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NONDESTRUCTIVE ASSAY OF HIGHLY ENRICHED SPENT FUEL*

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Abstract

Passive gamma-ray and neutron measurements have been made on irradiated MTR fuel at the Omega West Reactor. Detectors utilized include a Ge(Li) for gamma spectroscopy, a U-235 fission chamber for thermal neutrons, and a smaller U-235 fission chamber with a beryllium photoneutron converter for detecting gamma-rays above 1.7 MeV. Measurements confirm that burnup can be closely correlated with the Cs-134/Cs-137 ratio. A comparison of activity profiles taken with the three detectors indicates that the method based on (γ,n) reactions in beryllium is a promising technique.

Background

Under DOE sponsorship, and also under the U.S. Program for Technical Assistance to IAEA Safeguards, the Los Alamos Scientific Laboratory (LASL) has become involved in both international and domestic aspects of safeguarding spent reactor fuel. Our primary objective is to develop improved methods for nondestructive assay of LWR fuel. There is a need for ruggedized portable instrumentation for use by safeguards inspectors who must verify the content of spent fuel storage facilities. And, there is also a growing need for permanently installed instrumentation which achieves better assay accuracy with perhaps some trade off in size and complexity. Operators of commercial power reactors have shown interest in the nondestructive assay (NDA) of spent fuel, in part because of safeguards considerations, and also because of possible economic implications. We recently completed measurements on BWR fuel assemblies at a Consumers Power facility (Big Rock Point Nuclear Plant) and on PWR assemblies at a Commonwealth Edison facility (Zion Generating Station). However, the first measurements under this program were made on MTR fuel at the Omega West Reactor in Los Alamos.

NDA methods have been applied to spent fuel for a number of years, and there are several review papers which contain comprehensive lists of references.[1,2,3] Work on

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spent fuel has been an international undertaking with some of the most productive research being done in Europe. Measurement methods that have previously been discussed for the NDA of spent fuel include (1) passive gamma ray, (2) passive neutron, and (3) active neutron methods.[1] The most widely used technique today is high-resolution gamma spectroscopy, in which measurement of discrete gamma rays from fission products allows one to determine U-235 burnup. (In this paper, the burnup in a fuel element is defined as the mass of U-235 that has fissioned.) Passive gamma-ray methods have the advantage (compared with neutron methods) of requiring less information on the history of the fuel, since cooling times can generally be inferred by the ratio of gamma intensities from two fission products having different decay-times. However, gamma methods have the disadvantage (compared with neutron methods) of being less sensitive to the inner fuel rods in an assembly. A combination of gamma and neutron techniques would appear to provide the most promising approach for inspection purposes.

OWR Spent Fuel

The Omega West Reactor (OWR) is a research reactor, currently operated at a maximum power of 8 MW, one shift per day, 5 days per week. Fuel elements (which are MTR type) have overall dimensions of 108 cm x 7.6 cm x 7.6 cm and contain eighteen curved plates, 1.53 mm thick. Each plate holds UAl alloy fuel sandwiched between two 0.51 mm sheets of pure Al. The UAl alloy section is 61 cm long by 0.51 mm thick. It contains a mixture of 93% Al and 7% U (by weight), with the uranium having been enriched to 93% U-235. Each fuel element has an initial loading of approximately 220 grams of U-235, and under typical operation spends approximately 3 years in the reactor core.

Spent fuel elements are normally stored in the reactor containment vessel but can be removed to a nearby storage pool for experiments.[4] Fig. 1 shows a spent OWR fuel element in the storage pool together with the watertight, air filled aluminum collimator used with the Ge(Li) detector. Table I lists the fuel elements studied in the two exercises at OWR. Note that cooling time (or time elapsed from discharge) varied from 341 to 1445 days and that declared burnup varied from 61.36 to 73.87 grams. Asterisks in Table I indicate elements studied in both exercises.

Burnup Determination

Burnup measurements were made with the apparatus shown in Fig. 1; however, the collimator was rotated 90 degrees so that the entire fuel element could be viewed. Fig. 2 shows a typical gamma-ray spectrum taken with the Ge(Li) detector. Burnup can be determined from the Cs-134/Cs-137 isotopic ratio, since Cs-134 is a shielded isotope formed by neutron activation of Cs-133, and Cs-137 is a direct fission product. Whereas

Cs-137 formation is proportional to total integrated neutron flux, Cs-134 formation is proportional to the integrated flux squared. The advantages of using an isotopic ratio as a burnup indicator (rather than a single isotope) are clear: the ratio is independent of the amount of material in the assembly and independent of detector solid angle. Furthermore, by choosing gamma rays of nearly the same energy, gamma absorption in the assembly can be neglected or handled with a small correction. For determining the Cs-134/Cs-137 ratio, the 605 keV line of Cs-134 and the 662 keV line of Cs-137 were used.

Fig. 3 shows declared burnup for the ten MTR fuel elements studied in September, 1977 as a function of the Cs-134/Cs-137 activity ratio. The activity ratio has been corrected for cooling time using the declared values shown in Table I and the half-lives of Cs-134 (2.06 y) and Cs-137 (30.12 y). No correction was made for differing irradiation histories, and no correction was made for gamma absorption in the fuel assembly or beam attenuator (5.08 cm Pb).

In addition to a least squares fit to a straight line, Fig. 3 shows 95% confidence bounds. The bounds may be interpreted as defining a range of burnup values within which one is 95% confident that a burnup value will fall, given a measured Cs-134/Cs-137 ratio. These results indicate that the burnup of OWR fuel elements can be predicted using Cs-134/Cs-137 ratio techniques with a precision having 2.1% standard deviation (1σ). Measurements made on the same ten fuel elements, using the Eu-154/Cs-137 isotopic ratio, showed a slightly larger standard deviation, 3.3% (1σ). This is somewhat better precision than previous investigators have reported for MTR fuel, in which gamma-ray spectra were recorded for a single position on the MTR element.[5,6]

Activity Profile Measurements

Gamma-ray and neutron activity profiles give a measure of relative burnup as a function of axial position along the fuel assembly.[1] A combination of the activity profile and the Cs-134/Cs-137 ratio measured at one point on the assembly can be used to determine absolute burnup for the total assembly. For many inspection situations in which time is limited and/or experimental geometry is fixed, performing burnup measurements in this way is more practical than scanning complete assemblies using high-resolution gamma spectroscopy, even if there is some loss of precision. Thus, there is a need for both neutron and gamma-ray detectors for measuring activity profiles. For inspection purposes, such detectors should be rugged and reliable, should provide rapid data collection, and should give an activity profile that is known to be proportional to burnup.

Table III shows characteristics of detectors used for activity profile measurements at OWR. The activity profile determined by a Ge(Li) detector measurement of the 662 keV Cs-137 line was used as the standard against which activity profiles measured by other detectors were compared. In Fig. 4, three sets of activity profiles are plotted. These curves have been normalized at their peaks. At the top of Fig. 4, we compare the activity profile determined by the Cs-134/Cs-137 ratio with the standard Cs-137 profile. Both of these activity profiles were taken with the experimental arrangement shown in Fig. 1. Since Cs-137 and Cs-134/Cs-137 are both commonly used burnup indicators, their agreement is expected.

In the center of Fig. 4, we compare the Be(γ,n) fission chamber activity profile with the Cs-137 profile. The small fission chamber is encased in a 2 cm polyethylene sleeve, and placed on top of a Be disc 9 cm in diameter by 2.54 cm thick. In performing the experiment, the Be(γ,n) detector was lowered into the storage pond inside a vertical aluminum pipe and placed on top of the fuel element. Gamma rays exceeding the photoneutron threshold of beryllium (1.66 MeV) create neutrons which are thermalized in the polyethylene and detected in the fission chamber. Photoneutrons from Be considerably outnumber neutrons from the fuel element, giving a count rate at the center of the activity profile of 50 counts per second. As can be seen from the gamma spectrum in Fig. 2, the activity profile determined with the Be(γ,n) detector is due mainly to Pr-144. Considering the differences in experimental geometry, the agreement between the Be(γ,n) activity profile and the Cs-137 activity profile is excellent.

At the bottom of Fig. 4, we show the neutron activity profile measured with the larger fission chamber. The larger fission chamber was placed in a vertical aluminum pipe that was put in contact with the side of the fuel assembly for optimum detector solid angle. Because of the low neutron counting rate (2 counts per second maximum), statistical errors were significant. We expect much better results will be obtained with fission chambers measuring neutron profiles of spent LWR assemblies because of greater neutron emission arising from the spontaneous fission of curium and plutonium isotopes.

Conclusions

Gamma-ray measurements of fission products in irradiated MTR fuel assemblies may be more accurate than similar measurements on irradiated LWR fuel assemblies, because of less self-absorption in the fuel and insignificant fission product migration. Correlation of the Cs-134/Cs-137 isotopic ratio with declared burnup indicates that burnup can be determined with a high degree of precision, 2.1% (1σ). For measuring activity profiles, both Be(γ,n) detectors and fission chambers offer promise and should be further evaluated on LWR assemblies.

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FIGURE CAPTIONS

Fig. 1 Drawing of apparatus used with the Ge(Li) detector, positioned for measuring activity profiles of OWR fuel elements. The collimator was rotated 90 degrees for burnup measurements to view the entire fuel element. Water in the storage pond is 2.4 meters deep.

Fig. 2 Gamma-ray spectrum of irradiated Omega West Reactor fuel element showing prominent fission products and associated gamma-ray energies. The data, taken with the experimental arrangement shown in Fig. 1, was recorded on a 4,096-channel analyzer.

Fig. 3 Plot of declared burnup values in grams versus measured Cs-134/Cs-137 ratio for ten OWR fuel elements. A linear least squares fit and 95% confidence bounds are also shown.

Fig. 4 Activity profiles for OWR element #383 as measured by three types of radiation detectors.

TABLE I. OMEGA WEST REACTOR FUEL ELEMENTS STUDIED

	Fuel Element	Cooling Time (Days)	Initial 235-U (g)	Declared Burnup (g)
September 1977	356	1352	220.67	73.87
	359*	1269	219.66	69.46
	361	1205	222.90	67.12
	363	1206	223.52	69.27
	364	1092	219.47	63.40
	368*	948	223.50	65.15
	370*	892	221.20	61.36
	371	892	223.58	61.36
	372	725	223.00	70.28
	378	556	223.15	65.37
January 1978	357	1445	218.13	71.64
	359*	1374	220.67	69.46
	368*	1053	223.50	65.15
	370*	997	221.20	61.36
	373	856	219.25	70.54
	374	341	222.77	65.65
	375	683	222.51	68.38
	379	661	222.39	65.22
	383	513	223.23	63.37

TABLE II. DETECTOR CHARACTERISTICS

Ge(Li)

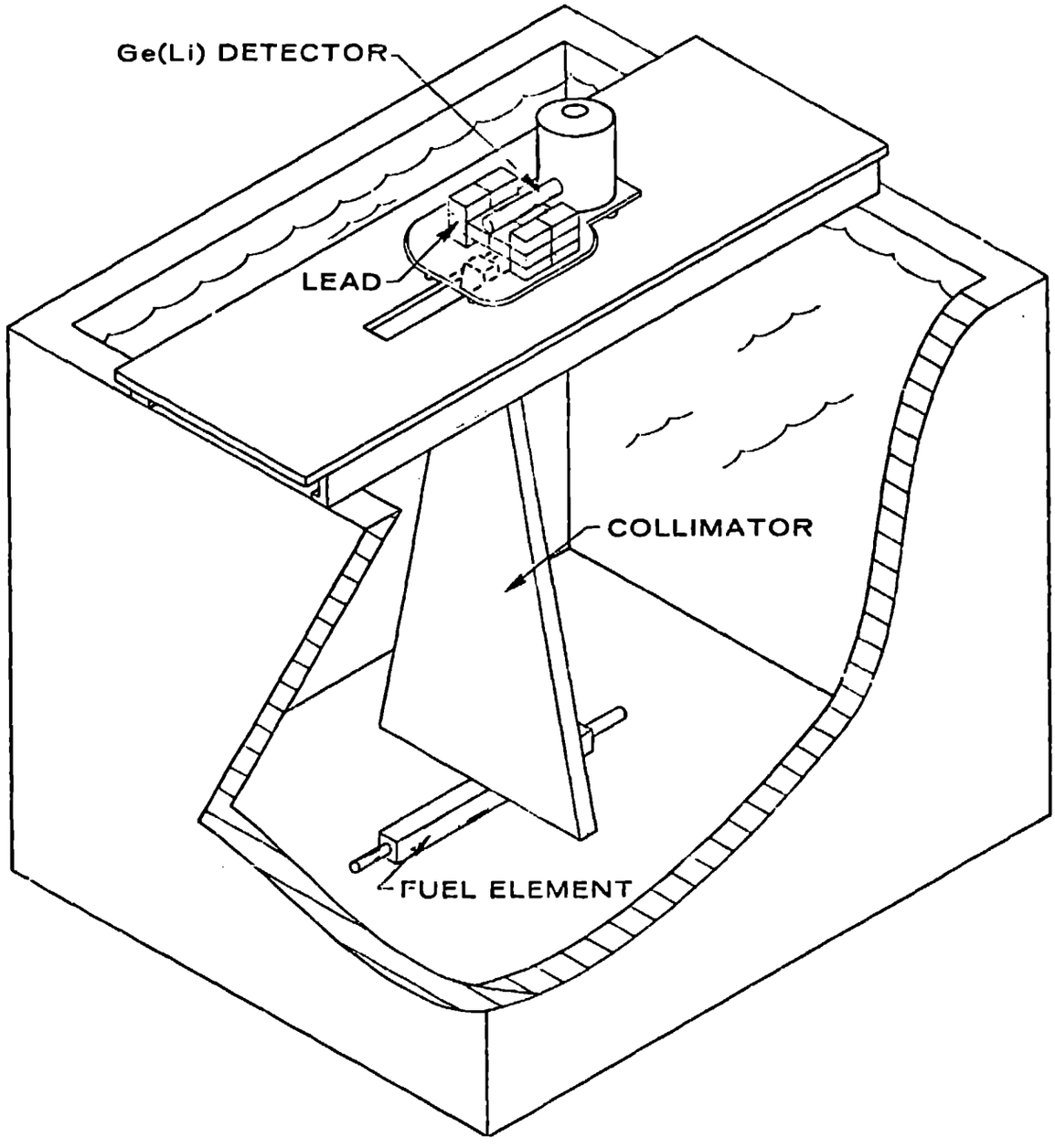
Volume:	90 cm^3
Efficiency:	17.5%
Resolution:	1.9 keV at 1.33 MeV
Manufacturer:	Princeton Gamma-Tech

Be(γ,n) Fission Chamber

Mass 93% U-235:	38.6 mg
Gas:	95% Argon, 5% Nitrogen
Active Length:	7.6 cm
Manufacturer:	Reuter-Stokes

Large Fission Chamber

Mass 93% U-235:	1.6 g
Gas:	95% Argon, 5% Nitrogen
Length:	30 cm
Manufacturer:	Reuter-Stokes



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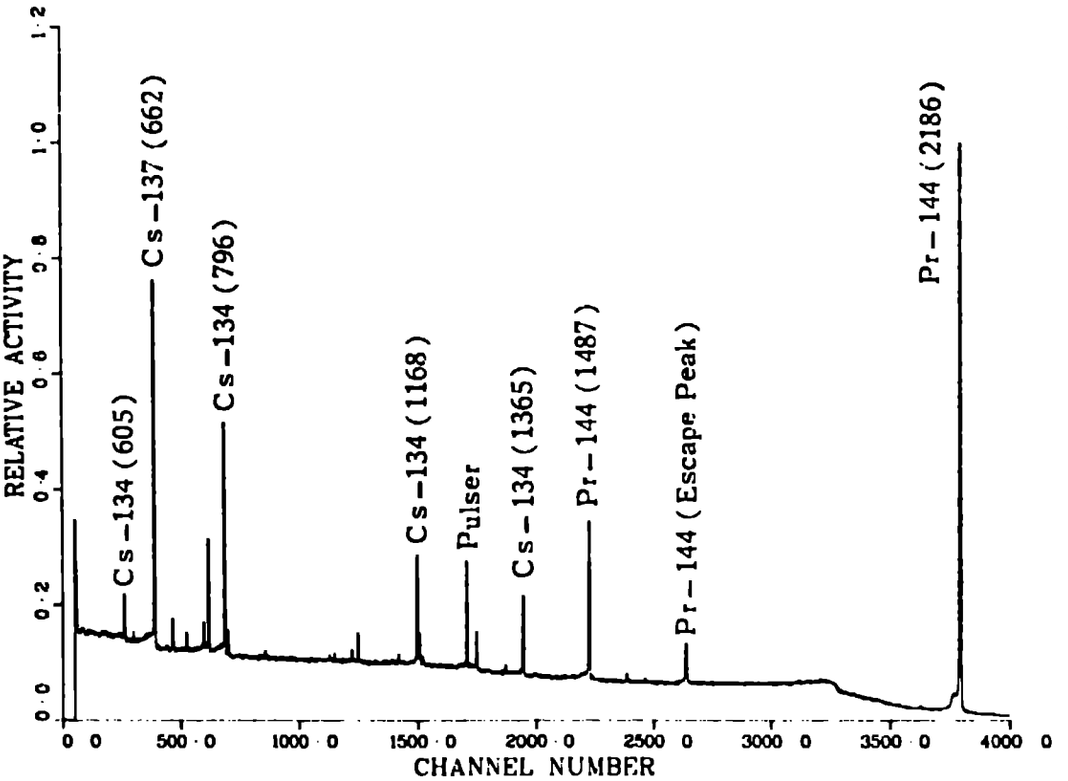

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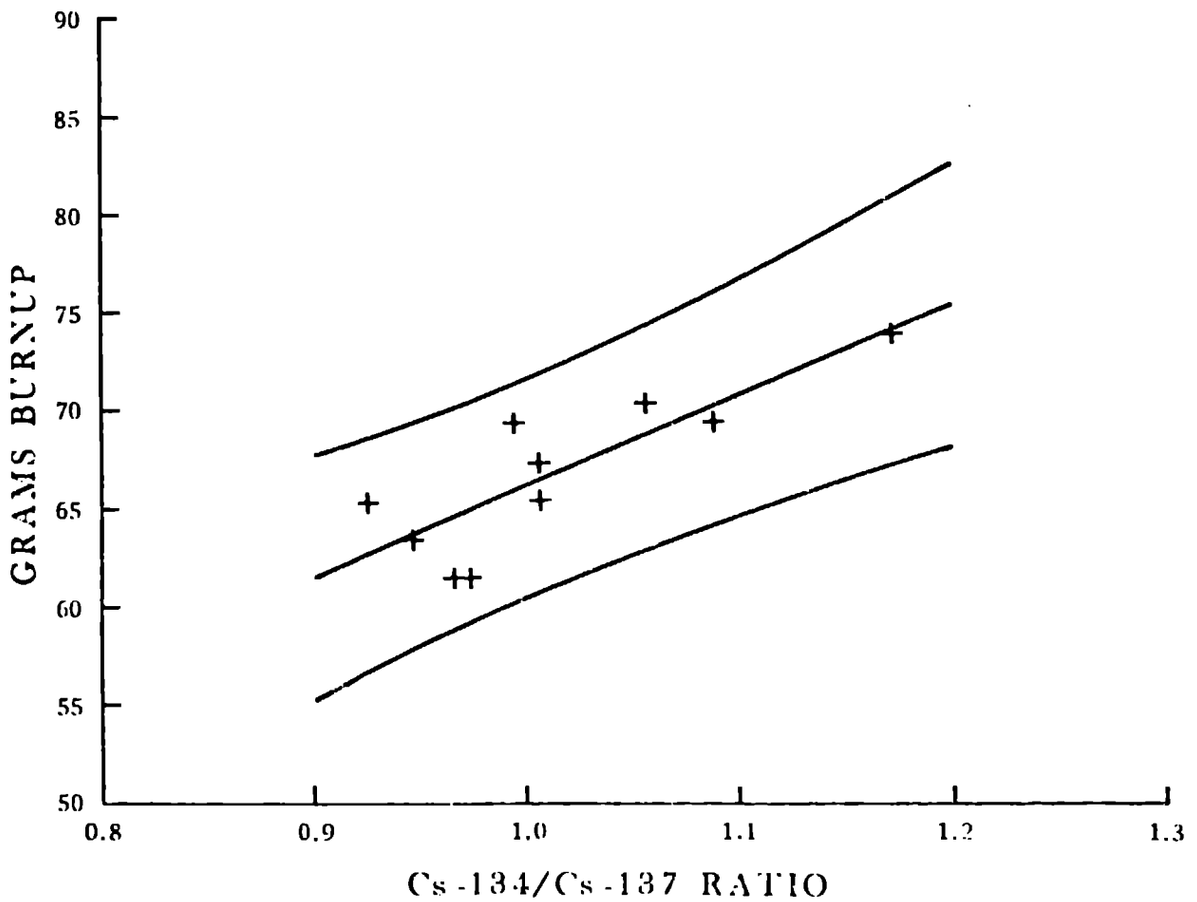

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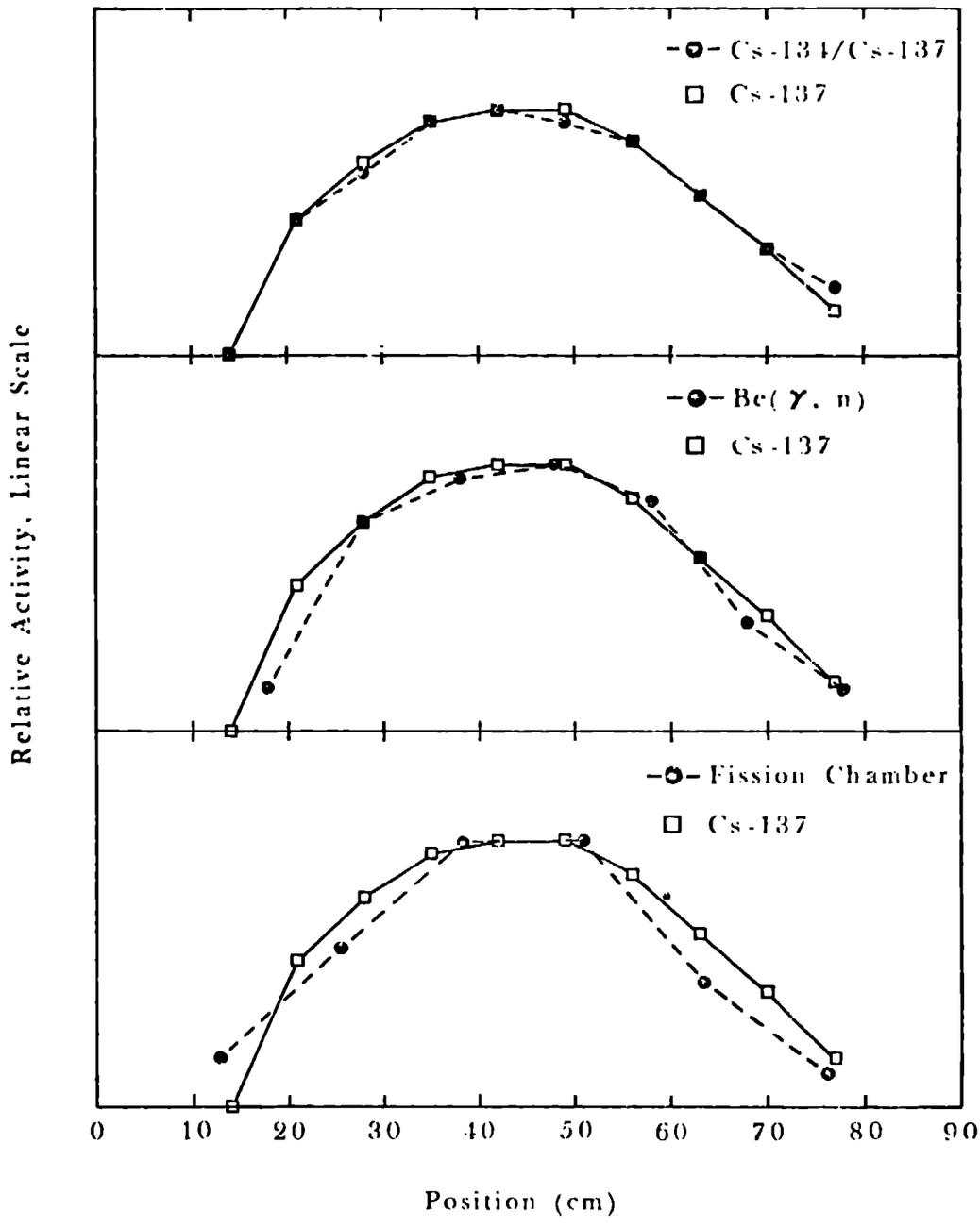

Fig. 4 Activity profiles for OWR element #383 as measured by three types of radiation detectors.