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MEASUREMENT OF FISSION PRODUCT LEAKAGE FROM FUEL ELEMENTS STORED IN WATER

D. W. Rhodes

MASTER

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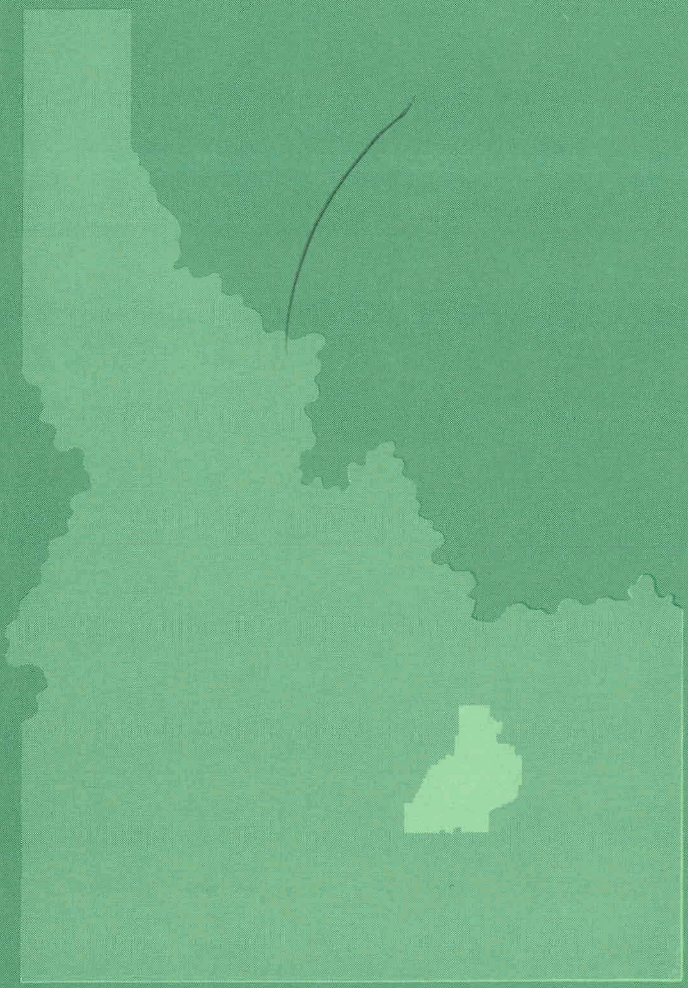

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ABSTRACT

A method for detecting cladding defects in fuel elements stored underwater by measuring the rate of increase in the concentration of radioisotopes in the water is described. The only special equipment needed is a stainless steel container with compressed air and water connections and conventional beta counting equipment. The method can be used for any type of solid fuel that is suitable for underwater storage.

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TABLE OF CONTENTS

LIST OF FIGURES

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I. SUMMARY

The identification of fuel elements with cladding defects which allow water to penetrate to the irradiated uranium alloy is essential to prevent excessive fission product contamination of storage basin water. Detection of fission product leakage from faulty fuel elements stored at the Idaho Chemical Processing Plant (ICPP) was accomplished by placing suspect fuel elements in a closed stainless steel cylinder containing uncontaminated water. The measured rate of increase in the fission product concentration of the water indicated the rate at which fission products were being leached from the fuel elements.

The leakage rate was measured for various uranium-aluminum alloy fuel elements clad with aluminum. The rate of fission product release from all defective fuel elements tested was found to be constant for at least 300 hours. A fuel element that was cut to deliberately expose 10 cm^2 of the irradiated uranium-aluminum alloy had a measured leakage rate of 335 μCi/hr. The measured background rate for the stainless steel cylinder and apparently undamaged fuel was 0.5 μCi/hr. The measured leakage rate of fuel considered to have minor cladding defects fell between these two values. This method of leak testing permitted the early identification of fuel elements with damaged cladding so that contamination of storage basin water was minimized.

II. INTRODUCTION

During the interim period between the discharge of fuel elements from a reactor and the recovery of uranium from the fuel, it is common practice in the atomic energy industry to store the fuel elements under water. Underwater storage is an economical and efficient means of providing both shielding and cooling of the irradiated fuel during the storage interval, which may be as long as several years.

Due to the wide variety and large number of fuel elements that are stored in the ICPP fuel storage basin, occasionally the cladding of one or more elements has been penetrated inadvertently by corrosion or by mechanical handling during the removal and subsequent transfer of the fuel to the storage basin, thus allowing fission products to leach from the irradiated fuel. The presence of fission products, particularly long-lived ones, in the water is undesirable because: (1) the exterior of the transfer casks, which often are shipped off-site, may become contaminated during the underwater loading and unloading operations, and (2) water that is discharged from the basin as a part of the corrosion control program must be decontaminated before it can be released to the environs. To detect fuel with defective cladding, a Fuel Element Leak Tester (FELT) was designed and tested at the ICPP.

III. EQUIPMENT

The FELT consisted essentially of a stainless steel cylinder, large enough to contain several fuel elements, with a stainless steel plate welded to the bottom and a flanged top that was covered by a hinged, stainless steel lid. An air- and water-tight seal was obtained by an O-ring recessed in the flange with swing bolts that clamped the hinged lid tightly to the flange. Flexible (polyethylene) tubing was attached through Swagelok fittings near the top on one side of the cylinder and near the bottom on the other side. Water or air was transported through this flexible tubing as needed. A cable was attached to arm-like projections on opposite sides of the cylinder to serve as a lifting bail for the entire apparatus. The weight of the cylinder, including the bottom and the lid, was such that the cylinder remained on the bottom of the basin even when all of the water was displaced by air. A schematic diagram of the FELT is shown in Figure 1 and a photograph of the unit is shown in Figure 2.

IV. OPERATION

The FELT was placed on the bottom of the fuel storage basin under about 20 feet of water. Raw water treated to contain about 0.5 ppm residual chlorine to inhibit microbiological growth and with sodium nitrate added to provide about 200 ppm nitrate to inhibit corrosion of fuel elements is used in the storage basin at the ICPP. The "suspect" fuel elements were placed in the unit by using underwater manipulators. The lid was closed and secured tightly with the swing bolts that were attached

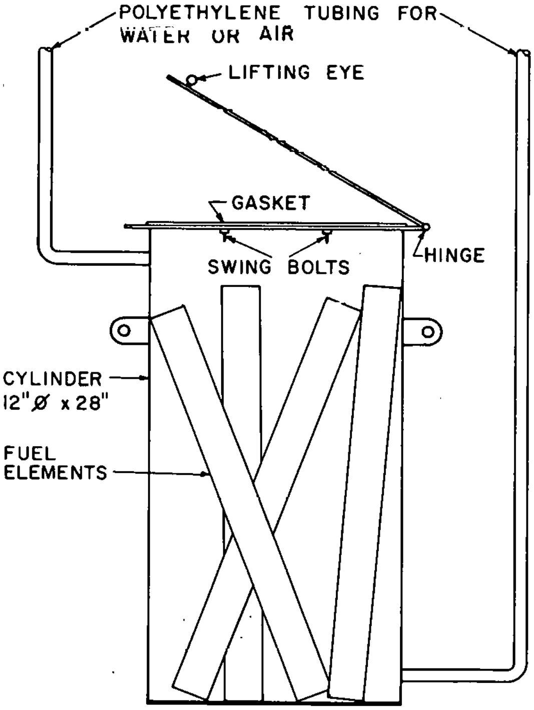

Fig. 1 Fuel Element Leak Tester (FELT)

Fig. 2 Fuel Element Leak Tester Vessel

to the cylinder flange. A fresh water purge was accomplished by adding fresh raw water through tubing connected near the bottom of the FELT to displace the contaminated basin water, which flowed out through the tubing connected near the top. Water samples were collected from the overflow and analyzed for total beta emitters. The purge was continued until the total beta emitter concentration became negligible or attained a constant value over several hours of sampling. The water purge was then discontinued and the fuel elements were allowed to stand in the water for several days. Water samples were removed from the FELT periodically by applying an air pressure of about 12 psig through the top connection and collecting a water sample as it flowed from the tubing attached to the bottom connection. A suitable interval was allowed to purge the water from the flexible line before taking the sample. An air sparge to agitate and thoroughly mix the water before sampling was tried, but no significant difference was obtained between sparged and unsparged samples. With the relatively long sampling intervals used, diffusion and convection apparently were sufficient to mix the water. Each sample had a volume of approximately 250 ml; since the total volume of water in the FELT was about 57 liters, the change in volume due to sampling was not considered to be significant, and no water was added to replace the samples.

The concentration of total fission products in the water samples was measured in a gas-flow proportional counter, and samples were also analyzed radiochemically to identify the fission products.

V. EXPERIMENTAL RESULTS

Four freshly stored aluminum-uranium alloy fuel elements from the Brookhaven Graphite Research Reactor (BGRR) were placed in the FELT and a leak test was conducted for about 400 hours. Curve I, shown in Figure 3, represents the results of the analysis of the water samples from this test. Initially, the concentration of total beta-emitters in the water was well below the background concentration of the basin water but increased slowly with time. This increase in fission product concentration with time may be partially due to the desorption of fission products from the walls of the FELT and the surface of the fuel, but the approximately constant increase in the quantity of fission products with time (0.5 μCi/hr) suggests that either a slow leak was occurring from the uranium alloy or that the cladding contained some activation products, resulting from the irradiation of impurities, which were being leached.

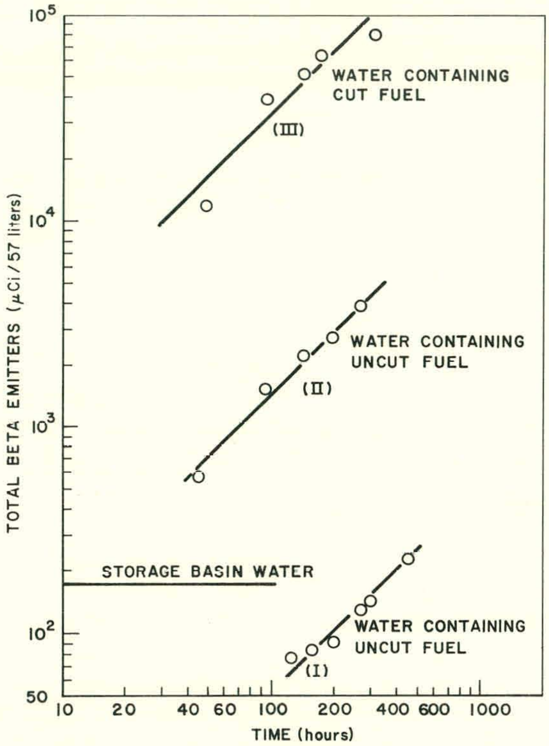

Fig. 3 Fission Product Leakage from Aluminum-Clad Fuel Elements (BGRR) Stored in Water

Curve II (Figure 3) represents four more elements of the same type which were obtained from a group of elements that were suspected to be leaking. The cladding very likely had been penetrated on at least one or more of these elements because the data, as indicated by a comparison of Curve II with Curve I, suggest that fission product leaching was occurring.

Curve III (Figure 3) gives data for the same four elements that were used to obtain the data for Curve II after each element had been cut into two sections on a cross section perpendicular to the long axis of the fuel. The uranium alloy area exposed by this cutting was about 10 cm^2. Using the known exposed surface area and the leaching data represented by Curve III, the indicated leach rate for Curve III was calculated to be about 35 μCi/(hr)(cm^2) of exposed uranium-aluminum alloy. With this value as a control, the apparent exposed uranium alloy area of this type of fuel can be estimated from a leach rate measurement. Thus, the leach rate of about 15 μCi/hr for the uncut fuel, represented by Curve II, indicates an apparent exposed uranium-aluminum alloy surface area of about 0.5 cm^2. Whether the break that produces the fission product leakage is due to mechanical handling, corrosion pitting, etc. can only be determined by more detailed hot cell tests; however, merely to know that an element or group of elements is susceptible to leaching is usually sufficient. The defective elements can then be processed immediately or encased in a secondary container, if the fission product leakage is detrimental to the storage system.

All curves approximated the general power form $y = ax^b$; however, the approximate slope of one for the curves in Figure 3 suggests that the movement of the fission product ions away from the metal surface is controlled by the mechanisms releasing the ion from the metal alloy and not by an equilibrium with the ions already in solution. Similar tests with other types of fuel elements produced similar curves but with different slopes. For example, Figure 4 shows the curves obtained with fuel elements of two different ages from the Materials Testing Reactor (MTR) and from three Sodium Graphite Experiment (SGE) capsules.

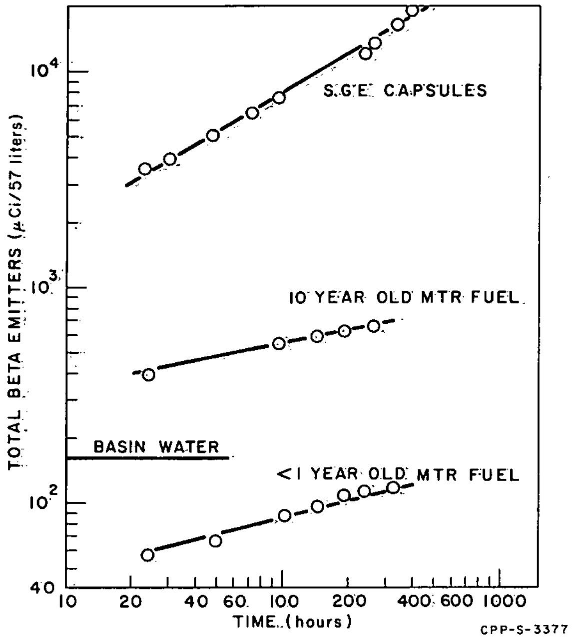

Fig. 4 Fission Product Leakage from MTR and SGE Fuel

Four of the MTR elements had been stored in the basin about 10 years and another four elements slightly less than one year; the SGE capsules had been in the basin about seven years. At least one of the SGE capsules was an obvious potential leaker as indicated by the fact that gas bubbles emanated from the capsule when it was loaded into the FELT. The 10-year-old MTR elements appeared to be leaking slowly, although because of the long time these elements had been stored in the basin water, the results may represent predominantly desorption of fission products from the fuel surfaces.

Radiochemical analysis of contaminated water from the FELT indicated that the measured radioactivity was essentially all due to the long-lived fission products, cesium-137 and strontium-90. Water contaminated by the cut fuel also contained about 8×10^{-5} g/l of uranium.

VI. CONCLUSIONS

The operation of this Fuel Element Leak Tester unit was both convenient and efficient. Very little time was required for the actual testing and no shielded facilities other than the water normally present in the storage basin was required. The equipment can be used to test nearly any type of solid fuel for defects; fuels of unusual dimensions or shapes can be accomodated by using a cylinder of the proper size for the fuel of interest.

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