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DEPOSITION OF CESIUM AND BARIUM
IN A
SODIUM - STAINLESS STEEL SYSTEM

AEC Research and Development Report

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By
J. GUON

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CONTENTS

	Page
Abstract	5
I. Introduction	7
II. Program Description	9
III. Facility Description	13
IV. Cesium Tests	18
A. Background	18
B. Experimental	18
C. Data Reduction	19
D. Test Results	21
1. Characterization Tests	21
2. Deposition Tests	23
3. Sample Station Tests	27
4. Draining and Flushing	28
E. Discussion of Cesium Results	28
1. Comparison with Other Work on Cesium Deposition	28
2. Cesium Deposition Model and Reactor Calculational Method	30
V. Barium Tests	34
A. Background	34
B. Experimental	34
C. Data Reduction	36
D. Test Results	37
VI. Conclusions	40
References	42
Appendices	
I. Reduction of Cesium Experimental Data	43
II. Cesium Deposition Test Data: Isothermal, Nonisothermal, and Kinetic	49
III. Calculation of Cesium Distribution in a Reactor System, Using the Equilibrium Constant (K) Approach	61
IV. Derivation of Analytical Expressions for Isothermal and Nonisothermal FPBL Operation, Barium Experiments . .	67

TABLES

	Page
1. Analogue Grouping and Expected Fate of Various Radioisotopes	10
2. Representative Radioisotopes for FPBL Experiments	10
3. Design Parameters for Fission Product Behavior Loop	13
4. Sequence of Cesium Experiments	20
5. Maximum Temperature Experienced by FPBL During Characterization Tests	23
6. Sample Station Drain Test Results	29
7. Parametric Test Series for Barium Deposition Tests	35
8. Intercomparison of Results from First Barium Series	38
II-1. Results of First Isothermal Test	49
II-2. Results of Second Isothermal Test	50
II-3. Results of Kinetic Test	52
II-4. Results of First Nonisothermal Test	53
II-5. Results of Third Isothermal Test	54
II-6. Results of Fourth Isothermal Test	56
II-7. Results of Second Nonisothermal Test	57
II-8. Results of Fifth Isothermal Test	58
II-9. Results of Third Nonisothermal Test	59

FIGURES

1. Fission Product Behavior Loop Schematic Drawing	14
2. Test Section Schematic Drawing	15
3. Fission Product Behavior Loop	16
4. Results of First Isothermal Test, 0.7-wppm Cs	22
5. Log K vs 1/T for 0.7-wppm Cs Isothermal Tests	24
6. Log K vs 1/T for 6-wppm Cs Isothermal Tests	24
7. Effect of Oxygen on K, Result from Sample Station Coring Tests (Circulation temperature = 500°F, samples quenched)	26
8. Mathematical Model of FPBL	36
9. Log K vs 1/T for Barium Deposition Tests [K(700°F) $\cong$ 500 cm]	39

ABSTRACT

Quantitative results have been obtained on the deposition of cesium and barium in a sodium - stainless steel system. For the first time, sufficient detail has been obtained to predict, with some confidence, the distribution of fission product cesium and barium released to a Liquid Metal Fast Breeder Reactor (LMFBR) system as a result of fuel cladding failure. Some insight has been developed into the processes occurring during deposition.

1

2

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I. INTRODUCTION

An economical Liquid Metal Fast Breeder Reactor (LMFBR) must operate at high temperatures, with high neutron fluxes, high heat fluxes, and high coolant flow rates. This combination of operating conditions will unquestionably lead to the release into the coolant of activated structural material through corrosion, and of fission products from fuel cladding failures. In time, the ability to maintain an LMFBR may depend upon the eventual deposition sites of these radioactive products in the primary coolant system. Because of the importance of knowing, or at least being able to intelligently predict, the fate of these radioactive products within the reactor system, Atomics International (AI) is carrying out a continuing study of the behavior of various radioisotopes in a sodium - stainless steel environment.

In order to predict the deposition sites of these radioactive materials, one must not only identify the physical and chemical processes involved in their transport, but must also develop kinetic data on both the short- and long-term disposition of the radioisotopes. Once the processes are known, and the kinetic data are available, mathematical coupling equations may be formulated to predict radioisotope distributions, not only in a particular test system, but also in a variety of other systems as well. In this way, it will be possible to include the effects of radioisotope deposition in plant optimization studies. In addition, once the deposition processes are understood, methods can be developed to control deposition and/or to decontaminate components.

The AI study of the behavior of radioisotopes in a sodium - stainless steel system includes three major areas: (1) Source Term Characterization (the focus of the AI effort), which involves the development of a synthesized mathematical code to compute the distribution, within a LMFBR system, of radioactivity originating from both fission products and activated corrosion products,⁽¹⁾ (2) Experimental Studies, in which experimental measurements are made of the deposition properties of a few selected fission and corrosion products, and (3) System Studies, whose goal is to apply the results of the other work to the evaluation of the radioactivity distribution in LMFBR systems under design.

The desired end product of the current program is the development of a computer code which will calculate, to the required accuracy, the radiation levels associated with any component in a LMFBFR system, as a function of time and operating condition. At present, simple, and as yet experimentally unverified, deposition models are assumed in this program, because of the almost complete absence of information on radioisotope behavior in a liquid metal system. Prior to the current experimental program, system parameters, such as solubilities, and equilibrium and rate constants used in the present version of the computer code (STP-1), were unknown, and there was no way of knowing whether predicted radiation levels were within orders of magnitude of what would occur in a real system. The experimental portion of this effort was directed toward the verification of the deposition models and the development of parameters required to define the system.

In order to obtain some of the needed experimental data, a well-characterized, forced-circulation sodium loop, the Fission Product Behavior Loop (FPBL), was designed, built, and operated. Basically, it is a simple loop, having two test sections in series with a pump and a surge tank, and side stream loops for chemical sampling, for adding materials to the main loop, and for cold trapping. The test program rationale and the loop design are described elsewhere.⁽²⁾ This report records the results of the test programs on the distribution and the rates of movement of cesium and barium in the FPBL. Both equilibrium partition data of cesium and barium between the liquid and the loop surfaces, and kinetic data on the movement of cesium and barium through the system were sought. A radioactive tracer technique was used, thereby permitting in-situ measurements. Data were obtained at two oxygen concentration levels in the sodium in the cesium tests, in order to determine the effect of oxygen on the transport parameters.

II. PROGRAM DESCRIPTION

The goal of this experimental work is to define the deposition behavior of radioisotopes which will be encountered in a LMFBR, in order to permit evaluation of the radiation hazard associated with plant operation and maintenance. In selecting the experimental approach to be used to define the deposition processes and to develop kinetic data, decisions were required, relative to the type of experiment to be performed and the source of fission product activity.

Because of the ability to measure deposition in-situ by gamma-ray counting, it was decided to use a forced circulation loop as the experimental facility, rather than to perform capsule experiments. Capsule experiments yield only one datum per capsule, but a loop with gamma-ray counting capability will rapidly and inexpensively yield considerable data relating deposition to such parameters as temperature and flow. Previous work in the Cold Trapping Apparatus⁽³⁾ (CTA) indicated that gamma-ray counting is a valid approach to the study of deposition; however, the sodium volume in the CTA (0.1 l) was too small to permit suitable control of the experimental variables. A loop containing ~ 2 l of sodium was considered optimum, being sufficiently large to overcome the problems encountered in the CTA and sufficiently small to provide the flexibility required for this program.

The most desirable source of fission products would be those extracted from failed fuel elements (or fuel material) by sodium of similar chemical composition, and operating at temperatures comparable to those planned for the reactor. The handling of high burnup fuel is difficult, because of the necessity for radiation shielding. Sophisticated counting techniques would be required to discriminate among the radioisotopes. Control of variables, such as concentration and sodium purity, could be difficult. Also, data interpretation could be an extremely difficult task, because of the wide and virtually uncontrolled variation in critical parameters. An alternate approach, and the one selected for the FPBL, is to dope the sodium with single elements containing a sufficient concentration of radioactive isotope to assure adequate counting statistics. For example, cesium containing radioisotope ^{134}Cs is used in lieu of fission product ^{137}Cs . Using this approach, fission product elements can be studied singly, circumventing most of the problems encountered when the source of

TABLE 1
ANALOGUE GROUPING AND EXPECTED FATE OF
OF VARIOUS RADIOISOTOPES

Analogue Grouping	Expected Fate in Stainless Steel System
Cr, Mn, Co, Fe	Isotopic exchange, precipitation, and/or diffusion into stainless steel
Cs, Rb, K	Volatization and/or surface adsorption
Ba, Sr	Deposition as oxides
Mo, Ru, Sb, Ag*	Diffusion into stainless steel
Ce, La, Nd, Y, Zr	Deposition as oxides or diffusion into stainless steel
Nb	Deposition as oxides or diffusion into stainless steel
H ³ , I, Te	Vaporization or diffusion into stainless steel

*May not be a successful analogue grouping. While the criteria used to group these four elements are satisfied, the differences in chemical properties in aqueous systems is such as to raise a serious question as to the similarity of behavior in sodium systems.

TABLE 2
REPRESENTATIVE RADIOISOTOPES
FOR FPBL EXPERIMENTS

Analogue Grouping	Radioisotope
Cr, Mn, Co, Fe	⁵¹ Cr, ⁵⁴ Mn, ⁶⁰ Co, ⁵⁹ Fe
Cs, Rb, K	¹³⁴ Cs
Ba, Sr	¹³³ Ba
Mo, Ru, Sb, Ag	¹⁰³ Ru, ¹²⁴ Sb
Ce, La, Nd, Y, Zr	¹⁴¹ Ce or ¹⁴⁴ Ce
Nb	¹⁸² Ta
³ H, I, Te	⁷⁵ Se

fission products is irradiated fuel. This approach, however, does raise the question as to whether the chemical form and deposition properties of the doped element are the same as for extracted fission products. It is believed, at least in the case of oxide fuel, that the chemical form of the fission products in reactor grade sodium will become similar, regardless of their source. Results from the FPBL will have to be verified by comparison with data from systems containing extracted fission products.

In order to reduce the analytical and experimental effort required for the prediction of radiation levels, the radioisotopes that will be present in the sodium have been grouped into seven categories (analogues), each category containing elements believed to have similar behavior in sodium. The ultimate fate of the radioisotopes may be to remain in solution in the sodium, vaporize into various gas spaces, deposit on system walls, or any combination of these. In the establishment of analogue groupings, the following factors were considered:

- 1) Do the isotopes form oxides that are stable, relative to sodium oxide?
- 2) Do the isotopes form intermetallic compounds with the major constituents of Type 304 stainless steel?
- 3) Are the isotopes volatile?
- 4) To what extent are the isotopes soluble in sodium?

Table 1 presents the present form of the analogue grouping.

Radioisotopes to be used to represent the seven analogue groups for the FPBL experiments were selected, based on the following considerations:

- 1) The radioisotopes were to be gamma emitters.
- 2) Radioisotopes were chosen that did not decay to gamma-emitting radioisotopes which might confuse data interpretation.
- 3) (n,γ) production reactions were preferred; no interfering radioisotopes being generated.
- 4) If no suitable radioisotope of the element was found, a suitable radioisotope was selected from a chemical analogue.

Table 2 lists the selected radioisotopes.

When designing the long-term program for the FPBL, it was decided to start with the best studied, and one of the important fission product elements observed in liquid metal reactors, cesium.⁽⁴⁾ The selection of barium as the second element to study was based on its observation as an important fission product in liquid metal systems and the relatively low gamma-ray energy of the radioisotope to be used in this test ($^{133}\text{Ba} - 0.36 \text{ Mev}$). The selection of the order for future tests will be based on the relative importance of the element to be studied, and, where practical, increasing gamma-ray energies. The selection of the order of isotopes to be studied is important to prevent Compton scattering from interfering with data reduction (in order to reduce experimental uncertainty and provide data on synergisms, it is desirable not to acid etch the FPBL between test series).

III. FACILITY DESCRIPTION

The facility used to study radioisotope distribution, the Fission Product Behavior Loop (FPBL), has been described in detail elsewhere.⁽²⁾ Design parameters for the loop are given in Table 3, and a schematic drawing is shown in Figure 1. The loop consists of, in the direction of sodium flow, a surge tank, the first test section, a dc Faraday electromagnetic pump, the second test section, and a dc Faraday electromagnetic flowmeter. The bypass leg consists of an off-line cold trap and three stations for material injection and chemical sampling. The loop is constructed entirely of Type 304 stainless steel, except for the valves, which are made of Type 316 stainless steel. The loop contains ~1800 cc of sodium; all but 180 cc is in the surge tank. Three NaI-Tl radiation detectors and single-channel analyzers are used to measure, in situ, the gamma-ray radiation from injected fission product elements — one monitoring each of the test sections, and one monitoring the surge tank. The test section detectors are mounted on hydraulic cylinders, permitting a scan to be made of virtually the entire length of each test section. A detailed drawing of the test sections is shown in Figure 2. Sodium flow in each test section is annular; the hollow center reduces axial heat transfer, and

TABLE 3
DESIGN PARAMETERS FOR FISSION
PRODUCT BEHAVIOR LOOP

Loop temperature (°F)	300 to 1200
Test section temperature* (°F)	300 to 1200
Sodium volume (cc)	~1800
Sodium flowrate (ft/sec)	
Loop tubing	0.15 to 1.5
Annular region of test section	0.15 to 1.5
Unpacked test section	0.02 to 0.2
Pump throat	0.3 to 3
Material of construction†	Type 304 stainless steel

*Independently variable

†Except valves, which are Type 316 stainless steel

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Figure 1. Fission Product Behavior Loop Schematic Drawing

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Figure 2. Test Section Schematic Drawing

Figure 3. Fission Product Behavior Loop

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permits installation of a number of thermocouples to monitor the sodium temperature. The inlet and outlet ends of the test sections contain integral heaters and coolers, permitting independent temperature operation of the central (isothermal) portion. The central portion is packed with Type 304 stainless steel mesh. The purpose of the mesh is to provide both a large surface-to-volume ratio and a minimum liquid phase diffusion distance. Figure 3 is a photograph of the loop installation.

IV. CESIUM TESTS

A. BACKGROUND

Cesium was the first of several fission product elements to be studied, because it has been observed in every sodium reactor where fuel elements have failed.⁽⁴⁾ Qualitative information from reactor operations, qualitative and semi-quantitative results from loop and loop-type experiments,^(3,5-7) and quantitative results from coupon experiments^(8,9) are in general agreement: cesium will transfer to and from stainless steel surfaces rapidly and reversibly with little hysteresis; the amount which will deposit on a surface decreases with increasing temperature, and deposition is probably enhanced by increased oxygen content. Prediction of the cesium distribution in a LMFBR is not possible from the preceding studies.

The purposes of the present work were: to provide sufficient insight into the cesium - sodium - stainless steel system to define the parameters that affect deposition, to develop quantitative information to allow prediction of cesium distribution in a LMFBR, and to formulate a deposition model. The first and third objectives have, at least in part, been achieved. The second objective, quantitative prediction of cesium deposition, is limited to the prediction of a realistic value for cesium deposition in the reactor piping and a reasonably good prediction (several factors) of cesium surfaces deposition in the cold trap.

B. EXPERIMENTAL

Elemental cesium, 99.99% pure, was used for these experiments. Approximately 0.2 gm was encapsulated in quartz, and irradiated to a ^{134}Cs specific activity of 150 mCi/gm Cs. The irradiated cesium was dissolved in thoroughly cold-trapped sodium in an inert atmosphere box, and several small extruders were filled with this (solid) stock solution (~1 wt% Cs in sodium). Cesium was added to the loop through one of the chemical sample stations, by filling a sample tube with a weighed amount of stock solution. The sample tube was pressurized to 50 psig with argon to prevent accidental injection of air into the loop, and to permit leak (bubble) testing of the tube fittings used to connect the sample tube to the sample station.

Oxygen was added to the loop through the surge tank:

- 1) A tube of known volume was pressurized to a predetermined pressure with oxygen
- 2) The tube, with isolation valves, was installed in one of the gas lines to the surge tank
- 3) The loop was evacuated to 15 in. Hg absolute, and back-filled through the tubing containing the oxygen with argon to 5 psig.

After each drain and flush, and prior to each cesium injection, the loop sodium was cold-trapped in the off-line cold trap for 18 to 24 hr at a temperature of 250 to 275°F. No operational problems were encountered during cold trapping, or during cesium or oxygen additions.

The three categories of runs in which in-situ data were obtained were the isothermal, kinetic, and nonisothermal runs. For isothermal runs, a minimum temperature gradient was maintained through the loop, in general <5°F. Temperatures were then varied stepwise. At the end of each day, the loop was returned to a baseline temperature (500°F for the first isothermal test, and 300°F for subsequent tests). The kinetic run was similar to the isothermal runs, except the loop was first operated at 1100°F, prior to each run at lower temperature. For nonisothermal runs, the loop was maintained at 700°F; one test section was then held at a lower temperature, and the temperature of the other test section was varied stepwise between the two. Sample station tests and chemical sampling procedures will be discussed later in the report.

Because cesium analysis in the sodium was done by radiochemical analysis for ^{134}Cs , it was necessary to determine the cesium present as impurity in the sodium. Using spark source mass spectrographic analysis (SSMS), cesium as an impurity of sodium was not detected (upper limit, 0.05 wppm). The enhancement factor for cesium over sodium was found to be 8. Table 4 is an outline of the test sequence.

C. DATA REDUCTION

Converting the radiation count rates from the three detectors to usable deposition parameters is by no means a trivial matter. One of the great worths of the FPBL as an experimental facility is the careful definition of system

TABLE 4
SEQUENCE OF CESIUM EXPERIMENTS

-
1. Cesium = 0 wppm, Oxygen <10 wppm
 Characterization Tests
 Chemical Samples 1, 2, 3
 2. Drain and Flush Loop with Clean Sodium
 3. Cesium = 0.7 wppm, Oxygen <10 wppm
 Chemical Sample 4
 First Isothermal
 Second Isothermal
 Kinetic
 Chemical Sample 5, 6
 First Nonisothermal
 4. Drain and Flush Loop with Clean Sodium
 5. Cesium = 6 wppm, Oxygen <10 wppm
 Chemical Sample No. 9
 Third Isothermal
 Fourth Isothermal
 Second Nonisothermal
 6. Cesium = 6 wppm, Oxygen = 25 wppm
 Fifth Isothermal
 Third Nonisothermal
 Chemical Samples 10, 11, 12
 7. Drain and Flush Loop with Clean Sodium
 Chemical Sample 13
 8. Cesium = 0.2 wppm, Oxygen <10 wppm
 Chemical Samples 14, 15, 16
 Sample Station Tests (by coring) – Chemical Samples 17 to 22,
 inclusive
 Sample Station Tests (by draining) – Chemical Samples 23 to 29,
 inclusive
 Chemical Sample 30
 9. Drain and Flush Loop with Clean Sodium. Cesium Tests Completed.
 Barium test series started.
 Chemical Sample 31
-

Sample No. 6 was a subsample of No. 5, and used for spark source mass spectrographic analysis. Sample 7 was a sample of sodium used to prepare the Cs-Na stock solution, and used as a standard for the SSMS analysis.

parameters (such as flow rates, volumes, and surface areas), and the resultant ease of mathematical modeling. Details of the analytical method used to reduce radiation data are reported in Appendix I.

Under the usual operating conditions, the FPBL is a closed system, so that, as cesium deposits on surfaces, its concentration in the liquid decreases. The surge tank has a small surface-to-volume ratio; therefore, it was assumed that the surge tank count rate is directly related to the cesium concentration in the sodium. (Neglecting surface deposition in the surge tank affects results by <20%.) The calculated relative cesium concentration in the sodium is used, along with test section count rate and surface-to-volume ratio, to calculate an "uncorrected" equilibrium constant for surface adsorption, K . K is defined as the ratio of cesium surface concentration (grams of cesium adsorbed per square centimeter) divided by the concentration of cesium in the sodium (grams of cesium per cubic centimeter). The calculated (and all reported) values of K are considered uncorrected, because one of the assumptions used in its development is that surface adsorption at 1100°F is negligible. K 's can be corrected for this assumption (see Appendix I-C); however, correction factors are within what is considered experimental accuracy (maximum factor of 2 for low values of K , and a maximum of a few percent for large values of K).

D. TEST RESULTS

1. Characterization Tests

Prior to the cesium injection into the FPBL, a number of tests were conducted to determine the loop's operating characteristics. Measured heat transfer characteristics were within engineering accuracy of the design calculations, and satisfied the design parameters listed in Table 3. Calibration of the flowmeter showed that the flow characteristics listed in Table 3 had been achieved. Thermocouples performed well; their response was rapid, and, judging from observed thermal arrests at 208 to 210°F (melting point of sodium), accurate. Stable operation of the loop was easily achieved, and temperature patterns and flow rates showed little drift over long periods of time (days). As was later found, in the cesium tests, deposition depends to a large extent on the temperature history of the stainless steel surfaces in contact with sodium. Table 5 presents the loop's maximum temperature and time of exposure prior to cesium injection.

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Figure 4. Results of First Isothermal Test, 0.7-wppm Cs

TABLE 5
MAXIMUM TEMPERATURE EXPERIENCED BY FPBL
DURING CHARACTERIZATION TESTS

Component	Temperature (°F)	Duration of Exposure (hr)
Test Section No. 1	1150 - 1200	1/2
Test Section No. 2	840	2
Loop, Excluding Test Sections	780	1-1/2

2. Deposition Tests

Figure 4 presents the results of the first cesium test. The numbers next to the data points indicate the order in which the data were obtained. After cesium injection and prior to this test, the loop had operated at 485°F for 260 hr. Because scalers had not as yet been installed, data were acquired from less accurate count rate meter records. "Loop, Excluding Test Sections" results are obtained from a cesium material balance. In subsequent runs, there was good correspondence between material balance and test section results. The loop was maintained at each temperature for a short time (see Table II-1), except for 16 hr (overnight) between Points 4 and 5 and 140 hr between Points 7 and 8. These results clearly show deactivation to occur after the high-temperature portion of the run, and, along with the information in Table 5 and from the Sample Station Tests (Section IV-D-3), indicate the importance of prior temperature history on cesium deposition.

The results from the cesium deposition tests are presented in tabular form in Appendix II. Additional narrative detail is also included.

An observation in common with all tests was that little or no change in deposition was measured after the first 1/2 hr (time required for a set of radiation readings). This indicates that the kinetics of deposition are rapid.

In order to express the results, an equilibrium constant (K) was defined, assuming the reaction:

Figure 5. Log K vs $1/T$ for 0.7-wppm Cs Isothermal Tests

Figure 6. Log K vs $1/T$ for 6-wppm Cs Isothermal Tests

If this "K" concept is analogous to chemical equilibria, an Arrhenius plot ($\log K$ vs $1/T$) should be a straight line. Figures 5 and 6 are such plots for data from the isothermal tests. Figure 5 shows the individual data points from the first isothermal test, and averaged selected data from other 0.7-wppm tests. These selected data were chosen based on minimal gain shift corrections, consistency of background counts, and consistency with mass balances. The scatter amongst points at the same temperature (at least 5 in number) is quite low; therefore, single-point representation is used, rather than a scatter bar. The diagonal line in this figure is obtained from a least squares fit of the average data point at each temperature. Figure 6 is a similar representation, except, because of the scatter amongst data, unaveraged selected data are presented, except at 300°F, where the data range is shown. The diagonal dashed line in Figure 6 was drawn parallel to the 0.7-wppm line. The horizontal dashed lines were drawn from the median 300°F value of K to the diagonal dashed line. When considering experimental accuracy, the fits to a straight line in Figures 5 and 6 are quite reasonable. The break in the Arrhenius plot will be discussed in Section IV-D-3.

The equations of the diagonal lines in Figure 5 are:

$$\log_{10} K = -3.41 + \frac{0.64 \times 10^3}{T} \quad \text{for deactivated surfaces} \quad \dots (1)$$

$$\log_{10} K = -6.45 + \frac{3.00 \times 10^3}{T} \quad \text{for "activated" surfaces} \quad \dots (2)$$

where:

K = equilibrium constant (cm)

T = absolute temperature (°K)

Based on the slopes of these lines, the heats of activation (ΔH) for "activated" and deactivated surfaces are -14 and -3 kcal/gm-mole, respectively. The latter value is consistent with ΔH 's associated with a physical adsorption process. The former value is too negative for physical adsorption, and suggests the possibility that two processes are involved in cesium depositions, the more energetic process being adversely affected by exposure of the stainless steel surfaces to high-temperature sodium.

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Figure 7. Effect of Oxygen on K, Result from Sample Station Coring Tests (Circulation temperature = 500° F, samples quenched)

3. Sample Station Tests

Two types of tests were conducted, using the sample stations, in an attempt to obtain corroborative results by independent methods. In one series, the sodium was circulated at 500°F through the sample stations for a period of time (usually 24 hr), the flow stopped, and the sample quenched as rapidly as practical. A portion of the sample was cored. The ^{134}Cs and sodium in both the sodium core and the remaining sodium and stainless steel were determined. From these analyses and the calculated (apparent) tube's internal surface area, a value of K was calculated. An attempt to correlate the values of K with surface preparation or cesium concentration in the sodium was unsuccessful. A plot (Figure 7) of K vs the oxygen concentration in the sodium (as determined by the amalgamation method) shows K decreasing as the oxygen concentration in sodium increases, when samples of the same circulation time (24 hr) are compared. The apparently low value of K calculated from Sample No. 9 (18-hr circulation) is not explained, the difference between 18- and 24-hr circulation times not being considered significant. It is not known whether the longer circulation times on Samples No. 8 (90 hr) and No. 30 (70 hr) affected the adsorbing capacity of the sample tubes. One possible explanation of the observed K dependence is that the increased sodium viscosity due to increased oxygen reduced cesium migration to the wall during the quench. However, when viewed with other evidence (the break in the lines in Figures 5 and 6, and the observation in the nonisothermal deposition tests that cesium deposition increases in the cold test section as the warmer test section temperature is decreased), it is believed that oxygen associated with the stainless steel surface decreases the ability of the surface to adsorb cesium. This conclusion is not inconsistent with the observation of Cooper⁽⁷⁾ that cesium can be associated with precipitated sodium oxide. Using the value obtained by Cooper, the gamma-ray count rates associated with cesium so deposited in the FPBL experiments would be at least an order of magnitude less than the gamma-ray count rate associated with surface-adsorbed cesium. This is one of the areas in which more work is needed.

A second set of sample station tests were performed in an attempt to verify, by an independent method, K's obtained from the deposition tests. In this series, sodium was circulated through the sample stations following a pre-determined

time-temperature program. The sample stations were drained, using an argon purge. The ^{134}Cs and sodium remaining in the tube were measured; and, from the nominal cesium concentration in the sodium and the tube's surface area, a value of K was calculated. The test results indicate (Table 6) vaporization of the cesium occurred during draining, yielding K 's lower than those predicted from deposition tests. Calculations indicate that the cesium must have had nearly unit activity to account for the amount of vaporization that occurred (assuming the validity of K 's obtained from deposition tests). The drain tests did not show a clear dependence of K on surface preparation, but did confirm surface deactivation, as observed in the first isothermal test.

4. Draining and Flushing

After tests at 0.7 wppm, 6 wppm, and 0.2 wppm, the loop was drained and flushed with clean sodium. Prior to each flush, the loop was circulated for a short time (<1 hr) at 500°F. Based on ^{134}Cs analyses, two flushes reduced the cesium from 0.7 to 0.004 wppm, two flushes reduced the cesium from 6 to 0.019 wppm, and four flushes reduced the cesium from 0.2 to 0.006 wppm. This indicates the applicability of draining and flushing to decontaminate sodium systems of cesium. A method would still be required to clean the sodium, unless the system were designed to handle the cesium-contaminated sodium.

E. DISCUSSION OF CESIUM RESULTS

1. Comparison with Other Work on Cesium Deposition

With the exception of the work of Cooper and Taylor,^(7,9,10) no quantitative data are available on cesium deposition in a sodium - stainless steel system, except for a few "points" from reactors operating with failed fuel. The temperature histories for these "points" are poorly defined, but calculated K 's are not inconsistent with the results obtained here for "activated" or partially deactivated surfaces. Cooper,⁽¹⁰⁾ in coupon tests* using ^{137}Cs † as the radioisotope

*Coupons were cold-rolled Type 304 stainless steel, washed in chloroform, acetone, and then in distilled water and dried. Coupons were "soaked" in sodium for a short time at 750°F to assure wetting, prior to low-temperature tests.

† ^{137m}Ba is the isotope actually counted. According to Cooper, the coupons were counted for a sufficiently long time, after being removed from the cesium-sodium mixture, so as to allow equilibration of the ^{137m}Ba with the ^{137}Cs .

TABLE 6
SAMPLE STATION DRAIN TEST RESULTS

TEST CONDITIONS				
Group	Series 1		Series 2	
First	500°F for 24 hr		400°F for 24 hr	
Second	500°F for ~24 hr		400°F for ~24 hr	
	1050°F for 24 hr		1050°F for 24 hr*	
	500°F for 70 hr		400°F for 24 hr	

TEST RESULTS, K(cm)				
Surface Preparation Method	Series 1		Series 2	
	500 (°F)	500-1050-500 (°F)	400 (°F)	400-1050-400 (°F)
Hydrogen Reduced				
Sample A**	0.0070	0.00049	0.085	0.00003
Sample B	0.0059	0.00026	0.26	0.0016†
Hydrogen Reduced and Air Oxidized				
Sample A	0.0041	0.00057	0.0026	0.0011§
Sample B	0.0048	0.00053	0.0041	0.0031†§
HNO ₃ -HF Etched and 1:1 HNO ₃ Treated				
Sample A	0.0090	0.00037	0.0045	0.00012
Sample B	0.0109	0.00045	0.0047	0.0012†
Expected K, FPBL	0.015	0.006	0.5	0.007

*The heater on the sample station with the air-oxidized tube failed during test. The high-temperature history for this tube is 200 min from 800 to 810°F and 110 min from 810 to 830°F. The heater was replaced after the loop was cooled to 400°F.

†Packed with stainless steel mesh

§Partial deactivation expected, based on FPBL results.

**Sample A — From inlet end

Sample B — From outlet end

tag (rather than the ¹³⁴Cs used in this experiment), obtained values of K between the "activated" and partially deactivated values shown in Figure 5. High-temperature exposure of the coupon to sodium (12 hr at 1000°F) did reduce the values of K, but not as much as would have been predicted from FPBL work (a factor of 2, as compared to a predicted factor of 100). It is believed that this

difference is related to the history of the stainless steel used (cold-rolled, degreased, and washed vs drawn and acid etched); and, with sufficient time at temperature, substantially lower values of K would be obtained from coupon tests.

Evans and Watson⁽¹¹⁾ performed cesium deposition tests in sodium in sealed capsules. Within the reported sensitivity of their test (2%), no cesium adsorption was observed. Stainless steel components were etched in a HNO_3 -HCl mixture, prior to test. The temperature program used was reported for only one cesium test (their No. 6). The first data point was obtained at 1200°F, with all subsequent data obtained at lower temperature. Based on FPBL results, the stainless steel in at least this capsule would have been deactivated, relative to cesium adsorption. The upper limit for K calculated from their data is 0.008 cm, assuming their capsule was half-full of sodium. This is not inconsistent with the K obtained from FPBL tests of 0.01 cm at 350°F, their lowest test temperature for Capsule No. 6.

Milstead and Zumwalt⁽¹²⁾ studied cesium plating, in vacuum, on stainless steel. They used ^{137}Cs as tracer. While their results show some similarity to FPBL results (deposition is rapid and reversible, with little or no hysteresis), details of the system are quite different (heats of activation decrease with increasing surface coverage, and oxidized stainless steel surfaces have much greater adsorptive capacity than unoxidized surfaces). These differences indicate that sodium does not act as an inert diluent, but is in competition with the cesium for deposition sites (whatever these may be). As a result, use of K's obtained in a sodium system may not be usable in other liquid metal systems, such as NaK or even sodium with substantial concentrations of impurities.

2. Cesium Deposition Model and Reactor Calculational Method

As is the case in most adsorption situations, cesium deposition in a sodium - stainless steel system cannot be represented by a simple, yet accurate, model. From FPBL work, and by comparison with the work of others, certain highlights of a suitable model now exist. There are at least two deposition mechanisms: one, physical adsorption, which is probably associated with the stainless steel surface in general; and the other, which is much more effective at adsorbing cesium, but sensitive to high-temperature exposure, and is either associated

with a portion of the surface or a particular character of the surface.* Sodium, and perhaps precipitated Na_2O , compete for the available deposition sites with the cesium. Precipitated Na_2O may also adsorb cesium from the sodium.

The formulation of a physical significance for the equilibrium constant, K , and comparison with established concepts is desirable. Based on barium (see Section V-D) and cesium results for deactivated surfaces, multilayer physical adsorption is occurring. One way of visualizing this process is to picture a condensed liquid phase forming at the sodium - stainless steel interface with an average composition different than that of the bulk sodium. The boundary between this condensed phase and the bulk of the sodium is probably undefinable, the composition of the liquid varying gradually as the surface is approached. There have been many mathematical treatments of this type of problem, based on a variety of driving forces. The most successful of these is the one developed by Brunauer, Emmett, and Teller⁽¹³⁾ (B.E.T.). Their development (made to explain adsorption isotherms of gases on solids) assumes the adsorbant surface can be divided into many regions, $S_0, S_1, S_2 \dots S_i$, each region representing the adsorbant area covered with 0, 1, 2, . . . i monolayers of gas. At equilibrium, the area of each region must remain constant. The well-known B.E.T. adsorption isotherm equations can be developed from a mass balance and the two assumptions that: (1) the heat of adsorption is the same for all layers except the first, and (2) the ratio of areas, S_i/S_{i+1} , is a constant when $i > 0$. This equation of interest to relate adsorption to K is the one which was developed for the formation of a finite number of monolayers, Equation B in Reference 13. A simple rearrangement of that expression yields:

$$\bar{n} = \frac{\zeta x}{(1-x)} \left[\frac{1 - (n+1)x^n + nx^{n+1}}{1 + (\zeta-1)x - \zeta x^{n+1}} \right], \quad \dots (3)$$

where:

$\bar{n}$ = average number of layers formed on the surface

$\zeta \cong \exp \left[(E_1 - E_L)R/T \right]$

E_1 = heat of adsorption of the first layer

*Once the details of this are known, highly efficient cesium traps may be developed.

E_L = heat of adsorption (or liquefaction) for all subsequent layers

R = gas constant

T = absolute temperature

x = in their development, the pressure divided by the saturation pressure

n = maximum number of layers that can form.

In our case, if we redefine x as the concentration (c) divided by the saturation concentration (c^*), (i. e., chemical activity, assuming unit activity coefficient), then:

$$K = \frac{\bar{n} a_m}{c} , \quad \dots (4)$$

where a_m is the number of grams adsorbed per monolayer per unit area.

Substitution yields:

$$K = \frac{a_m \zeta}{c^*} \left(\frac{1}{1-x} \right) \left[\frac{1 - (n+1)x^n + nx^{n+1}}{1 + (\zeta-1)x - \zeta x^{n+1}} \right] . \quad \dots (5)$$

There has not as yet been enough data reported to verify this predicted dependence of K on concentration.

The equation is consistent with the observations that a low-concentration K is essentially constant; and, at higher concentrations, K is finite; since:

$$\lim_{x \ll 1} K = \frac{a_m \zeta}{c^*} = \text{constant} , \quad \dots (6)$$

and

$$\lim_{x \rightarrow 1} K = \frac{a_m \zeta^n (n+1)}{2c^* (n\zeta + 1)} = \text{constant} . \quad \dots (7)$$

Because of the complexity of temperature effects, it is premature to estimate whether an Arrhenius plot of K should be a straight line.

While the available data do not permit a more quantitative description of the model, cesium deposition in a reactor system can be predicted with reasonable confidence. Appendix III presents the calculational method and four equilibrium sample cases. With the assumed surface areas, it is apparent that presently designed cold traps will not efficiently remove cesium from an LMFBR.

V. BARIUM TESTS

A. BACKGROUND

Barium has been observed to be one of the fission product elements that plates onto reactor piping. From measurements of the radioactive decay of ^{137m}Ba when coupon samples were removed from a ^{137}Cs -sodium mixture, Cooper⁽¹⁰⁾ inferred that barium is strongly adsorbed onto metal surfaces. A similar inference can be made from the work of Evans and Watson.⁽¹¹⁾ Quantitative data are completely lacking; therefore, FPBL tests were performed on barium adsorption. The standard free energy of formation of BaCl_2 is only slightly more negative than that of NaCl (-83 vs -76 kcal/gm-atom Cl at 1000°K); however, the standard state free energy of formation of BaO is appreciably more negative than that of Na_2O (-111 vs -66 kcal/gm-atom O). Therefore, if barium (as the chloride) were contacted with sodium containing an excess of oxygen relative to the barium, the barium should be converted to the oxide. (The system is, of course, not at standard state relative to oxygen; therefore, both the free energies of formation of Na_2O and BaO will be more negative than the standard state values. Unless Ba-O is a very unusual system, a sufficient difference in free energy should exist to drive the reaction toward the formation of the barium oxide.) No quantitative data are available on the solubility of barium in oxygen-rich sodium, except that it is very low. The experimental design for barium tests was therefore necessarily quite different than that for cesium.

B. EXPERIMENTAL

A sample station was prepared for barium injection, in which 0.3 mg of barium (as BaCl_2) was deposited on stainless steel mesh. The specific activity of ^{133}Ba in the BaCl_2 was 1.4 Ci/gm Ba. Cold-trapped sodium was first circulated through the loop and sample station for 1-1/4 hr at 300°F, then 1-1/2 hr at 500°F, with no barium migration, as indicated on the surge tank and test section detectors, and on a specially installed detector monitoring the sodium effluent from the sample station. The loop and station temperatures were increased to 700°F, and a gradual increase in radioactivity was observed on all detectors. There were no spikes or bursts monitored by the sample station detector; therefore, the barium was entering the loop in solution, rather than as particulates.

After 9 days, the barium content in the various components was still increasing. The sample station was valved from the loop, and the loop homogenized for 24 hr at 700°F. Based on a rather crude radiation measurement, an immeasurably small amount of barium had been removed from the sample station (<10%). A parameter test series was performed (Table 7), to provide at least two checks of the various system parameters needed to describe barium deposition in the FPBL (excluding surface area and sodium volume). The sample station was again valved into the system, and flow continued until the rate of change of barium measured by the various radiation detectors was very low (system nearly saturated). After completion of this test series, a second parametric test series was performed in the loop. The results from injection and second parametric tests will be reported at a later date.

TABLE 7
PARAMETRIC TEST SERIES FOR BARIUM DEPOSITION TESTS

Run Designation	Temperature (°F)			Sodium Flow Rate at 700°F (cc/sec)	Duration of Test (days)
	Barium Injection Sample Station	Loop and Test Section No. 1	Test Section No. 2		
-	300	300	300	15*	(1-1/4 hr)
-	500	500	500	15*	(1-1/2 hr)
-	700	700	700	15*	9
a	-	700	500	4.2	4
b	-	700	500	2.5	8
c	-	700	300	1.8	10
d	-	700	500	4.2	12
e	-	700	700	10.	1
-	700	700	700	15*	39
-	-	700	700	10.	14

*Total flow through Test Section No. 1. Flow was divided between Test Section No. 2 and sample station, in a ratio of ~1:2.

C. DATA REDUCTION

The method used to reduce data from the barium tests is necessarily different than the method used to reduce data from cesium tests. With cesium, surface adsorption was so low that the surface could reach equilibrium without appreciably changing the cesium concentration in the liquid. The solubility of barium in oxygen-rich sodium is very low, and the surface-liquid equilibrium very much favors the surface; therefore, appreciable time is required for the loop, as a closed system, to reach equilibrium. (This is not to be interpreted to mean that diffusion through the fluid boundary is limiting. As was shown for cesium, the test sections are not diffusion limited.) The problem in interpreting count rate data from the barium tests was to develop a suitable mathematical model of the loop to explain the observed time dependence of barium distribution. This development is given in detail in Appendix IV. In brief, for the barium test series reported here, the "black box" approach, as shown in Figure 8, was taken. It is assumed that a sodium stream containing barium enters a region, liquid-surface equilibrium takes place, and the sodium of the proper barium concentration leaves the region. One of the restrictions of this approach is that, at no time can the solubility limit of barium be exceeded. The solution to the differential equations obtained for the loop depends upon what is assumed about the exit concentration of barium in the sodium and the boundary conditions.

7708-4746

Figure 8. Mathematical Model of FPBL

Two concentration dependencies were assumed: (1) that the exit concentration is equal to the solubility limit of barium at the region temperature, and (2) that the ratio of surface-to-liquid concentration is a constant. The data did not fit the first assumption; therefore, barium distribution in the FPBL could not be explained by the constant exit concentration assumption. The second assumption is identical with that used to explain cesium distribution, and, as will be shown, provides an excellent fit and cross-fit to barium results.

D. TEST RESULTS

The results of the intercomparison from the first barium series is presented in Table 8. For these tests nothing inconsistent with the K-concept was found. The intercomparison of various parameters is sufficiently good, in the writer's opinion, to preclude the possibility of another deposition mechanism. Figure 9 is an Arrhenius plot of K's obtained for barium deposition. It is interesting to note that the heat of activation for barium deposition (-3.5 kcal/gm-mole) is virtually equal to the heat of activation for cesium deposition (-3 kcal/gm-mole) for similar surface condition. This tends to bear out prior judgments that the value of K for at least these two elements is related to how strongly the sodium is rejecting the element, with the surface being a passive participant. If, as more elements are studied, a similar behavior is observed, a generalized expression for K, perhaps based on solubility, will be developed.

TABLE 8
INTERCOMPARISON OF RESULTS FROM
FIRST BARIUM SERIES

<u>Source</u>	<u>K(500)/K(700)</u>
a	4.91
d	3.92
	<u>K(300)/K(700)</u>
c	22.9*
a & c	18.3*
	<u>K(700) (cm)</u>
e (1)	490
e (2)	530
	<u>v_a/v_b</u>
a & b	1.51
Heat Balance	1.63
	<u>I(700)/I(300)</u>
a & c	0.81
b & c	0.87
	<u>$\eta_1 \xi_1 / \eta_2 \xi_2$</u>
e	0.922
Cesium Deposition Tests	1.015
*Mass Balance

7708 4747

Figure 9. Log K vs $1/T$ for Barium Deposition Tests
 $[K(700^{\circ}F) \cong 500 \text{ cm}]$

VI. CONCLUSIONS

- 1) The kinetics of deposition and dissolution of cesium are rapid, with no apparent hysteresis. Based on preliminary results, the same is true of barium.
- 2) An equilibrium constant can be used to express the liquid-to-surface partition of cesium and barium.
- 3) For cesium (miscible in all proportions in sodium), the liquid-to-surface partition strongly favors the sodium phase. For barium (very slightly soluble in oxygen-rich sodium), the liquid-to-surface partition strongly favors the stainless steel surface. The heats of activation of adsorption (ΔH) for both the cesium - sodium - stainless steel and barium - sodium - stainless steel, for comparable surfaces, are virtually equal (-3.0 and -3.5 kcal/gm-mole, respectively). These values for ΔH suggest a physical adsorption process.
- 4) For cesium, surface adsorption was radically affected by the prior temperature history of the stainless steel surface. In FPBL tests, surface adsorption was reduced, by greater than an order of magnitude, by exposing the surface to high-temperature sodium. The surface deactivation was irreversible. To date, tests have not been conducted to determine whether barium adsorption follows a similar pattern.
- 5) For the surface preparations studied, no effect of surface treatment on cesium adsorption was measured.
- 6) Solid sodium oxide appears to reduce the ability of the stainless steel surfaces to adsorb cesium.
- 7) Draining and flushing has been calculated and demonstrated to be an efficient method for decontaminating a stainless steel system of cesium. Draining and flushing will probably not be an efficient method to decontaminate a system of barium.

- 8) For cesium, hot draining will not appreciably increase decontamination over warm draining; virtually all the cesium being in solution, and the majority of residual activity being associated with undrained sodium. It is not known, at this time, the degree of improvement in barium decontamination with a hot vs a warm drain.

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APPENDIX I

REDUCTION OF CESIUM EXPERIMENTAL DATA

A. DATA REDUCTION

The radiation count rate associated with the surface deposit must be discriminated from the total count rate, which also includes background and liquid phase counts. The data required to determine surface deposition are:

- 1) Deposition test section count rate, including fission product associated with the surface (s), fission product in the liquid phase (l), and background (b) at operating temperature (t), $C_1(s+l+b,t)$
- 2) Background radiation in the test section shield, $C_1(b,t)$
- 3) Count rate in the test section detector with a particular source (S), $C_1(S+b,t)$
- 4) Surge tank count rate; because the tank has a small surface-to-volume ratio, the surge tank count rate will reflect the total activity dissolved in the sodium in the surge tank, $C_3(l,t)$.

The following steps are used to reduce data:

- 1) Liquid Concentration of Fission Product at Temperature (t) Relative to Reference Temperature (t_1)^{*}

$$\frac{w(t)}{w(t_1)} = \frac{C_3(l,t)}{C_3(l,t_1)} \left[1 + \frac{V_L}{V_{ST}(t_1)} \left(1 - \frac{\rho(t)}{\rho(t_1)} \right) \right]^{-1}, \quad \dots (I-1)$$

where:

$w(t)$ = concentration by weight of fission product in sodium at temperature t

V_L and V_{ST} = volumes of sodium in the loop (exclusive of the surge tank) and the surge tank, respectively

$\rho(t)$ = the density of sodium at temperature t.

*Deposition at the reference temperature (1100°F) is assumed to be small. See Section B of this appendix for the derivation of this expression.

2) Normalization of Test Section Count Rate Data and Correction for Background

$$C'_1(s+l,t) = \left[C_1(s+l+b,t) - C_1(b,t) \right] \left[\frac{C(S)_{\text{NOM}}}{C_1(s+b,t) - C_1(b,t)} \right] , \quad \dots (I-2)$$

where $C(S)_{\text{NOM}}$ is the nominal source count rate.

3) Calculation of the Test Section No. 1 Equilibrium Constant $[K_1(t)]^*$

$$K_1(t) = \frac{V}{A} \left[\frac{C'_1(s+l,t)}{C'_1(l,t_1)} \frac{w(t_1)}{w(t)} \frac{\rho(t_1)}{\rho(t)} - 1 \right] , \quad \dots (I-3)$$

where V/A is the liquid volume - to - surface area ratio of the test section (cm).

4) Calculation of the Equilibrium Constant for the Loop Excluding Test Sections $[K_3(t)]^\dagger$.

$$K_3(t) = \left(\frac{V}{A} \right)_L \left[\frac{w(t_1)}{w(t)} - 1 \right] - \sum_{i=1}^2 \frac{A_{\text{TS}_i}}{A_L} K_i(t) , \quad \dots (I-4)$$

where:

$\left(\frac{V}{A} \right)_L$ = liquid volume - to - surface area ratio of the loop, excluding test sections

A_{TS} = apparent surface area of the test sections

A_L = apparent surface area of the loop, excluding test sections.

5) Calculation of Surface Concentration $[S(t)]$

$$S(t) = K(t)w(t)\rho(t) . \quad \dots (I-5)$$

*See Section C of this appendix for the derivation of this expression. A similar expression can be derived for Test Section No. 2, in which $K_2(t)$ is calculated, using $C'_2(s+l,t)$ and $C'_2(l,t_1)$.

†See Section D of this appendix for the derivation of this expression.

B. DERIVATION OF EXPRESSION $w(t)/w(t_1)$

The surge tank detector measures the total activity in the surge tank. With this, as well as all count rates, it is assumed that the count rate is sufficiently small that a live-time correction is not required. Since the weight of sodium in the surge tank depends upon the temperature of the loop, a density correction term is needed. The volume of the loop, excluding the surge tank, is assumed constant, the correction for stainless steel expansion being small.

The volume of sodium in the surge tank is

$$V_{ST}(t) = V(t) - V_L \quad , \quad \dots (I-6)$$

where $V(t)$ is the total volume of sodium at temperature t . Since

$$V(t) = \frac{W}{\rho(t)} \quad , \quad \dots (I-7)$$

where W is the mass of sodium in the loop, and

$$W = [V_{ST}(t_1) + V_L] \rho(t_1) \quad . \quad \dots (I-8)$$

Therefore:

$$V_{ST}(t) = \frac{\rho(t_1)}{\rho(t)} [V_{ST}(t_1) + V_L] - V_L \quad , \quad \dots (I-9)$$

and

$$\frac{V_{ST}(t)}{V_{ST}(t_1)} \frac{\rho(t)}{\rho(t_1)} = 1 + \frac{V_L}{V_{ST}(t_1)} \left[1 - \frac{\rho(t)}{\rho(t_1)} \right] \quad . \quad \dots (I-10)$$

The total weight of fission product in the surge tank is proportional to the corrected surge tank count rate; or:

$$\frac{w(t)}{w(t_1)} = \frac{\left. \frac{\text{wt. f.p.}}{\text{wt. Na}} \right|_t}{\left. \frac{\text{wt. f.p.}}{\text{wt. Na}} \right|_{t_1}} = \frac{C_3(\ell, t)}{C_3(\ell, t_1)} \frac{V_{ST}(t_1) \rho(t_1)}{V_{ST}(t) \rho(t)} \quad , \quad \dots (I-11)$$

or

$$\frac{w(t)}{w(t_1)} = \frac{C_3(\ell, t)}{C_3(\ell, t_1)} \left\{ 1 + \frac{V_L}{V_{ST}(t_1)} \left[1 - \frac{\rho(t)}{\rho(t_1)} \right] \right\}^{-1} \quad \dots (I-1)$$

C. DERIVATION OF EXPRESSION $K_1(t)$

The normalized and background corrected count rate from the test section $[C'_1(s+\ell, t)]$ is proportional to the mass of cesium dissolved in the sodium and absorbed on the surface, or:

$$C'_1(s+\ell, t) = \epsilon [w(t)\rho(t)V + SA] \quad \dots (I-12)$$

or

$$C'_1(s+\ell, t) = \epsilon w \rho V [1 + K_1(t) A/V] \quad , \quad \dots (I-13)$$

and

$$1 + K_1(t) \frac{A}{V} = \frac{C'_1(s+\ell, t)}{\epsilon w(t)\rho(t)V} \quad \dots (I-14)$$

Then

$$\frac{1 + K_1(t) \frac{A}{V}}{1 + K_1(t_1) \frac{A}{V}} = \frac{C'_1(s+\ell, t)}{C'_1(s+\ell, t_1)} \frac{w(t_1)}{w(t)} \frac{\rho(t_1)}{\rho(t)} \quad \dots (I-15)$$

Assuming the surface deposition at reference temperature t_1 is small,

$$1 + K_1(t_1) \frac{A}{V} \cong 1 \quad , \quad \dots (I-16)$$

$$C'_1(s+\ell, t_1) \cong C'_1(\ell, t_1) \quad \dots (I-17)$$

Then

$$K_1(t) = \frac{V}{A} \left[\frac{C'_1(s+\ell, t)}{C'_1(\ell, t_1)} \frac{w(t_1)}{w(t)} \frac{\rho(t_1)}{\rho(t)} - 1 \right] \quad \dots (I-3)$$

Of interest is the effect of the assumption that surface deposition at t_1 is small. If we let the K_1 's calculated above be denoted by K_{uncor} , and K_1 's calculated not assuming deposition at t_1 is small be denoted as K_{cor} , then:

$$1 + K_{\text{uncor}} \left(\frac{A}{V} \right) = \frac{C_1'(s+l, t)}{C_1'(\ell, t_1)} \frac{w(t_1)}{w(t)} \frac{\rho(t_1)}{\rho(t)} \quad \dots (I-18)$$

and

$$1 + K_{\text{cor}} \left(\frac{A}{V} \right) = \frac{C_1'(s+l, t)}{C_1'(\ell, t_1)} \frac{w(t_1)}{w(t)} \frac{\rho(t_1)}{\rho(t)} \left[1 + K_1(t_1) \left(\frac{A}{V} \right) \right] \quad \dots (I-19)$$

Dividing the two expressions:

$$\frac{1 + K_{\text{cor}} \frac{A}{V}}{1 + K_{\text{uncor}} \frac{A}{V}} = 1 + K_1(t_1) \frac{A}{V} \quad \dots (I-20)$$

Rearranging:

$$K_{\text{cor}} = K_{\text{uncor}} + K_1(t_1) + K_{\text{uncor}} K_1(t) \frac{A}{V} \quad \dots (I-21)$$

The small values of K_{uncor} obtained for deactivated surfaces (~ 0.01 cm at 300°F) and test section surface-to-volume ratio (12 cm^{-1}) means the last term will be small. The effect therefore of assuming $K_1(t_1)$ is small is to depress the value of K_{cor} by an amount equal to $K_1(t_1)$. Using the data from Figure 5, an iterative process was applied to correct for this assumption [by adding the extrapolated value of $K_1(1100)$ to each data point, redrawing the figure, and repeating the process using the newly obtained value of $K_1(1100)$]. The resultant line has a slightly lower slope than the uncorrected line, and the values for K_{cor} fall within a factor of two of those for K_{uncor} . Because of the uncertainty in the data, a factor of two is not considered significant.

D. CALCULATION OF EXPRESSION $K_3(t)$

The change in cesium concentration in the loop is proportional to the change in surface adsorption, assuming no absorption at t_1 :

1) Mass of cesium absorbed, in going from t_1 to $t =$

$$w(t_1)\rho(t_1)V_L(t_1) - w(t)\rho(t)V_L(t) \quad , \quad \dots (I-22)$$

where V_L is the total volume of the loop, which is almost equal to the total volume of the loop less the volume of the test sections.

2) Mass of cesium absorbed in the test sections =

$$\sum_{i=1}^2 K_i(t) A_{TS_i} w(t) \rho(t) \quad . \quad \dots (I-23)$$

3) Mass of cesium absorbed in the loop, excluding test sections =

$$\left[\frac{w(t_1)}{w(t)} \frac{\rho(t_1)}{\rho(t)} \frac{V_L(t_1)}{V_L(t)} - 1 \right] w(t) \rho(t) V_L(t) - \sum_{i=1}^2 K_i(t) A_{TS_i} w(t) \rho(t) \quad . \quad \dots (I-24)$$

Since there was no change in the mass of sodium in the loop,

$$\rho(t_1) V_L(t_1) = \rho(t) V_L(t) \quad . \quad \dots (I-24)$$

Then

$$K_3(t) = \frac{\left[\frac{w(t_1)}{w(t)} - 1 \right] w(t) \rho(t) V_L(t) - \sum_{i=1}^2 K_i(t) A_{TS_i} w(t) \rho(t)}{A_L w(t) \rho(t)} \quad , \quad \dots (I-25)$$

$$K_3(t) = \left(\frac{V}{A} \right) \left[\frac{w(t_1)}{w(t)} - 1 \right] - \sum_{i=1}^2 \frac{A_{TS_i}}{A_L} K_i(t) \quad . \quad \dots (I-4)$$

APPENDIX II
CESIUM DEPOSITION TEST DATA: ISOTHERMAL,
NONISOTHERMAL, AND KINETIC

A. FIRST ISOTHERMAL TEST (0.7 wppm Cs, <10 wppm O)

The results of this test are presented in Table II-1. As in the remainder of the tables in this appendix reporting test results, the order in which data are listed is the same as the order of the test.

TABLE II-1
RESULTS OF FIRST ISOTHERMAL TEST

Test Number (See Figure 4)	Temperature (°F)	Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop, Excluding Test Sections		Test Section No. 1		Test Section No. 2	
				S_3 (gm Cs/cm ² $\times 10^8$)	K_3 (cm)	S_1 (gm Cs/cm ² $\times 10^8$)	K_1 (cm)	S_2^* (gm Cs/cm ² $\times 10^8$)	K_2 (cm)
1	485	260	0.57	10.8	0.21	0.59	0.011	1.6	0.031
2	330	3.5	0.46	18.	0.43	1.22	0.030	2.2	0.053
3	725	1.3	0.72	0.76	0.013	-0.06	-0.001	0.026	0.0042
4	520	1.0	0.63	6.4	0.11	0.72	0.012	1.6	0.028
5	500	16.0	0.61	8.0	0.15	0.67	0.012	1.3	0.024
6	920	1.0	0.73	0.00	0.00	-0.03	0.000	-0.05	-0.001
7	1105	1.2	0.73	-	-	-	-	-	-
8	600	140	0.73	Low	Low	-	-	-	-
9	300	1.0	0.73	Low	Low	-	-	-	-

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K - Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

Based on counting statistics, 1σ corresponds to 0.15×10^{-8} gm Cs/cm².

The results from the first isothermal test are rather striking, the loop (excluding the test sections and Test Section No. 2) having K's* 10 and 2 times, respectively, that of Test Section No. 1 prior to the 920°F and/or the 1100°F runs. The deposition capacity of the surface (activity) was markedly reduced after the 1100°F run. The observed deactivation of the surface is irreversible,† and can only be related to a temperature effect. From Table 5, it appears that surface deactivation occurs to a marked degree when stainless steel is heated in

*Ratio of cesium surface-to-liquid concentration.

†As measured in later tests.

TABLE II-2
RESULTS OF SECOND ISOTHERMAL TEST

Temperature (°F)	Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop, Excluding Test Sections		Test Section No. 1		Test Section No. 2	
			S ₃ (gm Cs/cm ² $\times 10^8$)	K ₃ (cm)	S ₁ (gm Cs/cm ² $\times 10^8$)	K ₁ (cm)	S ₂ (gm Cs/cm ² $\times 10^8$)	K ₂ (cm)
300	390	0.67	2.6	0.043	2.1	0.034	1.0	0.016
400	4.0	0.70	1.3	0.021	0.76	0.012	0.25	0.004
300	0.8	0.69	1.9	0.031	1.3	0.021	1.2	0.020
300	18.0	0.67	2.7	0.044	2.0	0.033	1.1	0.017
500	4.5	0.71	0.95	0.015	0.07	0.001	0.42	0.007
300	0.8	0.70	1.4	0.022	1.0	0.015	0.46	0.007
300	18.4	0.67	2.6	0.043	1.9	0.032	1.5	0.024
600	3.2	0.72	0.55	0.009	0.44	0.007	0.37	0.006
300	0.8	0.70	1.4	0.022	1.4	0.022	0.37	0.013
300	91.2	0.67	2.7	0.044	2.1	0.034	1.3	0.021
700	4.5	0.73	0.12	0.002	-0.55	-0.009	-0.04	0.000
300	0.8	0.70	1.2	0.018	1.8	0.028	0.68	0.011
300	18.6	0.68	2.0	0.032	1.3	0.021	0.94	0.015
800	3.8	0.73	0.0	0.000	-0.34	-0.005	0.75	0.013
300	0.8	0.71	0.76	0.012	1.1	0.017	0.43	0.007
300	19.3	0.69	1.5	0.024	1.8	0.028	1.2	0.018
900	3.7	0.73	0.0	0.000	0.0	0.000	-0.04	0.000
300	0.8	0.72	0.61	0.009	0.2	0.003	0.73	0.011
300	19.3	0.69	1.6	0.025	1.8	0.028	1.7	0.027
1100	4.0	0.73	-	-	-	-	-	-
300	0.8	0.73	0.0	0.000	1.2	0.017	0.54	0.008
300	19.8	0.71	0.73	0.011	1.5	0.028	0.97	0.015

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

Based on counting statistics, 1σ corresponds to 0.25×10^{-8} gm Cs/cm².

contact with sodium above 800°F. One-half hour at 1150 to 1200°F reduced surface activity to within a factor of 2 of fully "deactivated"* FPBL stainless steel.

B. SECOND ISOTHERMAL TEST (0.7 wppm Cs, <10 wppm O)

The second isothermal test was essentially a repeat of the first isothermal test, except that, after each high-temperature test, the loop temperature was reduced to the baseline temperature. For this test only, collimators were used in the test section detector shields to reduce the field of view by a factor of 1.50 (2-1/4 to 1-1/2 in.). The results are summarized in Table II-2.

The scatter in the results from this test is, at least in part, due to the use of greater collimation, with the resultant decrease in count rate, the counting period having been kept constant (10 min for all 0.7-wppm Cs tests, and 4 min for all 6-wppm Cs tests). The tests did show that further deactivation of loop surfaces occurred after 4.5 hr at 700°F. Of interest, though, was the apparent "reactivation," each time the loop temperature was reduced to 300°F and maintained at this temperature for at least 18 hr. This test was the only test in which "reactivation" was observed. It is believed that an inadvertently low power setting on the surge tank pump, and the resultant loss of stirring action, is the explanation for this observation.

C. KINETIC TEST (0.7 wppm Cs, <10 wppm O)

The purpose of the kinetic test was to measure deposition kinetics and the kinetics of "reactivation." The loop was first heated to 1100°F to deactivate surfaces, then cooled to operating temperature. Results are reported in Table II-3.

Based on counting statistics, the scatter in data is not unreasonable. Test section results indicate some additional deactivation occurred after 12.3 hr at 1100°F, but it was slight; therefore, either the loop was fully deactivated, or deactivation was proceeding at a slow rate. As in all tests, deposition rates were too rapid to follow.

*As measured in later tests.

TABLE II-3
RESULTS OF KINETIC TEST

Temperature (°F)	Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop, Excluding Test Sections		Test Section No. 1		Test Section No. 2	
			S ₃ (gm Cs/cm ² $\times 10^8$)	K ₃ (cm)	S ₁ (gm Cs/cm ² $\times 10^8$)	K ₁ (cm)	S ₂ (gm Cs/cm ³ $\times 10^8$)	K ₂ (cm)
~300	670	-	-	-	-	-	-	-
1100	2.8	0.73	-	-	-	-	-	-
550	21.2	0.72	0.31	0.004	-0.28	-0.004	0.29	0.004
1100	3.0	0.73	-	-	-	-	-	-
450	21.0	0.71	0.76	0.010	0.33	0.005	0.67	0.009
1100	4.0	0.73	-	-	-	-	-	-
350	93.0	0.71	0.73	0.010	0.66	0.010	0.58	0.008
1100	2.5	0.73	-	-	-	-	-	-
300	21.5	0.70	1.2	0.017	0.76	0.011	0.59	0.008
1100	2.7	0.73	-	-	-	-	-	-
250	25.2	0.70	1.3	0.019	0.98	0.014	1.2	0.017

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

Based on counting statistics, 1σ corresponds to 0.20×10^{-8} gm Cs/cm²

D. FIRST NONISOTHERMAL TEST (0.7 wppm Cs, <10 wppm O)

During the period between the kinetic test and this test, the loop was twice cooled to room temperature — once to permit reradiographing of the test sections, to assure that the stainless steel mesh had not redistributed (no redistribution was measured), and once because of a 3-day power outage.

For the first nonisothermal test, the loop was heated to 1100°F to obtain reference data, then cooled to 700°F. Temperatures in the test sections were varied, with Test Section No. 1 always being the coolest. Results are reported in Table II-4.

The results from this test are interesting, because, with a constant temperature in Test Section No. 1, deposition increased with decreasing temperature in Test Section No. 2.

TABLE II-4
RESULTS OF FIRST NONISOTHERMAL TEST

Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Temperature Loop, Excluding Test Sections ($^{\circ}$ F)	Test Section No. 1			Test Section No. 2		
			Temperature ($^{\circ}$ F)	S ₁ (gm Cs/cm ² $\times 10^8$)	K ₁ (cm)	Temperature ($^{\circ}$ F)	S ₂ (gm Cs/cm ² $\times 10^8$)	K ₂ (cm)
2	-	1100	1100	-	-	1100	-	-
1	0.73	700	700	-	-	700	-	-
1	0.71	700	330	0.10	0.001	710	0.04	0.001
2	0.71	700	340	0.30	0.005	420	0.43	0.007
19	0.73	700	700	-	-	700	-	-
3	0.72	705	310	0.04	0.001	500	0.12	0.002
2	0.72	720	305	0.50	0.008	350	-0.06	-0.001
19	0.73	700	700	-	-	700	-	-
2	0.73	695	257	0.66	0.010	495	0.45	0.007
3	0.72	700	250	0.74	0.011	400	0.36	0.006
18	0.73	700	700	-	-	700	-	-
3	0.73	705	250	0.98	0.015	350	0.00	0.000
2	0.72	700	250	1.3	0.020	300	0.95	0.014
-	0.73	700	700	-	-	700	-	-

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant - $S/w\rho$ (ρ is the sodium density)

Based on counting statistics, 1σ corresponds to 0.20×10^{-8} gm Cs/cm²

E. THIRD ISOTHERMAL TEST (6 wppm Cs, <10 wppm O)

After completion of the first nonisothermal test, the loop was drained and rinsed twice with sodium from the fill tank. Prior to draining the second rinse from the loop, a data set was taken. The results indicate the cesium concentration in the loop was 0.004 wppm; counting statistics were such as to prevent calculation of surface deposition. After draining, a data set was taken, and the results indicated no cesium was associated with the surface.

The loop was refilled with sodium, heated to 500 $^{\circ}$ F, and cold trapped at from 250 to 275 $^{\circ}$ F for 6 hr. Cesium was added to the loop; and, after 24 hr at 500 $^{\circ}$ F, the third isothermal test was started. The loop temperature was reduced to 300 $^{\circ}$ F, and the collection of data sets started. As soon as the loop temperature was lowered, data from the surge tank detector indicated a gradual

TABLE II-5
RESULTS OF THIRD ISOTHERMAL TEST

Temperature (°F)	Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop, Excluding Test Section No. 2		Test Section No. 2	
			S_{1+3} (gm Cs/cm ² $\times 10^8$)	K_{1+3} (cm)	S_2^* (gm Cs/cm ² $\times 10^8$)	K_2 (cm)
295	20	5.6	8.6	0.016	6.4	0.012
395	2.5	5.6	6.9	0.013	5.8	0.011
503	2.2	5.6	3.3	0.006	4.7	0.009
598	1.3	5.7	1.2	0.002	3.4	0.007
298	1.0	5.6	6.5	0.012	4.3	0.008
302	17.0	5.6	6.4	0.012	5.0	0.009
695	2.2	5.6	4.5	0.009	2.4	0.005
800	3.7	5.8	-4.9	-0.010	4.9	0.010
300	1.0	5.6	7.5	0.014	4.1	0.008
298	65.6	5.6	7.5	0.014	4.9	0.010
905	3.0	5.7	1.4	0.003	-3.7	-0.008
1098	2.5	5.7	-	-	-	-
293	1.0	5.6	8.6	0.016	2.5	0.005
286	17.0	5.6	8.6	0.016	5.0	0.009
405	1.7	5.6	6.9	0.013	4.3	0.008
517	2.8	5.6	6.3	0.012	2.8	0.005
302	1.0	5.6	8.0	0.015	5.0	0.009

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

* Based on counting statistics, 1σ corresponds to 0.4×10^{-8} gm Cs/cm²

decrease in cesium concentration in the sodium. The surge tank pump was tested to assure the tank had adequate stirring. After 3 hr, test results indicated the cesium liquid concentration was 0.94 that at the start of the 300°F run, and remained unchanged after 20 hr. This change in liquid concentration would be expected, if no cesium were deposited on the loop surfaces at 500°F, and deposition to a K of 0.01 cm occurred at 300°F. When the same time-temperature program was followed later, with the addition of normally 1 wppm Cs, the change in the surge tank detector readings indicated deposition to values of K consistent with other tests at both temperatures.

The results from the third isothermal test are reported in Table II-5. Prior to completion of the test, the photomultiplier in Test Section No. 1 failed; inadequate data were collected to permit data reduction for this test section. K's calculated from this test are in good agreement with K's calculated from the 0.7-wppm tests. Considerable scatter, though, does exist, in results from the 800 and 900°F runs. One feature of the data reduction technique becomes apparent — with little surface deposition, test results are sensitive to small changes in surge tank data.

F. FOURTH ISOTHERMAL TEST (6 wppm Cs, <10 wppm O)

Because of the uncertainties in results from the preceding test, the fourth isothermal test was conducted. Shortly after the beginning of the test, the detector on Test Section No. 1 failed; apparently, when changing the photomultiplier, a solder connection within the preamplifier failed. After repair, the system was too unstable to consider the data from this portion of the test reliable.

Results of this test are reported in Table II-6. The large spread in the values of K calculated from the surge tank and test section data raises doubts as to the validity of the results. The 1100°F surge tank data are inconsistent with data from other runs. If they are assumed in error, and data from the third isothermal and second nonisothermal tests used, the calculated test section K's are approximately equal to the K's obtained in earlier runs.

TABLE II-6
RESULTS OF FOURTH ISOTHERMAL TEST

Temperature (°F)	Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop, Excluding Test Section No. 2		Test Section No. 2	
			S_{1+3} (gm Cs/cm ² $\times 10^8$)	K_{1+3} (cm)	S_2^* (gm Cs/cm ² $\times 10^8$)	K_2 (cm)
307	475	5.6	5.4	0.010	0.9	0.002
352	2.2	5.6	12.	0.026	2.5	0.005
402	2.5	5.6	2.1	0.004	0.3	0.001
502	2.5	5.6	15.	0.029	2.6	0.005
307	1.0	5.6	11.	0.021	2.0	0.004
300	17.3	5.6	15.	0.027	2.5	0.005
600	2.2	5.6	8.7	0.017	1.5	0.003
701	2.0	5.7	7.0	0.014	1.2	0.002
301	1.8	5.5	14.	0.026	2.5	0.005
300	17.2	5.4	16.	0.029	2.8	0.005
800	2.0	5.7	1.5	0.003	0.3	0.001
1100	2.5	5.7	-	-	-	-
300	2.5	5.6	18.	0.034	3.3	0.006
301	17.0	5.6	12.	0.023	2.2	0.004

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

* Based on counting statistics, 1σ corresponds to 0.4×10^{-8} gm Cs/cm²

G. SECOND NONISOTHERMAL TEST (6 wppm Cs, <10 wppm O)

The second nonisothermal test was conducted in much the same manner as the first nonisothermal test, except that Test Section No. 2 was always the coldest portion of the loop. Results of the test are reported in Table II-7. K's calculated for the loop, excluding the test sections, were consistent with previously obtained values, varying from 0.0003 to 0.0006 cm, at 700°F. Test section results show no trend, calculated K's being lower than would be expected from the results of the isothermal tests.

TABLE II-7
RESULTS OF SECOND NONISOTHERMAL TEST

Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop Temperature, Excluding Test Sections (°F)	Test Section No. 1			Test Section No. 2		
			Temperature (°F)	S ₁ (gm Cs/cm ² $\times 10^8$)	K ₁ (cm)	Temperature (°F)	S ₂ (gm Cs/cm ² $\times 10^8$)	K ₂ (cm)
1.3	5.6	700	700	2.9	0.006	700	1.1	0.002
2.7	5.6	700	700	2.8	0.006	350	2.4	0.005
2.4	5.6	700	500	3.3	0.006	350	2.1	0.004
1.5	5.6	700	400	2.7	0.005	350	1.4	0.003
9.5	5.6	700	700	0.9	0.002	700	3.1	0.006
2.3	5.6	695	500	0.6	0.001	300	1.6	0.003
1.7	5.6	690	405	1.1	0.002	300	1.2	0.002
3.7	5.7	690	350	0.8	0.002	310	1.1	0.002
1.3	5.6	700	700	0.9	0.002	700	1.2	0.002
17.2	5.6	700	700	1.1	0.002	700	-0.3	-0.001
3.0	5.7	700	400	1.2	0.002	250	1.9	0.004
2.2	5.7	700	350	0.2	0.000	250	2.3	0.004
2.2	5.7	700	300	0.6	0.001	250	2.2	0.004
18.8	5.6	700	715	2.2	0.004	705	1.1	0.002

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

*Based on counting statistics, 1σ corresponds to 0.4×10^{-8} gm Cs/cm²

H. FIFTH ISOTHERMAL TEST (6 wppm Cs, 25 wppm O)

After the second nonisothermal test, the temperature of the loop was reduced to 500°F and oxygen added. The oxygen concentration in the loop should have increased to 50 wppm, though chemical analysis indicated an increase to only 25 wppm. It is possible that some of the oxide did not make its way into the sodium, being left in the top of the surge tank. The oxygen could also have been at least partially adsorbed into the stainless steel surfaces of the loop. The addition went smoothly, with no indication of any change within the loop (i. e., temperature or flow rate). After three days at 500°F, the fifth isothermal run was conducted. Results are reported in Table II-8.

The results indicate that the oxygen added to the loop did not markedly change the amount of cesium that deposits. Based on surge tank data from this test, the reported test section K's are slightly low; if the data are recalculated, using surge tank data from other tests, K's are virtually equal to the K's obtained for the low-oxygen isothermal runs.

TABLE II-8
RESULTS OF FIFTH ISOTHERMAL TEST

Temperature (°F)	Time at Temperature (hr)	w (gm Cs/gm Na $\times 10^6$)	Loop, Excluding Test Sections		Test Section No. 1		Test Section No. 2	
			S_3 (gm Cs/cm ² $\times 10^8$)	K_3 (cm)	S_1 (gm Cs/cm ² $\times 10^8$)	K_1 (cm)	S_2 (gm Cs/cm ² $\times 10^8$)	K_2 (cm)
500	68	5.6	7.9	0.015	3.5	0.007	2.3	0.004
300	2.3	5.5	10.5	0.019	3.0	0.006	5.1	0.009
400	3.0	5.5	9.4	0.018	3.3	0.006	4.2	0.008
300	1.5	5.5	10.8	0.020	4.2	0.008	5.2	0.010
300	16.3	5.6	8.6	0.016	2.7	0.005	4.8	0.009
500	3.2	5.6	7.9	0.015	3.4	0.007	4.0	0.007
600	2.0	5.6	6.7	0.013	3.4	0.007	2.9	0.006
300	1.3	5.5	9.7	0.018	4.8	0.009	4.2	0.008
303	18.3	5.5	9.7	0.018	5.9	0.011	3.0	0.006
700	1.8	5.6	5.0	0.010	2.7	0.005	2.5	0.005
900	1.8	5.6	2.9	0.006	1.4	0.003	0.6	0.001
1100	1.5	5.7	-	-	-	-	-	-
300	25.3	5.5	10.2	0.019	4.5	0.008	3.0	0.006

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant - $S/w\rho$ (ρ is the sodium density)

*Based on counting statistics, 1σ corresponds to 0.4×10^{-8} gm Cs/cm²

I. THIRD NONISOTHERMAL TEST (6 wppm Cs, 25 wppm O)

Prior to starting the third nonisothermal test, the loop temperature was increased from 300 to 500°F. The elevations of the detector shield were changed, so that the detector on Test Section No. 1 monitored a volume immediately adjacent to the outlet from the isothermal portion of the section, and the detector on Test Section No. 2 monitored a volume immediately adjacent to the inlet to the isothermal portion of the section. The temperature of Test Section No. 2 was reduced to 306°F, and several data sets taken. The test section temperature was increased to loop temperature, and several more data sets taken. No deposition was measured at the inlet to Test Section No. 2 (after density correction, count rates at the two temperatures were within 0.1σ of each other). Background data indicated no substantial cesium deposition in the vicinity of the volumes being monitored.

The results of the third nonisothermal test are reported in Table II-9. These results are interesting, with low cesium deposition in the coldest test section and essentially constant deposition in the other test section. The large values of K (700°F) in Test Section No. 1 may indicate that all of these values are high.

TABLE II-9
RESULTS OF THIRD NONISOTHERMAL TEST

Time at Temperature (hr)	w (gm Cs/cm Na $\times 10^6$)	Loop Temperature, Excluding Test Sections (°F)	Test Section No. 1			Test Section No. 2		
			Temperature (°F)	S_1 (gm Cs/cm ² $\times 10^8$)	K_1 (cm)	Temperature (°F)	S_2 (gm Cs/cm ² $\times 10^8$)	K_2 (cm)
1.0	5.6	700	700	5.5	0.011	700	2.1	0.004
2.5	5.6	700	700	4.5	0.009	300	3.4	0.006
2.5	5.6	700	500	1.3	0.003	300	2.5	0.005
1.7	5.7	700	400	5.7	0.011	300	1.4	0.003
15.8	-	700	700	-	-	700	-	-
2.0	5.6	700	500	5.7	0.011	250	0.6	0.001
2.0	5.6	700	400	4.7	0.009	250	1.9	0.004
2.0	5.7	700	350	4.5	0.009	250	2.6	0.005
1.2	5.7	700	300	4.5	0.009	255	1.9	0.004
17.4	5.6	700	700	6.5	0.013	700	1.3	0.003
1.5	5.7	1000	1000	3.6	0.008	1000	1.3	0.003

w = Cesium concentration in the sodium

S = Cesium surface concentration, based on apparent surface area

K = Equilibrium Constant = $S/w\rho$ (ρ is the sodium density)

*Based on counting statistics, 1σ corresponds to 0.4×10^{-8} gm Cs/cm²

APPENDIX III
CALCULATION OF CESIUM DISTRIBUTION IN A REACTOR SYSTEM,
USING EQUILIBRIUM CONSTANT (K) APPROACH

SYSTEM DATA REQUIRED:

Total Sodium Volume
Surface Area of Individual Regions
Temperature of Each Region

SYSTEM INDEPENDENT DATA REQUIRED:

Equilibrium constant as a function of temperature
and degree of surface "activation"

This derivation assumes a burst release of cesium with sufficient circulating time to establish equilibrium in the reactor system. A similar derivation can be made without this restraint.

The mass of cesium adsorbed on the stainless steel in the i^{th} region is:

$$X_i = K_i A_i w \quad , \quad \dots \text{(III-1)}$$

where:

X_i = mass cesium adsorbed on the surface of the i^{th} region (gm Cs)

K_i = equilibrium constant for the i^{th} region $\left(\frac{\text{gm Cs ads/cm}^2}{\text{gm Cs sol/cm}^3} \right)$

A_i = surface area of i^{th} region (cm^2)

w = cesium concentration at equilibrium in the sodium (gm Cs sol/cm^3).

The total mass of cesium adsorbed on the walls is:

$$x = w \sum_{i=1}^n K_i A_i \quad , \quad \dots \text{(III-2)}$$

where:

x = total mass cesium adsorbed (gm Cs)

n = number of regions.

Since the total inventory of cesium in the reactor system (neglecting the cesium in the gas space) is the sum of the cesium adsorbed on the wall and in solution,

$$I = x + Vw \quad , \quad \dots \text{(III-3)}$$

where:

I = cesium inventory (gm Cs)

V = sodium volume (cc Na).

Substituting Equation III-2 into III-3 and rearranging,

$$w = \frac{I}{V + \sum_{i=1}^n K_i A_i} \quad \dots (III-4)$$

Rearranging Equation III-4, the fraction of the cesium inventory which is in solution in the sodium, f_ℓ , is:

$$f_\ell = \frac{wV}{I} = \frac{V}{V + \sum_{i=1}^n K_i A_i} \quad \dots (III-5)$$

Substituting Equation III-4 into III-1 and rearranging, the fraction of the cesium inventory adsorbed on the walls of the i^{th} region, f_i , is

$$f_i = \frac{X_i}{I} = \frac{K_i A_i}{V + \sum_{i=1}^n K_i A_i} \quad \dots (III-6)$$

Equations III-5 and III-6 are used to calculate the cesium distribution for the equilibrium case. They are readily and rapidly modified for the case where I is a function of time and/or a finite cold trap flow rate.

The following sample problems are presented to demonstrate the use of the equilibrium constant approach. It should be noted that core tank and fuel element surface areas should have been, but were not, included in the calculation, and no provision was made for cesium deposited on precipitated Na_2O .

SAMPLE PROBLEMS

CASE A – COLD TRAP SURFACES FULLY DEACTIVATED, REACTOR AT TEMPERATURE

Region	Temperature (°C)	Surface Area (cm ²)	K (cm)	KA (cm ³)	Fraction Cesium Inventory in Region
Hot leg	570	10 ⁷	0.002	2 x 10 ⁴	0.3 x 10 ⁻⁴
IHX, hot half	530	10 ⁸	0.002	3 x 10 ⁵	3 x 10 ⁻⁴
IHX, cold half	450	10 ⁸	0.003	3 x 10 ⁵	4 x 10 ⁻⁴
Cold leg	400	5 x 10 ⁶	0.003	1.5 x 10 ⁴	0.2 x 10 ⁻⁴
Cold trap	150	5 x 10 ⁶	0.010	5 x 10 ⁴	0.7 x 10 ⁻⁴
Sodium		Volume = 8 x 10 ⁸			~1.0
		$V + \sum K_i A_i = 8.00585 \times 10^8$			

CASE B – COLD TRAP SURFACES ACTIVATED TO SAME DEGREE AS FPBL, REACTOR AT TEMPERATURE

Region	Temperature (°C)	Surface Area (cm ²)	K (cm)	KA (cm ³)	Fraction Cesium Inventory in Region
Hot leg	570	10 ⁷	0.002	2 x 10 ⁴	3 x 10 ⁻⁵
IHX, hot half	530	10 ⁸	0.002	2 x 10 ⁵	3 x 10 ⁻⁴
IHX, cold half	450	10 ⁸	0.003	3 x 10 ⁵	4 x 10 ⁻⁴
Cold leg	400	5 x 10 ⁶	0.003	1.5 x 10 ⁴	2 x 10 ⁻⁵
Cold trap	150	5 x 10 ⁶	0.5	2.5 x 10 ⁶	3 x 10 ⁻³
Sodium		Volume = 8 x 10 ⁸			0.996
		$V + \sum K_i A_i = 8.03 \times 10^8$			

CASE C - COLD TRAP SURFACES HIGHLY ACTIVATED,
REACTOR AT TEMPERATURE

Region	Temperature (°C)	Surface Area (cm ²)	K (cm)	KA (cm ³)	Fraction Cesium Inventory in Region
Hot leg	570	10 ⁷	0.002	2 x 10 ⁴	2 x 10 ⁻⁵
IHX, hot half	530	10 ⁸	0.002	2 x 10 ⁵	2 x 10 ⁻⁴
IHX, cold half	450	10 ⁸	0.003	3 x 10 ⁵	4 x 10 ⁻⁴
Cold leg	400	5 x 10 ⁶	0.003	1.5 x 10 ⁴	2 x 10 ⁻⁵
Cold trap	150	5 x 10 ⁶	10.	5 x 10 ⁷	0.06
Sodium			Volume = 8 x 10 ⁸		0.94
			$V + \sum K_i A_i = 8.5 \times 10^8$		

CASE D - REACTOR COOLED TO 150°C, DEACTIVATED
SURFACES IN COLD TRAP

Region	Temperature (°C)	Surface Area (cm ²)	K (cm)	KA (cm ³)	Fraction Cesium Inventory in Region
Hot leg	150	10 ⁷	0.01	1 x 10 ⁵	1 x 10 ⁻⁴
IHX, hot half	150	10 ⁸	0.01	1 x 10 ⁶	1 x 10 ⁻³
IHX, cold half	150	10 ⁸	0.01	1 x 10 ⁶	1 x 10 ⁻³
Cold leg	150	5 x 10 ⁶	0.01	5 x 10 ⁴	6 x 10 ⁻⁵
Cold trap	150	5 x 10 ⁶	0.01	5 x 10 ⁴	6 x 10 ⁻⁵
Sodium			Volume = 8 x 10 ⁸		0.998
			$V + \sum K_i A_i = 8.02 \times 10^8$

APPENDIX IV
DERIVATION OF ANALYTICAL EXPRESSIONS FOR ISOTHERMAL AND
NONISOTHERMAL FPBL OPERATION, BARIUM EXPERIMENTS

Let:

I = total barium inventory, liquid + surface (gm Ba)

c = barium concentration in sodium (gm Ba/cc Na)

v = sodium flow rate (cc Na/hr)

K = barium liquid-surface equilibrium constant (gm Ba adsorbed/cm²/gm Ba in solution/cm³)

S = surface area (cm²)

V = sodium volume (cm³)

R = count rate Ba¹³³ from radiation detector (cpm)

ξ = surface area scanned by radiation detector (cm²)

η = sodium volume scanned by radiation detector (cm³)

ϵ = radiation detector net efficiency (cpm/gm Ba)

U = total barium associated with surface (gm Ba)

T = temperature (°F)

t = time (hr)

m = experimentally determined parameter, which, if the model works = $v(I/I_{\infty})/(KS + V)$

Subscripts:

1 = Region 1

2 = Region 2

0 = at zero time

∞ = as time $\rightarrow \infty$

a = temperature distribution for Run a (Table 7)

b = temperature distribution for Run b (Table 7)

Assumed temperature dependencies:

$$\eta \neq \eta(T)$$

$$S \neq S(T)$$

$$\xi \neq \xi(T)$$

$$\epsilon \neq \epsilon(T)$$

$$V_2 \neq V_2(T)$$

$$V_1 = V_1(T)$$

$$K = K(T)$$

$$I \neq I(T)$$

$$I_{1\infty} = I_{1\infty}(T)$$

$$I_{2\infty} = I_{2\infty}(T)$$

A. CASE 1

c_1 = solubility limit of barium at T_1

c_2 = solubility limit of barium at T_2

DATA DOES NOT FIT THIS MODEL

$$\frac{dI_1}{dt} = -v(c_1 - c_2) = \text{constant} = -\frac{dI_2}{dt}$$

$$I_{10} - I_1 = v(c_1 - c_2)t$$

$$I_2 - I_{20} = v(c_1 - c_2)t$$

B. CASE 2

Assume surface-liquid equilibrium constant can be used to express barium distribution; or

$$K_1 = \frac{U_1/S_1}{c_1} ,$$

$$K_2 = \frac{U_2/S_2}{c_2} ,$$

$$I_1 = K_1 c_1 S_1 + V_1 c_1 ,$$

$$c_1 = \frac{I_1}{K_1 S_1 + V_1} ,$$

$$c_2 = \frac{I_2}{K_2 S_2 + V_2} ,$$

$$\frac{dI_1}{dt} = -v \left(\frac{I_1}{K_1 S_1 + V_1} - \frac{I_2}{K_2 S_2 + V_2} \right) , \quad \dots \text{(IV-1)}$$

$$\frac{dI_2}{dt} = v \left(\frac{I_1}{K_1 S_1 + V_1} - \frac{I_2}{K_2 S_2 + V_2} \right) . \quad \dots \text{(IV-2)}$$

Equations IV-1 and IV-2 can be decoupled by

$$I = I_1 + I_2 ,$$

and solved to obtain:

$$\frac{I_1}{I_{10}} - \frac{I_{1\infty}}{I_{10}} = \left(1 - \frac{I_{1\infty}}{I_{10}}\right) \exp \left[-v \left(\frac{I/I_{1\infty}}{K_2 S_2 + V_2} \right) t \right] , \quad \dots (IV-3)$$

and

$$- \frac{K_2 S_2 + V_2}{v(I/I_{1\infty})} \frac{d}{dt} \left(\frac{I_1}{I_{10}} \right) + \frac{I_{1\infty}}{I_{10}} = \frac{I_1}{I_{10}} ; \quad \dots (IV-4)$$

$$\frac{I_{2\infty}}{I_{20}} - \frac{I_2}{I_{20}} = \left(\frac{I_{2\infty}}{I_{20}} - 1 \right) \exp \left[-v \left(\frac{I/I_{2\infty}}{K_1 S_1 + V_1} \right) t \right] , \quad \dots (IV-5)$$

and

$$- \frac{K_1 S_1 + V_1}{v(I/I_{2\infty})} \frac{d}{dt} \left(\frac{I_2}{I_{20}} \right) + \frac{I_{2\infty}}{I_{20}} = \frac{I_2}{I_{20}} . \quad \dots (IV-6)$$

Since

$$R = \text{surface count} + \text{liquid count} ,$$

$$R = \left(\frac{U}{S} \xi + c\eta \right) \epsilon .$$

Since

$$K = \frac{U/S}{c} ,$$

$$R = (K\xi c + \eta c) \epsilon .$$

However,

$$c = \frac{I}{KS + V} ,$$

$$R = \left(\frac{K\xi + \eta}{KS + V} \right) \epsilon I . \quad \dots (IV-7)$$

Therefore, for comparing data at constant temperature from the same detector,

$$\frac{I_1}{I_{10}} = \frac{R_1}{R_{10}} , \quad \dots (IV-8)$$

$$\frac{I_2}{I_{20}} = \frac{R_2}{R_{20}} \quad \dots \text{(IV-9)}$$

When comparing data from the same detector at two different temperatures,

$$\frac{R_1(T_1)}{R_1(T_2)} = \frac{I_1(T_1)}{I_1(T_2)} \left[\frac{K_1(T_1)\xi_1 + \eta_1}{K_1(T_2)\xi_1 + \eta_1} \frac{K_1(T_2)S_1 + V_1(T_2)}{K_1(T_1)S_1 + V_1(T_1)} \right] \quad \dots \text{(IV-10)}$$

$$\frac{R_2(T_1)}{R_2(T_2)} = \frac{I_2(T_1)}{I_2(T_2)} \left[\frac{K_2(T_1)\xi_2 + \eta_2}{K_2(T_2)\xi_2 + \eta_2} \frac{K_2(T_2)S_2 + V_2}{K_2(T_1)S_2 + V_2} \right] \quad \dots \text{(IV-11)}$$

The preceding equations can be used to compare data from different detectors at different temperatures with the addition of the factor, ϵ_1/ϵ_2 , or

$$\frac{R_1(T_1)}{R_2(T_2)} = \frac{I_1(T_1)}{I_2(T_2)} \frac{\epsilon_1}{\epsilon_2} \left[\frac{K_1(T_1)\xi_1 + \eta_1}{K_2(T_2)\xi_2 + \eta_2} \frac{K_2(T_2)S_2 + V_2}{K_1(T_1)S_1 + V_1(T_1)} \right] \quad \dots \text{(IV-12)}$$

Based on experimental data obtained from the barium tests, the numerical values of $K_i S_i + V_i$ were very much larger than V_i . Equations IV-10, IV-11, and IV-12 can therefore be simplified by letting $K_i S_i + V_i \cong K_i S_i$. Equation IV-7 then reduces to:

$$R = \frac{I \epsilon \xi}{S} \quad \dots \text{(IV-13)}$$

Then, Equations IV-10 and IV-11 reduce to

$$\frac{R_1(T_1)}{R_1(T_2)} = \frac{I_1(T_1)}{I_1(T_2)} \quad \dots \text{(IV-14)}$$

$$\frac{R_2(T_1)}{R_2(T_2)} = \frac{I_2(T_1)}{I_2(T_2)} \quad \dots \text{(IV-15)}$$

and Equation IV-12 reduces to

$$\frac{R_1(T_1)}{R_2(T_2)} = \frac{I_1(T_1)}{I_2(T_2)} \frac{S_2}{S_1} \frac{\epsilon_1 \xi_1}{\epsilon_2 \xi_2} \quad \dots (IV-16)$$

C. MATERIAL BALANCE:

$$I = I_1 + I_2$$

$$I = R_1 \left(\frac{S_1}{\epsilon_1 \xi_1} \right) + R_2 \left(\frac{S_2}{\epsilon_2 \xi_2} \right)$$

$$I_a = I_b$$

$$R_{1a} \left(\frac{S_1}{\epsilon_1 \xi_1} \right) + R_{2a} \left(\frac{S_2}{\epsilon_2 \xi_2} \right) = R_{1b} \left(\frac{S_1}{\epsilon_1 \xi_1} \right) + R_{2b} \left(\frac{S_2}{\epsilon_2 \xi_2} \right)$$

$$R_{1b} = R_{1a} + (R_{2a} - R_{2b}) \frac{S_2}{S_1} \frac{\epsilon_1 \xi_1}{\epsilon_2 \xi_2}$$

D. ANALYTICAL PROCEDURE

- 1) Subtract background radiation from countrate to obtain background corrected countrate, R_1 and R_2 .
- 2) Divide R_1 and R_2 by values of R_1 and R_2 at start of particular test to obtain R_1/R_{10} and R_2/R_{20} .
- 3) Plot R_1/R_{10} and R_2/R_{20} as function of time. (From Equations IV-8 and IV-9, equivalent to I_1/I_{10} and I_2/I_{20})

- 4) Using an analog computer, differentiate preceding plot, and obtain new plots of R_i/R_{i0} vs $\frac{d}{dt} (R_i/R_{i0})$. If the model works, this should be a straight line.
- 5) Extrapolate plots to $\frac{d}{dt} (R_i/R_{i0}) = 0$; and, according to Equations IV-4 and IV-6, the intercepts should be $I_{1\infty}/I_{10}$ and $I_{2\infty}/I_{20}$.
- 6) Calculate the parameters:

$$\ln \left(\frac{1 - \frac{I_{1\infty}}{I_{10}}}{\frac{R_1}{R_{10}} - \frac{I_{1\infty}}{I_{10}}} \right)$$

and

$$\ln \left(\frac{\frac{I_{2\infty}}{I_{20}} - 1}{\frac{I_{2\infty}}{I_{20}} - \frac{R_2}{R_{20}}} \right),$$

and plot against t .

- 7) If the model works, should obtain straight lines with slopes m_1 and m_2 , equal (according to Equations IV-3 and IV-5) to:

$$m_1 = v \left(\frac{I/I_{1\infty}}{K_2 S_2 + V_2} \right) ,$$

$$m_2 = v \left(\frac{I/I_{2\infty}}{K_1 S_1 + V_1} \right) .$$

E. OBTAINING $\epsilon_1 \xi_1 / \epsilon_2 \xi_2$ AND K

After nonisothermal tests, when the loop is operated isothermally, the following kinetic results are obtained:

$$\frac{I_1}{I_{10}} - \frac{I_{1\infty}}{I_{10}} = \left(1 - \frac{I_{1\infty}}{I_{10}} \right) \exp \left[-v \left(\frac{I/I_{1\infty}}{K S_2 + V_1} \right) t \right] ,$$

$$\frac{I_2}{I_{20}} - \frac{I_{2\infty}}{I_{20}} = \left(1 - \frac{I_{2\infty}}{I_{20}} \right) \exp \left[-v \left(\frac{I/I_{2\infty}}{K S_1 + V_1} \right) t \right] .$$

Parameters I/I_{∞} and m can be obtained as before.

If the model works, and again assuming $KS \gg V$,

$$\frac{m_1}{m_2} = \frac{v \left(\frac{I}{I_{1\infty} K S_2} \right)}{v \left(\frac{I}{I_{2\infty} K S_1} \right)} ,$$

$$\frac{m_1}{m_2} = \frac{I_{2\infty}/S_2}{I_{1\infty}/S_1} .$$

Since it is assumed that the barium is uniformly distributed,

$$\frac{I_{2\infty}}{S_2} = \frac{I_{1\infty}}{S_1} ,$$

or

$$\frac{m_1}{m_2} = 1 \quad .$$

From Equation IV-13,

$$\frac{I_{1\infty}/S_1}{I_{2\infty}/S_2} = \frac{R_{1\infty}/\epsilon_1 \xi_1}{R_{2\infty}/\epsilon_2 \xi_2} = 1 \quad , \quad \dots (IV-17)$$

$$\frac{\epsilon_2 \xi_2}{\epsilon_1 \xi_1} = \frac{R_{2\infty}}{R_{1\infty}} \quad . \quad \dots (IV-18)$$

Equation IV-18 permits evaluation of this critical parameter.

Now, from the value of m_1 or m_2 ,

$$m_1 = \frac{v}{KS_2} \left(\frac{I_{1\infty} + I_{2\infty}}{I_{1\infty}} \right) \quad ,$$

$$m_1 = \frac{v}{KS_2} \left(1 + \frac{I_{2\infty}}{I_{1\infty}} \right) \quad .$$

From Equation IV-17,

$$\frac{I_{2\infty}}{I_{1\infty}} = \frac{S_2}{S_1} \quad .$$

Substituting,

$$m_1 = \frac{v}{K} \left(\frac{1}{S_1} + \frac{1}{S_2} \right) \quad ,$$

or

$$K = \frac{v}{m_1} \left(\frac{1}{S_1} + \frac{1}{S_2} \right) \quad . \quad \dots (IV-19)$$

Using the experimentally obtained values of m , v , and the apparent system surface areas, an absolute value of K can be obtained at the loop isothermal temperature.

F. OBTAINING RELATIVE K 's FROM NONISOTHERMAL TESTS

For a particular nonisothermal test,

$$m_1 = \frac{v(I/I_{1\infty})}{K(T_2) S_2 + V_2} ,$$

$$m_2 = \frac{v(I/I_{2\infty})}{K(T_1) S_1 + V_1(T_1)} .$$

Again assuming $KS \gg V$, the preceding equations can be divided and rearranged to yield

$$\frac{K(T_2)}{K(T_1)} = \frac{I_{2\infty}}{I_{1\infty}} \frac{S_1}{S_2} \frac{m_2}{m_1} .$$

Substituting Equation IV-13,

$$\frac{K(T_2)}{K(T_1)} = \frac{R_{2\infty}}{R_{1\infty}} \frac{\epsilon_1 \xi_1}{\epsilon_2 \xi_2} \frac{m_2}{m_1} . \quad \dots (IV-20)$$

G. VELOCITY DEPENDENCE

Runs a and b (Table 7) were at the same temperature distribution, but at different sodium velocities. To determine the predicted velocity dependence:

$$m_{2a} = \frac{v_a(I/I_{2\infty})}{K(T_1)S_1 + V_1} ,$$

$$m_{2b} = \frac{v_b(I/I_{2\infty})}{K(T_1)S_1 + V_1} .$$

Dividing the two,

$$\frac{v_a}{v_b} = \frac{m_{2a}}{m_{2b}} \quad \dots (IV-21)$$

This can be compared to the velocity ratio determined from heat balances and flowmeter readings.

H. MATERIAL BALANCE BY INTERCOMPARISON OF RUNS

The purpose of this analytical test is to determine whether, within experimental accuracy, the total barium inventory (I) in the loop is constant.

Since

$$m_{2a} = \frac{v_a I_a / I_{2\infty a}}{K(700)S_1 + V_1} \quad ,$$

and

$$m_{2c} = \frac{v_c I_c / I_{2\infty c}}{K(700)S_1 + V_1} \quad ,$$

then

$$\frac{I_a}{I_c} = \frac{m_{2a}}{m_{2c}} \frac{I_{2\infty a}}{I_{2\infty c}} \frac{v_c}{v_a} \quad .$$

Substituting Equation IV-13,

$$\frac{I_a}{I_c} = \frac{m_{2a}}{m_{2c}} \frac{R_{2\infty a}}{R_{2\infty c}} \frac{v_c}{v_a} \quad \dots (IV-22)$$