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for Detecting Failure of Cladding in
Encapsulated Fast Reactor Fuel Pins

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THE USE OF HIGH-RESOLUTION GAMMA-RAY SPECTROMETRY
FOR DETECTING FAILURE OF CLADDING IN ENCAPSULATED
FAST REACTOR FUEL PINS*

by

J. R. Phillips, J. W. Schulte, and G. R. Waterbury

ABSTRACT

A nondestructive technique utilizing precision gamma-ray scanning has been developed for detecting fuel cladding failure in doubly encapsulated fast reactor fuel elements. Detection of fission products, in particular ^{137}Cs , outside the fuel pin envelope was used to indicate the failure. The method was tested by examining eleven mixed uranium-plutonium carbide and two mixed uranium-plutonium oxide fuel elements. Gamma-ray scanning indicated that six of the thirteen fuel elements had failed. These results were confirmed by mass spectrometric analysis of the gases in the outer encapsulation which were found to contain 11.7 to 65.0% xenon and 1.76 to 9.31% krypton. The cover gas from a seventh pin contained only 0.016% xenon and < 0.003% krypton, and the presence of ^{137}Cs outside the cladding was not conclusively detected by gamma-ray scanning. Cesium-137 was not found in the outer encapsulation of the six other fuel elements by gamma-ray scanning, and mass spectrometric analyses confirmed that the cladding had not failed.

INTRODUCTION

A definite need existed for nondestructive detection of failed cladding in doubly encapsulated fast reactor fuel elements. Extended periods of irradiation at very high neutron fluxes can cause swelling of the fuel and cladding, interactions of the fuel and fission products with the cladding, and progressive deterioration. Failure of the fuel cladding exposes the exterior cladding to fuel and fission products and increases the probability of fuel leakage into the surrounding reactor coolant. In helium-bonded mixed-oxide fuel pins, sodium may enter the pin from the capsule and react with the uranium and oxygen in the fuel to form a volumi-

nous product that can cause massive failure of the inner cladding. In the fuels development program, doubly encapsulated experimental fuel elements often are irradiated for several irradiation cycles and then given an interim examination in which nondestructive testing techniques, such as neutron radiography and dimensioning, are used to assess the condition of the fuel elements. Some fuel elements are re-inserted in the reactor to accumulate additional exposure and burnup. During the interim examination, positive identification of fuel pins in which the inner cladding is breached would aid significantly in the selection of pins for re-insertion.

Various diagnostic techniques including neutron and x-ray radiography as well as eddy current testing have been studied in attempts to detect

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failures in the pin cladding. Massive failure in fuel pins has been detected with neutron and beta-tron radiography. Although radiographic images can be improved by computer and photographic enhancement techniques, radiography probably cannot be made sufficiently sensitive to reveal small breaches in the cladding. As precision gamma-ray scanning determines distributions of individual isotopes at accurately measured locations along the length of the fuel elements,⁽¹⁾ this technique was promising for detecting volatile fission products or gas tracers escaping from the inner encapsulation into the outer capsule. Preliminary tests showed that detection of ^{137}Cs outside the fuel pin envelope reliably indicated breaching of a fuel pin and was a more sensitive test than measurement of ^{85}Kr . Efforts to locate other fission products outside the fuel pin or to consider the addition of a tracer gas were suspended at this time, and the reliability of the detection of ^{137}Cs as positive identification of breached pins was tested.

Theory

Fission products may escape from the fuel envelope to the coolant in the outer encapsulation either through minor cladding failures that permit the escape of gaseous and volatile fission products, or major cladding failures which permit the coolant to wash-out some of the non-volatile fission products.⁽²⁾ The gaseous and volatile fission products would move from the coolant into the gas plenum. The distribution of these fission products between the cover gas and the coolant would be dependent upon the distribution coefficients of the individual elements, the escape and re-entry probabilities, and the area of the interface. Once the nuclides were in the cover gas, they may remain in the gas, as do the isotopes of xenon and krypton, or they may adsorb onto an acceptable surface as does ^{137}Cs . The adsorption of fission products onto a surface is proportional to the numbers of unsaturated sites available and the probability of adsorption and release for the particular isotopes. The non-volatile fission products would remain in the coolant until they adsorbed on structural material of the fuel element or precipitated, generally at the coolest locations.

To be suitable as a monitor to detect fuel

cladding failure, a fission product must have certain physical and chemical characteristics. It should be gaseous or highly volatile at the temperature encountered during irradiation to facilitate the movement through minor (pinhole) failures as well as gross cladding failures. It should also have a sufficiently long half-life to allow time for examination, an easily resolved gamma-ray spectrum, and a reasonably high fission yield. These four characteristics are exhibited by ^{137}Cs . Its relatively long half-life of 30 years assures adequate gamma activity after extended periods of cooling, and it decays 93.5% of the time with a 661.6 KeV gamma photon⁽³⁾ which is in an energy region of the gamma-ray spectrum that is not complicated by other gamma rays from the irradiated fuels. A typical gamma spectrum of irradiated fuel taken with the anticoincidence Ge(Li) system used in these examinations (Figure 1) shows that the nearest significant photopeaks are at 605 KeV for ^{134}Cs and 765 KeV for ^{95}Nb which cause no interference as the gamma-scanning system has a resolution of 2.7 KeV. The fission yields for ^{137}Cs is 6.0% from uranium and 6.5% from plutonium. The ^{137}Cs isotope readily condenses on colder parts of the capsule structural material, thereby concentrating the gamma activity to make detection easier.

Use of the gaseous fission product ^{85}Kr also was considered because it was chemically inert, but its fission yields were too low, 1.3% from uranium and 0.54% from plutonium. The ^{85}Kr isotope emits a 514 KeV gamma ray 0.41% of the time.⁽³⁾ This particular region of the gamma spectrum has appreciable interference from annihilation gamma rays (511 KeV). Some method of concentrating the ^{85}Kr , such as freezing, would be necessary if this isotope were used as a monitor.

Method

The irradiated fuel element is positioned in the mechanical scanner in front of a set of precision gamma-ray collimators that defines the volume of the fuel element from which the unattenuated gamma rays may interact with the detector. The scanner has five degrees of freedom to allow scanning any type of sample; X, Y, Z, rotation, and tilt. The collimated gamma rays interact with an anticoincidence detector assembly consisting of

Figure 1. Typical Gamma-Ray Spectrum of End-plug of a Failed Irradiated Fuel Element.

a 50 cc Ge(Li) detector with 2.7 KeV resolution and 9.4% efficiency, surrounded by a NaI(Tl) annulus 8 in. in outside diameter, 2.5 in. in inside diameter and 12 in. long. (See Figure 2). A complete spectrum is accumulated for a preset time at each position. The spectrum is written on magnetic tape with two tagwords that record the time of day and the position of the fuel element. The scanner indexes a specified increment to a new position and another spectrum is accumulated. This continues until spectra at positions over the region of interest have been recorded and stored on magnetic tape.

The magnetic tapes containing spectra and position data are processed by computer programs to determine the area under the full energy photo-peaks of specific isotopes. There are two basic computer codes used for data analysis depending upon the complexity of the spectra. The areas may be determined by summing the total counts between a lower and upper limit and subtracting an average background. This method of analysis is satisfactory for spectra that do not have appreciable interference from other gamma peaks. In more complex spectra, the areas are determined by an iterative method that uses a summation function of analytical functions.⁽⁴⁾ In either case, isotopic distributions are obtained as a function of position

along the fuel element. The location of the cladding, capsule, and other structural material can be determined by examining the activation products, ^{58}Co , ^{54}Mn , and ^{60}Co , that have prominent gamma rays of 812 KeV, 835 KeV, and 1332 KeV, respectively. Once the position of the inner fuel envelope has been established, the detection of any fission product outside the envelope indicates that the inner cladding has been breached.

DISCUSSION AND RESULTS

During the past year at Los Alamos Scientific Laboratory, the integrity of the fuel cladding of each of thirteen fuel elements, containing either helium- or sodium-bonded mixed carbide or helium-bonded mixed oxide, was determined by gamma scanning. Following gamma scanning, the cover gas in the capsule outside the cladded fuel was analyzed by mass spectrometry. The results obtained from gamma scanning and cover gas analysis are compared in Table I.

The method appears to be equally applicable to helium-bonded or sodium-bonded fuel elements. In the six fuel elements in which ^{137}Cs was detected outside the cladding, two were helium-bonded and four were sodium-bonded.

The first fuel element in which ^{137}Cs was detected outside the cladding was a helium-bonded mixed uranium-plutonium oxide ($\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_2$), that had an accumulated burnup of 57,200 MWD/

Figure 2. Schematic Diagram of Gamma Scanning System.

TABLE I

Comparison of Gamma Scanning and Mass Spectrometric
Detection of Failed Fuel Elements

Fuel	Number of elements	Fission gases found in cover gas, atom %		^{137}Cs detected outside fuel cladding
		Krypton	Xenon	
$(\text{U}_{0.8}\text{Pu}_{0.2})\text{O}_2$	1	0.0	0.0	no
$(\text{U}_{0.8}\text{Pu}_{0.2})\text{O}_2$	1	9.31	65.0	yes
$(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$	5	1.76-5.46	11.8-34.7	yes
$(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$	5	0.0	0.0	no
$(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$	1	< 0.003	0.016	no

Tonne. The fuel pellets were contained in 0.015-in.-thick, 316 L, stainless steel cladding which was wrapped spirally with a 0.036-in.-diameter spacer wire. The fuel cladding was bonded to the inner surface of the capsule container by sodium.

On a routine axial scan ^{137}Cs was found to have deposited about one inch below the bottom of the fuel cladding. A diametral scan was taken at this position. A plot of the gross gamma activity, Figure 3, shows spikes at 2.645 in. and 2.678 in. on the arbitrary scale in this figure. These spikes were subsequently identified as the 661.6 Kev gamma ray of ^{137}Cs that was concentrated at the two locations. The outer surface of the fuel clad-

ding was located at 2.630 in. on the arbitrary scale. The ^{137}Cs deposits were located outside the fuel cladding by at least 0.015 in. which was much larger than the ± 0.003 -in. deviations in the measurements. Also, the spikes were separated by 0.033 ± 0.003 in. which corresponded to the diameter of the spiral spacer wire. These results indicated that the ^{137}Cs may have deposited on the surface of the 0.036-in. diameter spacer wire and appeared as a ring source viewed from a point perpendic-

Figure 3. Diametral Gamma Scan of Irradiated Fuel Pin in Non-Fueled Region.

Figure 4. Axial Distributions of ^{60}Co and ^{137}Cs in Failed Fuel element.

ular to the axis. The mass spectrometric analysis of the cover gas from this capsule showed that there were 9.31% krypton and 65.0% xenon present, thereby confirming the gamma scanning evidence.

Other regions of fuel elements were inspected to determine if the presence of ^{137}Cs could be reliably determined with a single scan. Because the cesium isotopes tend to migrate to the cooler regions, it was postulated that the inner surface of the capsule end plug would be the logical place to look for the deposition of ^{137}Cs . Generally, the capsule end plug was about 8 in. above the top of the fuel cladding. The presence of ^{137}Cs on the inner surface of the end plug would be indisputable evidence that the fuel cladding was breached. The area near the capsule end plug would be easy to gamma scan because interference from gamma emitting fission products in the fuel would be minimized.

The distributions of ^{137}Cs and ^{60}Co over the region of the capsule end plug of a mixed uranium-plutonium carbide fuel element, Figure 4, showed the relative positions of the structural material of the capsule, from the gamma activity of the activation product ^{60}Co , and of the ^{137}Cs . The ^{137}Cs apparently was deposited on the inner surface of the capsule end plug, 7.75 in. above the fuel cladding. These spatial distributions clearly indicated that the cladding failed, permitting fission products, in particular ^{137}Cs , to escape from the fuel pin envelope. The cover gas was found later to contain 4.61% krypton and 29.0% xenon, thus confirming that the inner cladding had been breached.

Conclusions

This technique may be used in the interim examination to screen experimental fuel elements and to assess the condition of the fuel pin cladding. If ^{137}Cs is found outside the fuel cladding, the fuel pin is breached, and it may not be advisable to return this pin to the reactor to accumulate additional exposure. The experience to date indicates that positive detection of ^{137}Cs outside the fuel pin proper will always be confirmed by the presence of fission gases in this same space. This latter condition is always associated with cracks or pinholes

in the pin cladding. The threshold for detecting a failed fuel pin by precision gamma-ray scanning is not known at this time. The method appears to be an effective, non-destructive screening process for detecting failed inner cladding in encapsulated fast reactor fuel pins.

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