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GADOLINIA FUEL PROPERTIES
FOR LWR FUEL SAFETY EVALUATION

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This Supplement to Topical Report XN-NF-79-56, Rev. 1, "Gadolinia Fuel Properties for LWR Fuel Safety Evaluation," provides ENC's responses to the additional information needs transmitted in the NRC's letter (R. L. Tedesco to G. F. Owsley) dated January, 1981. Each NRC identified item is repeated in the text, followed by ENC's response.

REACTOR FUELS SECTION, CORE PERFORMANCE BRANCH

490.1 *(a) Wada, Noro, and Tsukui (Topical Report Ref. 4) noted anomalous microstructure of irradiated* UO_2*-*Gd_2O_3 *as compared with irradiated* UO_2*. To what extent have the microstructures of irradiated Exxon* UO_2*-*Gd_2O_3 *pellets been examined ceramographically for potential anomalies,* Gd_2O_3 *redistribution, etc. (e.g., how many rods were examined, what burnups had they reached, what power levels were the rods operating at, what* Gd_2O_3 *concentrations were in the pellets, and what observations were made)? If the Exxon* UO_2*-*Gd_2O_3 *pellets were to exhibit the anomalies reported by Wada, et al., what potential effects might that have on poison rod performance? To what extent might the vastly different isotopic composition of Gd employed by Wada, et al. (91% Gd-160) compromise the applicability of their results to Exxon* UO_2*-*Gd_2O_3 *poison?*

Wada, et al. reported a lack of columnar grain growth in a 3 w/o gadolinia fuel pin irradiated at high power in the Japanese Materials Test Reactor (JMTR). Also reported was discontinuous grain growth along cracks in the cooler peripheral regions of the fuel pellets. The tendency for gadolinia to retard or prevent columnar grain growth is not a unique finding. For example, Flipot, et al.(1), has reported similar results for urania fuel containing 0.6 w/o dysprosium oxide (Dy_2O_3). A reduction in columnar grain growth is expected to result in lower fission gas release for gadolinia bearing fuel rods if they were to be irradiated at very high power levels. This follows since one mechanism for high fission gas release is postulated to be the liberation of fission gas trapped in the fuel matrix as the expanding boundaries of the columnar grains sweep through the fuel matrix.

An explanation of the discontinuous grain growth feature noted by Wada, et al., cannot be given. It is important to point out that both "anomalies" reported by Wada appear to be associated with the very high power levels that were achieved in the JMTR (centerline temperatures in excess of 2280°C).

These power levels are much higher than will occur in commercial reactors and hence the microstructural features reported by Wada are not expected to occur in practice and as noted below have not been seen in microstructural examinations of ENC gadolinia bearing fuel. Even if high power levels were to occur, the features reported by Wada do not appear to have any significant fuel performance impact other than an expectation of reduced fission gas release as noted above.

The isotopic composition tested by Wada, et al. (91% Gd-160) is not considered a significant factor with respect to the applicability of the data. It was notably low in burnable isotopes, presumably so as not to limit the power that could be achieved. On the other hand, the radial flux depression may have been non-typical considering that this was a test reactor irradiation with 8 w/o U-235 enriched material. High flux depression during the relatively short (28 day) irradiation period would have resulted in higher than normal fuel pellet temperatures in the peripheral regions and a relatively flat radial temperature distribution in the interior of the fuel pellet. Such a temperature distribution could partially account for the microstructural features noted by Wada.

To date, two ENC UO_2-Gd_2O_3 fuel rods have been subject to detailed destructive examination. The rods came from ENC 9x9 assemblies irradiated in the Big Rock Point boiling water reactor. The rods contained 1.0 w/o gadolinia bearing pellets. They attained a peak pellet burnups of 25,000 MWD/MTM and experienced peak power levels of about 12.0 kw/ft. Each rod was cut at a number of axial locations for detailed microstructure examinations. Figures 1.1 and 1.2 show representative photomicrographs from one of the rods. Figure 1.1 is from an axial cut just below the quarter core elevation at

X/L = 0.2, and is a location where peak power was experienced. Figure 1.2 is from mid-core height, X/L = .5, and corresponds to a region of where peak power levels were lower than in Figure 1.1. The microstructure in these figures shows no evidence of the discontinuous grain growth reported by Wada. The only noticeable features in these photographs are the typical cracking patterns and the slight darkening towards the center of Figure 1.1. This darkening marks a region of high temperature sintering and loss of porosity. The rod power level was not high enough to have expected any columnar grain growth.

(b) In general, it appears that post-irradiation examinations (PIE) of high Gd_2O_3 *content (≥5 w/o* Gd_2O_3*), high burnup (≥30,000 MWd/t), high power (≥8 kw/ft at some point during operation) gadolinia-uranium rods may be quite sparse (and may in fact be non-existent). Discuss the type of PIE program that Exxon will conduct for fuels over their full range of application.*

ENC will be monitoring the in-reactor performance of gadolinia-bearing fuel rods at the Prairie Island Unit 2 reactor. Five 14x14 fuel assemblies have been fabricated and will be initially irradiated in Spring 1981. Each of these assemblies has four fuel rods containing gadolinia. The four gadolinia-bearing rods in two of the assemblies have been highly characterized prior to irradiation; detailed data on the fuel pellet, cladding, and fuel rod characteristics have been measured and documented.

The five assemblies will be irradiated for three annual cycles

Visual examination of the assemblies will be performed after each of the first two irradiation cycles. After the third irradiation cycle, when the assemblies are discharged, a detailed examination will be performed on one of the highly characterized assemblies. During this examination, all four of the gadolinia rods and several of the UO_2 rods will be withdrawn from the assembly for non-destructive measurements. The length and diameter of the individual rods will be measured and compared to pre-irradiation values. The length of the fuel pellet stack will be determined by using an eddy current technique to measure the plenum length of each fuel rod. Eddy current measurements along the full length of each fuel rod will also be performed to assess the cladding integrity.

Hot cell examination of representative fuel rods will be considered if the non-destructive poolside inspection suggests the need for destructive examination.

490.2 *(a) Much of the information provided in report Section 3.7 on densification, while interesting, has no direct bearing on the prediction of in-reactor densification behavior of gadolinia-bearing fuel pellets. Thus, while it is true that the "initial sintering test" results presented in the report provide some evidence of the relative sinterability of some* UO_2-Gd_2O_3 *powder mixtures (and may also provide some indirect indication of likely in-reactor behavior), they do not allow one to conclude that "had the initial sintering conditions been adjusted to provide the same initial porosity for the pure* UO_2 *and the* Gd_2O_3 *bearing pellets, . . . then the resinter test would have shown essentially the same or less densification for the gadolinia bearing pellets as compared to the pure* UO_2 *pellets." Inasmuch as (we understand) you already have performed resintering tests on production lot* UO_2 *and* UO_2-Gd_2O_3 *pellets (as called for by Regulatory Guide 1.126), please provide the results for those tests for the* Gd_2O_3 *concentrations under consideration. Those results will show whether your as-fabricated gadolinia fuels densify more or less than your standard fuel.*

Resinter data from gadolinia-bearing pellet lots

are tabulated below. Included are all production lots to date

490.2 (b) *The in-reactor densification results presented in report section 3.7.2 are based on data on ten 1.0 w/o* Gd_2O_3 *and 90 pure* UO_2 *rods. Since it is indicated in the report that gadolinia concentrations up to 5 w/o are to be considered, we assume that Exxon intends to produce, irradiate, and subsequently examine rods containing up to 5%* Gd_2O_3*. And, while it is sufficient to follow the procedures specified in Regulatory Guide 1.126 for conservatively estimating maximum in-reactor densification, we should expect to receive the results of any confirmatory examinations that would be performed on the high* Gd_2O_3 *concentration rods. Such results would be useful for confirming the applicability of Regulatory Guide 1.126 to gadolinia fuels.*

Exxon Nuclear Company plans for post-irradiation examinations of gadolinia bearing fuel rods (as described in 490.1b above) include comparing resinter test data to data for fuel column slump. The post-irradiation data and its correlation with pre-irradiation resinter data will be made available to the NRC.

(c) *As a point of clarification, the anisotropic shrinkage model for axial shrinkage that is provided in Regulatory Guide 1.126 is intended to provide a conservative estimate for axial gap formation and concommitant local power peaking. Thus, half the pellet volume shrinkage is assumed to occur due to the change in pellet length. It is not, however, correct to use that model to infer the magnitude of in-reactor shrinkage from column slump data, except on a relative comparison basis.*

Agree.

POOR ORIGINAL

490.3 *Please provide quantitative data on fission gas releases from gadolinia bearing fuel rods as functions of gadolinia content and burnup, along with comparative results from urania fuel. If such data do not exist for rods with high Gd_2O_3 concentrations (e.g., 5% Gd_2O_3), high linear power levels (up to the licensed limit for such fuels) and high burnups, discuss your rationale for utilizing low Gd_2O_3 concentration, low power, low burnup rod data in the ENC fission gas release model to predict releases for conditions outside of the range of the data.*

Fission gas release data for ENC gadolinia bearing fuel currently exists only for gadolinia concentrations of about 1.0 w/o in BWR fuel assemblies. Fission gas release data for higher gadolinia concentrations will not be available for several years as assemblies with higher gadolinia concentrations have only recently begun irradiation. The available data is from ENC fuel bundles and is summarized in Figures 3.1 to 3.8. Four of the figures provide the rod locations for those rods for which fission gas release was measured. Also provided is a relative rod power at approximately the time in life that the bundle was at its highest power. The other four figures provide the fission gas release versus relative rod power.

The modification to relative rod power of the gadolinia rods has involved the following components which are based on known sensitivities:

Dashed lines in Figures 3.2, 3.4, 3.6 and 3.8 show the specific modifications to relative rod power that were made.

Figures 3.2, 3.4, 3.6 and 3.8 all show a common trend of no fission gas below a threshold power level. Above this threshold power level, fission gas release goes up sharply with increased power. The sharp increase in fission gas release above the threshold power level is due to the intercoupling between fission-gas-release and fuel temperature in which a small amount of fission gas release leads to a significant reduction in pellet-to-clad gap conductance for a non-prepressurized (BWR) fuel rod which, in turn, results in higher fuel temperatures and further increases in fission gas release. As shown by the trend curve in Figure 3.2, once the pellet-to-clad gap conductance has been degraded, the sensitivity of fission gas release to additional power increases is reduced.

The fuel temperatures reached by some of the rods are considered consistent with license limits of operation for present designs in view of the fact that the rods experienced power levels of about 12.0 kw/ft and had degraded gap conductance because of fission gas release. The results given in Figures 3.2, 3.4, 3.6 and 3.8 do not indicate any unusual fission gas release characteristics for the gadolinia rods other than the normal temperature dependence of release as found in urania fuel rods.

No unusual release characteristics are expected for higher gadolinia concentrations. In this regard it is noted that Manzel and Dorr report low fission gas release comparable to urania fuel for gadolinia concentrations up to 4 w/o. Further, as noted in our response to 490.1, at very high power levels gadolinia is expected to limit columnar grain growth which should reduce fission gas release.

With respect to fission gas release, it is important to note that gadolinia fuel pins employed in present fuel assembly designs are not limiting factors in accident and related safety analyses and would not be even if they had somewhat higher fission gas release than they presently are thought to have.

This enrichment reduction is to offset the reduced thermal conductivity of gadolinia bearing pellets relative to urania pellets so that at high exposure after depletion of the burnable gadolinia isotopes, both gadolinia and urania rods will have comparable pellet temperatures. The reduction in enrichment also insures that at high exposure the gadolinia rods will operate at lower linear heat generation rates than the urania rods in the fuel assembly. The reduced power for gadolinia rods makes them clearly non-limiting both in ECCS and plant transient licensing analyses.

490.4 *(a) How are sintered pellets tested for homogeneity?*

Homogeneity is determined by chemical analysis Homogeneity is tested to either about the nominal specified content for the pellet lot, whichever is greater.

(b) The statement is made that homogeneities which might remain in fabricated pellets should tend to disappear with increasing radiation. This presumes mobility of the gadolinium species. But reference is also made to the observed lack of migration of Gd due to a temperature gradient, which presumes a lack of mobility of the Gd species. How are these statements reconciled?

The statements in question refer to two different scales of observation. On a microscopic scale thermally activated intermolecular diffusion between adjacent gadolinia and urania particles in fabricated fuel pellets appears to take place over distances As a result of this diffusion, the concentration gradient (i.e., the driving force) between gadolinia and urania regions is reduced.

On a macroscopic scale the gadolinia concentration remains uniform under irradiation. More specifically, the temperature gradient from the centerline to the surface of a fuel pellet does not result in either higher or lower gadolinia concentrations at the pellet surface than at the pellet centerline.

490.5 *It is stated that the transient melting of small, fully contained Gd-rich dispersions will be of no significant consequence. What is the likelihood of such dispersions occurring at pellet-cladding contacts, and is transient melting in this instance also of no significance? What would be the calculated potential maximum pellet volume expansion and concommitant cladding plastic strain if the* Gd_2O_3*-rich dispersions were fully molten during a hypothetical transient?*

Since solid solution forms at the interface of adjacent urania and gadolinia particles during sintering, the size of gadolinia rich dispersions in fabricated fuel pellets can be expected to be smaller than the size of the individual gadolinia powder particles that are mechanically blended with urania powder in the pellet fabrication process. The maximum size of gadolinia powder particles is limited

After sintering, the size of any gadolinia rich dispersions is probably limited

Such dispersions would be distributed uniformly throughout the fuel and thus could occur at the pellet-clad interface. The melting point of these small dispersions might be as low as 2300°C versus a bulk material melting point typically at some point in the range of 2700°C to 2800°C.

Generally, for transients the peak power rod does not experience boiling transition and peak temperatures occur at the fuel centerline. Maximum temperatures near the pellet-clad interface are on the order of 600°C or less. Thus, localized melting (if it occurs at all) would be limited to central regions of the pellet where it should be of no consequence.

Pellet volumetric expansion with localized melting of gadolinia-rich dispersions is estimated on the following basis:

This level of linear expansion is not considered significant relative to clad strain, particularly since much of the expansion of molten material could be expected to fill intergranular cracks and other porosity within the fuel column rather than result in clad expansion, and since for the hypothetical transients the macroscopic region would be limited to a small fraction of the fuel about the fuel centerline.

490.6 *Curtis and Johnson (J. Am. Chem. Soc. __40__, 15 (1957)) indicate that Gd_2O_3 forms solid solutions with ZrO_2. Discuss the chemical compatibility of UO_2-5 w/o Gd_2O_3 with Zircaloy cladding at elevated temperatures. Have post-irradiation examinations been made to demonstrate the compatibility of gadolinia-bearing UO_2 with Zircaloy cladding?*

Sintered urania or urania/gadolinia fuel pellets are refractory materials in character and thus are relatively inert both physically and chemically. The paper by Manzel and Dorr shows that ceramic activity (sintering) in these materials begins to appear at 700-800°C in the initial heating of unsintered pellets. With sintering, however, the temperatures required for any further ceramic activity of significance become much higher. Thus, inducement of further sintering or densification in resinter tests for example requires temperatures of 1500°C or more.

Zirconium dioxide is likewise a refractory material, and the tendency for formation of solid solutions between it and gadolinia bearing fuel pellets will only occur at very high temperatures. Rouanet and Foex(2) have provided a phase diagram for the Gd_2O_3/ZrO_2 system that quantifies the degree of solubility between these materials. The temperatures at which the solid solutions were achieved, however, were typically above 1800°C. For temperatures of 1500°C and lower, little solid solution formation or other ceramic activity would be expected between ZrO_2 and sintered gadolinia-bearing fuel pellets.

During reactor operations, cladding temperatures remain below 500°C. Thus, no solid solution formation between gadolinia bearing fuel pellets and the clad would be expected. The destructive examination of ENC gadolinia bearing fuel rods from the Big Rock Point reactor (see Figures 1 and 2 in 490.1a) likewise does not indicate any incompatibility or other deleterious ceramic reaction between zircaloy clad and gadolinia bearing fuel pellets.

490.7 *Discuss the hydration, water corrosion, and disintegration resistance of gadolinia-urania fuel pellets under conditions akin to cladding rupture in an operating environment.*

The excellent corrosion resistance in water of urania (UO_2) and its chemical compatibility with cladding materials is one of the principal reasons UO_2 was selected as the fuel for LWRs(3,4). Gadolinia Gd_2O_3 is a similar refractory oxide with a high degree of physical and chemical inertness as noted in the response to 490.6. Curtis and Johnson(5) have found no reaction of sintered Gd_2O_3 with boiling water. As reported in Reference 6, other studies(7) by the Argonne National Laboratory have shown no reaction of gadolinia bearing fuel (up to 6 w/o) to water at 360°C and 2750 psi or to steam at 750°F and 1250 psi.

490.8 *What changes in fuel management procedures are required for routine use of gadolinia-urania fuel, and what safety significance might such changes have, if any?*

The routine use of gadolinia-urania fuel offers additional flexibility with respect to fuel management plans. However, the basic fuel management procedures and neutronic analysis requirements are unaffected by the inclusion of gadolinia bearing fuel. The gadolinia is modeled in the analyses and is therefore explicitly included in safety evaluations.

490.9 *Use of gadolinia fuel could be accompanied by modifications in control rod absorber content, control rod positioning, rod worths, and/or coolant boron concentrations. Discuss the potential implications of such changes on the relative severity of reactivity and power anomaly-type events such as control rod withdrawals and misoperations, control rod ejection (PWR) and rod drop (BWR), boron dilution, startup of idle loop, etc.*

A cycle specific safety evaluation is performed for each reload. The presence of the gadolinia is explicitly accounted for in the evaluation as are any accompanying effects on the reactivity control mechanisms. The results of these analyses demonstrate the safety margins expected with the inclusion of gadolinia bearing fuel.

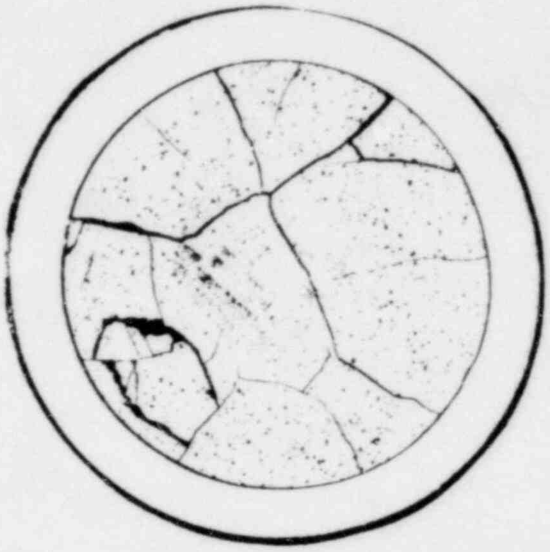

Figure 1.1 Gadolinia Rod Photomicrograph, X/L = .2

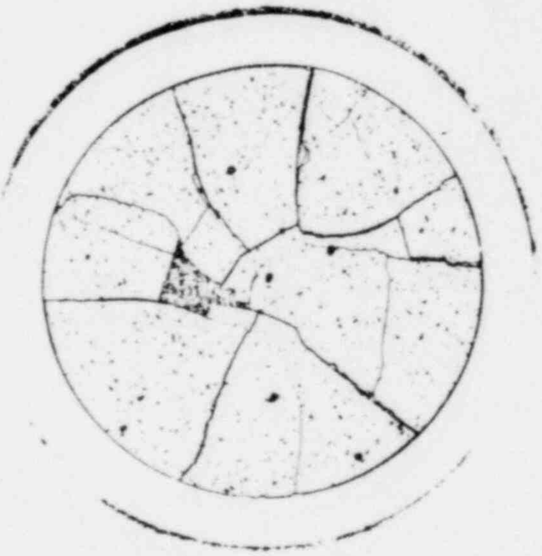

Figure 1.2 Gadolinia Rod Photomicrograph, X/L = .5

Figure 3.1 Fission Gas Release and Power for Assembly UD000A

Release Fraction, %

Relative Rod Power

Figure 3.2 Fission Gas Release versus Rod Power - Assembly UD000A

Figure 3.3 Fission Gas Release and Power for Assembly UD0008R

Release Fraction, %

Relative Rod Power

Figure 3.4 Fission Gas Release versus Rod Power - Assembly UD0008R

Figure 3.5 Fission Gas Release and Power for Assembly D-71

Release Fraction, %

Relative Rod Power

Figure 3.6 Fission Gas Release versus Rod Power - Assembly D-71

Figure 3.7 Fission Gas Release and Power for Assembly D-72

Release Fraction, %

Relative Rod Power

Figure 3.8 Fission Gas Release versus Rod Power - Assembly D-72

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