ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

EXPERIMENTAL IRRADIATIONS OF FUEL ELEMENTS
HAVING KNOWN CLADDING DEFECTS IN EBR-II

by

G. S. Brunson, R. M. Fryer,
and R. V. Strain

EBR-II Project

July 1971

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	7
I. INTRODUCTION.	7
II. BRIEF DESCRIPTION OF EBR-II	8
III. FISSION-PRODUCT MONITORING SYSTEMS.	9
A. Methods of Monitoring Fission Products	9
1. Fuel Element Rupture Detector (FERD)	9
2. Sodium Analysis	10
3. "Grab Samples"	10
4. Fission Gas Monitor (FGM)	10
5. Reactor Cover Gas Monitor (RCGM).	10
B. Operational Factors Affecting Data.	11
IV. TOP-LEAKING EXPERIMENT	12
V. BOTTOM-LEAKING EXPERIMENT	14
A. Preparation of Bottom Leaker	14
B. Calculation of Anticipated Effect of Bottom Leaker	16
C. Irradiation and Concurrent Observations	20
D. Consolidation of Observation	23
E. Results of Postirradiation Examination	28
VI. DISCUSSION AND CONCLUSIONS	42
APPENDIX. Summary of Isotopes Pertinent to the EBR-II	
Fission-product-monitoring Systems	43
ACKNOWLEDGMENTS	44
REFERENCES	45

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Mark-IA Driver-fuel Element	9
2.	Spade Piece Used in Bottom-leaking Experiment	14
3.	Neutron Radiograph of Bottom of Fuel Elements	15
4.	Calculated Surges in ^{135}Xe Activity, Assuming the 10 Leak Rates Listed in Table V	20
5.	^{135}Xe Activity in the Cover Gas and Reactor Power History during the Initial Irradiation of C-2250S, in Core Position 3B1	22
6.	^{135}Xe Activity in the Cover Gas at the End of Run 39B; C-2250S in Position 3B1	24
7.	^{135}Xe Activity in the Cover Gas during Runs 40A and 40B	25
8.	Comparison of Experimentally Observed ^{135}Xe Surges with Calculated Values	26
9.	Plot of Time from Turning Pumps Off to Arrival of Surge Peak vs Time Pumps Were Off.	26
10.	Comparison of ^{135}Xe Data from Grab Samples and from RCGM Using Delay Line with ~230-sec Sample-gas Age	27
11.	Concentration of ^{131}I in Primary Sodium during the Period Covered by These Experiments	28
12.	Eddy-current Bond-test Traces of Elements 46 and 47 from C-2250S	30
13.	Eddy-current Bond-test Traces of Elements 45 and 47 from C-2008.	31
14.	Photo of the Apparent Defect in the Lower-end-fitting Weld on Element 47 from C-2008	32
15.	Neutron Radiograph of Top of Elements 47, 50, 46, 39, and 45 from C-2250S and Element 47 from C-2008	32
16.	Photos Comparing the Weld on Element 47 from C-2008 and a Normal Lower-end-fitting Weld	35
17.	Micrograph Showing the Beginning of the Defect in the Weld at the OD of the Cladding.	35
18.	Series of Micrographs Showing the Progression of the Defect to the ID of the Cladding	36
19.	Photos Showing Cross Sections of Element 47 from C-2008	37

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
20.	Photos Showing Cross Sections of Element 46 from C-2250S . .	38
21.	Microstructures of Typical Areas near the Centerline of the Fuel Pin from Element 46 from C-2250S	39
22.	Microstructures of Typical Areas near the Edge of the Fuel Pin from Element 46 from C-2250S.	40
23.	Microstructures of Typical Areas near the Centerline of the Fuel Pin from Element 32 from C-2166	41

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	Irradiation History of Top-leaking Experiment	12
II.	Element Weights from Subassembly C-2194S.	13
III.	Equilibrium Volumes of Gas and Sodium in Bottom-leaking Fuel Element for Various Core Positions	16
IV.	Calculated Peak Surge Concentrations of ^{135}Xe for Various Positions of the Bottom Leaker in the Core.	17
V.	Effect of Leak Rate on Surge in ^{135}Xe Activity, Assuming Expulsion of I_0 Atoms of ^{135}I into Primary System	19
VI.	Irradiation History of Bottom-leaking Experiment	21
VII.	Summary of Surges Observed in ^{135}Xe Concentration.	21
VIII.	Weights of Suspect and Control Elements	33
IX.	Pressure and Volume Data for Elements from C-2250S and C-2008	33

EXPERIMENTAL IRRADIATIONS OF FUEL ELEMENTS HAVING KNOWN CLADDING DEFECTS IN EBR-II

by

G. S. Brunson, R. M. Fryer,
and R. V. Strain

ABSTRACT

The EBR-II program to develop techniques for detecting and identifying leaking fuel elements has included experimental irradiation of fuel elements having known leaks. This report covers two such tests: a "top leaker" having a small leak in the upper weld (which, however, did not leak enough to be detected) and a bottom leaker having a small laser-drilled hole (~3-mil diameter) at the bottom end of the fuel element. After a reactor shutdown, if the primary pumps are turned off, the reduction in external pressure permits the expulsion of bond sodium containing radioactive iodine from the fuel element. The daughter xenons (especially ^{135}Xe) growing out of the expelled iodine appear as a surge in primary-tank cover-gas activity, reaching a peak 11 or more hours after the reactor shutdown. This is a useful trademark of this type of leak; a semiquantitative model is presented to explain this behavior.

I. INTRODUCTION

Above-normal levels of fission-product activity in the primary system of EBR-II are indicative of leaking fuel cladding. Fission-product retention in metallic uranium is so high at the burnups incurred to date that no inconvenient radiation levels have been observed. In contrast, oxide fuel can release enormously greater (by about three orders of magnitude) quantities of fission products.¹ There is also good evidence that the thermal conductivity of uranium metal is so high that the Mark-IA driver-fuel element can tolerate substantial loss of bond sodium without significant danger of melting.² Thus some types of leaking fuel conditions are tolerable for sustained operation.

The purpose of the experiments reported here was to introduce known metallic-driver-fuel "leakers" into the core and observe the "symptoms" as reported by the operational monitoring systems. The association of symptoms with known defects makes it easier to annunciate

and identify unknown sources and fission products. Postirradiation examinations were made to assess the effect of the leaks on the structural integrity of the fuel pin and jacket. To date, two such experiments have been performed. The first was a "top leaker," which represented a realistic potential defect, that of an imperfect top weld, which somehow escaped detection. The second was a "bottom leaker," which simulated a similar defect in the bottom weld.

II. BRIEF DESCRIPTION OF EBR-II

This section covers the general characteristics of EBR-II that are necessary to explain the present work. Greater detail is available in the literature.³⁻⁵ EBR-II is a fast reactor using approximately 50%-enriched uranium fuel pins (0.144-in. diameter, 13.5 in. long) clad in 9 mils of stainless steel as the basic "driver" fuel. These fuel elements are packaged in hexagonal subassemblies containing 91 elements. A typical core loading (roughly 24 in. in diameter) comprises about 90 subassemblies of which perhaps one-fourth contain experimental material (frequently oxide fuel) rather than driver fuel. The core is blanketed by several rows of hexagonal subassemblies loaded with larger-diameter ($\sim 1/2$ -in.) pins of depleted uranium. The molten sodium coolant passes upward through the core and blanket. The core and blanket are contained in the reactor vessel, which is surrounded by a 2-ft-thick neutron shield of graphite. All the foregoing are immersed in 90,000 gal of sodium contained in the primary tank (about 26 x 26 ft). Also located in the primary tank are the two coolant pumps, which draw from the bulk sodium in the tank to force sodium up through the core and blanket, over to and down through the primary heat exchanger, and back into the bulk sodium. In the primary tank above the surface of the sodium is a free volume of about 700 ft³, filled with argon "cover gas"; this gas is at a slight positive pressure to inhibit the entrance of oxygen or other substances that would react with the sodium.

Figure 1 shows the details of the Mark-1A driver-fuel element. All fuel-element assembly operations are performed under an argon atmosphere. First a metered amount of extruded sodium is placed in the bottom of the jacket. The fuel pin is then inserted and comes to rest in a high position on the solid sodium. The restrainer is placed on top of the pin, and the whole is heated in a "settler" to well above the melting point of sodium. The pin and restrainer settle to their proper positions as the sodium flows up to occupy the annular space between the pin and jacket and reaches a level nominally 0.65 in. above the top of the fuel. Still under argon, the fuel element is placed in a jig, and the restrainer is discharge-arc-welded to the jacket.

The closure welds are tested by placing the top end of the fuel element in a fixture of very small volume. High-pressure (600-psi) helium is

applied for about 7 min, and a leak is identified by a decay in the system pressure due to in-leakage into the fuel-element plenum.

Fig. 1. Mark-IA Driver-fuel Element

Normally the sodium when melted does not uniformly fill the annulus, and a number of voids are formed. To remedy this, all fuel elements are "impact-bonded." This operation maintains the element at a temperature well above the melting point of sodium while the element is impacted 1000 times in the vertical position at the rate of one per second. The fuel element is then examined by an eddy-current test to determine the size and distribution of bond voids. In some cases, one cycle is not enough to bring the bond uniformity up to the rather stringent standard specified, and the element is subjected to another impact-bonding cycle. It is then re-examined for voids. If it does not pass after the second series, it is rejected because repeated impacting progressively deforms the "spade."

III. FISSION-PRODUCT MONITORING SYSTEMS

A. Methods of Monitoring Fission Products

There are five methods of monitoring fission products in the primary system of EBR-II

1. Fuel Element Rupture Detector (FERD)

The FERD monitors fission products in the sodium coolant. A side stream of sodium is taken from the outlet of the primary heat exchanger and pumped through a block of graphite (roughly a cubic meter) in which are embedded $^{10}\text{BF}_3$ proportional counters to detect delayed neutrons produced by fission products entrained in the sodium. Transit time for the sodium from the core to the counting position is of the order of 20 sec. Because of the short half-life of the delayed-neutron precursors and the relatively slow escape of fission products from a leaking jacket, the FERD did not respond during the experiments reported here. The FERD is described in full detail elsewhere.⁶

2. Sodium Analysis

Discrete samples of sodium are counted for ^{131}I . Because of the cooling time required for the ^{24}Na activity, there is a delay of several days between sampling and analysis. Samples are normally taken about every five days. Personnel exposure inhibits taking samples much more often, except under specific need. There have been wide gaps in the data because of operating difficulties with the sampling system.

The other three monitoring methods depend on counting noble-gas fission products that have diffused from the sodium to the argon cover gas or the deposited radioactive daughters of such noble gases. A gas sample stream of about $5\text{ ft}^3/\text{hr}$ is drawn through a sodium-vapor trap and a particulate filter to be shared by the following systems:

3. "Grab Samples"

These are discrete samples drawn from the gas-sampling system into a 10-ml glass bulb, which is carried to a nearby laboratory, where it is counted with a 3×3 -in. sodium iodide crystal into a conventional multichannel analyzer. The spectra are analyzed for ^{133}Xe and ^{135}Xe ($^{85\text{m}}\text{Kr}$ can also be observed, but the data are not used). The sampling rate is normally six per day during operation; this can be increased when needed to perhaps one per hour. Delay time from sampling to numerical results is of the order of 15-20 min.

4. Fission Gas Monitor (FGM)⁷

In the FGM, a small stream of cover gas flows through an electrostatic precipitator. The decay of noble-gas fission-product atoms (various xenons and kryptons) in this volume leaves ionized daughter atoms (cesium or rubidium), which are precipitated by an electric field onto a moving wire at the center of the cylindrical precipitator. The moving wire carries the precipitated atoms into a small volume of water, which readily picks up the alkali atoms. Two conventional scintillation detectors (2×2 -in. sodium iodide) count the gamma activity of the water sample. The associated discriminators are arbitrarily set to count all pulses above the energy of ^{137}Ce (662 keV). Of the various rubidiums and cesiums deposited in the water, some are stable or have very low specific activity. The bulk of the activity counted is from ^{88}Rb and ^{138}Cs --with some indication of ^{89}Rb . The FGM requires several minutes to react to a step increase in fission products in the cover gas.

5. Reactor Cover Gas Monitor (RCGM)⁸

The RCGM is based on a gamma scintillation detector (2×2 -in. sodium iodide), which views the flowing cover gas as it passes through a

counting chamber of about 50-ml volume. The pulses from the detector are fed to a bank of three single-channel analyzers, each of which is set to accept pulses in an energy interval associated with a specific isotope. In practice, it has been found best to observe ^{85m}Kr , ^{133}Xe , and ^{135}Xe . The RCGM suffers from the interference of ^{23}Ne (half-life, 38 sec; principal gamma, 440 keV), which is produced in relatively large quantities by the reaction $^{23}\text{Na}(n,p)^{23}\text{Ne}$ in the coolant. The diffusion of ^{23}Ne from the sodium to the cover gas is erratic because of the extreme sensitivity of the diffusion rate to the sodium temperature. The Compton continuum from the ^{23}Ne falls across the energies associated with the three isotopes of interest. As a result, the fluctuations in the ^{23}Ne activity appear in the channels dedicated to the fission products. This effect has been reduced to a few percent by aging the gas in a "delay line" before it is counted. The actual age of the gas when counted is about 4 min.⁸

B. Operational Factors Affecting Data

No matter how "clean" the core is, fission products still appear in the cover gas and in the primary sodium with an activity level corresponding to roughly 7 mg of unclad ^{235}U in the core. This slight, apparently irreducible contamination has been termed "tramp" uranium. Fission products from this source are quite helpful because they furnish reasonably constant, but unobtrusive, test sources for all the fission-product monitoring techniques enumerated above.

There are two further effects associated with routine reactor operation that have significance in interpreting fission-product data. The movement of fuel into and out of the reactor permits the escape of some cover gas with an apparent loss of activity due to dilution with clean makeup gas. This affects all three gas-counting systems. Perturbations in the gaseous fission-product level must be reconciled with the extent of fuel handling at the time in question. The effect is always in the direction of reduced activity.

The other operational perturbation has to do with the "cold trap," which is used to maintain coolant quality by trapping impurities (of which sodium oxide is the most important). Although the cold trap is designed to be in nearly constant operation, it has sometimes been shut down for extended periods. The problem here arises from the fact that the cold trap picks up various fission products, and the effect is apparent on those isotopes whose precursor chain includes a trappable isotope with at least a moderate half-life. Cold-trap operation affects the ^{131}I (8 days) directly, the reduction in activity approaching a factor of two. Its operation reduces the ^{133}Xe and ^{135}Xe levels by 20% or more, by trapping the precursors ^{133}I (21 hr) and ^{135}I (6.7 hr). The cold trap has no visible effect on the delayed-neutron level in the FERD or on the isotopes counted by the FGM, since their precursors all have relatively short half-lives.

IV. TOP-LEAKING EXPERIMENT

The fuel element chosen for this experiment was simply one found to be defective in the regular top-weld leak test. This fuel element is identified as pin No. 3, batch 115 IH. In normal practice, this element would have been discarded. For this experiment, it was returned to the production line and routinely impact bonded. Two treatments were necessary to obtain an acceptable bond test. The top weld was again leak-tested just before incorporation into the subassembly to ascertain if the leak had plugged. The estimated size of the leak was $\sim 1/2$ mil in effective diameter.

The fuel element was weighed, gauged, X-rayed, and neutron-radiographed, and was found to be entirely within specifications (except for the leak indication). It was loaded in position 46 (exact center of the hexagonal array) in an otherwise completely normal driver subassembly designated C-2194S. Two adjacent elements served as controls. This subassembly was loaded into the core on May 14, 1969, in grid position 3C1. In accordance with experimental procedure EX 27-68, the frequency of data taking was increased on all channels observing fission products. Over several days of operation, there was no indication of fission-product leakage. A second and much longer irradiation began on August 2, and again there was no effect attributable to this subassembly. It was concluded that the weld defect was plugged and that the subassembly should be burned up as any other driver subassembly. The complete irradiation history is given in Table I.

TABLE I. Irradiation History of Top-leaking Experiment

1. Subassembly No. C-2194S Burnup: Maximum 1.7673 Average 1.5196

2. Irradiation History:

Date to Grid	Grid Position	Date from Grid	Hours in Position	MWd in Position	Burnup		Maximum Reactor Power	Hours at Power
					Maximum	Average		
a. 5-14-69	3C1	5-28-69	336	330	0.1012	0.0071	50	135
b. 8-2-69	3F2	9-1-69	720	1196	0.3900	0.3388	50	564
c. 10-1-69	3B1	12-13-69	1752	400	0.1915	0.1450	50	278
d. 4-17-70	1A1	5-22-70	840	1344	0.4025	0.4025	50	639
e. 5-23-70	5B2	7-16-70	1296	2250	0.6341	0.5284	50	1060

3. Residence Time:

Storage Basket, hr		Primary Tank	
Before Grid Installation	0	Total Hours	10,800
After item a above	1384	Hours after Reactor Shutdown	552
After item b above	720		
After item c above	3008		
After item d above	24		
After item e above	528		
Total:	5856		

4. Total Reactor Operating Time While SA in Grid 2877 hours

5. Reactor Operating Data:

- Maximum Power Level of Reactor 50 MW
- Total Integrated Power of Reactor: Data 7-16-70 MWd 37,490

6. Subassembly Operating Data:

Grid Position	Average Power	Maximum Temperature, °F	Hours at Maximum Temperature
a. 3C1	0.733	851	135
b. 3F2	0.738	854	564
c. 3B1	0.713	851	278
d. 1A1	0.777	843	639
e. 5B2	0.601	901	1060

The postirradiation examination included bond testing, neutron radiographing and weighing of the element, and leak testing of the top weld after the element was sectioned.

The eddy-current bond-test traces of the elements indicated no major changes in the sodium levels in any of the three elements. All the levels were about 1 in. above the fuel pin. The level could not be measured accurately because of bubbles or shrinkage voids in the sodium above the fuel pins.

The neutron radiograph of the elements revealed no unusual features in any of the elements. Elements that have been punctured, exposing the bond sodium to the air, have exhibited a line of high-neutron-absorbing material at the top of the sodium. The absence of such a line in the radiograph of element 46 indicates that no significant amount of air leaked into the element after irradiation.

The pre- and postirradiation weights of the three elements are given in Table II. These data indicate that element 46 did lose about 0.3 g; but the accuracy of the balance used to weight the element after irradiation was only ± 0.2 g. Thus, this weight change is not a reliable indication of any change in the element. There could, however, have been a loss of fission gas from the element. The much lower viscosity of the gas could allow it to escape from the element with none of the higher-viscosity sodium being forced back into the element. Quantitatively, this might be a partial explanation of the weight loss if such a loss was real. The average burnup according to Table I was 1.52% or about 1 g of uranium. The yield of stable fission gases is about one-fourth mole of gas per mole of uranium fissioned. Thus, on the average, each uranium atom fissioned produces one-fourth atom of stable gas whose average atomic weight is ≈ 110 . The fission of 1 g of uranium would yield about $(1/4)(110/235)(1 \text{ g}) \approx 0.1 \text{ g}$ of noble-gas fission products. The principal uncertainty in this conjecture is the rate of diffusion of noble gas in uranium metal, about which there is little information. Table I shows that the experimental element spent nearly 15 months in the elevated temperature ($> 700^\circ\text{F}$) of the primary system, of which more than 20 days passed after the completion of irradiation. Under these circumstances, significant diffusion of the stable noble-gas fission products may have occurred.

TABLE II. Element Weights from Subassembly C-2194S

Element Number	Postirradiation Weight, g	Preirradiation Weight, g	Weight Change, g
45	84.1	84.0	+0.1
46	82.8	83.1	-0.3
47	84.1	84.0	+0.1

The experiment must be considered inconclusive, and future experiments are planned using simulated leaks drilled with a laser. The fact that no significant effect was observed from an "officially" defective element does, however, suggest that the specifications are more rigorous than necessary.

V. BOTTOM-LEAKING EXPERIMENT

A. Preparation of Bottom Leaker

Some effort was required to devise a method of producing a leak at the lower end of the fuel pin with a minimum of artificiality. The following

Fig. 2. Spade Piece Used in Bottom-leaking Experiment. At left, spade piece with milled slot and drilled hole; at right, same type of spade piece after dry run, showing peening at top. ANL Neg. No. 103-M5380A.

technique was used after it had been tried out in "dry run." The spade piece first had a slot milled across the top (see Fig. 2). Then a small hole was drilled from below until a faint bulge appeared in the bottom of the slot. The final opening was made through the bulge by a laser beam. The diameter of this orifice was estimated at 3 mils by photograph.

The spade with the prepared leak was then welded into a normal jacket tube. Leak-rate measurements confirmed the effective orifice diameter. The outer end of the hole was welded shut, and the jacket was loaded exactly as any regular jacket, including upper-weld testing and impact-bonding.

Several special measurements and examinations were made to identify changes that might occur during the experiment. Precise weight and

dimensions were taken. Two controls (i.e., completely normal fuel elements) were similarly characterized for comparison purposes. The experimental element was X-rayed and neutron-radiographed. An important aspect of this latter examination was to ensure that the inner orifice remained unobstructed. Note the peening apparent in the right-hand spade in Fig. 2, which had undergone a dry run. The purpose of the slot was to protect the orifice from being peened shut. The slot is visible end-on as a small white spot at the bottom of the pin in the neutron radiograph (Fig. 3); this confirms that the impact-bonding of the experimental fuel element did not affect the bottom of the slot and by inference did not affect the orifice.

Fig. 3. Neutron Radiograph of Bottom of Fuel Elements. Arrow points to white spot in element 46 that indicates slot in spade piece.

The leak, however, was still closed by the weld across the outside of the original hole. It was now necessary to open the outer end of the hole by drilling through the weld. Care was taken to minimize the time that the leak was exposed to the air.

On December 23, 1969, the experimental subassembly (C-2250S) was loaded with the first 45 elements, No. 45 being one of the controls. At this point, the experimental element was opened by drilling through the weld. Penetration was confirmed by extracting a grain of metal, which was clearly identified as sodium by its reaction with water. The experimental element was installed in

position 46, followed by the second control in position 47. Thereafter the subassembly was completed with normal elements as rapidly as possible. Assembly welding of the subassembly was completed about 20 min after the leak was drilled open. Gauging was completed, and the argon purge was begun in the temporary coffin approximately 1 hr after opening the hole. The subassembly was raised into the fuel-unloading machine to begin heating up at about $1\frac{1}{2}$ hr. Movement into the reactor started about 3 hr after the hole was opened. After about 1 hr at the "heating and blowing" position, the subassembly was immersed in the coolant and placed in the reactor storage basket.

The total elapsed time was roughly $4\frac{1}{2}$ hr, which is nearly minimum for all the required operations. During this time, great care was exercised to avoid any downward trend in temperature and to minimize exposure to air. These precautions were based on the following considerations: As the experimental fuel element is heated past the melting point of sodium, the increased pressure in the plenum expels a significant amount of sodium through the leak. This is unavoidable. What must be avoided is to allow a decrease in temperature while the element is in a gas atmosphere. If this were to happen, the contraction of the gas in the plenum would permit the ambient pressure to inject gas bubbles into the bond annulus, thereby defeating the effort to have the experimental element bonded exactly the same as a normal fuel pin. Of course, if a temperature decrease occurs while the element is immersed in sodium, no problem results; the element simply "inhales" more sodium to replace that originally expelled.

B. Calculation of Anticipated Effect of Bottom Leaker

With the reactor shut down and primary pumps turned off, the ambient pressure at the site of the bottom leak is 18.7 psia (12.2 psia local atmospheric pressure plus sodium static head) and the nominal temperature is 700°F. The actual pressure on the gas is 18.2 psia when its height above the leak is taken into account. Since the element was originally loaded with 0.672 ml of plenum gas at 70°F and 12.2 psia, the above shut-down conditions result in a gas volume of

$$(0.672) \left(\frac{12.2}{18.2} \right) \left(\frac{1160}{530} \right) = 0.986 \text{ ml}$$

when the system is stabilized by expulsion of sodium.

When the pumps are turned on, there is an increase in the effective external pressure at the leak, depending on the radial location of the subassembly in the core. In addition, the gas temperature increases to about 840°F (for 50-MWt operation). Under these conditions, the leaking element establishes a new equilibrium volume in accord with the values in Table III.

TABLE III. Equilibrium Volumes of Gas and Sodium in Bottom-leaking Fuel Element for Various Core Positions

Core Position	Ambient Pressure during Operation, psia	Equilibrium Gas Volume at 50 MW, ml	ΔV , ^a ml	Equilibrium Sodium Volume at 50 MW, ml
Row 1	44.2	0.455	0.535	0.987
Row 2	39.6	0.508	0.476	0.934
Row 3	32.4	0.621	0.359	0.821
Row 5	30.0	0.670	0.310	0.772

^a ΔV is the volume of sodium "inhaled" when the reactor goes from shutdown to 50-MWt operation and "exhaled" when the reactor is again shut down.

Of each sodium volume listed in Table III, 0.625 ml is contained in the annular space between the pin and the jacket. After shutdown, the reduced external pressure allows the plenum gas to expand again to the above-calculated 0.986 ml, leaving about 0.456 ml of sodium in the element. This is enough to fill the annulus to only about 10 in., assuming rather unrealistically that the sodium level is depressed uniformly. Actually, the pin is almost always tilted and not perfectly straight, so that surface tension may reasonably be expected to hold up an unpredictable quantity of sodium in the narrower part of the annulus.

The sodium expelled from the fuel element contains a number of fission products; however, in the present analysis, we are concerned only with the ¹³⁵I-¹³⁵Xe chain. The half-lives of ¹³³I and ¹³³Xe are too long to

offer useful data under the operating conditions of EBR-II. To calculate the amount of ^{135}I ejected into the primary system when the pumps are shut down, we first estimate the equilibrium inventory in the bond sodium. This is shown in column 1 of Table IV. These numbers are derived from a value of $4.0 I_0$ in the fifth row; I_0 is used to denote the equilibrium inventory at 50 MWt of ^{135}I atoms in the primary system from tramp uranium. The value for row 5 was established by bare-pin experiments.^{9,10} Similarly, X_0 is used to denote the equilibrium inventory of ^{135}Xe at 50 MWt.

TABLE IV. Calculated Peak Surge Concentrations of ^{135}Xe for Various Positions of the Bottom Lasker in the Core

	1	2	3 ^a	4	5 ^a	6 ^a	7 ^b	8 ^b	9 ^b
Core Position	Bond Inventory of ^{135}I , Units of I_0^c	Total Na In Element, ml	Concentration ^{135}I in Bond, I_0^c/ml	Na Expelled at Pump Shutdown, ml	Surge Inventory, Units of I_0^c	Predicted Peak Activity of ^{135}Xe , $\mu\text{Ci/ml}$ ($\times 10^3$)	Concentration ^{135}I in Bond, I_0^c/ml	Surge Inventory, Units of I_0^c	Predicted Peak Activity of ^{135}Xe , $\mu\text{Ci/ml}$ ($\times 10^3$)
Row 1	5.2	0.987	5.27	0.535	2.82	2.68	8.32	4.45	4.23
Row 3	4.9	0.934	5.25	0.476	2.50	2.38	7.84	3.73	3.95
Row 4	4.5	0.821	5.48	0.399	1.97	1.87	7.20	2.51	2.38
Row 5	4.0	0.772	5.18	0.310	1.61	1.53	6.40	1.98	1.88

^aThese values are based on the assumption that the ^{135}I is uniformly distributed in all bond sodium.

^bThese values are based on the assumption that the ^{135}I is uniformly distributed in the 0.625 ml of annular sodium only.

^c I_0 represents the total tramp equilibrium inventory of ^{135}I in the primary system.

Column 2 repeats from Table III the quantity of sodium present in the fuel element when the reactor is operating. Column 3 is the bond concentration of ^{135}I , assuming that the inventory in column 1 is uniformly distributed in the sodium volume shown in column 2. Column 4 is (again from Table III) the volume of sodium expelled when the pumps are shut down. This quantity times the concentration gives, in terms of I_0 , the inventory expelled (column 5).

If we assume that I_0 atoms of ^{135}I are introduced into the primary coolant at time 0, the equations for the ^{135}I and ^{135}Xe inventories are

$$I(t) = I_0 e^{-\lambda_I t}$$

and

$$\frac{dX(t)}{dt} = \lambda_I I_0 e^{-\lambda_I t} - \lambda_X X(t),$$

for which the solution is

$$X(t) = \frac{\lambda_I I_0}{\lambda_I - \lambda_X} (e^{-\lambda_X t} - e^{-\lambda_I t}).$$

The ^{135}Xe concentration due to the expulsion of ^{135}I at first increases because of the production from decay of iodine, passes through a maximum at about $t = 11.3$ hr, and thereafter decreases. Substitution of this value for t in the last equation yields

$$X(t)_{\max} = 0.427 I_0.$$

However, at equilibrium conditions for tramp activity,

$$\lambda_I I_0 = \lambda_X X_0 \text{ or } X_0 = \left(\frac{\lambda_I}{\lambda_X} \right) I_0 = 1.37 I_0.$$

Therefore, the maximum in the ^{135}Xe inventory above that due to tramp activity is

$$X_{\max} = \frac{0.427 I_0}{1.37 I_0} = 0.312 X_0.$$

That is, the peak inventory of ^{135}Xe associated with the expelled sodium is the above fraction of the tramp inventory. The equilibrium activity of ^{135}Xe from the tramp source is $3.0 \times 10^{-3} \mu\text{Ci/ml}$. Thus, after shutting the reactor down and turning off the primary pumps, we would expect to observe a surge above the normal decay of ^{135}Xe from equilibrium operation. The maximum would occur 11.3 hr after the pumps were turned off, and the peak amplitude would be

$$3.0 \times 10^{-3} \times 0.316 = 0.94 \times 10^{-3} \mu\text{Ci/ml}$$

for every I_0 atoms of ^{135}I atoms expelled. This factor can now be used to complete Table IV with the calculated peak amplitudes (column 6). In columns 7, 8, and 9, the calculations are repeated under the assumption that, because of the narrow (6-mil) diffusion channel, the ^{135}I atoms are entirely contained in the 0.625 ml of annular sodium.

Table IV has been calculated on the assumption that the orifice was large enough for the expulsion of sodium to be considered instantaneous on the time scale associated with ^{135}I - ^{135}Xe . We now examine some cases in which a slower leak rate is assumed. Assume that when the pumps are turned off at $t = 0$, there is a volume of sodium V_0 destined by the change in external pressure to be expelled; assume that the ^{135}I content of this volume is I_0 (the same I_0 as defined above). Assume further that the leak is exponential with decay constant ℓ .

Thus,

$$\frac{dV(t)}{dt} = -\ell V(t),$$

where $V(t)$ is that portion of V_0 remaining to be leaked. But the ^{135}I concentration of the leaking sodium is

$$\left(\frac{I_0}{V_0} \right) e^{-\lambda_I t}$$

because of the decay of the iodine, so that the equation for the ^{135}I input and loss in the primary sodium is

$$\frac{dI(t)}{dt} = I_0 \ell e^{-(\ell + \lambda_I)t} - \lambda_I I(t).$$

The equation for the ^{135}Xe added to the primary sodium as a result of the leak is

$$\frac{dX}{dt} = \lambda_I I(t) - \lambda_X X(t).$$

The solution for these two equations is

$$X(t) = \lambda_I I_0 \ell \left[\frac{e^{-(\ell + \lambda_I)t}}{\ell(\ell + \lambda_I - \lambda_X)} + \frac{e^{-\lambda_I t}}{\ell(\lambda_X - \lambda_I)} + \frac{e^{-\lambda_X t}}{(\ell + \lambda_I - \lambda_X)(\lambda_I - \lambda_X)} \right]$$

Since $\lambda_X X_0 = \lambda_I I_0$, and since

$$\frac{\lambda_X X_0}{3.7 \times 10^4 \cdot \text{cover-gas volume}} = 3.0 \times 10^{-3} \mu\text{Ci/ml},$$

we can write the predicted activity $A(t)$ in $\mu\text{Ci/ml}$ of cover gas:

$$A(t) = 3.0 \times 10^{-3} \mu\text{Ci/ml} \cdot \ell \left[\frac{e^{-(\ell + \lambda_I)t}}{\ell(\ell + \lambda_I - \lambda_X)} + \frac{e^{-\lambda_I t}}{\ell(\lambda_X - \lambda_I)} + \frac{e^{-\lambda_X t}}{(\ell + \lambda_I - \lambda_X)(\lambda_I - \lambda_X)} \right]$$

in which the numerical term normalizes the activity to unit gas volume. This equation has been used to calculate the activity for a number of different leak rates. The resulting curves are shown in Fig. 4. The characteristics of these curves are summarized in Table V; the 10 curves correspond to the 10 values of leak rate given in the table.

TABLE V. Effect of Leak Rate on Surge in ^{135}Xe Activity, Assuming Expulsion of I_0 Atoms of ^{135}I into Primary System

Leak Rate, hr^{-1}	Leak "Half-life," hr	Peak Activity, $\mu\text{Ci/ml}$ ($\times 10^3$)	Approximate Time of Peak after Pump Shutdown, hr
0.01	69.3	0.065	21
0.02	34.6	0.12	20
0.04	17.3	0.22	19
0.06	11.5	0.30	18
0.09	7.7	0.39	17
0.12	5.8	0.46	17
0.20	3.47	0.58	15
0.40	1.73	0.73	14
1.0	0.69	0.85	12
Instantaneous		0.94	11

Fig. 4. Calculated Surges in ^{135}Xe Activity, Assuming the 10 Leak Rates Listed in Table V. ANL Neg. No. 103-O5820 Rev. 1.

C. Irradiation and Concurrent Observations

The irradiation history of the bottom-leaker experimental subassembly (C-2250S) is given in Table VI. It was known from the ^{135}Xe data taken during and after the shutdown on January 10, 1970, that a "weak" bottom leaker was already present in the core. This is called X in Table VII, which tabulates surge data for the period of this experiment. On the basis of observations of C-2008, an earlier, inadvertent bottom leaker,¹¹ it was concluded that the experimental signal would be sufficiently large that the experiment could be carried out despite the competing signal. In a reactor such as EBR-II, operational requirements are paramount and ideal experimental circumstances are never approached.

Shortly after the first irradiation of the bottom leaker began on January 14, there was an unscheduled reactor shutdown, and the pumps were later shut off. This resulted in a small, but analyzable, surge in the concentration of ^{135}Xe in the cover gas. Virtually the entire analysis of this experiment is based on ^{135}Xe activity; discussion of the other activities is deferred until Section D below.

When the reactor started up again on January 16 (run 39B), the concentration of ^{135}Xe in the cover gas approached an equilibrium value of

about 8×10^{-3} $\mu\text{Ci}/\text{ml}$, as compared with 3.0×10^{-3} $\mu\text{Ci}/\text{ml}$ for the normal tramp level. This high activity during operation was entirely unexpected, because C-2008 (the inadvertent bottom leaker) had given a detectable signal only after pump shutdown. The ^{135}Xe data for the initial experimental irradiation of C-2250S are shown in Fig. 5. As shown by the solid lines, it

TABLE VI. Irradiation History of Bottom-testing Experiment

1. Subassembly No. C-2250S

Burnup: Maximum 0.6421 Average 0.5528

2. Irradiation History:

	Date to Grid	Grid Position	Date from Grid	Hours in Position	MWd in Position	Burnup		Maximum Reactor Power	Hours at Power
						Maximum	Average		
a.	1-11-70	3B1	1-19-70	192	285	0.0608	0.0526	50	83
b.	2-10-70	1A1	3-1-70	456	924	0.3142	0.2795	50	441.5
c.	3-1-70	5C4	3-27-70	624	961	0.2671	0.2207	50	455.5

3. Residence Time:

Storage Basket, hr		Primary Tank	
To Basket 12-23-69			
Before Grid Installation	456	Total Hours	2832
After Item a above	528	Hours after Reactor Shutdown	624
After Item b above	0		
After Item c above	576		
Total:	1560		

4. Total Reactor Operating Time While SA in Grid 1042 hours

5. Reactor Operating Data:

a. Maximum Power Level of Reactor 50 MW

b. Total Integrated Power of Reactor: Date 3-27-70 MWd 33,739

6. Subassembly Operating Data:

	Grid Position	Average Power	Maximum Temperature, °F	Hours at Maximum Temperature
a.	3B1	0.746	851	83
b.	1A1	0.794	841	441.5
c.	5C4	0.603	901	455.5

TABLE VII. Summary of Surges Observed in ^{135}Xe Concentration

Reactor Run	Date	Leaker(s)	Core Position	Approximate Time of Peak after Pump Shutdown, hr	Peak Intensity, $\mu\text{Ci}/\text{ml}$ ($\times 10^3$)	Peak Intensity Corrected to Saturation, $\mu\text{Ci}/\text{ml}$ ($\times 10^3$)	Time from Reactor Shutdown to Pump Shutdown, hr	Time Pumps Were Off, hr	Estimated Peak Intensity Corrected for Peak Delay, $\mu\text{Ci}/\text{ml}$ ($\times 10^3$)
39A	Jan 10, 1970	X	?	30	0.35	0.4 ± 0.15	-0	84	
39B	Jan 15, 1970	C-2250S (+ X)	Row 3 (7)	14	0.75	3.0 ± 0.8	12	4	3.1 ± 1.0
39B	Jan 20, 1970	C-2250S (+ X)	Row 3 (7)	28	4.1	4.1 ± 0.5	1	10	5.0 ± 1.2
39C	Jan 30, 1970	X	?	29	0.4	0.4 ± 0.15	2	33	
40A	Feb 10, 1970	X	?	20	0.4	0.4 ± 0.1	-0	12	
40B	Mar 1, 1970	C-2250S (+ X)	Row 1 (7)	24	2.0	2.0 ± 1.0^a $- 0.5$	-0	84	b
41A	Mar 15, 1970	C-2250S (+ X)	Row 5 (7)	15	2.2	2.2 ± 0.2	-0	4	2.3 ± 0.4
41A	Mar 27, 1970	C-2250S (+ X)	Row 5 (7)	roughly 34	0.5	0.5 ± 0.2	1	48	
32C	Dec 25, 1968	C-2008	Row 2	21	2.2	2.2 ± 0.4	2	12	2.75 ± 0.5
33B	Apr 18, 1969	C-2008	Row 3	16	2.5	2.5 ± 0.3	-0	7	2.7 ± 0.5
33B	Apr 23, 1969	C-2008	Row 3	28	1.4	1.9 ± 0.5	-0	63	2.3 ± 0.6

^aSurge data compromised by inadvertent dilution of cover gas.

^bIt is not feasible to make this estimate, because the pumps were off so long. The ^{135}Xe could not have diffused to the cover gas readily enough for comparison with those cases in which the pumps were turned on before the peak occurred.

Fig. 5. ^{135}Xe Activity in the Cover Gas and Reactor Power History during the Initial Irradiation of C-2250S, in Core Position 3B1. ANL Neg. No. 103-O5817 Rev. 1.

was possible to analyze the ^{135}Xe activity to show a contribution from a steady-state leaker having delayed-release characteristics. This technique, explained in Ref. 12, was developed to analyze the case of the inadvertent top leaker L-462. The anomalous level suggested that a top leaker had inadvertently occurred among the other fuel elements in the experimental subassembly. At the end of run 39B, a large surge in ^{135}Xe activity was observed, as had been anticipated for the bottom-leaking experiment (see Fig. 6).

For run 40A, the experimental subassembly was placed in the basket (fuel storage inside the primary tank) and the ^{135}Xe activity was observed to determine if the source of both signals (steady-state and surge) had been removed with the experimental subassembly. The ^{135}Xe activity during run 40A returned to the normal "tramp" level of about $3 \times 10^{-3} \mu\text{Ci/ml}$ (see Fig. 7). At the end of run 40A, the small, delayed surge associated with the inadvertent leaker X was again observed.

For run 40B, the experimental subassembly was placed in row 1 (center of core) and the ^{135}Xe signal again was high. The data at shutdown from run 40B were compromised by inadvertent dilution of the cover gas. A crude estimate of the dilution was made from the data for ^{133}Xe , but a large uncertainty must be understood.

The final irradiation of the experimental subassembly was in row 5 during run 41A. Again there was high steady-state activity. At the shutdown of March 15, a well-defined surge was observed. However, after the subassembly had been irradiated another few days in the same position, the reactor was again shut down (March 27), and on this occasion the only surge signal was small and delayed as had been observed to characterize the unknown leaker X.

At this point, we decided to terminate the experiment in the expectation that the postirradiation examination would explain the anomalous steady-state signal during operation and the absence of a significant surge signal at the end of run 41A.

D. Consolidation of Observation

The surge data obtained from experimental subassembly C-2250S and from the unknown leaker X are consolidated in Table VII. Also included in Table VII are the data from the surge releases emanating from the inadvertent bottom leaker C-2008. Figure 8 compares the experimental observations from Table VII with the surges predicted in Table IV. Before discussing this figure, we must comment on Fig. 9, in which the time from turning the pump off to the occurrence of the surge peak is plotted against the time that the pumps were off. With instantaneous expulsion of ^{135}I and immediate diffusion of ^{135}Xe to the cover gas, the peak would occur about

Fig. 6. ^{135}Xe Activity in the Cover Gas at the End of Run 39B; C-2250S in Position 3B1. ANL Neg. No. 103-O5818.

Fig. 7. ^{135}Xe Activity in the Cover Gas during Runs 40A (no leak) and 40B (C-2250S in position 1A1). ANL Neg. No. 103-05816.

Fig. 8. Comparison of Experimentally Observed ^{135}Xe Surges with Calculated Values. ANL Neg. No. 103-N5372 Rev. 1.

Fig. 9. Plot of Time from Turning Pumps Off to Arrival of Surge Peak vs Time Pumps Were Off. ANL Neg. No. 103-N5373.

11.3 hr after the pumps were turned off. This implies that ^{135}Xe diffusion to the cover gas is rapid when the pumps are on and quite slow when the pumps are off. It also justifies, in Table VII, analyzing the signal from C-2250S without correction for leaker X, since the surge from X consistently occurs so late as not to reinforce the experimental signal.

Returning to Fig. 8, we see that the rather crude model gives predictions in fair agreement with the surges actually observed. Reality should

lie somewhere between the two lines, since the annular sodium, while not containing all the ^{135}I , must necessarily be "richer" than the plenum because of the narrow (6-mil average) diffusion channel in the annulus. Two of the surges observed with the experimental leaker (C-2250S) are clearly larger than had been predicted. Despite the rather approximate data available, these high values may be both real and reasonable. One should bear in mind that the predictions were based on the expulsion of ^{135}I only. However, in these two cases--January 20 (row 3) and March 15 (row 5)--the pumps were turned off rather quickly after the reactor was shut down. The hole in C-2250S was known to be relatively large, which would permit rapid expulsion when pump pressure ceased. This expulsion so soon after 50-MWt operation would necessarily sweep out not only ^{135}I but also substantial quantities of ^{135}Xe that had not yet diffused upward to the fuel-element plenum. Supporting this hypothesis is the fact that the third surge plotted for C-2250S was delayed because the pumps remained in operation several hours after the reactor was shut down. In support, the leak in C-2008 is believed to have been sufficiently small to allow time for ^{135}Xe to diffuse upward into the plenum instead of being swept out into the primary sodium.

Before the results of the postirradiation examination are reported, it is appropriate to discuss more broadly the data taken during irradiation. All the results so far mentioned have been based on the ^{135}Xe activity in the cover gas as determined by counting discrete "grab" samples. The RCGM data on ^{135}Xe agree well with the grab-sample data, but lack precision (see Fig. 10). Grab-sample data are available for ^{133}Xe , but the half-lives of ^{133}I - ^{133}Xe are too long to be of much use on the time scale associated with EBR-II operation.* RCGM data on ^{133}Xe are only qualitative, since various isotopes of higher gamma energy contribute, through the Compton effect, counts in the ^{133}Xe channel.

Fig. 10. Comparison of ^{135}Xe Data from Grab Samples and from RCGM Using Delay Line with ~ 230 -sec Sample-gas Age. ANL Neg. No. 103-O5815.

*The appendix lists the isotopes (together with their characteristics) that either directly or indirectly enter into the problem of fission-product monitoring in EBR-II.

Data on the concentration of ^{131}I in the primary sodium (see Fig. 11) are spotty because of occasional malfunctions in the sampling equipment. At best, samples were taken rather infrequently to limit exposure of personnel. A further difficulty is that the operation of the cold trap has a marked effect on the equilibrium concentration. The effect of the leaker and of cold-trap operation are apparent, although no quantitative conclusion is possible.

Fig. 11. Concentration of ^{131}I in Primary Sodium during the Period Covered by These Experiments. ANL Neg. No. 103-N5396.

E. Results of Postirradiation Examination

The experimental leaker (element 46 of subassembly C-2250S) and its two normal controls (elements 45 and 47) were subjected to a detailed post-irradiation examination together with element 47 subassembly C-2008,*

*The search for and identification of this leaker are reported in Ref. 9. However, the faulty fuel element had been retained in the vault. Its inclusion in the postirradiation examination was considered advantageous since it was thought to be fairly similar to the experimental leaker in C-2250S.

which had been identified as the leaker in that subassembly. In addition, because of the suspicion that an inadvertent top leaker had occurred in C-2250S, the other 88 fuel elements were scrutinized to determine if one of them had been the source of the steady-state signal.

Subassembly C-2250S was subjected to a standard sodium-removal operation in the interbuilding coffin (IBC) before it was examined in the air cell of the Fuels and Examination Facility. The recirculating argon from the IBC was sampled and analyzed for fission products just before the sodium-removal operation. Only the nominal background activity was found in the gas. The effluent rinse water from the sodium-removal operation was also sampled and analyzed for fission products, with negative results.

The subassembly was transferred from the IBC to the air cell, and the following postirradiation examinations were performed: (1) visual examination of the elements during subassembly dismantling, (2) eddy-current bond testing of all the elements, (3) stereomacroscopic examination of the leaker and control elements, (4) neutron radiography of six elements including the leaker and control elements, (5) leak-testing of 90 elements from C-2250S, (6) weighing of leaker and control elements to the nearest 0.1 g, (7) diameter surveys of element 46 from C-2250S and the control elements, (8) fission-gas sampling and analysis of the leaker and control elements, (9) leak-testing of the spade ends of the leaker elements, and (10) metallographic examination of samples from the leaker elements.

The results were as follows:

1. Visual examination during the dismantling operation revealed the presence of sodium-reaction residue (the sodium-reaction product left after the sodium-removal operation) on the lower end of element 46. A small amount of the residue was observed on elements adjacent to element 46, but no other abnormal features were observed on any of the elements.
2. Eddy-current bond testing of the elements revealed changes in the sodium content in the two leaker elements only. Elements 30 and 50 from C-2250S also exhibited bond-test data slightly different from the other elements, but the variation was due to the formation of a gas bubble under the restrainer that displaced the sodium, causing a higher-than-normal postirradiation sodium level. The bond-test results indicated that there was no sodium in the leaker elements above the fuel pin, except possibly for a small amount on the restrainer knob. The bond-test traces illustrating this feature are shown in Figs. 12 and 13.
3. Examination of the leaker and control elements employing the stereomacroscopic revealed no abnormal conditions on the element cladding, such as heat tinting, surface defects, or stains, except at the lower-end-fitting (spade) weld on element 47 from C-2008. The apparent defect in the lower weld of element 47 is shown in Fig. 14.

Fig. 12. Eddy-current Bond-test Traces of Elements 46 and 47 from C-2250S (showing the pre- and postirradiation condition of the bond sodium in element 46 and the postirradiation condition in element 47)

Fig. 13. Eddy-current Bond-test Traces of Elements 45 and 47 from C-2008 (showing the pre- and postirradiation condition of the bond sodium in element 47 and the postirradiation condition in element 45)

SM6A1

Fig. 14. Photo of the Apparent Defect in the Lower-end-fitting Weld on Element 47 from C-2008. Mag. ~15X.

top of the elements is shown in Fig. 15. (The sodium features above the fuel pin are visible in the original negative.)

Fig. 15. Neutron Radiograph of Top of Elements 47, 50, 46, 39, and 45 from C-2250S and Element 47 from C-2008. Arrows show sodium level in nonleaker elements.

5. A total of 90 elements from C-2250S (all except element 46) were subjected to a fission-gas leak test. Each element was placed in a chamber, the chamber purged with argon, and the element heated to $850 \pm 50^\circ\text{F}$. The element was held at temperature for 30 min before it was opened to an evacuated sample bulb. After the pressure in the chamber and the bulb had equalized, the bulb was removed and the gas analyzed for fission products. No fission products were found in any of the bulbs, indicating that none of the elements had leaked.

4. Neutron radiographs were made of the leaker and control elements as well as elements 39 and 50. The radiographs showed no length changes in the fuel pins from C-2250S, but a lengthening of 0.1 in. was shown in element 47 from C-2008. This increase in length is normal for the burnup experienced by C-2008. The radiographs revealed a gas bubble just below the restrainer of elements 39 and 50 and a lack of sodium above the fuel pin in the leaker elements. The hole drilled in the spade of element 46 also could be observed in the radiograph. A radiograph of the

6. The leaker and control elements were weighed on a remote-readout electronic balance with a reported accuracy of ± 0.1 g. The pre- and postirradiation weights of the elements from C-2250S are given in Table VIII. These data indicate that element 46 from C-2250S lost approximately 0.5 g during its residence in the reactor. No estimate can be made as to the weight change of element 47 from C-2008 as no preirradiation weight of the element is available.

TABLE VIII. Weights of Suspect and Control Elements

Subassembly	Element Number	Postirradiation Weight, g	Preirradiation Weight, g	Weight Change, g
C-2250S	45	83.6	83.6	0.0
C-2250S	46	83.3	83.8	-0.5
C-2250S	47	83.6	83.4	+0.2
C-2008	47	83.3		

7. Diameter surveys were performed on the leaker and control elements from C-2250S using an optical gauge. Pre- and postirradiation diameter measurements for these elements are listed in the appendix. A small bulge was noted near the bottom of element 46, but no other diameter changes were noted. No apparent cause of the bulge has been determined.

8. The pressure and volume of the gas plenum were measured in the leaker and control elements, and the gas was sampled. Table IX gives the pressure and volume data. These data were derived by puncturing the element and letting the gas expand into a surrounding chamber while monitoring the chamber pressure with a transducer, then performing another monitored expansion from this chamber into a second chamber of known volume. The gas was then sampled by drawing it into an evacuated sample cylinder. Analysis of the gas showed that it contained argon, helium, and xenon, with trace amounts of oxygen and nitrogen. Helium had been used as a carrier gas in making the pressure and volume determinations, and the elements had been loaded in an argon atmosphere. The amount of gas collected was too small to make a quantitative determination of the composition of the gas in the element plenum.

TABLE IX. Pressure and Volume Data for Elements from C-2250S and C-2008

Subassembly	Element Number	Gas Pressure in Plenum, psia	Volume of Plenum, cc
C-2250S	45	13.5	0.573
C-2250S	46	10.0	1.182
C-2250S	47	11.9	0.538
C-2008	47	8.5	0.740

The pressure and volume data do indicate that sodium was lost from the leaker elements. The nominal gas-plenum volume of an "as-built"

element is 0.66 cc. After irradiation, both element 46 from C-2250S and element 47 from C-2008 had larger than the nominal "as-built" gas-plenum volume. The less than "as-built" gas-plenum volumes of the control elements are most likely an indication that some swelling occurred in the fuel pins in these elements. If the fuel pin in element 46 is assumed to have swollen the same amount as the pins in the control elements, the plenum-volume data indicate that 0.6 ± 0.1 cc of sodium was lost from element 46. The nominal volume of sodium loaded into a Mark-IA element is 0.75 cc. These data indicate that unbonding of the fuel occurred to an appreciable extent, but this unbonding would have occurred, as discussed earlier, only when the primary pumps were turned off. Considerable unbonding must have taken place when the subassembly was lifted above the sodium in the primary tank as it was being unloaded from the reactor. Almost certainly, the element would have been fully bonded during operation. It was not possible to determine accurately the amount of sodium lost from element 47 from C-2008, but the postirradiation bond trace and plenum pressure suggest an amount similar to that lost from element 46 in C-2250S.

9. The leaker elements were dissected for examination. The fuel was removed from the bottom 3 in. of the element by heating the section to melt the sodium and shaking the pin out of the cladding. The fact that the pin was removed from the cladding with ease indicates that the fuel pin had not swollen out tightly against the cladding.

The 3-in.-long sections of cladding, including the spade, were transferred to low-level caves, cleaned in deionized water, and pressurized at room temperature to check them for leaks. Neither of the sections leaked, so they were cleaned in hot acetic acid and pressure-tested again. Element 46 from C-2250S exhibited a leak of ~85 cc/min at a differential pressure of 10 psi of helium. No leak was detected in the cladding section from element 47 from C-2008. The photos in Figs. 16-18 show the defective weld at the lower-end fitting of this element and the defect in the weld where the leak probably occurred. The photos in Fig. 16 compare a normal weld with the weld made on element 47. The photo in Fig. 17 shows a defect at the outer surface of the weld. The photos in Fig. 18 show the progression of this defect to the inside of the jacket. These photos indicate that the leak path was tortuous and very small. The opening at the ID of the cladding was probably no larger than 3×10^{-4} in. in diameter. This leak could easily have been plugged by slag during pre-irradiation helium-leak tests and by sodium after irradiation.

10. Sections of the leaker elements for metallographic examination were cut 4, 7, 10, and 13 in. from the bottom of the fuel pins. These samples were mounted in epoxy resin and prepared by using standard metallographic procedures of grinding and polishing. During the early stages of sample preparation, the sodium in the annulus between the fuel pin and the cladding was lost. The epoxy mounting resin was drawn into the annulus by using vacuum impregnation. This technique maintains the general spacing between the fuel and the cladding, although some movement of the fuel in cladding may occur.

63C5-5

Normal Weld

Fig. 16. Photos Comparing the Weld on Element 47 from C-2008 and a Normal Lower-end-fitting Weld. Mag. 15X.

63C5-1

Fig. 17

Micrograph Showing the Beginning of the Defect in the Weld at the OD of the Cladding. Mag. 50X.

63C5-2

63C5-3

63C5-4

63C5-5

Fig. 18. Series of Micrographs Showing the Progression of the Defect to the ID of the Cladding. Approximately 0.0002 in. of material was removed from the sample before each succeeding micrograph. Mag. 250X.

Figures 19 and 20 show cross sections of the two elements. These cross sections show that both fuel pins retained their original round shape

and that no unusual swelling or slumping of the fuel had occurred. (The cracks shown in some of the photos are casting defects.) Figures 21 and 22 show the microstructure of fuel-pin samples from element 46 from C-2250S. These microstructures are typical for an element irradiated to less than 1 at. % burnup, as shown by comparing them to the microstructure of a standard element such as element 31 from subassembly C-2166 at 0.8 at. % burnup (Fig. 23). These microstructures indicate that element 46 from C-2250S lost none of its heat-transfer capability, even though it lost a significant amount of its sodium

63C1-6

13 in. above the Bottom

63C2-4

10 in. above the Bottom

63C3-2

7 in. above the Bottom

63C4-5

4 in. above the Bottom

Fig. 19. Photos Showing Cross Sections of Element 47 from C-2008. Mag ~16X.

64C1-5

13 in. above the Bottom

64C2-4

10 in. above the Bottom

64C3-4

7 in. above the Bottom

64C4-4

4 in. above the Bottom

Fig. 20. Photos Showing Cross Sections of Element 46 from C-2250S. Mag. ~16X.

64C1-3

Near the Top of the Pin

64C3-1

7 in. above the Bottom of the Pin

64C4-1

4 in. above the Bottom of the Pin

Fig. 21. Microstructures of Typical Areas near the Centerline of the Fuel Pin from Element 46 from C-2250S.
Mag. 600X.

64C1-2

Near the Top of the Pin

64C3-2

7 in. above the Bottom of the Pin

64C4-2

4 in. above the Bottom of the Pin

Fig. 22. Microstructures of Typical Areas near the Edge of the Fuel Pin from Element 46 from C-2250S.
Mag. 600X.

151A1-1

Near the Top of the Pin

151A2-1 7 in. above the Bottom of the Pin

151A3-1 3 in. above the Bottom of the Pin

**Fig. 23. Microstructures of Typical Areas near the Centerline of the Fuel Pin from Element 32 from C-2166.
Mag. 500X.**

VI. DISCUSSION AND CONCLUSIONS

Although these were preliminary experiments, several points can be educed. The lack of results from the top leaker means that the experiment should be repeated using larger and better-defined leaks. The present experience suggests that acceptance criteria for driver fuel are unnecessarily stringent.

The bottom leaker was more successful. It demonstrated that a leak could be simulated. During the early part of the experiment, it generated pump-off surges in cover-gas activity much as had been predicted by the model. At the same time it raised two questions. First, by what mechanism did the bottom leak cause a significant steady-state signal during operation? And, second, why did it quit generating surges after several cycles of operation?

One might reasonably conjecture that the steady-state signal simply represents a pulsative transport of bond sodium back and forth through the orifice, under minute variations in pump pressure. Since the orifice in the experimental leaker was two orders of magnitude greater in cross-sectional area than the leak in C-2008, it is possible that the effect might be observed in the former and not the latter. Clearly this is something to be clarified in future experiments using smaller leaks. The reduction in the pump-off surges is puzzling. No convincing hypothesis has been offered, but this effect might have been the result of some change in the fuel element, such as a loss of gas by channeling through the sodium bond.

Further experiments are planned with different-sized leaks. A high-resolution gamma spectrometer will be in operation for future experiments, and much more data are expected; in particular, data on ^{85m}Kr are expected to be especially useful, since its response to changes in leakage and power is much more prompt than that of ^{135}Xe .

Ultimately, we hope to introduce leakers containing Pu or ^{233}U . It appears possible to distinguish between various nuclear fuels from data on the "spectrum" of fission products.

APPENDIX

Summary of Isotopes Pertinent to the EBR-II
Fission-product-monitoring Systems

Isotope	Half-life	Predominant Gamma Energy, MeV	Relevant Monitoring System
^{85}Br	3.0 min	(β emitter)	RCGM
$^{85\text{m}}\text{Kr}$	4.4 hr	0.150	
^{88}Br	16 sec	(β emitter)	
^{88}Kr	2.80 hr	0.191	FGM
^{88}Rb	17.8 min	1.85	
^{89}Br	4.5 sec	(β emitter)	
^{89}Kr	3.2 min	(β emitter)	FGM
^{89}Rb	15.4 min	1.05	
^{131}I	8.05 days	0.36	Sodium sample
^{133}I	21 hr	(β emitter)	Grab sample; RCGM
^{133}Xe	5.27 days	0.081	
^{135}I	6.7 hr	Many gamma lines	
^{135}Xe	9.2 hr	0.250	Grab sample; RCGM
^{138}I	5.9 sec	(β emitter)	FGM
^{138}Xe	17 min	(β emitter)	
^{138}Cs	32.2 min	1.426	
^{23}Ne	38 sec	0.44	RCGM

ACKNOWLEDGMENTS

We are grateful to a number of associates for cooperation and helpful discussions. Special thanks are due to E. R. Ebersole, whose group provided the grab-sample data; to N. R. Grant and D. L. Mitchell, who prepared the experimental subassembly; and to the operating staff of EBR-II, who wholeheartedly supported the work.

REFERENCES

1. E. R. Ebersole and R. Villarreal, *Diagnostic Radiochemistry Following the Recent EBR-II Fission-product Release*, Trans. Am. Nucl. Soc 10(2), 634-635 (Nov 1967).
2. *Reactor Development Program Progress Report, August 1969*, ANL-7606, pp. 62-63 (Sept 26, 1969); *Reactor Development Program Progress Report, September 1969*, ANL-7618, pp. 66-69 (Oct 27, 1969); *Reactor Development Program Progress Report, November 1969*, ANL-7640, pp. 73-75 (Dec 31, 1969).
3. F. S. Kirn, *EBR-II as a Fast Reactor Irradiation Facility*, Nucl. News 13(3), 62-68 (Mar 1970).
4. L. J. Koch et al., *Hazard Summary Report, Experimental Breeder Reactor II (EBR-II)*, ANL-5719 (May 1957).
5. L. J. Koch, W. B. Loewenstein, and H. O. Monson, *Addendum to Hazard Summary Report, Experimental Breeder Reactor II (EBR-II)*, ANL-5719 (Addendum) (June 1962).
6. K. G. Porges, "EBR-II Fuel Element Failure Detector," *Reactor Physics Division Annual Report: July 1, 1963 to June 30, 1964*, ANL-7010, p. 231 (Jan 1965); K. G. Porges, "Design Improvements and Sensitivity Tests of the FERD System for EBR-II," *Reactor Physics Division Annual Report: July 1, 1964 to June 30, 1965*, ANL-7110, pp. 294-295 (Dec 1965).
7. R. R. Smith and C. B. Doe, *Fission Product Monitoring in EBR-I, Mark IV*, ANL-6783 (Jan 1964).
8. G. S. Brunson, *On-line Noble Gas Fission-product Monitoring in Experimental Breeder Reactor II (EBR-II)*, Nucl. Technol. 10, 33-43 (Jan 1971).
9. R. R. Smith and C. B. Doe, *Cladding Failure Simulation Tests in EBR-II*, ANL-7067 (Dec 1966).
10. R. R. Smith, C. B. Doe, and E. R. Ebersole, *Exposed-fuel Calibration Study in EBR-II, Second Series*, ANL-7558 (Jan 1970).
11. R. M. Fryer, R. R. Smith, E. R. Ebersole, and R. V. Strain, *Symptoms and Detection of a Fission-product Release from an EBR-II Fuel Element: Case II. Defect below Fuel Elevation*, ANL-7676 (June 1970).
12. R. M. Fryer, E. R. Ebersole, P. B. Henault, and R. R. Smith, *Symptoms and Detection of a Fission-product Release from an EBR-II Fuel Element: Case I. Defect above Fuel Elevation*, ANL-7605 (Jan 1970).