Conf-820406--32

POSTTEST EXAMINATION OF SODIUM LOOP SAFETY FACILITY EXPERIMENTS

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CONF-820406--32

DE83 009083

ABSTRACT

In-reactor, safety experiments performed in the Sodium Loop Safety Facility (SLSF) rely on comprehensive posttest examinations (PTE) to characterize the postirradiation condition of the cladding, fuel, and other test-subassembly components. PTE information and on-line instrumentation data, are analyzed to identify the sequence of events and the severity of the accident for each experiment.

Following in-reactor experimentation, the SLSF loop and test assembly are transported to the Hot Fuel Examination Facility (HFEF) for initial disassembly. Goals of the HFEF-phase of the PTE are to retrieve the fuel bundle by dismantling the loop and withdrawing the test assembly, to assess the macro-condition of the fuel bundle by nondestructive examination techniques, and to prepare the fuel bundle for shipment to the Alpha-Gamma Hot Cell Facility (AGHCF) at Argonne National Laboratory. During the macroexamination at the AGHCF, information is collected to permit assessment of cladding motion and redistribution; mode, extent, and location of fuel disruption; and blockage extent, location, and type. Specific microscopic examinations including optical and quantitative metallography, scanning electron microscopy, and electron microbeam analysis are often employed to satisfy the goals of the comprehensive postmortem examinations.

INTRODUCTION

The Sodium Loop Safety Facility (SLSF) program is being conducted at Argonne National Laboratory as part of the United States liquid-metal fast breeder reactor (LMFBR) safety assurance program. A series of experiments in the SLSF research program addressed the priority problem of various hypothesized whole-core accidents, which are of extremely low-probability but have potentially serious consequences.

The initiating phase of an unprotected, loss-of-flow accident was simulated in the SLSF experiment P3A, under prototypic LMFBR conditions, in order to extend current knowledge of the mechanisms involved in cladding relocation and initial fuel disruption[1]. The experiment was terminated prior to fuel melting, following sodium boiling and cladding relocation, to preserve a wide cladding and fuel-failure spectrum for the PTE, and for this reason, SLSF experiment P3A has been selected to provide a framework for discussing the various complexities of a comprehensive PTE. Each phase of the disassembly and examination involved formidable challenges that once overcome provided the necessary information for satisfying the P3A experiment objectives.

SLSF P3A EXPERIMENT DESIGN AND OPERATION

When inserted into the Engineering Test Reactor (ETR) at the Idaho National Engineering Laboratory (INEL), the SLSF loop is capable of providing a test subassembly with a thermal, hydraulic, and neutronic environment which simulates that within a fast reactor during some postulated accident conditions. In addition, the loop can operate at full-power, steady-state conditions for extended periods as required for the desired preconditioning of the fuel.

The main components of the 8.2-m long SLSF loop are (1) the test subassembly, (2) the annular linear-induction circulating pump, and (3) the sodium-to-helium heat exchanger that provides loop heat removal. Approximately half of the loop sodium flow passes through the fuel-bundle test section; the remaining half flows through an annular, orificed bypass that provides for continuous loop flow during tests in which the subassembly becomes blocked by relocated cladding and fuel. A cadmium filter, for hardening the neutron-flux spectrum, is bonded to the outer vessel of the doubly contained loop.

The P3A subassembly cross-section, illustrated in Figure 1, consisted of a bundle of thirty-seven, full-size LMFBR fuel elements containing mixed-oxide (25% PuO_2, 75% UO_2) fuel 237 cm long with a 91-cm active fuel length. The shroud assembly consisted of an inner niobium-1% zirconium hex-can, zirconia insulator pieces, and an outer Inconel 625 tube.

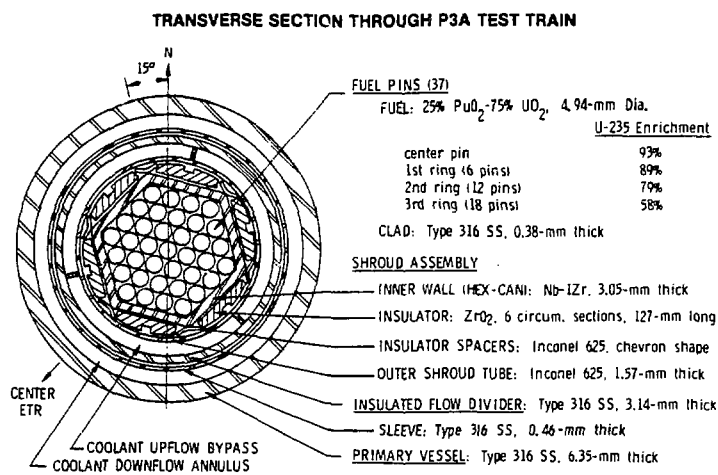


Fig. 1 Components of the P3A Subassembly Cross-Section

The multiphase P3A experiment consisted of testing phases, irradiation periods, and a fuel conditioning interval terminated with the loss-of-flow simulation. The P3A fuel elements were irradiated for an equivalent of twenty-six full-power days (bundle power = 1240 kW). Prior to the early-termination loss-of-flow (LOF) transient, a fuel-conditioning period was scheduled to insure proper fuel restructuring and fuel-cladding gap conductance. During this period, the P3A fuel bundle power of 1240 kW was maintained for approximately 90 hours. The LOF was performed by

reducing the fuel-bundle sodium flow to 30% of normal during a 13-sec period terminated by an ETR scram. The flow reduction simulated, as closely as possible, the flow coastdown that would result from loss of power to the pumps in a fast-reactor primary cooling system. The coolant flowrate was maintained at the 30% of normal value to remove residual decay heat prior to freezing the sodium within the loop. The loop sodium was frozen to stabilize the fragile cladding and fuel that were disrupted and relocated during the LOF accident simulation.

DISASSEMBLY AND NONDESTRUCTIVE EXAMINATION AT THE HFEF

Following the in-reactor experimentation, the P3A loop and test assembly were removed from the ETR and transported to the HFEF at the INEL for initial disassembly. The presence of mixed-oxide fuel and the loop's irradiated condition necessitated that the dismantling operation and all subsequent examinations be performed remotely in shielded cells.

The dismantling operation was initiated when the fission-product decay heat source had diminished to the point where removal of the test train from sodium into the HFEF argon atmosphere would not subject the fuel or structural materials to excessive temperatures. A controlled sodium-freezing sequence (top to bottom) had been used to maximize the void fraction in the test section prior to removal of the loop from the ETR, thereby minimizing possible degradation of experimental evidence due to expansion of loop sodium during the remelt operation. A controlled sodium-remelt sequence was used (also top to bottom) at the HFEF to decrease the stresses imposed on loop and test-assembly structures during sodium melting and expansion.

The test train was sectioned approximately in thirds as it was withdrawn from the loop. Sodium drained from the bottom test-train segment, containing the fuel, as it was removed from the loop leaving the fragile bundle components unstabilized. Steps were initiated to restabilize the fuel bundle prior to additional handling by sealing the lower end, immersing the test-train segment in the molten sodium within the loop, withdrawing the segment, and allowing the sodium to freeze within the bundle.

Full-length neutron radiographs were produced of the P3A fuel bundle at three azimuthal views (60° apart) aligned with the hex-can flats thus allowing sight down the rows of fuel pins. Two of these radiographs are shown in Figure 2 on either side of the axial-distribution-of-materials plot. Examination of the radiographs provided evidence of the formation of upper and lower metal blockages, the extent of fuel relocation and disruption, and the survivability of the shroud assembly. Knowledge of the macrocondition of the fuel bundle was essential in defining the remaining disassembly procedures at the HFEF.

To a certain extent, neutron radiographs can be used to identify various materials by their relative opacity to penetrating neutrons, but gamma scanning provides a more reliable method of identifying fuel and activated materials prior to destructive examination. Full-length, isotopic gamma scanning was performed at 6.4-mm steps corresponding to each of the three azimuthal neutron-radiographed orientations. Detailed

gamma scanning was performed throughout the fuel bundle, at the upper and lower metal blockages, at the original bottom of the active fuel, and at other selected regions where degradation of the hex-can was indicated by the radiographs.

Examination of the neutron radiographs indicated essentially complete upper and lower metal blockages at the ends of the fuel columns; therefore, the previous attempt to stabilize the fuel bundle with sodium was probably unsuccessful. A plan was devised to stabilize the fuel pellets adjacent to the voided region as identified in the radiographs of Figure 2. An end mill was used to remove the flow divider from the test assembly, and a hole was drilled through the shroud assembly into the cavity at the location indicated in Figure 2. The test, assembly was wrapped with electrical heating straps adjacent to the cavity, insulated to minimize heat loss, and heated to a temperature above the melting point of sodium. Molten sodium was poured into the hole and allowed to freeze, thus stabilizing loose pellets within the cavity.

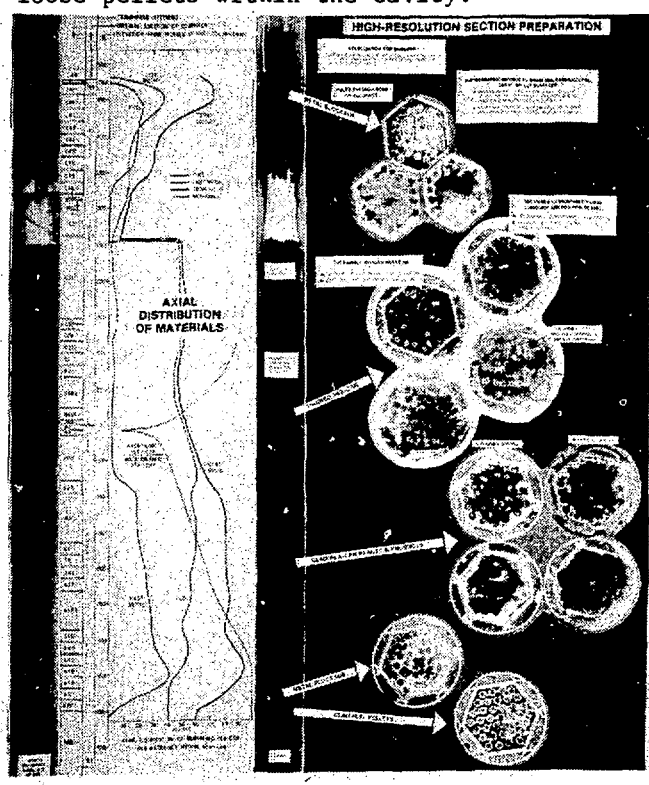


Fig. 2 Summary of P3A Macroexamination

Personnel at the AGHCF placed two requirements on the fuel-bundle shipment from the HFEF; no section could be longer than 0.9 m nor contain more than one-third of the active fuel. The radiographs were examined, and the sectioning sites were selected to satisfy the AGHCF requirements, to minimize the number of cuts and the resulting potential spread of contamination within the HFEF, to minimize damage to regions of interest, and to provide adequate stabilization during shipment.

The fuel bundle was sectioned with a high-speed diamond wheel; no coolant could be used. Cutting was periodically interrupted to minimize overheating fuel and sodium. The equipment did not perform satisfactorily since it was impossible with available facilities to section the piece without local over-heating resulting in melting sodium adjacent to the cut. Pillows of crumbled aluminum foil were secured to the ends of each segment to hold loose pellets in place during shipment to the AGHCF in Illinois.

MACROEXAMINATION AT THE AGHCF

The AGHCF has been in operation for 18 years and was designed to be used for metallurgical research of plutonium in support of the LMFBR program. In the macroexamination-phase of the PTE, information was collected to permit assessment of the following:

- cladding motion and distribution
- mode, extent, and location of fuel disruption, and
- blockage extent, location, and type.

It was necessary that the disassembly method retain the as-received condition and position of the fuel pellets and structural components. Also, macrofeatures on the as-cut surfaces had to be clearly delineated to facilitate the investigation of the condition, interaction, redistribution, and behavior of materials. The most effective method of examining the fuel bundle was found to be transverse sectioning throughout the regions of interest. The hex-can and outer-shroud tube provided radial support for the disrupted fuel, and 7-mm and 15-mm thick macrosections were made for metallography specimens. To make the necessary transverse sections, a sectioning machine was developed that is illustrated in Figure 3. It uses a high-speed 203-mm diameter by 1-mm thick SiC abrasive cutoff wheel without coolant. Functional characteristics to minimize heating the work and to obtain smooth, accurate ascut surfaces are outlined in Table I.

It was known that some regions of the fuel bundle contained disrupted fuel pellets that were not completely stabilized. At first, epoxy was used to secure the loose components, but that proved unsatisfactory during the micro-examination because the epoxy broke down at the temperature necessary to remove sodium by evaporation. The use of epoxy as a stabilizing agent was discontinued, and sodium was substituted. A suitable method of removing the sodium prior to metallography was developed and will be discussed later.

The fuel-bundle segments were sectioned at the axial positions identified in Figure 2 with the corresponding specimen labels tabulated in the left column of the figure. Photographs were made at 2X magnification of the top and bottom as-cut surfaces of all transverse sections using oblique illumination to highlight reflective surfaces (to distinguish metal from non-metal), and straight-on illumination for flat lighting. Representative macrophotographs are shown on the right of Figure 2. The various labels identify some of the pertinent features observed in each macrosection.

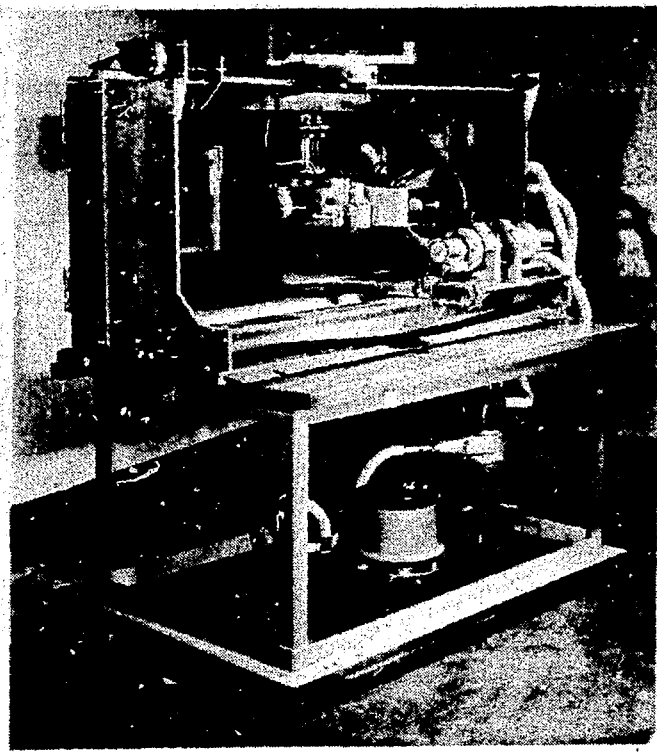


Fig. 3 The AGHCF Sectioning Machine - Mod. III

TABLE I

Functional Characteristics of the AGHCF Sectioning Machine - Mod. III to Optimize Transverse Sectioning of SLSF Loops

Minimize heating the work by -

- rotation of work to minimize surface contact between the cutting wheel and the work
- controlled slow feed of the wheel into the work
- extraction of heat by rotation of the high-speed cutting wheel, which drags hot-cell atmosphere past the work.

Accuracy and smoothness of cut surface obtained by -

- roller support of work under area being cut
- minimum play in cutting and holding mechanisms
- rotation of work in same direction as saw to keep the chain drive tight
- lock drive mechanism for saw on screw drive while cutting
- minimum cantilevered length of work being cut.

Examination at low magnification (2X) of macrosections throughout the P3A fuel bundle revealed two principal modes of fuel disruption: buckling

of stacks of declad fuel pellets, and large-scale swelling of unmelted fuel pellets. Many of the free-standing columns of declad fuel pellets from the inner fuel pins buckled throughout the bundle, and pellet-column segments relocated axially resulting in more than the original 37 pellets in a region extending from the fuel midaxis to 350 mm below. As a result the segments of the inner fuel columns at the upper end of the bundle were unsupported and fell leaving a large central cavity that can be observed in the Figure-2 neutron radiographs. Except for few a pellets tipped on their sides, most of the free-standing, pellet-column segments remained vertical throughout the rest of the fuel bundle.

Extreme radial swelling of pellets was found in an axial region extending from 80 mm to 160 mm above the fuel midaxis. Practically all of this swelling occurred in the relatively unrestructured outer rim of fuel. Fuel in the central pins can be seen to have swelled much more than fuel in the outer ring of pins. The amount of this type of swelling decreased with lower elevation. In the regions where gross fuel swelling occurred, the fuel occupied as much as 50% more cross-sectional area than would an equivalent number of unswelled pellets. Examination of the macrosections at 2X magnification also provided evidence of the type and extent of fuel restructuring throughout the bundle.

A Quantimet 720 image-analysis system, augmented with an image editor, was used to derive the axial distribution of materials within the fuel bundle plotted in Figure 2. Information such as this helped the experimenter understand changes in reactor reactivity and quantify the reduction of coolant flow area that were a result of the experimental transient.

MICROSCOPIC EXAMINATION

Highest priority was given to the microstructural characterization of the fuel with emphasis on the role fission gas played in initial fuel disruption. The examination of over 80 fuel pellets involved microstructural characterization by metallography and the determination of fission-gas behavior and effects by Quantimet, SEM, and laser analyses. The upper and lower metal blockages were examined by metallography and electron microprobe analysis to identify the sources of materials comprising the blockages.

Comprehensive metallography was a prerequisite for each phase of the P3A microexamination with specimen retrieval being the most time consuming operation of the examination. Metallic specimens and fuel pellets embedded in metal were removed by sectioning with an abrasive cutoff machine. Fuel pellets encased in sodium provided the most difficulty as removal from the 7-mm thick transverse sections was accomplished with a punch and die apparatus sized slightly larger than the pellet diameter.

The normal method of removing small amounts of sodium from specimens was by a water-alcohol dissolution process, but this method was unsatisfactory for the swelled P3A fuel mainly because of its extremely fragile condition. A technique was developed to remove sodium by evaporation in which the pellets were heated to 350°C in a pyrex tube for one hour at an absolute pressure of less than 10 microns mercury. After

cooling, the sodium-free pellets were vacuum cast in epoxy and allowed to cure for 24 hours. The lower-left micrographs in Figure 4 illustrate the results obtained by both sodium-removal techniques. The dissolution technique led to a nonrepresentative void fraction caused by enhanced grain pullout during preparation. A more representative microstructure was obtained by the vaporization technique which made it suitable for quantitative metallography and laser sampling.

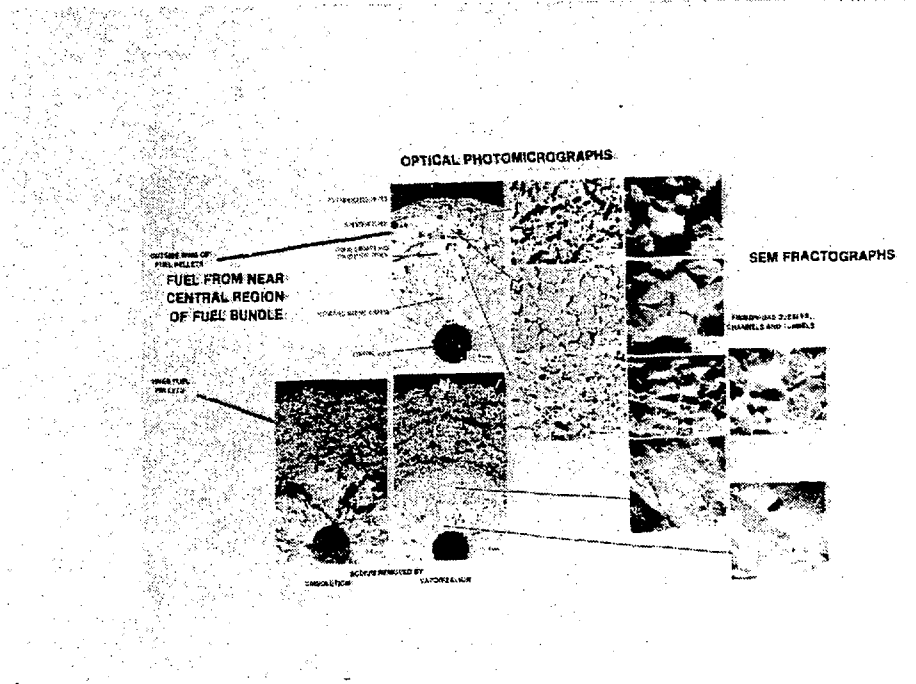


Fig. 4 Microexamination of Fuel from the Central Region of the Bundle.

The metallographic techniques used during this microexamination are described in Table II. The listed times were varied for different materials to obtain satisfactory microstructures, and the specimens were ultrasonically cleaned in ethyl alcohol after the fine grinding and each remaining operation.

The upper and lower metal blockages were examined to determine the origin of the metal. Samples of deposited metal throughout the dome and along the inside of the hex-can were prepared for semi-quantitative analysis for composition with a shielded electron microprobe. As expected, analysis showed the deposited metal to be principally Type 316 stainless steel indicating it originated mainly from the cladding. Likewise samples were prepared from the lower metal blockage. A metallographic examination indicated that the metal comprising the blockage derived from the interaction of Type 316 stainless steel cladding and the Nb-1% Zr hex-can. Electron microprobe analysis confirmed the presence of both host metals.

TABLE II

Summary of the AGHCF Metallographic Specimen Preparation

Course grinding:	320 grit SiC, dry.
Fine grinding:	600 grit SiC, 10-15 min.
Course polishing:	3μ diamond on nylon, Hyprez, 10-15 min.
Fine polishing:	1μ diamond on nylon, Hyprex, 10-15 min.
Etching: (when required)	$(U,Pu)O_2$ fuel - immersed in 90 ml of 30% hydrogen peroxide mixed with 10 ml concentrated sulfuric acid.

A comprehensive metallographic examination of the P3A fuel was undertaken to characterize the microstructures of pellets from three regions (bottom, central, and upper) within the fuel bundle. Representative micrographs showing varying degrees of swelling are illustrated in Figure 5. Fuel from near the central region of the bundle displayed a restructured microstructure with a central void, a region of columnar surrounded by equiaxed grains, and an unrestructured zone at the pellet periphery. In Figure 6a, micrographs of highly and extremely swelled fuel show the grain-boundary porosity caused as microscopic fission-gas bubbles migrated to the boundaries and coalesced there forming larger bubbles. This process continued within the unrestructured zone of the fuel; grain boundaries became weakened and many separated because of imposed thermal and mechanical stresses. Formation of large fission-gas bubbles in conjunction with grain-boundary separations explains the gross swelling observed in the P3A PTE. Significantly less restructuring occurred in the upper and lower regions of the fuel bundle than at the fuel midaxis. Restructuring had barely begun even though fission-gas pores decorated many of the grain boundaries. The polished surface of a metallography specimen was scratched with a glass-cutting tool to cause the grain-boundary fractures observed in the micrographs of Figure 6b. The SEM fractographs illustrated in Figure 6b were prepared basically to examine the fission-gas morphology at the grain boundaries.

Quantitative data were collected from fuel pellets of different axial and radial positions that represent a wide range of swelling in the region outside the columnar grain regions. The pore and bubble distribution were evaluated for representative fuel specimens by determining the void-volume fraction and the mean pore intercept using the Quantimet on 250X photos. The extremely large pores of some fuel specimens precluded the use of the Quantimet so the point count method, based on random tossing of a grid, was used to determine the void-volume fraction.

Direct radial swelling measurements were made on the 2X as-cut surface photos of selected pellets throughout the fuel bundle. Also, direct swelling measurements were made on 50.9X composite photos of four pellets. The swelling data from the 2X and 50.9X photos of the lesser swelled pellets were in excellent agreement. However, higher swelling was indicated in the 2X photos than in 50.9X composites for the more highly swelled pellets. The difference can be attributed mainly to the loss of fuel particles from the perimeter of the extremely friable fuel during

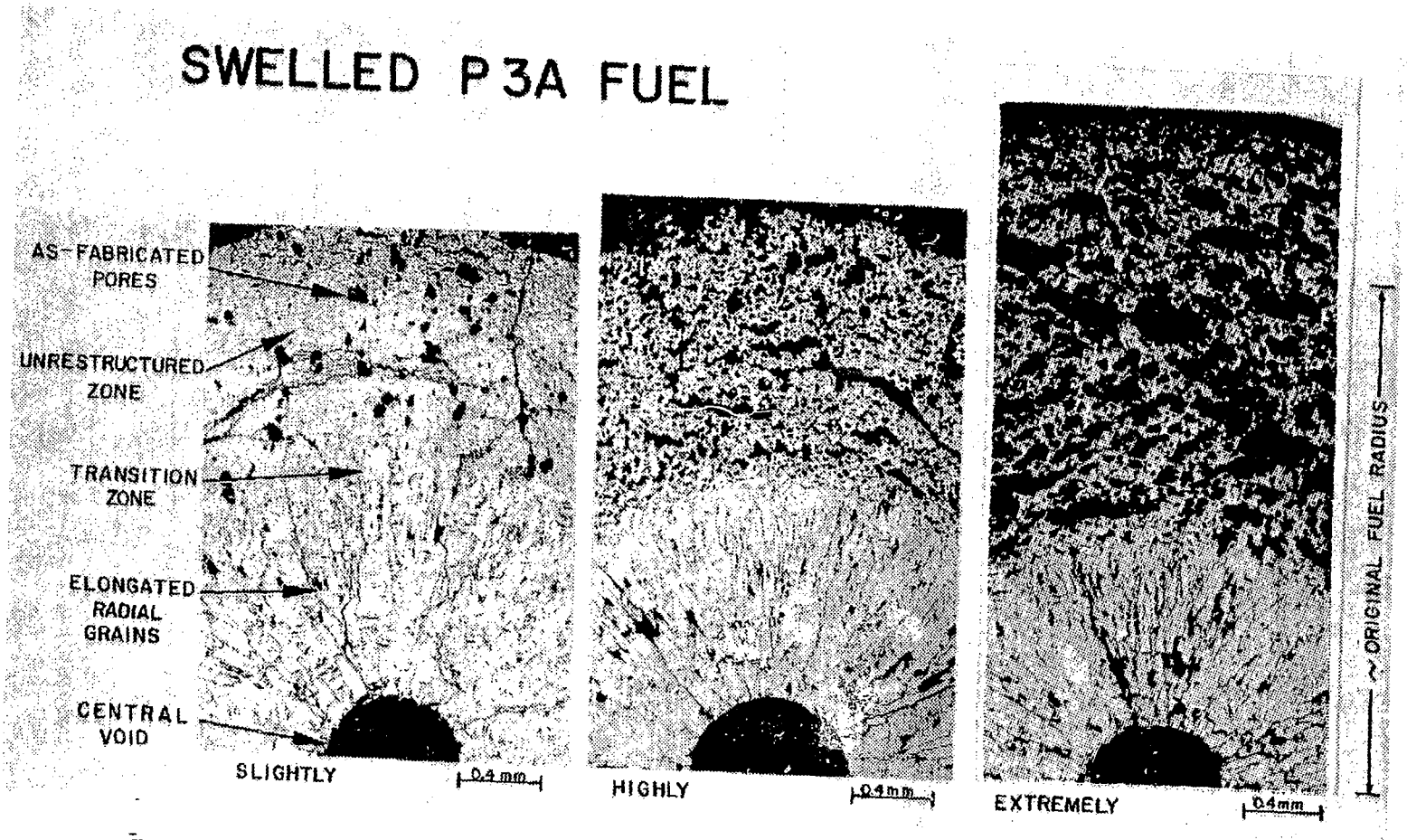
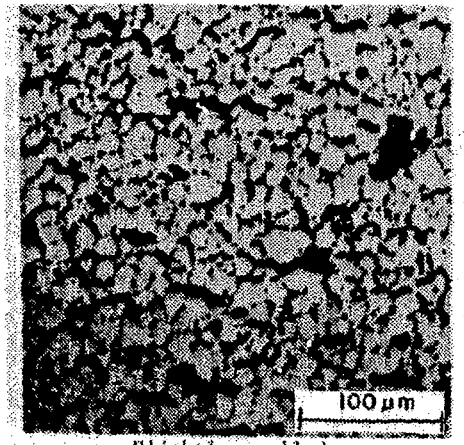
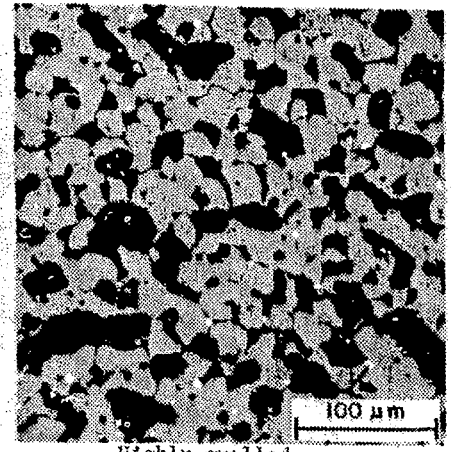


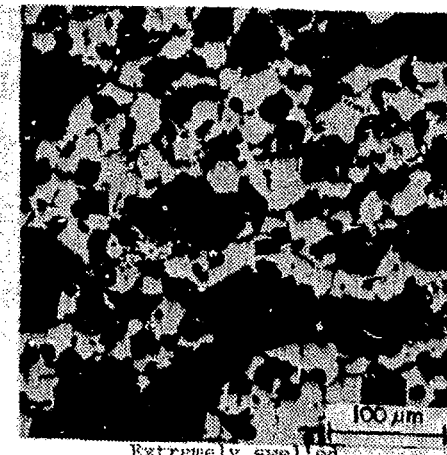
Fig. 5 Radial Strips from Fuel Pellets Showing Varying Degrees of Swelling Outside the Columnar Grain Region, ~+135 to +145 mm. As polished. Neg. No. MSD-226800.



Slightly swelled

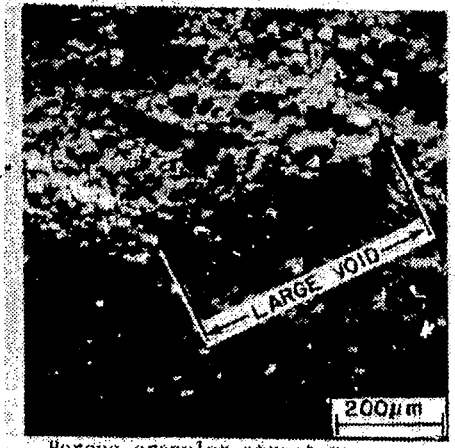


Highly swelled

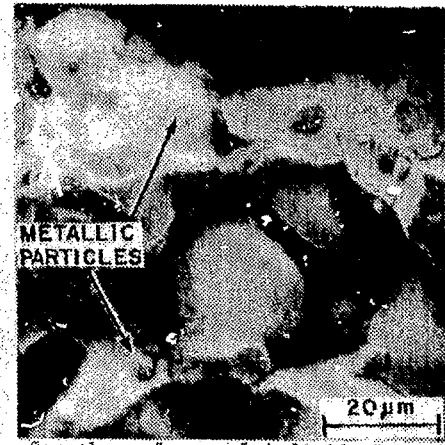


Extremely swelled

(a)



Porous granular structure showing several large voids



Smooth surfaces of individual grains that procrude into medium size void

Smooth surface of large void that extends over many grains

(b)

Fig. 6 Microstructures of Swelled Fuel. (a) Optical photos of polished fuel specimens that show a range of swelling; (b) SEM fractographs of extremely swelled fuel. ANL Neg. No. MSD-226801.

their metallographic preparation.

Also, grain-size distributions were determined of two archive fuel pellets and pellets from various axial positions in the posttest fuel bundle by the mean linear intercept method. The data were limited to relatively dense "tight" fuel since the pullout of grains from porous "loose" fuel made the data unreliable. In general, the grains near the center of the pellets and around mid-radius were larger than the grains near the outer edge, and all were two to three times as large as the grain size of the archive pellets. No other trends in the data were apparent.

The radial distribution of retained fission gas was determined for the three pellets by a laser sampling method. In this method krypton-85 is released by laser heating, and the volume of material from which release occurred is measured and used to calculate the concentration of retained krypton-85 at the sample site.

CONCLUSIONS

Posttest examination of the SLSF P3A experiment was a multiphased study to investigate the mode of fuel disruption during the initiating phase of an unprotected LOF accident in an LMFBR. The presence of disrupted $(U,Pu)O_2$ fuel and the loop's irradiated condition necessitated that the disassembly operation and the posttest examination be performed remotely in shielded cells. Particular care was taken during all handling and examination procedures to preserve the posttest condition of the fragile fuel-bundle components by providing for suitable stabilization of the components, by developing a unique cutoff machine to section the fuel bundle, and by developing a method to satisfactorily remove sodium from fuel samples prior to metallography. Optical metallography, scanning electron microscopy, quantitative metallography, and electron microprobe analysis were employed in this comprehensive posttest examination. The gross swelling observed in the P3A fuel was shown to be caused by the formation of fission-gas bubbles that coalesced at and weakened grain boundaries, resulting in extensive grain-boundary separations.

ACKNOWLEDGMENTS

The author expresses his appreciation to the HFEF/N crew for the disassembly and nondestructive examination of the P3A loop and test train and to the AGHCF crew under the supervision and direction of L. R. Kelman for the disassembly and detailed examination of the P3A fuel bundle, especially to G. H. Wolf for sectioning and J. C. Florek for metallographic preparation and examination. The contribution of R. C. Kuo in determining the axial distribution of materials and J. E. Sanecki for SEM and electron microprobe analyses are also acknowledged. This work was performed under the auspices of the U. S. Department of Energy.

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