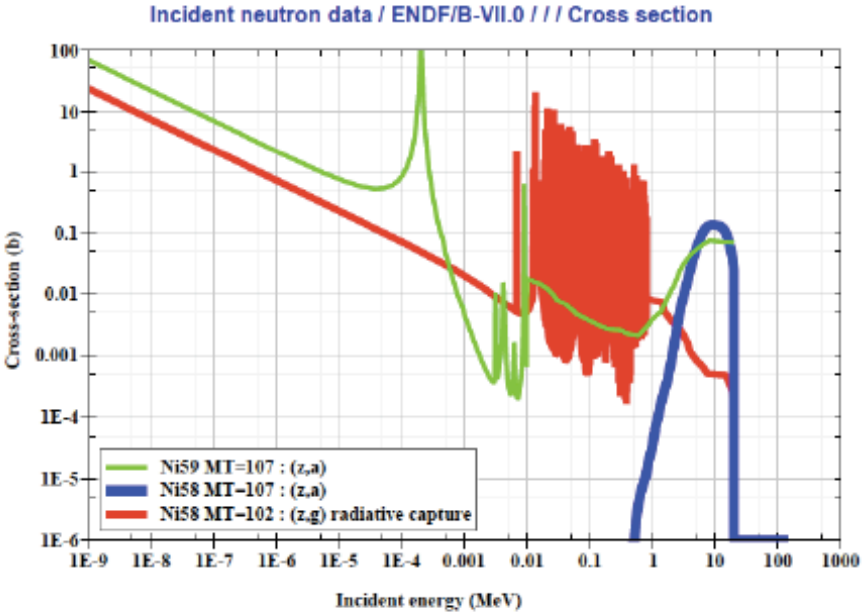
Nickel Vapor Deposition – Seamless, Weld-free Nickel liner

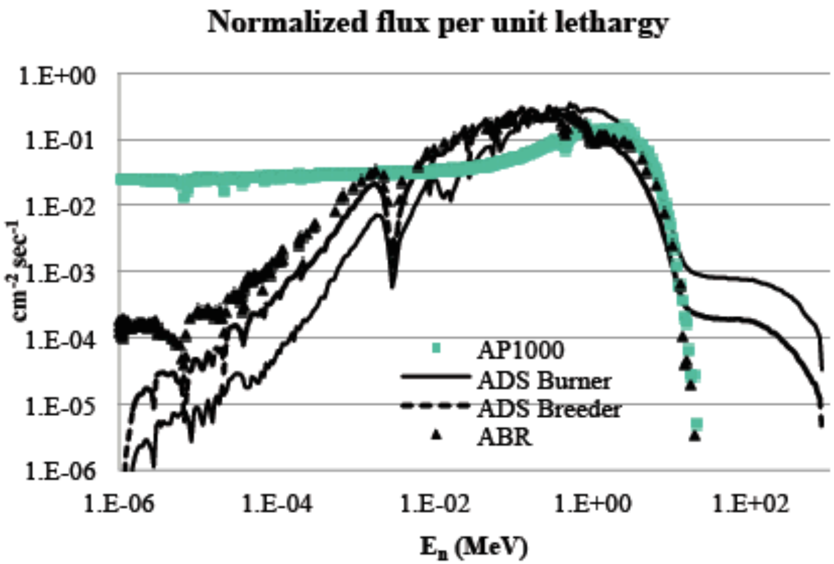


1. 0.25mm per 1 hr deposition rate
2. Uniform thickness during growth
3. Negligible residual stress
4. Cavities made sequentially from the same mandrel

Weber can fabricate a seamless hermetic liner for the entire core + PHX assembly. We have collaborated with them to develop improvements to their process that produce uniform high-temp-stable μm grain size, extreme toughness, no porosity.

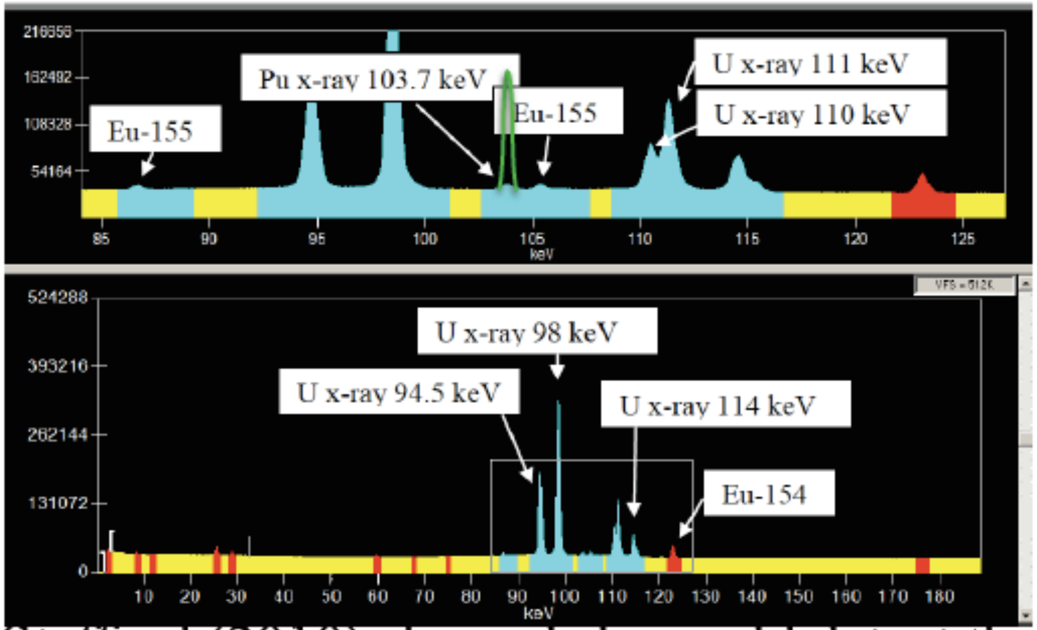
Pathway to embrittlement in Ni





^{58}Ni(n,γ)^{59}Ni, then ^{59}Ni(n,α)^{56}Fe,
energetic α produces lots of local damage in lattice.
But this chain requires thermal n flux, and we have very little.
We need to do experimental validation, looks probably OK for >20 y lifetime.

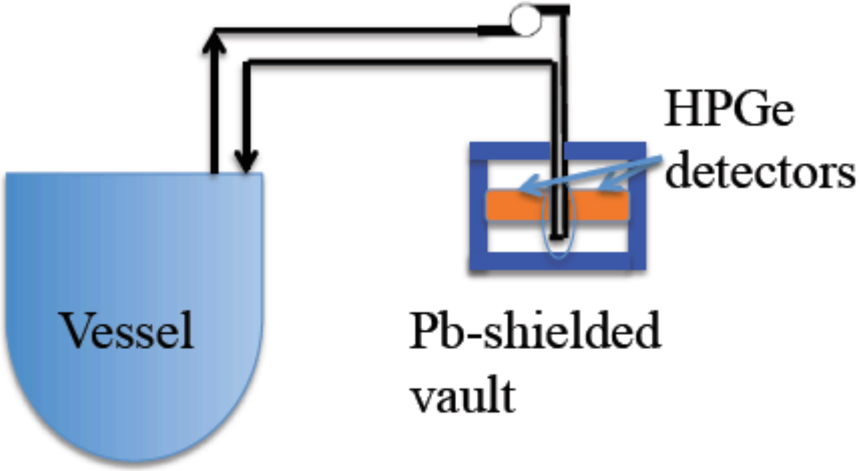
In situ verification of TRU element inventories using X-ray fluorescence

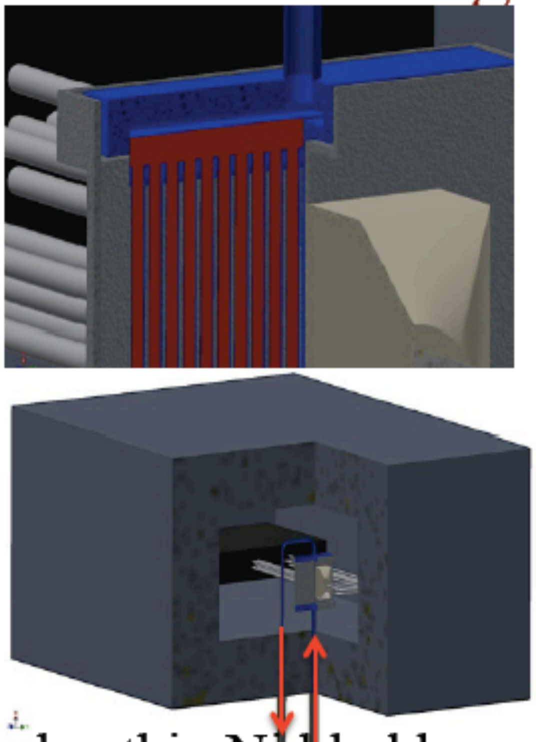


Note: ~1% Pu in pin; ADSMS fuel has ~18%!

- Stafford (2010) showed she could detect the K-shell X-rays of Pu in the spectrum of self-induced X-rays from SNF pins.
- But it would be extremely difficult to make any precise assay, because the mfp of the X-rays is only ~0.3 mm, Pu has a radial distribution that is non-uniform and variable, and any buildup of corrosion on pin cladding would produce an absorbing layer.

Recirculating sample flow of molten fuel salt through shielded detector *in situ* verification





Pump a closed-circuit sample flow of fuel salt through a thin Ni bladder: two thin Ni foils welded around their edges to make a hermetic bladder with supply and return tubes welded in the ends. Two HPGe detectors are located on the flat side faces to detect the X-rays.

We expect to be able to make continuous calibrated logging of concentrations of Pu, Am, and other TRU with accuracy <.5%.