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FISSION PRODUCT BEHAVIOR WITHIN TWO W-UO_2 CERMET FUEL ELEMENTS IRRADIATED IN A TEMPERATURE GRADIENT

August, 1968

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FISSION PRODUCT BEHAVIOR WITHIN TWO W-UO_2 CERMET FUEL ELEMENTS IRRADIATED IN A TEMPERATURE GRADIENT

by

D. A. Seifert, R. L. Stuart, P. K. Conn,
H. W. McLaughlin and G. J. Luersen

August, 1968

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TABLE OF CONTENTS

Page

ABSTRACT 3

INTRODUCTION 5

EXPERIMENTAL 6

- Specimen Description and Irradiation 6
- Gamma-Scan 7
- Mobile Gas Recovery 7
- Selective Dissolution of Fuel and Matrix 14
- Analysis of Fission Gases 19

RESULTS AND DISCUSSION 20

- Significance of Results for Operating Fuel Elements 34

CONCLUSIONS 35

ACKNOWLEDGEMENT 37

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ABSTRACT

A post-irradiation study on two 22.9-cm-long W-UO_2-ThO_2 cermet fuel specimens, one clad with T-111 and the other with W-Re-Mo, was carried out to determine the retention, distribution, and mobility of fission gas in cermet core fuel systems. The T-111- and W-Re-Mo-clad specimens, irradiated for 5000 hours to peak burnups of 5.6×10^{19} and 5.0×10^{19} fissions/cm^3 with temperature ranges of 530° to 1400°C and 770° to 1550°C, respectively, were gamma scanned and then punctured; fission gases which were free to migrate through flow channels between the fuel and matrix phases to the punctured holes were collected and analyzed for Kr^{85}. Thin transverse sections were cut from various positions along the lengths of the specimens, selectively dissolved using a technique developed at GE-NMPO, and their Kr^{85} and Cs^{137} contents determined for both the ceramic fuel and tungsten matrix phases.

Inventories and distributions of Cs^{137} and Kr^{85} between the UO_2 and tungsten phases were obtained as a function of temperature for both specimens by combining the normalized gamma scans with Kr^{85} recovery data and Cs^{137} and mass spectrometric burnup data. The analysis showed that 90 to 95% of the total fission gas inventories were either dissolved in the UO_2 and tungsten lattices or trapped in closed pores. The remaining 5 to 10% of the gases were mobile and free to migrate through channels between the fuel and matrix phases.

Fifteen to twenty percent of the fission products recoiled into the W matrix and were essentially immobile under the conditions of these experiments. The gas which was free to migrate was released only from the fuel phase; gas release was negligible below about 650° to 700°C, and at 1550°C amounted to about 20% of that produced in that temperature region of the specimen. Cs^{137} was quantitatively retained by both the fuel and matrix phase for the conditions of these tests and, therefore, did not contribute to the amount of mobile gas.

INTRODUCTION

An investigation was made of the effects of irradiation temperature on the distribution of gaseous fission products within two W-UO_2 cermet fuel specimens. Its intent was to better understand the influences of fission gas accumulation and retention and fission gas distribution and mobility on the swelling and leak-free life capabilities of refractory-metal-clad cermet core fuel systems. Previous[1] laboratory studies conducted on unirradiated cermet core specimens of this type indicated that rare gas percolated through the channels between the matrix and fuel phase at room temperature. Gas flow decreased with increasing temperature until the difference in thermal expansion between the fuel and matrix phases caused the gas flow to essentially cease.

A puncture technique was used to determine the amount of mobile gases (that fission gas free to migrate within the fueled core at room temperature) in the irradiated specimens. Data from previous[2] puncturing studies, made on short (3.8 cm fueled length) thin annular cermet fuel specimens, demonstrated that essentially all of the fission gases released to the channels between the fuel and matrix phases of cermet fuel cores were recovered from the short specimens by this technique. Selective dissolution of the two phases of the cermet core was used to determine the fission gas retention in and distribution between each phase of the core. A previous[3] selective solutioning study conducted

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[1] "Sixth Annual Report - High-Temperature Materials Program," GE-NMPO, GEMP-475B, February 28, 1967, pp. 182-3.

[2] "AEC Fuels and Materials Development Program Progress Report No. 68," GE-NMPO, GEMP-68, August 31, 1967, pp. 61-6.

[3] "AEC Fuels and Materials Development Program Progress Report No. 66," GE-NMPO, GEMP-66, May 31, 1967, p. 68.

on a similar cermet core specimen demonstrated the validity of the technique as a means for separating and recovering the fission gases contained in the fuel and matrix phases of irradiated cermet cores. It was not known, however, whether recoveries were quantitative because this specimen was not punctured and a mass balance could not be performed.

This study of the effects of temperature on fission gas distribution within the irradiated specimens was carried out using the radio-nuclide Kr^{85}. Chosen because it is easily measurable (not requiring sophisticated volumetric apparatus) and has a long half-life (10.76 yr), Kr^{85} best represents the stable fission gases which continuously accumulate throughout long irradiation times (the short lived fission gases reach an equilibrium concentration dependent on their halflives and production rates and quickly decay after irradiation is terminated).

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EXPERIMENTAL

Specimen Description and Irradiation - Two 22.9-cm long x 1.2-cm diameter W-54UO_2-6ThO_2(vol %) annular core samples, one clad with T-111 (Ta-8W-2Hf wt %) and the other with W-30Re-30Mo (at %) were irradiated in reactor experiments LT-710-2 and LT-710-3 respectively, for a total of 5000 hours each. The LT-710-2 specimen longitudinal temperature profile ranged from an estimated maximum of 1400°C near one end to 530°C at the other and the LT-710-3 longitudinal temperature profile ranged from a measured 1540°C at one end to 770°C at the other.

Gamma-Scan - In order to obtain detailed burnup profiles of the irradiated specimens without excessive cutting and chemical analyses, a gamma activity profile was made of each using a 400-channel gamma scintillation spectrometer operating in the multi-channel scaler mode and connected to a pulse generator and a scaler timer. The samples were traversed past the collimator-detector at constant speed using the motorized stage device shown in Figure 1. With the spectrometer-multi-scaler set to accumulate all gamma counts in a new channel at given preset intervals, gross activity profiles were made. Gamma-energy spectra were also collected at 1/2-inch intervals along the length of each specimen. $Zr\text{-}Nb^{95}$ and $Ba\text{-}La^{140}$ were the main fission product contributors to the gamma spectra. Longitudinal concentration profiles of these isotopes obtained by determining the counts under their respective photopeaks (0.75 to 1.60 Mev) are shown in Figure 2 along with the gross gamma scans.

Mobile Gas Recovery - Puncturing of the specimen claddings was accomplished remotely in the GE-NMPO hot cells using a vacuum puncturing and gas collection apparatus as shown in Figure 3a. The apparatus consisted of a vacuum drilling chamber mounted on a standard drill press, a 1 liter collection bottle containing 30 grams of 12/28 mesh activated charcoal, a mercury diffusion pump, and the necessary connecting manifolds, valves, and gauges. With a specimen sealed to the drilling chamber, the entire system was evacuated to $\sim 10^{-5}$ torr. The pumping station was then valved out of the system and the specimen drilled with a solid carbide

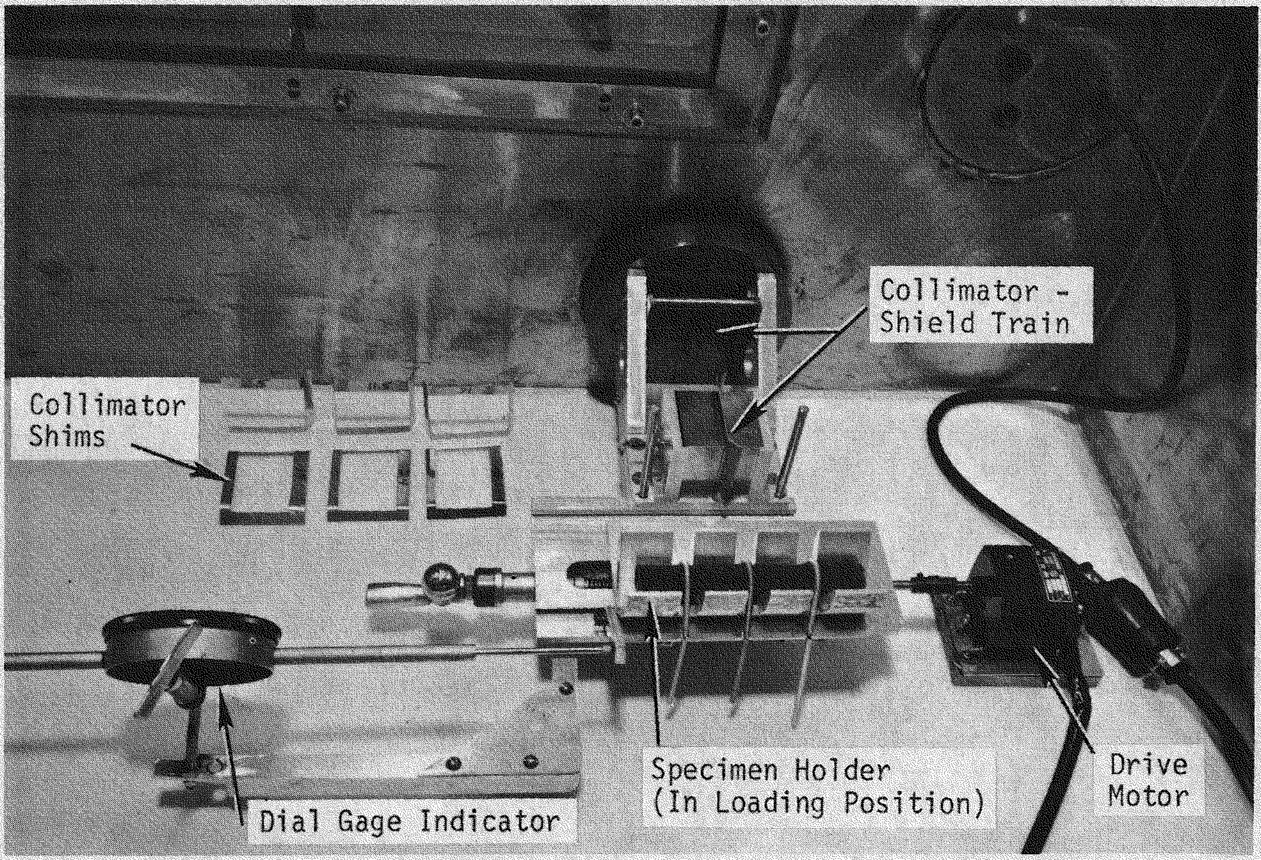

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Figure 1 - Motor-driven sample holder used for gamma scanning irradiated fuel samples.
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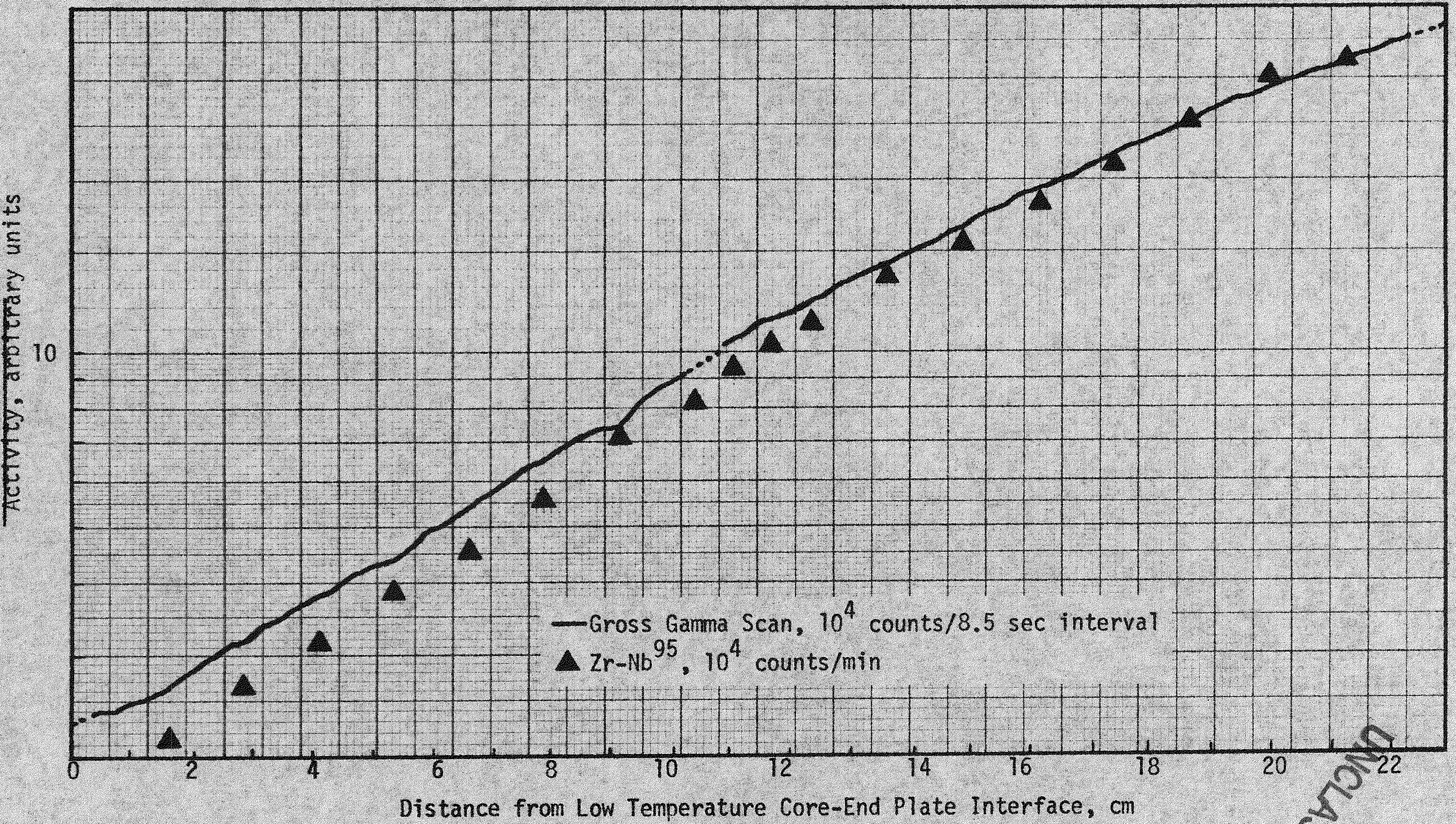

Figure 2a - LT-710-2 gamma scan

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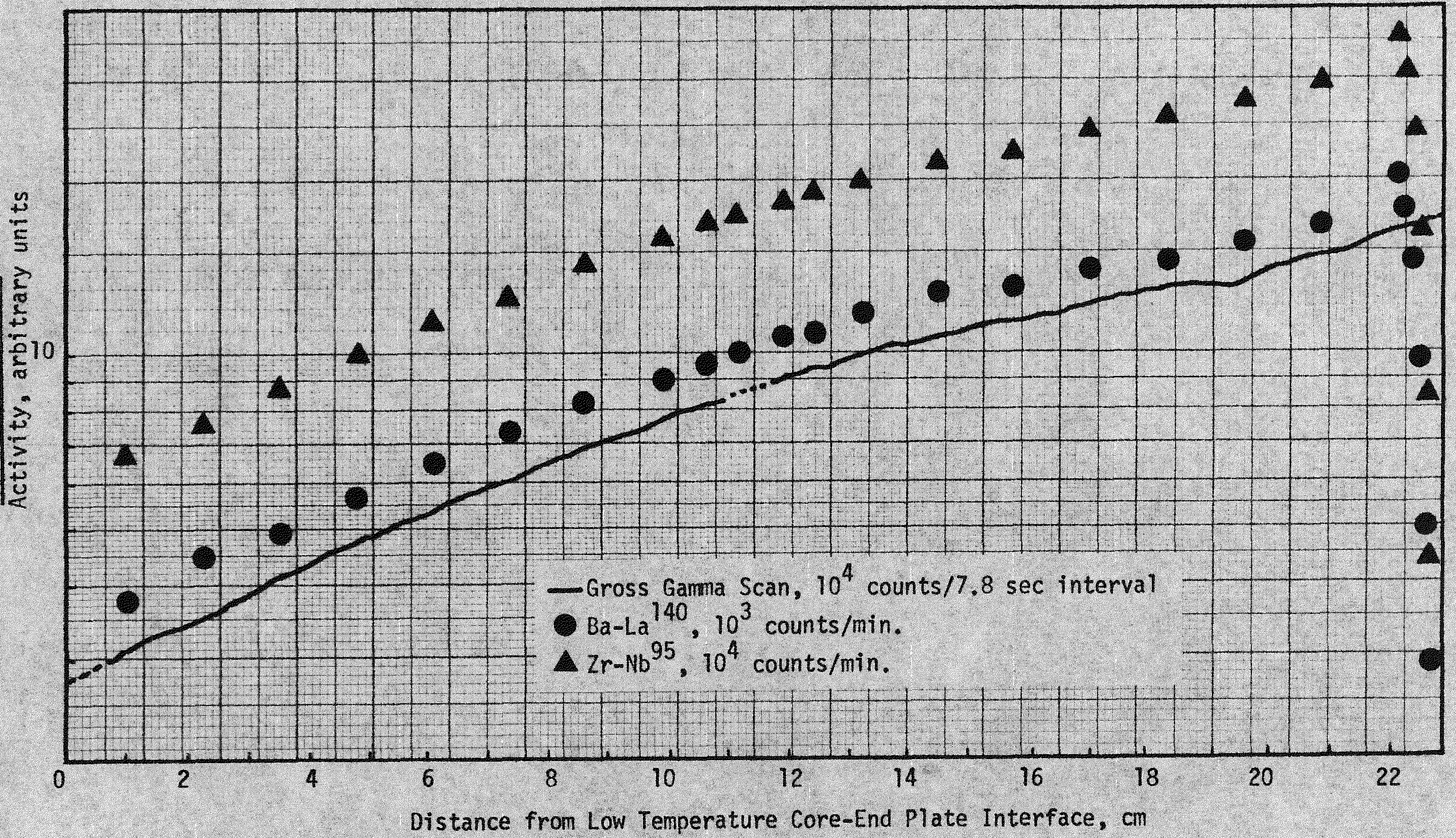

Figure 2b - LT-710-3 gamma scan

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Figure 3a - Remotely operated vacuum puncturing and gas collection apparatus.

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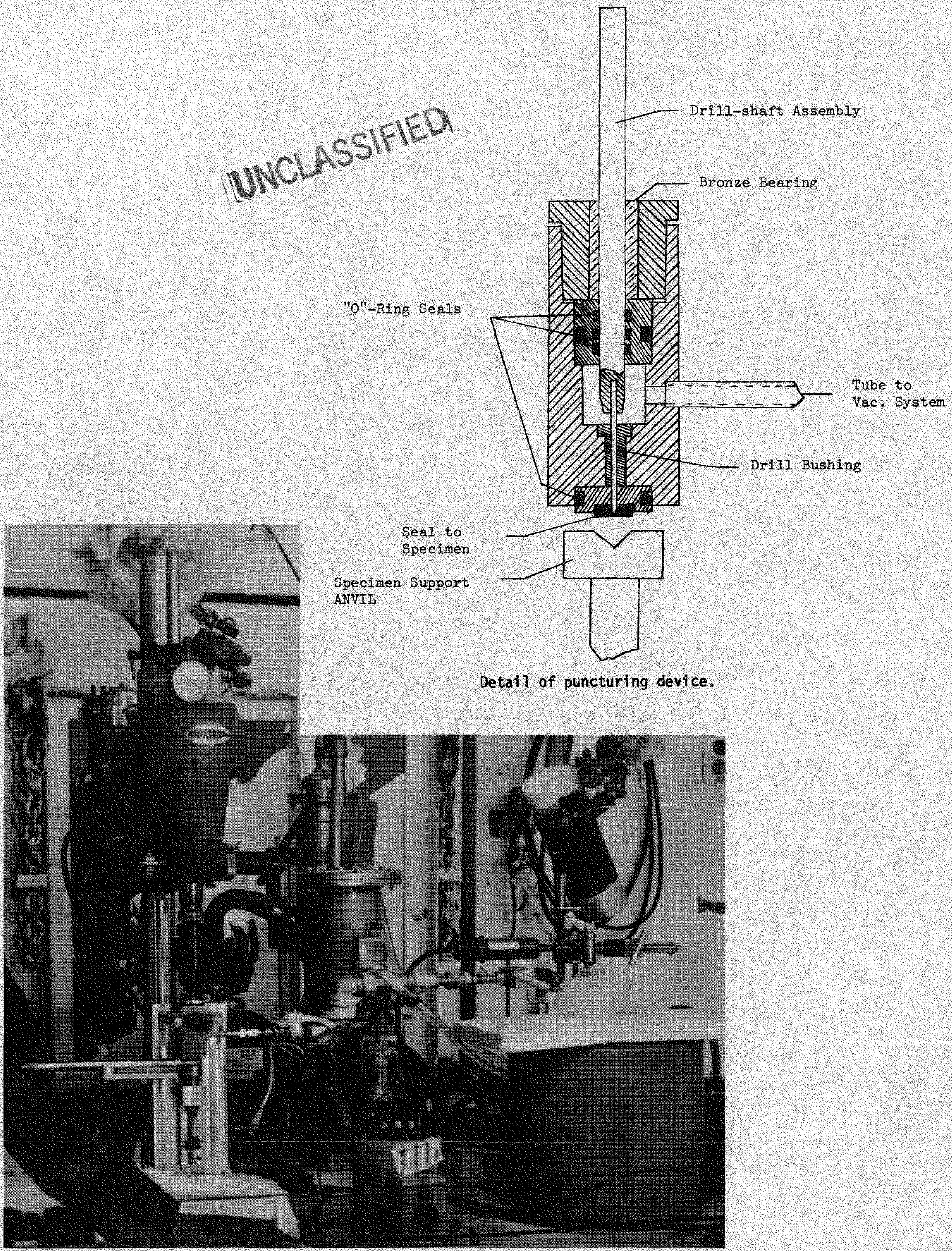
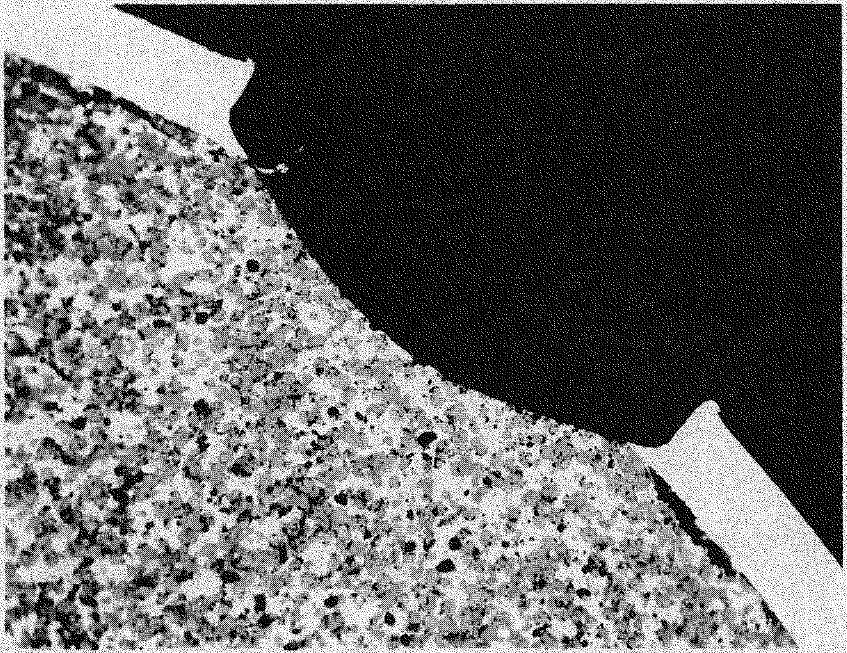


Detail of puncturing device.

straight-fluted drill to a great enough depth to penetrate the cladding as indicated by a dial gauge micrometer mounted in contact with the drill press spindle. A typical cross section of a drilled hole is shown in Figure 3b. The gases released on puncturing were concentrated in the collection vessel by absorption on the activated charcoal at liquid nitrogen (LN_2) temperature.

The LT-710-2 specimen was punctured three times (once near each end and once in the center), a 6-hour gas sample being collected from each hole. During the 3 days to 1 week that elapsed between punctures, the specimen was left exposed to the atmosphere and at the time it was thought that some gas may have been lost during the time lapse between punctures. To avoid this possibility of gas loss between punctures with the 710-3 specimen, the drilling chamber was sealed from the rest of the system after the initial puncture (near the low temperature end) and 6-hour gas collection interval. The gas collection vessel was then sealed, removed from the system, and replaced by a second collection vessel. Sampling of gases from the drilled specimen was continued for some 23 hours after re-evacuation of the connecting manifold between the puncturing device and the collection vessel. The specimen was then removed from the puncturing device and placed in a small-volume chamber which was evacuated and sealed and could be sampled from outside the hot cells. Two additional gas samples were collected from this chamber over a period of 282 hours. Three additional punctures (one near the high temperature end and single ones about 2-1/2 inches from each end) of the specimen were made and two gas samples, one from



LT-710-3 Drill Hole

35X
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Figure 3b - Transverse cross section of the initial puncture of the LT-710-3 specimen.

the puncture device and one from the vacuum chamber, taken from each over a total sampling time of 341 hours. At no time during the entire 650 hour gas sampling interval was the specimen allowed to remain disconnected from a gas collection device for more than 30 minutes.

Selective Dissolution of Fuel and Matrix

The technique developed for selectively dissolving the fuel and matrix phases of an ∿ 0.6 mm-thick slice of refractory metal clad W-UO_2 cermet involves first, treatment with concentrated nitric acid in a closed system to dissolve the fuel phase and then electrolysis in 20% NaOH to dissolve the W phase of the core. The effectiveness of nitric acid in removing the ceramic fuel without significantly affecting the metallic matrix was previously demonstrated.[3] Efficient use of this technique required that the cermet specimens be thin enough to permit complete removal of that portion of the ceramic fuel phase not completely enveloped by tungsten before matrix dissolution was begun. Transverse sections of the required thickness (0.5 to 0.8 mm) were cut from the cermet fuel elements using a thin silicon carbide wheel cooled by a water spray.

Fuel dissolution was carried out in the apparatus shown in Figure 4. The cleaned, weighed cermet fuel specimen was positioned between the opposing indentations in the dissolver vessel to maintain it in a vertical position directly above the sweep gas inlet nozzle. The system

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[3] "Sixth Annual Report - High Temperature Materials Program Part B," GE-NMPO, GEMP-475B, February 28, 1967, pp. 183-4.

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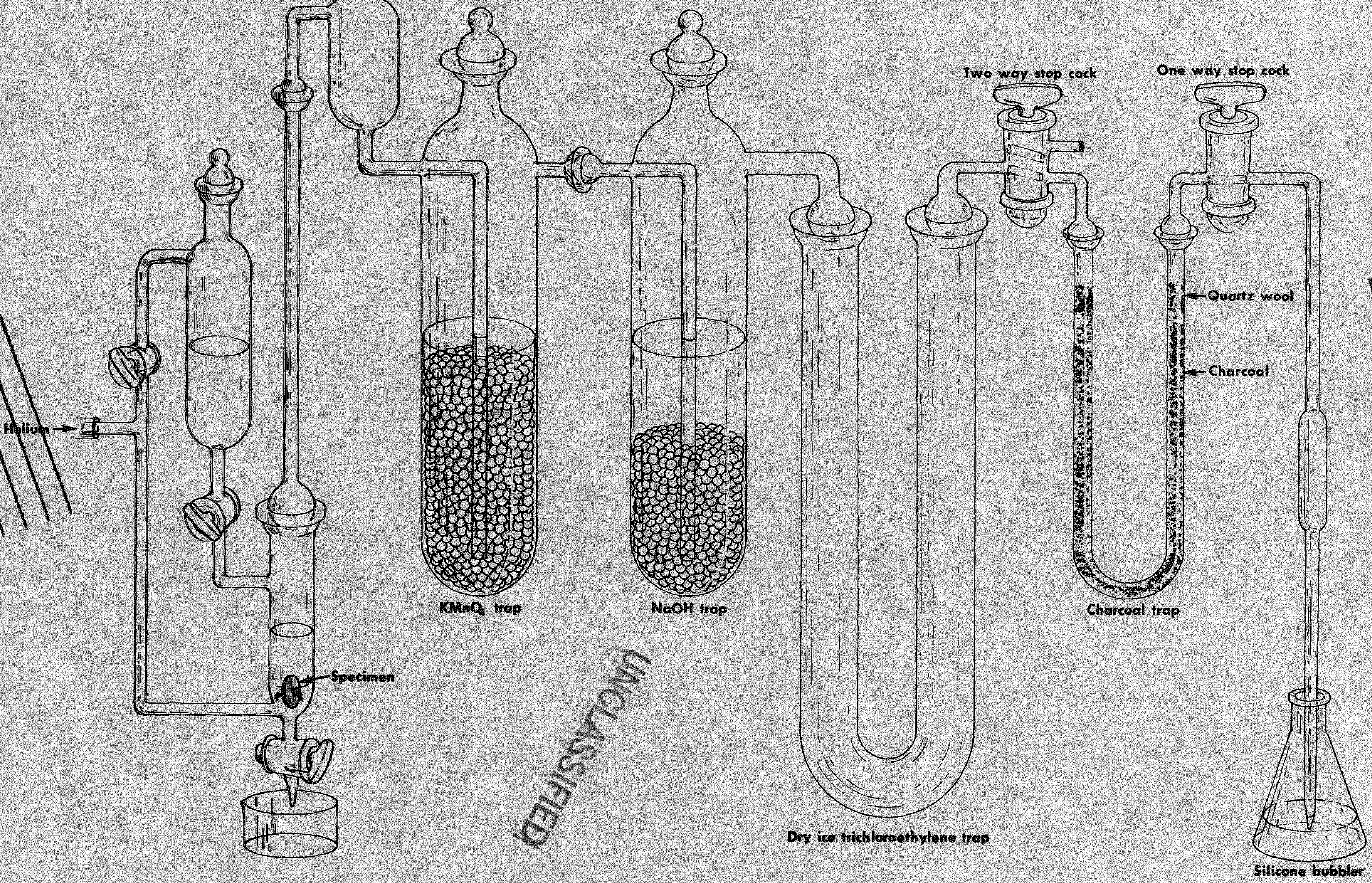

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Figure 4 - Fuel phase dissolution apparatus. (AS-782)

was then sealed and purged with the helium sweep gas for 45 minutes before diverting the flow through the charcoal-filled U-trap and adjusting it to about 1 bubble per second in the washing traps. Concentrated nitric acid to completely cover the specimen was introduced from the pressurized reservoir and the dissolver vessel heated gently with hot air to initiate the reaction. Dissolution, which initially proceeded with the evolution of gas bubbles from the specimen and the formation of strong brown fumes, was continued for a period of 20 to 24 hours. During this time the acid was drained, replaced and heated four or five times. The evolved oxides of nitrogen along with the released fission gases and the He sweep gas passed from the dissolver into a series of three cleansing traps. The first of these contained concentrated potassium permanganate to oxidize any N_2O to NO; the second contained 15N sodium hydroxide to remove NO, NO_2, and such fission products as iodine and ruthenium.[4] The third trap was a large U-tube chilled with a dry ice-trichloroethylene mixture to remove any water vapor from the gas stream before it passed into the LN_2 chilled U-trap where the fission gases were absorbed on activated charcoal. When dissolution of the fuel phase was complete, as evidenced by the absence of yellow color and very low radioactivity in the drained acid, the specimen was rinsed thoroughly with water and dried by heating the dissolver vessel with a stream of hot air. The He sweep was continued for 20 minutes after heating was discontinued to insure complete transfer of released fission gases to the charcoal trap before the apparatus was disassembled

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[4] B.F. Rider, C.P. Ruiz, J.P. Peterson, Jr., F.R. Smith, "Accurate Nuclear Fuel Burnup Analysis, Fourteenth Quarterly Progress Report," GE-VAL-GEAP-4893, June 1, 1965, p. 5.

and the specimen removed for weighing to determine fuel content by weight loss.

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Matrix dissolution was carried out in the apparatus shown in Figure 5. After determining the amount of fuel removed by the acid leach, the specimen was placed flat against the platinum wire loop in the bottom of the electrolytic dissolution cell. Carefully positioning the remaining clad-W matrix so that only the W matrix made direct contact with the wire electrode, the specimen was held in place from above and below by Teflon-coated bar magnets. The cell was filled with a 20% sodium hydroxide solution, sealed to the remainder of the apparatus and purged with the He sweep gas for 45 minutes before dissolution was started. A current of about 0.5 amp was maintained across the cell until the matrix was entirely dissolved. Hydrogen, liberated at the side-arm cathode, was allowed to escape to the atmosphere. As dissolution proceeded, oxygen was evolved from that portion of the platinum wire anode no longer in contact with the sample and carried through the system by the He sweep along with liberated fission gases. The evolved oxygen, which competes with the fission gas for absorption on the charcoal, was removed by passage of the sweep gas over hot copper (800°C). From the combustion furnace, the gas passed through a water-cooled condenser to lower its temperature before entering the large dry-ice-trichloroethylene-chilled U-trap for removal of any moisture content. The sweep then passed into the LN_2 chilled carbon bed where the fission gases were absorbed and held. Dissolution was usually complete within two hours.

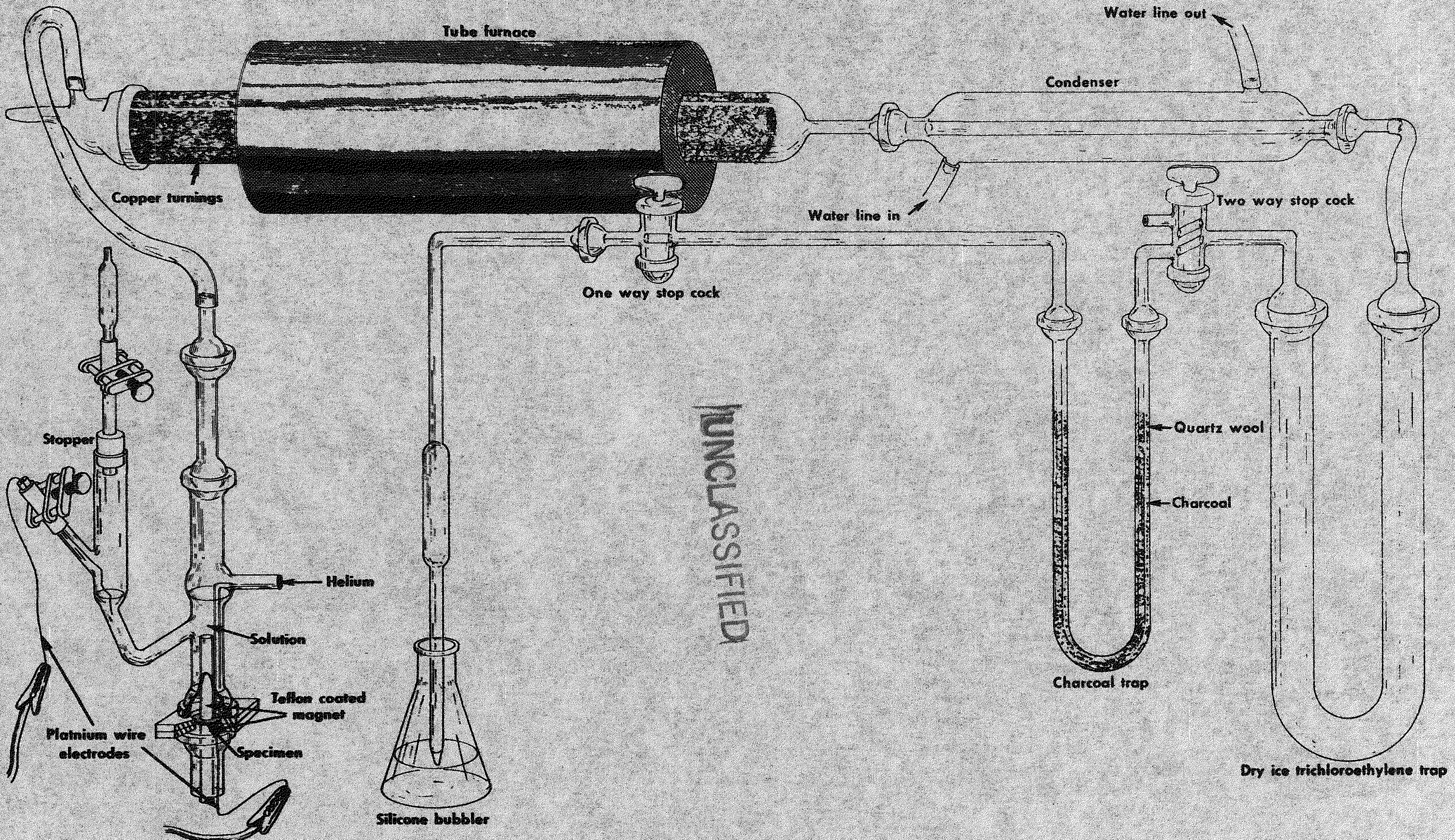

Figure 5 - Matrix dissolution apparatus. (AS-783)

The He flow was maintained for an additional hour to insure complete transfer of fission gas to the charcoal absorption bed.

The acid leach solutions from the fuel phase dissolution were combined and diluted to volume along with a few drops of cesium, cerium, and zirconium carrier solutions. The electrolyte from the matrix dissolution was decanted into a second volumetric flask. The remaining insolubles, less than 1% of the total sample weight, were washed and centrifuged, the wash solution being added to the electrolyte. Cesium carrier was also added to the electrolyte solution which was then diluted to volume. The insoluble fraction, consisting of undissolved fuel and matrix particles along with such fission products as Zr^{95}, Nb^{95}, Ce^{141}, Ce^{144}, Ru^{103}, and Ru^{106} which precipitate in NaOH, was boiled in concentrated nitric acid until only the W matrix particles remained undissolved. This leach solution was also diluted to volume after the addition of Cs, Ce, and Zr carriers. The three solutions thus obtained were analyzed for Cs^{137} content by standard radiochemical procedures.

Analysis of Fission Gases

All fission gases collected from both the puncturing and selective dissolution phases of the work were transferred to small ampoules suitable for radioassay for Kr^{85}. A small volume, diffusion pumped, vacuum manifold system was used for this purpose. Essentially the same technique was used to recover the gases from the stainless steel collection vessels used in puncturing and the Vycor U-traps used during selective dissolution. The U-traps were, however, maintained in a LN_2 bath and evacuated to less than 10^{-3} torr to remove the non-condensable He before fission gas recovery was begun. There was no apparent loss of fission gas (Kr^{85}) activity from the charcoal during this phase of the operation. The release of fission gases from both the collection vessels and the U-traps was driven to virtual completion by heating them to red heat to ensure removal of absorbed krypton from the charcoal. The released gases were collected in 15 mm dia. x 5 cm long Vycor ampoules containing about three-fourths gram of 12/28 mesh activated charcoal and chilled with LN_2. The ampoules, flame-sealed while still immersed in the LN_2 bath, were then analyzed for Kr^{85} content using the 400 channel scintillation spectrometer equipped with a well-type NaI (Tl) detector. Determination of the efficiency of this detector for measuring Kr^{85} in Vycor ampoules containing activated carbon was accomplished by transferring a known quantity of the gas from a standard source into one such ampoule and counting.[5]

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RESULTS AND DISCUSSION

The amounts of fission gas (Kr^{85}) recovered by both puncturing and dissolution are presented in Table 1 along with the burnup values obtained from uranium by double dispersion mass spectrometry and Cs^{137} radiochemical analyses.

The quantities of Kr^{85} recovered by selective dissolution, expressed

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[5] D.A. Seifert, R.L. Stuart, accepted for publication in "Analytical Chemistry".

Table 1

Summary of Burnup and Fission Product Recovery Analyses From LT-710-2 and LT-710-3

Spec. I.D.	Spec. Location, Cm From Low Temp. End of Core	Approx. Irrad. Temp., °C	Analyzed Burnup (fis/cm^3)		Kr^{85} Recovered^c	
			Mass Spec.	Cs^{137} Analysis^b	By Dissolution atoms/cm^3	By Puncture Atoms
LT-710-2:	Fueled Length = 22.86 cm					5.36×10^{16}
1	0.33	530	$2.5\text{-}3.1 \times 10^{18}$	-	-	
2A-1	5.79	650	-	4.64×10^{18}	1.30×10^{16}	
4A-1	16.94	1230	-	2.60×10^{19}	6.27×10^{16}	
5	22.45	1375^a	-	5.15×10^{19}	1.41×10^{16}	
5A	22.56	1370^a	5.41×10^{19}	-	-	
LT-710-3:	Fueled Length = 22.86 cm					1.54×10^{17}
1	0.45	790	-	8.17×10^{18}	2.08×10^{16}	
1A	0.55	795	-	8.29×10^{18}	2.15×10^{16}	
1A-1	0.61	800	7.92×10^{18}	-	-	
3	12.10	1325	-	2.48×10^{19}	5.86×10^{16}	
3A	12.21	1330	-	2.57×10^{19}	(d)	
3A-1	12.32	1335	2.65×10^{19}	-	-	
5A-1	22.25	1540	-	4.77×10^{19}	1.05×10^{17}	
5A	22.35	1540	4.82×10^{19}	-	-	
5	22.45	1540	-	4.62×10^{19}	1.09×10^{17}	

[a] These irradiation temperatures were determined from a heat transfer analysis based on available thermocouple data and comparison of microstructural details of these specimens with those of similar specimens for which accurate temperature data were available.

[b] Burnup values derived from Cs^{137} analyses on the basis of 6.15 atom % fission yield and $T_{1/2}$ = 30 years.

[c] Expression of quantities of 10.76 yr Kr^{85} as atoms was obtained by analysis of the 0.51 MeV photon emission rates and a gamma conversion ratio of 4.31×10^{-3} as determined by T.A. Eastwood, et al. Can. J. Phys., <u>42</u>, 218 (1964).

[d] Recovery incomplete because of He flow stoppage in dissolution apparatus.

as fission densities by dividing by the Kr^{85} fission yield,[6] were plotted along with the mass spectrometer and Cs^{137} burnup values against their respective specimen locations along the lengths of the LT-710-2 and LT-710-3 fuel elements. Burnup and Kr^{85} recovery profiles, presented in Figures 6 and 7, were then determined for each fuel element by rescaling the gamma scans of Figure 2 to fit the analytical data. A better fit to the LT-710-2 data was obtained by using the $Zr\text{-}Nb^{95}$ profile rather than the gross gamma scan, which was dominated by the $Ta^{181}(n,\gamma)Ta^{182}$ activation product present in the clad material. Since no Ta was present in the cladding alloy of LT-710-3, both the $Zr\text{-}Nb^{95}$ profile and the gross gamma scan were found to fit the analytical data equally well.

The total number of fissions and the total amount of Kr^{85} that remained dissolved or trapped in bubbles in each fuel element core were determined by mechanical integration under the respective profiles and multiplication by the fuel element core cross sectional areas. Material balances determined from these data and the amounts of Kr^{85} recovered from puncturing are presented in Table 2. The agreement between the quantities of gas which could be experimentally accounted for and the inventories calculated from analytical burnup and gamma activity profile data are excellent. The 2 to 3% by which the gas recoveries were deficient may easily be accounted for either by uncertainties in the exact shapes of the burnup and Kr^{85}

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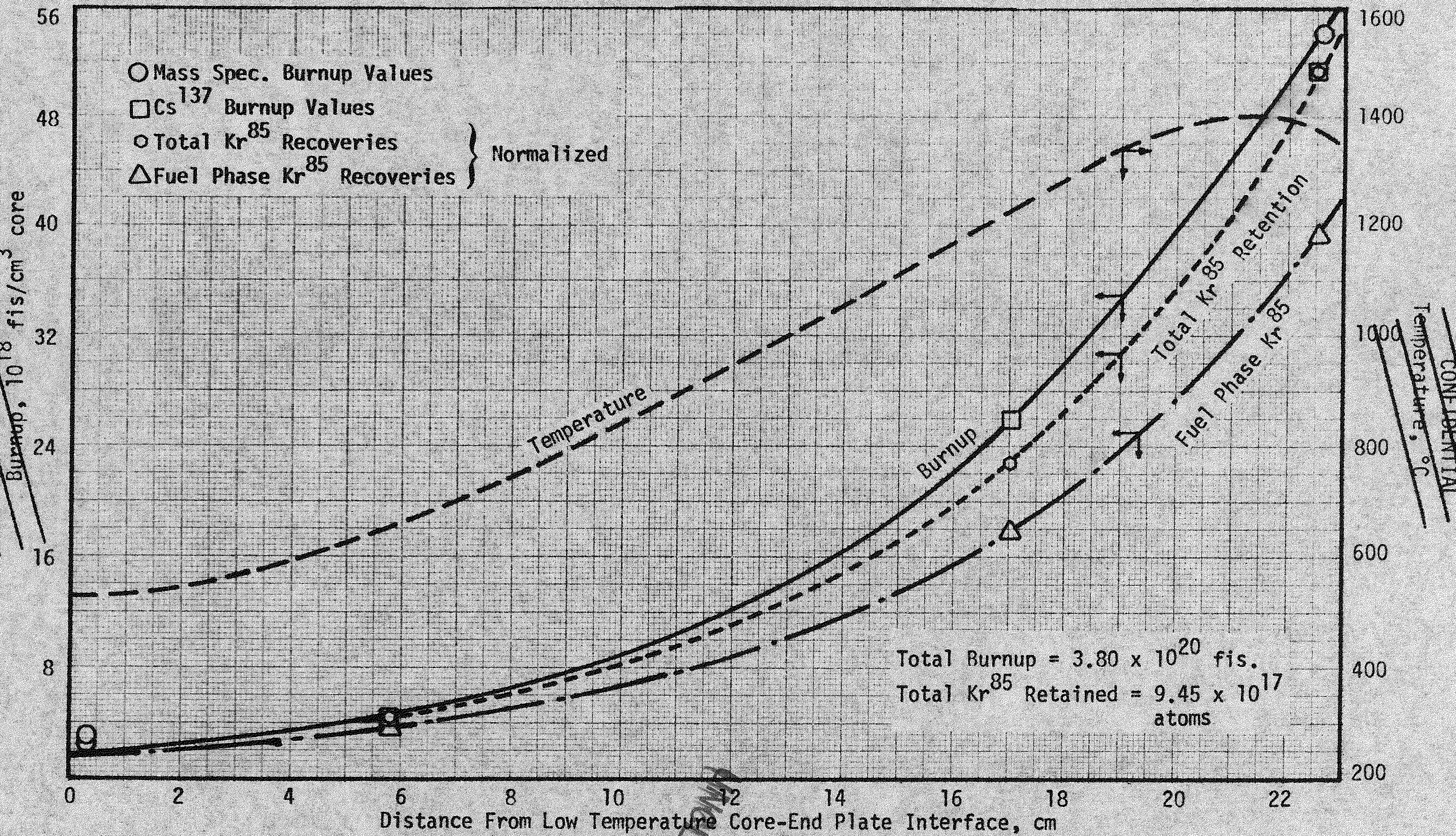
[6] S. Katcoff, W. Rubinson, "Yields of ^{85}Kr in Thermal Neutron Fission of ^{235}U and ^{239}Pu," J. Inorg. Nucl. Chem., 27, 1447 (1964).



Total Burnup = 3.80 x 10^{20} fis.
Total Kr^{85} Retained = 9.45 x 10^{17} atoms

Figure 6 - LT-710-2 burnup, Kr^{85} retention, and temperature profiles.

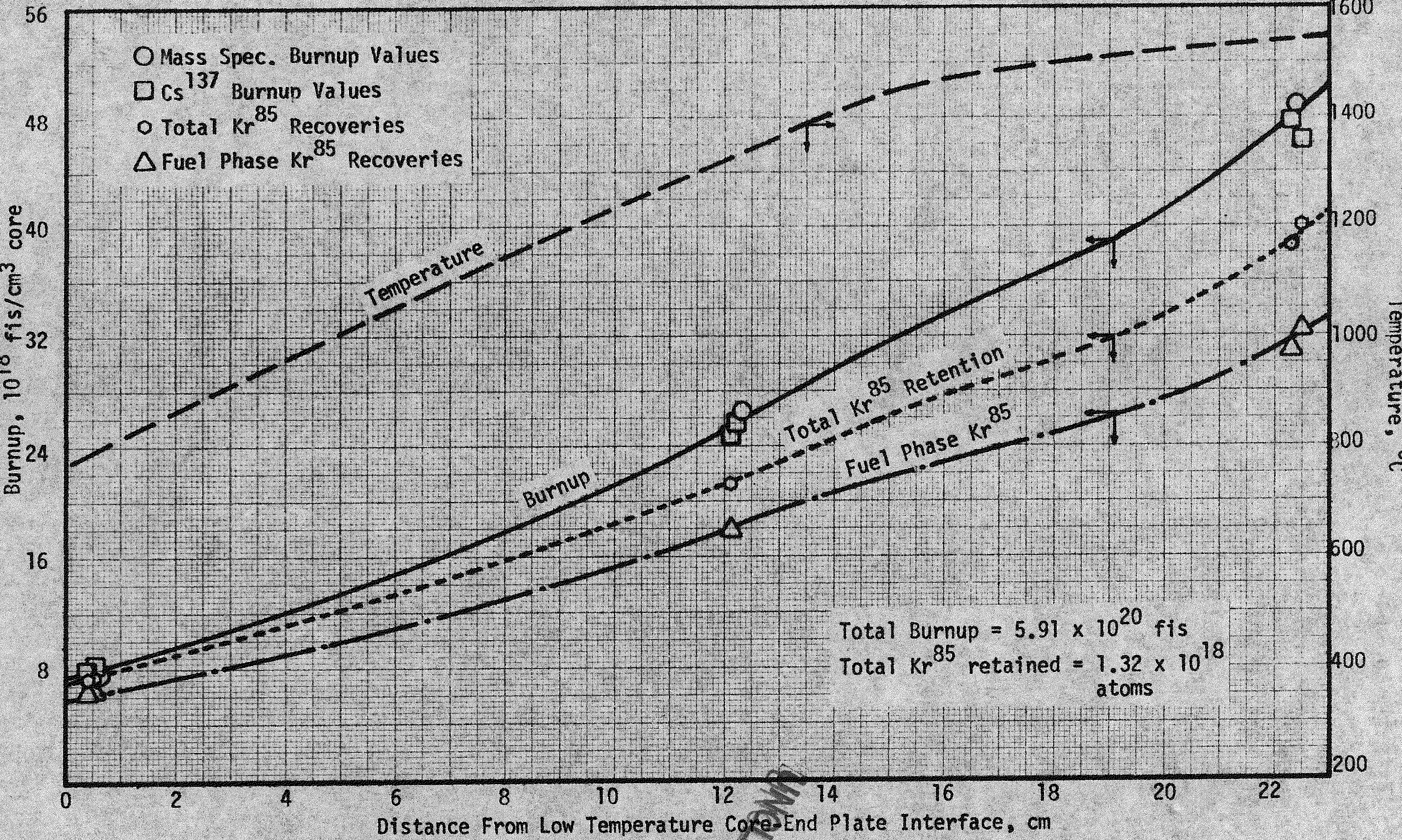


Figure 7 - LT-710-3 burnup, Kr^{85} retention, and temperature profiles

Table 2

Material Balance for Kr^{85}

Specimen	Kr^{85} Inventory at End of Irrad.[a]	Kr^{85} Recovered by Puncturing		Kr^{85} Retained in Specimen[b]		Total Kr^{85} Accounted for	
	(Atoms)	(Atoms)	(% Inv.)	(Atoms)	(% Inv.)	(Atoms)	(% Inv.)
LT-710-2	1.02×10^{18}	5.37×10^{16}	5.3	9.45×10^{17}	92.6	9.98×10^{17}	97.9
LT-710-3	1.58×10^{18}	1.54×10^{17}	9.8	1.37×10^{18}	86.7	1.53×10^{18}	96.5

[a] Values obtained by integration under the respective burnup profiles and adjusting for fission yields.

[b] Values obtained by integration under the respective Kr^{85} recovery profiles.

recovery profiles or by small losses of gas during the intervals between punctures of the fuel elements.

A total of 5.36 x 10^{16} Kr^{85} atoms (Table 1) was recovered by puncturing the LT-710-2 fuel element. Of these, 21.9% was from the first puncture (low temperature end), 78.1% from the second puncture (high temperature end), and only 2.5 x 10^{-2} % from the third puncture (center). The small fraction of Kr^{85} obtained from the third puncture indicated that the mobile gas was gone from the LT-710-2 specimen, but it was not known whether the recovery was quantitative until the materials balance was completed. The results, Table 2, show that 5.3% of the inventory was mobile and recovery of the gas was quantitative within experimental error; apparently gas transport is very slow from regions far from the puncture hole and to some extent, air which enters when the holes in the fuel elements are re-exposed to atmospheric pressure effectively prevents further loss of fission gas except by some limited diffusion.

The results of the extensive gas sampling program previously described for LT-710-3 are presented in Figure 8 as Kr^{85} recovered versus sampling time. Of the total of 1.54 x 10^{17} Kr^{85} atoms recovered (Table 1), the first four data points, accounting for some 99.5% of the total, represent gas collected from the first puncture hole (low temperature end) and suggest that gas could communicate through the channels between the fuel and matrix over the entire length of the specimen if sufficient sampling time were allowed, as shown in Figure 8. The remaining three pairs

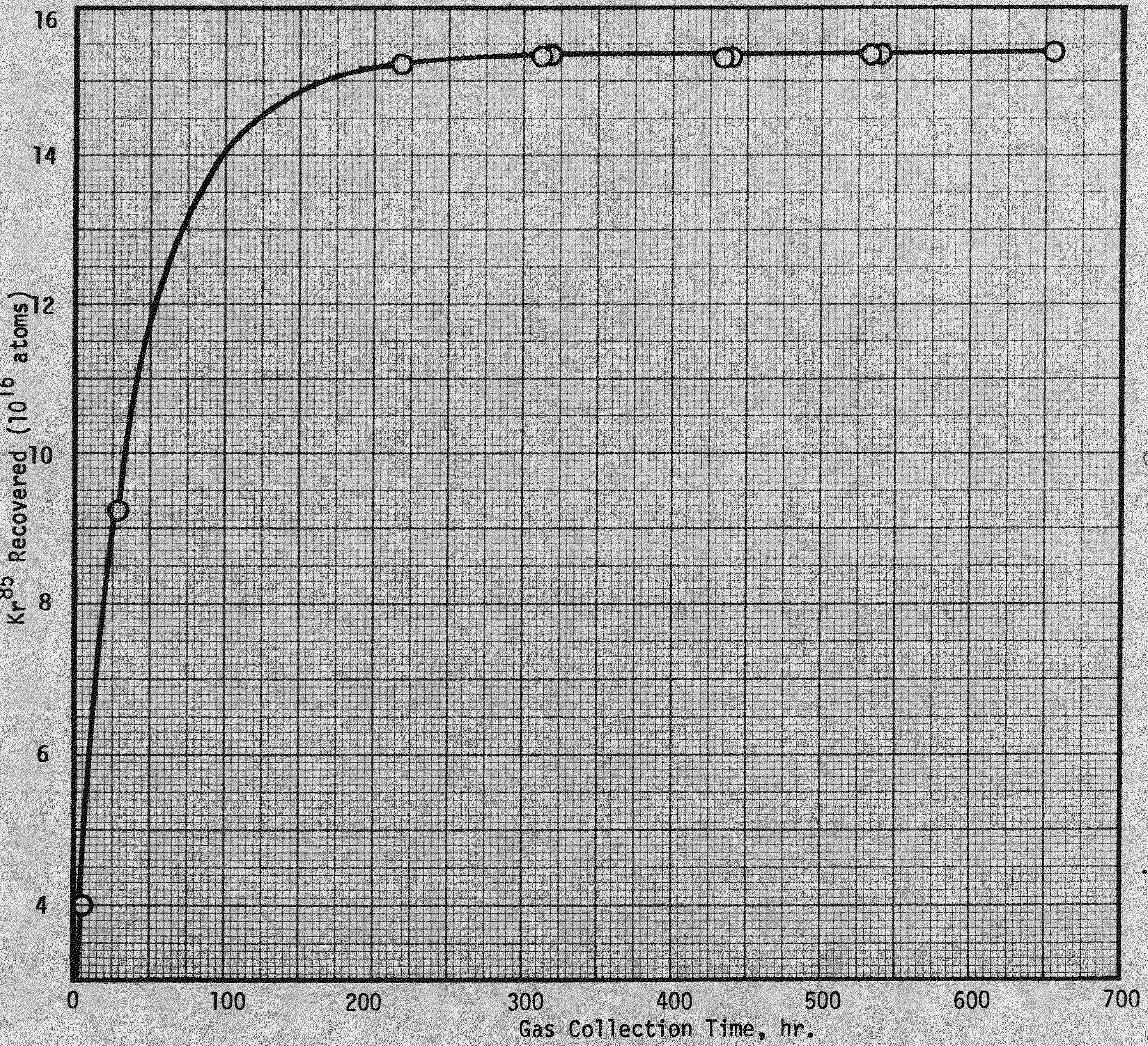


Figure 8 - Kr^{85} recovery from LT-710-3 punctures.

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of data points, each spanning an interval of 100 to 125 hours, represent gas recovered from the second (higher temperature end), third, and fourth (intermediate positions) puncture holes, respectively, and account for the remainder of the mobile gas. Based on the materials balance, recovery of the mobile gas was again complete within experimental error and amounted to 9.8% of the Kr^{85} inventory in the fuel element.

The first gas sample taken from LT-710-3, representing some 25.8% of the total mobile gas recovery, can be directly compared with the first puncture of LT-710-2 where a similar fraction (21.9%) of the mobile gas inventory was recovered during a comparable sampling interval. This indicates a similarity in the extent and size of flow paths from the low to high temperature ends of the two specimens. The amounts of mobile gas found in the LT-710-2 and -3 specimens were less than obtained previously for shorter specimens tested under more nearly isothermal conditions, probably because a substantial portion of each 710 specimen was at a lower temperature than the nominal test temperataure of the shorter specimens. The shorter specimens were irradiated at a maximum temperature of 1590°C to burnups of 4.4×10^{19} to 7.3×10^{19} fissions/cc and 10.8 to 16.2% of the gas inventory was found to be mobile.[7]

Examination of the distribution of Cs^{137} and retained Kr^{85} between the fuel and matrix phases of the cermet cores, Table 3, indicates that the fraction of both isotopes retained by the W matrix remained

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[7] "Seventh Annual Report - AEC Fuels and Materials Development Program," GE-NMPO, GEMP-1003, February 20, 1968, p. 110, Table 3.1. The burnups noted in the table, namely 7.7×10^{19} and 4.0×10^{19} fissions/cc for the ORRF tests are incorrect. They are correct as stated above.

Table 3

Distribution of Kr and Cs Between Fuel and Matrix Phases of Cermet Cores

Specimen No.	Approx. Irradiation Temp., °C	Inventory From Burnup Profile (atoms/cm^3)		Fuel Phase Retention % Inv.		Matrix Retention, % Inv.		Total Retention, % Inv.	
		Kr^{85}	Cs^{137}	Kr^{85}	Cs^{137}	Kr^{85}	Cs^{137}	Kr^{85}	Cs^{137}
LT-710-2									
2A-1	655	1.31×10^{16}	3.15×10^{17}	80.1	76.5	18.3	19.4	98.4	95.9
4A-1	1230	6.96×10^{16}	1.69×10^{18}	70.7	84.7	19.4	15.3	90.1	100.0
5	1375	1.44×10^{17}	3.50×10^{18}	75.0	78.1	23.1	17.6	98.1	95.7
LT-710-3									
1	790	2.17×10^{16}	5.33×10^{17}	81.1	78.4	14.7	21.2	95.8	99.6
1A	795	2.20×10^{16}	5.40×10^{17}	80.5	78.9	17.4	20.9	97.2	99.8
3	1325	6.83×10^{16}	1.66×10^{18}	71.9	78.3	14.0	18.6	85.9	97.0
3A	1330	-	1.68×10^{18}	-	80.8	-	18.6	-	99.4
5A-1	1542	1.28×10^{17}	3.10×10^{18}	66.3	80.9	16.1	19.1	82.4	100.0
5	1543	1.30×10^{17}	3.14×10^{18}	64.8	76.6	15.9	19.0	80.7	95.6

approximately constant over the entire specimen lengths. Recoil calculations based on spherical fuel particles having an average diameter of 50-60 μ, as shown in the microstructure of Figure 9, would place 15% to 17% of these fission products in the W matrix.[8] The recovery of 15-20% of the fission products (Cs^{137} and Kr^{85} for the LT-710-2 and Cs^{137} for LT-710-3) from the matrix, considering the presence of an appreciable number of smaller fuel particles, as shown in the figure, is in good agreement with the calculated recoil and indicates that fission gases recoiled into the matrix are essentially immobile at temperatures up to ∿ 1550°C. The difference between the amounts of Kr^{85} and Cs^{137} retained in the matrix of the LT-710-3 specimen is not understood.

The fraction of Cs^{137} retained in the fuel phases of both specimens remained constant over their lengths and did not vary with temperature. The lack of cesium mobility in both the fuel phase and tungsten matrix for these test times and temperature profiles indicates that, in this burnup range, cesium probably did not contribute to the internal gas pressure of the fuel elements during operation at temperature even though operating temperatures were above the boiling point of cesium (690°C).

The amount of Kr^{85} retained by the fuel phase decreased with increasing

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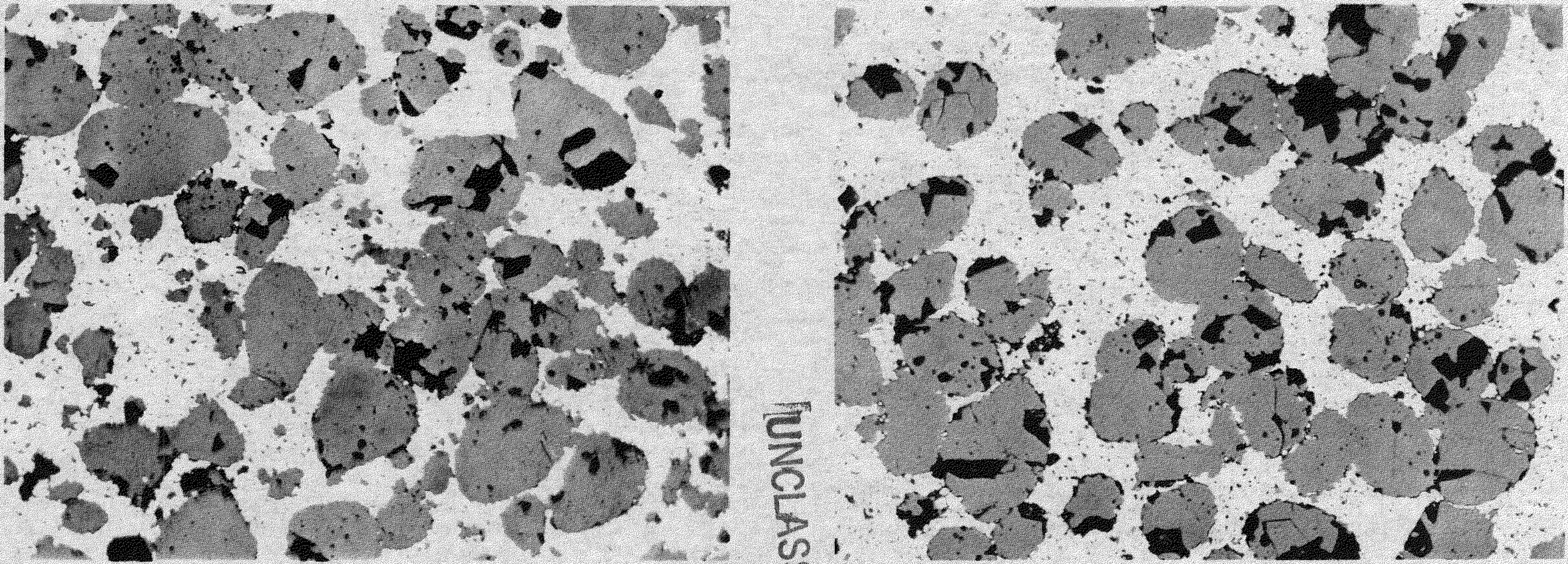
[8] A fuel particle size of 60 μ is obtained by taking the average of the sieve openings used during preparation of the fuel for this test.



LT-710-2 Core structure 250 X Neg. No. R-222

LT-710-3 Core structure 250 X Neg. No. R-2212

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Figure 9 - Typical core structure of cermet fuel elements irradiated during in-pile experiments LT-710-2 and LT-710-3.

irradiation temperature. This and the previous conclusion that the fission gases which recoiled into the W matrix remained immobile at the temperatures in question indicate that all of the mobile fission gas within the fueled cores was released from the fuel phase. Figure 10 shows the temperature dependence of Kr^{85} release from the fuel phase on an Arrhenius-type plot calculated on the basis of the gas which was retained in the fuel phase below 650°-700°C for each specimen. The agreement between LT-710-2 and LT-710-3 is excellent up to about 1400°C, the estimated maximum for LT-710-2 test. The apparent abnormal decrease in release from the LT-710-2 specimen fuel phase in the region of the temperature inversion shown in Figure 6 is not understood. The lower portion of Figure 10, showing the dependence at low temperatures, indicates that fission gas retention by the fuel phase is complete below 650°-700°C. This is in good agreement with other investigators[9,10] whose studies in bulk UO_2 suggest that fission gas retention is complete up to temperatures from 600° to 800°C.

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By assuming a constant rate over the entire test time for the temperature-dependent release of Kr^{85} from the fuel phase of LT-710-2 and LT-710-3, an activation energy of 6.1 kcal/mole was determined from data in Figure 10. This is in good agreement with the 7.8 kcal/mole calculated

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[9] R.M. Carroll, R.B. Perez, O. Sisman, "Release of Fission Gas During Fissioning of UO_2," Journal Am. Ceram. Soc. 48, February, 1965, pp. 55-59.

[10] R.S. Barnes, R.G. Bellamy, B.T. Bradbury, J.B. Sayers, A.D. Whapham, "The Behavior of Fission Product Gases in Uranium Dioxide," Trans. Am. Nucl. Soc. 7, June 1964, p. 89.

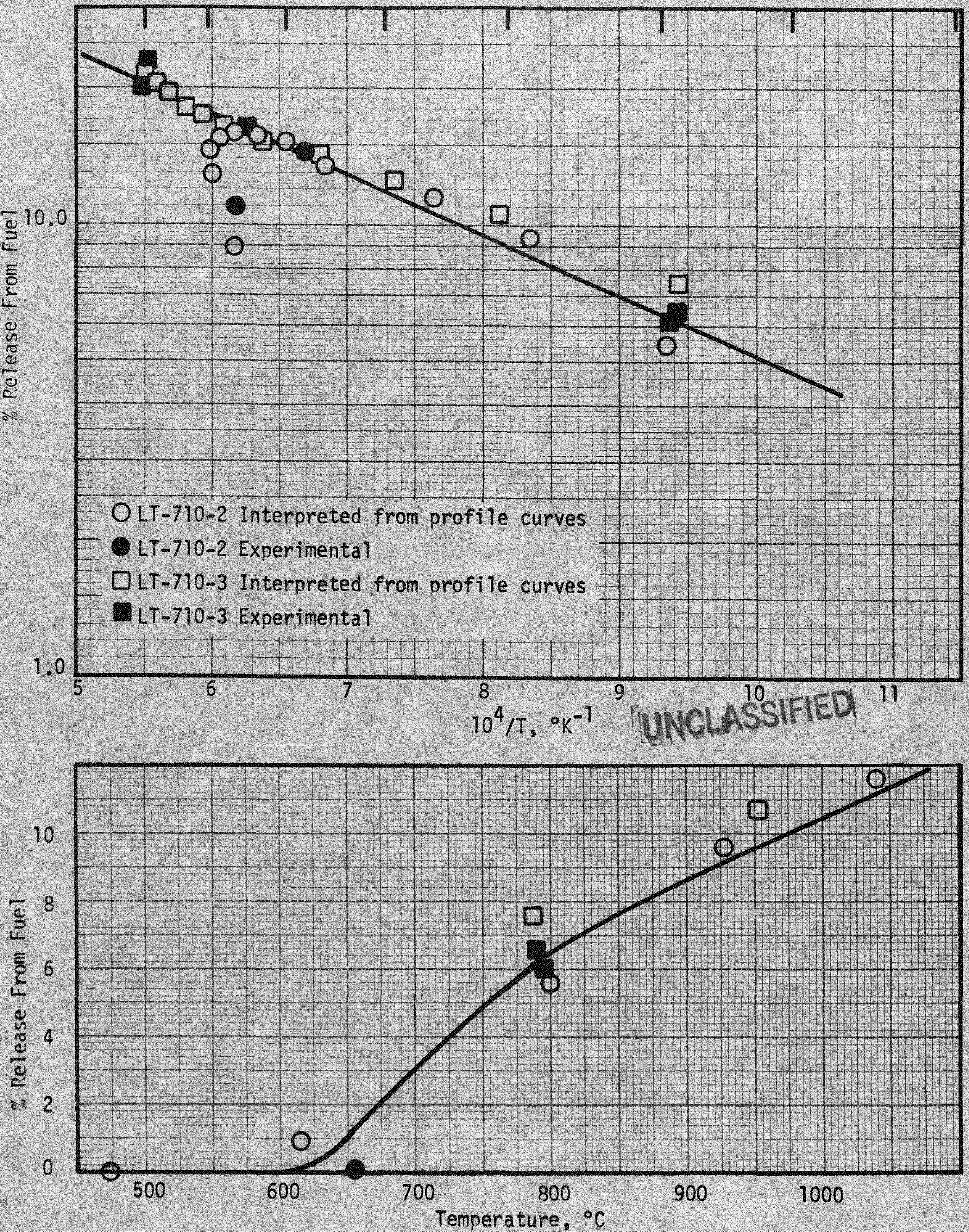

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Figure 10 - Temperature dependence of fission gas release from fuel phase

from data collected during in-pile operation of a leaking specimen at temperatures between 400° and 1350° to 1400°C,[11] from which gas release through open channels from W-UO_2 cermet cores was thought to be occurring, but much lower than either the 51-57 kcal/mole suggested by Carroll, et al[9], for diffusion from UO_2 or the 35 kcal/mole proposed by Greenwood and Speight[12] for the mechanism controlling bubble migration in UO_2.

It is probable, however, that a constant gas release rate did not exist throughout the test so that the calculated activation energy has no significance. Large quantities of gas may have been released from the fuel phase as a result of thermal cycle induced cracking of fuel particles in the high temperature portions of the specimens (some cracks are apparent in the photomicrographs of Figure 9 and the pullout during polishing would indicate even more cracks). Resolution of some of the fission gases, as suggested by Lewis[13], may also have occurred in regions of high localized pressure (∿ 10's of atmospheres).

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Significance of Results for Operating Fuel Elements

The probability of localized deformations occurring in high density (∿ 97%) cermet core fuel elements is greatest in elements possessing burnup and temperature gradients whose maxima occur in the same region,

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[11] Loc cit, GEMP-1003, p. 99.

[12] G.W. Greenwood, M.V. Speight, "An Analysis of the Diffusion of Fission Gas Bubbles and Its Effect on the Behavior of Reactor Fuels," J. Nucl. Mat. 10, 1963, pp. 140-144.

[13] W.B. Lewis, "Behavior of Fission Gases in UO_2 Fuel," Atomic Energy of Canada, Ltd., AECL-1402, November 1961.

since the greatest gas pressure buildup occurs in the region of highest temperature. It is beneficial for fission gases to be highly mobile in such fuel elements so that pressure can equalize throughout the element. Even though these data indicate that gas could flow to the cold end of the 710-2 and 710-3 specimens through the channels between the fuel and W phases, 85% or more (15% in the W and 70-85% in the fuel) is retained in the core, depending on the temperature, for the burnups obtained in this study. If this large fraction of retained gas does not change with additional burnup, it is likely that localized deformation will occur in the high temperature, high burnup regions of high density cermet core fuel elements because of localized fission gas retention. Further, a simple vent hole may not effect release of enough fission gas to significantly improve the temperature-burnup capability of this fuel system beyond that already demonstrated, i.e., $\sim 7.3 \times 10^{19}$ fissions/cc at 1600°C for W-Re-Mo clad W-UO$_2$ cermets.[14] Microstructural changes in the as-fabricated core appear to be necessary to allow increased gas redistribution rates within the fuel element at elevated temperatures and to allow a much larger fraction of total gas inventory to be mobile in the core.

CONCLUSIONS

1. Depending on the temperature of the fueled specimens, between 85% and 95% of the fission gases generated in 60 volume % fuel phase

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[14] Ibid, p. 107, Figure 3.1.

cermet fuel systems at temperatures up to 1550°C remain "dissolved" in the lattice and/or trapped in isolated pores, 15 to 20% being recoiled into the tungsten matrix.

2. The remaining 5 to 15% of the generated gases are released by the fuel phase, and at room temperature, are free to migrate through channels between the fuel and matrix phases. These flow channels, while completely closed at elevated temperatures, are extensive enough to permit complete communication throughout the specimen lengths at room temperature.

3. The fuel phase does not begin to release its fission gases until the irradiation temperature exceeds ~ 700°C. Above this temperature fission gas release increases with increasing temperature in a manner which, up to about 1550°C, seems to be controlled by a low-activation-energy (6-8 kcal/mole) process.
4. Although most or all of each fueled specimen was irradiated at temperatures above 690°C, the boiling point of cesium, the retention data for Cs^{137} indicate no mobility during the test time as there was for Kr^{85}; thus Cs^{137} can be used to supplement mass spectrometric burnup data for long specimens irradiated under these test conditions.
5. Based on information obtained from the combined techniques described and the results of other studies cited, it appears that fission gas retention by high density cermet fuel elements will cause localized swelling, particularly in regions of simultaneous high temperature and burnup. Basic microstructural changes in the cores of these

fuel systems before irradiation may be necessary either to permit redistribution of mobile fission gases at elevated temperatures or to accommodate fuel phase swelling caused by retained fission products.

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