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POSTIRRADIATION EXAMINATION OF LIGHT WATER REACTOR FUEL: A UNITED STATES PERSPECTIVE

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L.A. NEIMARK, MS, Senior Metallurgist, Argonne Mational Laboratory, Argonne, Illinois N. OCKEN, PhD, Project Manager, Electric Power Research Institute, Palo Alto, California Postirradiation examination of light water reactor fuel: a United States perspective

Poolside and hot-cell postirradiation examination (PIE) have played and will continue to play a significant role in the U.S. LNR program. The principal uses of PIE are in fuel surveillance, fuel improvement, and frilure analysis programs and in the postmortem analysis of safety-related tests. Institutional problems associated with fuel shipping, waste disposal, and fuel disposal can be expected to pose obstacles to hot-cell examinations and likely result in more sophisticated poolside examinations.

INTRODUCTION

The role of postirradiation examination (PIE) in the United States Light Water Reactor fuel program and the facilities in which the eyaminations are conducted have evolved along with the industry itself. From the early development programs on small specimens, geared to understanding fuel behavior, has grown the need and the capability to examine full-size BVR and PWR assemblies and rods and to perform detailed metallurgical investigations. Hot cells now are basically metallurgical laboratories replete with modern, sophisticated instruments. Concurrently, detailed nondestructive poolside examination has come along to augment hot-cell PIE, and, as a screening tool, to minimize the need for handling large numbers of full-size assemblies in hot cells.

The use of PIE in the U.S. can be divided into five segments. First, early fuel development programs took a conservative approach in pursuing the unknowns of fuel irradiation behavior. Numerous specimens and small-scale prototype assemblies irradiated in test reactors were given detailed postirradiation examinations with the limited capabilities that were then available. From these examinations evolved the fuelrod designs of the early commercial reactors. Second, licensing requirements of the then Atomic Energy Commission and now the U.S. Nuclear Regulatory Commission (NRC) state the implied need for surveillance of LWR fuel to assure that the fuel assemblies perform in the anticipated manner. These surveillance programs are conducted by the fuel vendors and are supported by then and the Electric Power Research Institute (EPRI).

The third area for PIE is the analysis of fuel-element failures. Over the years a number of generic failure mechanisms have appeared that have been the object of intensive PIE investigations. The principal ones have been hydriding of Zircaloy cladding, cladding collapse owing to fuel densification, and pellet-cladding interaction (PCI). The PIE campaigns associated with

these mechanisms will be discussed later to highlight the role of PIE in LUR fuel development. Coming full circle, the fourth area for PIE is fuel improvement. The impetus for design optimization in the U.S. is principally economic; from increasing reactor operating efficiency by minimizing fuel failures (after the failure mechanism has been identified) to stretching the world's uranium resources. These fuel improvement programs are supported by the U.S. Department of Energy (DOE) and EPRI.

The fifth area of PIE is the extensive examination given in-reactor tests that simulate hypothetical accident situations. This work is supported by the NRC and is conducted at the Power Burst Facility (PBF) and the Loss-of-Flow Test (LOFT) facility at the Idaho National Engineering Laboratory (INEL). The purpose of this work is to confirm the adequacy of the safety margins implicit in the licensing criteria and of the evaluation models specified in regulatory statutes.

Because the PIE data generated in fuel improvement, fuel surveillance, and fuel safety programs are used in the quasi-legal proceedings of the NRC, postirradiation examinations have essentially become "forensic metallurgy". The legalistic aspect of PIE is underscored by the application in the U.S. of strict quality assurance procedures for the collection of PIE data and the calibration of the instruments that are used. Sound engineering practice has thus been married to the need for quality data that must withstand the scrutiny of both proponents and opponents of nuclear power.

The following sections of this paper will provide an overview of the current U.S. capabilities for LWR postirradiation examinations, the role of PIE in fuel development, and the future of PIE in the U.S. LWR program.

CURRENT U.S. PIE CAPABILITIES

The capability to perform PIE in the U.S. falls into two categories: (1) poolside examinations at the reactor site, and (2) hot-cell examinations. Prolside examinations are performed to obtain data on whole assemblies and as many individual rods as possible, given the constraints of the bundle design. These examinations provide surveillance data during reactor refueling periods, act as a screening tool for selecting fuel rods for in-depth hot-cell examination, and provide an early, albeit limited, characterization of fuel-rod failures. Poolside examination provides the earliest information, with excellent turnaround time, on the condition of the assembly and fuel rods. One of the most useful procedures for identifying assemblies containing failed rods is "sipping" for fission-product activity. Visual examination also plays a major role in assembly interrogation. Typical poolside examination capabilities in the U.S. are given in Table I.

Table I. Poolside examination capabilities in the ${\tt U.S.}$

Visual inspection and photography
Channel measurement
Spacer/grid measurement
Bundle disassembly
Rod length
Profilometry
Ultrasonic and eddy-current inspection
Crud sampling
Fission-product sipping
Fission-gas puncturing and collection
Rod disassembly or sectioning

Because of their high cost, hot-cell examinations are generally performed only when generic failures are encountered or destructive examination data are required for fuel surveillance and improvement programs.

The two commercial hot-cell facilities in the U.S. that can handle an intact fuel assembly are the Battelle Memorial Institute Hot Cell Laboratory at West Jefferson, Ohio (ref. 1) and the Babcock and Wilcox Hot Cell Facility at Lynchburg, Virginia (ref. 2). Both facilities can unload under water large casks containing full-size assemblies of either BWR or PWR design. When possible, however, only individual full-length rods are brought into the hot cells. The General Electric Co. hot-cell facility at the Vallecitos Nuclear Center in Pleasonton, California can handle fuel-rod segments up to 2.2 m long (ref. 3). The MRC's safety tests are examined in the DOE hot cells near the test reactors at INEL, which have capabilities similar to those of the commercial hot cells.

Once in any of these hot cells, the assembly and individual rods can be examined by a variety of nondestructive and destructive examination techniques. A composite summary of the available hot-cell capabilities is given in Table II.

The degree of sophistication of these techniques compared with those available in the early days of the program is most apparent in the electron-beam area. Electron microprobe analysis and scanning electron microscopy are two tools that have helped change PIE from a qualitative guessing game to a quantitative scientific investigation. These techniques have been invaluable in studies of fuel chemistry and fuel/cladding chemical interactions, and in cladding failure analysis. Electron microprobe analysis, in particular, has added a new dimension to metallography/ceramography with the ability to identify specific microconstituents.

Table II. Composite of hot-cell capabilities in the U.S.

Examination

Visual inspection and photography Gross and specific isotope gamma scanning Profilometry Rod length Eddy-current inspection Ultrasonic wall thickness measurement Leak detection Fission-gas sampling and analysis Rod internal void volume Metallography/ceramography Quantitative metallography Fuel bulk density measurement Fuel burnup analysis Neutron dosimetry Retained or dissolved gas analysis Alpha and beta/gamma autoradiography Electron microprobe analysis Scanning electron microscopy/EDAX Replica and transmission electron microscopy Radiochemistry Neutron radiography Ion microprobe analysis* Scanning Auger microprobe analysis* Laser sampling for retained gas* Precision gamma spatial analysis*

Testing

Tensile and bend Tube burst Hardness and microhardness Charpy impact Fracture toughness

*Available at national laboratories.

These hot-cell capabilities are augmented by those of the U.S. national laboratories. These additional sophisticated capabilities include ion microprobe analysis, scanning Auger microprobe analysis, and laser sampling techniques at Argonne National Laboratory (ANL), and analitical chemistry techniques and precision two-dimensional garma scanning at Los Alamos Scientific Laboratory.

When available, neturon radiography has proven to be an exceptional tool for PIE. This technique, which is used extensively by General Electric at Vallecitos, can be used to determine fuel shrinkage, fuel swelling, fuel restructuring and cracking, and cladding hydriding. Unfortunately, it is not available at commercial reactor sites.

PIE ROLE IN FUEL DEVELOPMENT PROGRAMS

Identification of fuel-rod failure mechanisms and surveillance of current and new fuelred and assembly designs are currently the prin-. ipal functions of PIE in the U.S. Garzarolli ; al. (ref. 4) concluded that three phenomena ti.e., primary hydriding, PC1-induced failures, and cladding collapse due to fuel-pellet densification) have led to the highest incidence of fuel-rod failures in commercial LNRs. On a core-wide basis, representative failure rates were in the 0.1 to 1% range at some plants, while exceeding 1% in some batches of fuel. We will briefly review (1) the role played by PIE in identifying the factors responsible for the failures noted above, (2) the techniques used in the examinations, and (3) the changes in fuel-rod design or operating conditions that were introduced as a consequence of PIE. Also, we will show how PIE is being used in programs to extend the burnup capability of the UO2-Zircaloy system.

Primary hydriding

Internal hydride attack was first observed at significant levels in powder and pellet fuel in the Big Rock Point reactor. Klepfer et al. (ref. 5) have discussed the diagnostic techniques that were used at the Vallecitos Nuclear Center to identify the failure mechanism. Neutron radiography established that high local concentrations of hydrogen were present in failed rods. Metallographic and chemical analyses led to the identification of these regions as "sunbursts" of zirconium hydride that initiated at the ID surface of the cladding. Local hydriding was also observed at the ID surface in unfailed rods, indicating an internal source of hydrogen. Cracking was attributed to the following: The hydride phase is quite brittle, hydride formation produces an increase in volume, and the local stress field tends to align hydrides radially.

The source of hydrogen leading to the local hydriding was subsequently found to be residual moisture in the fuel pellets, introduced by wet grinding to final dimensions and by pickup from the atmosphere. Measurements of out-of-reactor release rates suggested that moisture would be released during reactor startup. Hydrogen then would be liberated by radiolysis or as a consequence of the moisture reacting with the fuel.

The fuel vendors have solved this probelm by implomenting fabrication procedures that are designed to reduce the moisture content in the fuel rod to innocuous levels. Fuel pellets

are now dry ground to final dimensions and absorption of moisture from the atmosphere is minimized by outgassing of the fuel rod immediately prior to welding of the end caps. Also, General Electric inserts a hydrogen getter in each fuel rod.

These changes in fuel-rod design and fabrication appear successful in eliminating primary hydriding as a source of fuel-rod failures. Based on sipping of bundles containing a total of over 200,000 rods after at least one cycle of operation, Proebstle et al. (ref. 6) reported no evidence of hydride failures in newer General Electric designs, and Boman et al. (ref. 7) reported that hydride-related failures in Westinghouse designs occurred only in two regions that contained lowdensity fuel pellets.

Fuel-pellet densification

Flattened fuel rods were observed in five Westinghouse PWRs in the early 1970s. Flattening of the fuel rods was found to follow axial gap formation in the fuel-pellet column. Gap formation itself was of concern since the absence of fuel in a localized region causes a dicrease in the absorption of thermal neutrons and a consequent increase in local flux and press. Jordan (ref. 8) noted that while the incidence of flattening was about 5%, about 10% of the flattened rods failed.

Insight into the processes leading to rod flattening came, in part, from a PIE campaign at Windscale. The most direct evidence that fuel densification was the factor responsible for fuel-rod flattening was provided by fuel-pellet density values as determined by mercury pycnometry: the as-fabricated initial fuel-pellet density of 92% T.D. increased to 94.7-96.0% T.D. following irradiation.

The observation of detrimental consequences that could be ascribed to fuel densification led to many studies aimed at establishing the mecahnism of, and factors controlling, this process. Freshley et al. (ref. 9) have reported in detail the results of an extensive program that was supported by the Edison Electric Institute, EPRI, and the nuclear industry. Its goal was to establish those features of the fuel pellets that lead to densification and to relate the kinetics of the densification process to fuel microstructure and irradiation conditions. Similar procedures were used to characterize both pre- and postirradiation attributes of the fuel pellets. Individual pellet densities were determined by using water and mercury pycnometry. Grain size was determined from optical microscopy measurements on ceramographic cross sections. Pellet microstructure, as reflected in the pore size distribution, were determined by using optical and scanning electron microscopy. Pore size distributions in unirradiated fuel pellets were measured at Battelle Northwest Laboratories while measurements on irradiated fuel pellets were performed at ANL.

The key discovery that emerged from the PIE study was a significant decrease in the number of as-fabricated small pores in fuel-pellet types that are prone to densify (i.e., unstable fuel). The major portion of the increase in pellet density resulted from the elimination of pores less than $\sqrt{2}~\mu m$ in diameter. Grain size and initial pellet density were found to be of less significance in determining the propensity towards densification of a given pellet type. However, pore-size distribution, grain size, and pellet density are determined by the pellet fabrication process and should not be viewed as truly independent variables. Of equal importance from a practical perspective, the microstructural features of stable (i.e., densification-resistant) fuel types were identified and a simple out-of-reactor test (the so-called resintering test) that provides a means of inferring in-reactor behavior was established.

The results from this program and other vendor-sponsored programs have marked an end to densification as a fuel-performance concern. Boman et al. (ref. 7) report that over a two-year period beginning in the fall of 1974, 35 reactor refuelings were performed in Westing-house plants with no indications of flattened sections. Furthermore, the initial stringent licensing procedures have been relaxed to the point (ref. 10) where, for stable fuel, accounting for fuel densification in licensing calculations has little effect on plant capacity or availability factor.

Pellet-cladding interaction

Examination of fuel discharged from the Dresden 1 reactor in 1971 revealed the presence of some rods with longitudinal cladding splits. These failures were attributed to localized stresses and strains that were imposed on the cladding once the as-fabricated gap had closed. It was also suspected that the strain-to-failure capability of the cladding had decreased as a result of PCI. Extensive laboratory, test reactor, and commercial power reactor studies have subsequently been undertaken to identify the factors responsible for this type of fuel failure. The consensus now appears to be that aggressive fission products released from the fuel, as well as stress, are required for cladding failure. The phenomenon leading to failure is believed to be stress-corrosion cracking (SCC) of the Zircaloy.

PIE has played a significant role in developing this consensus. An important piece of evidence is the macroscopic appearance of a typical SCC crack: it is oriented radially, has a high length-to-width ratio, is branched, and shows macroscopically flat fracture surfaces, with little cladding strain. In addition, examinations of fracture surfaces by scanning electron microscopy show differences between fractures that occur in the presence and absence of a chemically aggressive environment. Intergranular cracking and cleavage-plus-fluting features occur only in the presence of aggressive species, and these features are typical of in-reactor PCI failures.

Fuel-rod failure is not necessarily a consequence of high cladding stresses induced by large power increases: Failures occurred in the Maine Yankee reactor even though the fuel had been subjected to only modest power increases. PIE data reported by Fuhrman et al. (ref. 11) pointed out the significant role played by the release of fission products in contributing to the observed failures. The releases of fission products were believed to have been a consequence of an old fuel-rod design (unpressurized rods and the use of densification-prone fuel) that led to a reduction in the gap conductance, a rise in fuel temperature, and the subsequent release of fission products. The reduced role of stress in the failure process is suggested by the postirradiation test results of Yaggee et al. (ref. 12), who showed that neutron irradiation increases the susceptibility of Zircaloy to SCC failure based on tube burst testing with an iodine environment.

The difficulty of locating and characterizing PCI defects during PIE is exemplified by at least one examination campaign of BWR and PWR fuel (ref. 13). Because of the tight nature of the PCI defect, eddy-current techniques were only marginally successful in locating incipient defects. Nevertheless, the evidence from the limited number of defects that were examined in this program strongly supports SCC as the principal defect mechanism in PCI failures.

These PIE results, supplemented by out-ofreactor work, have suggested directions for remedies to PCI-induced failures. These remedies include: annular fuel pellets, large grain size fuel, and fuel rods incorporating a barrier between the fuel pellets and the inner surface of the cladding. Programs to irradiate lead test assemblies incorporating these new fuelrod designs are being supported by DOE and EPRI. Full assemblies are being tested in commercial power reactors, while smaller numbers of rods will undergo ramping tests in test reactors. The PIE plans call for a nondestructive examination of all rods, with destructive examination of a fraction of these. Only a few rods will be given a very detailed destructive examination. This selective approach to detailed destructive examination is common practice because of the high costs of these examinations.

Extended burnup capability

Current U.S. policies on plutonium recycle in LWRs provide a strong incentive to extend the burnup capability of the UO2-Zircaloy system. Extending core average burnups to \$50,000 MWd/T could result in significant savings in the costs of uranium, separative work, and fuelrod storgae and transportation (ref. 14). Accordingly, current EPRI programs on fuel-assembly surveillance are being extended to achieve discharge burnups of 40-50,000 MWd/T in the 1980-1983 period. Also, DOE currently has in place a number of cooperative programs between fuel vendors and utilities to investigate extending the burnup of LWR fuels. The DOE and EPRI programs include designs that increase the H/U ratio by decreasing the cladding OD or its

thickness and that reduce parasitic materials by replacing luconel grids with Zircaloy. Poolside and hot-cell examinations are an integral part of each program. Plans include the hot-cell PIE of at least one fuel assembly in each program. Of principal interest are data on bundle mechanical stability, fuel rod thermal performance, and correston.

A corollary program in the planning stage is the study of fission-product release from high-burnup fuel. This program is a joint venture among RPRI, DOE, the U.S. reactor vendors, and utilities from a number of countries. Detailed postirradiation characterization of both the gaseous and volatile fission products is planned. In addition, EPRI fuel surveillance projects with M.S. vendors are being expanded to obtain fission-gas release data from fuel assembli's irradiated for four, and even five, cycles.

The performance of gadolinia-containing fuel is receiving increasing attention because of the need to make greater use of burnable poisons when going to higher fuel burnups in PWRs. It is anticipated that one or more programs will soon be in place to investigate the behavior of UO2 containing various concentrations of Gd₂O₃. Because fuel additives can significantly affect fuel and fission-product behavior, these programs should make use of detailed destructive examinations to evaluate possible deleterious effects on fuel thermal performance and fission-product/cladding interactions.

Safety-related programs

The NRC program being carried out at the PBF and the LOFT facility addresses the response of individual fuel rods or small bundles to simulated loss-of-coolant, power-coolant mismatch, and reactivity-initiated accidents. The objective of this work is to confirm that the safety margins implicit in the licensing criteria and the evaluation models specified in regulatory statutes are adequate. Following the tests, the fuel rods are given detailed destructive examination in the INEL hot cells to determine cladding and fuel damage mechanisms. The techniques that have provided most of the data are metallography and posttest diametral measurements. It can be expected that such safety-related programs will be expanded in light of the recent concerns over the safety of light-water reactors.

Special mention should be made of the expected detailed interrogation of the Three Mile Island reactor fuel. As of this writing, no detailed plans have been made for this PIE because the condition of the core is still unknown. It can be said with assurance, however, that this fuel will receive the most intensive examination ever given any fuel in the U.S. Attempts will be made not only to determine the present condition of the core but also to reconstruct the sequence of events that took place during the accident. The evidence suggests many fuel-rod failures and extensive oxidation and embrittlement of the cladding. Emphasis will be

placed on determining the maximum achieved fuel and cladding temperatures, fuel and cladding oxidation, cladding hydriding, fuel restructuring, and fission-product release. The condition of the core will determine the success that can be achieved in the PIE. Problems can be expected in removing the brittle fuel rods from the core and in handling them during the subsequent examinations in the hot cells. The condition of the core has been inferred from interviews with plant operating personnel and analyses of out-of-core and in-core instrument readings. These analyses will help direct the PIE in evaluating the applicability of the analytical models.

FUTURE OF PIE

From the current and planned fuel surveillance and improvement programs, the safety-related programs, and the need to identify the causes of generic fuel failure, it can be seen that PIE is expected to play a vital role in the U.S. LWR industry. This role, however, will be significantly more difficult than in the past because of new problems in the shipping of radioactive materials, the availability of licensed shipping casks, and waste disposal. Recently imposed restrictions on shipping that require approved routes, escorts, and concurrence of local authorities have already caused delays of more than five months in shipping fuel rods from reactors to hot cells. At present the licenses of all LWR shipping casks in the U.S. are undergoing review to determine their conformance to new structural standards. The closing of commercial low-level waste disposal sites is causing waste to back up at the hot cells. Likewise, there are no commercial outlets that will accept examined fuel from the cells. These considerations pose significant problems for hot-cell PIE and will assuredly raise the cost of such examinations. Since hot-cell PIE is already an expensive examination technique, examination of failed fuel is considered only if there is strong evidence that the vendor and utility will incur higher costs or that safety issues may develop if the source of failure is not promptly identified

The current problems associated with hotcell PIE will likely result in greater reliance on poolside examinations. For many programs this could mean the loss of valuable information on the physical and chemical state of the fuel, fission products, and cladding, the study of which requires a controlled examination environment, even in a hot cell. In spite of this, poolside examination techniques must be further developed. Fruitful development areas would be defect detection, precision gamma scanning, fuel imaging, and dry specimen isolation and sectioning.

SUMMARY AND CONCLUSION

Poolside and hot-cell examination of LWR fuel have played a significant role in establishing the response of fuel and cladding to normal and abnormal irradiation conditions. PIE diagnoses of failed fuel rods have led to fuel design changes that have resulted in re-

duced failure rates and increased plant capacity factors. Significant use of PIE techniques is planned in achieving the objectives of fuel-improvement programs now in place and anticipated in the U.S. However, institutional problems besetting the entire nuclear industry could have an especially negative impact on the use of hot cells for detailed postirradiation examinations. This would seriously hamper the development of improved fuels and the achievement of the goals that have been envisioned for nuclear power.

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