Plasma Mass Filters for Nuclear Waste Remediation

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PPPL 2/20/18

Overview of a draft report based on LDRD work 2012-2018:

1. Introduction and motivation
2. Mechanisms for plasma separation
3. Generic physics issues
4. Generic technology issues
5. R&D plan

Hanford Nuclear Waste Site

- Waste comes from plutonium production reactors run from 1943-1987, designed in part by Fermi, Wigner, and Wheeler

- 177 large underground waste storage tanks totaling ~2×10^5 m^3 of waste, with ~2×10^8 Ci of radioactive material (mainly Cs and Sr), plus many toxic, corrosive, and explosive chemicals use for processing Pu

tank under construction

present surface of tank waste

Present Waste Remediation Plan

- Tank waste is chemically "pretreated" to separate high-level radioactive waste from a much larger volume of low-level radioactive waste
- High level waste to be vitrified in glass canisters and buried (e.g. at Yucca mountain); low level waste to be mixed with cement and stored in shallow burial on site

present waste treatment plant at Hanford

high-level waste canisters (SRS)

D. Kramer, Physics Today July 2017

Plasma Mass Filter Idea for Nuclear Waste Pretreatment

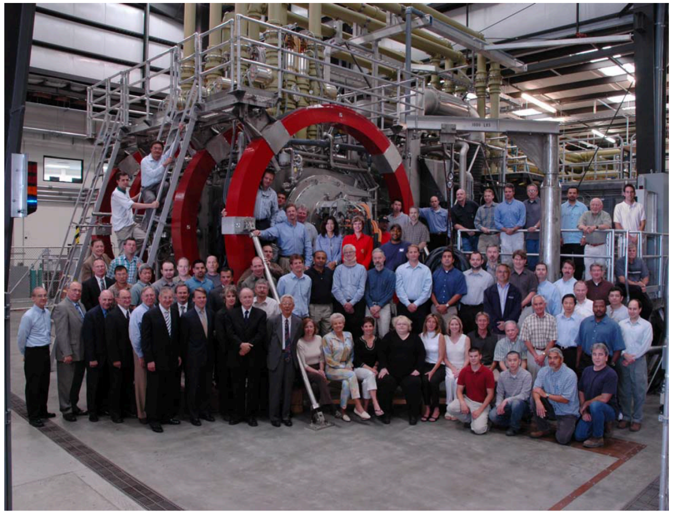
- Almost all radioactive species have high mass, e.g. ^{137}Cs, ^{90}Sr, ^{239}Pu, ^{235}U
- First convert all the nuclear waste into singly-charged ions inside a plasma
- Separate high mass ions ($M \geq 90$) from low mass ions using plasma physics
- Remove high mass stream for disposal with high-level radioactive waste

this does not (and should not) separate isotopes !

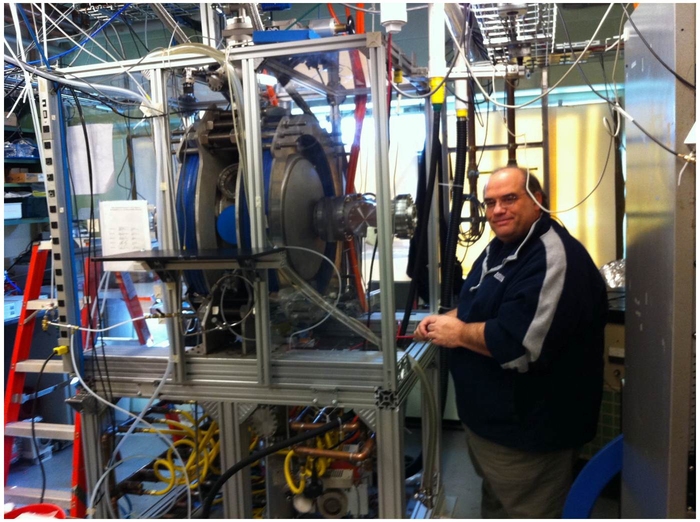
$M \geq 90$ separation does not have to be perfect !

History of Plasma Ion Mass Separation Experiments

Device (country)	species	year(s)	Ref's
calutron (Berkley, ORNL)	U isotopes	1941-present	5-8
FI torus (Sweden)	H/Ar	1966-71	9
ICR (US, USSR, France)	many isotopes/elements	1976-present	10-13
plasma centrifuge (Yale)	metal isotopes and elements	1980-87	14-16
vacuum arc centrifuge (Australia)	Cu/Zn and their isotopes	1989-99	17-19
PCEN vacuum arc centrifuge (Brazil)	C, Al, Mg, Zn, Cd, Pb etc.	1987-98	20-23
Archimedes filter (San Diego)	Xe/Ar and Cu/Ag/Au ?	1998-05	24-31
linear device with electrodes (Kyushu)	Ar, Xe	2007	32
POMS-E-3 (Irkutsk)	N, Ar, Kr	2010-15	33,34
vacuum arc separator (Irkutsk)	Ni, Cr, Fe, W	2011-15	35,36
PMFX (PPPL)	Ar/Kr	2013-14	37,38
SNF separator (JIHT Moscow)	U, Gd, He	2013-16	39-44



Archimedes Technology (2005)



PMFX @ PPPL (2014)

Required Throughput and Energy Requirement

- To process 25% of total Hanford inventory ($\sim 10^8$ kg) over 30 years (24/7)

$\Gamma \sim$ 100 g/sec average throughput

- Assuming energy for ionization and plasma heating $\sim$500 eV/atom

P $\sim$ 100 MW average power requirement

Plasma processing of any significant fraction of Hanford waste would be a huge task, comparable to ITER

R. Gueroult et al, PPCF 60, 014018 (2018)

Is This Throughput Possible in a Plasma ?

- Maximum possible throughput for collisionless ions is roughly:

$$\Gamma \sim 1/2\, n_i\, v_i\, A$$

- Assuming optimistic scenario: $n_i = 10^{14}$ cm^{-3}, $T_i \sim 10$ eV, Z=1, M=40, $A = 10^4$ cm^2

$$\Gamma \sim 2.5\text{x}10^{23} \text{ atoms/sec} \sim 20 \text{ gr/sec}$$

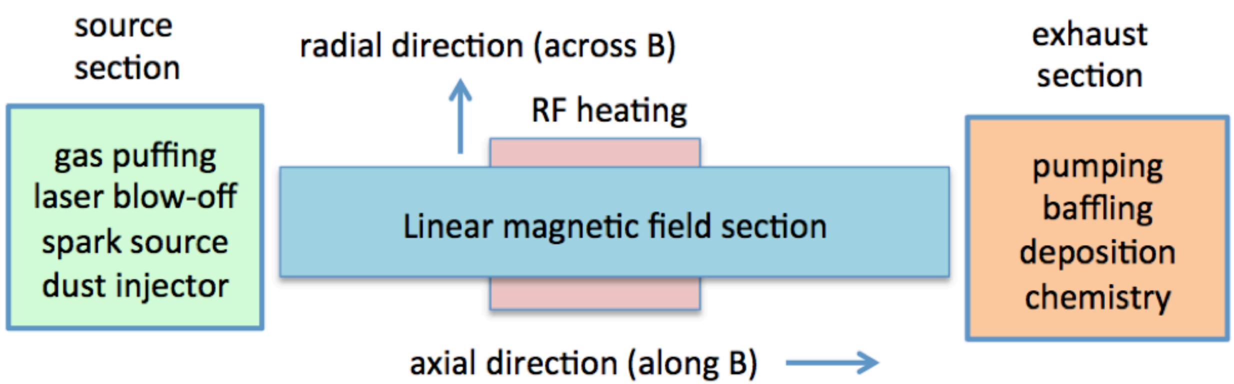
- But mean free path for ion-ion collisions would be roughly

$\lambda_{ii} \sim \nu_{ii}\, v_i \sim 1$ cm (so ions will *not* be collisionless)

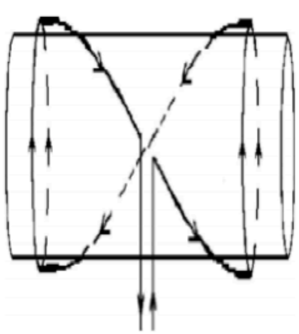
Perhaps 100 g/sec throughput might be obtained with ~ 20 plasma devices of ~ 5 MW each

Generic Plasma Mass Separation Device

- Many possible options based on *differential ion confinement* in magnetic fields

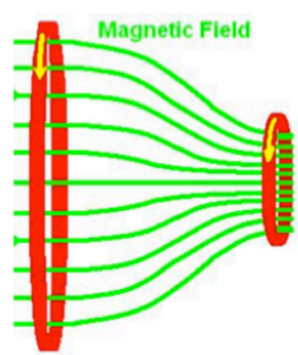


RF heating end



Electrode end

Mirror end



Curved end

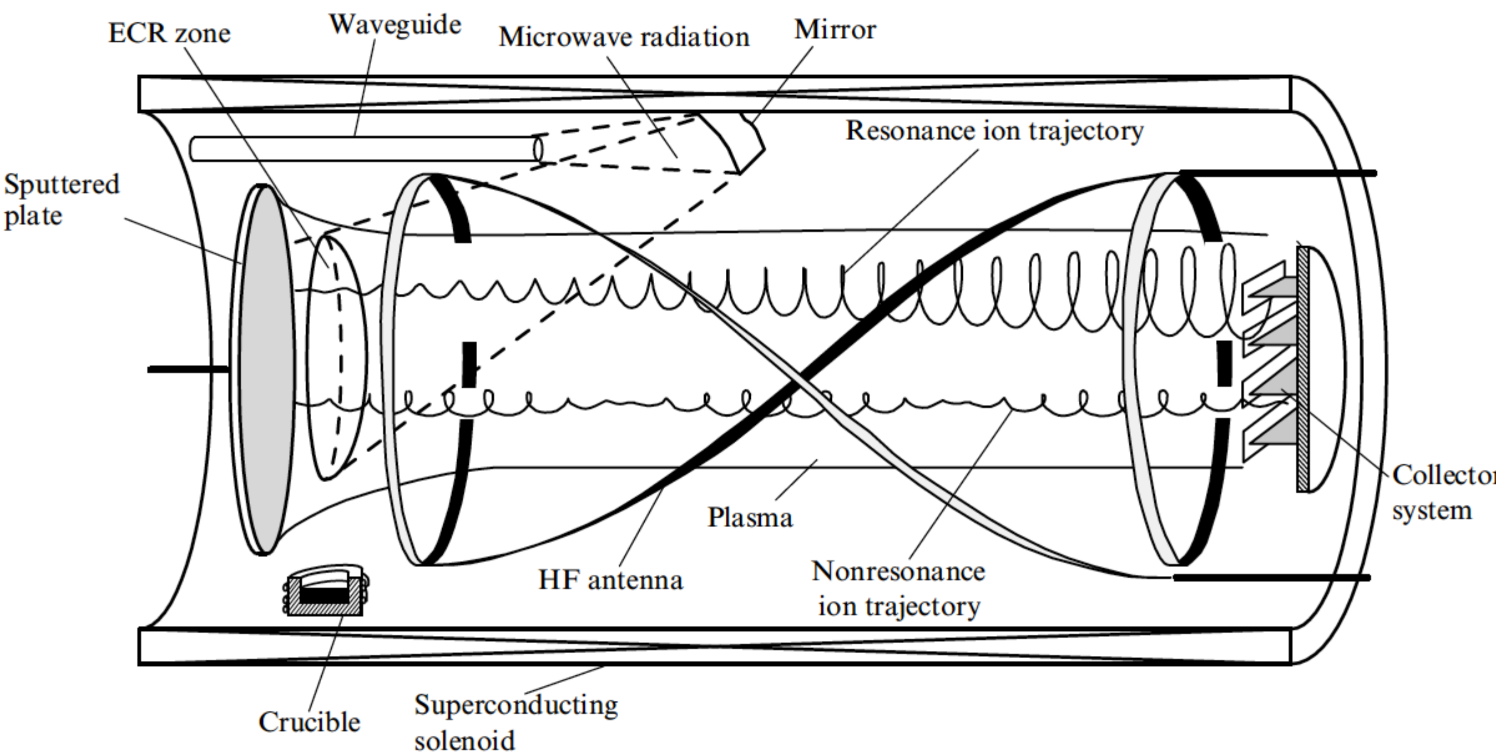
2. Mechanisms for Plasma Mass Separation

- Each has advantages and disadvantages, e.g. in simplicity and throughput

Section	Separation mechanism
2.1	ion gyroradius
2.2	ion drifts in a curved magnetic field
2.3	plasma centrifuge (vacuum arc centrifuge)
2.4	rotating plasmas (uniform magnetic field)
2.5	rotating plasmas (variable magnetic field)
2.6	rotating plasmas (azimuthal magnetic field)
2.7	ion mobility in an electric field
2.8	radial advection in rotating plasmas
2.9	ionization energy
2.10	dusty plasma separation
2.11	radial diffusion in a linear magnetic field
2.12	transit time separation
2.13	collisionality gradient

Ion Gyroradius Separation

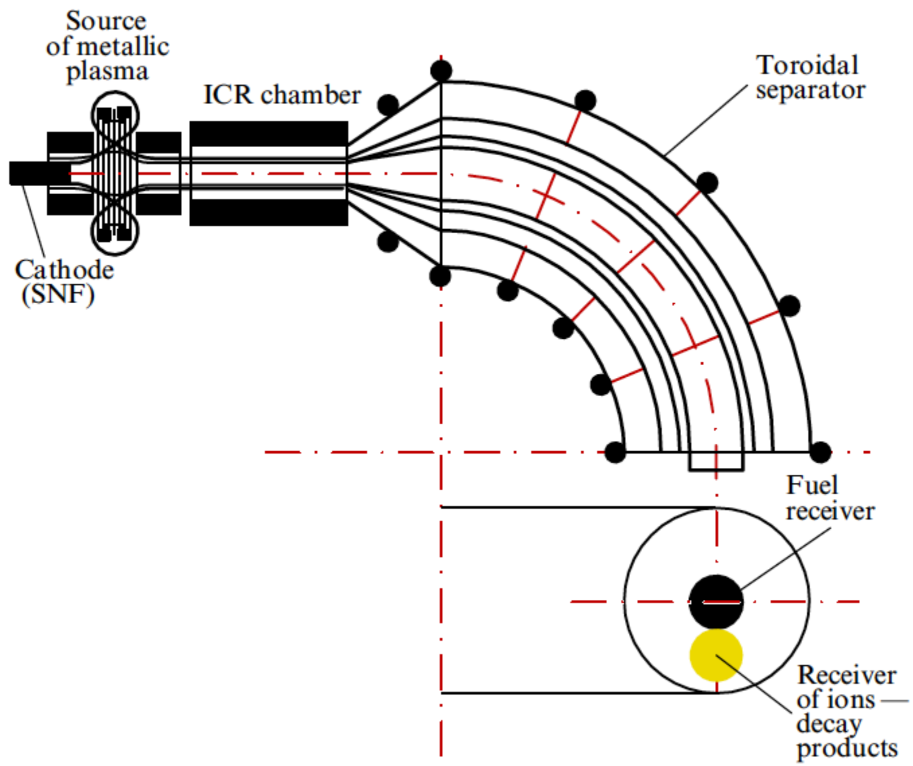
- Heat ions in selected mass range and physically separate using baffles or slits
- Several devices used for isotope/chemical separation in US, Russia, and France



D.A. Dolgolenko and Yu. A. Muromkin, Physics-Uspekhi 52, 345 (2009)

Ion Drifts in Curved Magnetic Field

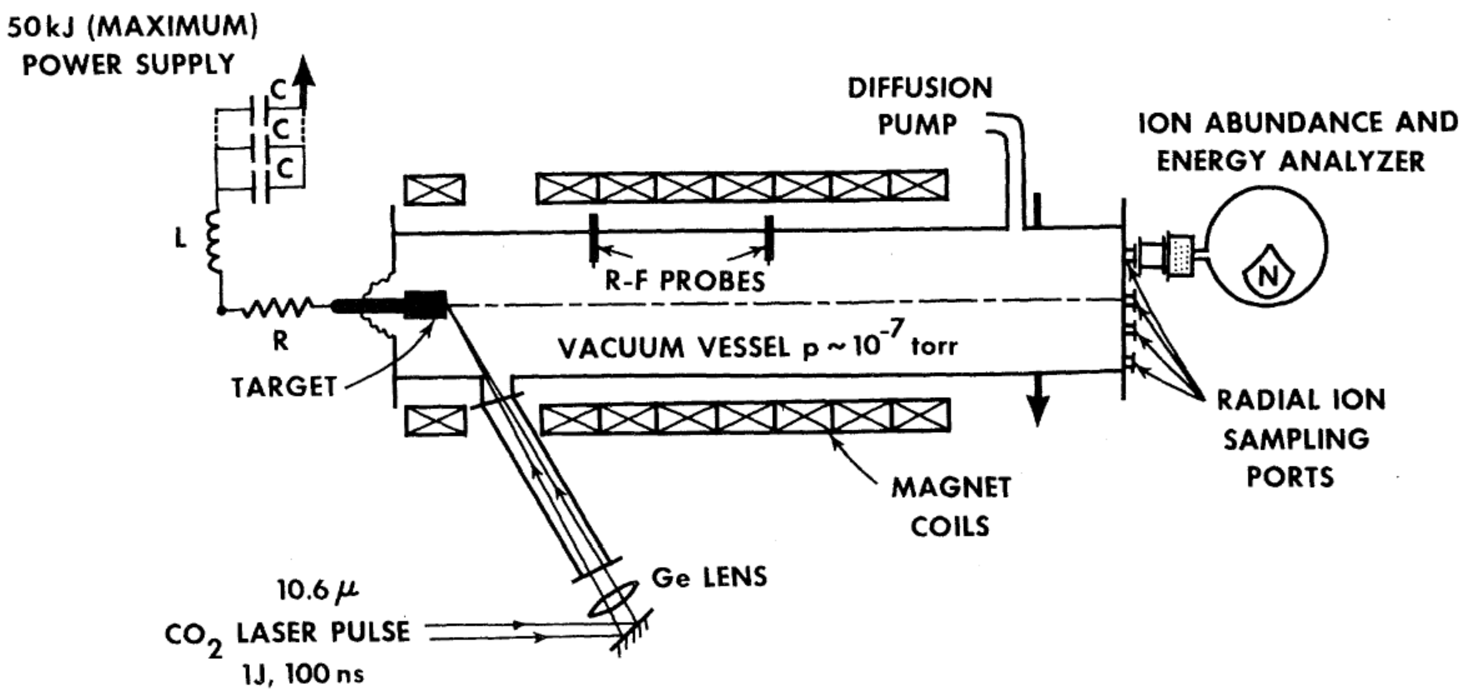
- Heat ions and use curvature/grad-B drifts and separate vertically in curved B field
- Small tests in 1960's with some separation, but self-generated E field is a problem



A.V. Timofeev, Physics-Uspekhi 57 990 (2014)

Plasma Centrifuge (a.k.a. Vacuum Arc Centrifuge)

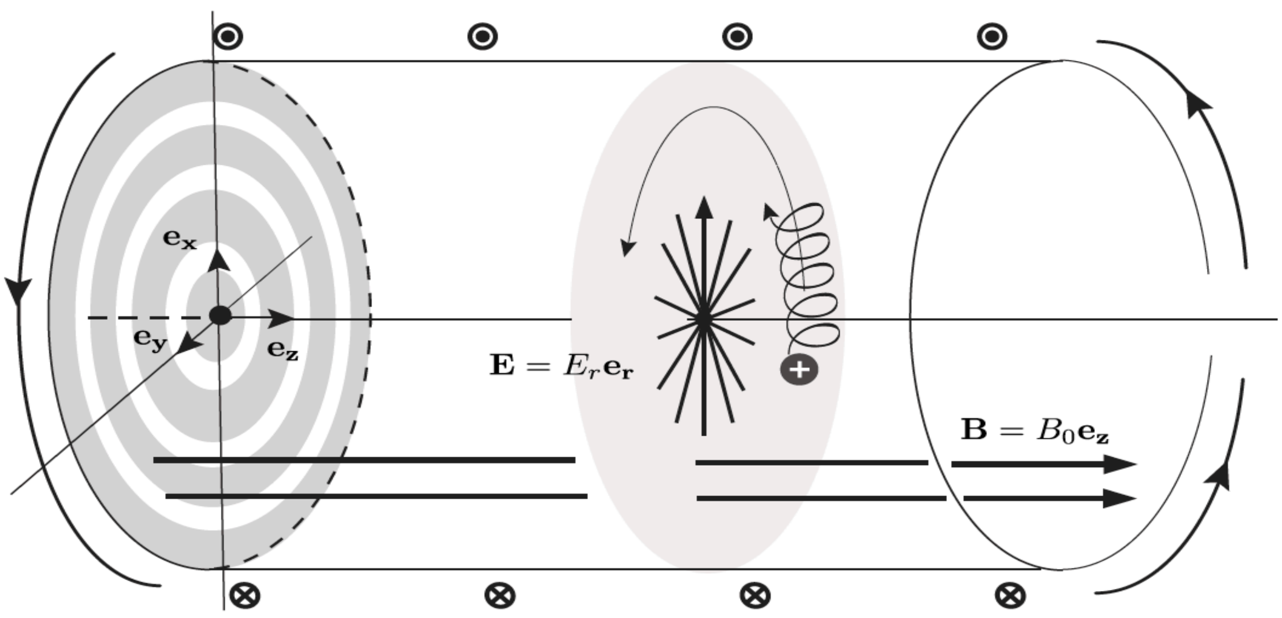
- Pulsed current from arc creates strong azimuthal rotation and radial ion separation
- Some isotope/metal separation obtained in experiments at Yale, Australia and Brazil



M. Krishnan, PRL 46, 36 (1981)

Rotating Plasmas (Uniform Magnetic Field)

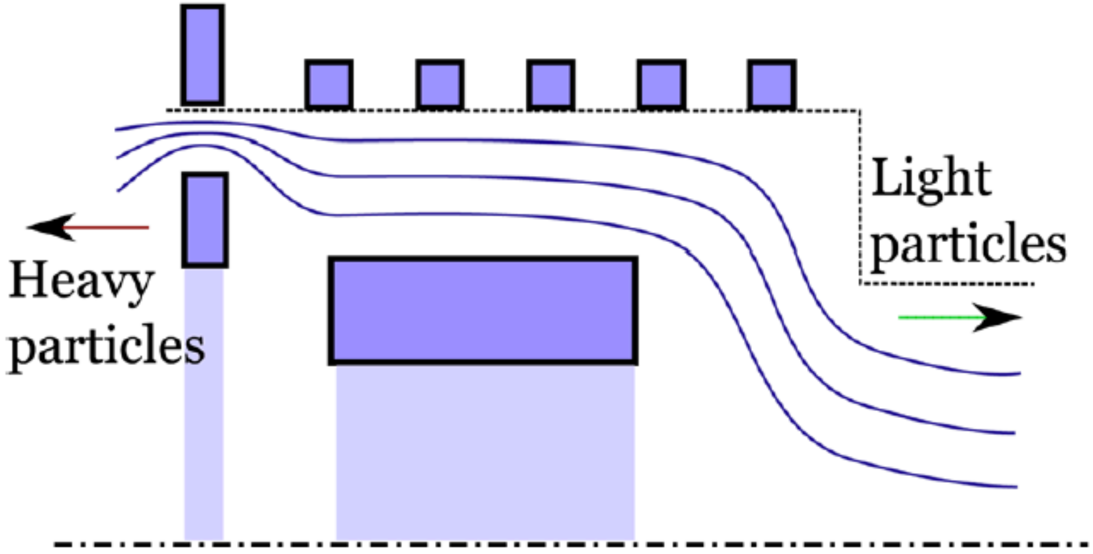
- Radial electric field from end electrodes for ExB rotation and differential orbit loss
- ExB rotation obtained in some experiments, but magnitude is difficult to control



R. Gueroult et al, PPCF 014018 (2018)

Rotating Plasmas (Non-uniform Magnetic Field)

- Asymmetrical centrifugal trap combines ExB rotation and magnetic mirror effects
- Allows axial separation of heavy and light ions at moderate ion collisionality

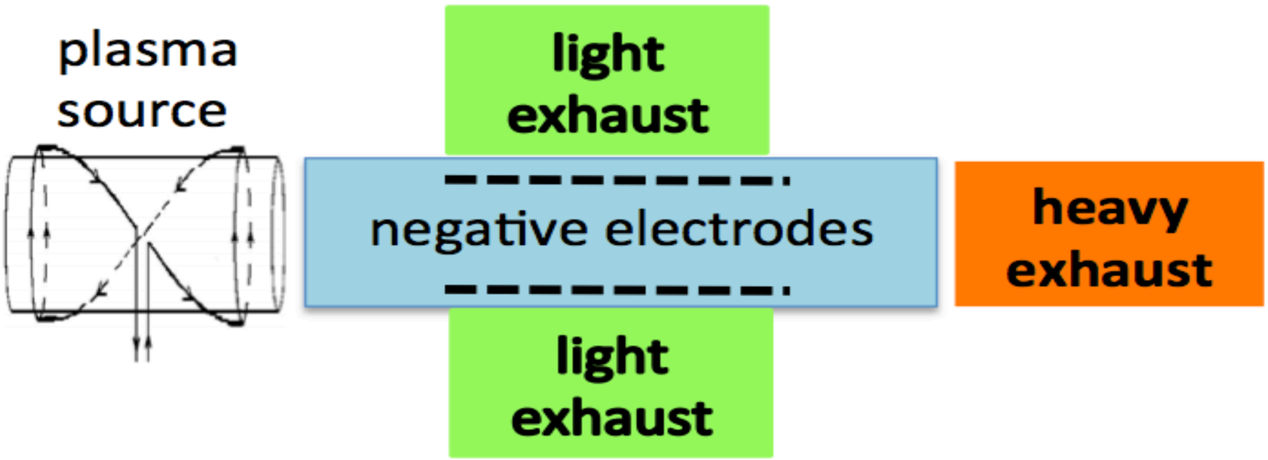


MCMF (magnetic centrifugal mass filter

Fetterman and Fisch, Phys. Plasmas 094503 (2011)

Separation by Ion Mobility or Ionization Energy

- Ion drift speed in a plasma with an electric field should be higher for low mass ions
- Might separate heavy from light ions without a magnetic field, if E field penetrates
- Low ionization potential of Cs may allow differential collection of radioactive ions



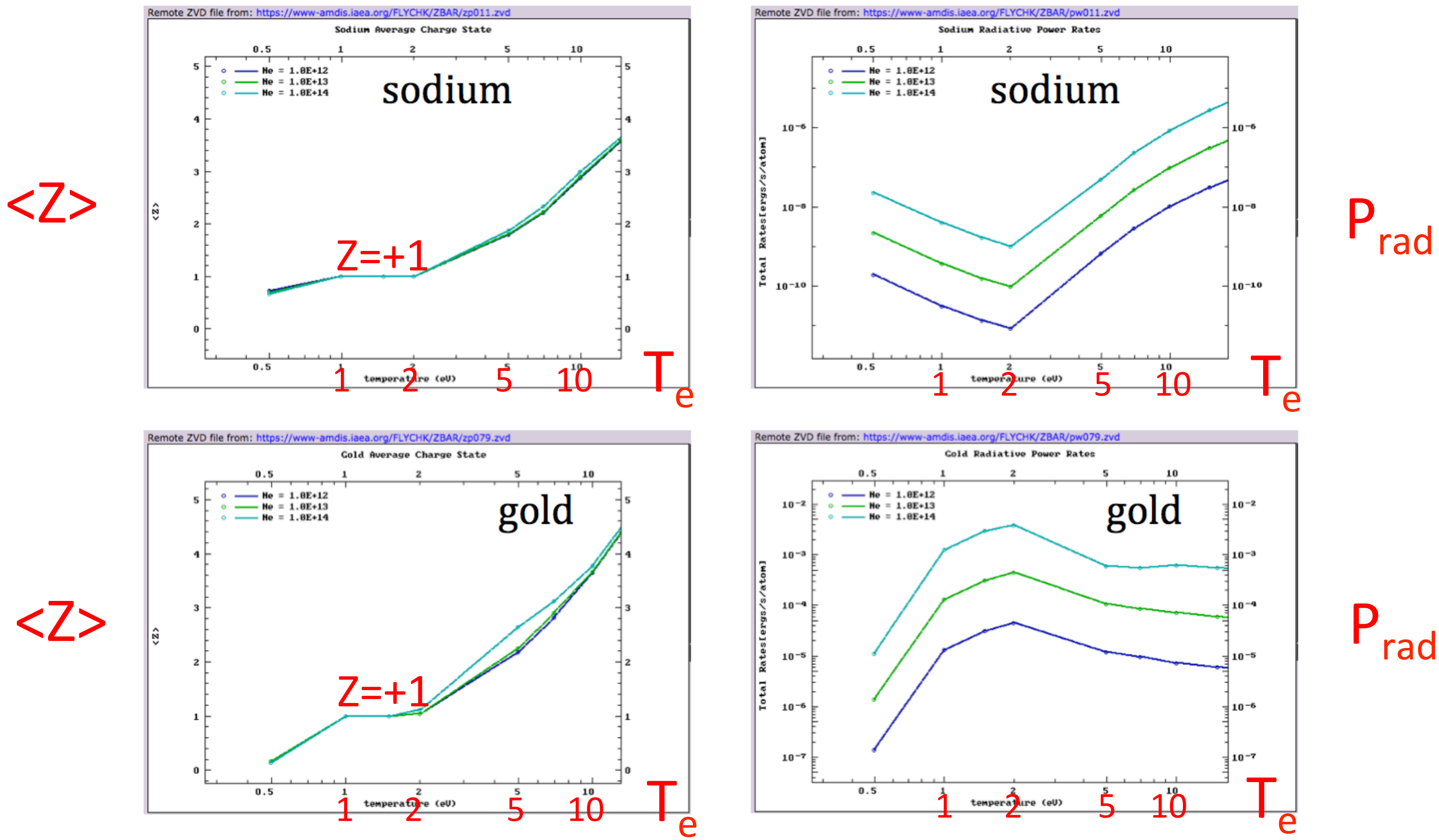
3. Generic Physics Issues

- Almost all ion mass separation mechanisms have these generic physics issues:

Section	generic physics issue
3.1	charge state and radiated power
3.2	molecular ions and plasma chemistry
3.3	charge exchange and recombination
3.4	neutral gas transport
3.5	droplets, dust, and nanoparticles
3.6	collisional effects
3.7	electric fields and rotation
3.8	plasma fluctuations and mixing
3.9	energy loss due to plasma transport
3.10	ion throughput and separation efficiency

Charge State and Radiated Power

- Average ion charge is +1 for only a limited range of T_e ~ 1-2 eV for most atoms
- Radiated power very high even at T_e = 2 eV, e.g. 1% gold @ $n_e=10^{13}$ cm^{-3} ~5 MW/m^3



IAEA Atomic Molecular Data https://www-amdis.iaea.org/FLYCHK/

Molecular Ions and Plasma Chemistry

- Most molecules not dissociated below T_e ~ 2 eV, so will remain ± ions in the plasma
- Total mass of molecules with of light atoms might be significantly changed
- Plasma chemistry is complicated and will make modeling plasma difficult

dissociation
energy ~10 eV
for most molecules

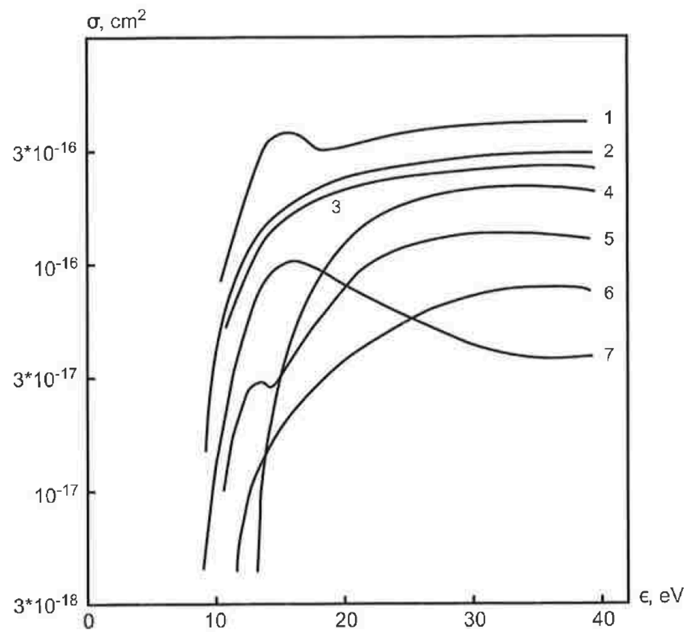


Figure 2–25. Cross sections of dissociation of molecules through electronic excitation as a function of electron energy: (1) CH_4, (2) O_2, (3) NO, (4) N_2, (5) CO_2, (6) CO, (7) H_2.

1 – CH_4
2 – O_2
3 – NO
4 – N_2
5 – CO_2
6 – CO
7 – H_2

A. Fridman, Plasma Chemistry, Cambridge 2008

Charge Exchange and Recombination

- Cs and Sr (radioactive species) have relatively high cross-sections for charge exchange
- Recombination can also be a common process for molecular ions, e.g. $e + N_2^+ = N$
- Both process could interrupt and compromise any of the ion separation mechanisms

for Cs @ 4 eV
$\sigma(cx) \sim 3 \times 10^{-14}$ cm^2
Lcx => 0.3 cm
@ $n_{Cs} = 10^{14}$ cm^{-3}

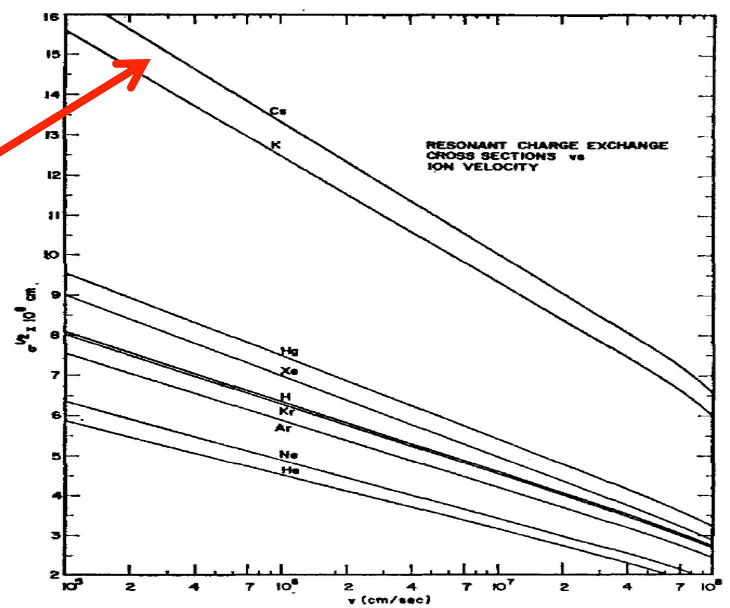


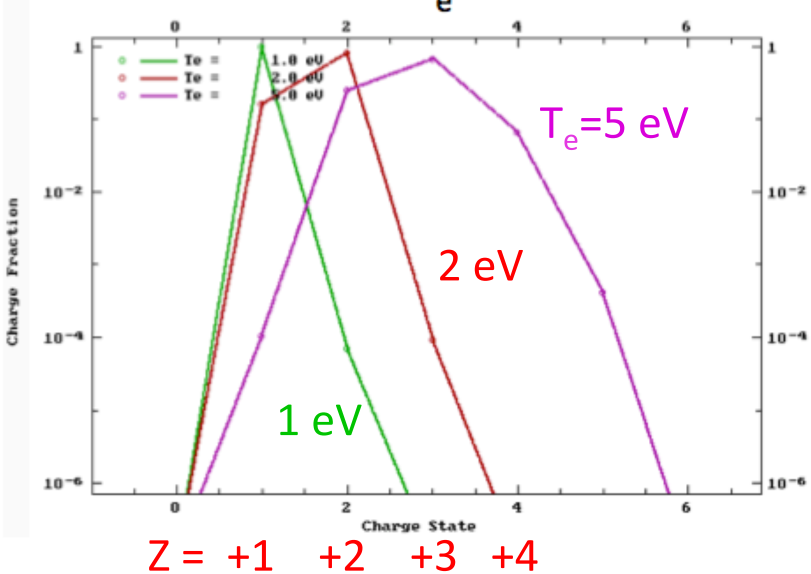
FIG. 2. Calculated cross sections for resonant charge exchange between monatomic ions and their parent gases. Interpolation can be made for other gases in terms of their ionization potentials (listed in Table I).

[3] D. Rapp and W.E. Francis, J. Chem. Phys. 37, 2631 (1962)

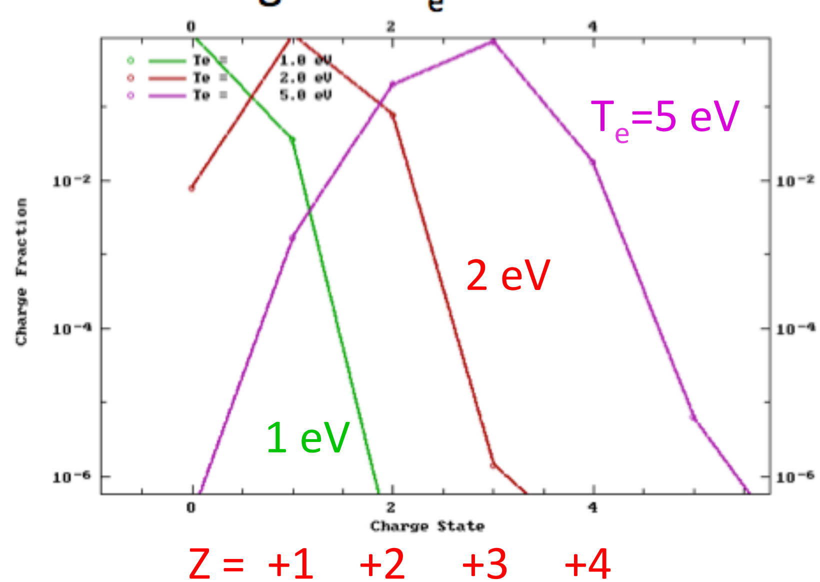
Neutral Gas Transport

- Neutrals will dominate the charge distribution of *some* species in T_e = 1-2 eV range (e.g. at T_e=1 eV, 100% Cs @ Z=1 but 3% Ar @ Z=1)
- Neutral transport will compromise total separation efficiency, depending on n_o/n_i
- Difficult in practice to maintain $n_o/n_i < 1$ in most low temperature plasmas

cesium at $n_e = 10^{13}$ cm^{-3}



argon at $n_e=10^{13}$ cm^{-3}



IAEA Atomic Molecular Data https://www-amdis.iaea.org/FLYCHK/

Collisional Effects

- Ions collisions will be important for devices operated at desired high n and low T
- PMFX helicon device at PPPL was strongly dominated by collisions, with $\nu_{ii}/\Omega_i > 1$
- Collisional effects can be reduced at lower n, higher T_i, and higher magnetic field

parameters for PMFX-like devices

	PMFX*	PMFX-U	PMFX-LD
n_j (cm^{-3})	10^{13}	10^{13}	3×10^{11}
n_n (cm^{-3})	10^{14}	10^{14}	10^{13}
B (G)	950	15 000	2500
E (V/cm)	2	5	5
μ_i (a.m.u.)	80	80	80
μ_j (a.m.u.)	40	40	40
T_i (eV)	1	3	1
T_e (eV)	5	5	5
$L_{\|}$ (cm)	40	40	40
$L_{\perp}$ (cm)	8	12	12

collisional effects for PMFX-like devices

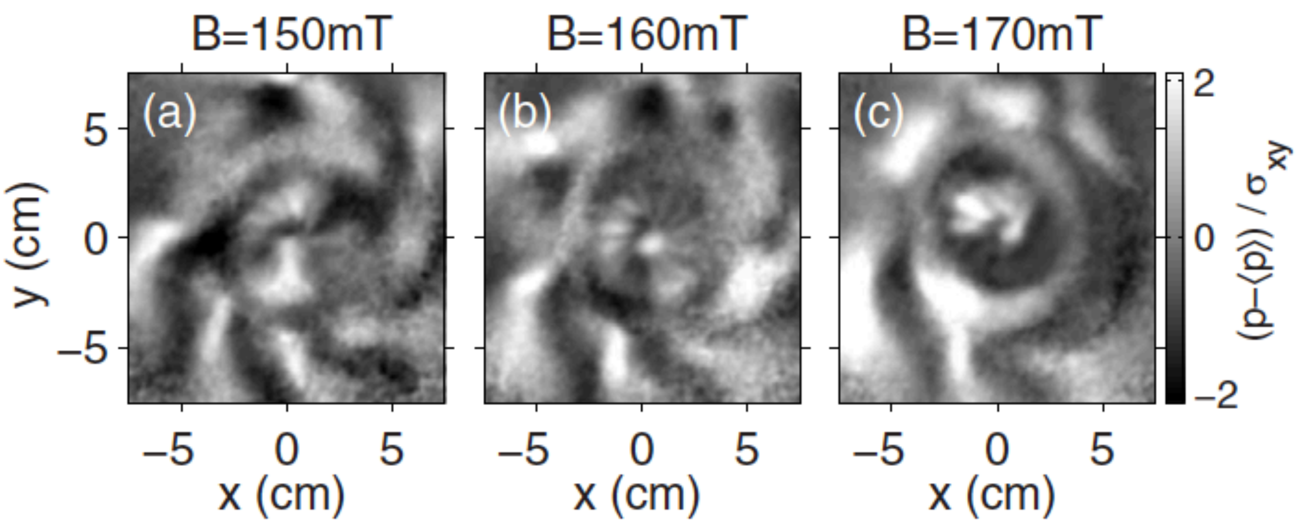
	PMFX*	PMFX-U	PMFX-LD
v_{thi} (cm/s)	1.1×10^5	1.9×10^5	1.1×10^5
t_{pD} (s)	3.2×10^{-2}	2.1×10^{-3}	9.7×10^{-4}
ρ_i (cm)	0.94	0.10	0.36
$v_{E\times B}$ (cm/s)	2.1×10^5	3.3×10^4	2.0×10^5
Ω_i (s^{-1})	1.1×10^5	1.8×10^6	3.0×10^5
ν_{ii} (s^{-1})	1.5×10^6	2.8×10^5	4.4×10^4
ν_{in} (s^{-1})	2.9×10^4	2.9×10^4	2.9×10^4
τ_M	0.12	9.6	10
τ_c	4.0×10^{-2}	12	13
τ_n	4.0×10^{-2}	8.1	4.8
τ_B	1.5×10^{-2}	8.3	3.0

I. Ochs et al, Phys. Plasmas 244, 043503 (2017)

Plasma Fluctuations and Mixing

- Almost all plasmas have self-generated E and B fluctuations which can drive transport
- Fluctuating ExB drifts will not directly separate different masses, but can mix plasma
- These effects can be minimized by using fast separation mechanisms, e.g. orbit loss

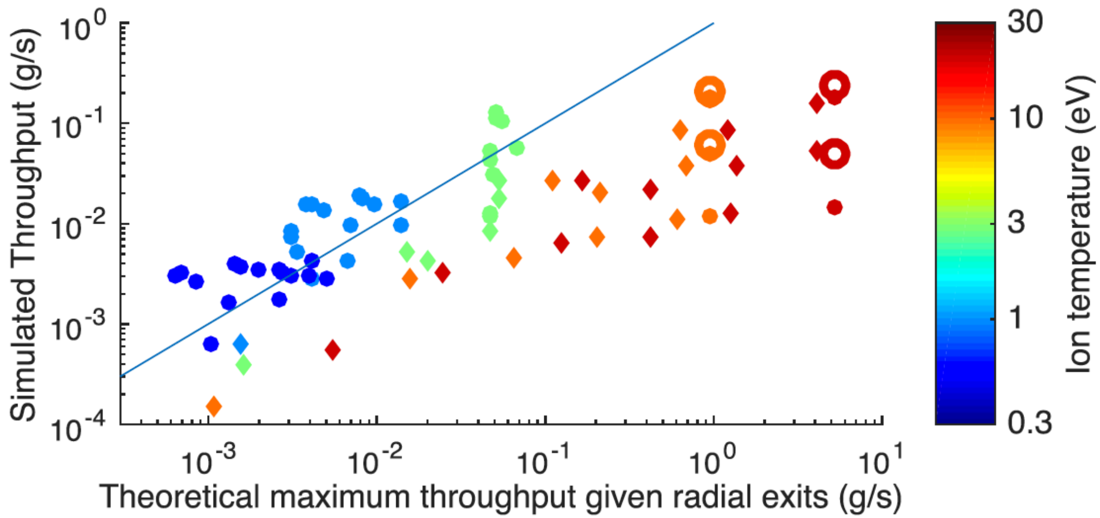
instabilities in the CSDX linear helicon device (UCSD)



S.C. Thakur et al, Phys. Plasmas 23, 082112 (2016)

Ion Throughput and Separation Efficiency

- Ion throughput *does not* increase with density in the collisional limit where $\nu_i \sim n$
- Ion throughput *does* generally increase with T_i, so desirable temperature $T_i \geq 10$ eV
- System should be designed for highest throughput at specified separation efficiency



I. Ochs et al, Phys. Plasmas 244, 043503 (2017)

4. Generic Technology Issues

4.1 Plasma sources

- avoid droplets, dust, and nanoparticles with too-low Z/M for separation
- avoid generation of plasma fluctuations, such as occur in vacuum arcs
- best options may be thermal evaporation and ion sputtering

4.2 Waste handling

- batch transfer with vacuum interlocks needed for feeding/extraction
- vacuum pumping and cleaning of vacuum chamber will be difficult
- whole operation may have to be remotely controlled due to radiation

4.3 Plasma heating and magnets

- RF antennas in vacuum may become coated and are subject to arcing
- RF antennas outside vacuum may be shielded by conducting coatings
- magnets above few kG may need to be superconducting (or permanent)

4.4 ES&H issues

- exhaust gases from vacuum pumps need to be processed safely
- vacuum leaks relatively harmless but may need to be fixed remotely
- water coolant leak inside hot vessel could be serious risk, as in ITER

5. R&D plan

- Possible Staged goals
- Some criteria for evaluating separation mechanisms
- Needs for theory and simulation
- Process diagnostics
- Possible proposals

Possible Staged Goals

1) evaluate various plasma mass separation mechanisms with flexible small experiments

   use gas mixtures (e.g. Ne/Ar or WF_6) and small samples of solids (e.g. FeO)

2) demonstrate plasma mass separation at low throughput with simulated nuclear waste

   try for ~1-10 mg/sec for long pulses (≥ 1 hour) with realistic simulants

3) demonstrate plasma mass separation at low throughput with real nuclear waste

   will need thorough ES&H review to use ~10-100 gr of radioactive waste

4) demonstrate plasma mass separation at high throughput with simulated nuclear waste

   try for ~0.1-1 g/sec for long pulses (≥ 1 hour) with realistic simulants

5) demonstrate plasma mass separation at high throughput with real nuclear waste

   most likely needs to be done onsite to avoid transportation of waste

Some Criteria for Evaluating Separation Mechanisms

- This preliminary evaluation is based on intuition, not quantitative analysis
- Scale of: 1=good, 2=average, 3=poor, lowest total => most likely to succeed
- Best mechanisms seem to be: ion gyroradius, curved B, and simple diffusion

Section	mechanism	history	simplicity	robustness	throughput	efficiency	total
2.1	gyroradius	1	1	2	2	2	**8**
2.2	curved B	1	1	1	1	3	**7**
2.3	centrifuge	1	2	3	3	1	**10**
2.4	rotate in B	2	3	2	2	2	**11**
2.5	rotate vary B	3	3	2	2	3	**13**
2.6	azimuthal B	3	3	2	2	3	**13**
2.7	mobility	3	2	3	1	2	**11**
2.8	advection	3	3	3	2	2	**13**
2.9	ionization	2	2	2	1	3	**10**
2.10	dust	2	2	3	1	3	**11**
2.11	diffusion	2	1	1	2	2	**8**
2.12	transit time	2	2	1	1	3	**9**
2.13	collisionality	3	2	2	2	2	**11**

Needs for Theory and Simulation

- Atomic physics – charge state, radiation, molecules, CX and recombination
- Particle transport – orbit effects, collisions, neutrals, charged dust
- Plasma dynamics – electric fields, rotation, fluctuations, plasma energy loss
- Engineering - ion source, waste removal, plasma heating, control systems

Section	generic issue	theory/simulation code
3.1	ion charge and radiation	atomic physics
3.2	molecules and chemistry	atomic physics
3.3	CX and recombination	atomic physics
3.4	neutral gas transport	particle transport
3.5	large particles	particle transport
3.6	collisional effects	particle transport
3.7	electric fields and rotation	plasma dynamics
3.8	plasma fluctuations	plasma dynamics
3.9	plasma energy loss	plasma dynamics
3.10	throughput and efficiency	atomic+particle+dynamics+engineering
4.1	ion source	engineering systems
4.2	waste handling	engineering systems
4.3	plasma heating	engineering systems
4.4	ES&H	engineering systems

Process Diagnostics

- Plasma diagnostic needed to understand physics and validate codes
- Surface diagnostics needed to demonstrate separation and throughput

diagnostic	measurement
Langmuir probe	electron temperature and density
Mach probe	ion rotation speed
visible spectroscometer	ion flow and temperature
gridded energy analyzer	ion distribution function
survey spectrometers	species location and charge state
bolometer	radiated power
fast camera	plasma fluctuations
residual gas analyzer	gas composition at a few points
laser ellipsometry	thin film thickness
quartz microbalance	deposition thickness
LIBS	surface chemical composition
x-ray flourescence	surface chemical composition
Raman spectroscopy	surface chemical composition
removable coupons	standard chemical analysis
laser scattering	dust in plasma
gamma detector (ex-vessel)	Cs^{137} content and location
beta detector (in vessel)	Cs^{137} and Sr^{90} content
vessel wall temperature	thermocouples, IRTV

Possible Proposal(s)

Theory and Simulation

- Develop necessary codes (atomic physics, particle transport, plasma dynamics)
- Develop new ideas which could make this process simpler and easier

Experiments

- Develop and test suitable ion sources (evaporation, sputtering, dust…)
- Start on flexible stage 1 experiments with appropriate diagnostics

Engineering

- Consult with engineers at Hanford to understand reality of waste handling
- Develop technology needed for RF heating, vacuum and control systems