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Laser Enhanced Microwave Plasma Isotope Separation

Progress Report

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ABSTRACT

We are developing an innovative method of isotope enrichment using laser enhanced microwave/electron cyclotron resonance (ECR) discharges. Experimental research is focusing on laser excitation of a low abundance isotope and then ionization and separation using a microwave/ECR discharge at 2.45 GHz. The first six months of the grant period have focused on modifying the plans for the ion source to include accessibility for laser excitation. Construction of the ion source has begun and is close to completion. During the next six months we plan to test the performance of the ion source. During the next fiscal year we plan to purchase a dye amplifier which will be pumped by our copper vapor laser. The dye laser, tuned to a wavelength of 670.8 nm, will excite ^6Li which will then be preferentially ionized by the ECR source and collected with a charged grid. The degree of enrichment will be determined using thermal ionization mass spectrometry. The Dept. of Geology is currently calibrating their mass spectrometry system for $^6\text{Li}/^7\text{Li}$ ratios. The final objective of this project will be to assess the feasibility of this system to large-scale production of stable isotopes.

INTRODUCTION AND BACKGROUND

Enriched stable isotopes are used for the fabrication of nuclear targets in both basic and applied research as well as for industrial applications. The demand for these isotopes continues to expand particularly with the growth of the medical field.¹ Calutrons have traditionally been used to manufacture the needed enriched stable isotopes. A calutron is a very energy intensive isotope separation method however. Several other isotope separation techniques have been proposed to help meet the demand for enriched isotopes at a lower energy cost than calutrons.¹

For the past 15 to 20 years several plasma based methods have been developed to address the problems of the current isotope separation technology.² They include centrifuge techniques from rotating plasmas, atomic vapor laser isotope separation techniques (AVLIS), selective ion cyclotron resonance methods, ion waves and gas discharges.² We are proposing to study an innovative new method based in part on some of these methods as well as technology developed for non-nuclear applications. Selective laser excitation will be used in conjunction with microwave plasma ion sources. This method combines a laser selectivity method similar to AVLIS with an efficient microwave/electron cyclotron resonance source used for extraction.

Atomic Vapor Laser Isotope Separation techniques (AVLIS) use laser light (typically from a tunable dye laser) set at a very precise wavelength to photo-ionize a particular isotope, leaving the remaining isotopes unaffected. A charge collection system is used to extract the charged isotopes. A typical method is to use one laser pulse to excite the selected isotope to a long lived state and then use a second laser wavelength to ionize the isotope.^{3,4} The reason for the two (or three) photon method is that wavelengths needed for single photon ionization are in the ultraviolet region of the spectrum. It is far more difficult to find efficient tunable

lasers in the ultraviolet compared to the visible lasers used for the two photon technique.

Recent plasma/laser techniques have been developed to combine both the selective ionization of lasers and the extraction efficiency of plasmas.⁵⁻⁸ A tunable laser is used to selectively *excite* the neutral isotopes of interest in a positive column glow region of a d.c. discharge. These excited isotopes are preferentially ionized by the discharge because it takes less energy for the free electrons to ionize excited atoms compared to ground state atoms. These ions of the selected isotope are attracted towards the cathode where they are deposited. In a similar technique, a sheath forms at the walls of the discharge chamber which enhances diffusion of the ions to the walls, where the isotopes are then collected and removed. A theoretical treatment of the method estimates a 10,000% enrichment of isotopes of hydrogen.⁵

There are some limitations to this method due to charge exchange effects between the ions of the isotopes to be enriched and the neutral abundant isotope. This can be minimized by keeping the radius of the discharge tube small compared to the collisional mean free path⁹ and by controlling the electric and, when available, external magnetic field.⁴ Another problem is that resonance exchange collisions (transferring the energy from the excited scarce isotope to the ground state abundant isotope) can also occur. This effect can be reduced by keeping the overall pressure low and in particular by keeping the concentration of the isotope to be enriched low compared to the abundant isotope.⁵ Plasma sources other than d.c. glow discharge may be able to more easily overcome these problems. One possibility is a microwave plasma source with electron cyclotron resonance regions.

Electron cyclotron resonant (ECR) microwave plasma sources can have many configurations.¹⁰ In general, microwave energy is coupled to the natural resonant

frequency of an electron in the presence of a static magnetic field. This resonance occurs when the electron cyclotron frequency defined as $\omega_{ce} = eB/m$ (where e and m are the electron charge and mass and B is the magnetic field strength) equals the microwave frequency used to generate the plasma. The electrons accelerate as they gyrate around the magnetic field lines in the ECR region and subsequently ionize and excite the neutral gas. These systems operate at low pressures, 10^{-3} to 10^{-1} Torr and produce an "almost collisionless plasma that can be varied from weakly to highly ionized state by changing discharge pressure, gas flowrates and input microwave power".¹⁰ These inexpensive electrodeless systems are finding a wide variety of applications in the plasma processing arena ranging from depositing thin films to etching semiconductors.

These ECR microwave plasmas have several attractive features for isotope separation techniques. One is that they can be very efficient ion sources. For example, Hofer and Reist¹¹ have used such a source in an apparatus to separate ^{123}I isotopes by using an ECR system to remove xenon after the reaction $^{127}\text{I}(p,5n)^{123}\text{Xe}$ has occurred in an accelerator. Total efficiencies of 40 - 50% removal have been achieved for powers as low as a few tens of Watts. Also microwave sources can be very efficient, (greater than 99%), at delivering microwave power to the gas, depending upon source configuration.¹⁰

Another advantage of microwave ECR sources is that the electron and ion energy distribution can be controlled via input parameters such as power and pressure.¹²⁻¹⁴ It would be advantageous to tailor the electron energy distribution function to preferentially ionize the pre-excited scarce isotopes over ground or low energy atoms. Thus, pre-excitation of a select isotope with a laser would have a greater probability of achieving ionization over atoms which have not been pre-excited and which would require a larger energy transfer to be ionized.

ECR microwave plasmas operate at a fairly low pressure range of 10^{-3} to 10^{-1} Torr. This is the pressure range suggested by Silfvast⁵ to minimize charge and resonance exchange collisions. The collisional mean free paths in these discharges are large and hence these sources can be considered "collisionless".¹⁰

Another advantage to microwave plasmas in general is that they can easily vaporize many solids as well as ionize most gases. We have recently¹⁵ shown that microwave plasmas can easily heat and vaporize even high temperature metal compounds such as copper chloride. Thus many different types of compounds and gases could be used without pre-heating or pre-vaporization. In addition to the high efficiencies of ECR microwave sources, the technology is well established and relatively inexpensive, particularly at 2.45 GHz, the microwave oven frequency.

We had planned on using our Asmussen resonant cavity retro-fitted with rare earth magnets to produce an ECR region at one end of the cavity. We have decided to use a smaller, compact ERC source based upon a similar design instead.²⁴ A description of this source is given in the following section. A copper laser-pumped dye laser will be used to excite select isotopes we plan to separate. Thermal ionization mass spectrometry of our collected samples will determine the degree of enrichment. See the following section for progress.

DESCRIPTION OF PROGRESS

Our research plan (as described in the original research proposal) was broken up into three years. During the first year we had planned on modifying an existing microwave resonant cavity by adding rare earth magnets to provide an electron cyclotron resonance region as well as add a processing vacuum chamber. We decided to build a completely new ion source which will be described below.

The design of the ion source which will be used to ionize samples of lithium is based upon a device of L. Mahoney and J. Asmussen.²⁴ This ion source is very

similar in design to our resonant cavity, see Figure 1. The microwave cavity is designed to excited various electromagnetic modes at 2.45 GHz depending upon the length of the cavity and the position of the co-axial probe which couples the microwaves from the source to the cavity. The microwave resonance cavity ion source can produce vaporization, heating and ionization of lithium samples.

In addition to some design considerations like placement of the permanent magnets, the major difference between the old cavity and the new source is that the new source is 3.5 inches in diameter whereas the old cavity was 8.9 inches in diameter. The advantages of the new source include:

- smaller plasma volume - this means that the laser excitation volume to overall plasma volume will be much larger than in the previous system. This will improve the total efficiency of the system.
- this source is based upon an 8 inch conflat vacuum flange, making it very compatible with conventional vacuum systems. This is important for future adaptability of this system to a production setting.
- this ion source has a long working lifetime, requires little maintenance and produces high beam current densities (5 - 10 mA/cm²).²⁴

A drawing of the ion source²⁴ is shown in Figure 1. Our major modification has been to move the gear assembly which controls the sliding short to an off axis position. This means that two gears and a ball bearing assembly are now required instead of the one gear. This allows for a hole to be drilled in the center of the sliding short and end wall of the cavity. We plan to shine the laser down the center axis of the cavity rather than at an oblique angle from below (as described in the original proposal). See Figure 2 for a schematic of the isotope enrichment system. Figure 3 shows a photograph of the ion source to date. We anticipate that it should be complete in about four weeks. The ion source will be attached to the vacuum processing chamber shown in Figure 4. The ion source will be attached to the side

of the vacuum chamber so that sputtered material will fall into the vacuum pumps rather than onto the collection grid.

Additional progress includes the following: We have purchased and assembled the vacuum chamber, pumps and vacuum components. We are in the process of leak checking this system. Calibration studies for lithium have been initiated in the Radiogenic Isotope Geochemistry Laboratory in the Geological Sciences Department.

During the next year of our project we plan to excite the lithium samples with a copper vapor pumped dye laser. We plan to purchase a Lambda Physik FL3001E one stage amplifier which is designed to be pumped by our Oxford copper laser (output power 10 Watts). This dye laser can be pulsed with the same repetition rates as the copper vapor laser, i.e. 2 to 15 kHz. The dye laser is 6% efficient at 6700Å resulting in a total average output power of 600 milliwatts. The advantage of using a copper vapor laser to pump the dye laser is the capability for high repetition rates compared to excimer pumped dye lasers (in the tens of Hertz range). These high repetition rates will allow for increased efficiency over the excimer pumped dye lasers.

The dye laser will provide selective ionization of ^6Li . Samples both with and without the laser excitation will be taken and analyzed for $^6\text{Li}/^7\text{Li}$ ratio. The performance of the plasma system will be diagnosed with electrostatic probes, laser induced fluorescence, gridded energy analyzer, and optical emission spectroscopy.

Cylindrical shell, (2) sliding short, (3) stainless-steel base plate, (4) adjustable coaxial input probe, (5) finger stock, (6) screened window, (7) assembly screws, (8) water cooling lines, (9) silicone O-ring, (10) fused quartz cap, (11) gas input channel, (12) annular gas channel, (13) gas pinholes, (14) rare-earth magnets and iron keeper, (15) brass cap for magnets, (16) air cooling input channel, (17) air cooling pinholes.

Figure 1. Drawing of the compact ECR microwave-cavity ion source, taken from Rev. Sci. Instrum 61(1), 285, 1990

Figure 2. Schematic of the laser/ECR plasma isotope separation technique.

Figure 3. Photograph of ion source to date. Diameter of the chamber is 3.5 inches.

Figure 4. Photograph of vacuum processing chamber. Diameter of stainless steel chamber is eight inches.

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