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THE AVAILABILITY OF ENRICHED STABLE ISOTOPES: PRESENT STATUS AND FUTURE PROSPECTS

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The Availability of Enriched Stable Isotopes: Present Status and Future Prospects

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Introduction

The history of electromagnetic separation of isotopes at Oak Ridge National Laboratory (ORNL) began over 40 years ago as one of the great wartime efforts of the Manhattan Engineering District. In 1943, a contract was written with the Tennessee Eastman Corporation to operate a large and secret electromagnetic facility known as Y-12 near the newly created town of Oak Ridge, Tennessee. The purpose was to obtain large quantities of 235U free of the more abundant isotope 238U. By 1945, the electromagnetic isotope separations program was functioning at its peak with nearly 25,000 people and more than 1100 separating units. About this time, however, the gaseous diffusion process was demonstrated as a cheaper way of obtaining 235U and soon after the cessation of hostilities, the electromagnetic plant was declared obsolete. During this period discussions were held to consider the possibility of using a part of the plant to supply quantities of enriched isotopic materials for basic and applied research. When the shutdown operation was finished, only two of the nine Y-12 buildings were retained intact for this purpose - the pilot plant with four separators or "calutrons" and a production building containing 72 additional separators.

What began as a limited effort, just the four separators in the pilot plant were to be used for the enrichment of various stable isotopes other than uranium, rapidly grew in scope and importance. By extending the temperature range of the ion source from 400 degrees C (as used in the uranium separations) up to as high as 2800 degrees C, all elements with naturally occurring multiple stable isotopes (except osmium) had been processed at least once in the facility by the mid 1950's. During the 1960's, the elements osmium, americium, and curium were added to the list of elements whose isotopes had been enriched. In the period 1946-1973, it is estimated that the facility had been used to produce 200 kg of enriched isotopes, including over 250 stable nuclidic species and with isotopic impurities as low as < 0.1 ppb. As stated by L. O. Love, whose paper on the electromagnetic separation of isotopes at Oak Ridge [1] is the source of much of the preceding information, "the electromagnetic process is undoubtedly the most versatile known means of separating isotopes".

The present ORNL facility, which is now referred to as the Electromagnetic Isotope Enrichment Facility (EMIEF), is operated with a total of 16 separator positions devoted to stable isotope enrichment (for which there are some 30 different separator units available) and with 8 separator positions used to enrich radioactive species. The EMIEF is currently used to produce 225 enriched stable isotopes of 50 elements. Among these are included most of the known elements with stable isotopes except for the noble gases, certain light elements, monoisotopic elements, etc. The EMIEF can also be used to produce enriched samples of radioactive species, most notably the isotopes of uranium and plutonium. These enriched materials are placed in either the Sales Inventory or in the Research Materials Collection (RMC). The materials in the Sales Inventory are for sale to anyone on a first come, first served basis. Prices in the most recent catalog [2] range from \$0.05/mg for 99.8% 140Ce to \$1,267/mg for 98.5% 176Lu. The materials in the RMC are made available to U.S. researchers (or groups that include a U.S. investigator) on a loan basis for use in non-destructive experiments and applications. In addition, certain samples have been provided to European investigators for cross-section studies through the auspices of EURATOM and the European-American Nuclear Data Committee.

Recent operating history of the EMIEF

Some statistics on the recent operating history and financial support of the EMIEF are given in Table 1. The effect of declining budgets for the EMIEF is evident in the figures for operation during the past few years. From the period including FY82 and FY83 where annual operations were at the level of 94,000 vacuum tank-hours, the rate declined to 46,000 tank-hours in FY84 and again to 24,000 tank-hours in FY85 (see Fig. 1). Total support for the EMIEF operation was down to \$2.5M (including approximately \$1M revenue from isotope sales) in FY85. At this point, the operating level of the EMIEF had fallen to something like one-quarter of the rate required just to balance isotope sales and maintain an equilibrium amount of material in inventory. Thus, projections were made at the end of FY85 of the number of isotopes with zero inventory for the coming years based on an annual operating rate of 24,000 tank-hours and average sales rates from the previous 5 years. Although the number of isotopes with zero inventory during the period FY82 to FY86 was actually reasonably low (see Fig. 2 - note that there is some lag time before the effects of reduced operations appear in the inventory status), it is clear that the number of unavailable isotopes was predicted to rise alarmingly, reaching a level of more than 100 species or nearly 50% of the total by the end of FY89. Some financial relief was obtained in FY86 funding and that, coupled with certain economies, has made it possible for the facility to be operated for 46,000 tank-hours this year or at the same level as in FY84. A comparison is shown in Fig. 3 where one can see that in spite of this increase in operating time, the FY86 operating level is still about a factor of 2 less than the operating capacity represented by annual sales.

NAS/NRC workshop, February 1982, Washington, D. C.

In February 1982, a workshop was held under the sponsorship of the National Academy of Sciences/National Research Council (NAS/NRC) to assess the needs for (enriched) stable isotopes in the scientific, medical, and industrial communities. The co-chairmen of the steering committee were G. Friedlander (Brookhaven National Laboratory) and H. N. Wagner, Jr. (Johns Hopkins University). At the workshop, which was held in Washington, D.C., summary papers were presented and panels were convened to discuss applications of enriched stable isotopes in the following areas of specialization: 1) Physics, chemistry, and geoscience research, 2) Commercial, 3) Biomedical research, and 4) Clinical. In the first topical area, as a part of his presentation as plenary speaker, M. S. Zisman (Lawrence Berkeley National Laboratory) made a comprehensive survey to assess current usage and future needs for enriched stable isotopes. Of the more than 1000 questionnaires that were mailed out, he received 553 responses representing 231 separate institutions. For the three-year period covered by the survey (1979-81), total usage involved 220 different isotopes, 2,350 gram-atoms of material, and a cost of $1.6M. In an attempt to estimate future needs, Zisman made the point that quantitative estimates were not possible due to the unpredictable nature of fundamental physical research programs, but that it appeared the demand for isotopes would remain approximately constant over the next five-year period. Zisman estimated the highest demands would be for 1) rare earths, 2) elements with the widest range of stable isotopes, i.e. Ca, Ni, Zr, Mo, Sn, Sm, and Pb, and 3) isotopes in the Mg-Si region. He discussed various trends in the areas of physical research, including the need for high isotopic purity for isotopes with the lowest natural abundances, the use of certain enriched isotopes as exotic projectiles in accelerators, etc. The workshop report [3] concludes with the following recommendations:

1.) The production rate of the ORNL electromagnetic separation facility should be increased to utilize more fully the existing physical plant. ...to replenish, as soon as is practical, the rapidly diminishing supplies in both the Research materials Collection and the Sales Inventory, and subsequently to maintain a complete range of isotopes in both inventories at adequate levels.

2.) Mechanisms are needed to ensure adequate supplies and equitable distribution of electromagnetically separated isotopes to all segments of the user community, as well as to avoid violent price fluctuations. To this end, the appointment of an advisory committee responsible to the administrative level in the U. S. Department of Energy (USDOE) that has authority for the policy in the Stable Isotopes Program is recommended.

3.) Research and development efforts should be directed toward the demonstration of alternative technologies which might in the future complement ORNL's electromagnetic separations for certain isotopes. ...may have the potential for achieving adequate enrichment of specific isotopes at lower cost.

The actual outcome of recommendation 2 was the formation of two committees. The first, the Technical Policy Committee, advises ORNL management on questions of policy with regard to managing the facility, relationships with the USDOE including questions of pricing policy, etc. The second is a Users Group for the EMIEF which was formed upon the recommendation of the Technical Policy Committee. Its function is to assess future needs for enriched stable isotopes, to provide information to the users regarding isotope availability, to provide information to the ORNL management regarding users needs, etc.

EMIEF Users Group

The first organizational meeting of the EMIEF Users Group was held at ORNL in April 1984. A charter for the organization was drawn up, as follows:

1.) To provide a formal channel for information exchange between the EMIEF administration and the isotope user community.

2.) To provide a means of including the priorities of the users in the production scheduling of the facility.

3.) To provide a forum for sharing information on the needs for and uses of enriched isotopes.

Currently, membership in the organization totals approximately 500. The present Executive Committee is made up of the following persons: D. K. Evans, Chairman (Chalk River Nuclear Laboratories), R. F. Casten (Brookhaven National Laboratory), H. Folger (GSI, Darmstadt, West Germany), J. A. Harvey (ORNL), L. L. Riedinger (U. Tennessee), S.N. Suchard (TRW), and J. W. Terry, Secretary-Liaison Officer (ORNL). Serving as Committee Chairmen are the following persons: R. L. Hahn (ORNL), Communications, and L. L. Riedinger, Government Affairs Liaison.

In October 1985 at ORNL, a second meeting of the Users Group was held at which several items were identified for emphasis in discussions with the users and as goals of the group's activities, as follows:

1.) The necessity to make the users more aware of the implications of reduced levels of EMIEF operation,

2.) The need to obtain more information regarding projected requirements for enriched stable isotopes in the future,

3.) A reminder to the users that through the Users Group their needs can have some impact on the schedule for enrichments in the EMIEF,

4.) The Users Group can serve as a clearinghouse for information regarding the exchange of rare isotope samples,

5.) The Users Group and the ORNL Isotope Sales organization should document all requests for enriched stable isotopes, even if the item in question is unavailable,

6.) The development of an improved pricing policy that features adiabatic price changes (increases), which is desirable from the users point of view, and pricing according to current replacement costs rather than original production costs, which is desirable with respect to greater support of the EMIEF operating costs from isotope sales revenue.

The Users Group has received many letters during the past year that discuss the problems and concerns of individual users with respect to the availability and use of enriched stable isotopes. The problems outlined included questions of availability, e.g. "When will enriched 41K become available; the catalog merely indicates after 1985?", problems caused by severe, often unannounced, price increases, e.g. cost increases in the period between January and April 1985 for 58Fe, from $28/mg to $150/mg, for 29Si, from $3/mg to $27/mg, and for a list of varied isotopes ordered for use in a chemical study of transition metal behavior, from a total order of $3,400 to a total of $6,800. It is clear that such drastic price increases, which ranged up to as much as a factor of 9, create severe budgeting problems and can often be the cause for certain research proposals to be withdrawn or to be reduced in scope. Since the enriched isotopes must be recognized as essential tools in the hands of capable researchers, a mechanism should be found to permit gradual price increases that do not exceed some reasonable limit when they become necessary.

Present status of isotope availability

The status of the enriched isotopes included in the Sales Inventory is shown in Table 2 where isotopes are listed that are either not available of are in insufficient quantity or quality to meet current requests, as of 6/30/86. These can be summarized in the following subcategories:
1) Isotopes with zero inventory (22), 2) Isotopes of insufficient quantity (17), and 3) Isotopes with insufficient enrichment quality (10). Of these 49 species, the supplies of 10 will be replenished by the scheduled FY86 enrichments in process (isotopes of bromine, calcium, nickel, potassium, rubidium, and strontium). In Table 3 are listed isotopes where the current inventory is less than the average annual sales level for the past five years. There are 47 isotopes listed, representing 25 different elements. Thus, there exists considerable potential for a substantial increase in the number of isotopes with zero inventory.

Support for the EMIEF

As for support of the EMIEF from the user community, two important reports were generated in the past; the first is from an Ad-Hoc Panel on National Uses and Needs for Separated Stable Isotopes, edited by G. C. Phillips, NRC, 1968. In this report, the EMIEF was termed "a national asset", a description that was later amended by the Danish scientist, J. Koch, to read "an international asset" for scientific research. The second report was the product of the aforementioned NAS/NRC Workshop in February 1982.

More recently, the EMIEF and the enriched stable isotopes that it produces were evaluated by the Nuclear Science Advisory Committee (NSAC). In a letter (December 1984) from J. P. Schiffer, then chairman of NSAC, to A. W. Trivelpiece, Director of the Office of Energy Research, USDOE, it was stated that "We (NSAC) regard the preservation of the Research Materials Collection as a most important need for our science, with a major potential impact on the future."

In another letter to the USDOE from the National Academy of Sciences (Frank Press, November 1985) and written by the co-chairmen of the Board on Chemical Sciences and Technology of the National Research Council, the importance of enriched stable isotopes to many areas of experimental science was emphasized and it was urged that 1) the EMIEF be provided with increased funding over a few years to restore the isotope inventory to an adequate level, e.g. a FY86 budget of $5M (including an estimated $1.2M in sales), and

2) a pricing policy be developed that will maintain the inventory by allowing true replacement costs to be charged.

The result of this show of support for the EMIEF has been a modest increase in the EMIEF operating budget (which is not yet up to the level of $5M) and the resultant increase in operations, as shown graphically in Fig. 3. It seems clear from the workshop recommendations and letters of support that the case has been made by the U. S. scientific community for operating the EMIEF at a level that avoids serious shortages among the 225 product isotopes. It remains only for the USDOE to find a mechanism for bringing the funding of the EMIEF up to the desired level.

References

* Work performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

1. L. O. Love, Science 182, 343 (1973).

2. The isotopes in the Sales Inventory are listed in the ORNL Electromagnetic Separated Stable Price List, available from the Isotope Distribution Office, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, TN 37831.

3. "Separated Isotopes: Vital Tools for Science and Medicine", National Academy Press, Washington, D. C., 1982.

Table 1. Summary of EMIEF operations, materials processed, and inventory status for FY82 through FY86.

Period -	FY82	FY83	FY84	FY85	FY86
Operation:					
Vacuum tank hours	93,300	93,300	46,700	23,300	46,700
Operating time	12 mo.	12 mo.	6 mo.	6 mo.	12 mo.
Total funding*				$2.5M	$3.3M
Materials processed:					
Number of elements	6	12	5	3	5
Number of isotopes	23	55	17	6	15
Inventory status:					
Number of additions	16	16	39	24	17
Available isotopes**	206	195	187	200	206
Unavailable isotopes**	19	30	38	25	19

* Including $1-1.2M revenue from isotope sales.

** As of the beginning of the fiscal year.

Table 2. Isotopes not available or insufficient in enrichment quality or quantity to meet current requests (6/30/86).

Antimony-121[A]	Dysprosium-163[B]	Potassium-40[A,D]
Barium-132[A]	Erbium-162[A]	Potassium-41[A,D]
Barium-134[A]	Gadolinium-152[C]	Rhenium-185[A]
Barium-137[A]	Gadolinium-157[B]	Rubidium-87[A,D]
bromine-79[A,D]	Germanium-76[C]	Ruthenium-96[A]
Bromine-81[A,D]	Hafnium-176[B]	Samarium-144[C]
Cadmium-108[C]	Hafnium-179[B]	Selenium-80[C]
Cadmium-110[B]	Lead-204[B]	Silver-107[C]
Cadmium-112[C]	Molybdenum-97[A]	Strontium-84[B]
Cadmium-114[C]	Nickel-58[C,D]	Sulfur-36[C]
Calcium-42[A,D]	Nickel-60[C,D]	Tantalum-180[A]
Calcium-46[A,D]	Nickel-62[C,D]	Tin-112[B]
Cerium-140[A]	Osmium-186[C]	Tin-119[A]
Cerium-142[C]	Osmium-187[B]	Titanium-48[B]
Dysprosium-156[A]	Osmium-189[C]	Tungsten-180[C]
Dysprosium-162[A]	Palladium-102[A]	Tungsten-183[A]
		Tungsten-186[C]

A. Zero inventory

B. Insufficient enrichment quality

C. Insufficient quantity

D. In process

Table 3. Isotopes where the current inventory is less than the average annual sales for the past 5 years (6/30/86).

Antimony-123	Mercury-201	Selenium-77,78
Calcium-44	Molybdenum-92,94,95,96	Silver-109
Copper-63	Neodymium-144,145,146,150	Strontium-88
Dysprosium-160,161,164	Nickel-64	Sulfur-33,34
Erbium-167,168,170	Potassium-39	Tellurium-128
Europium-151	Rhenium-187	Thallium-203
Gadolinium-158,160	Rubidium-85	Tin-114,116,117,122,124
Gallium-71	Samarium-147,149,150,152,154	Zirconium-90,92
Iron-58		

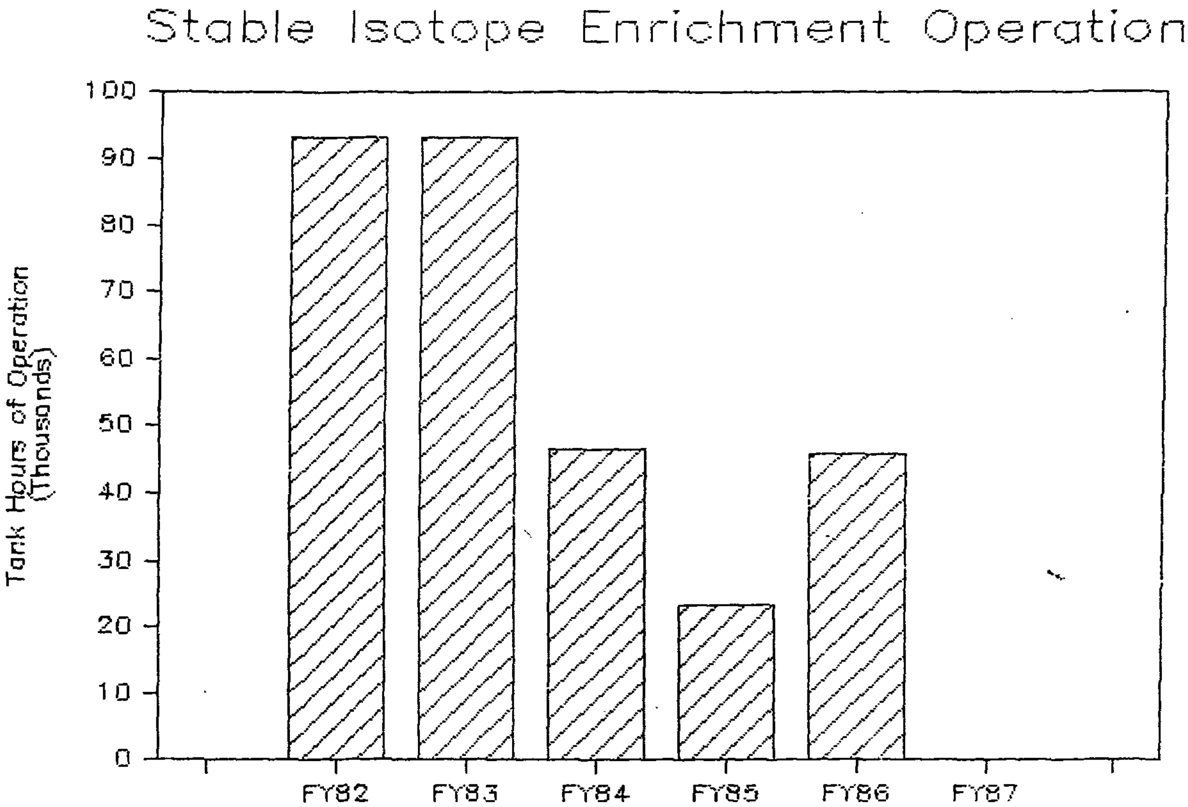

Figure 1. Isotope separation effort at the EMIEF, expressed as annual vacuum tank-hours of operation.

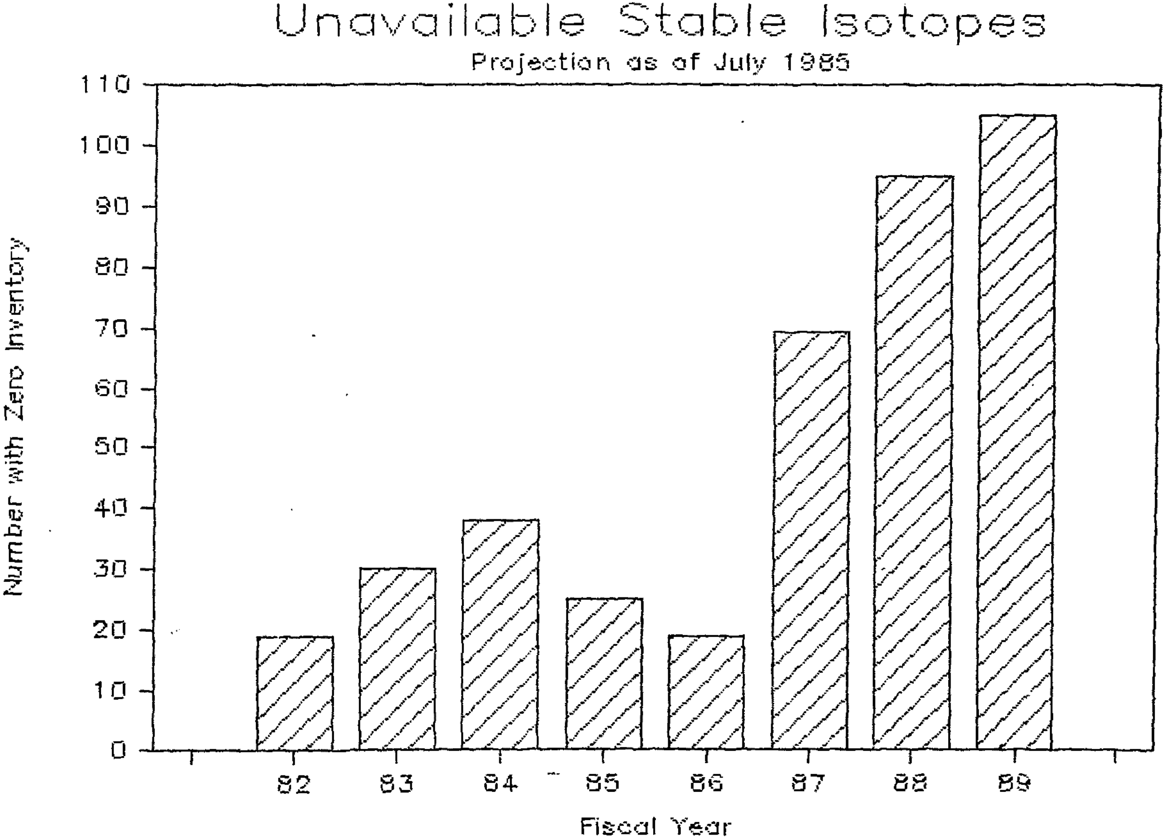

Figure 2. Number of unavailable isotopes (those with zero inventory), as of the beginning of the fiscal year.

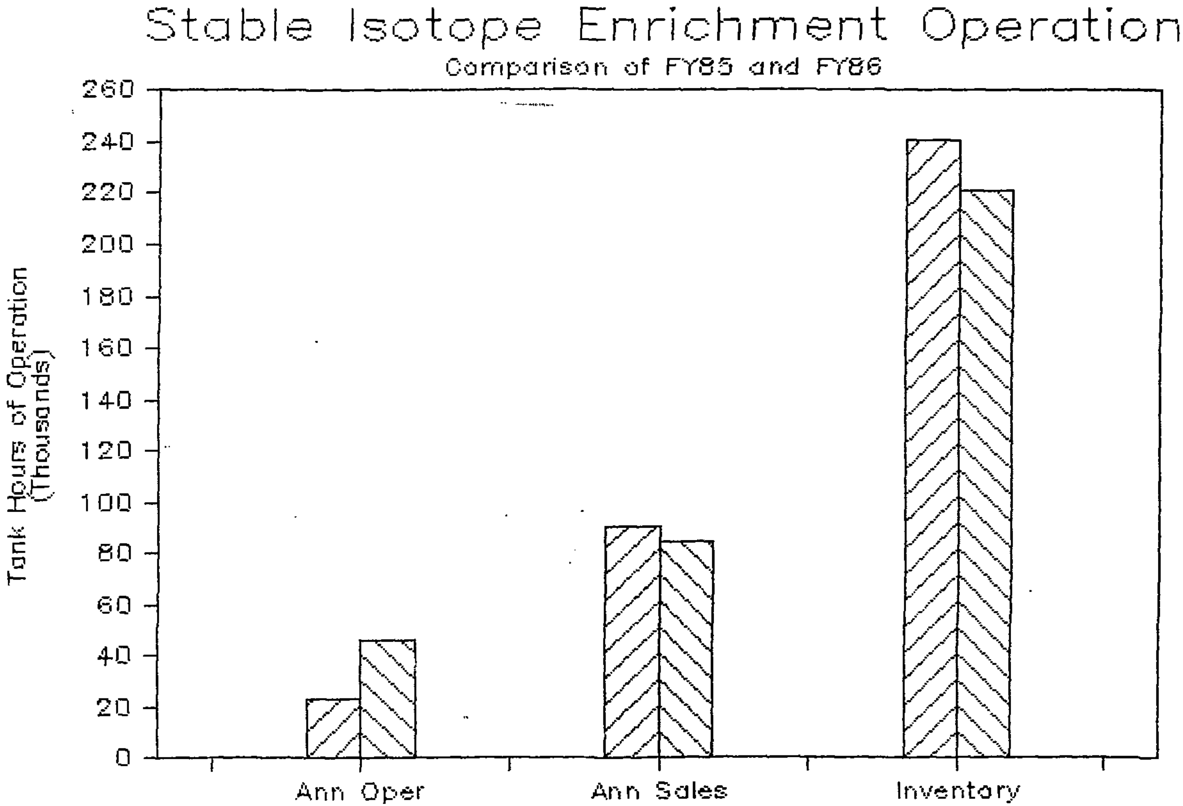

Figure 3. Comparison of FY85 and FY86 annual EMIEF operation, annual isotope sales, and inventory at the end of the fiscal year, expressed either directly or in equivalent units of vacuum tank-hours of operation.