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MASTER

FISSION PRODUCT BEHAVIOR DURING OPERATION OF THE SECOND PEACH BOTTOM CORE*

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ABSTRACT

The Peach Bottom high-temperature, gas-cooled reactor began operation June 1, 1967 and continued power production until October 9, 1969, accumulating 452 equivalent full power days (EFPD) operation. After reload, power production with Core 2 began July 14, 1970 and terminated October 31, 1974 after 899 EFPD operation.

Surveillance of fission product release and behavior was intensified during Core 2 operation to permit a wider range of measurements to be made. In addition to monitoring the fuel element purge system and noble gas content of the coolant circuit, the program was extended to include measurements of radioactive and other condensible species (including dust)

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entering and exiting the core and steam generator, and of surface concentrations of gamma-emitting nuclides deposited on the primary coolant surfaces. These data, which were obtained over the operating period April, 1971-October, 1974, are summarized and discussed. The data demonstrate that cesium behavior in the coolant circuit during the first two-thirds of Core 2 life was primarily governed by cesium released during Core 1 operation. The data also indicate that whereas the steam generator surfaces attenuate molecular cesium concentrations in the coolant, the dust-borne component is remarkably persistent.

Driver fuel elements were removed from the reactor after 384 EFPD, 701 EFPD, and at end of life. These fuel elements are at various stages of an intensive post-irradiation examination. Some of the axial and radial concentration profiles of fission products which have been obtained are likewise presented. Although these profiles indicate varied fission product behavior, the observations can in general be qualitatively described on the basis of the operational histories of the fuel elements.

INTRODUCTION

The Peach Bottom high-temperature, gas-cooled nuclear reactor (HTGR) is a 40-MW(e) power station which was placed in commercial operation on June 1, 1967. The reactor operated until October, 1969 with its initial core loading, accumulating 452 equivalent full power days (EFPD) operation, and was then shut down for core changeout. Power production resumed on July 14, 1970, and continued with Core 2 through October 31, 1974. During this period Core 2 experienced 899 EFPD operation.

Surveillance of fission product behavior in the Peach Bottom reactor was intensified during Core 2 operation. Coolant sampling probes were emplaced in the Loop 1 circuit to sample and characterize the helium coolant impurities at entrance and exit locations of the steam generator, and monitoring stations were established to measure γ-emitting fission products which had become deposited on the Loop 1 primary coolant circuit surfaces. A layout of the reactor plant which identifies the locations of the coolant sample probes and γ-activity monitoring stations is presented in Figure 1.

The coolant sample probes were designed to withdraw samples of the helium coolant from the Loop 1 circuit under isokinetic conditions, separate the molecular and particulate coolant impurity species, and classify the particulates with respect to size. Although the sample probes were in place throughout a given reactor operating cycle, coolant sampling was actually performed only toward the end of the cycle, to minimize dust accumulation (which could have rendered the probes inoperable) and to make the sample time correspond as closely as possible to the end of cycle time at which the sample probes were removed.

When practicable, the γ-scanning stations which are identified in Figure 1 were monitored during reactor shutdowns with a Ge(Li)

detector whose shield was designed to transmit gamma-rays from only a limited area.

In addition, selected driver fuel elements of Core 2 were removed after 384 EFPD and 701 EFPD operation and subjected to extensive post-irradiation examination (PIE), to determine the extent and nature of fission product transport within the fuel element. Moreover, five driver fuel elements of the 804 elements which comprised Core 2 at end-of-life have also been selected for extensive PIE; each of these five driver elements has experienced 899 EFPD operation.

FISSION PRODUCTS IN THE COOLANT CIRCUIT

The sample probes to monitor impurities in the helium coolant at the entrance and exit sides of the Loop 1 steam generator were operated in May 1973 and in May 1974. Concentrations of I-131, Cs-134, Cs-136, and Cs-137 in the helium coolant as deduced from the sample probe determinations are listed in Table I. For each nuclide it was in general possible to differentiate between a molecular form of the nuclide and a form associated with particulate matter. This was accomplished by analyzing the characteristic deposition profiles which were observed in the diffusion tube sections of the probes, and from γ-scans of the activity associated with the particulates which were collected on impactor plates and filters in the probes. In addition, the sample probes also contained special adsorber traps for the collection of iodine species which could penetrate both the diffusion tube and cascade impactor regions of the probes. The iodine collected in these traps, which is designated as "penetrating iodine," is believed to be organic in nature, probably primarily methyl iodide.

Analysis of the May, 1973 sample probe data is complicated by two factors, the relatively large releases of cesium which were experienced during Core 1 operation, and the relatively large quantities of debris which were in the coolant circuit at that time.

Because the deposition of large amounts of iodine associated with particulate matter was observed in the diffusion tube portions of the May, 1973 sample probes, as was evidenced by a nearly uniform deposition pattern, it was not possible to unequivocally decouple the contribution from particulate matter from an apparently smaller, exponentially decreasing concentration profile which is characteristic of the deposition of molecular species. On the other hand, the special adsorber traps in the probes both upstream and downstream of the steam generator were found to contain a penetrating iodine species which we tentatively identify as organic iodide. The organic iodide species was also observed in both of the May, 1974 sample probes; moreover, at this time it was also possible to differentiate between molecular and dust-associated iodine.

Both the source and the behavior of the organic iodide species remain unresolved. The May, 1973 results suggest that the organic

iodide is formed in the steam generator and destroyed in the reactor core, but neither conjecture is supported by the May, 1974 data. Since all of the sample probes operated at similar ambient temperatures, it appears unlikely that organic iodide generation occurred within these devices; however, it is not unreasonable for some of the penetrating species to be distributed among the molecular and dust-associated iodine forms as well, so that the amounts collected on the sorber traps represent only minimum values.

Like the iodine, significant perturbations are observed in the May, 1973 cesium data due to the relatively large dust-associated fractions which were present. Nonetheless, it is evident that much of the cesium in the coolant stream is re-entrained material which had been deposited on the coolant circuit surfaces during Core 1 operation. The activity ratio of the two isotopes Cs-137/Cs-134 is 2.6 overall for species collected in May, 1973 in the upstream probe, and 3.6 for species collected in the downstream probe. In contrast, the corresponding results from the May, 1974 probes are 0.66 and 0.67, respectively. Moreover, most of the cesium associated with Core 1 release appears to be dust-borne, and it is tempting to conclude that cesium re-entrainment arises from abrasion of the surface by dust particles or by spallation of surface coatings.

Unlike the molecular cesium form, which appears to experience significant deposition on the steam generator surfaces, dust-borne cesium seems to be remarkably persistent. Thus, for example, the May, 1973 probe data suggest that the steam generator surfaces function as cesium sources rather than as sinks. This may in fact be partly correct, as is evidenced by the much smaller attenuation in molecular cesium concentration on passage through the steam generator in 1973 as compared with 1974, and the increase in dust-borne cesium concentration downstream of the steam generator in 1973.

The May, 1974 sample probe data indicate a loss of approximately 90% of molecular cesium to steam generator surfaces, and deposition of about 70% of molecular iodine. Almost negligible amounts of iodine were associated with particulates in the 1974 determinations, whereas persistence of the dust-borne cesium component decreases net cesium loss to steam generator surfaces to about 70% of entrance inventory.

In addition to the sample probe data taken in May, 1973 and in May, 1974, coolant sampling upstream of the steam generator was performed in April, 1971, and sampling downstream of the steam generator also occurred in January, 1972. Interpretation of the results was somewhat obscured by the sampling procedures, and in Table II the Cs-137/Cs-134 ratios that are presented are differentiated in terms of Cs species found on the diffusion tubes and on the filters rather than as molecular and particulate forms.

Other significant sampling events which have transpired in the course of the Peach Bottom Reactor fission product surveillance program are likewise identified in Table II; these involved determinations of radio-

active species on the particulate debris collected by the dust collectors (Dust), γ-spectrometer determinations of activity deposited on Loop 1 coolant circuit surfaces (Large Duct), and fuel element PIEs. Also presented in Table II are values of the Cs-137/Cs-134 ratio which were determined as part of the surveillance effort.

The effect of cesium released during Core 1 operation is evident from a consideration of the Cs-137/Cs-134 ratios which are given in Table II. If it is assumed that the dust which was collected at the end of Core 1 operation, at 452 EFPD, had a Cs-137/Cs-134 distribution which was typical of the cesium deposited on the coolant circuit surfaces, then decay of the two nuclides, with no release from Core 2, would yield a value of 2.9 for the Cs-137/Cs-134 ratio on April 24, 1971. This value is in excellent agreement with the measurements made on the dust in the sample probe, cesium deposited on the coolant circuit surfaces, and dust removed from the dust collectors. Similarly, by January 6, 1972 one would anticipate an increase in the ratio to 3.8; this is only slightly larger than the 3.4 value which was determined from the dust collected on the sample probe filters and external γ-scans of the Loop 1 coolant circuit.

In contrast, breakthrough of Core 2 cesium is obvious in the September 14, 1973 scans of the coolant circuit.

FISSION PRODUCT DISTRIBUTIONS IN FUEL ELEMENTS

A schematic representation of a Peach Bottom HTGR fuel element is presented in Figure 2. Although primary coolant flow is upward in the core, a small purge stream is diverted through the top of the individual fuel elements and made to flow in the downward direction inside the element. In the course of its downward passage, the purge flow sweeps past both surfaces of the cylindrical fuel compacts, through a charcoal fission product trap at the base of the fuel element, and thence to an external cleanup system.

A post-irradiation program has been developed to examine fission product distributions in a selected number of these driver fuel elements. The detailed examination of one of these elements has been completed; this element, which is designated E06-01, experienced 384 EFPD operation. More limited data have been obtained to date on fission product distributions in fuel element E11-07, which was removed after 701 EFPD, and an end-of-life element, E14-01.

In assessing the extent of cesium release from the fuel compacts, it is convenient to mitigate the effects of fluence by comparing the Cs concentrations in the fuel with the corresponding values for Ce-144, since cerium is immobile at nominal fuel temperatures. Thus, for example, in Figure 3, values of the Cs-137/Ce-144 ratio for each of the thirty compacts comprising fuel element E06-01 are presented graphically as a function of position (compact number) within the element. (Compact number 30 is at the top of the fuel element as positioned within

the reactor; the purge flow is thus in the direction from compact number 30 to compact number 1.) Also presented in the figure are preliminary estimates of the average temperature distributions at the inner (ID) and outer (OD) diameters of the fuel compacts; more refined, but still preliminary estimates suggest that the temperatures presented in Figure 3 may be too high by as much as 100-150°C.

As is readily apparent in the data displayed in Figure 3, no discernible redistribution of cesium occurred within the E06-01 fuel compact stack, partly because of the time at power, but more likely because of the temperatures involved.

Some cesium release from the fuel compacts had in fact occurred in fuel element E06-01, as is evidenced by the radial concentration profiles of Cs-134 and Cs-137 which were obtained in the spine and sleeve graphite at compact position number 16. These profiles are displayed in Figure 4. That these profiles result from release of cesium from the E06-01 compacts rather than from re-entrained cesium from Core 1 releases is readily demonstrated. If the latter possibility applied, the profiles in the sleeve graphite would be parabolic in shape, since nearly identical cesium concentrations would prevail at both the inner and outer surfaces of the sleeve. Moreover, the Cs-137/Cs-134 activity ratio in both the spine and the sleeve varies about the value 1.1; this value is more in accord with the value of 1.4 which was determined for E06-01 fuel rather than the 3.4 result which was found for deposited cesium.

The profile data of Figure 4 also serve to establish that transport of cesium through this sleeve graphite is due to the cesium itself, and not to the corresponding xenon precursors. This is evident from the similarity in profile shape as contrasted with the orders of magnitude difference in half-lives of the two relatively short-lived xenon precursors involved. Moreover, detailed calculations of predicted sleeve inventories of the two cesium isotopes as calculated on the premise of xenon transport through the sleeve are orders of magnitude smaller than that observed.

The observed axial distribution of ^{3}H activity in the spine graphite of fuel element E06-01 is compared with corresponding estimated distributions of temperature and thermal neutron fluence in Figure 5. (As remarked previously, the estimated temperatures may be too high; in like manner, more recent, but still preliminary fluence estimates indicate that the values presented in Figure 5 may be slightly low. In either case, the distributions themselves do not appear to be markedly different.) The similarity between ^{3}H activity and neutron fluence suggests that some fraction of the ^{3}H born in the spine (probably due to ^{6}Li impurity) remains immobile. This aspect merits further study.

Fuel element E11-07 was removed from the reactor after 701 EFPD; moreover, the fuel element was expected to operate at higher temperatures than those experienced in the operation of fuel element E06-01. As a result of both factors, it is reasonable to anticipate redistribution

of cesium within the fuel element. This expectation is verified by the data displayed in Figure 6, in which the Cs-137/Ce-144 activity ratio and estimated fuel compact temperatures are plotted as a function of position within the element. (Additional, but still tentative, calculations suggest that E11-07 may have operated slightly hotter than the temperatures designated.)

The data presented graphically in Figure 6 indicate significant loss of cesium in compacts 15-23. If compacts 26-30 are assumed to have retained the cesium born in these compacts, then almost half of the cesium that was born in compacts 15-23 has been released. Much of this released cesium has been swept by the purge stream toward the base of the fuel element, and has deposited on the cooler fuel compacts. In contrast, the Zr-95/Ce-144 ratio has been found to be sensibly constant along the fuel compact stack.

Fuel element E14-01 is one of five end-of-life driver fuel elements which are to be subjected to extensive PIE. This element, which contains ZrB_2 rods in the spine region, was expected to operate at somewhat lower temperatures than E11-07 (but no temperature distributions have been estimated thus far). Thus, it is not altogether surprising that no significant redistribution of cesium has been observed from γ-scans of the fuel compacts, as is seen from the data which are presented graphically in Figure 7. What is surprising is the manner in which the activity ratio varies with position; in regions of higher neutron fluence, it appears that more Cs-137 is born relative to Ce-144. Since the Peach Bottom HTGR fuel consists of pyrocarbon-coated $(U,Th)C_2$ microspheres, a possible explanation of the observed activity ratio variation is that at 899 EFPD, significant fissioning of bred U-233 is occurring, so that the variation of Cs-137/Ce-144 activity ratio is now perturbed by differences in fission yield of the two nuclides from U-235 and U-233 fission.

DISCUSSION

Surveillance of fission products during Core 2 operation of the Peach Bottom HTGR has been extensive, and a considerable body of data has been amassed (and is still being collected). Thus this paper serves to highlight only some of the data obtained to date. More detailed descriptions of selected aspects of the surveillance effort will be presented elsewhere.

Also, no attempt has yet been made to correlate the data with existing fission product transport computational programs. While the presence of the purge flow complicates the mathematical analysis, the surveillance data nonetheless present an excellent opportunity for the verification of the many computational programs which have been developed for purposes of describing fission product transport in gas-cooled reactors.

The most striking aspect attending our surveillance effort has been the cleanliness of the coolant circuit. As a result, it was possible to obtain data in some instances at minimal cost, such as the γ-scans of deposited activity. The cleanliness of the circuit is unquestionably due to excellent fuel performance and the highly efficient fuel element purge system. Indeed, as the data in Table II indicate, cesium behavior in the coolant circuit through May, 1973 (approximately two-thirds of design core life) was largely due to releases which had occurred during operation of Core 1. Total activity in the primary system coolant averaged a few hundred millicuries throughout most of Core 2 operation; this is in marked contrast to the 4200 curie design value.

ACKNOWLEDGEMENT

It is a pleasure to acknowledge the excellent cooperation that has been given to us by the staffs of Philadelphia Electric Company and General Atomic Company in the conduct of the surveillance program. Their advice and assistance are deeply appreciated.

TABLE I. IODINE AND CESIUM RADIONUCLIDE CONCENTRATIONS IN THE PEACH BOTTOM REACTOR COOLANT

	Concentration, pCi/L(STP)							
	May 18, 1973				May 28, 1974			
Form	^{131}I	^{134}Cs	^{136}Cs	^{137}Cs	^{131}I	^{134}Cs	^{136}Cs	^{137}Cs
	Entrance to Steam Generator (720°C)							
Molecular	?	0.078	0.0254	0.095	0.214	5.20	0.623	3.32
Particulate	?	0.149	0.0291	0.499	6.5×10^{-5}	0.93	0.139	0.74
Penetrating	7.6×10^{-4}	-	-	-	0.0381	-	-	-
Total	0.26	0.227	0.0545	0.594	0.252	6.13	0.76	4.06
	Exit from Steam Generator (325°C)							
Molecular	?	0.0379	ND[a]	0.053	0.066	0.53	0.061	0.37
Particulate	?	0.280	ND	1.09	0.00162	1.14	0.122	0.75
Penetrating	0.0106	-	-	-	0.00114	-	-	-
Total	0.112	0.318	ND	1.14	0.068	1.67	0.183	1.12

[a]Not detected.

TABLE II. SIGNIFICANT SAMPLING EVENTS IN THE PEACH BOTTOM REACTOR FISSION PRODUCT SURVEILLANCE PROGRAM

Date	Sampling Event	EFPD	Cs-137/Cs-134
10-03-69	Dust	452 (Core 1)	1.8
04-24-71	Upstream Coolant	252 (Core 2)	
	Diffusion Tube		1.3
	Filters		2.7
	Large Duct		3.0
	Dust		2.5
01-06-72	Downstream Coolant	384	
	Diffusion tube		3.9
	Filters		3.4
	Large Duct		3.4
	E06-01 Fuel		1.4
05-18-73	Upstream Coolant	629	
	Molecular		1.2
	Particulate		3.3
	Downstream Coolant		
	Molecular		1.4
	Particulate		3.9
09-14-73	Large Duct	701	1.4
	E11-07 Fuel		0.92
05-28-74	Upstream Coolant	818	
	Molecular		0.64
	Particulate		0.80
	Downstream Coolant		
	Molecular		0.70
	Particulate		0.66
10-31-74	Large Duct	899	0.81
	E14-01 Fuel		1.2

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Figure 1. Peach Bottom HTGR Plant Layout (ORNL-DWG-71-8680 R1).

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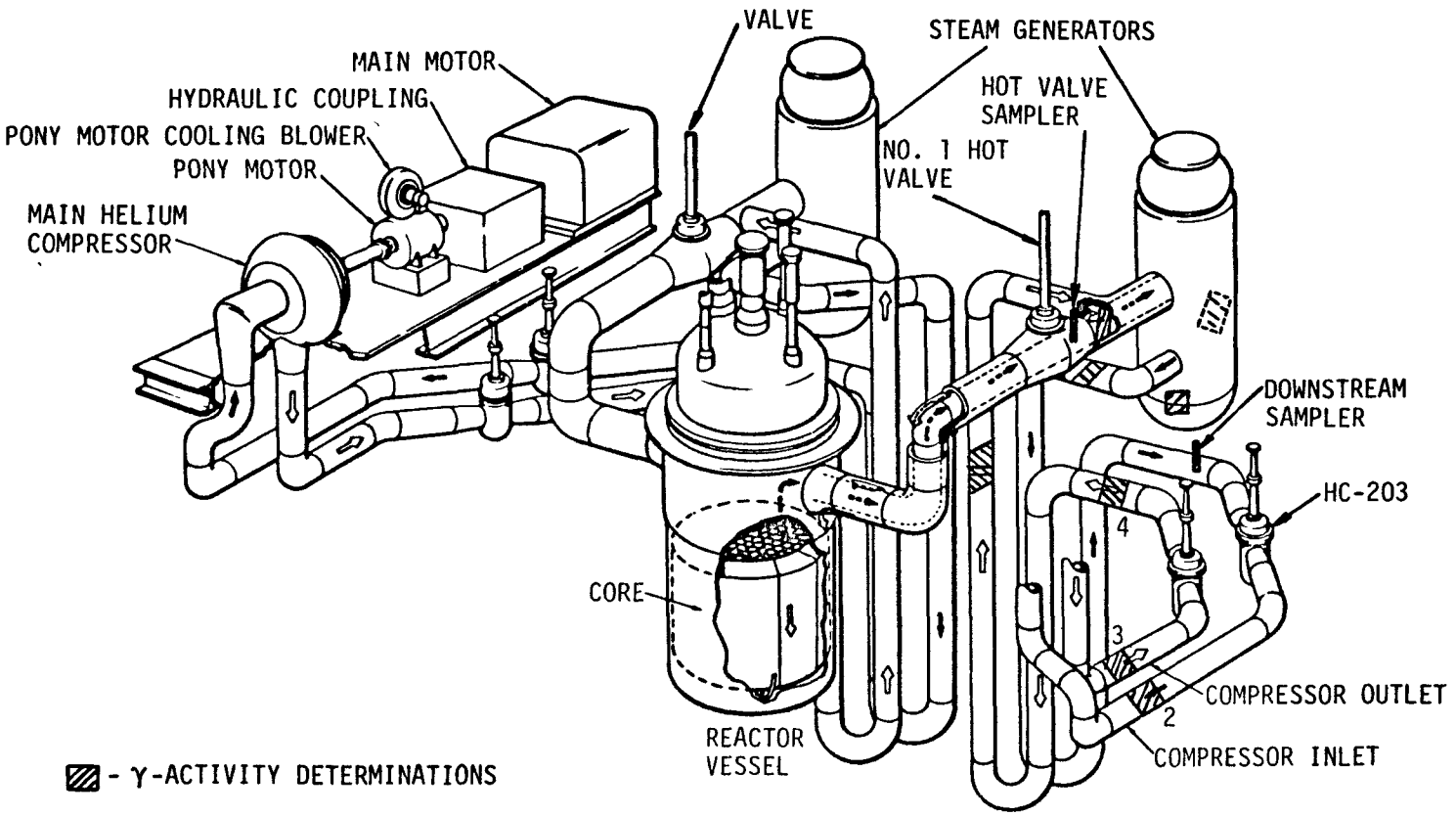
Figure 4. Radial Concentration Profiles of ^{134}Cs and ^{137}Cs at Compact 16, Fuel Element E06-01 (ORNL-DWG-75-8271).

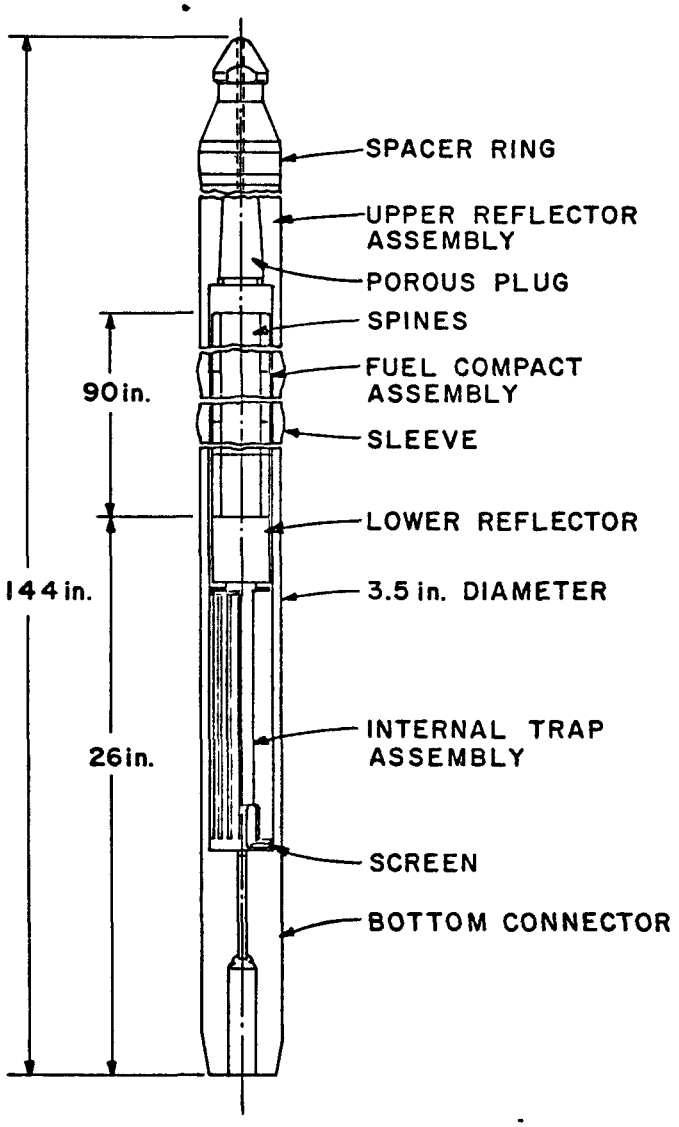
Figure 5. Axial Distribution of ^{3}H in Fuel Element E06-01 Spine Graphite (ORNL-DWG-75-8269).

Figure 6. $^{137}Cs/^{144}Ce$ Activity Ratio (September 14, 1973) in E11-07 Fuel Compacts (ORNL-DWG-75-8267).

Figure 7. $^{137}Cs/^{144}Ce$ Activity Ratio (October 31, 1974) in E14-01 Fuel Compacts (ORNL-DWG-75-8270).

ORNL-DWG 71-8680




SPACER RING
UPPER REFLECTOR ASSEMBLY
POROUS PLUG
SPINES
FUEL COMPACT ASSEMBLY
SLEEVE
LOWER REFLECTOR
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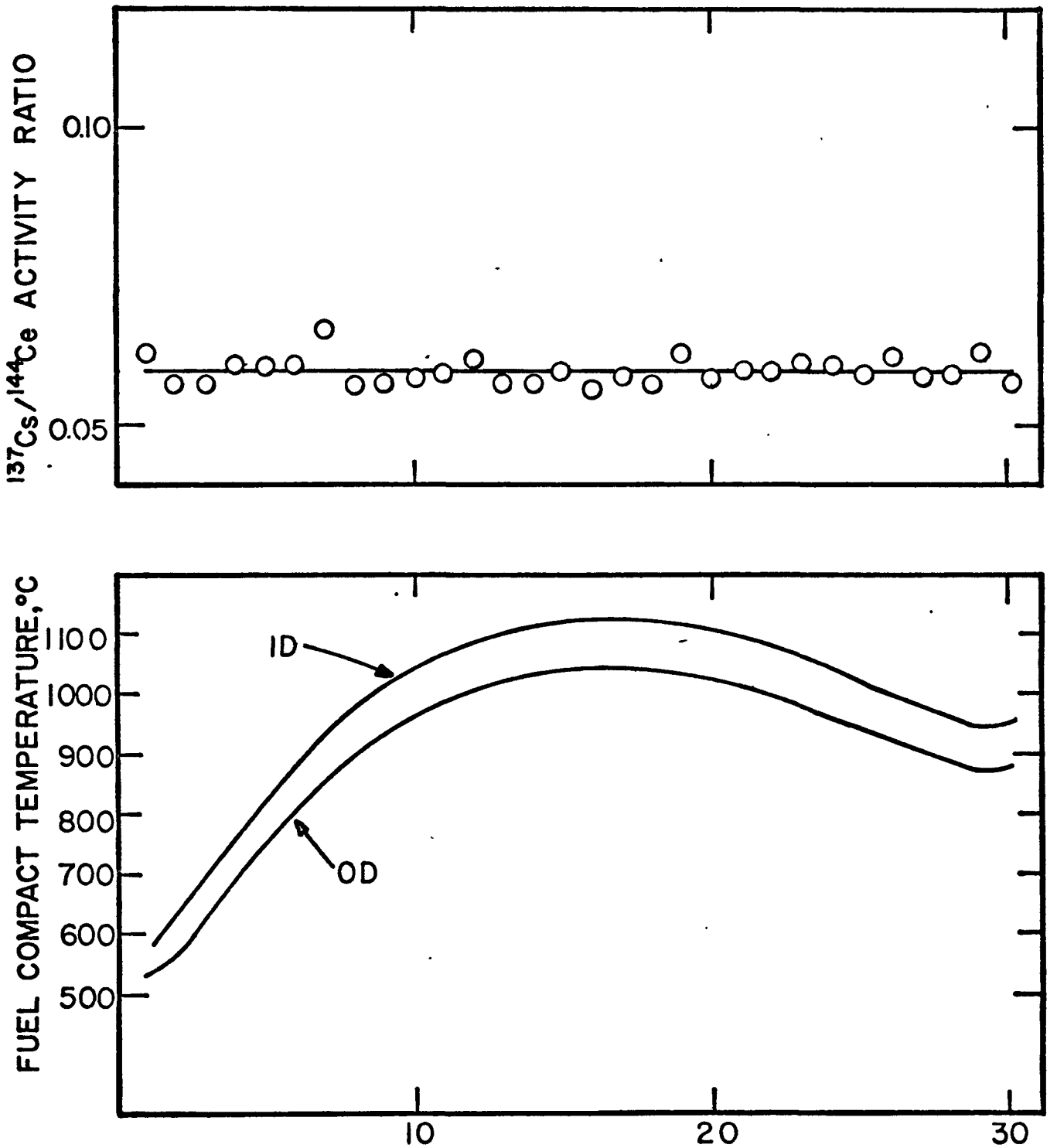
INTERNAL TRAP ASSEMBLY
SCREEN
BOTTOM CONNECTOR
90 in.
144 in.
26 in.

ORNL DWG 75-8268



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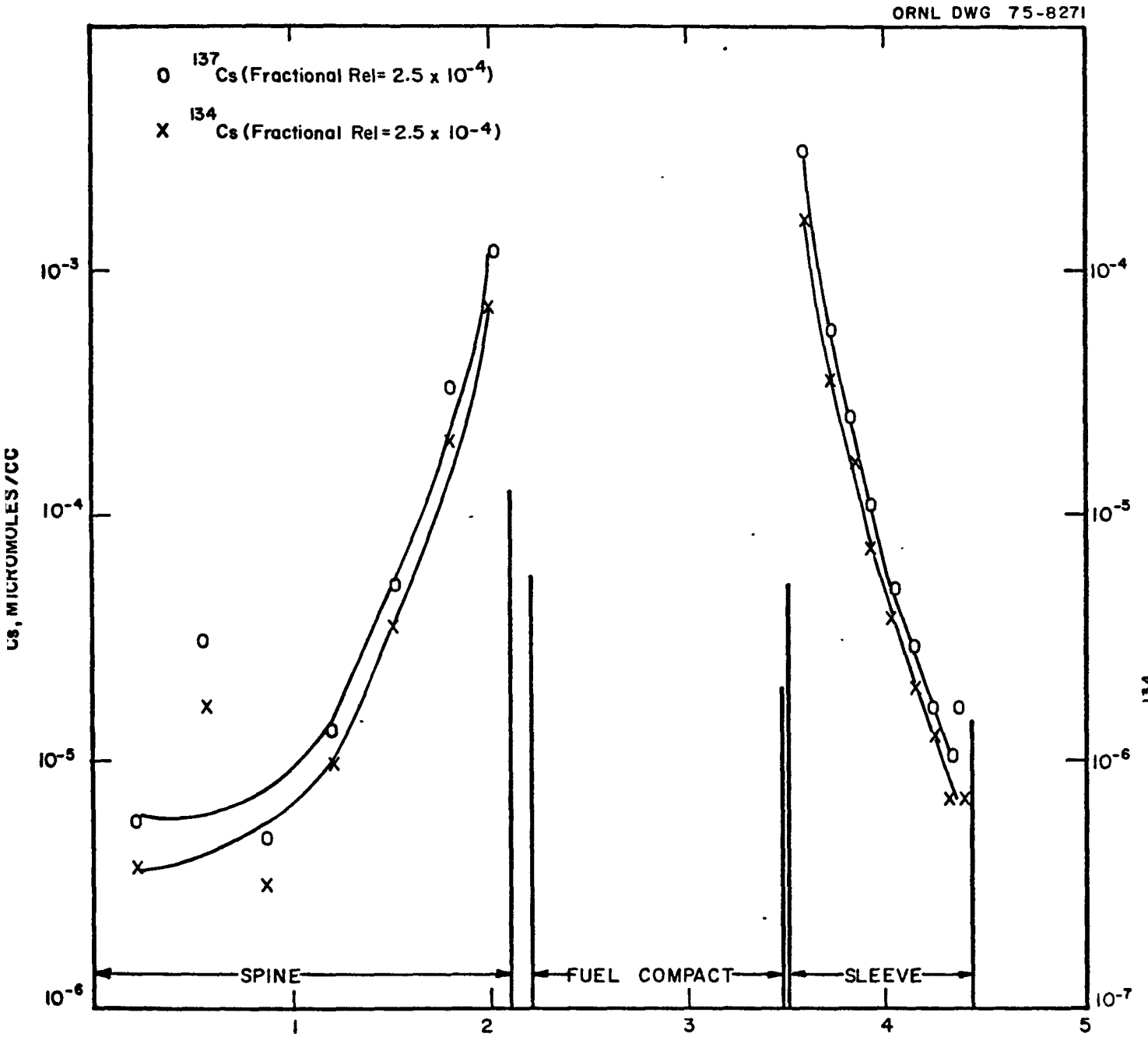
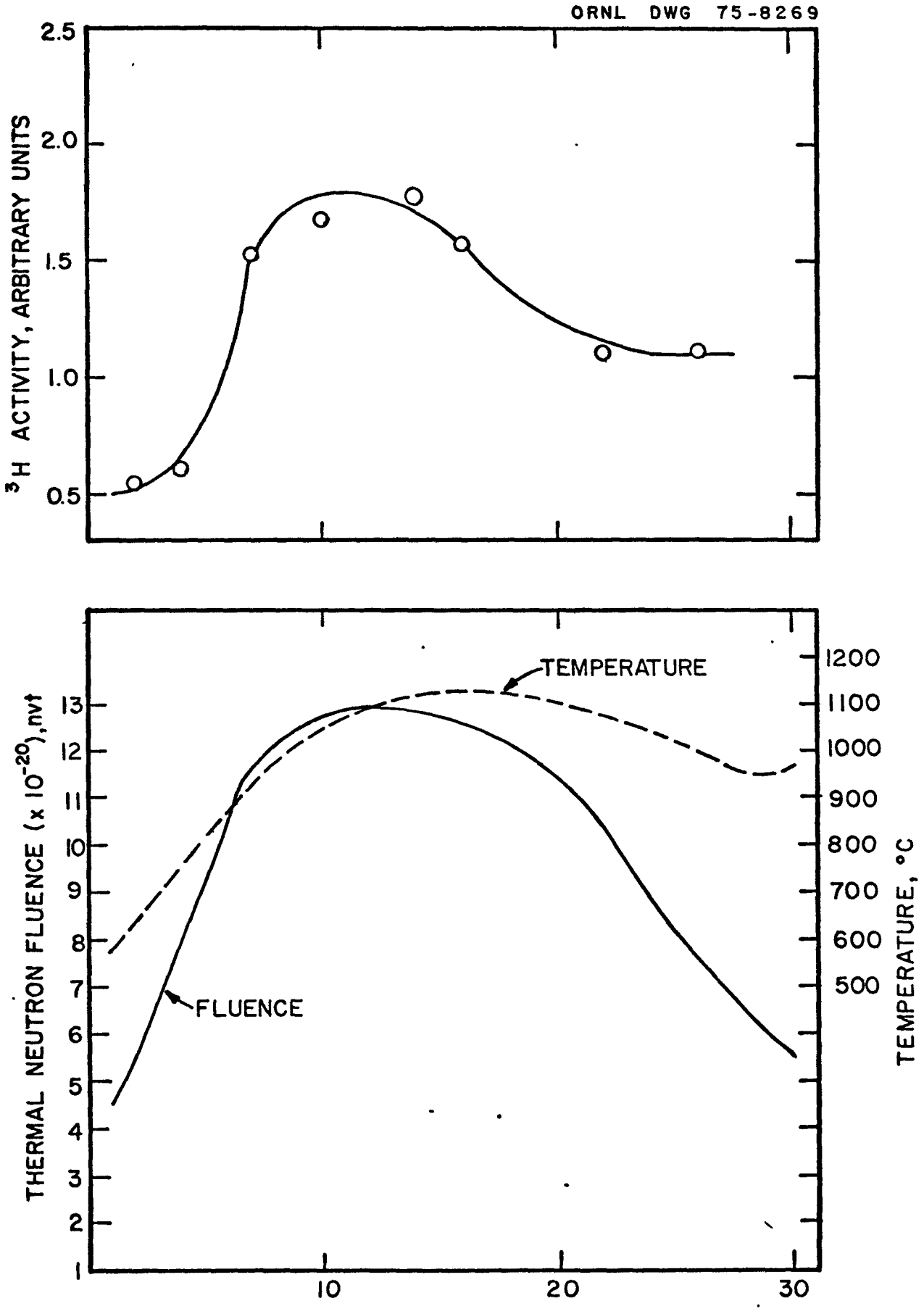
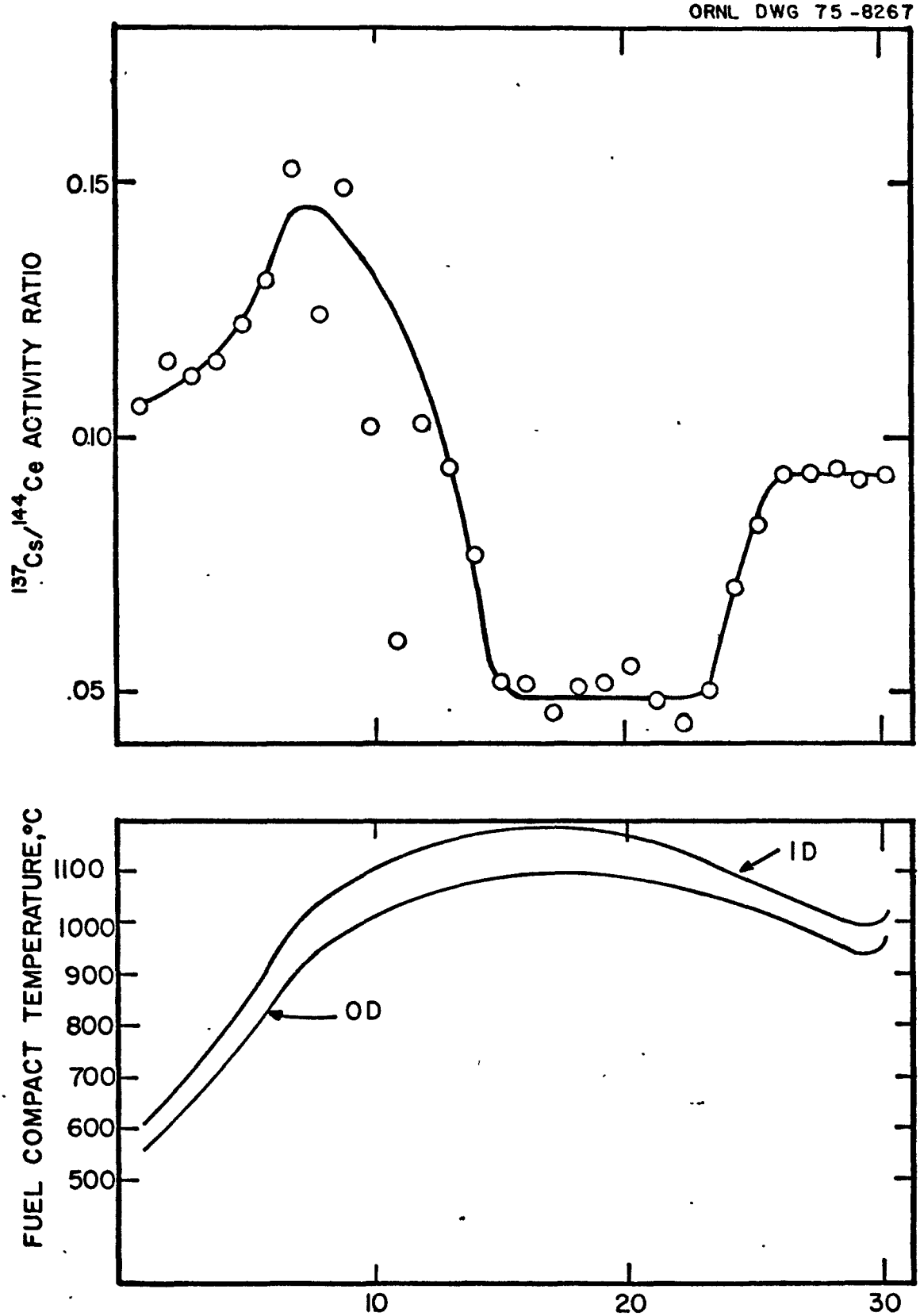


RADIAL DISTRIBUTION OF ^{137}Cs AND ^{134}Cs AT COMPACT 16, E06-01

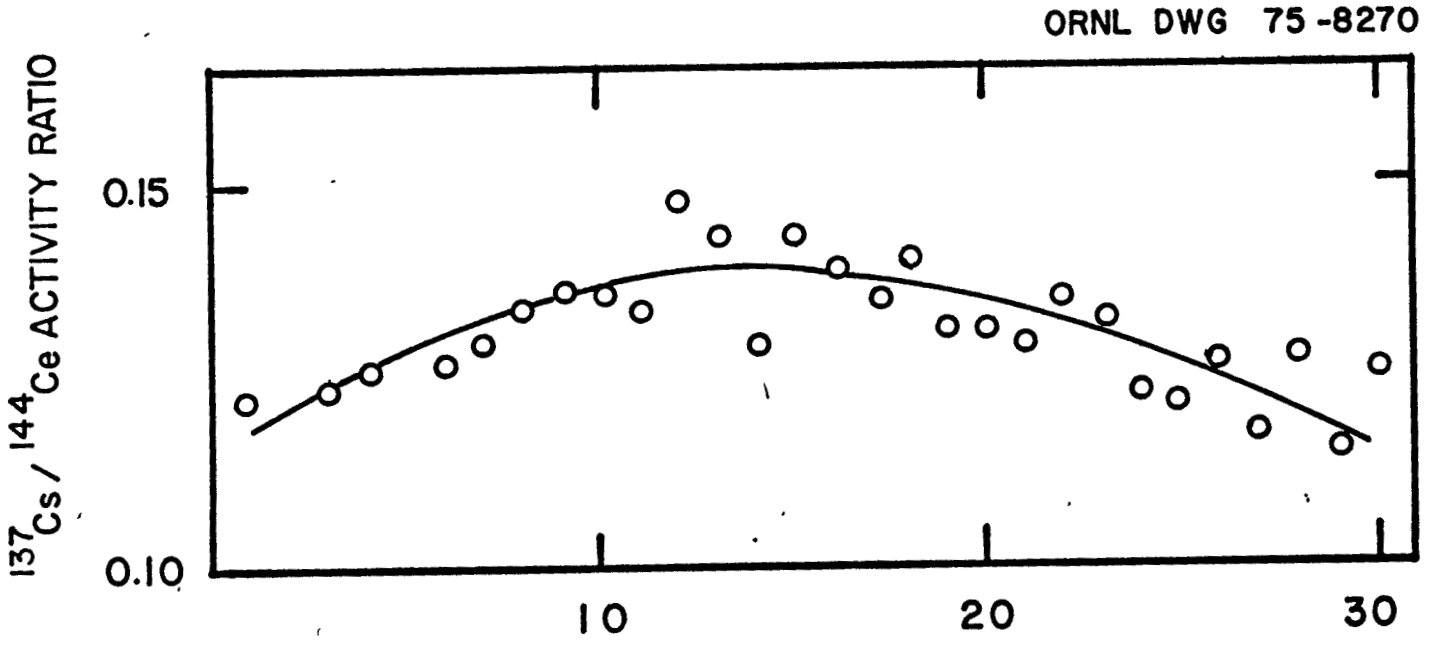


AXIAL DISTRIBUTION OF TRITIUM IN E06-01 SPINE

ORNL DWG 75-8267



$^{137}Cs/^{144}Ce$ ACTIVITY RATIO (SEPT. 14, 1973) IN E11-07 FUEL COMPACTS



$^{137}Cs / ^{144}Ce$ ACTIVITY RATIO

(OCT. 31, 1974) IN E14-01 FUEL COMPACTS