

## Current Status of the MLIS Uranium Enrichment Process

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### 1. Introduction

Well-established energy security is extremely important for the national economy and future prosperity of the country. Many countries are trying to develop and use renewable energy sources such as solar, wind, and tidal power to keep the natural environment clean and safe. Although some have disputed calling nuclear power (which produces ~40% of domestic electricity) "green energy", it deserves credit for this label because of its low CO<sub>2</sub> emission and fuel efficiency. While fuel costs in nuclear power generation are less than 30% and uranium enrichment is only ~40% of front-end fuel loop, uranium enrichment is a critical step in nuclear power generation. It is also politically sensitive worldwide due to potential proliferation aspects. Currently, gaseous diffusion and centrifuge are the technologies for uranium enrichment. Since the 70s, many countries have been trying to develop a more advanced and economic technology after the gaseous diffusion process started to lose its economic viability. Among the developed laser-assisted technologies, MLIS (Molecular Laser Isotope Separation) is still under investigation for industrial application while most other laser uranium enrichment technologies have been terminated. In this regard it is worthwhile to review the MLIS processes being developed currently, SILEX (Separation of Isotopes by Laser EXcitation) and CRISLA (Condensation Repression by Isotope Selective Laser Activation). Recently the Canadian company Cameco joined the GE-Hitachi venture by paying \$123.8M for a 24% holding, and GE-Hitachi-Cameco is now running the GLE (Global Laser Enrichment) facility using SILEX technology in Wilmington, North Carolina to evaluate commercialization of the process.

### 2. Brief Reviews of MLIS

#### 2.1 MPD (Multiple Photo-Dissociation)

After lasers had been developed in the 1960s, there was a strong belief that laser technology could replace the old and inefficient gaseous diffusion process for uranium enrichment. Two different laser methods, AVLIS (Atomic Vapor Laser Isotope Separation) and MLIS have been developed, with the former using the vapor of uranium and the latter using gaseous uranium hexafluoride (UF<sub>6</sub>) as an initial feed material. For AVLIS, atomic vapor produced by an electron gun is isotope-selectively ionized by 2-colors or 3-colors laser irradiation. Selected U-235 ions are collected by a

positively charged collector. For MLIS, selective dissociation of UF<sub>6</sub> via  $^{235}\text{UF}_6 + \text{nhv} \rightarrow ^{235}\text{UF}_5 + \text{F}$  using intense pulsed 16 $\mu\text{m}$  irradiation was the most popular process among various MLIS schemes. Since no fundamental 16 $\mu\text{m}$  laser was available, 16 $\mu\text{m}$  IR (Infra-Red) photons were produced by Raman conversion in optically pumped para-H<sub>2</sub> gas using a pulsed CO<sub>2</sub> laser. Reactant CH<sub>4</sub> was usually added to scavenge F atoms and inhibit the back reaction of UF<sub>5</sub> to UF<sub>6</sub>. This particular MLIS scheme was often called the MPD (Multi-Photon Dissociation) process. Since the isotope shift  $\Delta\nu_3$  between  $^{235}\text{UF}_6$  and  $^{238}\text{UF}_6$  is only 0.6 cm<sup>-1</sup>, it is necessary to cool UF<sub>6</sub> gas to a sufficiently low temperature to shrink and separate the overlapping  $\nu_3$  vibrational bands located around 16 $\mu\text{m}$  (626 cm<sup>-1</sup>). To achieve this, a jet of UF<sub>6</sub> gas diluted in an Ar buffer gas was supercooled by supersonic expansion through a nozzle into a vacuum chamber and cross-irradiated with 16 $\mu\text{m}$  laser radiation. It was found that low-intensity two-photon resonant IR excitation followed by high-intensity red-shifted laser irradiation gave the highest selectivity. In some MPD experiments, UV irradiation was used to enhance dissociation efficiency [1].

#### 2.2 Background Information of SILEX

Unfortunately both the AVLIS and MPD uranium enrichment processes were not industrialized. The latter was terminated in the mid-80s in the US and in the mid-90s by other countries including Japan, Russia, France, and South-Africa. This was due to inefficient laser excitation, complicated expensive chemistry, and problems with low pulse repetition rates of 16 $\mu\text{m}$  lasers. The AVLIS project was continued until the 90s in the US. However USEC (United States Enrichment Corporation) halted AVLIS in favor of SILEX developed by an Australian company in 1999. Currently no technical information except some media announcements are available since SILEX technology is classified under the protection of the US and Australian governments. However, a report written by John L. Lyman, former scientist from LANL (Los Alamos National Laboratory), evaluates the uranium separative capacity of SILEX [2]. It contains some procedural factors such as the process materials, laser wavelength and specification, nature of photon-matter interaction, and current handicaps of the process to overcome, etc. The present paper is focused on the evaluation of some technical aspects of SILEX by looking into those factors and the historical background from publications available in the open literature.

### 2.3 Laser Assisted Condensation Repression Method

Besides MPD's vicissitudes, experiments in another MLIS scheme which uses laser-induced dissociation of VanderWaals molecules generated in supersonically nozzle-expanded free jets have shown some successes. The concept of this scheme originated with Becker's pre-laser "Nozzle Process" of 1957, which used radial diffusion and mass difference effects that cause  $^{235}\text{UF}_6$  to flee out of the core of a free supersonic jet slightly faster than  $^{238}\text{UF}_6$ . After the advent of the laser and the discovery that hexafluoride gases form, sooner than expected, dimers in free jets due to VanderWaals forces, Y.T. Lee (1986 Nobel Laureate) at LANL suggested using photo-dissociation of these dimers by isotope-selective laser excitation [3]. Since the mass of non-excited dimerized  $^{238}\text{UF}_6\text{:UF}_6$  (or G = carrier) and laser-maintained  $^{235}\text{UF}_6$  monomers now differ by  $\Delta M = 355$  instead of by  $\Delta M = 3$  as in Becker's scheme, radial escape of  $^{235}\text{UF}_6$  from a free-jet core is greatly enhanced. K. C. Kim et. al. of LANL first confirmed the early onset of  $\text{UF}_6$  dimer formation in supersonic expansions [4] which had already been observed earlier for supersonic expansions of  $\text{SF}_6$  and other gases. Because VanderWaals binding energies ( $\sim 100 \text{ cm}^{-1}$ ) in dimers are much smaller than the energy of molecular vibration ( $\sim 1000 \text{ cm}^{-1}$ ), absorption of one IR laser photon by a dimer is all that is needed to cause a near-instant dimer break-up according to molecular pre-dissociation physics. The first experimental proof that laser suppression of dimer formation can be used to separate  $\text{SF}_6$  isotopes was carried out by H VandenBergh at the Ecole Polytechnique in Lausanne, Switzerland [5]. He observed the separation of  $^i\text{SF}_6$  isotopomers using selective  $\text{CO}_2$  laser excitation of supersonically expanded  $\text{SF}_6/\text{Ar}$  mixtures in a free jet. The importance of VandenBergh's experiments was not immediately recognized by most MLIS researchers because at the time it appeared that MPD would be a winning MLIS technique. However after subsequent disappointments with MPD, condensation repression schemes were vigorously investigated by scientists in the CRISLA/US program and presumably by SILEX/Australia. CRISLA developed by Jeff W. Eerkens [6] from the University of Missouri-Columbia appears to be similar to SILEX and to have the same historical background. Technical aspects of the discussed MLIS processes are briefly compared in the next section.

### 3. SILEX vs. CRISLA

Although not completely confirmed for SILEX, based on Lyman's report, it appears to use the nozzle expansion and dimer condensation concept. Non-excited heavier dimers ( $^{238}\text{UF}_6\text{:UF}_6$  or  $^{238}\text{UF}_6\text{:G}$ ) tend to stay in the core of the expanding jet while selectively excited  $^{235}\text{UF}_6$  monomers migrate outwards of the supersonic jet into the subsonic background or "rim" gases. The  $^{235}\text{UF}_6$  acquire some additional kinetic energy during (pre-) dissociation events when laser-deposited vibrational energy in  $^{235}\text{UF}_6\text{:G}$  dimers is

liberated and converted into translational recoil energy that slightly enhances out-of-the-jet-core escape rates of  $^{235}\text{UF}_6$  [6]. This driving force is in addition to mass-dependent differential radial pressure diffusion mentioned before. For SILEX, Lyman repeatedly mentions that the repetition rate of the 16 $\mu\text{m}$  laser is the main problem lowering process efficiency. Fig.1 and Table 1 shows the simplified diagram and some operating factors/conditions of the processes available from the open literature.

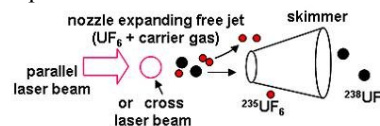


Figure 1. Simplified schematic diagram of the processes

Table 1. SILEX vs. CRISLA comparison [2,6]

Process Factors	SILEX	CRISLA
Laser type	Pulsed $\text{CO}_2$ Raman laser	N/A
Laser efficiency	$\sim 0.2\%$	$\sim 5\%$
Wavelength	$\sim 16\mu\text{m}$	N/A
Irradiation power	$\sim 12\text{W}$	$> 1.0\text{kW}$
Pulse rep rate	$\sim 300\text{Hz}$	CW
Feed/Product/Tail	$\text{UF}_6/\text{UF}_6/\text{UF}_6$	$\text{UF}_6/\text{UF}_6/\text{UF}_6$
Process pressure	classified	N/A
Enrich. Factor	2.0 ~ 20	$> 2.0$

### 4. Conclusions

While many aspects of SILEX have not been released to the public, there are many indications that the basic concept is similar to CRISLA, except for laser type and excitation technique. It is too early yet to tell which process is more economical and non-proliferation-proof. SILEX representatives have stated that if pilot runs are successful, they can supply the fuel for power plants at half the price of that from 2<sup>nd</sup> generation centrifuges. It may not be long before it will be announced if SILEX is commercially viable. In the meantime it is possible to assess the potential of condensation repression MLIS by evaluating CRISLA publications. Even if little more will be disclosed about SILEX, it may be possible that CRISLA can overcome some of the major problems of SILEX pointed out by Lyman by using a different IR laser excitation scheme.

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