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REACTOR IN-CORE REGENERATIVE
NEUTRON DETECTORS INTERIM
DEVELOPMENT REPORT

C. N. Jackson, Jr.

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INTERIM DEVELOPMENT REPORT

By

C. N. Jackson, Jr.

Instrumentation Section
Applied Physics and Electronics Department

October 1967

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INTRODUCTION

Rapid burnout of nuclear reactor in-core neutron flux detectors at high thermal flux levels and other limitations of existing detectors are well recognized. A candidate technique for considerably extending the useful, in-core lifetime of neutron sensing detectors is the employment of regenerative materials within an ionization chamber, for example. The regenerative material comprises both fertile and fissile atoms, where the fissile atoms cause generation of an electrical signal proportional to the thermal neutron flux level. In this method, appropriate combinations of isotopes, ^{234}U and ^{235}U in this case, are used such that the fertile atoms transmute, by absorption of thermal neutrons, to fissile atoms at approximately the same rate as the fissile material depletes. Thus, long-term detection sensitivity remains relatively constant for extended nuclear reactor in-core exposure periods.

This report describes nuclear reactor in-core investigations at high thermal neutron flux levels being conducted under the sponsorship of the United States Atomic Energy Commission Division of Reactor Development and Technology. These experiments are determining the operational and life-test characteristics of developmental, regenerative neutron flux detectors utilizing an appropriate mixture of ^{234}U and ^{235}U as the coating material within the detector. With such in-core detectors, desirable optimization of reactor performance can be achieved by continuously monitoring the detector output in order to provide useful data concerning the spatial distribution and level of neutron flux throughout the reactor core. The obtained flux information yields detailed

knowledge of core conditions and thus allows the fuel elements to be irradiated at an optimum rate with assurance that critical material design limitations will not be exceeded. Economy in reactor operating costs and improved operational safety can thus be obtained by using in-core flux monitoring.

Currently, ^{235}U fission chambers and self-powered rhodium detectors⁽¹⁾ are two typical devices utilized for in-core neutron flux monitoring. The fission chamber suffers from an excessively rapid burnup characteristic which results in a significant reduction in output signal strength during a reasonable life expectancy. With ^{235}U as a target material, 10% of the emitter atoms are burned up in 17 days at a possible commercial reactor thermal neutron flux level of 1×10^{14} nv. Relative sensitivity may significantly change during one reactor operating cycle, thus requiring frequent replacement of detectors. Compared with fission chambers, the electrical signal strength of the rhodium device is smaller and correspondingly more difficult to use.

Earlier regenerative detector reports⁽²⁻⁴⁾ describe initial phases of this program, including the initial test where two experimental detectors were first installed in-core in a graphite-moderated, light water cooled Hanford production reactor. For these regenerative detectors, the optimum concentrations for the two isotopes were determined to be 90.1% ^{234}U and 9.9% ^{235}U by a calculational experimental technique⁽⁵⁾ involving computer analysis. Formulation of the emitter material was matched to the in-core requirements. Extended life and operational characteristics of these regenerative chambers observed during the subsequent sustained in-core testing are described and analyzed in this report, and technical details are presented for both the detector and capsule design. Future plans are presented for a second in-core test capsule that will feature a more comprehensive test package,

which includes several types of neutron detectors in order to fully demonstrate, on a comparative basis, the performance of this regenerative detector concept.

SUMMARY

In order to circumvent the problems of rapid performance degradation and other limitations of nuclear reactor in-core neutron flux detectors operating at high thermal neutron flux levels, development of regenerative ^{234}U - ^{235}U detectors is being conducted. The sustained, relatively constant sensitivity of two experimental detectors, inserted within the core of a Hanford plutonium production reactor, demonstrated that extension by a factor greater than ten in the useful in-core life can be achieved with a regenerative emitter material as compared to a typical fission chamber. The sensitivity remained within essentially 10% of a median value throughout an exposure of 5×10^{21} nvt for a calculated optimum regenerative emitter formulation of about 10% ^{235}U and 90% ^{234}U . This performance is equivalent to more than ten years operation in a possible nuclear commercial reactor having a neutron flux level of 1×10^{13} nv. Typical of fission chambers, these experimental devices exhibited a robust signal strength, prompt response, and a dynamic range of about four decades. The detector sensitivity was about 1×10^{-16} A per nv for thermal neutron and about 8×10^{-13} A per R/hr for gammas with a chamber sensitive volume one inch in diameter by one inch in length. Noted reductions in chamber dielectric characteristics were attributed to deficiencies in the test chamber vehicle.

To demonstrate the practicality of these regenerative chambers for use in the nuclear industry and to overcome the dielectric problems, a second test capsule has been prepared using commercially fabricated detectors. This capsule will

be irradiated in the ETR at the National Reactor Test Site in a thermal neutron flux level of about 4×10^{15} nv. A comprehensive test package, this capsule includes ten individual detectors and will permit comparative performance evaluation of regenerative, ^{235}U , gamma and rhodium beta current devices. The test capsule features chambers of nearly identical design, differing principally in the material contained within the detectors.

DISCUSSION

REGENERATIVE MATERIAL DESIGN FACTORS

The optimum regenerative mixture of about 90% ^{234}U and 10% ^{235}U utilized in the experimental samples was determined by a MIN of INT computer program.⁽⁵⁾ In this calculational method, the isotopic concentrations were optimized to achieve a maximum "useful" lifetime during which the sensitivity remains within 10% of a median value.

Of four potential fertile isotopes, shown in Table I, ^{234}U exhibits the most desirable characteristics for use in a regenerative chamber. Briefly reviewing the characteristics, the ^{232}Th and ^{238}U ⁽⁶⁾ both have undesirable long half-life delay factors and the ^{240}Pu with a relatively high cross section would burnup too rapidly. The ^{234}U has desirable prompt response and an effective burnup cross section of about 100 b that will achieve long life characteristics.

CHAMBER MECHANICAL CONFIGURATION

The detector chamber was designed along conventional fission chamber lines with significant emphasis being placed on the achievement of a maximum neutron to gamma signal ratio. The quantity of fissile emitter coating determines the neutron sensitivity; it is limited ultimately by the self-absorption experienced by the fission recoil products. The regenerative

^{235}U - ^{234}U emitter coating was plated by electro-chemical deposition on the inner surface of the emitter cylinder. A relatively heavy coating of regenerative material was applied to the emitter to achieve a high neutron sensitivity and a correspondingly high neutron to gamma signal ratio. This high signal ratio was reflected in the large dynamic range of five decades observed in the neutron sensitivity of the detectors during early phases of the testing. Gradually, the neutron dynamic range dropped to about four decades as the residual gamma background built up in the chamber due to exposure.

TABLE I. *Comparative Characteristics of Possible Isotope Combinations for Regenerative Thermal Neutron Flux Detector*

Fertile Isotope	Half Life, Days	Fissile Isotope	Cross Section, b	
			Capture	Fission
^{232}Th	27	^{233}U	~ 10	~ 500
^{234}U		^{235}U	~ 100	~ 600
^{238}U	2-1/2	^{239}Pu	~ 10	~ 750
^{240}Pu		^{241}Pu	~ 1000	~ 950

Detector sensitivity and the details of interelectrode spacing and detector configuration including cable and electrical connectors are shown in Figure 1. The active chamber volume is an annular section about 0.015 in. thick, 0.89 in. diameter, and 1 in. long, formed by concentric coaxial cylinders supported at each end by ceramic insulators. A limiting factor in the closeness of interelectrode spacing to reduce chamber volume and corresponding gamma sensitivity is posed by voltage breakdown limits. A polarizing potential of plus 200 V and a 0.015 in. spacing is required for efficient charge collection in the chamber.

Neutron Sensitivity - 1×10^{-16} a/nv Nominal
Neutron Flux Range - 10^9 to 10^{14} n/s
Gamma Sensitivity - 8×10^{-13} a/hr Nominal
Operating Voltage - 100 to 200 VDC
Temperature Range - 0 to 40 °C
Pressure Maximum - 500 psig
Insulation Resistance - 10^{10} ohms Min at 100 VDC and 70 °C

Cable Sheath - AISI 304 Stainless Steel
Chamber Shell - AISI 304 Stainless Steel
Cable Insulation - Alumina
Chamber Insulation - Alumina
Filling Gas - Argon at 91.5 cm Mercury
Cathode - Uranium Coating {0.2% U²³⁵, 89.8% U²³⁴ - 18.2 cm² Active Area}
Active Volume - Annular Region 0.015 in. by 0.89 in. Diam

Fission Chamber in Cross Section

NOTE: Dimensions are in Inches
Unless Noted Otherwise

FIGURE 1. Design Details of Experimental In-Core Regenerative Neutron Chamber
(Sensitive coating material is a uranium isotope mixture in which ^{234}U transmutes to ^{235}U to sustain signal strength during in-core exposures to large thermal neutron fluence.)

TEST CAPSULE

The two regenerative detectors were mounted in a water cooled test capsule as shown in Figure 2. The capsule featured relatively simple configuration and used proven structural materials as identified. The capsule contains provision for the insertion of a dry cobalt aluminum flux wire for measuring neutron particle fluence and estimating flux levels.

This long slender assembly penetrates the reactor core and locates the test samples in a flat flux region of the core. Numerous support blocks and relatively rugged construction are required in-core to position and hold the experimental chamber firmly and restrain possible motion caused by flow of cooling water. This test capsule firmly positioned the two samples side by side in the reactor core throughout the course of the investigation.

TEST CIRCUITS

At the start of the tests, the electrical output signals of both chambers were continuously recorded on a strip chart recorder. As shown in the earlier report,⁽²⁾ a range switch selected the signal developing resistors to match the increases or decreases in the signal output over a four decade current range. Periodically, about twice a month, chamber plateau characteristics and insulation resistance values were measured on both chambers. By using an x-y recorder, a complete saturation curve (I-V curve) was machine drawn in a few minutes.

The details of the I-V test circuits for accurately and efficiently accumulating this performance data are shown in Figure 3. An x-y recorder simultaneously plots the detector voltage on the abscissa and the corresponding signal current

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FIGURE 2. *Regenerative Chamber Test Capsule (This is an instrumented test capsule in which the leads from two chambers are brought out from the reactor core. The integral chamber and lead assemblies are positioned firmly in place by aluminum supports and flow-holes are provided for cooling water. The 0.025 in. dry traverse tube permits ready access for insertion of flux measuring wires.)*

9

Neg. PNL-0670909-7

FIGURE 3. Instrumentation Circuit for IV Plateau Curves

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output on the ordinate. A polarizing potential from zero to plus 400 volts is supplied by an adjustable power supply that is highly isolated from circuit ground to prevent leakage current error. Good circuit grounding practices were used to isolate the signal current from possible error-inducing, circulating ground currents. A 4000 Ω current limiting resistor is placed in series with the chamber to protect the chambers against possible short circuits.

EXPERIMENTAL RESULTS

BURNUP PERFORMANCE

Constancy of the neutron output signal and comparative theoretical performance characteristics of both a regenerative and a ^{235}U coated chamber as a function of increasing nvt are shown in Figure 4. This signal output data was normalized against reactor coolant temperature change to correct for variations in reactor power level. Despite this correction a sizable spread ($\sim \pm 15\%$) in individual data points occurred. This spread is attributed to unavoidable changes in reactor neutron flux level caused by shifts or oscillations in the flux profile. The regenerative chamber characteristic is a fitted curve (least squares fit) over the $20\text{--}70 \times 10^{20} \text{ n cm}^{-2}$ range, and the intervals shown are the 95% confidence limits on the fitted linear curve. A mathematical test for linearity indicated that no significant departure from the simple relation occurred. Over this exposure range the sensitivity (average of both test samples) decreased with a slope of $-0.373 \pm 0.049\%$ per $1 \times 10^{20} \text{ n cm}^{-2}$. The anomalous decrease in signal current at the start of the exposure, though similar in character to ^{235}U burnup, has not been satisfactorily accounted for and is thought to be not representative to the optimum 10% ^{234}U -90% ^{235}U regenerative emitter. Compared to the ^{235}U curve, the ^{234}U - ^{235}U regenerating chamber characteristic demonstrates a substantially improve constancy of signal output to achieve at least tenfold

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FIGURE 4. Effect of In-core Exposure on Chamber Sensitivity

improvement in useful life. An exposure of 5×10^{21} nvt is equivalent to ten years operation in a commercial reactor at a flux level of 1×10^{13} nv.

CHAMBER DIELECTRIC

Both regenerative chamber samples experienced a signal strength decrease during the first 1.5×10^{21} fluence of the neutron exposure as well as severe degradation of insulation resistance ($\sim 3X$ reduction) as shown in Table II. Coupled with the resistance decrease a dielectric breakdown occurred with polarization voltages over a range of 100 to 400 Vdc. Interestingly, there occurred a periodic restoration of dielectric strength during each reactor shutdown and by an exposure of 5×10^{21} nvt the dielectric strength had been partially restored, as shown in Figure 5. Restoration occurred to the extent that both chamber samples were capable of operation with up to 200 Vdc polarizing potential without a dielectric breakdown occurring. We consider this anomalous behavior to be not typical of the regenerative emitter material and not typical of a fission chamber vehicle of this type. The quality of the seal between the integral cable and chamber is under question and possible migration of gas from the cable mineral insulation (MgO) into the chamber volume proper is postulated as a cause of this anomalous behavior.

PLATEAU CHARACTERISTICS

Degradation in insulation resistance is also evident in Figure 6. The two upper curves (obtained after substantial in-core exposure) exhibit a high slope characteristic above 100 Vdc indicating a poor chamber dielectric condition.

TYPICAL SIGNAL PERFORMANCE

Typical chamber performance, measuring neutron flux level during transient and steady state operation of the nuclear

FIGURE 5. *Effect of In-core Exposure on Regenerative Chamber Plateau Characteristics (The curves are numbered to indicate time sequence with "0" being the initial detector characteristics. A neutron exposure of about 1×10^{21} fluence was incurred between each successive curve. The plateau characteristics tended to stabilize as shown after an exposure of 2×10^{21} fluence)*

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FIGURE 6. Typical Saturation Curves

reactor, are shown in Figure 7. The recorded signals exhibit prompt proportionate sensitivity for both regenerative chambers.

TABLE II. Effects of Sustained In-Core Exposure on Electrical Resistance of Regenerative Neutron Fission Chamber

Exposure, nvt	Relative Flux Level	Sample #1, Ω	Sample #2, Ω
0	0*	1.5×10^{11}	1.1×10^{11}
0	0	2×10^{10}	1.4×10^{10}
1.2×10^{21}	0	1.1×10^9	1.1×10^9
0.1×10^{21}	1	3×10^5	7×10^4
1.5×10^{21}	1	8×10^4	2.5×10^4
3×10^{21}	1	6×10^4	2.2×10^4
5×10^{21}	1	6×10^4	2.2×10^4
3×10^{21}	0.4	3×10^5	1×10^5
7×10^{21}	1	6×10^4	2.5×10^4

* Out of reactor

ADVANCED EXPERIMENTAL REGENERATIVE NEUTRON CHAMBERS

Preparations are established for a second test capsule that includes a comprehensive package of ten devices. This composite assembly includes four types of chambers: regenerative, a standard ^{235}U , an uncoated gamma unit, and two rhodium type devices. Except for the rhodium self-powered units, all chambers are similar in configuration. The type or omission of sensitive emitter material is the principal difference. All chamber configurations are representative of designs that

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FIGURE 7. Typical Regeneration Chamber
Recorded Signal Output.

can be obtained from leading detector manufactureres in the nuclear industry. The dimensions of the in-core fission chambers are 0.25 in. (or less) diameter by 3.5 in. (or less) in length, and the regenerative chambers incorporate the optimum regenerative mixture of 10% ^{235}U and 90% ^{234}U , as did the previous experimental chambers.

To accelerate the in-core life testing of this new capsule, plans are established and approvals granted to conduct the irradiation at the Engineering Test Reactor (ETR). This capsule, irradiated in a neutron flux of 5×10^{14} nv, will provide an excellent source of accelerated comparative performance data. Hermetic seals are incorporated between the integral cables and chamber assemblies to prevent gas migration into the sensitive chamber volume. These prototype units will provide improved statistical data for establishing the performance of the regenerative emitter material and also provide needed additional information about chamber insulation and dielectric characteristics. Concurrent measurements of all ten chambers will be made for neutron sensitivity, response time, and dynamic range. This composite test arrangement will provide an excellent comparison of performance characteristics for the various types of chambers operating under nearly identical in-core conditions.

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