

Atomic Energy of Canada Limited

IRRADIATION OF DEFECTED SAP CLAD UO_2 FUEL
IN THE X-7 ORGANIC LOOP

Exp-NRX-5903

by

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R.D. MacDONALD

Chalk River, Ontario

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S U M M A R Y

This report describes an experiment designed to test the behaviour under irradiation of a UO₂ fuel specimen clad in a defected SAP sheath and cooled by recirculating organic liquid.

The specimen containing the defect was irradiated in the X-7 loop in the NRX reactor from the 25th of November until the 13th of December 1960. Up to the 13th of December the behaviour was analogous to that seen with defected UO₂ specimens clad in zircaloy which were irradiated in water loops. Reactor power transients resulted in peaking of gamma ray activities in the loop, but on steady operation these activities tended to fall to a steady state level. Over this period the pressure drop across the fuel increased by a factor of two, the increases occurring after reactor shut downs and start ups. On 13th December the pressure drop increased rapidly, after a reactor shut down and start up, to over five times its original value and the activities in the loop rose to a high level. The specimen was removed and examination showed that the sheath was very badly split and that the volume between the fuel and the sheath was filled with a hard black organic substance.

This report gives full details of the irradiation and of the post-irradiation examination. Correlation of the observed phenomenon is attempted and a preliminary assessment of the problems which would be associated with defect fuel in an organic reactor is given.

Chalk River, Ontario
October, 1961

* Attached to AECL from CGE Peterborough

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FIGURES

FIGURE 1 Fuel Specimen AHR before irradiation.

FIGURE 2 Element AHR showing the relative position of the drilled defect and thermocouple beads.

FIGURE 3 Fuel Specimen assembly showing element and thick walled flow tube.

FIGURE 4 Schematic Representation of X-7 Loop Test Section

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FIGURE 17 }

FIGURE 18 }

FIGURE 19 } Micrographs of Pellet Cross Section Showing
Organic Buildup in Cracks

1. INTRODUCTION

The fuel element proposed for Canada's first organic liquid cooled reactor, OCDRE consists of compacted and sintered UO₂ pellets contained in a sheath made of Sintered Aluminum Product (SAP). The irradiation behaviour of UO₂ is now well known but the technology of SAP is yet only in an early stage of development*. As part of the agreement, it was possible to obtain the initial fuel specimens from AI, to OCDRE approximate design and performance parameters, in exchange for an early defect test in the X-7 Loop.

From the inception of their program to develop organic cooled UO₂ fuel, the participants have been concerned with the performance of defected fuel especially for the AI reactor concept which does not allow removal of the fuel under power.

Out-reactor tests at AI (1) had indicated that organic coolant would enter through a hole in the sheathing and would penetrate the oxide pellets, the penetration occurring at a low temperature portion of a temperature cycle. They found that the coolant would decompose in regions of the oxide exceeding the decomposition temperature and that alternative filling and decomposition of the coolant would lead to catastrophic failure of the sheath from stresses caused by decomposition products alone. These results led to the recommendation that in-reactor tests be carried out to investigate the phenomenon of organic penetration, decomposition and final rupture of the fuel specimen.

Therefore an irradiation of a defected SAP-clad UO₂ specimen in organic coolant was proposed by AECL (2) with the following major objectives:

- * In September, 1959, a cooperative program on the irradiation of SAP clad UO₂ was started between the following.

Atomics International - Canoga Park, California (AI)
 Atomic Energy of Canada Ltd. - Chalk River, Ontario

(AECL)

Canadian General Electric Co. - Peterborough, Ontario
 (CGE)

1. To study the behaviour of defected SAP-clad UO_2 fuel under irradiation in organic coolant and to measure the release rate of fission products through the defect to the coolant.
2. To study the effect on the coolant and loop facility of irradiating a defected SAP-clad UO_2 fuel specimen.

2. PRE-IRRADIATION DATA

2.1 The Fuel Element

2.1.1 UO_2 Pellets

The UO_2 pellets were fabricated by the Davison Chemical Co. from oxide with a starting enrichment of 3.23 wt.% U-235 in U. The density, calculated from weights and volumes of individual pellets, ranged from 9.98 to 10.2 g/cm³. Metallographic examination of representative pellets at Chalk River revealed a non-homogeneous oxide structure consisting of an outer high density skin and an inner porous region. The reason for this has not yet been determined (3). Pre-irradiation length and diameter measurements of individual pellets are given in Table 1.

2.1.2 Sheath and End Plugs

The cladding material was M-257 (APM)* impact extruded by Alcoa with a tube wall thickness of approximately 0.5 mm. (0.020 in.). Metallographic examination by AECL (4) and CGE (5) showed that the tubing was of substandard quality, but since it was the only material available it was decided to use it. More details can be found in the report of the first SAP-clad UO_2 irradiation (4).

The end plugs were machined from the aluminum alloy AA 6061 (equivalent to Alcan 65S) and electroplated with silver to improve bonding.

* APM (Aluminum Powder Metallurgy) is the name given by US Manufacturers for sintered aluminum product.

2.1.3 Assembly and Testing of the Element

The element was fabricated at the Canoga Park facilities of AI. A stack length of eight UO₂ pellets (each pellet dished 0.36 mm(0.014 in)at one end)was loaded into the SAP tubing. The end plugs were pressure bonded to the sheath in a helium atmosphere at a temperature of 590°C (1100°F). Two weld beads were built up on the surface of the SAP to hold the sheath thermocouples. A photograph of the specimen designated AHR, is shown in Figure 1. Pre-irradiation dimensions of the fuel element are included in Table 2. Prior to drilling the artificial defect, the specimen was leak tested by both AI and AECL using two methods. The first was thermal cycling between 150°C (300°F) and 480°C (900°F) and sampling for He gas, and the second was by dipping in liquid nitrogen followed by alcohol at room temperature (4). The absence of bubbles in the second test and no observable He leakage during the first indicated that the specimen was sound.

A 0.51 mm (0.020 in.) diameter hole was drilled through the SAP sheath of specimen AHR at a point mid way on the diagonal joining the thermocouple beads, see Figure 2.

2.1.4 Instrumentation and Flow Tube

All thermocouples associated with the test section were made from 0.13 mm (0.005 in.) diameter Chromel and Alumel wires swaged inside a 1 mm (0.040 in.) OD stainless steel sheath. MgO was used as the insulant. Two thermocouples were peened into the weld beads and the leads were wired firmly to the upper end of the specimen (Figures 2 and 3).

The element was mounted by means of a spider ring inside a thick walled 7.6 mm (0.3 in.) aluminum flow tube of 3.3 cm (1.3 in.) outside diameter as shown in Figure 3. The gap between the flow tube and the fuel was 2.03 mm (0.080 in.) giving a coolant velocity of 9.1 meters/sec. (30 ft/sec.) over the element at the normal loop flow rate of 57 litres/min. (15 gal(US)/min.).

Thermocouples which measured the temperature of the coolant upstream and downstream of the specimen were also attached to the flow tube. The numbering of thermocouples associated with temperature measurements in the vicinity of the fuel was as follows:-

<u>Thermocouple No.</u>	<u>Location</u>
1	Coolant at inlet
2	Coolant at outlet
3	On sheath below defect
4	On sheath above defect
5	Coolant at inlet
6	Coolant at outlet

2.2 The X-7 Loop

2.2.1 Loop Characteristics

The X-7 Loop is a facility for recirculating an organic coolant at high temperature past an experimental fuel assembly held in the core of the NRX reactor.

The loop has been described by Wilson and Delaney (6). Briefly an organic liquid can be recirculated at temperatures up to 427°C (800°F) at a maximum pressure of 21.0 kg/cm² (300 lb/in²) and a flow rate of 144 l/min. (38 gal(US)/min). Immersion heaters provide 60 KW of heat in addition to the nuclear heat from the fuel. The loop coolers are in a circuit parallel to the main circuit. In a second parallel circuit, a flow of up to 7.6 l/min. (2 gal(US)/min) can be passed through sintered stainless steel filters. The pore size of these filters can be varied from 5 µm to 40 µm. The sampling valves are also located in this circuit. In another circuit a flow of coolant from the lowest temperature point in the system, just after the loop coolers, can be maintained to the surge tank.

2.2.2 The Test Section

The test section of the loop consists of a stainless steel pressure tube, 0.125 cm (0.049 in.) thick and 3.57 cm (1.402 in.) internal diameter which is inserted through a fuel rod position in NRX reactor. Coolant is circulated from the bottom of the pressure tube upwards to the top. Figure 4 shows a schematic representation. The coolant entrance and exit are connected to the circulating system by welded carbon steel pipe. The top closure of the tube is made by a "Unibolt" closure adapted so that thermocouple leads can be taken out through a modified Conax seal.

The aluminum flow tube containing the fuel specimen fits in the pressure tube as indicated in Figure 4. The assembly is held in position by a locking nut. The thermocouple leads are carried up the outside of the flow tube and exit through the Unibolt closure via the Conax seal.

2.2.3 The Coolant

The coolant used in the X-7 loop was a mixture consisting principally of ortho- and meta-terphenyl with high boiling tars (obtained from the OMRE* reactor) added. At the start of the irradiation a typical composition of the coolant in wt.% was

o - terphenyl	35
m - terphenyl	18
p - terphenyl	2.5
Biphenyl	5.5
Tars	33
Unaccounted	6

This mixture of isomers was sufficiently fluid at room temperature that it could be pumped through the loop. The only trace heating used on the loop was on stagnant lines leading to pressure or flow measuring instrumentation.

3. IRRADIATION DATA

3.1 Loop Start-up

Prior to 25 November, while the NRX reactor was shut down, the flow tube containing the fuel specimen was inserted in the test section of the loop and the thermo-couple wires were brought out through the Conax fitting. The Unibolt closure was sealed, the test section was filled with organic fluid and circulation was started.

During the filling stage the test section was flushed with N_2 in an attempt to prevent O_2 coming in contact with the coolant.

* The Organic Moderated Reactor Experiment reactor located at the NRTS Idaho, U.S.A.

After the loop had been pressure tested to ensure leak-tightness, the temperature of the coolant was raised by the loop heater to about 370°C (700°F) and with the reactor still shut down the readings of both loop and fuel thermocouples were recorded under constant temperature conditions. This procedure was in essence a rough calibration of the thermocouples and gave "base-line" readings at operating conditions but with no nuclear heat coming from the fuel. Readings of thermocouples associated with the fuel assembly are given in Table 3.

One of the two sheath thermocouples (couple No. 4) failed at this time because of an electrical short circuit below the top closure of the loop.

When the calibration was completed the loop temperature was lowered to 260°C (500°F) and the reactor was started. At this time the loop operating parameters were as outlined below. These values remained essentially unchanged throughout the irradiation.

Flow Rate of Circulating Organic -
58 l/min (15.3 gal(US)/min.)

Linear Velocity past fuel -
9.1 m/s (30 ft/s)

Flow to filter circuit -
2.85 l/min (0.75 gal(US)/min.)

Flow to surge tank -
0 to 1.5 l/min (0 to 0.4 gal(US)/min.)

Pressure at surge tank -
8.8 kg/cm² (125 lb/in²)

Pressure at inlet to test section -
18.6 kg/cm² (265 lb/in²)

3.2 Log of Events

This section gives the sequence of events throughout the irradiation period with some brief comments. Behaviour of the various parameters is discussed in more detail in subsequent sections. A plot of several parameters is given in Figure 5.

3.2.1 Period 25-28 November 1960

The reactor was started at 1850 hours, 25 November, 1960 but was shut down shortly after reaching a power of 20 MW because of a fuel element failure in another loop. The reactor did not start again until 2000 hours, 26 November when the power was raised to 20 MW, where it was held constant for about one hour. During this time flow and temperature readings were taken. Over the next 24 hours the power was raised in incremental stages to 40[★] MW.

At midnight 27 November, the power was lowered to 30 MW, for reasons not associated with this experiment. The monitor reading increased at this time and then fell. At 0945 hours 28 November, the reactor tripped. The reactor was started and by 1030 hours was again at full power.

The only unusual occurrence up to this time was the increase in the gamma ray monitor counting rate on the decrease in reactor power at midnight 27 November. Loop and fuel temperatures were reasonably steady and about as expected, and the pressure drop across the fuel was steady at 1.41 kg/cm² (20 lb/in²).

Several unusual occurrences were observed during and after the reactor trip and start-up of 28 November.

- (a) The gamma-ray activity increased sharply when the reactor tripped and then decayed rapidly.
- (b) After the reactor start-up the pressure drop across the test section rose gradually from 1.41 kg/cm² (20 lb/in²) to 1.97 kg/cm² (28 lb/in²) over a period of about 24 hours.
- (c) The temperature reading of the one remaining sheath thermocouple fell to a value below that of the loop coolant.

These three occurrences are all indicative of damage to the fuel element.

[★] Subsequent to this experiment, the NRX reactor power was recalibrated and operating power was found to be 42 MW rather than 40 MW.

3.2.2 Period 28 November to 14 December, 1960

From 28 November to 13 December the reactor operated at a power of 40 MW, with brief shutdowns on 2 and 8 December. After the start-up from each shutdown the pressure drop across the test section increased by a few lb/in^2 . Gamma-ray activities increased on start-up or shutdown but under steady conditions fell to reasonably steady levels of about 3000 counts per minute.

On 13 December the pressure drop across the test section appeared to be rising more rapidly than normal. After a reactor trip at noon on that day, this parameter increased rapidly. By noon on 14 December, it had increased to over 110 psi (5 times its initial value) and a decision was made to terminate the test.

The fuel was removed from the loop on 15 December 1960 and was taken to the Metallurgical Cave for examination.

Operation of the loop was continued with no fuel in the test section.

3.3 Thermal Data - Fuel Power Output

3.3.1 Coolant Temperatures

Figure 5 shows the variation of the coolant temperature at the outlet of the test section, as read by a thermocouple on the loop piping. The low temperature periods on 29-30 November and 14 December were necessary to allow maintenance work on other loops.

From 27 November to 29 November the coolant outlet temperature was maintained at 354°C (670°F). After the reactor start-up on November 30, it was raised to 377°C (710°F), and was maintained at this level for the remainder of the test.

3.3.2 Sheath Temperatures

As noted earlier, one sheath thermocouple failed prior to start-up. The reading of the other one is indicated on Figure 6 for the period 25-29 November. It can be seen that this thermocouple, since it indicated

a temperature of only 3°C (5°F) above the average bulk coolant temperature, was not reading the true sheath temperature but some intermediate temperature. After the reactor power cycle of 28 November this reading decreased still further and for the remainder of the test actually indicated a temperature below that of the bulk coolant. The drop in the indicated temperature was not abrupt but took place over about 18 hours.

3.3.3 Power Output

The power output from the fuel specimen was calculated from a knowledge of the temperature rise across the fuel and the flow rate of the loop coolant. Because of the small nuclear heat output, the loop was operating very close to its upper limit of temperature control when the outlet coolant temperature was 376°C (710°F). Hence the oscillations in coolant temperature made measurement of the temperature rise across the fuel very difficult. On December 5, a large number of readings were averaged to give the results tabulated in Table 4. In this table, measurements at other reactor powers are also given. The measured power output of $8 \pm 0.5 \text{ KW}$ at 40 MW reactor power is in agreement with the calculated value of 7.9 KW .

3.4 Pressure Drop Results

Values of the pressure drop across the test section (ΔP) are recorded in Figures 5, 6 and 7. Up to the reactor trip on 28 November, the value, corrected to 371°C (700°F), was steady at 1.41 kg/cm^2 (20 lb/in^2). After the reactor start-up on that date it rose over a 24 hour period to 1.97 kg/cm^2 (28 lb/in^2). This rise was coincident with the drop in sheath temperature noted previously and may be indicative of swelling of the fuel element causing the thermocouple to come into contact with the flow tube.

For the remainder of the period, while the reactor power was steady, the ΔP was steady, and only showed increases after a reactor start-up. Again, these changes were not abrupt but occurred during several hours after start-up.

On December 13, the ΔP started to rise at a much greater rate. The solid dots on Figure 7 represent actual ΔP readings. Unfortunately, intermediate readings

were not taken and it is not known whether the ΔP rose continuously over the period from 1300 hours to 1800 hours or whether it was a discontinuous rise, coincident with the increase in reactor power as suggested by the dotted line. The plateau from 0001 hours to 1000 hours on 14 December followed by a further increase in ΔP on reactor start-up again would tend to substantiate the discontinuous pressure drop increase. When the ΔP had reached 8.4 kg/cm² (120 lb/in²), it was decided to terminate the test and remove the fuel from the loop.

3.5 Radiation Monitor Behaviour

3.5.1 During Test

A Geiger-Muller probe was located on the loop piping, in a water cooled castle, at a point downstream of the test section. At a flow rate of 58 l/min. (15 gal(US)/min) the time delay between the fuel and the monitor was about 25 seconds. Activity levels are shown in Figure 5 and on an expanded time scale in Figures 8 and 9.

The activity level recorded by this gamma monitor showed sharp spikes at each power increase at the beginning of the test. The gamma-activity decreased until the next power increase, when it spiked again. Secondary spikes were also visible at about three minute intervals following each primary spike. This interval was taken to be the re-cycle time for the loop coolant.

During the first part of the period, the highest activity level recorded was estimated to be about 40,000 to 50,000 c/min. The actual value is not known, because, by the time the recorder had settled back after a range change, the activity was less than 25,000 c/min.

At midnight 27 November, an increase in count rate from 6000 to 13,000 c/min was observed when reactor power was lowered. Immediately following a reactor trip on 28 November, a small spike from 3500 c/min to 5000 c/min was observed on the monitor.

On the next start-up a maximum of 60,000 c/min was recorded. The reactor was shut down on 29 November after about 31 hours of operation, and this time, a sharp, much greater spike (~50,000 c/min) was observed on the gamma recorder.

Following this reactor shutdown, all further reactor start-ups, trips, and shutdowns produced spikes on the activity monitor. Peak activity levels ($\approx 50,000$ c/m) produced by reactor start-ups were, in general, not quite as great as those produced by shutdowns. Invariably, when reactor power reached steady conditions, gamma activities fell, and if operation were long enough levelled off at 40 MW at about 4000-5000 c/min.

Radiation levels recorded around the loop equipment during the course of the test are recorded in Table 5. Both the mean levels and the highest recorded levels are shown for each location. The high values shown were recorded following reactor start-ups, shutdowns or trips.

3.5.2 Activity Levels Subsequent to Test

After removal of the defect fuel specimen, activity levels around the loop frame decayed within 24 hours to the low levels of ~ 30 mr/hr that had been observed when no defected fuel was present. When the loop coolant was recirculated with no fuel in the test section and the NRX reactor was once more at 40 MW, the activity shown by the gamma-monitor was only 1000-2000 c/min and fields around the loop piping were ~ 30 mr/hr.

3.6 Particulate Matter in Coolant

3.6.1 Behaviour of Filters

The periods of operation for the various loop filters are shown in Figure 5. The solid bars represent the length of time a filter of nominal porosity (shown on the ordinate) was in operation. Removal from operation was occasioned by inability to pass any flow through the filter. The notation "5 μ CP" refers to a small disc of nominal pore size 5 μ m which was used to determine particulate matter concentration (see Section 3.6.2.2). At the start of the test 10 μ m filters were in operation and by 5 December, 5 μ m filters could be used for reasonably long periods without plugging. However, by 13 December, smaller pore size filters started to plug very rapidly and on 13 and 14 December, 10 μ m filters were plugging after only a few hours operation. After the fuel was removed on 14 December, a 20 μ m filter lasted only a few hours before clogging.

It should be noted that all filters had been used on the loop for some months. Subsequent tests have shown that the cleaning methods used at that time were inadequate to remove all the filtered matter from the pore filters. This and subsequent comparative tests have shown that the effective pore size of the filters was probably considerably smaller than the nominal pore size.

3.6.2 Particulate Matter Concentrations

3.6.2.1 Ash and Fe Content

The ash content (the inorganic residue after ignition of the sample) of the organic coolant in the loop has always been low and difficult to determine with any accuracy. A sample of coolant taken on December 17 (a few days after the test ended) and sent to AI for analysis showed the presence of 5 ppm ash.

Instead of ash determinations, AECL has been determining the Fe content of X-ray fluorescence analyses. Table 6 shows the results of samples taken during the test. The "Equivalent ash content" represents the ash concentrations calculated on the basis that all the ash is Fe_{203} . The Fe content of the coolant was very low: on December 12 it was < 2 ppm, the lower limit of detection by the apparatus used.

3.6.2.2 Organic Particulates

Towards the end of the test, the filters plugged with increasing rapidity. The Fe or ash content of the coolant showed no significant change. Thus it can be assumed that towards the end of the irradiation the concentration of organic particulate matter was increasing.

3.6.2.3 Fouling Potential

AI have developed a Pyrolytic Capsule Fouling Test (PCFT) as a measure of the fouling tendency in an organic loop. The test essentially consists of keeping a standard-size filament immersed in a sample of organic liquid for a given time at a given temperature. The amount of material that deposits on the filament is weighed. AI have set the rather arbitrary specifications for a "non fouling" organic coolant at a PCFT number of < 5 mg and an ash content of < 5 ppm. It should be noted that no reactor has yet operated with organic coolant within these specifications. Attempts are now being made to clean up the OMRE coolant to these conditions.

A PCFT determination by AI on a sample of X-7 coolant taken on 17 December showed a PCFT number of 95 mg. Thus at the end of the test, the ash content was below that specified by AI but the coolant showed a very high "Fouling Potential". This sample was taken after the badly failed fuel was removed from the loop.

3.7 Coolant Chemistry

In this section the results of various chemical analyses of the loop coolant are given. Methods have not been included but will be covered in a report to be issued shortly. (Exp-NRX-5707)

3.7.1 Isomeric Composition

Table 7 shows the coolant composition at various stages of the test. Analyses for biphenyl and o-, m-, and p-terphenyl were performed with a Loe Engineering Company high-temperature gas chromatograph using 0.1% Apiezon L on 60-80 mesh micro glass beads as the adsorbent. The column was operated at 175°C (347°F). High boiler determinations were performed using a microsublimation technique.

Results indicate that there was no significant changes in coolant composition during the irradiation. The AI determination on 17 December shows gratifying agreement with AECL results.

It will be noted that the "Remainder" figure from the gas chromatographic results is 5 to 6 wt.% higher than the "High Boiler" figure. This discrepancy can be accounted for by measurement errors but may also be in part due to the presence of intermediate boiling materials.

3.7.2 Water Content

Water determinations throughout the test showed the presence of 190-250 ppm water. The AI analysis on the coolant sample of 17 December showed 153 ppm, in reasonable agreement.

3.7.3 Dissolved Gas Analyses

Table 8 shows the concentrations (ml(STP)/kg coolant) measured during the test.

N_2 concentrations were dependent on surge tank temperature. H_2 , CH_4 and "Other Hydrocarbon gases" showed significant increases in concentration throughout the irradiation.

3.7.4 Radio-chemical Data

Table 9 shows the activities of various radio-nuclides determined throughout the irradiation.

The gamma-ray spectra of coolant samples were also examined on a 100-channel pulse-height analyzer. A typical spectrum is shown in Figure 10. The spectra showed the presence of fission product gases, their daughters, or radio-iodines. Any other fission products were present only to a minor extent.

AI radiochemical analyses of the sample taken on 17 December are shown in Table 10. They confirm that the levels of fission products such as Ce or Zr were very low compared to radio-iodines or fission-product-gas daughters. The AI activity for I-131 is considerably higher than any AECL figure. This is probably because the last AECL sample was taken on 14 December. Subsequent to this there were several bursts of radioactivity released to the loop due to fluctuation in reactor power.

The AI activity of Zr-95, calculated back to 14 December is roughly 1×10^3 d/min-g. This is comparable with results of 9.4×10^3 and 2.8×10^2 d/min-g obtained for these nuclides by AECL on samples taken on 11 October and 20 October, 1960. At this time no defected fuel had been present in the loop and the source of these activities was probably the OMRE tars used. Thus it can be concluded that the release of activities such as Zr-95 to the loop coolant was negligibly small.

4. EXAMINATION OF FUEL ELEMENT

4.1 Disassembly

The twelve-inch section of flow tube containing element AHR was examined in the Metallurgy Cave on December 19, 1960. The assembly was first dipped in trichlorethylene to remove the superfluous coolant. An attempt was then made to determine whether the coolant annulus was blocked; if so, it would account for the large increase in ΔP measured across the test section.

Trichlorethylene was poured into the flow tube and flowed readily through the section indicating a gap did exist between the fuel and tube. Direct viewing through the annulus showed a light passage of 190° to 210° around the channel. It was difficult to see through the annulus, indicating that the passage was quite narrow. The flow tube was then split longitudinally with a milling cutter.

4.2 Appearance of the Element

When the top half of the flow tube was removed the element appeared to be in one piece with a large circumferential bulge near the position of the drilled defect. However, when the element was lifted from the tube it was found to be broken into three sections. A longitudinal crack extended along the entire specimen and a second shorter crack appeared at 180° to the first. A number of circumferential cracks branched from the long split; two of these had propagated around the specimen. A composite photograph of the cracked specimen is shown in Figure 11. The uranium oxide pellets, with the exception of the two end compacts, were loose inside the sheath and were removed intact.

Diameter measurements were taken across the sheath at 90° to the main longitudinal split. These are recorded along with the pre-irradiation values in Table 2.

It was difficult to remove the two thermocouple wires from under the retaining wire at the top end. The lower thermocouple bead had been milled flat when the flow tube was split.

4.3 Films on the SAP Sheathing and Flow Tube

Examination of the inside of the flow tube showed virtually no organic deposit; the machining marks were still clearly visible - see Figure 12. The outer surface of the SAP sheath was clean (see Figure 11). The inside surface of the sheath contained a black deposit near the circumferential splits. This deposit was made up of two films, the first very dense and hard (see Figure 13), and the second loose and granular. The inner surface of the sheath, away from the cracks was reasonably clean. Discolouration rings (see Figure 13) which correspond to pellet interfaces were observed on the inside of the sheath.

4.4 UO₂ Fuel Pellets

The UO₂ pellets were heavily coated with a hard black deposit believed to be polymerized organic coolant (see Figure 14). The thickness of the deposit varied over the surface of the pellets and was in the range 0.13 mm (0.005 in.) to 0.89 mm (0.035 in.). The pellet ends were also coated but not as heavily as the outside surfaces. One pellet showed evidence that the deposit had been built up in layers. The cracking pattern of the deposit matched the cracks in the pellets (Figure 14). The pellet fragments were probably cemented together by the polymerized organic coolant that had deposited in the oxide cracks. Diameter and length measurements were taken on most pellets to determine swelling and film thickness after irradiation. These readings are tabulated in Table 1. Straight cracks, occasionally at right angles (see Figure 15) were found at the ends of some pellets. It is not certain if the location of these cracks correspond to the axial splits in the sheath. An attempt was made to remove the deposit by solution in chloroform but after 24 hours the film was still intact. Chipping the deposit was also unsuccessful and succeeded only in breaking the pellets.

The interior of the end and center pellets were examined under the stereomicroscope. The entire cross section of both end pellets was covered with a thin film of organic coolant (Figure 16). In contrast, the organic film on the cross-section of the center pellet coated only the outer 1/3. The uranium oxide showed slight cracking and no central voids. The center pellet displayed what is thought to be grain growth (Figure 16).

Metallographic samples from both the sheath and fuel were retained. A section of the SAP sheath adjacent to the middle break and across the long split will be examined for its microstructure. Two complete pellets were examined by G.W. Parry of General Metallurgy Branch. One of the pellets was mounted transversely and the other was sectioned longitudinally and mounted. Examination of the latter was not possible since it was too badly cracked for retention to the mount. The transverse section was polished with 1/4 μ m diamond-impregnated cloth and etched in a solution of 1 part H₂SO₄, 9 parts H₂O₂.

Photographs are shown in Figures 17, 18, 19. Microscopic examination showed that the coolant had penetrated the cracks. A discontinuous layer of average thickness 0.004 cm (0.0015 in.) was found around the periphery of the pellet.

The organic had the appearance of being deposited in layers in the cracks. The layers adjacent to the UO_2 were slightly darker than the adjoining layers. There was no evidence of reaction between the UO_2 and the coolant. The oxide was of high density, was completely single phase and showed slight equiaxed grain growth towards the centre. The actual limits of the area that showed no grain growth could not be easily determined.

5.

RADIOLYTIC AND PYROLYTIC DAMAGE

Table 7 indicates that during the irradiation no gross changes occurred in the coolant composition. However, the thick organic film found on the UO_2 pellets does indicate that severe damage had occurred in the vicinity of the fuel.

At the start of the irradiation, the loop was operated at temperature and pressure before reactor start-up. Thus it can be assumed that organic coolant entered the defect hole and surrounded the fuel pellets before irradiation commenced.

As the reactor came to power, temperature of the fuel would increase and the average temperature of the coolant in the gap between the fuel and the sheath would be higher than that of the bulk coolant. Hence, the liquid would expand and some would be forced out the defect hole. This would account for the spike in gamma-activity seen at each reactor power increase. The surface temperature of the fuel at 40 MW reactor power has been estimated to be $527^\circ C$ ($1000^\circ F$) and vapour formation may assist the expulsion of organic coolant through the hole. At $527^\circ C$ ($1000^\circ F$) pyrolytic damage rates of the coolant are known to be high, and the rate of high boiler formation is probably 10% per hour (7). Thus pyrolysis of the coolant inside the sheath could occur at an appreciable rate.

Radiolytic damage from fission-fragment bombardment could also play a significant role. Landsman and Truswell have irradiated Santowax-R (8) in the presence of fission recoils from U_3O_8 and find G(polymer), the number of terphenyl molecules converted to polymer per 100 eV energy absorbed is about 2 at $350^\circ C$. They found the U_3O_8 which was deposited on foils was always coated after irradiation with a layer of coke 5-7 μm thick.

The dose rate from fission fragments at the surface of the fuel may be estimated roughly as follows:

The average range of a fission fragment in UO_2 is about 7 mg/cm^2 (9). Assume a cylinder of oxide 16.3 cm. long and 1.28 cm. O.D. The weight of oxide at a density of 10.3 g/cm^3 is 216 g. If fission fragments emanate from a skin around the outside of the cylinder of thickness equal to the average range of the fission fragments, then the weight of this skin is about 0.48 g. Assuming only $1/4$ of the fission fragments leave the surface, and taking the power output from the fuel to be 8 KW, the rate of energy release due to fission fragments is:

$$\frac{1}{4} \times \frac{0.48}{216} \times 8 \times 10^3 = 4-1/2 \text{ watts}$$

The surface area of the fuel is approximately 68 cm^2 and hence the dose rate is 0.066 watt/cm^2 or $\sim 4 \times 10^{17} \text{ eV/cm}^2\text{-sec}$. It is assumed that the energy associated with the fission fragments is all absorbed in the coolant.

From their experiments, in which Pb foils coated with U_3O_8 were exposed in a capsule containing Santowax R to reactor irradiation, Landsman and Truswell (8) estimate that the first immobile skin of high polymer forms at a dose of $\sim 1 \times 10^{21} \text{ eV/cm}^2$. By the time the dose has reached $5 \times 10^{22} \text{ eV/cm}^2$ the film will have attained a thickness sufficient to stop most fission fragments. On this basis the first film would form on the oxide fuel in this test in less than one hour's irradiation at 40 MW reactor power. It would be thick enough to stop fission fragments ($\sim 10 \mu\text{m}$) after 2 days irradiation.

The film observed after irradiation was 130 to $890 \mu\text{m}$ thick. This thickness would completely prevent release of fission products by recoil although they might still diffuse through it. This very thick film probably indicates that pyrolytic damage also plays an important role in filling the gap between the fuel and the sheath with coked organic.

In Landsman and Truswell's experiments, the surface of the oxide was washed freely by the coolant and all the polymerized coolant did not deposit on the fuel.

In this irradiation, with the coolant, at least initially, in a more confined space, more polymerized organic may have stayed on the oxide surface. Using the dose rates calculated above and assuming $G(\text{polymer})$ is two molecules of terphenyl damaged per 100 eV. the rate of destruction of the coolant is roughly 0.8 g/hr. Assuming polymerization occurs with negligible loss in weight, then after one half hour's irradiation 0.4 grams of polymerized coolant should be available for deposition. Assuming a density of 2 g/cm³ for the polymerized coolant, a film 30 μm thick could form on the surface of the oxide in this time.

The value for G (gas) is also about unity for fission fragment bombardment at this temperature. This means that one molecule of gas (mainly H₂) is formed per 100 eV of energy absorbed. Using this value, it can be calculated that about 20 cm³ H₂ (at STP) would be formed from one half hour of reactor operation at 40 MW.

The above calculations are only very rough but they do indicate that radiolysis by fission fragment damage and pyrolysis play an important role in film formation.

The behaviour of the filters can now perhaps be explained. It will be recalled that after 12 December they start to plug rapidly and larger and larger pore sizes had to be used. It will also be recalled that the ash content or the Fe content of the loop coolant did not increase during this period.

This was the period during which the ΔP was rising rapidly and it is believed the sheath was swelling and splitting severely. One can postulate that the increase in particulate matter was due to fragments of highly polymerized coolant which were flaking off the surface of the UO₂ (which was now exposed) and being carried to the filters. These fragments also contributed to the high PCFT number determined by AI for the coolant at the end of the irradiation.

6. RELEASE OF FISSION PRODUCTS TO COOLANT

6.1 Escape-Rate Coefficients

The primary mechanism for escape of the longer-lived fission products from defected UO₂ fuel elements is considered to be diffusion. The rate-controlling step is believed to be diffusion through the UO₂ matrix. Holdups in the open porosity and in the fuel-sheath gap are thought to be small compared to the diffusion rate in the UO₂ itself (10).

In order to compare the release of fission products from defected fuel elements in pressurized-water loops a parameter called the "escape-rate coefficient (ν)" has been used. This coefficient is based on the assumption that the release is a first-order process, i.e., is proportional to the concentration of the nuclide in the fuel.

The escape rate coefficient is defined by the equations:

$$(1) \text{ in the fuel } \frac{dN_f}{dt} = F.Y - \lambda N_f - \nu N_f$$

$$(2) \text{ in the loop coolant } \frac{dN_l}{dt} = \nu N_f - \lambda N_l - k N_l$$

where N_f = number of atoms of the nuclide in the fuel

N_l = number of atoms of the nuclide in the loop coolant

F.Y = fission rate in the fuel x fission yield of the nuclide

λN_f = disintegration rate of nuclide in the fuel

λN_l = disintegration rate of nuclide in the loop coolant

N_f = release rate of the nuclide from fuel element to loop coolant

kN_l = total loss-rate of nuclide to the ion exchange resins, system surfaces, surge tank, and through leakage, sampling etc.

t = equivalent time at full reactor power.

At equilibrium $dN_f/dt = dN_l/dt = 0$ and assuming that $\nu \ll \lambda$

The equations reduce to

$$\nu = \frac{(\lambda N_l)}{F.Y} (k + \lambda)$$

When the irradiation time is insufficient to produce saturation of the nuclide in the fuel, the term $(1 - e^{-\lambda t})$ is required in the denominator.

In the organic loop, there is no purification of the coolant other than removal of particulate material by filtration. The only loss of fission products from the coolant other than decay is by any process which removed coolant from the system, i.e., leakage, sampling, changing filters etc. Losses of this kind are indicated by changes in the surge-tank level, and were generally only 3.8 litres per day (1 gal.(US)) per day over the periods when reactor operation was steady. The loss-rate coefficients (loss rate total vol.) works out to be $1 \times 10^{-7} \text{ S}^{-1} \text{ sec.}$ and this has been used in calculating for I-131 and Xe-133. Another loss of Xe-133 which should be allowed for is diffusion to the vapour phase of the surge tank. Unfortunately, this rate is not known and no correction can be applied.

The escape-rate coefficients which have been calculated are given in Table 11 along with average values obtained from defect tests in the X-2 pressurized-water loop.

In calculations for the iodines only the results of the samples taken on December 7 and 13 were used since other samples were taken at times too close to reactor startups or power fluctuations.

It can be seen that generally the escape-rate coefficients for the fission-product gases were in the same order as those found in the water systems. However, the results for the two radio-iodines were considerably lower than those found in the X-2 tests.

The lower rate of radio-iodine release to the coolant may have been due to absorption in the film around the oxide. In the last section evidence was produced to show that this film is probably produced early in the irradiation, and Landsman and Truswell (8) have shown that I-131 release was consistent with significant hold-up in the organic film.

This film was undoubtedly the reason that other fission products such as Zr, Nb, the rare earths, and in fact UO₂ itself, were not found to any significant extent in the coolant.

6.2

Behaviour of Gamma-ray Monitor

Experience in water loops with UO₂ fuel has indicated generally (10) that when only a small defect is present, the gamma-ray monitor count rate peaks

sharply with each increase in reactor power. This phenomenon is attributed to the "Water-logging" effect. Presumably a similar "Organic-logging" effect similar to that described in Section 5 could be present and hence account for the peaking of gamma-ray activities on reactor start-up.

In the water systems, as a general rule, increase in gamma-ray count rate with reactor shutdown or lowering in power is associated with a defect considerably larger than a 0.020-inch-diameter hole (10). If the same is true in the organic system, then the first indication of an enlargement of the small defect hole is at midnight 27 November when the gamma-ray counting rate increased with a decrease in reactor power.

7. DISCUSSION

7.0 General Discussion

It is very difficult to deduce the mechanism which caused the rupture of the fuel specimen from the results of this one experiment. Further irradiations will be necessary to obtain a clearer understanding of the processes involved. However, an attempt is made to explain the behaviour of a defected specimen in organic coolant based on the evidence from this test.

7.1 First Indication of Damage

The first indication of anything unusual in the behaviour of the test specimen was a peak in γ activity coincident with a decrease in reactor power at midnight on November 27. Experience with defected UO_2 fuel in water cooled loops has shown that, while peaking in γ activity at reactor startup is normal behaviour for the original deliberate defect, peaking in activity at a reactor power decrease is indication of a more serious defect or rupture of the fuel specimen. Therefore it is possible that a crack developed in the SAP sheath on or shortly before the reactor power decrease at midnight, November 27.

Cracking could have initiated in a number of places besides the defect itself in this sub-standard sheathing material, e.g. at a deep pit or extrusion die mark (4). The crack would then propagate due to stress concentration.

Furthermore, the γ activity peaks at each reactor startup after November 27 were much greater than before, indicating that there was less restraint to the emission of fission gas in the coolant.

Following this first indication of damage, there was a sudden decrease in the sheath thermocouple reading coincident with the beginning of a rise in pressure drop across the test assembly at a reactor startup at 1000 hours, November 28. The pressure drop rise occurred over about one day until noon, November 29, when the reactor was again shut down and there was another peak in γ activity. It is very likely that the specimen swelled during the one-day period, pressing the sheath thermocouple into contact with the relatively cool flow tube and constricting the flow annulus. The fact that the sheath thermocouple appeared to be in contact with the flow tube after irradiation supports this assumption.

7.2 Propagation of Rupture

Pressure-drop increases signifying further swelling of the specimen occurred over three- to twelve-hour periods at each reactor startup until December 14 when the pressure drop became excessive necessitating removal of the fuel.

The loop filters did not plug significantly until about midnight, December 12. Shortly after this, on December 13, the pressure drop across the test assembly rose quite sharply. These two occurrences indicate gross swelling of the specimen and opening of the crack allowing particles of decomposed organic formed on the UO₂ fuel to enter the coolant stream.

7.3 Proposed Mechanism of Rupture

There are several (possible) mechanisms that could have caused the initial rupture of the SAP sheath.

1. Internal pressure on the sheath due to the buildup of decomposed organic on the fuel combined with differential thermal expansion of the fuel and sheath.
2. A coolant-logging mechanism, i.e. a sudden hydraulic pressure exerted by coolant trapped between the UO₂ fuel and sheath due to thermal expansion or vaporization of the coolant on increases in reactor power.

3. Pressure of contained hydrogen gas in a porous deposit of decomposed organic on the fuel.

There is no evidence as yet to support the third mechanism. Photomicrographs show no signs of porosity in the decomposed organic.

The coolant logging mechanism is well established for UO_2 -water systems, and peaks in γ activity at reactor startup are evidence of the second coolant-logging mechanism. The sudden increase in γ activity indicates a sudden discharge of coolant and fission gases through the defect in the specimen due to the building of pressure. However, it is still not known whether this mechanism caused the initial cracking of the sheath.

Based on the evidence of this experiment and the AI out-of-reactor tests, it is perhaps a better assumption that the buildup of decomposed organic and differential expansion during a reactor power cycle (mechanism 1) was the most likely cause of the rupture of the specimen.

Preliminary stress-strain calculations indicate that a deposit of decomposed organic 80 to 130 μm (0.003 to 0.005 in.) thick is sufficient to rupture the sheath. A deposit up to 1040 μm (0.040 in.) thick was observed on the UO_2 after the irradiation. Calculations from section 5 show that, at 40 MW reactor power, combined radiolysis and pyrolysis could easily account for a deposit 80 to 130 μm thick in one day. Therefore it is likely that the first crack which occurred after one day's irradiation at midnight, November 27, could easily have been caused by this mechanism.

Evidence of the mechanism of alternate filling of a space with organic coolant and decomposition was seen in the cracks in the UO_2 fuel. Thermal stresses in the UO_2 could start cracks which would open up at reactor shutdown allowing coolant to enter. Even with the fuel expansion at reactor startup some of this organic might be retained in the cracks and decompose rapidly (up to 30 μm in 1/2 hour).

7.4

Seriousness of Rupture

In estimating the seriousness of a rupture of a SAP-clad UO_2 fuel specimen in organic coolant, it is noted that there was no evidence at any time of UO_2 fuel

particles in the coolant. The examination of the fuel after irradiation showed that the pellets were cracked but cemented together with organic material. The cementing and coating of the UO₂ pellets with organic implies that, if a rupture of the SAP sheath occurs, the possibility of small UO₂ particles getting into the coolant stream is low.

Because it is desirable for reactor operation to know how soon after the first indication of a defect it is necessary to remove the fuel, an attempt was made to correlate the pressure drop with the appearance of the specimen and flow passage after irradiation. Some fairly broad assumptions were made on the extent of the swelling of the specimen, the friction factor and the final shape of the specimen in situ. Based on these assumptions, the final pressure drop was found to be in reasonably good agreement with the final appearance of the specimen and the flow passage. By a similar calculation, the increase in pressure drop from 20-38 psi between 1000 hrs November 28, and 1000 hrs November 29, could indicate swelling of .082 in. in diameter of the specimen. This would be a dangerous condition for a fuel-bundle design (such as OCDRE) with 0.040 in. spacing between elements and would necessitate immediate removal of the fuel. However, the stress causing the swelling and rupture of the fuel was most likely due to the differential expansion of fuel and sheath during a reactor power cycle. Therefore, it is conceivable that the fuel could remain in reactor much longer provided there were no reactor power cycles.

8. CONCLUSIONS

8.1 Time of Rupture

From the evidence available, it is concluded that cracking of the specimen sheath was initiated at or shortly before a decrease in reactor power combined with a peak in activity at midnight on November 27. Approximately 10 hours after this, the pressure drop first increased, indicating swelling of the specimen. It is possible that the sheath first split at this time. Subsequent pressure-drop increases always coincided with reactor startups. Excessive pressure drop and plugging of filters on December 13 could indicate gross splitting of the sheath allowing particles of decomposed organic off the UO₂ to enter the coolant stream.

8.2 Mechanism of Rupture

The results observed cannot be attributed definitely to any one cause. Most probably several phenomena contributed to the final appearance of the specimen. It is known that during reactor power cycles a coolant-logging phenomenon was operative as demonstrated by the peaks in γ activity. It is also known that a thick film of polymerized organic material covered the UO_2 . The increases in ΔP always occurred after increases in reactor power. This effect in itself is very hard to explain. It is possible that, each time the reactor shut down, coolant entered between the fuel and sheath. At each start-up and during subsequent operation some coolant would remain which would polymerize to a hard film filling the space and effectively increasing the OD of the oxide. On subsequent shut downs the sheath would tend to contract but would be restrained by this organic layer and hence be strained. Eventually it would split. This may not be the true or the complete mechanism but the presence of the thick layer of polymerized organic undoubtedly contributed to the final sheath failure.

8.3 Seriousness of Rupture

Since no UO_2 particles were evident in the coolant, since the activity level was about the same as that from a water-cooled defect, and since the fuel pellets were cemented together by organic, it is concluded that the hazard of small UO_2 fuel particles contaminating the coolant stream is low.

It may be necessary to remove defective fuel specimens 40 to 60 hours after the first signs of defect based on the results of this one test. However, if there are no reactor power cycles, the fuel could possibly remain in reactor much longer.

9. FUTURE WORK

9.1 Conduct further irradiations to determine if this type of specimen failure is typical.

9.2 Assess more carefully the feasibility of SAP sheathing under conditions of defect in organic coolant.

9.3 Analyze the mechanism for progressive deposition of decomposed organic on the UO₂. Determine if this condition which is dangerous to sheath integrity can be overcome by design or operation.

9.4 Apply the results of this test of a single element to a fuel bundle design for an organic cooled reactor.

10. ACKNOWLEDGEMENTS

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11. REFERENCES

1. J.E. Kinzer and A.N. Mellott "Organic Coolant UO₂ Compatibility" NAA-SR-Memo 5167 (Apr. 1960).
2. A.G. Cracknell and R.D. Page "Final Proposal for the First Irradiation of Defected SAP-clad UO₂ Fuel in the X-7 Organic Loop for NRX". Exp-NRX-5901 (Nov. 1960).
3. G.H. Chalder Private Communication.
4. G.M. Allison and R.D. Delaney "The First Irradiation of SAP-clad UO₂ Fuel in the NRX X-7 (organic) Loop" Exp-NRX-5805 (1961) (In publication).
5. J.W. Standish "A Report on the Examination of Sheath and End Closure Materials as used in the X-7 SAP-clad Fuel Irradiation" CGE Metallurgical Report No. 128 (Dec. 1960).
6. A.H. Wilson and R.D. Delaney "Design Specifications for the NRX Organic Loops" Exp-NRX-3905 (Oct. 1959).
7. A.W. Boyd "An Assessment of the Data on the Radiolysis and Pyrolysis of Organic Coolants" CI-218 (1961).
8. D.A. Landsman and A.E. Truswell "The Irradiation of Santowax R with Fission Fragments" AERE-M715 (1960).
9. J.D. Eichenberg et al "Effects of Irradiation on Bulk UO₂" WAPD-183 (1957).
10. G.M. Allison and R.F.S. Robertson "The Behaviour of Fission Products in Pressurized Water Systems" RLI-4 (Revised AECL No. (In publication).

TABLE 1
DIAMETER AND LENGTH MEASUREMENTS ON UO₂ PELLETS FROM ELEMENT AHR

Pellet No.	D I A M E T E R (in.)			L E N G T H (in.)		
	Post- Irrad. Min.	Post- Irrad. Max.	Pre-Irrad.	Post- Irrad. Min.	Post- Irrad. Max.	Pre- Irrad.
29	0.525	0.527	0.505	0.020	0.022	(1)
71	0.525	0.543	0.505	0.020	0.038	0.830
27	0.536	0.553	0.506	0.030	0.047	0.837
33	0.531	0.551	0.506	0.025	0.045	0.842
39	0.524	0.541	0.506	0.018	0.035	0.840
68	0.530	0.540	0.506	0.024	0.034	0.8425
80	0.533	0.571	0.506	0.027	0.065	0.836
34	(2)		0.506	(2)		0.802

AHR (3) (1) Length measurements not taken as pellet split into two sections.
 (2) Pellet badly cracked length and diameter measurements not taken.
 (3) Axial clearance 0.044 in. diametral clearance 0.0025 in.
 pellets dished on one end.

TABLE 2
 PRE- AND POST-IRRADIATION DIAMETERS OF
 ELEMENT AHR

<u>Position</u>	<u>D I A M E T E R (in.)</u>		
	<u>Pre-Irradiation</u>	<u>Post-Irradiation 90° to Split</u>	<u>Change</u>
Top	0.547	0.555	0.008
1/8		0.576	0.028
1/4		0.595	0.047
3/8		0.617	0.069
Middle	0.548	0.665	0.117
5/8		0.650	0.102
3/4		0.660	0.112
7/8		0.576	0.028
Bottom	0.5485	0.565	0.0165

TABLE 3
THERMOCOUPLE INTER-CALIBRATION

(Readings taken November 25, 1960)
All readings are $^{\circ}\text{F}$
Reactor at Zero Power

Thermo-couples	Location	Reading 1		Reading 2		Reading 3		Average	Normal [†]	Deviation From Normal
		1800 hrs.	1818 hrs.	1800 hrs.	1830 hrs.	1800 hrs.	1830 hrs.			
1	Coolant in	692	692.5	693	694.5	694.5	696	697	697	-3.1
2	Coolant out	694	695	697	697	698	700	700	701	-0.1
3	Sheath	694	695	696	697	699	699.5	700	701	-0.2
5	Coolant in	695	697	696	698	700	699	700.5	701	+0.4
6	Coolant out	694.5	696	696	698	698	699	700	700.5	-0.1

[†] Normal is the average of the most accurate thermocouples (numbers 2,3,5 and 6).

TABLE 4
LOOP FUEL POWER OUTPUT
Calorimetric heat output using specimen instrumentation

Time	Date	Average Bulk		Average Coolant		Loop Power (kW)	Fuel Power (MW)	Reactor Power (MW)	Estimated Sheath Temp. (°F)
		Coolant Temp. °F	p cp	Temp. °F	ΔT (°F)				
1930-2110	Nov. 27	665	0.496	7.0		7.6		40	833
0310-0450	Nov. 28	655	0.494	4.7		5.1		28	770
1820-2320	Nov. 28	667	0.496	6.9		7.5		40	834
0000-1030	Dec. 5	706	0.499	7.73		8.54		40	887
AVERAGE						7.7		40	

II Loop Fuel Power = $0.14671 p cp (flow) \Delta T$

where p is specific gravity

cp is specific heat

flow is approximately steady at 14.9-15.1 USGPM

ΔT is the average coolant temperature rise across fuel.

TABLE 5
X-7 LOOP RADIATION LEVELS

<u>Location</u>	<u>Mean Level mr/h</u>	<u>Highest Recorded Level mr/h</u>
At sample dry box	30	180
At roof over surge tank	60	170
On walk way on roof over loop frame	100	400
Loop piping below valve VD-108 just inside the loop room door	300	1400
At loop filters	600	1500
At loop piping below gamma monitor	80	700
Unibolt, top of test section	500	1500
Test section at header level in upper header room	700	3000

TABLE 6
IRON CONTENT OF X-7 LOOP COOLANT

<u>Date</u> <u>1960</u>	<u>Iron Content</u>	<u>Equivalent</u> <u>Ash Content</u> <u>ppm</u>
Nov. 28	4	6
Dec. 5	4	6
Dec. 12	< 2	< 3
Dec. 17*		5

* Atomics International determination

TABLE 7
MAJOR COMPONENTS OF X-7 LOOP COOLANT

<u>Date</u>	<u>Biphenyl</u>	<u>Terphenyl</u>			<u>Remainder</u>	<u>High Boiler</u>	<u>O/M Ratio</u>
		<u>o</u>	<u>m</u>	<u>p</u>			
1960							
Nov. 28	5.5	35.3	17.9	2.75	38.55	33.6	1.97
Dec. 1	5.5	35.0	19.1	2.3	38.1		1.83
Dec. 5					33.4		
Dec. 8	5.5	33.7	18.8	2.2	39.8		1.79
Dec. 12	5.9	34.3	17.65	2.6	39.55	33.3	1.94
Dec. 17*	5.3	33.1	17.7	2.5	41.4	34.4	1.87

* Atomics International Determination

TABLE 8
X-7 DISSOLVED GAS ANALYSES (ml (STP)/kg)

Date	Coolant Temp.		Surge Tank Temperature °F °C	Flow Rate		Total [†] Gas	H ₂	N ₂	CH ₄	CO	Other ^{††} Hydro- Carbon Gases
	Inlet to Test Section °F °C	Surge Tank Temperature °F °C		Cold Flow to Surge Tank Gal (US)/ min	ml/s						
Nov. 29	660	349	266 130	0.42	26	310	29	221	27	2.2	30
Dec. 2	710	377	307 153	0.45	28	319+	71	194	52	2.6	+
Dec. 7	700	371	316 158	0.46	29	501	74	231	98	3.3	94
Dec. 9	700	371	461 238	0.0	0	586	92	282	109	4.4	99
Dec. 13	705	374	463 239	0.0	0	605	92	323	125	4.0	62

[†] "Total Gas" is that fraction of the coolant which is not condensable at -77°C at 0.005 mm Hg pressure.

^{††} "Other Hydrocarbon Gases" is that fraction of the coolant which is condensable at -196°C but not at -77°C at 0.005 mm Hg pressure.

+ "Hydrocarbon Gas" fraction lost. Total gas figure does not include this fraction.

TABLE 2
RADIOCHEMICAL DATA ON X-7 ORGANIC COOLANT (d/min- g)

<u>Date</u> <u>1960</u>	<u>Time</u>	<u>Gross</u> <u>Activity</u> <u>**</u>	<u>Kr</u> <u>88</u>	<u>Xe</u> <u>133</u>	<u>Cs</u> <u>138</u>	<u>I</u> <u>131</u>	<u>I</u> <u>133</u>
Nov. 27	1530	1.20 \times 10 ⁶					
	0915	3.42 \times 10 ⁵					
Nov. 28	1040						
	0905						
Nov. 29	0930	5.0 \times 10 ⁶		3.8 \times 10 ⁶			
	1020	8.20 \times 10 ⁵					
Dec. 1	1330						
Dec. 2	1335						
Dec. 5	1335	3.83 \times 10 ⁵					
Dec. 6	1330						
Dec. 7	0915						
	1030	1.5 \times 10 ⁶		~6.0 \times 10 ⁶			
Dec. 8	1340	1.70 \times 10 ⁶					
	0915						
Dec. 9	1405	3.46 \times 10 ⁵		3.4 \times 10 ⁶			
Dec. 12	0840						
Dec. 13	1020						
Dec. 14	1320	1.30 \times 10 ⁶		1.3 \times 10 ⁶			
	1435						

** Total Count on second shelf of β -proportional counter in c/min- g ,
one hour after sampling.

TABLE 10RADIONUCLIDE DETERMINATIONS BY ATOMICS INTERNATIONAL

<u>Nuclide</u>	<u>d/min-g</u> <u>Feb. 7, 1961</u>	<u>Calculated</u> <u>d/min-g</u> <u>Dec. 17, 1960</u>
Ba-140	9.7×10^3	1.6×10^5
Zr-95	5.4×10^2	9.3×10^2
Ce-141	5.8×10^2	1.8×10^3
Mn-54	$\sim 6.0 \times 10^2$	$\sim 6.8 \times 10^2$
Co-60	$\sim 1.2 \times 10^2$	$\sim 1.2 \times 10^2$
I-131	2.7×10^4	2.3×10^6 †
Cs-137	1.2×10^3	1.2×10^3

† 2.7×10^6 d/min-g calculated to 14 Dec. 1960.

TABLE 1.1
ESCAPE-RATE COEFFICIENT

Test	Density of U_0^2 g/cm ³	$\int k d\Theta$ w/cm	Escape-Rate Coefficient s ⁻¹					
			Kr^{88}	Xe^{138}	Cs^{138}	Xe^{133}	I^{133}	I^{131}
X-7-59	10.3	39	Max.	2.1×10^{-7}	5.0×10^{-8}	3.1×10^{-8}	1.5×10^{-8}	
			Min.	5.5×10^{-8}	2.6×10^{-8}	1.4×10^{-8}	4.3×10^{-9}	
			Av.	1.2×10^{-7}	3.8×10^{-8}	2.0×10^{-8}	8.8×10^{-9}	1.5×10^{-10}
X-2-q	10.2	48		6.7×10^{-8}	6.9×10^{-8}	3.7×10^{-8}		1.9×10^{-10}
X-2-s	10.3	45		1.0×10^{-7}	1.0×10^{-7}	5.5×10^{-8}	6.4×10^{-9}	1.0×10^{-8}
X-2-y	10.6	39		3.8×10^{-8}	8.6×10^{-8}	4.5×10^{-8}	4.3×10^{-8}	4.8×10^{-9}

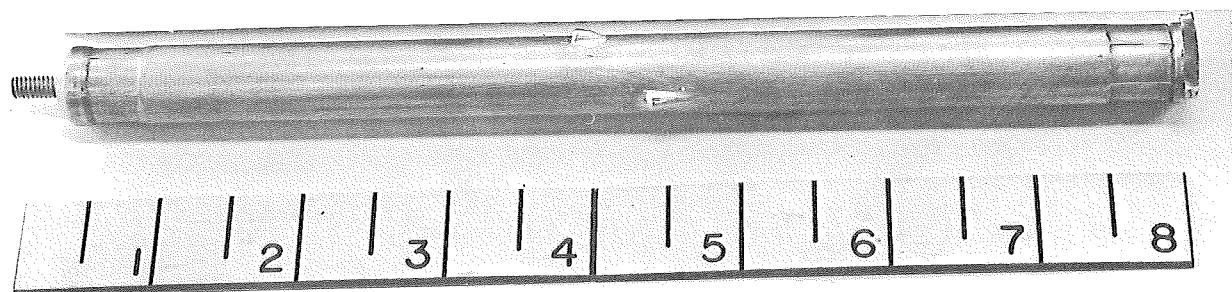


Figure 1 Fuel Specimen AHR before irradiation

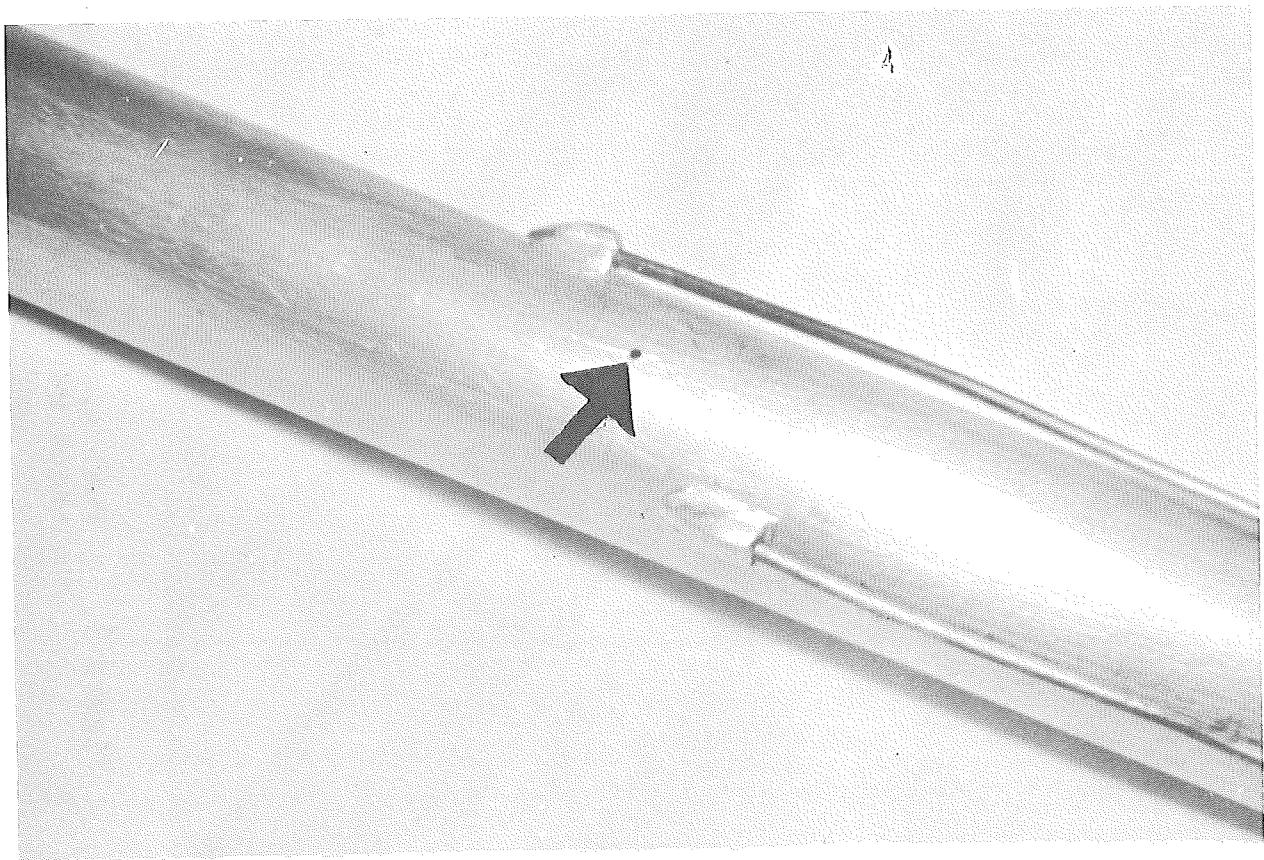


Figure 2 Element AHR showing the relative positions of the drilled defect (arrow) and thermocouple beads.

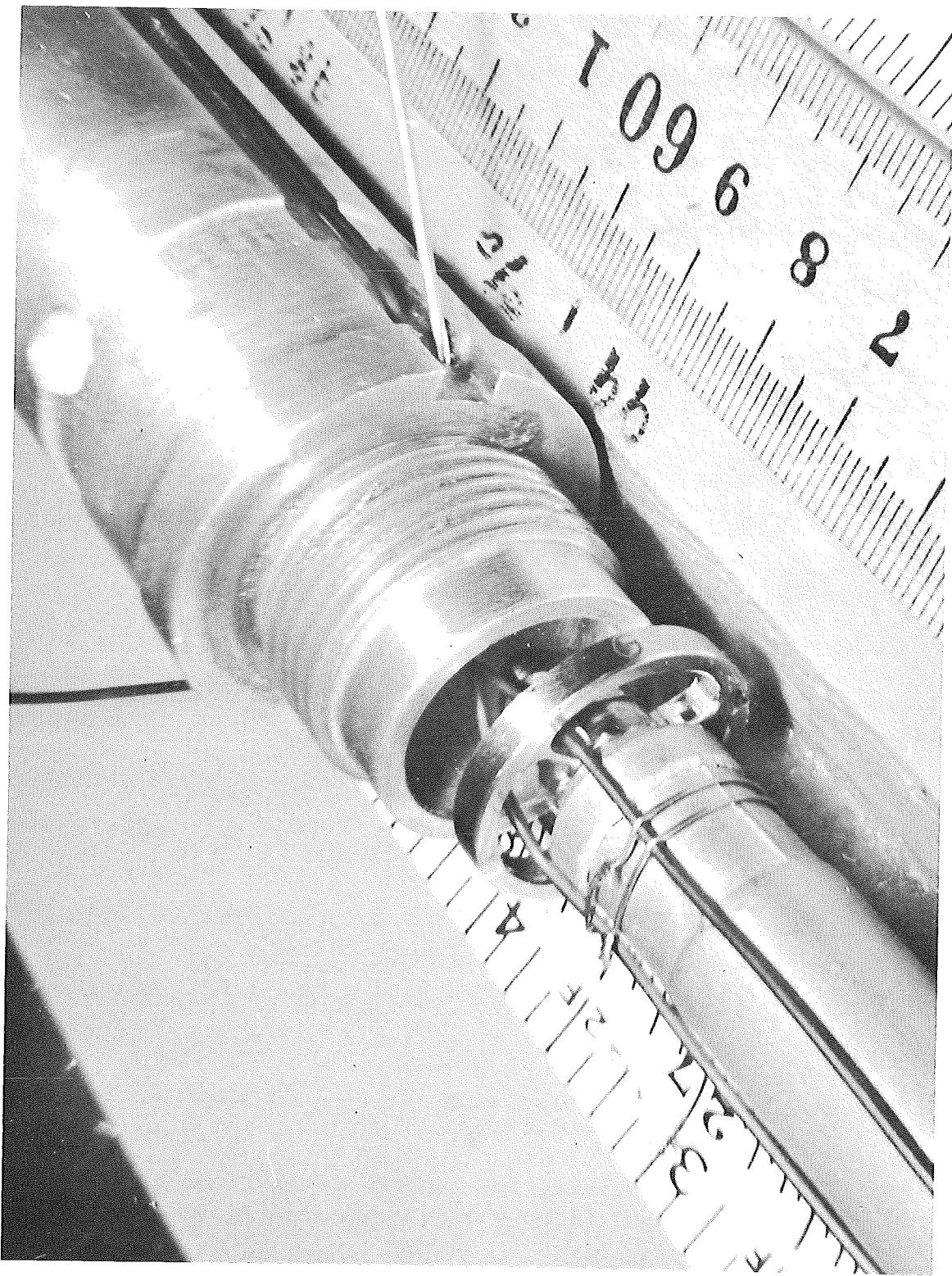
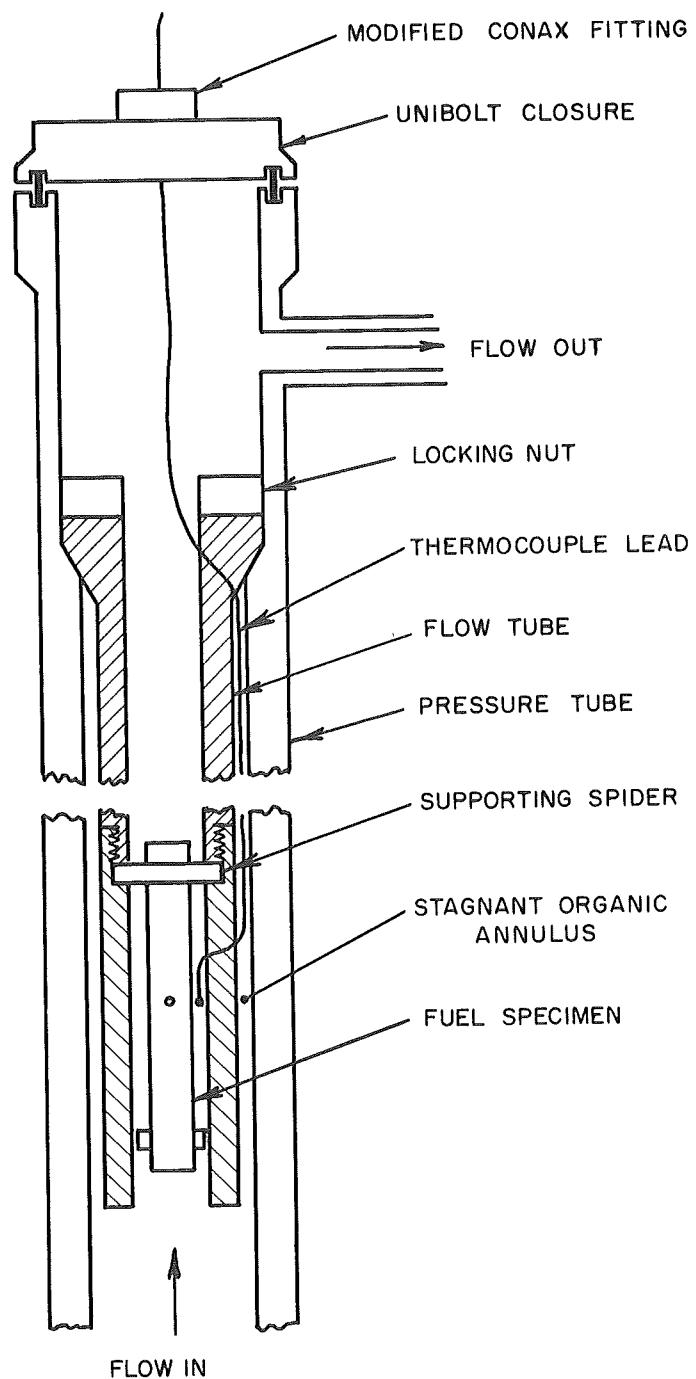


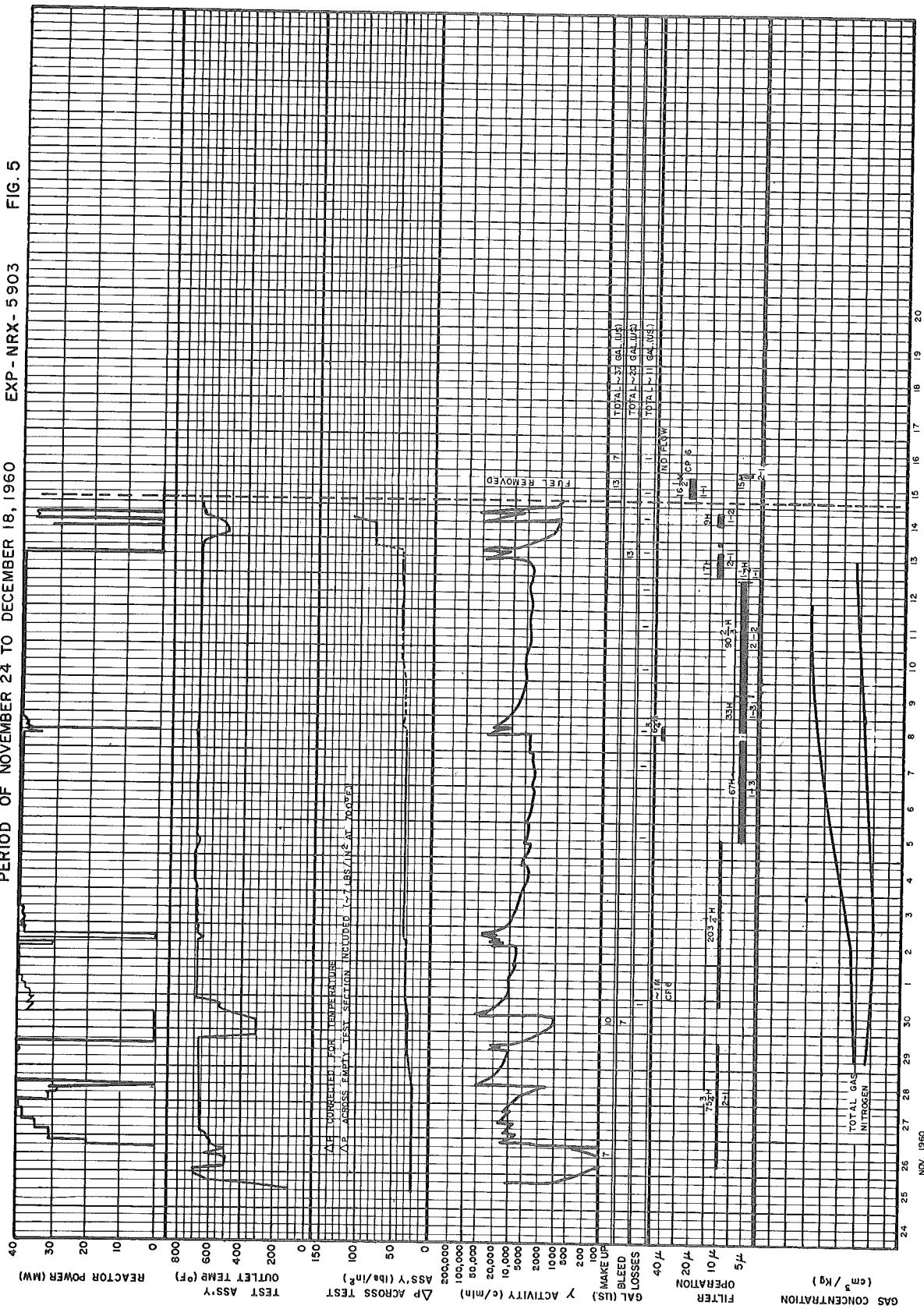
Figure 3 Fuel specimen assembly showing element and thick walled flow tube.



SCHEMATIC REPRESENTATION OF X-7 LOOP
TEST- SECTION

FIG. 4

PERIOD OF NOVEMBER 24 TO DECEMBER 18, 1960 X-7 LOOP OPERATION LOG EXP-NRX-5903 FIG. 5



EXP-NRX-5903

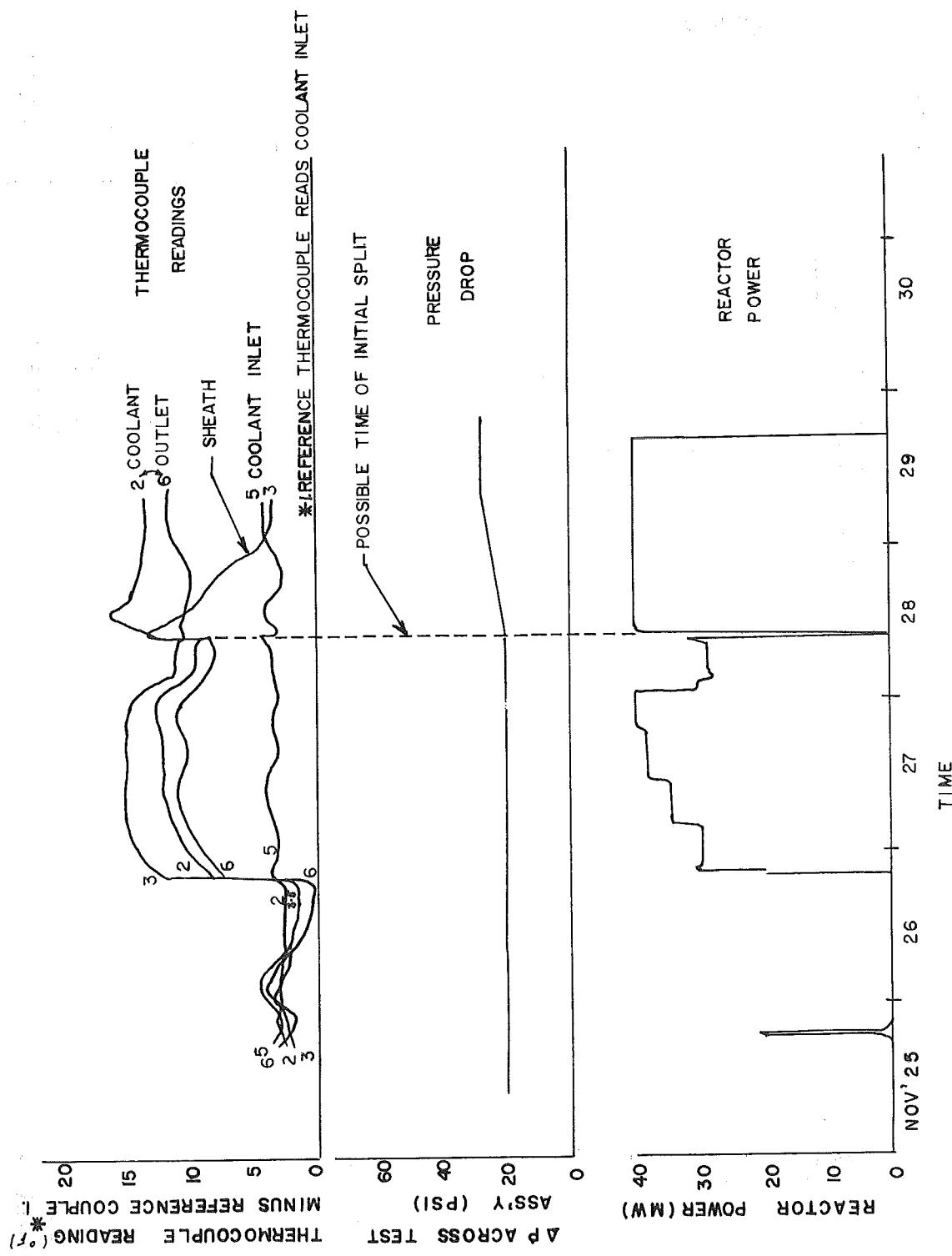
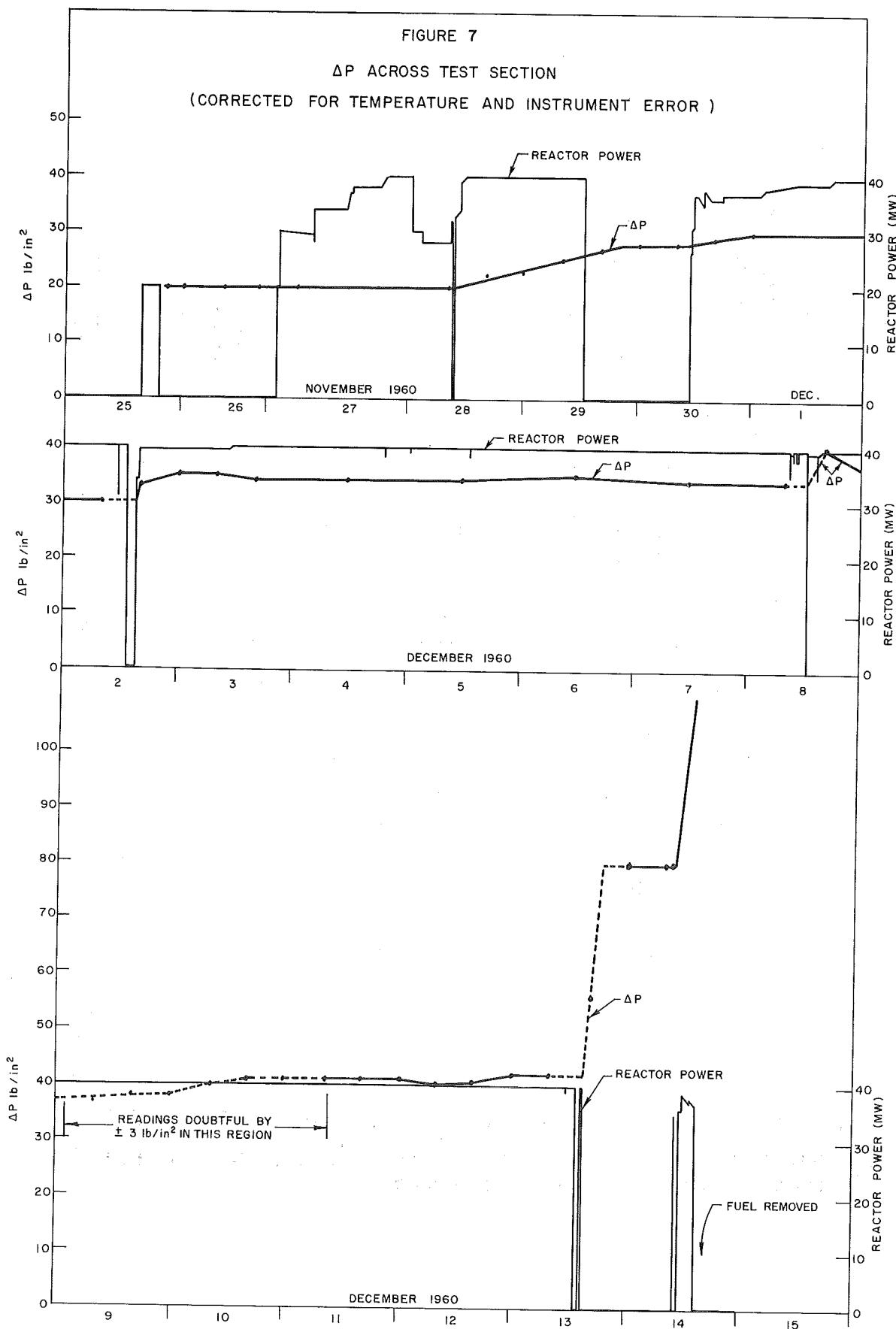
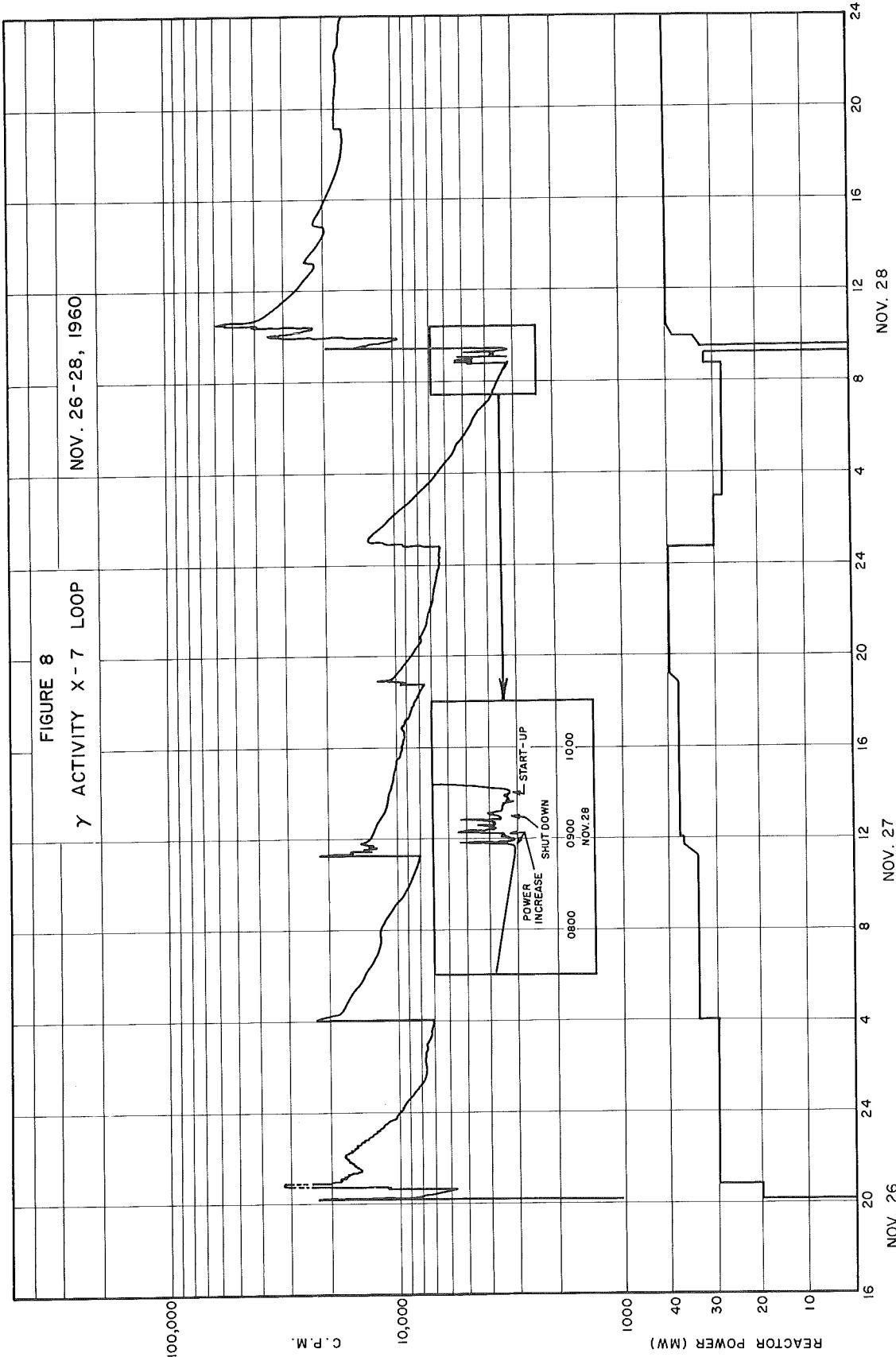


FIGURE 6 FUEL THERMOCOUPLE READINGS



EXP. NRX- 5903



EXP. NRX- 5903

FIGURE 9
 γ ACTIVITY X-7 LOOP
DEC. 13 - 14, 1960

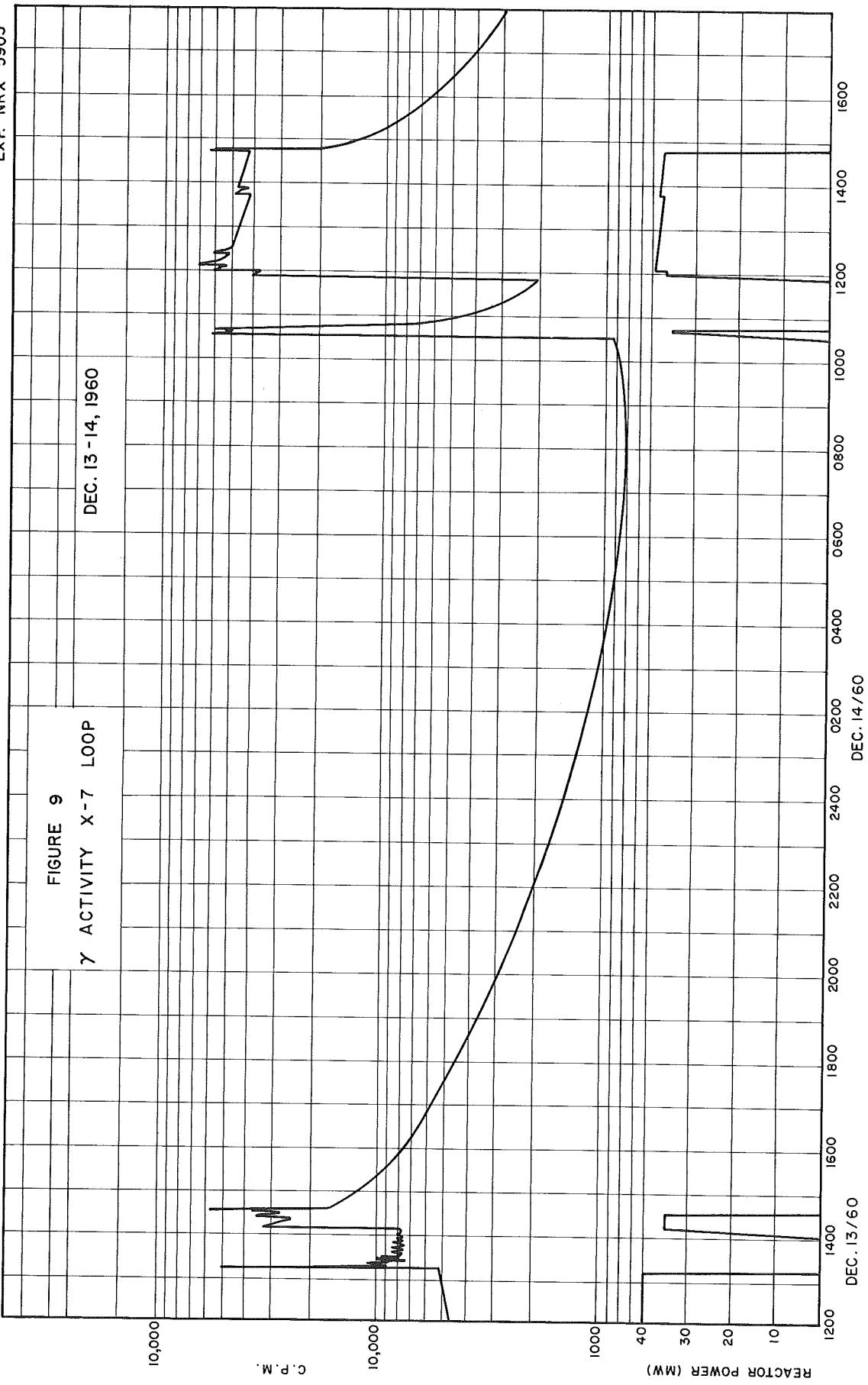
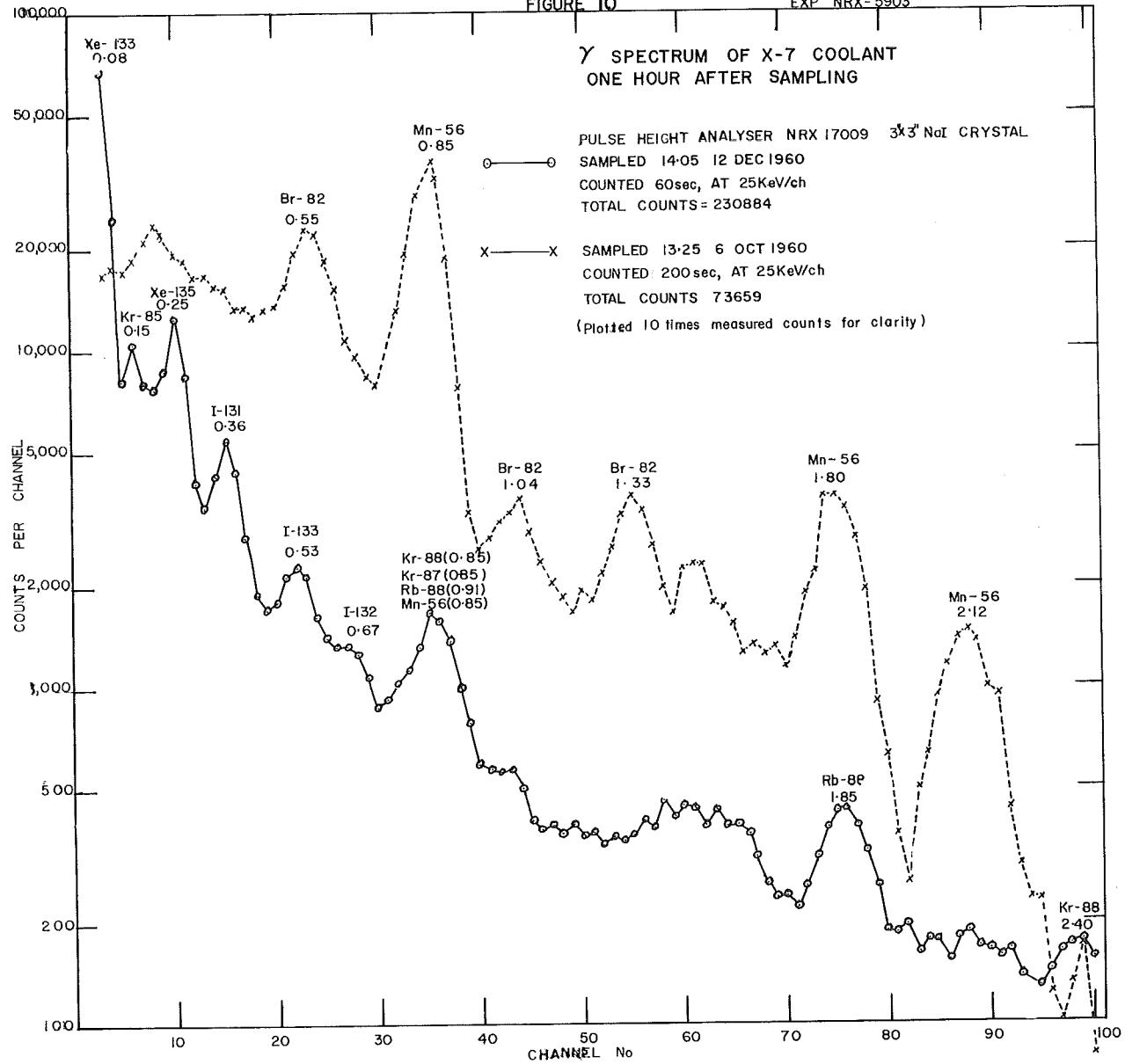


FIGURE 10 EXP NRX-5903

 γ SPECTRUM OF X-7 COOLANT
ONE HOUR AFTER SAMPLING

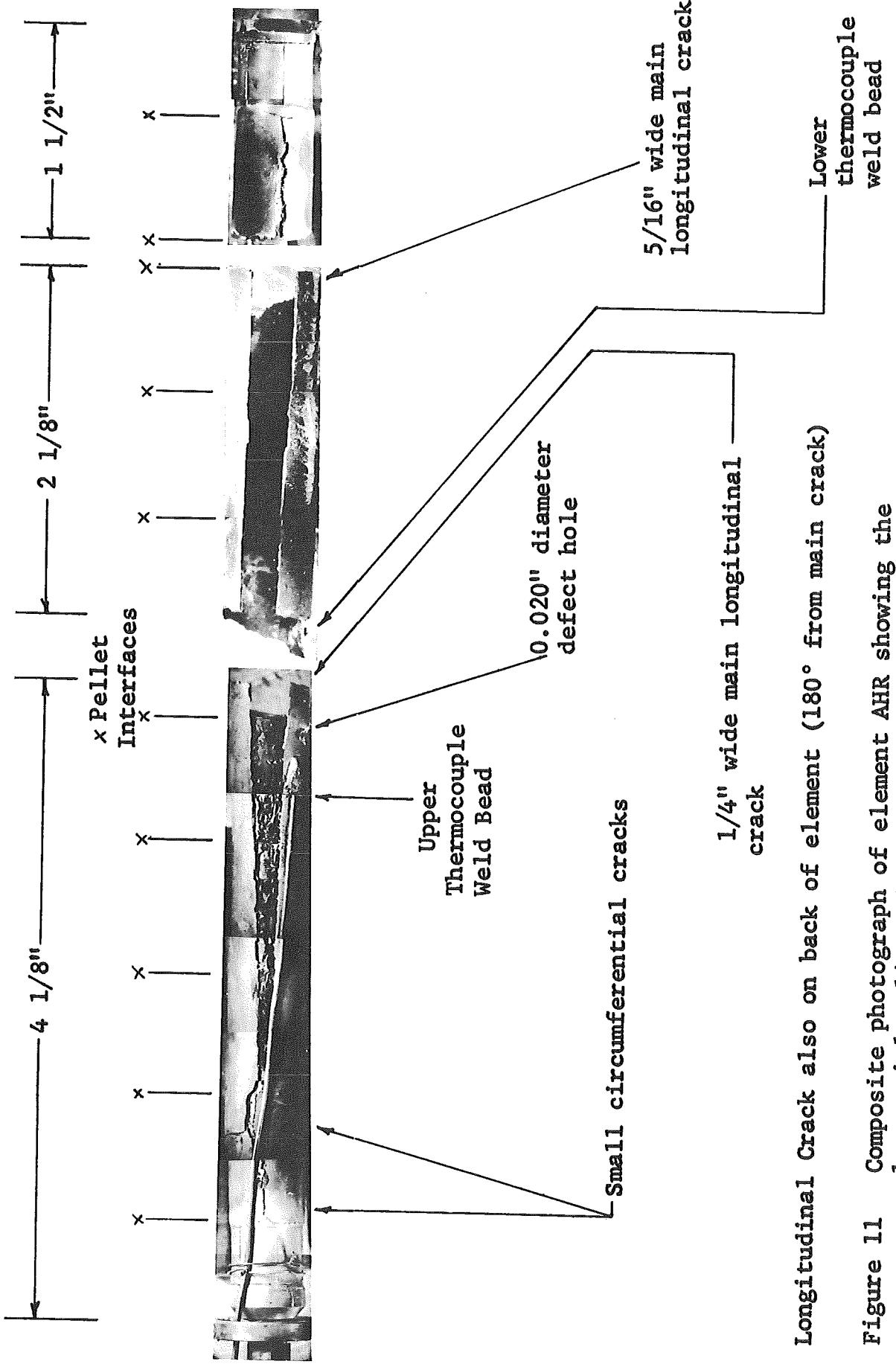


Figure 11 Composite photograph of element AHR showing the long axial split and UO_2 pellets - note surface free from film or deposit - Approximately full size.

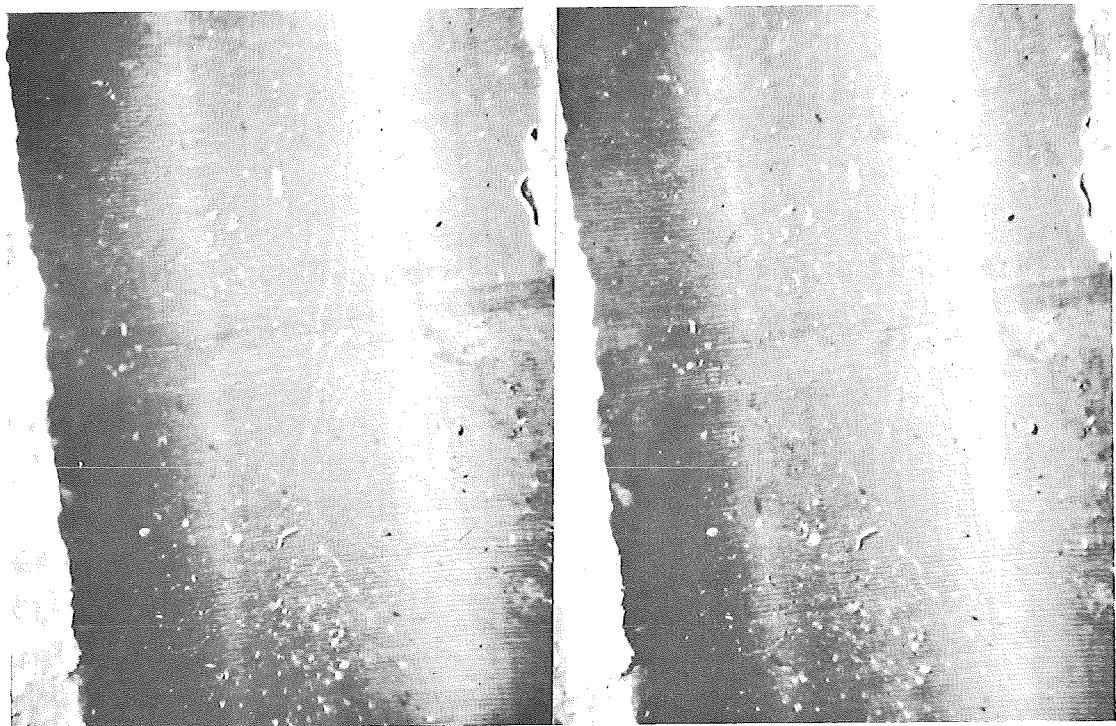


Figure 12 Inside surface of flow tube showing very slight film and machining rings - Magnification 4X - Reference No. 4542.

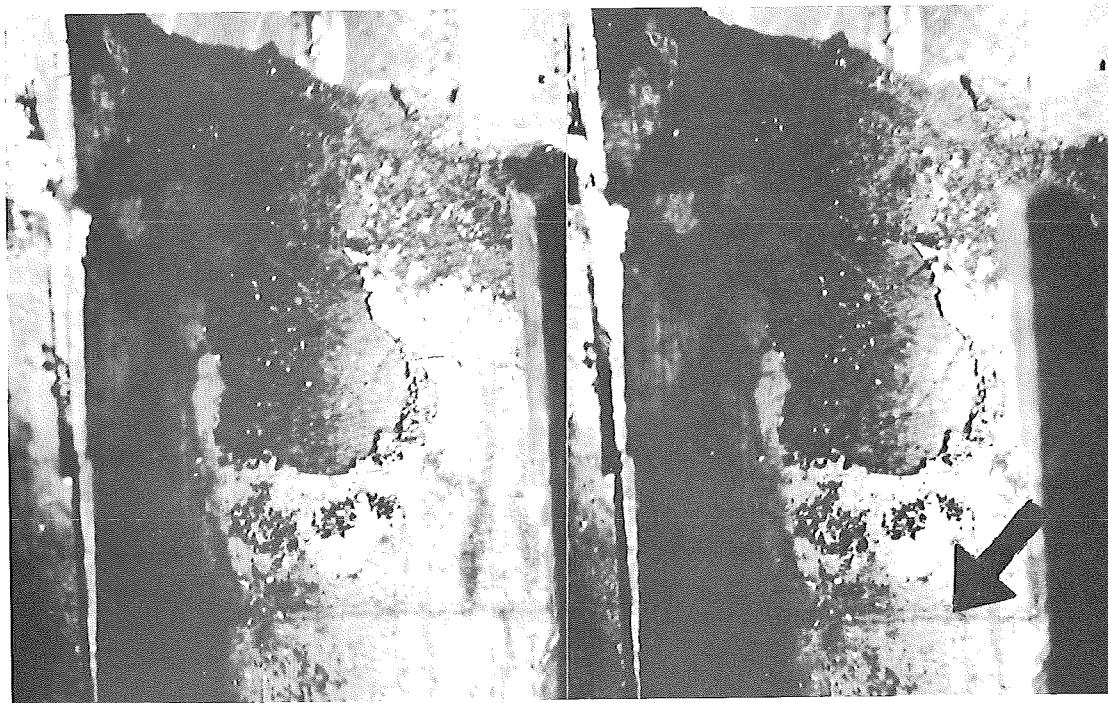


Figure 13 Inside surface of SAP sheath near middle crack with coating of organic deposit and discoloration ring (arrow) at pellet interface - Magnification 4X- Reference No. 4539.

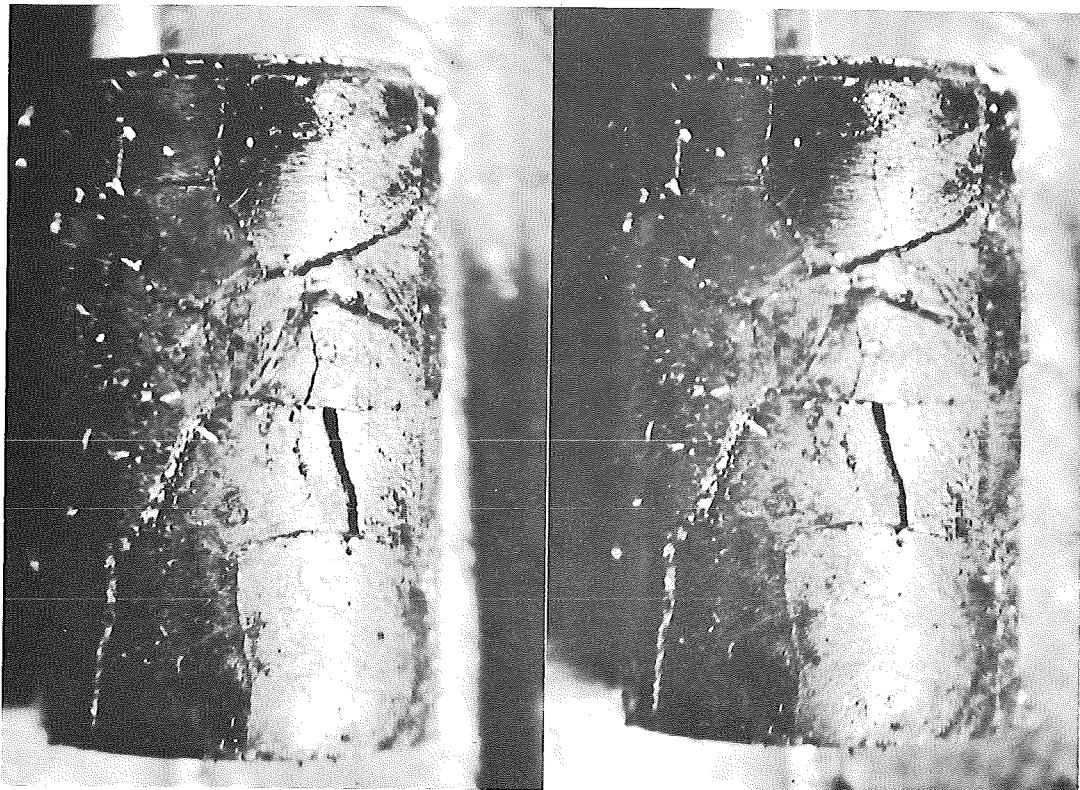


Figure 14 Organic deposit on pellet No. 33 note cracks in pellet match those of the deposit. Magnification 4X - Reference No. 4544.

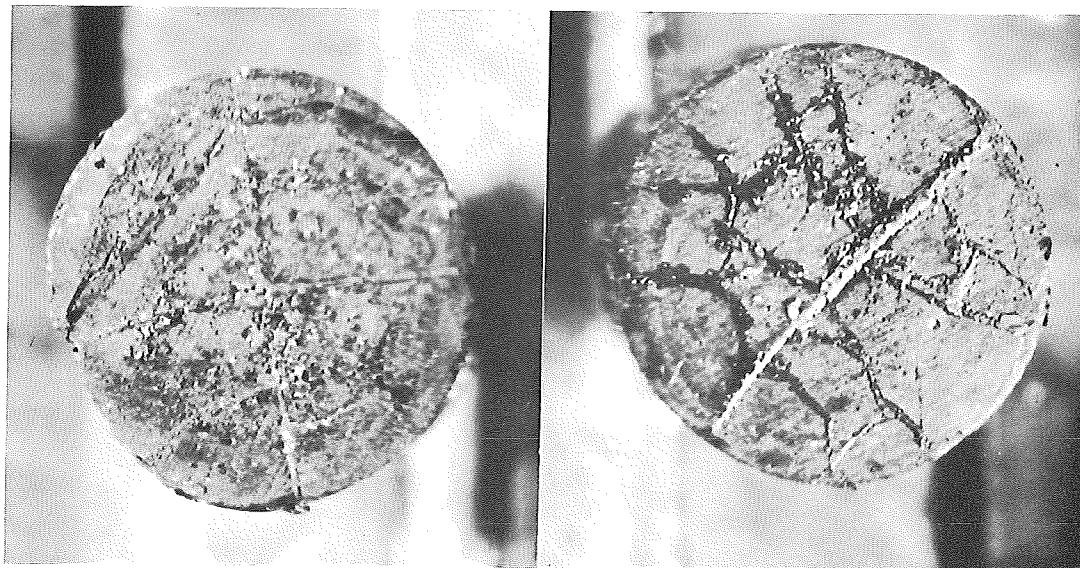


Figure 15 Right photograph shows the end of pellet No. 33 with straight line crack filled with deposit. Left photograph the flat end of pellet No. 80 showing cracking and deposit in a cross pattern. Magnification 4X - Reference Nos. 4545 and 4515.



Figure 16 Left photograph cross-section of pellet No. 34 showing organic penetration and slight cracking. Right photograph cross-section of pellet No. 39 with organic penetration $1/3$ radius (arrow) and coarse grains at center. Magnification 4X - Reference Nos. 4558 and 4562.

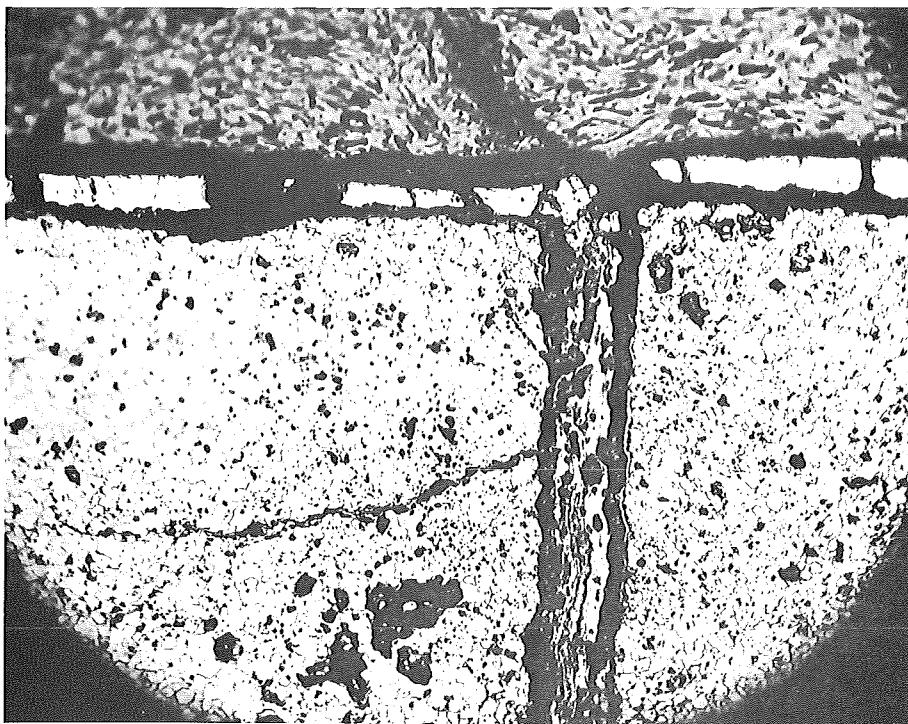


Figure 17 Layer of organic along periphery of pellet also showing penetration into crack. Magnification 100X - Reference No. Z50A4.

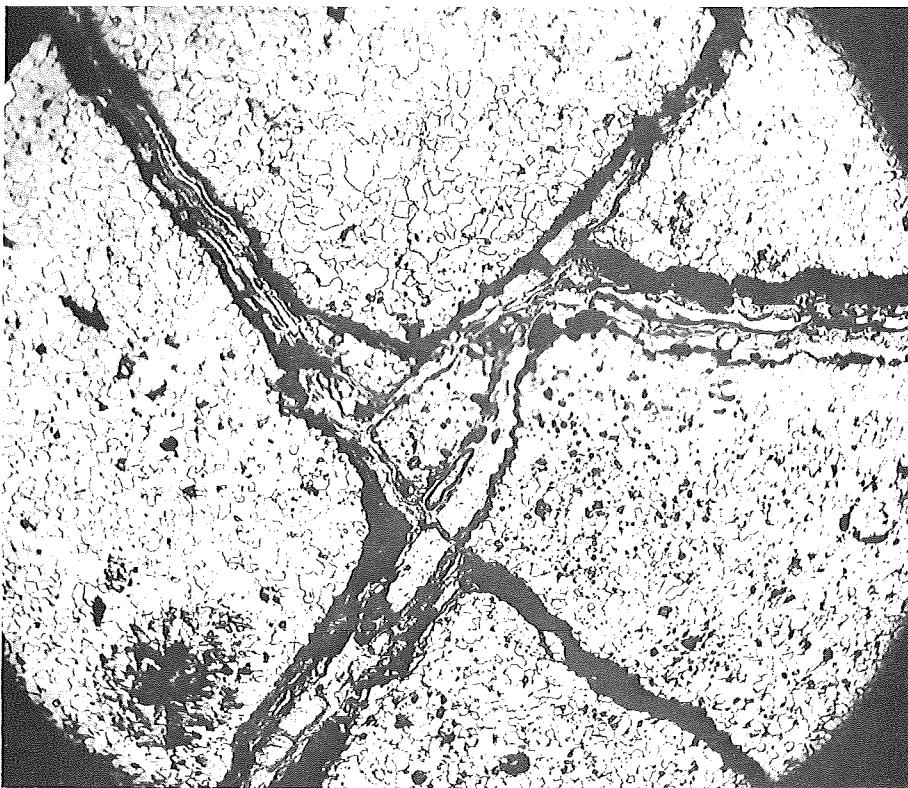


Figure 18 Deposit of organic along cracks near center of pellet. Magnification 100X. Reference No. Z50A3.

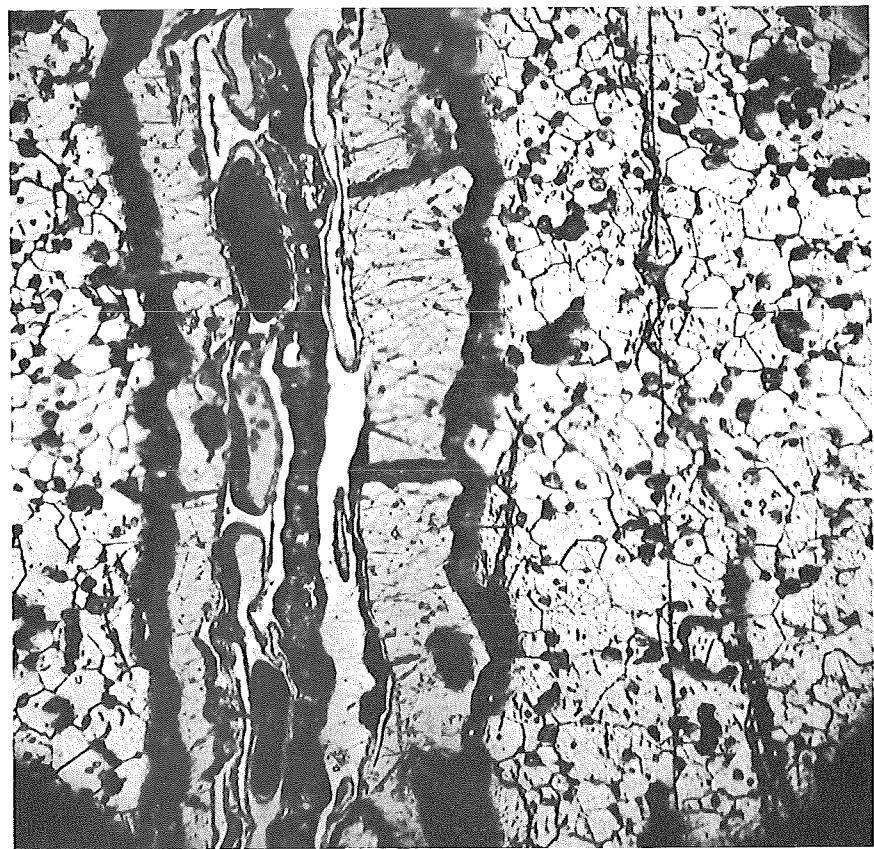


Figure 19 Appearance of organic layers in cracks.
Magnification 500X - Reference No. Z50A2.

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