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MEMORANDUM

EPSX 105

August 23, 1950

TO: T. F. Furlong
FROM: W. H. Bowman, Leader, D. L. James, and J. D. Roarty
SUBJECT: The Decay of Induced Activity in Portland, Barytes,
and Brookhaven Cements.

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I. INTRODUCTION

The materials used in pile construction must be of such a nature that little response is shown to neutron bombardment and that the induced activity decay rapidly. Maintenance is facilitated since personnel may enter the irradiated regions shortly after pile shut-down.

Three types of cements, Barytes, Brookhaven, and Portland, were exposed in the ORNL Reactor for periods of seven and twenty-eight days, and the induced activity was measured in a 100% -geometry ionization chamber responding only to gamma radiation. The activity was plotted as a function of the length of decay and the half lives of the active materials determined therefrom. Using these data the probable identity of the elements giving rise to this activity was determined from a search of the isotope tables.

The absorption of radiation by lead from these samples was measured with a Geiger-Mueller counter. The absorption coefficient in the expression,

$$\frac{I}{I_0} = e^{-\alpha x}$$

where

- I = radiation transmitted by lead
- I_0 = incident radiation
- α = absorption coefficient
- x = thickness of lead,

was determined by plotting the ratio $\frac{I}{I_0}$ on a logarithmic scale against the lead thickness on a linear scale and measuring the slope of the resulting line.

II. RESULTS

Decay Rates

The decay rates of the irradiated samples are presented in Table I. When possible, the element producing the activity has been determined and its concentration reported. The concentration of the element as determined from the spectrographic analysis is also reported. In most cases no identification could be made because of insufficient information concerning the quantity and intensity of gamma radiation resulting from the decay of the active constituents. The activity curves for these cements are to be found in Figs. 1 to 6.

Absorption Data

The absorption data are shown in Table II. It will be noted that the absorption coefficients vary both with time of irradiation and length of decay. This phenomenon results from the composite nature of the radiation from the sample. The coefficients determined are for an average value of the emitted photon energy. This average value is a function of the amount and type of active material in the sample and, hence, is dependent on the previous history of the cement. The absorption curves are shown in Figs. 7 to 10.

Table I
ANALYSES OF DECAY CURVES OF IRRADIATED CEMENT

Sample	Measured Half-Lives of Radioactive Isotopes	Probable Radioactive Isotope	Calculated Weight Percent of Element in Samples Based on Activity	Spectrographic Value Percent
Brookhaven	47.6 da.	?	?	?
	1.06 da.	14.8 hr Na ²³	0.06	0.04
	7.60 hr.	?	?	?
Portland	37.6	?	?	?
	3.05 da.	?	?	?
Barytes	14.4 da.	?	?	?
	1.68 da.	1.12 da As ⁷⁵	0.12	-
	3.82 hr.	2.60 hr Mn ⁵⁵	00.097	0.08T

A question mark (?) indicates uncertainty in the chemical identification.

T = Trace

Table II

Sample	Period of Irradiation	Length of Decay	Absorption Coefficient
	hr	hr	in. ⁻¹
Brookhaven	168	535	1.64
	168	750	1.68
	672	32	1.50
	672	250	1.73
Portland	168	535	3.58
	168	750	2.98
	672	32	1.41
	672	250	3.36
Barytes	168	535	4.07
	168	750	4.38
	672	32	3.36
	672	250	4.13

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Table III

Spectrographic Analysis *

<u>Element</u>	<u>Brookhaven</u>	<u>Portland</u>	<u>Barytes</u>
Ag	< 0.04	< 0.04	< 0.04
Al	2.5	2.5	0.15
B	< 0.004	0.015	< 0.004
Ba	0.08	2.5	> 10
Be	0.0003	< 0.0003	< 0.0003
Bi	--	--	--
Ca	> 5	> 5	2.5
Cd	< 0.04	< 0.04	< 0.04
Co	< 0.04	< 0.04	< 0.04
Cr	0.04	< 0.04T	< 0.04T
Cs	--	--	--
Cu	0.02	< 0.02T	< 0.02T
Dy	--	--	--
Eu	--	--	--
Fe	10	0.6	0.6
Ga	< 0.04	< 0.04	< 0.04
Hf	< 0.5	< 0.5	< 0.5
Gd	--	--	--
Hg	--	--	--
Ho	--	--	--
In	< 0.15	< 0.15	< 0.15
K	0.15	0.63	0.04
La	--	--	--
Li	< 0.04T	< 0.04T	< 0.04

<u>Element</u>	<u>Brookhaven</u>	<u>Portland</u>	<u>Barytes</u>
Mg	0.63	0.63	0.15
Mn	0.31	< 0.08T	< 0.08T
Mo	< 0.04	< 0.04T	< 0.04
Na	0.04	0.15	< 0.04T
Nd	--	--	--
Ni	0.08	0.08	< 0.08T
P	--	--	--
Pb	< 0.08	< 0.08	< 0.08
Pr	--	--	--
Pt	--	--	--
Rb	< 0.04	< 0.04	< 0.04
Si	5	10	1.3
Sm	--	--	--
Sn	< 0.04T	< 0.04T	< 0.04
Sr	< 0.06	< 0.06	0.27
Te	--	--	--
Th	--	--	--
Ti	0.04	0.15	0.01
V	< 0.04	< 0.04	< 0.04
Zn	< 0.31	< 0.31	< 0.31
Zr	< 0.15	< 0.15	< 0.15
W	--	--	--
Y	--	--	--

Amounts are in percentages.

T = Trace

-- = Not looked for

* Y-12 Spec. Lab Report No. 3376, plate A-811.

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NOTE: SAMPLE IRRADIATED FOR 672 HOURS

MASSACHUSETTS INSTITUTE OF TECHNOLOGY			
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GAMMA AND GAMMA-CORRELATED SYSTEMS			
DECAY CURVE OF BROOKHAVEN CONCRETE			
IRRADIATION IN ORNL PILE			
DATE	BY	FILE NO.	PAGE
9-24-50	J.D.R.	EPD-K105	2

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NOTE: SAMPLE IRRADIATED FOR 168 HOURS

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CARBIDE AND CARBON CHEMICALS DIVISION			
DECAY CURVE OF PORTLAND CONCRETE			
IRRADIATION IN ORNL FILE			
DATE	DRAWN BY	FILE NO.	PAGE
9-24-50	J.D.R.	EPS-X-105	3

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TIME-HOURS FROM END OF IRRADIATION

NOTE: SAMPLE IRRADIATED FOR 672 HOURS

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DEPARTMENT OF CHEMISTRY			
DECAY CURVE OF PORTLAND CONCRETE			
IRRADIATION IN ORNL PILE			
DATE	BY	FILE NO.	PAGE
8-24-50	J. B. R.	EPS-3-102	4

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NOTE: SAMPLE IRRADIATED FOR 168 HOURS

MASSACHUSETTS INSTITUTE OF TECHNOLOGY			
ENGINEERING PRACTICE SCHOOL			
CARBIDE AND CARBON CHEMICALS DIVISION			
DECAY CURVE OF BARYTES CONCRETE			
IRRADIATION IN ORNL PILE			
DATE	BY	FILE NO.	PAGE
8-14-50	J.D.R.	EP-1-N-105	5

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○ AFTER 536 HOURS DECAY TIME, 7 DAYS IN ORNL PILE

A	"	755	"	"	"	7	"	10	"	10
B	"	32	"	"	"	20	"	10	"	10
X	"	250	"	"	"	20	"	10	"	10

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UPON CARBIDE AND CARBON CLAYTON			
ABSORPTION OF GAMMA RADIATION			
FROM BROOKHAVEN DEPT			
DATE	DRAWN BY	FILE NO.	FIG.
8-23-60	D L J	EPSX-105	7

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O	AFTER	536	HOURS	DECAY	TIME,	7	DAYS	IN	ORNL	PILE
A	"	755	"	"	"	7	"	"	"	"
X	"	265	"	"	"	25	"	"	"	"

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UNION CARBIDE AND CARBON CORPORATION			
ABSORPTION OF GAMMA RADIATION			
FROM PORTLAND CEMENT			
DATE	DRAWN BY	FILE NO.	FIG.
8-25-50	D. L. J.	EPS X-105	6

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B AFTER 32 HOURS DECAY TIME, 20 DAYS IN ORNL PILE

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CARBIDE AND CARBON CHEMICALS DIVISION			
UNION CARBIDE AND CARBON CORPORATION			
ABSORPTION OF GAMMA RADIATION			
FROM PORTLAND CEMENT			
DATE	DRAWN BY	FILE NO.	TITLE
8-25-50	D L J	EPSX-105	9

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O AFTER 636 HOURS DECAY TIME, 7 DAYS IN ORNL PILE

A " 766 " " " 7 " " " "

B " 88 " " " 26 " " " "

X " 260 " " " 26 " " " "

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ABSORPTION OF GAMMA RADIATION

FROM BARYTES CEMENT

DATE	DRAWN BY	FILE NO.	FILE
8-23-60	D L J	EPSX-125	10

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III. DISCUSSION OF RESULTS

Activity

It will be observed that the Portland cement has the least activity and that the activities of the Barytes and Brookhaven cements are about 10 times greater. Other properties being equal, the Portland is the most promising for use in irradiated regions. It has an additional advantage in that there are no extremely active short lived materials in it although this is of no consequence if a cooling off period of two or three days is available.

In order to make these results easy to compare with results obtained from samples irradiated with a different neutron flux, the activities are presented for a standard radiation of 10^{12} neutrons/(sq cm)(sec).

Selection of Parent Isotopes

The selection of elements having half lives similar to those measured in this test is somewhat arbitrary. The selection was limited to naturally occurring isotopes producing gamma emitters as a result of neutron bombardment. Preliminary selections were narrowed down to those elements probably present as shown in a spectrographic analysis. This approach resulted in an identification of only three of the eight observed isotopes. The others may be the result of other nuclear reactions or of an (n,γ) reaction for which data are not available.

The sample of Portland cement which was irradiated for 28 days was not amenable to this type of analysis since the period of observation was not sufficient to allow the short lived materials to decay to insignificance as required.

Both samples of the Brookhaven cement were found to have an isotope with a half-life of 27.6 days. The samples also contained 10% iron. The probability of Fe^{59} (half life of 47 days) being present would seem to be high. When the amount of iron required to produce the measured activity was computed, the result was about five times the total sample weight. If Fe^{59} is indeed the active element, this high value could result from using an improper value of the neutron absorption cross section.

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Absorption Data

The absorption data indicate the amount of shielding required to reduce the radiation from the sample to any given value. It can be shown by several methods that the fraction of radiation transmitted is an exponential function of the thickness of the shield. This is true only for radiation composed of photons of a single energy level. The approximation that it holds for all levels is usually quite good for gamma radiation since the photon energies do not vary over a wide range for the materials found at any one time in the cement.

The distinct curvature of these plots near the zero thickness of lead is quite possibly due to the presence of beta rays. These are completely absorbed by a very thin section, and hence, the absorption coefficient is quite large in this range. This hypothesis is supported by the data gained from using aluminum as a shield. Thirty-four percent of the incident radiation is transmitted by 0.048 in. of aluminum, whereas thirty-three percent passes through 0.120 in. This transmitted portion is probably all gamma radiation.

Accuracy

Jones and Overman (2) in their report on the calibration of the 100% geometry ion chamber state that the accuracy of the instrument is about 1%. The scatter of data about a smoothed curve is of the same order. On this basis, using a standard differentiation technique (5), the error in half life determinations is about 10% for the longest half-life. Because the determination of the shorter half-lives involves subtracting two values of about the same size from each other, the errors in these determinations are far greater, probably about 20 or 30%.

Spectrographic Analysis

The samples were analyzed spectrographically for the non-volatile metallic elements. This analysis is shown in table III. Since this method of analysis is not accurate for high concentrations, the lower limit of some elements such as calcium is given. The sum of the weight fractions is much less than 1.00 because of this fact and because of the volatile materials such as CO_2 are not reported.

IV. PROCEDURE

Activity Measurement

After removal from the pile the samples were placed in glass test tubes and stored in a lead cylinder with two inch walls. The radiation from a sample was determined by placing it in a 100% - geometry ionization chamber and measuring the voltage developed. To increase the precision of this measurement an opposing voltage was placed in series with the leads from the chamber. This potential was obtained from a potentiometer which was calibrated from an internal standard cell once each day. The potentiometer drift was found to be negligible. The difference of the output of the chamber and the bucking voltage was then amplified with a vibrating reed electrometer and read on a Speedomax. In this way the output could be determined to one millivolt.

Absorption Measurement

The absorption curves were found by placing the sample in a lead chamber containing a Geiger-Mueller tube and interposing sheets of lead between the cement and the tube. Four different thicknesses of lead were used for each sample. Care was exercised to prevent movement of the sample once it had been placed in the chamber since the reading on the counter was found to depend on the placement of the sample.

Method of Analysis

The measured activity, based on the disintegrations of Co^{60} per second for an irradiation of 10^{12} neutrons/(sq cm)(sec), was plotted on a logarithmic scale against length of decay on a linear scale. After all but one of the isotopes had decayed to insignificance as evidenced by a straight line relationship, the straight line was extrapolated to zero time. The difference between this line and the measured activity was the activity due to the other isotopes in the sample. The process was repeated until the last difference was a straight line. (No more than three isotopes can be determined in this fashion since any errors rapidly accumulate.)

The activity at zero time is found for each of the isotopes and the number of disintegrations per second that produce such an activity is determined from published data on the amount and energy of the radiation from a single disintegration. This information in turn can be used to find the weight of the active material producing the radiation. Once it is known, the weight of the parent isotope can be easily found. (See Sample Calculations.)

V. LOCATION OF ORIGINAL DATA

The original data are located in M. I. T. Practice School Data Book No. 1555, pages 11 to 28 inclusive, and the log sheets of pile operations, July 10 to August 7, 1950, which can be found in the files of the Operations Division, ORNL.

VI. CONCLUSIONS AND RECOMMENDATIONS

Pending additional studies as to the composition of the active materials in the cements, the following conclusions can be drawn:

1. The Portland cement has the least induced activity of the cements tested and the absorption coefficient is nearly as high as that of Barytes. The former is the best suited of the three samples with respect to a low induced activity.
2. If the 47-day isotope found in the Brookhaven cement, which is probably iron, can be economically eliminated, this cement will be preferable to either of the others.
3. The majority of the active materials appear to be present in trace amounts with the possible exception of iron in the Brookhaven cement, and their elimination will probably prove difficult.

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A. NOMENCLATURE

- A activity at time t , disintegrations of $\text{Co}^{60}/\text{sec}$
- A_0 activity at end of irradiation, disintegrations of $\text{Co}^{60}/\text{sec}$
- A'_0 activity at end of irradiation, disintegrations of actual isotope/sec
- E efficiency of ionization
- I transmitted radiation
- I_0 incident radiation
- MW molecular weight
- N_0 Avogadro's number, 6.02×10^{23} molecules per gram mole
- ϕ neutron flux, neutrons/(sq cm)(sec)
- t length of decay, hr or days
- t' length of irradiation, hr or days
- $t_{1/2}$ half life, hr or days
- x thickness of lead shielding, in.
- α absorption coefficient, in.^{-1}
- λ decay constant $\ln 2/t_{1/2}$
- σ neutron absorption cross section, sq cm

B. SAMPLE CALCULATIONS

Activity

Sample	Activity Photons/sec based on Co ⁶⁰	Pile Level kv	% of Maximum Flux at "Rabbit Hole"
Portland 2.166 g.	0.94×10^7	2780	57

In the ORNL pile a power level of 3600 kw is equivalent to a maximum flux of 0.57×10^{12} neutrons/(sec)(sq cm), (2). This flux has been found to vary in a linear manner with the power level. The average neutron flux is then:

$$\frac{2780}{3600} (0.57 \times 10^{12}) = 4.40 \times 10^{11} \text{ neutrons/(sec)(sq cm)}$$

The flux at the "Rabbit Hole" is:

$$(4.40 \times 10^{11}) (0.57) = 2.51 \times 10^{11} \text{ neutrons/(sec)(sq cm)}$$

The activity is a linear function of the flux (2). The activity for a flux of 10^{12} neutrons/(sec)(sq cm) is:

$$\frac{10^{12}}{2.51 \times 10^{11}} (0.94 \times 10^7) = 3.75 \times 10^7 \text{ photons/sec or } 1.74 \times 10^7 \text{ Photons/sec}$$

for one gram of the cement

Half-Life

It can be shown that:

$$t_{1/2} = t \frac{\ln 2}{\ln(A_0/A)}$$

Sample	Length of Decay	Photon ^A / ₀ (sec)(g)	Photon ^B / ₀ (sec)(g)
Portland	267 hr	1.79×10^7	1.03×10^7

Then:

$$t_{1/2} = 267 \frac{\ln 2}{\ln \frac{1.79 \times 10^7}{1.03 \times 10^7}}$$

$$= 334 \text{ hr or } 13.9 \text{ days}$$

Absorption

Sample	Shield Thickness	I/I_0
Portland	0.0375 in.	0.845
	0.1130	0.657

$$I/I_0 = e^{-\mu x}$$

$$\frac{(I/I_0)_1}{(I/I_0)_2} = e^{-\mu(x_1 - x_2)}$$

$$\frac{\ln \frac{(I/I_0)_1}{(I/I_0)_2}}{x_1 - x_2} = -\mu$$

$$\frac{\ln \frac{0.845}{0.657}}{0.1130 - 0.0375} = -\mu$$

$$\mu = 3.36 \text{ in.}^{-1}$$

Isotope Identification

<u>Sample</u>	<u>Assumed Isotope</u>	A_0 <u>Photons/sec</u>	<u>Radiation Intensity mev.</u>	<u>Percent of Disintegrations</u>
Barytes	Mn ⁵⁶	9×10^7	0.84	100
			1.81	25
			2.13	15

The paper of Jones and Overman (3) contains a plot of ionizing efficiencies for various energy gammas as compared to those of Co⁶⁰. A procedure is outlined for determining the number of disintegrations per second from the activity measured in terms of Co⁶⁰.

$$A_0' = \frac{200 A_0}{E(E) (\%)}$$

<u>Radiation Energy mev.</u>	<u>Efficiency</u>	<u>Percent of Disintegrations</u>	<u>(E)(%)</u>
0.84	0.80	100	80
1.81	1.30	25	52.5
2.13	1.40	15	21

$$A_0' = \frac{(200)(9 \times 10^7)}{133.5}$$

$$= 13.6 \times 10^7 \text{ disintegrations per second}$$

Salisbury (4) presents an equation by which the weight of the parent isotope may be determined.

$$W = \frac{(A_0') (MW)}{N_0 (1 - e^{-\lambda t'})}$$

$$= \frac{(15.6 \pm 10^7) (55)}{(10^{12})(12.8 \pm 10^{-24})(6.02 \pm 10^{23})(1 - e^{-0.268 \pm 0.12})}$$

$$= 9.72 \pm 10^{-4} \text{ g.}$$

Since the prevalence of Mn^{55} from which the isotope is formed is 100%, the weight of Mn in the sample is 9.72 ± 10^{-4} g. The spectrographic analysis showed a weight of less than 8.0×10^{-4} g.

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END

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