NOVEL ^{125}I PRODUCTION AND RECOVERY SYSTEM

A Thesis

by

ADWITIYA KAR

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

August 2007

Major Subject: Health Physics

NOVEL ^{125}I PRODUCTION AND RECOVERY SYSTEM

A Thesis

by

ADWITIYA KAR

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

Approved by:

Chair of Committee, Warren Dan Reece
Committee Members, Leslie Braby
Michael Walker
Head of Department, John Poston

August 2007

Major Subject: Health Physics

ABSTRACT

Novel ^{125}I Production and Recovery System. (August 2007)
Adwitiya Kar, B.Sc., University of Calcutta;
M.Sc., University of Calcutta
Chair of Advisory Committee: Dr. Warren Dan Reece

This research suggests ways of reducing contamination of iodine-126 in iodine-125 and lays out a simpler iodine-125 production technique to increase the yield.

By using aluminum irradiation vessels the yield of iodine-125 produced by neutron irradiation of Xe-124 can be doubled compared to using stainless steel vessels. Because of increased yields irradiation times are shorter, the chance of I-126 contamination is reduced. Solidified iodine within the aluminum vessels can be extracted using 0.1 N sodium hydroxide solution, however the solution also reacts with the vessel walls. These impurities in the extracted solution are then removed by distillation that concentrates and purifies the extracted solution. High recovery, ranging from 88 to 96 percent, was typical for the experiments described. Gamma spectroscopic results suggest that the distillate is free from any impurities such as aluminum or sodium ions. Distillation can reduce the extracted solution to at least one third or less of its original volume. The work described here provides the basis for I-125 production at the Texas A&M Nuclear Science Center.

DEDICATION

To my mother

ACKNOWLEDGMENTS

I would like to thank my committee chair, Dr. Dan Reece, and my committee members Dr. Leslie Braby and Dr. Michael Walker, for their guidance and support throughout the course of research.

Thanks also to my friends and colleagues at the Nuclear Science Center at Texas A&M University. I also want to extend my gratitude to the employees of the Nuclear Science Center for their constant support.

Finally, thanks to my mother for her love and encouragement.

TABLE OF CONTENTS

Page

Page

LIST OF FIGURES

LIST OF TABLES

INTRODUCTION

There is an increasing need for iodine-125 in the world market for research, medical and diagnostic use. In radio-biological assays, I-125 is used as a tracer, often tagged to protein molecules and imaged by auto radiography. Its 35 keV gamma and 27 keV X- rays makes it a widely used brachytherapy source for prostrate cancer, breast cancer and brain tumor treatments. Recent journal articles and reports from a Department of Energy (DOE) expert panel for last few years have pointed out that there is limited domestic availability of radionuclides for laboratory and clinical research during the early stages of developing and testing new radiopharmaceutical products (DOE 1999, Tenforde 2004). At present, a considerable amount of iodine-125 comes to US from international sources (Studsvik 1998, ANS 2004). Furthermore a report from DOE's symposium in 2004 showed McClellan/UC Davis as the only producer of I-125 under DOE's isotope program in U.S. (OSTI 2004). This is not enough to support the domestic demand of iodine-125. The USA alone accounts for about 36000 procedures requiring radioisotopes. This includes medical procedures such as brachytherapy, radioactive tracing as well as diagnostic procedures like radio-immuno-assay (RIA), all of which depend to some extent on the supply of iodine-125.

The current processes followed by most companies producing I-125 from Xe-124 are have problems. Most producers use stainless steel irradiation cylinders for activating Xe-124 because iodine's chemical reactivity makes the removal of iodine from the decay cylinder challenging for anything other than stainless steel. However, stainless steel reduces the neutron flux by half and lowers the yield. This prolongs the irradiation time for stainless steel containers that in turn can increase the level of I-126. Iodine-126 is an undesired contaminant in I-125. An alternative to this is to use aluminum irradiation cylinders. Aluminum cylinders are essentially transparent to thermal neutrons and double the flux compared to stainless steel. Nevertheless, it is difficult to extract iodine from aluminum cylinders because of aluminum's reactivity with extracting solution such as sodium hydroxide. The chemical product of the reaction contaminates the extracted iodine solution.

This thesis follows the style of Health Physics.

One objective of this research is to suggest ways of reducing such contaminants and lay out a simpler I-125 production technique to increase the yield. McMaster’s production process, for example uses stainless steel capsules for Xe-124 irradiation, transfers the irradiated gas to a second stainless steel capsule where it decays to I-125. The process of transferring of highly radioactive Xe-125 is complicated and has to be carried out underwater within a second vessel to preclude escape of the radioactive gasses. In addition, reusing the irradiation chamber for succeeding cycles raises the chance of making I-126 from any I-125 remaining from previous cycles. Instead, we suggest using a single cylinder for irradiation and decay obviating the need for a separate decay chamber. Using a new aluminum cylinder for every cycle or aggressively cleaning the single cylinder can reduce the chance of I-126 contamination resulting from any in buildup of I-125. Besides avoiding a complex valve system, this method eliminates the risk associated with a transfer process like McMaster’s. As mentioned before, aluminum irradiation canisters, unlike stainless steel, are essentially transparent to thermal neutrons and allow a higher thermal flux during irradiation. This in turn reduces the irradiation time for a desired yield and can help in limiting the possibility of I-126 formation from I-125.

Another objective of this research was to devise a suitable method for extracting plated iodine-125 from aluminum cylinders. Experimental results suggest that 0.1N sodium hydroxide can be used as an extracting solution for recovering iodine-125. Sodium hydroxide etches the walls of the aluminum and extracts the iodine along with it. The aluminum hydroxide formed as a result of aluminum’s reaction with sodium hydroxide renders the recovered solution unusable without further purification. The third objective of the research was to find a way to concentrate and purify the extracted I-125. The current radiopharmaceutical market typically sells I-125 as sodium iodide after being recovered from the irradiation cylinders with sodium hydroxide solution. The target specific activity is typically 1 Ci/ml and this complicates the removal of the iodine from the vessel because of the small volumes of solute that can be used. No one has published procedures to concentrate the solution of I-125 after its removal from the irradiation cylinders. We suggest using distillation to purify and concentrate the recovered solution. The main part of the research effort was to work out the details associated with iodine distillation. The findings suggest that distillation of 30ml of solution can result in distillate of about 7ml while carrying over 90% of the iodine. Distillation therefore can reduce the solution to almost one-third or less of its original volume. Thus, distillation serves both as a means of increasing of the I-125 concentration and removes impurities such as sodium and aluminum ions.

THEORY

The history of iodine started when Bernard Courtois accidentally discovered it in the 1800s. Since then iodine has become an important element in medicine, photography, dyes, and industry. Only one isotope of iodine, I-127, occurs naturally. The other iodine isotopes are artificial and are made in a reactor or a cyclotron. Iodine is similar with other group VII elements chlorine, fluorine and bromine and like all halogens forms a diatomic molecule. However iodine is the least reactive of all halogens and has the special property of sublimation. Iodine has a meting point of 113°C and on heating turns into vapor without the intermediacy of liquid. It has a high vapor pressure and plates out as crystals on cooling. The vapor pressure of iodine, shown in Table 1, increases rapidly with temperature making it an extremely volatile element. Many of the measures adapted in the present research are directly related to this particular property of iodine. Since the chemistry of naturally occurring I-127 is the same as any of its radioisotopes, the reader should be aware of some basic chemical reactions of iodine in order to understand the analytical methods applied in this research.

Table 1 Change in vapor pressure with temperature.

Pressure (Torr)	10^{-5}	10^{-4}	10^{-3}	10^{-2}	10^{-1}	1	10	100	760
Temperature (^{0}C)	-61.3	-46.9	-30.2	-10.8	12.1	39.4	73.2	115.8	182.8

Iodine dissolves slowly in water but dissolves faster in a solution of potassium iodide because of the formation of tri-iodide ions.

$$I_2 + I^- = I_3^- \qquad (1)$$

The excess of iodide in the potassium iodide in the solution displaces the equilibrium from left to right. Iodide ions have much less vapor pressure than iodine and presence of tri-iodide ions reduces the volatility of iodine in the solution. Iodine can act as an oxidizing or reducing agent and has an oxidation potential of -0.53 volts. Equation 2 shows the reversible reaction of an iodine-iodide system. Iodine acts as oxidizing agent for substances with an oxidation potential much lower than that of the iodine-iodide system.

$$I_2 + 2e^- = 2I^- \qquad (2)$$

On the other hand iodide exerts a reducing action upon strongly oxidizing systems, with the formation of equivalent amount of iodine. Oxidizing agents like hydrogen peroxide (H_2O_2) react with iodide in an acidic medium to form iodine according to the reaction:

$$H_2O_2 + 2I^- + 2H^+ \longrightarrow I_2 + 2H_2O \qquad (3)$$

The oxygen content of hydrogen peroxide is decreased during reduction. In such cases, the oxidation potential increases as a function of hydrogen ion concentration in the solution (Kolthoff et al 1952). This is because hydrogen ions take part in the reduction process and, as a result, raise the capability of hydrogen peroxide to accept electrons. Although the oxidation potential of the iodine-iodide system is virtually independent of the pH of the solution, for higher alkaline solution like sodium hydroxide (NaOH) with pH above 8, iodine reacts with hydroxyl ions to form hypoiodite and iodide. The hypoiodite is extremely unstable and is rapidly transformed into iodates:

$$I_2 + 2OH^- \longrightarrow IO^- + I^- + H_2O \qquad (4)$$

$$3IO^- \longrightarrow 2I^- + IO_3^-$$

Upon reaction with potassium hydroxide, which is also strong base, iodine follows the same reaction as in Equation 4 to form potassium iodide and water, thereby reducing the volatility of the iodine in solution. Such reactions prove to be of great value for analytical methods of iodine estimation and have been used extensively in the present research.

MEDICAL HISTORY AND RADIOISOTOPES OF IODINE

Iodine entered medical history shortly after its discovery in 1820 when a Swedish scientist Jean François Coindet (1774–1834) suggested that iodine compounds might cure goiters, an enlargement of the thyroid gland in humans. But his studies were mostly observational and were not experimentally tested until Gaspard Adolph Chatin (1813–1901) began his extensive series of iodine analyses. In 1833 a French scientist named Jean

Boussingault (1802-1887) also recommended the use of dietary iodine for the prevention of goiter but both Chapin's and Jean's method of treatment was confirmed by a German chemist Eugen Baumann (1846-1896) in 1896. Baumann discovered that the thyroid gland was rich in iodine and also determined that the thyroid was the only tissue containing iodine (Rosenfield 2000). Just two years later, Austrian psychiatrist, Julius Wagner von Jaurreg (1857-1940), established that goiter could be prevented by taking iodine tablets regularly and proposed that iodized salt be sold in areas where goiter was widespread. This made iodine a very sought after element and soon it gained a widespread acceptance in treating hyperthyroidism.

Perhaps because iodine was so successful in treating goiters, the idea of using iodine to treat other diseases such as thyroid related cancers developed in 1900s with the discovery and development in the field of radioactivity. The first conscious urge to make radioactive iodine isotopes probably arose around 1934 when Enrico Fermi reported the production of several radioisotopes with small half-life in his laboratory (Fermi 1934). The paper was published in the reputed English journal 'Nature' and energized the Physics world to produce artificial radio-isotopes that would be of value in health and medicine. In 1937 a group of scientist headed by Robley Evans in MIT produced I-128 by bombarding ethyl iodide with neutrons (Hertz et al 1938, Chapman 1983) and in 1939 published a paper on the role of I-128 in thyroid physiology. Around the same time in 1938, I-131 was first produced at the Lawrence- Berkeley University of California by using Lawrence's cyclotron. In a bid to produce radioisotopes with shorter half-life that can be used in medicine without many side effects, Glenn Seaborg and Jack Livingood bombarded tellurium with deuterons in the lab's 37-inch cyclotron producing I-131 (Livingood 1938).Of the 35 isotopes of iodine that can be produced artificially, I-131 was the first isotope to be used in medicine and since has become the most popular in treating thyroid and other types of cancers. But interests soon turned to other low energy emitting radio-isotopes of iodine and this led to production of iodine-123 and iodine-125. I-123 replaced I-131 in radio-imaging and it was not long before the medical community started looking into I-125's feasibility in curing prostrate and breast cancers along with its use in radioimmunoassay.

PRODUCTION OF IODINE-125 AND MARKET DEMANDS

Iodine-125 was first produced by Allen Reid and Albert Keston in 1946 (Stabin) using a cyclotron. The usual method was to bombard natural Te in a cyclotron with 14 MeV deuterons.

Since natural tellurium is a mixture of eight different nuclides, contamination from radio-iodines such as I-131 and I-126 would occur. I-125 was the longest living contaminant and it could be separated by allowing the product to decay for months after formation (Myers 1984). However, this method was inconvenient and could not be used profitably for a steady supply of iodine-125 in the world of medicine. In 1960 in another paper published in the 'New Journal of Nuclear Medicine' Myer and his colleagues suggested that I-125 might also be produced from Xe-124 following the sequence:

$$^{124}Xe\,(n,\gamma)\,^{125}Xe \xrightarrow{EC,\beta+} {}^{125}I$$

He conjectured that because a neutron rich isotope will be formed, the procedure can be carried out in a reactor instead of a cyclotron and noted that even if Xe-124 is only 0.096% abundant such a reaction might be possible because of the high absorption cross section of Xe-124 to thermal neutrons. By this process xenon-124 absorbs a thermal neutron undergoing an (n, γ) reaction in which a new isotope Xe-125 is produced. Xe-125 which has a 16.9 hr of half-life will in turn undergo electron capture (EC) and beta plus decay (β^+) to produce iodine-125. The changes in a nucleus following a beta plus decay and electron capture are explained in equations 5 and 6.

$$p^+ \longrightarrow n^0 + e^+ + v_e \qquad (5)$$

$$p^+ + e- \longrightarrow n^0 + v_e \qquad (6)$$

In both cases the new isotope will carry the same mass number but a different atomic number than its parent. The iodine-125 formed will now undergo electron capture and decay to stable tellurium-125 completing the radioactive decay process. Soon after Myer's proposition in 1960, Lathrop and Harper (1961) explored this method of iodine-125 production (Harper at al 1963). They irradiated natural xenon (Xe-124 0.095%) gas in a Zircaloy irradiation vessel using the ANL CP5 reactor (Lathrop and Harper 1963). Apart from the contamination from Cs-137 which resulted from Xe-136 in the natural Xe-124, another possible side reaction seemed to be the formation of iodine-126 from I-125. In order to be certain Lathrop and Harper carried out a series of experiments to determine the neutron activation cross sections of xenon-124 and iodine-125 and soon confirmed chances of heavy contamination in I-125 with I-126. After studying their data of the neutron absorption cross section for the two nuclides (reproduced in Table 2) they concluded that iodine-125's high neutron activation cross section increases its possibility of a neutron capture during the irradiation of Xe-124. But the authors also pointed out that because iodine-126 arises as secondary product from iodine-125, I-126 contamination could be prevented to a great extent by irradiating huge of quantities of Xe-124 for a short period of time.

Table 2 Neutron activation cross sections from ^{124}Xe (n, γ) ^{125}Xe and ^{125}I (n, γ) ^{126}I in barns (Lathrop and Harper 1963).

Pile neutron activation cross section		
Target	Xe-124 (σ)	I-125 (σ)
Xe-124	169	
Xe-124	179	
Xe-124…I-125	173	1160
Xe-124…I-125	175	1138
I-125		1180
I-125		1110

The current thermal neutron cross section for Xe-124 is about 168 barns and the thermal cross section for I-125 is 900 barns with a 14,000 barn resonance integral. Harper's production method was a milestone in the history of radioactive I-125. Finally there was chance of producing of iodine-125 free from other radioactive contaminants like I-131 and I-126 and provided a profitable opportunity for using it in methods such as radioimmunoassay (RIA). Researchers soon realized that I-125's 60-day half-life over I-131's 8 day half-life would prove advantageous in studying protein structures and function and its low energy gamma could be effectively used in autoradiography and interstitial radiation therapy. But to be used in iodination of proteins and as radioactive seeds, the specific activity of the radioactive source was of primary importance. When producing iodine-125 from Xe-124 the specific activity of I-125 could be decreased by the contamination from iodine-126. When xenon-124 is irradiated it initiates a chain of side reactions shown in Figure 1. The main side reactions arising from the process were two double neutron captures and a direct neutron capture (Martinho et al 1984).

$$^{125}I\ (n, \gamma)\ ^{126}I\ (n, \gamma)\ ^{127}I$$

$$^{125}Xe\ (n, \gamma)\ ^{126}Xe\ (n, \gamma)\ ^{127}Xe \xrightarrow{EC} {}^{127}I$$

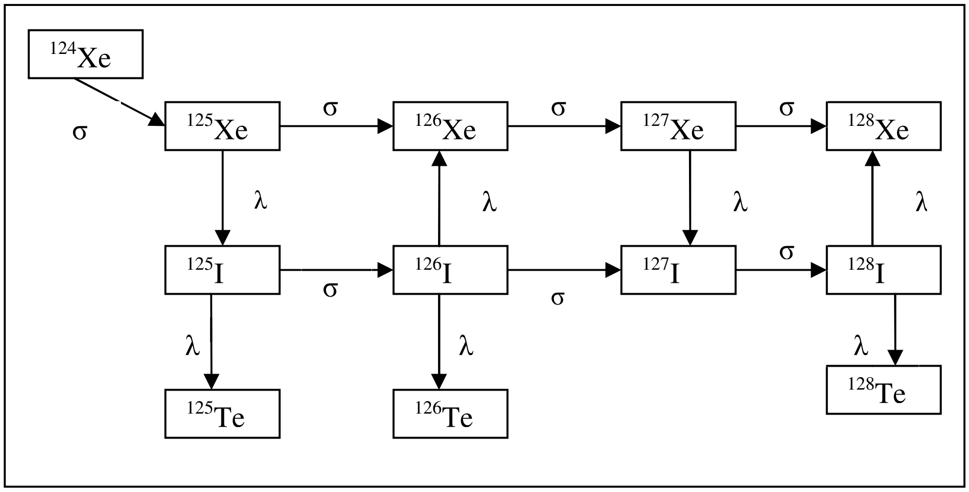

Figure 1 Growth and decay of nuclides arising from neutron irradiation of natural xenon.

Because the maximum specific activity and the minimum radionuclide contamination depended on the branching reactions, the neutron flux, the time of irradiation and the decay time, high specific activity could only be achieved by a proper planning of the radioisotope production scheme. Martinho et al. addressed the problem by using the computer program ISOTOP to graphically show the changes in iodine-125 activity and I-125 radionuclide contamination as a function of irradiation time for different thermal neutron fluxes. He studied iodine-125 activities as a function of irradiation time for changing thermal neutron fluxes as well as iodine-125 radionuclide contamination as a function for irradiation time for varying fluxes and presented his observations as shown in Figures 2 and 3.

Apart from controlling the time and the flux there were other concerns in making iodine-125. Using natural xenon for irradiation raised the level of contaminants such as Cs-137 and Cs-134. Since natural xenon is a mixture of several isotopes, its irradiation produced unwanted radionuclides such as Cs-137. But such problems were soon resolved and the industry quickly replaced natural xenon with 99.9% enriched xenon-124.This cut down the percentage of other isotopes of xenon and restricted the contamination to negligible quantities. Also as already suggested by Harper, irradiation of huge quantities of enriched Xe-124 in a short period of time with a thermal fluence roughly around 10^{13} helped to limit the production of I-126 to a considerable extent. In the early 90s with the advancement of computers such complex calculations were made relatively easy and now various computer programs such as MCNP are being used to calculate optimum irradiation conditions for a desired specific activity of the isotope.

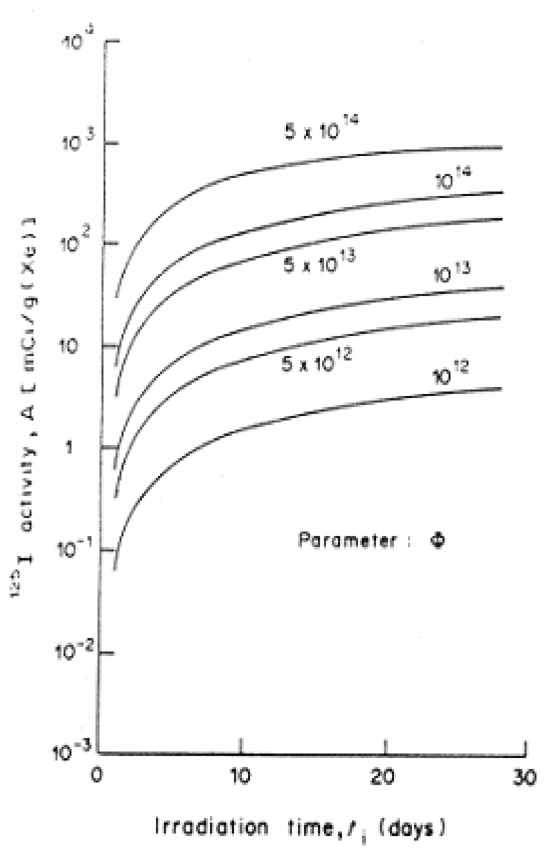

Figure 2 Iodine-125 activity as a function of irradiation time for different thermal neutron fluxes.

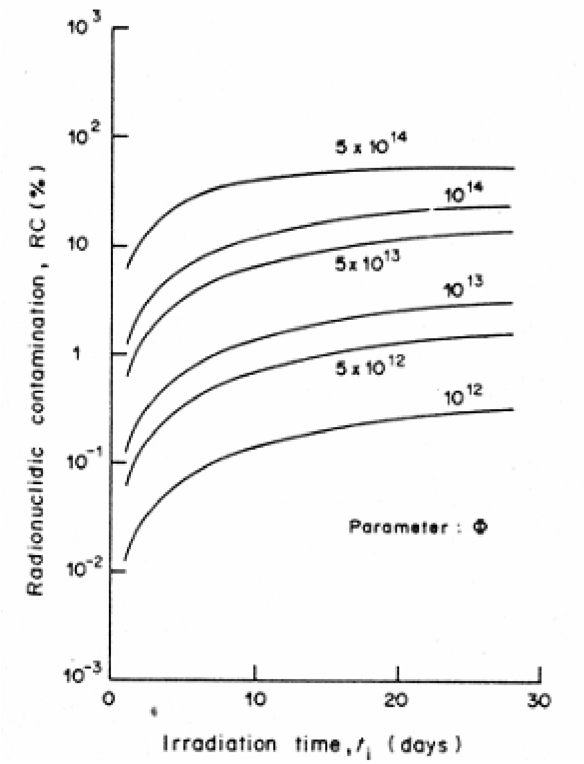

Figure 3 Iodine-125 radionuclide contamination as a function of irradiation time for different thermal neutron fluxes.

After optimizing the irradiation procedures, a suitable process must be developed to recover the iodine-125 produced from Xe-125.The recovery of I-125 from the irradiation vessel has not been easy since the process's conception. For a radionuclide that sublimes on the wall of the irradiation vessel and has a high vapor pressure, it was best recovered by Lathrop and Harper in 1961using potassium bisulphite adjusted to pH 7-8 with potassium hydroxide solution (Harper et al 1963). However, because of the reaction of the alkali with the walls of the irradiation vessel, the recovered solution wasn't useful for medical applications. Since then the technique of I-125 recovery has changed based on the market demands and technical feasibility. To minimize contamination, today most I-125 producers use stainless steel as an irradiation vessel. Since stainless steel has minimal reaction with sodium hydroxide, the standard practice is to recover the iodine-125 using an aqueous solution of sodium hydroxide and ship it as a sodium iodide solution.

At the Nuclear Science Center continuing efforts have been made to develop novel techniques in iodine-125 production and recovery. Interest in this research has largely been fueled by the current market requirements for iodine-125. In the last few years the use of iodine-125 has increased as the source in brachytherapy seeds for cancer and in treatment of breast tumors and as a source for eye plaques. Although initially overshadowed by iodine-131, I-125 has become one of the most used radioisotopes of our time. Because of its low energy gamma, I-125 seeds are now being extensively used in interstitial tissue irradiation, in treating carcinomas of small glands such as the thyroid and prostrates. New innovative techniques are also calling for pure iodine-125 with high specific activity rather than its solution in sodium hydroxide. None of the major pharmaceutical suppliers have so far claimed to have produced iodine-125 in such form. Nowhere in the current literature is a suitable method for concentrating the I-125 solution obtained from the irradiation vessel. It is this lack that motivates the search for a new technique to produce iodine-125 solutions with high specific activity.

A shortage in market supply against demands of I-125 has acted as another incentive for the present endeavor. Current journal papers and Department of Energy's (DOE) symposia have consistently pointed out that the United States market is facing a shortage in radionuclide supply compared to demand. A review of the sales of pharmaceutical products by the consulting firm Frost and Sullivan from 1998 to 2002 has showed that the compound annual revenue growth rate of the total United States radiopharmaceutical market will be 10.2% during the period 2001–2008 (Tenforde 2004). Recent journal papers and reports from a DOE expert panel for last few years have continuously stressed on the problem of limited domestic availability of radionuclides for laboratory and clinical research during the early stages of developing and testing new radiopharmaceutical products (DOE 1999). In 1998 an expert panel assembled by DOE was formed to monitor the nation's isotope program, to maintain a steady isotope supply as per demand. But subsequent reports through 2005 have shown that DOE is facing a crisis in providing radioisotopes for research and medicinal purposes (DOE 2005). This supply shortage fuels a large import rate from international companies (Adelstein et al 1995, Nordion 2007). Studsvik, partnered with McMaster claims to be the second largest producer of iodine in the world (Studsvik 2004) and is providing most of I-125 required by the US market. Furthermore, a report from DOE's symposium in 2004 had shown McClellan/UC Davis as the only producer of I-125 under DOE's isotope program in United States. U.C Davis has currently stopped iodine-125 production leaving foreign companies to be the only suppliers of iodine-125 in United

States. All these statistics has prompted the Nuclear Science Center to take an avid interest in producing iodine-125.

PRODUCTION METHODS OF IODINE-125 BY MAJOR COMPANIES

A literature review of the United States patent on iodine-125 production from 1976 was carried out in order to gain an insight into the current production methods utilized by the companies holding U.S patents. The search revealed that McMaster University reactor of Canada to be the sole patent holder in iodine-125 production and recovery methods (Hassal et al 1997) in United States. Other references indicate that the Swedish company Studsvik utilizes its R2 reactor at Sweden to produce Iodine-125, and supplies iodine-125 in U.S through a partnership with the McMaster. MDS Nordion, the Canadian company is also a supplier of iodine-125. Although their production and recovery method has not been published, the radiopharmaceutical data shows that Nordion carries out dry distillation of the target to recover iodine-125 (Nordion 2007) and the iodine ships as a solution in sodium hydroxide.

Iodine production at McMaster has been described in detail in U.S patent 5633900. The McMaster Nuclear Reactor (MRC) uses a thermal flux of 10^{12} $cm^{-2}s^{-1}$ to irradiate Xe-124 in a stainless steel irradiation vessel. Figure 4 is a schematic reproduction of their production system explained in US patent 5633900. In this method , xenon-124 is transferred cryogenically into the irradiation vessel from storage of Xe-124. At the end of irradiation the decay of Xe-125 is moved to a second chamber connected to the irradiation vessel through a system of valves. The second chamber is detachable and is transferred to safe holding to keep iodine-125 away from neutron fluence. Once the iodine-125 is formed, the second chamber is attached to a vessel of degassed solution of NaOH and hydroxide solution is allowed to flow into the second chamber by a vertically attached needle through an opening controlled by a valve. The entire chamber is then inverted such that hydroxide solution is accumulated near the valve and the iodine-125 on the wall of the chamber is dissolved by refluxing the sodium hydroxide solution.

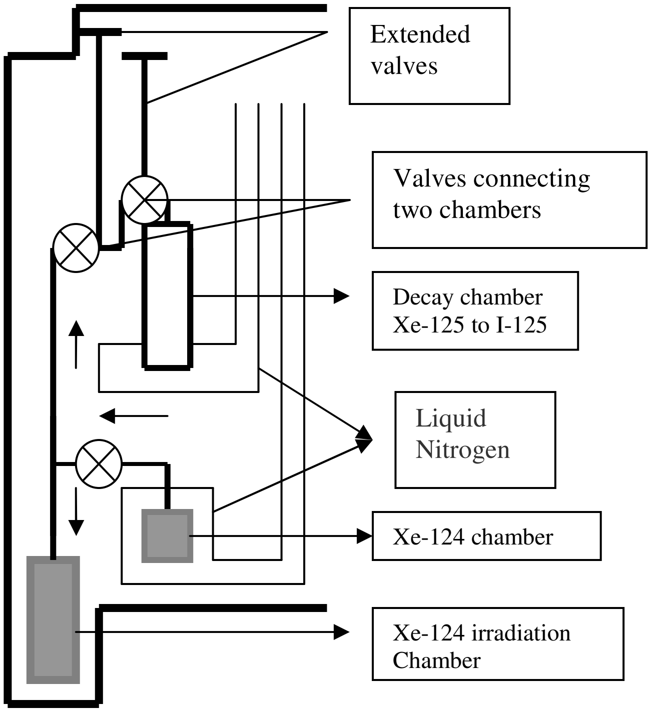

Figure 4 Iodine-125 production scheme at Mc Master (US 5633900).

Although McMaster's production system has been in operation since 1993, their method of production suffers apparent engineering problems. The method of gas transfer between the two chambers is complicated and has to be carried out in a tightly controlled environment. For a one week cycle, the McMaster irradiates 5 grams of Xe-124 for 15 hours per day for a period of three to five days. A problem with this method is that iodine-125 produced at the beginning of the week becomes I-126 contaminant at end of irradiation. Since McMaster re-uses the stainless steel irradiation chamber, the residual I-125 plated inside the chamber from the first irradiation contributes to produce iodine-126 in subsequent cycles. This builds up and over time has the potential of exerting enough vapor pressure to contaminate the Xe-125 transferred into the decay chamber with iodine-126. Moreover McMaster uses stainless steel as their material for irradiation vessel. Stainless steel has a high absorption cross section for thermal neutron and can reduce the flux to almost half requiring a longer irradiation time to produce the desired iodine activity. This can lead to higher levels of I-126 and eventually defeat the purpose of transferring Xe-125 into the second chamber away from any neutron flux to control the in growth of I-126.

The United States's own iodine-125 production facility, an iodine-125 production assembly designed as a self contained unit, was first designed, fabricated and tested at the Los Alamos National Laboratory in 1992. But it was never tested with radioactive material and was eventually transferred to Sandia National Laboratories. Afterwards it was again sold to U.C Davis and in 2004 U C Davis became the sole producer of iodine-125 in the United States. The MNRC reactor of U C Davis produced iodine-125 from xenon-125 using the production assembly sold by Sandia Laboratories. Although this production method did not suffer from apparent problems of contamination with I-126, the recovery process from the decay chambers was complicated and had to be accomplished through network fine capillary tubes. A schematic portrayal of their production system in Figure 5 shows the way the iodine-125 was produced and recovered (Parma 1999)[1]. The assembly had a primary containment which was inside a secondary containment and was attached to a glove box. The primary containment as shown in Figure 5 was used for irradiation, transfer and decay of xenon-125 to iodine-125. The elution process from the decay chamber was done by allowing a flow of 0.005N sodium hydroxide solution through a system of capillary tubes leading directly to the product bottle. The decay storage vessel was surrounded by electrical heaters to keep the sodium hydroxide solution warm. One problem with such a method of recovery rose from the need to maintain a constant flow rate in the capillary tubes during the process of elution. A small accidental crack in any of the tubes meant a total dispersion of the radioactive solution and radioactive iodine vapors. Also because it was closed system there was a regular need of removing air from the product bottle to maintain the driving pressure. Apparently, this whole method of iodine production and recovery did not work out as planned and by 2006 U C Davis discontinued their iodine-125 production unit.

---

[1] "1-125 Production Assembly Design Specifications and Operating Theory," Unpublished Report, Sandia National Laboratories, Albuquerque, NM, October 27, 1999.

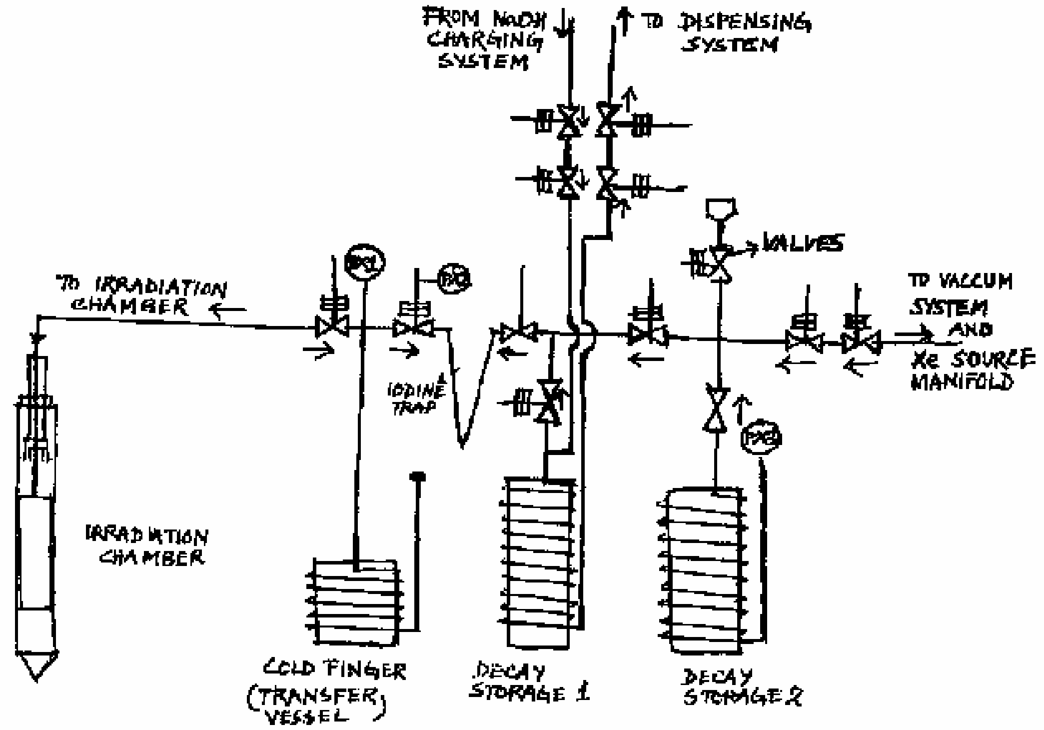

Figure 5 Iodine-125 production unit at U C Davis.

The research at NSC is centered on simplifying this process of transfer and recovery of pure iodine-125.The irradiation vessel at NSC is made of aluminum because aluminum is essentially transparent to thermal neutrons unlike stainless steel. The irradiation time at NSC can be kept reasonably short and this helps to prevent iodine-126 formation. Figure 6 gives a short schematic of the iodine-125 production at NSC. Comparing the scheme with the McMaster and U C Davis processes, it is evident that the NSC process is much simpler and does not require any complex containment. Use of a single chamber for irradiation and decay precludes the complex valve system needed for connecting two chambers as used by McMaster. Once the valves 1, 2, 3 and 4 are opened a vacuum pump removes the air inside the aluminum irradiation vessel. After sufficient time valve 1 is closed, the aluminum cylinder is immersed in liquid nitrogen, and valve 6 is opened allowing the flow of Xe-124 from its storage cylinder. Xe-124 is now transferred cryogenically to the irradiation chamber solidifying inside the vessel. When the transfer is complete, the valves are closed and the tube containing the irradiation vessel is detached from the rest of the system. The irradiation vessel is now transferred to the core where it is irradiated for about 14 hours to make Xe-125. At the end of irradiation the vessel is removed from the core to a safe storage away from neutron flux inside the pool. Here the Xe-125 decays to iodine-125.

Table 3 shows the activity of xenon-125 at the end of irradiation and the also the activity from the gradual buildup of iodine-125 inside the irradiation vessel during decay of xenon-125 to iodine-125. 7.65 grams of Xe-124 produces 546 Ci of Xe-125 after 14 hours of irradiation. Since the irradiation time at NSC is less than 17 hrs there is much less possibility of producing iodine-126 from iodine-125. A calculation of the t_m value which is the time when the daughter and parent's activity equals show that in about 5 days following end of irradiation the activity of Xe-125 equals that of I-125.

Table 3 Activities of Xe-125 and I-125 during decay.

Days	**Xe-125 activity in Ci**	**I-125 activity in Ci**
0	546.3128	0
0.01	541.0236	0.0627
0.05	520.375	0.3074
0.1	495.6688	0.6
0.5	335.8863	2.4869
0.7	276.4984	3.1844
1	206.3128	4.0016
2	78.0629	5.4684
3	29.5085	5.9774
4	11.1545	6.1250
5	4.2165	6.1365
6	1.5939	6.097
7	0.6025	6.0388
8	0.2277	5.974
9	0.0861	5.9071
10	0.0325	5.84

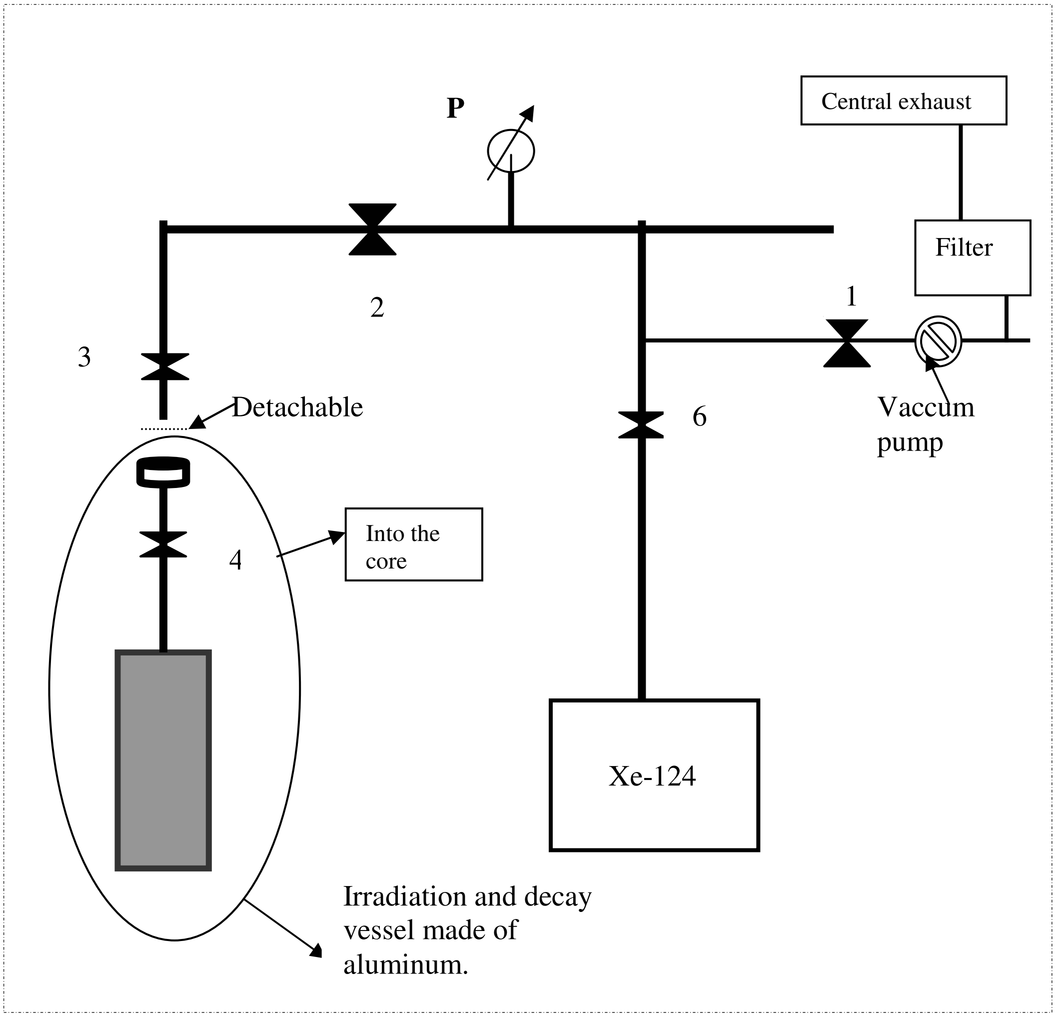

Figure 6 Proposed scheme for I-125 production at NSC.

An activity curve showing the in growth of the daughter iodine-125 from its shorter-lived parent xenon-125 is shown in Figure 7. Following the table and the exponential decay curve note that between 4 and 5 days post-irradiation the activity of I-125 builds up and reaches a maximum when the activity of iodine-125 equals the activity of xenon-125. This is the maximum amount of iodine-125 that can be recovered from a single irradiation cycle. Although the activity of iodine-125 starts falling thereafter, the iodine is not extracted until the activity of Xe-125 falls to very low levels. This ensures proper practice of ALARA and limits the chances of iodine-126 contamination for the next irradiation cycle. A 10 day decay time is suggested before iodine-125 is recovered. At the end of 10 days the irradiation vessel is removed from the pool and attached to the system shown in Figure 6. The system is evacuated and the Xe-124 storage vessel is now placed in liquid nitrogen. Valves 4,3,2,6 are opened and Xe-124 and a small amount of Xe-125 remaining in the irradiation vessel is transferred back into the storage vessel for reuse in subsequent irradiation cycles. Once the gas is transferred, valves 4, 3, 2, 6 are closed; the tube connected to the irradiation vessel is detached from the rest of the system and is transported to the glove box for extraction and distillation.

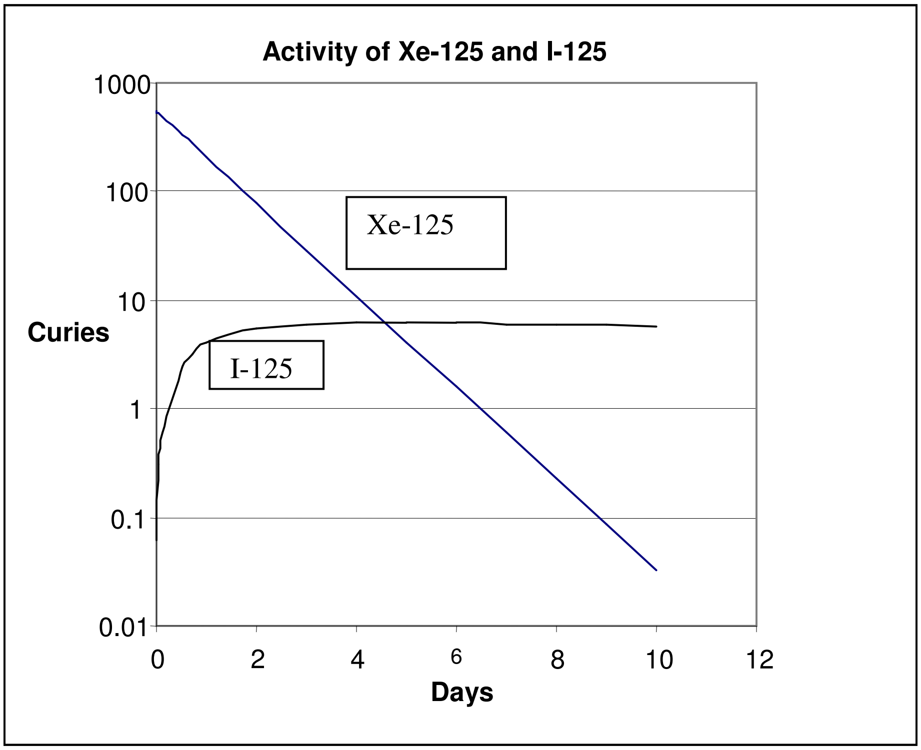

Figure 7 Quantitative relationship between activity vs. time for Xe-125 and I-125.

RECOVERY METHOD FOR IODINE-125

This research centers on finding a suitable method of recovering iodine-125. The concentration of the iodine-125 available in the market is low and can be improved. A search into iodine-125 specifications data of the current suppliers reveals that McMaster sells (800-1200) mCi of iodine ml^{-1} and Nordion, has three ranges of concentration, the highest being 500 mCi ml^{-1} and the lowest being less than 200 mCi ml^{-1}. This research shows that a high yield of pure and concentrated iodine-125 is possible with distillation. Since iodine has a high vapor pressure, a considerable amount of iodine can be recovered with minimum amount of water in a short period of time. So the distillate obtained would be pure iodine-125 dissolved in a small amount of water distilled over with the iodine vapors. Although distillation of iodine has been attempted in the past, it has not been applied on an industrial scale for radioactive iodine recovery. In the past distillation has been used for separating carrier free iodine-125 from irradiated tellurium dioxide in which the aluminum capsule containing the target was dissolved in a mixture of aqueous sodium hydroxide and sodium nitrate (Khan et al.1977). The dioxide was then dissolved in concentrated nitric acid and iodine distilled into sodium hydroxide solution.

At the Nuclear Science Center the aim is to keep the distillation procedure short and simple. The plated iodine inside the aluminum canisters is washed out with sodium hydroxide solution. However, the alkali will react with the aluminum and these contaminates are carried with the sodium hydroxide. This renders the solution unusable without further purification. Distillation of the extracted solution can remove these purities and can concentrate the iodine in the final solution. This can prove to be a milestone in iodine recovery if properly conducted. The core of this research has been to work out the associated details of this distillation process. Analytical procedures like Neutron Activation Analysis (NAA) have been carried out to ensure that the distillate contains no other impurities such as aluminum or sodium. Iodine has a tendency to plate out on the condenser of the distillation unit during its passage to the collector vessel. This research has found ways to improve the distillation procedure so that iodine passes from the distillation flask to the collector without plating. Findings of this research suggest that Nuclear Science Center can now produce and recover iodine by a unique method and can compete with companies such as McMaster.

METHODS AND MATERIALS

The research was divided into two phases: a non-radioactive phase to work out the details of the iodine chemistry and a radioactive phase to prove the principals developed in the first phase. Because stable I-127 and its radioactive isotopes share the same chemistry, chemical challenges anticipated in the production of iodine-125 were simulated with I-127 as closely as possible. Such a plan allowed working out the experimental details without the health concerns that would otherwise arise from working with radioactive iodine. Analytical procedures adapted in the non-radioactive phase included spectrophotometric analysis and analysis by neutron activation (NAA). Radioactive iodine-128 was used for experiments in the radioactive phase to corroborate the findings in the non-radioactive phase. Again, because the chemistry of all iodine isotopes is the same, iodine-128 was chosen as a suitable tracer. Iodine-128 has a 24.9 minutes half-life and is less hazardous than iodine-125 which has a 59.4 days half-life. Results obtained in radioactive phase were analyzed using gamma spectroscopy with high purity germanium detectors and GENIE 2000 software.

ANALYTICAL METHODS APPLIED IN NON-RADIOACTIVE PHASE AND RADIOACTIVE PHASE

Spectrometric analysis was used as a quick and easy way to determine the amount of recovered iodine during the initial stages of research. A Spectronic 20 was used for all spectrophotometric measurements. The spectrophotometer measures the concentration of the substance by measuring its absorbance or transmittance at a particular wavelength. The Spectronic 20 used in this project reports both percent transmittance and absorbance. Percent transmittance is the ratio of intensity of the light passing through the sample to the light incident on the sample multiplied by 100. Absorbance is described by Beer-Lambart's Law shown in Equation 7, where A is the absorbance, I_o is the intensity of the incident light, and I_1 is the intensity of the light after passing the material. Absorbance is the log of transmittance.

$$A = \log_{10} \left(I_o / I_1 \right) \qquad (7)$$

The Spectronic 20 instrument is shown in Figure 8. The control knobs are black and the meter has a mirror scale graduated from 0-100% in 1% divisions and from 0 to infinite optical density (logarithmic scale is in red). The wavelength scale is graduated from 350nm-1000 nm by 5nm increments with numbered major divisions every 10nm. The wavelength control knob lies on top of the spectrophotometer while the zero control and the 100% transmittance control lie on the front panel.

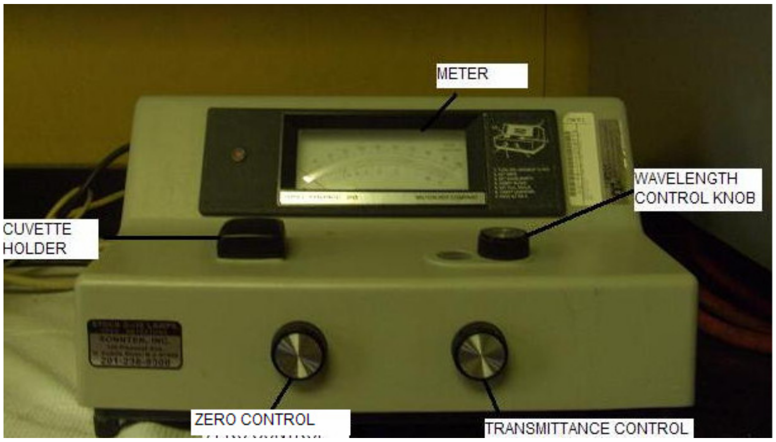

Figure 8 Spectrophotometer spectronic 20.

In order to measure the iodine concentration in a sample the maximum absorption wavelength of iodine had to be determined. The maximum absorption wavelength was found by measuring a diluted standard iodine solution. The sample was measured at wavelengths between 380nm to 650 nm. Before every measurement the machine was adjusted for 100 percent transmittance with a cuvette of deionized water. The absorbance for each of the wavelengths was noted and a plot of absorption versus wavelength was constructed using Excel. The wavelength at which the maximum absorption occurred was used to measure all iodine samples.

A calibration curve was constructed for measuring iodine concentrations in the sample solutions. A standard solution of iodine was prepared using the method of Kolthoff and Sandell (Kolthoff et al. 1952) described later. A series of dilutions were prepared from the standard solution and their optical densities were measured at the maximum absorption wavelength of 420nm. The data obtained were used to plot the change in optical density against iodine concentration. A linear curve fit for the data points generated a linear equation, as expected, and the equation of this line was used to find the concentration of iodine in unknown solutions.

Some experiments were also analyzed using neutron activation analysis. The reactor at the Nuclear Science Center was used activate sample solutions using a pneumatic system shown in Figure 9. During neutron activation I-127 undergoes an n, γ reaction to form I-128 and the activity of I-128 is compared against standards activated the same way to find the concentration of iodine.

Figure 9 Pneumatic system at the laboratory of NSC.

Using the pneumatic system, the samples were transport to reactor core position D2. There they were subjected to a thermal flux of about $10^{13}cm^{-2}sec^{-1}$ for about 1 min and then transported back to the laboratory receiver. All the samples were homogenous and there were at least two representative sample units for a single sample. Since liquid samples could vaporize by thermal and radiation heating during irradiation, an expanse volume was included within the sample vials. Each sample vial (shown in Figure 10) was packed inside a bigger transfer vial.

The space between the sample vial and the mouth of transfer vial was packed with foam to maintain proper geometry during irradiation. A standard sample with a known amount of iodine was irradiated under the same conditions as the unknown sample solutions. Since the samples were irradiated for short period of time, the standard and the sample were exposed within few minutes of each other insuring negligible differences in the exposures.

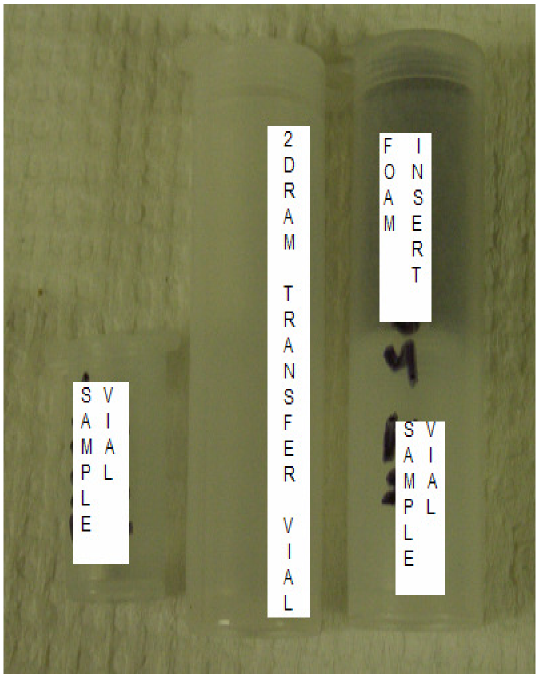

Figure 10 Pneumatic irradiation sample configuration.

After activation the samples were counted to determine I-128 activity. Counting geometries were chosen to collect as many counts as possible while limiting dead time. Both the standard and the sample were counted on the same geometry and at same distance from the detector. The distance between the sample and the detector can be varied by using different stands as shown in Figure 11. The GENIE2000 software was used for analyzing the data obtained from the detector. Besides the usual energy and absolute calibrations against NIST tracable sources, the simple ratio between the known and unknown samples gave the amount of I-128.

Figure 11 Stands of different heights for DET4.

REAGENTS USED FOR ANALYSIS

0.1N Sodium hydroxide (NaOH) solution: 1 ml of a 10N sodium hydroxide reagent solution from Sigma Aldrich was diluted with 99 ml of distilled water to prepare 0.1N sodium hydroxide solution.

1N Hydrochloric acid (HCl): 4 ml of concentrated hydrochloric acid (36% w/w, $\rho = 1.79$ gm.cc^{-1}) from Merck was diluted with 46 ml of distilled water to prepare a solution of 1N hydrochloric acid.

3% Hydrogen peroxide solution (H_20_2): 3% grade hydrogen peroxide was used.

0.01N Standard I-127 solution: 1.27 gram of reagent quality iodine was placed in a 250 ml beaker. 4 grams of potassium iodide was added to it followed by 2.5ml water. The iodine was stirred occasionally to dissolve. When all the iodine had dissolved the solution was diluted to approximately 1liter.

Potassium iodide (KI) solution: 10 ml. of 10% (w/v) potassium iodide solution from Fisher Scientific was added to 90 ml of water to prepare a potassium iodide solution with concentration of 10 milligrams of potassium iodide ml^{-1}.1 ml of this was diluted with 9 ml of water to form KI solution with a concentration of 1 milligram of KI ml^{-1}.

EXPERIMENTAL METHODS ADAPTED FOR THE SECTROPHOTOMETRIC AND NEUTRON ACTIVATION ANALYSIS IN NON-RADIOACTIVE PHASE

Few milligrams of natural iodine was carefully weighed into an aluminum canister. Figure 12 shows the aluminum canister that was used. About 0.5ml of water was added to the canister and the canister was heated with a hot air gun to about 180 C to allow the iodine vapors to react with the cylinder walls. The cylinder was allowed to cool and 10 ml of 0.1 N sodium hydroxide solution was added to the canister and the solution was left to equilibrate and then shaken to rinse the walls with sodium hydroxide. The washes were collected and the process repeated at least three times to ensure complete removal of the iodine from the canister. Throughout the text, the word ‘washing’ or ‘extracted solution’ has been used to refer to such cylinder washes.

Figure 12 The aluminum cylinder.

Possible reactions occurring in the extraction are given in Equations 8 and 9.

$$2NaOH + Al_2O_3 + 3H_2O \rightarrow \left[Al(OH_4)\right]^- + 2Na^+ \qquad (8)$$

$$AlCl_3 + 3NaOH \rightarrow Al(OH)_3 + 3NaCl \qquad (9)$$

To measure the iodine content, the iodide and the iodate ions in the washes were converted to I_2 using hydrochloric acid. The aluminum hydroxide precipitate was dissolved using about 2 ml of 1N hydrochloric acid.

Since aluminum chloride is a Lewis acid, it reacts with the excess of sodium hydroxide to reform aluminum hydroxide. Therefore care was taken to completely neutralize the sodium hydroxide solution and to leave an excess of H^+ ions in the solution. The H^+ ions react with I^- and IO_3^- according to Equation 10 to form elemental iodine.

$$IO_3^- + 5I^- + 6H^+ \rightarrow 3I_2 + 3H_2O \qquad (10)$$

However, the chance of forming iodate from hypoiodite is usually quite low for such low concentration of sodium hydroxide. The chain of reactions preceding the formation of I- and IO- ions has been explained in Equation 4.

The excess acid changed the solution to orange-yellow, typical of free iodine. The solution was well mixed and a 7 ml aliquot was transferred into a cuvette and the absorption measured at 420nm. To account for the interference from aluminum and sodium ions, blanks were prepared. For the blanks, the aluminum canisters were washed using 10ml of 0.1N sodium hydroxide, 2ml of 1N HCl was added to the wash and stirred. This dissolved the aluminum hydroxide precipitate and provided a clear solution. The solution was then well mixed and a 7 ml aliquot was used as the blank. The transmittance was set to 100% for the blank and the test solutions measured against this blank. The optical density was noted and the concentration of the sample was calculated using the iodine calibration curve. This process was repeated several times with 5 and 6 milligrams of iodine in aluminum cylinder to examine the accuracy of the experimental results.

Distillation: Distillation was carried out with solutions extracted from aluminum cylinders. NAA was performed on the distillate and the results were used to calculate the amount of iodine in the distillate. Comparison of the iodine in the distillate to the amount of iodine weighed into the aluminum cylinder gave the percentage recovery of iodine. Table 6 sets forth the NAA results for the extracted solution. Another set of distillations (Table 7) was carried out with potassium iodide solution instead of the extracted solution. The exact amount of iodine in the initial solution must be known in order to calculate the percentage iodine recovery by the process of distillation alone. To avoid the error associated with weighing milligram quantities of iodine into aluminum cylinders, a volumetric approach was adapted. A potassium iodide solution was prepared according to the procedure described before and the iodide in KI was converted to iodine using acidified solution of hydrogen peroxide. Hydrogen peroxide reacts with iodide according to Equation 3 Since 1gram of potassium iodide results in 0.76 gram of iodide ions; about 1ml of the prepared KI solution having about 0.76 milligrams of iodide was transferred into the distillation flask with an automated pipette. Ten ml of sodium hydroxide solution washed out from empty aluminum canisters was added to replicate the presence of aluminum hydroxide and sodium ions expected to be present in the washes during actual I-125 production. About 3 ml of 1N HCl was then added to neutralize the alkali and make the solution acidic. Under acidic condition the iodide is converted to elemental iodine following Equation 3.The volume was made up to about 24 ml and a distillation unit as shown in Figure 13 was assembled to distill the iodine over into a 50 ml round bottom flask.

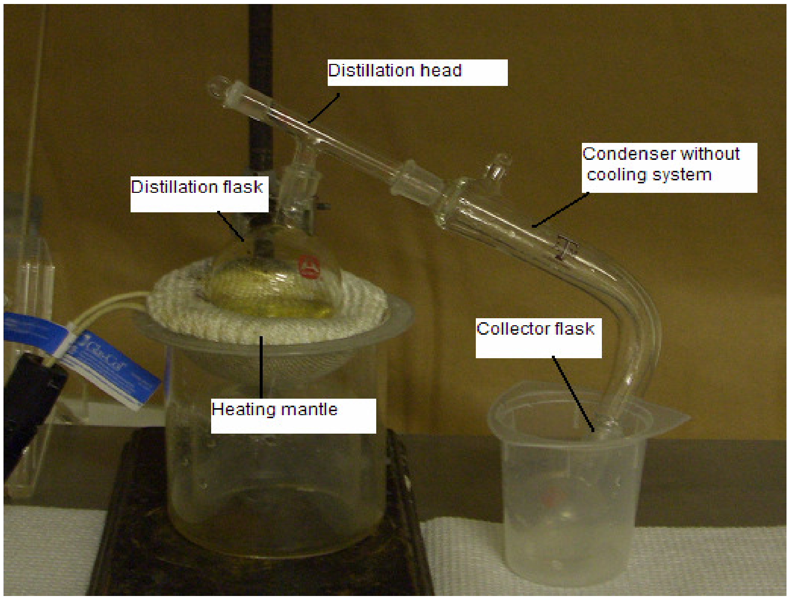

Figure 13 The distilling unit.

Distillation was carried out using Sigma Aldrich's mini distillation kit and a special order condenser. The heating mantle showed in the picture is from Glass-Kol and is used with a variac to control the heating rate. During distillation of iodine there was no forced cooling (water flow) through the condenser. Heat was removed from the distillation head with wet tissue paper occasionally. Care was taken to maintain a high temperature inside the distillation unit to prevent the condensation of iodine on the walls of the distillation head or the condenser. The small plastic beaker (Figure 13) holding the collector vessel was filled with liquid nitrogen. The liquid nitrogen created a very low temperature inside the collector vessel and this allowed the iodine vapors to be drawn in quickly to the collector vessel once there was enough vapor pressure inside the distillation flask. The variac was maintained at a 130 V for the initial 12 minutes of distillation but was turned down 110V once there was enough vapor pressure inside the flask. At the end of distillation, the distillate was collected and 0.01N potassium hydroxide solution was added to provide a homogenous solution of I^- ions for ease of sampling.

NAA of distillate: At the end of distillation, 0.1 ml aliquots of the distillate were transferred into the small polyethylene vials shown in Figure 11 and packed as described before. Two such vials were prepared for a single set of distillate and analyzed by activation. The vials were irradiated in the core for about 1 min. A standard sample having a known amount of iodine was prepared and analyzed also. The activated samples were counted with HPGe detectors in standard geometries.

Once the constituents of sample were identified the activity of I-128 was obtained from the spectrum setting the appropriate region of interest. The software calculates the activity for the sample acquisition time and corrects for decay while counting.

The concentration of I-128 was estimated from the specific activities for the sample and the standard as shown in Equation 11. C_{sam} and C_{std} are the amounts of the sample and standard respectively. A_{sam} denotes the specific activity of the sample and A_{std} is the specific activity for the standard.

$$C_{sam} = \frac{A_{sam}}{A_{std}}.C_{std} \tag{11}$$

The estimated concentration of I-128 was for the mass of sample solution in the vial. Adjusting it to the total weight of the distillate collected gave us the amount of iodine that could be recovered following distillation. A sample calculation for the above has is explained in Appendix A.

EXPERIMENTAL METHODS APPLIED FOR GAMMA SPECTROSCOPY IN THE RADIOIACTIVE PHASE

Distillation was carried out with a radioactive tracer. A glove box as shown in Figure 14 was used for distilling radioactive iodine. 1 ml of 20 milligrams potassium iodide sample solution was activated using neutron activation. After 1 min irradiation, neutron activation produced trace quantities of iodine-128.After sufficient decay time the sample was analyzed in a HPGe detector using GENIE 2000 software. The sample acquisition time was noted and the activity was recorded after correcting to the end of irradiation time following equation in Appendix B. For distillation, 0.5 ml of the irradiated potassium iodide solution was transferred to a glass vial filled with 4ml of sodium hydroxide solution .The solution was again counted using point source geometry and appropriate efficiency data. The time of acquisition was noted and the activity was recorded after correcting to the end of irradiation time for the sample. Thereafter the solution was transferred to a distillation flask followed by addition of 0.1 ml of 1N HCl acid and 0.5 ml of 3% hydrogen peroxide. The peroxide oxidized the iodide into iodine and gave the solution a rich yellow color.

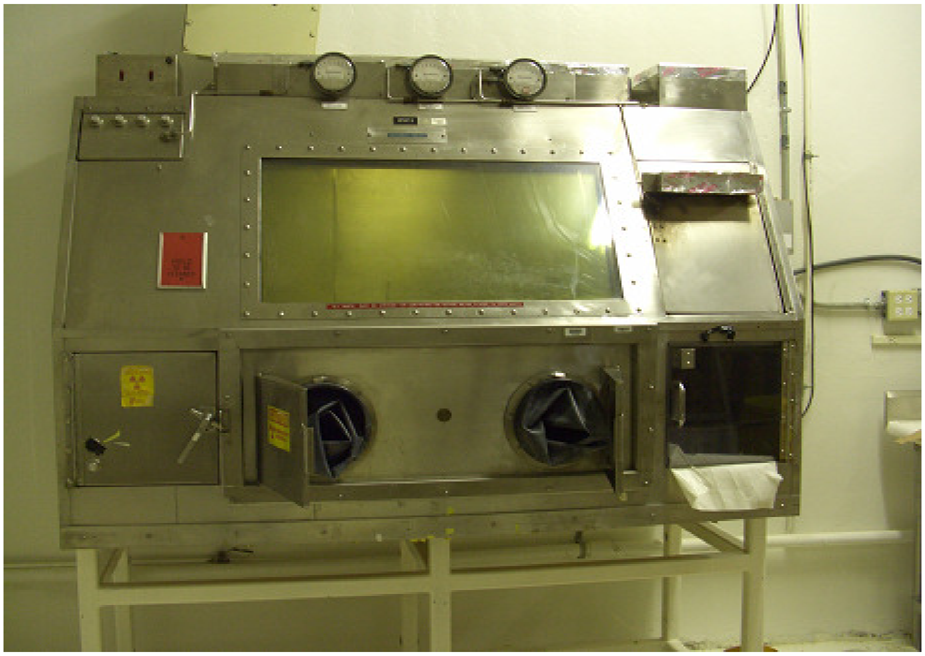

Figure 14 Glove box used to carry out distillation of radioactive iodine.

Distillation was carried out and about 7 ml of distillate was collected in a 50ml round bottom flask with a diameter of 2.4 inches. In order to ensure a uniform distribution of iodine, few milliliters of 0.1 N sodium thiosulfate solution was added to the distillate. The whole distillate in the flask was counted on a 24 inch stand which placed the source (distillate) at a distance of 24 inches from the face of the detector. The efficiency calibration file for 24 inch stand was loaded into the sample spectrum before counting. Counting was carried out for 10 mins, the acquisition time was noted and corrected to the end of irradiation time for the sample. Two sample vials with 0.1 ml of the distillate were also counted to substantiate the results from whole distillate counting. Comparison of the activities for the distilling solution and the distillate provided an estimate of the recovered iodine in the distillation.

RESULTS

The transmittance as a function of wavelength was measured using a diluted standard solution of iodine. The transmittance was converted to absorbance by taking the logarithm of the transmittance and Figure 15 shows a plot of these measurements. Inspection shows 420 nm to be the maximum absorption wavelength for the iodine solution.

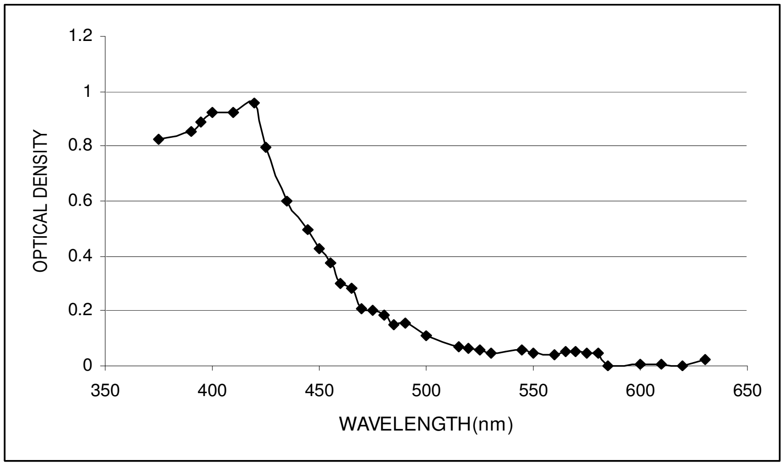

Figure 15 Plot of optical density versus wavelength for iodine solution.

A calibration curve was constructed using 420 nm and differing concentrations of iodine solutions. Table 4 shows the different concentrations of the standard iodine solutions that were measured. Percent transmittance was measured for each concentration and the absorption was calculated as before. Because absorbance varies directly as the concentration, a linear curve fit the data points. The graph generated in Figure 16 shows the linear equation for the plot. Concentration of iodine for unknown samples was measured using the plot.

Table 4 Absorbance versus iodine concentration.

Absorbance	Measured iodine concentration (µg/7ml).
1.045	889
1	825.2
0.959	762
0.92	698
0.886	635
0.795	571.5
0.769	508
0.6989	381
0.638	317
0.585	254

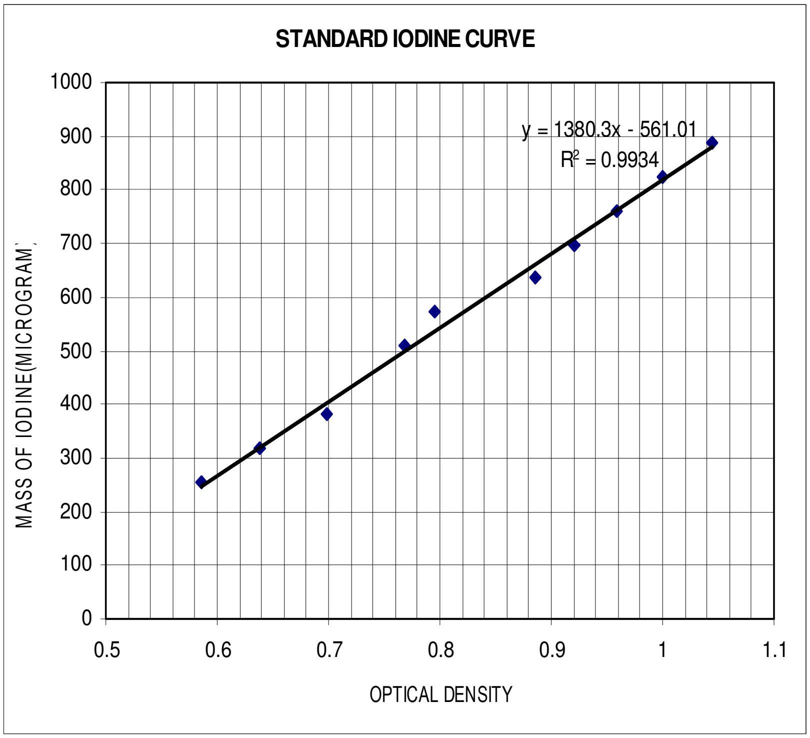

Figure 16 Iodine calibration curve.

To measure iodine content in extracted solution from aluminum cylinders, the absorbance for the particular solution was recorded following procedures described above. The amount of iodine was calculated using the linear equation in the above plot. At least three replicate measurements were recorded for each experimental run and the average of the three was taken as the expected concentration of iodine. This value, adjusted for the total volume of the extracted solution, was the total amount of iodine that was washed out. Experiments were repeated with 5 and 6 milligrams of I-127. Table 5 shows the percent recovery of iodine following spectroscopic measurements. A more explicit table for the process is shown in appendix C.

Table 5 Recovery of iodine versus initial mass (measured by spectrometry).

Experiment no.	Initial mass of iodine in cylinder (mg)	Recovered amount of iodine in washout (mg)	Percent recovery of iodine
1.00	6.01	5.52	
2.00	6.00	4.89	
3.00	6.10	5.19	
4.00	6.02	4.89	
Average	**6.03**	**5.120**	**84.86**
5.00	5.13	4.37	
6.00	5.02	4.86	
7.00	5.00	4.11	
8.00	5.00	4.24	
Average	**5.04**	**4.397**	**87.28**

Iodine extracted from the aluminum cylinder was also measured using neutron activation analysis. The extracted solution was distilled and NAA was performed on the distillate. Table 6 shows the recovery percent following distillation. Minor details of the experiment are shown in appendix D. For neutron activation, two sample vials having 0.1 ml of distillate (represented by 'D' in appendix D) were transported into the reactor core by pneumatic system, irradiated for 1 minute and counted using the Genie2000 software. The activity was recorded. A standard KI solution with a known amount of iodine was also irradiated. The average of the specific activities for distillates (D) was compared with the average of the specific activities of KI standard (represented by KI Std.I and KI Std.II in appendix D). The amount of iodine in the sample vials was deduced using Equation 11. Concentration of iodine for the total volume of distillate was calculated by multiplying by the ratio of total distillate volume and aliquot volume. This provided for a measure of the amount of iodine recovered following the process of washing and distillation.

Table 6 Recovery of iodine following distillation of extracted solution (measured by NAA).

Experiment no.	Amount of iodine weighed into aluminum cylinders (mg)	Recovered amount of iodine in total volume of distillate (mg)	Percent recovery of iodine
1	6.99	5.066	72.448
2	7.001	5.254	75.023
3	6.995	4.914	70.249
Average	**6.995**	**5.078**	**72.591**
5	5.012	4.010	80.005
6	5.001	3.576	71.514
Average	**5.007**	**3.793**	**75.76**

A new set of experiments was conducted to establish the percent recovery of iodine by distillation alone. Iodide in potassium iodide solution was converted to iodine using hydrogen peroxide and distilled. Table 7 is the recovery percent of the distillation. An explicit table for the experiments has been shown in Appendix E. The letter S in the appendix represents the solution of potassium iodide used for distillation and the letter D stands for distillate. The distillate collected was made up to the same volume as the KI solution used for distillation. This made it easy to compare the specific activity of the solution and the distillate after NAA. NAA was done with two representative sample vials (having 0.1 ml) from each of ‘S’ and ‘D’. A standard sample with known quantity of iodine (KI std.I and KI std. II) was used for calculating iodine amount in the distillate and solution. Relative iodine concentration in total volume of sample and distillate provided the percent recovery of iodine.

Table 7 Iodine recovery from distilling I-127 in potassium iodide solution (measured by NAA).

Experiment no.	Measured initial mass of iodine in KI solution (mg)	Standard error	Recovered mass of iodine in distillate (mg)	Standard error	Percent recovery	Error in percent recovery
1	1.13E+00	4.62E-02	1.05E+00	4.46E-02	9.30E+01	5.50E+00
2	1.09E+00	4.48E-02	1.01E+00	4.26E-02	9.34E+01	5.49E+00
3	1.11E+00	4.64E-02	9.79E-01	4.12E-02	8.85E+01	5.26E+00
4	1.11E+00	4.70E-02	9.50E-01	3.87E-02	8.53E+01	5.00E+00
Average	**1.11E+00**		**9.98E-01**		**9.00E+01**	

Experiments for the radioactive phase were carried out with an I-128 tracer. Table 8 shows the recovery for distillation carried out using this tracer. Four experiments were carried out with initial masses of iodine. The recovery percent for the different masses is shown in Table 8. Details for each experiment are in Appendices F, G, H and I. For each of the four radioactive experiments, 1 ml of known quantity of potassium iodide was irradiated. This formed trace quantities of iodine-128.A few milliliters of this iodine solution with tracer was then transferred to a distillation flask to be distilled. This solution is the ‘potassium iodide solution for distillation’ in the appendices. From the measurement of the activities for each, the mass of iodine can be calculated. Distillation was carried out with the ‘KI solution for distillation’ and the distillates were counted and compared by two different methods. Counting two aliquots designated ‘Distillate-I’ and ‘Distillate-II’ was one way of measuring the activity for the whole distillate. The other was counting the entire volume of distillate in a round bottom flask (D-24). To maintain point source geometry the flask was placed about 24 inches away from the face of the detector. All activities were corrected to the end of irradiation time. Comparison of the total mass of iodine in KI solution with the amount of iodine in total mass of distillate specified the percent recovery of iodine through distillation.

Table 8 Recovery of iodine from distillation of I-128 tracer in KI solution (measured by gamma spectroscopy).

Experiment No.	Initial iodine in KI solution		Recovered iodine in distillate				Recovery percent		Percent error in recovery	
	Measured mass (mg)	Standard error	Average mass from D1 and D2 (mg)	Standard error	Measured mass from D-24 (mg)	Standard Error	D1 and D2	D-24	D1 and D2	D-24
1	**7.60E+00**	4.34E-01	**6.95E+00**	3.89E-01	**7.36E+00**	4.35E-01	**9.07E+01**	**9.38E+01**	7.26E+00	7.96E+00
2	**7.80E+00**	2.45E-01	**6.98E+00**	5.58E-01	**7.17E+00**	4.88E-01	**8.95E+01**	**9.19E+01**	7.68E+00	6.89E+00
3	**1.37E+01**	4.40E-01	**1.21E+01**	9.89E-01	**1.20E+01**	5.95E-01	**8.86E+01**	**8.75E+01**	7.77E+00	2.82E+00
4	**7.60E-01**	2.02E-02	**6.94E-01**	0.0218454	**7.09E-01**	0.026739	**9.13E+01**	**9.33E+01**	3.76E+00	4.30E+00
Average							9.00E+01	9.16E+01		

DISCUSSION

This research focused on testing a novel I-125 recovery technique. Using aluminum cylinders for irradiation and using distillation as a purifying procedure are relatively new ideas in the field of iodine-125 production. Results suggest both ideas work well in terms of productivity and simplicity. Sodium hydroxide can be used as an extracting solvent in spite of its reactivity with the aluminum walls.

Spectrometry was used to quantify the amount of iodine that was extracted with sodium hydroxide. This was a quick, simple way of estimating percent iodine recovery. Experiments were conducted with relatively large amount of iodine owing to the difficulty in measuring very small amounts of solid iodine. Percent recovery was checked for differing masses of I-127 in the cylinder. The average iodine content in aluminum wash for an average initial amount of 6.01 milligram (Table 5) is 5.12 milligrams with a standard deviation of 0.301 milligrams. This shows an average recovery of about 84 percent. Recovery is 87 percent for an average initial amount of 5 milligrams. But, it is difficult to quantify the exact amount of iodine weighed into the cylinders. Calibration data suggest the OHAUS balance has at least 20 percent error in reporting milligram quantities of material. Apart from the volatility of iodine which can cause an error in weighing, there is al least 20 percent error in the weights of iodine recorded by OHAUS. This is likely to change the calculation of percent recovery. Considering 20 percent error, the percent recovery for the average initial amount of 5 milligrams probably ranges from 83 percent to 90 percent. For the same reason, the percent recovery for the initial amount of 6.01 milligrams ranges from 82 percent to 87 percent.

Iodine content in the extracted solution was also measured by NAA. The washout was distilled and NAA of the distillate was performed. This was an indirect way of estimating the iodine amount. A more direct way would be neutron activation of the solution itself, but the extracted solution was inhomogeneous and created considerable difficulty in sampling. As before, recovery percent was checked for differing quantities of iodine in the cylinder. The amount of iodine in the distillate was compared to the initial amount of iodine that was weighed into the cylinders. Table 6 shows that for an average of 7.11 milligrams of iodine the recovery is 5.07 milligram, which is about 72 percent. But the rest 30 percent loss includes the loss incurred in cylinder washing as well as the loss in distillation. It was important to know the loss incurred in each of the processes. A new set of experiments was conducted to assess the recovery percent for distillation alone. Table 7 suggests that the percent recovery of iodine for distillation is quite high. Examination of all four experiments indicates that recovery varies from 85 percent to 93 percent. This varying range is likely because of variation in the process of distillation rather than any statistical error in the measurements. As shown in Appendix E, the standard error in the specific activity of the distilling solution and the distillate is largely consistent and centers around 3 percent. Error propagation shows that the error associated with measured iodine content in the distillate is reasonably low and is within 1 percent. The error in percent recovery for all four experiments in Table 7 is also remarkably consistent. Any noteworthy difference in recovery has therefore resulted from the process of distillation itself. Nevertheless, if the mean iodine content in the solution (Table 7) is compared to the mean iodine content in the distillate the overall recovery percent by distillation is about 90%.

Although it is difficult to avoid the inherent variation between any two distillations, careful distillation promises even higher yield. The chemical nature of iodine causes it to plate on the surface of the condenser while passing from distillation flask to the collector vessel. The non-radioactive phase was designed to work out such details associated with iodine chemistry. Experimental trials in the course of the research have shown that plating can be prevented by controlling the temperature in the condenser during the course of distillation. The usual practice for a typical distillation is to allow water jacketed cooling of the condenser. This ensures condensation of the vapors and prevents bursting of the distillation head from excessive vapor pressure. But iodine would plate on the walls of the condenser under such circumstances. Distillation techniques were therefore modified to suit this research. Distillation was performed without any forced cooling of the condenser; instead the collector vessel was placed on liquid nitrogen. The intention was to bring down the temperature inside the collector vessel so that the iodine vapors get drawn into the collector vessel driven by high pressure in the distillation flask. The liquid nitrogen also condensed the vapors once it reached the collector vessel. Adjustments were also made for a suitable distillation head. Figure 17 below shows a two way distillation adapter placed vertically into the distillation flask. Iodine distillation is difficult with a vertically placed distillation head. Because the vapor pressure of iodine increases rapidly with temperature, as distillation proceeds, a sudden high pressure is created inside the distillation head. This exerts enough pressure on the lid to make it act as a safety valve release, causing huge loss of iodine.

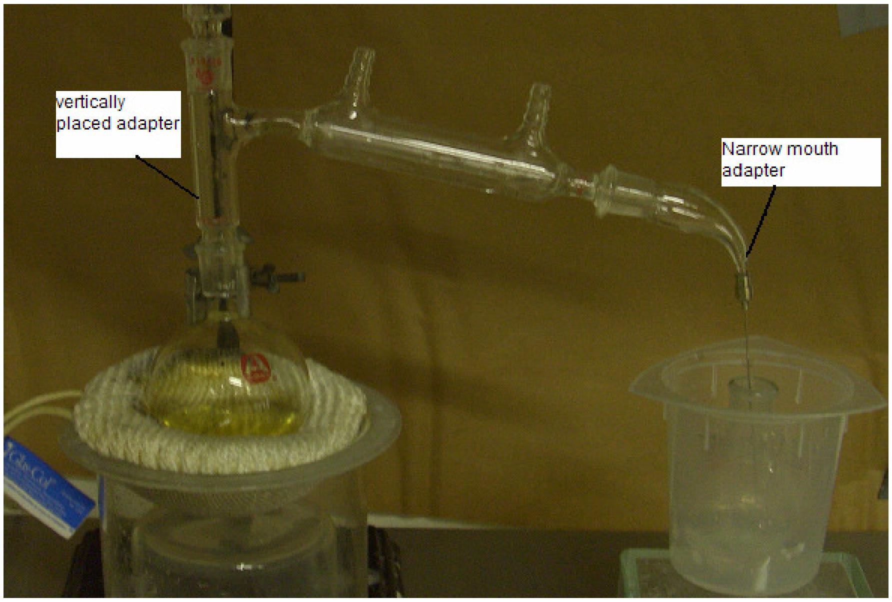

Figure 17 Distilling unit not suitable for iodine distillation.

Placing the distillation head horizontally as in Figure 18 prevents such bursting. Also, a narrow mouthed adapter attached with a needle (Figure 17) is not suitable for collection of iodine. Since iodine immediately transforms from the vapor phase to the solid phase, any narrow passage on its course allows the element to plate. The best alternative to a needle is therefore a wide mouthed condenser tube connecting the distillation head directly to the collector vessel through ground joints. This ensures a wide passage for the iodine vapors and prevents any loss.

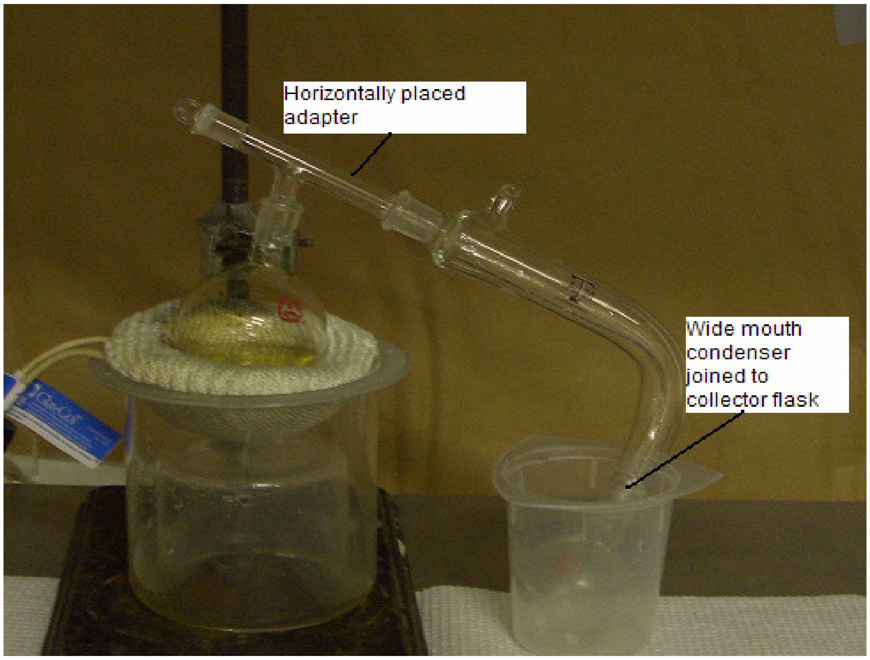

Figure 18 Distillation unit used for iodine distillation.

Once these details were worked out, experiments were conducted with iodine-128 tracer. The results for radioactive iodine distillation strongly corroborate the data for non-radioactive iodine distillation. The experiments were the same as that used in non-radioactive phase. Four sets of distillation were carried out with differing quantities of initial iodine (Tables 8, Appendices F,G,H and I). The results from all four suggest that recovery percent of iodine ranges from 88 to 96. For each experiment, two types of measurements were made. Counting two aliquots, designated ‘Distillate-1’ and ‘Distillate-2’ was one way of measuring the activity for the whole distillate. The other was counting the whole volume of distillate (named D-24) maintaining a point source geometry. Appendices F,G, H, I gives a detailed insight into how these measurements were made. Note, however, that there is a difference in the percent recovery calculated by the above two ways for the same amount of iodine distilled. For example, in experiment 1, the recovery percent for D-24 is 96 but the recovery percent for D1 and D2 is 90. Such difference exists for all four experiments done with I-128 tracer. It may have been caused by the difference in aliquot size for the two vials or because of error associated with weighing the vials. Since the average quantity of iodine (measured by D1 and D2) was dependent on the specific activities of the representative sample vials, any small difference in recorded sample mass for D1 and D2 could have caused a difference in calculation of specific activity. This would eventually make a difference in calculating average quantity of iodine. The measurements for D-24 were for the whole distillate and were free of such possibilities. D-24 therefore provided a better accuracy in iodine estimation.

In conclusion it can be said that higher yield of radioactive iodine is possible with simple distillation. The procedure for distillation is simple and can be carried out safely in a glove box. Spectroscopic data suggests that the distillate obtained is 100 percent pure iodine. There is no evidence to show that recovery varies with initial iodine mass. Recovery can be high with masses as high as 7.6 mg (Appendix F and I) and as low as 0.76 mg. Moreover, distillation can concentrate the amount of iodine extracted from the cylinders. A careful observation of the data presented in appendix F, G and H bears proof to the concentration achieved by distillation. The mass of the distillate collected is much less than the mass of the solution that was used for distilling. For experiment 1 in Appendix F, a mass of 38 grams of initial distilling solution resulted in 12 grams distillate. This shows that distillation can reduce the solution to almost one third or less of its original volume. The concentrated iodine can now be dissolved in buffered solution and the concentration can be adjusted based on market demands. Although the present research could not provide any conclusive evidence for the amount of iodine in the extracted solution results suggest that the recovery of iodine in the extracted solution is at least 84%. Future work with iodine-125 can provide more convincing data on the process.

CONCLUSION

There is a demand for iodine-125 in the present market. The present I-125 production companies suffer from certain problems that can be overcome using the results of this research. Using aluminum irradiation cylinder can reduce irradiation time for Xe-124 and this reduced irradiation time will help minimize I-126. Iodine in these aluminum cylinders can be extracted using sodium hydroxide. The concentration of the iodine can be increased by distillation. Thus distillation is a suitable means for both purifying and concentrating iodine-125. Distillation can reduce the extracted solution to one third or less of its original volume. The recovery percent of iodine during distillation ranges from 88 to 96 but careful distillation promises even higher recovery. There is no evidence to show that recovery varies with varying mass. Recovery can be high with mass as high as 7.6 milligrams (Appendix F) and mass as low as 0.76 milligram (Appendix I). The difference in recovery is mostly because of the process of distillation itself and not because of any statistical error in measurement. Distillation for I-125 differs slightly than conventional techniques. No forced cooling of the condenser is necessary during distillation, for example.

Although results indicate that extraction of iodine is possible with sodium hydroxide this research could not provide conclusive evidence for the amount of iodine extracted. Spectrometric results suggest that recovery percent is high and is at least 84. But better analytical means are necessary to quantify the exact amount of iodine in the extracted solution. Making I-125 in the reactor and extracting it using procedures discussed in this work will help in better analysis. The whole process of extraction and distillation needs to be automated for large scale production. The tested principles should be applied on an industrial scale for successful production of iodine-125.

REFERENCES

Adelstein SJ, Manning FJ, eds. Isotopes for medicine and life sciences. Washington, DC: The National Academic Press; 1995.

American Nuclear Society. U.S radioisotope supply position statement # 30. 2004. Available at: http://www.ans.org/pi/ps/docs/ps30.pdf. Accessed 13 May, 2007.

Chapman EM. History of the discovery and early use of radioactive iodine**.** JAMA 250: 2042-2044; 1983.

Fermi E. Radioactivity induced by neutron bombardment. Nature 133: 757; 1934.

Harper PV, Siemens WD, Lathrop KA, Endlich H. Production and use of iodine-125. J Nucl Med 4: 277-89; 1963.

Hassal, Scott B. Method and apparatus for production of radioactive iodine. 1997. Available at http://www.wikipatents.com/5633900.html. Accessed 13 May 2007.

Hertz S, Roberts A, Evans RD. Radioactive iodine as an indicator in the study of thyroid physiology. Proc Soc Exp Biol Med 38: 510; 1938.

Kahn M, Kleinberg J. Radiochemistry of iodine. Virginia. National Academy of Sciences National Research Council; 1977.

Kolthoff IM, Sandell EB. Textbook of quantitative inorganic analysis. 3rd ed. New York: The Macmillan Company; 1952.

Livingood JJ, Seaborg GT. Radioactive isotopes of iodine. Phys Rev 54: 775-782; 1938.

Martinho E, Neves MA, Freitas MC. Iodine-125 production: Neutron irradiation planning. J Appl. Radiat. Isot 35: 933-938; 1984.

MDS Nordion. Regulatory decision ensures supply of essential medical isotopes [news release online].Available at: http://www.nordion.com/documents/newsreleases/1999/NRCMDSN_Decicion_on_HEUExpor tPermit_06_29_99.pdf. Accessed 13 May 2007.

MDS Nordion. I-125 radiochemical sodium iodide solution. 2007. Available at http://www.mdsnordion.com. Accessed 13 May, 2007.

Myers WG. Radioiodine-125 in biomedicine. J Nucl Med 25: 1389-1391; 1984.

Nuclear Energy Research Advisory Committee. Expert panel: Forecast future demand for medical isotopes. March 1999. Available at www.ne.doe.gov/nerac/neracPDFs/isotopedemand.pdf. Accessed 13 May, 2007.

Rosenfield L. Discovery and early uses of iodine. J Chem Ed 77: 984-987; 2000.

Stabin M. Nuclear medicine dosimetry. Phys Med Biol 51: R187-R202; 2006.

Studsvik. Studsvik 1998. Available at: http://www.studsvik.se/files/Studsvik_Annualreport1998_en.pdf. Accessed 13 May, 2007.

Tenforde TS. Medical radionuclide supplies and national policy: time for a change? AJR 182: 575-577; 2004.

U.S. Department of Energy. Office of Inspector General Office of Audit Services. Available at: http://www.ig.energy.gov/documents/CalendarYear2005/ig-0709.pdf. Accessed 13 May, 2007.

U.S. Department of Energy. Office of Scientific and Technical Information. Overview of U.S Department of Energy isotope program. 2004. Available at http://www.osti.gov/bridge/servlets/purl/840075-Qk6OFq/native/840075.pdf. Accessed 13 May, 2007.

APPENDIX A

SAMPLE CALCULATION OF I-128 CONCENTRATION FROM SPECIFIC ACTIVITY

$$M_{sam} = \frac{A_{sam}}{A_{std}} . M_{std} = \frac{7.20E+1}{2.16E+2} * 0.152 = 0.051$$

Here M_{sam} and M_{std} are the mass of the sample and standard respectively. A_{sam} denotes the specific activity of the sample and A_{std} is the specific activity for the standard.

S.N	Sample ID	Type	Sample mass (mg)	Irradiation time (sec)	Counting time (sec)	Average specific activity of I (µCi/g)		Amount of iodine in sample mass (mg)
						Wt. mean	Error	
1	KI Std I	KI Std	100.5000	60	240	2.19E+02	4.97E+00	
	KI Std II	KI Std	100.9000	60	240	2.13E+02	3.76E+00	
Average iodine content in potassium iodide standard						**2.16E+02**	4.36E+00	**1.52E-01**
S.N	Amount of iodine weighed into aluminum cylinders (mg)	Total volume of distillate collected (ml)	Specific activity for measured volume of distillate (µCi/g)	Amount of iodine in measured volume of distillate (mg)	Iodine content in total volume of distillate (mg)	Average of iodine content in total volume of distillate (mg)	Average of iodine weighed in aluminum cylinders (mg)	Percentage recovery of iodine
1	6.99	10	7.40E+01	0.052	5.208			
		10	7.00E+01	0.049	4.924			
Average iodine content in D-1			**7.20E+01**	**0.051**	**5.066**			72.448

APPENDIX B

SAMPLE CALCULATION FOR ACTIVITY CORRECTION TO END OF IRRADIATION TIME

$$A = A_0 e^{-\lambda t} \rightarrow \frac{A}{e^{-\lambda t}} = \frac{8.52E+2}{e^{-0.693/25 * 47}} = 3.10\text{E+3}$$

Where A is the activity at the time of sample acquisition, A_o is the activity at the end of irradiation time for the sample, λ is the disintegration constant, t is the time difference between end of irradiation time and acquisition time. The acquisition time for example below is 10:18 AM and the end of irradiation time is 9:31AM.

Experiment number	Sample ID	Mass used for analysis (g)	Specific activity (µCi/gm) at 10:18:02AM		Specific activity (µCi/gm) at 9:31:29AM	
			Activity	Uncertainty	Activity	Uncertainty
1	Irradiated KI soln.	0.9937	8.52E+02	2.16E+01	3.10E+03	7.87E+01

APPENDIX C

SPECTROMETRIC MEASUREMENT OF IODINE CONTENT IN EXTRACTED SOLUTION

Experiment no.	Initial iodine mass in cylinder (mg)	Total cylinder wash (ml)	Volume of sample measured (ml)	No. of measurements	Optical density of sample	Iodine in sample (mg)	Iodine mass in total cylinder wash (mg)	Standard error	Mean of iodine mass in wash(mg)	Mean of initial iodine mass in cylinder(mg)	Percent recovery of iodine
	6.01	54.00	7.00	I	0.92	0.70	5.38				
1				II	0.96	0.75	5.79				
				III	0.92	0.70	5.38				
Average							**5.52**	0.13			
	6.00	54.00	7.00	I	0.85	0.62	4.78				
2				II	0.85	0.62	4.78				
				III	0.89	0.66	5.09				
Average							**4.89**	0.10			
	6.10	54.00	7.00	I	0.89	0.66	5.09				
3				II	0.92	0.70	5.38				
				III	0.89	0.66	5.09				
Average							**5.19**	0.10			
	6.02	54.00	7.00	I	0.89	0.55	5.09				
4				II	0.85	0.55	4.78				
				III	0.85	0.55	4.78				
Average							**4.89**	0.10			
									5.12	**6.03**	**84.86**

Continued.

Experiment no.	Initial iodine mass in cylinder (mg)	Total cylinder wash (ml)	Volume of sample measured (ml)	No. of measurements	Optical density of sample	Iodine in sample(mg)	Iodine mass in total cylinder wash (mg)	Standard error	Mean of iodine mass in wash(mg)	Mean of initial iodine mass in cylinder(mg)	Percent recovery of iodine
	5.13	45.00	7.00	I	0.82	0.55	4.24				
5				II	0.80	0.60	4.63				
				III	0.82	0.55	4.24				
Average							**4.37**	0.13			
	5.02	45.00	7.00	I	0.85	0.62	4.78				
6				II	0.85	0.62	4.78				
				III	0.89	0.65	5.01				
Average							**4.86**	0.08			
	5.00	45.00	7.00	I	0.80	0.60	4.63				
7				II	0.77	0.50	3.86				
				III	0.77	0.50	3.86				
Average							**4.11**	0.26			
	5.00	45.00	7.00	I	0.82	0.55	4.24				
8				II	0.82	0.55	4.24				
				III	0.82	0.55	4.24				
Average							**4.24**	0.00	**4.40**	**5.04**	**87.28**

APPENDIX D

NAA OF DISTILLATE FROM DISTILLATION OF CYLINDER WASH

S.N	Sample ID	Type	Sample mass (mg)	Irradiation time (sec)	Measuring time (sec)	Specific activity of I (µCi/g)		Amount of iodine in sample mass (mg)						
						Wt. mean activity	Standard deviation							
1.00E+00	KI Std I	KI Std	1.01E+02	6.00E+01	2.40E+02	2.19E+02	4.97E+00							
	KI Std II	KI Std	1.01E+02	6.00E+01	2.40E+02	2.13E+02	3.76E+00							
	Average iodine content in potassium iodide					**2.16E+02**	**4.36E+00**	**1.52E-01**						
S.N	Initial iodine content in aluminum cylinders (mg)	Sample ID	Type	Total volume of distillate (ml)	Sample volume for NAA (ml)	Sample mass (mg)	Specific activity of I (µCi/g)		Iodine content in sample volume (%)		Total iodine content in distillate volume(mg)	Average of iodine in distillate content(mg)	Average of initial iodine content in aluminum cylinders(mg)	Percent recovery of iodine
							Wt. mean activity	Standard error	Amount(mg)	Standard error				
1.00E+00	6.99E+00	D-Ia	Distillate	1.00E+01	1.00E-01	1.02E+02	7.40E+01	1.63E+00	5.21E-02	1.55E-03	5.21E+00			
		D-Ib	Distillate	1.00E+01	1.00E-01	1.04E+02	7.00E+01	1.73E+00	4.92E-02	1.57E-03	4.92E+00			
Average iodine content in D-I							**7.20E+01**	**1.68E+00**	**5.07E-02**	**1.56E-03**	**5.07E+00**			7.25E-01
2.00E+00	7.00E+00	D-IIa	Distillate	1.15E+01	1.00E-01	1.05E+02	7.10E+01	1.55E+00	4.99E-02	1.49E-03	5.74E+00			
		D-IIb	Distillate	1.15E+01	1.00E-01	1.05E+02	5.89E+01	1.31E+00	4.14E-02	1.25E-03	4.76E+00			
Average iodine content in D-II							**6.49E+01**	**1.43E+00**	**4.57E-02**	**1.37E-03**	**5.25E+00**			7.50E-01
3.00E+00	7.00E+00	D-IIIa	Distillate	1.10E+01	1.00E-01	1.02E+02	7.29E+01	1.59E+00	5.13E-02	1.52E-03	5.64E+00			
		D-IIIb	Distillate	1.10E+01	1.00E-01	1.06E+02	5.41E+01	5.70E-02	3.81E-02	7.70E-04	4.19E+00			
Average iodine content in D-III							**6.35E+01**	**8.22E-01**	**4.47E-02**	**1.15E-03**	**4.91E+00**			7.02E-01
												5.08E+00	**7.00E+00**	**7.26E-01**

Continued.

S.N	Initial iodine content in aluminum cylinders (mg)	Sample ID	Type	Total volume of distillate (ml)	Sample volume for NAA (ml)	Sample mass (mg)	Specific activity of I (µCi/g)		Iodine content in sample volume (%)		Total iodine content in distillate volume(mg)	Average of iodine in distillate content(mg)	Average of initial iodine content in aluminum cylinders(mg)	Percent recovery of iodine
							Wt. mean activity	Standard error	Amount(mg)	Standard error				
5.00E+00	5.01E+00	D-IVa	Distillate	1.40E+01	1.00E-01	9.95E+01	4.03E+01	9.95E-01	2.83E-02	9.05E-04	3.97E+00			
		D-IVb	Distillate	1.40E+01	1.00E-01	1.01E+02	4.11E+01	9.93E-01	2.89E-02	9.11E-04	4.05E+00			
Average iodine content in D-IV							**4.07E+01**	**9.94E-01**	**2.86E-02**	**9.08E-04**	**4.01E+00**			8.00E-01
6.00E+00	5.00E+00	D-Va	Distillate	1.50E+01	1.00E-01	1.04E+02	3.10E+01	8.02E-01	2.18E-02	7.16E-04	3.27E+00			
		D-Vb	Distillate	1.50E+01	1.00E-01	1.02E+02	3.68E+01	9.11E-01	2.59E-02	8.28E-04	3.88E+00			
Average iodine content in D-V							**3.39E+01**	**8.57E-01**	**2.38E-02**	**7.72E-04**	**3.58E+00**			7.15E-01
												3.79E+00	**5.01E+00**	**7.58E-01**

APPENDIX E

NAA RESULTS FROM DISTILLING NON-RADIOACTIVE IODINE

Sample ID	Type	Sample mass (g) of 0.1 ml	Specific activity of I (µCi/g)		Amount of iodine in 0.1 ml of Std.(mg)
			Wt. mean activity	Error	
KI Std I	KI Std	9.95E-02	5.91E+04	1.47E+03	
KI Std II	KI Std	9.96E-02	5.71E+04	1.43E+03	
Average I Content in KI Std			5.81E+04	1.45E+03	1.20E-02

Sample ID	Type	Mass of iodine in solution to be distilled (mg)	Total volume of solution to be distilled (ml)	Total volume of distillate collected (ml)	Sample volume for NAA (ml)	Sample mass (g)	Specific activity of iodine (µCi/g)		Iodine Content in sample		Mass of iodine in distillate (mg)	Standard error	Percent recovery of iodine	Error in percent recovery
							Wt. mean activity	Standard error	Amount (mg)	Standard error				
S Ia	Solution	1.13E+00	2.40E+01		1.00E-01	1.00E-01	2.27E+04	7.35E+02	4.70E-03	1.92E-04				
S Ib	Solution	1.12E+00	2.40E+01		1.00E-01	1.00E-01	2.26E+04	7.41E+02	4.68E-03	1.93E-04				
Average I Content in Solution I							**2.27E+04**	7.38E+02	4.69E-03	1.92E-04	**1.13E+00**	4.62E-02		
D-Ia	Distillate			2.40E+01	1.00E-01	9.90E-02	2.10E+04	6.91E+02	4.34E-03	1.79E-04	1.04E+00			
D-Ib	Distillate			2.40E+01	1.00E-01	9.87E-02	2.12E+04	7.63E+02	4.38E-03	1.92E-04	1.05E+00			
Average I Content in D-I							**2.11E+04**	7.27E+02	4.36E-03	1.86E-04	**1.05E+00**	4.46E-02	**9.30E+01**	5.50E+00
S IIa	Solution	1.09E+00	2.40E+01		1.00E-01	9.89E-02	2.23E+04	7.24E+02	4.61E-03	1.89E-04				
S IIa	Solution	1.06E+00	2.40E+01		1.00E-01	9.79E-02	2.15E+04	7.14E+02	4.45E-03	1.85E-04				
Average I Content in Solution II							**2.19E+04**	7.19E+02	4.53E-03	1.87E-04	**1.07E+00**	4.48E-02		
D-IIa	Distillate			2.40E+01	1.00E-01	1.00E-01	2.14E+04	7.02E+02	4.43E-03	1.83E-04	1.06E+00			
D-IIb	Distillate			2.40E+01	1.00E-01	1.01E-01	1.95E+04	6.76E+02	4.03E-03	1.72E-04	9.67E-01			
Average I Content in D-II							**2.05E+04**	6.89E+02	4.23E-03	1.77E-04	**1.01E+00**	4.26E-02	**9.44E+01**	5.59E+00

Continued.

Sample ID	Type	Mass of iodine in solution to be distilled (mg)	Total volume of solution to be distilled (ml)	Total volume of distillate collected (ml)	Sample volume for NAA (ml)	Sample mass (g)	Specific activity of iodine (µCi/g)		Iodine Content in sample		Mass of iodine in distillate (mg)	Standard error	Percent recovery of iodine	Error in percent recovery
							Wt. mean activity	Standard error	Amount (mg)	Standard error				
S IIIa	Solution	1.11E+00	2.40E+01		1.00E-01	9.90E-02	2.21E+04	7.57E+02	4.56E-03	1.94E-04				
S IIIa	Solution	1.12E+00	2.40E+01		1.00E-01	9.79E-02	2.25E+04	7.45E+02	4.65E-03	1.93E-04				
Average I Content in Solution IV							**2.23E+04**	7.51E+02	4.61E-03	1.93E-04	**1.11E+00**	4.64E-02		
D-IIIa	Distillate			2.40E+01	1.00E-01	9.75E-02	1.72E+04	6.14E+02	3.55E-03	1.55E-04	8.51E-01			
D-IIIb	Distillate			2.40E+01	1.00E-01	9.86E-02	2.23E+04	7.20E+02	4.61E-03	1.88E-04	1.11E+00			
Average I Content in D-IV							**1.97E+04**	6.67E+02	4.08E-03	1.71E-04	**9.79E-01**	4.12E-02	**8.81E+01**	5.22E+00
S IVa	Solution	1.11E+00	2.40E+01		1.00E-01	9.80E-02	2.24E+04	7.57E+02	4.63E-03	1.95E-04				
S IVb	Solution	1.11E+00	2.40E+01		1.00E-01	9.78E-02	2.25E+04	7.69E+02	4.65E-03	1.97E-04				
Average I Content in Solution V							**2.25E+04**	7.63E+02	4.64E-03	1.96E-04	**1.11E+00**	4.70E-02		
D-IVa	Distillate			2.55E+01	1.00E-01	1.00E-01	1.95E+04	6.80E+02	4.03E-03	1.73E-04	1.03E+00			
D-IVb	Distillate			2.55E+01	1.00E-01	1.00E-01	1.66E+04	5.92E+02	3.42E-03	1.49E-04	8.73E-01			
Average I Content in D-V							**1.80E+04**	6.36E+02	3.73E-03	1.61E-04	**9.50E-01**	3.87E-02	**8.53E+01**	5.00E+00

APPENDIX F

DISTILLATION OF I-128 IN EXPERIMENT 1

Sample ID	Total mass of the solution that was distilled(g)	Total mass of distillate collected (g)	Mass used for analysis (g)	Iodine content (mg)	Counting geometry	Specific activity (µCi/gm) at 9:31:29AM		Total activty (µCi) at 9:39:29AM		Recovered amount of iodine in total mass of distillate (mg)	Standard error	Expected iodine content in total mass of distillate (mg)	Percent recovery of iodine
						Activity	Uncertainty	Activity	Uncertainty				
Irradiated KI soln.			9.94E-01	1.52E+01	36 inch stand point source	3.10E+03	7.87E+01	3.08E+03	7.87E+01				
Soln. for distillation	3.86E+01		4.97E-01	7.60E+00		1.55E+03	7.87E+01	1.54E+03	7.87E+01				
Distillate in vial D1		1.20E+01	9.70E-02		Stand 1 point source	1.23E+02	4.70E+00	1.47E+03	5.65E+01	7.27E+00	3.35E-01	7.60E+00	
Distillate in vial D2		1.20E+01	9.70E-02		Stand 1 point source	1.12E+02	4.84E+00	1.34E+03	5.82E+01	6.62E+00	4.44E-01	7.60E+00	
Average iodine content										**6.95E+00**	3.89E-01	**7.60E+00**	9.07E+01
Distillate in round bottom flask D24		1.20E+01	1.20E+01		24 inch stand point source			1.49E+03	7.96E+01	**7.36E+00**	4.35E-01	**7.60E+00**	9.68E+01

APPENDIX G

DISTILLATION OF I-128 IN EXPERIMENT 2

Sample ID	Total mass of the solution that was distilled(g)	Total mass of distillate collected (g)	Mass of sample analyed (g)	Iodine content (mg)	Counting geometry	Specific activity (μCi/gm) at 10:34:00 AM		Total activty (μCi) at 10:34:00 AM		Recovered amount of iodine in total mass of distillate (mg)	Standard error	Expected iodine content in total mass of distillate (mg)	Percent recovery of iodine
						Activity	Uncertainty	Activity	Uncertainty				
Irradiated KI-II soln.			1.00E+00	1.52E+01	24 inch point source	1.80E+03	4.02E+01	1.80E+03	4.02E+01				
Soln. for distillation	3.93E+01		5.21E-01	7.80E+00	Stand 5 point source	1.80E+03	4.02E+01	9.36E+02	2.07E+01				
Distillate in vial D1		1.36E+01	9.70E-02		Stand 1point source	6.19E+01	4.94E+00	8.41E+02	6.70E+01	7.09E+00	5.87E-01	7.80E+00	
Distillate in vial D2		1.36E+01	9.40E-02		Stand 1point source	6.06E+01	4.47E+00	8.24E+02	6.07E+01	6.87E+00	5.29E-01	7.80E+00	
Average iodine content										**6.98E+00**	5.58E-01	**7.80E+00**	8.95E+01
Distillate in round bottom flask D24		1.36E+01	1.36E+01		24 inch point source			8.51E+02	5.47E+01	**7.17E+00**	4.88E-01	**7.80E+00**	9.19E+01

APPENDIX H

DISTILLATION OF I-128 IN EXPERIMENT 3

Sample ID	Total mass of the solution that was distilled(g)	Total mass of distillate collected (g)	Mass of sample analyed (g)	Iodine content (mg)	Counting geometry	Specific activity (µCi/gm) at 10:31:23 AM		Total activty (µCi) at 10:31:23 AM		Recovered amount of iodine in total mass of distillate (mg)	Standard error	Expected iodine content in total mass of distillate (mg)	Percent recovery of iodine
						Activity	Uncertainty	Activity	Uncertainty				
Irradiated KI-III soln.			9.84E-01	1.52E+01	36 inch stand point source	3.70E+03	8.93E+01	3.63E+03	8.78E+01				
Soln. for distillation	4.60E+01		8.85E-01	1.37E+01	Stand 5 point source	3.70E+03	8.93E+01	3.27E+03	6.94E+01				
Distillate in vial D1		1.69E+01	1.00E-01		Stand 1point source	1.65E+02	1.24E+01	2.79E+03	2.09E+02	1.17E+01	9.19E-01	1.37E+01	
Distillate in vial D2		1.69E+01	9.80E-02		Stand 1point source	1.78E+02	1.45E+01	3.01E+03	2.45E+02	1.26E+01	1.06E+00	1.37E+01	
Average iodine content										**1.21E+01**	9.89E-01	**1.37E+01**	8.86E+01
Distillate in round bottom flask D24		1.69E+01	1.69E+01		24 inch point source			2.86E+03	1.24E+02	**1.20E+01**	5.95E-01	**1.37E+01**	8.75E+01

APPENDIX I

DISTILLATION OF I-128 IN EXPERIMENT 4

Sample ID	Total mass of the solution that was distilled(g)	Total mass of distillate collected (g)	Mass of sample analyed (g)	Iodine content (mg)	Counting geometry	Specific activity (µCi/gm) at 10:12 AM		Total activty (µCi) at 10:12AM		Recovereda mount of iodine in total mass of distillate (mg)	Standard error	Expected iodine content in total mass of distillate (mg)	Percent recovery of iodine
						Activity	Uncertainty	Activity	Uncertainty				
Irradiated KI-II soln.			9.99E-01	1.00E+00	Stand 5 point source	2.34E+02	4.09E+00	2.34E+02	4.09E+00				
Soln. for distillation	4.01E+01		7.69E-01	7.70E-01	Stand 5 point source	2.34E+02	4.09E+00	1.74E+02	3.64E+00				
Distillate in vial D1		1.57E+01	9.70E-02		Stand 1point source	9.10E+00	3.80E-01	1.43E+02	5.96E+00	6.32E-01	2.86E-02	7.70E-01	
Distillate in vial D2		1.57E+01	9.40E-02		Stand 1point source	1.04E+01	3.82E-01	1.63E+02	5.99E+00	7.21E-01	3.05E-02	7.70E-01	
Average iodine content										**6.77E-01**	2.96E-02	**7.70E-01**	8.79E+01
Distillate in round bottom flask D24		1.57E+01	1.57E+01		24 inch point source			1.62E+02	5.09E+00	**7.18E-01**	3.48E-02	**7.70E-01**	9.33E+01

VITA

Name: Adwitiya Kar

Address: Department of Nuclear Engineering
Texas A&M University- Mail Stop 3133
College Station, Texas-77843-3133

E mail address: adwitiya_kar@yahoo.co.in

Education: B.Sc. 2001, University of Calcutta, M.Sc. 2003, University of Calcutta, M.S., Health Physics, Texas A&M University.