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**CHEMISTRY OF DEFECT TEST ON HIGHLY RATED
SINTERED UO₂ FUEL TEST X-2-q**

EXP-NRX-1806

by

G.M. ALLISON and I.H. CROCKER

Chalk River, Ontario

June 1959

AECL No. 906

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ABSTRACT

A highly rated ($\int k.d\theta = 48 \text{ W/cm}$) sintered UO_2 specimen, 0.8 in. O.D., with a 0.010-in. diameter hole through the Zircaloy sheath, was irradiated in the X-2 loop in the NRX reactor for approximately 16 days.

For this irradiation period the escape-rate coefficients calculated for the fission products Kr-88, Cs-138, I-135, I-133 and I-131 were similar to those found in previous defect tests on UO_2 fuel operated at considerably lower heat ratings.

The test was terminated due to high activity in the loop water which occurred during a reactor trip-startup cycle. Post-irradiation examination of the defected fuel element indicated no reason for the activity surge.

CHEMISTRY OF DEFECT TEST ON HIGHLY RATED SINTERED UO₂ FUELTest X-2-q1. INTRODUCTION

In a previous test in the X-2 loop in the NRX reactor (test X-2-p) (1) an uranium oxide fuel specimen of relatively large diameter, i.e. 0.75 in. was irradiated at a heat rating of 630 W/cm length, slightly above the maximum expected in NPD-2. In that test no defected fuel elements were present. The behaviour of defected UO₂ fuel has been studied previously in NRX in AECL and BAPL* loop tests (2,3) but most of this fuel was rated at considerably less than 600 W/cm length. It was therefore considered important to test a UO₂ fuel element of NPD-2 heat rating in a defected condition.

The original proposal and addendum for test X-2-q (4) provided for the irradiation of flat UO₂ specimens (5) and U₃Si specimens as well as the defected sintered UO₂ element. It was also intended to test a defected swaged UO₂ specimen of high heat rating. Due to the failure of a swaged specimen and a U₃Si specimen at the beginning of the test (6), the defect test was finally carried out with only the defected sintered UO₂ element present in the loop test section.

* - Bettis Atomic Power Laboratory, Pittsburgh, Penn.

2. OBJECTIVES OF THE TEST

There were three main objectives of this defect test, viz.-

- (a) To determine if highly-rated sintered UO_2 fuel of this large diameter could survive irradiation in a defected condition.
- (b) To study the release of fission products to the loop water and to compare this release with that observed in other defect tests on fuel of lower heat rating.
- (c) To determine by post-irradiation examination of the oxide the effect of the defect on such things as grain growth and O/U ratio.

With regard to objective (a) the worst conditions for survival of a defected element prevailed due to the large number of reactor trips and startups which occurred during the test. For objective (b) the lack of steady reactor operation made it difficult to achieve equilibrium conditions with respect to the concentration of fission products in the loop water except for the shorter-lived nuclides.

3. PREPARATION AND DETAILS OF THE DEFECTED UO_2 FUEL SPECIMEN

The uranium oxide was prepared by hydrogen reduction of ammonium diuranate at 950°C. The pellets were cold compacted at 20,000 psi using a wax binder and were sintered in hydrogen for 1-2 hours at 1700°C(7). The pellets were ground in a centreless grinder

to provide a cold diametral clearance when assembled in the sheath of 0.007 inches. An axial clearance of approximately 2% was allowed between the oxide and the Zircaloy-2 end cap.

Details of the assembled fuel element are given in Table I and heat rating data in Table II.

TABLE I

DETAILS OF DEFECTED SINTERED UO₂ FUEL ELEMENT

| | |
|------------------------------------|------------------------|
| Length of UO ₂ | 5.75 inches |
| Diameter of UO ₂ | 0.746 inches |
| Average Density of UO ₂ | 10.2 g/cm ³ |
| Weight of UO ₂ | 419 g |
| Enrichment | 1.9 w/o U-235 in U |
| Oxygen to Uranium Ratio in Fuel | 2.007 |
| O.D. of Fuel Element | 0.803 inches |
| Cold Diametral Clearance | 0.007 inches |
| Cold Axial Clearance | ~2% of fuel length |
| Cladding | Zircaloy-2 |
| Cladding Thickness | 0.025 inches |
| Diameter of Defect Hole | 0.010 inches |

TABLE II

HEAT RATING DATA*

| | |
|---------------------------------------|--|
| Heat Rating per unit length | 628 W/cm |
| T_{center} k.dθ $T_{surface}$ | 48 W/cm |
| Surface Heat Flux | |
| (1) Fuel/Sheath | 105 W/cm ² |
| (2) Sheath/Water | 99 W/cm ² (310,000 BTU/ft ² ·hr) |
| Heat Output | 9.1 kilowatts |

* - Obtained from calorimetric measurements and datatron program (8)

4. THE X-2 LOOP

The X-2 loop is a stainless steel system in which water can be recirculated at high temperature and pressure through a test section positioned in the NRX reactor. A flow diagram of the loop is given in Figure 1. A complete description of the system can be found in a previous report (9).

5. LOOP WATER CONDITIONS

The water conditions which prevailed in the loop during the defect test are given in Table III.

TABLE III

LOOP WATER CONDITIONS

| | |
|--------------------------|--------------------------------------|
| Temperature | |
| Inlet to test section | 520°F |
| Outlet from test section | 527°F |
| Pressure | 1800 psi |
| Main loop flow rate | 10 U.S.gpm |
| Ion Exchange flow rate | 0.18 U.S.gpm |
| Oxygen concentration | <0.1 cm ³ /kg (<0.14 ppm) |
| Hydrogen concentration | >10 cm ³ /kg |
| pH | 9.5 - 10.0 |
| Conductivity | 10 - 30 μ mhos cm ⁻¹ |

The pH was maintained with a column of mixed-bed resin in the K-OH form (Rohm and Haas XE-149). A column of cation resin in the potassium form (Rohm and Haas XE-151) was used upstream from the iodine monitor.

6. OUTLINE OF THE TEST

The fuel element was inserted in the loop test section in the reactor during the week of December 7, 1958. The loop water was degassed until the oxygen concentration was 0.05 ppm. When degassing was stopped sufficient hydrogen was added to the loop to give a concentration in the water of about 50 cm³/kg.

The reactor started on December 13 and the test proceeded satisfactorily until December 29. On that day, after a reactor trip and startup, very high activity in the loop water and high radiation fields at the loop piping indicated that the test should be terminated and the fuel was removed from the loop.

7. BEHAVIOUR OF THE MONITORS

Three activity monitors were in use during this defect test, namely, the delayed neutron (D.N.), gamma and iodine monitors. These monitors have been described in detail in previous reports (9,10) but very briefly the D.N. monitor employed a BF_3 tube; the gamma monitor was simply a Geiger tube held against a section of the loop piping and housed in a lead castle; the I_2 monitor consisted of a NaI scintillation crystal viewing a coil of loop piping downstream from a cation resin column. The pulse amplitude distribution from the I_2 monitor was biased so that only gamma rays with energies above 0.9 MeV were counted. The outputs from all the monitors were recorded on Esterline Angus recorders.

The record of the counting rates of the three monitors is shown in Figure 2 along with reactor power. It can be seen that all the monitors showed the usual peaking at reactor start-ups associated with waterlogging of the fuel (2). The following points should be noted:

- (1) The lack of steady reactor operation made it difficult to estimate the steady-state activity level of the monitors. An approximate estimate is given in Table IV.
- (2) In the period December 22-23 oscillation of the loop water temperature occurred due to failure of the automatic temperature controller requiring the temperature to be adjusted manually. At this time the activity as recorded by the three monitors also oscillated. This is shown in more detail in Figure 3.
- (3) When the reactor was shut down at 1600 hours on December 24 the activity as recorded by the γ and I_2 monitors rose somewhat but there was no spike on the D.N. record (Fig. 2). A rise in activity on reactor shutdowns has been noted in other tests but has always been associated with an enlargement of the defect hole or a split in the sheath. In these instances a spike was also apparent on the D.N. monitor record.

The monitor records for the reactor startup on December 26 looked normal and did not indicate a large defect hole or the presence of an additional defect.

- (4) When the reactor tripped at 1624 hours on December 29 the gamma and iodine-monitor counting rates rose and the D.N. activity spiked slightly. The monitor records for this period have been plotted in more detail in Figure 4.

(5) At 1642 hours on December 29 the reactor started again and reached a power of about 5 MW at 1652 hours, when another trip occurred. On this startup the monitors indicated very high activity in the loop water even though the reactor power reached only 5 MW. The D.N. monitor recorded a maximum of 6×10^5 cpm. On the trip the slow drop in the D.N. counting rate indicated that the BF_3 tube was being affected by the high γ field in the loop water. The iodine monitor rose to 10^6 cpm and then stopped counting as the activity got too high for resolution of the pulses by the electronic equipment. The gamma monitor went off scale ($>25,000$ cpm).

TABLE IV

STEADY-STATE MONITOR READINGS (cpm)

| | |
|--------------|-------------------|
| D.N. | 1.2×10^4 |
| γ | 1000 |
| I_2 | 4×10^5 |

8. RADIOCHEMICAL RESULTS

8.1 Methods:

Cesium and iodine determinations were made using standard radiochemical methods and techniques (11). The separated activity was mounted on filter paper and counted with a proportional counter. I-131, I-133 and I-135 were determined by resolution of decay curves. No correction was made for growth of Xe activities. I-131 results are considered to be accurate to $\pm 5\%$ but the other two iodines to only $\pm 20\%$.

Gross beta-gamma activity was determined by counting a source prepared by evaporating an aliquot of loop water to dryness on an aluminum planchet. A proportional counter was again used.

Fission-product gas activities were determined with a 20-channel pulse height analyser employing a 1-1/2" x 1" NaI(Tl) scintillation crystal (12). The loop water was sampled into an evacuated flask and the gas was pumped from the water using a Toepler system (13). A known fraction of the total gas was then transferred to a calibrated counting bulb.

8.2 Fission-Product Results:

Fission-product results are given in Table V and are plotted in Figure 5. Whenever possible samples were taken sufficiently long after a reactor startup to avoid high results due to the surge of activity caused by the waterlogging effect. The effect of reactor power changes on the fission product activity level is shown in Figure 5 by the results on the samples of December 18 which were taken 10 minutes after the reactor power had been raised by 7 MW.

Up to December 29 the Kr-88 and Cs-138 activities remained fairly level but the iodine activities increased very sharply during the period Dec. 19-21 and then more slowly until the end of the test. The sharp increase between Dec. 19-21 is considered to be due to the large number of

reactor trips and startups in this period (see Figure 2), the effect being greatest for the longest lived, i.e. I-131.

At 1624 hours on December 29 the reactor tripped. At 1642 it started up and tripped again at 1652 after reaching about 5 MW power. It was at this startup that the monitors began indicating very high activity in the loop water. The loop was sampled 78 minutes after the reactor had shut down. Due to the high radiation field coming from the sample (~ 40 R/h at contact with a bottle containing 50 ml) no determination of fission-product gases was made. Only Cs-138 and Cs-137 were determined at that time. Later I-131 and I-133 were also run. These results are given in Table V.

Because of this high activity in the coolant and the high radiation fields at the loop piping (10 R/h in the upper header room) the test was terminated.

TABLE V

FISSION-PRODUCT RESULTS (dpm/ml)

| Sample Time | <u>Kr</u> ⁸⁸ | <u>Cs</u> ¹³⁸ | <u>Cs</u> ¹³⁷ | <u>I</u> ¹³¹ | <u>I</u> ¹³³ | <u>I</u> ¹³⁵ | <u>Gross</u> $\beta\gamma$ |
|-----------------------------|-------------------------|--------------------------|--------------------------|-------------------------|-------------------------|-------------------------|----------------------------|
| 1038 Dec. 16 | 2.10×10^7 | 8.95×10^6 | 79 | 1.2×10^4 | 6.5×10^4 | 2.9×10^5 | 5.5×10^6 |
| 0950 Dec. 18 | 7.38×10^6 | 1.00×10^6 | 330 | 5.5×10^3 | 3.6×10^4 | 8.4×10^4 | |
| 0915 Dec. 19 | | 1.62×10^6 | | 3.3×10^3 | 2.3×10^4 | 7.2×10^4 | |
| 1109 Dec. 19 | 9.91×10^6 | | | | | | |
| 1158 Dec. 21 | | 7.30×10^5 | 280 | 5.5×10^4 | 1.6×10^5 | 6.1×10^5 | |
| 0954 Dec. 22 | | 1.04×10^6 | 570 | | | | |
| 1100 Dec. 22 | | | | 3.9×10^4 | 1.1×10^5 | 7.3×10^5 | |
| 1224 Dec. 22 | 1.18×10^7 | | | | | | |
| 0929 Dec. 24 | 9.08×10^6 | | | | | | |
| 0933 Dec. 29 | 1.52×10^7 | | | | | | |
| 1024 Dec. 29 | | 1.97×10^6 | 50 | | | | |
| 1336 Dec. 29 | | | | 3.4×10^5 | 7.5×10^5 | 1.7×10^6 | 7.2×10^5 |
| 1652 Dec. 29 (Trip Time) | | 6.80×10^7 | 1.2×10^5 | 5.9×10^7 | 5.1×10^7 | | |

* - Gross $\beta\gamma$ results are cpm/ml.

8.3 Escape-Rate Coefficients

The escape-rate coefficient, ν , is defined as the proportionality constant relating the total amount of an isotope in the fuel to the rate of escape to the external system. The complete equations for calculating ν have been given in other reports (14,2). For this test ν has been calculated for Kr-88, Cs-138 and three iodine isotopes using the expression

$$\nu = \frac{(\lambda N)_w (\beta + \lambda)}{N^{235} \sigma \phi Y (1 - e^{-\lambda t})}$$

where $(\lambda N)_w$ = total activity of a specific nuclide in the loop water, dps

λ = radioactive decay constant of the nuclide, sec^{-1}

β = $\frac{re}{v} \text{ sec}^{-1}$, purification constant

r = flow rate to resin column, g/sec

e = efficiency of resin

v = mass of flowing water in loop, g

N^{235} = U-235 in fuel, atoms

σ = fission cross section of U-235 for thermal neutrons, cm^2

ϕ = thermal neutron flux in fuel specimen at full reactor power, $\text{n/cm}^2\text{-sec}$

Y = fission yield of nuclide

t = equivalent time at full reactor power, sec

For steady-state conditions, i.e. $t > 4$ half-lives of the nuclide, $(1 - e^{-\lambda t}) \approx 1$. Of the nuclides considered here,

I-131 ($t_{1/2} = 8$ d) is the only one for which this factor had to be evaluated. Decay during reactor shutdown periods has been neglected. The values for these parameters used in calculating the escape-rate coefficients are given in Table VI.

TABLE VI

NUMERICAL VALUES OF PARAMETERS FOR CALCULATION OF ν

| | | | | |
|--|---|------------------------|------------------------|-----------------------------|
| U-235 in defected specimen | 1.80 x 10 ²² atoms | | | |
| ⁺ Average 2200 m/sec thermal neutron flux in the specimen | 3.1 x 10 ¹³ n/cm ² -sec | | | |
| 2200 m/sec fission cross section of U-235 | 565 x 10 ⁻²⁴ cm ² | | | |
| Flow rate to resin column (measured at 375°F) | 9.9 g/sec | | | |
| Mass of flowing water in loop | 60 kg | | | |
| Equivalent time at full power as of 0000 hours Dec. 19 | - 88 hours | | | |
| 0000 hours Dec. 23 | - 152 hours | | | |
| 1336 hours Dec. 29 | - 267 hours | | | |
| | | | | |
| <u>Kr⁸⁸</u> | <u>Cs¹³⁸</u> | <u>I¹³⁵</u> | <u>I¹³³</u> | <u>I¹³¹</u> |
| Fission yields, % | 3.7 | 5.8 | 6.4 | 6.5 |
| λ , sec ⁻¹ | 6.95x10 ⁻⁵ | 3.62x10 ⁻⁴ | 2.89x10 ⁻⁵ | 9.25x10 ⁻⁶ |
| β , sec ⁻¹ | 0 | 1.49x10 ⁻⁴ | | 1.65x10 ⁻⁴ |

* - Efficiency of resin for removing Cs-138 is taken as 0.9, for iodines, 1.

+ - The thermal neutron flux in the specimen was determined from the measured heat output of the fuel using the expression

$$\phi = 2.40 \times 10^{10} \times \frac{\text{W/g U}}{\text{fraction U-235 in U}} \text{ n/cm}^2\text{-sec} \quad (15)$$

Since the nuclides considered were gaseous or very soluble it was assumed that the amount depositing on the system surfaces was sufficiently small to be neglected. Similarly, no correction has been applied for loss through leakage since the leak rate was only ~ 1 kg water per day or less (2). Table VII contains the values of v calculated.

TABLE VII
ESCAPE-RATE COEFFICIENTS

| <u>Period in Test</u> | <u>Nuclide</u> | <u>dpm/ml</u> | <u>v (sec$^{-1}$)</u> |
|---------------------------------|----------------|------------------------------|---|
| * Average for test | Kr-88 | $(1.07 \pm 0.4) \times 10^7$ | $(6.7 \pm 2.5) \times 10^{-8}$ |
| " " " | Cs-138 | $(1.27 \pm 0.5) \times 10^6$ | $(3.7 \pm 1.5) \times 10^{-8}$ |
| Dec. 18-19 | I-135 | 7.8×10^4 | 7.5×10^{-10} |
| | I-133 | 3.0×10^3 | 2.7×10^{-10} |
| | I-131 | 4.4×10^3 | 3.1×10^{-10} |
| Dec. 21-23 | I-135 | 6.7×10^5 | 6.4×10^{-9} |
| | I-133 | 1.3×10^4 | 1.1×10^{-9} |
| | I-131 | 4.8×10^4 | 2.1×10^{-9} |
| Dec. 29 before final startup | I-135 | 1.7×10^6 | 1.6×10^{-8} |
| | I-133 | 7.5×10^5 | 6.4×10^{-9} |
| | I-131 | 3.4×10^5 | 1.0×10^{-8} |

* - The averages for Kr-88 and Cs-138 do not include the first result, which was suspected to be high due to the reactor power change, nor the results obtained after the activity surge on Dec. 29.

9. POST-IRRADIATION EXAMINATION OF THE FUEL SPECIMEN

Some details of the post-irradiation examination of the defected fuel specimen have been given by A.S. Bain in a Chemistry and Metallurgy Divisional Progress Report (16) and will be presented in more detail later. His findings may be summarized as follows:

- (a) The drilled defect had not altered in size or shape; the edge of the hole was still sharp and there was no sign of corrosion.
- (b) There were no changes in the length or diameter of the specimen.
- (c) There was no other defect visible on the sheath.
- (d) The uranium oxide had cracked radially and circumferentially.
- (e) There was a large amount of grain growth along the length of the specimen. No central void was observed but near the top of the specimen the oxide appeared quite porous.
- (f) There was a liberal amount of yellow deposit throughout the specimen.

10. DISCUSSION OF RESULTS

Escape-rate coefficients, obtained by BAPL from defect tests on "reference PWR" UO_2 in the X-1 loop in the NRX reactor, have been compared with results from this test in Table VIII.

TABLE VIII

COMPARISON OF BAPL AND X-2-q ESCAPE-RATE COEFFICIENTS

| | <u>BAPL</u> | <u>X-2-q</u> |
|---|---------------------------------------|----------------------|
| $\int k(\theta) d\theta$ W/cm | 24 | 48 |
| UO ₂ density g/cm ³ | 10.2 | 10.2 |
| Grain growth | Trace | Extensive |
| ν (sec ⁻¹) | | |
| Kr-88 | $1 \times 10^{-8} - 2 \times 10^{-8}$ | 6.4×10^{-8} |
| Cs-138 | $2 \times 10^{-8} - 4 \times 10^{-8}$ | 3.5×10^{-8} |
| I-131 | $10^{-9} - 10^{-8}$ | 1×10^{-8} * |
| I-133 | $1 \times 10^{-8} - 5 \times 10^{-8}$ | 6×10^{-9} * |

* - Values obtained just prior to activity burst which terminated the test.

It can be seen that the values of ν obtained in this test were quite similar to those found by BAPL in the X-1-L test (17). Considering the difference in heat rating and therefore the operating temperature of the fuels, one would have expected a considerably higher rate of escape of fission products from the X-2-q fuel than from the PWR UO₂ since this rate is dependent on diffusion in the oxide lattice and thus on the central fuel temperature. It is quite possible, however, that the large rise in activity in the loop water on December 29 was the beginning of a considerably higher fission product escape rate which might have continued with further irradiation.

Post-irradiation examination of the fuel (sec. 9) indicated no obvious change which could have caused the rise in activity release. It is clear that a further defect test on a similar fuel element will be required to clarify the situation.

11. SUMMARY

A highly rated ($\int k.d\theta = 48 \text{ W/cm}$) UO_2 fuel specimen with a 0.010 in. diameter hole drilled through the sheath has been irradiated in the X-2 loop in the NRX reactor.

For about two weeks the release rates of fission products were similar to those found by BAPL for defected "reference PWR" UO_2 operated at a fuel temperature below that required to give extensive grain growth. The test was then terminated due to a surge of activity which began with a reactor trip and which continued during the subsequent startup.

Post-irradiation examination of the fuel specimen revealed no change in the defect hole or in the dimensions of the specimen and no further defects. Extensive grain growth had occurred but there was no central void formed. The reason for the surge of activity is not obvious and a further test on a similar defected specimen is required.

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NOTE:

Many of the reports in the Exp-NRX-series, particularly those offering proposals for irradiations and methods of

fabrication of fuel specimens, are preliminary reports published before the experiment has concluded. They are printed in very limited quantities and hence are not generally available. Others, reporting results on chemistry and irradiation examinations, are final reports on the test and are given a much wider distribution.

FIGURE 1
LOOP X-2 FLOW DIAGRAM

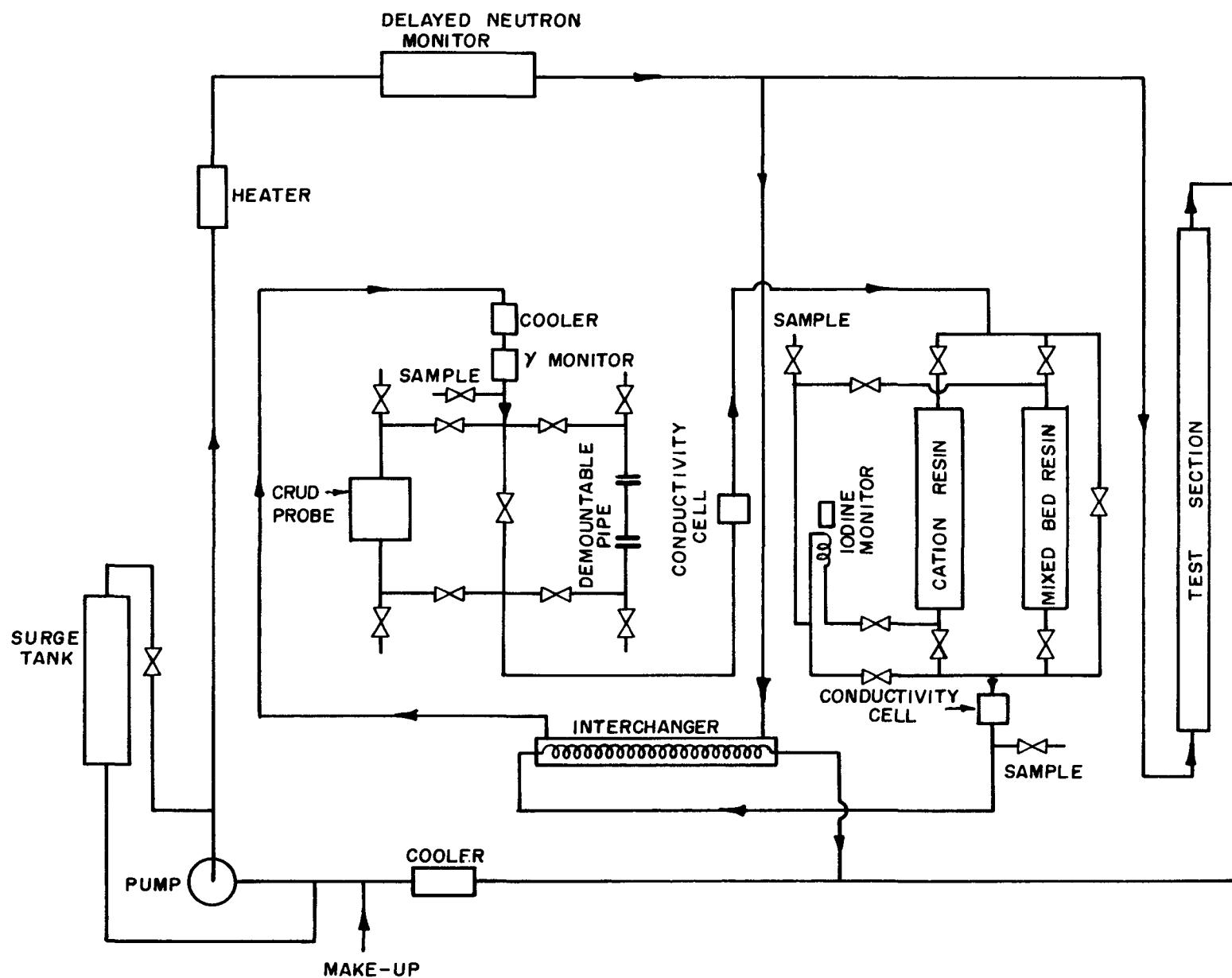


FIGURE 2 REACTOR POWER & MONITOR RECORDS

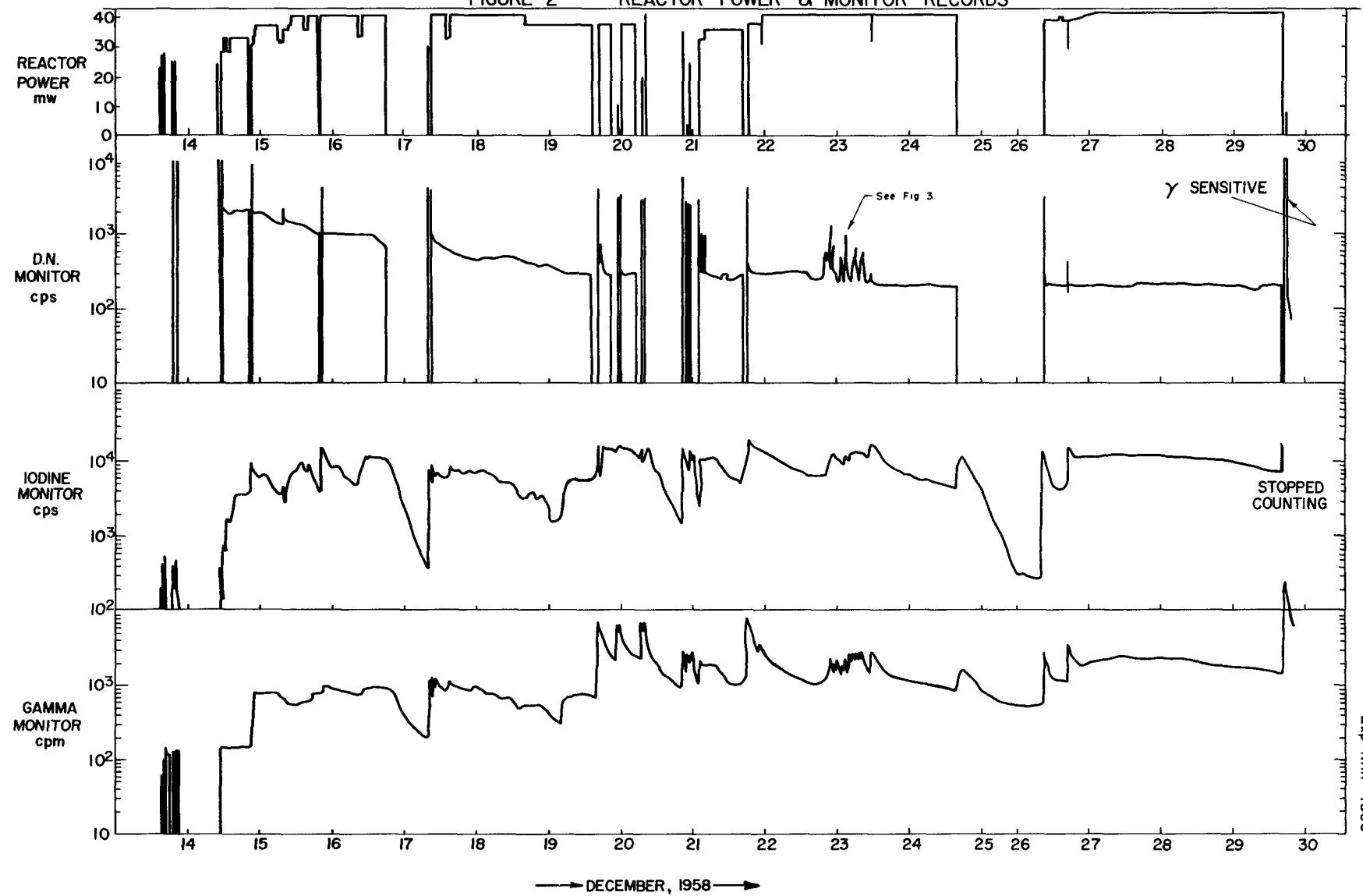
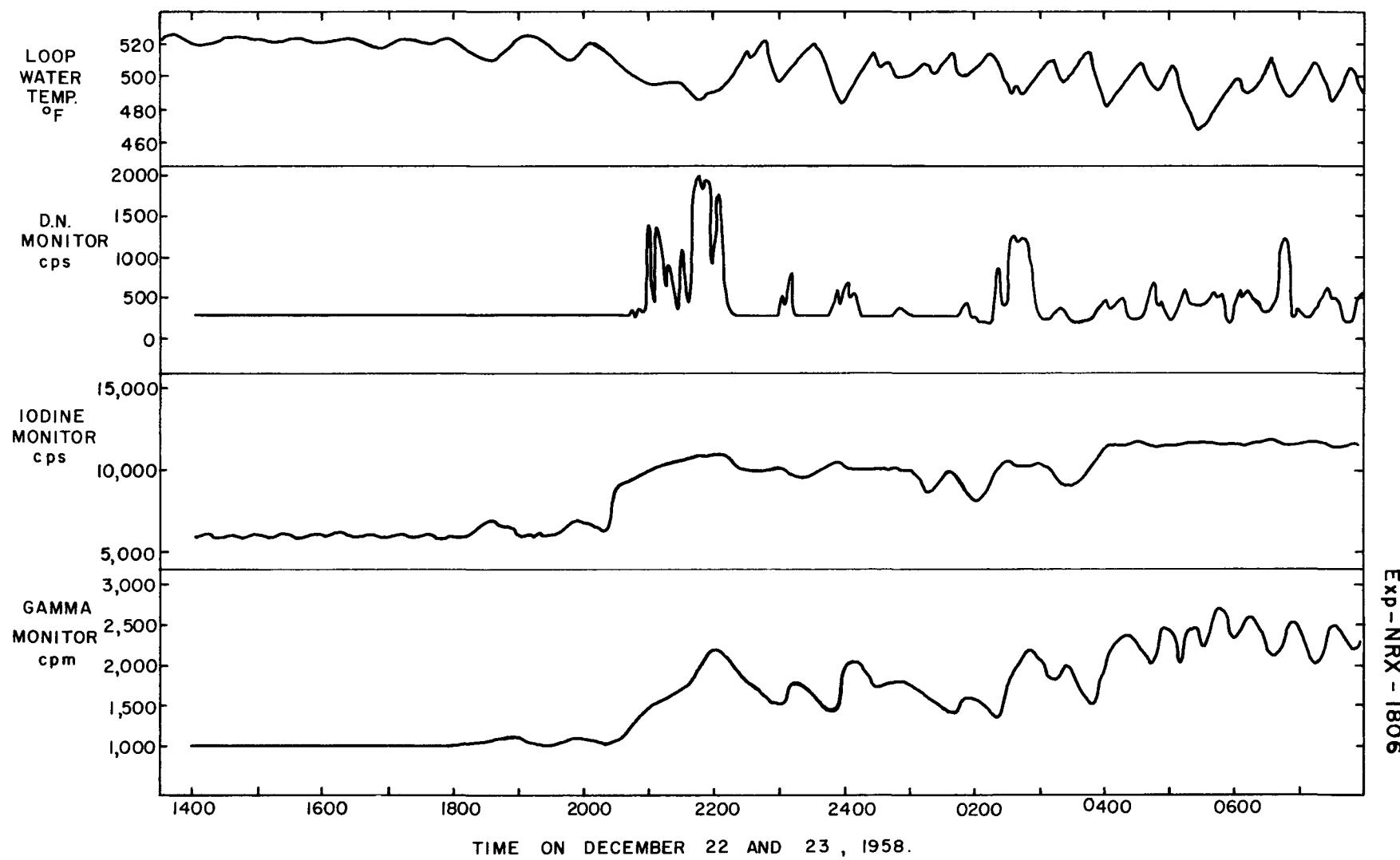


FIGURE 3

EFFECT OF TEMPERATURE CYCLING ON LOOP WATER ACTIVITY



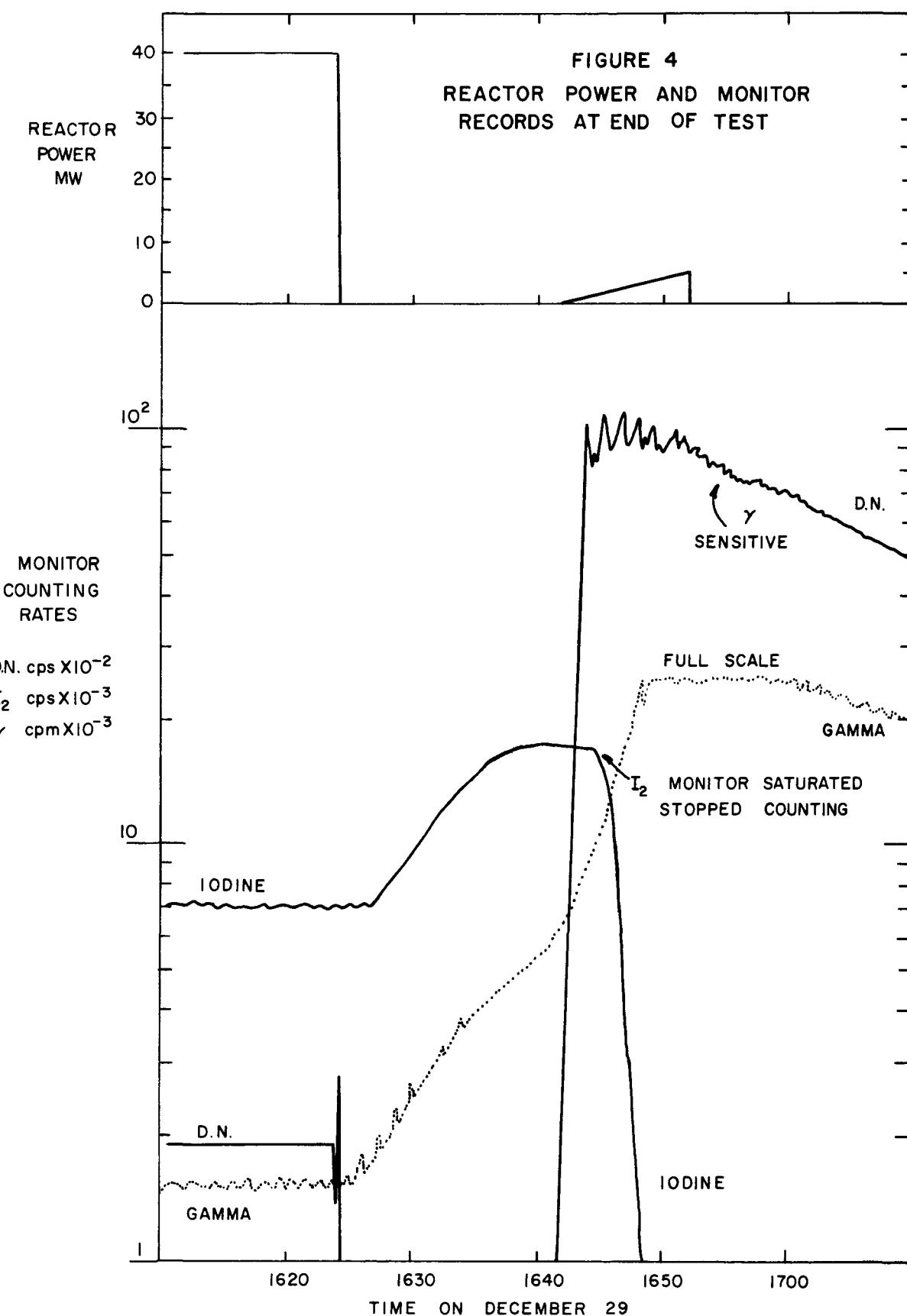


FIGURE 5
FISSION PRODUCT RESULTS