THE CALUTRON AS A SOURCE OF TARGET MATERIALS*

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L. O. Love

Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830

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On the occasion of the twenty-fifth anniversary of Oak Ridge, General Leslie R. Groves (who had been head of the wartime Manhattan District) visited the old calutron pilot plant and made the comment that when evaluations of the various methods for separating uranium were made, it was thought that the electromagnetic process would be impractical. He also commented that the committees assigned to the task of evaluating the various processes would have recommended against building an electromagnetic facility except that it was backed by Professor E. O. Lawrence. In time, of course, the faith in Professor Lawrence's method was justified although it was proven that another process was found to be superior for providing large quantities of ^{235}U, and the Y-12 Plant, consisting of over 800 alpha and almost 300 beta calutrons, was shut down. Fortunately for the nuclear industry, however, that was not the end of the story. Some farsighted people in what is now the U.S. Atomic Energy Commission and the Oak Ridge National Laboratory made the decision to use some of the machines for isotope separations and certain research activities, and, as a result, the pilot plant, containing two alpha 122-cm radius and two beta 61-cm radius calutrons, and one beta building, containing 72 calutrons, were retained. Today both of these buildings are used for isotope separations, and together they represent the most versatile isotope separating facility in existence. They have the capability of providing enriched samples of every naturally occurring isotope in the periodic chart and significant quantities of many radioactive species. It is from these unique materials that many of the targets for nuclear research are made.

The first stable element (copper) was separated in late 1945 and since then over 200 kg of enriched isotopes have been collected for use in science and medicine. The elements, both stable and radioactive, that have been processed to acquire these isotopes are shown in Figure 1. The separated material has been divided into three categories:

1. Research Materials Collection Pool for the AEC
2. Sales inventory
3. Unprocessed inventory

The first category contains the separated isotopes retained by the AEC (the prime customer and program supporter) for use in their sponsored programs; these are usually obtained on a loan basis for nondestructive experiments.

Category two contains material available for sale and is purchased by investigators who will normally use it in experiments that frequently destroy its identity. These isotopes are listed with prices and purities in the ORNL Research Materials Catalog (Separated Isotopes, Radioisotopes, Special Preparations), a copy of which can be obtained by request to Oak Ridge National Laboratory, Isotopes Sales Department, Building 3037, P. O. Box X, Oak Ridge, Tennessee 37830.

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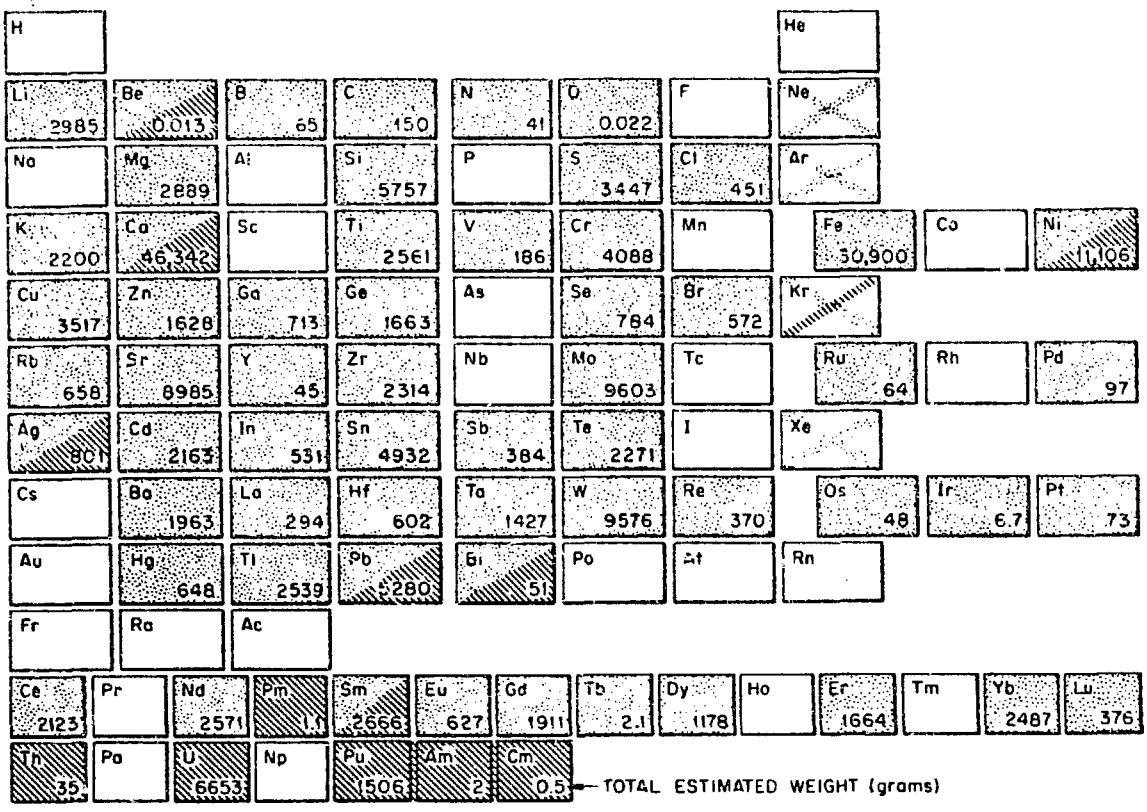


Fig. 1. Calutron Processed Elements.

The third category contains an estimated 80 kg of separated isotopes which are in various stages of chemical purification and are supplied to investigators when they express a need for them. These enriched isotopes are available because the electromagnetic separator is a mass discriminator and separates the "unwanted" isotopes as well as the "wanted" ones. These "unwanted" materials, their estimated weight, and isotopic purity are listed in Table I.

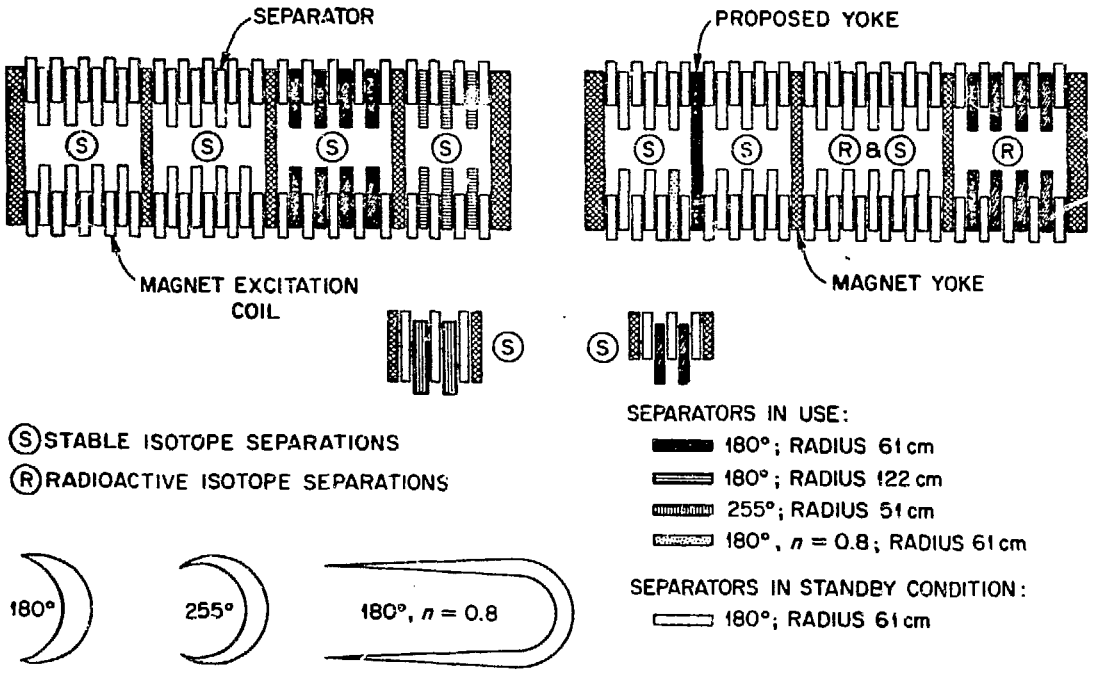
The ORNL calutron facility as it exists today is depicted schematically in Figure 2. The pilot plant, with its two alpha and two beta calutrons, was used in the electromagnetic development program for the Y-12 Plant during the World War II effort to get ^{235}U, but since has been involved in the separation of stable isotopes for peacetime uses. The remainder of the facility was placed into similar usage about 1960. While the use of the ORNL calutron facility may have been rather inconspicuous, many people believe that the separation of stable isotopes is one of the most important single programs for exploiting peaceful uses of the atom sponsored by the AEC; they also believe that the pilot plant, like the graphite reactor, should eventually be dedicated as one of the nation's noted landmarks.

In order to increase the collection rate of the calutron, each machine was equipped with magnetic focusing lenses called linear shims. In addition to these original linear shims, two other types of magnetic focusing,

$$B = B_o \left(\frac{r}{r_o} \right)^n$$

Table 1. Unprocessed Stable Isotopes

Isotope	Approx Purity (%)	Approx Quantity (g)	Isotope	Approx Purity (%)	Approx Quantity (g)
^{26}Mg	99.9	1.160	^{92}Mo	97.5	940
^{28}Si	99.8	3,500	^{94}Mo	92.0	600
^{32}S	99.7	750	^{95}Mo	96.4	1,030
^{39}K	99.9	1,340	^{96}Mo	96.6	1,140
^{40}Ca	<99.9	32,695	^{97}Mo	94.0	630
^{44}Ca	98.5	79	^{100}Mo	97.2	620
^{48}Ti	99.2	1,200	^{110}Cd	97.0	65
^{52}Cr	99.8	2,500	^{111}Cd	95.0	65
^{54}Fe	95.0	820	^{116}Cd	97.0	40
^{56}Fe	98–99.8	15,000	^{120}Sn	98.0	425
^{58}Ni	99.8	4,750	^{128}Te	99.2	345
^{60}Ni	99.5	790	^{130}Te	99.5	475
^{64}Zn	99.5	580	^{138}Ba	99.8	750
^{66}Zn	98.5	340	^{139}La	<99.95	172
^{78}Se	98.6	48	^{140}Ce	99.8	640
^{80}Se	99.4	184	^{142}Nd	97.5	195
^{86}Sr	96.4	750	^{144}Nd	97.5	225
^{87}Sr	85.0	350	^{146}Nd	97.5	160
^{88}Sr	99.8	5,270	^{181}Ta	<99.99	1,200



The Upper Blocks Represent the Two Tracks of Separators Housed in Bldg. 9204-3, Indicating the Relative Positions of the Three Types in use, While the Lower Blocks Represent the Four Separators (Two Types) Contained in Bldg. 9731. The Geometrical Designs in the Lower Left Depict the Beam Profile Produced by Each of the Three Types in use.

Fig. 2. Schematic Representation of ORNL EM Isotope Separation Facilities.

have been incorporated into the overall program in order to increase the versatility of the separation facility. The first of these was the 255° modification ($n = 0.5$) which has an effective radius of ~40 inches (~102 cm) and increases the dispersion between masses by 1.4 times. These machines have been an excellent addition to the program since total ion output is similar to the calutron while assay purity is usually higher especially for masses greater than 100.

The second system added is a sector which is unique to the regular calutron in that the ion beam focuses outside the magnetic field. The sector, which we refer to as the Oak Ridge Sector Isotope Separator (ORSIS), was mounted in a spare calutron tank and has a beam deflection of 180° with inhomogeneity of $n = 0.8$. A schematic of this machine is shown in Figure 3. The objective of building this instrument was to wed the high-throughput features of the calutron with the high-purity capabilities of the European type separators. In addition to the focal point being almost 3 meters from that of the calutron ion beam, and as seen from the insert in Figure 3, the mass dispersion is over seven times as great. Currently, the ORSIS is being equipped with automatic controls for computer-assisted operation. A second machine of this type is under construction, and, when completed, will materially increase the capability of the electromagnetic facility to provide more samples of highly enriched isotopes in a single pass. Typical of the material expected is indicated in Table II, which compares the isotopic purities of the calutron with those obtained in the 180° sector separator.

Another desirable feature of the sector is its ability to make ion implants at energies ranging from a few volts to 160 keV. A holder that can accomodate 60 substrates is shown in Figure 4; similar devices have been used for making as few as two and as many as 100 targets in one run. It is not generally known, but between 15-20 thousand ion implants have been produced in the Oak Ridge separators. So far, ≤ 50 $\mu g/cm^2$ is the quantity usually deposited per target, but efforts are being made to increase the quantity deposited; some progress has been made as can be seen in Table III. To the extent that this can be done, target costs will be reduced because the separation and deposition can be made in one step.

The electromagnetic separator will, if conditions are right, resolve the isotopes of each ion beam of an element into its respective masses. However, the ion production mechanism is complex and the degree of separation can be no better than that dictated by the energy spread existing in the positively charged ion beam. Since this spread is not reduced to zero the separation is not perfect. Figure 5 shows actual pictures of oscilloscope traces that recorded beam fluctuation during the initial stages of a calcium run. The time interval involved in the start-up of this run was about four hours and the monitored beam was ^{40}Ca. The pictures show the beam intensity fluctuations and corresponding focal property as it increased from an output of about 15 mA in the early stages to approximately 150 mA at its maximum. The disturbances during this period are one of the reasons for the less-than-perfect isotopic purities that we obtain. But even with imperfection such as this, the calutrons still provide isotopic purities that are very high when compared to other methods. For example, ^{48}Ca was enriched from 0.18% to >96%, and ^{46}Ca from 0.0033% to about 50% in runs similar to those from which the pictures were made.

In most instances the attainment of high chemical purity presents greater problems than those associated with attaining the desired isotopic purity. For example, we collected about 100 grams each of >95% ^{29}Si and ^{30}Si in 1963 for growing single crystal wafer and for use in fabricating lithium-coated barrier-layer detectors. It was thought that single crystal ^{30}Si would greatly improve the counting efficiency of neutron spectrometers if the boron content could be held to less than one part per billion. The problem of reducing the boron content in these isotopic samples to an acceptable level for these uses has not yet been solved.

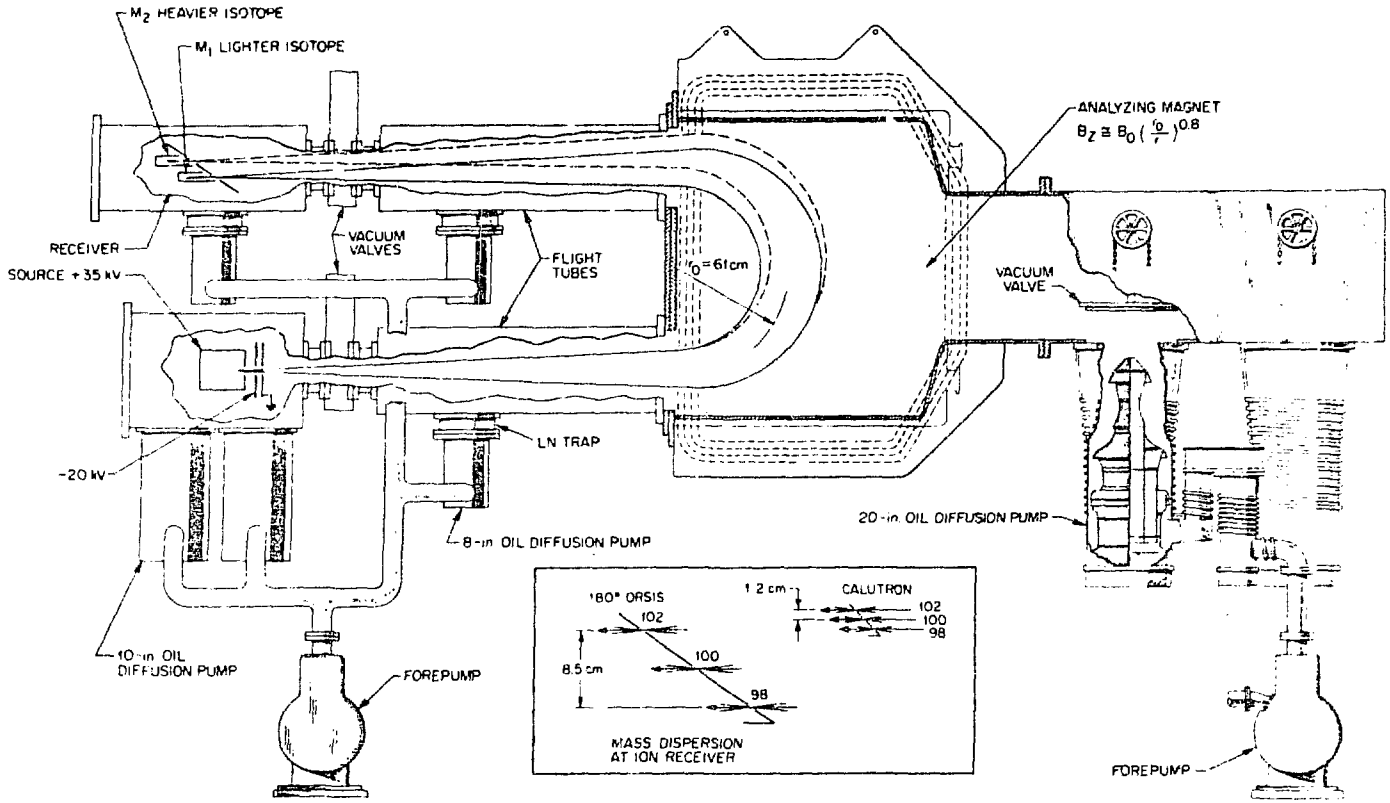


Fig. 3. Schematic of the 180° ORIS.

Table 2.

COMPARISON OF SEPARATIONS MADE WITH THE ORNL 180° (n = 0.8) SECTOR AND THE CALUTRON

Isotope	NA (%)	180° ORSIS		CALUTRON	
		Assay (%)	Total Beam Current (mA)	Assay (%)	Total Beam Current (mA)
^{84}Sr	0.56	99.72	4.6	83.6	46
^{119}Sn	8.58	99.34	2	89.8	20
^{198}Hg	10.02	99.36	1.5	97.3	3
^{156}Dy	0.056	99.53	1.5	20.9	17

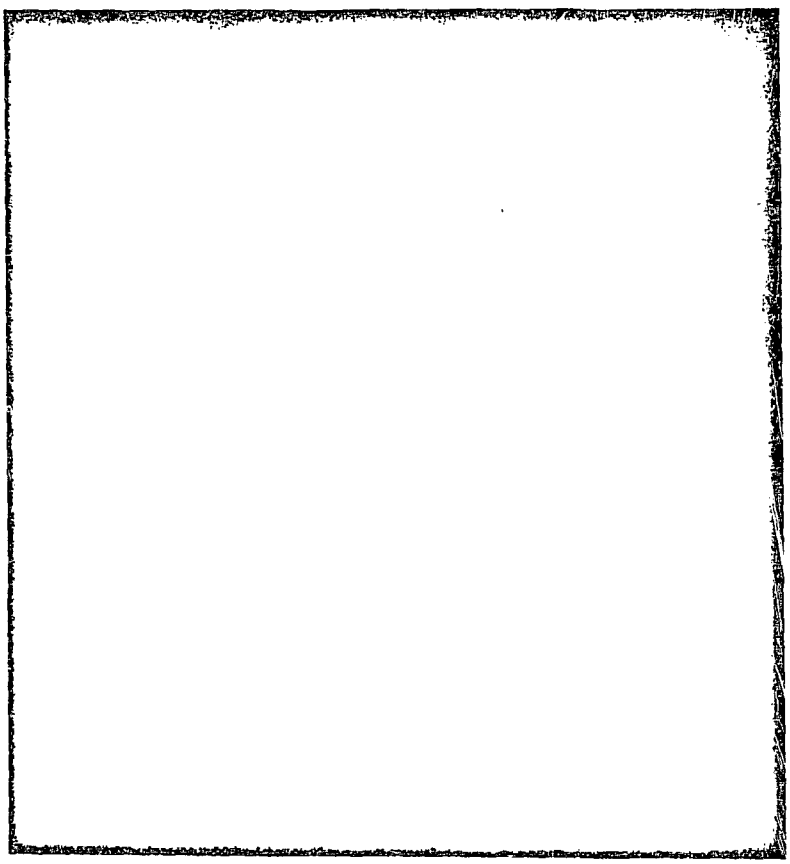


Fig. 4. Target holder.

1 of

FIG-4 Reduce 70% of original

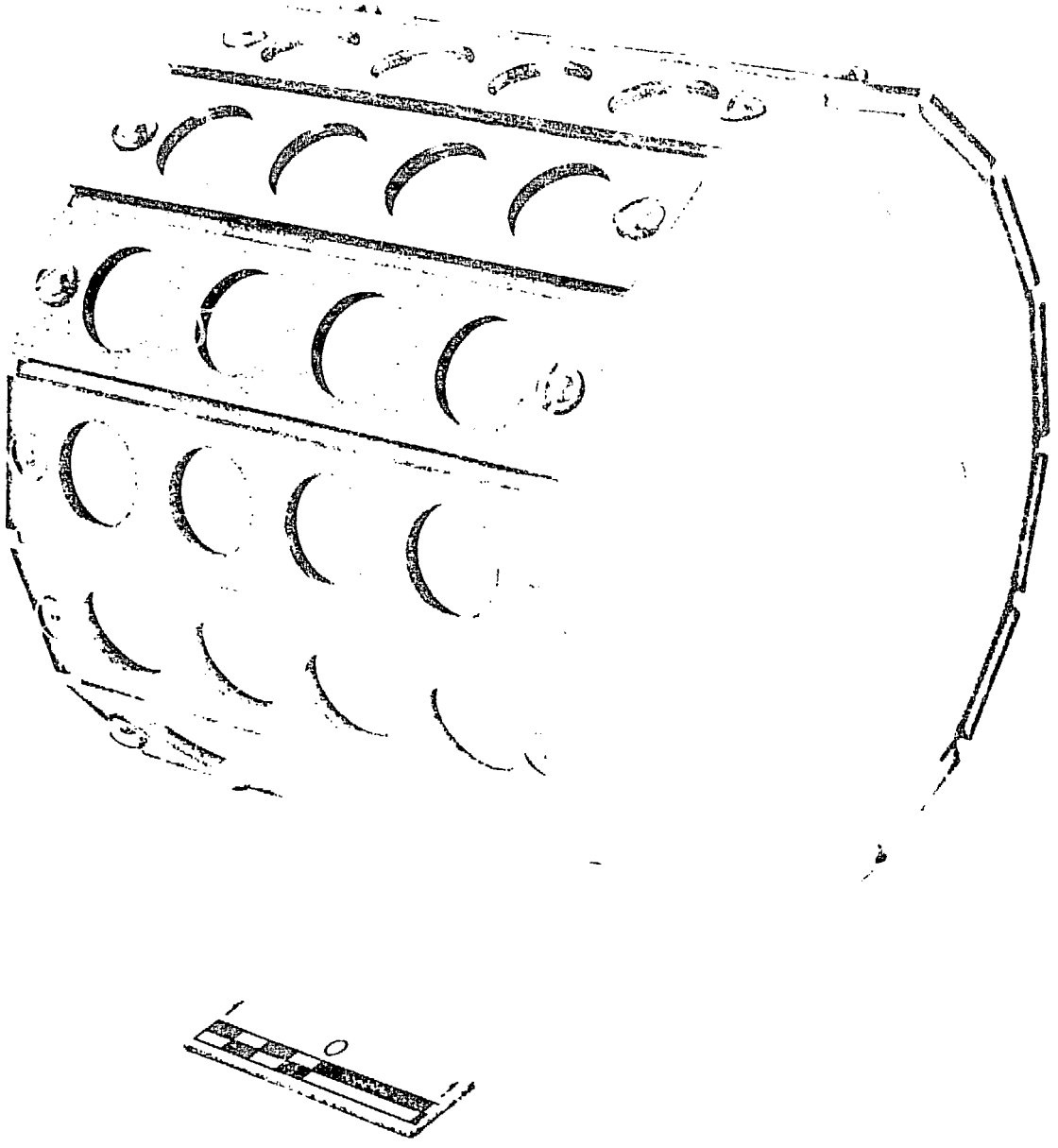


Table III

Samples of Targets Prepared in Oak Ridge Electromagnetic Separators

Isotope	Backing	Energy (keV)	Atoms/cm^2
^{119}Sn	SS	0.050	10^{19}
^{158}Gd	C	0.200	10^{17}
^{176}Hf	Ni	0.850	10^{18}
^{3}H	Au	2.5	10^{13}
^{85}Kr	Al	5.0	10^{13}
^{69}Ga	Ge	10.0	10^{16}
^{184}Pt	Ni	40.0	10^{17}
^{11}B	Si	80.0	10^{16}
^{31}P	Si	160.0	10^{16}

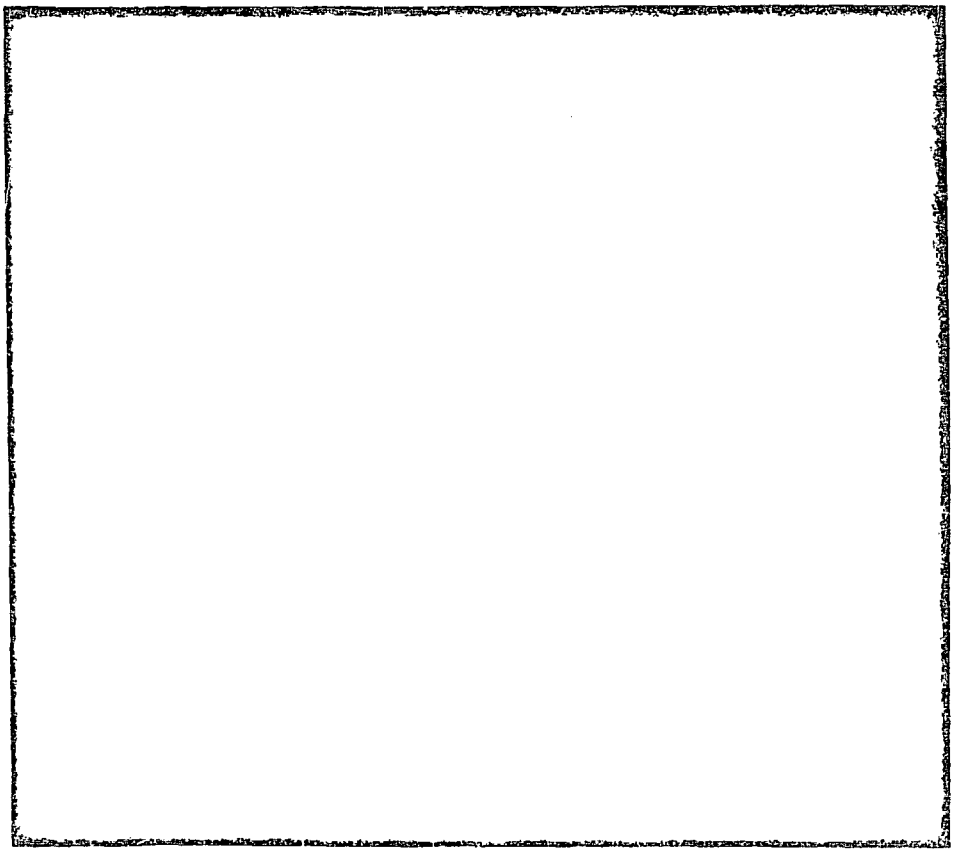
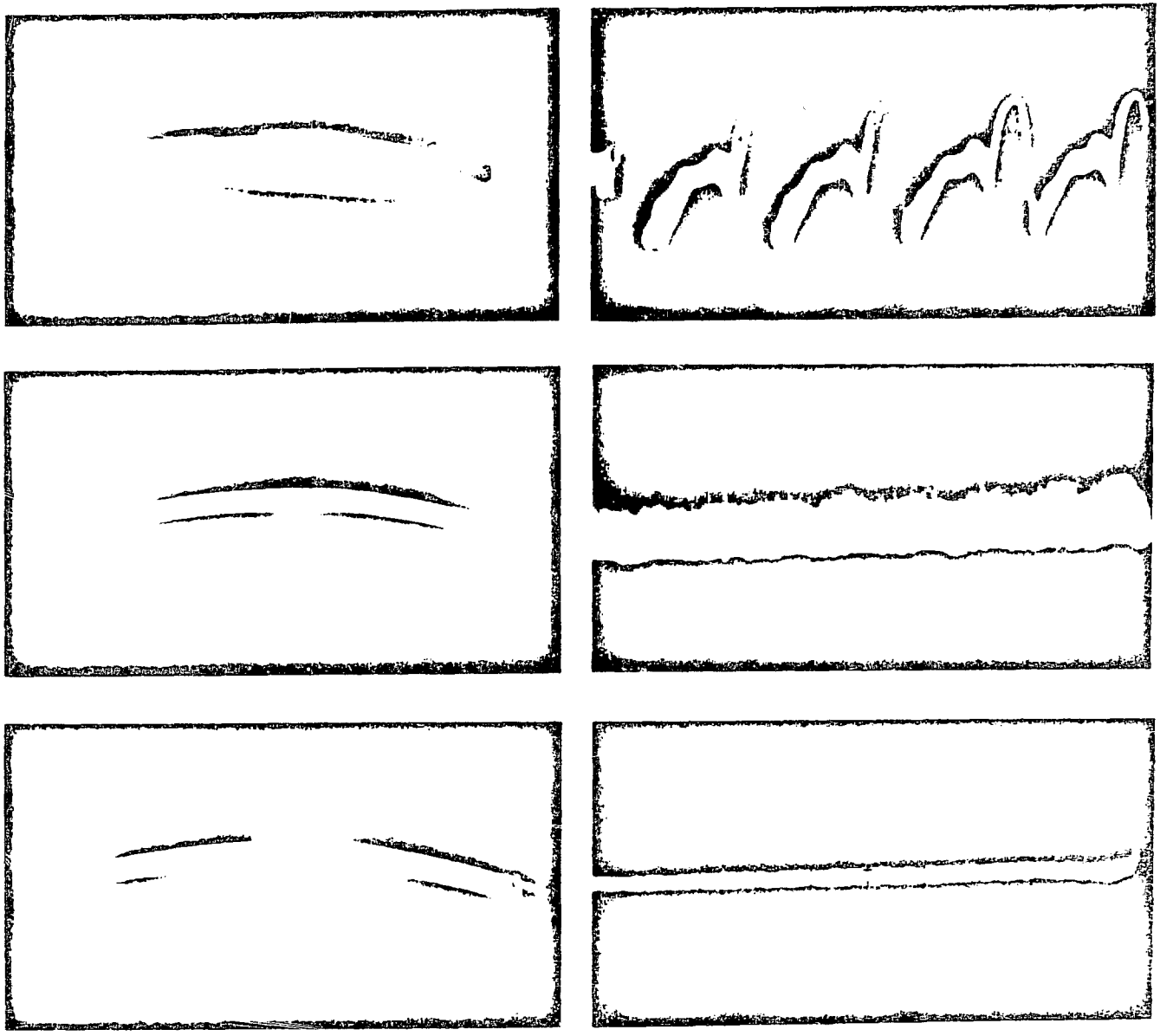


Fig. 5. Oscilloscope Trace of ^{40}Ca Beam During Calutron Start-Up.

Fig 3 Reduc. 11%
of original



Some sources of chemical impurities are inherent in the electromagnetic process. For example, elements present in the ion source components are heated to their vapor state and find their way into the ion production arc. These spurious ions are accelerated into the ion beam along with the wanted ions. Energy changes and charge exchange result in trace amounts of these unwanted atoms finding their way into the collector pocket, making problems for the chemist to remove, and if not removed, for the user to tolerate. The same thing happens when there are chemical impurities in the charge material. It is impractical to try to purify each charge to analytical grade material and, as a result, impurities in trace amounts may find their way into the collector pocket and again make problems for the chemist and the user.

On the other hand, to meet high isotopic purity requirements, it is only necessary to make second or third passes in the spearator using for charge that material which was separated before. It must be added that this approach to high purities is not without penalties because the enriched charge material for these runs is usually expensive and the low efficiency of the calutron (10-20%) means that the cost of a second and third-pass sample may be quite high. An example of this cost is seen by examining the economics of a second-pass run made with about 12 grams of 6% ^{180}W to obtain approximately 20 mg of >95% ^{180}W at the nominal price of $200 per mg. In this case the charge was made up of material sputtered from the ^{180}W collector pocket and deposited on the receiver defining plate, and was incidental to a 70,000-hour collection series made for the purpose of getting pound quantities of 95% ^{184}W. The actual time of the second-pass collection involved only a little more than 10 hours, but getting the 12 grams of 6% charge would have made the collection prohibitively expensive if the runs had been made for the 0.14% naturally occurring ^{180}W alone.

An example of the technical ease with which the isotopic impurities can be reduced is a case where we had a request for 50 grams of ^{233}U with the ^{232}U content reduced to 1-10 ppb. The separation was made and the 232 was reduced to 4 ppb. The investigators began their experiment, then called back and said that they had missed their calculation — the 232 content was eight times too high. All that was necessary was to make another pass and the unwanted isotope was reduced to 0.1 - 0.2 ppb.

It has been gratifying to watch the interest grow in separated isotopes since the first separation was made in the mid-1940's. Figure 6 is a graph showing the trend in the use of separated isotopes based on the number of shipments made by years. The decline in usage in 1973 parallels a forced decline in separation effort. In 1969 we used 225,000 separator hours, and by 1974 the effort had dropped to less than 100,000 hours.

Another interesting, but not surprising, aspect of isotope separation has been the changing emphasis on various isotopes from year to year, with no predictable way of determining which will become important at a given period of time. Figure 7 is a graph showing the variation in interest with time in the iron isotopes. When Professor Mössbauer made the discovery that some energy was absorbed in the nuclei after a nuclear reaction, interest in ^{57}Fe exploded. Several investigators wanted this 2.4% abundant isotope in hundred-gram quantities and since then we have collected kilograms of the iron isotopes. When the separation got under way, I called an investigator at one of the national labs and asked if he did not need kilogram quantities of ^{56}Fe. He said no, but he would like to have two pounds of ^{54}Fe and gave me the name of a friend in another lab to contact about the ^{56}Fe. On learning that kilogram quantities of this 92% abundant isotope were going to be available, he wrote to the AEC Division of Physical Research and said that he could do an experiment with 7 kg, but he could do a useful experiment if he had 15 kg. Figure 8 is a picture of the chemist preparing a 15-kg sample of 99.9% ^{56}Fe.

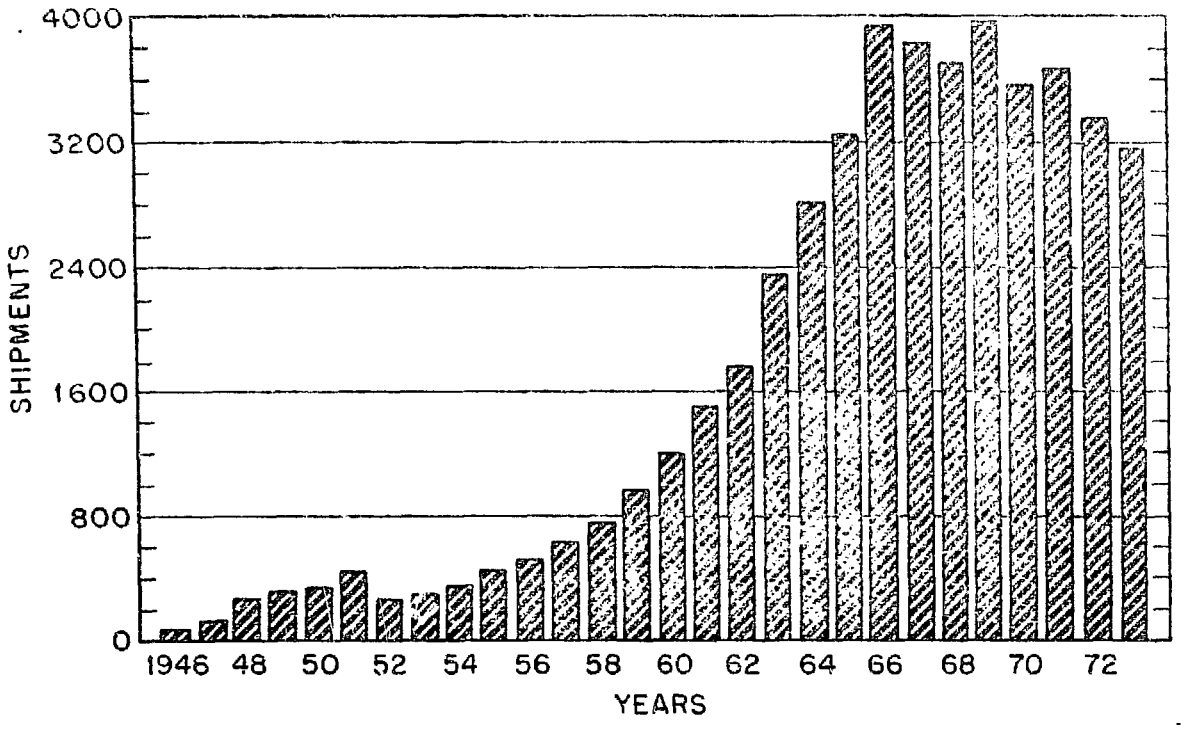


Fig. 6. Shipments of Electromagnetically Separated Stable Isotopes. (1946-1973)

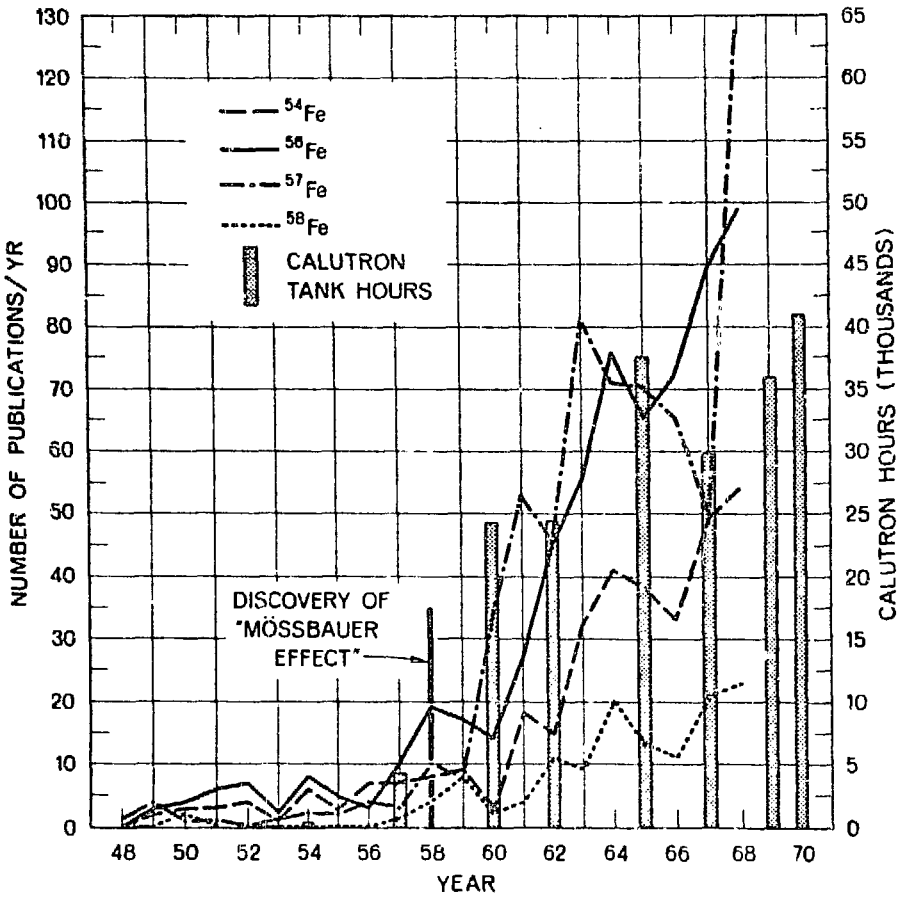


Fig. 7. Trend in Separation and Use of Iron Isotopes Enriched in the ORNL Calutron.

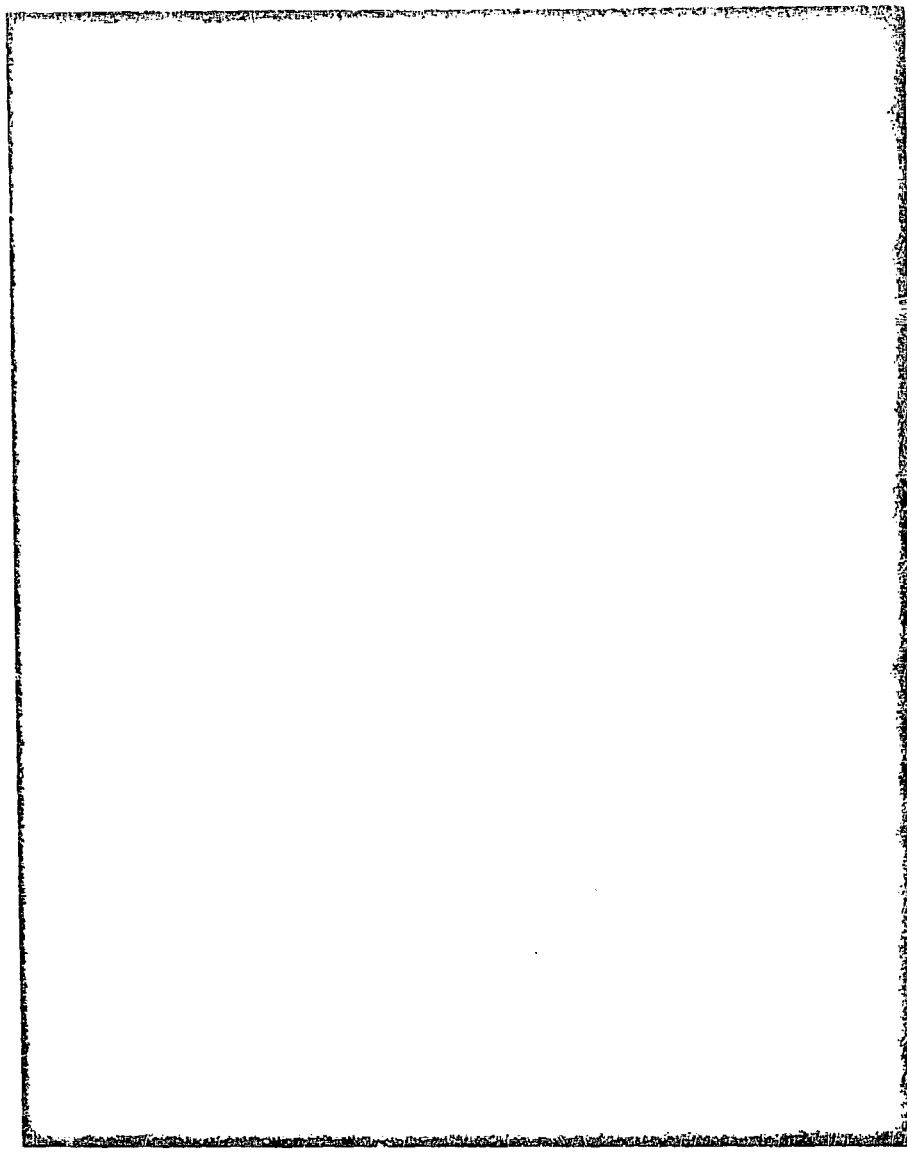
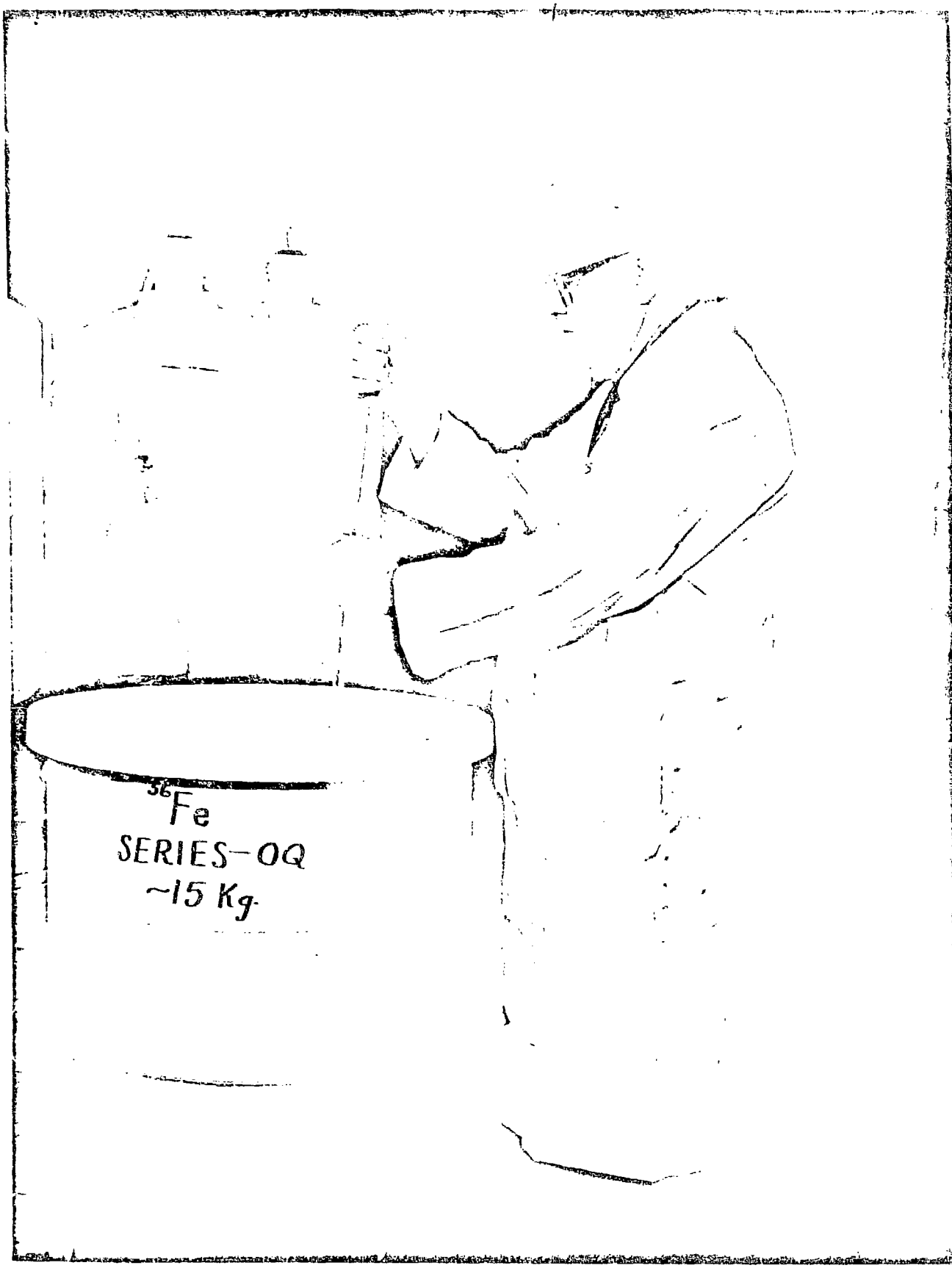


Fig. 8. Recovery and Refinement of ^{56}Fe.

I have mentioned milligram to kilogram quantities of separated isotopes and 50 $\mu g/cm^2$ targets from the calutrons, but the true versatility of the machine is perhaps more vividly illustrated by the following story. At the time that we were preparing for the iron separation, we were also interested in seeing what we could do with milligram quantities of charge. An experiment had been planned that was to use 1 mg of uranium and we were discussing it when the charge preparation chemist commented that he had just ordered a ton of iron chloride and would order another ton when the separation got under way. The milligram charge of uranium ran for one hour and forty minutes, and the ton quantities of iron for almost 100,000 hours. Thus, the calutron is a versatile machine and illustrates the extent to which the ORNL Electromagnetic Program is usually capable of providing useful quantities of highly enriched isotopes from milligram to kilogram sizes of charge of a specific element for use in nuclear research and medicine. We dispense these isotopes primarily from an on-hand inventory, but if there are items needed which are not listed in the stock catalog, inquiries concerning those materials will be welcomed.


^{56}Fe
SERIES–OQ
~15 Kg.