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CARBON-14 EXTRACTION FROM REACTOR GAS
PROGRESS REPORT
PRODUCTION TEST AUTHORIZATION #124

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CARBON-14 EXTRACTION FROM REACTOR GAS
PROGRESS REPORT
PRODUCTION TEST AUTHORIZATION #124

J. P. Cooke
Engineer
Equipment Systems Engineering

January 24, 1969

DOUGLAS UNITED NUCLEAR, INC.
RICHLAND, WASHINGTON

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INTRODUCTION

Carbon-14 is by far the most popular of the labeled isotopes because of its adaptability to so many chemical compounds and its relatively harmless characteristic beta radiation. Because the inert gas atmosphere contains up to 30 percent nitrogen, carbon-14 is generated within the K Reactors by the conversion of nitrogen-14, and if not immediately oxidized, carbon-14 may remain in the reactor core for some time. It is eventually oxidized and escapes when the outward gas leakage from the reactor is vented to the stack. Production Test 124 (Ref. DUN-3825) was authorized to determine if carbon-14 could be extracted from the K gas as a profitable by-product.

SUMMARY & CONCLUSIONS

As much as 500 or 600 curies per year of carbon-14 could be recovered if both K Reactors were provided with relatively simple extraction facilities. This is about twice the amount of this product for which there is a current market at the present price level of approximately \$3000/curie.

The grade (as specific activity) of the product recovered in the production test was low - ranging from 3 to 26 millicuries per mol. Substantial upgrading (perhaps by a factor of 10X to 100X) is possible by skimming techniques and reduced oxidation of normal carbon at Hanford, but economic parameters of upgrading vs total production rates will require additional market volume and price data. Two further methods of upgrading the product after it leaves Hanford are discussed briefly.

DISCUSSION

Test Results

Small quantities (approximately 300 grams) of 8 mesh "Ascarite" and 1/8" pellets of 5A Molecular Sieve were placed in a screen-lined perforated cylindrical holder of 1 3/4" diameter which was inserted in the circulating gas stream of KE Reactor. An insertion port had previously been provided in the housing of the filter unit, downstream of the gas dryers, where suitable isolation and bypass valving would permit samples to be changed during operation. The fact that the gas stream was pre-cooled and dry, therefore, made the entire adsorptive capacity of the reagent available for CO₂. Details of the sample collections are shown in Table I. Analytical results from the ARHCO Purex Lab are shown in their letter report (Appendix A). These may be compared with results from ORNL in Appendix B.

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Market Requirements for Carbon-14 & Present Production Status

The pure carbon-14 isotope would have an activity of 4.42 curies per gram, or $4.42 \times 14 = 62$ curies per mol. (This is usually expressed as millicuries/millimol). The percentage concept is also useful. (Percent of above theoretical level.)

Carbon-14 is difficult to produce in pure form, and the 95+ percent grade currently available is prepared by capsule irradiation of beryllium or aluminum nitride. Two years or more are required in the K irradiation facility

In many chemical compounds, only a selected site in a chemical structure is labeled with carbon-14 and the maximum possible specific activity of such compounds is thus further limited.

Most research activities involve no more than millicurie or microcurie amounts of carbon-14. Interest in higher specific activities stems from two sources:

1. A desire for labeling of one particular carbon atom position in the molecule to determine biological result of a particular molecular structure. This technique is also useful in synthesis chemistry, etc.
2. A need to provide better beta-counter response and level of carbon-14 in clinical examinations or dissections. This would be the case for studies with certain plant or animal organisms in which an extremely toxic or powerful compound was being used, and it is not desired to poison the test organism with the compound before enough carbon-14 can be deposited to yield usable research results.

There are however, many research activities involving greenhouse or plant uptake studies in which radio-carbon-14 in the form of carbon dioxide would be quite applicable even though percentage of carbon-14 were low. These levels of specific activity may also be useful as instrument calibration standards.

Product-Upgrading & Extraction Processes

It is also possible to upgrade the product if the raw material is cheap enough and in plentiful supply. As the first step in the present processing of carbon-14, material obtained from the capsule is converted to carbon dioxide. Since we start with carbon dioxide this step would not be necessary. (See Appendix C for details of extraction processes at the various sites.)

*Personal comment from FN Case, ORNL

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From the difference between molecular sieve samples #1 and #2, it would appear that some graphite from the central core is being oxidized during operation and that carbon-14 is also being removed with these oxidation products. The fact that the highest level of carbon-14 was found in the samples collected over the three week operating period is contrary to previous expectations. Thus, if collection is to be scheduled over this portion of the operating cycle, efforts to improve the grade of carbon-14 will be most productive if directed toward reduced oxidation of normal carbon. Alternatively, if optimum collection conditions can be programmed for part of the startup cycle, as originally outlined in PT-124, it will suffice merely to control or limit the central zone oxidation (and carbon monoxide formation.)

Carbon Monoxide Problem & the Influence of Graphite Burnout

Carbon monoxide is adsorbed only one tenth as strongly as CO₂ on 5A molecular sieve, and not at all on alkaline adsorbents such as ascarite or lithium hydroxide. The carbon-14 yield will therefore suffer unless means are provided to deal with carbon monoxide.

Carbon monoxide is assumed to be formed within a K Reactor by three mechanisms:

1. Water-gas reaction from traces of moisture at temperatures above 750°C.

The amount of water present in the hottest part of the reactor is difficult to assess (the reactor inlet content is less than 10 ppm and the outlet content is usually less than 50 ppm.) It is probable that most of the 50 ppm of H₂O comes from the rear vanstone zone but the presence of this 50 ppm makes it impossible to measure the depletion of the inlet moisture content that might be attributed to the water gas reaction. The formation of hydrogen cannot be used as a measure of the reaction, since most of the H₂ is converted to ammonia*. This NH₃ is removed in the dryer dessicant (3A² Molecular Sieve.) Only through measurement of volume and concentration of NH₃ in the dryer condensate could a rough approximation of the hydrogen formation rate and water gas reaction rate be obtained, this would be a totalized value for a period of several weeks. The value would also be subject to bias from possible variations in the dryer regeneration cycles. Such measurements, though of much theoretical interest to burnout evaluators, have never been made.

As an alternative estimate of the extent of the water-gas reaction at KE the following bases are offered:

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Gas circulation 1000 cfm = 1,440,000 cu ft/day
Assume 10 ppm inlet moisture content is entirely
reacted to form CO.

$10 \text{ ppm} \times 1.4 \times 10^5 = 14 \text{ cu ft CO.}$

This would account for practically all of the CO
in the 3000 cu ft daily gas loss. $.5\% \times 3000 =$
15 cu ft CO.

A substantial reduction in water gas burnout should be possible by reducing the flow rate to the 100 cfm minimum value allowed by the process standards. Such an expedient would demonstrate the extent to which the CO in K Reactors originated from the water gas reaction vs from reduction of CO₂ by hot graphite. (See discussion in next paragraph.) The presence of NH₃ in the dryer condensate favors the water gas origin theory.

As a point of immediate practical importance, the above chemical reaction is promoted by the rapid removal of the hydrogen (as NH₃), and could best be inhibited by additions of CO.

2. Conversion of CO₂ to CO in the presence of carbon at temperatures above 750-800 C. (With a maximum stringer graphite temperature of 850°C this may probably be the source of about half the CO, (especially when the rate temporarily exceeds the .5 percent mentioned above.)

This reaction is temperature-dependent and goes to completion at about 1000 C. At lower temperatures it can be inhibited by adding CO. Hydrogen would also inhibit by a reverse water gas reaction, but as has been mentioned above, the ammonia generation effectively removes almost all the hydrogen. Because this reaction does not go to completion at the K graphite temperature, the CO percentage from this source will change less with reduced gas circulation than CO from water gas. There will be some change however, because complete stopped circulation with stagnant gas in the reactor core would stop CO formation at the localized equilibrium level for lack of more CO₂.

3. A combination of the above reactions with the carbon dioxide reduction being the more important, and localized hot spot temperatures playing a very important role.
4. Radiolysis of CO₂ may also play a role.

The 850° front to rear stringer temperature limit is monitored in only four channels. The hottest spot moves around from day to day according to rod position and other factors. Historically, the unofficial 903°C temperature record was attained in one of these stringers when first installed, with a different fuel configuration than currently used. This was accompanied by a CO concentration of three percent in the bulk gas. By the next day the temperature had been reduced to 860° and the CO promptly followed it down

to approximately two percent. A similar phenomenon was noted at D Reactor when a test coupon burnout channel was instrumented with thermocouples and provision made to sample the gas at corresponding points. (Reference DUN-1375 - Production Test Authorization - Investigation of Operating Conditions of Air Channel 3478-D and DUN-3295, the final report of that test.)

In the above test, the local CO content in the 3478-D "hot" channel was two to four times that in the bulk gas, ranging to eight percent on one occasion.

When the K Reactors were equipped with Zirconium tubes the channel straightening tooling left small zones with greater than normal clearances between process tubes and the graphite. There are no means to measure graphite temperatures in the adjacent filler blocks. Since total burnout is being monitored as well within the two pounds/day adopted as an arbitrary limit*, there is little incentive to try to measure or estimate such localized temperatures.

Since no more than 30 cubic feet of CO is currently formed per day in routine operation, this may be assumed to be approximately the equilibrium percentage (.6 to .8) percent) corresponding to the prevailing temperature or average temperature pattern. Addition of 60 cubic feet per day of CO should completely reverse the reaction, and if carefully metered, need not upset the dual burnout monitoring required by process standards.

Carbon monoxide is present in approximately equal amounts to carbon dioxide during equilibrium operation. Since carbon monoxide can be formed only in local hot spots or in higher temperature core graphite where the carbon-14 would be deposited, it appears that to maintain the richest possible fraction of carbon-14 isotopes it will be desirable:

1. To reduce gas circulation to the minimum, (or stop it altogether).
2. To operate the gas system on practically zero oxidation throughout the operating period.
3. To minimize the formation of carbon monoxide either by control of local graphite temperatures and by deliberate additions of normal carbon monoxide to prevent and inhibit formation of carbon-14 as monoxide. This may be possible because of the definite and predictable ratio of carbon monoxide to carbon dioxide depending on the temperature, when both gases are in contact equilibrium with graphite. Probably no more than 50 or 60 cubic feet per day of carbon monoxide would be required to inhibit the formation of this carbon-14 monoxide gas within the reactor by reversing the reaction.

*Two pounds of carbon burnout/day has been determined as well below the level at which burnout could possibly be the limiting factor in useful life of the Hanford Reactors. Ref. K Reactor Process Standards HW-46000-K.

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The addition of CO will have to be carefully controlled for a metasthetical process is possible and such an exchange of carbon-14 from one gas to the other would probably be enhanced by a large excess of CO.

An on-line means for analysis of CO and CO₂ for carbon-14 content would be necessary to permit optimization of the carbon-14 grade and collection rate. The next logical step in this test program will involve study of the effect of flow and moisture content plus the ratio of carbon oxides on the carbon-14 in the gas.

Beta radioactivity analysis for carbon-14 is complicated by the gamma radiation from the Argon-40 isotope present in the gas. Fortunately the CO₂ and CO fractions can be separated easily by gas chromatography. A typical chromatographic analysis of K gas during equilibrium operation is as follows:

CO₂ - .6 to 1%
O₂ + AR = 200 - 400 ppm (mostly Argon)
H₂ - 30%
CO - .6 to 1%

Experiments with the tail gas from the spare gas chromatograph in KE during the early periods of the carbon-14 collection showed that a radiation detector mounted outside of the gas instrument room could respond to the successive fractions of gas coming through the chromatograph. Since the sample size on the standard reactor gas instrument is quite small, the larger sample possible with a laboratory model gas chromatograph (recently obtained from the 300 Area) should be used in future tests. Use of longer columns and larger samples will give a better signal than was obtained from the first experiments. The high gamma background in the gas instrument room makes it necessary that such a beta counter be heavily shielded. The detector chamber could also be placed in a lead shield with shorter tubing runs so the sample cuts would be sharper and less diffused.

An alternative and perhaps more flexible analytical technique uses a 7/8 inch sample wafer of 4A molecular sieve mounted on a Beta counter planchette. This can be used to analyze for carbon-14 by impinging the chromatograph tail gas at the center with a close fitting plate orifice (.010" - .015" separation.) A planchette of this type will absorb CO₂ from a dozen or so analyses and can then be removed and counted in the RM lab remote from the gamma background of the gas instrument room.

After each analysis, the planchette pellet can be regenerated by heating at 500 - 600°C followed by cooling in a sealed container under a bed of molecular sieve pellets. A standardized sample of carbon-14 in carbon

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dioxide form will be required for calibration with both of these techniques. [REDACTED] has some accurately standardized Barium Carbonate which presumably can be used to provide satisfactory CO₂ standard gas by treating it with acid and passing the stream over a dessicant.

Preliminary tests with the sample planchette have not been encouraging, as there seems to be enough affinity for radio-argon for its gamma to obscure the beta from carbon-14. Inherent requirements for a prompt on-line analysis will not allow enough time for argon decay.

G. B. Barton of BNW chemistry labs suggests use of a Barium Hydroxide solution and beta counting the precipitated Barium Carbonate on a small filter paper. Such an analysis would require less than 10 minutes and the geometry of the beta counter could be accurately compensated by counting a weighed amount of the standard ORNL material in the same setup.

If the Hanford product can be upgraded by a factor of 5 or 10 through skimming techniques, this could possibly make an attractive feed material for [REDACTED] and they also might be able to separate a carbon-14 product.

Engineer
Equipment Systems Engineering

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TABLE ICIRCUMSTANCES OF SAMPLE COLLECTIONS:

Ascarite #1 was on the line approximately 5 hours at startup of June 4-5. The previous shutdown had been made with gas circulation stopped. The following is from the operation log sheet:

	<u>Time</u>	<u>Power Level</u>	<u>CO₂</u>	<u>CO</u>	<u>N₂</u>	<u>GT</u>
	8:30 pm	1350	.70	0	35	No record
	9:30 pm	3400	.25	0	35	" "
June 4	10:30 pm	3400	.25	0	35	" "
	11:30 pm	3450	.52	0	35	" "
June 5	12:30 am	3550	.68	.04	35	708

The sample holder was placed on bypass for 6 hours until the gas activity died off enough so that it could be removed. The next sample was put in @ 8:30 am. Interim log data is of interest in that it shows the steady trend and concurrent appearance of CO and graphite temperatures of over 750°.

	<u>Time</u>	<u>Power Level</u>	<u>CO₂</u>	<u>CO</u>	<u>N₂</u>	<u>GT</u>
	2:30 am *	3750	.78	.15	37	766
	4:30 am	3950	.86	.14	37	802
Ascarite #2 sample in	6:30 am	4100	.90	.14	37	802
	8:30 am	4100	.90	.12	37	778
	10:30 am	4200	.88	.32	37	788
	12:30 pm	4280	1.08	.32	37	806
	2:30 pm	4360	.55 **	.24 **	33	830
	4:30 pm	4370	1.14	.46	33	836
	6:30 pm	4370	1.18	.54	33	846
	8:30 pm	4370	1.20	.54	33	850
	10:30 pm	4370	1.24	.54	33	854
June 6	12:30 am	4370	1.23	.70	33	850
	6:30 am	4380	1.14	.70	33	837
	8:30 am	4400	1.14	.79	33	848
thru 6-9	Midnite ***	4400	.82-.95	.72-.90	38-41	825-844

Sample holder was bypassed and sample removed 8:30 am Monday June 10.

* The paramagnetic O₂ analyzer showed .05% O₂ at this point.
No readings taken before this.

** Readings anomalous.

*** Stable operation.

TABLE I (Continued)

Ascarite #3 covered the remainder of the operating period, including an unscheduled outage June 16. During the outage the sample was placed on bypass and not restored to service until full power was again reached July 1.

<u>Date</u>	<u>Power Level</u>	<u>CO₂</u>	<u>CO</u>	<u>N₂</u>	<u>GT</u>
6-7	4400	.82-.95	.72-.90	38-41	825-844
6-8	4400	.78-.81	.92-.97		850-853
6-9	4400	.80-.94	.82-.92	40-41	848-850
6-10	4400	.88-.92	.80-.92		853
6-11	4400	.83-.86	.72-.78	39-40	848
6-12	4400	.87-.88	.63-.70		850
6-13	4400	.78-.86	.70	37	846
6-14	4400	.74-1.00*	.68-1.11*		852
6-15	4400	1.00	1.14		845
6-16	4400	.97-1.02	1.08-1.15		842
6-23	4370	1.0	.64-70	38	848-850
- - - Unscheduled outage and startup - - - - -					
7-1	4350	.98	.50-.52	37.5	833
7-4	4320	.95-1.0	.46-.50	37.5	838
7-7	4210	1.0	.40	38.5	846
7-17	4250	1.0	.31	33	826

* Recalibrated

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TABLE II

Molecular Sieve Sample #1 was on stream for 4 hours after startup of 7-23-68 and was put on bypass at approximately 2 a.m. at a power level of 3100 and 664° maximum graphite temperature. The startup was interrupted by a scram from 2350 power level at 8:30 p.m. The following data are from the gas log sheet:

<u>Date & Time</u>	<u>PL</u>	<u>CO₂</u>	<u>-O₂+A=.1</u>	<u>N₂</u>	<u>Max. GT</u>
Prior to startup (7pm)	0	0		.4	
7-23 - 8:30 pm	2350 (Scram)	0		19	
11:30 pm	1450	.08=24 cu.ft.	0	19	.48 O ₂ +A=150 cu.ft.
7-24 -12:30 am	3100	.10= 30 cu.ft.	0	19	.005 O ₂ +A=1.5 cu.ft.
2:30 am	3100	.65=195 cu.ft.	0	20	(664°)033 O ₂ +A=10 cu.1
		6.5 #			

Molecular Sieve Sample #2 was left on for the balance of the operating period with 6 hours off line between samples. Gas log data continues:

<u>Date & Time</u>	<u>PL</u>	<u>CO₂</u>	<u>CO</u>	<u>N₂</u>	<u>Max. GT</u>	<u>O₂+A</u>
4:30	3225	.70	0	20	707	
6:30	3425	.64	0	19.2	742	
8:30	3615	.72	0	19.8	756	.045
10:30	3725	.84	0	20	786	.038
12:30	3845	.92	0	20	813	.036

Sample #2

2:30	3935	X	.34	29	823	
4:30	3975	.92	.22	37	856	
6:30	3975	.91	.22	37	840	
8:30	4055	.90	.4	32	832	
10:30	4100	.92	.44	36.2	845	
7-25	4125-4200	.90-.97	.42-.48	34-36	800-848	
7-26	4170-4180	.89-.92	.45-.54	33	857	
7-27	4170-4135	.96-1.0	.38-.42	34	814	
7-28	4135-4100	.90-1.0	.29-.3	34	804	
7-29	4100	.85-.90	.29-.30	34	834	
7-30	4100	.83-.91	.25-.26	34	802	
8-1	4100	.82-.89	.21-.24	34	798	
8-2	1	.86-.97	.17-.21	34-35	792	
8-3	4100	.96-.98	.20-.26	36	828	
8-4	4100-4090	.98-1.02	.24-.28	36	791	

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APPENDIX A

DUN-4745-DE

Date: October 21, 1968
To: Philip J. Cooke
From: LP McRae *Larry P. McRae*
Subject: ANALYSIS OF CO₂ FOR C¹⁴ CONTENT

Ascarite Analysis

	<u>uci C¹⁴/mole C¹²O₂</u>	<u>uci C¹⁴/g ascarite</u>
Sample #1	2700	8.2
Sample #2	3300	16
Sample #3	4600	36

The material appears to be approximately 89.0% NaOH and approximately 11% inert material (asbestos).

Molecular Sieve Analysis

	<u>uci C¹⁴/mole C¹²O₂</u>	<u>uci C¹⁴/g sieve</u>
Sample #1	9400	.82
Sample #2	26000	2.8

Methods Utilized

Ascarite:

The samples were ground and then dried in 110° C oven for 24 hours. A sample was weighed out and dissolved in distilled water. An aliquot was analyzed for CO₂ by the Winkler Method using a pH meter for endpoint detection.

Another aliquot of solution was mixed with a POPOP-PPO liquid scintillation solution and analyzed for C¹⁴ content. The beta detection system functioned at about 92% efficiency. A quench curve for measurement of efficiency was obtained using a solution of ascarite and caustic in approximately equal M strength of OH ion. Counting was performed on a Beckman CPM 200 model liquid scintillation analyzer.

Molecular Sieve:

Several methods were investigated for the release and trapping of the CO₂ held by this material. A gas train was fabricated for acid release and caustic trapping with or without N₂ transfer of the CO₂. The results were poor

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due to design of the system as C^{14} was found in all down stream traps and we were not sure all CO_2 was being retained by the traps. Another drawback was the analysis of more than one trap to obtain a measure of total C^{14} activity.

A final method of release was attempted whereby a dilute 0.04 M NaOH solution was used as both a generator and as a trapping solution for CO_2 . After several attempts, the technique was formalized. The measure of complete recovery was taken to be the maximum activity attainable from a sample.

Generally, a sample is placed into a gas-tight flask of 0.04 M NaOH and heated for about 1 hour under a heat lamp and allowed to cool for 1 hour or until the gas stops bubbling. An aliquot is then analyzed for CO_3 by the Winkler Method as before and an aliquot counted for C^{14} in the liquid scintillation system.

Discussion

The ascarite material being very hydrosopic, was hard to grind with a mortar and pestle to get a uniform sample. When placed into a drying oven, the samples extracted a large amount of water from the air inside the oven. Therefore, we feel these samples were contaminated with CO_2 and thereby diluted the C^{14}/C^{12} ratio.

After placing these samples into a dry oven and drying at $110^\circ C$ for 48 hours, we began the analysis for CO_3 and C^{14} . Both types of material as handled, suffer from an atmospheric CO_2 interference. The ascarite interference can be lessened some by increasing the concentration considerably above the levels we used. Another approach for either material would be to use an inert atmosphere of N_2 . This, however, is somewhat unhandy for us.

LPM/kav

cc: GL Gurwell
JD Anderson
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APPENDIX BReport of Mass Spectrometer Analyses of C-14 Samples Sent to ORNL

(Residues of sample containers previously analyzed at Purex Lab.)

Molecular Sieve	Content as CO ₂		Variety
	C-12	C-13	C-14
Control	98.92%	1.08%	≤ 0.05%
#1	98.16%	1.65%	.19%
#2	98.16%	1.57%	.27%

The Ascarite samples showed higher values but on rechecking the analyses for extraneous peaks, the Ascarite was found to yield a wide range including Boron, Fluoride and several others not specifically identifiable.. The following ORNL results should therefore be regarded as maximum figures:

	<u>C-12</u>	<u>C-13</u>	<u>C-14</u>
1	97.34	2.33	.33
2	97.64	2.06	.30
3	97.67	2.04	.29

Reducing the results to the same terms and ignoring the ORNL results with Ascarite,

Sample #1 - .19% C-14 specific activity =

$$.0019 \times \frac{62. \times 10^6}{1000} = \underline{11.7 \text{ millicuries/mol.}}$$

 compared to 9.4 millicuries/mol. by beta activity methods.

Sample #2 - .27% $.0027 \times \frac{62 \times 10^6}{1000} = \underline{16.8 \text{ millicuries/mol.}}$
 compares to 26.0 millicuries/mol. by beta activity methods.

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APPENDIX C

Extraction Processes

Preparative gas chromatographic techniques are suggested by work of Professor A. Liberti of University of Rome (Anal. Chemistry Nov. 1967 P 1618) who made separations of methane (deuterium and tritium forms) in this manner.

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APPENDIX D

Potential Hanford Production Level

For evaluation of potential production rates of carbon-14 the following bases may be found useful:

1. In Production Test 124, the carbon dioxide content in the gas system during startup (first four hours) was approximately 3/4 of 1 percent by the time the graphite temperature had reached 500 500 or 600 degrees centigrade. (No measurable carbon monoxide had been formed as yet.) At a gas system volume of approximately 30,000 cubic feet this means that approximately 210 cubic feet of carbon dioxide had then been generated. This represents seven pounds of carbon or approximately 3200 grams. In PT-124 the specific carbon-14 content found was .8 millicuries per gram. Thus, 2.56 curies would have been collectable over this four hour operating period without interfering with normal operations.
2. The current nitrogen content in the gas atmosphere is now up to 30 percent vs the 10 percent maximum which prevailed at the time the Barton study reported a level of .3 curie/day in documents HW-75618 and HW-84569. With present higher power levels, the carbon-14 formation should now be at least approximately 1 curie/day in each K Reactor.
3. Accepting a carbon-14 content of 2.16 millicuries/gram in the 1 percent carbon dioxide portion of the average gas composition over the three week normal operating period and assuming a gas leakage loss of 3,000 cubic feet per day to the stack, then 30 cubic feet per day of CO₂ will be formed and one pound carbon burned out. Then:

$$\frac{2.16 \times 453 \text{ grams/lb.}}{1000} = .99 \text{ curies/day}$$

is indicated over the three week normal operating period.

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