U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

PRESENTATION TO THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT: SPENT FUEL CLADDING DEGRADATION

PRESENTER: HARRY D. SMITH

PRESENTER'S TITLE AND ORGANIZATION: SENIOR RESEARCH SCIENTIST
BATTELLE, PACIFIC NORTHWEST LABORATORIES
RICHMOND, WASHINGTON

PRESENTER'S TELEPHONE NUMBER: (509) 376-3588

AUGUST 28-29, 1990

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TOPICS COVERED

- CARBON-14 RELEASE FROM ZIRCALOY CLADDING
- C-RING FAILURE TESTING - SCOPING STUDIES
- PRESSURIZED TUBE TESTING
  - OXIDE FILM DEGRADATION
  - HYDRIDE STRESS-DEPENDENT ORIENTATION
  - HIGH TEMPERATURE AND PRESSURE TESTING
- ZIRCALOY-FLUORIDE CORROSION TESTING

WHY ZIRCALOY CLADDING DEGRADATION STUDIES

- CARBON-14 RELEASE DATA ARE NEEDED SO THAT A QUANTITATIVE EVALUATION OF ITS BEHAVIOR UNDER PRE-EMPLACEMENT AND POST-EMPLACEMENT (REPOSITORY) CONDITIONS CAN BE MADE

- THE CLADDING CAN BE CONSIDERED PART OF THE BARRIER SYSTEM TO RADIONUCLIDE RELEASE IF THE CLADDING DEGRADATION CHARACTERISTICS CAN BE PREDICTED. IF NOT, AN ENGINEERED BARRIER (OR BARRIERS) WILL BE NECESSARY AS AN ALTERNATIVE TO THE CLADDING TO MITIGATE THE EFFECTS OF THE PULSE RELEASE OF SOLUBLE OR GASEOUS RADIONUCLIDES THAT WOULD OCCUR AT THE END OF THE CONTAINMENT PERIOD

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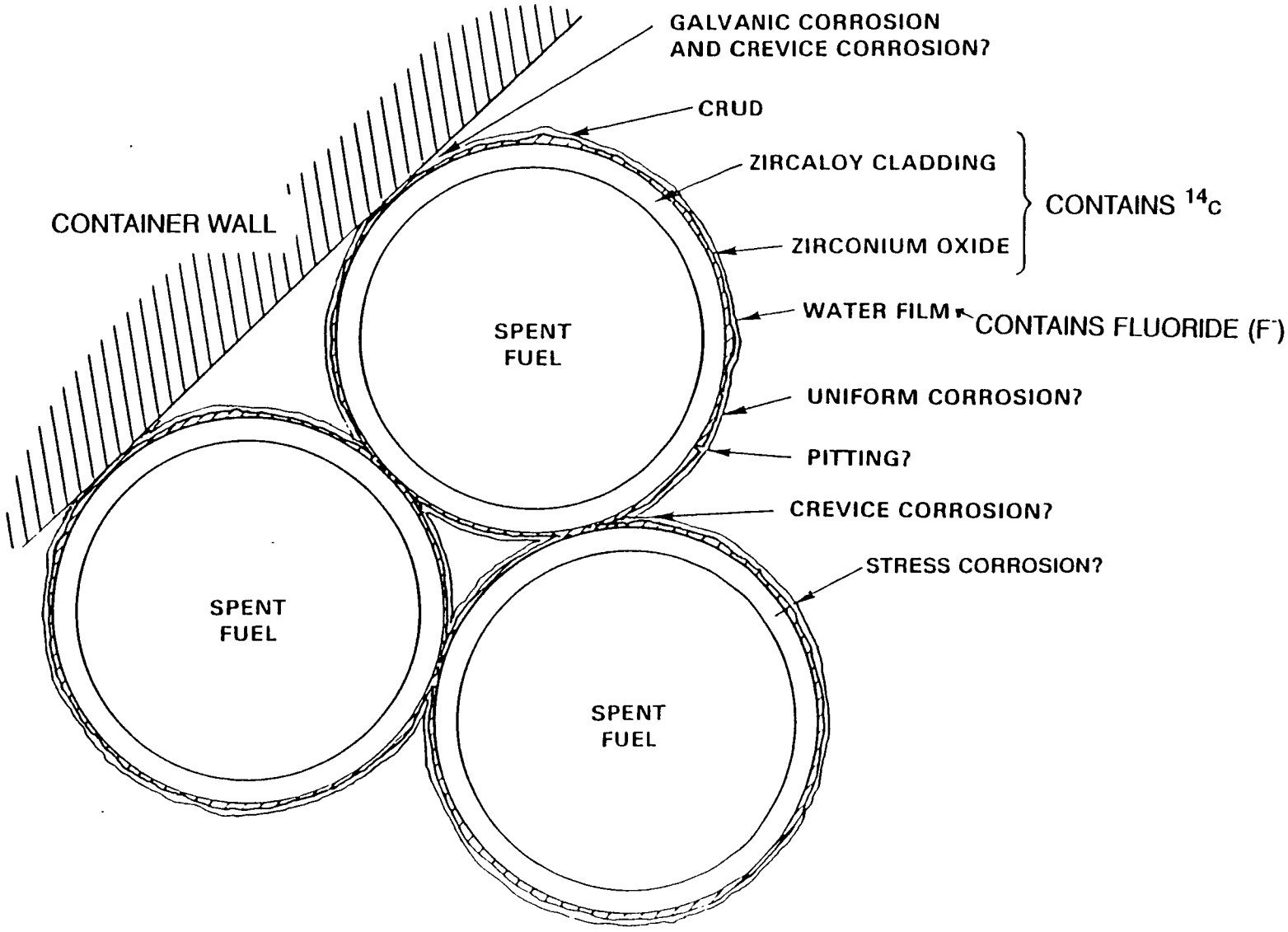
ZIRCALOY CLADDING IN THE REPOSITORY SETTING



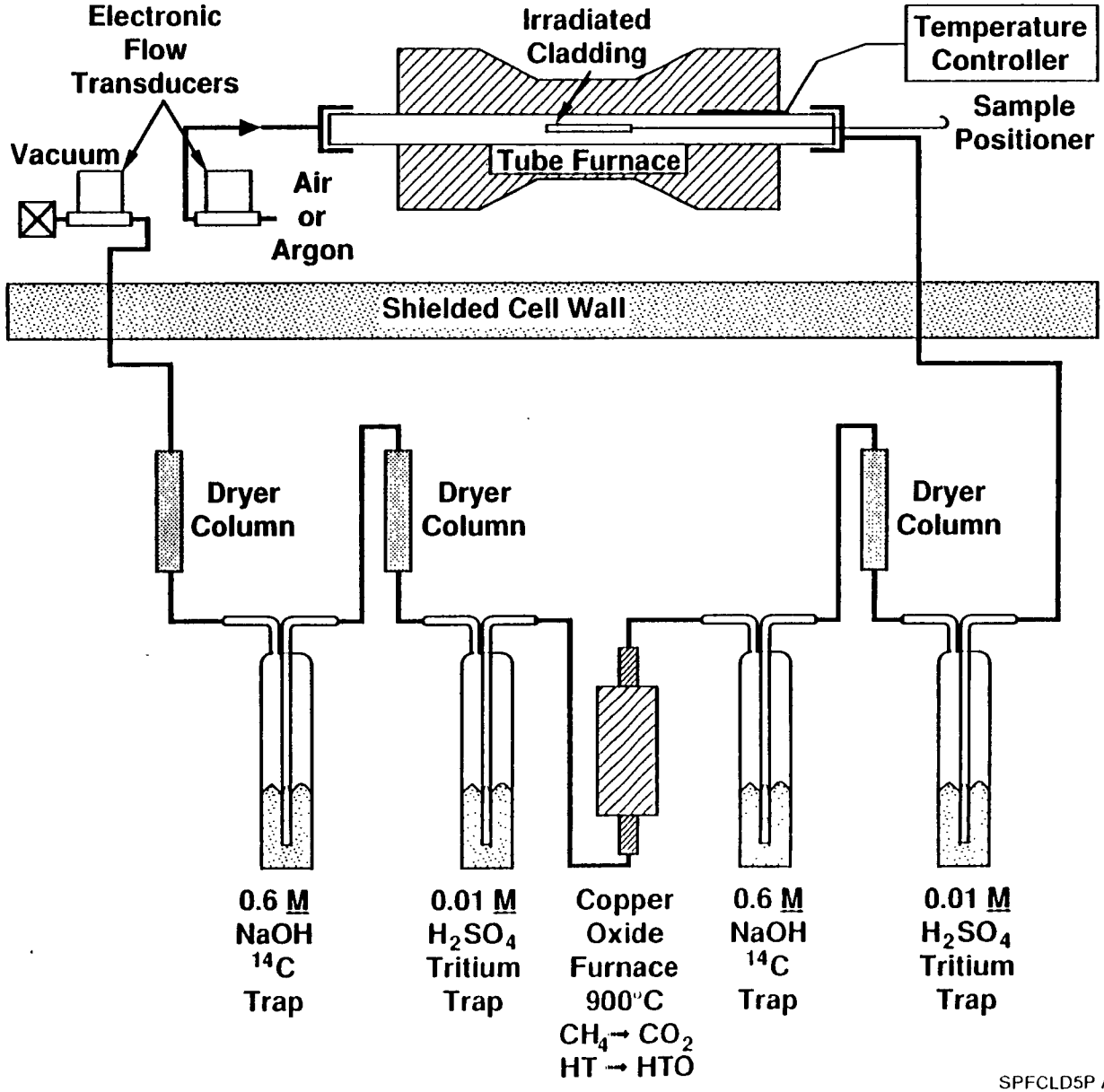
EXPERIMENTAL MATERIAL: ZIRCALOY-4

- PWR REACTORS
  (HB ROBINSON, TURKEY POINT)
- 27 TO 30 MWd/kgHM
- THICK OXIDE - 12 TO 20 μm
- THIN OXIDE - 3 TO 6 μm

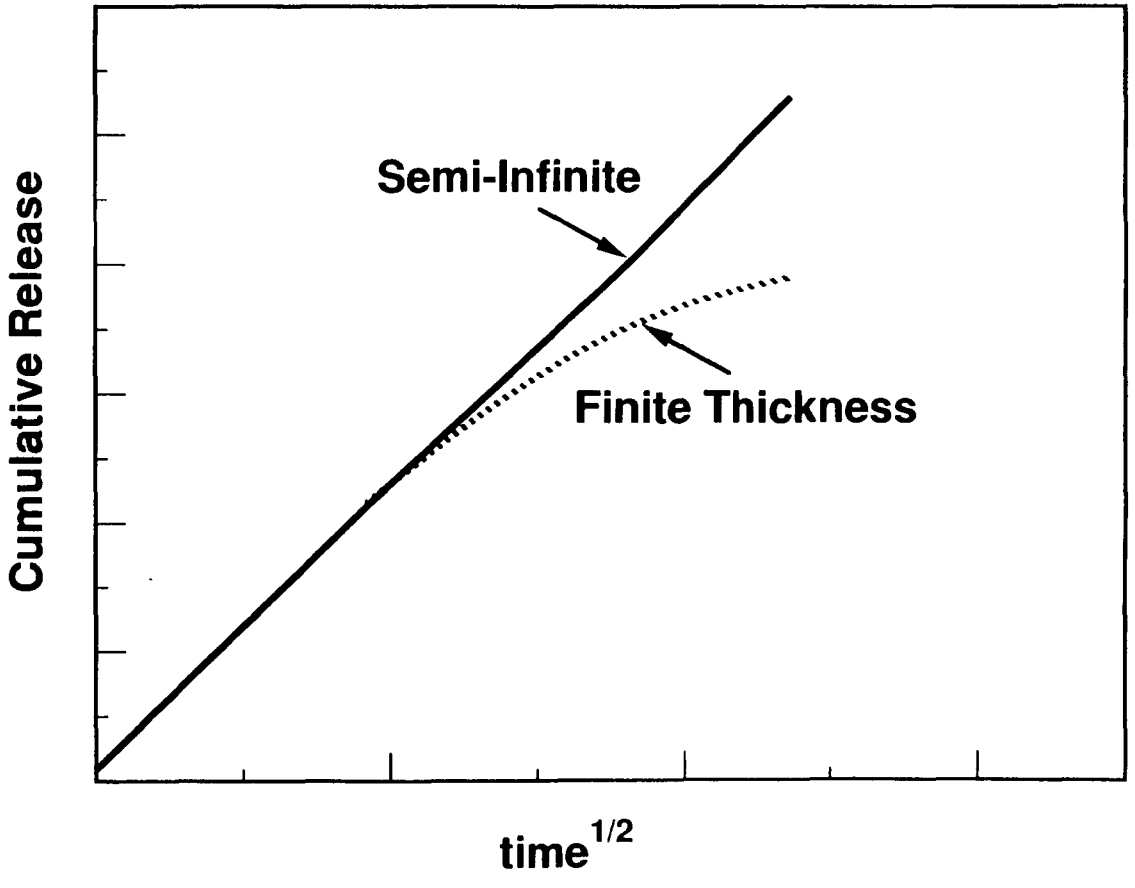
THERMAL RELEASE OF ^{14}C FROM SPENT FUEL CLADDING

- DEVELOP AND DEMONSTRATE THE TECHNIQUES TO QUANTITATIVELY MEASURE RELEASE
- APPLY THE TECHNIQUES TO SPENT FUEL CLADDING AND REACTOR HARDWARE

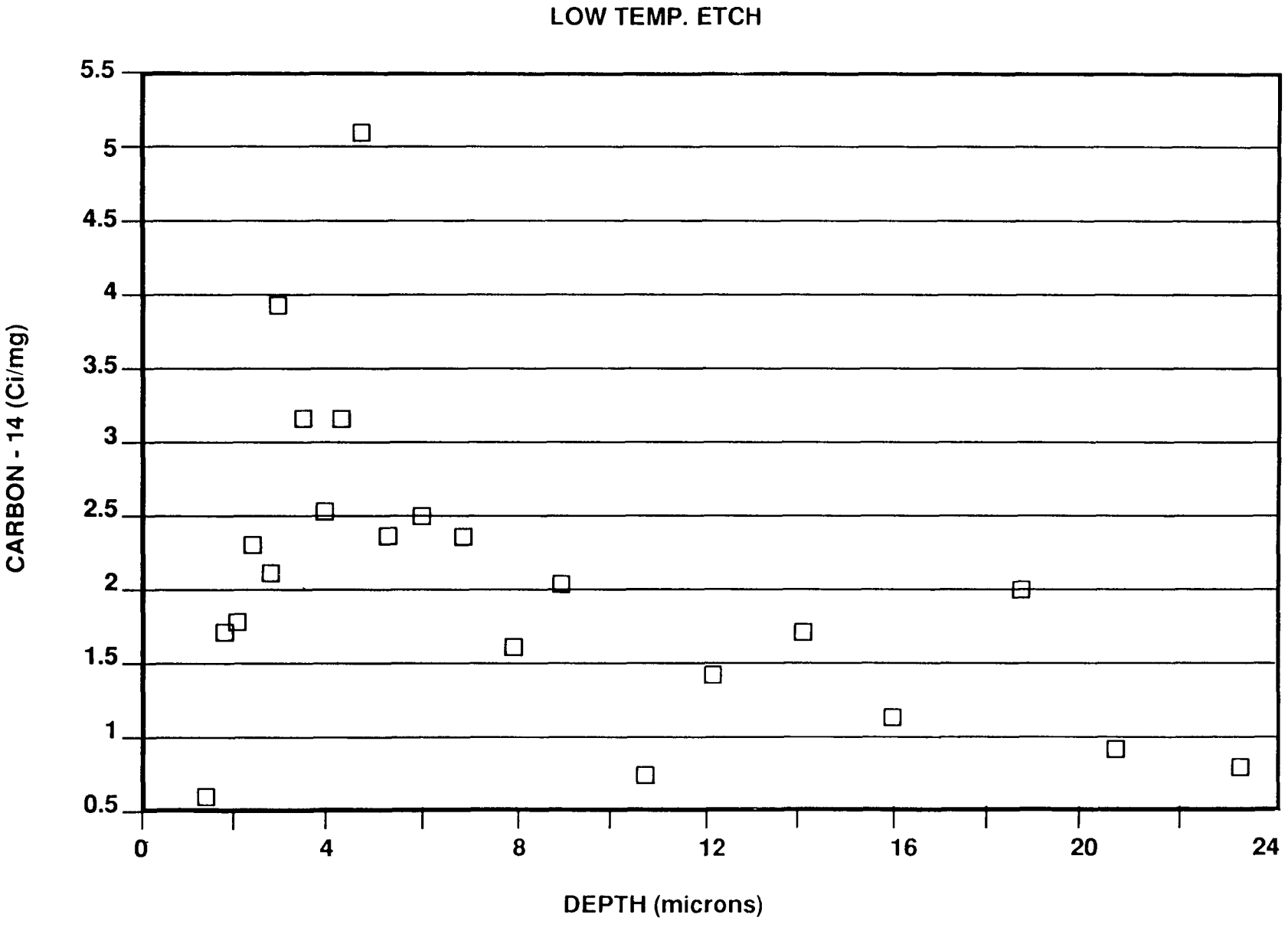
APPARATUS FOR THERMAL RELEASE STUDIES



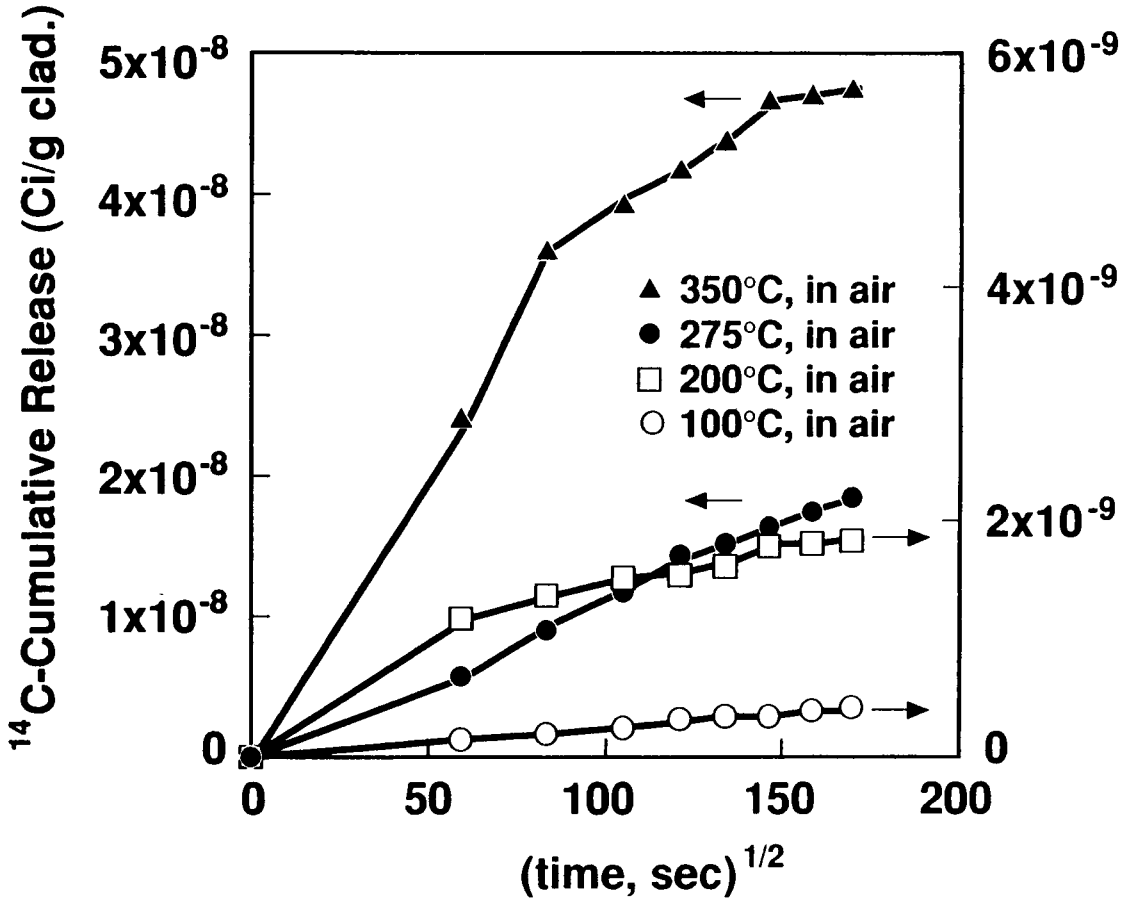
RELEASE MODEL COMPARISON FOR TWO ^{14}C SOURCE GEOMETRIES



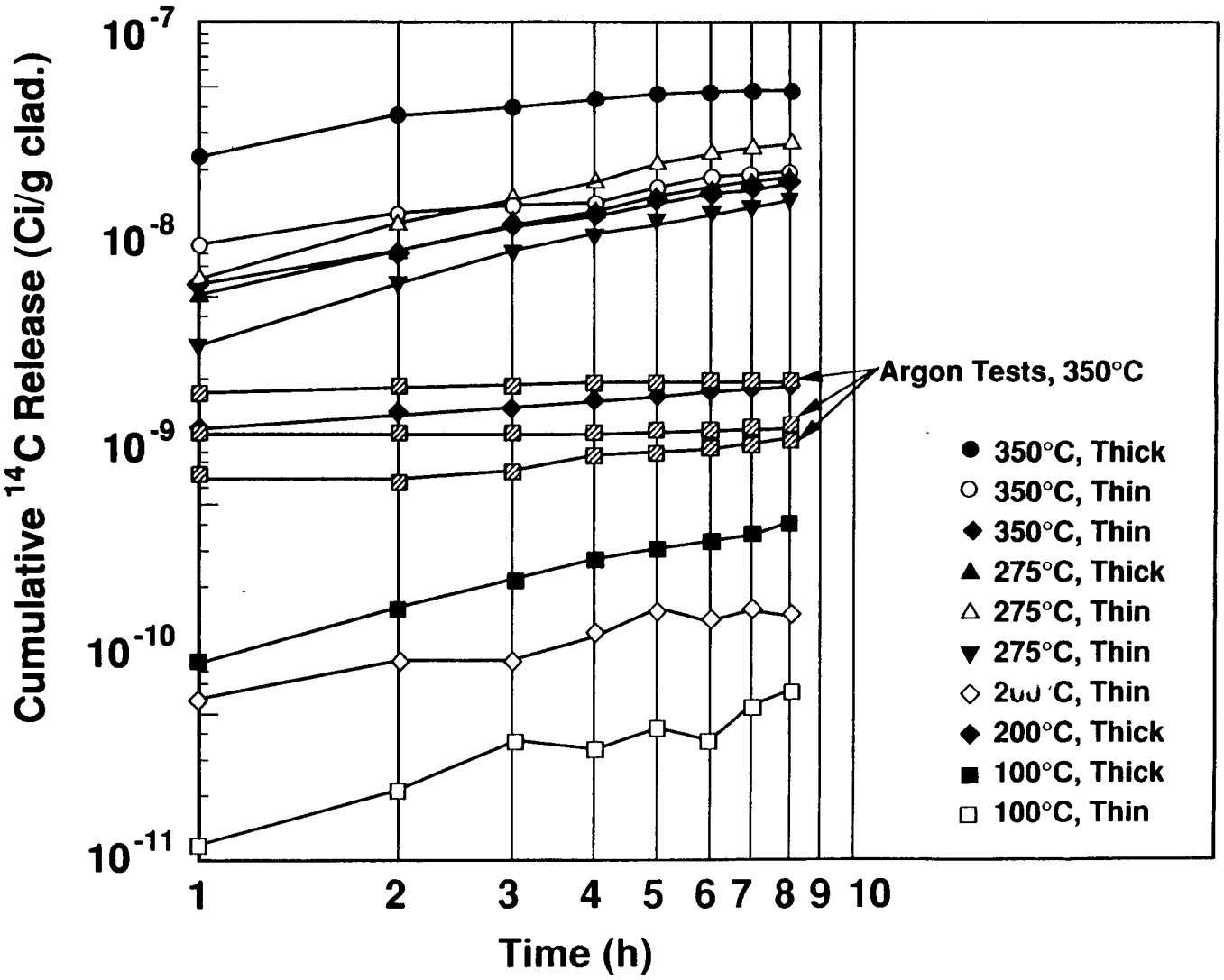
ZIRCALOY CLADDING PROFILE



CONSTANT TEMPERATURE TESTS, THICK OXIDE



OBSERVED ^{14}C RELEASE FROM ZIRCALOY-4 SPENT FUEL CLADDING



SUMMARY/CONCLUSIONS

- **A SYSTEM HAS BEEN DEVELOPED THAT ALLOWS THE THERMAL RELEASE OF ^{14}C TO BE DETERMINED AS A FUNCTION OF**
  - TEMPERATURE
  - ATMOSPHERE, INCLUDING MOISTURE
  - OTHER ENVIRONMENTAL FACTORS (RADIATION)

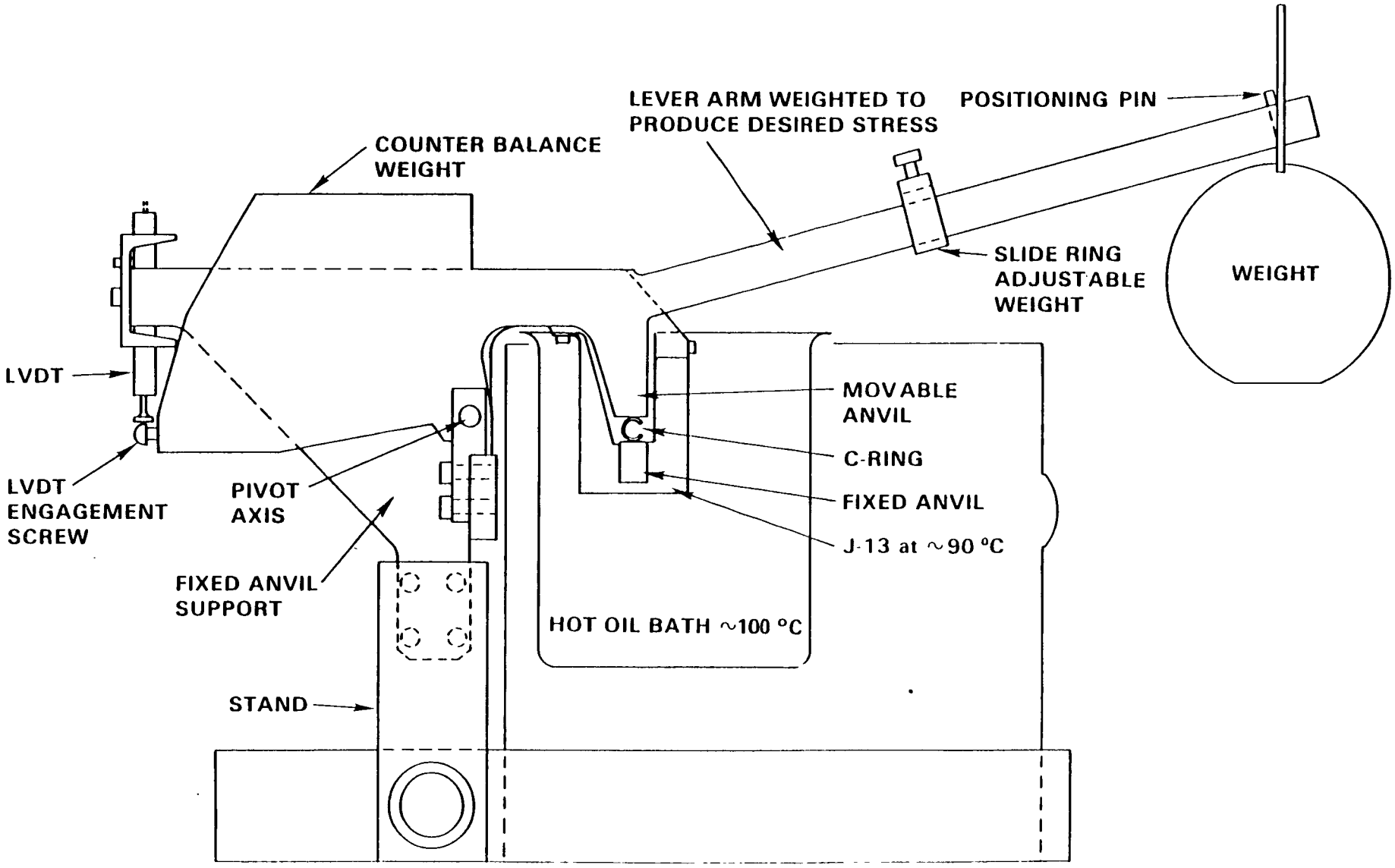
- **INITIAL DATA ON ^{14}C RELEASE FROM SPENT FUEL CLADDING HAVE INDICATED**
  - UP TO 10% OF THE ^{14}C CLADDING INVENTORY IS RELEASED IN 8 HOURS AT 350°C IN AIR
  - ONE ORDER OF MAGNITUDE LOWER RELEASES WERE OBSERVED UNDER AN ARGON ATMOSPHERE
  - ^{14}C RELEASE APPEARS TO FOLLOW A FINITE PLANE SOURCE MODEL

SPENT FUEL CLADDING DEGRADATION: CRACKING STUDIES OF ZIRCALOY-4

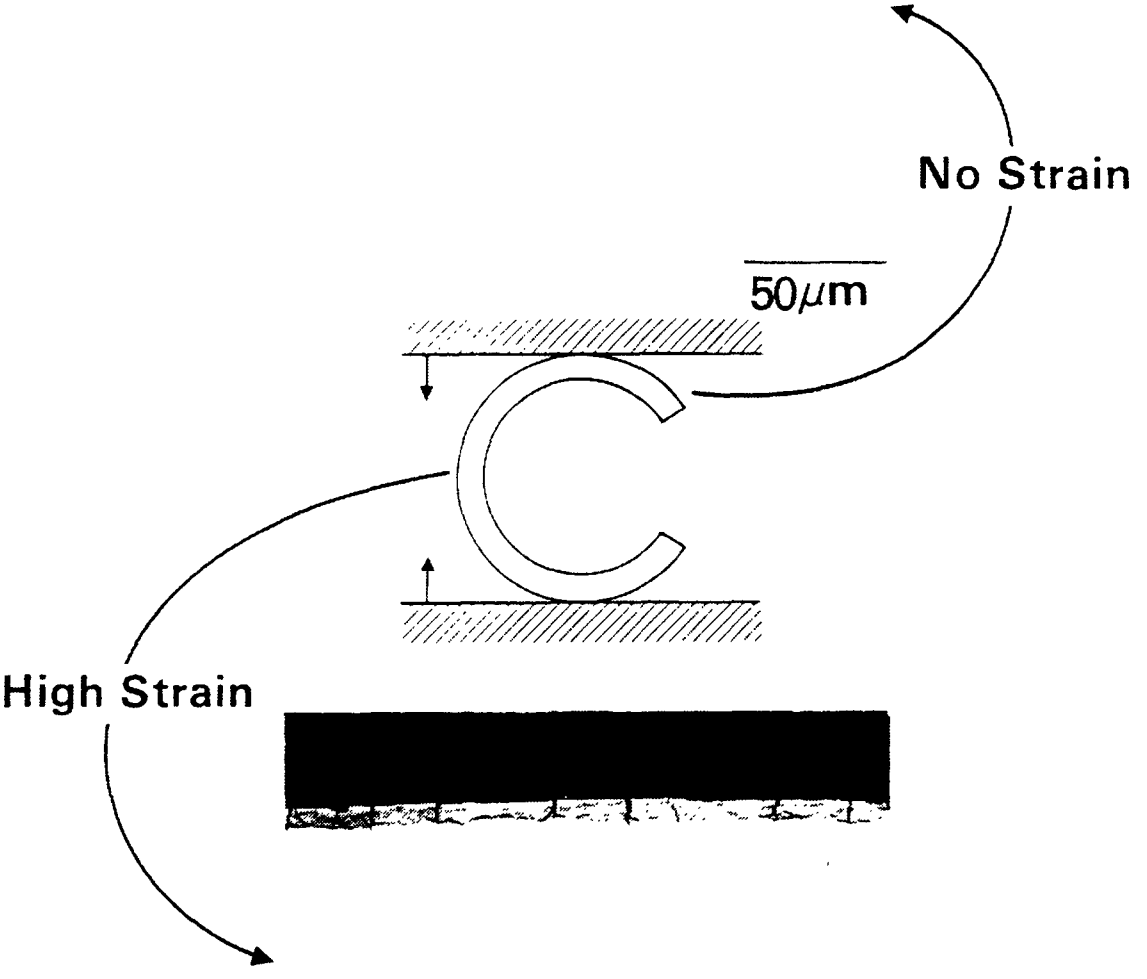
- C-RING EXPERIMENTS
  - DETERMINE IF CRACKING OCCURS
  - DETERMINE RELATIONSHIP BETWEEN "TIME TO FAILURE" AND "STRESS"

- PRESSURIZED TUBE TESTING
  - DETERMINE STRAIN REQUIRED TO CRACK PROTECTIVE OXIDE FILM
  - DETERMINE CONDITIONS REQUIRED FOR HYDRIDE REORIENTATION (CIRCUMFERENTIAL TO RADIAL)

"C-RING" CRACKING EXPERIMENT APPARATUS

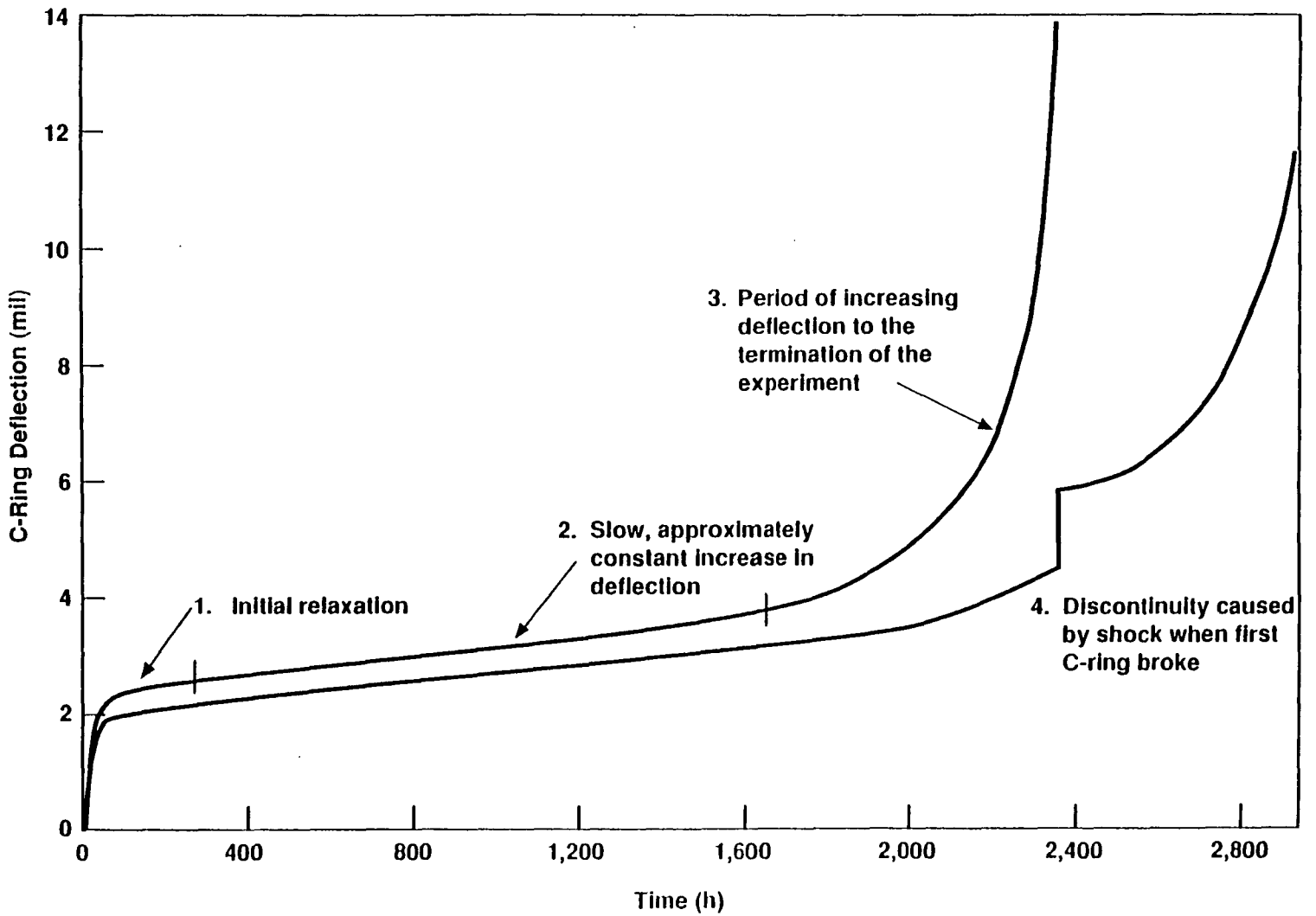


OXIDE FILM CRACKING IN STRAINED ZIRCALOY-4 C-RING

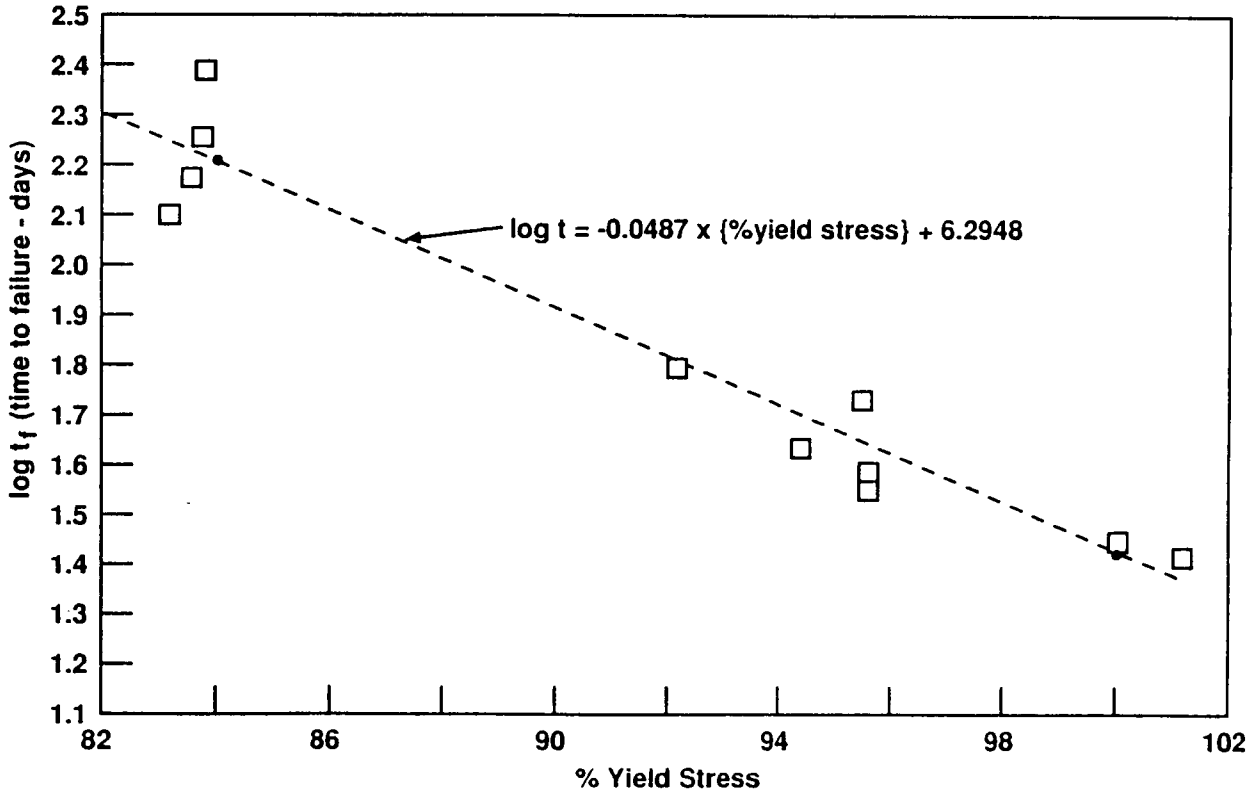


50μm

TYPICAL C-RING DEFLECTION PROFILES



OBSERVED RELATIONSHIP: log t_f (TIME TO FAILURE) vs. STRESS (% OF YIELD STRESS)



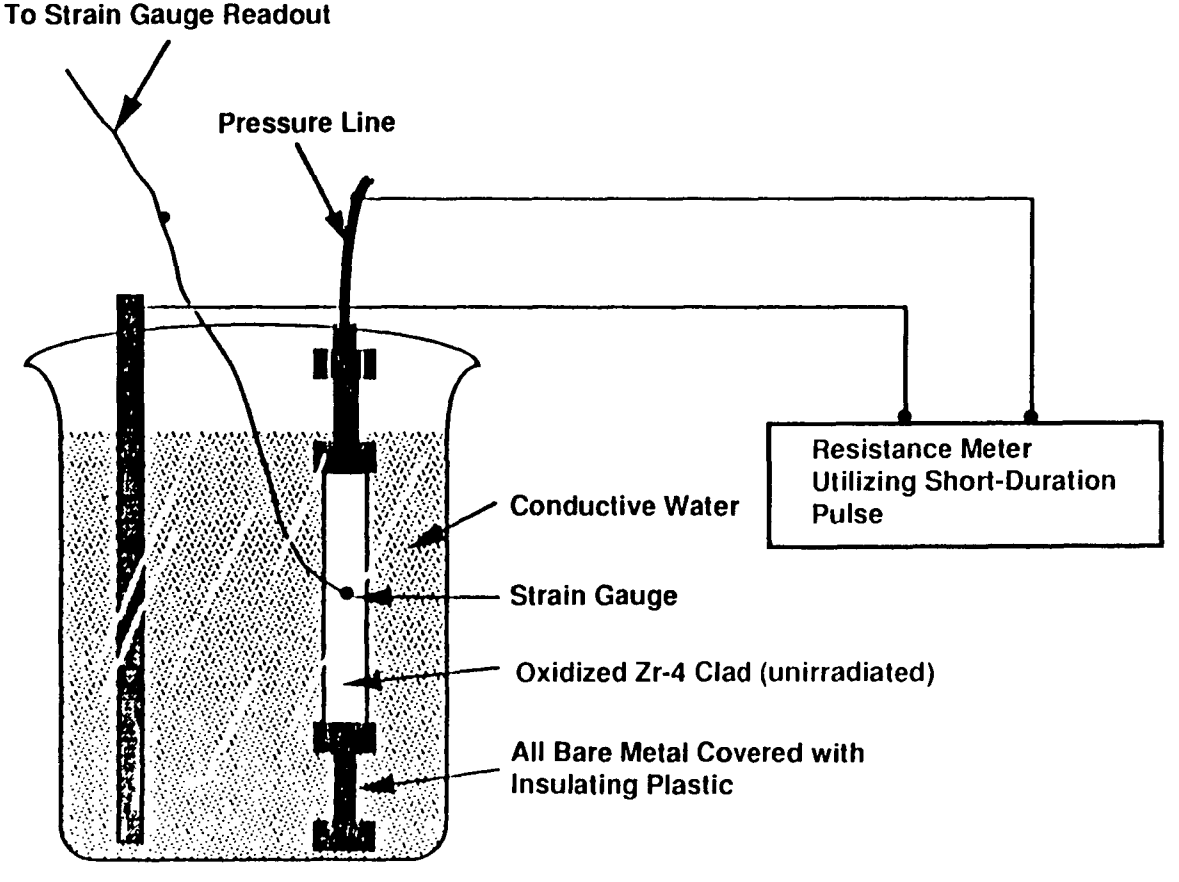
C-RING EXPERIMENTS: SUMMARY/CONCLUSIONS

- DELAYED HYDROGEN CRACKING IS BELIEVED TO BE THE PRINCIPAL MECHANISM OF FAILURE BASED ON COMPARISONS WITH OTHER EXPERIMENTAL WORK
- LOG (TIME TO FAILURE) α % YIELD STRESS
- THE OBSERVED TIME-TO-FAILURE WAS ABOUT TWICE AS LONG IN AIR AS IN WATER UNDER THE EQUIVALENT STRESS CONDITIONS
- C-RING HIGH STRESS DATA EXTRAPOLATED TO EXPECTED REPOSITORY LEVELS FOR RODS WITH HIGH INITIAL PRESSURIZATION (PWR FUEL WITH HIGH FGR) SUGGEST THAT THE MINIMUM TIME TO FAILURE WILL BE ON THE ORDER OF HUNDREDS TO THOUSANDS OF YEARS

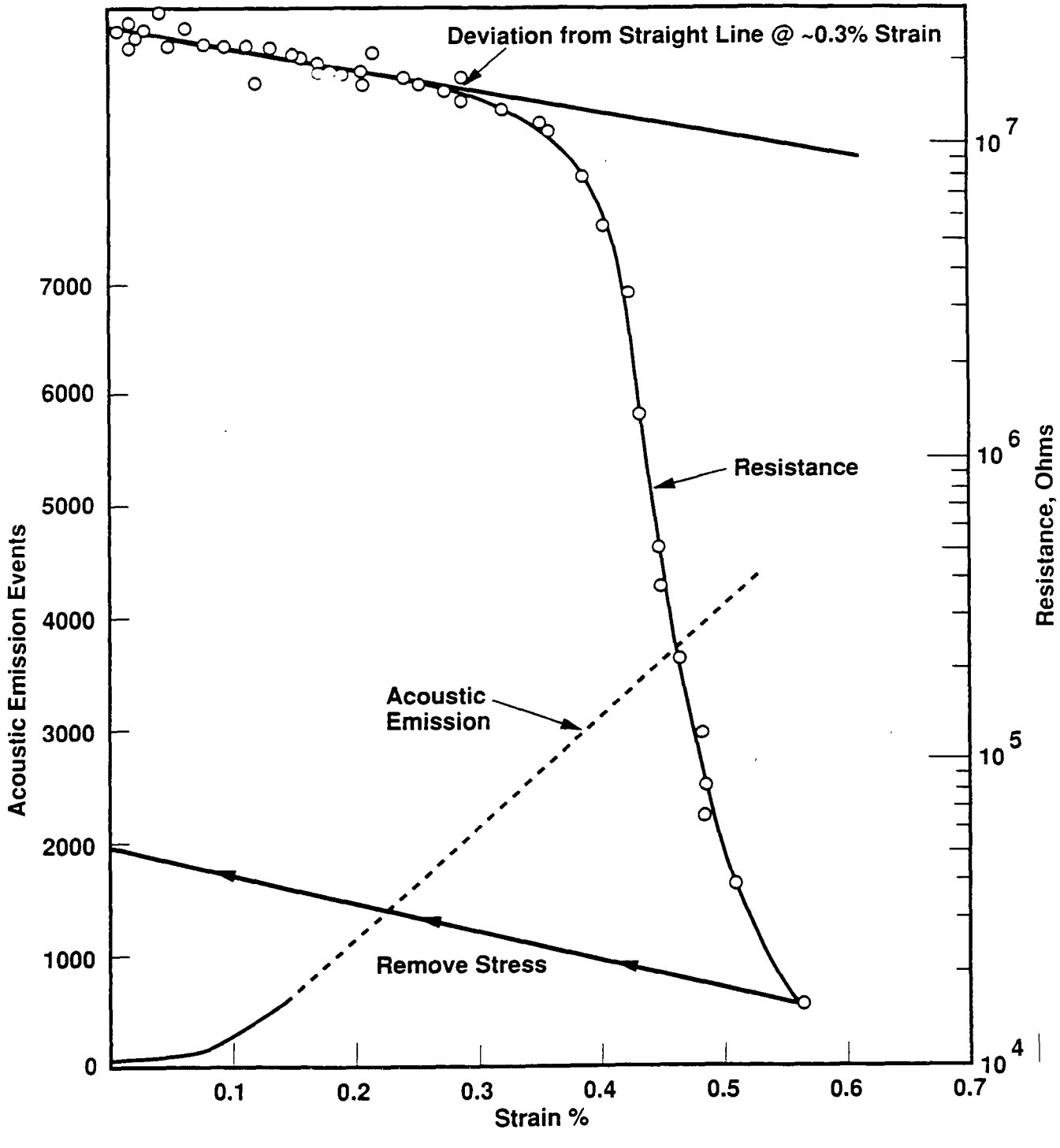
PLANNED PRESSURIZED TUBE TESTING

- DETERMINE STRAIN TO PRODUCE CRACKS THROUGH THE OXIDE FILM
- CONFIRM AND EXTEND THE RESULTS FROM THE C-RING EXPERIMENTS
- DETERMINE THE POTENTIAL FOR HYDRIDE REORIENTATION UNDER POSSIBLE REPOSITORY CONDITIONS
- DETERMINE IMPACT OF HYDRIDE REORIENTATION ON CLADDING CRACKING BEHAVIOR

SCHEMATIC OF SYSTEM FOR INDUCING AND DETECTING OXIDE FILM CRACKS



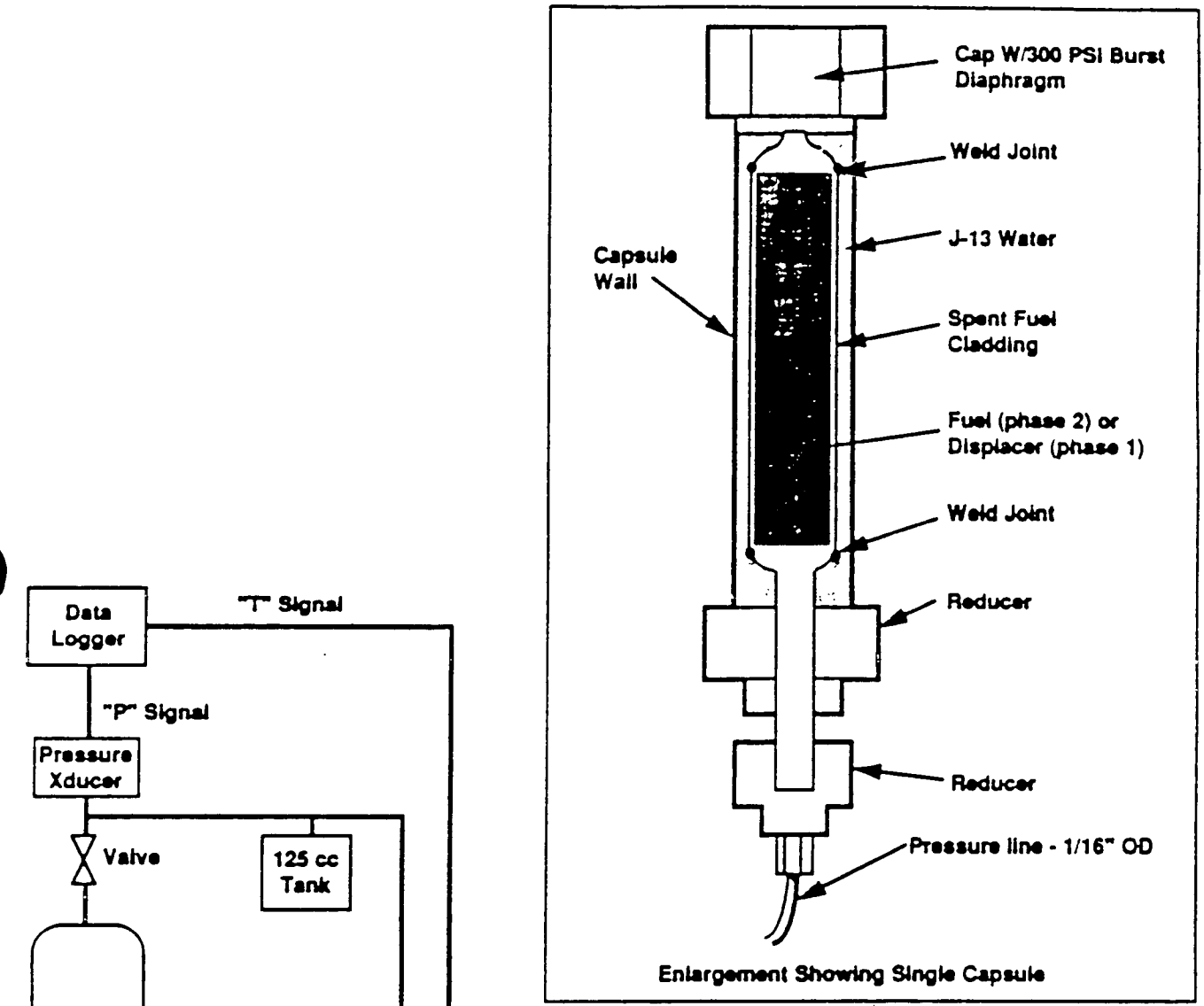
ACOUSTIC EMISSION AND ELECTRICAL RESISTANCE RESPONSE AS A FUNCTION OF STRAIN IN A SECTION OF UNIRRADIATED PRESSURIZED ZIRCALOY CLADDING



OXIDE FILM CRACKING EXPERIMENTS: CONCLUSION

- OXIDE FILM CRACKING BEST DETECTED BY RESISTIVITY TECHNIQUE
- ACOUSTIC EMISSION - SENSITIVE TO TOO MANY OTHER EVENTS

SCHEMATIC OF SYSTEM FOR ADDITIONAL PRESSURIZED TUBE TESTING



HYDRIDE REORIENTATION

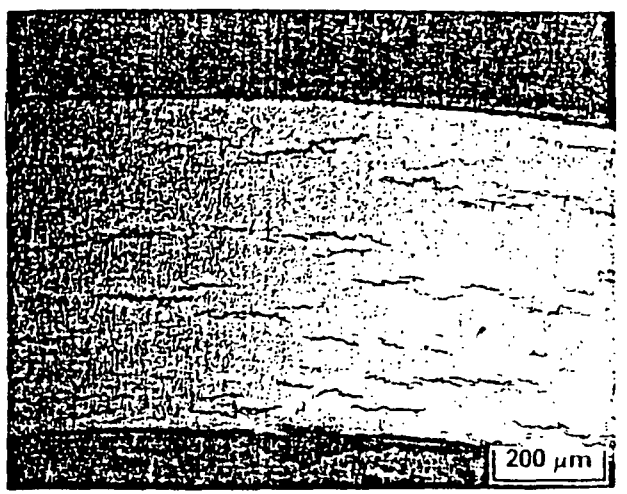
- MAY OCCUR DURING SLOW COOL DOWN AFTER CLADDING REACHES PEAK TEMPERATURE
- REORIENTATION OF HYDRIDES TO RADIAL ORIENTATION WILL DEGRADE THE MECHANICAL PROPERTIES OF THE CLADDING

PWR FUEL ROD HYDRIDE REORIENTATION

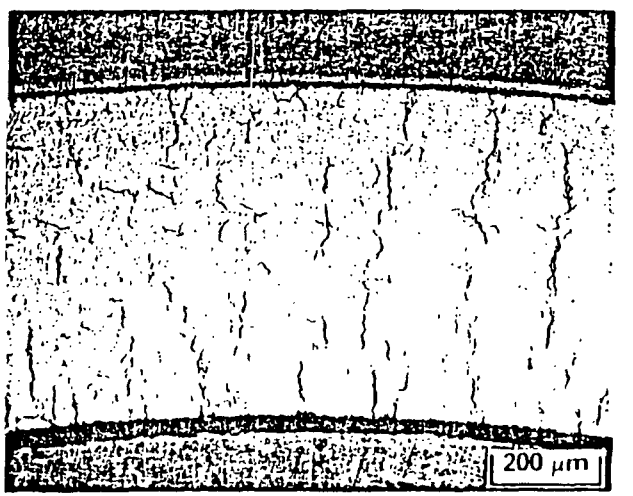
TYPE OF TEST	ROOM TEMP. PRESSURE (MPa)	STRESS (MPa) AT 290°C	SOAK TEMP. (°C)	COOLDOWN RATE (°C/h)	RADIAL HYDRIDES (%)
REACTOR SHUTDOWN*	3.45	~50.4	~350	30-50	0
HIGH PRESSURE WHOLE ROD TEST	9.25	135	323	5	50-90
	0	0	323	5	0

*BECAUSE OF REACTOR SYSTEM PRESSURE, DURING COOLDOWN THE CLADDING WOULD BE IN A COMPRESSIVE STATE

HYDRIDE REORIENTATION



NORMAL



REORIENTED

ZIRCALOY/FLUORIDE CORROSION TESTING: BACKGROUND

- FLUORIDE HAS BEEN RECOGNIZED FOR ITS CORROSION ENHANCEMENT OF ZIRCONIUM FOR MANY YEARS
- FLUORIDE IS PRESENT AT 2.2 ppm LEVELS IN J-13 WATER
- NO QUANTITATIVE STUDIES EXIST AT THESE LEVELS

ZIRCALOY/FLUORIDE INTERACTION

- DEVELOP AND DEMONSTRATE A TECHNIQUE FOR MEASURING THE CORROSION RATE OF ZIRCALOY IN WATER CONTAINING LOW LEVELS OF FLUORIDE

- APPLY THIS TECHNIQUE TO ZIRCALOY SPENT FUEL CLADDING TO OBTAIN THE DATA NECESSARY FOR PREDICTING EFFECT OF FLUORIDE ON THE CORROSION RATE OF ZIRCALOY SPENT FUEL CLADDING

ZIRCALOY/FLUORIDE INTERACTION: SUMMARY/CONCLUSIONS

- pH STAT TECHNIQUE SHOULD BE A PRACTICAL METHOD FOR MAKING CORROSION MEASUREMENTS ON IRRADIATED CLADDING SAMPLES

- ZIRCALOY-4 CORROSION RATE IN WATER IS A FUNCTION OF pH, TEMPERATURE, AND FLUORIDE CONTENT. THESE FACTORS WILL NEED TO BE KNOWN IF THE POST-CONTAINMENT CLADDING CORROSION RATE IS TO BE PREDICTED

- PITTING WAS UNIVERSALLY PRESENT, BUT SPECIFIC CONDITIONS REQUIRED TO PRODUCE PITTING WERE NOT DETERMINED

ZIRCALOY/FLUORIDE INTERACTION: SUMMARY/CONCLUSIONS

(CONTINUED)

- **RESULTS ARE CONSISTENT WITH TWO DISTINCT REACTIONS MAKING UP THE CORROSION REACTION:**
  - FILM (SCALE)-FORMING REACTION
  - FILM-DESTROYING REACTION
- **BEHAVIOR OF IRRADIATED CLADDING NEEDS TO BE COMPARED WITH THE BEHAVIOR OF UNIRRADIATED MATERIAL (AS DETERMINED IN SCOPING STUDIES CONDUCTED TO DATE)**

ZIRCALOY CLADDING DEGRADATION INVESTIGATIONS: SUMMARY

- ^{14}C THERMAL RELEASE - ZIRCALOY SPENT FUEL CLADDING
  - FUNCTIONAL APPARATUS HAS BEEN DEVELOPED
  - SCOPING RESULTS INDICATE ^{14}C RELEASE RATES AND LEVELS ARE TEMPERATURE AND ATMOSPHERE DEPENDENT
- ZIRCALOY CLADDING CRACKING STUDIES
  - C-RING HIGH STRESS DATA EXTRAPOLATED TO EXPECTED REPOSITORY LEVELS SUGGEST THAT THE MINIMUM TIME TO FAILURE WILL BE HUNDREDS TO THOUSANDS OF YEARS
  - PRESSURIZED TUBE TESTS WILL EXTEND C-RING RESULTS TO LOWER PRESSURES AND ALLOW EVALUATION OF THE POTENTIAL FOR (AND IMPACT OF) HYDRIDE REORIENTATION

ZIRCALOY CLADDING DEGRADATION INVESTIGATIONS: SUMMARY

(CONTINUED)

- **FLUORIDE-INDUCED ZIRCALOY CORROSION**
  - CORROSION IS OBSERVED AT LOWEST FLUORIDE LEVEL INVESTIGATED (100 ppm)
  - CORROSION RATE IS STRONGLY DEPENDENT ON pH AND FLUORIDE CONCENTRATION
- **RESULTS OF CLADDING DEGRADATION STUDIES WILL BE UTILIZED IN WASTE PACKAGE AND TOTAL SYSTEM PERFORMANCE MODELS**