STABLE ISOTOPES

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INTRODUCTION

Stable isotopes used in nuclear medicine and elsewhere come mainly from one supplier, Oak Ridge National Laboratory (ORNL) in the United States. They supply about 75% of the stable isotopes used in the western world with most of the remainder coming from the Soviet Union. This paper will describe the operation at ORNL and its limitations, present an overview of the various methods of enriching isotopes and conclude with a description of the programs at CRNL which involve research in isotope enrichment.

PRESENT SITUATION

Enriched stable isotopes have been supplied by ORNL since shortly after the end of the Second World War. The equipment used there is a modification of part of the CALUTRON (CALifornia University cycloTRON) facility constructed to provide uranium-235 for the Manhattan Project.

This facility consists of electromagnetic separators which have a small production capacity but are versatile enough to be tuned to allow enrichment of most of the stable isotopes, with varying degrees of purity. Isotopes of small natural abundance whose mass lies near that of a very abundant isotope are difficult to enrich to high isotopic purity using the CALUTRONS.

For the last few years, funding has not been sufficient to operate enough CALUTRONS to maintain a diverse inventory of isotopes and each time a price list is published many isotopes are not available. For example, the April 1984 price list showed that, of the about 60 elements with two or more isotopes, ORNL was out of stock for at least one isotope of 21 elements. Different isotopes are out of stock at different times, and it is likely that the situation will get worse. Our concern as Canadian users of stable isotopes is that it is very likely that foreign needs will be met only after US domestic needs are satisfied. An official of the United States Department of Energy (USDOE) has stated that while the users of stabler isotopes are international, the US Congress/DOE produces these isotopes for domestic needs and interests. Thus any shortages in supply will be felt outside the United States first.

The pricing policy at ORNL is such that when a new separation and recovery of an isotope is completed, there will be a sudden large increase in price. For example, new supplies of silicon and iron were added to the inventory between the January 1985 and April 1985 prices lists. Table 1 shows the changes in price for isotopes of these elements and checking with ORNL staff confirms that this is the pattern to expect.

TABLE 1. PRICES FOR STABLE ISOTOPES FROM ORNL

Isotope	Jan. 85 Price ($US/mg)	Apr. 85 Price ($US/mg)	Apr. Price/Jan. Price
Fe-54	1.89	5.75	3.0
-56	0.09	0.40	4.4
-57	11.02	16.25	1.5
-58	28.00*	150.00	5.4 (2.1*)
Si-28	0.84	0.90	1.1
-29	3.07	27.00	8.8
-30	5.62	48.00	8.5

* Fe-58 was $70.13 US/mg in 1983 and 1984.

ISOTOPE ENRICHMENT TECHNIQUES

There are many ways of enriching one particular isotope of an element. For simplicity these have been divided (somewhat arbitrarily) into three types which are shown in Table 2.

TABLE 2. METHODS OF ISOTOPE ENRICHMENT

1. Mechanical Methods:
   - centrifuge
   - diffusion
   - nozzle
   - distillation
2. Physical Property Based:
   - atomic vapour laser isotope separation
   - plasma centrifuge
   - plasma separation process
   - electromagnetic
   - molecular laser isotope separation
3. Thermodynamic Processes:
   - hydrogen sulphide-water exchange (heavy water)
   - electrolysis

The centrifuge, diffusion and nozzle techniques are used or proposed for use in enriching uranium in U-235 for use in light water reactors. A gas centrifuge spins a high vapour pressure compound, usually uranium hexafluoride, very rapidly to get a very small difference in the concentration of the isotopes across the machine. By cascading many of these machines, an enrichment of three or four fold is obtained. Similarly, a diffusion process repeats a very small enrichment step many times by forcing the gas through tiny holes. The nozzle process also requires cascading of many steps. Distillation takes advantage of the small difference in the boiling point of different isotopic species and is often used in the final stages of heavy water production.

Atomic vapour laser isotope separation is based on the small shift in the absorption frequency of the different isotopes of elements in the gas phase. Precisely chosen wavelengths of light will be absorbed by only one isotope which becomes ionized. An electric field can then be used to remove that isotope from the mixture. Because there is a high probability of collisions with the other isotopes leading to charge exchange, the initial high selectivity of the ionization steps may be lost if the density of the atomic vapour is too high. Thus very pure isotopes can only be obtained by this process if the throughput is low. The two "plasma" processes each involve accelerating charged particles through a magnetic field. In each case, high single step enrichment is difficult to achieve at high throughput. Molecular laser isotope separation is still in the research phase and while there is a large potential, more development is required before such a process can be applied.

The exchange of deuterium between hydrogen sulphide and water is the basis for the first stages of heavy water plants. This is a well understood process involving many small enrichment steps and is economic for ton quantities of heavy water. Whether similar processes could be developed, especially for the heavier elements, is yet to be determined. Electrolysis is an alternative process for the final purification of heavy water to reactor grade.

In all of these processes the main goal has been enrichment of very large quantities of isotopes, usually uranium-235 or deuterium. Production of small quantities of a wide variety of isotopes is only possible using an electromagnetic separator and this is the process that AECL is investigating as the basis of a proposed enrichment facility.

In many cases, one can see that the different processes could be placed under one of the other headings depending on the characteristic of the system which is used for the decision. All of these processes also tend to fall into two other groupings: large quantities combined with relatively small enrichments or small quantities combined with large enrichments. Because small amounts of a radioisotope do a large job in diagnosis or therapy and often because minimizing the dose to the patient requires high isotopic purity, the latter case is usually the one of interest in medical procedures. A high price for an isotope does not rule out its use. For example, a mass of tellurium-124 equal to the mass of a five-cent coin would cost almost $70 000.00 CDN. A great deal of research has gone on around the world for the last few years into new means of producing slightly enriched uranium-235. This research is not likely to provide cheap, high purity isotopes of other elements.

RELATED RESEARCH PROGRAMS AT CRNL

A variety of research and development programs at Chalk River are related to the enrichment of isotopes. In the Physical Chemistry Branch there are programs in solution phase thermodynamic equilibrium chemistry of isotope partitioning including the relevant kinetic studies to determine the rate of reaching equilibrium and a fairly extensive fundamental program of catalysis research. In Chemical Engineering there is a large catalysis development program, which includes other aspects of application of the catalysts developed at CRNL in addition to isotope enrichment. Also in the Physical Chemistry Branch, there is a program of fundamental research in molecular laser isotope separation which includes a modest understanding of laser physics. Complementing all these programs is the necessary expertise in analytical chemistry in the General Chemistry Branch which gives us the ability to develop new methods of determining isotopic concentrations in samples. Finally, AECL is the world leader in ion source development and has experience with large magnets. This combined with the chemical skills on site allows us to consider building an electromagnetic isotope separator in Canada which would remove the uncertainties in supply because of the situation at ORNL. In the near future, it is very likely that AECL will decide to enter the business of supplying enriched stable isotopes.

SUMMARY

There is a great deal of uncertainty in the supply of stable isotopes for users outside of the United States (the internal supply could also be in trouble, but that is less likely). Many isotopes are not available at any particular time; however, development of new procedures requires small quantitites of the suggested isotope and such development will not be possible if all the available production capacity is used to keep up with the demand for the currently used isotopes.

Development of new uses of different stable isotopes requires more than the medical research. The list in Table 3 shows some of these requirements. At CRNL, we can provide most of the first four requirements and workshops or meetings such as this could provide the fifth. Working together, we may provide better means of diagnosis and treatment of illnesses.

TABLE 3. ADDITIONAL REQUIREMENTS FOR GENERATING NEW TECHNIQUES USING STABLE ISOTOPES

1. Availability of stable isotopes as precursors
2. Irradiation mechanism - reactors or accelerators
3. Chemical processing before and after irradiation
4. Design of measuring equipment - detectors, tomographs
5. Continuing interaction of 1-4 with the medical community