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A Proliferation Assessment of Third Generation Laser Uranium Enrichment Technology

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ABSTRACT

Long-standing efforts to develop a commercially viable laser-based process for uranium enrichment, initially with atomic and later molecular isotope separation, have had limited success. This article discusses a model for a third generation of laser enrichment technology where CO₂ laser light is Raman scattered to generate 16 μm photons that excite a vibrational mode in uranium-235 hexafluoride molecules within an adiabatically expanding free carrier gas jet, allowing for the partial separation of uranium isotopes by condensation repression. The SILEX (Separation of Isotopes by Laser Excitation) process being developed as part of the Global Laser Enrichment project may be one example of this separation technique. An ideal, asymmetric cascade for enriching uranium to weapon-grade levels is presented, and an analysis of the minimum laser performance requirements is included. Optimal running parameters, physical space constraints, and energy efficiency estimates are discussed. An assessment of the technical skills required is also provided. Finally, material available in an online supplement discusses possible lasers that may be utilized in such a process, and offers an introduction to dimer formation, a laser-based enrichment cascade, and a model for estimating the enrichment factor.

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Introduction

The search for an economical laser-based method for enriching uranium has thus far had limited success. Different excitation techniques with either atomic or molecular species of uranium along with different methods of harvesting the enriched product have been investigated, but no laser technique has proven competitive with the gas centrifuge. Third generation laser enrichment systems currently under development, however, may challenge the dominance of the gas centrifuge in the uranium enrichment market, and the consequences for the proliferation of nuclear weapons have yet to be fully assessed.

One third generation laser enrichment process under consideration for commercialization is known as separation of isotopes by laser excitation (SILEX).¹ It was

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developed at laboratory scale by Silex Systems Ltd., an Australian company, and has been licensed to Global Laser Enrichment (GLE), a consortium comprised of General Electric (51 percent), Hitachi (25 percent), and Cameco (24 percent), for commercial development. GLE was issued a construction and operating license for a laser enrichment plant at General Electric-Hitachi's nuclear fuel fabrication facility in Wilmington, North Carolina, by the United States Nuclear Regulatory Commission (NRC) in 2012.² GLE has stated that a decision about whether to proceed with this project will be based upon market considerations, but Silex Systems Ltd. claims key technology improvements that may lower the operating and capital costs of a commercial production facility.³ Other countries pursuing research related to this technology⁴ could also build plants if they believe it is cheaper to enrich uranium with lasers than with centrifuges.

Concerns about the proliferation risks of SILEX technology were raised in a 2010 petition to the NRC requesting that proliferation assessments be included as part of the licensing process.⁵ A 2012 letter to the NRC by nineteen nuclear experts also requested a proliferation assessment.⁶ The only assessment done was internally commissioned by General Electric-Hitachi but was never released to the public. It was reported to conclude that while a SILEX facility could require less physical space and use less power than a comparable centrifuge plant, a SILEX enrichment plant nonetheless may be easier to detect, and in any case the laser technology was beyond the technical means of most proliferant states.⁷ While the most likely detection signal referred to in this assessment is a market signature from laser-related equipment purchases, it is unclear if the assessment considered all possible laser systems that could be used with SILEX, as identifying all such lasers and related equipment could be complicated by other commercial and research applications for them. In addition, it is unclear if the judgment that the laser technology would not be accessible to most states considered all possible laser systems.

This article aims to provide an initial analysis of some key technical features of a possible third generation laser uranium enrichment system and their relevance for nuclear weapons proliferation. The first section discusses what is currently known about SILEX and the laser that could be used in this process. These details are then used to analyze the minimum performance requirements of a laser used to vibrationally excite all uranium-235 in a gas flowing through a low-pressure vessel. The second section discusses the isotopic separation method known as condensation repression.

The third section of the article presents possible operating parameters and estimated enrichment factors for a third generation laser enrichment system and results from a model for an ideal, asymmetric cascade to produce enough highly enriched uranium (HEU), enriched to 90 percent in the uranium-235 isotope (also known as weapon-grade HEU), suitable for a nuclear weapon. The results of this analysis were used to estimate the physical area required for a laser enrichment facility to produce more than 30 kg of 90 percent enriched HEU annually, and the minimum laser performance requirements and other electricity needs were used to calculate the energy consumption per unit of

enrichment capacity. A comparison with a similar scale centrifuge facility is provided.

The online supplement describes some laser technologies that may be suitable for uranium enrichment by condensation repression, as well as some other applications for them. It also provides more details about dimer formation, a model of the enrichment factor of a third generation laser enrichment system building on the work of J. Eerkens, and a model for an ideal, asymmetric laser enrichment cascade for producing weapon-grade uranium.

Third generation laser uranium isotope separation technology

Atomic and molecular versions of laser isotope separation have been used for uranium enrichment with limited success. Atomic Vapor Laser Isotope Separation (AVLIS) involves the selective excitation and ionization of uranium-235 from atomic uranium vapor using two- or three-color visible or ultraviolet laser irradiation, and the most popular scheme of Molecular Laser Isotope Separation (MLIS) involves the dissociation of a fluorine atom from uranium-235 hexafluoride ($^{235}\text{UF}_6$) followed by uranium-235 pentafluoride ($^{235}\text{UF}_5$) precipitating out of the gas. Ionization of uranium-235 by AVLIS involves a minimum input of 6.1 eV of energy, and dissociation of a fluorine atom in MLIS requires at least 2.7 eV. Both of these techniques were investigated over the past forty years, and both were abandoned due to cost challenges such as expensive chemical methods, materials corrosion, low laser pulse repetition rates, or inefficient laser excitation.⁸

By contrast with these previous techniques, third generation laser isotope separation systems use UF_6 for feed, product, and tails streams.⁹ The general physical concept involves the laser excitation of a vibrational mode in $^{235}\text{UF}_6$ molecules diluted in a carrier gas, which results in two forces that cause different migration rates between excited $^{235}\text{UF}_6$ ($^{235}\text{UF}_6^*$) and $^{238}\text{UF}_6$ away from the core of an expanding supersonic free jet.¹⁰ This allows the partial separation of $^{235}\text{UF}_6$ before the gas condenses.¹¹ SILEX is only one method of such a condensation repression approach to laser isotope separation. Other variations of the concept may challenge SILEX in the future.

Photons with wavelengths of 16 μm provide the 0.08 eV energy required to excite the ν_3 vibrational mode of $^{235}\text{UF}_6$.¹² Such photons can be produced by the Raman scattering of 10.2 μm wavelength photons from a pulsed high-power CO_2 laser off optically pumped para- H_2 gas within a conversion cell.¹³ Raman scattering is the inelastic scattering of photons, and scattering from an excited rotational state of para- H_2 allows for the loss of enough energy to emit 16 μm light. This is needed because no high-power 16 μm laser currently exists to directly excite the ν_3 vibrational mode of $^{235}\text{UF}_6$.

While there are multiple suggestions that the SILEX process uses the 16 μm laser, others suggest that a carbon monoxide (CO) laser emitting 5.3 μm light could be used to excite the $3\nu_3$ band instead. The energy deposited in the excitation would be three times higher (0.24 vs. 0.08 eV), and the cross section for the $3\nu_3$

transition is $\sim 5,000\text{--}10,000$ times smaller than that for ν_3 , raising the required energy for successful operation. Repetition rates with CO lasers are claimed to be as high as 10 MHz, which would easily irradiate all $^{235}\text{UF}_6$ molecules flowing through a separation unit. Possible CO lasers designed to compensate for $3\nu_3$'s smaller cross section are discussed in the online supplement.

Laser performance characteristics

The laser likely employed by GLE with SILEX is the Transversely-Excited Atmospheric (TEA) CO_2 Raman-shifted system. This laser is available commercially (under export controls), but the Raman-shifting must be accomplished separately. There are two performance characteristics of a Raman-shifted $16\ \mu\text{m}$ pulsed laser that are most relevant when considering its effectiveness at separating isotopes: its repetition rate and energy fluence per pulse. The repetition rate is the number of pulses emitted by the laser per second and indicates what fraction of the UF_6 is irradiated with the laser. The energy fluence per pulse is the energy per cross-sectional area of the laser beam and is an important parameter in both achieving the most economical use of photons and in reaching the required peak power threshold for Raman scattering (see online supplement).¹⁴

When laser light irradiates UF_6 , there is a minimum amount of energy that each laser pulse must have to vibrationally excite all $^{235}\text{UF}_6$ molecules.¹⁵ The beam's energy fluence (energy per cross-sectional area) must reach a minimum threshold for excitation of all $^{235}\text{UF}_6$ flowing from the nozzle. The reason is that the vibrational mode has a cross section σ , an area around the $^{235}\text{UF}_6$ molecule into which a photon must enter for excitation to occur, and there must be a photon distribution that is geometrically sufficient for excitation of all $^{235}\text{UF}_6$. Shorter pulses with the same energy are therefore better than longer ones assuming they are long enough to avoid Dicke superradiance.¹⁶ If those photons are spread along the length of a longer pulse, there will be less energy per cross-sectional area than minimally required along each point of it. The cross-sectional dependence of the minimum energy required suggests this is not as efficient as if the photons were densely packed into the beam's cross-sectional area in a shorter pulse. This would require less energy and make the technique more economical. Development to improve the energy fluence by shaping the laser pulse is a focus of research groups around the world.¹⁷

Calculating the minimum energy fluence needed to excite all $^{235}\text{UF}_6$ molecules is done using the rate equation

$$\frac{dN_2}{dt} = W_{12}N_1 - W_{21}N_2 \quad (1)$$

where N_1 and N_2 are the respective molecular densities in cm^{-3} of the ground and excited ν_3 vibrational state of $^{235}\text{UF}_6$, W_{12} is the transition probability between the ground and excited ν_3 vibrational state, and W_{21} is that between the excited vibrational and ground state. Most possible processes that may deexcite $^{235}\text{UF}_6^*$, represented by the term $W_{21}N_2$ in Equation 1, may be neglected here as they are not

likely to make a significant contribution during the transit time through the beam. This includes collisions between a carrier gas G and $^{235}\text{UF}_6^*$, where the energy can be absorbed by many rotational energy changes of $^{235}\text{UF}_6$ that allow it to avoid deexcitation by stimulated emission¹⁸ and spontaneous emission.¹⁹ Significant relaxation due to direct vibration to vibration transfers in collisions between $^{235}\text{UF}_6$ and $^{238}\text{UF}_6$ can be avoided if the concentration of UF_6 in G is kept low (<0.05).²⁰ The only other deexcitation mechanism for $^{235}\text{UF}_6^*$ is dimer formation, but this process is desired for better enrichment. The probability and rates of these deexcitation mechanisms are included in the model discussed in the online material.

The term $\frac{dN_2}{dt}$ in Equation 1 represents how many $^{235}\text{UF}_6$ molecules can be excited per unit time, and depends upon multiple parameters of the SILEX separation unit, including the laser's pulse repetition rate. If a new laser pulse appears as frequently as it takes one UF_6 molecule to cross the diameter of the beam, then almost all of the UF_6 will be irradiated. When considering a circular beam spot, there will be some UF_6 left unirradiated at the top and bottom of the beam, but as an approximation the laser coverage with such repetition rate capability can be assumed to irradiate nearly all UF_6 . This allows $\frac{dN_2}{dt}$ to be written as

$$\frac{dN_2}{dt} = \frac{n_{235}}{2R_{\text{beam}}/U} \quad (2)$$

where $n_{235} = 0.97 \times 10^{19} p_{\text{UF}_6}/T \times 0.007$ is the molecular density (cm^{-3}) of excited $^{235}\text{UF}_6^*$ after irradiation (0.007 is the concentration of uranium-235 in natural uranium),²¹ R_{beam} is the radius of the beam spot, and U is the velocity of the jet core crossing the path of the beam.

The probability of excitation W_{12} in units of s^{-1} is equal to

$$W_{12} = \frac{\sigma(\nu_3)I}{h\nu} \quad (3)$$

where $\sigma(\nu_3) \approx 10^{-18} \text{ cm}^2$ is the strongly temperature-dependent cross section of the ν_3 transition,²² I is the laser intensity in W/cm^2 , and $h\nu = \frac{hc}{\lambda} = 1.25 \times 10^{-20} \text{ J}$ is the $16 \mu\text{m}$ photon energy. Given the goal of exciting all $^{235}\text{UF}_6$ crossing the beam, $N_1 = n_{235}$ in Equation 1. What follows is a simple expression for calculating the minimum laser intensity needed to excite all $^{235}\text{UF}_6$ molecules:

$$I_{\text{min}} = \frac{Uh\nu}{2\sigma(\nu_3)R_{\text{beam}}} \quad (4)$$

For a free jet with a supersonic velocity of $U = 3 \times 10^4 \text{ cm/s}$ and $R_{\text{beam}} = 0.5 \text{ cm}$, the minimum laser intensity needed to excite all $^{235}\text{UF}_6$ is $374 \text{ W}/\text{cm}^2$, with a repetition rate of 30 kHz. Given the assumption of a circular beam spot, only 78.5 percent of $^{235}\text{UF}_6$ may be excited where only $374 \times 0.785 \approx 294 \text{ W}$ of power is needed.

Returning to energy fluence, 294 W means that only $\sim 10 \text{ mJ}$ of energy is needed to excite all $^{235}\text{UF}_6$, but the energy distribution within a pulse determines whether this is an accurate estimate. Equation 4 is effectively a calculation of the number

of incident photons required to excite a single $^{235}\text{UF}_6$ molecule flowing through a separation unit. The number required depends only upon $\sigma(\nu_3)$, as R_{beam} and U only determine the transit time through the beam. If all of these photons do not arrive at the same time, there exists a greater chance that the molecule will remain unexcited, as the required fluence of photons was too small to account for all of its possible locations within the beam. This is why photon flux is not accurate, and why longer pulses are not desired; the spacing of photons will not be as ideally distributed for the highest chance of excitation. With sufficiently short pulses, reported to be as short as 80 ns from Raman-shifted lasers,²³ the minimum energy fluence ψ_{min} needed to excite all $^{235}\text{UF}_6$ can be expressed as the photon energy divided by the cross section:

$$\psi_{\text{min}} = \frac{2I_{\text{min}}R_{\text{beam}}}{U} = \frac{h\nu}{\sigma(\nu_3)}. \quad (5)$$

This article will assume that 80 ns pulses are sufficiently short to approximate the total pulse energy per unit area as a measure of energy fluence, and not only of energy flux.

Another consideration is that the energy fluence is high enough so that the minimal level is maintained over the entire length of the separation unit. This requires an understanding of how much energy is absorbed over the distance the laser light is traveling. For a single pulse, the energy absorbed E_{abs} when penetrating through a depth z is

$$E_{\text{abs}} = h\nu\pi R_{\text{beam}}^2 n_{235}z \quad (6)$$

where n_{235} , introduced earlier in Equation 1 as N_1 and N_2 , is the molecular density of $^{235}\text{UF}_6$. The parameters p_{UF_6} and T on which n_{235} depends are those at the location where the laser is irradiating the jet. For a laser beam of $R_{\text{beam}} = 0.5$ cm, the best cross-axial overlap with the jet core is achieved at $p_G = 0.026$ torr and $T = 81$ K. This gives $n_{235} = 2.18 \times 10^{13} \text{ cm}^{-3}$ and $E_{\text{abs}} = 2.18 \times 10^{-5}$ J of energy absorbed for every meter of a separation unit at these running parameters. The length of a separation unit refers to the total penetration depth of UF_6/G gas that a laser travels through in cross-axial irradiation of a free jet. If Equation 6 is simply divided by πR_{beam}^2 , adding the resulting energy fluence lost to absorption gives a more complete expression for ψ_{min} in Equation 5 of

$$\psi_{\text{min}} = h\nu \left(\frac{1}{\sigma(\nu_3)} + n_{235}z \right) \quad (7)$$

which is the minimum energy fluence needed to excite all $^{235}\text{UF}_6$ in a separation unit of length z . The power absorbed over this length is simply E_{abs} multiplied by the laser's pulse repetition rate.

If approximating the laser beam spot as one square centimeter, the minimum pulse repetition rate f_{min} in Hz needed to irradiate all uranium is the magnitude of the free-jet core's velocity U calculated in cm/s :

$$f_{\text{min}} = |U| \quad (8)$$

Using a more realistic circular beam spot, a rate of f_{\min} would only irradiate 78.5 percent of the uranium flowing through the separation unit. A well-placed mirror should be able to solve that issue, however, and allow for 100 percent irradiation.²⁴ This formalism could be applied to any pulsed laser system. The online supplement discusses the design of a pulsed CO laser within this context when considering how to accommodate the smaller cross section of the $3\nu_3$ vibrational mode.

In 2005 it was revealed that Silex Systems had lasers operating at a 50 Hz repetition rate, but that the lasers could produce pulses at 300 Hz,²⁵ still insufficient to irradiate the entire feed stream. What capabilities currently exist is unknown, but there are no physical constraints on being able to produce kilowatts of 16 μm light pulsed at tens of kHz. A laser emitting 6 kW of power at 2 kHz repetition rates was reported by a group from South Africa in 1991,²⁶ and this remains the most advanced performance of a Raman-shifted TEA CO₂ laser cited in the public literature. Much higher repetition rates may currently be out of reach by all but the most sophisticated of engineering workforces, but it is not clear that it will long remain beyond the capabilities of a number of states. Aside from this possibility, multiple lasers with only a fraction of these capabilities could be combined in various ways to achieve this goal, and other laser systems may be developed for the same purpose.

The enrichment factor for a 5.3 μm CO laser may be higher than with the 16 μm laser, which would require fewer stages and possibly less space to cascade to 90 percent HEU. Fewer or more conflicting market signals could also complicate tracking purchases of a CO laser or its components, while the possibility exists that such a system may be indigenously assembled without the assistance of an external supplier. It is important to note that the Nuclear Suppliers Group did add CO lasers operating between wavelengths of 5 and 6 μm to its list of dual-use equipment in 2013 as equipment that is now controlled by all participating governments in accordance with its Guidelines for Nuclear Transfers.²⁷ The details of suggested CO laser designs are discussed in the online supplement.

Condensation repression by laser excitation

The likely SILEX process feeds UF₆ gas through a nozzle into a low-pressure chamber where supersonic free jets are formed by adiabatic expansion. The vibrational excitation of ²³⁵UF₆ allows it to resist condensing by causing the nearly instant dissociation of dimers upon their formation, and the recoil energy acquired by these molecules allows for physical separation from ²³⁸UF₆ for collection. The small isotope shift $\Delta\nu_3$ of 0.6 cm^{-1} requires cooling the gas to low temperatures to narrow and separate the overlapping absorption spectra and improve the selective excitation of ²³⁵UF₆. This section will outline this condensation repression process and provide details about nozzle design, the relationship between parameters during free jet expansion, and identifiable constraints that affect design choices.

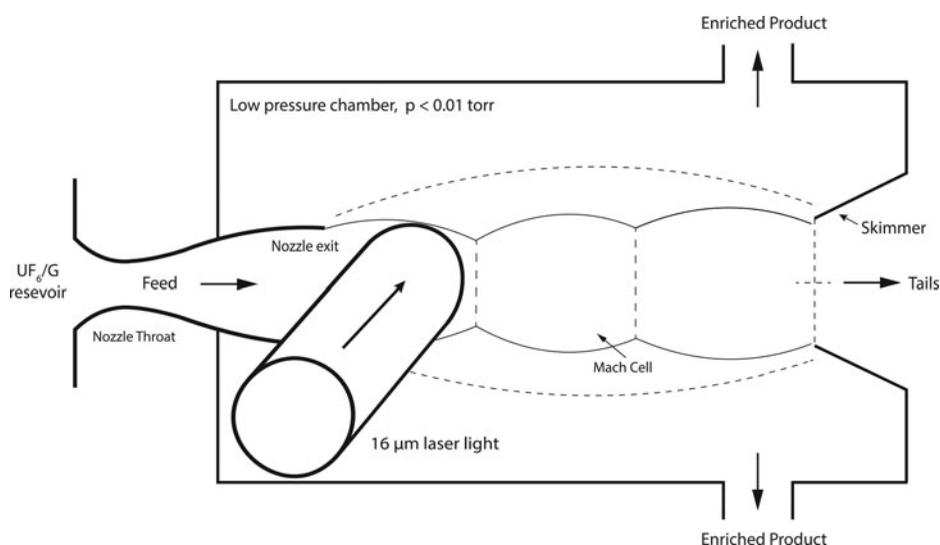


Figure 1. Schematic of a third generation laser isotope separation unit for uranium enrichment. A high repetition rate laser tuned to excite a vibrational mode of $^{235}\text{UF}_6$ cross-axially irradiates a UF_6/G free jet flowing from a supersonic nozzle into a low pressure chamber. The enriched product is collected with a downstream skimmer.

Nozzle expansion and free jet flow

The importance of high isotopic selectivity requires that the UF_6 gas be cooled to narrow and separate the absorption spectra. This is achieved by feeding the gas into an evacuated chamber through a nozzle to form a supersonic jet, which cools as it adiabatically expands and accelerates. Absent such cooling, thermal excitations would result in the unwanted excitation of some $^{238}\text{UF}_6$ molecules due to the overlap of its vibrational absorption spectrum with $^{235}\text{UF}_6$.²⁸

Supersonic speeds can be achieved when a gas passes through a converging and diverging duct, typically called a Laval nozzle (Figure 1). The downstream speed for the free jet core depends upon both the ratio of the nozzle exit and throat cross-sectional areas and the ratio of the specific heats at constant pressure and volume of the jet gas. Figure 1 displays the evolution of the free jet as it expands into a low-pressure chamber, and the oscillating expansions and contractions that form due to adjustments in pressure (shock waves) from the jet flow are called Mach cells. It is estimated that the temperature of the jet gas can increase by 50–60 percent after a shock compared to before due to the work done on the free jet by the pressure change.²⁹ This may be a limiting factor in dimer formation, which is what allows the SILEX process to work, and could complicate deciding on the optimal operating parameters like temperature and pressure.

For successful condensation repression, the low-pressure chamber typically contains background gases pumped down to less than 0.01 torr that provide a boundary layer for the expanding free jet to prevent excessive flaring of the gas from the jet.³⁰ Using torr for the units of pressure may seem antiquated, but given that 1 torr equals approximately 133.3 pascals (Pa) and that a UF_6/G gas mixture of only 5–200 torr is

manageable in the upstream reservoir before nozzle expansion,³¹ using torr seems appropriate for more workable numbers.

Before nozzle expansion and laser excitation, the UF₆ is mixed with a carrier gas G that acts to suppress vibration to vibration energy transfers between uranium isotopes. The concentration of UF₆ is typically kept below 5 percent for this reason,³² but experiments are needed to determine the ideal UF₆ concentration.³³ It is important that G be mixed prior to the expansion, as low-energy collisions between UF₆ and G within a supersonic beam will make dimer formation possible in a gas at low temperature. Conservation of energy will prevent their formation due to the large difference in relative velocities between supersonic UF₆ colliding with G already present in the irradiation chamber. All streams (feed, product, and tails) are therefore really a UF₆/G mixture for all stages in a cascade of laser separation units producing higher enrichments.

The design parameters of a Laval nozzle must balance choices between better isotope separation and processing a high volume of UF₆. For example, a larger nozzle exit would allow more gas to be processed, but it would come at the expense of a smaller enrichment factor, as there would be more molecules to collide with as ²³⁵UF₆ travels out of a thicker free jet. One possible way around this tradeoff is to orient the laser cross-axially to the free jet and irradiate the gas along a longer separation unit as shown in [Figure 1](#). This geometry is likely the most preferred for industrial-scale production, and if a nozzle of such depth could be manufactured, there would be advantages in processing large volumes. If such a nozzle is not workable, multiple smaller axi-symmetric nozzles could be aligned along the separation unit to achieve the same advantage, although with some cost of occupying a larger area. The nozzle design must also consider where along the free jet the laser will irradiate the gas, as the jet pressure and temperature when irradiated will affect the level of enrichment in the product. A nozzle with a longer neck may be preferable (a greater distance between the throat and exit in [Figure 1](#)) so that irradiation within the nozzle aids in dimer formation (see online supplement) and a smooth pre-expansion of the gas prevents flaring.³⁴

Understanding the relationship between the nozzle geometry and downstream flow parameters and shock waves is desirable, but the theoretical calculations for two-dimensional free jets are unfortunately complex.³⁵ Average order-of-magnitude values, however, can be obtained from the following one-dimensional supersonic expansion relations:³⁶

$$\frac{T}{T_0} = \left(\frac{p}{p_0} \right)^{(\gamma-1)/\gamma} \quad (9)$$

$$\left(\frac{A}{A_t} \right)^2 = \frac{\gamma - 1}{2} \frac{\left(\frac{2}{\gamma+1} \right)^{(\gamma+1)/(\gamma-1)} \left(\frac{p_0}{p} \right)^{2/\gamma}}{\left[1 - \left(\frac{p}{p_0} \right)^{(\gamma-1)/\gamma} \right]} \quad (10)$$

$$\left(\frac{U}{U_{so}} \right)^2 = \frac{2(1 - T/T_0)}{\gamma - 1} = M^2. \quad (11)$$

The subscripts o and t refer to conditions in the upstream reservoir (before nozzle compression and expansion) and throat (Figure 1), respectively, and A_t is the throat's cross-sectional area. Parameters with no subscripts indicate downstream conditions in the supersonic free jet, where A is the cross-sectional area of the jet and U is the supersonic velocity of the jet's bulk flow. T and p are the downstream temperature and pressure of free jet, and T_o and p_o represent those parameters in the upstream reservoir. U_{so} is the velocity of sound in the reservoir and can be calculated with

$$U_{so} = 9,116.5\{\gamma T_o(\text{K})/m(\text{g/mol})\}^{1/2} \quad (12)$$

where m is the molar mass of G in g/mol.³⁷ The ratio U/U_{so} in Equation 11 is referred to as the Mach number M . The gas coefficient $\gamma = c_p/c_v$, where c_p and c_v are the specific heats at constant pressure and volume, is an important parameter in choosing G.

The idea with Equation 9 is to find optimal values for T and p by first choosing one and allowing the other to vary to find the highest enrichment factor. Once T and p are selected, a choice for T_o (typically ~ 300 K) determines p_o . Equation 10 is used to choose the area of the nozzle throat A_t and what pressure p or area A is desired in the free jet downstream. Choices about p at the location of product collection, what A best overlaps with the laser beam spot, and what nozzle dimensions (A_t and A) need to be mechanically engineered are all related through Equation 10. U is determined by T in Equation 11 and depends upon U_{so} within G. These relations will be referred to again in later sections, but this provides an introduction to how they could be used.

Dimer formation

For the UF_6 molecule to remain intact and the collected product enriched in the ^{235}U isotope, interaction with another gas is necessary. This begins as collisions between UF_6 and G form weak bonds between them. The bonded units are collectively referred to as a dimer, and are typically held together by a hydrogen or Van der Waals bond. Although dimers are defined as two molecules or atoms bonded together, the study of them is dominated by those with weak bonds, as they are more likely to break apart from collisions below the equilibrium vapor pressure and questions about these dynamics remain.³⁸ The key idea is that while $^{238}\text{UF}_6$ and $^{235}\text{UF}_6$ form dimers at the same rate, they are much shorter lived if they include laser-excited $^{235}\text{UF}_6$ ($^{235}\text{UF}_6^*$). The binding energy of a Van der Waals bond (~ 0.01 eV) is much less than the energy of the excited ν_3 mode of $^{235}\text{UF}_6$ (~ 0.08 eV), and it takes many fewer collisions for $^{235}\text{UF}_6^*:\text{G}$ rather than $^{238}\text{UF}_6:\text{G}$ dimers to dissociate, as the attractive potential well is not as deep. Upon dissociation, the laser-excited vibrational energy from $^{235}\text{UF}_6^*$ is converted to translational recoil energy and $^{235}\text{UF}_6$ begins to flee the jet core at a faster rate. When the outer rim of the gas is collected, the product is enriched in $^{235}\text{UF}_6$. This process is displayed in Figure 2. More details are provided in the online material together with an introduction to nucleation and particle

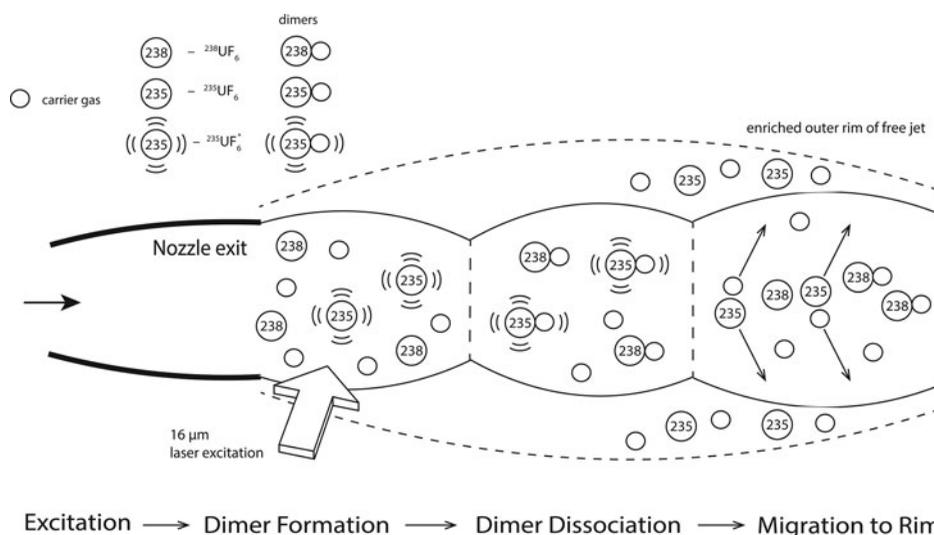


Figure 2. The process of laser excitation, dimer formation, dimer dissociation, and migration to produce enriched product.

growth, which along with dimer formation are the beginning processes of condensation. Successful enrichment with third generation laser-based systems involves separating uranium isotopes before nucleation and particle growth occur in a cooling free jet.

The running parameters for SILEX will likely include a carrier gas pressure p_G between 0.03 and 0.002 torr during the expansion through the separation unit, and assuming a low UF_6 concentration of 2–10 percent to limit vibration to vibration energy transfers, a UF_6 partial pressure p_{UF_6} between $\sim 2 \times 10^{-4}$ to 2×10^{-5} torr. The equilibrium vapor pressures $p_e(T)$ at 100 K, a likely temperature at which laser irradiation will occur, and at 45 K and 25 K, two possible temperatures at which product collection will occur, are respectively much lower at 3.97×10^{-19} , 1.54×10^{-82} , and 5.53×10^{-249} torr.³⁹

The time for irreversible particle growth to set in t_c is defined as the time for 20 percent of the UF_6 to reach “critical embryo” size. At $p_{\text{UF}_6} \sim 2 \times 10^{-4}$ torr, $t_c \sim 95$ s at $T = 25$ K due to the smaller number of collisions at that low temperature, but is ~ 0.1 s at 45 K. At 100 K, t_c would be longer since more UF_6 molecules would be required to bond together before irreversible growth could occur. Thus if the transit time from nozzle to skimmer is $t_{\text{tr}} < 0.1$ s significant cluster growth can be avoided.⁴⁰ The possibility of cluster growth is important when considering whether UF_6 should be more or less diluted with the carrier gas G at a particular temperature, an important factor when considering whether processing a higher volume of UF_6 is possible. A transit time $t_{\text{tr}} < 0.1$ s is likely needed to avoid 20 percent cluster growth, and suggests that $p_{\text{UF}_6} \sim 2 \times 10^{-4}$ torr is the upper limit of allowed partial pressures for adequate separation. Section B of the online material provides more details.

Enrichment factors and optimal running parameters

A measure of the efficacy of an enrichment process is the change in isotopic concentrations between the feed and product materials. This net change can be quantified by the enrichment factor β and the depletion factor γ defined as

$$\beta = \frac{N_P/(1 - N_P)}{N_F/(1 - N_F)} = \frac{R_P}{R_F} \quad \text{and} \quad \gamma = \frac{N_F/(1 - N_F)}{N_T/(1 - N_T)} = \frac{R_F}{R_T} \quad (13)$$

where N_P , N_F , and N_T are the respective concentrations of $^{235}\text{UF}_6$ in the product, feed, and tails, and $R_P = N_P/(1 - N_P)$, $R_F = N_F/(1 - N_F)$, and $R_T = N_T/(1 - N_T)$ are the respective relative isotopic abundances of $^{235}\text{UF}_6$ to $^{238}\text{UF}_6$ in those streams. The factors β and γ can be multiplied to give the overall separation factor α between the product and tails,

$$\alpha = \beta\gamma = \frac{N_P/(1 - N_P)}{N_T/(1 - N_T)} = \frac{R_P}{R_T} = \frac{\beta(1 - \theta)}{1 - \beta\theta}. \quad (14)$$

Unlike β , α depends on the cut θ , which is the fraction of feed material collected as product. The figure of merit used to assess the economics for an isotope separation process typically uses α and θ , where the economics improves as α and θ increase.⁴¹ These two parameters, however, are not independent. Increasing one may decrease the other, and will affect β as well. Its value determines how many stages are needed to acquire 90 percent HEU, how much time would be needed to acquire it, and is a key element in any proliferation assessment.

A conservative estimate of $\beta = 1.5$ to 2.5 for third generation condensation repression laser isotope separation is given by Eerkens and Kim (a similar estimate not taking into account the dynamic of free jet expansion is provided in the online supplement).⁴² Values of $\beta \sim 2$ are likely underestimates, as they do not dynamically model the change in $^{235}\text{UF}_6$ migration rates to the rim of the free gas jet as its pressure lowers from expanding and cooling, thereby increasing β . Michael Goldsworthy, the CEO of Silex Systems Ltd., has claimed that the enrichment factor for SILEX is between 2 and 20.⁴³ These enrichment factors are much higher than for typical gas centrifuges and gaseous diffusion systems.⁴⁴

The previous discussion permits an estimate for the uranium flow rate through a single separation unit, which in turn will provide a basis for estimating the separation length needed to produce 90 percent HEU. The flow rate F may be calculated from

$$F(\text{g UF}_6/\text{s}) = \frac{d_{\text{jet}} U n_{\text{UF}_6} m(\text{g/mol}) z}{6.025 \times 10^{23} \text{ molecules/mol}} \quad (15)$$

where d_{jet} is the diameter of the free jet, U is the velocity of the jet's bulk flow (Equation 11), n_{UF_6} is the molecular density of UF_6 , m is the molar mass of UF_6 in g/mol, and z is the penetration depth of the separation unit. If $d_{\text{jet}} = 2$ cm at the skimmer ($p_G = 0.002$ torr and $T = 35$ K), $U = 3.5 \times 10^4$ cm/s and $n_{\text{UF}_6} = 2.217 \times 10^{13}$ molecules/cm³, giving a flow rate $F = 0.0907$ g/s, or 2859 kg/yr of UF_6 for a one-meter long separation unit (Table 1). The carrier gas is assumed to be SF_6 .

Table 1. The Performance Characteristics and Processing Parameters at the Skimmer of a Single, One-meter Long SILEX Separation Unit.

Enrichment factor β	2
Cut θ	0.25
Pressure of carrier gas G	0.002 torr
Temperature	35 K
Concentration of UF ₆ in carrier gas	0.04
Flow rate of UF ₆	2859 kg/yr
Flow rate of Uranium	1933.4 kg/yr
Separative capacity	257.8 kg·SWU/yr

The enrichment factor β also depends on what fraction of the free jet flow is cross-axially irradiated by the laser, as the skimmer (Figure 1) will still cut some product from the rim of the free jet even if that gas has not been irradiated. This results in different cuts θ for irradiated and unirradiated sections, respectively referred to here as “on” and “off.” A general expression for β ’s dependence on these “on” and “off” sections is

$$\beta = \frac{\theta_{\text{on}}\beta_{\text{on}}f_{\text{rep}} + \theta_{\text{off}}\beta_{\text{off}}(f_{\text{min}} - f_{\text{rep}})}{(\theta_{\text{on}} + \theta_{\text{off}})f_{\text{min}}} \quad (16)$$

where f_{rep} is the real laser pulse repetition rate and f_{min} is the minimum laser pulse repetition rate required to irradiate all uranium. As f_{rep} approaches f_{min} in Equation 16, β approaches β_{on} .

This analysis assumes that all uranium is irradiated, as this would be the more desirable choice if cascading to 90 percent enriched uranium in a shorter time was more important than the higher costs of either acquiring more lasers or a more advanced laser. If only 78.5 percent of the uranium is irradiated with a circular beam spot, β will be affected as indicated in Equation 16. The enrichment level may not be significantly lower, however, if θ_{off} is small compared to θ_{on} .

Furthermore, the amount of material capable of being processed in one separation unit or irradiation chamber is an important constraint on the SILEX process. The need to irradiate gaseous UF₆ at a low temperature means that the molecular density of UF₆ cannot exceed 1×10^{15} molecules/cm³,⁴⁵ as rapid condensation can occur at higher densities. There are multiple parameters that can be manipulated to process different amounts of ²³⁵UF₆ over a given time, but this molecular density appears to be an upper limit.

Table 1 lists the performance characteristics of a single, one-meter long SILEX-type separation unit. The separation unit length is once again the penetration depth of the laser through the gas in its cross-axial orientation with respect to the expanding free jet. Based upon the model presented in the online material, the values of $\beta = 2$ and $\theta = 0.25$ ⁴⁶ are achieved at different pressures, which more accurately reflect the behavior of an expanding free jet than the static parameters assumed in the model. As already mentioned, the enrichment factor in Table 1 is likely underestimated, but is valuable as a conservative input into a cascade to 90 percent HEU. As a result, the total separation unit length in the following section represents the higher-end estimate to produce a significant quantity of 90 percent HEU in one year.⁴⁷

Table 2. Summary of Uranium Resource and Space Requirements for Weapon-grade Material Production Using Ideal Asymmetric Cascades with Uranium Feedstock Concentrations $N_F = 0.035$ and $N_F = 0.007$.

	$N_F = 0.035$	$N_F = 0.007$
90 percent HEU production rate	36.4 kg/yr	32.5 kg/yr
Feed-to-product ratio	27.5 kg per kg	153.8 kg per kg
External feed of uranium (mass, volume ⁴⁸)	1000 kg, 0.05 m ³	5000 kg, 0.26 m ³
External feed of UF ₆ (mass, volume ⁴⁹)	1480 kg, 16.29 m ³	7400 kg, 81.47 m ³
Required total separation unit length	11.37 m	35.25 m

Cascading to 90 percent highly enriched uranium

A summary of the two cascades modeled in the online material is shown in Table 2. The required separation unit length is based upon 1,933.4 kg of uranium flowing through a one-meter long separation unit per year (Table 1). These requirements assume no interstage mixing in the cascade. This assumption conserves separative work but may not lead to the most energy efficient cascade. Such alternate arrangements have not been considered in this analysis.

As higher enrichments are achieved in moving up the cascade, a higher concentration of uranium can likely be used in the carrier gas, as adverse effects to the selectivity due to vibration to vibration transfers between ²³⁵UF₆ and ²³⁸UF₆ become less of a limitation. There is no clear model about limits of this deexcitation mechanism, and if concentrations > 0.05 in G are possible, less separation unit length would be required. Whether this is true depends upon what can be measured on separation units in a lab.

Batch recycling

Batch recycling allows for 90 percent HEU production in less time than does cascade interconnection, but this advantage requires more uranium feedstock to start, as it uses only the product of one stage as feed for the next. This is an inefficient use of uranium since the tails are not recycled, and this is particularly true in SILEX, as the product cuts tend to be fairly low. The attractiveness of batch recycling with SILEX, however, depends upon the size of β , and would clearly be the quickest route to a nuclear weapon if $\beta \sim 10$.

Using the model in the online material, where θ depends upon the feed concentration and $\beta = 2$, 25 kg of 90 percent HEU is acquired in 8 stages. This requires 49,382 kg of 3.5 percent uranium feedstock, with $\theta = 0.36$ in the first stage and $\theta = 0.43$ in the final one.⁵⁰ This is 1975 kg of 3.5 percent enriched feed for every 1 kg of 90 percent HEU produced. Centrifuges, by contrast, need only 90 kg of 3.5 percent feed for every kilogram of weapon-grade HEU produced with batch recycling.⁵¹ This performance reflects a clear preference for centrifuges, as the uranium requirements appear intolerably high.

As β increases, however, fewer stages and less uranium are required. Table 3 shows the uranium requirements with batch recycling for $\beta = 2, 3$, and 10. If

Table 3. A Summary of the Required Uranium Resources and Separation Unit Lengths for the Production of 25 kg of 90 Percent HEU Using Batch Recycling with Uranium Feedstock Concentrations $N_F = 0.035$ for $\beta = 2, 3$, and 10 and $N_F = 0.007$ for $\beta = 10$.

Enrichment factor β	$\beta = 2$	$\beta = 3$	$\beta = 10$	$\beta = 10$
Feed Concentration N_F	0.035	0.035	0.035	0.007
Enriching stages	8	6	3	4
Feed per kg of 90 percent HEU	1,975 kg	1,562 kg	42 kg	151 kg
External uranium feedstock	49,382 kg	39,061 kg	1,049 kg	3,774 kg
Required separation unit length	38.7 m	26.4 m	0.7 m	2.53 m

$\beta = 3$, the required uranium feedstock of 1562 kg per kg of weapon-grade material still seems intolerably high, but for $\beta = 10$, 42 kg per kg 90 percent HEU would provide an attractive pathway to a nuclear weapon. Yet without a declared SILEX-type facility, the 3.5 percent feedstock would likely have to be diverted from a declared centrifuge facility, making this pathway less likely from a clandestine plant. If natural uranium feedstock is used, however, requiring only 151 kg per kg of weapon-grade material with $\beta = 10$ might be very tolerable for a proliferator. The additional need for only 2.53 m of separation unit length would make weapon-grade material acquisition in a such a small space perhaps the most attractive aspect. The likelihood of this pathway would seem to depend on where β falls between the range of 2 and 20.

Minimum space required for the SILEX process

The estimated area required to operate a third generation laser enrichment facility based on a SILEX-type model assumes that a clandestine facility would employ a three-up, two-down cascade as described in the online material, and would use only 5000 kg of natural uranium, or 7400 kg of UF_6 . Figure 3 displays a possible arrangement. The 39 stages are arranged in three parallel product streams, with the product collected at stages 37, 38, and 39. The size of the stages are not drawn to scale, as each stage requires a different flow rate, but with a total separation length of 32.5 m each of product streams was assumed to be 13 m long to allow space between the stages and room for the mirrors at the end. Raman shifters (discussed in the online supplement) are each 4 m in length. The width of each product stream was allowed 2 m to allow access to the equipment for a total width of 6 m. Adding 2 m between the Raman shifters and the wall to allow for laser beam propagation and worker access, the total required space by the product streams and Raman shifters was 20 m \times 6 m, or 120 m².

The optics bench in Figure 3 was assumed to be 5 m \times 2 m, and each laser crate was designed to stack 4 lasers vertically for each product stream. Each laser is roughly 2 m \times 1 m based upon data provided by commercial suppliers. The optics bench could support additional TEA lasers to be used as amplifiers and any optics necessary to tune and narrow the laser light. The holding tank for solid UF_6 could have a variety of dimensions and could be oriented vertically to save floor space, but allowing for two cubic meters would suffice at a density of 5090 kg/m³. The

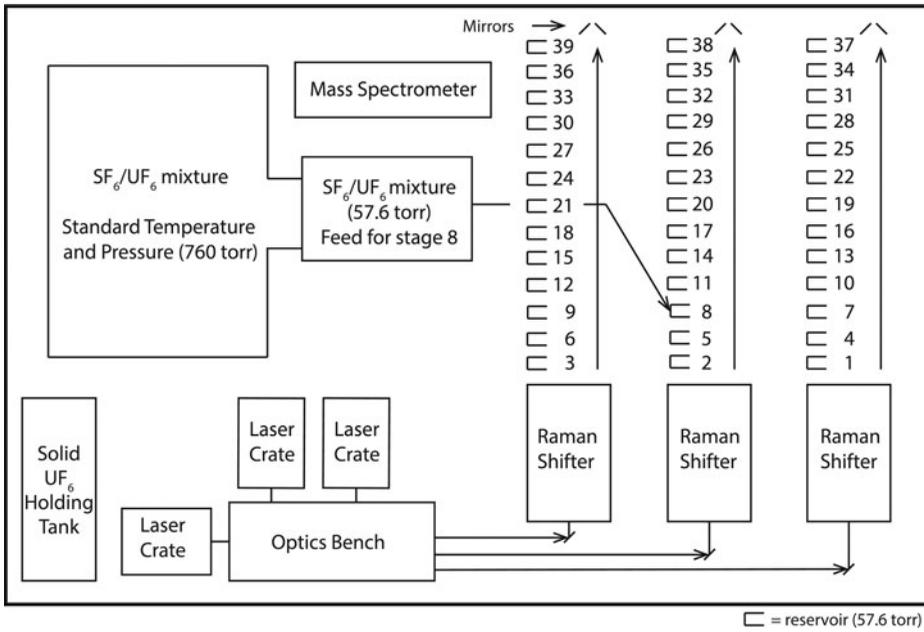


Figure 3. The area footprint ($15 \text{ m} \times 20 \text{ m} = 300 \text{ m}^2 = 3229 \text{ ft}^2$) of SILEX equipment capable of producing 32.5 kg of 90 percent HEU annually.

remaining equipment consists of a mass spectrometer to measure the $^{235}\text{UF}_6$ concentration and storage vessels to prepare the UF_6 for nozzle expansion. Compressors and vacuum pumps could easily find space, as all adequate equipment could fit within a couple square meters. A vacuum pump is needed to lower the pressure of the SF_6/UF_6 mixture from 760 to 57.6 torr, and **Figure 3** indicates a connection between the two that shows only a tank at lower pressure prior to nozzle expansion. Its size is unimportant, as storage at 760 torr is possible in a smaller space. All stages in the cascade would require a reservoir at 57.6 torr prior to nozzle expansion, and this is indicated with a small rectangle opened at one end next to each stage number. Assuming a rectangular room, an additional 117 m^2 meters remains to include this equipment. This leaves a room with dimensions $20 \text{ m} \times 15 \text{ m}$, or 300 m^2 . It is worth mentioning that the three product streams could be placed on top of each other if a proliferator desired to conserve floor space. This would remove 4 m from one dimension and leave a minimum space of 220 m^2 .

If the assumption is made that the same space for holding tanks, low-pressure storage vessels, a mass spectrometer, and compressors and vacuum pumps is required for centrifuges ($\sim 117 \text{ m}^2$), direct comparisons between the two technologies are possible. For a 5000 SWU facility with ~ 24 SWU capacity centrifuges (the claimed separative capacity of Iran's IR-8 centrifuge),⁵² 208 m^2 is required if one square meter is assumed per centrifuge. Adding one meter on each side for machine access and subtracting the space required for an optics bench and lasers for a SILEX-type system, a total of $\sim 370 \text{ m}^2$ is required for an IR-8 centrifuge facility. If URENCO centrifuges, each with 100 SWU capacity, were used in a 5000 SWU facility, a total area of only 189 m^2 would be required.

The estimate that a SILEX-type facility would require a space between those for advanced IR-8 and URENCO centrifuges suggests that it could be hidden in a building similar in size to buildings used for multiple other purposes. Different plausible assessments for the sizes of holding tanks are irrelevant considering that such equipment could be designed vertically to limit the floor space utilized. The area estimate for such a facility assumes an enrichment factor $\beta \sim 2$, and would decrease accordingly for higher values as discussed previously.

Minimum energy consumption

The minimum power consumption P_{\min} for a third generation laser enrichment system similar to SILEX is likely to be dominated by the required minimum number of photons to excite all $^{235}\text{UF}_6$. It can be expressed as the minimum energy fluence multiplied by the free jet core's velocity:

$$P_{\min} = \psi_{\min} \cdot U = h\nu \left(\frac{1}{\sigma(\nu_3)} + n_{235z} \right) \cdot U \quad (17)$$

For a separation unit of one meter with a laser irradiating at $p_G = 0.026$ torr and $T = 81$ K, the laser power needed is 374 Watts, an energy consumption of 3276 kWh/yr. With the separative capacity for such a unit being 257.8 kg-SWU/yr, the laser energy consumed is then only 12.7 kWh/SWU. For ten meters of separation length, the minimum power needed to irradiate all uranium raises to 381 Watts. This lowers the energy consumed to 1.29 kWh/SWU.

For current Enrichment Technology Company (ETC)-supplied centrifuges to Urenco plants or American Centrifuge Plant (ACP) centrifuges, about 50 kWh/SWU is required.⁵³ The 12.7 kWh/SWU (or 1.29 kWh/SWU for ten meters) estimate is low considering that 10.2 μm light must be Raman-shifted, and only 55 percent energy efficiency is claimed in the shift to 16 μm .⁵⁴ In addition, the electro-optical efficiency of the TEA CO_2 laser is only 15 percent⁵⁵ to 26 percent⁵⁶ prior to Raman-shifting. These inefficiencies change the energy consumed to 88.8–154 kWh/SWU for one meter and 9.02–15.6 kWh/SWU for ten.

In addition to the laser energy requirements, vacuum pumps and gas compressors are also required for SILEX operation. There are two needs for the vacuum pumps: to pump the UF_6/G mixture from atmospheric pressure to the desired reservoir pressure just prior to nozzle expansion and to pump the irradiation chambers within the separation units down to just below 0.01 torr. Using a commercially available liquid ring vacuum pump with a suction capacity of 68 m^3/hr ,⁵⁷ 1.5 kW of power is needed to pump 68 m^3 from a standard pressure of 760 torr to the desirable reservoir pressure of 57.6 torr. This translates into an energy consumption of 0.41 kWh/SWU.⁵⁸ To pump the irradiation chambers down to 0.005 torr, a rate of only 6×10^{-6} kWh/SWU is required.⁵⁹ Compressing the gas from 0.002 torr upon collection down to 57.6 torr for the next stage was modeled by as a polytropic process, and energy was calculated to be consumed at 0.37 kWh/SWU.⁶⁰

Table 4. A Summary of Electricity Needs Related to Lasers, Vacuum Pumps, and Gas Compressors with One-meter and Ten-meter Long Separation Units. The Energy Consumption of URENCO (or American Centrifuge Plant) Centrifuges is Listed for Comparison in the Last Column. (N. A. = Not Applicable).

Electricity need	One-Meter (kWh/SWU)	Ten-meter (kWh/SWU)	URENCO (kWh/SWU)
Laser energy consumption	88.8	9.02	N. A.
Pump for reservoir (760 to 57.6 torr)	0.41	0.41	N. A.
Pump for irradiation chamber (760 to 0.005 torr)	6×10^{-6}	6×10^{-6}	N. A.
Compression for reservoir (0.002 to 57.6 torr)	0.37	0.37	N. A.
Total energy consumption	89.6	9.80	50

A summary of the electricity needs for a SILEX-type system is shown in Table 4. The 50 kWh/SWU shown for URENCO represents the state-of-the-art energy efficiency for centrifuge technology. The energy consumption for a SILEX-type plant is higher or lower than this depending upon the length of the separation unit. Units of only a few meters would consume less energy than URENCO centrifuges. For example, a 5000 SWU SILEX-type plant operating at 10 kWh/SWU would consume as much energy as a grocery store in the United States that was only 100 m² in size.⁶¹

Expertise in laser technology

Lasers are used widely in multiple fields of research and industry, and there is a broad and growing understanding of laser physics and technologies. In principle, this puts laser enrichment systems within reach of many countries. Some third generation laser enrichment systems, however, have specific requirements that may require sophisticated expertise.

There are a number of lasers with the performance characteristics necessary for use with third generation laser enrichment technology (see online supplement). As previously mentioned, the laser likely employed by GLE for the SILEX process is the TEA CO₂ Raman-shifted system. This laser requires that the peak power incident on the Raman cell be high enough so that ~ 10 mJ pulses are produced that are capable of selectively exciting all uranium-235 flowing cross-wise through the laser beam. This might require combining multiple CO₂ laser beams in various ways depending on what lasers are accessible to a proliferator, and tuning the laser's wavelength and narrowing its linewidth may also be needed. These skills do involve techniques that require knowledge about light and optics, but not necessarily detailed knowledge about lasers. Raman-shifting, however, likely requires engineers skilled in arranging highly reflective mirrors that allow for multiple passes of laser light. Such lasers also run at high pressure (~ 5 – 8 atm), and while there are commercially available systems that claim a user would not be required to manage the high-pressure gas, this appears to be the most demanding technical skill needed with such systems. It is a skill not typically required of those familiar with lasers, and managing the gas stability is a reported challenge. Mastery of other techniques could lessen the need for this skill, but future laser innovations that do not require sophisticated

high-pressure gas management would limit the number of technical skills required for successful SILEX-type operation.

It remains possible that advanced Raman-shifting techniques would limit the need for high-pressure laser mediums by lowering the peak power threshold for Raman emission or that other lasers could be utilized with SILEX-type systems. These lasers include other pulsed CO₂ systems or CO lasers where the UF₆ is irradiated within the laser cavity. While skill with mirrors, wavelength tuning, and line-narrowing would likely be necessary, it would be a mistake to consider these skills inaccessible by those without formal technical training. Other commercially available lasers could be combined in various ways to enrich uranium with sufficient determination, and laser capabilities are always improving. Many lasers could have both their pulse length shortened and pulse energy increased by increasing the pressure in the laser's amplifying medium and adding more advanced RF pulsing or Q-switching techniques.

A more detailed technical discussion about these matters is provided in the online supplement.

Conclusion

Third generation laser uranium enrichment technology, with the SILEX process being a likely example, may create new proliferation risks. It appears that such a system could be designed and built to enrich uranium to weapon-grade levels with higher energy efficiency than state-of-the-art URENCO centrifuges. With some laser enrichment and cascade designs the efficiency could be greater by a factor of 5, and possibly much higher. This has direct impact on the possible detection of a clandestine laser enrichment facility based on size or energy use.

The space required by a laser enrichment plant capable of producing about thirty kilograms per year of 90 percent enriched uranium (sufficient for more than one weapon a year) is estimated to be about 300 m². This estimate is almost certainly generous. Satellite surveillance intended to distinguish a building hosting such an activity among other buildings this size should not be expected to provide useful information. The energy use would be less than a comparable centrifuge plant.

These conclusions suggest that third generation laser enrichment provides a new technological pathway to weapon-grade uranium and nuclear weapon development, with the acquisition of a usable laser being the main technological hurdle. The risk is that continued laser development and successfully demonstrated commercial deployment of laser enrichment may encourage more states to seek indigenous laser research programs that could provide them with a latent nuclear weapons capability. Research programs on lasers relevant to third generation uranium enrichment, however, may also be used for other applications that complicate identifying the intended purpose of equipment and programs. Attention needs to be focused on laser systems capable of enriching uranium to weapon-grade levels which may come to pose proliferation concerns comparable to if not greater than gas centrifuge development or plutonium reprocessing today.

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14. The laser linewidth could also be a factor, as one broader than the $^{235}\text{UF}_6$ absorption spectrum would be an inefficient use of photons and would adversely affect the selectivity if it overlapped with the spectrum of $^{238}\text{UF}_6$. This does not appear to be a concern with the TEA CO_2 laser, but could be an issue with other lasers. See From A. V. Kunets et al., “Diode Laser Measurements of a Pulsed TEA CO_2 Laser Linewidth,” *Optics Communications* 84 (1991): 37–41. The spectral linewidth Γ of a TEA CO_2 laser was measured to be $\Gamma = 0.013 \pm 0.002 \text{ cm}^{-1}$ at the full width at half maximum of the linewidth. The relevance of this narrow linewidth, however, depends upon centering the output from the TEA CO_2 laser on the correct wavelength prior to Raman shifting, but once this technical challenge is met there are no indications in the scientific literature of isotopic selectivity being significantly adversely affected by the unwanted vibrational excitation of $^{238}\text{UF}_6$ molecules. There is more discussed about TEA CO_2 lasers in the online supplement.
15. For a molecule like $^{235}\text{UF}_6$ with many rotational sublevels, downward stimulated emission to the ground level is avoided by changing its rotational state from J to $J \pm 1$ in a few collisions without changing its excited vibrational state. This allows the maximum fraction of vibrationally-excited $^{235}\text{UF}_6$ molecules to exceed 50 percent, with 100 percent possible if the loss of excited vibrational energy k_V is minimized with the carrier gas G and the laser excitation rate k_A is large compared to it.
16. Dicke superradiance occurs when a short pulse excites a collection of molecules too quickly (called “bleaching”) before any collisions occur, resulting in lasing out photons and thereby affecting the selective excitation. In such a case, the population of $^{235}\text{UF}_6$ that may be excited will not exceed 50%. This phenomena can be avoided if pulses long enough to allow changes in the rotational state are allowed to occur (typically $> 50 \text{ ns}$).
17. For an example of research in China, see D. J. Li et al., “Stimulated Rotational Raman Scattering at Multiwavelength under TEA CO_2 Laser Pumping with a Multiple-Pass Cell.” For research by a South African group in an Indian physics journal, see E. Ronander, H. J. Strydom, and L. R. Botha, “High-pressure Continuously Tunable CO_2 Lasers and Molecular Laser Isotope Separation,” *Pramana—Journal of Physics* 82 (2014). This is also an issue with Raman scattering, as a threshold power must be reached for any possible emission of $16 \mu\text{m}$ light. This is discussed in more detail in the online supplement, but the main idea is that if two pulses both contain 1 J of energy, the pulse with the shorter pulse duration will have the higher peak power.
18. Eerkens, “Separation of Isotopes by Laser-Assisted Retardation of Condensation (SILARC).”
19. Eerkens, “Laser-induced Migration and Isotope Separation of Epi-Thermal Monomers and Dimers in Supercooled Free Jets.”
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22. This temperature dependence originates from the broadening of acceptable wavelengths that may vibrationally excite $^{235}\text{UF}_6$ due to collisions between $^{235}\text{UF}_6$ and the carrier gas G. From J. W. Eerkens, *Rocket Radiation Handbook, Volume II. Model Equations for Photon Emission Rates and Absorption Cross-Sections* (United States Department of Commerce, 1973), the absorption cross section can be written as $\sigma_{nm} = 0.0960 \nu_{mn} \left(\frac{C_{mn}}{w_n}\right)_{\text{vib}} (R_{mn}^2)_{\text{vib}} g_c(\nu, \nu_{mn}, \Delta\nu_{mn})$, where C_{mn} , w_n , and R_{mn} are the respective nondimensional “connection” factors, matrix transition factors, and degeneracy “weighting” factors. The collisional broadening function g_c is given by $g_c(\nu, \nu_{mn}, \Delta\nu_{mn}) = \frac{1}{(\Delta\nu_{mn})} \left[\frac{[\frac{(\Delta\nu_{mn})_c}{\pi}]^2}{(\nu - \nu_{mn})^2 + [\frac{(\Delta\nu_{mn})_c}{\pi}]^2} \right]$, where ν , ν_{mn} , and $\Delta\nu_{mn}$ are the respective incident laser photon frequency, vibrational transition frequency, and broadened absorption spectrum at the Full-Width at Half Maximum (FWHM) of the spectrum. The subscript “c” simply indicates “collisional.” The broadening $(\Delta\nu_{mn})_c = 3.3841 \times 10^{26} \frac{\sigma_{1-2} p}{\sqrt{\mu_{1-2} T}}$, where p and T are

the pressure and temperature and σ_{1-2} and μ_{1-2} are the collisional cross section and reduced mass of molecules 1 and 2. Assuming the incident laser photon frequency $\nu = \nu_{mn}$, $\sigma_{nm} \propto \sqrt{T}$. Doppler broadening also depends upon temperature, but this is assumed to be negligible with cross-axial laser irradiation. It may be a significant factor with a coaxial geometry, however.

23. Eerkens, "Separation of Isotopes by Laser-Assisted Retardation of Condensation (SILARC)."
24. The use of mirrors may be an important factor in the geometrical design of SILEX technology. After laser light passes through once, two mirrors at a 90-degree included angle could direct the light back to the laser source at any desired position parallel to the incident beam. The only constraints would be the size of the mirrors and how the jet parameters at additionally irradiated locations may affect separation. The number of additional laser beam passes would raise the effective laser repetition rate by a factor of $n + 1$, where n is the number of additional passes. This would not be an accurate estimate if the only goal was to increase the 78.5 percent of the uranium irradiated in a circular beam spot to 100 percent. In such a case, some overlap with the incident beam would be required, making additional laser passes not so easily related to the effective pulse repetition rate.
25. Lyman, "Enrichment Separative Capacity for SILEX," 2–3.
26. D. M. Kemp et al., "Uranium Enrichment Technologies in South Africa," Atomic Energy Corporation of South Africa, Ltd. (1991).
27. IAEA, "Communication Received from the Permanent Mission of the Czech Republic to the International Atomic Energy Agency regarding Certain Member States' Guidelines for Transfers of Nuclear-related Dual-use Equipment, Materials, Software and Related Technology," INFCIRC/254/Rev.9/Part 2, November 13, 2013, <http://www.iaea.org/sites/default/files/publications/documents/infcircs/1978/infcirc254r9p2.pdf>.
28. It should be noted that the exact dependence of thermally excited UF_6 molecules on temperature is uncertain. If an undesirable number remain vibrationally excited at a temperature intended for irradiation, then selectivity will be adversely affected. One option may be to shift the laser wavelength to selectively excite dimers at a gas temperature where a significant fraction of both monomers and dimers exist. There are advantages and disadvantages to exciting monomers versus dimers, but choosing to excite both may still be more energy efficient than centrifuges. Most importantly for a proliferator, it may be a significant advantage in the separative performance of a third-generation system.
29. From L. D. Landau and E. M. Lifshitz, *Fluid Mechanics, 2nd edition* (Butterworth Heine-
mann, 1987), the expression $T_2/T_1 = \{2\gamma M_1^2 - (\gamma - 1)\} \{(\gamma - 1)M_1^2 + 2\} / (\gamma + 1)^2 M_1^2$ for a gas experiencing a shock compression from a polytropic process on p. 335 was used to calculate T_2/T_1 , the ratio of the temperature after the shock T_2 to that before the shock T_1 . For a Mach number M of 2, with $\gamma = 1.33$ for SF_6 as the carrier gas G, T_2/T_1 was calculated to be 1.57, or T_2 is 57 percent higher than T_1 . For a shock experienced around $T_1 = 100$ K, the temperature T_2 after the shock would be 157 K.
30. Eerkens, "Laser-induced Migration and Isotope Separation of Epi-Thermal Monomers and Dimers in Supercooled Free Jets," 225–253.
31. *Ibid.*, 246.
32. Eerkens, "Separation of Isotopes by Laser-Assisted Retardation of Condensation (SILARC)," 308.
33. Email communication with Jeff Eerkens.
34. Eerkens, "Laser-Induced Migration and Isotope Separation of Epi-Thermal Monomers and Dimers in Supercooled Jets," 246.
35. T. C. Adamson and J. A. Nichols, "On the Structure of Jets from Highly Under Expanded Nozzles Into Still Air," *Journal of Aerospace Sciences* 23 (1959).

36. H. W. Liepmann and A. E. Puckett, *Introduction to Aerodynamics of a Compressible Fluid* (John Wiley & Sons, 1947).
37. Ibid.
38. J. W. Eerkens, "Equilibrium Dimer Concentrations in Gases and Gas Mixtures," *Chem Physics* 269 (2001):189–241.
39. From J. J. Katz and E. Rabinowitch, *The Chemistry of Uranium* (Dover, 1951), the vapor pressure for solid UF₆ was calculated using the expression $\ln p_e(T) = -132.87 - \frac{344,490}{T^2} + \frac{707.31}{T} + 26.436 \ln T - 0.0387T$ on pg. 407 for $T = 100, 45, \text{ and } 25 \text{ K}$.
40. Eerkens, "Laser-Induced Migration and Isotope Separation of Epi-Thermal Monomers and Dimers in Supercooled Free Jets," 251.
41. R. J. Jensen et al., "Separating Isotopes with Lasers," *Los Alamos Science* 4 (1982).
42. Eerkens and Kim, "Isotope Separation by Selective Laser-Assisted Repression of Condensation in Supersonic Free Jets," 2333.
43. A range for β between 2 and 20 was claimed by Michael Goldsworthy, CEO of Silex Systems Ltd., see *Reference: Developing Australia's non-fossil fuel energy industry: Hearing before the Standing Committee on Industry and Resources, Commonwealth of Australia House of Representatives* (2006) (statement of Michael Goldsworthy, Chief Executive Officer, Silex Systems Ltd., Sydney, NSW, Australia), <http://www.aph.gov.au/binaries/hansard/reps/commtee/r9041.pdf>.
44. Example of enrichment factor values for gas centrifuges are given in: Alexander Glaser, "Characteristics of the Gas Centrifuge for Uranium Enrichment and Their Relevance for Nuclear Weapon Proliferation," *Science & Global Security* 16 (2008): 1–25. Values for gaseous diffusion are given in: Sébastien Philippe, Alexander Glaser, "Nuclear Archaeology for Gaseous Diffusion Enrichment Plants," *Science & Global Security* 22 (2014): 27–49.
45. Lyman, "Enrichment Separative Capacity for SILEX," 5.
46. Ibid.
47. The IAEA defines a significant quantity as 25 kg of uranium-235 included in HEU enriched to a level of 20% or greater. For 90% HEU, a significant quantity would be $\sim 27.8 \text{ kg}$.
48. Based on density of natural uranium = 19.1 g/cm^3
49. Based on density of UF₆ vapor, $\rho_{\text{UF}_6} = 4.291 \times 10^2 (P/T)(1 + 1.2328 \times 10^4 P/T^3) \text{ g/cm}^3$ where P is in kPa. In reservoir, $P = 57.57 \text{ torr} = 7.67 \text{ kPa}$ and $T = 300 \text{ K}$. From IAEA, *Interim Guidance on the Safe Transport of Uranium Hexafluoride*, IAEA-TECDOC-608, June 1991.
50. The product cuts θ in the author's model are slightly higher than in the ones provided by Jeff Eerkens that were used when calculating the separative capacity and required space for SILEX. The author used the definition of β in Equation 13, and this provides higher cuts when the carrier gas pressure $p_G = 0.002 \text{ torr}$ at the skimmer. The use of $p_G = 0.003 \text{ torr}$ gives $\theta \sim 0.25$, which is closer to Eerkens' model when $p_G = 0.002 \text{ torr}$. The use of Eerkens' model as opposed to the author's in this regard requires SILEX to occupy a larger space and use energy less efficiently. The real performance characteristics, however, need to be measured experimentally to clear up what p_G at the skimmer allows for the desired θ .
51. A. Glaser, "Characteristics of the Gas Centrifuge for Uranium Enrichment and Their Relevance for Nuclear Weapon Proliferation (corrected)" *Science & Global Security* 16 (2008): 1–25.
52. "Iran develops new generation of centrifuges: AEOI chief," PressTV, September 10, 2014, <http://www.presstv.ir/detail/2014/08/27/376767/iran-develops-ir8-centrifuges/>
53. "Uranium Enrichment," World Nuclear Association, last modified in September 2015, <http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Conversion-Enrichment-and-Fabrication/Uranium-Enrichment/>.
54. P. Rabinowitz et al., "Efficient Tunable H₂ Raman Laser," *Appl. Phys. Lett.* 35 (1979).
55. G. Yang et al., "High Power Repetitive TEA CO₂ Pulse Laser," *Laser Physics* 22 (2012): 1173–1176.

56. V. P. Singal et al., “A Highly Efficient Electron Beam Controlled Switchless Multi-joule TEA CO₂ Laser,” *Infrared Physics and Technology* 44 (2003): 69–73.
57. “Two-stage vacuum pump DV0076B-DA,” Dekker Vacuum Technologies, Inc., <http://www.dekkervacuum.com/>. This is a two-stage pump which can be used for both pumping down to the reservoir pressure of 57.6 torr and the much lower irradiation chamber pressure of 0.005 torr. There is a .pdf document with its performance characteristics that shows a suction capacity of 68 m³/hr near atmospheric pressure and this was assumed to be the average suction capacity in pumping down to the reservoir pressure of 57.6 torr.
58. This real driving power is that required in an isothermal process divided by a thermal efficiency of 0.25. The isothermal power $P_{is} = P_a \cdot S \cdot \ln(P_b/P_a)$, where P_a and P_b are the compression and suction pressure in Pascals, and S is the suction capacity in m³/hr. The energy in kWh needed for a one-meter long separation unit was calculated and divided by the annual separative capacity for such a unit, 257.8 kg · SWU/yr.
59. Same calculation as in the previous note. The only difference is that the suction capacity was lowered to 0.5 ft³, as the irradiation chamber is being pumped down to 0.005 torr. This takes less than an hour with 35.25 m of separation unit length. This would not need to happen continuously during operation, but the energy consumption is so small that it can be neglected. Again, the calculated isothermal power P_{is} was divided by a thermal efficiency of 0.25.
60. For a polytropic process where the volume of a gas is compressed increasing both the pressure and temperature, the work done on a gas when compressing from volume and pressure V_1 and p_1 to volume and pressure V_2 and p_2 is $W = \int_{V_1}^{V_2} p dV = \frac{p_1 V_1^n}{1-n} (V_2^{1-n} - V_1^{1-n})$. The polytropic index n is determined from the requirement that pV^n is unchanged after compression. For a one-meter long separation unit, the work done on the gas to compress a constant flow from 0.002 torr to 57.57 torr is only 0.011 kWh over a duration of 1 hour, or 96.36 kWh over a single year. Given that this is for a unit with a separative capacity of 257.8 kg · SWU/yr, energy is consumed at a rate of 0.37 kWh/SWU.
61. It is widely reported that a U. S. grocery store consumes about 50 kWh of electricity per square foot annually. Available at: “Buildings Energy Data Book,” U. S. Department of Energy, last updated March 2012, <http://buildingsdatabook.eren.doe.gov/CBECS.aspx>.