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S. G. Thornton
Environmental Management Division
OAK RIDGE K-25 SITE
for the Health Studies Agreement

July 1995

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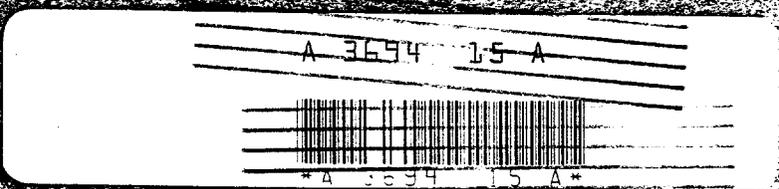
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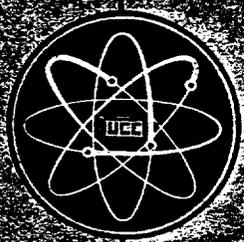
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AEC RESEARCH AND DEVELOPMENT REPORT



DECAY PRODUCTS OF U-235 AT THE K-25 PLANT

REPORT



K-25 GASEOUS DIFFUSION PLANT

Operated by

UNION CARBIDE NUCLEAR COMPANY
DIVISION OF UNION CARBIDE AND CARSON CORPORATION

for the Atomic Energy Commission

Under U.S. Government Contract W740-06-0001

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CARBIDE AND CARBON CHEMICALS CORPORATION
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DECAY PRODUCTS OF U-234 IN THE K-25 PLANT

R. L. Macklin

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CLINTON ENGINEER WORKS
CARBIDE AND CARBON CHEMICALS CORPORATION

Research Laboratory

DECAY PRODUCTS OF U-234
IN THE K-25 PLANT

R. L. Macklin

C. K. Beck
Director Research Laboratory

A. D. Callihan
Physics Department

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S E C R E T

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RESEARCH LABORATORY

Date: January 30, 1947
Serial No.: A-3694
Written by: R. L. Macklin

DECAY PRODUCTS OF U-234
IN THE K-25 PLANT

1 - SUMMARY

The quantities of ionium and radium deposited in the K-25 plant have been estimated from plant inventory and disintegration constant data. The radon gas from this source (Ra) in the product and in laboratory samples has been calculated to lead to no more than one alpha particle per minute in a standard counted film.

11 - INTRODUCTION

The natural radioactive decay products of process material have occasionally been considered as possible causes of a number of phenomena. For example, the UX-complex presents, because of its concentration in certain operations, a potential health hazard.

The single gaseous member of the uranium disintegration series, radon, has been proposed as a possible product contaminant.

S E C R E T

Its low molecular (and atomic) weight of 222 indicates a fairly rapid rise to the top of the diffusion cascade where its boiling point of -62° C and freezing point of -71° C would lead to its inclusion in "frozen out" samples of product. In this condition the short-lived active deposit (Ra A, Ra B, Ra C, Ra C¹, Ra C¹¹) would approach transient equilibrium in about two hours and therefore contribute twice as much alpha radiation as the radon. Traces of radon amounting to a few micrograms would produce decided irregularities in analysis because of alpha emission. For example, one microgram of radon plus active deposit emits seventeen billion (1.7×10^{10}) alpha particles per second. In comparison, a product sample film for laboratory analysis is expected to emit at most a few thousand alpha particles per second.

In May, 1946, the concentration of radon in a product cylinder was roughly calculated and shown to be far too small to account for the observed gamma radiation from such a cylinder. The present paper presents similar calculations undertaken in an attempt to explain discrepancies appearing in fission and alpha counting of product material in December, 1946.

III - CALCULATIONS

Radon is the third generation decay product of U-234: U-234 decays into ionium, ionium into radium, radium into radon. In order

S E C R E T

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to calculate the amount of radon to be found in the cascade and the quantity of radiation resulting therefrom, it is first necessary to calculate the quantities of ionium and radium, from which the radon derives. Having this quantity established, the amount of radiation is calculated from the decay of radon and from the activity of short-lived daughters of radon.

During the 20 months of plant operation the U-234 inventory has increased from the

In the absence of intermediate analysis of the U-234 inventory we assume a linear increase:

(1)

where t is the time in months since the start of full scale operations.

For the calculation of ionium and radium formation, the period of 20 months considered is a tiny fraction of the half-lives involved (See Table I).

S E C R E T

TABLE I

	Half life	λ / second	λ / month
U-234	2.4×10^5 yrs.	9.15×10^{-14}	2.37×10^{-7}
Io	8.3×10^4 yrs.	2.65×10^{-13}	6.87×10^{-7}
Ra	1.59×10^3 yrs.	1.38×10^{-11}	3.58×10^{-5}
Rn	3.825 days	2.097×10^{-6}	---

For this reason the second term of the equation (2) for the rate of growth of ionium is small compared with the first term. In the following calculations it is assumed negligibly small.

$$\frac{d(Io)}{dt} = (U-234) \lambda U-234 - (Io) \lambda Io \quad (2)$$

Where (Io) is the number of moles of ionium
(U-234) is the number of moles of U-234
 λ is the instantaneous decay rate
t is time

Substituting the expression (1) for the amount of U-234 present and integrating from time zero to time t gives:

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This is then the calculated weight of radium distributed throughout the cascade.

Predictions of future radium accumulation based on the pres-

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Since the half-life of radon is quite short compared with 20 months and the short-lived active deposit formed from it is nearly at equilibrium in two hours, it is not possible to use the simple approximation employed in (4) and (6) to calculate quantities formed. Instead, it is convenient to find the activity of the radon and active deposit as fractions of the activity when in equilibrium with radium. This is quite simple as the equilibrium activity (alpha disintegrations per minute) is equal to that of the parent radium for each alpha emitter in equilibrium with the radium. The alpha activity (disintegrations per second) from a gram of radium is given as 3.7×10^{10} .

The top cell of the cascade will tend to accumulate only a small fraction of the equilibrium quantity of radon, for two reasons. Much of the radon will be passed on to the purge cascade since radon is only about two thirds as heavy as process gas. Radon from the lower portions of the cascade may have largely disintegrated (forming solid Ra A) before reaching the top cell. Thus, it is unreasonable to expect even as much as half the equilibrium quantity of radon to collect in the top cell even if withdrawals were made only once every two or three weeks.

S E C R E T

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A laboratory sample of a few grams at most could hardly contain more than one percent of this quantity. A single counted film of eight milligrams could in turn be expected to have less than one hundredth percent of the quantity of radon and active deposit in a product cylinder. Since an alpha counter counts but half of the alpha disintegrations in a film, these impurities should contribute less than one-half alpha count per minute.

III - DISCUSSION

The conservative over-estimations made above for the cases of product cylinders and laboratory samples may amount to a factor of a hundred. The negligible alpha rates found even under these conditions make it unnecessary to calculate the fractions of equilibrium radon

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more accurately for the purposes of this paper.

IV - CONCLUSIONS AND RECOMMENDATIONS

The radon and active deposit alpha activity of counting section product films is less than, probably considerably less than, one half count per minute. In comparison with the observed rates (circa 100,000 ~~cpm~~/min.) from product films this is entirely negligible.

V - ACKNOWLEDGMENTS

The request that the effect of radon content on product alpha activity be calculated was made by F. W. Hurd. The encouragement and advice of F. W. Hurd and E. W. Bailey were invaluable in promoting the writing of this paper.

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