Princeton Plasma Physics Laboratory

PPPL- 4627

PPPL- 4627

Plasma Mass Filters For Nuclear Waste Reprocessing

Abraham J. Fetterman and Nathaniel J. Fisch

May, 2011

Prepared for the U.S. Department of Energy under Contract DE-AC02-09CH11466.

Princeton Plasma Physics Laboratory
Report Disclaimers

Full Legal Disclaimer

Trademark Disclaimer

PPPL Report Availability

Princeton Plasma Physics Laboratory:

http://www.pppl.gov/techreports.cfm

Office of Scientific and Technical Information (OSTI):

http://www.osti.gov/bridge

Related Links:

U.S. Department of Energy

Office of Scientific and Technical Information

Fusion Links

Plasma mass filters for nuclear waste reprocessing

Abraham J. Fetterman[a,b], Nathaniel J. Fisch[a,b]

[a] *Department of Astrophysical Sciences, Princeton University, Princeton, New Jersey 08540, USA*
[b] *Princeton Plasma Physics Laboratory, Princeton, New Jersey 08543, USA*

Abstract

Practical disposal of nuclear waste requires high-throughput separation techniques. The most dangerous part of nuclear waste is the fission product, which contains the most active and mobile radioisotopes and produces most of the heat. We suggest that the fission products could be separated as a group from nuclear waste using plasma mass filters. Plasma-based processes are well suited to separating nuclear waste, because mass rather than chemical properties are used for separation. A single plasma stage can replace several stages of chemical separation, producing separate streams of bulk elements, fission products, and actinoids. The plasma mass filters may have lower cost and produce less auxiliary waste than chemical processing plants. Three rotating plasma configurations are considered that act as mass filters: the plasma centrifuge, the Ohkawa filter, and the asymmetric centrifugal trap.

1. Introduction

The recent disaster at the Fukushima Daiichi nuclear power plant has highlighted issues in the storage of spent nuclear fuel [1]. While the immediate danger to civilians seems to be contained, officials may face an even larger challenge in finding a permanent solution to the high level nuclear waste that is now strewn about the site [2]. Leaving the waste on-site for a century or more until it becomes less active seems both unwise and irresponsible, particularly after the existing safeguards offered such inadequate environmental protection. Yet there are few options for disposal of the solid wastes that are likely to be collected: a mix of concrete, graphite, activated metal, and a thousand tons of spent nuclear fuel.

The lack of attractive and practical disposal methods has apparently also delayed the treatment of high level nuclear waste at the Hanford nuclear disposal site in Washington. There are over a hundred tanks at Hanford, many of which are well past their useful lifetime. The Department of Energy is undertaking an $56 billion dollar project to remove and vitrify the waste from these containers over the next 40 years [3]. Currently, there is no geological storage facility available or under construction to store high level nuclear waste [4].

In general, nuclear waste can be divided into three groups by mass: the light elements that make up the majority of the mass, the fission product that produce the most heat and radiation, and finally the radioactive actinoids that may still be useful as nuclear fuel. The danger of the fission product has been underscored by findings of caesium-137 in significant quantities in Yamagata and Fukushima prefecture [5]. This isotope along with other highly radioactive fission products should be isolated and stored permanently as soon as possible to reduce the possibility of accidents and public exposure. This isotope only makes up 4% of the spent fuel mass, so this is much less costly than disposing of all nuclear fuel [6]. The remaining actinoids produce little heat and mostly emit alpha particles, which do not penetrate the skin.

The aim of this paper is to review and compare the opportunities for state-of-the-art plasma separation techniques. Plasma mass separation presents unique opportunities for the separation of nuclear waste by acting on the mass categories as groups rather than targeting specific elements [7]. This process would allow isolation of the fission product from nuclear waste for permanent storage. Plasma filters can do this with a smaller ecological footprint and have a simpler design than chemical separation processes. Plasma filters are not sensitive to adverse chemical interactions, and no solvents or input streams are used that will increase the total mass of nuclear waste.

Plasma mass filters will have continuing use as nuclear power plants continue to produce spent nuclear fuel [8]. In the US, nuclear fuel used once is treated as high level waste and is stored for disposal at a geological storage facility. However, more than 96% of the spent fuel mass is made of actinoids like uranium and plutonium, which are still useful as nuclear fuel. The other 4% is the fission product, which absorbs neutrons and prevents the spent nuclear fuel from being efficiently burned in a reactor. By separating these two parts, the total high level waste can be dramatically reduced. Even if the actinoids are not

reformed into fuel, they present a less hazardous radiation risk than the fission product as they emit less radiation and significantly less heat [6]. The Department of Energy allows transuranic waste (TRU) to be treated differently than high level waste, and there is an active disposal site for TRU in the US (the Waste Isolation Pilot Plant) [9]. It might be an adequate disposal option to store TRU indefinitely to use as nuclear fuel in the future.

The idea of using plasmas to separate ions based on mass is not new. In fact, the calutron has been used since the Manhattan project to produce concentrated fuel for nuclear weapons. Only recently has the possibility of separating nuclear waste with plasmas been developed, primarily by the Archimedes Technology Group [10]. Archimedes made important steps in developing technology and demonstrating separation effects before operations were halted in 2006. Good separation of heavy elements was demonstrated, although the throughput was limited by the inefficiency of the plasma source. We will show that plasma mass filters can be remarkably effective, with almost arbitrarily high purity of the output streams and large throughput comparable to chemical separation schemes.

In this paper, we will first describe the problems of Hanford nuclear waste and spent nuclear fuel. In the last part of Section 2, we will quantify the separation requirements to reduce the environmental impact of waste. In Section 3 we will discuss the advantages of plasma mass separation techniques and compare a few promising methods.

2. Nuclear waste separation

2.1. Hanford Nuclear Waste

At the beginning of the nuclear era, the Department of Energy pursued the large scale production and purification of plutonium for nuclear science, energy, and weapons. To produce the plutonium, breeding reactors were built to process millions of tons of nuclear fuel. While the plutonium and uranium were extracted from most of this fuel, 94 million gallons of radioactive fission product and irradiated materials remain mixed with hazardous processing chemicals, and are stored in hundreds of storage tanks in a few sites across the nation [4].

The largest disposal site is in Hanford, Washington, where 177 tanks contain 54 million gallons of high level waste with 194 MCi total radioactivity [11]. Solutions are urgently needed as 149 of these tanks are decades past their useful lifespan. Already 67 of these single-shell tanks are assumed or confirmed to have leaked radioactive waste into the environment. Decades remain before the final decomissioning of all single-shell tanks on site [4].

The Department of Energy expects that clean up of the Hanford site tank waste will have a total cost of $56 billion [12], but this figure does not include indirect costs that bring the total to over $86 billion [3]. The plan for disposing of this waste is referred to as the River Protection Project (RPP), named after the Colombia River that is adjacent to the Hanford site. The latest version of this plan (Revision 5, 2010) calls for the separation of tank waste into two primary streams, a high level waste (HLW) stream and a low activity waste (LAW) stream. The HLW stream and part of the LAW stream would be vitrified, with the LAW stored permanantly on site and the HLW glass shipped to a geological storage site to be determined.

The waste at Hanford is highly inhomogenous [4]. Each group of tanks has a unique origin and history of previous treatments. Each tank has settled into three chemical layers with distinct chemical properties: sludge with a peanut-butter consistency, a crystallized saltcake, and liquid supernatant. Because these layers are themselves inhomogenous, it is difficult to obtain a representative sample of the waste.

The classification of waste as HLW or LAW is not entirely intrinsic to the waste (ie, based on radionucleide concentrations), but is instead related to the personal history of the waste. The result is that separation requirements are not always universal or clear. The DOE has imposed its own standards at Hanford to keep personel radiation exposure levels from LAW handling "as low as reasonably achievable" (ALARA). In general at Hanford, supernate in a tank will be filtered for solids and ^{137}Cs and then treated as LAW, while the filtered solids, salts and sludge are treated as HLW [11].

The RPP plan does include a facility to remove aluminum and chromium from HLW solids in order to reduce the loading on the pretreatment plant, the HLW vitrification plant, and the eventual geological storage facility. The Aluminum Removal Facility (ARF) is still being tested at the bench scale, although it is expected to begin processing waste in 2022. Because of the chemical leaching and washing processes used to extract the aluminum, strontium and caesium, the tradeoff to less HLW is an increased amount of LAW production. To meet these needs, it is expected that a second facility for liquid effluent treatment will be necessary [4].

With a plasma mass filter, it might be possible to replace or support some operations of both the ARF and waste treatment plant. The plasma filter does not produce as much LAW byproduct as the ARF. In addition, removing the light elements (e.g., aluminum, iron, chromium) reduces the amount of HLW mass to be processed. Because HLW vitrification is the rate-limiting step in the RPP plan, this will lead to substantial savings in time and money [4]. As an added benefit, reducing HLW glass production will decrease the need for geological storage, an open-ended and costly disposal method.

2.2. Ongoing Waste Production

Currently, the United States uses a once-through fuel cycle: nuclear fuel is used once and then stored for disposal as HLW. A number of other countries, such as France, the United Kingdom, India, Russia and Japan reprocess fuel to recover uranium and plutonium, which can be used in

another fuel cycle. The current method for removing these elements, PUREX (plutonium uranium extraction), is a chemical extraction process using nitric acid as a solvent.

There are a number of substantial issues surrounding the PUREX process that drive a search for alternatives. The primary criticism is that reprocessed fuel is not economically competitive with directly disposing of waste and using fresh uranium fuel. For example, a Japanese reprocessing plant in Rokkasho capable of reprocessing 800 MT per year cost $20 billion to construct. This works out to a reprocessing cost of $3750/kg, almost ten times the expected cost to directly dispose of HLW [13].

A second issue related to PUREX is that by separating uranium and plutonium into different streams, there is greater risk for nuclear proliferation. Because tons of plutonium are produced every year by a reprocessing plant and plutonium is highly radioactive, controlling plutonium inventory on the kilogram scale is not practical. This poses a significant risk as only 20 kg plutonium are needed to produce a nuclear weapon. Proliferation could therefore occur if a non-nuclear country constructs a reprocessing plant, or if a small part of the plutonium stream is diverted by a hostile organization.

A final argument against PUREX is that while a large fraction of waste mass is converted to fuel, the total waste mass is not drastically reduced, and the resulting waste may be more difficult to dispose of. This is because the PUREX process requires nitric acid to be combined with the waste, and the nitric acid used for separation becomes a part of the waste stream along with corrosion products from storage and transportation vessels.

A useful way to leverage plasma mass separation technology is to reduce the volume of SNF requiring permanent geological disposal. The filter could be set to separate fission products (mass 80-160) from actinoids (mass 225-250, including uranium, plutonium, americium and other transuranics). SNF is removed from reactors because of the presence of fission products, rather than an absence of fertile fuel [6]. Therefore, the actinoids may be stored as potential fuel and the fission product can be immediately and permanently disposed of. For this to be economically productive, separation should cost less than about $200/kg [13]. This corresponds to a $2 billion budget to process 500 MT/year for 20 years, which is not unreasonable for the plasma devices described in Section III.

Alternatively, the filter could be used after the UREX (uranium extraction) process which would reduce the throughput requirements on the filter. This was suggested for the Archimedes filter [8]. Another possiblity is to use the filter after the UREX process and the removal of strontium and cesium, therefore reducing the mass and reactivity of the feed to the filter.

2.3. Separation requirements

Isotopes in nuclear waste can be divided into three groups by mass: the lightest group (1-65 amu) is bulk mass that entered the waste stream through reprocessing or leaching, the intermediate group (80-160 amu) is the highly radioactive fission product, and the heavy group (225-250 amu) is the series of actinoids, moderately radioactive and potentially fissionable. The composition of Hanford high level waste and spent nuclear fuel in terms of these categories are shown in Tables 1 and 2. In both cases, the fission product produces more than 99% of the radioactivity, despite making up a small fraction of the total mass.

	Bulk elements 1–65 amu	Fission product 80–160 amu	Actinoids 225-250 amu
Mass (kg)	1.49×10^8 98.9%	1.05×10^6 0.7%	5.82×10^5 0.4%
Radioactivity (Ci)	1.51×10^5 0.1%	1.93×10^8 99.7%	3.56×10^5 0.2%

Table 1: Inventory of Hanford high level waste divided into three mass categories.[14]

	Bulk elements 1–65 amu	Fission product 80–160 amu	Actinoids 225-250 amu
Mass (kg)	–	8.8×10^3 3.4%	3.1×10^2 96.6%
Radioactivity (Ci)	–	2.2×10^7 99.8%	4×10^4 0.2%

Table 2: Spent nuclear fuel produced per year from a 1 GW(electric) light water reactor, divided into three mass categories. Assumes a holding period of 5-10 years for ^{144}Pr decay.[6]

This demonstrates that there is an opportunity to separate the waste into high and low radioactivity streams. While this seems to have significant value, regulations regarding nuclear waste are ambiguous about the benefit from this type of separation. In this section, we will determine to what extent separation can impact disposal costs, and compare the behavior of a plasma mass filter with current chemical extraction methods.

A major problem in any attempt to separate waste is that high-level waste, as defined by the DOE in its *Radioactive Waste Management Manual* [9], is directly related to waste origin rather than waste contents,

> High-level waste is the highly radioactive waste material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and other highly radioactive material that is determined, consistent with existing law, to require permanent isolation.

The definition bears the condition that waste "contains fission products in sufficient concentrations," although the concentrations are never specified. The manual proceeds to define low level waste as HLW that has been processed

to remove key radionuclides to the extent "technically and economically practical." The result is that individual agreements and case-by-case determinations control the separation requirements at each site, with a broad upper bound on LLW radioactivity defined by NRC regulations (Table 3).

At Hanford, the waste disposal requirements are set by an agreement between the DOE, the NRC, and Washington state [4]. The major separation problem is removing ^{137}Cs from the supernate (liquid LAW) stream. On average in the Hanford waste, Caesium is responsible for 0.2 Ci/L. For waste with radioactivity levels greater than 0.05 Ci/L, the activity must be reduced to 9.0×10^{-5} Ci/L before vitrification, requiring reduction by a factor of 2,200 (removal of 99.95% from waste on average). Waste with ^{137}Cs radioactivity less than 0.05 Ci/L is exempt from reprocessing to remove that isotope [11].

The NRC grades low level nuclear waste from A to C (Table 3). Although these are lumped together in the DOE's implementation, they are useful standards to measure further separation of low-level waste. Because the Hanford waste exceeds limits on TRU α material, it is all classified as high level waste. Once separated into the three mass categories, the fission product would be the only HLW product, the actinoids could be classified as TRU waste (requiring less supervision than HLW [9]), and the light product could be level A waste.

We can use Table 3 to determine the fraction of each isotope that must be removed from the Hanford stream to purify the LLW stream to a given level. The key elements are ^{90}Sr, ^{137}Cs, and the TRU α-emitters. To maintain LLW as class C, on average 93% of the TRU material must be removed. To reach class B, 99.3% of the TRU material, 81% of ^{137}Cs and 38% of the ^{90}Sr must be removed. Finally, to produce class A LLW, 99.98% of the ^{90}Sr must be removed, along with 99.6% of the ^{137}Cs and 99.3% of the TRU material. Higher removal efficiencies than stated here will be necessary, as waste contents can vary dramatically between containers.

We will find that these separation requirements are easily met by several plasma separation schemes. In addition, plasma filters offer some immunity from the whims of regulating agencies, as radioactive isotopes can be categorically removed from the low level waste streams, and the separation factor can be varied on a batch-to-batch basis.

3. Plasma mass separation

Plasma mass separation is an ideal method to separate nuclear waste because the waste can separated into radioactive categories in a single step. Chemical methods only separate specific elements one at a time. For example, we would like to remove all fission products (rather than just caesium-137, for example) from spent nuclear fuel so these can be vitrified and stored safely and permanently. This will make the remaining waste much safer as there is less heat produced and less potential for dispersed waste to interact with the environment.

In addition to replacing several separation steps with one, a plasma filter can reduce the low level waste production compared to chemical separation. After spent nuclear fuel is processed to remove plutonium and uranium by PUREX extraction, 95% of the waste mass is nitric acid introduced by reprocessing [15]. With plasma reprocessing, no working fluid is introduced, and output streams are solids which are easier to store and vitrify than liquid wastes [14].

Economics may be the determining factor in choosing plasma separation methods. Plasma separation techniques require a much smaller footprint than chemical facilities, which reduces the cost of construction, maintainence, and operation. The magnetic field coils, often a major cost for plasma devices, are relatively simple and produce fields smaller than those in an MRI machine. By contrast, chemical separation is very expensive: the Hanford project clean up is estimated to cost over $86 billion, with each year of operation past 2047 increasing the cost by more than $1 billion [3]. Thus, shortening the timeline with a supplemental plasma filter clearly leads to significant cost savings.

There are two important factors in determining the utility of a separation method: throughput and separation factor. Throughput is the rate at which waste can be processed by a single device. The separation factor is a measure of the effectiveness of separation. Any separation problem has requirements on both factors, for example processing 1 MT of waste per day to decrease the ^{137}Cs concentration by a factor of 1,000.

We can formally define the separation factor for a group of species i as,

$$\alpha = \frac{\text{Fraction of product in group } i}{\text{Fraction of waste in group } i} \tag{1}$$

Mass filters based on a rotating plasma have a separation factor that is exponential in the rotation speed and mass difference. This means that for moderate rotation speeds (10 km/s) and large mass differences (25 amu), separation factors from one hundred to one million are possible in a single stage. Because of the high separation factor per stage, only a single stage is necessary to completely separate bulk waste from the fission product. This minimizes the overall cost and complexity of separation.

We classify 10 km/s as moderate because rotation speeds over 100 km/s have been achieved recently in a Maryland experiment, and past rotating plasma experiments have reached 2,000 km/s [16, 17]. However it is common for rotating plasmas to be limited to the Alfven critical ionization velocity (CIV), which is around 1 km/s for a plasma containing transuranic elements [18, 19, 20]. It was proposed that CIV limit may be avoided by using radio frequency waves instead of electrodes to drive rotation [21, 22]. It is crucial to the future of plasma mass filters that the CIV limit is overcome.

	^{3}H	^{14}C	^{60}Co	^{63}Ni	^{90}Sr	^{99}Tc	^{129}I	^{137}Cs	TRU α*	^{241}Pu *	^{242}Cm *
A	40	0.8	700	3.5	0.04	0.3	0.008	1	10	350	2,000
B	–	0.8	–	70	150	0.3	0.008	44	10	350	2,000
C	–	8	–	700	7000	3	0.08	4600	100	3,500	20,000
Hanford	0.05	0.015	0.04	0.63	240	0.14	2.3E-4	230	1,500	830	1

Table 3: NRC classification of low level waste in Ci/m^3. Starred columns are in nCi/g. TRU α indicates total radiation from transuranic alpha emitters (mostly ^{241}Am and ^{231}Pu) The last row indicates the average radioactivity of all high level waste at Hanford.

The throughput of a plasma separation device is directly related to device cost: it scales with the cross-sectional area of the device and the density (which is limited by the magnetic field),

$$F \approx mnA\frac{v_{th}}{4}, \tag{2}$$

$$\approx 4.5\left(\frac{n}{10^{14}\,\mathrm{cm}^{-3}}\right)\left(\frac{r}{\mathrm{m}}\right)^{2}\left(\frac{m}{10\,\mathrm{amu}}\right)^{1/2}\left(\frac{T}{10\,\mathrm{eV}}\right)^{1/2}\ \mathrm{MT/day}. \tag{3}$$

Depending on the separation scheme used and other device parameters, a 1 m radius device with a moderate density (10^{14}cm^{-3}) might process 1,500 MT of Hanford waste/year. This is comparable to the throughput at chemical separation plants.

Many of the auxiliary engineering issues for preparing, injecting, and collecting the waste into the plasma have been solved by the Archimedes Technology Group in developing the Ohkawa filter [23, 24, 10]. For injection, the waste is ground into sub-micron particles and launched into the plasma for ionization. The ions leaving the plasma device are collected on a condenser plate which is periodically heated to liquify and remove waste. The efficiency of the plasma source is an outsanding issue for development [25].

We will discuss three different rotating plasma geometries that could be used to filter nuclear waste based on mass. These are the plasma centrifuge, the Ohkawa filter, and the asymmetric centrifugal trap. We will also mention plasma technoligies that can separate single elements or isotopes, which could support another waste treatment scheme.

3.1. Plasma centrifuge

Plasma centrifuge based mass separation is similar to separation in a gas centrifuge in many ways [26]. In a plasma centrifuge, a plasma column is produced in an axial magnetic field and rotation is produced using a radial electric field. The radial electric field produces rotation through balancing of the Lorentz force ($F = q\left(\mathbf{E} + \mathbf{v}\times\mathbf{B}\right) = 0$). The centrifugal force from rotation acts on particles to cause an azimuthal drift relative to the plasma rotating frame. The centrifugal drift is proportional to the mass, and the differential drift leads to a drag that pushes heavy ions outward and light ions inward.

The resulting equilibrium is comparable to a gas centrifuge: the separation factor is proportional to $\exp\left[\left(m_2 - m_1\right)\Omega^2 r^2/2T\right]$, where Ω is the rotation frequency, r is the radius, T is the thermal energy, and m_1 and m_2 are the masses of species 1 and 2 respectively. Unlike a gas centrifuge, the total ion density profile is arbitrary because the magnetic pressure can be used to balance centrifugal pressure. Put another way, azimuthal currents in the axial magnetic field produce a centripetal force to maintain the density profile.

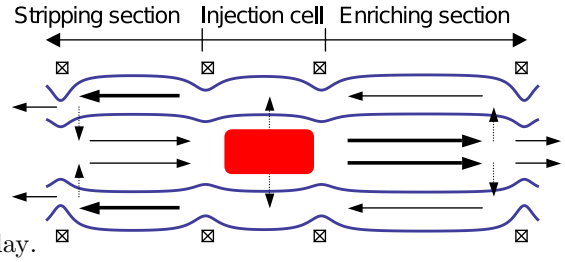

Figure 1: A countercurrent flow pattern in a plasma centrifuge [22]. Flow along field line is indicated by solid arrows. Dashed lines indicate cross-field diffusion induced by radio frequency waves.

Plasma centrifuges have been studied for isotope separation because they produce high separation factors even for small mass differences (a few amu). For example, a radial separation factor of 17 has been measured in a plasma centrifuge removing ^{90}Zr from ^{96}Zr [27]. Because the separation factor is exponential in the mass difference, we could extrapolate this result to a separation factor of 1,000 removing fission product from bulk ions (65 amu from 80 amu).

In steady state plasma centrifuges, the separation factor can be multiplied within a single device by using a countercurrent flow pattern (Fig. 1). For this flow pattern, the plasma near the axis flows in one direction along the field line, and the edge plasma flows in the other direction. The effect is that one centrifuge is divided into many units, and the feed for each unit is a mix of the product of a downstream unit and the waste of an upstream unit. This flow pattern can be produced in plasmas using radio frequency waves [22].

Another benefit of the countercurrent flow pattern is that product and waste streams are at opposite sides of the device. This reduces the radioactivity at the collection point of the low level waste stream, leading to more simple maintainence and handling operations. In a single

stage centrifuge without countercurrent flow, the product and waste streams would be separated radially, and both streams exit at each end of the mirror. However, countercurrent flow limits the number of output streams to two, while a single stage output can be divided into an arbitrary number of mass groups.

The throughput of a plasma device is directly related to the density of the plasma (Eq. (3)). The density in a plasma centrifuge is technically only limited by the magnetic field, which must produce magnetic pressure to oppose the centrifugal pressure in the plasma. We express the maximum density very approximately using the separation factor α,

$$n\frac{1}{2}\Omega^2 a^2 < B^2/2\mu_0 \tag{4}$$

$$n \lesssim 10^{16}\left(\frac{B}{\text{Tesla}}\right)^2\left(\frac{T}{10\,\text{eV}}\right)^{-1}\frac{1}{\log_{10}\alpha}\,\text{cm}^{-3} \tag{5}$$

This density limit is significantly higher than in "collisionless" plasmas, in which the density is limited by requirements on the collision frequency in the plasma. However, there are many potential issues that may lead to lower density limits than calculated by Eq.(5). Waves that can be coupled to the plasma to produce rotation have yet to be determined, and these might require a nearly collisionless plasma.

It is also not clear whether the plasma will be stable to magnetohydrodynamic (MHD) instabilities. Because of the collisions, the plasma will probably behave like a rigidly rotating body, with no sheared rotation [28]. Sheared rotation is the most common way of stabilizing a rotating plasma [29, 30]. On the other hand, the plasma may be stabilized by other effects like the large orbits of heavy particles or by escaping particles, or it may be possible to use newly proposed stabilization methods [31, 32].

A drawback of this separation technique is that it does not prevent nuclear proliferation. A country posessing this waste separation technology could to modify the components to isolate plutonium from nuclear waste, or to separate ^{235}U from ^{238}U. Either of these products provide high quality fissile material for nuclear weapons. Because the apparatus requires relatively low power and has a small footprint, it may be particularly difficult to control. On the other hand, significant scientific expertise would be required to modify the high-throughput, large mass difference filter into a separator suited for purification of ^{235}U from ^{238}U. The proliferation risk must be weighed against other separation methods such as atomic vapor laser isotope separation or PUREX.

Nonetheless, plasma centrifuges are a simple concept that can have relatively high throughput and high separation factors. They have a large experimental basis to draw from and could produce large separation factors by countercurrent flow even if the CIV limit is not overcome.

3.2. Ohkawa filter

A different kind of filter was proposed by Tihiro Ohkawa for the purpose of separating nuclear waste at Hanford [7]. A demonstration unit was constructed by Archimedes Technology Group, which developed unique methods for creating and processing plasmas from nuclear waste. However, the project never published any results demonstrating mass filter capabilities.

The Ohkawa ion mass filter is based on a radial confinement condition dependent on the charge to mass ratio. A radial electric field is applied to the plasma, causing rigid rotation about the axis. The electric field is oriented outward, so both electrostatic and centrifugal forces point outward. The rotation frequency is chosen so that above a critical mass, the outward forces exceed the inward magnetic force ($v_\theta B_z$) for all values of v and r. Critical particles satisfy $\Omega_i = -4\Omega$, where Ω_i is the ion cyclotron frequency and Ω is the plasma rotation frequency. The Ohkawa filter rotates in the opposite direction from a plasma centrifuge because the electric field direction is reversed. Heavy ions are pulled toward the outer wall rather than confined in the plasma column.

The throughput of the Ohkawa filter is limited by the collision frequency. If the collision frequency exceeds the cyclotron frequency, the motion of particles is diffusive and it is more likely that heavy particles will exit along the axis. This differentiates the Ohkawa filter from the plasma centrifuge, and implies that the Ohkawa filter will have a lower throughput. On the other hand, it is possible to achieve better separation of light and heavy elements with an Ohkawa mass filter than with a plasma centrifuge at the same rotation speed.

A major issue with the Ohkawa design is that the heavy radioactive particles must be collected across most of the plasma facing surface. A large and critical surface area is therefore made radioctive. The collection area surrounds the plasma, so any maintainence on the plasma injection apparatus, diagnostics, magnetic coils, or vacuum vessel require remote handling equipment. A potential way to mitigate this issue is to create a non-rotating buffer region around the rotating core to collect heavy particles. The heavy particles would be trapped in this region and could be removed along the axis.

Overall, the Ohkawa filter provides the potential for higher separation factors than a similar plasma centrifuge. The poor performance of the Ohkawa filter when separating particles with a mass ratio near 1 can limit the potential for nuclear proliferation. However, these benefits may be negated by the lower throughput and the broad disbursement of radioactive heavy particles, requiring significant remote maintainence.

3.3. Asymmetric centrifugal trap

The asymmetric centrifugal trap (ACT) has been proposed as an advanced fuel fusion device [33]. A diagram of such a trap is shown in Figure 2. Similar to the plasma

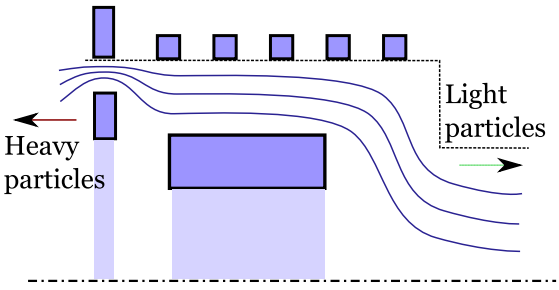

Figure 2: An asymmetric centrifugal trap. The solid lines indicate magnetic field lines, shaded squares indicate magnetic field coils, the dashed line is the vacuum boundary, and the dash-dotted line is the axis of symmetry.

centrifuge, a radial electric field is introduced that causes the plasma to rotate much faster than the ion thermal speed. The centrifugal force from rotation can be used to confine or deconfine ions, with the magnitude of the effect varying with the ion mass.

A unique feature of the asymmetric centrifugal trap is that the two sides have different confinement conditions. On one side (the right in Fig. 2), the magnetic field decreases along the axis, which would usually propel particles outward. At the same time, the field line radius is made smaller, causing ions to be pushed back to the midplane by the centrifugal force. Because heavier particles experience a stronger centrifugal force, they will be confined better than light particles. The other side (left in Fig. 2) has the reverse configuration: a larger radius and an increased magnetic field. Heavy particles will be accelerated toward this exit and a larger fraction of heavy particles will escape through this end of the filter.

This design has a number of advantages over previously mentioned separation methods. It requires a collisional plasma, so the throughput may exceed that of the Ohkawa filter. On the other hand, the throughput may be reduced as the entire vacuum vessel is not used for plasma processing.

Another avantage of the ACT filter is that the heavy and light particles exit on opposite sides of the device. This limits the activation of the device and simplifies the separate handling of the light and heavy streams. The surface area over which the ions are collected is also smaller than in the Ohkawa filter.

The ACT filter produces similar separation factors to a plasma centrifuge with no countercurrent flow. The unusual magnetic field shape allows good field line curvature at the midplane, producing natural MHD stability. In addition, the heavy and light product streams exit at opposite ends of the filter. Because the magnetic field monotonically increases along the plasma axis, there is not a simple way to add additional separation stages, which may make the ACT filter more proliferation resistant than the plasma centrifuge.

The ACT filter appears to be the most promising plasma technology for mass filter problems requiring only one separation stage (ie, separation of ions with a large mass difference or separation requiring a low separation factor). Although it does not have a significant experimental base, most of the components are simple and commonly used in plasma experiments.

3.4. Other plasma separation methods

There are several other plasma technologies used for isotope separation that are not well suited to filtering many species by mass, but may be used to remove single species.

One is ion-cyclotron resonance (ICR), in which the plasma is heated at one species' cyclotron frequency [34]. This method simply isolates ions with cyclotron frequencies near the excitation frequency–it will not effect heavy particles in a collective way. If used in combination with the Ohkawa filter, ICR waves will produce "band gaps," narrow mass ranges in which particles are not confined [7]. ICR waves might also be used with a plasma centrifuge or ACT filter to remove a multiply charged ion species, or to enhance removal efficiency of a specific species.

Likewise, AVLIS (atomic vapor lasor isotope separation) targets specific isotopes rather than entire mass ranges. In AVLIS, lasers tuned to the excitation energy of a specific isotope are passed through a partially ionized plasma. Those isotopes that are ionized by the laser are easily separated from the neutrals. This is more useful for nuclear waste after the main separation has occured, for example in removing any remaining ^{137}Cs from the light stream. Because nuclear waste contains many types of ions, it may be impractical to find resonances that are only absorbed by the target species.

4. Conclusion

We have shown that there are significant opportunities for plasma mass separation in nuclear waste disposal. The plasma mass filter could greatly simplify clean up of the Fukushima Daiishi nuclear plant in Japan, and could also save time and money in the clean up of high level nuclear waste at the Hanford facility. It is not feasible with chemical processes to remove the lighter elements that make up the majority of the waste mass. Aluminum, iron, chromium, and other metals are especially difficult. However, these elements may be removed in a single step using a plasma mass filter.

The same mass filter technology could be used to remove the highly radioactive fission product from spent nuclear fuel, which makes up less than 4% of the fuel mass. This greatly increases the safety of the remaining mass, which can be stored safely and be used as nuclear fuel. By isolating the fission product, the total amount of nuclear waste requiring permanent geological disposal is reduced significantly.

Plasma mass filters could significantly alter the treatment of nuclear waste because they filter many elements at once, require a lower cost and footprint than chemical methods, and do not increase the total waste volume.

More than that, to the extent that plasma filters make reprocessing of nuclear fuel cost effective, they could be revolutionary to the nuclear fuel cycle. By grouping elements together by mass, the plasma filter can efficiently remove all bulk elements in a single step. This greatly reduces the mass of high level waste that must be vitrified and finally stored. Because there is no need for large volumes of recirculating solvent used in chemical processing, plasma processing requires less physical space. This reduces construction and operation costs many ways. Finally, the elimination of added solvents reduces the total low level waste and effluent produced by processing.

Plasma mass filters do still require significant development to determine if they are economically viable. In particular, the ability to exceed the Alfven critical ionization velocity by using wave driven rotation should be established [21]. Plasma metal sources must be developed to keep up with the high throughput of plasma filters [25]. In addition, with respect to spent nuclear fuel, plasma filters still suffer from some shortcomings of reprocessing. For example, the separation does not diminish the total heat output produced by the nuclear waste; the reforming of waste actinoids into fuel is costly; and some potential for nuclear proliferation remains.

Plasma mass separation could facilitate processing and storage of the most dangerous nuclear waste without sacrificing undue resources. It will be especially important quickly and permanently to provide clean up solutions to the Fukushima nuclear accident. In this era of environmental consciousness, establishing a dependable and economical solution to nuclear waste could help reshape public attitudes toward nuclear power.

Acknowledgements

This work was supported by DOE contract nos. DE-FG02-06ER54851 and DE-AC02-09CH11466.

[1] G. Brumfiel, The meltdown that wasn't, Nature 471 (7339) (2011) 417. doi:doi:10.1038/471417a.

[2] G. Brumfiel, Fukushima set for epic clean-up, Nature 472 (7342) (2011) 146. doi:doi:10.1038/472146a.

[3] G. Aloise, Tech. Rep. GAO-09-913, United States Government Accountability Office (2009).

[4] P. J. Certa, M. N. Wells, River protection project system plan, Tech. Rep. ORP-11242 rev 5, Office of River Protection (2010).

[5] D. Butler, Radioactivity spreads in japan, Nature 471 (7340) (2011) 555. doi:doi:10.1038/471555a.

[6] C. on Separations Technology, N. R. C. Transmutation Systems, Nuclear Wastes: Technologies for Separations and Transmutation, National Academies Press, 1996.

[7] T. Ohkawa, R. Miller, Band gap ion mass filter, Phys Plasmas 9 (12) (2002) 5116. doi:10.1063/1.1523930.

[8] K. Czerwinski, Role of the separator in snf applications, in: Archimedes General Atomics Workshop, 2006.

[9] Radiactive waste management manual, Tech. Rep. DOE M 435.1-1, Department of Energy (1999).

[10] R. Freeman, S. Agnew, F. Anderegg, B. Cluggish, J. Gilleland, R. Isler, A. Litvak, R. Miller, R. O'Neill, T. Ohkawa, S. Pronko, S. Putvinski, L. Sevier, A. Sibley, K. Umstadter, T. Wade, D. Winslow, Archimedes plasma mass filter, AIP Conference Proceedings 694 (2003) 403–410.

[11] W. R. Wilmarth, G. J. Lumetta, M. E. Johnson, M. R. Poirier, M. C. Thompson, P. C. Suggs, N. P. Machara, Review: Waste-pretreatment technologies for remediation of legacy defense nuclear wastes, Solvent Extr Ion Exc 29 (1) (2011) 1–48. doi:10.1080/07366299.2011.539134.

[12] W. R. Swick, Challenges to achieving potential savings in doe's high-level waste cleanup program, Tech. Rep. GAO-03-593, US General Accounting Office (Jun 2003).

[13] M. Bunn, S. Fetter, J. P. Holdren, B. van der Zwaan, The economics of reprocessing vs direct disposal of spent nuclear fuel, Tech. Rep. DE-FG26-99FT4028, Project on Managing the Atom (2003).

[14] R. Alvarez, Reducing the risks of high-level radioactive wastes at hanford, Sc. & Glob. Security 13 (1) (2005) 43–86. doi:10.1080/08929880590961871.

[15] G. Choppin, J.-O. Liljenzin, J. Rydberg, Radiochemistry and Nuclear Chemistry, Butterworth-Heinemann, 2002.

[16] G. F. Abdrashitov, A. V. Beloborodov, V. I. Volosov, V. V. Kubarev, Y. S. Popov, Y. N. Yudin, Hot rotating plasma in the psp-2 experiment, Nuclear Fusion 31 (1991) 1275.

[17] R. F. Ellis, A. Case, R. Elton, J. Ghosh, H. Griem, A. B. Hassam, R. A. Lunsford, S. J. Messer, C. Teodorescu, Steady supersonically rotating plasmas in the maryland centrifugal experiment, Physics of Plasmas 12 (2005) 055704. doi:10.1063/1.1896954.

[18] J. Bergstrom, Experiments on the velocity distribution of a rotating plasma, Nuclear Instruments and Methods 133 (2) (1976) 347–353. doi:10.1016/0029-554X(76)90629-7.

[19] B. Bonnevier, Experimental evidence of element and isotope separation in a rotating plasma, Plasma Physics 13 (1971) 763. doi:10.1088/0032-1028/13/9/007.

[20] S. T. Lai, A review of critical ionization velocity, Reviews of Geophysics 39 (4) (2001) 471–506.

[21] A. J. Fetterman, N. J. Fisch, Wave-driven rotation in supersonically rotating mirrors, Fusion Sci Technol 57 (2010) 343–350.

[22] A. J. Fetterman, N. J. Fisch, Wave-driven countercurrent plasma centrifuge, Plasma Sources Science and Technology 18 (2009) 045003. doi:10.1088/0963-0252/18/4/045003.

[23] S. Putvinski, S. Agnew, T. Ohkawa, L. Sevier, Induction plasma torch liquid waste injector (2002).

[24] T. Ohkawa, Collector cup (2001).

[25] S. F. Agnew, Efficient metal ion sources for 10^13/cc density plasmas, in: Archimedes General Atomics Workshop, 2006.

[26] B. Bonnevier, Diffusion due to ion-ion collisions in a multicomponent plasma, Arkiv Fysik 33 (1967) 255.

[27] M. Krishnan, Centrifugal isotope separation in zirconium plasmas, Physics of Fluids 26 (9) (1983) 2676. doi:10.1063/1.864460.

[28] M. J. Hole, S. W. Simpson, Analytical description of a collisional plasma column, Journal of Physics D: Applied Physics 34 (2001) 3028. doi:10.1088/0022-3727/34/20/305.

[29] B. Lehnert, Rotating plasmas, Nuclear Fusion 11 (1971) 485–532.

[30] V. I. Volosov, Mhd stability of a hot rotating plasma: A brief review of psp-2 experiments, Plasma Phys. Rep. 35 (9) (2009) 719–733. doi:10.1134/S1063780X09090025.

[31] A. D. Beklemishev, P. A. Bagryansky, M. S. Chaschin, E. I. Soldatkina, Vortex confinement of plasmas in symmetric mirror traps, Fusion Science and Technology 57 (2010) 351–360.

[32] D. Ryutov, Magneto-hydrodynamically stable axisymmetric mirrors, Bulletin of the American Physical Society 55 (2010) BAPS.2010.DPP.CT3.1.

[33] V. I. Volosov, Asymmetric centrifugal magnetic confinement device, Plasma Physics Reports 23 (1997) 9.

[34] J.-M. Rax, J. Robiche, N. Fisch, Autoresonant ion cyclotron isotope separation, Phys Plasmas 14 (4) (2007) 043102. doi:10.1063/1.2717882.

The Princeton Plasma Physics Laboratory is operated
by Princeton University under contract
with the U.S. Department of Energy.

Information Services
Princeton Plasma Physics Laboratory
P.O. Box 451
Princeton, NJ 08543

Phone: 609-243-2245
Fax: 609-243-2751
e-mail: pppl_info@pppl.gov
Internet Address: http://www.pppl.gov