RADIOISOTOPIC INVESTIGATION
OF OMR COOLANTS, PART I
OMRE OPERATION
JANUARY, 1958 THROUGH AUGUST, 1960

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ABSTRACT

Over 30 radioisotopes were identified in the coolant of the OMRE during the period from January, 1958 through August, 1960. The principal gamma-emitting isotopes measured were the corrosion products: Mn^{56} , Fe^{59} , and Mn^{54} ; the principal beta emitters were: P^{32} , H^3 , S^{35} , Fe^{55} , and C^{14} . The total (gamma plus beta) activity of the coolant during normal reactor operations was estimated to have been less than $1.08 \mu\text{c/gm}$ of coolant, with Mn^{56} contributing approximately 80% of the detected process radiation.

I. INTRODUCTION

A. PURPOSE

The low induced radioactivity postulated for a chemically pure liquid hydrocarbon has provided one of the foremost arguments for its use as a moderator-coolant for power plant reactors. The first experimental data on the performance of a hydrocarbon in a reactor have been obtained through the operation of the Organic Moderated Reactor Experiment (OMRE), located at the National Reactor Testing Station near Idaho Falls, Idaho. The purpose of this report is to present the information obtained on the radioisotopes detected in the coolant of the OMRE during the period from January, 1958 through August, 1960.

Prior to September, 1960, over 30 radioisotopes had been detected in the OMRE coolant. The second section of this report enumerates those radioisotopes, the third suggests their probable sources, and the fourth (and most important section) presents the radioactivity levels of those radioisotopes which were measured in the coolant.

B. PRELIMINARY INFORMATION

The period from January, 1958 through August, 1960 included power operation of the OMRE with Core I (January, 1958 to November, 1958) and Core II (June, 1959 to March, 1960) as fuel. Figure 1 shows the power levels of the reactor during this period; the nature of the OMRE, as an experimental reactor, required that the power level vary according to prescribed testing conditions.¹

Prior to December, 1959, a mixture of polyphenyls, mainly ortho- and meta-terphenyl with small amounts of diphenyl and para-terphenyl, was used as a moderator-coolant in the reactor, while after that date, Santowax OMP was used; both coolants were mixtures of diphenyls and terphenyls purchased from Monsanto Chemical Company. The coolant was circulated essentially as shown in the flow diagram, Figure 2. It passed through the core, where it was irradiated and radiolytic decomposition occurred. Most of the decomposition products are of higher molecular weight and of lower volatility than the undecomposed coolant; these decomposition products are called high boilers, or HB.^{2,3} Carbon steel pipes then carried the greater portion of the coolant to the air blast heat exchanger and back to the reactor core; smaller portions passed either through the particulate removal loop, composed of a centrifuge and various filters, or through the

Figure 1. Power Levels of OMRE from February, 1958 to September, 1960

Figure 2. Coolant Flow Diagram of OMRE

purification system, designed to control the heavy polymer content of the system. In the purification system, the low boiling, unpolymerized polyphenyls distilled over and were returned to the core, but the heavy polymers and other high boilers were collected in the still bottoms and removed as waste. By this means, the high-boiler concentration in the coolant was usually held at 30 wt %.

II. MEASURED RADIOISOTOPES

During the period from January, 1958 to August, 1960, over 30 radioisotopes were detected in the OMRE coolant. Principal among these were the gamma emitters: Mn^{56} , Fe^{59} , Mn^{54} , As^{76} , and Se^{75} ; and the beta-emitting isotopes: P^{32} , H^3 , S^{35} , Fe^{55} , and C^{14} . A more exhaustive enumeration of the isotopes detected, their characteristic radiations, and their half-lives are given in the first three columns of Table I.

TABLE I

SUMMARY TABLE FOR RADIOISOTOPES DETECTED IN OMRE COOLANT

Radioisotopes	Mode of Decay and Radiation*	Half-Life*	Source	Maximum Specific Activity in Core II Coolant ($\mu\text{c/gm}$)
Mn ⁵⁶	β^- , γ	2.6 hours	Corrosion, Recoil	3.8×10^{-1}
P ³²	β^-	14.22 days	Activation of Trace Impurity	2.8×10^{-1}
H ³	β^-	12.2 years	Ternary Fission Product (?) Activation of Organic	1.8×10^{-1}
S ³⁵	β^-	87 days	Activation of Trace Impurity	3.8×10^{-2}
Fe ⁵⁵	EC	2.60 years	Corrosion, Recoil	3.0×10^{-2}
C ¹⁴	β^-	5568 years	Activation of Organic, Activation of Blanket Gas	2.5×10^{-2}
Fe ⁵⁹	β^- , γ	44.3 days	Corrosion, Recoil	1.7×10^{-2}
As ⁷⁶	β^- , γ	26.5 hours	Activation of Trace Impurity	9.9×10^{-3}
Mn ⁵⁴	EC, γ	300 days	Corrosion, Recoil	7.3×10^{-3}
Se ⁷⁵	EC, γ	121 days	Activation of Trace Impurity	7.2×10^{-3}
Na ²⁴	β^- , γ	14.90 hours	Activation of Trace Impurity, Corrosion	1.7×10^{-3}
Co ⁵⁸	EC, β^+ , γ	71.3 days	Recoil	1.6×10^{-3}
Cu ⁶⁴	EC, β^- , β^+ , γ	12.8 hours	Corrosion	1.6×10^{-3}

TABLE I (CONT'D)

SUMMARY TABLE FOR RADIOISOTOPES DETECTED IN OMRE COOLANT

Radioisotopes	Mode of Decay and Radiation*	Half-Life*	Source	Maximum Specific Activity in Core II Coolant ($\mu\text{c/gm}$)
Ba ¹⁴⁰	β^- , γ	12.80 days	Fission	8.5×10^{-4}
Sb ¹²²	β^- , EC, γ	2.75 days		8.1×10^{-4}
I ¹³¹	β^- , γ	8.08 days	Fission	8.1×10^{-4}
Co ⁶⁰	β^- , γ	5.24 years	Corrosion, Recoil	7.8×10^{-4}
Zn ⁶⁵	EC, β^+ , γ	246 days	Corrosion	5.9×10^{-4}
Cs ¹³⁷	β^- , γ	30.0 years	Fission	4.5×10^{-4}
Zr ⁹⁵	β^- , γ	65 days	Fission	3.3×10^{-4}
Sr ⁹⁰	β^-	27.7 years	Fission	7.7×10^{-5}
Cl ³⁸	β^- , γ	37.29 min.	Activation of Trace Impurity	Not measured
I ¹³²	β^- , γ	2.30 hours	Fission	Not measured
Kr ⁸⁸	β^- , γ	2.77 hours	Fission	Not measured
I ¹³⁵	β^- , γ	6.8 hours	Fission	Not measured
Xe ¹³⁵	β^- , γ	9.13 hours	Fission	Not measured
K ⁴²	β^- , γ	12.46 hours		Not measured
I ¹³³	β^- , γ	20.9 hours	Fission	Not measured
La ¹⁴⁰	β^- , γ	40.27 hours	Fission	Not measured
Te ¹³²	β^- , γ	77.7 hours	Fission	Not measured
Cr ⁵¹	EC, γ	27.8 days	Corrosion	Not measured

* Reference: Revs. Modern Phys. 30, 2 Pt. 2 (1958)

III. SOURCES OF RADIOISOTOPES

The majority of the radioisotopes found in the OMRE originated from one or more of the following sources:

- a) Activation of the hydrocarbon
- b) Activation of trace impurities in the vendor-supplied coolant
- c) Corrosion of structural materials in contact with the coolant
- d) Recoil of activated nuclei into the coolant
- e) Fission

The probable source, or sources, for the isotopes listed in Table I are given in Column 4 of that table. In general, the activation products listed are the results of (n, γ) , (n, p) , or (n, α) reactions.

A. ACTIVATION OF HYDROCARBON

Although the pure coolant was composed of approximately 6% hydrogen and 94% carbon, by weight, the pure coolant is not the prime source of activity, because of the low neutron cross section of these elements. Nevertheless, a portion of the C^{14} and the H^3 activity can be considered to originate from activation of the hydrocarbon.

B. ACTIVATION OF TRACE IMPURITIES

In the production, shipping, and transferring of the hydrocarbon, certain impurities were introduced into the coolant. For example, sulfur was present as a result of the method of production of the coolant; and manganese, the precursor of the major radioisotope (Mn^{56}) in the OMRE, has been found in very high concentrations in the dregs (rust, moisture, etc.) in the coolant drums. Impurity concentrations for the major precursors in the coolant, as received from the vendor, are listed in Column 2 of Table II.

The impurity content of the vendor coolant was found to be greatly reduced by distillation; therefore, during Core II operations, the coolant was distilled before being added to the reactor. However, certain impurities (particularly selenium, sulfur, and chlorine) were not effectively removed by this process;

TABLE II
IMPURITY CONTENT OF SAMPLES OF VENDOR COOLANT
AND CIRCULATING COOLANT OF OMRE

Impurity	Impurity Content of Vendor Coolant, Mixed Polyphenyls (ppm)	Impurity Content of Circulating Coolant Removed August, 1959 (ppm)
Aluminum	0.27	1.0
Arsenic	<0.3	0.14
Antimony	-	2
Barium	<0.5	-
Chlorine	1.5	1.5
Cobalt	0.005	0.01
Chromium	0.05	0.65
Copper	0.53	0.2
Iron	4	35
Manganese	0.05	0.08
Nickel	0.18	-
Phosphorus	<0.3	0.5
Selenium	0.05	0.06
Silver	<0.005	0.2
Sodium	0.11	0.2
Sulfur	64	54
Zinc	<0.05	0.6

such impurities have been termed "trace impurities". The radioisotopes produced by activation of coolant impurities were P^{32} , S^{35} , Na^{24} , Se^{75} , and Cl^{38} .

C. CORROSION OF STRUCTURAL MATERIALS

The primary source of the isotopes Fe^{55} , Fe^{59} , Co^{60} , Mn^{56} , and Mn^{54} was corrosion of structural materials in contact with the coolant; the mechanisms included both activation of corrosion products and corrosion of activated isotopes from the core region.

The elements expected to have been introduced by corrosion of the carbon steel, stainless steel, and aluminum were iron, manganese, carbon, chromium, nickel, cobalt, and probably some zinc, copper, and aluminum. Column 3 of

Table II indicates the major elements present in the circulating coolant of the OMRE.

D. RECOIL OF ACTIVATED NUCLEI

Recoil of activated nuclei into the coolant from the stainless steel cladding of the fuel elements represented a further source of the isotopes Mn^{54} , Mn^{56} , Co^{58} , and Co^{60} . However, it was impossible to determine the percentage of these isotopes that were produced by recoil.

E. FISSION

There have been negligible fission products present in the OMRE during normal reactor operations; the major portion of the observed fission products could be attributed to trace amounts of uranium which were present in the materials of construction. However, a cladding failure of an experimental test element (October, 1958) introduced fission products, some of which remained in the Core II coolant.

F. OTHERS

The activation of the nitrogen (used to pressurize the OMRE) and its trace impurities represent another source of activity in the coolant. C^{14} is one of the radioisotopes resulting from this source.

Tritium has been shown to be a ternary fission product in both plutonium and uranium fuels.⁴ The tritium can get into the coolant through the cladding, by either diffusion or penetration of high energy tritium particles.

IV. ACTIVITY LEVELS OF RADIOISOTOPES

A. FACTORS INFLUENCING ACTIVITY LEVELS

In the preceding section, the radioisotopes detected in the coolant were enumerated. These isotopes were not necessarily present in the coolant at the same time, nor always at the listed activity levels. The specific activity ($\mu\text{c/gm}$ of coolant) of a radioisotope at any particular time was a function of the operation of the purification still and the centrifuge, the quantity of fresh coolant added to the reactor, the half-life of the isotope, the power level of the reactor, and, for certain isotopes, the concentration of the precursor. The effect of these parameters will be seen upon examination of the activity data obtained from the coolant samples.

B. ANALYSES OF CORE I COOLANT

Only a limited number of quantitative radioactive analyses were performed on coolant samples during the period from January, 1958 (reactor startup) to August, 1958. However, Mn^{56} , Fe^{59} , and Cl^{38} had been identified in the coolant as early as February, 1958. By June of that year, the following isotopes had been positively identified: Cl^{38} , Mn^{56} , K^{42} , Na^{24} , As^{76} , Sb^{122} , Ba^{140} , P^{32} , Cr^{51} , Fe^{59} , Co^{58} , S^{35} , Mn^{54} , and Co^{60} . At that time, Mn^{56} was the primary contribution to the radiation detected in the process area of the reactor. The early occurrence of radioisotopes in the coolant was due to the high impurity content of the circulating coolant.

The specific activities of the primary radioisotopes present in the coolant, after a total of 765 Mwd of reactor operation, are given in Column 2 of Table III. Note that Mn^{56} remained the primary contribution.

In the period from January, 1958 to October, 1958, fission products were not detected in the coolant in significant quantities. However, the cladding failure of October released gross amounts of these radioisotopes. Among those detected were I^{131} , I^{132} , I^{133} , I^{135} , Ba^{140} - La^{140} , Te^{132} , Kr^{88} , and Xe^{135} . Column 3 of Table III presents the isotopes measured in a sample 21 days after the fission product release. At the time of removal of this sample, most of the fission products had decayed to almost undetectable levels, although Cs^{137} and Sr^{90} remained.

TABLE III
SPECIFIC ACTIVITIES OF RADIOISOTOPES PRESENT IN TWO
SAMPLES OF OMRE CORE I PRIMARY-LOOP COOLANT*

Isotope	Specific Activity ($\mu\text{c/gm}$) (Sept. 15, 1958; 765 Mwd)*	Specific Activity ($\mu\text{c/gm}$) (Nov. 13, 1958; 926 Mwd)*
Mn ⁵⁶	6.8×10^{-1}	Not measured
P ³²	4.1×10^{-1}	5.4×10^{-1}
As ⁷⁶	2.0×10^{-2}	Not measured
Fe ⁵⁹	1.8×10^{-2}	1.1×10^{-1}
S ³⁵	1.4×10^{-2}	8.1×10^{-2}
Mn ⁵⁴	4.1×10^{-3}	3.7×10^{-3}
Co ⁶⁰	7.7×10^{-4}	5.9×10^{-4}
Co ⁵⁸	4.5×10^{-4}	$<1.9 \times 10^{-4}$
C ¹⁴	Not measured	2.0×10^{-2}
Cs ¹³⁷	Not detected	3.3×10^{-3}
Sr ⁹⁰	Not detected	2.2×10^{-3}
Se ⁷⁵	Not measured	5.4×10^{-4}

* The samples are identified by their removal date from the reactor and the accumulated exposure at the time of removal.

On November 24, 1958, the reactor was shut down for fuel replacement; and remained at zero power level until June, 1959, when Core II operations began.

C. ANALYSES OF CORE II COOLANT

The activity levels at the beginning of the irradiation of Core II were low, in comparison with those at the end of the irradiation of Core I. This change in activity resulted from distillation of the entire coolant supply, as well as decay during the period from November to June. During the irradiation of Core II, continual analyses were made, not only on primary-loop coolant samples, but also on distillate and high-boiler samples.

1. Comparison of Primary-Loop, High-Boiler, and Distillate Samples

Comparison of the samples from a set of primary-loop, high-boiler, and distillate samples yielded information on the relative effectiveness of the purification still in removing various radioisotopes from the coolant. Table IV shows

TABLE IV
SPECIFIC ACTIVITIES OF RADIOISOTOPES
PRESENT IN A SET OF OMRE CORE II COOLANT SAMPLES*

Isotope	Specific Activity ($\mu\text{c/gm}$) (Primary Loop)	Specific Activity ($\mu\text{c/gm}$) (High Boiler)	Specific Activity ($\mu\text{c/gm}$) (Distillate)	Purification Still Efficiency†
P ³²	2.6×10^{-1}	9.0×10^{-1}	1.2×10^{-2}	0.97
S ³⁵	3.6×10^{-2}	5.9×10^{-2}	1.3×10^{-2}	0.73
C ¹⁴	2.0×10^{-2}	3.8×10^{-2}	9.9×10^{-3}	-
As ⁷⁶	8.8×10^{-3}	2.0×10^{-2}	5.4×10^{-4}	0.96
Fe ⁵⁹	3.4×10^{-3}	1.2×10^{-2}	$<8.1 \times 10^{-6}$	0.98
Na ²⁴	1.7×10^{-3}	2.5×10^{-3}	$<6.8 \times 10^{-5}$	0.97
Mn ⁵⁴	1.4×10^{-3}	5.0×10^{-3}	$<4.3 \times 10^{-5}$	0.98
Cu ⁶⁴	9.9×10^{-4}	2.7×10^{-3}	$<4.0 \times 10^{-5}$	0.97
Co ⁵⁸	9.0×10^{-4}	3.4×10^{-3}	$<2.3 \times 10^{-5}$	0.98
Co ⁶⁰	4.4×10^{-4}	1.2×10^{-3}	$<1.4 \times 10^{-5}$	0.98
Se ⁷⁵	2.4×10^{-4}	5.2×10^{-4}	9.0×10^{-5}	0.73
Sr ⁹⁰	3.3×10^{-5}	1.6×10^{-4}	$<1.8 \times 10^{-6}$	-

* The set was removed from the OMRE in August, 1959, after an accumulated exposure of 309 Mwd. Mn⁵⁶ was not measured in these samples.

† Still efficiency is fraction removed in HB during distillation.

the results of the analysis of one such set which indicated, as others, that Fe⁵⁹, Co⁵⁸, Co⁶⁰, and Mn⁵⁴ are among those radioisotopes most responsive to operation of the purification still. These isotopes are thought to be present in particulate form in the coolant.

2. Comparison of Primary-Loop Samples

The combined effect of distillation, centrifugation, and variation of power level is evident from comparison of the activity levels of several primary-loop samples. The graphs of the specific activities of P³², C¹⁴, and Mn⁵⁴ in Figure 3 exemplify the effect on the individual radioisotopes.

Mn⁵⁴ (300 day half-life) exhibited a radical variation of specific activity, due to its immediate response to distillation and centrifugation, but only moderate decay during shutdown periods. Conversely, P³² possesses a short half-life

Figure 3. Variation of Specific Activities of P^{32} , C^{14} , and Mn^{54} in OMRE Core II Coolant

(14.2 days) and was moderately affected by distillation; so that its specific activity essentially followed the power level of the reactor. In contrast, the activity of C^{14} was relatively unaffected by reactor shutdown, because of its long half-life (5600 years). The almost insignificant change due to distillation and filtering operations presupposes attachment of C^{14} to the hydrocarbon itself.

Although the Mn^{54} specific activity of the coolant varied radically and attempts to predict it were unsuccessful, it was found that the Fe^{59} to Mn^{54} ratio in the coolant could be predicted quite accurately and quickly, since the prime precursor of both isotopes is iron, and they are not separated by any of the physical removal mechanisms employed at the OMRE. The theoretical ratio can be calculated from the ratio of Fe^{58} (precursor of Fe^{59}) and Fe^{54} (precursor of Mn^{54}), the ratio of thermal neutron flux to fission neutron flux, and the ratio of the half-lives. Part A of Figure 4 shows the agreement of the theoretical and measured ratios, as a function of the time of irradiation of the coolant during Core II operations. Part B of this figure compares the measured ratio to the theoretical ratio for the zero power level, which began after approximately 180 days of irradiation.

3. Maximum Specific Activities in Coolant

The maximum specific activity values for P^{32} , C^{14} , Mn^{54} , and Fe^{59} , as well as for other isotopes measured in the Core II primary loop coolant, are given in Column 5 of Table I. These figures indicate that the total maximum specific activity was approximately $0.98 \mu\text{c/gm}$ of coolant, with Mn^{56} contributing about 40% of this quantity.

4. Process Area Radiation

In order to estimate the percentage of the process radiation due to Mn^{56} , the pure beta emitters were eliminated from the total indicated by Column 5 of Table I. Computation then showed that Mn^{56} should have accounted for about 85% of the radiation detected in the process area. This result is consistent with the radiation data obtained at the OMRE.

Under normal reactor operating conditions during Core II, the maximum gamma radiation in the process area, as measured 4 in. from a 16-in. primary coolant pipeline, was slightly less than 150 mr/hr, while the reactor was operating at 6 Mw. Observations of the radiation field, immediately following scrams and other shutdowns, indicated that isotopes having half-lives of about 2-1/2 hr

Figure 4. Comparison of Theoretical and Experimental Fe^{59} to Mn Ratios for OMRE Core II Coolant

(i.e., Mn^{56}) accounted for 75 to 80% of the gamma field. Twelve to fifteen percent of the radiation was due to very short-lived isotopes, i.e., isotopes with half-lives less than 30 sec (and consequently not measured or listed in Column 5). The remaining 5 to 10% was due to isotopes with relatively long half-lives (12 hr or greater).

V. SUMMARY AND CONCLUSIONS

Assuming that the total maximum specific activity ($0.98 \mu\text{c/gm}$) represented 90% of the coolant activity, while the other 10% was due to short-lived isotopes, the total specific activity in Core II coolant has been estimated to have been less than $1.08 \mu\text{c/gm}$ of coolant.

Of the isotopes measured, Mn^{56} was the principal isotope. It contributed 70 to 85% of the process radiation, although it accounted for only about 40% of the total (beta plus gamma) radiation measured in the coolant. Fe^{59} and Mn^{54} are also considered as major gamma emitting isotopes; H^3 , C^{14} , S^{35} , and P^{32} are regarded as the major beta emitting isotopes.

The major gamma emitters arise as a result of activation of impurities which have corroded from the process piping, and from corrosion and recoil of the isotopes produced in the core of the reactor. Neutron activation of the hydrocarbon produced a portion of the H^3 and C^{14} . However, C^{14} was also produced by activation of nitrogen dissolved in the coolant. S^{35} and P^{32} were probably produced by activation of impurities which were present in the hydrocarbon, as purchased from the vendor.

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