PHOTO-INDUCED CATAPHORETIC ISOTOPE SEPARATION

Progress Report

for Period June 15, 1976 - June 14, 1977

A D I T I O N

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and will be used to show to the public as a demonstration of the progress of the research and development work. The results of the work will be published in the open literature and will be available to the public. The work was performed by J. A. Carruthers, L. M. Chanin and H. J. Oskam.

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Abstract

The studies of the feasibility of radiation-induced cataphoretic segregation of isotopes were started on June 15, 1976 under ERDA Contract E(11-1)2961. The results obtained during the initial eight months period are summarized. More detailed feasibility calculations showed that significant isotope separation efficiencies can be obtained for discharges at low pressures. This may make it desirable to use a buffer gas in the discharge in order to manipulate the electron energy distribution. Requirements for the properties of the buffer gas have been established. The experimental system for studying laser-induced cataphoretic separation of neon isotopes has been completed. Preliminary studies showed that the system is capable of detecting less than 1% change in the $\text{Ne}^{20}/\text{Ne}^{22}$ isotope ratio. It is anticipated that after about three months a systematic study can be made with respect to the photo-induced isotope separation efficiency using this method. The other experimental system relates to the separation of mercury isotopes in a direct current gaseous discharge using radiation from external monoisotopic mercury light sources. Initial experimental feasibility studies of this technique are expected to be started after about three months. The ultra-high vacuum and gas handling system has been completed.

I. Introduction

The studies of the feasibility of radiation - induced cataphoretic segregation of isotopes were started on June 15, 1976 under Contract No. E(11-1)2961. This progress report summarizes the results obtained during the initial eight month period, while the results expected to be achieved during the remainder of the first year are mentioned.

Section II describes the theoretical studies, while sections III and IV summarize the experimental efforts. The percentage of time which the principal investigators have devoted to the project since the beginning of the contract (projected through the remainder of the first year) are given in section V.

II. Theory of Photo-Induced Cataphoretic Isotope Separation

The term cataphoresis refers to the relative enhancement in the negative electrode region (cathode region) of a direct current discharge of the gas component having the largest ionization probability. This process has successfully been used for the separation of different atoms and molecules and is believed to be also applicable as an isotope separation method.

If the isotope shift is such that the absorption of radiation for one isotope of an atom or molecule occurs at frequencies where the other isotopes are more transparent, one can selectively excite that species with radiation of a sufficiently narrow line width. This isotope then will be ionized preferentially by electrons and/or metastable particles in the direct current discharge. The ionic isotope moves to the cathode and is neutralized there. The resulting degree of isotope separation can be calculated if the relevant reaction rate constants for the collision processes involved are known.

Feasibility calculations of photo-induced cataphoretic isotope separation were presented in the proposal which led to the funding of the studies described in the present progress report. During the period covered by this report additional calculations were made and new insights in the experimental conditions needed for achieving a significant isotope separation efficiency were obtained. They are summarized in this section and can be divided in the following two parts:

2.1 More accurate feasibility calculations

In the original calculations the production of the excited and ionized state of the desired isotope by excitation and charge

transfer from the other isotopes was neglected since it was believed that this would lead to a lower limit of the isotope separation efficiency. When these processes are included four coupled equations are obtained. The solution of these equations shows that the ionization degree of the desired isotope is indeed underestimated. However, the same holds for the other isotopes and the ratio of the relevant ionization degrees is more than sufficient for isotope separation applications but smaller than expected.

The recent calculations also show that for sufficient intensity of selective radiation the limiting factor is charge exchange between the desired isotopic ions and the other neutral isotopes. This seems to limit the number density of the isotopes to be separated and may require the use of a buffer gas in the discharge. This was anticipated and included in the original calculations.

2.2. The influence of the gaseous environment

The recent feasibility calculations indicate that, even when the ionization degree of the desired isotope is sufficient, charge exchange processes may limit the degree of isotope enhancement. If the required density leads to the need to produce discharges in very low pressure discharges other complications, such as high electron temperatures, may appear. A simple solution to this possible problem is the use of a buffer gas.

A study of the interaction of a buffer gas with the isotope gas shows that the buffer gas has to satisfy the requirement that cataphoretic segregation of the isotope gas (independent of the isotope) should not occur in the resulting gas mixture. If this happens, this effect would most probably mask the isotope separation

due to selective isotope excitation. The buffer gas to be used in the studies of mercury-isotope separation (section III) is xenon. In a xenon-mercury mixture the metastable xenon atom can not ionize a ground state mercury atom. However, the selectively excited isotope will be ionized by this metastable atom. Therefore, xenon is the proper buffer gas for the mercury studies. The same holds for the helium-neon studies at low pressure (section IV).

During the remainder of the first year of the studies the effect of variations in buffer gas pressures, etc. on isotope enrichment efficiencies will be investigated.

III. Mercury Studies

The original proposal suggested that initial studies to test the feasibility of the isotope separation technique be undertaken using Hg as well as parallel investigations involving Ne. To perform the Hg measurements a high vacuum gas handling system has been designed and is nearing the final stages of assembly. The system includes a mechanical forepump, high vacuum diffusion pump, appropriate valves, ionization and thermocouple gauges and control panels. The system will be capable of high temperature bakeout at about 400°C and should readily achieve the vacuum conditions necessary for the studies. The initial attempt to detect isotopic enrichment in Hg will involve using radiation from monoisotopic Hg resonance lamps for selective excitation of the mercury isotopes in the discharge. The lamps will use Xe as a buffer gas and the mercury isotope Hg.¹⁹⁹ Figure 1 shows a cross sectional view of the proposed geometry. Initially, measurements will be made using a single resonance lamp to determine appropriate operating conditions. To provide a high uniform level of irradiation of the discharge from the Hg resonance lamps a series of Hg discharge tubes may be used to surround the central cataphoresis discharge tube. Various discharge parameters such as current and pressure of both the Hg lamps and the discharge tube will be adjusted to achieve optimum conditions. The initial tests to detect Hg isotopic separation will involve observations of the fluorescent radiation at various positions along the discharge tube using fiber optic methods. Later studies using mass analysis to detect isotopic enrichment will be undertaken on the same high vacuum gas handling system.

Within the next three months we hope to complete the vacuum station, the discharge tube and to obtain initial data concerning the Hg isotope enrichment.

Discharge Tube (DT): Xe-Hg (natural) mixture
24 mm O.D. ultra-violet glass

Resonance Lamp (RL): Xe-Hg¹⁹⁹ mixture
6 mm O.D. ultra-violet glass

Figure 1 Proposed Resonance Lamp and Discharge Tube Configuration
for Photochemical Separation of Hg¹⁹⁹.

IV. Neon Studies

The studies are designed to explore laser-induced cataphoretic separation of the Ne isotopes using a He-Ne 6328A° laser. The laser serves as the radiation source for selectively exciting one isotope. Two discharge tubes are within the laser cavity, a laser gain tube with a $\text{He}^3\text{-Ne}^{20}$ gas mixture, and a cataphoresis tube with a $\text{He}^3\text{-Ne}^{20}\text{-Ne}^{22}$ mixture.

The approach depends on using the laser light to significantly alter the population of the $3S_2$ level, - the upper level of the 6328 A° transition, - for the Ne^{20} isotope in the cataphoresis tube, and therefore affect the ionization rate for that isotope. Since the population of the $3S_2$ level of the Ne^{22} isotope should not be appreciably affected by the laser, a means is provided for obtaining a differential ionization rate between the Ne^{20} and Ne^{22} isotopes. One then depends on cataphoretic action to separate the isotopes.

By using a laser gain tube separate from the cataphoresis tube we should be able to work over a wide range of pressures and mixtures in the cataphoresis tube, with the objective of optimizing the conditions for cataphoretic separation.

A schematic of the laser cavity, the diagnostic apparatus, etc., is shown in Fig. 2. The general arrangement now appears to be finalized, although some changes in the details of the laser cavity, the cataphoresis tube, and in the confocal Fabry-Perot spectrometer are anticipated. The system is capable of detecting less than 1% change in the $\text{Ne}^{20}/\text{Ne}^{22}$ ratio.

The pump and filling station was assembled from an existing system, with a Granville-Phillips differential capacitive manometer

and calibrated leak valves added. Both the laser and cataphoresis tubes are attached to the pumping station, and are replaced as the need arises. The laser cavity is about 1.2 m long, and uses high-reflectivity mirrors of 1.47 and 10.0 meters radii of curvature. The laser operates with high-order off-axial modes, and the cavity is designed to fill the bore of the cataphoresis tube as completely as possible. Some further improvement in the laser cavity are anticipated, as discussed below.

The shape of the cataphoresis tube has evolved considerably. At the present time it is about 50 cm long, with a bore diameter of 6 mm, and is filled to about 0.8 Torr with a $\text{Ne}^3\text{-Ne}^{20}\text{-Ne}^{22}$ mixture of 10-1-1. Very sensitive observation of the spontaneous fluorescence is made using a side tube of about 3 inches in length, which is in the discharge path but not in the path of the laser beam. Thus we are able to observe the relative fluorescent intensity of the Ne^{20} and Ne^{22} isotopes near one end of the discharge, and to note any change which occurs when the laser beam is interrupted. The fluorescence sampling tube produces a relatively intense fluorescent beam of small source area which is fed through a $6328 \text{ \AA}$ spike filter to the wide-angle confocal spectrometer. The spectrometer is scanned repetitively through its free spectral range, and the double-isotope fluorescent line displayed on an oscilloscope. A small AC scan at 90 Hz, with narrow-band signal amplification and synchronous detection permits the slope of the line-shape to be simultaneously traced out on a strip chart recorder. This slope tracing is very sensitive to small changes in the isotope ratio, and direct calibration by adding a known amount of one isotope shows that a 1% change in the isotope ratio is easily observable.

The isotope splitting at $6328 \text{ \AA}$ for Ne^{20} and Ne^{22} is about 900 MHz. At the temperature of an ordinary discharge tube the Doppler broadening is appreciably greater than this, and is sufficient to keep the two isotope lines from being resolved. By cooling the fluorescence sampling tube by means of liquid nitrogen (the main laser-irradiated section of the cataphoresis tube is not cooled) the Doppler width is reduced and the isotope lines are resolved. The cooled sampling tube in combination with the scanning confocal spectrometer system thus provide a very sensitive method for observing changes in the isotope ratio.

Another scanning Fabry-Perot spectrometer is also available for occasional diagnostic use, but is not an integral part of the system. This parallel-mirror scanning interferometer, with variable mirror spacing and wide-band coatings, is invaluable for looking at other Ne lines. It uses photon counting and multi-channel storage, and can detect very low photon intensities. Using the spectrometer we have looked at the $5852 \text{ \AA}$ Ne line, and are interested in taking advantage of the greater isotope splitting for this line than for the $6328 \text{ \AA}$ line.

Isotope separation in neon has not yet been observed, although it is only recently that we have reached the point where observable cataphoretic separation might be expected. We anticipate two to three months work on the system before a systematic search for isotope separation will be undertaken.

During the approximately 3 months remaining of the first year of the program there is an excellent possibility that cataphoretic isotope separation in Ne will be observed. However, we still have the following changes and improvements in the apparatus

to make, and these may occupy us most of this period before systematic changes in the current density, gas pressure, He-Ne ratio etc. are carried out.

(a) By using the 5852 Å fluorescent line for observing the isotope ratio instead of the 6328 Å line we hope to avoid cooling the sampling tube. Parts for a confocal Fabry-Perot spectrometer at 5852 Å have been procured but the instrument is not yet assembled and aligned.

(b) In order to have a higher laser intensity we want to use a longer laser tube. We are planning a folded-cavity arrangement probably more than two meters in length. The cataphoresis tube will also be lengthened. More complete filling of the cataphoresis-tube cross-section by the laser beam is also a primary objective.

NEON ISOTOPE SEPARATION SCHEMATIC
FIGURE 2

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Effort: term 15%: summer 50% (1 1/2 months)

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