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A CLADDING FAILURE MONITOR FOR LIQUID METAL-COOLED REACTOR SYSTEMS

MASTER

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I. INTRODUCTION

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In the event of a fuel element cladding failure, minute quantities of fission products are released directly to the coolant. If the coolant is water or a gas such as helium or carbon dioxide, the failure may be annunciated by an activity monitor installed at some downstream location. Interference from activities generated in such coolants is usually either of minor significance or can be satisfactorily reduced by relatively simple methods of discrimination.¹

If the coolant is sodium or a sodium-potassium alloy (NaK), the problem of annunciation is more complicated, since the monitor must be capable of detecting a small fission product signal in the presence of an overriding background originating from the decay of Na^{24} which is generated continuously in the coolant. The quality of information afforded by any conceivable device designed to separate a true fission product signal from an overriding Na^{24} activity is certain to be questionable.

The necessity of blanketing the primary coolant in a fast reactor (usually Na or NaK) with an inert gas such as argon, however, suggests an interesting possibility, since it is known that rare gas fission products (Kr and Xe) diffuse rapidly into the cover gas system.² In principle, some suitable beta-gamma detector which monitors the activity of the cover gas should be capable of annunciating a cladding failure. In practice, it is necessary to effect an almost complete discrimination against A^{41} , which is present in the cover gas at activity levels many orders of magnitude higher than those associated with credible concentrations of rare gas fission products.

A simple and extremely efficient method of discrimination against A^{41} has been developed, a method which utilizes the selective electrostatic fixation of

rare gas daughter nuclides.³ The principle of the fixation process is premised on the fact that at the instant of beta decay, each of the daughter species becomes a positively charged ion. Accordingly, cover gas is passed through an electrostatic precipitation unit which consists primarily of a cylinder maintained at a potential of 2500 volts positive. A closed-loop wire driven axially through the cylinder is maintained at ground. Upon beta decay, the ionized daughter species migrate to the wire and become electronically neutralized. The wire carrying the neutralized daughters passes through a mercury trap-type gas seal which has a few milliliters of water floating on its surface. The strongly electropositive Rb and Cs daughters react instantly with the water and become permanently fixed in the water layer in the hydroxide form. K^{41} , the daughter of A^{41} , also reacts with the water; but since K^{41} is radioactively inert, the activity associated with the water fraction is almost exclusively that arising from the decay of various Rb and Cs fission products. It follows that the output from a detector which monitors the activity level of the water fraction may be used as a criterion of the fission product concentration in the cover gas.

II. EQUIPMENT

The principal item of interest is the electrostatic precipitation chamber illustrated schematically in Fig. 1. The unit consists essentially of two concentric cylinders: an inner, 2 in. in diameter, of aluminum; and an outer, 4 in. in diameter, of stainless steel. Running axially through the inner cylinder (the anode) is a 6-mil, closed-loop, stainless steel wire driven at a speed of approximately 18 in. per minute by a 3 rpm Bodine induction motor. High voltage (2500 volts positive) is supplied to the anode through a conventional Teflon-insulated high-voltage fitting. Plexiglass end caps, not illustrated, insulate

the anode from the outer cylinder. The remainder of the precipitation unit, including the collection wire, is maintained at ground potential.

Cover gas under 5 psi and flowing at a rate of approximately 100 ml/min is admitted to the chamber through 1/4-in. copper tubing and is exhausted at near atmospheric pressure to the reactor stack system. Cover gas enters and leaves the cylindrical anode through 1/4-in. holes drilled at opposite ends. Short circuit gas flow between inlet and outlet ports is prevented by a Plexi-glass baffle plate.

To avoid the leakage of active cover gas to the surroundings from the wire inlet and outlet ports, two mercury seals in the form of U-tubes are used. One of these, along with other components, is illustrated photographically in Fig. 2; the other, the downstream trap, is located behind lead shielding bricks. Floating on the surface of the mercury in the upstream leg of the downstream trap is a layer of water approximately 1 in. in depth. The water, through its chemical reaction with the Rb and Cs daughters, serves two purposes: to integrate the activity with respect to time, and to decontaminate the wire continuously. A system of pulleys in each U-tube enables the moving wire to negotiate the necessary bends. Two idler pulleys supply tension to the wire.

To prevent active cover gas from diffusing into the downstream trap, the region between the water layer and the precipitation unit is flushed continuously under positive pressure with uncontaminated argon from the reactor cover gas feed system. Flowmeters and valves installed in the flush and active cover gas inlet lines serve as a means of flow control.

The detector used to monitor the various activities collected in the water fraction is a 2 x 2-in. NaI (Tl activated) crystal optically coupled to a Dumont 8219 photomultiplier tube. To reduce the effects of radiations emitted from the reactor and from the precipitation chamber, the crystal and photomultiplier

tube assembly are enclosed in a cylindrical lead shield approximately 4 in. in thickness.

During earlier stages of the tests, the output from the photomultiplier tube was fed to a Nuclear Data 256 channel transistorized pulse height analyzer equipped with both photo-oscillographic and digital readouts. Periodic measurements with Cs¹³⁷ and Co⁶⁰ sources were used to establish the energy calibration of the spectrometer.

In later stages of the tests, after the feasibility of the method was established, the spectrometer was replaced with more conventional equipment. Pulses from the photomultiplier tube were amplified by a linear amplifier and fed to a conventional count rate meter. A block diagram of the various components is given in Fig. 3.

III. EXPERIMENTAL RESULTS

The testing of the equipment in EBR-I was complicated by the fact that under normal operating conditions, substantial quantities of rare gas fission products are released continuously to the cover gas from the following sources: (1) vented blanket thermocouple rods, and (2) a small, unknown, uranium contamination in the NaK coolant.⁴ Obviously, the natural existence of fission products in the system reduces the sensitivity and the effectiveness of the system. On the other hand, the continuous release of fission products from these sources provided an opportunity to evaluate the performance of the equipment under realistic conditions of fission product release.

A. Discrimination Against A⁴¹

In a system such as the one described above, the possibility exists that A⁴¹ may be carried over into the downstream (monitored) trap, either through diffusion, sorption into the stainless steel wire, or through some unsuspected

mechanism. To test for this possibility and to assess the degree of discrimination against A^{41} , 0.1 ml of water was taken from the trap after 2 hours of full-power operation and analyzed with a Nuclear Data 256 pulse height analyzer. The results are given in Fig. 4. Each of the gamma peaks may be identified with radiations associated with specific isotopes of Rb and Cs. From the spectrum, it is clear that the 1.29 Mev gamma of A^{41} is almost completely missing. In contrast, a pulse height analysis of an active cover gas sample (not illustrated) reveals the existence of a single peak at 1.29 Mev. Radiations from rare gas fission products and their respective daughters, while present, are completely masked by the much higher activity associated with A^{41} . It is clear from these considerations that the degree of discrimination against A^{41} is satisfactorily high.

As the spectrum of Fig. 4 indicates, essentially all of the activity collected in the water fraction is the result of decay in three fission product species, i.e., Rb^{88} , Rb^{89} , and Cs^{138} . The half lives of each of these species are short, being respectively 18, 15, and 32 minutes. In addition, the respective half lives of their precursors are also short, i.e., 2.8 hr, 3.2 min, and 32 minutes. It follows that the decay of precursors and daughters will be essentially complete for shutdown periods longer than a few hours. During a startup following a prolonged shutdown -- overnight, for example -- it is clear that the activity level of the water fraction will increase rapidly from zero and will eventually reach equilibrium at a time dictated by the half lives of the rare gas precursors and their respective daughters. The time base along which saturation proceeds is the subject of a later discussion.

B. Background Components

To evaluate the feasibility of differentiating the effects of fission product release during a cladding failure from those associated with

normal release, it was necessary to separate the saturated normal response into the following components: (1) environmental background, (2) reactor background, (3) A^{41} background, and (4) the normal Rb-Cs signal. The first component was evaluated from measurements conducted with the reactor off. The second component was evaluated from measurements conducted with the reactor on, but with anode voltage and cover gas flow off. The difference between these two components gives the reactor background. The third component was separated from measurements carried out with the reactor operating and cover gas and seal gas flowing, but with the anode voltage off. The difference between these results and the sum of the environmental and reactor backgrounds gives the effect originating from A^{41} contained in the precipitation unit. The final component was established through measurements conducted with all systems operating. The difference between these results and the sum of all other background effects gives the normal Rb-Cs signal. A comparison of the relative intensities of the various components is given in Table I.

Table I

INTENSITY OF BACKGROUND COMPONENTS IN EBR-I, Mark IV

<u>Component</u>	<u>Source</u>	<u>Intensity</u>
1	Environmental	4 c/sec
2	Reactor	36
3	A^{41}	0
4	Rb-Cs	23
Total	All sources	63

From this information, it may be seen that the ratio of normal Rb-Cs activity to the sum of all other background components is approximately 0.58. Similar measurements carried out with a previous loading (Mark III)⁵ resulted in a ratio value of 8.0. This striking difference reflects the removal of 12 vented thermocouple rods which released approximately 14-fold greater quantities

of fission products to the coolant.

C. Simulated Fuel Rod Failure

To obtain some realistic measure of the time response and the strength of the signal resulting from a fuel rod cladding failure, a simulation experiment was carried out with the Mark IV loading. Twenty unclad enriched uranium foils, each 1 x 1 cm, were packed into a well^{Nak}ventilated basket which in turn was installed near the edge of the core in a^{Nak}ventilated thimble. The combined area of the twenty foils was chosen to coincide with the exposed area of fuel in a typical Mark IV fuel rod.

The results of measurements conducted at 900 kw (full power, 1200 kw) are illustrated in Fig. 5, which gives the activity associated with the water layer as a function of time after reaching operating power. The lower curve, obtained under normal operating conditions, gives a true measure of the background level. As expected, the activity, extremely low shortly after startup, increases rapidly and eventually reaches saturation approximately 3 hours after startup. Repeat measurements conducted on three successive days demonstrated the reproducibility of the results.

The upper curve of Fig. 5 gives the results of measurements carried out with the unclad enriched uranium foils in position and with all other conditions unchanged. Hence, for any given time the intensity difference between the two curves gives a measure of the fission product release from the unclad foils. To permit a reasonable evaluation of the effect expected from the failure of a single Mark IV fuel rod, corrections for the difference between Pu^{239} and U^{235} fission cross sections have been applied to the data illustrated.

From the results of Fig. 5, it is clear that the strength of the fission product signal from the simulated failure is approximately five times that of the normal background signal. It is reasonable to conclude from the

results of this somewhat idealized, yet still credible, experiment that the release of fission products from a Mark IV fuel rod cladding failure would be readily detectable.

In conclusion, it seems clear that the charged-wire cover gas monitoring principle may find useful application in other sodium-cooled fast reactor systems.

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REFERENCES

1. R. L. Heath, Fission Product Monitoring in Reactor Coolant Stream, IDO-16213, 1956
2. Private Communication, R. O. Haroldsen, July 1, 1963
3. "Catching Burst Slugs", Engineering, pp 599-600, April 28, 1961
4. R. O. Haroldsen, et al, Hazards Summary Report, EBR-I Mark IV, ANL-6411, September, 1961
5. R. E. Rice, et al, EBR-I Mark III Design, ANL-5836, March, 1958

FIG. 1
PRECIPITATION CHAMBER
FOR
CLADDING FAILURE MONITOR

FIG. 3
BLOCK DIAGRAM
CLADDING FAILURE MONITOR

Fig. 4

FIG. 5
FUEL ROD FAILURE SIMULATION
EBR-I, MARK-IV

END