

THE PLASMA SEPARATION PROCESS FOR GENERIC ISOTOPE SEPARATION

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ABSTRACT

Spent nuclear fuel is a high-grade ore source for the noble metals Palladium (Pd), Rhodium (Rh) and Ruthenium (Ru). After its recovery from the spent fuel, the radioactive isotope(s) of each of these elements must be removed to reduce its specific activity. This paper will describe the Plasma Separation Process (PSP) for isotope separation and discuss its applicability for spent fuel, byproduct recovery missions.

The fissioning of nuclear materials results in a waste product "ash" unlike the ashes of any other "fires" known to man. These ashes consist of a wide variety of isotopes of many different elements which were not present in the original unfissioned material. Nuclear ashes, specifically those for the fissioning of uranium, have been shown to be a high-grade ore source of several strategic materials for which our nation's ore reserves are low.

The knowledge that these materials were present in high concentration in spent nuclear fuel has been known since the 1940's; however, a method of recovering them in an elementally pure, non-radioactive form has not been available. This unavailability has been caused by two forces. The first of these forces has recently been addressed in the Nuclear Waste Isolation Bill. While this Bill does not specifically approve the full-scale reprocessing of spent nuclear fuels, it is the first step along these lines. The reprocessing of spent nuclear fuel is important for strategic material recovery in that this is the first step in the processing of the "ore" source containing these valuable metals. The second force which has prevented the recovery and sale of these valuable materials have been their radioactivity level, even when elementally pure. The fissioning process is not selective in the atomic weight and number of its daughter nuclei. Consequently, in the formation of daughter materials, such as the noble metals, Palladium (Pd), Rhodium (Rh) and Ruthenium (Ru), isotopes of these metals are formed which are radioactive. In order to recover these metals from the nuclear waste and sell them commercially, it is necessary that the radioactive isotopes of each of these elements be removed. Here we come to understand the second force which prevents us from recovering materials from nuclear waste - at the present time there is no fully-developed process for the large-scale isotopic separation of the elements, except of the very lightest and very heaviest.

The purpose of this paper is to present a description of the Plasma Separation Process (PSP) under development at TRW; to describe how this process has been developed for the large-scale separation of the isotopes of uranium, to describe other separations that have been performed on diverse elements from the Periodic Table, to describe the generic applicability of this process for isotope separation and to provide specific insight into the use of PSP for the purification of strategic materials recovered from nuclear wastes.

The PSP process has already demonstrated its ability to separate the isotopes of a wide variety of materials. Figure 1 enumerates the elements that have been isotopically separated on a macroscopic scale and samples collected; the listing of elements which have been "scientifically" separated but for which collections were not made, range over the entire Periodic Table of chemical elements.

ΔM	ISOTOPES	RELATIVE DEPLETION FACTOR								
		1	2	3	4	5	6	7	8	9
1	Pb ²⁰⁷ FROM Pb ²⁰⁸ Pb ²⁰⁸ FROM Pb ²⁰⁷ U ²³⁴ FROM U ²³⁵		████████							
2	In ¹¹³ FROM In ¹¹⁴ Pb ²⁰⁶ FROM Pb ²⁰⁸		████████							
3	U ²³⁵ FROM U ²³⁸		████████	████████						
4	Pb ²⁰⁴ FROM Pb ²⁰⁸				████████	████████	████████	████████	████████	

DEMONSTRATED PERFORMANCE IS ADEQUATE FOR MANY GENERIC ISOTOPE SEPARATION MISSIONS

Fig. 1. General Applicability of PSP Verified by Separation of Several Species

The following is a description of the PSP technology, with specific reference to its principles of operation and description of material separations performed. The process, as depicted in Fig. 2, employs the principle of ion cyclotron resonance to selectively energize a desired isotope in a plasma containing ions of several isotopic species. The various isotopes can then be differentiated and the desired isotope preferentially collected based on the fact that it is more energetic than the other ions.

Ion cyclotron resonance is a well-characterized physical principle which has been used for many years to selectively energize ions in particle accelerators and in very high resolution mass spectrometry. In the

PSP, a plasma source generates ions of the element of interest which are confined by a magnetic field and introduced into a resonance, or excitation, region in the process chamber. The electric field signal at the desired ion cyclotron frequency is also generated and introduced into the excitation region where ion cyclotron resonance causes the desired ions to be selectively energized.

As the ions pass out of the excitation region, the selected ions possess more energy than the other ions, and therefore the cyclotron orbits of the selected ions are larger than those of the other ions. A physical separation is then accomplished making use of this difference in geometric trajectory and/or energy.

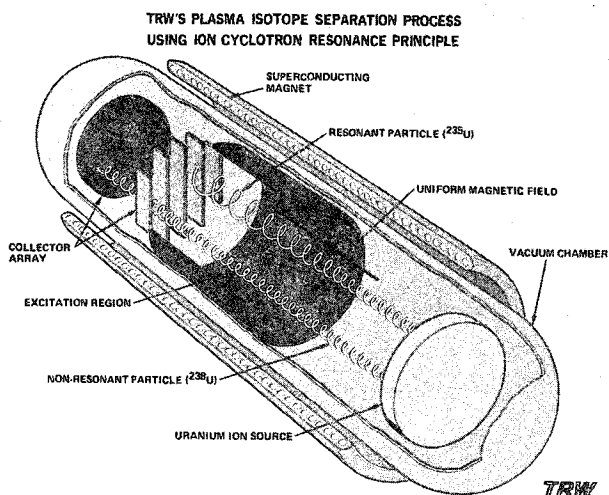
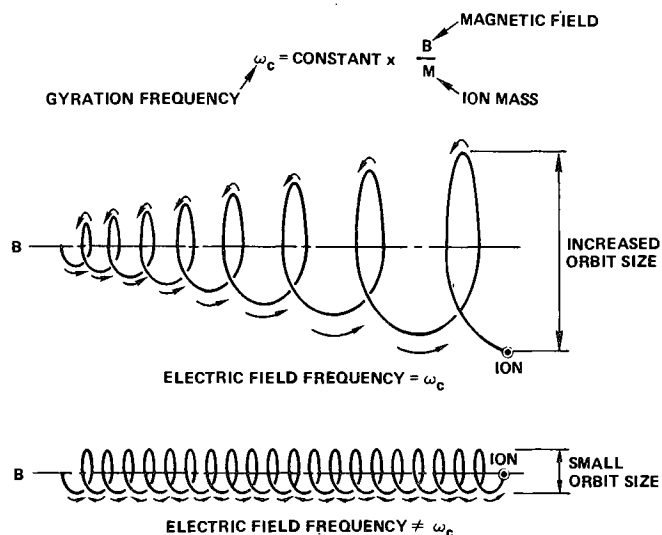


Fig. 2. TRW's Plasma Isotope Separation Process Using Ion Cyclotron Resonance Principle

The physical principle upon which the Plasma Separation Process (PSP) is based are purely classical. When ions are put into a parallel magnetic field, they are precess, or rotate, around the magnetic field lines. The rate at which a specific ion precesses about a magnetic field line is proportional only to the strength of the magnetic field and inversely to the mass and charge of the ion. (see Fig. 3). Consequently, no detailed information about the particular isotope to be separated is needed, except the mass of the ion (as in the Calutron).

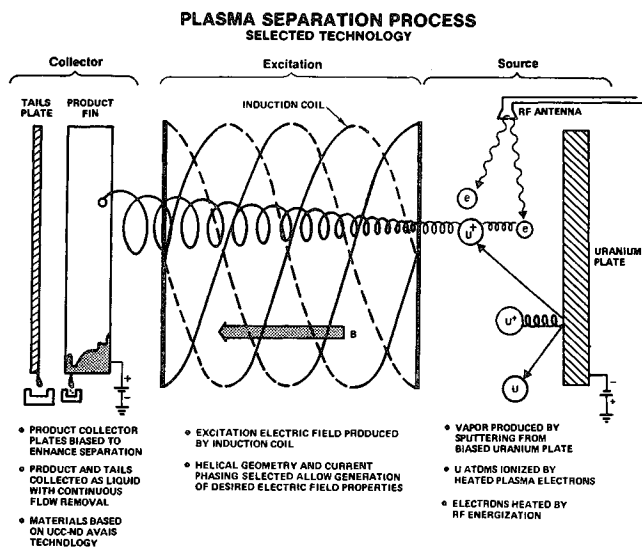
In order to use the ion cyclotron principle to separate isotopes, a PSP separation module must vaporize the feed material, then produce an electrically neutral plasma of ions and electrons. Once the plasma is formed and the isotopic ions are selectively heated, the separator uses the difference in average energy between the different ionic species to effect a physical separation.

The PSP configuration used to perform these functions for the separation of uranium ions is shown schematically in Fig. 4. The process takes place in a uniform magnetic field. At one end of a cylindrical vacuum vessel housing the process hardware is a feed plate, biased negatively at several thousand volts. The highly mobile electrons in the plasma which is produced shield the negative bias, so that it does not significantly affect particle motions in the plasma over a distance of more than a millimeter from the plate. However, particle-particle collisions cause a



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Fig. 3. PSP "Spectroscopy" is Determined Entirely by Isotope Mass and Magnetic Field



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Fig. 4. Plasma Separation Process. Selected Technology

fraction of the ions to drift toward the plate, into the thin region where the negative potential drop is felt. These ions are accelerated into the plate by the negative bias, striking the plate with sufficient energy to sputter off several atoms.

The sputtered neutrals pass through the thin plasma sheath in front of the plate, from which electrons are effectively excluded, and into the plasma where they are bombarded by electrons with sufficient energy to strike off bound electrons, producing ions. The plasma electrons are heated to ionization energies by microwaves injected into the cavity at the electron cyclotron frequency. As the ions are produced, they retain the initial velocities of the neutrals from which they are formed. Components of velocity transverse to the magnetic field are constrained by the field to follow circular paths, rotating about their

"guiding centers" at the ion cyclotron frequency. Components of velocity parallel to the magnetic field are retained, causing the ions to have an average directed velocity away from the source plate. Thermalizing collisions between the ions cause a fraction of them to have thermal velocities towards the source plate larger than the mean drift speed, providing the ion reflux necessary to maintain the neutral flux.

The interactive nature of the source requires a startup procedure for process turn on: if there is no plasma present, there are no ions to produce vapor, so so plasma can be formed. In practice this is overcome by providing an ion background gas pressure (typically argon) during start-up. An argon plasma forms, providing the ions to produce the initial sample vapor. After sample ions are formed, the argon can be valved off, and the sample plasma becomes self-sustaining. By using fast-response valves, this start-up process can be made virtually instantaneous and fully automatic.

After they are formed, the majority of the ions drift away from the source into the excitation region where isotopic ions of interest are selectively heated. Electric field at the proper frequency are produced by an induction coil surrounding the plasma. As the ions drift through the excitation coil, the energies and orbit diameters of the heated ions are increased relative to the unheated ions.

The collector takes advantage of both the energy separation and the difference in orbit diameters to effect a physical separation between the species. The collector consists of an array of plates, extending across the plasma like a venetian blind, backed by a plate which terminates the plasma column. The plates are separated by a distance smaller than the average orbit of the unexcited ions. Thus excited ions are more likely to contact the array of plates which the unexcited ions are more likely to pass between them. Material collected on the plate array is enriched in the isotope(s) of interest while material reaching the plate behind them is depleted in this isotope(s). To further increase the discrimination, a small positive bias imposed on the array rejects some of the low energy, unexcited ions that would otherwise intersect the surface due to the proximity of guiding centers.

For production separation of materials such as uranium, the collector can be operated at temperatures slightly above the uranium melting temperature, so that enriched product and depleted tailings are deposited and withdrawn continuously as liquid films. The collected liquid streams are cast into billets and subsequently withdrawn from the modules through vacuum locks.

The current development status of PSP for uranium isotope separation is shown in Fig. 5. The general applicability of the PSP process has also been demonstrated during the development of this process over the last six years. The most significant examples of this process' general applicability occurred during the summer of 1982, when the separation of the isotopes of the elements of uranium, lead and indium were all demonstrated within a timespan of one month; these separations were all performed using the same separation module. A few weeks later, again with a total span time of one month from start of development of completion of materials collections, the process was used to enrich ^{58}Ni from its natural abundance of 67.8% to over 90% in a single pass. A total of approximately 3 grams of isotopically enriched nickel was collected during a one week period near the end of this effort. This material was delivered to the user, Los Alamos National Laboratory, almost three

months before the promised delivery date, which had been selected before the nickel tests began.

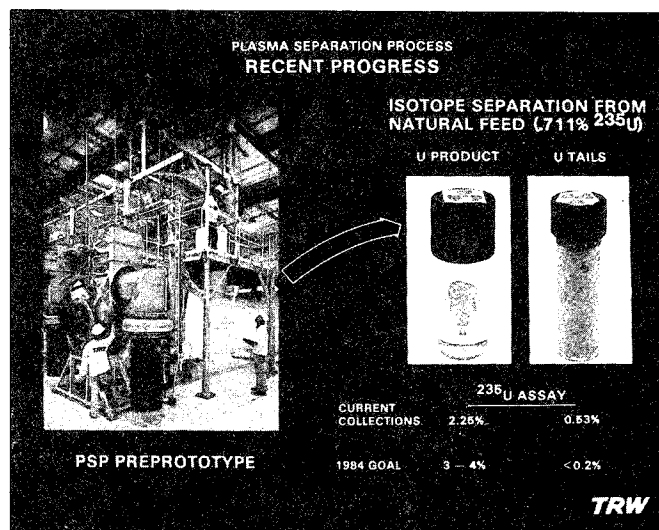


Fig. 5. Plasma Separation Process. Recent Progress

With this description of how the PSP works, it is easy to understand its generic applicability. The excitation and subsequent separation of a specific isotope is a resonance phenomena; at a fixed magnetic field, each individual isotope of a given element will become "excited," or resonate at a different frequency input by the excitation coil. Hence, in order to change the specific isotope being separated, the only change that needs to occur in the entire PSP system is the frequency of the excitation coil. Carrying this analysis one step further, changing the operation of a PSP separation facility from one element to another is almost exactly the same as changing from the separation of one isotope of a given element to another; the frequency of the isotope which is to be separated. In going from one element to another, the feed material changes and hence the exact settings of the machines parameters that were utilized for one element will need to be modified for optimized operation with a different element. The basic physics of the separation process, however, is exactly the same. Therefore, one separation facility can be utilized for the generic separation of the isotopes of many elements.

Many other methods of isotope separation exist besides the PSP process, but none of these processes appear to offer the possibility of separating the large quantities of material that would in order to make strategic material recovery from nuclear wastes appear economically attractive, while still offering the possibility of separating a range of different materials with a single facility. Molecular processes are limited by the availability of a molecular species containing an isotope of the element of interest co-ordinated with ligand ions which will not mask the identify of the isotope to be separated. Other electromagnetic separation techniques are limited in species throughput of these techniques is severely limited. The atomic vapor laser separation technique has demonstrated its utility for uranium isotope separation; however, the manner in which this process has been demonstrated to operate precludes its use for all but the very heavy and very light elements.

Let us now look more specifically at what the requirements of the isotope separation mission for the separation of the noble metals from reprocessed re-

actor waste. Inspection of the data in Table I provides us with the isotopic composition of the reactor-produced material. Recovered Rhodium and Ruthenium both contain trace quantities of radioactive isotopes, but these isotopes all have rather short half-lives (Rh-102^m T_{1/2} - 3 years, Ru-103, T_{1/2} - 39 days; Ru-106 T_{1/2} - 368 days), consequently the specific activity of each of the elementally pure elements is large. Recovered Palladium, on the other hand, contains 18 mole percent of the radioactive isotope Pd-107. The half-life of this isotope is T_{1/2} - 6 x 10⁶ years though, consequently the specific activity of this material is much less than that of either the Rhodium or Ruthenium. The specific activities of all three of these noble metals as they would be in recovered materials is also presented in Table I. The isotopic separation factor that would be required to "strip" these materials to the specific activity level below which materials of commerce can be sold on an unrestricted basis is simple to calculate technically, but exceedingly hard to determine politically. The current standard for unrestricted distribution is <2 nCi/gm - this standard covered materials for all uses. For semi-restricted uses, however, such as for use as a forming catalyst in a hydrocarbon-cracking plant it has been suggested that this standard should actually be ~10 μCi/gm. The difference between these two levels of specific activity for unlimited distribution is large and most certainly can become the deciding factor in the determination of the economic viability of these separations.

TABLE I. SPECIFICATIONS OF REACTOR-PRODUCED NOBLE METALS

	Palladium	Rhodium	Ruthenium
Radioactive Isotopes Produced	107	102 m	103, 106
Isotope Half-lives	6 x 10 ⁶ years	3 years	39.4 days, 368 days
Specific Activity (Ci/gm)	8.6 x 10 ⁻⁵	6.4 x 10 ⁻⁴	2.5 x 10 ⁻²

There are several other nuclear waste recovery missions of immediate interest for PSP separations. These missions are the isotopic enrichment of recovered Strontium (Sr) in the ⁹⁰Sr isotope and recovered Krypton (Kr) in ⁸⁵Kr. The primary interest for these enrichments is for remote lighting and RTG applications. Any operating PSP facility would be applicable for performing these enrichments; a shift in the RF excitation frequency and source optimization is all that is required. Other uses of the PSP technology for separations will also be discussed.