The Possibility of Release of Cesium in a Spent-Fuel Transportation Accident

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In this report we consider the hazard associated with the release of cesium in a hypothetical event in the transportation of spent reactor fuel. We find that release of Cs presents a very grave problem, much more important than the release of krypton and iodine considered in previous studies. An accident in the transportation of light water reactor fuel to the reprocessing plant is assumed which results in loss of the cooling water from a cask. This eventuality has been previously considered,[1,2,3,4] but detailed attention was restricted to the gases krypton (Kr^{85}) and iodine (I^{131}). The loss of coolant will also lead to escape of some volatile and soluble fission products. The most important fact which brought us to the present study is that the Cs migrates in the fuel pellet during irradiation and substantial quantities escape and are deposited on the cladding. Furthermore, the cladding of the fuel pins in the postulated accident would experience much higher temperatures than in normal reactor operation. In this report, we estimate the amount of release of Cs from the fuel, from the fuel pin, and from the cask into the atmosphere, and find it to be substantial. We then consider the exposure to which a population might be subject at a distance from the cask. The isotopes involved, Cs^{134} and Cs^{137}, are among the most potent known. The ensuing somatic effects and implied effects on land use are briefly discussed.

Quantity of Cesium Released from the Cask:

One metric ton, MT, of spent LWR fuel (33,000 MWD/MT average burn-up) contains, after cooling off 90 days, $2.3x10^5$ curies of Cs^{134}, $1.1x10^5$ Ci of Cs^{137}, $1.1x10^4$ Ci of Kr^{85}, and 381 Ci of I^{131} in addition to other radioactive materials.[5] Under typical licenses, the fuel must cool off at least 90 or 120 days depending on the cask. Spent fuel may be sent for reprocessing by road or rail. Typical large casks are water filled and contain .5 MT and 3.2 MT respectively[6].

An accident in transit could result in various degrees of damage to a water filled cask: 1) There could be zero release or no substantial release of cooling water (or radioactivity) in a period of, say one day. 2) There could be seepage of coolant or flow of coolant through a small orifice or pressure crack such as might result from damage to a pressure release system, to distortion of the cask in the neighborhood of a gasket, or to excess heating of the coolant, resulting in substantial loss of cooling water over an hour to a few hours. This would typically result in drying out the cask as steam is released via some pressure release mechanism. 3) There could be a major breach leading to rapid loss of coolant.

It is difficult to predict what kinds of accidents or impact, with what probability, would characterize the various possibilities mentioned above. The major cask test standard involves impact against a flat surface. Impalement at substantial velocity, as may occur in rail accidents, or broadside impact at substantial velocity against curved objects, such as a bridge abutment, may be important. Accidents may involve multiple impact. In other words, there is a significant difference between test and design conditions and real accident conditions. In addition, we stress that quality of actual casks and the handling of these casks will not correspond to theoretically considered systems. Failure of workmanship, such as failure to secure bolts properly on loading, may result in substantial loss of cooling water in a minor accident or even in the absence of a road or rail accident. In the latter case the loss would in many cases be unobserved, especially in the case of rail transport.

An important possibility is fire with or without associated impact. Fire could distort the cask or it could heat the coolant, causing its release. It could cause fuel rod rupture.[4] It could heat the spent fuel. Combustion gases could transport radioactive materials. In this report we are principally interested in the consequences of loss of cooling water.

To put this study in perspective, consider at turn of the century the typical prediction of 1000 LWR's in the 1000 Mw region which will involve about ten million miles per year of rail transport of large casks containing spent fuel, and/or 50 million miles of truck transport. Probabilities per mile of accident at various velocities and with various times of duration of fire have been reviewed by the AEC[7]. In a ten year period surrounding the year 2000, several rail accidents and/or several tens of road accidents involving casks would be expected with fires lasting well over one-half hour and/or impact at well over 30 miles per hour, as shown in Table I. Considering this and the possibility of faulty workmanship we conclude that accidents resulting in damage of category (2), as defined three paragraphs above, will not be unusual in a ten year period[8]. Furthermore, the loss of cooling water is a design basis accident considered by the AEC for water cooled casks.[1,2,4] The results of loss of cooling water accidents are the subject of the rest of this report.

One uncertainty in an accident is the extent and degree of breakage of fuel pin cladding. (We are not interested in fuel pins which were perforated before transportation to the extent the available Cs was removed in core or storage pool.) The cladding is brittle after irradiation. An impact such that the cask if breached may or may not impart considerable physical shock to the pins. It is not surprising that different authorities differ on this point.[2,4] We assume that 10% of the pins will be fractured at the time of the accident. (This number is not critical in our final result.)

Table I

Expected Number of Serious Accidents in Transport of Spent Fuel Casks in Ten Year Period Surrounding the Year 2000*
(Based on reference 7)

Accident Severity Category	Vehicle Speed at Impact (mph)	Fire Duration (hr)	Expected Road Accidents	Expected Rail Accidents
"Moderate"	0-30	1/2-1	150	8
	30-70	< 1/2		
"Severe"	0-50	> 1		
	30-70	1/2-1	4	0.2
	> 70	< 1/2		

*Very severe and more moderate accidents have been omitted.

In addition to possible damage to cladding upon impact and shortly afterwards, fuel pins which remain intact are subject to creep rupture at the high temperatures which develop after the cask is substantially dried out.[9]

Routes for Cs release associated with the two types of breach of cladding are shown in Figure 1. Of major interest is the amount of Cs which had already escaped from the fuel during irradiation. In typical LWR experience 30-35% of the noble gases escape from the fuel. Laboratory tests and calculations are the main source of information on escape of Cs. These can be correlated with rather limited direct evidence from actual reactor experience. The diffusion of cesium in sintered UO_2 is slow at temperatures such as characterize the outer curved surface of the pellet, but is comparable or greater than that of the noble gases in irradiated fuel at higher temperatures as shown in Figure 2.[10] Some Cs will diffuse out of the pellet; most will remain in a cylindrical distribution very near the outer curved surface. See Figure 3.[11] The migration of Cs to the surface shown in this figure would be even greater at higher, typical burn-up levels. In one experiment it is stated: "The Cs^{137} migrated both radially and axially and in the case of the highest rated rod it is estimated that about 30% of the total rod inventory of Cs^{137} is deposited on the cladding."[12] As remarked here, the Cs which escapes from fuel pellets during irradiation condenses on the relatively cold pellet surface and inner wall of the cladding. The fraction escaping is sensitive to temperature and thus to location in the reactor. We assume that 20% of the Cs produced in the fuel escapes from the fuel pellet into the interior of the rod before transportation. Our final result scales with this number.

A further source of Cs escape from fuel is leaching of the fuel in those pins which are breached at the time of impact. If the outer 1.2×10^{-5} inch of the fuel pellets is leached,[1,2] 0.01% of the pellet volume is involved, and we conclude that about 1% of the Cs is leached on the basis of its high surface concentration. (This number is not important in our final result.)

Under normal conditions in a large water cooled cask the temperature of the cladding may be in the range 320-460°F.[13] After substantial loss of coolant, the temperature of the hottest rod will ultimately rise to 1250-1500°F depending on the cooling off time of the fuel at the reactor and other considerations.[9] Similar calculations for the large IF 300 cask indicate the even higher hottest-pin temperature of 1576°F.[4,13]

The fraction of intact pins which ultimately rupture under these conditions, and the time to rupture, is discussed in the Cask Designer's Guide.[9] If the cooling off period has been short, e.g., 90 days, most pins are expected to rupture.

If it is longer, e.g., 150 days, a substantial fraction of Zircaloy clad pins are still expected to rupture within 10 hours. For the hypothetical accident to be discussed, we assume 1/2 of all pins creep rupture.[4]

Two release routes appear most interesting: 1) For those rods breached at impact, Cs released from fuel during irradiation will dissolve in the cooling water and some more will be leached out of the pellet surfaces. Some of this Cs will be carried out of the cask with the steam and enter the atmosphere. 2) For those rods that creep rupture later, Cs released from the fuel during irradiation, if in volatile form, will enter the cask cavity as gas or an aerosol. The metallic Cs vapor pressure is high at temperatures well below its boiling point of 1253°F at one atmosphere. Some of the Cs free in the cavity will be carried out of the cask with further pressure releases by the cask.

1) Release from pins breached on impact: We have assumed above that 10% of the pins are breached and that 21% of the total Cs burden from these pins is taken up by the cooling water. As this water boils, a fraction of this Cs will be contained in particulates and escape from the cask and be carried off in the atmosphere as the steam escapes. Unlike quiescent vaporization, the bursting of bubbles results in an aerosol which has roughly the same content of solute as the body of liquid.[14] The fraction of Cs compounds entrained in the steam

in the boiling process and escaping into the outer atmosphere is taken to be 10%. This may be a significant underestimate. (Our final result is not sensitive to this number.) The remaining 90% will be left on surfaces in the cavity. The product of these factors implies an escape of about 2,200 Ci of Cs into the atmosphere from a cask containing 3.2 MT of fuel.

2) Release after creep rupture of pins: We have assumed above that 50% of the pins creep rupture at high temperature, and that 20% of the total Cs burden from these has escaped the fuel pellets. We further assume that 1/2 of this available cesium is in volatile chemical form. The chemical situation is complex and is the subject of current research.[15] Thus 10% of the total Cs inventory of ruptured pins is assumed to go into the cask cavity atmosphere as a gas or aerosol. We do not explicitly consider further outgassing of the fuel as a source. There are three sources of mass flow out of the cask at this time: boiling off of any remaining cooling water, ultimate increases in temperature, and the gases released from the ruptured pins. The available fission gases per metric ton are about 300 liters STP which implies over 1/2 atmosphere partial pressure at conditions under consideration. The mass flow which will carry some Cs out and into the atmosphere will depend on particular conditions such as cavity pressure and the pressure release mechanism at this stage. On the basis of these three sources, we estimate that 10% of the Cs available in the cavity atmosphere will escape from the cask in pressure releases into the outer atmosphere. The product of these factors implies an escape of about 5,300 Ci of Cs.

Dispersal of Cesium and Its Somatic Effects

In a loss of coolant accident involving a railroad cask containing 3.2 MT of spent fuel we have estimated a combined release of 7500 Ci of Cs (roughly 2/3 Cs^{134} and 1/3 Cs^{137}). This release may take place in intervals over a time of some hours. The problem of estimating the damage due to such a release is difficult because it depends in detail on conditions which will differ widely from case to case. Rather than examining all kinds of weather conditions and all population distributions we will examine two simple interesting cases.

First, we consider an individual 1 km downwind from the accident, remaining there while the radioactive cloud passes, or in a building with ventilation such that the exposure is the same. We consider stable weather conditions with a wind speed of 1 m/sec (about 2 mi/hr) and a low degree of gustiness such that at 1 km there is a vertical spread $\sigma_z = 8$m and a horizontal spread $\sigma_y = 42$m.[16] Thus the cloud would sweep out a volume like a long narrow plume (subtending an angle of about 1/10). We assume that the release is not carried high into the air by fire. (If it were, our second case below would be more applicable.) We assume that particulates are small and weather conditions are such that the Cs is not largely deposited out closer to the cask. The concentration of the cloud downwind at 1 km is then

$$\frac{Q}{\pi\sigma_z\sigma_y V} = 7\ \frac{\text{curie-sec}}{m^3} .$$

We only consider the dose to an individual associated with inhalation. Breathing at a rate of $3.5x10^{-4}$ m^3/sec an individual would have an intake of $2.5x10^{-3}$ Ci. Assuming 75% uptake of Cs [17] and dose conversion of $.86x10^{-4}$ millirem for each picocurie uptake[18] (i.e. body burden) of Cs^{134} + Cs^{137} we find a dose commitment to the individual at one kilometer of 160 rem. Assuming subsequent evacuation, we do not consider further exposure. Nevertheless, the radiation dose calculated is not received all at once, but is governed by a biological half-life of roughly 70-100 days.

At the dose level calculated, serious ill effects and some fatalities can be expected. A 300 rem sudden dose causes 50% fatalities.[19] The extended dose considered here would be much less lethal. Fetuses[20] and young children and other susceptible people[21] might be stricken. The ultimate dose of 160 rem implies a dose of about 1 rem/day for some time.[17] This would have significant impact on health with depression of bone marrow and lymphatic system and changes in immune responses. The probability of induced cancer within 25 years would be about 2%.[20]

The second case we consider is a windy, gusty day. Here it is most significant to consider the somatic effect of small doses of radiation suffered by people over a wide area and over a long period of time. Assuming the linear hypothesis, the BEIR report[20] concludes that the effect with respect to cancer of the sum of small doses averaging 0.1 rem/year to the population of the United States would be roughly 3500 cancer deaths/ year (within 25 years).

According to analysis of Cs^{137} from weapons testing the dose commitment conversion factor is 1.44 mrem/mCi/km^2.[22] That is, deposition at one time of Cs^{137} at the level one mCi/km^2 results in a typical person living indefinitely in that environment acquiring a dose of 1.44 mrem from external decays. To calculate the dose due to Cs^{137}, factors for shielding by buildings and body tissue and for internal decays must be included.[22] The net result is a dose commitment factor of 0.67 mrem/mCi/km^2. The internal decay dose depends on particular food chains and might not apply at the site in question; about 50% of the deaths quoted below are due to the external exposure alone. The dose from (1/3 Cs^{137} + 2/3 Cs^{134}) is less than for the same number of curies of Cs^{137}. Using an effective mean life of 14 years for Cs^{137} for external irradiation,[23] we calculate that the Cs^{134}-Cs^{137} mixture is 0.80 times as effective per Ci as Cs^{137} alone. The dose in man-rems is thus $.80 \times .67\ \rho Q$ where Q is the total Cs release in Ci and ρ is the average population per km^2.

Assuming a suburban population of 1000 persons/km^2 distributed over a very wide region downwind from the accident the dose in man-rems resulting from the assumed Cs release of 7500 Ci is $.40 \times 10^7$. This implies roughly 700 added cancer deaths within 25 years according to the BEIR result quoted above. We do not consider effects other than cancer. The calculation in this second case has the virtue of being related to empirical evidence from fallout which integrates the dose from varied pathways under actual conditions.

Land usage will be compromised by deposited cesium. The deposit of Cs^{134} is governed by its half-life of 2 years and that of Cs^{137} by an effective mean-life of 14 years on the surface. The external dose considering shielding effects of buildings and body tissues associated with radiation from ground level Cs^{137} is roughly 33 mrad per Ci/km^2 on the surface,[22] i.e., four Ci/km^2 yields an external dose comparable to background. The impact of deposited Cs on crops is small, but the effect on milk and on meat from grazing animals is extremely large during the first six months or so of deposit. A maximum dose commitment of 8 rems per Ci/km^2 of Cs^{137} is indicated for a child via the milk path.[24] About four times as much Cs is concentrated in a kg of beef as in a liter of milk.[23]

Conclusion:

A semi volatile fission product, in particular cesium, has been shown to present a serious hazard in transportation of spent fuel. The hazard is greater than that related to the noble gases and halogens at this stage of the nuclear fuel cycle. Thus, with rather similar assumptions, a release of 850 Ci of Kr^{85} is projected in a loss of cooling water event involving the IF-300 cask.[4] This amount of Kr^{85} will have very minor impact compared to the cesium release discussed in the present report. There are a variety of possibilities for corrective action at this time. For example, two aspects of the release of Cs to which the amount of release is sensitive are the use of a fluid under pressure as heat transfer medium and the decay heat rate, which in turn depends on the cooling off period for the fuel. This latter aspect would appear to create an even more serious problem for transport of spent LMFBR fuel. A more sweeping modification has been proposed to eliminate all problems associated with long range transport: geographical concentrations of nuclear facilities.[25]

Many of the assumptions made in this report are subject to great uncertainty and to variation from case to case. We have attempted to strike middleground between underestimating the problems or overestimating them. Some of the numbers estimated here are subject to considerable refinement through detailed calculation. It is hoped that these refinements will be undertaken.

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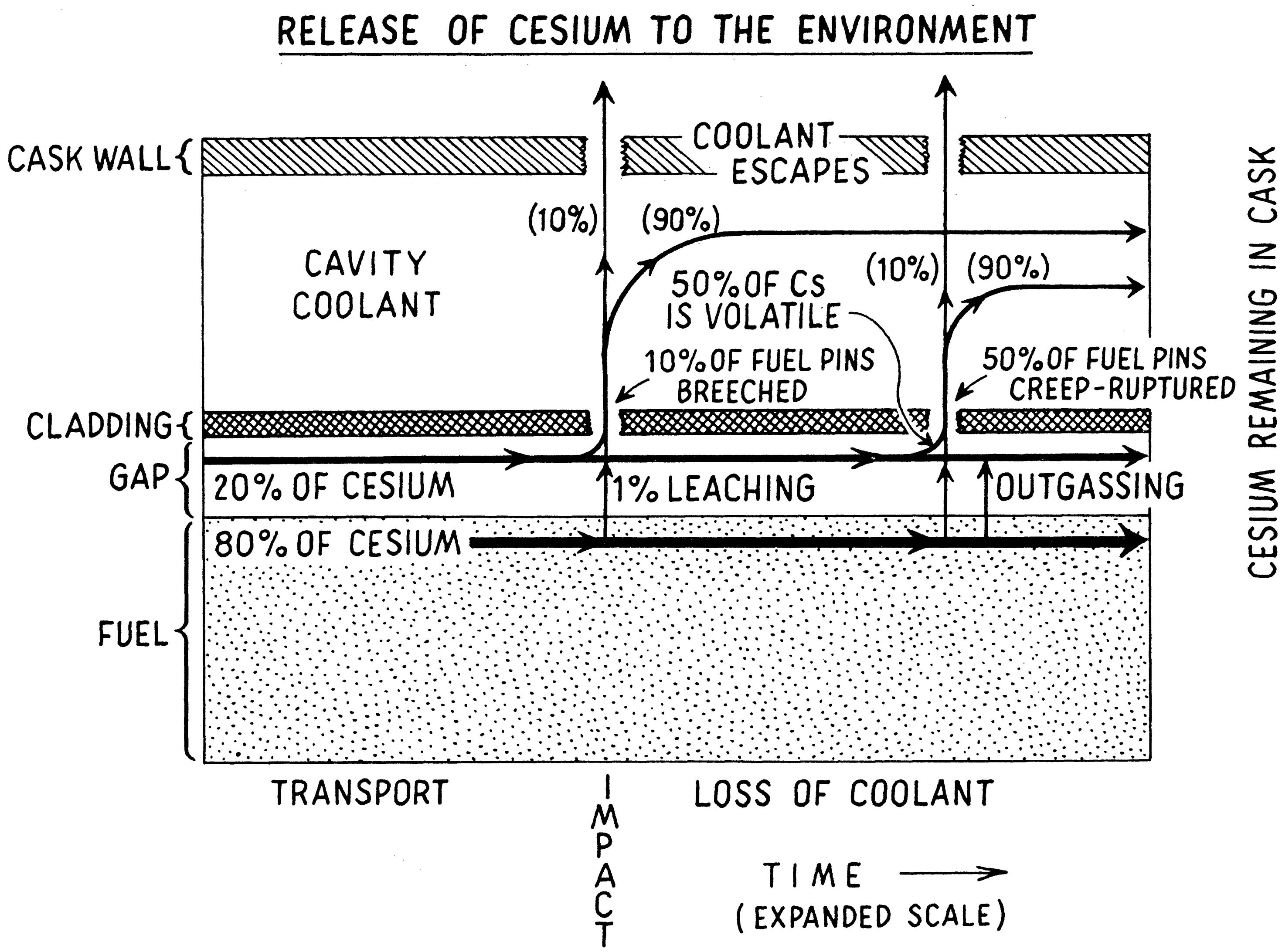

Fig. 1. Processes Considered on this Report.

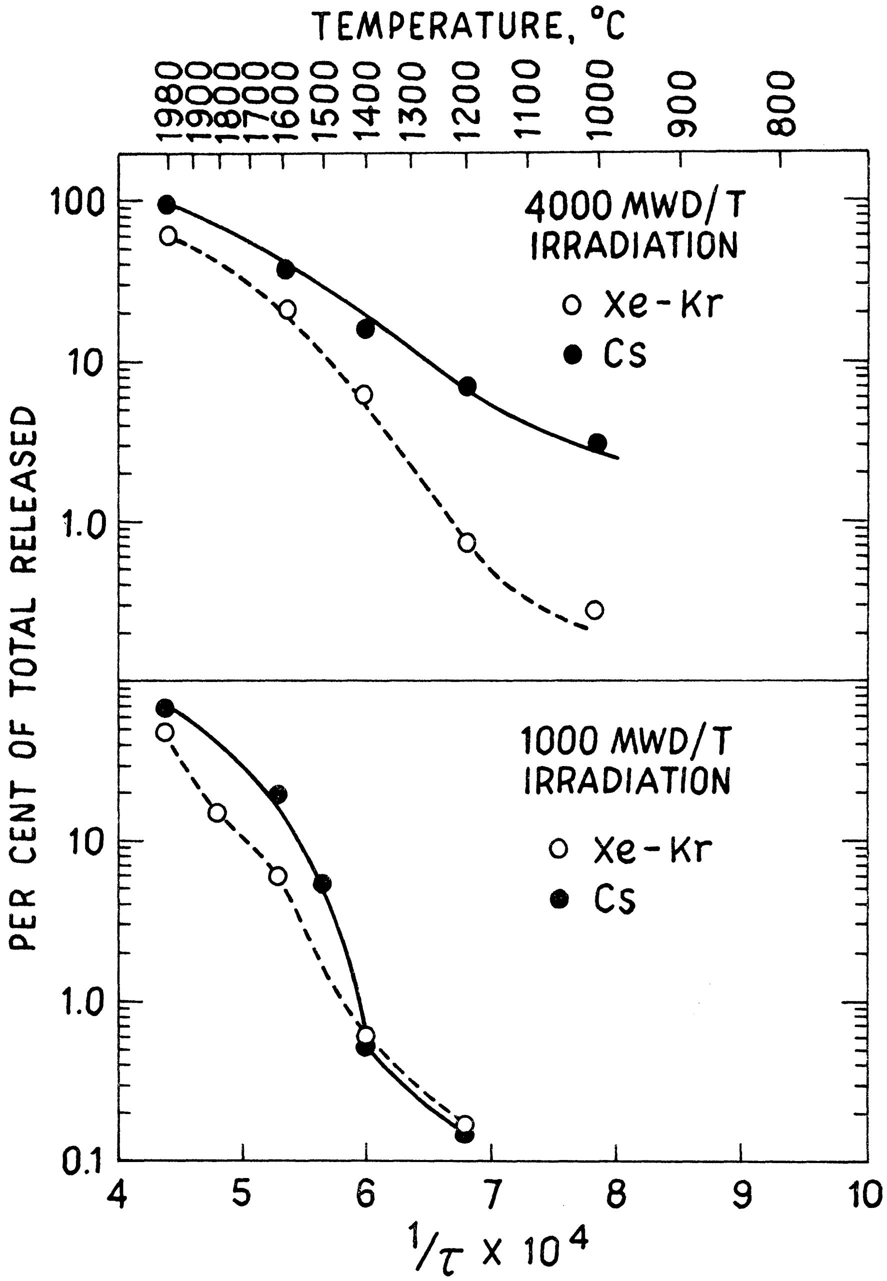

Figure 2. Release of Fission Products by Diffusion from Highly-Irradiated PWR-Type UO_2 Heated 5.5 Hours in Purified Flowing Helium. Source, reference 10.

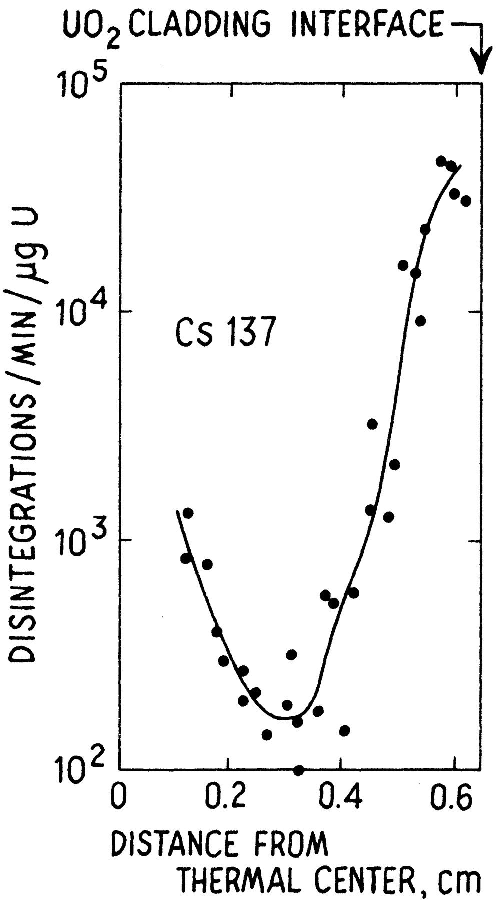

Fig. 3. Radio Chemical Analysis of Microdrilled UO_2 Sample Showing Radial Variation in Cs Distribution at Irradiation Level of 1000 to 2000 MWD/MTU. Source, reference 11.