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SODIUM TECHNOLOGY AT EBR-II

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SODIUM TECHNOLOGY AT EBR-II

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ABSTRACT

Since the installation of purity monitoring systems in 1967, the control of the purity of the primary and secondary sodium and cover gas systems at the Experimental Breeder Reactor II (EBR-II) has been excellent.

A rigorous monitoring program is being used to assure that operating limits for more than 25 chemical and radioactive impurities are not exceeded. The program involves the use of sophisticated sampling and analysis techniques and on-line monitors for both sodium and cover gas systems.

Sodium purity control is accomplished by essentially continuous cold trapping of a small side stream of the total circulating sodium. The cold traps have been found to be very effective for the removal of the major chemical impurities (oxygen and hydrogen) and tritium but are almost ineffective for ^{131}I and ^{137}Cs that enter the sodium from fuel cladding breaks. Purging with pure argon maintains the cover gas purity.

INTRODUCTION

The ability to maintain low levels of impurities in the EBR-II sodium and cover gas systems has been a major positive factor in the almost 12 years of safe and reliable plant operation. In 1975 the plant capacity factor was over 65%. EBR-II, initially a demonstration fast breeder reactor power plant with integral fuel reprocessing and refabrication, is currently being used as an experimental irradiation test facility for fuels, materials, and nuclear instrumentation. The important design and current operating parameters for the EBR-II's primary and secondary sodium systems are given in Table I.

For the first three years of power operation (1964-1967) sodium purity was monitored with plugging temperature indicators and infrequent sodium samples. Cover gas purity was monitored by grab samples and on-line gas chromatographs. After discovery of high copper concentrations in the primary sodium in early 1967⁽¹⁾, a rigorous routine of sodium and cover gas

Table I. EBR-II Design and Operating Parameters

	Primary System	Secondary System
Power	62.5 MWe	19.5 MWe*
Volume of Sodium	341,000 ft^3 (90,000 gal)	37,900 ft^3 (10,000 gal)
Pumping Rate	34,100 lpm (9,000 gpm)	22,000 lpm (5,800 gpm)
Hot Leg Temperature	480°C (900°F)	470°C (880°F)
Cold Leg Temperature	370°C (700°F)	290°C (520°F)
Cold Trap Flow Rate	95 lpm (25 gpm)	45 lpm (12 gpm)

* Electricity is produced in a conventional steam power plant.

purity monitoring was implemented.^(2,3) Sodium purification by cold trapping has been changed from the original as-needed basis to the currently used, nearly continuous, cold-trap operation. Sodium sampling capabilities have been improved from the early flow-through tube samplers to the current use of overflow cup-type samplers and specimen equilibrium techniques.⁽⁴⁾ The use of in-sodium meters for continuous hydrogen and oxygen monitoring was initiated in 1972.^(5,6) Other than the copper impurity increase prior to 1967, EBR-II has operated with only minor upsets in sodium and cover gas purity.

The sodium and cover gas monitoring activities at EBR-II have proved useful in detecting and diagnosing abnormal conditions. For example, measurements of ^{131}I and ^{137}Cs in primary sodium are routinely used as an aid in diagnosing fuel cladding ruptures; increases of Sn in primary sodium have indicated losses of the tin-bismuth alloy from the seal trough of the rotating plug; increases of hydrogen and oxygen in sodium during cold-trap outages have been used to estimate source rates and to identify sources of these impurities. The presence of helium in the primary cover gas has signaled leakage from experimental subassemblies. Erratic increases of methane in the primary cover gas have been correlated with hydrocarbon contamination from the fuel handling activities.

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SODIUM AND COVER GAS PURITY MONITORING

Monitoring Program

The basic purpose of our purity monitoring program is to assure uninterrupted plant availability, and long component life. Sodium and cover gas purity is assured by holding to specified impurity limits through the use of a vigorous monitoring program. The program requires periodic sampling and analysis and in some cases, continuous monitoring of sodium and cover gas for the radioactive and nonradioactive impurities. In the primary sodium 13 nonradioactive species and 12 radioisotopes are analyzed monthly. In the secondary sodium only one radioisotope (^{24}Na) and three nonradioactive species are monitored monthly using flow-through sampling techniques. In the near future, additional nonradioactive species and ^3H will be monitored in secondary sodium using a new overflow cup sampler and specimen equilibration device. The primary cover gas is monitored continuously for He, H_2 , O_2 , and N_2 by a gas chromatograph. The secondary chromatograph monitors H_2 , O_2 , and N_2 .⁽⁶⁾

Sodium Samplers

Two techniques are used for taking samples of EBR-II sodium for impurity analysis. These are termed the "flow-through" and "overflow" techniques.⁽²⁾

In flow-through sampling, a length of tubing or other vessel is installed directly in-line in the sampling system. Sodium flow is initiated through the sampler and continued for a period of time to clean up or "condition" the sample tube or vessel. The sample is then isolated and frozen. The sample vessel is removed from the system and transported to the laboratory for analysis.

The principal disadvantage of analysis on flow-through samples is that either the total sample is not analyzed, or the sample must include a segment of tubing. The first case is subject to errors caused by impurity segregation. The second case is subject to errors inherent in a tubing "blank". Flow-through samples have been used mostly for oxygen, hydrogen, and carbon analysis by mercury amalgamation, amalgum reflux, and oxyacidic flux methods respectively.⁽⁴⁾ With the development of equilibration methods for the analyses of these impurities, the flow-through technique now receives little use.

The preferred sampling method for most other impurities is now the overflow technique. In overflow sampling, one or more cups of 3 to 60 g capacity are placed on a rack in a vessel inserted with argon. Sodium flows from spigots into the sample cups until they overflow. Sodium level in the vessel is maintained below the sample cups by adjusting the argon cover gas pressure. Following the desired flushing period, the sampler is isolated and the samples are allowed to freeze. Samples are removed from the vessel and transported to the laboratory for analysis. The total sample contained in the cup is used in the analysis to avoid potential errors due to impurity segregation.

Equilibration Devices

Since 1972 the EBR-II primary sodium system has been equipped with a device for equilibrating O, H and C impurities with appropriate metal specimens.⁽⁸⁾ The equilibration device exposes specimens (wires or foils) to a flowing stream of 750°C sodium. An equilibration device has recently been put into service in the secondary sodium system. Combined exposures of vanadium (for oxygen), low-carbon 18-Cr, 8-Ni steel (for carbon), and nickel-foil wrapped scandium (for hydrogen) are performed at 750°C for 24 hours. These specimens are equilibrated with the impurity content in the sodium; after cooling, they are removed, cleaned in alcohol and analyzed by standard chemical methods. The impurity content of the sodium is related to the impurity content in the specimens from known chemical activity distribution relationships.⁽⁴⁾ The advantages of the equilibration method are that (1) it measures chemical activity rather than total impurity content, (2) the exposed specimens are not contaminated by exposure to air, (3) the specimens are specific for the impurity of interest, (4) the method is not subject to errors caused by impurity segregation, and (5) the analytical sensitivity of the method is enhanced because the concentration of the impurity in the equilibrated specimen is as much as 1000 times greater than in the sodium.

Cover Gas Samplers

Carbonaceous gases (hydrocarbons) in both the primary and secondary cover gas are periodically sampled in a small diameter copper tube about 100 ml in volume. Analysis is by a laboratory chromatograph that uses a flame ionization-type detector.⁽⁴⁾ Sensitivities of <1 ppm by volume are achieved.

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Xenon fission product gases (^{133}Xe and ^{135}Xe) and ^{41}Ar , an activation product of the cover gas, are measured six times daily by direct gamma spectrometry on samples taken from the primary cover gas in 10 ml glass bulbs. These Xe analyses provide a method of monitoring for fuel cladding breaks. The normal background, due to tramp fissionable material in the sodium, is about $1.2 \times 10^3 \text{ nCi/l}$ and $2.3 \times 10^3 \text{ nCi/l}$ for ^{133}Xe and ^{135}Xe respectively and about 3.2 nCi/l for ^{41}Ar .

Tritium is measured periodically in both primary and secondary cover gases using 2l glass sample flasks. The tritium is counted by scintillation techniques after being converted to water. (4)

On-line Monitors

The primary sodium (since 1972) and secondary sodium (since 1975) have been equipped with on-line oxygen and hydrogen meters which are used to continuously monitor those systems. (6)

The oxygen meters are electrochemical cells with thoria-yttria ceramic electrolytes and air reference electrodes. (9) Calibration is provided monthly by analysis of the oxygen in sodium by the equilibration of vanadium specimens.

The hydrogen meter uses a 0.25-mm thick nickel membrane to allow diffusion of hydrogen into an ion-pumped vacuum system. (7) The hydrogen equilibrium pressure is periodically measured using a vacuum gauge. The equilibrium pressure is related to the hydrogen content in sodium via Sievert's Law. An alternative (and continuous) method for measuring hydrogen is to measure the ion pump current which can be related to hydrogen-in-sodium by calibration via the equilibrium pressure method.

The continuous signals from the oxygen and hydrogen meters are monitored and are used to detect trends so that corrective action can be taken prior to reaching the limits for the reactor operating impurity concentrations.

The primary cover gas is monitored continuously for He, H_2 , O_2 , and N_2 by an on-line gas chromatograph. The secondary chromatograph monitors all but helium since there is no potential source of helium to the secondary cover gas. (6)

SODIUM AND COVER GAS PURITY CONTROL

Operating Limits

The operation of EBR-II requires the maintenance of sodium and cover gas purity to below set operating limits for the various impurities. Table II gives the normal impurity concentrations and the plant operating limits. Exceeding the limits uncontrollably for plugging temperature, oxygen, or hydrogen requires plant shutdown until the source is identified and minimized and the purity is returned to less than the limiting values.

Exceeding the limits for other impurities in sodium or cover gas requires an investigation into the cause of the high concentration, its impact on continued reactor operation and the minimization of the source of contamination. These limits do not automatically call for a reactor shutdown.

Sodium Purity Control

Impurities considered important and routinely analyzed in EBR-II sodium and cover gas are given in Table II. Many of these (e.g., O, H, Bi, Mn, ^3H) are controlled by cold trapping of a small side stream from the primary and secondary sodium inventories. Other impurities, evidently not controlled by cold trapping, are simply monitored for unexpected changes in concentration. Observed changes are investigated to identify the cause and to eliminate the source, if possible.

Cold Trap Description

EBR-II cold traps are essentially the same design as those developed in U. S. reactor programs in the early 1950's. The crystallizer tank is a chamber fitted with stainless steel mesh. The chamber is divided into two nearly equal volumes by a cylindrical flow divider. The in-flowing sodium is cooled as it passes downward through the outer, mesh-filled annulus. The sodium returns upward through the center, mesh-filled section. The primary cold trap crystallizer tank has about a 1100 liter volume and is about 0.95 meters in diameter. The crystallizer for the secondary trap has a 550 liter volume and is about 0.7 meters in diameter. Heat is regenerated between inlet and outlet streams by a toroidal-shaped, tube-and-shell economizer located immediately above the crystallizer tank.

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Table II. Impurity Concentrations for EBR-II Sodium and Cover Gas Systems

<u>SODIUM SYSTEMS (By weight)</u>					
Parameter [†]	PRIMARY SYSTEMS			SECONDARY SYSTEMS	
	1975 Normal Value	Limit	1975 Normal Value	Limit	
Plugging Temperature	<115°C	150°C	<115°C	160°C	
Oxygen	1.1 ppm	2.0 ppm	0.8 ppm	3.5 ppm	
Hydrogen	100.0 ppb	200.0 ppb	120.0 ppb	300.0 ppb	
Carbon	0.2 ppm	0.7 ppm	0.3 ppm (1976)	0.7 ppm	
Boron	<0.05 ppm	10.0 ppm		**	
Bismuth	2.9 ppm	5.0 ppm		**	
Chromium	<0.02 ppm	10.0 ppm		**	
Iron	0.07 ppm	10.0 ppm		**	
Lithium	<0.0005 ppm	10.0 ppm		**	
Manganese	<0.005 ppm	10.0 ppm		**	
Nickel	<0.04 ppm	10.0 ppm		**	
Silicon	0.2 ppm	10.0 ppm	0.5 ppm	10.0 ppm	
Tin	37.0 ppm	50.0 ppm		**	
Uranium	<2.0 ppb	10.0 ppb		**	
Tritium	85.0 nCi/g	300.0 nCi/g		**	
²² Na	90.0 nCi/g	200.0 nCi/g		**	
²⁴ Na	2.6 mCi/g	5.0 mCi/g	20.0 nCi/g	**	
⁵⁴ Mn	0.2 nCi/g	1.0 nCi/g		**	
^{110m} Ag	2.0 nCi/g	5.0 nCi/g		**	
^{113m} In	22.0 nCi/g	50.0 nCi/g		**	
^{117m} Sn	11.0 nCi/g	50.0 nCi/g		**	
¹²⁵ Sb	3.6 nCi/g	10.0 nCi/g		**	
¹³¹ I	60.0 pCi/g	2 X normal ⁺		**	
¹³⁷ Cs	64.0 nCi/g	2 X normal ⁺		**	
²¹⁰ Po	160.0 pCi/g	250.0 pCi/g		**	
²³⁹ Pu	<0.3 pCi/g	10.0 ppb		**	
<u>COVER GAS SYSTEMS (By volume)</u>					
Nitrogen	3000.0 to 6000.0 ppm	7000.0 ppm	100.0 to 500.0 ppm	*	
Oxygen	0 ppm	1400.0 ppm	0 ppm	*	
Hydrogen	0-200 ppm	280.0 ppm	<10.0 ppm	100.0 ppm	
Helium	<10.0 ppm	280.0 ppm	--	--	
Carbon Compounds					
Methane	1-10 ppm	*	1-5 ppm	*	

* No limit established at this time.

** Not sampled and analyzed since 1972 when the overflow cup sampler became inoperable due to a sodium leak.

+ ¹³¹I and ¹³⁷Cs may increase dramatically following a fuel cladding breach.

† Other impurities analyzed routinely are: Ag, Al, Ca, Co, Cu, K, Mg, Mo, Pb, N, F, Cr

The primary sodium cold trap is cooled by a circulating-NaK to silicon-oil, to water, heat transfer system. The secondary sodium cold trap is cooled by a static-NaK to Dowtherm, to water, heat-transfer system.

The thermal performance of both EBR-II cold traps is less than design specification. As a result, both traps must be operated at one-quarter to one-half their

design flowrates. The design deficiency appears to be in the heat transfer that can be achieved in the economizer.

Cold Trap Performance History

The initial cleanup of primary sodium was accomplished with a temporary cold trap similar in design to the permanent trap. The first permanent trap was used

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from March 1963 to June 1968. It processed approximately 87 million liters (23 million gallons) of sodium. From 1963 through 1966 it was operated approximately 15% of the time. From 1966 until it was removed in 1968, it operated 75% of the time. A more rigorous operation of the cold traps followed the discovery of copper in primary sodium in early 1967.

The second primary trap began operating in November, 1968, and still remains in service. At the end of January, 1976, it had processed 216 million liters (57 million gallons) of sodium and has been operated 66% of the time.

The first cold trap used for the secondary sodium plugged during initial system cleanup. The temporary cold trap used to purify primary sodium was installed in the secondary system to complete the initial clean-up.

The second permanent secondary cold trap operated from 1963 to April, 1972. It is estimated that this trap was on-line less than 50% of the time.

The third, and current, secondary cold trap began operation in June, 1972. At the end of January, 1976, it had processed 72 million liters (19 million gallons) and operated 86% of the time.

Oxygen and Hydrogen Purification

Oxygen and hydrogen purity control is of major concern to plant operations because of potential detrimental effect of the oxygen on materials and because both form low solubility sodium compounds that can cause blockages in coolant channels. EBR-II cold traps are effective in controlling oxygen to less than 1 ppm in the primary and secondary sodium. Hydrogen is controlled to less than 100 ppb and less than 140 ppb in the primary and secondary sodium respectively.

The primary and secondary cold traps have been purposely taken out of service on occasion to measure the oxygen and hydrogen source rates. The results are given on Table III.

The source rates for hydrogen and oxygen to the primary sodium essentially correspond stoichiometrically to water. The maximum source rates are observed only during periods of fuel insertion and removal from the primary sodium. At other times these source rates are negligible. It is believed that water is being introduced from residual moisture on the fuel or the fuel handling equipment that is water-washed and dry-

Table III. Oxygen and Hydrogen Source Rates to EBR-II Sodium Systems

		Source Rates	Time Plant Can Operate Without Na Purification	Probable Nature of Source
PRIMARY SODIUM	Oxygen	0-10 g/day	5 to 6 weeks with normal fuel handling, longer without fuel handling	Source is moisture from intermittent fuel insertion & removal operations
	Hydrogen	0-1 g/day		
SECONDARY SODIUM	Hydrogen	0-0.5 g/day	10 days if reactor is operating at full power, longer if reactor is shut down	Hydrogen is produced from steam-power, longer side corrosion that diffuses into the sodium

argon dried as part of normal operating procedures. Reactor operating limits for oxygen, hydrogen and plugging temperature will be reached in 5 to 6 weeks if the primary cold trap is inoperable during fuel handling activities.

During periods when the reactor is operated at full power, hydrogen is produced in the steam generators by water-side corrosion at a fairly rapid rate. The hydrogen diffuses through the tube walls into the sodium at a rate of about 0.5 g/day. This source rate causes the hydrogen concentration to change from its normal 80 ppb when the plant is shut down to 120-130 ppb when the reactor is at power and the cold trap is operating. Without the cold trap operating, the concentration would continue to rise and reach its operating limit in about ten days. The plugging temperature limit would be reached at the same time. There appears to be no significant source of oxygen to the secondary sodium.

Although EBR-II has not experienced water-to-sodium leaks, such leaks are a potential source of both oxygen and hydrogen contamination. The normal hydrogen source rate would be equal to a water leak rate of about 20 ug H₂O/sec. A system of ten hydrogen meters that are capable of alarming water leak rates of <2 mg H₂O/sec (a rate that is well below those that cause rapid damage) is described in a companion paper. (10)

The normal source rate of hydrogen is too small to interfere with the water-to-sodium leak detection capabilities.

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Impurity Removal by Cold Trapping

The effectiveness of EBR-II cold traps for removing various impurities has been measured using data during periods when the source rate of the impurity is essentially zero or after the cold trap has been put into operation.

The general mathematical equation that describes the bulk concentration of any species is:

$$\frac{dc}{dt} = \frac{I}{M} - \left(\frac{FE}{M} (C - C_e) + \lambda C \right) \quad (1)$$

Production Rate Removal Rate

I is the total source rate by in-leakage, diffusion, fission, neutron capture, decay or other mechanism. M is the total mass of sodium (assumed to be well-mixed), F is the crystallizer flow rate, E is the fraction of the impurity removed per pass through the crystallizer, C is concentration of impurity, and λ is the decay constant for the isotope of interest. The solution of equation (1), normalized to $C_o - C_e$ (initial concentration minus the solubility concentration at the cold trap temperature), is:

$$\frac{C - C_e}{C_o - C_e} = \frac{C_o - C_e}{C_o - C_e} e^{-\left(\frac{FE}{M} + \lambda\right)t} + \frac{I}{FE(C_o - C_e)} \left(1 - e^{-\frac{FE}{M}t}\right) \quad (2)$$

Evaluating equation (2) for hydrogen, oxygen, ^{131}I , ^{137}Cs , ^{54}Mn and ^3H led to the following conclusions:

1. Hydrogen is trapped effectively and rapidly. The fraction removed per pass is about 3/4 for the primary cold trap and about 1/2 for the secondary trap.

2. Oxygen is trapped rapidly but the fraction removed per pass appears to be less than 1/2. Cleanup from an oxygen excursion takes longer than from a hydrogen excursion.

3. ^{131}I is not removed effectively by the primary cold trap. The fraction removed per pass is less than 1/20. The major removal mechanism for ^{131}I is radioactive decay (8.0 day half-life).

4. ^{137}Cs shows anomalous behavior. During the first year of its operation (1968-1969) the present cold trap removed about 30% of the ^{137}Cs from the primary sodium. Subsequently, ^{137}Cs was not effectively removed following the stepwise increase caused by certain fuel cladding failures. A single measurement of the $^{137}\text{Cs}/^{22}\text{Na}$ concentration factor (made in June

1972) showed that the ratio of the $^{137}\text{Cs}/^{22}\text{Na}$ activities in the cold trap were a factor of 50 greater than the same ratio in the 371°C bulk sodium. This is approximately the enhancement of activity that would be caused by the initial (30%) removal of ^{137}Cs .

5. ^{54}Mn appears to be effectively removed by the primary cold trap. About 3/5 is removed per pass. The normal bulk sodium concentration is maintained at <1 nCi/g.

6. The primary cold trap controls the tritium concentration in the primary sodium to less than 100 nCi/g. The effectiveness of the primary cold trap varies from about 1/10 to over 1/2 fractional removal per pass.

7. Although quantitative data for fractional removal estimates is not available the following general conclusions have been reached:

a. Impurities that appear to be effectively controlled by cold trapping include:

Bismuth	< 2 ppm
Copper	< 20 ppb
Manganese	< 5 ppb

b. Impurities that do not appear to be well controlled by cold trapping include all other impurities listed on Table II and not previously discussed.

Cover Gas Purity Control

The EBR-II primary and secondary cover gas is argon. Its purity is maintained within the limits given on Table II by replacing argon lost due to leakage and occasionally purposely adding and venting fresh argon. Within the next year a cryogenic gas-purification system will be installed for the removal of fission product gases (mainly Xe and Kr). The new system will permit continuous reactor operation with defected fuel cladding.

The sources and behavior of various impurities in the EBR-II cover gas systems are as follows:

1. Oxygen is introduced to the cover gas by air which enters during fuel handling and certain maintenance operations. It is normally not detected by the gas chromatograph since it rapidly reacts with the sodium.

2. Nitrogen is also introduced by air. A residual of 1000-3000 ppm is normal in the primary cover gas but can exceed 6000 ppm during times when fuel

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handling activities are intensified. A residual of 100-300 ppm nitrogen in the secondary cover gas is normal.

3. Hydrogen is introduced from moisture in air or the fuel handling operation. Hydrogen in the primary cover gas can reach 200 ppm during fuel handling but it reacts with the sodium and is normally <10 ppm. Moisture introduction is the likely cause of the hydrogen excursions in the primary cover gas. Hydrogen is normally less than 4 ppm in the secondary cover gas. Higher levels in the secondary cover gas might indicate the presence of a water-to-sodium leak. No water-to-sodium leaks have occurred.

4. Methane in the primary and secondary cover gas is normally less than 5 ppm and is traceable to impurities in the argon supply. At times, concentrations up to 100 ppm have been observed in the primary cover gas. These appear to be correlated with fuel handling operations. The source is thought to be lubricants from the argon blowers associated with the fuel handling equipment. Another potential source is lubricants from the primary pumps. The methane is removed by the natural cover gas leakage and, possibly, by its slow reaction with sodium.

5. Helium is normally not detectable in the primary cover gas but can exceed 50 ppm if certain helium blanketed fuel elements or experiments leak or are purposely vented.

6. Radioisotopes including those of Xe, Kr and Ne are always present in the primary cover gas. The Xe and Kr are produced from background concentrations of fissionable material in the sodium or on the fuel cladding or structural materials. Ne is a short-lived activation product of sodium that is detected only by the Germanium Lithium Argon Scanning System. When a fuel cladding ruptures the Xe and Kr activities can exceed safe limits in the reactor building and the reactor is shut down. Removal of the excess activity is achieved by purging with fresh argon.

7. Activities of tritium in the primary and secondary cover gases are variable. They are dependent on a large number of parameters, not all of which can be measured accurately. In general, the activity of tritium in the primary cover gas is 4-20 times greater than its activity in the secondary cover gas. A typical example is given by the samples of primary and secondary cover gas taken on Aug. 3, 1975. These samples showed tritium activities of 75 and 12 nCi/l,

respectively. In this instance, the reactor had been at full power and the cold traps had been operating without interruption for at least 11 days.

RADIOACTIVITY BEHAVIOR IN SODIUM SYSTEMS

The concentration of radionuclides in the EBR-II sodium systems has in general increased over the operating life of the plant.⁽³⁾ The reasons are:

1. Long-lived radionuclides are still approaching saturation activities.
2. Increased neutron activation, as the maximum reactor power was raised from 30 Mwt in 1964 through a number of steps to 62.5 Mwt in 1970.
3. Softening of the neutron spectra by replacing the depleted uranium blanket with a stainless steel reflector in 1972.
4. Release of fission products from ruptured fuel elements.
5. Loss of tin-bismuth alloy to the sodium providing for an increase in certain activation products.
6. Progressive, slow corrosion of the stainless steel components, adding activated corrosion products to the sodium.

In-Sodium Results

Some specific examples of radioisotope behavior are as follows:

1. Sodium-22 and ²⁴Na are the predominant activities in the EBR-II primary sodium system. ²²Na, with its 2.6 year half life has increased steadily over the life of the plant. Its current activity is near 100 nCi/g. It is the principal source of radioactivity after the decay of 15 hr ²⁴Na. ²⁴Na has shown a number of step increases in activity related to reactor power changes and the use of a stainless steel reflector. It now reaches saturation at about 3 mCi/g at full power operation. This activity is over 10,000 times greater than any other radioactivity in the sodium. Sodium sample handling or direct maintenance on the primary system requires about five days of decay of the ²⁴Na activity.

2. Cesium-137 is a dominant fission product in EBR-II sodium. It is introduced by fuel cladding breaks and was first noted in 1967. Measurable step increases have been noted and related to specific sub-assembly ruptures. Its activity is now near 70 nCi/g.

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^{137}Cs tends to concentrate in the cover gas regions and in the cold trap but in spite of this the ^{137}Cs activity continues to increase as additional fuel cladding ruptures take place.

3. Iodine-131 is analyzed frequently as an indicator of the seriousness of a fuel cladding rupture. The iodine measured is possibly related to the size or nature of the cladding breach. ^{131}I reaches a saturation activity of 20-100 pCi/g during reactor operation and is produced by background fissionable materials. Activities close to 10,000 pCi/g have been measured following fuel cladding breaks.

4. Tin-113 and ^{113}In are produced by neutron capture or electron capture in material from the tin-bismuth rotating plug seal alloy. A step increase in the activity of these isotopes was noted in 1972 and a steady increase has been noted since that time to their present activity of about 20 nCi/g. The increases are due to the ingress of the seal alloy which has been confirmed by a fourfold increase in the nonradioactive tin concentration over a ten-year period.

5. Polonium-210 is another activity that results from the tin-bismuth seal alloy getting into the sodium. It is an activation and decay product of ^{209}Bi . ^{210}Po is an alpha emitter with a 138-day half life. Its activity has steadily increased since it was first detected in 1971 to its current level of about 200 pCi/g.

6. Uranium and plutonium are both below their analytical detection limits (<1 ppb and <0.5 pCi/g, respectively). This is not surprising since subassemblies with ruptured fuel cladding are removed immediately after the break is detected and essentially all cladding breaks to date have been rather innocuous.

RESULTS OF COMPONENT EXAMINATIONS

The principal radionuclides found, more or less generally, on components in the EBR-II primary sodium system are ^{22}Na , ^{54}Mn , ^{137}Cs and ^{60}Co . Somewhat less pervasive is ^{182}Ta . Other radionuclides found in minor amounts or in localized concentrations are ^{210}Po (in deposits on the primary-pump impeller) and ^{58}Co (in stainless steel from the SURV-3 subassembly and on capscrews from primary pump No. 1). Not listed here are relatively short-lived (<3 days) radionuclides, or radionuclides on in-core materials.

Results from examinations of specific components are summarized below: (11)

1. The irradiated-fuel monitor thimble had been exposed to sodium in the primary tank for almost two years. Axial gamma scans with a GeLi detector measured relative activities of ^{54}Mn , ^{137}Cs , and ^{60}Co . Steam cleaning of the thimble removed 23% of the ^{54}Mn activity, 93% of the ^{137}Cs activity, and 0.3% of the ^{60}Co activity.

2. Primary pump No. 1 was removed in 1971 after 32,000 hrs of service and almost seven years in the primary tank. Radiation levels, measured on the uncleaned pump generally ranged from 15 mR/hr at 0.3 m (near the shield plug) to 130 mR/hr at 0.3 m (near the discharge). These radiation levels posed no serious problem during the repair of the pump.

The $^{137}\text{Cs}/^{22}\text{Na}$ ratios found on $\text{Na}/\text{Na}_2\text{O}$ deposits on pump surfaces that resided in the cover-gas region were as high as 100-400X the ratios of $^{137}\text{Cs}/^{22}\text{Na}$ in the bulk sodium before shutdown. This finding indicates that ^{137}Cs is volatile and tends to condense in the cover gas regions of the primary tank.

Axial scans of the pump assembly and of the pump components were made with a GeLi detector. Besides ^{22}Na , the major activities found on uncleaned pump surfaces were ^{54}Mn , ^{60}Co , ^{137}Cs , and ^{182}Ta . Decontamination of the pump components by scrubbing in hot baths of organic acids removed 44-67% of the ^{54}Mn activity, 42-92% of the ^{60}Co activity, and about 65% of the ^{137}Cs activity (only one case studied). The ^{182}Ta activity was essentially not removed, but appeared to be redistributed.

Decontamination of the pump and successive acid treatments of the baffle-subassembly capscrews show that ^{54}Mn and ^{137}Cs are strongly attached to the stainless steel. Roughly half the activity from these radionuclides was removed by nondestructive decontaminating solutions; the remainder yielded only to vigorous attack by hot mineral acids.

3. The intermediate heat exchanger (IHX) is the original IHX installed in 1962. Since wet criticality, an estimated $60 \times 10^6 \text{ m}^3$ of primary sodium have passed through it.

In 1971, several radiation scans were made along the axis of the IHX by lowering probes into the secondary inlet pipe after the sodium was pumped out.

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Essentially all the observed radiation originated on the primary side of the IHX. Maximum dose rates measured near the lower end of the IHX were 230 mR/hr. Dose rates measured below the level of the primary sodium were generally greater than 140 mR/hr. The contribution of ^{22}Na to these measurements is estimated to be less than 77 mR/hr. Thus, 50% or more of the measured dose rate must have originated from deposits of fission products and activated corrosion products.

4. The pump duct of the fuel-element-rupture-detector (FERD) loop continuously samples a 380 lpm stream of primary-sodium effluent from the IHX. This stream of sodium is monitored for delayed-neutron activity and then returned to the primary tank. The pump duct was replaced in April, 1970. A radiometric scan of the drained, but uncleaned, duct with a GeLi detector revealed only ^{54}Mn and ^{60}Co activities. An alcohol-water rinse removed 28% of the ^{54}Mn and 52% of the ^{60}Co activities. Cesium-137 was not detected. Presumably the ^{137}Cs diffused from the pump surfaces into the sodium when the pump duct was heated before the sodium was drained.

5. The SURV subassemblies are a set of 10 materials-irradiation subassemblies that were loaded into the EBR-II on March 1, 1965. On Feb. 24, 1971, subassembly SURV-3 was removed from the reactor core for examination. Wash water from its cleaning showed a high ^{182}Ta activity. Tantalum is one of the test materials exposed to sodium. Specimens were cut from the Type 304 stainless steel shroud that guided the flow of primary sodium past the test materials. One of these specimens was examined by electropolishing successive 1.3×10^{-2} mm layers from the surface until 13×10^{-2} mm of material had been removed. Measurements of the activities of the principal radionuclides in the electropolishing solutions showed that ^{182}Ta was totally removed with the first 1.3×10^{-2} mm of steel. The concentrations of ^{54}Mn , ^{58}Co and ^{60}Co were distributed fairly uniformly throughout the top 13×10^{-2} mm of surface, although ^{58}Co and ^{54}Mn activities were slightly higher in the outer 1.3×10^{-2} mm layer.

Results of External Cold Trap Gamma Scans

Beginning in 1971 and at intervals thereafter, radiation levels around the perimeter of the primary cold trap were measured using survey meters. The radiation level of the trap has increased roughly in

proportion to the cumulative energy produced by the reactor. Isodosic maps of the radiation show two persistent "hot spots". One, near the bottom of the tank at the NaK-coolant inlet, was to be expected because this area is where precipitating impurities should collect. The other "hot spot" was 3.14 radians around the tank from the first and near the top of the tank. This locus of radioactivity was not expected and, at the moment, remains unexplained. The most recent survey, made in April 1975, showed radiation levels of <1 R/hr at both of these "hot spots" 31 days after the trap had been isolated from flowing primary sodium (^{24}Na was essentially completely decayed).

One gamma-energy scan of the cold trap, made in June, 1972, using a GeLi detector, 81 days after shutdown, identified ^{22}Na , ^{54}Mn , ^{60}Co , ^{65}Zn , ^{124}Sb , ^{134}Cs and ^{137}Cs . The concentration factor for $^{137}\text{Cs}/^{22}\text{Na}$ in the cold trap vs the 370°C (700°F) bulk sodium was 50. This shows that the cold trap has removed some (~30%) of the ^{137}Cs arising from fission-product releases to the sodium. The principal radioisotopes found in this scan, listed in descending order of activity, were: ^{137}Cs , ^{54}Mn , ^{125}Sb , ^{134}Cs and ^{22}Na .

SUMMARY

EBR-II has operated in a safe and reliable manner for almost 12 years. Excellent sodium purity control supported by a rigorous purity monitoring program has been instrumental in achieving this history. Sodium technology efforts at EBR-II will continue to contribute to the high availability and operability of the reactor and coolant systems.

REFERENCES

1. Olson, W. H., Miles, C. C., and Cissel, D. W., "Impurities in EBR-II Sodium and Cover Gas," ANL-7520 Part I, pp. 222-226 (1968).
2. Olson, W. H., "Sampling and Analysis of EBR-II Sodium," ANL-7844 (1971).
3. Smith, C.R.F. and Holmes, J. T., "Purity of EBR-II Sodium: 1967-1974," ANL 75-26 (1975).
4. "Methods for the Analysis of Sodium and Cover Gas," RDT Standard F3-40T, (January 1976).
5. Holmes, J. T. and Haroldsen, G. O., "The Utilization of On-line Monitors at EBR-II for Sodium

INTERNATIONAL CONFERENCE ON LIQUID METAL TECHNOLOGY IN ENERGY PRODUCTION

Purity," Nuclear Technology 21, pp 228-234 (March 1974).

6. Smith, C.R.F., Richardson, W. J. and Holmes, J.T., "On-line Sodium and Cover Gas Purity Monitors as Operating Tools at EBR-II," International Conference on Liquid Metal Technology in Energy Production, Session VIII A, Champion, Pa. (May 3-6, 1976).
7. Vissers, D. R., Holmes, J. T., Bartholme, L. G., and Nelson, P.A., "A Hydrogen Activity Meter for Liquid Sodium and Its Application to Hydrogen Solubility Measurements," Nuclear Technology 21, pp. 235-244 (March 1974).
8. Kolba, V. M., Slawecki, M. A., Filewicz, E. C., McKee, J. M., Holmes, J. T., and Nelson, P. A., "Carbon Meter - Equilibration Module for LMFBR's: Design and Operation," ANL-7941 (May 1972).
9. McKee, J. M., Vissers, D. R., Nelson, P. A., Grundy, B. R., Berkey, E., and Taylor, G.R., "Calibration Stability of Oxygen Meters for LMFBR Sodium," Nuclear Technology 21, pp 217-227 (March 1974).
10. Wrightson, M. M., McKinley, K., Ruther, W. E., and Holmes, J. T., "EBR-II Water-to-Sodium Leak Detection System," International Conference on Liquid Metal Technology in Energy Production, Session VI B, Champion, Pa. (May 3-6, 1976).
11. Smith, C.R.F., "Radioactivity of EBR-II Components: I. Examinations in FY 1971," ANL-7873, (1972).