APAE NO. 50

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SM-1
RESEARCH AND DEVELOPMENT PROGRAM

FINAL REPORT ON FISSION PRODUCT ACTIVITY IN THE SM-1 PRIMARY COOLANT
TASK XIII

ALCO

ALCO PRODUCTS, INC.
NUCLEAR POWER ENGINEERING DEPARTMENT
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FINAL REPORT ON FISSION PRODUCT ACTIVITY IN THE SM-1 PRIMARY COOLANT
TASK XIII

Contract No. AT(30-3)-326

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ABSTRACT

Fission product measurements were made on the SM-1 primary coolant. The airborne activity observed during sampling of the primary system was identified. An analysis was made on the primary coolant for alpha activity and on the secondary water for fission product iodine.

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1.0 INTRODUCTION

1.1 General

The Army Package Power Reactor (SM-1) is operated by Alco Products, Incorporated under Contract No. AT(30-3)-326 with the United States Atomic Energy Commission. As part of this contract, a research and development program is being carried out. The results of experimental work performed under Task XIII of this program are described in this report. Task XIII is an extension of Task III, which concluded with the issuance of APAE No. 44.(1)

Between the completion of work on Task III and the beginning of work on Task XIII, the SM-1 pressure vessel was opened. A stationary fuel element and a control rod element were replaced at that time.

"Examination by periscope of the removed elements at Fort Belvoir showed that a crack may exist in the cladding of the stationary fuel element. This would support the conclusion in the Task III final report (APAE No. 44) that a cladding defect was present in the core."

1.2 Scope of Work

The work to be performed under the Task XIII program consisted of the following operations:

(1) Continuing on a routine basis the measurement started on Task III of several of the high yield fission products in the primary system.

(2) Monitoring the primary water sampling operation and the stack for airborne fission product activity and evaluating the environmental hazard it presented.

(3) Performing periodic measurements on the secondary water for fission products.

(4) Investigating the presence of alpha activity in the primary water.

(5) Continuing an analytical program.

(6) Developing detailed procedures, to be used in the

SM-1 and applicable to other pressurized water reactors of the Army Nuclear Power Program for the routine measurement and analysis of primary and secondary water for fission products.

1.3 Studies

To carry out the program outlined, the following studies were planned:

(1) The behavior of the steady state level of the high yield fission products in the primary system (as determined by the measurements of (1) above), and a startup and shutdown run for I^{131} and I^{133}, were to be integrated into an analytical program. This would be a development of the program initiated by Task III.

(2) An analysis for I^{133} was to be performed on the secondary water once each month to detect the presence of fission products in the secondary system.

(3) An analysis for uranium isotopes on the water and crud was to be performed to detect the presence of alpha activity in the primary system.

(4) A nuclide identification followed by measurements of nuclide levels was to be performed to evaluate the hazard from airborne activity.

The work outlined above was not completed. This was the result of extensive low power operation, an unusual number of scrams during startup, and the short time at full power available, in which to accomplish this task. In particular, sufficient data were not obtained to continue development of the analytical program initiated in Task III.

2.0 CONCLUSIONS AND RECOMMENDATIONS

Conclusions

The following conclusions are derived from the work described in this report:

(1) The rate of fission product addition to the primary system has decreased slightly since December 1958.

(2) Although the rate of addition has decreased, the steady state level of several fission products has increased since December. This is possibly explained by a difference in fission product deposition rates since December 1958, caused by a higher pH of the primary water.

(3) Uranium was not found in the primary system water.

(4) Fission products were not found in the secondary system.

(5) The airborne activity consists of fission product gases and their daughters. Major daughter activities identified were Cs^{138}, Rb^{88}, and Rb^{89}. No immediate hazard exists under existing sampling procedures.

(6) Cesium-137 is present in the primary makeup tank. Indications of other long-lived activities were also found.

Recommendations

The following recommendations are made as a result of the information obtained from this task:

(1) Continue the routine analysis for Cs^{137}, Cs^{138}, I^{131}, I^{133} and Sr^{91} in the primary water. Iodine-131 should be run weekly, and the others on a routine monthly basis.

(2) Continue the iodine-133 and iodine-131 analysis on the secondary water on a routine monthly basis.

(3) Perform startup and shutdown tests for I^{131} and I^{133} to fully characterize the behavior of these isotopes as a possible routine test for defective fuel elements.

(4) Continue efforts to detect any alpha activity in the primary system, particularly in the crud.

(5) Continue efforts to establish any health hazard presented by airborne activity. An effort should be made to measure the gaseous component as well as the particulate.

(6) An analysis of activity found in the primary make-up tank should be carried out to determine the efficiency of the demineralizers for specific nuclides. In particular, the studies should determine whether the nuclides contributing the most to dose rate buildup are being completely removed by the demineralizers.

(7) Studies should be initiated concerning the installation of instrumentation to monitor fission product activity in the primary coolant.

(8) A thorough study of core 2 for surface and cladding contamination should be made. This should be followed by a determination of fission product levels obtained in the primary system after the core has been inserted. This will allow a correlation to be made between a known surface contamination and fission product levels to be expected. For cores subsequent to core 2 at SM-1, and for cores in future Army pressurized water reactors, the amount of fission products to expect would be known before startup. If the expected fission products level in the core is greatly exceeded, a leak may be suspected.

3.0 PRESENTATION AND DISCUSSION OF DATA

The data presented in the following sections are those considered to be most representative of the conditions measured. All data collected from the primary system are included in the Appendix, Section 5.0.

3.1 Fission Product Level in the Primary Coolant

The measurement of fission product levels in the primary coolant was carried out as described in APAE No. 44.(1) Briefly, it consisted of drawing a sample from the primary system, performing the appropriate chemical separation and, with the exception of strontium -90, measuring the amount of the nuclide present by counting with a scintillation spectrometer. The strontium-90 was measured with a G-M tube and scaler. The primary water was not filtered.

The fission product levels observed are listed in Table 3.1. The levels observed during October 1958 and December 1958 are included for comparison.

TABLE 3.1 FISSION PRODUCT LEVELS IN THE PRIMARY COOLANT

Isotope	dpm/ml (Oct. 1958)	dpm/ml (Dec. 1958)	dpm/ml (June 1959)
Sr^{90}	15	27	37.5
Sr^{91}	3.7×10^4	$4.2 \times 10^{4*}$	$5.8 \times 10^{4*}$
I^{131}	$1.1 \times 10^{4*}$	2.1×10^4	1.8×10^4
I^{133}		$1.3 \times 10^{5*}$	$1.0 \times 10^{5*}$
Cs^{137}	33	93	74
Cs^{138}		$2.2 \times 10^{5*}$	$1.7 \times 10^{5*}$
Ba^{139}		1.5×10^5	$2.4 \times 10^{5*}$
Ba^{140}	$3.2 \times 10^{3*}$	4.5×10^3	7.1×10^3

* May be considered steady state levels.

3.1.1 Discussion of Fission Product Levels

The levels recorded for Sr^{91}, I^{133}, Cs^{138}, and Ba^{139} are equilibrium levels. An interesting phenomenon is apparent.

The cesium and iodine levels show a small decrease since December 1958. The overall accuracy in determining the concentration of a long-lived isotope is about 20 percent. This is limited by the accuracy with which the purification rate may be set. In contrast to the small decrease observed for iodine and cesium, the strontium and barium levels show a definite increase since December 1958. The purification rate during this power run has been within 20 percent of that maintained during December. The behavior of these fission products requires an explanation.

It is known from earlier studies$^{(1)}$ that cesium does not deposit on the primary system surfaces to any appreciable extent. Barium and strontium are found in the circulating crud and can be assumed to deposit.$^{(1)}$ The pH of the primary coolant during the May-June power run had been higher than the normal operating pH. The pH was 8.5 during the initial startup on May 17 and slowly decreased to the normal pH of about 7.5 by June 15. Most of the analyses were run previous to June 15. The abnormal pH in the primary system could have altered the deposition rate of strontium and barium. If this rate were decreased, an increase in their levels in the coolant could be observed even though a small decrease occurred in their rates of addition to the primary system.

No other explanation for this phenomenon is apparent. The buildup of fissionable plutonium-239 does not explain it, since this would increase the iodine and cesium level as well.

A decrease in the iodine and cesium levels is apparent from the data. There is an uncertainty of ± 20 percent in the purification rate. For this reason, a decrease in the rate of iodine-133 addition is uncertain.

Since cesium-138 level is unaffected by the purification system, as a result of its short half life, the only uncertainty in its level is the uncertainty in its chemical analysis of ± 10 percent. Thus, a definite decrease in the rate of addition is observed in the case of cesium-138. One stationary fuel element and one control rod element were replaced in the core between the December and the May-June power run. It appeared there was a cladding defect in the stationary element. This could offer an explanation for a decrease in the rate of fission product addition to the primary system.

It is possible that some surface contamination has been released from the core and deposited outside the core area or removed by the purification system. This could account for part of the decrease in the rate of fission product addition to the primary system.

It is recommended that studies be intitated concerning the installation of instrumentation to monitor fission products present in the primary system. Such instrumentation would reduce the amount of routine laboratory work necessary to safely monitor reactor operation.

3.1.2 Startup and Shutdown Tests

It had been planned to conduct a startup and shutdown run for iodine-131 and iodine-133. However, this was not possible. The reactor could not be maintained at a steady power level from initial startup to a steady state level of the iodine isotopes. Because no shutdown was scheduled for the duration of this task, a shutdown test could not be conducted.

Startup and shutdown tests are of value in evaluating the source of fission products. It has been observed,(2) for PWR type fuel elements, that iodine-131 continues to enter the primary coolant after shutdown if a leak exists in the core. This has been observed at the SM-1.(1) Also, if a leak exists, the time for iodine-131 to reach a steady state level in the coolant is greater than that predicted for surface contamination. Thus, a startup and shutdown test could be a method of determining whether a leak exists.

Another phenomenon observed, for a fuel element with a known leak and no meat to cladding bond, is the peaking of fission products after startup.(2) That is, the fission product level will increase sharply after startup to a certain level, then decrease to a steady state level. The explanation offered is that coolant leaks into the fuel element during shutdown and is forced out during reactor operation. As the coolant is forced out, it contains a high concentration of fission products. This peaking is also observed for fission product delayed neutrons present in the primary coolant. Since a cladding to meat bond does exist in the SM-1 type fuel element, it is doubtful if this phenomenon would be observed in SM-1 if a leak existed.

3.2 Fission Products in the Secondary System

A four liter sample of secondary water was analyzed for iodine-133 and iodine-131 while the reactor was at full power. No iodine activity could be found. It may be concluded from this that no fission products exist in the secondary system at present.

3.3 Alpha Activity in the Primary System

A chemical separation for uranium was performed on 4 liters of primary water. The final sample was analyzed for alpha activity. The results were negative. Time available did not permit a similar analysis on primary system crud.

3.4 Airborne Particulate Activity

3.4.1 Observation of Airborne Activity

The levels given in Table 3.2 are those observed in the immediate vicinity of an open sample point upstream of the demineralizers. The open sample point refers to a steady flow of primary water into a drain pipe open to the atmosphere. The water was flowing into a drain through a length of rubber tubing inserted about one inch into the drain pipe. The particulate activity was collected on a Hollingsworth filter using a portable air monitor with a 15-liter per minute flow through the filter. The level of each nuclide was measured by counting a characteristic photopeak using a scintillation spectrometer. A collection efficiency of 90 percent was assumed. The levels in Table 3.2 should be considered approximate, since they are based on a single measurement.

TABLE 3.2 AIRBORNE PARTICULATE ACTIVITY

Isotope	Level (μc/cc)	MPC (μc/cc)*
Mn^{56}	Not detectable	3×10^{-7}
Rb^{88}	5×10^{-8}	
Rb^{89}	9×10^{-9}	
Sr^{90}	$\ll Cs^{137}$**	1×10^{-9}
I^{131}	Inconclusive	9×10^{-9}
I^{133}	Inconclusive	
Cs^{137} (calculated from Rb^{89})	1×10^{-14}	6×10^{-7}
Cs^{138}	1×10^{-8}	
Ba^{139}	$< Cs^{138}$**	

* Code of Federal Regulations, January 1, 1958.

** Concluded by comparing the half lives of the respective gaseous precursors.

Chemical separations were attempted on the particulate activity without success. The short half lives of the material collected, combined with the small amount it was possible to collect, made chemical separation impractical. Nuclide identification was made by following the decay of characteristic photopeaks of the material collected.

3.4.2 Discussion of Airborne Activity

No induced activity was found in the airborne material. From this, the conclusion is reached that no particulate activity is being carried out of the primary water by escaping gases. The particulate component of the airborne activity consists entirely of the decay products of the escaping radioactive gases. The presence of iodine has not been confirmed. Only one attempt of the four made to detect it gave positive results and there is some doubt concerning its reliability. An activated charcoal filter was used to collect any iodine activity. The possibility exists that the filter used in the test giving positive results was contaminated before it was used. All subsequent filters were checked for contamination immediately before use.

The levels of activity given in Table 3.2 are those observed in the immediate vicinity of the open sample point defined previously. For a given volume of gas escaping from the sample point, the total particulate content will increase with time as the gas decays. However, since this original volume is also expanding and thus diluted with increasing time, two opposing effects occur as the escaping activity moves away from the sample point. No conclusion was reached as to which effect is greatest. It is concluded, however, that no hazard will exist under the normal sampling procedures now used at SM-1. Cs^{137}, Sr^{90} and Ba^{139} were not detected experimentally. Their presence was calculated from the half lives of their gaseous precursors(1) and an assumed transit time of 40 seconds from core to sample point. The gaseous precursors have sufficiently long half lives to exist in the primary coolant at the upstream sample points and will, therefore, escape in the same manner as the other gases. The level quoted for Cs^{137} was calculated on the basis of the level of Rb^{89}. Their gaseous precursors have approximately the same half lives.

Since it has been determined that no particulate activity is being carried out of the primary system, the level of the radioactive gases, rather than the particulate component, will determine the health hazard present. An effort should be made to measure this level.

3.5 Activity in the Primary Makeup Tank

A determination of the Cs^{137} level in the makeup tank was performed. The level was found to be 10.6 dpm/ml. The cesium enters the makeup tank as its gaseous precursor Xe^{137}, which has a half life sufficiently long to pass through the demineralizers.

While performing this analysis, an activity with a 0.85 mev gamma ray was found. This gamma ray is characteristic of Mn^{54}, Mn^{56}, and Co^{58}, significant contributors to the primary system activity. It is felt that a complete analysis of activity present in the primary makeup tank should be performed. This will determine the efficiency of the demineralizers for the various nuclides.

4.0 FISSION PRODUCT PROCEDURES

The routine measurement of the fission product level may be accomplished by the determination of the iodine and cesium levels. The only source of iodine and cesium in the primary system is the fission process. Complete procedures for routine radio-iodine and radio-cesium determinations in the SM-1 are given in Sections 4.1 and 4.2.

4.1 Fission Product Iodine

If a gamma-ray spectrometer is available, an analysis for iodine-133 and iodine-131 may be made. If a spectrometer is not available, an analysis for iodine-131 only is possible. In either case, a waiting period of several days after drawing the sample is necessary before counting.

<u>Reagents</u>

Carbon tetrachloride

Sodium hypochlorite - 5% solution

Hydroxylamine hydrochloride-10% solution

Sodium carbonate- 2$\underline{M}$ solution

Concentrated nitric acid

6$\underline{N}$ nitric acid

Sodium bisulfite ($NaHSO_3$)-1$\underline{M}$ solution

Standard iodide solution-6.53 gm KI per 500ml water (10 mg I^- per ml)

One-tenth percent silver nitrate solution

Absolute ethanol

Equipment

Two 250 ml separatory funnels

Two 60 ml separatory funnels

Two 50 ml Ehrlenmeyer flasks

Eight droppers (2 ml capacity)

One 2 ml volumetric pipette

Filter chimney apparatus

Whatman No. 42 filter circles (to fit chimney apparatus)

Drying oven (100 C)

Sample mounts (planchette plus retaining ring) and Mylar film

Hot plate

Hood

Counting apparatus - G-M tube plus scaler or gamma-ray spectrometer

Procedure

Caution: All steps through step (6) should be performed in the hood.

Duplicate samples should be run.

(1) Place 100 ml of primary water in a 250 ml separatory funnel.

Add by pipette 2 ml of the standard iodide solution.

Add 10 ml of saturated sodium carbonate solution.

(2) Add with a dropper 2 ml of sodium hypochlorite solution and mix. Stopper the funnel and wait at least ten minutes.

(3) Slowly add 5 ml of concentrated nitric acid, allowing the acid to slide down the side of the funnel. Care should be taken not to agitate the solution which would facilitate the release of CO_2. This would reduce the iodine yield by carrying any free iodine formed out of the funnel, and it would

create a health hazard in the laboratory. Add 4 ml of hydroxylamine solution in the same manner. Then add 10 ml of carbon tetrachloride and stopper the funnel.

(4) Being careful to hold the stopper firmly in place, shake the funnel thoroughly. A pressure will build up inside the funnel due to the release of CO_2. After shaking for about one minute, release the pressure by holding the funnel upside down and carefully opening the drain stopcock. After releasing the pressure, agitate for an additional 30 seconds and release the pressure again. Allow the funnel to stand until the water and carbon tetrachloride layer (lower layer) have separated completely.

(5) Drain the lower layer (carbon tetrachloride) into a 60 ml separatory funnel. Add 10 ml of fresh CCl_4 to the 250 ml separatory funnel and repeat step (4). Add the lower layer to that in the 60 ml separatory funnel.

(6) To the carbon tetrachloride in the 60 ml funnel, add 10 ml of H_2O. Then add 4 drops of the sodium bisulfite solution and agitate until the mixture is colorless. Allow the layers to separate. Discard the lower layer (CCl_4) and drain the upper layer into a 50 ml Ehrlenmyer flask or if the sample is to be counted with a G-M tube, add 10 ml Na_2CO_3 and repeat steps (2), (3), (4), (5) and (6) using a clean 250 ml separatory funnel.

(7) Add 1 ml of 6<u>N</u> HNO_3 and boil on a hot plate for 2 minutes. Remove the flask from the hot plate and allow to cool for about 5 minutes. Place a previously washed (water and then ethanol), dried and weighed filter circle in the chimney apparatus. To the solution in the 50 ml flask, add dropwise with swirling 20 drops of the silver nitrate solution. Boil for one minute and then filter through the chimney apparatus using a vacuum. Wash the precipitate with 5-10 ml of water and then 5-10 ml of ethanol.

(8) Place the filter circle in the drying oven and dry for 10 minutes. Remove and weigh.

(9) Place the filter circle on a planchette, cover with Mylar film and secure with the retaining ring. Count according to instructions in (10)A or (10)B below.

Counting

(10) A. Spectrometer

I^{133}: Wait 30 hours after the sample is drawn. Count the 0.53 mev gamma. Operation of the spectrometer is described in the handbook supplied with the instrument and in AP Note 165.

I^{131}: Wait at least 5 days after the sample is drawn. Count the 0.365 mev gamma.

B. G-M Tube (See Note 1 at end of this section)

I^{131}: Count the sample long enough to collect at least 10,000 counts.

Calculations

(11) A. Spectrometer

I^{133}:

$$\text{dpm/ml} = \frac{\text{cpm X C.F.}}{\frac{\text{sample weight in mg}}{37.0} \text{ X } 100 \text{ X D.F. X } 0.93}$$

cpm = Observed counts per minute — background counts per minute.

D.F. = Decay Factor = $e^{-\lambda t}$

$\lambda = 3.33 \times 10^{-2}\ \text{hr}^{-1}$

t = number of hours elapsed from the time the sample was drawn to the time the sample was counted.

C.F. = Correction Factor

The correction factor is obtained from a plot of C.F. versus gamma energy. The C.F. is defined as $\frac{\gamma\text{pm}}{\text{cpm}}$. Here, γpm is the gammas per minute at the particular energy emitted by the sample and cpm is the counts per minute observed at the peak at a given window width (see spectrometer handbook for definition of window width). It is necessary that the calibration standards and samples be mounted in essentially the same manner, since the C.F. includes the geometry correction.

I^{131}:

$$\text{dpm/ml} = \frac{\text{cpm x C.F.}}{\frac{\text{sample weight in mg}}{37.0} \text{ x } 100 \text{ x D.F. x } .80}$$

$$\text{D.F.} = e^{-\lambda t}$$

$$\lambda = 3.59 \times 10^{-3} \text{ hr}^{-1}$$

(11) B. G-M Tube

I^{131}:

$$\text{dpm/ml} = \frac{\text{cpm x C.F.}}{\frac{\text{sample weight in mg}}{37.0} \text{ x } 100 \text{ x D.F. x S.A.S.}}$$

C.F. = Correction Factor

Since all the I^{131} beta particles are being counted, a net correction factor must be used. A plot of particle energy vs. C.F. is determined using standard samples. Correction factor is defined as $\frac{\text{dpm}}{\text{cpm}}$, where dpm is the beta disintegrations per minute in the sample, and cpm the observed counts per minute. Since I^{131} has four beta particles, the net correction factor is the weighted sum of the individual beta particle correction factors. Iodine-131 decays with 0.7 percent 0.815 mev beta, 87.2 percent 0.608 mev beta, 9.3 percent 0.335 mev beta, and 2.8 percent 0.250 mev beta.

$$\text{C.F.} = .007 \text{ (C.F. for 0.815 mev)} + .872 \text{ (C.F. for 0.608 mev)}$$
$$+ .093 \text{ (C.F. for 0.335 mev)} + .028 \text{ (C.F. for .250 mev)}$$

S.A.S. = Self-absorption and self-scatter correction for beta particles. This correction is a function of beta energy and sample thickness $\left(\frac{\text{sample weight mg}}{\text{area sample cm}^2}\right)$.

The correction may be estimated by using curves given by Nervik and Stevenson.(3) The net S.A.S. is determined in the same manner as the correction factor.

NOTES

1. If a G-M tube is used to count iodine-131, it is necessary to wait 5 days after the sample is drawn before the iodine separation is performed. The sample may be counted immediately after separation.

2. When performing an analysis for iodine on the secondary system, draw duplicate 4-liter samples. Add the iodine carrier and 1 ml of 1 M $NaHSO_3$ solution. Evaporate the 4-liter samples to 100 ml and then proceed as for a primary system sample. Remember to delete the addition of the iodide carrier in step (1) since this has already been done.

4.2 Fission Product Cesium

The separation of cesium must be carried out quite rapidly if a determination of cesium-138 is to be performed. Cesium-138 has a half life of 32 minutes. For this reason the sample must be counted within 2 hours after drawing the sample. The procedure below is designed for Cs^{138} although it may be used for Cs^{137} as well. (See Note 1 at end of this section). The separation should take no longer than 1.5 hours.

Reagents

Cs carrier - 10 mg Cs/ml (see procedure for standardization of carrier)

Fe carrier - 10 mg Fe/ml (as $Fe(NO_3)_3$ in H_2O to which 2 or 3 drops of concentrated HNO_3 per 100 ml of solution has been added)

Hydrochloric acid - concentrated and 6M

Nitric acid - concentrated

Perchloric acid - 70 percent

Sodium hydroxide - pellets

Ammonium hydroxide - 6M

Silicotungstic acid - solid

Absolute ethanol

Equipment

Drying oven (110 C)

Centrifuge

Centrifuge tubes - 40 ml conical, glass

Ehrlenmeyer flasks - 50 ml, 125 ml

Ice bath

Planchettes and retaining rings

No. 42 Whatman filter circles

Filter chimney apparatus

Sintered glass Gooch crucibles for standardization of carrier

Pipettes - 5 ml, 25 ml

Droppers - 2 ml capacity

Preparation of Carrier

Dissolve 12.7 grams of CsCl in water and dilute to 1 liter in a volumetric flask. Pipette 5 ml of the solution into a 125 ml Ehrlenmeyer flask and add 1 ml of conc. HNO_3 and 5 ml of $HClO_4$. Boil until dense white fumes appear. Cool to room temperature and add 15 ml of absolute ethanol. Cool for 15 minutes in an ice bath. Filter on a weighed sintered glass Gooch crucible (fine porosity) and wash three times with 5 ml portions of absolute ethanol. Dry at 110 C for 15 minutes, cool, and weigh as $CsClO_4$. Repeat the standardization until results agree within one percent. The weight of $CsClO_4$ should be 88.0 mg if the CsCl solution was made accurately.

Procedure

Caution: Always work in the hood when using $HClO_4$. Do not allow it to come in contact with any organic material.

(1) Before obtaining the primary water sample, pipette 5 ml of

the cesium carrier into a 125 ml Ehrlenmeyer flask. Now draw the primary water sample and as rapidly as possible, pipette 25 ml of the sample into the 125 ml flask and bring to boiling. Boil for about 2 minutes and then transfer to a 40 ml centrifuge tube.

(2) Add 0.5 ml of Fe carrier and add 6 M NH_4OH dropwise until the $Fe(OH)_3$ precipitates. Centrifuge and transfer the supernate to a clean 40 ml centrifuge tube.

(3) Repeat step (2).

(4) To the supernate add 10 ml of conc. HCl. Add 2 gm of silicotungstic acid dissolved in 2 or 3 ml of water. Stir and allow to stand for 5 minutes. Centrifuge and discard the supernate. Wash the precipitate twice with 10 ml portions of 6 M HCl, discarding the washes.

(5) To the precipitate add 2-5 ml of water and 3 pellets of NaOH. Heat cautiously over a bunsen flame until the precipitate dissolves. Add more water on NaOH pellets as needed to complete the dissolution. Pour into 20 ml of hot 6 M HCl contained in a 125 ml Ehrlenmeyer flask and boil. Add 0.5 ml conc. HNO_3 and continue to boil. Add 10 ml of $HClO_4$ and boil until dense white fumes appear. Then cool and add 10 ml of water and heat almost to boiling. Centrifuge in a 40 ml centrifuge tube.

(6) Pour the supernate into a 50 ml Ehrlenmeyer flask, add 1 ml of conc. HNO_3 and boil until dense white fumes appear. Cool and transfer to a 40 ml centrifuge tube with 30 ml of absolute ethanol. Cool in an ice bath for 15 minutes and centrifuge. Pour the supernate into the sink into running water. Wash the precipitate twice with 10 ml portions of absolute ethanol.

(7) Collect the precipitate on a filter circle, previously washed with ethanol, dried, and weighed, using a filter chimney apparatus. Use absolute ethanol to make the transfer. Dry at 110 C for 15 minutes, cool and weigh. Mount on a planchette, cover with Mylar film and secure with a retaining ring. Count according to one of the methods below.

Counting

(8) A. Spectrometer

Cs^{138}: Count the 1.43 mev gamma ray immediately after separation

Cs^{137}: Count the 0.662 mev gamma ray

(8) B. G-M Tube

Cs^{137}: Collect at least 10,000 counts

Cs^{138}: Since the beta particle energies and percentages are not known, it is not possible to determine Cs^{138} with a G-M tube.

Calculations

(9) A. Spectrometer

Cs^{138}:

$$\text{dpm/ml} = \frac{\text{cpm X C.F.}}{\text{Yield x D.F. x } 0.73 \text{ x } 25}$$

$$\text{Yield} = \frac{\text{sample weight in mg}}{\text{weight of CsClO}_4 \text{ per 5 ml of carrier}}$$

D.F. = $e^{-\lambda t}$

λ = $2.15 \times 10^{-2}\ m^{-1}$

t = number of minutes elapsed from time sample was drawn to time sample was counted.

C.F. = See iodine procedure.

Cs^{137}:

$$\text{dpm/ml} = \frac{\text{cpm X C.F.}}{\text{Yield x } 0.92 \text{ x } 4\text{x}10^3}$$

(9) B. G-M Tube

Cs^{137}:

$$\text{dpm/ml} = \frac{\text{cpm X C.F.}}{\text{Yield x } 4\text{x}10^3}$$

C.F. = Determined the same way as for I^{131}

Cs^{137} decays 92% by 0.514 mev beta and 8% by 1.17 mev beta.

NOTES

1. If cesium-137 is being determined, a 4-liter sample is taken. Five ml of the carrier are added and the sample is boiled down to less than 25 ml and transferred to a 40 ml centrifuge tube. The separation is started with step (2).

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APPENDIX

5.0 TABULATION OF FISSION PRODUCT DATA

The results of all fission product analyses on the primary coolant are tabulated in Table 5.1.

TABLE 5.1 TABULATION OF FISSION PRODUCT DATA

Isotope	Sample No.	Date & Time Sampled	dpm/ml	
Sr^{90}	Sr-102A	June 9, 1959 (1315)	38.2	
	Sr-102B	June 9, 1959 (1315)	35.9	
Sr^{91}	Sr-101A	June 9, 1959 (1315)	---	
	Sr-101B	June 9, 1959 (1315)	5.77 x 10	
			I^{131}	I^{133}
I^{131}, I^{133}	I-102E	May 19, 1959 (0810)	2.41×10^3	--
	I-102F		2.81×10^3	--
	I-111A	May 22, 1959 (1526)	9.71×10^3	--
	I-112A	May 23, 1959 (1415)	9.66×10^3	--
	I-112B	May 23, 1959 (1415)	1.02×10^4	--
	I-114A	June 2, 1959 (1910)	1.5×10^3	1.05×10^4
	I-115A	June 4, 1959 (1600)	9.23×10^3	6.35×10^4
	I-115B	June 4, 1959 (1600)	8.10×10^3	6.35×10^4
	I-117B	June 5, 1959 (2000)	8.95×10^3	6.12×10^4
	I-119A	June 8, 1959 (1400)	1.25×10^4	8.40×10^4
	I-119B	June 8, 1959 (1400)	1.34×10^4	9.42×10^4
	I-120A	June 11, 1959 (1445)	1.58×10^4	1.01×10^5
	I-120B	June 11, 1959 (1445)	1.61×10^4	1.04×10^5
	I-121A	June 12, 1959 (2100)	1.75×10^4	1.07×10^5
Cs^{137}	Cs-106A	June 8, 1959 (1400)	72.3	
	Cs-106B	June 8, 1959 (1400)	75.2	
	Cs-108	June 19, 1959 (1500)		
		Makeup Tank	10.6	
Cs^{138}	Cs-104A	June 2, 1959 (1427)	1.67×10^5	
	Cs-104B	June 2, 1959 (1427)	1.83×10^5	
	Cs-107A	June 8, 1959 (2015)	1.20×10^5	
	Cs-107B	June 8, 1959 (2015)	1.28×10^5	

TABLE 5.1 TABULATION OF FISSION PRODUCT DATA (CONT'D)

Isotope	Sample No.	Date & Time Sampled	dpm/ml	
			Ba^{139}	Ba^{140}
Ba^{139}, Ba^{140}	Ba-101A*	June 10, 1959 (1525)	2.17×10^5	6.07×10^3
	Ba-101B	June 10, 1959 (1525)	2.43×10^5	7.08×10^3

* Some sample material lost after yield determination

6.0 REACTOR OPERATING LOG DURING STARTUP 17 MAY TO 24 MAY, 1959

17 May, 1959

1908 - reactor critical, primary system heat up

18 May

0317 - scram
0905 - raising rods
1002 - scram
1028 - raising rods
1108 - scram
1755 - bringing reactor critical

19 May

0001 - heating up
0959 - scram
1029 - raising rods
1432 - manual scram
2055 - reactor critical

20 May

0001 - critical from previous day
0142 - scram
0242 - critical
1224 - 500 kw load
1526 - 1000 kw load
1626 - scram

21 May

0001 - down from previous day
1748 - critical

22 May

0001 - critical from previous day
0938 - 350 kw load
1325 - 2000 kw load
1448 - station load
1521 - scram
1930 - critical
2305 - 500 kw load
2330 - 2000 kw load

23 May

1710 - scram
2032 - critical
2318 - 2000 kw load

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