PHYSICS DIVISION

PRODUCTION OF CARBON 14 FROM AMMONIUM NITRATE SOLUTION IN THE CHAIN REACTOR

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A B S T R A C T

The apparatus in which the first useable quantities of Carbon 14 were produced is described. Ammonium nitrate solutions were bombarded with slow neutrons in the Oak Ridge Nuclear Reactor, and the resulting Carbon 14, appearing chiefly as carbon monoxide and carbon dioxide, was vented off to be ultimately precipitated as barium carbonate. A total of 440 mc, or 88 mg, were taken from the extraction trains.

The isotopic concentration was for the most part in the neighborhood of two to five per cent, although samples as rich as forty per cent in Carbon 14 were obtained from the carbon monoxide fraction.

Introduction

Carbon 14 was discovered several years ago,[1] and has since held the interest of physicists, chemists, biochemists, and biologists as a tracer isotope of far-reaching possibilities. At the time of its discovery it was known to have a half-life of some thousands of years, a soft beta emission with a maximum energy of approximately 0.15 Mev, and no gamma rays. In addition, it was soon learned that Carbon 14 could be prepared by slow neutron capture in nitrogen, $N^{14}(n,p)C^{14}$, with a cross section somewhat larger than 10^{-24} cm^2.[2] The existence of this reaction has become a particularly fortunate circumstance since the development of the chain-reacting pile, with its abundant supply of thermal neutrons. Irradiation of nitrogen in the nuclear reactors has made possible the preparation of Carbon 14 samples thousands of times stronger than those which heretofore could have been made with long bombardments by the largest cyclotrons. The nitrogen reaction also has another important advantage in that it results in Carbon 14 which should be isotopically pure, no other carbon being necessarily involved in the process. Consequently, material of high specific activity can be expected -- an important consideration when a long half-life is involved.

Early in 1945 at the Oak Ridge National Laboratory a Carbon 14 "factory" utilizing the $N^{14}(n,p)C^{14}$ reaction was designed and installed in the uranium-graphite reactor. This unit provided for the continuous circulation through the pile of a concentrated solution of ammonium nitrate. Since it was fairly well established the most of the Carbon 14 would combine with oxygen to form CO or CO_2, a separation tank was placed in the external piping system in order that the product Carbon 14 plus the radiation decomposition gases could be vented off through an extraction train and the Carbon 14 removed.

Such a method for producing Carbon 14 was decided upon because (1) a reasonable quantity of nitrogen could be introduced into the pile by virtue of the high water-solubility of ammonium nitrate, (2) parasitic neutron capture would not be objectionably large and no competing side reactions would be likely to give conflicting radioactivities requiring special decontamination procedures, (3) extensive radiative decomposition of the solution was considered unlikely, (4) thermal decomposition of the solution at the contemplated operating temperature would be negligible, (5) ammonium nitrate is inexpensive, and easily obtained pure and in quantity, and (7) the method seemed capable of working in a continuous fashion with minimum attention, yielding Carbon 14 which would be available almost immediately as it was produced. In addition, the necessary apparatus could be rapidly constructed from readily available materials.

The Carbon 14 Production Unit

Fig. 1(a) is a schematic drawing of the production unit; Fig. 1(b) being an alternate arrangement of the extraction train, which was used in the early stages of operation. This will be discussed later in more detail. The pump was of the glass centrifugal type, with a nominal rating of 10 gpm. The ammonium nitrate solution was circulated through the irradiation tube in the pile, led into the tall separation tank as indicated, and finally returned to the suction side of the pump to be recycled. The irradiation tube was 30 feet in length, and constructed of 1-5/8" OD 2S aluminum. It might be described as a twisted U-tube in which the legs of the U lay tightly against each other, the twist providing for complete drainage of solution during periods of inoperation. The separation tank, 6 feet tall and 6 inches in diameter, was also constructed of aluminum, and served (1) as a venting off point for the gaseous products, and (2) as a heat exchanger for controlling the solution temperature. To provide protection against any

build-up of stray induced activities in the solution, the separation tank was encased in a lead shield one inch think. The reservoir, a converted stainless steel drum, was used to supply or store solution as needed. The circulating system was constructed only of aluminum, stainless steel, and glass in order to minimize corrosion. All the connecting piping was 3/4" ID 2S aluminum. In ordinary operation the circulating system, including the irradiation tube, contained approximately 52 liters, with an active volume in the pile of 17 liters.

Gases evolved from the solution at the top of the separation tank forced their way through the diagrammed extraction trains. The several different arrangements of extraction train ultimately performed the same basic operations of (1) converting to $C^{14}O_2$ that portion of Carbon 14 present in the gas stream in other forms, and (2) precipitating the $C^{14}O_2$ as $BaC^{14}O_3$ by means of saturated barium hydroxide solution, which is a highly efficient reagent for isolating even very small quantities of carbon dioxide.

The Circulating System

The ammonium nitrate solution as made up contained 850 g of chemically pure NH_4NO_3 per liter at room temperature, which corresponded to 98% of saturation. From this a figure of 5.1 kg is obtained for the amount of nitrogen in the active volume of the irradiation tube. Samples were taken from the circulating system every month and analyzed for total nitrogen. No depletion in nitrogen concentration with time could be observed.

Besides its usefulness in removing the gaseous decomposition products, the rapid (20 l per min) circulation of the ammonium nitrate solution had an added advantage in that it facilitated control of the solution temperature. Since the temperature of the reactor structure in the vicinity of the irradiation tube was

known to be somewhat more than 100°C, it was felt that if for any reason the solution temperature should approach that range, an explosive accident might occur. By adjusting the flow of cold water through the cooling coil in the separation tank, and by ensuring sufficient circulation, the normal operating temperature of the solution was maintained at 25 - 30°C with no trouble, the temperature being observed continuously by means of a recorder connected to four thermocouples placed at equal distances along the irradiation tube. Upon shutting the unit down, the solution temperature was sometimes allowed to rise to 50°C in order to collect as much gas as possible, but prolonged operation at such temperature was avoided because of increased corrosion. For a warning device a large alarm buzzer was used. This buzzer could be activated by either the thermocouple circuits or the flowmeter (see Fig. 1(a) . If flow of the solution ceased, or if the solution temperature reached 55°C, the buzzer called attention by making violent noises. An additional factor of safety was provided by connecting the reactor trip circuits into the thermocouple network so that a temperature of 85°C would cause an immediate shutdown.

The problem of corrosion was not considered serious at the time of installation of the factory. Experiments had indicated that the effects of saturated ammonium nitrate solution on aluminum and stainless steel were negligible. Unfortunately it was soon discovered that radiation effects in the reactor result in a rapid fall in the pH of such a solution. Solution samples, taken at intervals of two weeks during a three month period of almost continual operation, showed a decrease in pH from 4.8 (fresh, unbombarded solution) to 2.4. In Fig. 2 the fall in pH during this period is plotted against bombardment time in arbitrary units. Because of this increasing acidity, considerably more dissolution of the aluminum pieces of the circulating system occurred than was anticipated,

and although such corrosion created no serious mechanical difficulty, the Aluminum 28 activity induced in the solution materially increased the gamma radiation background around the separation tank pump and piping. Increased shielding and semi-yearly solution changes were resorted to in order to keep the radiation level within tolerable bounds.

The Gaseous Products

The stream of evolved gas amounted to about 20 cc per minute. Qualitative tests showed that it contained mainly oxygen, nitrogen, hydrogen, and small amounts of nitrogen oxide and ammonia. The suggestion that appreciable amounts of air could be leaking into the system ran counter to the design feature by virtue of which the hydrostatic pressure in the system was nearly everywhere slightly above atmospheric; any inward leak would almost certainly have to be near the inlet side of the circulating pump. Tests on this point will be described under discussion of the appearance of inactive carbon in the product.

In order to take gas samples for counting, a set of mica window gas samplers was constructed, of identical shape and volume, with windows of equal thickness (3.0 mg/cm^2). These samplers were evacuated, attached to the gas line at the desired point and filled. For counting the gas samples mica window GM tubes were employed, the mica window end of the gas sampler being directed towards that of the counter tube. Decay curves obtained by collecting and counting samples of the gas stream were found to resolve into four components, one of long life which was assumed to be Carbon 14, and others with periods of approximately 14.5 hours, 2 hours, and 17.5 minutes. It is likely that the two hour period is Argon 41, but no attempt was made to identify the activities chemically.

The $BaC^{14}O_3$ Product

Removal from Extraction Train -- As can be seen from the sketches of the extraction trains, the bottom sections of the bubblers in which the barium carbonate was precipitated were removable from the train by means of large taper joints. In ordinary operation these bubbler bottoms were changed about once a week.

Upon removing a bubbler bottom from the train, the supernatant saturated barium hydroxide solution was filtered off through the built-in sintered glass filter, and the precipitate washed on the filter with warm water and alcohol. After removing the alcohol with a gentle stream of air, the precipitate was transferred to a vial and stored in a desiccator.

Chemical Purity -- Various attempts to generate CO_2 by decomposing barium carbonate taken from the extraction train indicated that the material was not chemically pure. Indeed, the average chemical purity was found to be about 85%, the chief impurities being silica, barium hydroxide, and a difficulty soluble compound of barium which was never identified. It has since been observed that the preparation of barium carbonate of 98% purity or better must be done by precipitation from warm, dilute barium hydroxide solution.

Counting -- Thin samples, prepared from the first barium carbonate collected, gave absorption curves which demonstrated the presence of a low energy beta particle emission. The range of the beta particles, as estimated visually from the absorption curves, was approximately 19 mg/cm^2 of aluminum. A Feather analysis gave the "Feather" range as being 28 mg/cm^2, corresponding to an energy maximum of about 0.15 Mev. Since no decay was observed over a period of weeks, it was assumed that the activity present in the barium carbonate was the desired Carbon 14. Mass spectrometric analyses of several samples by M. G. Inghram of the

Argonne National Laboratory, established beyond doubt that the species responsible for the activity was Carbon 14. One of the mass spectrograms obtained by Inghram is reproduced in Fig. 3.

Assay -- Routine assay of barium carbonate from the extraction train was accomplished as follows: One portion of barium carbonate was arbitrarily selected as standard material. From this thin samples were prepared and counted in a known geometry in a "low -absorption" counter.(3) By making the proper scattering and absorption corrections, the specific activity of the standard material was thereby ascertained. All other active barium carbonate was then compared to the standard material by "thick" sample counting, i.e., by comparing similar counting samples made from the unknown and from the standard material, wherein the thickness of barium carbonate in the samples substantially exceeded the range of the beta particles. For preparing such "thick" samples, aluminum plates were machined with centered wells, the wells being 3/16" in diameter and 1/16" deep. Into these wells the carbonate was tightly tamped until flush with the top of the plate. The surfaces of the plates were then wiped clean, and counts taken with a mica-window GM tube. The thickness of carbonate in the wells approximated 250 mg/cm^2, which is considerably greater than the beta particle range. When the amount of barium carbonate available for the assay was limited, plates with somewhat smaller wells were sometimes used.

This "thick" sample technique had several definite advantages. Besides being simple and rapid, counting data reproducible to 2% were obtained. Ordinarily, to attain even a minimum degree of precision when counting low energy beta particles by conventional "thin" sample methods, great care must be taken in preparing samples for counting and in making the corrections necessitated by

prominent absorption and scattering effects. Hence "thick" sample counting would seem advisable in those cases where a choice can be made.

Despite the internal consistency of the "thick" sample counts, the probable error in the reported specific activities of our $BaC^{14}O_3$ samples is estimated to be 10%. This resulted from the uncertainty in the calibration of the standard material, and from sampling errors. It should be noted that the chemical impurities present in the product barium carbonate would make isotopic concentrations calculated from specific activity data appear somewhat low. This caution applies to practically all Carbon 14 samples prepared in this apparatus.

Another matter of interest recently has concerned the possibility that Carbon 14 might be lost from active barium carbonate through exchange reactions with ordinary carbon.[4,5] In our own case, no statistically significant depletion of activity was observed. Some of the samples were followed for as long as two years, and most of them were exposed for considerable periods to the atmospheric conditions which prevail in a conventional counter room.

Operating Data

The operation of the Carbon 14 production unit can be conveniently divided into four periods. These periods, which will be described in chronological order, are distinguished by differences in the operation or construction of the extraction train. In Table 1 typical production figures are given for each period. Such typical production figures were obtained from those intervals in each period during which the unit was operating in a continuous and trouble-free fashion. Note that isotopic concentrations given in the table are denoted as "nominal", being calculated from the respective specific activities and subject to the caution mentioned above concerning the effect of chemical impurities in the samples

upon such calculations. These concentrations were obtained using a value of 350 μC per mg of 100% $BaC^{14}O_3$, this value being based on a half-life for Carbon 14 of 5100 years recently reported by L. D. Norris and M. G. Inghram.(3)

Operating Period 1 -- In the earliest arrangement of the extraction train, the gas stream was passed through a pyrex tube, 0.8 cm ID and 30 cm long, containing Hopcalite (a catalyst for oxidizing CO to CO_2 at room temperature in the presence of oxygen), and then bubbled through saturated barium hydroxide solution. This can be seen in Fig. 1B.

Since the mica window gas samplers described previously were not available at this time, gas sample counting to determine the efficiency of the train for removing Carbon 14 was not possible. Having little information at hand about Hopcalite, it was soon decided to replace it with copper oxide.

Operating Period 2 -- In this arrangement, the gas stream was passed through a quartz CuO combustion tube, 0.8 cm ID and 30 cm long, at a temperature of 700°C. It was then again led to barium hydroxide bubblers, as shown in Fig. 1B.

With the mica window gas samplers now at hand, it was possible to measure the efficiency of this train by comparing the activity of a sampler filled from the entrance gas stream with the activity of a sampler filled from the exit gas stream. It was found that 15-20% of the Carbon 14 in the gas entering the train was passing through and appearing in the exit gas.

Operating Period 3 -- To improve the efficiency of the extraction system a combustion tube of 2.5 cm ID was substituted for the previous one, and the furnace temperature was raised to 900°C. The larger diameter combustion tube served to increase the contact time of gas and CuO by a factor of 10.

Measurement of the efficiency of this arrangement of extraction train indicated that only 1.0-1.5% of the Carbon 14 entering was now being lost with the exit gas. A corresponding increase in yield of Carbon 14 was also observed, as can be seen in Table 1.

Operating Period 4 -- Since it was thought that a portion of the Carbon 14 in the gas stream would be in the form of carbon monoxide, the extraction train was altered so as to permit precipitation of the carbon dioxide component of the gas stream in front of the combustion tube (see Fig. 1A). The carbon monoxide fraction, being free to pass on to the combustion tube, could there be converted to carbon dioxide, and precipitated separately. By such an arrangement it was hoped to obtain some Carbon 14 of high specific activity. Table 1 shows that the barium carbonate resulting from the carbon monoxide fraction was considerably more active than any previous material, one sample being as much as 40% Carbon 14.[6]

By comparing the amount of Carbon 14 removed from the carbon dioxide fraction with the amount recovered from the carbon monoxide fraction, figures of 56% and 44% are obtained for the percentage of Carbon 14 present in the gas stream as carbon monoxide and carbon dioxide respectively. The percentage as carbon monoxide was considerably larger than expected, since it had been thought that no more than 30% of the Carbon 14 would be in that form.

Measurement of the efficiency of this extraction train showed that it was somewhat more efficient than the other arrangements, less than 1% of the Carbon 14 being found in the exit gas stream.

Presence of Ordinary Carbon in Extraction Train Barium Carbonate -- By making Carbon 14 from nitrogen it would appear possible to prepare samples containing only Carbon 14. Nevertheless, Table 1 shows that barium carbonate removed from the extraction train during the first three operating periods always yielded isotopic concentrations of 5% Carbon 14 or less. Although 3% Carbon 14 material is suitable for nearly all tracer work, it was felt from the beginning that attemps to improve the specific activity of the product should be made. Two possible sources of the unwanted ordinary carbon were considered, the first being carbon contamination in the ammonium nitrate solutions, and the second air leakage into the circulating system.

Since facilities were not available for working with large volumes of solution, little effort was made to remove dissolved CO_2 and carbonate contamination from the ammonium nitrate solutions before introducing them into the circulating unit. As a consequence, large amounts of low activity barium carbonate were taken from the extraction train immediately after changing the ammonium nitrate solution. However, in about five days the amount collected would fall to less than 100 mg. per day, and, thereafter neither a decrease in the amount of carbonate formed per day, nor an increase in its specific activity could be observed. Because of this, and in view of the unexpectedly high rate of gas evolution from the ammonium nitrate solution, it seemed possible that leakage of air into the circulating unit was contributing the ordinary carbon. All welds and couplings were examined, especially these at or near the suction side of the pump. The pump was submerged in water, and later placed in a box through which CO_2-free nitrogen was flushed. Despite these efforts, the specific activity of the barium carbonate showed a singular lack of improvement. Hence we can propose no satisfactory explanation concerning the presence of inert carbon in our Carbon 14 samples.

Active Carbon in the Irradiated Solution

Since the possibility existed that significant amounts of Carbon 14 in the form of carbon monoxide and carbon dioxide remained dissolved in the ammonium nitrate solutions, these solutions, having been saved and stored in a capped stainless steel drum, were examined several months after the production unit was dismantled. Aliquots were taken, from which the dissolved gases were collected by acidifying and boiling. After adding a small amount of CO_2 carrier to these gases, they were passed through a copper oxide combustion tube, and finally into bubblers containing saturated barium hydroxide solution. Upon counting the resulting barium carbonate, 0.7 mc of Carbon 14 was accounted for from the total volume of bombarded ammonium nitrate solutions. We take this as good evidence that less than 1% of the Carbon 14 formed as a gas remained dissolved in the target solutions.

The work of Yankwick, Rollefson and T. H. Norris[7] suggested that some of the Carbon 14 might be present in the solution in the form of formic acid, methanol, and (less probably) formaldehyde. W. B. Leslie of the Chemistry Division of this laboratory analyzed the ammonium nitrate solutions from our unit for the presence of these three compounds. This work has since been described in another paper.[8] Using Leslie's results, we find that our bombarded solutions contain approximately 2.8 mc, 0.24 mc, and 0.31 mc of Carbon 14 as formic acid, methanol, and formaldehyde respectively. It can be seen that Leslie found the formaldehyde fraction more active than the methanol fraction, which was not the case in the earlier reported work. The Carbon 14 remaining in the ammonium nitrate solutions as formic acid, methanol, and formaldehyde comprised 0.8% of the total amount collected.

Since extensive scanning of the extraction trains and the ammonium nitrate solutions made it impossible for large amounts of Carbon 14 to escape notice, it was assumed that the presence of significant quantities of Carbon 14 in any other

chemical forms than those already discussed was quite unlikely. Attempts were made to detect the $C^{14}N$ molecule in the gas stream and in the solutions, with negative results.

The Theoretical Yield

It is of interest to see if the yields quoted in the last column of Table I are those to be expected from cross-section and neutron flux data. The Nitrogen 14 capture cross-section for "thermal" neutrons is 1.7 barns,[9] which should be corrected for the somewhat higher effective temperature (530°K[10]) of the neutrons in the Oak Ridge reactor. The corrected value is 1.1 barns. The neutron flux, averaged over the length of the pipe in the reactor, could be estimated from foil measurements taken in the empty hole by Dr. Haydn Jones and his colleagues. The amount of nitrogen under irradiation was of course known from the volume of the pipe and the concentration of the solution. When these factors were put together, the estimated yield turned out to be 0.31 mc per megawatt-day of pile operation. The agreement with the observed yields given in Table I is satisfactory, when one remembers that the neutron flux has been assumed to be unperturbed by the presence of the solution.

Summary and Conclusions

The equipment was removed from the reactor primarily because it was wasteful of pile reactivity. Ten times the production could be achieved by loading large amounts of calcium nitrate as canned solid around the periphery of the reactor, with a negligible consumption of reactor inhours. The manpower and facilities had become available for the extra work involved. As secondary considerations, the radiation from the Aluminum 28 in the external piping was a nuisance, and unsteadiness in reactor operation caused by air bubbles in the solution was occasionally disturbing.

The apparatus manufactured a total of 440 millicuries of Carbon 14 at a time when only microcurie amounts had previously been available. Some of the material was used for the first mass spectroscopic observations and the first reasonably accurate half-life determinations. The high concentration material (40 per cent) was useful for the spin determination,[11] for beta-spectrum studies,[12] and for other nuclear physics experimentation.[13] The tracer experiments for which the bulk of the Carbon 14 was used are too numerous to mention.

Acknowledgements

We feel honored to have been associated with the late Dr. Louis Slotin in the early phases of this work. We are also indebted to E. Meiners, Jr., and F. Schuler, who aided considerably during the initial operation of the unit. A great deal of credit is due Miss Thelma Arnette and Mrs. Ida Coveyou for assistance in the preparation and counting of samples. The majority of the work was done under the auspices of the Manhattan District.

TABLE 1: Typical Carbon 14 Production Figures for Various Operating Periods

Operating Period	Extraction Train Arrangement	Total C^{14} Collected During Reported Interval	Accumulated Bombardment (Megawatt-days of Reactor Power)	Average Spec. Act. of C^{14} Collected	(Nominal) Average Isotopic Concentration of C^{14} Collected	Spec. Act. of Best Sample Collected During Reported Interval	(Nominal) Isotopic Concentration of Best Sample	Average Yield per Megawatt-day
1	Hopcalite Oxidation Tube Room Temp. No separation of $C^{14}O$ and $C^{14}O_2$ fractions	61 mc	207	8.1 μc/mg	2.3%	16.8 μc/mg	4.8%	0.29 mc
2	CuO Combustion Tube 700°C. No separation of $C^{14}O$ and $C^{14}O_2$ fractions	59 mc	217	13.3 μc/mg	3.8%	16.2 μc/mg	4.6%	0.27 mc
3	CuO Combustion Tube 900°C. No separation of $C^{14}O$ and $C^{14}O_2$ fractions	65 mc	178	13.4 μc/mg	3.8%	15.1 μc/mg	4.3%	0.36 mc
4	CuO Combustion Tube 900°C. $C^{14}O$ and $C^{14}O_2$ fractions precipitated separately	CO Fraction 48 mc	241			(1) 111 μc/mg (2) 78 "	See Footnote 6	0.20 mc
		CO_2 Fraction 37 mc	"	7.1 μc/mg	2.0%	8.5 μc/mg	2.4%	0.16 mc

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(3) L. D. Norris and W. G. Inghram, Phys. Rev. 73, 350 (1948)

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(6) As calculated from the specific activities, one gets 32% and 22% respectively for these two samples. But F. A. Jenkins, Department of Physics, University of California, has measured band intensities on grating spectrograms prepared from these materials, and has found that the ratio of Carbon 14 to total carbon is 40% in the one case and 27% in the other (Private Communication, 1948).

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CAPTIONS

Figure 1A: Schematic drawing of Carbon 14 Production Unit. The extraction train shown permitted separate precipitation of the $C^{14}O$ and $C^{14}O_2$ fractions.

Figure 1B: An extraction train arrangement used in the early stages of operation of the Carbon 14 Production Unit. No separation of the $C^{14}O$ and $C^{14}O_2$ fractions was accomplished, all of the Carbon 14 being first converted to $C^{14}O_2$, and then precipitated.

Figure 2: Fall in pH of ammonium nitrate solution plotted against pile bombardment (in arbitrary units).

Figure 3: Recorder tracing of the isotopes in normal carbon dioxide and in carbon dioxide containing Carbon 14. The regions about mass 45 and 46 are recorded at 40 times the sensitivity used in recording the mass 44 peaks.

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FIG 1 - A

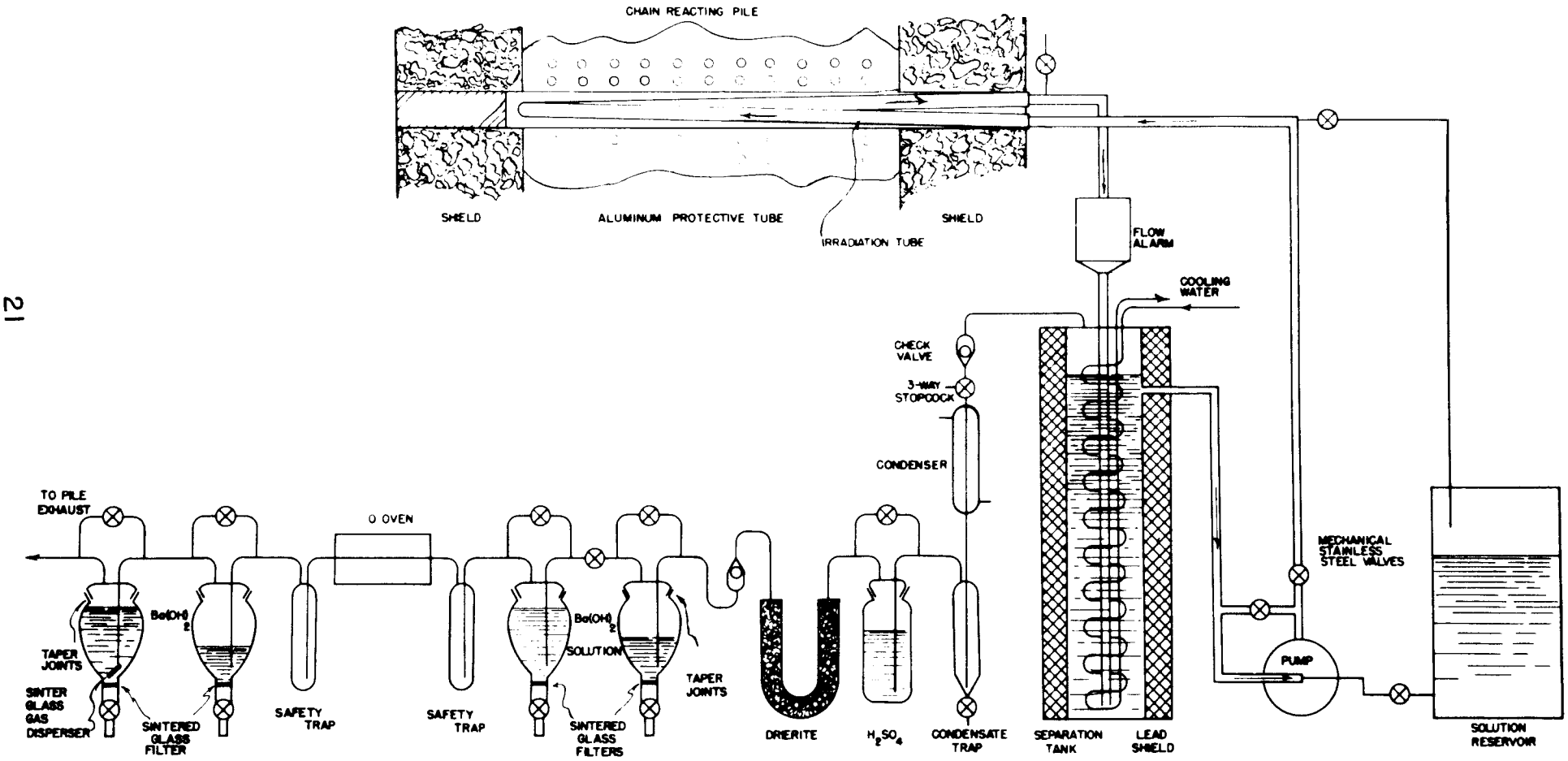
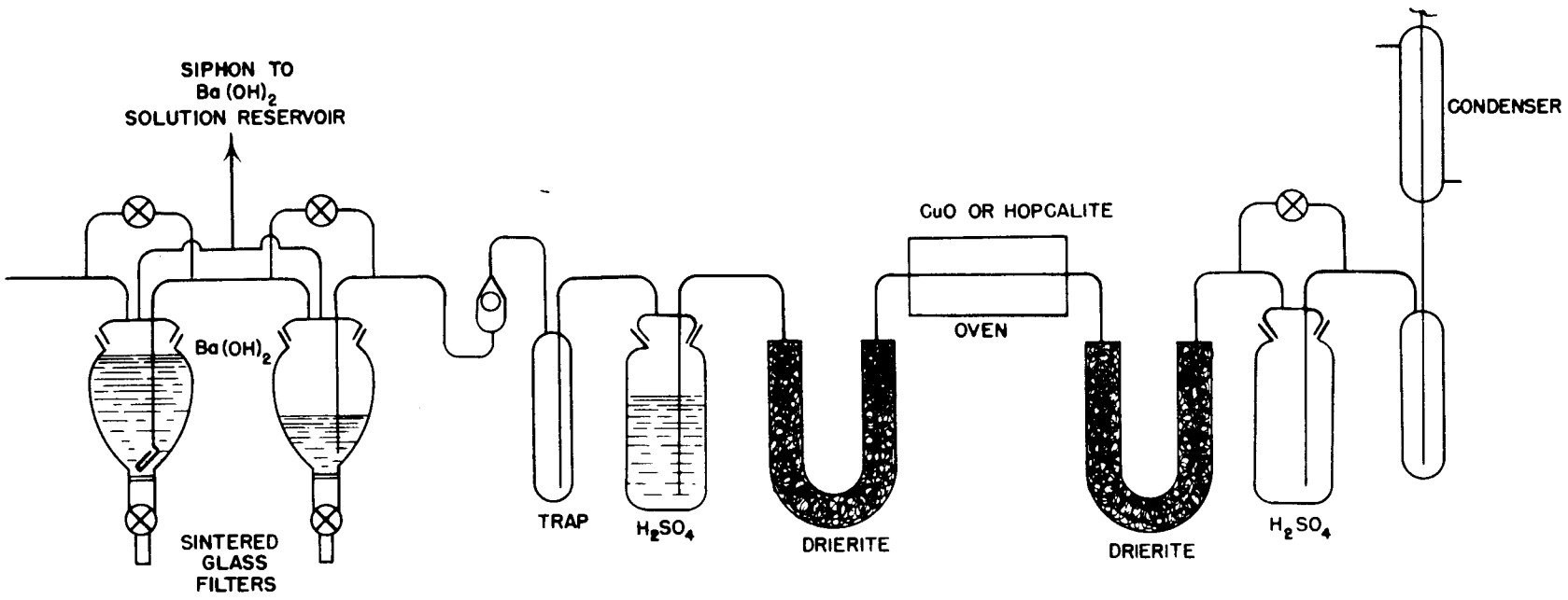


FIG. I – B


SIPHON TO
Ba (OH)2
SOLUTION RESERVOIR
CONDENSER
CuO OR HOPCALITE
OVEN
Ba(OH)2
SINTERED
GLASS
FILTERS
TRAP
H2SO4
DRIERITE
DRIERITE
H2SO4

UNCLASSIFIED

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FIG. 2

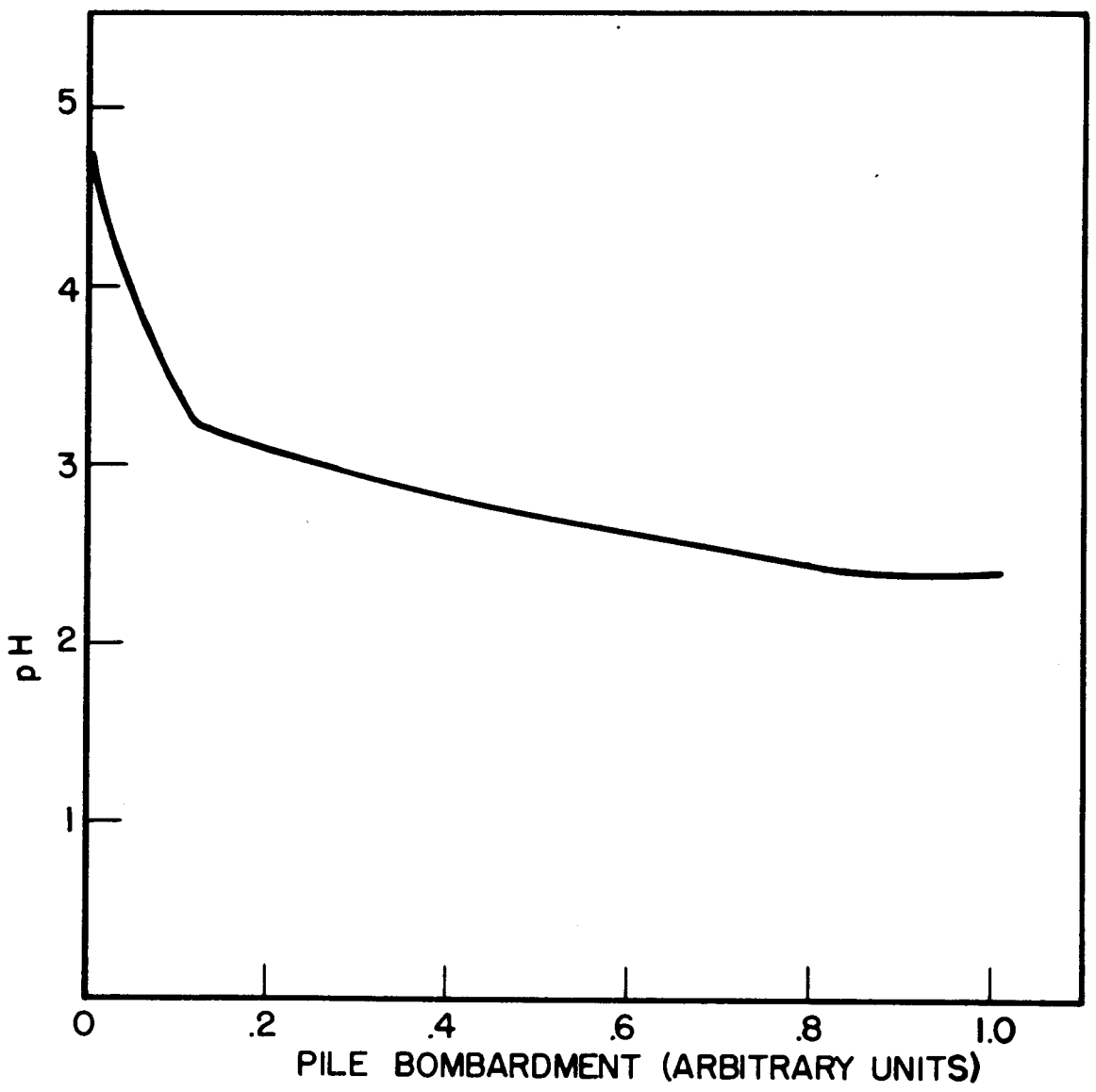

FIG. 3 TRACINGS OF MASS SPECTROMETER CURRENTS FOR CO_2