FORENSIC SIGNATURES FROM LASER ISOTOPE SEPARATION

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FORENSIC SIGNATURES FROM LASER ISOTOPE SEPARATION

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LIST OF SYMBOLS AND ABBREVIATIONS

AVLIS	Atomic Vapor Laser Isotope Separation
CEA	Atomic Energy Commission (French)
IAEA	International Atomic Energy Agency
LIS	Laser Isotope Separation
LLNL	Lawrence Livermore National Laboratory
MLIS	Molecular Laser Isotope Separation
NNSA	National Nuclear Security Administration
NRC	Nuclear Regulatory Commission
SILEX	Separation of Isotopes by Laser Excitation

SUMMARY

As part of its nonproliferation mission, the National Nuclear Security Administration (NNSA) notes, “Developing and maintaining the technical means to monitor whether the terms of a nuclear arms control treaty or other international agreement are fulfilled is a critical factor in ensuring that such agreements are successful.” As recognition grows that laser isotope separation (LIS) is commercially feasible, organizations such as the NNSA and IAEA are likely to require means of detecting and inspecting LIS facilities.

It has long been recognized that LIS poses a proliferation risk, since the lower energy requirements and smaller physical parameters associated with its efficiency makes an LIS facility harder to detect. It is therefore necessary to determine novel ways of detecting the existence of an illicit LIS facility and confirming whether undeclared or safeguarded material was produced in an LIS facility. This work uses a mathematical model of LIS to determine the likely isotopic ratios in uranium enriched using an LIS process and provides a comparison to the isotopic ratios in centrifuge-enriched uranium. It also uses the model to determine several key operating parameters of an LIS device from the feed, tails, and product streams. This work is combined with an isotopic chronometer, and an analysis of hypothetical samples of uranium is given to demonstrate the qualities of an LIS facility that can be determined solely from the enriched and depleted uranium it produces.

CHAPTER 1. INTRODUCTION

This chapter introduces the history of laser isotope separation and the associated proliferation concerns. It discusses the motivation and the goals of the thesis.

1.1 Motivation

The potential for the commercial development of laser isotope separation for uranium enrichment has been apparent since 1974, when researchers at Lawrence Livermore National Laboratory successfully enriched uranium using the technique [1]. The potential advantage is clear; such an efficient process could vastly decrease the cost of nuclear fuel production. However, this increased efficiency has prompted concerns by scientists and policymakers about nuclear proliferation. The lower energy and space requirements of a laser enrichment facility would theoretically make it more difficult to detect with conventional means than a centrifuge enrichment plant or a gaseous diffusion facility. [2, 3] In addition, LIS processes involve airtight vessels filled with gasses that will not remain gaseous at room temperature. [4] This suggests that an LIS device is unlikely to leak anything into the surrounding environment that might be detectable from a distance.

Therefore, laser isotope separation is both an attractive option for commercial development and a potential contributor to nuclear proliferation. For this reason, increased understanding of the systems involved should be useful in the future, both for the detection of clandestine facilities diverting enriched uranium and the inspection of any commercial facilities that may be licensed. In particular, methods for determining machine operating parameters without analysis of an entire machine design will be useful for inspectors, and

methods for determining the capabilities of an LIS device from a sample of enriched uranium may be useful for determining the existence, capabilities, and rough location of a clandestine LIS facility.

1.2 Objectives

The goal of this work is to develop a mathematical model of the laser isotope separation process that can be used to determine LIS machine parameters.

1. This model should demonstrate the expected isotopic assays of the three main isotopes of uranium (^{238}U, ^{235}U, and ^{234}U) in the enriched product.
2. It should allow back-calculation from the enriched product to estimate operating parameters of the LIS device, as well as allow for exact calculation of machine parameters with access to the tails stream.
3. It should also be possible to use the model to estimate the time of most recent enrichment.
4. It should provide a method to calculate the $^{234}U/^{238}U$ ratio in the feed to determine possible origins of the feed. This will aid facility inspectors examining any potential production facility and create the means for the detection of a clandestine laser enrichment facility.

1.3 Thesis Outline

Chapter One of this thesis reviews the history of the development of uranium enrichment methods, culminating in the development of laser isotope separation and the associated benefits and drawbacks proposed by the scientific community.

Chapter Two of this thesis contains an in-depth discussion of the basic physics of laser isotope separation, comparison of different types of LIS (notably AVLIS and MLIS), and comparison of LIS to conventional enrichment methods.

Chapter Three discusses the development of AVLIS theory by Lawrence Livermore National Laboratory (LLNL) and the French Alternative Energies and Atomic Energy Commission (CEA), comparing their approaches and highlighting important differences in nomenclature. It also contains equations derived using the preexisting theory to determine important machine operating parameters based on measurements of tail and product assays.

Chapter Four will compare the isotopic ratios resulting from enrichment by gaseous diffusion, centrifuge, and LIS, and the way these ratios are influenced by LIS design. Specifically, it gives equations for determining the feed assays of every isotope present in an enriched product (given those isotopes' product assays) and shows that the minor isotopic product assays distinguish a product laser enrichment from a product of centrifuge or diffusion enrichment processes.

Chapter Five discusses the forensic signatures from ^{234}U, including an isotopic chronometer that provides an estimate of the time since last enrichment and a comparison to the same chronometer when applied to conventionally enriched uranium. It also provides a correction to the previously stated equation for determining minor isotopic feed assays based on the decay of ^{234}U into ^{230}Th over time.

Chapter Six concludes the thesis, summarizes the results, and provides recommendations for the continuation of the work.

1.4 Literature Review

The following literature review discusses the history of enrichment methods, the development of laser enrichment technology, and various uses of laser enrichment (both currently implemented and theoretically practical), followed by a discussion of nuclear proliferation concerns related to laser enrichment technology.

1.4.1 Thermal Diffusion

In the 1940s, World War II sparked interest in the development of a bomb based on nuclear fission, which required developing methods for producing kilogram quantities of enriched uranium. Some of the first successful attempts took place in the U.S. at Oak Ridge in Tennessee and involved electromagnetic separation, thermal diffusion, and gaseous diffusion. Separate facilities existed at Oak Ridge for each of these methods: the S-50 plant for thermal diffusion, the Y-12 plant for electromagnetic separation, and the K-25 plant for gaseous diffusion. [1]

The first thermal diffusion column was constructed in Germany in 1938. Enrichment by thermal diffusion relies on the fact that a mixture subjected to a temperature gradient will develop a concentration gradient, with lighter isotopes diffusing towards the heating element and heavier elements diffusing towards the cooling element. Unfortunately, this effect is relatively small, and researchers at Oak Ridge found it unable to significantly enrich uranium, even in very large thermal columns. [1]

1.4.2 Electromagnetic Separation

Electromagnetic separation, which functions on principles like that of a mass spectrometer, was achieved in a device known as a Calutron (for California university cyclotron) by Ernest O. Lawrence at the University of California in 1942; a plant based on the Calutron was then constructed in late 1943 at Oak Ridge. The Calutrons produced the highly enriched uranium used in the "Little Boy" bomb dropped on Hiroshima; however, like thermal diffusion, electromagnetic separation was found to be expensive and inefficient at producing large quantities of enriched uranium. The difficulty of keeping the ion beams sufficiently collimated and the low throughput of the few machines built suggested that electromagnetic separation would be a poor option for mass production of enriched uranium, and like thermal diffusion, electromagnetic separation was largely abandoned as a technique to produce reactor fuel and weapons-grade uranium. [1]

1.4.3 Gaseous Diffusion

Researchers at Oak Ridge abandoned attempts at enrichment by thermal diffusion after the K-25 gaseous diffusion plant was able to produce significant quantities of low enriched uranium, and shortly thereafter abandoned electromagnetic separation in 1947 after the K-25 plant was able to produce significant quantities of high enriched uranium. [1] The successful use of gaseous diffusion to produce enriched uranium cemented its position as the global standard for uranium enrichment during the first decades of nuclear power.

1.4.4 Gas Centrifuges

U.S. government interest in using centrifuges to enrich uranium also began during World War II as part of the Manhattan Project. Tests by Westinghouse in 1943 were

ultimately unsuccessful after an oil gland, which sat between the centrifuge's driveshaft and casing, began to leak, leading the centrifuge to crash while running. Therefore, the U.S. turned to gaseous diffusion for enriched uranium instead. Since gaseous diffusion technology proved successful and economical in producing large quantities of enriched uranium, the U.S. saw no need to further develop centrifuge technology, especially given the problems posed by thermally isolating the centrifuge. [5]

However, in 1956, U.S. government officials in the Office of Naval intelligence became aware of the work of Gernot Zippe, an Austrian scientist. Soviet forces captured him during the war, during which time he and his coworkers developed an original centrifuge design that outperformed the American design. This discovery reignited U.S. government interest in centrifuges as a potential source of enriched uranium, and after release from the USSR, Zippe was employed to recreate the Soviet technology at the University of Virginia. [5]

In the 1970s, predictions by DOE researchers that demand for reactor fuel would greatly increase in the coming decades fueled desire for economically competitive centrifuge plants. As lifetime-extension programs were applied to existing gaseous diffusion plants, the cost of building new centrifuge enrichment plants remained uncompetitive in the U.S. [5] However, this marked the first decade in which gas centrifuge enrichment became competitive with gaseous diffusion on an international scale, as the increased efficiency of gas centrifuge enrichment compared to gaseous diffusion made it an attractive option for countries without existing plants. Construction began on centrifuge plants in the U.S., U.K., Germany, the Netherlands, and Japan during the 1970s, and

subsequent decades saw the increasing dominance of gas centrifuge plants in the industry of uranium enrichment. [1]

1.4.5 Laser Enrichment Methods

The concept of isotope separation based on selective photoexcitation goes back many years. As far back as 1922, researchers recognized that it might be possible to separate chlorine isotopes by irradiating atoms with light filtered through the more prevalent isotope (although this was unsuccessful). [6] However, it was not until LLNL scientist Mary Spaeth invented the tunable laser, which produces a reliable, high-intensity source of photons with a single, specified wavelength, that isotope separation in large quantities through photoionization became feasible.

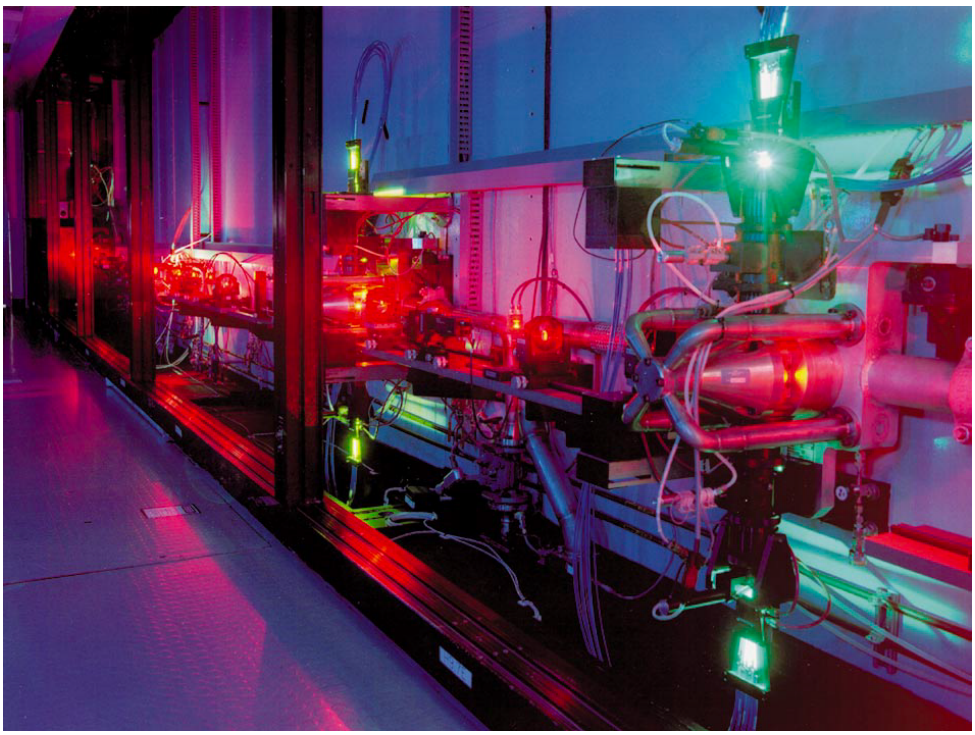


Figure 1 – AVLIS Laser Setup at LLNL [7]

The two main types of laser isotope separation are Atomic Vapor Laser Isotope Separation, or AVLIS, and Molecular Vapor Laser Isotope Separation, or MLIS. As the names suggest, one process acts on uranium in an atomic form, while the other enriches uranium in a molecular form.

1.4.5.1 AVLIS

In 1974, a successful AVLIS experiment at Lawrence Livermore National Lab delivered milligram quantities of enriched uranium and demonstrated that laser isotope separation had the potential to be a viable method of uranium enrichment to produce reactor fuel. Improvements in the design led to the U.S. Department of Energy's decision in 1982 to favor AVLIS for future reactor fuel production. [1] After a federal investment of $1.8 billion, the U.S. AVLIS program was privatized in 1998 and continued under the direction of the United States Enrichment Corporation (USEC) at LLNL. While USEC invested approximately $100 million in AVLIS, they decided to abandon the program in 1999 after projecting that bringing AVLIS to readiness for commercialization would cost additional hundreds of millions of dollars. [3] Instead, USEC pivoted to investment in SILEX (discussed below), which functionally ended the possibility of commercially available, AVLIS-produced reactor fuel at that time.

1.4.5.2 MLIS

Various MLIS methods have undergone significant development in the U.S. and Australia. The variation that has come closest to commercial deployment is Separation of Isotopes by Laser Excitation (SILEX), developed principally by the Australian company Silex Systems Ltd. and licensed to Global Laser Enrichment, "a consortium comprised of General Electric

(51 percent), Hitachi (25 percent), and Cameco (24 percent)". [8] Increasing costs, difficulty scaling the technology, and the requirement of a proliferation assessment in 2012 to proceed with commercial development appear to have halted any plans for commercial deployment at present time. [8] It is unclear whether there are any plans to reattempt commercial deployment. This is likely dependent on demand for reactor fuel, which if increased could result in renewed investment in SILEX.

1.4.6 Alternate Uses of Laser Enrichment

The selectivity and low cost of laser isotope separation when compared to other enrichment methods have generated interest in several different fields that rely on isotopically pure elements; these include medical isotope production, the aforementioned nuclear fuel production, and enriching isotopes recovered from reprocessing spent fuel. Laser enrichment has broad potential applicability, since any element with isotopes possessing photoionization peaks without significant overlap is a valid candidate for the process. For medical applications, the production of ^{99m}Tc is of particular interest, since this isotope is a valid candidate for laser enrichment, requires a high level of purity (greater than 99.99%), and is extensively used in medical tomography. [4]

In general terms, AVLIS is a good candidate for any desired isotope separation, since it can likely efficiently separate isotopes for more than half of all elements. [9] In the realm of nuclear power, as well as the obvious use of uranium enrichment, laser isotope separation has applications for nuclear and non-nuclear materials. For example, AVLIS could be used for enriching isotopes recovered from reprocessing spent fuel, since these processes could enrich plutonium or other isotopes for use as fuel. Laser isotope separation

could also be used to produce isotopically pure elements for use in reactors. For instance, ^{206}Pb produces a substantially lower concentration (by a factor of 10^3-10^4) of ^{210}Po and ^{207}Bi over long periods of irradiation than lead with naturally occurring isotopic ratios. These radionuclides emit substantial amounts of alpha and gamma radiation respectively, so it would be desirable to prevent their production. [4]

1.4.7 Nuclear Proliferation Concerns Related to Laser Enrichment

In *Nature*, Francis Slakey and Linda R. Cohen argue that the drawbacks of the proliferation risks posed by laser enrichment outweigh the benefits of cheaper nuclear reactor fuel. Specifically, they suggest that the smaller size of an LIS plant would make it difficult or impossible to detect from satellite imagery. They write, "If laser enrichment is as efficient as has been suggested, then it could occupy a space substantially smaller than a warehouse (75% smaller than centrifuge technologies) and draw no more electricity than a dozen typical houses." [2] Slakey and Cohen argue that this level of efficiency would prevent detection by existing surveillance technology, thereby posing a grave proliferation risk, as countries that currently lack nuclear weapons might use laser enrichment to covertly enrich enough uranium for a weapon before they could be detected by the IAEA and stopped. [2]

The IAEA defines a significant quantity (SQ) as "the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded." In the case of enriched uranium, the IAEA defines a significant quantity as 25 kg of HEU (> 20% U^{235}) material. [10] Given estimated throughput for a standard AVLIS facility, an SQ of enriched uranium could likely be produced within a

year. [2] While Slakey and Cohen specifically address their concerns at the prospect of a SILEX enrichment plan (then under review for licensing by the NRC to Global Laser Enrichment), these concerns are broadly applicable to other laser enrichment technologies, which boast similar efficiency. [2] It has been noted that commercial development of laser technology and electron gun heating has lowered the barrier to AVLIS specifically, though export controls and monitoring training and work experience relevant to AVLIS can help reduce the risk of proliferation. [11]

CHAPTER 2. BACKGROUND

This chapter contains an overview of commercially implemented and proposed enrichment technologies, with special attention paid to the development of laser isotope separation techniques and their benefits and drawbacks.

2.1 Conventional Enrichment Technologies

2.1.1 Principles of Thermal Diffusion

Uranium enrichment by thermal diffusion functions based on the principle by which a fluid subjected to a temperature gradient develops a concentration gradient opposing the temperature gradient. In practice, this means that in a gas containing both ^{238}U and ^{235}U, atoms of ^{235}U will increase in concentration near the heating element, while atoms of ^{238}U will decrease in concentration near the heating element. Uranium enrichment by thermal diffusion takes place in a thermal diffusion column consisting of a central pipe containing high-temperature steam and an outer pipe cooled by water, so ^{235}U in a thermal column diffuses towards the center. [1]

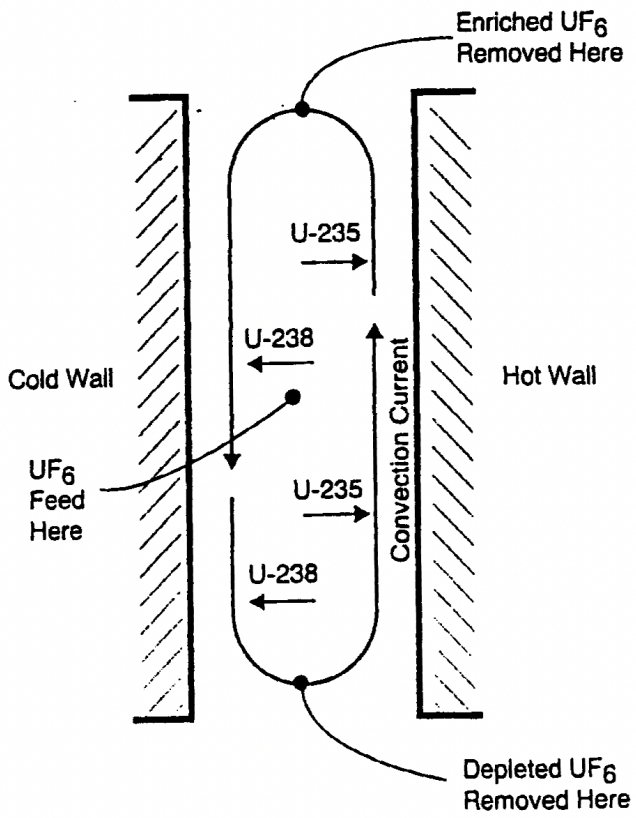


Figure 2 – Thermal Diffusion Column Diagram [1]

The light uranium hexafluoride (UF_6) stream containing more ^{235}U then moves upwards by convection and can be collected at the top of the column. Unfortunately, experimental data from thermal diffusion enrichment experiments demonstrated a much smaller single stage separation than gaseous diffusion enrichment, resulting in the abandonment of thermal diffusion as a method of uranium enrichment. [1]

2.1.2 Principles of Electromagnetic Separation

Electromagnetic separation is based on the same principles as an ordinary mass spectrometer. When an ion travels with a velocity perpendicular to a magnetic field, it experiences a force perpendicular to both the original velocity and the magnetic field, causing the ion to move in a circular path. Because the force is proportional to the velocity

but independent of the mass, lighter isotopes will travel in a circular path with a smaller radius than heavier isotopes moving with the same initial velocity through a magnetic field. [1]

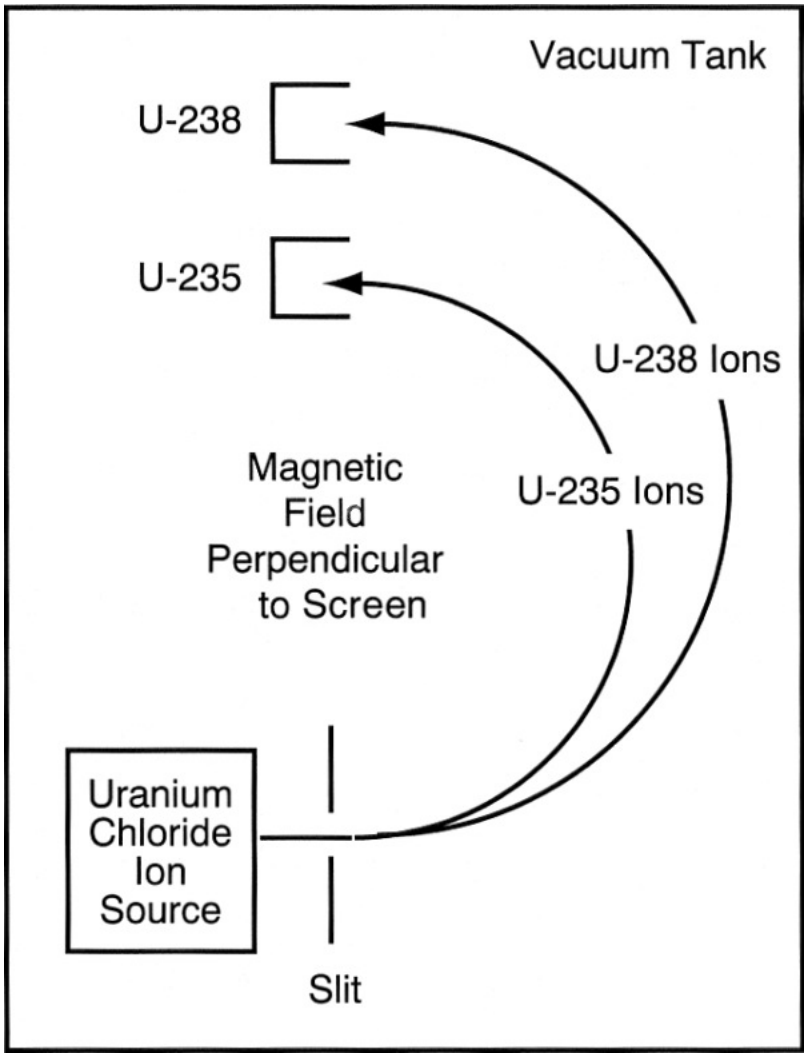


Figure 3 – Electromagnetic Separation Diagram [1]

2.1.3 Principles of Gaseous Diffusion Enrichment

Gaseous diffusion was the first uranium enrichment technology used for large-scale production of enriched uranium. It was followed by centrifuge enrichment, which is the most common enrichment technology in use today. [5] Both gaseous diffusion and

centrifuge enrichment are mass-based enrichment technologies, relying on the difference in mass between ^{235}U and ^{238}U to separate the isotopes. In a gaseous diffusion enrichment facility, this is accomplished by exploiting the fact that particles with a higher speed will diffuse across a semipermeable membrane with greater frequency than particles with a lower speed and that in a gas at equilibrium temperature, lighter isotopes have a higher average speed than heavier isotopes.

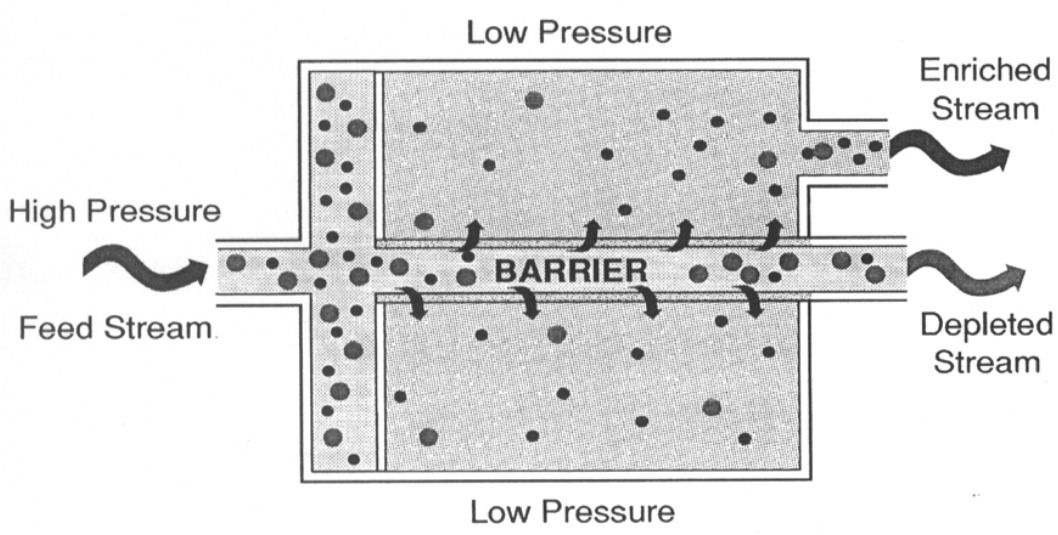


Figure 4 – Simplified Gaseous Diffusion Process [1]

As the name suggests, gaseous diffusion enrichment requires that the isotope of interest be present in a gaseous form. However, as a metal, uranium does not exist in gaseous form at anything close to room temperatures. For this reason, uranium hexafluoride (UF_6), which has a boiling point of about 57 ºC, was chosen as the process medium for the original attempts at gaseous diffusion enrichment. [12] Along with the ability to exist in gaseous form at relatively low temperatures, UF_6 is useful because it can also exist "as a liquid for filling or emptying containers or equipment, and as a solid for storage, all at temperatures and pressures commonly used in industrial processes." [13]

As previously mentioned, lighter molecules will diffuse through a membrane faster than heavier molecules. Specifically, the rate of diffusion is inversely proportional to the square root of the particle's mass.

2.1.4 Principles of Gas Centrifuge Enrichment

In centrifuge enrichment, isotopes are separated by spinning UF_6 in a centrifuge at high speeds, creating a density gradient oriented towards the outer wall of the centrifuge. This concentrates the gas at the edge of the centrifuge, leaving a near vacuum near the axis of rotation. The heavier UF_6 (containing ^{238}U) will tend to accumulate further from the axis, while the lighter UF_6 (containing ^{235}U) remains closer to the axis. Modern centrifuges also incorporate a countercurrent flow produced either by heating one end of the centrifuge or by scoops and baffles that interrupt the flow created by the centrifuge's rotation. This flow concentrates the enriched gas at one end of the centrifuge. [5] Centrifuge enrichment represents a major improvement over gaseous diffusion, since centrifuge enrichment requires substantially less energy and operates with greater separation efficiency when compared to gaseous diffusion enriching to the same product assay.

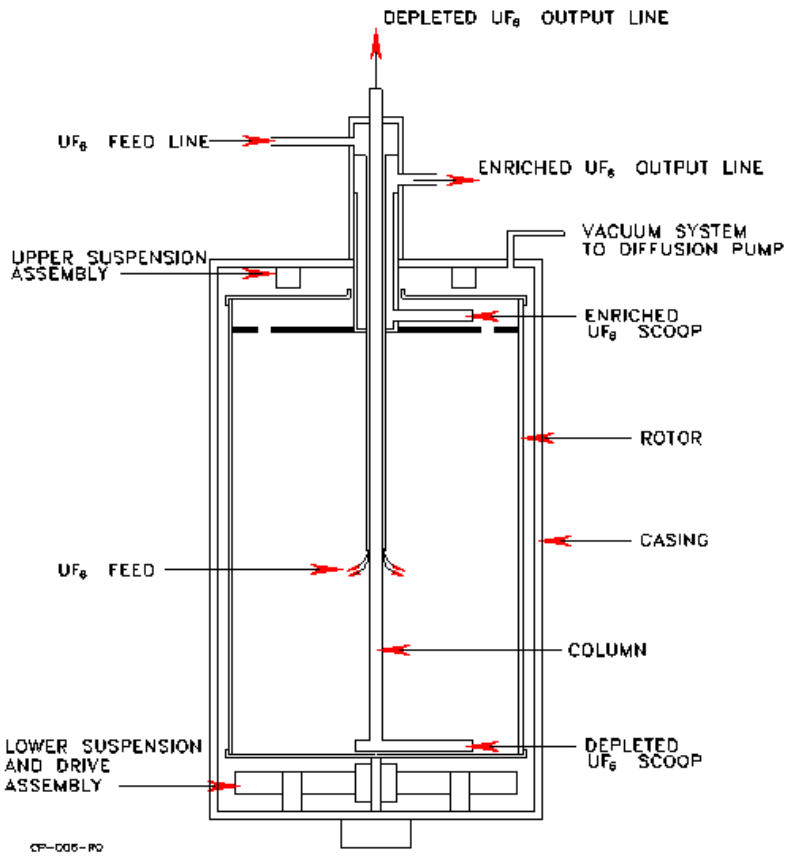


Figure 5 – Gas Centrifuge Schematic [1]

2.2 Laser Enrichment Methods

The two best-developed laser enrichment methods are molecular laser isotope separation (MLIS) and atomic vapor laser isotope separation (AVLIS). The primary differences between these two methods are the feedstock form and laser-produced isotopic-selective excitation mechanisms. MLIS enriches a molecular form of uranium (typically UF_6), which is naturally gaseous above approximately 57ºC. [12] In contrast, AVLIS enriches atomic uranium, which has a much higher boiling point and so must be vaporized by some means (typically an electron beam). MLIS takes advantage of the fact that the existing nuclear fuel supply chain is set up to provide UF_6, since all commercial enrichment

facilities currently require UF_6. If AVLIS were widely implemented for commercial purposes, it would instead require a supply of metallic uranium. These two methods of LIS function using a similar physical mechanism, namely, selective excitation through exposure to a laser. The difference comes from the choice of whether to use tuned lasers to photoionize individual uranium atoms or induce dissociation in molecules containing uranium.

2.2.1 Principles of AVLIS

The electrons of an atom each reside at different, quantized energy levels with specific values dependent on the number of protons and neutrons in the atom's nucleus.

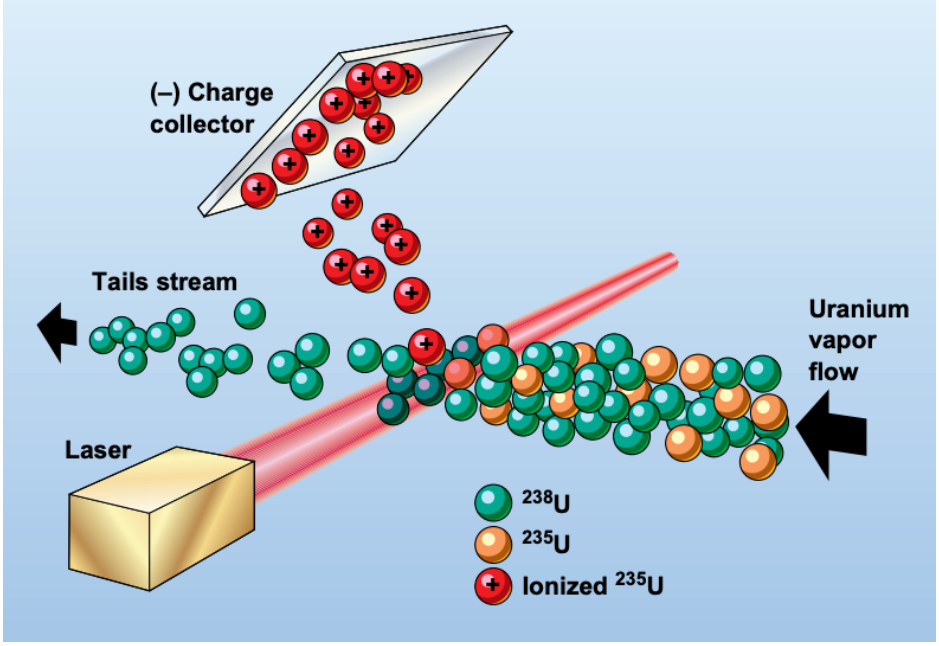


Figure 6 – Simplified Photoionization Diagram [7]

This gives the valence electrons specific resonant ionization frequencies, which are excited by photons of the same frequency. Because different isotopes have different resonant ionization frequencies for their valence electrons, it is possible to tune a laser to a

frequency that will ionize one isotope but not another. In the case of uranium enrichment, this means ionizing ^{235}U while leaving ^{238}U and ^{234}U unaffected. [4]

In the AVLIS process, a three-step photoionization process is employed in which the atoms sequentially absorb three separate photons of different wavelengths. One of the principal challenges in developing an AVLIS program capable of efficiently producing reactor fuel is the determination of the most efficient wavelengths, since "uranium possesses a branched system of atomic levels and has many valence electrons." [4] The first two photons each excite a valence electron to a higher energy state, while the third and final photon completes the process by fully liberating the electron from the atom, producing a free electron and a positively charged photo-ion. Once ionized, the ^{235}U atoms have a charge of +e (1.6 E-16 Coulombs) and can be affected by electric fields. A negatively charged collection plate can therefore be used to collect the ionized atoms, since it produces an electric field oriented toward the plate and attracts the positively charged ions. These ions collide with the collection plate, are neutralized, and condense on the plate's surface. [4] This effectively produces an enriched product stream.

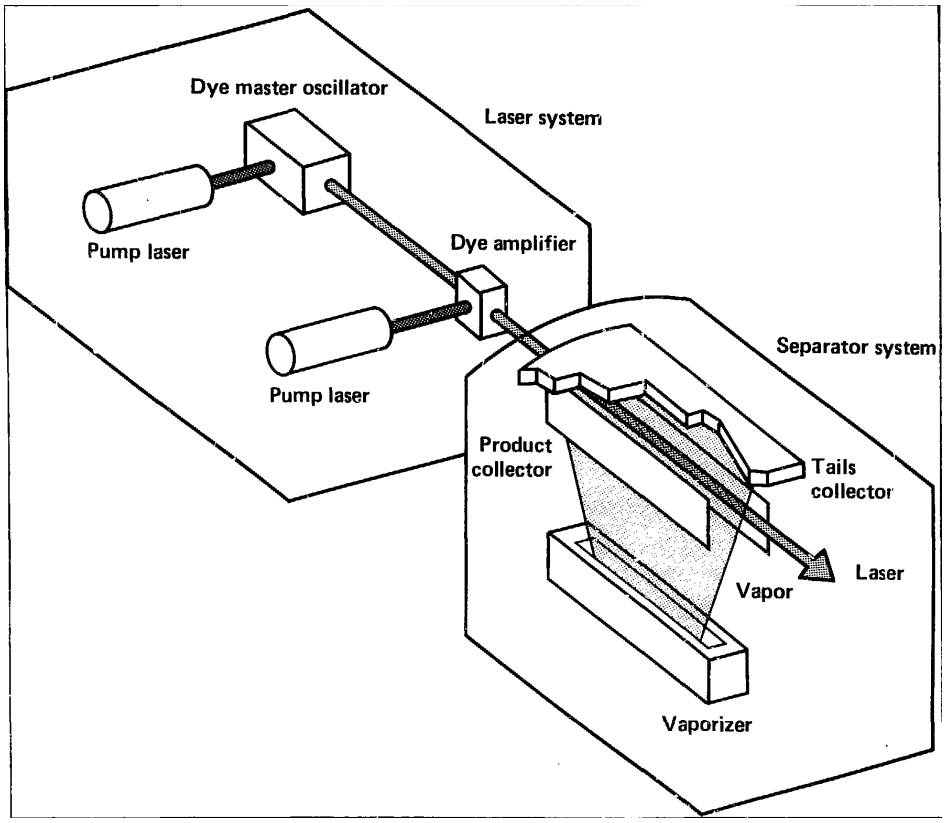


Figure 7 – Simplified AVLIS Diagram [14]

AVLIS is only a feasible method of uranium enrichment due to the development of tunable lasers with sufficiently narrow bandwidth. Typically, an AVLIS setup uses tunable dye lasers pumped by a copper vapor laser, which provide the required selectivity of tuning and the high average power and efficiency necessary to ionize large quantities of ^{235}U. [4]

2.2.2 Principles of MLIS

In contrast to AVLIS, which uses lasers to selectively ionize ^{235}U, MLIS uses lasers to induce photodissociation in $^{235}UF_6$ molecules. All molecules have certain vibrational modes determined by their mass. Specifically, lighter molecules vibrate at higher frequencies. [6] Therefore, a photon with a frequency high enough to excite a $^{235}UF_6$

molecule will have a frequency too high to excite a $^{238}UF_6$ molecule. As with AVLIS, MLIS requires multiple photons to achieve a sufficient excitation level. UF_6 molecules have an isotopically selective transition near 16 μm wavelength, in the infrared. [1]

After using infrared photons to excite a vibrational mode in $^{235}UF_6$, the molecules can then be dissociated by the addition of 3.2 eV per molecule, also delivered by photons. While this additional energy can be delivered by the same infrared photons used to excite the vibrational mode (several MLIS programs have used this technique), it does require many additional photons, since the energy of these photons is on the order of 0.01 to 0.1 eV. The U.S. MLIS program instead utilized a two-step process involving both infrared and ultraviolet frequencies: an infrared laser excites vibrations in $^{235}UF_6$ molecules, and the excited molecules are then dissociated by an ultraviolet laser into $^{235}UF_5$ and F. [6] The $^{235}UF_5$ molecules then precipitate out of the gaseous UF_6, creating a ^{235}U-rich product.

While SILEX functions similarly to standard MLIS in that it utilizes lasers to excite vibrational modes in $^{235}UF_6$ molecules, the method of separation is different. In the SILEX process, rather than causing photo-dissociation, the uranium remains in the form of UF_6 throughout the process. This is desirable since it obviates the need for refluorination of the product to continue enrichment, making it faster and cheaper to repeat passes through the device. The UF_6 streams through the chamber with a carrier gas, taking the form of an expanding supersonic free jet, and the excitation causes $^{235}UF_6$ molecules to move away from the core of the jet at a different rate from the $^{238}UF_6$ molecules. [8] This allows for separation of the isotopes. Not much information is publicly available about the specifics of this process since the technology is privately held. However, the enrichment factor of SILEX is estimated to be approximately 2, which, while higher than conventional

enrichment methods, is somewhat lower than the estimated enrichment factor for AVLIS. Discussion of the need for a SILEX enrichment cascade similarly suggests a lower enrichment factor than AVLIS. [8]

CHAPTER 3. LIS THEORY

Researchers attempting to implement laser isotope separation to confirm the fundamental physics of the process and determine whether implementation at production scale is feasible have developed systems of notation and equations that describe important parts of the LIS process. These parts include the various feed, tail, and product assays and the ways in which they change under the influence of the separative process.

3.1 LLNL and CEA: Difference in Nomenclature

3.1.1 Lawrence Livermore National Laboratory (LLNL): Nomenclature

In the United States, AVLIS was pioneered by researchers at Lawrence Livermore National Laboratory. This thesis uses the conventions and notation developed by these researchers to represent the various quantities and parameters associated with an AVLIS device, such as isotopic assays, non-selective pickup, and stripping efficiency. LLNL researchers modelled the separation process conducted by AVLIS as composed of three separate streams: the feed, tails, and product streams. This is consistent with the designation of different streams in other enrichment methods such as centrifuge enrichment. The assay, or percentage of the stream composed of ^{235}U, is designated by X. The assays of the different streams are designated by subscript as follows: X_F for the feed assay, X_T for tails assay, and X_P for product assay. [14]

LLNL nomenclature also refers to two important machine parameters: stripping efficiency, designated η, and non-selective pickup, designated ϕ. Stripping efficiency is a measure of the fraction of the target isotope (in this case ^{235}U) fed into the machine which

is successfully ionized in the photoionization zone and collected by a charged collection plate. [14] This parameter is a key measure of any LIS device's efficiency; a stripping efficiency of one means that a device is perfectly selective, and therefore more efficient than a device with a lower stripping efficiency.

Stripping efficiency is affected by several factors, including the size of the photoionization zone, the frequency of the laser pulses, the geometry of the device, and the fraction of ^{235}U atoms that reach the photoionization zone in their lowest energy state. For this reason, stripping efficiency η is sometimes decomposed into the fraction of ^{235}U atoms available in their lowest energy state to be ionized (f_a), the fraction of those atoms that actually are ionized (f_i), and the fraction of ionized ^{235}U atoms that are successfully collected (f_c), as in the following equation [14]:

$$\eta = f_a \bullet f_i \bullet f_c \tag{1}$$

Non-selective pickup is a measure of the fraction of the feed which is deposited on the product collection plate unmodified in assay. Non-selective pickup is primarily an aerodynamic effect that deposits material at feed assay on the collector without regard for the presence of lasers. [14] By varying the geometry, it is possible to modify the nonselective pickup. Generally, a low non-selective pickup is desired for a product with a higher ^{235}U assay. However, a higher non-selective pickup may be acceptable if low-enriched uranium with high throughput is the desired product.

The enrichment factor, which is the ratio of the product assay to the feed assay, is designated α. A higher enrichment factor indicates more effective extraction of ^{235}U atoms.

The depletion factor β is the ratio of the feed assay to the tails assay. A high depletion factor also indicates effective extraction of ^{235}U atoms. The separation factor, designated γ, is the product of the enrichment factor and the depletion factor [14]:

$$\gamma = \alpha\beta \tag{2}$$

3.1.2 French Alternative Energies and Atomic Energy Commission (CEA): Nomenclature

While the analysis conducted by CEA researchers is like that conducted by LLNL researchers in many respects, the nomenclature used differs in several key areas. When using CEA research as a reference on AVLIS, it is important to be aware of these differences. In some instances, the differences can cause unnecessary confusion, while in others, the same symbol may be used to refer to the inverse of the quantity associated with that symbol by LLNL researchers. This could potentially lead to serious errors in calculation.

CEA documents typically decompose the stripping efficiency into two separate components: the rate of ionization and the rate of extraction (or separation). They are designated ρi and ρs, meaning that the stripping efficiency is represented by the product [15],

$$\rho = \rho i \bullet \rho s \tag{3}$$

The non-selective pickup is referred to as the rate of pollution and is designated π. The principal point of concern lies in the CEA description of the depletion factor and the separation factor. In CEA documents, the term "depletion factor" is represented by β, as is standard in LLNL documentation. However, in CEA usage, the term "depletion factor" is used to refer to the inverse of the quantity designated as the depletion factor by LLNL documents. That is, CEA documents identify the depletion factor as follows [15]:

$$\beta = 1 - \rho i \bullet \rho s \tag{4}$$

Or, using LLNL nomenclature:

$$\beta = 1 - \eta \tag{5}$$

As a result of this difference, the separation factor needs to be redefined as

$$\gamma = \frac{\alpha}{\beta} \tag{6}$$

where β is the CEA quantity. The chief confusion arises from the use of the same symbol for the reciprocal quantity. Note that β as defined by LLNL varies from one to infinity, with a depletion factor of one indicating that no ^{235}U is taken into the product stream, and an increasing β indicating an increasing removal of ^{235}U. In contrast, β as defined by CEA varies linearly with stripping efficiency from one, which again indicates no removal of ^{235}U, to zero, which indicates perfect stripping.

Table 3 – LLNL Variables and Corresponding CEA Representations

Parameter	*LLNL Nomenclature*	*CEA Nomenclature*
Stripping Efficiency	η	$\rho i \bullet \rho s$
Non-selective pickup	ϕ	π
Enrichment factor	α	α
Depletion factor	β	$1/\beta$
Separation factor	$\gamma = \alpha\ \beta$	$\gamma = \alpha/\beta$

3.2 International LIS Development

The U.S., France, Russia, Japan, and Australia have all seen significant development of LIS programs with the goal of producing enriched uranium to be used as reactor fuel. Brazil, Iran, India, China and South Korea have also produced milligram (or greater) quantities of enriched uranium using LIS in the past but are not known to have continued any attempts to implement LIS for uranium. [1]

Japan's attempts at LIS have included research on both AVLIS and MLIS. Japan's AVLIS endeavours are largely similar to those of the U.S., while its MLIS process is unique in that it cools UF_6 below 100 K using a supersonic nozzle. The commercial success of gas centrifuge enrichment and the Fukushima disaster have both decreased the urgency for a cheaper source of reactor fuel. [1]

France has conducted AVLIS research similar in many ways to that conducted by the U.S. Known as “SILVA”, for “Séparation Isotopique par Laser de la Vapeur Atomique d'Uranium”, the French process has focused on determining the most efficient shapes of the uranium crucible and photoionization zone. According to a description of the process published by the NRC, the depleted uranium is collected on a “dome” past the photoionization zone and the product collection plates. [1] This appears to be a difference in machine geometry, as diagrams from U.S. and Russian sources appear to show flat collection plates; however, the diagrams may be conceptual in nature. [4, 14] SILVA was successful at producing 5% enriched uranium in 1990. [1]

Russia’s work on AVLIS produced promising results, including milligram quantities of HEU. [16] Work on AVLIS has continued into the 2000s for industrial and scientific applications. While recent experiments have focused on elements other than uranium, they have demonstrated high levels of enrichment, including producing milligram quantities of mercury enriched in ^{196}Hg up to 90%. [16] As seen in Figure 8, Russian AVLIS work cells utilize an electron beam to vaporize uranium in a crucible, which then streams upwards. The uranium atoms pass by ion traps—negatively charged collectors that attract any unintentionally ionized atoms—before reaching the photoionization zones and the adjacent product collectors that attract the ^{235}U ions. The remainder of the vapor streams past into a “dump collector”, also known as a tails collector. This is one of the most complete AVLIS diagrams currently available; due to the classified nature of the research, exact diagrams and dimensions are difficult to come by. However, simplified AVLIS diagrams from US publications demonstrate similar features and geometry, suggesting broadly similar construction and processes between countries (see Figure 7).

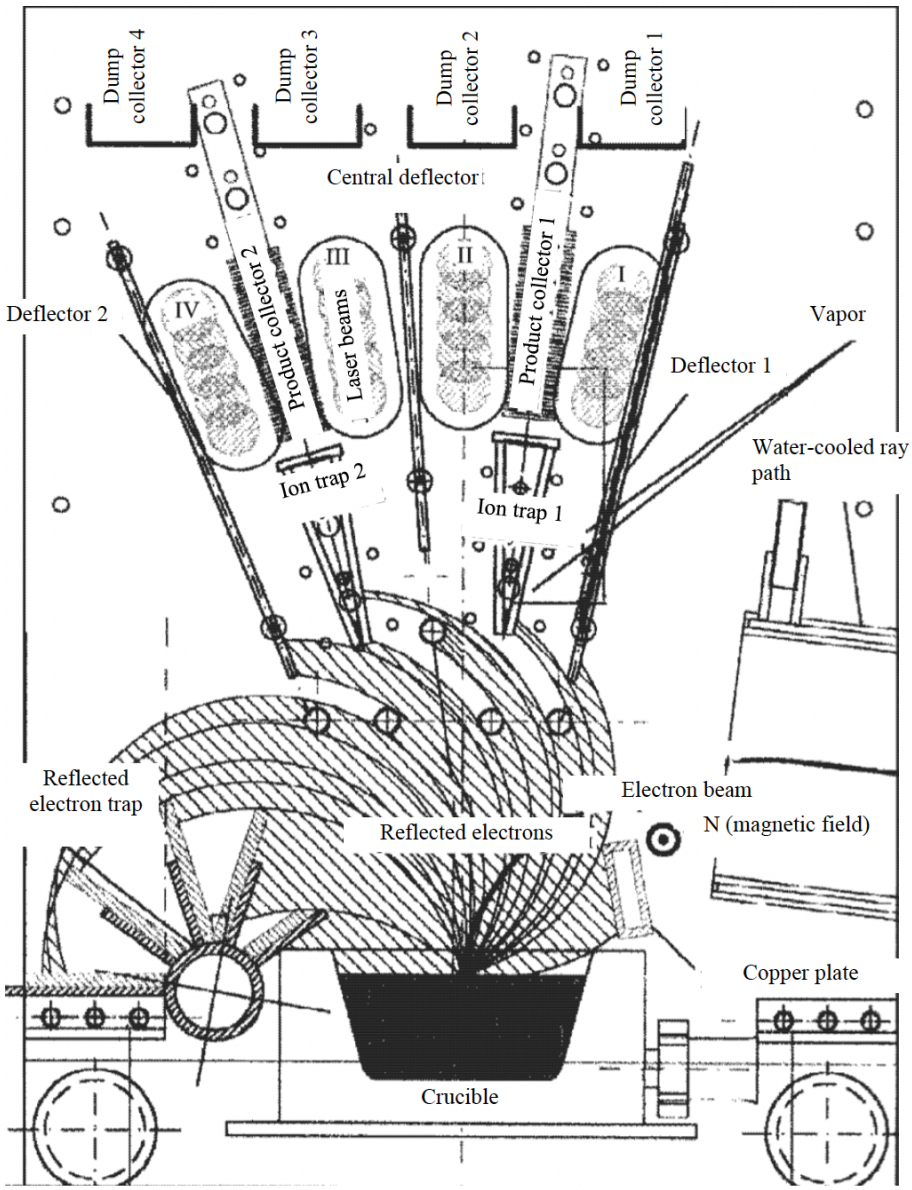


Figure 8 – Diagram of AVLIS Work Cell at the Kurchatov Institute [16]

Australia has focused on MLIS, developing a process known as SILEX (for "Separation of Isotopes by Laser Excitation"). The process is proprietary and so little is known about the specific machine parameters or any innovations that may have been made. However, it is known to be a monochromatic process, unlike the U.S. MLIS process developed at Los Alamos, which utilizes both infrared and ultraviolet photons. [1, 6]

3.3 Fundamental AVLIS Equations

This section contains several equations that describe fundamental elements of the AVLIS process, such as the feed and product masses and assays. Explanations for the structures of these equations are also given. It is important to note that these equations only apply for a good implementation of AVLIS. When the lasers are appropriately designed and tuned properly, they effectively ionize only ^{235}U atoms. However, a laser improperly given a wider frequency may also ionize ^{238}U atoms, leading to a selectivity $S \ll \infty$. In this case, a non-zero stripping efficiency for ^{238}U would need to be introduced, producing different equations.

LLNL documents on AVLIS describe the mass of the product as a result of the combination of mass from non-selective pickup and the mass of the ionized and collected ^{235}U. This gives the following equation for product mass, where P is product mass, F is feed mass, and T is tails mass [14]:

$$P = F(\phi + (1 - \phi)\eta X_F) \tag{7}$$

The tails mass is simply the feed mass F minus the product mass P:

$$T = F - F(\phi + (1 - \phi)\eta X_F) \tag{8}$$

$$T = F(1 - \phi - (1 - \phi)\eta X_F) \tag{9}$$

$$T = F(1 - \phi)(1 - \eta X_F) \tag{10}$$

The product assay is given by the total mass of ^{235}U in the product divided by the total product mass P. The total mass of ^{235}U in the product comes from the nonselective pickup plus the amount present from ion extraction, which gives [14]:

$$X_P = \frac{X_F\left(\phi + \eta(1-\phi)\right)}{\phi + \eta X_F(1-\phi)} \tag{11}$$

The tails assay is given as

$$X_T = \frac{(1-\eta)X_F}{1-\eta X_F} \tag{12}$$

The enrichment factor α is given as

$$\alpha = \frac{\eta(1-\phi)}{\phi} + 1 \tag{13}$$

The depletion factor β is given as

$$\beta = \frac{1}{1-\eta} \tag{14}$$

From these fundamental equations, several useful equations describing feed assays, product assays, and machine operating parameters may be derived.

Table 4 – Fundamental AVLIS Equations in LLNL and CEA Notation

Fundamental Equation	*LLNL Nomenclature*	*CEA Nomenclature*

Product Mass	$P = F(\phi + (1-\phi)\eta X_F)$	$P = F(\pi + (1-\pi)(\rho i)(\rho s)X_F)$
Tails Mass	$T = F(1-\phi)(1-\eta X_F)$	$T = F(1-\pi)(1-(\rho i)(\rho s)X_F)$
Product Assay	$X_P = \frac{X_F(\phi + \eta(1-\phi))}{\phi + \eta X_F(1-\phi)}$	$X_P = \frac{X_F(\pi + (\rho i)(\rho s)(1-\pi))}{\pi + (\rho i)(\rho s)X_F(1-\pi)}$
Tails Assay	$X_T = \frac{(1-\eta)X_F}{1-\eta X_F}$	$X_T = \frac{(1-(\rho i)(\rho s))X_F}{1-(\rho i)(\rho s)X_F}$
Enrichment Factor	$\alpha = \frac{\eta(1-\phi)}{\phi} + 1$	$\alpha = \frac{(\rho i)(\rho s)(1-\pi)}{\pi} + 1$
Depletion Factor (note that these are reciprocal quantities)	$\beta = \frac{1}{1-\eta}$	$\beta = 1-(\rho i)(\rho s)$

3.4 Derived AVLIS Equations for Determining η and ϕ

Starting with Equation 12, η can be isolated to determine an equation for stripping efficiency based on the observed tails assay and feed assay:

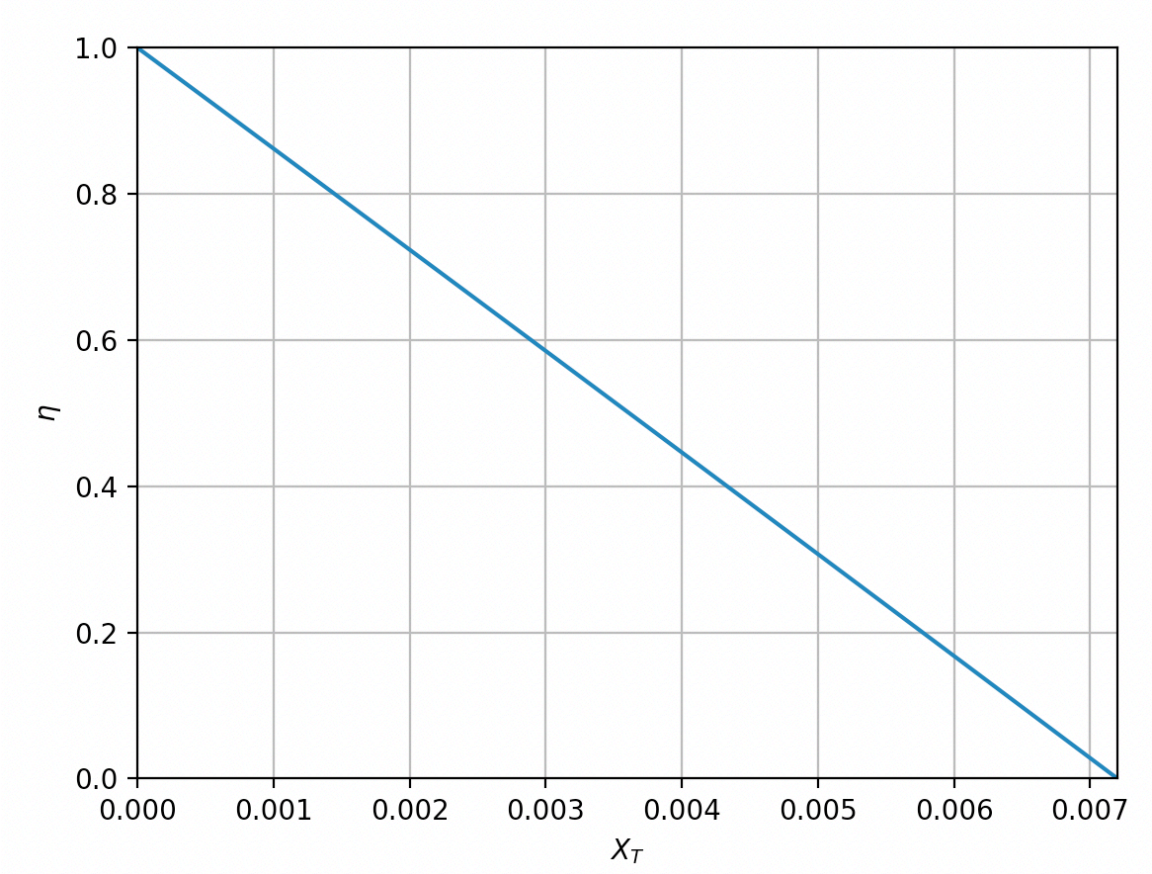
$$X_T - \eta X_F X_T = X_F - \eta X_F \tag{15}$$

$$\eta X_F - \eta X_F X_T = X_F - X_T \tag{16}$$

$$\eta = \frac{X_F - X_T}{X_F(1-X_T)} \tag{17}$$

This is a useful equation for facility inspection since it allows an inspector to determine the stripping efficiency of an AVLIS device by examining only the tails and feed assays. The dependence of η on X_T can be seen in the following figure:

Figure 9 – Stripping efficiency (η) vs. tails assay (X_T)



Similarly, Equation 11 can be used to isolate ϕ:

$$X_P\phi + \eta X_P X_F(1-\phi) = X_F\phi + \eta X_F(1-\phi) \tag{18}$$

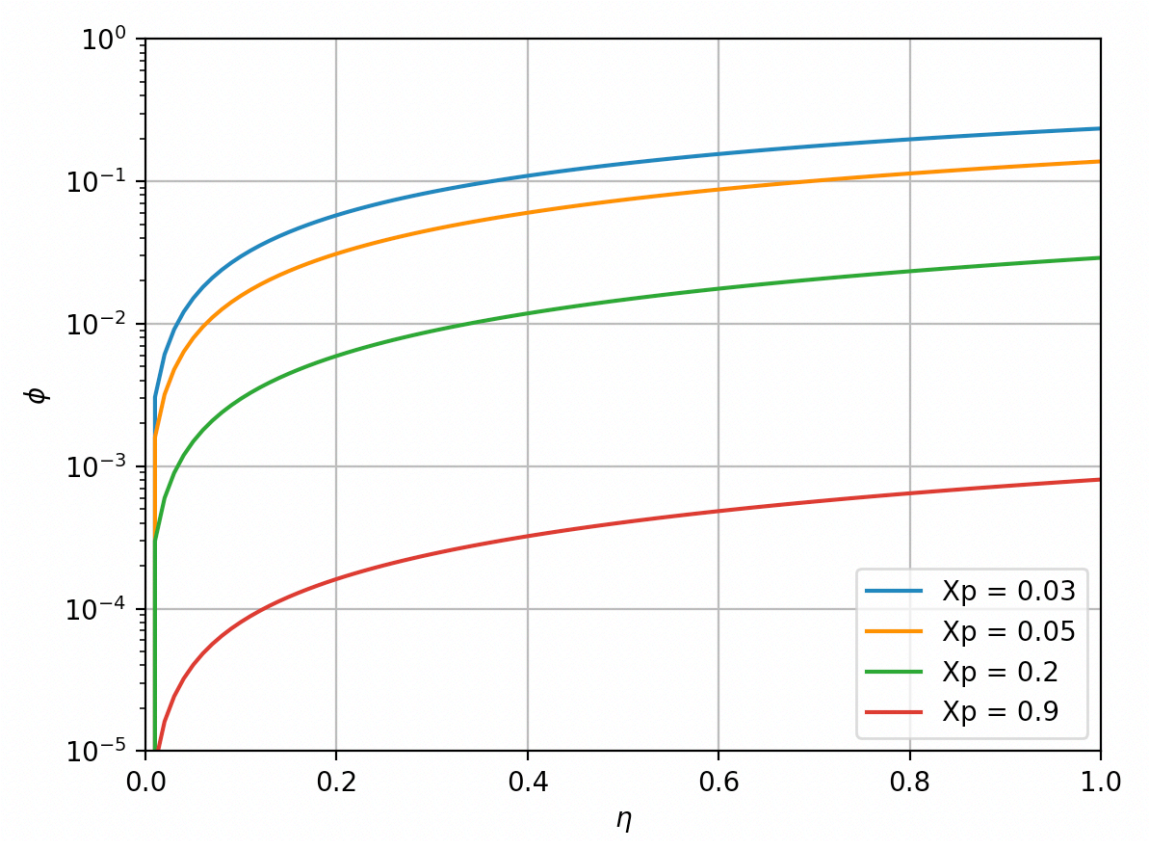
$$X_P\phi + \eta X_F X_P - \eta X_F X_P\phi = X_F\phi + \eta X_F - \eta X_F\phi \tag{19}$$

$$X_P\phi - X_F\phi + \eta X_F\phi - \eta X_F X_P\phi = \eta X_F - \eta X_F X_P \tag{20}$$

$$\phi = \frac{\eta X_F (1 - X_P)}{(X_P - X_F) + \eta X_F (1 - X_P)} \tag{21}$$

The relationship between ϕ, η, and X_P can be seen in the following figure, where a natural feed assay of 0.0072 is assumed, and the nonselective pickup is calculated for different values of the product assay:

Figure 10 – Nonselective pickup (ϕ) vs. stripping efficiency (η)



For an idealized AVLIS setup with perfect stripping efficiency (η = 1), this simplifies to

$$\phi = \frac{X_F (1 - X_P)}{X_P (1 - X_F)} \tag{22}$$

Otherwise, to obtain a more useful expression (entirely in terms of assays), Equation 17 can be substituted into Equation 21, which gives the following:

$$\phi = \frac{\frac{(X_F - X_T)(1 - X_P)}{1 - X_T}}{(X_P - X_F) + \frac{(X_F - X_T)(1 - X_P)}{1 - X_T}} \tag{23}$$

$$\phi = \frac{(X_F - X_T)(1 - X_P)}{(X_P - X_F)(1 - X_T) + (X_F - X_T)(1 - X_P)} \tag{24}$$

It should be noted that the non-selective pickup cannot be determined without access to the product, since product assay varies with non-selective pickup. However, in most cases the feed can safely be assumed to be uranium at natural assay, so reasonable estimates of stripping efficiency and non-selective pickup can be made without access to the feedstock.

If it were desirable to determine the nonselective pickup without reference to the product, it might be possible to use the following equation, derived from Equation 10:

$$\frac{T}{F} = (1 - \phi)(1 - \eta X_F) \tag{25}$$

$$\frac{T}{F}\left(\frac{1}{1 - \eta X_F}\right) = (1 - \phi) \tag{26}$$

$$\phi = 1 - \frac{T}{F}\left(\frac{1}{1 - \eta X_F}\right) \tag{27}$$

This equation can be further simplified by replacing η with the expression from Equation 17:

$$\phi = 1 - \frac{T}{F}\left(\frac{1}{1 - \frac{X_F - X_T}{1 - X_T}}\right) \tag{28}$$

$$\phi = 1 - \frac{T}{F}\left(\frac{1 - X_T}{1 - X_F}\right) \tag{29}$$

This equation has the advantage of relying solely on quantities pertaining to the feed and tails. If used in a facility inspection, this could be useful since it would not require access to the enriched uranium. In one sense, this quantity may be inconvenient to determine during an inspection of an AVLIS device, since unlike the various isotopic assays, it cannot be determined by taking a sample. Instead, the total mass of the feed and the total mass of the tails must be measured for an entire batch. However, given sustained access to the device, this equation could prove useful to determine the non-selective pickup. Assuming the feed assay is known, this equation only requires one assay measurement (tails), rather than the two assay measurements (tails and product) required by Equation 24.

CHAPTER 4. ISOTOPIC RATIOS RESULTING FROM ENRICHMENT

This chapter discusses the isotopic ratios of enriched uranium resulting from different enrichment methods and the physical qualities of enrichment devices that can be determined from these ratios. This work focuses on identifying the operating characteristics of AVLIS facilities but includes notes on generalizing the work to MLIS facilities. Uranium in its natural (unenriched) state has the following isotopic composition, given in terms of atomic percentage [17, 18]:

Table 3: Isotopic percentages in natural uranium

Isotope	*Percentage by number of atoms (at%)*
^{234}U	0.0055
^{235}U	0.7200
^{238}U	99.2745

The following work assumes enrichment takes place starting with uranium composed of isotopes at these concentrations.

4.1 Ratios from Gaseous Diffusion and Centrifuge Enrichment

Conveniently, gaseous diffusion and centrifuge enrichment are both mass-based enrichment processes. As such, the minor isotope product assays produced by these two

methods are quite similar. In particular, the ^{234}U product assay increases as the ^{235}U product assay increases. This is due to the fact that, as an isotope even lighter than ^{235}U, mass-based enrichment processes enrich ^{234}U as well as ^{235}U.

4.1.1 Gaseous Diffusion

The enrichment factors for the various minor isotopes can be determined through the equation

$$\alpha_i = \sqrt{\frac{M_k}{M_i}} \tag{30}$$

where M_i is the molecular weight of the minor isotope and M_k is the molecular weight of the most numerous isotope. [18] In the case of gaseous diffusion, this would be the molecular weight of $^{238}UF_6$, M_k = 352. Based on the derived enrichment factors, Wood includes the resulting isotope concentrations obtained with matched abundance ratio (or M* cascade modelling) theory. In the ideal cascade described by this theory, "the assay of the desired isotope in the up-flowing stream from a stage is matched to the assay of the down-flowing stream from the stage above". [18] To calculate isotopic assays using the M* program, four inputs are required: "(1) the concentrations of all the isotopes in the feed stream, (2) the concentration of ^{235}U in the both the product and tails stream, (3) either the feed rate or the product rate, and (4) the overall stage separation factor for each isotope". [18] Using stage separation factors calculated using the above equation, natural uranium assays (given in Table 3) as feed, a tails assay of 0.3% ^{235}U, and a feed rate varying from 1 to 100 mg/s, the following isotope percentages were calculated [18]:

Table 4: Isotopic percentages in LEU (5% ^{235}U) produced by gaseous diffusion enrichment

Isotope	*Percentage by number of atoms (at%)*
^{234}U	0.047791
^{235}U	5.0
^{238}U	94.952209

Table 5: Isotopic percentages in HEU (93% ^{235}U) produced by gaseous diffusion enrichment

Isotope	*Percentage by number of atoms (at%)*
^{234}U	0.934694
^{235}U	93.0
^{238}U	6.06531

4.1.2 Gas Centrifuge

The enrichment factors for the various minor isotopes can be determined through the equation

$$\alpha_i = 1 + \frac{\Delta M_i}{3}(\alpha_{235} - 1) \tag{31}$$

where ΔM_i is the difference in molecular weight between the minor isotope *i* and ^{238}U. [18] Based on these derived enrichment factors, Wood calculates the resulting minor isotope product concentrations for various ^{235}U enrichment levels. Wood does not include the product concentration of U^{238}, but it can be easily calculated by subtracting the minor isotope concentrations from one.

Wood shows that after gas centrifuge enrichment from natural uranium to a concentration of 0.05 ^{235}U, the ^{234}U concentration is $4.5739 \cdot 10^{-4}$, significantly higher than the natural concentration of approximately $5.5 \cdot 10^{-5}$. [18] Similarly, when enriched from natural uranium to a concentration of 0.93 ^{235}U, the ^{234}U concentration is $8.89859 \cdot 10^{-3}$. [18] Wood also includes an analysis of the minor isotope product concentrations in uranium enriched from reprocessed uranium, which contains a higher proportion of U^{234}. [18] Tables 6 and 7 show the resulting isotopic percentages in uranium enriched by gas centrifuge with feed at natural assay.

Table 6: Isotopic percentages in LEU (5% ^{235}U) produced by gas centrifuge enrichment

Isotope	*Percentage by number of atoms (at%)*

^{234}U	0.045739
^{235}U	5.0
^{238}U	94.954261

Table 7: Isotopic percentages in HEU (93% ^{235}U) produced by gas centrifuge enrichment

Isotope	*Percentage by number of atoms (at%)*
^{234}U	0.889859
^{235}U	93.0
^{238}U	6.110141

4.2 Ratios from Laser Isotope Separation

This work uses the LLNL conventions previously referenced in section 3.1. The assay notation (X) is slightly modified for the purposes of this work. When not specified, assay is assumed to refer to ^{235}U. However, the assays of ^{238}U and ^{234}U are also of importance to this work. For this reason, this work will sometimes specify a specific isotope; for instance, $X_{P,234U}$ refers to the assay of ^{234}U in the product stream, in contrast to the standard X_P, which refers to the product assay of ^{235}U.

It should be noted that the product assay of a minor (non-enriched) isotope is equivalent to the total mass of the minor isotope in the product divided by the total mass of the product. For a well-designed and operated AVLIS system with high stripping efficiency, almost all minor isotopes present in the product are due to the non-selective pickup, and the total mass of a minor isotope (^{234}U in this instance) in the product is equivalent to:

$$M_{P,234U} = F\phi X_{F,234U} \tag{32}$$

Relying on equation 7, which gives the total product mass, an equation for the product assay of ^{234}U can be derived:

$$X_{P,234U} = \frac{M_{P,234U}}{P} = \frac{\phi X_{F,234U}}{\phi + \eta X_F(1-\phi)} \tag{33}$$

While this equation is easily utilized if the non-selective pickup and stripping efficiency are already known, it cannot be effectively utilized if inspectors only have access to a product sample. Therefore, it is worthwhile to rewrite the equation in terms of isotopic assays. Substituting Equation 21 into Equation 33 gives the following:

$$X_{P,234U} = \frac{\dfrac{\eta X_F X_{F,234U}(1-X_P)}{(X_P - X_F) + \eta X_F(1-X_P)}}{\eta X_F + \dfrac{\eta X_F(1-X_P)(1-\eta X_F)}{(X_P - X_F) + \eta X_F(1-X_P)}} \tag{34}$$

$$= \frac{\eta X_F X_{F,234U}(1-X_P)}{\eta X_F(X_P - X_F) + \eta^2 X_F^2(1-X_P) + \eta X_F(1-X_P)(1-\eta X_F)} \tag{35}$$

$$= \frac{\eta X_F X_{F,234U}(1 - X_P)}{\eta X_F X_P - \eta X_F^2 + \eta^2 X_F^2 - \eta^2 X_F^2 X_P + \eta X_F - \eta^2 X_F^2 - \eta X_F X_P + \eta^2 X_F^2 X_P} \quad (36)$$

$$= \frac{\eta X_F X_{F,234U}(1 - X_P)}{\eta X_F - \eta X_F^2} \quad (37)$$

$$X_{P,234U} = \frac{1 - X_P}{1 - X_F} X_{F,234U} \quad (38)$$

As can be seen in simplified Equation 38, the product assay of ^{234}U can be written entirely as a function of the product and feed assays for ^{235}U and ^{234}U. Importantly, the product assay of minor isotopes can be estimated without first determining the stripping efficiency, nonselective pickup, or tails assay in the AVLIS process. As a result, the assay of ^{234}U in the feed stream can be estimated through spectrometry of the enriched product (assuming a feed of natural uranium). It should be noted that this equation can also be applied to any other isotope which is not the isotope targeted for enrichment; the product and feed assays of ^{234}U simply need to be replaced by the product and feed assays of a different minor isotope.

Table 8: Isotopic percentages in LEU (5% ^{235}U) produced by AVLIS

Isotope	*Percentage by number of atoms (at%)*
^{234}U	0.005263
^{235}U	5.0

^{238}U	94.994737

Table 9: Isotopic percentages in HEU (93% ^{235}U) produced by AVLIS

Isotope	*Percentage by number of atoms (at%)*
^{234}U	0.000388
^{235}U	93.0
^{238}U	6.999612

As can be seen from the product assays given in the above tables, it is straightforward to distinguish between the product of a well-designed and operated AVLIS system with high stripping efficiency and the product of gas centrifuge enrichment or gaseous diffusion enrichment. Results from an AVLIS system with poor isotopic selectivity could be much less clear.

Also of interest is the ^{234}U/^{238}U ratio, which is sometimes measured instead of the individual assays. This ratio is surprisingly easy to calculate for AVLIS-enriched uranium. As suggested by Equation 32, the mass of ^{238}U present in the product will be almost entirely due to the non-selective pickup:

$$M_{P,238U} = F\phi X_{F,238U} \tag{39}$$

Therefore, the mass ratio of $^{234}U/^{238}U$ is easily given by

$$\frac{M_{P,234U}}{M_{P,238U}} = \frac{X_{F,234U}}{X_{F,238U}} \tag{40}$$

In other words, the $^{234}U/^{238}U$ ratio is preserved from the feed to the product. This is in sharp contrast to mass-based enrichment processes, which noticeably increase the $^{234}U/^{238}U$ ratio. This provides a potentially straightforward means of differentiating between uranium enriched by AVLIS and uranium enriched by gaseous diffusion or centrifuge. Any enriched uranium sample in which the $^{234}U/^{238}U$ ratio remains the same as the naturally occurring $^{234}U/^{238}U$ ratio, while seeing a decrease in both ^{234}U and ^{238}U assays, must come from a non-mass-based process such as laser enrichment; any enriched uranium sample in which the $^{234}U/^{238}U$ ratio increases (due to increased ^{234}U assay and decreased ^{238}U assay) must come from a mass-based enrichment process. In the event that undeclared, enriched uranium is intercepted, inspectors will likely be able to determine whether they should concentrate their attention on gaseous diffusion plants, gas centrifuge plants, or as yet undiscovered laser enrichment facilities.

CHAPTER 5. FORENSIC SIGNATURES FROM ^{234}U

This chapter discusses the specific forensic signatures produced by ^{234}U, with special attention given to a method of determining the feedstock source of an AVLIS device and an isotopic chronometer to determine the time of last enrichment.

5.1 Determining the Feedstock Origin

It is well-documented that, due to a variety of geological phenomena, the ratio of ^{234}U to ^{238}U varies slightly depending on geographic location "due to natural isotope fractionation, nuclear reactions or anthropogenic contamination". [17] This suggests that the list of possible origins of a sample of uranium can be narrowed down, potentially to a single mine, given sufficiently accurate spectroscopy and a reference table of ^{234}U/^{238}U ratios measured at different geographic locations.

As previously mentioned, the ^{234}U and ^{238}U product assays are decreased by laser isotope separation. However, the ^{234}U/^{238}U ratio in the enriched product remains the same as that of the original feedstock since the minor isotopes are depleted at the same rate. In order to effectively utilize the ^{234}U/^{238}U ratio as a forensic signature determining the place of origin of the feedstock, this ratio must be large enough to detect, and the sample of uranium must be representative of the enriched product, i.e., the enriched uranium produced by an AVLIS device must feature a homogeneous distribution of isotopes. Unfortunately, no experimental data could be found confirming the isotopic homogeneity of AVLIS-enriched uranium. In the absence of such data, this work will use the assumption of homogeneity.

Assuming homogeneity, the main concern would be that the ^{234}U product assay would be too low to perform an accurate measurement. Considering the case of weapons-grade uranium (enriched to 93% ^{235}U) enriched from uranium at natural assay, the total ^{234}U product assay would be given by the following:

$$X_{P,234U} = \frac{1 - 0.93}{1 - 0.00711} X_{F,234U} \tag{41}$$

$$= 0.070501\, X_{F,234U} \tag{42}$$

So for weapons-grade uranium, the highest enrichment one would expect to see from an AVLIS device, the product assay of ^{234}U is reduced by approximately a factor of ten. The typical assay of ^{234}U at natural feed is about $5.5 \cdot 10^{-5}$, so the reduced product assay would be about $3.9 \cdot 10^{-6}$. Luckily, modern mass spectrometry techniques still allow for accurate measurement at this level. Richter *et al.* report measuring the $^{236}U/^{238}U$ ratio in uranium at natural assay, at which point ^{236}U has an assay on the order of 10^{-8}–10^{-10}. [17] More recent improvements in thermal ionization mass spectrometry (TIMS) show even more promise, exhibiting measurements of minor uranium isotope ratios on the order of 10^{-9} with an error of only 0.1–0.5%. [19] Therefore, a measurement of ^{234}U in enriched uranium, with an assay on the order of 10^{-6}, should not pose a significant problem.

Richter *et al.* give the ratios of ^{234}U to ^{238}U for six different uranium ore samples, all taken from different locations. [17] These ratios vary on the order of 10^{-5} to 10^{-7}, and the largest uncertainty in measurement is of the order $5 \cdot 10^{-9}$. The table below shows the product assay of ^{234}U in hypothetical samples of uranium enriched by AVLIS to 93% from the different samples of natural uranium analyzed by Richter *et al*. [17]

Table 10 – Derived ^{234}U Assays for Different Feed Assays

	$^{234}U/^{238}U$ Feed [17]	*Derived ^{234}U Feed Assay*	*Derived ^{234}U Product Assay*
Sample 1	$5.434 \cdot 10^{-5}$	$5.395 \cdot 10^{-5}$	$3.804 \cdot 10^{-6}$
Sample 2	$8.355 \cdot 10^{-5}$	$8.294 \cdot 10^{-5}$	$5.848 \cdot 10^{-6}$
Sample 3	$5.444 \cdot 10^{-5}$	$5.404 \cdot 10^{-5}$	$3.811 \cdot 10^{-6}$
Sample 4	$5.460 \cdot 10^{-5}$	$5.420 \cdot 10^{-5}$	$3.822 \cdot 10^{-6}$
Sample 5	$5.154 \cdot 10^{-5}$	$5.117 \cdot 10^{-5}$	$3.608 \cdot 10^{-6}$
Sample 6	$5.340 \cdot 10^{-5}$	$5.301 \cdot 10^{-5}$	$3.738 \cdot 10^{-6}$

5.2 Determining Time of Last Enrichment

As with standard enriched uranium, the most useful chronometer will be a $^{230}Th/^{234}U$ chronometer. This is due to the reasonably short half-life of ^{234}U and the low incidence of ^{230}Th in the feedstock (given standard purification of uranium ore). In a study of reactor fuel production, purification by ion exchange and solvent extraction was shown to produce a low level of residual thorium which resulted in about 0.5% positive bias in age determination. [20] There should be an even lower level of ^{230}Th in the product stream of the AVLIS process, since any that is present in the feedstock will be further depleted by the LIS process. It is therefore justified to assume that using the $^{230}Th/^{234}U$ chronometer

will allow a fairly accurate estimation of the time since last enrichment with a low positive bias. If the uranium feed is enriched significantly later than the time of purification, there will of course be some ^{230}Th ingrowth, which, while reduced by the enrichment process, will contribute a small positive bias to the estimate of age.

The equation used to determine time since last enrichment is

$$T = \frac{1}{\lambda_{234U} - \lambda_{230Th}} \bullet \ln\left(1 - \left(\frac{N_{230Th}}{N_{234U}}\right)\left(\frac{\lambda_{230Th} - \lambda_{234U}}{\lambda_{234U}}\right)\right) \tag{43}$$

where T is the time since last enrichment, N_a is the number of atoms of species *a* in the sample, and λ_a is the activity constant for species *a*. [21] The following figure demonstrates time dependence as a function of the nuclide ratio:

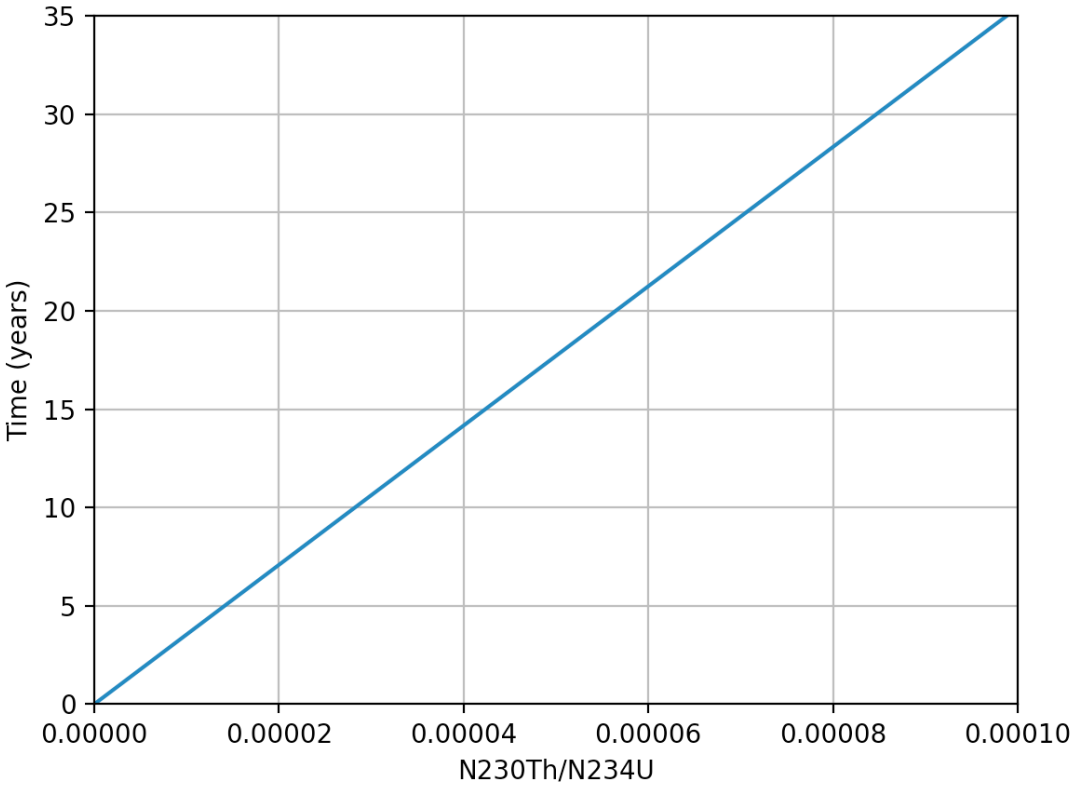


Figure 11 – Time Since Last Enrichment Based on $^{230}Th/^{234}U$ chronometer

The use of this chronometer also suggests a way to make previous equations more accurate. The $^{234}U/^{238}U$ ratio in a sample of enriched uranium is still the primary quantity of concern when analyzing an enrichment center product on-site. However, when analyzing a product that was enriched some time ago, it would be better to consider the ratio ($^{234}U + {}^{230}Th)/^{238}U$. This is due to the fact that any ^{230}Th present in the product can be assumed to be the result of the decay of ^{234}U over time, provided that the initial feedstock undergoes appropriate chemical purification. Therefore, a more accurate initial $^{234}U/^{238}U$ ratio can be calculated by assuming that the ^{230}Th present in a sample of enriched uranium was originally ^{234}U in the product directly after enrichment. This results in a new set of corrected equations. Rather than considering solely the ^{234}U product assay, the ^{230}Th product assay plus the ^{234}U product assay should be used:

$$X_{P,234U} + X_{P,230Th} = \frac{1 - X_P}{1 - X_F} X_{F,234U} \tag{44}$$

Similarly, rather than using the $^{234}U/^{238}U$ ratio to determine the original $^{234}U/^{238}U$ feed ratio, the sum of the ^{234}U and ^{230}Th assays should be used:

$$\frac{X_{P,234U} + X_{P,230Th}}{X_{P,238U}} = \frac{X_{F,234U}}{X_{F,238U}} \tag{45}$$

This will result in a further reduction in error when attempting to determine qualities of the original feedstock.

CHAPTER 6. CONCLUSION

This section provides a summary of the work conducted and recommendations for further research.

6.1 Conclusions

A system-level mathematical analysis of AVLIS has been conducted, and the resulting applications to nuclear forensics are given. This work demonstrates that the stripping efficiency of an AVLIS device can be determined solely by analyzing the tails assay and assuming a feed of uranium at natural assay. Based on this stripping efficiency, this work shows that the non-selective pickup can also be determined, although this requires access to the product.

This work also demonstrates that the $^{234}U/^{238}U$ ratio expected to result from a laser enrichment process differs substantially from the $^{234}U/^{238}U$ ratio observed in the products of gaseous diffusion enrichment and centrifuge enrichment. In addition, the work demonstrates that a $^{230}Th/^{234}U$ isotopic chronometer can be used to determine the approximate time of the most recent enrichment by an AVLIS device, although the uncertainty is greater than that observed when this technique is applied to uranium enriched by a mass-based process.

6.2 Recommendations

This work assumes that the samples analyzed by inspectors, namely product and tails, have not been altered in an attempt to frustrate inspection. This may be a reasonable

assumption for a sample of enriched uranium intercepted in transit from an as yet unknown AVLIS facility; the operators of such a facility may reasonably expect to remain undetected and therefore fail to take further steps to complicate analysis. However, if multiple countries begin to operate commercial laser enrichment facilities, it is possible that one of them might decide to use the existence of a civilian enrichment program as cover to divert nuclear material to a covert weapons program. In this case, they could expect IAEA inspectors to have access to the facility, and may take steps to alter the feed, tails, or product in order to frustrate analysis. For instance, the product or tails could be adulterated with uranium of a different assay or purity, or otherwise tampered with in order to throw off the isotopic ratios or isotopic chronometer. For this reason, it is recommended that future work in this area focus on methods of detecting such tampering and accounting for it when applying the methods of analysis detailed in this work.

Future research should also focus on modelling the expected isotopic ratios in the tails of LIS devices, since this may reveal information similar to that revealed by an analysis of the product and may be easier to access (in an inspection) or discover (when searching for a clandestine facility) than the enriched product. It should also be noted that this work rests entirely on a mathematical model that, while developed and tested by researchers with access to a functional AVLIS device, has not been tested specifically with regards to various isotopic assays in the product and tails. The use of the $^{230}Th/^{234}U$ chronometer on AVLIS-enriched uranium also has not been experimentally verified. If possible, it would be worthwhile to obtain a sample of AVLIS-enriched uranium and AVLIS-depleted tails and, using mass spectrometry methods, verify that the isotopic ratios

are as expected and that the $^{230}Th/^{234}U$ chronometer accurately gives the latest time of enrichment.

As a final measure, it may be useful to complete a similar analysis of LIS processes that feature relatively low selectivity, such as some MLIS processes. Such an analysis was not undertaken in this work for two reasons. First, Livermore researchers assert that AVLIS, properly implemented, features selectivity so high as to result in the fundamental modeling equations used in this work. LIS processes that feature significantly lower selectivity require a different set of modeling equations. Second, such processes are unlikely to be economical, as processes with lower selectivity result in significantly higher cost per SWU. Nevertheless, recent years have seen some interest in developing MLIS processes for commercial use, and if this is achieved, an analysis of the process like that shown in this work would prove useful.

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