NUREG/CR-3474

Long-Lived Activation Products in Reactor Materials

Prepared by J. C. Evans, E. L. Lepel, R. W. Sanders, C. L. Wilkerson,
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Prepared for
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Manuscript Completed: July 1984
Date Published: August 1984

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Prepared for
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Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
NRC FIN B2296

ABSTRACT

The purpose of this program was to assess the problems posed to reactor decommissioning by long-lived activation products in reactor construction materials. Samples of stainless steel, vessel steel, concrete, and concrete ingredients were analyzed for up to 52 elements in order to develop a data base of activatable major, minor, and trace elements. Large compositional variations were noted for some elements. Cobalt and niobium concentrations in stainless steel, for example, were found to vary by more than an order of magnitude. A thorough evaluation was made of all possible nuclear reactions that could lead to long lived activation products. It was concluded that all major activation products have been satisfactorily accounted for in decommissioning planning studies completed to date. A detailed series of calculations was carried out using average values of the measured compositions of the appropriate materials to predict the levels of activation products expected in reactor internals, vessel walls, and bioshield materials for PWR and BWR geometries. A comparison is made between calculated activation levels and regulatory guidelines for shallow land disposal according to 10 CFR 61. This analysis shows that PWR and BWR shroud material exceeds the Class C limits and is, therefore, generally unsuitable for near-surface disposal. The PWR core barrel material approaches the Class C limits. Most of the remaining massive components qualify as either Class A or B waste with the bioshield clearly Class A, even at the highest point of activation. Selected samples of activated steel and concrete were subjected to a limited radiochemical analysis program as a verification of the computer model. Reasonably good agreement with the calculations was obtained where comparison was possible. In particular, the presence of ^{94}Nb in activated stainless steel at or somewhat above expected levels was confirmed.

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EXECUTIVE SUMMARY

This report presents the results of a three-phase project that was designed to assess the potential problems posed to light water reactor (LWR) decommissioning by long-lived activation products produced in the major construction materials of the reactor. Reactor components investigated included the bioshield, pressure vessel, vessel cladding, and stainless steel internals.

In the first phase of the study, samples were solicited from all utilities in the United States with reactors under construction, and a large number of appropriate samples were obtained. A few samples were obtained from operating and shutdown reactors, also. Samples were analyzed for up to 52 elements by a combination of analytical techniques including instrumental neutron activation analysis, energy dispersive X-ray fluorescence analysis, and a number of special single element techniques. The stainless steel samples analyzed showed little variability in their major element composition, but varied widely in the content of activatable trace elements, notably cobalt and niobium, both of which showed concentration ranges of more than a factor of 10. Selenium, bromine, and molybdenum also showed very wide variability, but are not present in sufficient quantities to result in the production of significant levels of activation products. The vessels' steels showed no unusual compositional features. Niobium levels were, in general, much lower than in the stainless steel samples. The long-lived activation inventory should be dominated primarily by activation of iron, cobalt, and nickel. With the exception of niobium, trace elements are of negligible importance at the concentration levels found.

A wide range of compositional variation was seen in the concrete samples studied, reflecting geologic differences in the quarry sites used for the aggregate. The composition of the concrete samples studied was, on the average, very similar to that of typical crustal rock for most elements of interest. A few of the reactor sites sampled used fly ash in small amounts (4%) as a concrete additive. While this did increase the concentration of some trace elements up a factor of 2 or more, it does not appear to represent any significant problem with respect to the inventory of long-lived radionuclides. The 48 rebar samples studied showed a relatively constant composition typical of carbon steel. The rebar component dominates the bioshield composition for nickel and adds significant amounts of iron, cobalt, and molybdenum. The inventory of other elements was completely dominated by the concrete itself with the rebar of negligible importance.

The second phase of the study consisted of the calculation of expected levels of long-lived activation products in the major reactor components for various time scales. A thorough evaluation was made of all possible activation products that could be produced in sensible quantities in a LWR. Fifty-two isotopes with half-lives greater 5 five years were considered. It was concluded, that with a few exceptions the activation inventory is overwhelmingly dominated by thermal neutron capture with N-P and N-α reactions of importance only for ^{54}Mn, ^{14}C, and ^{3}T production. Photonuclear reactions are of negligible importance. A thorough review of all available cross-section information was carried out for both slow and fast neutron reactions. This information was incorporated into a multigroup computer code specifically written for that purpose. The code was used to calculate expected levels of activation products in reactor internals, vessel cladding, vessel walls, and at various depths in the bioshields. Average materials compositions taken from the first phase of the study were used in the calculations. A series of calculations was carried out for both pressure water reactor (PWR) and boiling water reactor (BWR) geometries. The majority of the activation products' inventory of the reactor was found to reside in the core shroud with only a few isotopes dominating the total activity levels. Cobalt-60 was found to dominate the total activity from 5 years to about 20 years; it dominated the gamma dose for up to 100 years. After 100 years, ^{94}Nb will be the dominant gamma emitter with a minor contribution, because of its small inner bremsstrahlung branch, from ^{59}Ni. The nickel isotopes dominate the total activity after about 20 years. A similar pattern is seen in the vessel wall itself but with a somewhat diminished importance for Co and Ni. Activation levels in the vessel wall and vessel cladding were found to be reduced by a factor of 15 for the BWR geometry. The activation patterns for the bioshield concrete was, of course, quite different with a much more complex mix of isotopes possible. On the 100-year time scale, the total activity of concrete is dominated by tritium, while on the long-term disposal time scale of hundreds to thousands of years the total activity is totally dominated by ^{41}Ca representing about 99% of the total activity. Maximum activation levels for ^{41}Ca do not appear to represent any serious hazard potential, however. Gamma activity from the bioshield is mainly due to ^{152}Eu with ^{60}Co of relatively minor importance after the first 20 years. Rebar was found to be a minor contributor to the bioshield activation inventory. For a 10% rebar component, the ^{60}Co contributions from the rebar and concrete are approximately equal. A comparison was made between calculated activation levels at various points in PWR and BWR systems and the regulatory guidelines stated in 10 CFR 61 for shallow land disposal of that material following decommissioning. Using those specifications, in general, ^{63}Ni levels become the dominant consideration. On that basis, the PWR shroud exceeds the Class C limits for near-surface disposal while the core barrel approaches these limits. A similar situation exists for the BWR shroud. Control rods are difficult to model and will require a separate detailed study. At least from a direct activation standpoint, the remaining massive components will qualify as Class A or B waste. Some surface decontamination may be required to remove other sources of activity, however. Activity inventories for the total reactor system were calculated for shutdown after 30 EFPY of operation. For the PWR case, the total was 1.7 million curies of isotopes with half lives greater than 5 years with only 1700 Ci in the bioshield. For the BWR case, 530,000 Ci were estimated for the reactor internals with 146 Ci in the bioshield. The results of this work are fully consistent with a number of previous studies. It appears that all significant activation products have been satisfactorily considered in the decomissioning strategies currently under consideration.

The third phase of the work consisted of a radiochemical analysis of neutron activated samples of steel and concrete. This was intended as a check on the calculation method.

Within the framework of the samples available, only a limited verification of the predictive capability was possible. In the steel samples, ^{59}Ni, ^{63}Ni, and ^{93}Mo were found to be present at approximately the expected levels based on a neutron flux normalization using the measured ^{55}Fe levels to adjust the predictions. The presence of ^{94}Nb in stainless steel samples was verified, and its measured levels were somewhat higher than expected. The discrepancy appears to be attributable to epithermal capture in the core region for those particular samples. For the concrete samples, the dominant role of the europium isotopes was verified with good agreement between measurement and calculation obtained for a number of shorter-lived isotopes. It was not possible to determine ^{41}Ca activation in the rather low-level samples used in this study.

1.0 INTRODUCTION

This report presents the results of a study sponsored by the U.S. Nuclear Regulatory Commission (NRC) to investigate the problems posed to eventual decommissioning of light water reactors (LWR) by nuclear activation of major reactor construction materials to isotopes with half-lives of importance on the time scale for decommissioning and disposal. This information is of importance in the design of decommissioning strategies and in the formulation of regulatory guidelines for implementing those strategies. The program is divided into three major tasks. The first task consists of the assessing of the levels of major, minor, and trace elements in the major construction materials used in pressure water reactor (PWR) and boiling water reactor (BWR) construction. These materials consist primarily of concrete (cement, fly ash, sand and aggregate), rebar, vessel steel and stainless steel internals. Samples of these materials were solicited from all utilities in the United States with reactors under construction, as well as from a number of operating reactor sites. A comprehensive program of chemical analysis was carried out on these samples for up to 52 elements. The second task consists of estimating expected levels of activation products produced in materials of representative composition as defined by the chemical analysis work. A thorough assessment was made of all potentially important nuclear activation pathways for more than 40 isotopes with half-lives ranging from a few years to tens of millions of years for incorporation into a computer model so that no important activation product would be overlooked. Finally, samples of activated material were obtained from several operating or decommissioned reactors and were subjected to radiochemical assay for selected long-lived isotopes.

1.1 REVIEW OF PROPOSED DECOMMISSIONING STRATEGIES

According to a recent U.S. Department of Energy (DOE) survey (U.S. DOE 1981) there are now a total of 75 central station electric power reactors operating in the United States with an additional 81 under construction. Although that number is now reduced due to project terminations, it is still large. The operating life span of these reactors is estimated to be of order of 40 years. These reactors will, at some time in the future, represent a problem with regard to the ultimate disposition of the massive components of the reactor station. This problem is complicated by the presence of substantial amounts of activation products formed over the life of the reactor. These activation products begin decaying immediately upon shutdown and have a wide range of half lives. The type of problem caused by specific isotopes is to a large extent a function of the particular decommissioning strategy chosen. According to Smith, Konzek and Kennedy (1978) "...decommissioning is defined, for a nuclear facility, as the measures taken at the end of the facility's operating life to assure the continued protection of the public from any residual radioactivity or other potential hazards present in the facility." Several basic approaches have been considered. The first approach involves immediate dismantlement over a 4-year period, shortly after final cessation of power production operations. This was basically the approach used in the Elk River decommissioning, and more recently, in the Sodium Reactor experiment. A number of decommissioning projects currently being initiated have also opted for immediate dismantlement. Examples include the British Windscale Advanced Gas-Cooled Reactor and the Japanese JPDR, as well as the Shippingport station, which will be the largest such project attempted to date. A significant advantage of this procedure is the continuity on site of staff familiar with all aspects of the reactor operation. An alternative to the first approach is some type of safe storage with a deferred dismantlement. The deferrment would typically be of the order of 30 to 50 years. The principal purpose of the deferral period is to allow decay of residual radioactivity to more manageable levels. The first option may, in fact, be more cost effective, while the second offers reduced risk of exposure to both the decommissioning workers and the public. The goal of both of these options is the removal of a potential long-term hazard from the immediate environs and, ideally, the return of the land to unrestricted use. The remaining option to be considered is permanent in-place entombment. This option involves sealing all of the remaining highly radioactive components in a structural integral within the biological shield after removal of fuel assemblies and other readily removable high activity components. This option was generally considered to be rather attractive until recently. This type of option was, for example, originally implemented at the Pathfinder reactor near Sioux Falls, South Dakota, with conversion of the turbine to oil-fired peak load use. Permanent entombment does, however, require a long-term commitment to a surveillance and monitoring program, as well as the guarantee of complete structural integrity for an indefinitely long period of time. This is necessary because of the presence of a large inventory of isotopes with very long half-lives, such as ^{59}Ni ($T_{1/2}$ = 80,000 y) and ^{94}Nb ($T_{1/2}$ =20,000 y) which do not undergo significant decay on a historical time scale. Thus, permanent onsite entombment is no longer generally considered to be a viable ultimate solution to the decommissioning problem.

In the interest of simplicity and clarity the NRC has recently adopted the following definitions and pseudoacronyms to define the options involved (Calkins 1982):

- Decommission means to remove the property safely from service and dispose of the radioactive residue to allow unrestricted use of the property.

- DECON means to immediately remove all radioactive material to permit unrestricted release of the property.

- SAFSTOR means to fix and maintain property so that risk to safety is acceptable for the period of storage, followed by decontamination and/or decay to the unrestricted level.
- ENTOMB means to encase and maintain property in a strong and structurally long-lived material (e.g., concrete) to assure retention until radioactivity decays to an unrestricted level.

A thorough review of the practical considerations involved in nuclear facilities decommissioning is given in the U.S. Department of Energy (DOE) Decommissioning Handbook (Manion and LaGuardia 1980). A technical and economic assessment has been carried out for PWR's by Smith, Konzek, and Kennedy (1978) and for BWR's by Oak et al. (1980).

In reviewing the expected problems posed to decommissioning by activation products, it becomes readily apparent that the particular isotopic mix responsible for a given level of problem is to a large extent related to the type of decommissioning scenario considered. If immediate dismantlement is chosen, isotopes with half-lives down to one year or less will be present in significant quantities. With the deferred dismantlement option it is unlikely that isotopes with half-lives shorter than that of ^{60}Co ($T_{1/2}$ = 5.3 y) would be of significant interest. In both cases, much longer-lived isotopes must be considered as posing a potential disposal hazard. In general, most of the effort in the area of nuclear waste disposal has been focused on problems associated with stabilization of wastes generated by the fuel cycle. While the activity levels produced by the fuel cycle are of course many times higher than those resulting from activation products alone, fuel cycle waste is inherently more compact and thus amenable to a different type of disposal technology, such as vitrification or retrievable storage. The massive components comprising the bioshield, vessels, and internals by their very bulk require a completely different disposal treatment such as near-surface burial. Thus, each component must be considered independently with regard to the risk posed by release of long-lived isotopes.

1.2 LONG-LIVED ACTIVATION PRODUCTS

1.2.1 Scope of Problem

Long-lived activation products are defined in this context as any radionuclide product of a half-life greater than 5.3 years which can be produced in sensible quantities in a nuclear reactor by the bombardment of neutrons or gamma rays upon a suitable target material. The choice of 5.3 years, the half-life of ^{60}Co, is somewhat arbitrary. It is based on the expectation that on a time scale of a few years, ^{60}Co will be of dominant importance. This simply serves to delimit the discussion somewhat. Table 1.1 gives a list of 52 isotopes conforming to that definition. A wide variety of nuclear reaction exit channels were considered. These include N-γ, N-2N, N-P, N-α, N-D, N-T, N-N^1, neutron induced fission (N-F) and photonuclear reactions. The list is by no means exhaustive since a number of the more exotic reactions (including reactions induced by secondary protons and complex multiple neutron capture reactions) are in principle possible, but of negligible importance. Most of the isotopes shown in Table 1.1, are in general, of negligible importance because of a variety of nuclear properties and target abundance considerations. This listing simply serves to define the magnitude of the problem. A complete discussion of the nuclear considerations involved is given in Section 2.1. It is evident from an examination of Table 1.1 that a wide variety of isotopes can be produced from the abundant neutron fluxes present under typical nuclear reactor operating conditions. In practice, only a few isotopes are expected to totally dominate the inventory of activation products for any given type of construction material. This is due primarily to the abundance patterns of target elements found in the materials of interest. It should be noted that the product isotope is not necessarily the same chemical element as the target isotope even for direct neutron capture reactions. Technetium-99 ($T_{1/2}$ - 213,000 y) for example, can be produced in good yield from Mo, a major constituent of Type 304 stainless steel, the most commonly used component of reactor internals. This isotope is normally considered as a fission product in fuel cycle considerations. It is the function of this program to sort out various considerations of that type while ensuring that no important activation product is overlooked in future discussions of reactor decommissioning strategy.

1.2.2 Review of Existing Information

Comprehensive information on inventories of long-lived activation products in power reactors is relatively limited to date. Emphasis has tended to be placed mainly on a short list of isotopes, such as ^{60}Co and ^{55}Fe. One reason for this is that practical experience with decommissioning of large nuclear facilities is rather limited; however, DOE and a number of utilities are planning several large dismantling projects so this may change somewhat in the near future. Most of the smaller power reactors that have been decommissioned to date used at least temporarily the SAFSTOR or ENTOMB options. Estimates of total radioactive inventory are made as part of the radiological safety analysis for entombment. For example, the Hallam Nuclear Power Facility was estimated to contain 300,000 Ci of activation products at the time of its entombment (Atomics International 1970). The total radioactivity sealed inside containment at the Piqua Nuclear Power Facility was estimated at 260,000 Ci (Atomics International 1969). The analysis conducted for the entombment of the BONUS Facility concluded that about 50,000 Ci of activity were entombed consisting of 71% ^{55}Fe, 29% ^{60}Co, and <1% ^{63}Ni (Puerto Rico Water Resources Authority 1970). These are fairly representative examples of the type of

TABLE 1.1. Long-Lived Activation Products with Half-Lives Greater than 5 years

Isotope	Half-Life (Years)[(a)]	Target Element	Principal Production Mode
^{3}H	12.3	Li	N-α
^{10}Be	1.6E6	Be	N-γ
		B	N-P
^{14}C	5730	N	N-P
^{26}Al	7.2E5	Al	N-2N
^{36}Cl	3.01E5	Cl	N-γ
		K	N-α
^{39}Ar	269	K	N-P
^{41}Ca	1.03E5	Ca	N-γ
^{53}Mn	3.7E6	Fe	N-D
^{59}Ni	80000	Ni	N-γ
^{60}Co	5.272	Co	N-γ
^{63}Ni	100	Ni	N-γ
		Cu	N-P
^{79}Se	65000	Se	N-γ, N-2N
		Br	N-P
		U, Th	N-F
^{81}Kr	2.1E5	Sr	N-α
^{85}Kr	10.7	U, Th	N-F
		Rb	N-P
^{90}Sr	29	U, Th	N-F
^{93}Zr	9.5E5	Zr	N-γ
		U, Th	N-F
^{92m}Nb	2.7E7	Nb	N-2N
		Mo	N-P
^{93m}Nb	12	Nb	N-N^{1}
^{93}Mo	3500	Mo	N-γ
^{94}Nb	20000	Nb	N-γ
		Mo	N-P
^{97}Tc	2.6E6	Ru	N-γ
^{98}Tc	4.2E6	Ru	N-P
^{99}Tc	2.13E5	Mo	N-γ
		U, Th	N-F
^{107}Pd	6.5E6	Pd	N-γ
^{108m}Ag	130	Ag	N-γ
^{113m}Cd	14.6	Cd	N-N^{1}
^{121m}Sn	50	Sn	N-γ
		Sb	N-P
^{129}I	1.59E7	Te	N-γ
^{133}Ba	10.4	Ba	N-P
^{135}Cs	2.3E6	Ba	N-P
		U, Th	N-F
^{137}Cs	30.1	Ba	N-P
		U, Th	N-F
^{137}La	60000	Ce	N-γ
^{145}Pm	18	Sm	N-γ
^{146}Sm	1.0E8	Sm	N-2N
^{150m}Eu	36	Eu	N-2N
^{150}Gd	1.8E6	Eu	N-2N
^{151}Sm	93	Sm	N-γ
^{152}Eu	13	Eu	N-γ
^{154}Eu	8.6	Eu	N-γ
^{158}Tb	150	Tb	N-2N
^{163}Ho	33	Er	N-2N
^{166m}Ho	1200	Ho	N-γ
^{178m}Hf	30	Hf	N-γ
^{192m1}Ir	241	Ir	N-γ
^{193}Pt	50	Pt	N-γ
		Pt	N-2N
^{205}Pb	1.4E7	Pb	N-γ
		Pb	N-2N
^{208}Bi	3.68E5	Bi	N-2N
^{210m}Bi	3.5E6	Bi	N-γ
^{233}U	1.58E5	Th	N-γ
^{236}U	2.3E7	U	N-γ
^{237}Np	2.14E6	U	N-2N
^{239}Pu	24390	U	N-γ

(a) E6 = X 10^{6}

information available from past decommissioning activities. The one exception is the Elk River Reactor, which until recently was the only power reactor in the United States to be completely dismantled. The dismantling operation of the 58.2 MW_{Th} reactor was successfully completed between 1972 and 1974 (United Power Association 1974). An estimated 10,000 Ci of activated material was disposed of during the decommissioning (Rural Cooperative Power Association 1971). About 90% of the activity was present in the core shroud with most of the remaining 10% in the vessel. In addition, 1224 m^3 of concrete and 1377 m^3 of soil containing low levels of activity were removed for disposal (Rural Cooperative Power Association 1971). As part of that work, a core was taken of the bioshield and subjected to gamma ray analysis. Approximately equal amounts of ^{60}Co and ^{152}Eu were found with minor amounts of ^{22}Na. Smith, Konzek and Kennedy (1978) in their reevaluation of that data have commented on the improbability of finding ^{22}Na under those conditions and were surprised they did not see ^{154}Eu. They were correct on both accounts, because the principal gamma ray energy of ^{154}Eu is within 0.1 KeV of the principal gamma ray of ^{22}Na and unresolvable even with the high resolution detectors available today. This work is mainly of importance in that it verifies the importance of the europium isotopes in activation of bioshield concrete.

More recently, the Sodium Reactor Experimental (SRE), a small experimental power reactor located near Los Angeles, was successfully decommissioned (Kittinger, Ureda and Conners 1982). In the course of this work it was determined that only the inner 25 cm of the bioshield contained sufficiently high levels of activation products to require disposal as low level waste. The unactivated portion could thus be safely buried in place. The total volume of contaminated solids removed to offsite burial was 3850 M^3.

Perhaps the most complete combined experimental and theoretical analysis of activation products was carried out by Woolam (1978) and Woolam and Pugh (1978) for British Magnox reactors. These reactors are gas-cooled graphite reactors of vastly different design from the PWR and BWR designs used in the U.S. Nevertheless, some aspects of this study are very relevant. The study consisted of two parts. In the first phase reported by Woolam (1978), several samples of activated mild steel and concrete were taken from accessible areas during a scheduled outage. The steel samples were subjected to radiochemical analysis to determine the levels of ^{59}Fe, ^{54}Mn, ^{60}Co, ^{110m}Ag, $^{124,125}Sb$, ^{63}Ni, ^{94}Nb, ^{108m}Ag, ^{152}Eu and ^{166m}Ho. The concrete samples were analyzed by gamma spectrometry for $^{152,154,155}Eu$, ^{60}Co, ^{59}Fe, ^{160}Tb, ^{182}Ta, ^{181}Hf, ^{51}Cr, ^{124}Sb, ^{134}Cs, ^{85}Sr, ^{46}Sc and ^{65}Zn. A partial chemical analysis was also carried out on the samples to attempt to reconcile the measured activities with neutron transport calculations. It was only possible to do this in a somewhat qualitative manner since the samples available were from regions not readily amenable to accurate neutron transport calculation, a persistent difficulty in the present work as well. It is rather clear that this work could be best accomplished during an actual decommissioning project. Woolam (1978) used a pair of indices which he defines as the dose contribution and disposal toxicity indices for prioritizing the relative importance of various isotopes on different time scales. The indices were used to treat independently the problems of dismantling and disposal. The results of this study indicated that on a 10 year time scale the dismantling dose would be dominated in mild steel by ^{60}Co, but in concrete, ^{152}Eu would dominate. On a 100-year time scale ^{108m}Ag and ^{94}Nb tend to dominate in steel, whereas ^{152}Eu is still the most important dose producing isotope in concrete. The situation is considerably different from the standpoint of debris disposal, with ^{63}Ni dominating the situation for a 10 to 100-year time scale in steel, and ^{59}Ni important on a greater than 1000-year time scale. Europium-152 dominates the disposal problem for concrete on a 10-year scale, but on the longer time scale of 100 years or more, the very long-lived ^{41}Ca dominates. The indices for dose and disposal toxicity advocated by Woolam (1978) appear to be useful for prioritizing the relative importance of various isotopes. The dose toxicity index approach has consequently been adapted to the present work as well. The second stage of the Magnox Project as reported by Woolam and Pugh (1978) is more theoretical in nature. Representative chemical compositions of major construction materials were combined with appropriate cross sections and neutron transport information to calculate expected activation levels in stainless steel, mild steel, graphite, and concrete. These data were then used to calculate activation levels and dose rates as a function of shutdown time. A detailed inventory of radioactive components was worked out for time scales of 10 and 100 years. Isotopes included in the inventory include ^{14}C, ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{94}Nb, ^{108m}Ag, ^{3}H, ^{36}Cl, ^{41}Ca, ^{151}Sm, ^{152}Eu and ^{154}Eu.

A similar, though more limited study, has recently been carried out on the decommissioned Gudremmingen Unit A Nuclear Power Station in the Federal Republic of Germany (Bergemann et al. 1982). This study included chemical and radiochemical analysis of bioshield cores, Charpy-V-notch samples of vessel steel and samples of the austenitic steel feedwater sparger ring. Steel samples were analyzed for ^{60}Co, ^{54}Mn and ^{63}Ni. Bioshield samples were analyzed for ^{60}Co, ^{54}Mn, ^{63}Ni, ^{134}Cs and ^{152}Eu. Neutron flux calculations by one- and two-dimensional neutron diffusion codes were used to calculate expected activation levels in those samples. In general, excellent agreement was obtained. This demonstrated the reliability of the calculation method provided adequate chemical information is available. This study showed a total activity inventory for the reactor pressure vessel and internals of 1.4×10^{6} Ci 6 years after shutdown. It was also found that activation of the bioshield above the limit for unrestricted release in the Federal Republic of Germany (FRG) was confined to the inner concrete ring, crossing the 10^{-10} Ci/g level at about 110 cm shield thickness.

The most comprehensive approach to the activation products problem carried out in the United States was that adopted by Smith, Konzek, and Kennedy (1978) as part of their PWR Decommissioning Assessment Program. A similar approach was also used by Oak et al. (1980) in the parallel program applied to a BWR. Detailed neutron transport calculations were carried out for the reference reactors using the one-dimensional diffusion code ANISN (Engle 1967). Representative compositions of mild steel, stainless steel and concrete were obtained from a variety of literature sources. The neutron transport data, collapsed into three energy groups, were combined with the composition data to calculate expected activation levels for 30 effective power years (EFPY) via code ORIGEN (Bell 1973). By integrating over the axial flux distribution, an approximate radionuclide inventory was obtained for a variety of long-lived activation products including ^{3}H, ^{14}C, ^{36}Cl, ^{39}Ar, ^{41}Ca, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{93m}Nb, ^{94}Nb, ^{93}Mo, ^{99}Tc and ^{152}Eu as well as a number of shorter-lived isotopes. The total estimated activity in neutron-activated components at shutdown after 30 EFPY was about 5 million curies for a PWR and slightly higher for a BWR. From these calculations it was shown that the decay of ^{63}Ni controls the total specific activity after about 15 years, while ^{60}Co dominates the dose rate for up to 80 years. After that time the dose rate is determined by ^{94}Nb. They concluded that the radiation dose rate from the internal components of the reactor would remain above acceptable levels for thousands of years and for that reason permanent entombment (ENTOMB) is not a satisfactory decommissioning approach. These two studies provided a reasonably good understanding of the activation products situation in large LWR nuclear generating stations. It is the purpose of the present program to supplement that information with a better data base of compositional information, as well as to provide confirmation of the presence of such isotopes at ^{94}Nb.

1.2.3 Description of Research Program

To supplement information gaps in the programs previously described in Section 1.2.2, a three-phase program was carried out to provide a more focused approach to the long-lived activation products problem. Central to the goals of the program was the need to assure that no important mode of activation capable of having an adverse effect on decommissioning planning was being overlooked. One of the major areas of uncertainty in the Smith, Konzek and Kennedy (1978) assessment was the lack of comprehensive compositional information on the major construction materials of interest. In particular, information on key trace elements such as europium was unavailable. In addition, where reference compositional information could be inferred, the range of variability was poorly known. The present program was designed to correct some of these deficiencies by providing major, minor, and trace element analyses on samples of concrete, rebar, structural steel, vessel steel, vessel cladding, and stainless steel internals. All utilities in the country with reactors under construction were contacted and samples of opportunity obtained with good geographical diversity. In addition to whole concrete samples, individual concrete components including cement, sand, aggregate, and fly ash were obtained to permit assessment of the relative importance of various components in the concrete mix in producing long-lived activation products. In addition to the construction or archive samples obtained through the utilities, a number of samples including activated concrete and steel were obtained from operating or shutdown reactors. These samples were taken as part of a second related program aimed at assessing all other sources of radioactive contamination present in LWR stations. Shutdown reactors sampled include Pathfinder, Humboldt Bay, and Indian Point No. 1. Samples were also taken at two operating plants, Turkey Point and Monticello, during scheduled shutdowns. A chemical analysis for up to 52 elements was carried out on these samples by a combination of X-ray fluorescence and instrumental neutron activation analysis supplemented by special single element techniques on selected samples.

To utilize the chemical analysis results obtained in the first task for calculating expected activation levels, a computer code (ACTIV) was written to satisfy the needs of that task. Initial attempts to carry out these calculations using the three group generalized activation code ORIGEN proved unsatisfactory due to the inadequacy of its data base for long-lived isotopes. Rather than attempt to adapt this rather large and cumbersome code to the needs of this program, a simpler series of semi-interactive codes were written to run on a PDP-11 or VAX minicomputer. The program accesses a series of composition, flux, and cross-section libraries, which can be combined in any appropriate order, allowing complete flexibility of use and relative ease of data base maintenance. A careful evaluation of the best available cross-section information was made in constructing and maintaining these libraries and reevaluated several times before carrying out final calculations. Neutron flux information used in these calculations was taken from the ANISN calculations of Smith, Konzek and Kennedy (1978) and Oak et al. (1980) supplemented by the code DOT calculations of Gritzner et al. (1977).

The third phase of the program was designed to verify the presence of long-lived activation products in selected samples representative of the major construction materials involved. Samples obtained for radiochemical analysis include stainless steel internals, vessel steel and bioshield concrete. Samples were first subsampled in order to reduce them to manageable levels of activity for laboratory analysis. They were then subjected to nondestructive gamma ray analysis. Following nondestructive analysis, the samples were digested and subjected to a series of single element radiochemical separations to assay levels of ^{60}Co, ^{63}Ni, ^{59}Ni, ^{94}Nb, ^{108m}Ag, ^{152,154}Eu, ^{93}Mo and ^{151}Sm.

2.0 REVIEW OF NUCLEAR PROPERTIES

To fully understand the setting in which nuclear activation occurs in a power reactor, a thorough understanding of the nuclear considerations involved is useful. This will be reviewed both from the standpoint of general reaction systematics as well as from a standpoint of isotope-specific properties for some of the more important activation products.

2.1 ACTIVATION REACTIONS

2.1.1 N-γ

In general, the most important type of activation reaction is thermal neutron capture in which an isotope of mass A captures a neutron to produce a product of mass A+1, which can, in some instances, be the short-lived parent of a longer-lived daughter at atomic number Z+1 or Z-1. Capture rates are normally maximal in the thermal region, which, at room temperature, is 0.025 eV. Neutron capture cross sections are defined for a thermal region (0 to 0.5 eV) and an epithermal region (0.5 eV-1 MeV). Neutron capture rates can vary enormously from isotope to isotope, depending upon nuclear structure factors and can span some nine to ten orders of magnitude in extreme cases. This range of variability is illustrated in Figure 2.1 for some selected cases.

In the epithermal region between 1 eV and a few KeV, especially for elements with intermediate and high mass numbers, there are often particular energies for which the interaction rate is exceptionally high. The rate of occurrence of a particular reaction in this resonance region is proportional to the integral of the cross section as a function of neutron energy multiplied by the flux density. This is referred to as the resonance integral. In the case of LWRs, neutrons are not well thermalized, particularly near the in-core region. This results in a rather large contribution from the resonance integral for some isotopes. Unfortunately, resonance integrals are not always well known, particularly for very long-lived isotopes. In that case it is necessary to assume a smooth background contribution to the resonance integral comprising a factor of 0.45 times the thermal cross section. This represents a minor source of uncertainty in calculating expected capture rates. Figure 2.2 shows the relative importance of the resonance contribution for some key isotopes.

2.1.2 N-2N Reaction

Neutrons produced by fission have a distribution of energies ranging up to about 15 MeV. These high-energy neutrons are eventually slowed down to thermal energies by repeated elastic and inelastic scattering interactions with the moderating medium. At most points in the reactor outside of the core, the flux in the high-energy region is considerably reduced over the low energy region.

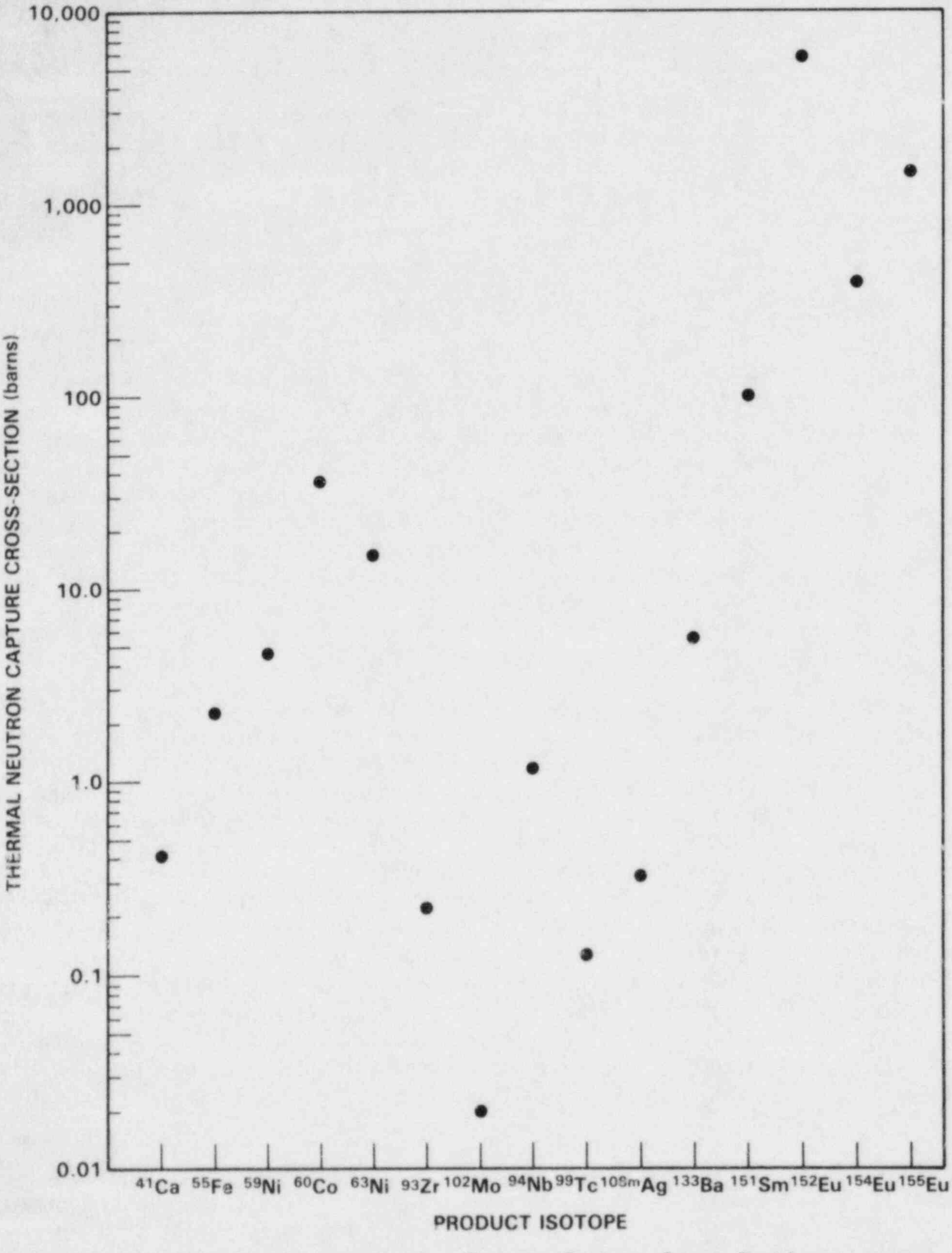

FIGURE 2.1. Representative Neutron Capture Cross-Sections for Selected Isotopes

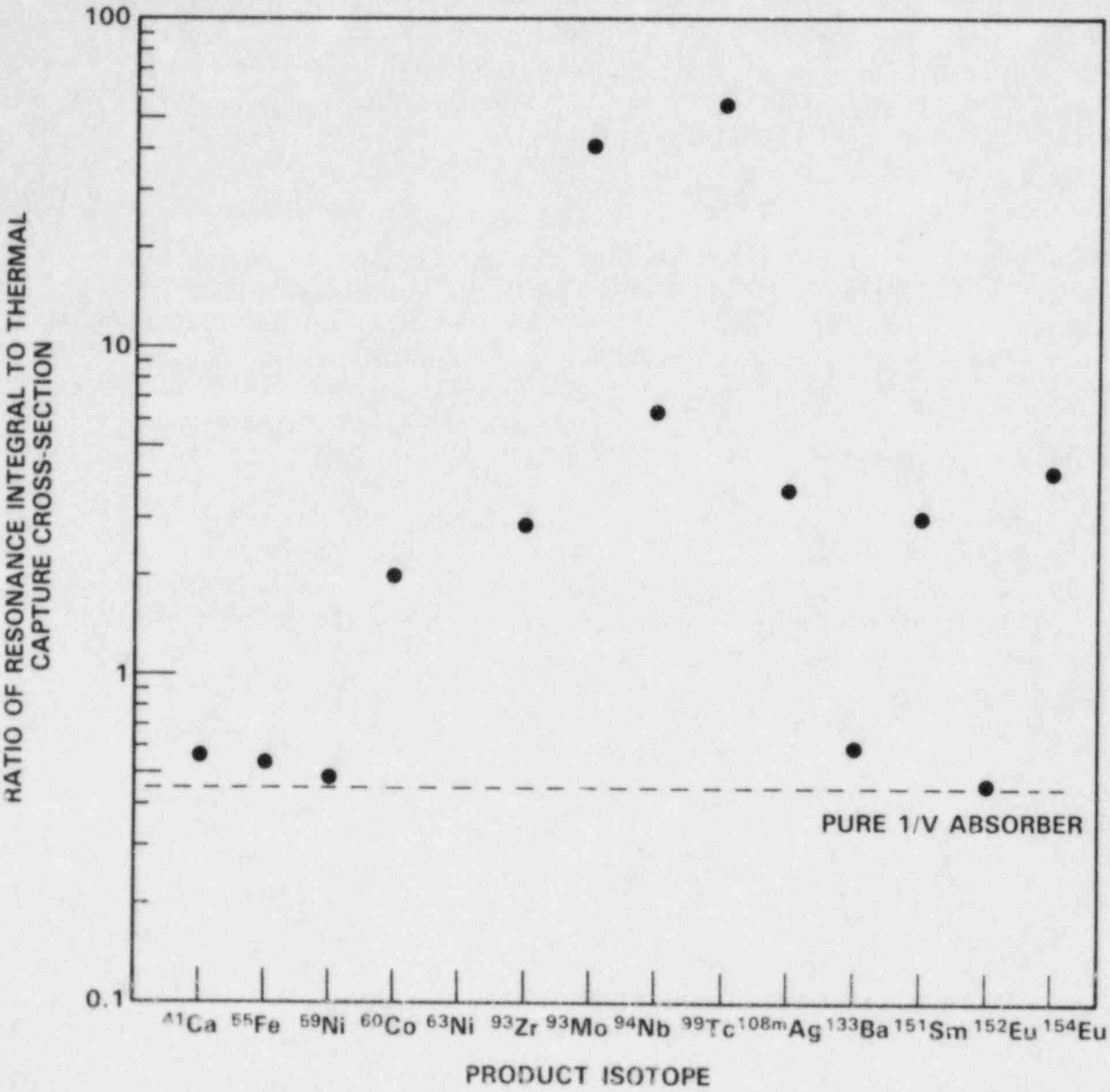

FIGURE 2.2. Relative Contribution of Resonance Capture for Selected Isotopes

Nevertheless, neutrons in that region are capable of inducing activation reactions, one of the more prominent of which is the N-2N reaction with the product nucleus of mass A-1. The threshold for the reaction is dominated by the neutron-binding energy, consequently, this type of reaction is typically limited to energies above about 8 MeV and is thus relatively unimportant compared to neutron capture. N-2N reactions are mainly of interest in the high mass region where cross sections above the threshold can exceed two barns.

Table 2.1 shows a number of possible N-2N reactions and their respective threshold energies. Most involve relatively exotic target elements. In addition, some of the possible N-2N reactions shown compete with thermal neutron capture and thus result in a trivial contribution. An example would be ^{60}Ni(n,2n)^{59}Ni versus ^{58}Ni(n,γ)^{59}Ni. The latter reaction is expected to dominate by at least three or four orders of magnitude.

2.1.3 γ-N

In addition to the copious flux of neutrons present in an operating reactor, there is also a very high flux of photons or gamma rays produced by a variety of processes including fission, thermal neutron capture, radiative neutron capture, radioactive decay and beta induced bremsstrahlung. In principle, the high energy end of the photon spectrum is capable of producing some minor level of activation by the photonuclear evaporation process. The same threshold considerations apply as for N-2N reactions since the net reaction is identical. Photonuclear reactions are coupled via the electromagnetic rather than the strong interaction and consequently have much lower interaction rates per particle. Neutron transport calculations carried out by Gritzner et al. (1977) for both PWR and BWR geometries show that the photon flux per energy group above 8 MeV is approximately the same as the neutron flux per energy group in the same region. Photonuclear reactions can thus be safely ignored because they are in any case in a practical sense indistinguishable from the equivalent N-2N reaction.

TABLE 2.1. N-2N Reactions

Product Isotope	Target Isotope	Abundance (%)	Q (MeV)
^{26}Al	^{27}Al	100	13.06
^{55}Fe	^{56}Fe	91.8	11.20
^{59}Ni	^{60}Ni	26.1	11.39
^{79}Se	^{80}Se	49.8	10.02
^{92}Nb	^{93}Nb	100	8.83
^{133}Ba	^{134}Ba	2.4	9.47
^{146}Sm	^{147}Sm	15.0	6.35
^{158}Tb	^{159}Tb	100	8.13
^{163}Ho	^{164}Er	1.6	8.84
^{193}Pt	^{194}Pt	32.9	8.38
^{205}Pb	^{206}Pb	24.1	8.09
^{208}Bi	^{209}Bi	100	7.46
^{237}Np	^{238}U	99.3	10.11

2.1.4 N-X (Charged Particle Reactions)

A number of nuclear activation reactions are possible at reactor neutron energies involving charged particles in the exit channel. This includes N-P, N-D, and N-α reactions primarily. These reactions typically have much lower energy thresholds than N-2N reactions and are, in most cases, exoergic. This is to some extent counter-balanced by the necessity for the outgoing particle to overcome the coulomb repulsion barrier to escape from the nucleus. Thresholds for charged particle reactions thus consist of two parts, an energy threshold term representing the mass difference between initial and final states and a coulomb barrier term. The coulomb barrier is not in itself an absolute threshold because particles of lower energy can escape through the barrier by quantum mechanical tunneling; however, it severely limits the interaction cross section below that level. Table 2.2 lists a number of representative activation reactions involving charged particles in the exit channel together with the energy threshold Q, the coulomb barrier V and the total energy threshold T. The coulomb repulsion term V is given by

$$V = \frac{Z_1 Z_2 e^2}{R_1 + R_2}$$

where $R = 1.6 \times 10^{-13}A$ 1/2 cm. Z represents the charge of the product nucleus and the outgoing particle, respectively, and A is the corresponding atomic number. R is the appropriate nuclear radius. The subscripts on Z and R are used to denote the respective nuclei. From this it can be seen that this type of reaction is mainly of importance for target elements of low atomic number. Probably the best known N-P rection is $^{14}N(n,p)^{14}C$ which, due to its relatively low total energy threshold, has an appreciable interaction cross section even at thermal energies. The most prolific N-α reaction is the well known $^{6}Li(n,\alpha)^{3}H$ reaction. This reaction has the minimum possible coulomb barrier and is highly exoergic due to the stabilizing effect of the alpha particle binding energy. N-α thresholds go up rapidly with atomic number due to the coulomb term. The only other isotope of importance potentially produced by this mechanism is ^{36}Cl. The only N-D reaction of note is that producing ^{53}Mn. It has a very high threshold due to the fact that two nucleons are removed. For the same reason N-T reactions have even higher thresholds and are not of significance at reactor energies except for the special case of the aforementioned ^{6}Li reaction in which tritium is the residual nucleus.

2.1.5 N,N^1,γ-γ^1 (Inelastic Scattering)

Another possible type of activation reaction is inelastic scattering of either neutrons or photons in which a portion of the energy of the incoming particle is absorbed by the target nucleus with no net change in nucleon number.

TABLE 2.2. Activation Reactions Involving Charged Particles

Product Isotope	Target Isotope	Abundance (%)	Reaction	Q(MeV)	V(MeV	T(MeV)
^{10}Be	^{10}B	19.8	N-P	0.23	1.14	0.91
^{14}C	^{14}N	99.6	N-P	-0.63	1.58	0.95
^{63}Ni	^{63}Cu	69.1	N-P	-0.72	5.07	5.17
^{79}Se	^{79}Br	50.7	N-P	-0.62	5.79	5.17
^{92}Nb	^{92}Mo	14.8	N-P	-0.42	6.70	6.28
^{94}Nb	^{94}Mo	9.1	N-P	1.26	6.70	7.96
^{121}Sn	^{121}Sb	57.3	N-P	-1.16	7.58	6.42
^{135}Cs	^{135}Ba	6.5	N-P	-0.58	8.09	7.51
^{137}Cs	^{137}Ba	11.2	N-P	0.39	8.09	7.70
^{53}Mn	^{54}Fe	5.8	N-D	6.63	4.74	11.37
^{3}H	^{7}Li	92.5	N-α	-5.60	0.59	-5.01
^{36}Cl	^{39}K	93.3	N-α	-1.36	6.26	4.90
^{81}Kr	^{84}Sr	0.56	N-α	-2.66	10.97	8.31
^{14}N+^{3}H	^{16}O	99.8	N-T	14.50	1.60	16.10
^{54}Mn+^{3}H	^{56}Fe	91.8	N-T	11.93	4.20	16.23

The target nucleus is raised to an excited state which can be a long-lived metastable. Two such cases are shown in Table 1.1, ^{93m}Nb ($T_{1/2}$ = 12 y) and ^{113m}Cd ($T_{1/2}$ =14.6 y). They are included for completeness.

2.1.6 N-F (Neutron Induced Fission)

The remaining type of activation process considered is neutron induced fission. This is primarily of interest in bioshield materials, which being of geologic origin can contain significant amounts of uranium and thorium. Fission activation will thus either occur due to direct fission of naturally abundant ^{235}U, or due to multiple neutron capture. Thorium-232 can capture a neutron to produce fissionable ^{233}U which produces fission products by capturing a second neutron. The same consideration applies to the ^{238}U-^{239}Pu system. Fission products produced in this manner are likely to be of minor importance compared to fuel cycle derived fission product contamination but are included for completeness.

2.2 ISOTOPE PROPERTIES

A number of activation products studies including this one have demonstrated that a few isotopes tend to dominate hazard potential in reactor construction materials. These isotopes deserve special attention with respect to production mode and decay properties.

2.2.1 Gamma Emitters

The primary level of concern about activation products from a standpoint of dismantlement revolves around isotopes emitting energetic gammas. Fortunately, this involves a relatively limited list of isotopes with half-lives on the greater than 5-year time scale.

2.2.1.1 ^{60}Co

Cobalt-60 is produced almost entirely by thermal and epithermal neutron capture on 100% abundant ^{59}Co. Although other production modes, including ^{60}Ni(n,p)^{60}Co and ^{63}Cu(n,α)^{60}Co are in principle possible, the relatively high neutron capture cross section (37 barns) and the fact that cobalt is a rather common impurity particularly in stainless steel render alternate production mechanisms insignificant. Cobalt-60 ($T_{1/2}$ = 5.27 y) decays by emission of a medium energy beta particle (0.32 MeV, 99%) to the 2.5057 MeV level of ^{60}Ni which, in turn, decays to the ground state with the emission of 1.173 and 1.332 MeV gamma rays with close to 100% abundance. This combination of properties makes ^{60}Co the dominant dose producing isotope in the reactor interior on the 10-year time scale. The production rate of ^{60}Co is sufficiently high in the high flux region near the core that a substantial portion (up to one-third) of the stable cobalt may be transmuted over the life of the reactor.

2.2.1.2 ^{152,154}Eu

The two europium isotopes are the dominant activation products in bioshield concrete on a time scale of 10 to 20 years or longer. Both have very large neutron capture production cross sections, 5900 barns for 152 and 390 barns for 154. Europium-152 is produced primarily by thermal neutrons, whereas 154 also has a rather substantial resonance integral (1635 barns). To further complicate matters, europium-152 itself has an extremely high burn-up cross section (13,000 barns) so that in regions of high flux, essentially all of the europium can be converted to 153 and 154 with the 151 and 152 totally depleted. Under lower flux conditions typical of the bioshield area, burn-up is far less important, but the expected isotope ratio is nonetheless variable, depending upon thermalization parameters. Both isotopes have particularly complex decay schemes. Europium-152 ($T_{1/2}$ = 13 y) has the somewhat unusual property of decaying both by beta emission and by electron capture. Beta decays to a number of excited states in ^{152}Gd account for 27% of the decay rate with most of the remainder populating excited states in ^{152}Sm by electron capture. The positron branch is of negligible importance. The resulting gamma cascade is very complex with intense gamma rays ranging in energy from 122 KeV to 1408 KeV. A typical gamma ray spectrum of ^{152}Eu shows more than 30 peaks greater than 1% abundant. The decay scheme of ^{154}Eu ($T_{1/2}$ = 8.5 y) is somewhat less complex decaying primarily by beta emission to a number of excited states in ^{154}Gd. Intense gamma ray peaks include those at 123, 724, 876, 996, 1005 and 1278 KeV. There are, as well, numerous less intense peaks.

2.2.1.3 ^{94}Nb

Niobium-94 ($T_{1/2}$ = 20,000 y) is produced by thermal neutron capture from 100% abundant ^{93}Nb with a cross section of 1.15 barns. It decays by beta emission with a maximum beta energy of 473 KeV to a single level in ^{94}Mo at 1574 KeV. A cascade of two 100% abundant gammas of 703 and 871 KeV each results. Since Stevens and Pohl (1977) pointed out that presence of high levels of niobium in reactor internals stainless would lead to production of significant amounts of the very long-lived ^{94}Nb, a good deal of attention has been focused on this isotope. For long deferral intervals prior to dismantlement, it may in fact represent the principal contributor to personnel exposure during dismantlement.

2.2.1.4 ^{108m}Ag

Silver-108m ($T_{1/2}$ = 130 y) is produced by thermal neutron capture on 52% abundant ^{107}Ag. The neutron capture cross section for this isotope has been revised downward considerably in recent years. Woolam (1978b) concluded from a survey of the literature that the generally accepted value of three barns was about an order of magnitude too high. The currently accepted value according to Mughabghab, Divadeenam and Holden (1981) is 0.33 ± 0.08 barns. Radiochemical measurements made by Woolam (1978a) on activated Magnox reactor components resulted in a factor of five disagreement between silver content estimates based on ^{110m}Ag and ^{108m}Ag, suggesting that the cross section may still be lower yet. It is not clear, however, in that work whether or not the rather large resonance integral to produce ^{110m}Ag was properly accounted for in the calculation. The reduced cross section considerably diminishes the importance of this isotope for general decommissioning considerations, however, the use of large amounts of silver in PWR control rods will nonetheless result in a large inventory of ^{108m}Ag in the reactor at the time of shutdown. Silver-108 m decays primarily by electron capture (91%) to ^{108}Pd resulting in a series of energetic gammas of 434, 614, and 722 KeV, respectively.

2.2.1.5 ^{133}Ba

Barium-133 ($T_{1/2}$ = 10.7 y) is produced by a combination of epithermal neutron capture reactions on 0.1% abundant ^{132}Ba. Barium-132 has a neutron capture cross section of only 0.76 barns and a resonance integral of eight barns. Normally this isotope should be of negligible importance, however, significant amounts of ^{133}Ba can be produced in concrete containing barite as a high density aggregate. This apparently was the case with the North Carolina State University research reactor currently being decommissioned, which shows relatively high levels of ^{133}Ba in the bioshield concrete. Barium-133 emits a series of relatively low energy gammas, at 276, 302, 356, and 382 KeV.

2.2.1.6 Other Gamma Emitters

In addition to the isotopes mentioned above, a number of the other potential activation products shown in Table 1.1 emit energetic gamma rays. These include ^{26}Al, ^{92}Nb, ^{137}Cs, ^{158}Tb, ^{166}Ho, ^{178m}Hf, ^{192m2}Ir, ^{193}Pt, and ^{208}Bi. These isotopes should result in a negligible dose contribution due to direct activation. There is, of course, the possibility of a large inventory of ^{137}Cs that may be present due to fuel ruptures. Other sources of gamma radiation include beta-induced bremsstrahlung and inner bremsstrahlung from electron capture isotopes. The latter effect is rather important for ^{59}Ni and will be discussed separately.

2.2.2 Pure Beta Emitters

Studies of dose effects related to decommissioning activities such as those carried out by Smith, Konzek and Kennedy (1978) have amply demonstrated that pure beta emitters do not represent a serious contribution to dose during dismantling operations due to the low penetrating power of beta radiation. In considering potential disposal hazards, however, all abundantly produced isotopes must be considered.

2.2.2.1 ^{3}T

Tritium can be produced in a reactor by several mechanisms. Neutron capture on deuterium is possible though of relatively low yield in LWR's due to the low natural abundance of deuterium (0.015%) combined with its extremely low neutron capture cross section (0.53 mb). This reaction is, of course, considerably more important in D_2O-moderated reactor designs such as the Canadian CANDU for example or in the numerous D_2O moderated research reactors around the world. N-T reactions by fast neutrons are of negligible importance. The thresholds for this reaction on the two most abundant targets present, ^{56}Fe and ^{16}O, are both over 16 MeV. Bioshield materials will, of course, contain small amounts of Li. Tritium is thus expected to be present in the bioshield due to the highly sensitive $^{6}Li(n,\alpha)^{3}T$ reaction. Tritium ($T_{1/2}$ = 12.33 y) decays by emission of a beta with an 18.6 KeV endpoint. The rather low beta energy results in a relatively low hazard potential for tritium. However, it is very mobile in the environment.

2.2.2.2 ^{10}Be

Beryllium-10 is something of an interesting curiosity as an activation product. Production by direct neutron capture on 100% abundant ^{9}Be is unlikely except perhaps in research reactors that use beryllium reflectors to produce neutron beams. In any case, the thermal capture cross section is small (8 mb). Table 1.3 reveals that the N-P reaction on ^{10}B [$^{10}B(N,P)^{10}Be$] has a similar threshold to that of the well-known N-P reaction producing ^{14}C. The best experimental value for the reactor spectrum-averaged cross section is an upper limit of 0.18 barns. Since the atomic number of beryllium lies outside of the semiempirical formulation used in this work, code THRES2, (Pearlstein 1975, Pearlstein 1973) it is difficult to estimate. The abundant use of boron for reactor power level control suggests that a considerable amount of ^{10}Be may be produced in control rods, shim rods, and in primary reactor coolant when boric acid is used as soluable poison. Beryllium-10 ($T_{1/2} = 1.6 \times 10^6$ y) decays by beta emission with a 556 KeV beta endpoint.

2.2.2.3 ^{14}C

Carbon-14 is produced in a reactor primarily by the N-P reaction on nitrogen. As the principal component of the atmosphere, nitrogen occurs as a highly variable trace impurity in virtually all materials including steel and concrete. Carbon-14 ($T_{1/2} = 5730$ y) decays by beta emission with a 156 KeV endpoint.

2.2.2.4 ^{63}Ni

Nickel-63 is by far the most abundant activation product expected to be present in a LWR on the time scale of deferred dismantlement. Its half-life of 100 years makes its production optimal for that time scale. It is produced by direct neutron capture on nickel with a 14.6 barns cross-section. The problem is somewhat minimized by the relatively low abundance of the target isotope, ^{62}Ni (3.6%). This is offset by the presence of large amounts of Ni in stainless steel in the high flux region. Nickel-63 decays by emission of a relatively low energy beta with a 66 KeV endpoint energy. Bremsstrahlung production is thus negligible for this isotope.

2.2.2.5 ^{99}Tc

Technetium-99 is normally considered in most reactor contexts as a fission product since the mass 99 isobar is essentially at the low mass peak of the ^{235}U fission yield curve at 6.1% abundance. It is a major source of concern from a standpoint of nuclear waste disposal due to its long half-life (213,000 y) and its potentially high environmental mobility. Its detection and identification is also a bit difficult. Technetium-99 can be produced as an activation product as well by neutron capture on 24% abundant ^{98}Mo to produce 66h ^{99}Mo. Molybdenum-99 in turn decays to the much longer-lived ^{99}Tc. The neutron capture cross-section of 0.14 barns and resonance integral of 6.6 barns are small but significant since molybdenum like nickel, is a major constituent of the stainless steel used in reactor internals in the high flux region. Technetium-99 decays by emission of a beta with a 298 KeV endpoint energy.

2.2.2.6 Other Beta Emitters

In addition to the isotopes specifically discussed above, Table 1.1 contains a large number of other isotopes which are essentially pure beta emitters. These include ^{36}Cl, ^{79}Se, ^{81}Kr, ^{90}Sr, ^{93}Zr, ^{121}Sn, ^{129}I and ^{135}Cs. None of these are expected to be of major importance as activation products although, as was the case with ^{137}Cs, ^{90}Sr will be present in some materials due to translocation effects from ruptured fuel and is of major importance due to its relatively high biohazard potential.

2.2.3 Electron Capture Isotopes

A number of activation products decay by capture of orbital electrons to isotopes of atomic number Z-1. A few of these deserve special discussion due to their unusual properties.

2.2.3.1 ^{59}Ni

Nickel-59 is certainly one of the most important activation products with extremely long half-life (80,000 y). It can be produced by a variety of mechanisms, however, the most important overwhelmingly is thermal neutron capture on 68.3% abundant ^{58}Ni. The high abundance of the target isotope, the large amounts of nickel present in reactor internals, and the moderately high production cross-section (4.6 barns) combine to produce ^{59}Ni in abundance particularly in reactor internals. Nickel-59 decays purely by electron capture 100% of the time to the ground state of ^{59}Co. These decays are accompanied by low energy X-rays and electrons characteristic of cobalt. Most of the transition energy is carried away by a mono-energetic neutrino. In a very small percentage of decays in the electron capture process, the transition energy is divided between a gamma ray and neutrino, a process known as inner bremsstrahlung. Normally that branch is very minor. Nickel-59 has the unusual property of having an exceptionally high transition energy for its half life, 1.07 MeV. The total number of quanta per electron capture disintegration due to inner-bremsstrahlung is approximately $7.4 \times 10^{-4}\ E^2$ where E is the transition energy in MeV. Nickel-59 thus produces a continuous spectrum of gamma rays up to 1.07 MeV energy about one in every thousand decays. Due to its rather substantial production rate in all parts of the inner containment, ^{59}Ni is thus one of the limiting isotopes for dose considerations after decay of other shorter-lived residual activity has occurred.

2.2.3.2 ^{93}Mo

Molybdenum-93 is of special interest due to the presence of large amounts of molybdenum in reactor internals. Its half-life is thought to be around 3500 years, but is poorly known. Molybdenum-93 has the unusual property that it is produced primarily by epithermal neutron capture. Since the neutron shell of the target, 14.8% abundant ^{92}Mo, is closed at 50 neutrons, its thermal capture cross-section is very low (0.02 barns). The resonance integral of 0.81 barns is also moderately low. It can also be produced by N-2N on 9.3% abundant ^{94}Mo. Since the threshold for this reaction is nearly 10 MeV it also has a rather low yield. Molybdenum-93 is thus likely to be of less significance as an activation product than originally anticipated. It decays greater than 90% of the time to a 30.4 KeV level of ^{93}Nb. That level is itself a long-lived activation product, ^{93m}Nb, which decays to the ground state by internal transition with a 13.6 y half-life.

2.2.3.3 ^{41}Ca

Calcium-41 is of particular interest due to its long physical and biological half-life. Bioshield concrete contains extremely high levels of calcium. Calcium-41 is thus likely to dominate the long-term disposal considerations with respect to bioshield concretes, the most voluminous activated component of the reactor. Calcium-41 is produced entirely by thermal neutron capture on 96.9% abundant ^{40}Ca. Since the target nucleus is doubly magic in neutron and proton number (twenty each) its neutron capture cross-section is not very large (0.41 barns) and there are no important resonances. The production of ^{41}Ca in bioshield material is minimized by the relatively low neutron flux escaping from the vessel in a LWR. Presumably, this situation would be somewhat worse for deuterium moderated or fast breeder designs. Calcium-41 decays purely to the ground state of ^{41}K with an approximate 103,000 year half-life. [The half-life is poorly known and is shown in various publications ranging from 80,000 to 130,000 y. In this work the half-life values as given in the Seventh Edition of the Table of Isotopes, (Browne, Dairiki, and Doebler 1978), have generally been adopted.] Its transition energy of 0.43 MeV is carried away by the neutrino with inner bremsstrahlung of minimal importance. Decay energy available in the form of ionizing radiation amounts to only 3.6 KeV mostly in the form of Auger electrons. According to the ICRP model, (ICRP 1959) however, calcium has a biological half-life of forty-four years and a fraction to the bone of greater than 50%. This results in a rather low calculated MPCw for this isotope (1.1×10^{-4} mCi/cm^3). In spite of its low decay energy, the MPCw is considerably lower than that of

^{60}Co, for example. The problem is further complicated by the fact that this isotope has very unfavorable decay properties for routine detection at that level. The rather low specific activity found in PWR and BWR bioshield concrete does not appear to represent any significant biohazard. However, the hazard potential of this isotope is worth noting. Materials containing significant levels of calcium such as concrete should be carefully excluded from exposure to high neutron fluxes if possible.

2.2.3.4 Other Electron Capture Isotopes

Other isotopes shown in Table 1.1 decaying purely by ground state electron capture include ^{54}Mn, ^{137}La, ^{145}Pm, ^{193}Pt, ^{205}Pb and ^{208}Bi. These isotopes are expected to be of minor importance but are included for completeness.

2.2.4 Alpha Emitters

Several isotopes included in Table 1.1 decay by alpha emission, including ^{146}Sm, ^{233}U, ^{236}U, ^{237}Np and ^{239}Pu. In general, yield of these isotopes should be very low with the possible exception of ^{239}Pu. The ^{239}Pu inventory of the reactor containment is in all probability dominated by other sources of Pu contamination unless the reactor operation is exceptionally clean and the concrete used is exceptionally high in uranium.

3.0 CONSTRUCTION MATERIALS

3.1 SOURCE OF SAMPLES

Reference analytical samples of construction materials were solicited from a variety of sources. All utilities with reactors in the construction phase were contacted and requested to provide appropriately representative samples of bioshield materials and major reactor components. A total of 27 utilities representing 69 different construction projects were contacted. The actual yield of samples proved somewhat less than anticipated, but appears adequate for the goals of the program. Samples were also rather slow to arrive due to the complex organizational structure involved in a typical reactor construction project. An alternative source of samples was operating or shutdown reactors. These samples were taken during onsite visits on a related program. In addition to utility sources, steel samples were also made available through vessel surveillance programs. A selection of Type 304 and 316 stainless steel for nuclear use was also provided by the steel manufacturers.

3.2 CONCRETE

Concrete samples were obtained from 13 reactor sites with good geographical diversity. A full listing of the samples obtained is given in Table 3.1. Samples were either test pourings used for compressive strength testing or core samples taken with a 3 in. diamond coring bit. The samples were in several cases accompanied by matching ingredients for analysis in order to determine the origin of any unusually high concentrations of trace elements. The diversity of materials used is represented by the samples obtained. The coarse aggregate used in concrete manufacture at nuclear plants is generally typical of whatever rock can be quarried locally economically. The high costs associated with the use of high density aggregate has resulted in a far more limited use than was common in the early days of nuclear reactor development. Two samples of high density aggregate appearing to be illmenite and magnetite were provided from the Susquehanna Project. Several reactor projects are also using coal fly ash as an additive to their cement. Samples were obtained from the Enrico Fermi No. 2, Hartsville , Bellefonte and Palo Verde No. 2 projects. Since coal fly ash typically tends to concentrate some normally rare trace elements, these samples are of some interest.

3.3 REBAR

In this work rebar is estimated to comprise at least 10% by weight of the bioshield. In some newer reactors it may be considerably higher. The trace element composition of rebar is thus of some interest, particularly since the

TABLE 3.1. Bioshield Materials (Unirradiated)

Reactor	Type	Rebar	Concrete	Cement	Sand	Fly Ash	High Density Aggregate	Ordinary Aggregate
Palo Verde No. 2	PWR	6	2	1	1	1		2
Enrico Fermi No. 2	BWR	4	2	1	1	1		
Wolf Creek	PWR	10	3	1	1			2
Susquehanna	BWR	1		1	1		2	1
Hartsville	BWR		2	1	1	1		3
Bellefonte	PWR	4	4	1	1	1		3
Rancho Seco	PWR	1	1					
Pathfinder	PWR		3					
WPPSS No. 1	PWR	4	4	1	2			4
Turkey Point No. 4	PWR		2					
Humboldt Bay	BWR		4					
Waterford No. 3	PWR	3	2		1			2

use of recycled materials in its composition can, in principle, result in an unusual trace element distribution. A good selection of rebar was obtained as shown in Table 3.1.

3.4 CARBON STEEL

Samples of carbon steel used for vessel wall fabrication were obtained from several sources. These samples are in general Charpy impact specimens used for vessel surveillance programs. In several cases matching irradiated samples were also available.

3.5 STAINLESS STEEL

A selection of stainless steel was obtained from several sources. Most reactor internals are constructed of Type 304L stainless steel with occasional use of 316 or Inconel for special purposes.

3.6 CONTROL RODS

No attempt was made to obtain control rod material. Due to the nature of the material it would be a formidable analytical problem and very difficult to model from an activation standpoint. BWR control rods are composed of boron carbide. They should contain a substantial inventory of ^{10}Be and ^{14}C, as well as the isotopes associated with the stainless steel cladding. PWR control rods are a mixture of Ag, Cd, and In. The inventory of activation products in PWR control rods is thus likely to be largely dominated by 252 day ^{110m}Ag in the short term and by 130 year ^{108m}Ag on the longer time scale. It would be worthwhile determining these inventories empirically; however, the sampling and analysis of control rods was beyond the resources of this program.

TABLE 3.2. Steel Samples (Unirradiated)

Supplier	Vessel Steel	Vessel Cladding	Carbon Steel Internals	Stainless Steel Internals	Inconel
Enrico Fermi No. 2				2	
Susquehanna			1	1	1
Bellefonte	1	1			
Northanna				1	
Pathfinder				1	
Battelle Columbus	1				
U.S. Steel				6	
Westinghouse	8				
Waterford No. 3				1	
Turkey Point No. 3				1	

4.0 CHEMICAL ANALYSIS PROGRAM

4.1 ANALYTICAL TECHNIQUES

An analytical program was implemented to obtain information on up to 52 elements. The program relied mainly on two multielement instrumental methods; instrumental neutron activation (INAA) and energy dispersive X-ray fluorescence (XRF). The analytical procedures used are described below.

4.1.1 Instrumental Neutron Activation Analysis (INAA)

Sample Preparation

Geological material such as sand, cement, aggregate materials, and concretes were obtained for analysis from various reactor construction sites. Aliquots of each solid material were pulverized and ground to less than 150 μm (100 mesh) using a Spex Shatterbox. The prepared samples were then stored in polyethylene bottles from which aliquots were taken for analysis by both INAA and XRF. Aliquots of 100 to 200 mg were weighed into acid-cleaned 2/27 dram polyvials for INAA analysis.

Metal construction materials such as rebar, stainless steels, and various other metal materials were prepared for analysis by machining. Aliquots of each metal material were taken as shavings or drill turnings. The samples were cleaned with alcohol, air dried, and stored in polyethylene bottles. Aliquots of 100 to 200 mg for "longs" and ~500 mg for "shorts" were then weighed into acid-cleaned 2/5 dram polyvials for analysis by INAA.

Sample Activation and Counting

The strategy used for INAA analysis of these reactor construction materials involved two irradiations and five counting periods. The first, a short irradiation ("shorts") , was followed by two counts. The second irradiation was followed by three counts with an increasing decay time for each counting period. A summary of the nuclear properties involved is given in Tables 4.1 and 4.2. A thorough review of the procedures for INAA used in this laboratory and others is given by Laul (1979).

Shorts Analysis

Samples and standards were irradiated in the ^{252}Cf-neutron multiplier facility of Pacific Northwest Laboratory. The samples were irradiated for 3 minutes at a flux of $\sim 2 \times 10^9$ n$\cdot$cm$^{-2}\cdot$s^{-1}. The irradiation and counting was controlled by a PDP 11/10 computer which was part of a Tracor Northern computer-based data acquisition system (TN-11 system) (Wogman et al. 1978).

TABLE 4.1. Production and Properties of the Nuclides Observed in a "Shorts" Irradiation

Element	Reaction	Isotopic Abundance (%)	Cross-Section (barns)	Half-Life t 1/2(min)	Intensity (γ's per 100 dis)	Major γ-Rays (keV)	Possible Interferences
First Count							
Al	$^{27}Al(n,\gamma)^{28}Al$	100.	0.231	2.24	100	1778.7	
Ca	$^{48}Ca(n,\gamma)^{49}Ca$	0.187	1.1	8.719	92	3084.4	
Ti	$^{50}Ti(n,\gamma)^{51}Ti$	5.2	0.179	5.80	93.4	319.7	316.5 (^{102}Ir), 3200 (^{51}Cr)
V	$^{51}V(n,\gamma)^{52}V$	99.75	4.88	3.746	100	1423.06	
Cu	$^{65}Cu(n,\gamma)^{66}Cu$	30.8	2.17	5.10	8	1039.2	
Second Count							
Na	$^{23}Na(n,\gamma)^{24}Na$	100	0.53	901.2	100	1368.6	
					100	2754.1	
Cl	$^{37}Cl(n,\gamma)^{38}Cl$	24.23	0.428	37.29	31.	1642.2	
					42	2167.6	
K	$^{41}K(n,\gamma)^{42}K$	6.71	1.46	741.7	18.8	1524.6	
Mn	$^{55}Mn(n,\gamma)^{36}Mn$	100	13.3	154.7	98.87	846.7	841.5 (^{152m}Eu), 843.8 (^{27}Mg)
					27.2	1810.7	
					14.3	2113.0	2112.1 (^{116m}In)
Sr	$^{86}Sr(n,\gamma)^{87m}Sr$	9.8	0.84	168.3	82	388.4	
Ba	$^{138}Ba(n,\gamma)^{139}Ba$	71.7	0.4	82.9	22	165.8	
Dy	$^{164}Dy(n,\gamma)^{155}Dy$	28.1	2700	140.0	3.58	94.7	

TABLE 4.2. Production and Properties of the Nuclides Observed in a "Long" Irradiation

Element	Reaction	Isotopic Abundance (%)	Cross-Section (barns)	Half-Life $T_{1/2}$ (days)	Intensity (γ's per 100 dis)	Major γ-Rays (keV)	Possible Interferences
First Count							
Na	^{23}Na(n,γ)^{24}Na	100	0.53	0.6258	100	1368.6	
K	^{41}K(n,γ)^{42}K	6.71	1.46	0.5151	18.8	1524.6	
Ga	^{71}Ga(n,γ)^{72}Ga	39.9	4.6	0.5883	95.6	833.95	834.8 (^{54}Mn)
As	^{75}As(n,γ)^{76}As	100	4.4	1.097	45	559.1	554.3(^{82}Br), 563.1(^{134}Cs), 564.1(^{122}Sb)
					6.38	657.0	657.6 (^{110m}Ag)
					3.41	1216.0	1221.4 (^{182}Ta)
Br	^{81}Br(n,γ)^{82}Br	49.3	2.7	1.4725	70.7	554.3	558.5(^{77}Ge), 559.1(^{76}As), 551.5(^{187}W)
					43.0	619.0	618.2 (^{187}W)
					83.4	776.5	778.8(^{152}Eu), 773.7(^{131}Te)
Mo	^{98}Mo(n,γ)^{99}Mo(β^-)	24.1	0.13	2.751	88.7	140.5	142.6(^{59}Fe) 145.4 (^{141}Ce)
	(β^-)^{99}Tc				12.6	739.6	744.2 (^{110m}Ag)
Sb	^{121}Sb(n,γ)	57.3	6.2	2.681	70	564.1	559.1+562.8 (^{76}As), 564.0+566.4 (^{152}Eu), 563.2+569.3 (^{134}Cs)
La	^{139}La(n,γ)^{140}	99.9	9.2	1.678	42.98	487.0	482.2(^{181}Hf), 492.3 (^{115}Cd)22.4815.8817.9 (^{110m}Ag)
					95.5	1596.5	1596.7(^{72}Ga)
Sm	^{152}Sm(n,γ)^{153}Sm	26.6	204	1.95	28.3	103.2	103.3(^{153}Gd)
Yb	^{174}Yb(n,γ)^{175}Yb	31.6	65	4.19	2.83	282.5	277.6(^{239}Np), 279.2(^{203}Hg), 279.5(^{75}Se), 283.3(^{192}Ir)
					6.0	396.3	392.5(^{160}Tb), 398.5(^{233}Pa), 400.6(^{75}Se)
W	^{186}W(n,γ)^{187}W	28.6	38	0.9938	21	479.5	482.2(^{181}Hf), 484.6(^{192}Ir)
					26.2	685.8	686.8(^{110m}Ag), 682.3(^{160}Tb), 692.8(^{233}Sb), 688.6(^{152}Eu)
U	^{238}U(n,γ)^{239}U						
	(β^-)^{239}Np	99.275	2.7	2.346	11.4	228.1	228.5(^{177m}Lu), 227.9(^{125}Sb), 229.3(^{182}Ta)
					15.0	277.6	279.2(^{203}Hg), 279.6(^{75}Se)
Second Count							
Cr	^{50}Cr(n,γ)^{51}Cr	4.35	15.9	27.70	10.2	320.1	316.5(^{192}Ir), 319.7(^{147}Nd)
Rb	^{85}Rb(n,γ)^{86}Rb	72.17	0.447	18.82	8.79	1077.2	
Ba	^{130}Ba(n,γ)^{131}Ba	0.106	8	12.0	21.67	216.0	215.7(^{160}Tb)
					42.1	496.2	497.1(^{103}Ru)
Ce	^{140}Ce(n,γ)^{141}Ce	88.5	0.56	32.55	40.4	145.4	142.4(^{59}Fe)
Sm	^{152}Sm(n,γ)^{153}Sm	26.6	204	1.950	28.3	103.2	100.1(^{182}Ta), 103.2(^{153}Gd), 106.1(^{239}Np)
Yb	^{168}Yb(n,γ)^{169}Yb	0.135	3500	32.02	21.7	177.2	179.4(^{182}Ta)
Yb	^{174}Yb(n,γ)^{175}Yb	31.6	65	4.19	2.83	282.5	277.6(^{239}Np) 279.2(^{203}Hg), 279.5(^{75}Se), 283.3(^{112}Ir)
					6.0	396.3	392.5(^{160}Tb), 398.5(^{233}Pa), 400.6(^{75}Se)
Lu	^{176}Lu(n,γ)^{177}Lu	2.61	2000	6.71	18.4	208.4	205.8(^{192}Ir)

TABLE 4.2. (contd)

Element	Reaction	Isotopic Abundance ($)	Cross-Section (barns)	Half-Life $T_{1/2}$(days)	Intensity (γ's per 100 dis)	Major γ-Rays (keV)	Possible Interferences
Third Count							
Sc	$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	100	26	83.80	100	889.2	$884.7(^{110m}\text{Ag})$
Cr	$^{50}\text{Cr}(n,\gamma)^{51}\text{Cr}$	4.35	15.9	27.70	100	1120.5	$1115.5(^{65}\text{Zn})$, 1120(Bkgd), $1121.3(^{182}\text{Ta})$
					10.2	320.1	$316.5(^{192}\text{Ir})$, $319.7(^{147}\text{Nd})$
Fe	$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	0.29	1.14	44.56	56.5	1099.2	$1102.9(^{160}\text{Tb})$
					43.5	1291.6	$1289.1(^{182}\text{Ta})$, $1292.0(^{152}\text{Eu})$
Co	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	100	37	1925.5	100	1173.2	$1177.8(^{160}\text{Tb})$
					100	1332.5	
Ni	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	68.3	4.6	70.78	99.4	810.8	
Zn	$^{64}\text{Zn}(n,\gamma)^{65}\text{Zn}$	48.6	0.78	244.0	50.75	1115.5	$112.0(^{152}\text{Eu})$ $113.2(^{182}\text{Ta})$, $1115.2(^{160}\text{Tb})$, $1120.5(^{46}\text{Sc})$
Se	$^{74}\text{Se}(n,\gamma)^{75}\text{Se}$	8.7	52	118.4	54.0	136.0	$133.0(^{181}\text{Hf})$, $133.5(^{131}\text{Ba})$, $136.3(^{181}\text{Hf})$, $136.2(^{192}\text{Ir})$
					58.0	264.7	$261.0(^{169}\text{Yb})$, $264.1(^{181}\text{Ta})$
Rb	$^{85}\text{Rb}(n,\gamma)^{86}\text{Rb}$	72.17	0.447	18.82	8.79	1077.2	
Sr	$^{84}\text{Sr}(n,\gamma)^{85}\text{Sr}$	0.56	0.3	64.35	99.3	514.0	511.0 (Annihilation peak)
Zr	$^{94}\text{Zr}(n,\gamma)^{95}\text{Zr}$	17.4	0.055	63.98	43.1	724.2	
					54.6	756.7	
Ag	$^{109}\text{Ag}(n,\gamma)^{110m}\text{Ag}$	48.7	4	252.2	94.4	657.8	$656.5(^{152}\text{Eu})$, 661.6(Bkgd)
					72.8	884.7	$884.5(^{192}\text{Ir})$, $889.3(^{46}\text{Sc})$
					34.3	937.5	
Sb	$^{123}\text{Sb}(n,\gamma)^{124}\text{Sb}$	42.7	4.0	60.20	49.0	169.10	
Cs	$^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$	100	27	753.1	85.44	795.8	
Ba	$^{130}\text{Ba}(n,\gamma)^{131}\text{Ba}$	0.106	8	12.0	21.67	216.0	$215.7(^{160}\text{Tb})$
					42.1	496.2	$497.1(^{103}\text{Ru})$
Ce	$^{140}\text{Ce}(n,\gamma)^{141}\text{Ce}$	88.5	0.56	32.55	48.4	145.4	$142.4(^{59}\text{Fe})$
Eu	$^{151}\text{Eu}(n,\gamma)^{152}\text{Eu}$	47.9	5800	4821.2	14.3	964.0	$962.3(^{160}\text{Tb})$, $966.2(^{160}\text{Tb})$
					10.1	1085.8	
					20.7	1408.1	
Tb	$^{159}\text{Tb}(n,\gamma)^{160}\text{Tb}$	100	25	72.1	4.0	215.7	$216.0(^{133}\text{Ba})$
					30.0	879.4	
Yb	$^{168}\text{Yb}(n,\gamma)^{169}\text{Yb}$	0.135	3500	32.02	21.7	177.2	$179.4(^{182}\text{Ta})$
Hf	$^{180}\text{Hf}(n,\gamma)^{181}\text{Hf}$	35.2	14	42.45	32.4	133.0	$130.5(^{169}\text{Yb})$, $133.5(^{131}\text{Ba})$, $136.0(^{75}\text{Se})$
					81	482.0	$480.1(^{131}\text{Ba})$, $484.6(^{192}\text{Ir})$
Ta	$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	99.99	21	115.0	16.4	1189.0	
					27.1	1221.4	
Th	$^{232}\text{Th}(n,\gamma)^{233}\text{Th}$ $(\beta^-)^{233}\text{Th}$	100.	7.4	26.95	37	311.9	$307.7(^{169}\text{Yb})$, $308.4(^{192}\text{Ir})$, $309.6(^{160}\text{Tb})$, $316.5(^{192}\text{Ir})$

The computer based TN-11 system operated a pneumatic transfer rabbit shuttle which by operator input and program control irradiated the sample for a set length of time, allowed the sample to decay for a set time, and counted the sample for a preset length of time. The cycle was then repeated as many times as necessary. In particular, after a 3-min irradiation, the sample was allowed to decay for 5-min whereupon a 5-min count was initiated. Additionally, a 30 minute decay was also observed before a count of 30-min was initiated.

The detector system consisted of 2 Ge(Li) diodes of 105 cm^3 and 110 cm^3 volume in an opposed geometry resulting in an exceptionally high counting efficiency. Acid-cleaned polyvials were used to contain the sample and as spacers in the acid-cleaned rabbit tube. Data was stored on floppy disks for subsequent data reduction.

Longs Analysis

Samples and standards were irradiated for 7 hours at a flux of 1 x 10^{12} $n \cdot cm^{-2} \cdot s^{-1}$ in the rotating rack (lazy susan) of the 250 kW Triga reactor operated by Westinghouse-Hanford. The samples and standards were returned to the laboratory and repackaged in clean polyvials for counting on several high efficiency germanium diodes. The activated samples and standards were counted three times following delays of 3, 7, and 25 days. The data was collected on 4096 channel analyzers which were interfaced to a PDP 11/44 computer. The computer was used both for data manipulation and storage, as well as final data reduction.

Data Reduction

The spectral data was reduced with the use of a PDP 11/44 computer and the program CANGAS (Laul, Wilkerson and Crowe 1978). This program uses an operator created library with the parametric information necessary for each nuclide peak sought (see Table 4.1 and 4.2). The sample specific activity was then compared to the standard specific activity to obtain concentrations in µg/g. Spectral interference corrections were also made through the use of the CANGAS program.

Standard Results

The following standards were analyzed to obtain specific activities for each nuclide peak and as a means of quality assurance:

NBS SRM 1633 Coal Fly Ash
NBS SRM 123C Stainless Steel
NBS SRM 1155 Stainless Steel
IAEA Soil-5
USGS BHVO-1, Hawaiian Basalt

USGS PCC-1, Peridotite, and
Various single element standards (Zn, U, Mo and Ag)

Table 4.3 shows the average result obtained by the "shorts" analysis of NBS 1633 fly ash, NBS 123C stainless steel, and NBS 1155 stainless steel. Table 4.4 shows the average result obtained for the "longs" analysis of NBS 1633 fly ash, NBS 123C stainless steel, NBS 1155 stainless steel, IAEA soil-5, USGS BHVO-1, and USGS P[illegible]. There is excellent agreement between the average of the experimental results for "shorts" and "longs" with the recommended or best values.

4.1.2 X-ray Fluorescence Analysis (XRF)

The X-ray fluorescence procedure utilized in this work is a rather highly automated multielement technique capable of producing a rather large amount of high quality information on a short time scale at relatively minimal cost. It is applicable to a wide range of sample types including geological materials, waters, oils, metals, biological materials, glasses, etc. In most cases sample analysis proved to be a routine matter. A modest level of technique development was required for cement samples due to their very high calcium levels. A modification of the fundamental parameters data reduction code was also required for the steels analysis particularly for the highly radioactive samples (Nielson, Sanders, and Evans 1982).

Sample Preparation

Sample preparation for XRF is a relatively simple matter. Samples were initially treated as described in Section 4.1.1. Powdered samples were then pressed into 500 mg pellets and mounted in 35 mm slide holders. Steel samples in the form of shavings or turnings were sandwiched between 0.1 mil polypropylene films on 35 mm slide holder mounts. In a few cases steel discs were also machined for comparison with the turnings. In general equivalent results were obtained and the simpler steel turnings method was adopted for all other samples. This comparison is described in more detail in Nielson, Sanders and Evans (1982).

Analytical Procedure

Three energy dispersive X-ray fluorescence systems were utilized in this work. For elements with low and intermediate atomic numbers a pair of secondary source systems manufactured by Kevex were used. The two systems, employing 60 kV and 80 kV X-ray tubes respectively are normally used for three separate runs with Ti, Zr, and Ag secondary sources producing characteristic radiation to fluoresce the sample. A fourth bombardment with low energy gamma rays from a ^{241}Am isotopic source is used for elements with higher atomic numbers such as Ba. This system was developed in-house and shares data acquisition and

TABLE 4.3. Comparison of Experimental and Reported Standard Values for "shorts" Analysis

		NBS 1633 Coal Fly Ash		NBS 123C Steel		NBS 1155 Steel	
Element		Expt.	Reported[a]	Expt.	Reported[b]	Expt.	Reported[b]
		(19 Samples)		(5 Samples)		(6 Samples)	
Al	%	12.9 ± 0.3	12.7 ± 0.5	0.049 ± 0.007	---	0.022 ± 0.004	---
Ca	%	4.9 ± 0.5	4.7 ± 0.6	---	---	---	---
Ti	%	0.70 ± 0.04	0.74 ±0.03	---	---	---	---
V	ppm	235 ± 1	235 ± 13	294 ± 8	---	472 ± 14	470
Cu	ppm	---	---	1160 ± 150	1030	1640 ± 60	1690
Na	ppm	3400 ± 600(17)	3200 ± 400	---	---	---	---
Cl	ppm	44 ± 9 (14)	42 ± 10	---	---	---	---
K	%	1.7 ± 0.3(13)	1.61 ± 0.15	---	---	---	---
Mn	ppm	497 ± 22 (17)	496 ± 19	14900 ± 300	17000	15000 ± 300	16300
Sr	ppm	1350 ± 140(15)	1700 ± 200	---	---	---	---
Ba	ppm	2740 ± 120(15)	2700 ± 200	---	---	---	---
Dy	ppm	8.9 ± 0.5(15)	8.8 ± 2.3[c]	---	---	---	---

(a) Data taken from Ondov et al. (1975).
(b) Data taken from NBS Standard Certificates.
(c) Data taken from Gladney (1980).

TABLE 4.4. Comparison of Experimental and Reported Standard Values for "Longs" Analysis

Element	NBS 1633 Fly Ash		IAEA Soil-5		BHVO		PCC-1		NBS Steel 1230		NBS Steel 1155	
	Expt.	Reported$^{(a)}$	Expt.	Reported$^{(c)}$	Expt.	Reported$^{(d)}$	Expt.	Reported$^{(e)}$	Expt.	Reported$^{(f)}$	Expt.	Reported$^{(f)}$
	(8 Samples)		(5 Samples)		(5 Samples)		(4 Samples)		(4 Samples)		(4 Samples)	
Na (ppm)	5190 ± 70	3200 ± 340	18900 ± 500	18800	17100 ± 500	16400 ± 600		50	---	---	---	---
K (ppm)	17000 ± 600	17000 ± 900	18500 ± 1500	19800 ± 1000	4600 ± 200	4600 ± 700		30	---	---	---	---
Ca (ppm)	49000 ± 4000	46000 ± 4000	28000 ± 500	25000 ± 4000	81000 ± 7000	81600 ± 1300	3600 ± 900	3600	---	---	---	---
Sc (ppm	26 ± 1	26 ± 2	13 ± 1	15.3 ± 0.3	30 ± 2	30 ± 2	7.5 ± 0.3	6.9	---	---	---	---
Cr (ppm)	128 ± 1	129 ± 11	28 ± 3	31 ± 4	290 ± 20	290 ± 30	2670 ± 160	2730	164000 ± 17000	174000	191000 ± 5000	184000
Fe (ppm)	63000 ± 3000	62000 ± 4000	44300 ± 900	46000 ± 1000	86000 ± 2000	85100 ± 1900	56700 ± 1700	58400	666000 ± 40000	---	655000 ± 25000	---
Co (ppm)	40 ± 2	41.5 ± 1.2$^{(b)}$	15.5 ± 0.8	14.6 ± 0.3	44 ± 2	45 ± 2	111 ± 6	112	1210 ± 80	1200	1080 ± 30	1000
Ni (ppm)	133 ± 30	98 ± 11	35 ± 15	<10	130 ± 20	120 ± 16	2120 ± 120	2340	106000 ± 9200	113000	119000 ± 10000	121000
Cu (ppm)	---	127 ± 8	---	---	---	130 ± 13	---	---	1030 ± 260	1030	1420 ± 80	1690
Zn (ppm)	200 ± 20	213 ± 15	340 ± 30	400 ± 40	140 ± 20	102 ± 6	57 ± 9	56	104 ± 24	---	---	---
Ga (ppm)	46 ± 7	49 ± 14	28 ± 3(2)	---	---	22 ± 3	---	---	---	---	---	---
As (ppm)	57 ± 4	58 ± 4$^{(b)}$	94 ± 8	110 ± 5	---	---	---	---	57 ± 4	---	99 ± 9	---
Se (ppm)	9.3 ± 0.5	9.6 ± 0.8	2.5 ± 0.1(2)	2.0 ± 0.2	---	---	---	---	---	---	---	---
Br (ppm)	12.1 ± 0.3	12 ± 4$^{(b)}$	10.4 ± 0.4	6.8 ± 0.5	---	---	0.73 ± 0.05	---	---	---	---	---
Rb (ppm)	115 ± 4	114 ± 10	117 ± 4	140 ± 20	18 ± 3	---	---	0.063	---	---	---	---
Sr (ppm)	1400 ± 60(6)	1380 ± 190	390 ± 40	400 ± 50	420 ± 40	440 ± 70	---	0.4	---	---	---	---
Zr (ppm)	290 ± 20	300 ± 75	170 ± 20	240 ± 20	140 ± 20	177 ± 26	---	0.7(7)	---	---	---	---
Mo (ppm)	---	29 ± 6	---	---	---	---	---	---	2200 ± 200	2200	22000 ± 2000	23800
Ag (ppm)	---	---	---	---	---	---	---	---	---	---	---	---
Sb (ppm)	6.8 ± 0.3	6.9 ± 0.4	14.5 ± 0.5	14.7 ± 0.4	0.23 ± 0.0[illegible]	0.17 ± 0.01	1.5 ± 0.1	1.4	8.5 ± 1.4	---	19 ± 4	---
Cs (ppm)	8.5 ± 0.5	8.6 ± 1.8	51 ± 2	50 ± 5	0.18 ± 0.02	---	---	0.006	---	---	---	---
Ba (ppm)	2800 ± 120	2700 ± 170	650 ± 60	550 ± 40	260 ± 60	143 ± 20	---	---	---			
La (ppm)	78 ± 2(7)	79 ± 7	28.4 ± 0.8	30 ± 1	16.6 ± 0.9	16.7 ± 0.6	---	0.15	---	---	---	---
Ce (ppm)	145 ± 8	146 ± 15$^{(b)}$	51 ± 2	59 ± 2	58 ± 1	40 ± 4	---	0.09	---	---	---	---
Sm (ppm)	12.3 ± 0.5	12.4 ± 1.1	4.8 ± 0.2	5.60 ± 0.04	5.5 ± 0.4	6.2 ± 0.7	---	0.008	---	---	---	---
Eu (ppm)	2.6 ± 0.1	2.6 ± 0.2	1.07 ± 0.07	1.15 ± 0.05	2.0 ± 0.1	2.1 ± 0.4	0.0086 ± 0.0007	0.002	---	---	---	---
Tb (ppm)	2.1 ± 0.1	2.1 ± 0.6	0.76 ± 0.05	0.70 ± 0.03	1.11 ± 0.07	1.0 ± 0.3	---	0.001	---	---	---	---
Yb (ppm)	6.4 ± 0.2	6.4 ± 1.1	2.4 ± 0.1	2.3 ± 0.1	2.3 ± 0.1	2.1 ± 0.5	---	0.02	---	---	---	---
Lu (ppm)	1.51 ± 0.04	1.5 ± 1.2	0.35 ± 0.04	0.37 ± 0.03	0.43 ± 0.07	---	---	0.006	---	---	---	---
Hf (ppm)	7.6 ± 0.5	7.8 ± 1.1	6.5 ± 0.6	6.5 ± 0.2	4.8 ± [illegible]	4.1 ± 0.3	---	0.06	---	---	---	---
Ta (ppm)	2.2 ± 0.1	2.2 ± 0.5	0.83 ± 0.05	0.76 ± 0.05	1.38 ± 0.06	1.08 ± 0.18	---	<0.1	5.9 ± 0.5	---	---	---
W (ppm)	5.5 ± 0.4	5.5 ± 2.6	6.4 ± 0.5	---	1.3 ± 0.3	---	---	---	92 ± 11	---	1040 ± 160	---
Th (ppm)	25 ± 1	25 ± 1	11 ± 1	10.9 ± 0.6	1.25 ± 0.07	1.0 ± 0.2	---	0.01	---	---	---	---
V (ppm)	14 ± 2	11.4 ± 1.4	3.8 ± 0.8	---	0.58 ± 0.10	0.40 ± 0.06	---	---	---	---	---	---

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(a) Data taken from Gladney et al. (1980a)
(b) Data taken from Ondov et al. (1975)
(c) Data taken from Standard Certificate for IAEA Soil-5
(d) Data taken from Gladney (1980b)
(e) Data taken from Flanagan (1973)
(f) Data taken from NBS Standards Certificate

reduction with the two Kevex systems. Data acquisition utilizes a Canberra System 80 multichannel analyzer with multiple analog to digital converters interfaced to a DEC PDP 11/34 computer. All data reduction is done on line by the PDP 11/34. A fundamental parameter method is used which utilizes coherent to incoherent peak scattering ratios to calculate the absorption properties of the matrix. The program (SAP3) makes peak overlap corrections, grain size corrections, self-absorption corrections, etc. These results are then reconciled against a library of thin film standards to calculate a final result. The SAP3 program has undergone continual improvement and upgrading. A description of the most recent version of this work is given by Nielson and Sanders (1982).

Standard Results

In order to verify the accuracy of the method quality assurance samples of standard reference materials are run periodically. Tables 4.5 and 4.6 show comparisons with accepted values for NBS steel and NBS fly ash standards. In general, agreement is excellent.

TABLE 4.5. Comparison of Experimental and Reported Values for XRF Analysis of Steel

Element	NBS 1155 Expt. (4 samples)	Reported(a)
Ti (%)	0.148 ± 0.017	
V (ppm)	456 ± 86	470
Cr (%)	18.1 ± 0.05	18.45
Mn (%)	1.76 ± 0.063	1.63
Fe (%)	64.83 ± 0.096	64.5
Ni (%)	12.1 ± 0.082	12.18
Cu (%)	0.158 ± 0.0096	0.169
Zn (ppm)	200 ± 62	
As (ppm)	108 ± 21	
Br (ppm)	<25	
Rb (ppm)	<14	
Sr (ppm)	<12	
Y (ppm)	<11	
Zr (ppm)	<11	
Nb (ppm)	<14	
Mo (%)	2.22 ± 0.019	2.38

(a) Data taken from NBS Certificate of Analysis.

TABLE 4.6. Comparison of Experimental and Reported Values for XRF Analysis of Fly Ash

Element	NBS 1633A Expt. (4 samples)	Reported
Al (%)	15.9 ± 1.3	14.0 ± 0.2
Si (%)	23.4 ± 1.1	22.8 ± 0.80
S (%)	0.342 ± 0.013	0.27 ± 0.02
K (%)	1.88 ± 0.018	1.88 ± 0.06
Ca (%)	1.17 ± 0.038	1.11 ± 0.01
Ti (%)	0.816 ± 0.026	0.84 ± 0.01
V (ppm)	302 ± 32	300 ± 40
Cr (ppm)	194 ± 58	196 ± 6
Mn (ppm)	174 ± 36	190 ± 15
Fe (%)	9.53 ± 0.29	9.40 ± 0.10
Ni (ppm)	139 ± 11	127 ± 4
Cu (ppm)	122 ± 15	118 ± 3
Zn (ppm)	241 ± 13	220 ± 10
Ga (ppm)	58 ± 4	58 ± 6
As (ppm)	156 ± 12	145 ± 15
Se (ppm)	10.0 ± 1.7	10.3 ± 0.6
Rb (ppm)	133 ± 5	131 ± 2
Sr (ppm)	842 ± 19	830 ± 30
Y (ppm)	88 ± 3	
Zr (ppm)	239 ± 5	
Nb (ppm)	28.7 ± 1.3	
Mo (ppm)	33.7 ± 0.37	29 ± 3
Ba (ppm)	1642 ± 199	1500 ± 200
Pb (ppm)	73.9 ± 5.05	72.4 ± 0.4

4.1.3 Other Analytical Methods Used

In a few selected cases specialized analytical procedures were utilized to add a limited amount of data to the total information base. These include flame atomic absorption for Li, graphite furnace atomic absorption for Ag, and a combustion train method for nitrogen. A few samples were also sent to the National Bureau of Standards for analysis by prompt gamma activation analysis.

4.2 CHEMICAL ANALYSIS RESULTS

4.2.1 Stainless Steel

Chemical analysis results for thirteen samples of nuclear grade Type 304L stainless steel are given in Table 4.7. This data is summarized in Table 4.8. As might be expected, elements involved in the alloy process control show very little variability. These include Fe, Cr, and Ni. Activatible trace elements on the other hand show a surprisingly large variation. Of particular note is cobalt which shows a range of more than an order of magnitude with the average value about seven times higher than the lowest value. This suggests that had greater attention been given to process control and batch selection of steels prior to construction, a considerably reduced ^{60}Co inventory would result, thus both reducing translocation problems during operations and minimizing doses during dismantling. The niobium results showed a very wide variability. The two samples from the Enrico Fermi project from different heats were very low in niobium (<5 ppm) with other samples ranging up to 200 to 300 ppm, somewhat above the 160 ppm now adopted by the steel manufacturers as an upper bound for nuclear grade steel. Several other activatible elements also showed marked variability including Se, Br, and Mo. Europium was in general below detection limits for that matrix; however, at least one other more abundant rare earth, cerium was found at highly variable levels suggesting that there may be some Eu present also. The origin of these large variations in trace element concentration is a bit unclear. At least for some elements such as Co, Nb, and Mo, it may be in part due to cross contamination between batches of ordinary and specialty stainless steels produced in the same crucibles.

4.2.2 Other Reactor Internals Steels

Table 4.9 tabulates the chemical compositions of some steels used in a more limited application for reactor internals. The three Type 316 samples analyzed contained cobalt levels similar to the average for the 304. They were relatively low in niobium but about ten times higher in molybdenum. The inconel samples were very high in niobium, particularly the Point Beach sample which contained about 5% Nb. This was an activated sample from the fuel support structure which did in fact contain a substantial level of ^{94}Nb.

4.2.3 Vessel Steels

Table 4.10 lists the compositions of 10 vessel steels. The data is summarized in Table 4.11. Eight of the samples were received very late in the program and consequently were analyzed only using XRF. The Bellefonte sample is interesting in that it shows a small amount of europium present in spite of being very low in cerium, in contrast to some of the stainless steel samples. Carbon steel is in general rather low in cobalt. Variations in cobalt in

TABLE 4.7. Chemical Analyses of Type 304 Stainless Steel

Element		U.S. Steel (1)	U.S. Steel (2)	U.S. Steel (3)	North Anna	Susquehanna	Enrico Fermi	Enrico Fermi	Monticello	Humboldt Bay	Westinghouse	Point Beach	Belle-Fonte	Waterford
Li	ppm	0.13												
N	ppm	402	525	430										
Na	ppm	3	5	9	37	4.7	5.1	4.0						
Al	ppm	<50	<50	<50	130		165	200						
Cl	ppm				<50	<60	<70	<60					130	
K	ppm	<3	<3											
Ca	ppm	19												
Sc	ppm				<0.3	<0.1	<0.03	<0.05						
Ti	ppm	<200	<200	<200	1000	1030	1350	<1000	410	<200	<500	<500		
V	ppm	580			680	720	260	280	190	570	<300	200	690	
Cr	%	19.9	19.2	19.4	17.7	19.0	19.0	18.0	17.6	18.0	16.5	17.3	20.2	18.3
Mn	%	1.66	1.76	1.76	1.13	1.30	1.24	1.57	1.63	1.89	1.85	1.20	1.11	1.58
Fe	%	68.0	68.0	69.0	74.1	76.4	72.6	70.7	71.0	68.7	69.9	69.4	71.2	69.9
Co	ppm	750	820	1240	1720	2265	229	2570					1720	
Ni	%	10.3	10.0	9.5	10.6	10.8	9.6	10.4	9.1	11.0	9.4	10.8	10.4	8.8
Cu	ppm	5000	2700	2300	2850	2860	504	300	2220	1270	6620	2900	2510	8150
Zn	ppm	44	51	86	136	<20	134	<15	1520	310	740	200	142	2230
Ga	ppm	60	50	70	<40	<40	56	74	50	160	430	600	52	140
As	ppm	95	57	86	94	81	187	96	200	80	370	1500	78	90
Se	ppm	<10	<10	<10	<2	<5	2.5	25	70	<20	<40		<9	<30
Br	ppm	8	<2	<2	<1	<2	1.4	<0.9					<1	<1
Rb	ppm	<10	<10	<10	<9	<13	<18	<9					<10	<10
Sr	ppm	0.20												
Y	ppm	<4	<4	<4	<7	<7	<7	<6	<10	<10	<10		<7	<10
Zr	ppm	7	<4	<4	<7	<7	<6	<6	<20	<10	<10	<20	<7	20
Nb	ppm	190	16	110	71	300	<6	<5	70	10	30	120	63	200
Mo	ppm	2200	2200	1900	4500	5500	510	78	2300	2300	3200	2300	3900	2400
Ag	ppm	<6	<6	<7	<1.4	<5	<2	<2					<7	
Sb	ppm	17	7.3	12	16.1	15.4	11.1	6.9					12.6	
Cs	ppm				<0.3	<0.7	<0.4	<0.3					<0.3	
Ba	ppm	<1000	<1000	<1000	<500	<500	<500	<700					<500	
La	ppm	<0.2	<0.2	<0.2	<0.05	0.094	<0.07	<2.1					<0.2	
Ce	ppm				<2	440	446	419					550	
Sm	ppm	<0.2	<0.2	<0.2	<0.05	0.15	<0.05	<0.06					0.054	
Eu	ppm	<0.05	<0.07	<0.08	<0.07	<0.06	<0.03	<0.1					<0.1	
Tb	ppm				0.48	<0.9	0.22	0.71					<1	
Dy	ppm				<0.1	<1	<1	<1					<1	
Ho	ppm	<1	<1	<1										
Yb	ppm	<2	<2	<2	<5	<3	<2	<2					<4	
Lu	ppm	<0.8	<0.7	<3	1.4	<0.9	<0.9						<0.5	
Hf	ppm				<3	<2	<1	<1				<3		
Ta	ppm	<0.8	<0.8	<0.8	<0.36	1.1	<0.2	0.42				<1.5		
W	ppm	142	157	140	220	520	110	7.2				194		
Pb	ppm	<10	<10	<10	93	139	118	111				50		
Th	ppm	<9	<9	<9	<0.6	<1.5	<0.8	<0.9				<2		
U	ppm	<4	<5	<5	<2	<3	<3	<0.8				<1		

TABLE 4.8. Type 304 Stainless Steel Composition Summary

Element	Average	% S.D.[a]	Range	Range Factor[b]	No. of Measurements Used
Li ppm	0.13				1
N ppm	452 ± 64	14	402 to 525	1.3	3
Na ppm	9.7 ± 12.2	126	3 to 37	12.3	7
Al ppm	100		<50 to 200		6
Cl ppm	70		<50 to 130		5
K ppm	<3				3
Ca ppm	19				1
Sc ppm	<0.03				1
Ti ppm	<600				10
V ppm	456 ± 235	52	140 to 690	4.90	9
Cr %	18.4 ± 1.1	6	16.5 to 20.2	.2	13
Mn %	1.53 ± 0.27	18	1.11 to 1.76	1.6	13
Fe %	70.6 ± 2.6	4	68.0 to 76.7	1.1	13
Co ppm	1414 ± 800	57	229 to 2570	11.2	8
Ni %	10.0 ± 0.7	7	8.8 to 11.0	1.3	13
Cu ppm	3080 ± 2270	74	300 to 8150	27.2	13
Zn ppm	457 ± 717	157	<15 to 2230	>1.3	12
Ga ppm	129 ± 143	90	<40 to 450	>11.3	13
As ppm	194 ± 259	168	<57 to 1010	>17.7	13
Se ppm	≈35		<2 to 70	>35	13
Br ppm	~2		<0.9 to 8.0	78.9	8
Rb ppm	<10				9
Sr ppm	0.2				1
Y ppm	<5				12
Zr ppm	≈10		<4 to 20		13
Nb ppm	89 ± 90	101	<5 to 300	>60	13
Mo ppm	02.600 ± 1500	58	80 to 5500	68.8	13
Ag ppm	<2				8
Sb ppm	12.3 ± 3.8	31	6.9 to 17.0	2.5	8
Cs ppm	<0.3				5
Ba ppm	<500				8
La ppm	≈0.2		<0.05 to 2.1	>42.0	8
Ce ppm	371 ± 212	57	<2 to 550	>275.0	8
Sm ppm	≈0.1		<0.05 to 0.15	>3	7
Eu ppm	≈0.02				inferred
Tb ppm	0.47 ± 0.25	53	0.22 to 0.71		3
Dy ppm	<1				5
Ho ppm	<1				3
Yb ppm	<2				5
Lu ppm	<0.8				7
Hf ppm	<2				3
W ppm	186 ± 149	80	7.2 to 520		8
Pb ppm	67 ± 50	75	<10 to 139	>13.9	9
Th ppm	<1				3
U ppm	<2				3

(a) Relative standard deviation expressed as a percentage of the average value.

(b) Range factor shown is the ratio of the highest to the lowest value measured.

TABLE 4.9. Chemical Analysis of Steel Used for Reactor Internals

Element		U.S. Steel SS 316	U.S. Steel SS 316	U.S. Steel SS 316	Susquehanna Inconel SB 166	Point Beach Inconel 710	Susquehanna SA 508
Li	ppm	0.18					
N	ppm	357					
Na	ppm	5	6	<2	4.1		6.8
Al	ppm	<50			1160		120
Cl	ppm				26		63
K	ppm	<3			<60		
Ca	ppm	14					
Ti	ppm	<200	<200	<200	2280		690
V	ppm	630			270		121
Cr	%	16.8	17.3	17.2	15.2	15.1	0.38
Mn	%	1.30	1.30	1.52	0.23		0.65
Fe	%	65	65	64	9.96	17.5	115
Co	ppm	1630	1340	1450	574		115
Ni	%	13.2	12.8	12.5	74.5	57.3	0.93
Cu	ppm	2600	2900	2200	2240	450	1000
Zn	ppm	71			<9	2330	<9
Ga	ppm	60	50	60	13	40	<40
As	ppm	95	75	86	33		96
Se	ppm	<9	<9	<9	<3		<1
Br	ppm	<2	<2	<2	<0.7		16
Rb	ppm				<11		32
Sr	ppm	0.23					
Y	ppm	<5	<5	<5	<11		<7
Zr	ppm	6	6	<6	<8		<7
Nb	ppm	12	64	12	385	49600	<8
Mo	%	2.06	2.12	2.16	0.19	2.72	0.76
Ag	ppm	<5			<2		<6
Sb	ppm	11	13	12	2.3		1619
Cs	ppm				<20		<30
Ba	ppm				<200		<300
La	ppm	<0.2	<0.2	<0.2	<0.03		0.064
Ce	ppm				400		4.1
Sm	ppm	<0.2	<0.2	<0.2	0.045		<0.2
Eu	ppm	<0.07			<0.05		0.042
Tb	ppm	<9	<9	<9	<0.2		0.40
Dy	ppm				<0.5		1.2
Ho	ppm	<1	<1	<1			
Yb	ppm	<2	<2	<2	<2		<0.7
Lu	ppm	<0.8	<0.8	<0.8	<1		<0.4
Hf	ppm				<0.9		<0.5
Ta	ppm	<0.7			1.2		1.2
W	ppm	105	218	108	2.8		60
Pb	ppm	<30	<30	<30	<60		<60
Th	ppm				<1		<0.5
U	ppm	<4	<5	<4	<0.8		<1

TABLE 4.10. Chemical Analysis of Vessel Steels

Element		SA 302	Bellefonte	P34	P147	GP55	P21	Pu	P337	RR-1	GP85
Li	ppm	0.30									
N	ppm	84									
Na	ppm	40	6.6								
Al	ppm	<50	613								
Cl	ppm		<40								
K	ppm	<3	20								
Ca	ppm	14	<400								
Sc	ppm		0.26								
Ti	ppm	<2									
V	ppm	<30	74	<90	<90	<80	<90	<90	<90	<80	70
Cr	%	0.06	0.36	0.083	0.078	0.11	0.34	0.35	0.09	0.073	0.13
Mn	%	1.49	0.76	1.20	1.23	1.32	0.20	0.44	1.	1.16	1.23
Fe	%	98	98	97	97	97	98	97	96	97	97
Co	ppm	93	151								
Ni	%	<0.3	0.72	0.62	0.66	0.68	0.75	0.76	0.66	0.70	0.72
Cu	ppm	1900	675	1480	1410	1140	800	1000	1450	1780	1100
Zn	ppm	22	<4	<70	210	170	<70	<70	140	160	<70
Ga	ppm	<20	24	<50	<50	<50	<50	<50	300	150	<50
As	ppm	160	63	80	50	260	200	190	3600	250	470
Se	ppm	<9	<0.7	<20	<20	<20	<20	<40	<30	<20	<20
Br	ppm	<2	0.85								
Rb	ppm	72	23								
Sr	ppm	0.15	<8	<10	<10	<10	<20	<10	<20	<10	<10
Y	ppm	<4	<7	<20	<10	<10	<10	<10	120	<10	<10
Zr	ppm	<4	<7	<10	<10	20	<10	<10	<10	<10	20
Nb	ppm	40	<8	10	20	20	10	10	<10	40	20
Mo	%	0.45	0.64	0.56	0.56	0.54	0.57	0.63	0.53	0.57	0.59
Ag	ppm	<2	<7								
Sb	ppm	16	6.1								
Cs	ppm		<0.2								
Ba	ppm	<800	<270								
La	ppm	<0.2	0.10								
Ce	ppm		<1								
Sn	ppm	<0.09	0.017								
Eu	ppm	<0.2	0.031								
Tb	ppm		0.45								
Hf	ppm	<0.6	<1								
Yb	ppm	<1	0.81								
Lu	ppm	<0.3	0.12								
Hf	ppm		0.21								
Ta	ppm	<0.3	0.13								
W	ppm	5	5.9								
Pb	ppm	<40	<50	<50	<40	<40	<40	<50	7730	60	90
Th	ppm	<4	0.18								
U	ppm	<2	0.20								

TABLE 4.11. Vessel Steels Composition Summary

Element		Average	% S.D.[a]	Range	Range Factor[b]	No. of Measurements Used
Li	ppm	0.30				1
N	ppm	84				1
Na	ppm	23 ± 24	102	6.6 to 40	6.1	2
Al	ppm	330 ± 400	102	<50 to 613	712	2
Cl	ppm	40				1
K	ppm	12 ± 12	100	<3 to 20		2
Ca	ppm	14				1
Sc	ppm	0.26				1
Ti	ppm	<2				1
V	ppm	<80		<30 to 90		10
Cr	%	0.17 ± 0.13		0.06 to 0.36		10
Mn	%	1.02 ± 0.41		0.2 to 1.49		10
Fe	%	98				10
Co	ppm	122 ± 41	34	93 to 151		2
Ni	%	0.66 ± 0.13	20	<0.3 to 0.76		10
Cu	ppm	1274 ± 400	31	675 to 1900		10
Zn	ppm	≈100		<4 to 210	753	10
Ga	ppm	≈80		<20 to 300		10
As	ppm	532 ± 1085	204	50 to 3600	72.0	10
Se	ppm	0.7				1
Br	ppm	0.85				1
Rb	ppm	48 ± 35		23 to 72		2
Sr	ppm	0.15				1
Y	ppm	<20				10
Zr	ppm	<10				10
Nb	ppm	18.8 ± 12.2	65	<8 to 40		10
Mo	ppm	0.56 ± 0.05	10	0.45 to 0.63		10
Ag	ppm	<2				1
Sb	ppm	11 ± 7	64	6.1 to 16		2
Cs	ppm	<0.2				1
La	ppm	0.10				1
Ba	ppm	273				1
Ce	ppm	<1				1
Sm	ppm	0.017				1
Eu	ppm	0.031				1
Tb	ppm	0.45				1
Dy	ppm					
Ho	ppm	<0.8				2
Yb	ppm	<1				2
Lu	ppm	<0.2				2
Hf	ppm	0.21				1
Ta	ppm	0.13				1
W	ppm	5.5 ± 0.6				2
Pb	ppm	820 ± 2430				10
Th	ppm	0.18				1
U	ppm	0.20				1

(a) Relative standard deviation expressed as a percentage of the average value

(b) Range factor shown is the ratio of the highest to lowest value measured carbon steel are probably related to the original ore body rather than any manufacturing artifacts, as may be the case with stainless.

4.2.4 Rebar

Table 4.12 lists results for the analysis of a total of 48 rebar samples with a summary in Table 4.13. The composition is relatively constant both within a given site as well as between sites. The cobalt is about the same as for the vessel steels at about 100 ppm. Except for the Wolf Creek samples, all are low in niobium. An interesting feature of the rebar is the presence of measurable amounts of silver in a number of the samples. This may be caused by the use of recycled materials in the manufacture of rebar. The amounts involved are too small to result in any significant amount of activation products.

4.2.5 Concrete

Table 4.14 lists concrete composition results for 36 samples taken at 12 sites. In most cases, compositional variation was rather minimal at a given site while large differences are seen between sites reflecting geological differences between quarry sites. The range of variation is summarized in Table 4.15. Range factors for key elements are between one and two orders of magnitude. The europium isotopes, for example, would be much more abundant at the WPPSS site which uses a basalt aggregate than at sites such as Turkey Point or the two TVA reactors which use a limestone aggregate. The limestone-based concretes, however, are higher in other activatible elements such as Ca, and U. Each reactor would thus have to be handled in a site-specific fashion if a detailed activation products inventory is required. This situation is illustrated in Table 4.16 which tabulates the cobalt to europium ratios for several concretes. This ratio varies by about a factor of three thus making a significant difference in whether ^{60}Co or ^{152}Eu would dominate.

The bioshield concrete obtained in this study can be regarded as representative of rather ordinary rock. This is illustrated in Figure 4.1 in which the average elemental abundances given in Table 4.15 are normalized to the average crustal abundance pattern given by Mason (1966). The large error bars simply reflect the average range of variability. The dashed line in the figure at unity represents the composition of typical crustal rock. Most of the elements plotted fall close to or below this line. Calcium and sulfur are higher due to the addition of cement. A few other elements were up to an order of magnitude higher. These include As, Se, Mo, Sb, and Pb, none of which influence the activation products inventory significantly. Interestingly, Eu and Co both plotted below crustal abundance.

TABLE 4.12. Chemical Analysis of Rebar

Element		WPPSS #1 (4)	Rancho Seco (1)	Bellefonte (4)	Wolf Creek (10)	Enrico Fermi (4)	Susquehanna (1)	Palo Verde (11)	Waterford (3)
Li	ppm	0.10							
N	ppm	77 ± 5							
Na	ppm	6.5 ± 0.9	10.5	6.9 ± 2.4	6.5 ± 1.5	6.2 ± 0.1	5.6	8.1 ± 2.3	
Al	ppm	<400	<500	59 ± 12		<100	296		
K	ppm	<3	<100	<30	<50	<30	73	<60	
Ca	ppm	50		900 ± 700	<400	<400	<1000	<400	
Sc	ppm	<0.1	<0.1	<0.05	<0.05	<0.04	<0.06	0.18 ± 0.12	
Ti	ppm	<3000	500	<600	<600	<700	<700	<600	<400
V	ppm	223 ± 38	280	79 ± 22	149 ± 211	220 ± 190	334	182 ± 157	<140
Cr	%	0.11 ± 0.02	0.12	0.16 ± 0.015	0.15 ± 0.05	0.12 ± 0.04	0.16	0.14 ± 0.05	0.09 ± 0.01
Mn	%	1.30 ± 0.05	0.80	0.90 ± 0.13	0.79 ± 0.06	0.93 ± 0.31	0.77	1.1 ± 0.2	0.94 ± 0.012
Fe	%	98	98	98	98	98	98	98	98.5
Co	ppm	117 ± 9	126	85 ± 11	80 ± 4	83 ± 14	115	115 ± 9	
Ni	ppm	1258 ± 83	1150	1130 ± 150	1250 ± 300	780 ± 300	1200	1200 ± 200	590 ± 60
Cu	ppm	2765 ± 280	4000	3200 ± 320	2670 ± 1690	2633 ± 790	2530	2670 ± 1060	3550 ± 110
Zn	ppm	59 ± 11	160	40 ± 25	50 ± 36	51 ± 6	56	53 ± 30	<90
Ga	ppm	<30	<50	<40	<40	<40	<10	<40	<70
As	ppm	148 ± 67	162	117 ± 8	121 ± 16	93 ± 11	101	156 ± 59	133 ± 6
Se	ppm	<3	<3	<1.5	<1	<1	<1.5	<0.7	<30
Br	ppm	<0.6	<0.7	<0.4	<0.5	<0.4	<3.7	1.6 ± 1.0	
Rb	ppm	<30	<30	16 ± 5	<15	<20	<40	<10	<20
Sr	ppm	0.40	<10	<9	<7	<8	<20	<8	<10
Y	ppm	<10	<20	<7	<7	<7	<10	<7	<10
Zr	ppm	<5	<200	<6	<6	<6	<10	<6	<10
Nb	ppm	<5	<150	<6	151 ± 31	<6	<10	<7	<10
Mo	ppm	193 ± 20	<150	261 ± 75	274 ± 140	216 ± 80	263	231 ± 48	157 ± 6
Ag	ppm	5.0 ± 1.4	6	3.5	3.4 ± 1.2	<3	<2	6.7 ± 4.2	
Sb	ppm	48 ± 3	101	36 ± 6	32 ± 8	32 ± 13	43	36 ± 5	
Cs	ppm	<1	<1	<0.2	<0.3	<0.3	<0.6	<0.3	
Ba	ppm	<200	<200	<500	<300	<500	<400	<400	
La	ppm	<0.1	<0.1	<0.2	<0.2	<0.03	<0.21	<0.3	
Ce	ppm	<0.3	<4	<2	2.2 ± 0.8	2.1 ± 0.4	2.6	<3	
Sm	ppm	<0.04	<0.04	<0.02	<0.02	<0.01	<0.02	<0.03	
Eu	ppm	<0.08	<0.09	0.055 ± 0.014	0.05 ± 0.01	<0.04	<0.08	<0.05	
Dy	ppm	<3	<2	<1	<1	<1		<0.9	
Ho	ppm	<0.03	<0.4						
Yb	ppm	<1	<1	<0.7	<0.7	<0.6	<0.8	<0.7	
Lu	ppm	<0.1	<0.1	0.49 ± 0.23	<0.2	<0.6	<0.2	<0.1	
Hf	ppm	<0.7	<0.8	<0.5	<0.3	<0.4	<0.4	<0.3	
Ta	ppm	<0.5	<0.5	<0.6	<0.3	<0.5	0.51	<0.5	
W	ppm	7.7 ± 1.0	12	19 ± 7	13 ± 7	12 ± 10	17.1	26 ± 16	
Pb	ppm	150	<110	<60	<60	<60	100	<60	<80
Th	ppm	<0.6	<0.6	0.3 ± 0.1	<0.3	<0.3	<0.6	0.32 ± 0.09	
U	ppm	<3	<4	<0.6	<0.7	<0.4	<0.9	<0.5	

Note: Numbers in parenthesis indicate number of samples analyzed at each site. Numbers in the table are averages and standard deviations for that number of samples.

TABLE 4.13. Rebar Composition Summary

Element	Average	%(a) S.D.	Range	Range Factor(b)	No. of Measurements Used
Li ppm	0.10				1
N ppm	77				1
Na ppm	7.2 ± 1.7	23	5.6 to 10.5	1.9	7
Al ppm	180 ± 170	94	59 to 296	5.0	2
K ppm	≈40		<3 to 73	724	7
Ca ppm	≈50				1
Sc ppm	<0.1				7
Ti ppm	<700				7
V ppm	201 ± 81	41	74 to 334	4.2	8
Cr %	0.131 ± 0.025	19	0.09 to 0.16	1.8	8
Mn %	0.94 ± 0.18	19	0.77 to 1.30	1.7	8
Fe %	~98				8
Co ppm	103 ± 19	19	80 to 126	1.6	78
Ni ppm	1070 ± 250	23	590 to 1260	2.1	8
Cu ppm	2980 ± 550	18	2530 to 4000	1.6	8
Zn ppm	67 ± 41	62	40 to 160	4.0	7
Ga ppm		<50			7
As ppm	129 ± 25	20	93 to 162	1.7	8
Se ppm		<2			7
Br ppm	<1.1		<0.4 to 3.7	<9.3	8
Rb ppm	<20				8
Sr ppm	0.40				1
Y ppm		<10			8
Zr ppm		<10			7
Nb pm		<40	<5 to 151		8
Mo ppm	218 ± 48	22	150 to 274	1.8	8
Ag ppm	4.2 ± 1.7	40	<2.0 to 6.7	3.4	7
Sb ppm	47 ± 24	51	32 to 101	3.2	7
Cs ppm	<0.6				5
Ba ppm	<400				5
La ppm	<0.3				7
Ce ppm		<3			6
Sm ppm	<0.04				7
Eu ppm	<0.09				7
Dy ppm		<1			4
Ho ppm	<0.04				2
Yb ppm		<1			7
Lu ppm	<0.6				7
Hf ppm	<0.8				7
Ta ppm	<0.6				7
W ppm	15 ± 6	39	7.7 to 26.0	7	
Pb ppm		<90			8
Th ppm	<0.6				7
U ppm	<0.9				5

(a) Relative Standard Deviation expressed as a percentage of the average value.

(b) Range factor shown is the ratio of the highest to lowest value measured.

TABLE 4.14. Chemical Analysis of Bioshield Concrete

Element		WPPSS (4)	Rancho Seco (1)	Bellefonte (4)	Hartsville (3)	Wolf Creek (3)	Palo Verde (2)	Enrico Fermi (2)	Turkey Point (2)	Pathfinder (3)	Humboldt Bay (4)	Waterford (2)	Susquehanna (5)
H	ppm	6100 ± 30	6000										
B	ppm	15.3 ± 0.2	26										
N	ppm	107 ± 47	140										
Na	ppm	15500 ± 1300	13500	650 ± 160	510 ± 30	390 ± 10	18200 ± 1600	5240 ± 1950	176 ± 23	6320 ± 1480	18940 ± 1690		1830
Al	%	5.5 ± 0.3	5.7	1.5 ± 0.3	1.3 ± 0.3	1.8 ± 0.1	6.1 ± 0.1	2.0 ± 0.4	0.53 ± 0.01	2.2 ± 0.3	5.1 ± 0.5	1.6 ± 0.1	2.1
Si	%	21.5 ± 1.7	23.9	3.9 ± 0.7	4.2 ± 0.5	12.3 ± 2.2	26.7 ± 2.7	9.2 ± 4.0	8.5 ± 1.3	25.4 ± 1.6	20.3 ± 2.71	32.4 ± 2.4	12.7
P	%	0.81 ± 0.33	0.92	<0.5	<0.5	<0.9	<0.5	<0.2	<0.2	<0.1	<0.1	<0.4	<0.2
S	%	0.46 ± 0.12	0.44	0.26 ± 0.05	0.33 ± 0.02	0.25 ± 0.08	0.27 ± 0.12	0.42 ± 0.06	0.20 ± 0.04	0.20 ± 0.04	0.20 ± 0.07	0.43 ± 0.11	0.3
Cl	ppm	59 ± 12	56		23 ± 4		56 ± 22	51 ± 6	11.0 ± 1.5	46 ± 19	58 ± 31		
K	%	1.2 ± 0.4	1.1	0.29 ± 0.04	0.31 ± 0.05	0.93 ± 0.06	2.5 ± 0.3	0.60 ± 0.34	0.047 ± 0.002	0.61 ± 0.11	0.84 ± 0.12	0.080 ± 0.001	0.5
Ca	%	14.0 ± 2.2	12.3	32.4 ± 1.9	34.7 ± 2.8	26.8 ± 1.7	9.2 ± 2.1	17.5 ± 3.5	29.4 ± 0.9	8.3 ± 1.9	8.5 ± 2.8	12.0 ± 1.6	15.0
Sc	ppm	17.4 ± 0.6	16.2	2.9 ± 0.4	2.3 ± 0.6	1.09 ± 0.03	5.3 ± 1.1	3.0 ± 0.6	0.73 ± 0.01	2.4 ± 0.6	14.7 ± 2.0		6
Ti	ppm	7900 ± 1400	4100	840 ± 80	850 ± 130	660 ± 360	2100 ± 300	1030 ± 260	230 ± 40	620 ± 30	2850 ± 140	600 ± 14	3600
V	ppm	180 ± 8	150	39 ± 4	37 ± 12	28.5 ± 1.4	34 ± 7	32 ± 7	13.3 ± 0.6	16 ± 3	115 ± 8		490
Cr	ppm	37 ± 4	209	30 ± 2	19 ± 4	31 ± 2	47 ± 12	26.7 ± 1.3	19.1 ± 0.2	40 ± 16	540 ± 330		200
Mn	ppm	990 ± 100	710	127 ± 11	214 ± 15	340 ± 70	290 ± 50	270 ± 20	56 ± 5	214 ± 6	590 ± 60	80 ± 4	640
Fe	%	5.0 ± 0.2	3.6	0.9 ± 0.3	1.1 ± 0.2	1.05 ± 0.12	1.9 ± 0.4	1.1 ± 0.3	0.50 ± 0.01	1.2 ± 0.1	4.4 ± 0.7	0.70 ± 0.09	24
Co	ppm	24 ± 2	15	4.1 ± 0.5	1.0 ± 0.7	1.6 ± 0.6	5.6 ± 2.0	3.8 ± 1.0	1.1 ± 0.10	2.1 ± 1.7	17.0 ± 2.6		31
Ni	ppm	23 ± 3	70	35 ± 6	31 ± 8	28 ± 3	30 ± 4	14 ± 4	<14	44 ± 3	87 ± 4	11.9 ± 0.4	70
Cu	ppm	44 ± 19	46	21 ± 4	16 ± 4	11 ± 2	28 ± 7	10 ± 4	10 ± 4	12 ± 2	32 ± 4	10.9 ± 1.1	60
Zn	ppm	340 ± 80	68	77 ± 13	33 ± 8	15.0 ± 1.4	90 ± 60	33 ± 6	8.4 ± 0.5	30 ± 6	106 ± 18	15.2 ± 15	80
Ga	ppm	14 ± 2	12	3.3 ± 0.4	3.7 ± 0.8	7 ± 6	13 ± 1	5.5 ± 1.0	1.05 ± 0.7	5.9 ± 0.6	18 ± 4	1.8 ± 0.3	20
As	ppm	29 ± 50	6.6	13.6 ± 5.1	4.6 ± 1.2	4.2 ± 0.4	13.6 ± 1.7	4.9 ± 1.9	0.89 ± 0.08	3.0 ± 0.3	6.2 ± 1.1	2.8 ± 1.2	9
Se	ppm	<1	<1	3.99 ± 0.14	0.57 ± 0.32	<0.3	2.0 ± 0.3	0.71 ± 0.26	0.26 ± 0.04	0.71 ± 0.21	1.2 ± 0.3	<1.1	0.56
Br	ppm	<1	<1	2.4 ± 1.0	1.4 ± 0.4	1.0 ± 0.2	2.0 ± 0.9	5.6 ± 1.6	1.7 ± 0.3	3.3 ± 1.3	3.5 ± 1.8	<0.8	1
Rb	ppm	35 ± 4	29	15 ± 2	13 ± 2	32.7 ± 1.5	170 ± 110	22 ± 3	2.5 ± 0.4	20 ± 3	38 ± 6	2.92 ± 0.06	40
Sr	ppm	367 ± 11	422	560 ± 120	490 ± 10	940 ± 50	220 ± 40	310 ± 80	680 ± 40	260 ± 100	240 ± 40	361 ± 15	410
Y	ppm	26 ± 6	16	10 ± 1	6.9 ± 1.7	6 ± 1	96 ± 97	6.7 ± 2.4	3.0 ± 1.1	4.5 ± 2.2	18 ± 3	4.7 ± 0.4	20
Zr	ppm	144 ± 6	92	35 ± 6	33 ± 9	40 ± 4	160 ± 6	52 ± 17	27 ± 4	54 ± 13	90 ± 14	40 ± 4	90
Nb	ppm	9.3 ± 1.0	5.4	1.4 ± 0.8	2.0 ± 0.7	1.3 ± 0.2	9.2 ± 1.1	1.8 ± 0.7	<2	2.1 ± 0.1	5.6 ± 0.5	3.7 ± 0.05	7
Mo	ppm	8.0 ± 7.4	2.5	3.0 ± 1.4	4.8 ± 1.2	1.8	7.2 ± 0.9	3.5 ± 0.5	26 ± 16	36 ± 5	16 ± 3	4.35 ± 0.27	10
Pd	ppm	<3	<3										
Ag	ppm	0.10 ± 0.11	<0.4	<0.4	<0.3	<0.2	<0.3	<0.2	<0.2	<0.3	<0.5		<0.5
Cd	ppm	0.20 ± 0.04	0.35										
Sn	ppm	6.8 ± 3.7	<3	<9	<9	<8	<8	<6	<10	<9	<4		
Sb	ppm	13 ± 24	1.0	1.0 ± 0.3	0.8 ± 0.3	0.3 ± 0.1	1.4 ± 0.1	0.5 ± 0.4	0.16 ± 0.01	0.40 ± 0.11	0.9 ± 0.2		0.8
Cs	ppm	1.1 ± 0.3	1.3	0.7 ± 0.2	0.7 ± 0.2	0.36 ± 0.02	6.2 ± 2.2	0.54 ± 0.01	0.52 ± 0.01	0.32 ± 0.02	0.94 ± 0.20	<1.4	
Ba	ppm	480 ± 80	630	112 ± 22	93 ± 12	360 ± 30	885 ± 7	7060 ± 9750	<20	280 ± 40	840 ± 100	434 ± 47	160
La	ppm	21 ± 4	15	11.6 ± 1.3	7.9 ± 1.4	8.7 ± 0.4	28 ± 2	9.4 ± 0.6	2.9 ± 0.2	12.5 ± 0.3	16.4 ± 0.7		10
Ce	ppm	42 ± 6	28	18 ± 2	14.1 ± 2.5	14.1 ± 0.9	52 ± 1	15.5 ± 0.7	6.2 ± 0.7	26.4 ± 0.4	31 ± 4		20
Nd	ppm	20 ± 2	14					<17	<20	<23	<20		
Sm	ppm	4.2 ± 0.3	0.42	1.8 ± 0.5	1.4 ± 0.3	1.34 ± 0.05	3.9 ± 0.3	1.7 ± 0.2	0.61 ± 0.04	1.55 ± 0.12	3.4 ± 0.5		
Eu	ppm	1.2 ± 0.1	1.0	0.38 ± 0.04	0.27 ± 0.06	0.31 ± 0.02	0.77 ± 0.01	0.34 ± 0.06	0.11 ± 0.015	0.21 ± 0.18	1.01 ± 0.13		
Gd	ppm	4.0 ± 0.3	3.0										
Tb	ppm	0.69 ± 0.03	0.48	0.29 ± 0.03	0.24 ± 0.04	0.19 ± 0.03	0.78 ± 0.14	0.29 ± 0.06	0.11 ± 0.01	0.27 ± 0.05	0.77 ± 0.11		
Dy	ppm	0.3 ± 0.3	2.9				3.0 ± 0.6	1.15 ± 0.07	0.55 ± 0.20	1.8 ± 0.3	2.6 ± 0.5		
Ho	ppm	1.0 ± 0.2	<0.4				0.85 ± 0.08						
Yb	ppm	2.4 ± 0.1	1.6	0.98 ± 0.07	0.79 ± 0.11	0.64 ± 0.05	3.0 ± 0.8	0.9 ± 0.2	0.38 ± 0.01	0.88 ± 1.11	2.4 ± 0.2		
Lu	ppm	0.36 ± 0.03	0.31	0.23 ± 0.03	0.18 ± 0.04	0.15 ± 0.03	0.15 ± 0.11	0.19 ± 3.01	0.20 ± 0.05	0.19 ± 0.06	0.49 ± 0.05		
Hf	ppm	3.6 ± 0.1	2.7	0.80 ± 0.10	0.98 ± 0.07	1.39 ± 0.15	5.7 ± 0.8	2.0 ± 1.2	0.65 ± 0.04	1.9 ± 0.6	3.1 ± 0.8		
Ta	ppm	0.73 ± 0.07	0.46	0.22 ± 0.03	0.20 ± 0.04	0.17 ± 0.03	0.90 ± 0.32	0.24 ± 0.04	0.092 ± 0.06	0.23 ± 0.03	0.66 ± 0.15		
W	ppm	1.3 ± 0.7	1.1	0.8 ± 0.4	0.5 ± 0.3	<0.5	3.9 ± 0.4	0.6 ± 0.3	0.39 ± 0.16	2.1 ± 1.7	1.3 ± 0.6		
Pb	ppm	24 ± 32	8.0	9.6 ± 3.8	5.4 ± 3.2	7.8 ± 8.3	34 ± 6	5.9 ± 5.2	<6	15 ± 13	560 ± 660		
Th	ppm	4.2 ± 0.6	3.3	2.4 ± 0.3	1.9 ± 0.5	1.9 ± 0.1	12 ± 2	2.11 ± 0.06	0.75 ± 0.09	3.36 ± 0.01	4.4 ± 1.0		
U	ppm	1.40 ± 0.05	1.5	4.0 ± 0.5	2.8 ± 0.7	2.8 ± 0.4	2.7 ± 0.8	3.0 ± 0.7	4.4 ± 0.3	1.8 ± 0.6	2.1 ± 0.3		

(a) Based on analysis of ingredients.
Note: Number in parenthasis.

TABLE 4.15. Bioshield Concrete Composition Summary

Element		Average	% S.D.(b)	Range	Range Factor(a)	No. of Measurements(a) Used
H	ppm	≈6100				2
Li	ppm	20				Crustal Abundance
B	ppm	≈20				2
N	ppm	≈120				2
Na	ppm	7390 ± 7640	103	17[illegible] to 18940	108	11
Al	%	3.1 ± 2.0	67	0.53 to 6.1	11.5	11
Si	%	16.8 ± 9.5	[illegible]7	3.9 to 32.4	8.3	12
P	%	<0.5				12
S	%	0.31 ± 0.10	32	0.20 to 0.46	2.3	12
Cl	ppm	45 ± 18	40	11 to [illegible]	5.9	8
K	%	0.75 ± 0.61	8[illegible]	0.047 to 2.5	53.2	12
Ca	%	18.3 ± 9.7	5[illegible]	8.3 to 34.7	4.2	12
Sc	ppm	6.5 ± 6.3	97	0.73 to 17.4	23.8	11
Ti	ppm	2121 ± 2320	105	230 to 7900	34.4	12
V	ppm	103 ± 140	136	13.3 to 490	36.8	11
Cr	ppm	109 ± 159	146	29.0 to 540	28.4	11
Mn	ppm	377 ± 290	77	56 to 990	17.7	12
Fe	%	3.9 ± 6.4	172	0.50 to 24.0	48.0	12
Co	ppm	9.8 ± 10.3	105	1.1 to 31.0	2[illegible]	11
Ni	ppm	38 ± 25	65	11.9 to 37.0	[illegible]	12
Cu	ppm	25 ± 17	68	10 to 6[illegible]	6.[illegible]	12
Zn	ppm	75 ± 90	120	8.4 to 340	40.5	12
Ga	ppm	8.8 ± 6.4	73	1.05 to 20	19.1	12
As	ppm	7.9 ± 7.9	100	0.89 to 29	32.6	12
Se	ppm	0.92 ± 0.56	61	0.26 to 2.0	7.7	7
Br	ppm	2.4 ± 1.5	61	1.0 to 5.6	5.6	9
Rb	ppm	35 ± 44	127	2.5 to 170	68.0	[illegible]
Sr	ppm	[illegible] ± 208	47	220 to [illegible]	4.3	[illegible]
Y	ppm	18.2 ± 25.6	147	3.0 to 96	32.0	12
Zr	ppm	71 ± 44	62	27 to 160	5.9	12
Nb	ppm	4.3 ± 3.0	69	1.3 to 9.3	7.2	12
Mo	ppm	10.3 ± 10	104	1.8 to 36	10.0	12
Pd	ppm	<3				
Ag	ppm	<0.2				
Cd	ppm	≈0.3				2
Sn	ppm	<7				
Sb	ppm	1.8 ± 3.7	202	0.16 ± 13.0	[illegible]1.3	11
Cs	ppm	1.3 ± 1.8	139	0.32 ± 6.2	19.4	10
Ba	ppm	950 ± 1950	[illegible]	<20 to 7060	353	[illegible]
La	ppm	13.0 ± 6.9	53	2.9 to 28	9.7	1[illegible]
Ce	ppm	24.3 ± 13.5	55	6.2 to 52	8.4	1[illegible]
Sm	ppm	2.0 ± 1.3	63	0.42 to 4.2	10.0	[illegible]
Eu	ppm	0.55 ± 0.38	69	0.11 ± 1.2	10.9	1[illegible]
Tb	ppm	0.41 ± 0.24	59	0.11 to 0.78	7.1	11
Dy	ppm	2.3 ± 1.3	54	0.55 to 4.3	[illegible].8	7
Ho	ppm	≈0.9				R.E. Ratio
Yb	ppm	1.4 ± 0.9	63	0.38 ± 3.0	7.9	1[illegible]
Lu	ppm	0.27 ± 0.13	48	0.15 to 0.50	3.[illegible]	[illegible]
Hf	ppm	2.2 ± 1.5	71	0.65 to 5.7	8.[illegible]	11
Ta	ppm	0.44 ± 0.31	70	0.092 to 0.90	9.8	[illegible]
W	ppm	1.4 ± 1.1	81	0.39 to 3.9	10.[illegible]	[illegible]
Pb	ppm	61 ± 158	258	5.4 to 560	104	12
Th	ppm	3.5 ± 3.0	85	0.75 ± 120	16.0	[illegible]
U	ppm	2.7 ± 0.9	36	1.4 ± 4.4	3.1	11

(a) Range factor is the ratio of the highest to lowest value measured.

(b) Relative standard deviation expressed as a percentage of the average value.

TABLE 4.16. Cobalt to Europium Ratio in Bioshield Concrete

WPPSS	20.0
Ranch Seco	15.0
Bellefonte	10.8
Hartsville	15.2
Wolf Creek	9.7
Palo Verde	7.2
Enrico Fermi	11.5
Turkey Point	10.0
Pathfinder	10.0
Humboldt Bay	17.0
Average	12.6 ± 4.0 (31%)
Range	7.2 to 20.0
Range Factor	2.8

4.2.6 Concrete Ingredients

In addition to the whole concrete samples, a selection of concrete ingredients was also collected in order to obtain a better understanding of the origin of the trace elements in the concrete. These ingredients are summarized in Tables 4.17 to 4.20 for cement, coarse aggregate, sand aggregates, and fly ash, respectively. These were retabulated for the Palo Verde, Enrico Fermi, and Hartsville sites respectively on Tables 4.21 through 4.23. In most cases the coarse and fine aggregates are of very similar composition. Since concrete is normally about 75% aggregate, the aggregate tends to dominate the composition for most elements. A few elements are contributed mainly by the cement. These include S, Ca, Sr, and in some cases Zn. The use of coal flyash as a cement substitute has become more common in recent years. Several of the reactor sites included in this study provided fly ash along with other ingredients. For the Hartsville samples, a regression analysis was carried out to determine the relative contribution of each component on an element by element basis. This data gave a good fit to an aggregate component of 73.4 ± 9.6%, with 19.7 ± 4.5% cement and 4.0 ± 0.5% fly ash. This is in reasonably good agreement with the engineering specifications for the concrete which require 4.9% fly ash. The fit is quite good for most elements as shown in Figure 4.2 which gives a plot of the standardized residuals. Although the fly ash is a relatively small fraction of the total concrete by weight it has a disproportionately large influence on the trace element content. This situation is graphically illustrated in Figure 4.3 with the dashed line representing the weight percent contribution of fly ash. All elements except Ca, Mn, and Sr plot well above the line, with typical values ranging from 20 to 40%. Elements whose concentrations are more than doubled by the addition of fly ash include

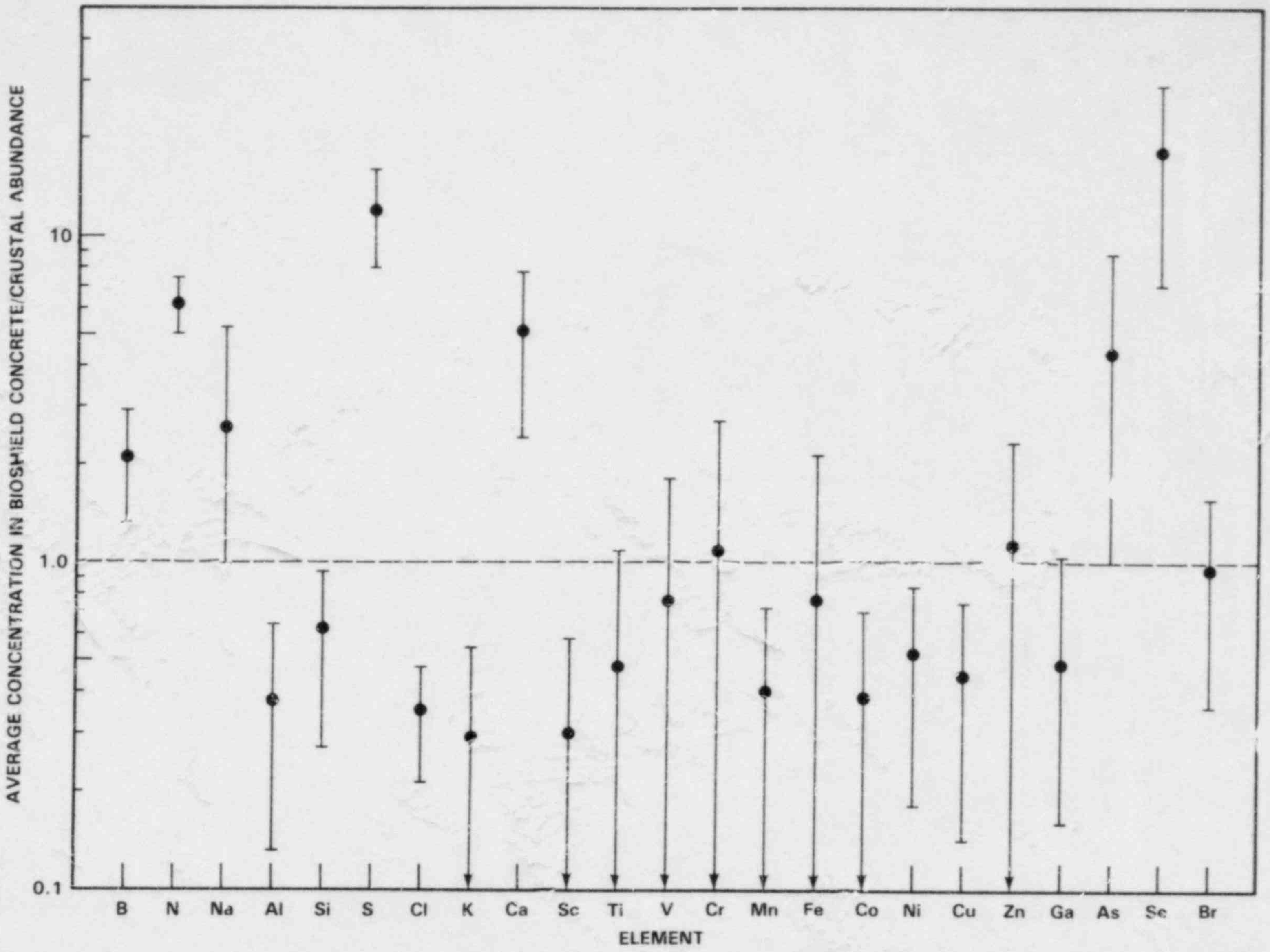

FIGURE 4.1a. Average Elemental Concentrations in Bioshield Concrete Normalized to Average Crustal Abundance

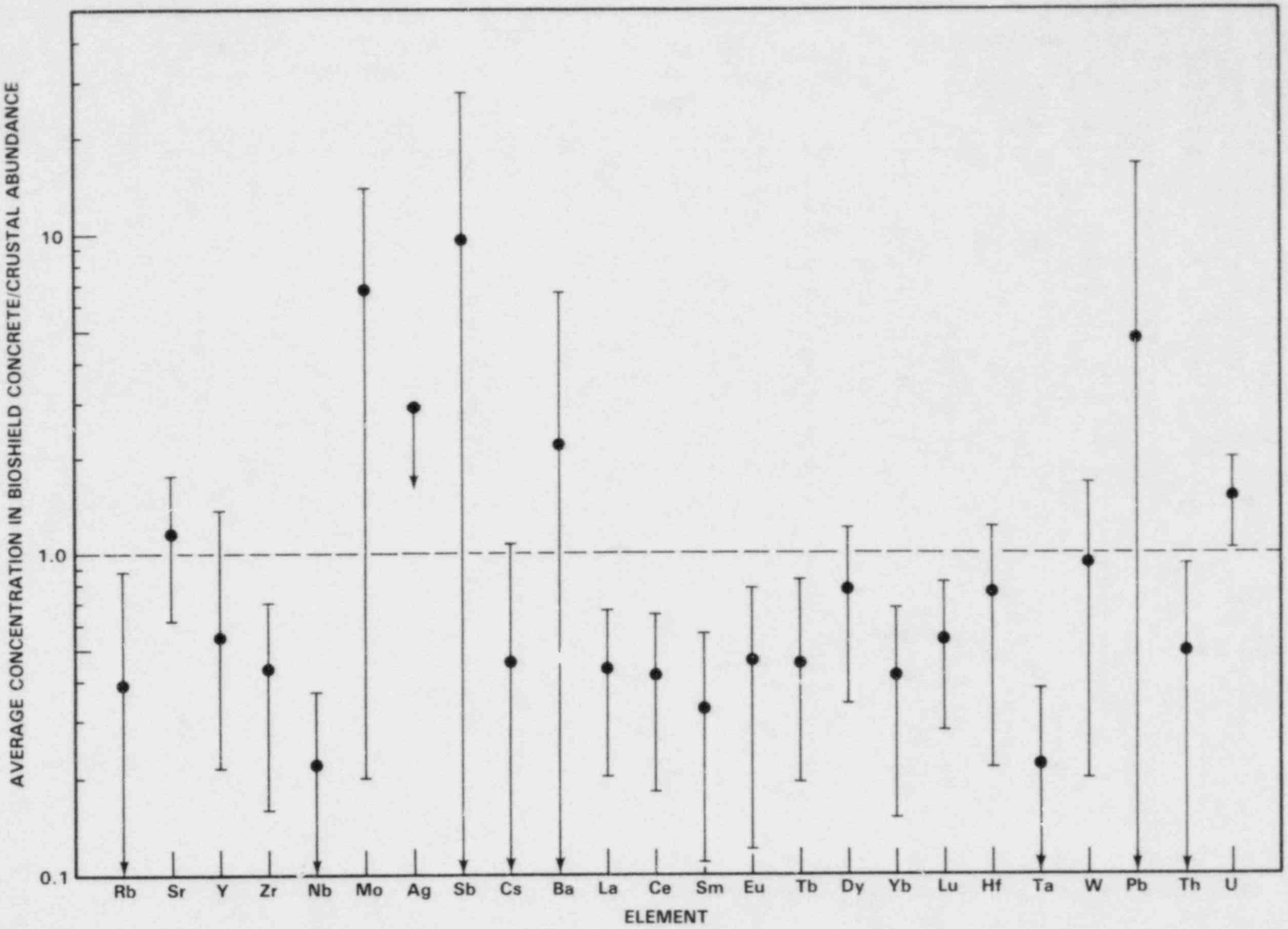

FIGURE 4.1a. (contd)

TABLE 4.17. Chemical Analysis of Cement Used for Bioshield Concrete

Element		WPPSS	Belle Fonte	Hartsville	Wolf Creek	Palo Verde	Enrico Fermi	Susquehanna
Na	%	0.076	0.090	0.14	0.25	0.10	0.24	0.20
Al	%	2.3	2.5	2.4	2.2	2.2	2.6	2.2
Si	%	11.0	10.4	11.4	10.9	10.0	10.9	10.7
P	%	<0.6	<0.6	<0.6	<0.7	<0.6	<0.6	<0.6
S	%	1.4	1.3	0.88	1.0	1.6	1.1	1.1
K	%	0.56	0.49	0.40	0.27	22	0.7	60
Ca	%	41.6	45.5	46.1	46.0	48.5	45.3	46.1
Sc	ppm	2.99	4.3	3.5	3.9	4.3	4.2	3.1
Ti	ppm	1100	960	850	1060	1810	1210	1040
V	ppm	28	44	41	51	43	43	36
Cr	ppm	66	52	28	72	105	67	272
Mn	ppm	1090	280	191	658	347	414	616
Fe	%	2.17	1.86	2.00	3.35	2.12	1.78	2.29
Co	ppm	29	7.0	5.3	4.4	9.9	5.5	7.8
Ni	ppm	32	32	34	43	62	36	108
Cu	ppm	77	72	27	17	85	18	43
Zn	ppm	988	230	56	33	470	37	31
Ga	ppm	8.0	6.3	5.1	7.1	5.8	7.1	7.5
As	ppm	2.9	8.4	6.5	12.5	22	6.8	5.5
Se	ppm	1.6	0.62	0.52	1.1	1.3	<0.2	1.7
Br	ppm	<0.3	0.56	0.42	<0.4	0.73	<0.4	<0.2
Rb	ppm	14	18	12	7.5	16	22	28
Sr	ppm	853	1140	789	2210	460	205	1500
Y	ppm	9.0	15	8.6	9.7	11	15	7.6
Zr	ppm	17	<3	<3	<4	76	55	32
Nb	ppm	4.1	3.0	3.1	2.8	7.1	3.9	2.2
Mo	ppm	4.1	<1	<1	<1.5	4.7	5.0	20
Ag	ppm	<0.3	0.2	<0.5	<0.3	<0.7	<0.4	<0.4
Sb	ppm	3.8	0.62	0.48	0.61	1.29	0.59	0.63
Cs	ppm	<4	1.2	<5	1.4	<3	<4	1.32
Ba	ppm	514	134	159	2120	210	266	128
La	ppm	12	18	11	13	17.8	16	10.8
Ce	ppm	22	28	20	25	29	26	21
Sm	ppm	1.7	3.2	1.7	2.3	2.4	2.5	1.6
Eu	ppm	0.38	0.64	0.38	0.50	0.58	0.51	0.33
Tb	ppm	0.27	0.51	0.29	0.36	0.35	0.38	0.26
Dy	ppm	1.6	1.4					
Yb	ppm	0.96		1.0	1.3	1.9	1.4	0.87
Lu	ppm	0.40	0.31	0.18	0.30	1.13	0.34	0.18
Hf	ppm	1.6	1.6	1.2	3.0	2.2	1.9	1.1
Ta	ppm	0.28	0.32	0.26	0.37	0.63	0.35	0.26
W	ppm	1.4	0.54	0.47	1.0	0.72	0.86	3.3
Pb	ppm	6.9	17	<3	<3	33	<3	20
Tb	ppm	3.5	3.8	2.5	4.1	3.2	3.3	2.6
U	ppm	2.2	2.8	1.9	3.5	1.5	6.0	1.7

TABLE 4.18. Chemical Analysis of Coarse Aggregate for Bioshield Concrete

Element		WPPSS (4)	Belle Fonte (3)	Hartsville (3)	Wolf Creek (2)	Palo Verde (2)	Susquehanna (Normal)	Susquehanna (High Density)
Na	%	2.15 ± 0.06	0.043 ± 0.018	0.045 ± 0.019	0.052 ± 0.03	2.07 ± 0.09	0.12	0.0071
Al	%	7.08 ± 0.13	0.41 ± 0.18	0.43 ± 0.02	0.43 ± 0.20	7.1 ± 0.5	1.11	0.36
Si	%	29.1 ± 1.6	2 ± 0.2	<2	<5	33.3 ± 0.2	4.7	
P	%	<0.6	0.2	<0.5	<1	<0.5	<0.5	
S	%	<0.2	<0.2	0.18 ± 0.07	<0.1	<0.2	0.13	
K	%	1.67 ± 0.13	0.135 ± 0.066	0.32 ± 0.04	0.11 ± 0.06	2.9 ± 0.6	0.37	<0.1
Ca	%	4.4 ± 0.3	35.5 ± 1.4	34.4 ± 3.6	38.3 ± 0.6	0.98 ± 0.06	30.6	0.55
Sc	ppm	23.1 ± 1.7	0.85 ± 0.36	1.03 ± 0.13	0.67 ± 0.26	5.0 ± 0.05	1.44	1.5
Ti	%	1.08 ± 0.13	0.036 ± 0.005	0.051 ± 0.013	0.054 ± 0.127	0.20 ± 0.03	0.060	0.088
V	ppm	243 ± 29	12.3 ± 6.1	9.9 ± 2.0	9.2 ± 2.6	29.5 ± 0.7	16	50
Cr	ppm	35 ± 2	16.8 ± 5.7	7.8 ± 0.9	9.5 ± 0.9	50 ± 11	13.7	
Mn	ppm	1128 ± 75	87 ± 19	253 ± 118	537 ± 118	384 ± 23	192	670
Fe	%	6.17 ± 0.24	0.26 ± 0.11	0.36 ± 0.03	0.60 ± 0.008	1.8 ± 0.07	0.57	68
Co	ppm	26 ± 3	0.95 ± 0.43	0.98 ± 0.18	1.08 ± 0.03	3.98 ± 0.75	2.0	8.4
Ni	ppm	22 ± 4		19.3 ± 1.5	25 ± 6	9.7 ± 4.7	31	89
Cu	ppm	26.3 ± 3.7	10.3 ± 4.1	6.4 ± 1.1	6.7 ± 0.4	15.5 ± 3.5	10.2	42
Zn	ppm	93 ± 6	40.7 ± 2.9	15.3 ± 5.0	10.4 ± 6.2	43.1 ± 3.0	16.1	22
Ga	ppm	19.0 ± 0.8	0.97 ± 0.85	1.6 ± 0.7	2.9 ± 0.9	13.1 ± 5.4	14.3	21
As	ppm	2.4 ± 0.2	2.1 ± 1.7	0.85 ± 0.12	2.6 ± 1.3	8.6 ± 2.5	1.6	11.8
Se	ppm	1.5	0.3 ± 0.3	0.5 ± 0.4	0.29 ± 0.18	<0.3	<0.2	<0.3
Br	ppm	<0.8	2.5 ± 0.8	0.89 ± 0.03	0.77 ± 0.49	0.95 ± 0.49	2.6	<0.3
Rb	ppm	43 ± 4	6.8 ± 3.4	10.6 ± 1.4	5.8 ± 2.5	109 ± 17	16	40
Sr	ppm	315 ± 13	471 ± 28	426 ± 28	1065 ± 78	131 ± 6	557	5.7
Y	ppm	28 ± 3	6.8 ± 0.6	3.9 ± 0.7	3.2 ± 0.1	39 ± 12	5.3	<3
Zr	ppm	186 ± 22	11.9 ± 3.1	27 ± 3	18 ± 6	198 ± 9	32	10.8
Nb	ppm	12 ± 3	<1	<1	<1	12.5 ± 2.1	1.7	<3
Mo	ppm	3.9 ± 0.7	<2	<1	<1	7.1 ± 2.7	1.9	19
Ag	ppm	0.046	<0.2	<0.2	<0.1	<0.3	<0.2	<0.2
Sb	ppm	0.6 ± 0.5	0.25 ± 0.13	0.068 ± 0.02	0.20 ± 0.01	1.0 ± 0.4	0.12	2.3
Cs	ppm	1.15 ± 0.06	<1	<2	<1.4	<4	0.55	<0.4
Ba	ppm	557 ± 25	42 ± 29	49 ± 10	63 ± 4	825 ± 199	71	212
Ca	ppm	24.8 ± 3.5	6.3 ± 2.5	5.03 ± 0.90	3.1 ± 1.1	30.5 ± 0.4	7.4	2.6
Ce	ppm	49 ± 8	6.6 ± 2.7	8.95 ± 1.20	1.5 ± 1.5	63.0 ± 6.0	12.2	4.4
Sm	ppm	5.2 ± 0.4	0.90 ± 0.33	0.79 ± 0.15	0.57 ± 0.18	4.8 ± 0.3	1.2	0.62
Eu	ppm	1.7 ± 0.05	0.21 ± 0.07	0.16 ± 0.02	0.096 ± 0.017	0.88 ± 0.17	0.25	0.18
Tb	ppm	0.90 ± 0.05	0.17 ± 0.05	0.14 ± 0.02	0.090 ± 0.005	0.94 ± 0.13	0.18	0.34
Dy	ppm	5.5 ± 0.6	0.95	0.73 ± 0.15	0.505 ± 0.049	6.0 ± 1.6	1.2	0.50
Ho	ppm	1.05 ± 0.17						
Yb	ppm	2.87 ± 0.15	0.52 ± 0.13	0.50 ± 0.13	0.27 ± 0.06		0.53	0.58
Lu	ppm	0.49 ± 0.049	0.12 ± 0.03	0.091 ± 0.018	0.096 ± 0.003	3.5 ± 0.4	0.13	1.2
Hf	ppm	4.5 ± 0.7	0.65 ± 0.81	0.83 ± 0.03	0.24 ± 0.13	6.9 ± 0.3	0.78	0.31
Ta	ppm	0.93 ± 0.15	0.067 ± 0.15	0.11 ± 0.02	0.064 ± 0.031	0.92 ± 0.01	0.13	0.85
W	ppm	<1	<0.3	0.18 ± 0.03	0.165 ± 0.007	2.0 ± 0.6	0.20	7.7
Pb	ppm	9.3 ± 0.5	<7	2.8 ± 0.7	<6	15.5 ± 0.7	5.0	28
Th	ppm	5.0 ± 0.95	0.78 ± 0.42	1.0 ± 0.1	0.50 ± 0.21	13.8 ± 3.9	1.25	0.87
U	ppm	1.6 ± 0.5	2.6 ± 0.7	0.82 ± 0.16	2.6 ± 0.7	3.4	1.42	5.95

Note: Number in parenthesis indicate number of samples analyzed at each site. Numbers in the table are averages and standard deviations for that number of samples.

TABLE 4.19. Chemical Analysis of Sand Aggregate for Bioshield Concrete

Element		WPPSS	Bellefonte	Hartsville	Wolf Creek	Palo Verde	Enrico Fermi	Susquehanna (Normal)	Susquehanna (High Density)
Na	%	2.2	0.038	0.045	1.05	2.03	0.73	0.41	0.35
Al	%	7.5	0.39	0.75	3.84	6.79	2.7	4.9	2.7
Si	%	27.1	<1.8	2.4	37.8	34.0	22.6	32.6	
P	%	<0.6	<0.4	<0.5	<1	<0.4	<0.5	<0.4	
S	%	<0.2	0.12	0.12	<0.3	<0.2	0.15	<0.2	
K	%	1.5	0.12	0.29	2.72	2.85	0.82	1.41	0.10
Cu	%	4.6	38.0	36.6	0.46	0.72	14.4	0.36	0.90
Sc	ppm	27	0.95	0.97	0.54	6.0	3.4	7.1	18.1
Ti	ppm	12400	230	450	740	1900	1180	12200	
V	ppm	280	11	8.8	8.2	40	32	73	2510
Cr	ppm	33	12	8.4	48.3	13.2	43	73	815
Mn	ppm	1220	83	237	47	225	420	720	1120
Fe	%	7.0	0.24	0.35	0.58	1.85	1.36	2.92	38.2
Ce	ppm	27	0.59	0.96	1.91	4.9	4.3	10.4	139
Ni	ppm	23	9	19	6.9	15	28	32	187
Cu	ppm	23	6.5	7.4	9.9	13	16	24	24
Zn	ppm	102	30	15.5	9.3	31	33	58	297
Ga	ppm	20	0.94	1.0	8.4	14	6.3	9.7	43
As	ppm	2.4	3.5	0.95	2.4	11.1	4.4	11.2	<1.2
Se	ppm	2	0.36	0.91	0.82	2.3	<0.2	<0.4	<0.8
Br	ppm	<0.8	2.[illegible]	0.86	1.5	<0.8	4.2	<0.5	<0.8
Rb	ppm	46	6.0	9.4	101	126	27	73	35
Sr	ppm	354	485	422	124	180	170	47	131
Y	ppm	33	7.0	4.5	8.0	26	10	28	5.2
Zr	ppm	193	12	25	60	210	69	263	162
Nb	ppm	12	<1	<1	2.8	7.4	2.6	15	16
Mo	ppm	2.1	<1	1.5	<1	6.0	2.2	4.3	4.7
Ag	ppm	0.039	0.17	<0.1	<0.2	<0.3	<0.3	<0.4	<0.5
Sb	ppm	0.30	0.37	0.089	0.29	1.0	0.32	0.71	0.11
Cs	ppm	1.2	0.60	0.77	<0.8	<2	<4	<0.2	<0.1
Ba	ppm	611	37	40	1007	1000	232	335	54
La	ppm	25	8.5	5.1	16.4	25	8.5	33.7	1.3
Ce	ppm	47	7.6	8.6	22.8	48	16.3	60.8	4.1
Sm	ppm	5.7	1.3	0.77	2.2	3.1	1.5	5.2	0.27
Eu	ppm	2.0	0.30	0.16	0.66	0.79	0.40	1.14	0.17
Tb	ppm	0.93	0.27	0.13	0.33	0.61	0.30	0.86	0.15
Dy	ppm	6.3	1.3	0.83	1.6				<0.4
Ho	ppm	0.9							
Yb	ppm	3.1	0.78	0.50	1.05	2.1	0.89	2.90	0.32
Lu	ppm	0.58	2.2	0.91	0.19	0.40	0.20	0.59	0.10
Hf	ppm	4.2	1.9	0.85	2.5	7.1	2.1	8.6	6.0
Ta	ppm	0.89	0.063	0.10	0.28	0.62	0.20	1.08	1.22
W	ppm	1	0.11	0.16	0.48	2.5	1.4	1.8	<0.7
Pb	ppm	9	<2	3.5	12.5	15	9.2	12	165
Th	ppm	4.5	0.88	0.98	3.6	7.7	1.9	8.4	0.12
U	ppm	1.2	1.5	0.90	1.9	1.7	1.7	3.0	<0.4

TABLE 4.20. Chemical Analysis of Fly Ash Used in Bioshield Concrete

Element		Bellefonte	Hartsville	Enrico Fermi	Palo Verde
Na	%	0.25	0.35	0.28	1.44
Al	%	15.1	10.1	13.8	3.3
P	%	<0.8	<0.8	<0.8	<0.8
S	%	<0.25	1.0	0.45	0.41
K	%	2.78	1.78	1.63	1.34
Ca	%	0.87	2.10	2.98	1.14
Sc	ppm	37	23	26	10.5
Ti	ppm	7800	6000	7040	2600
V	ppm	320	630	216	42
Cr	ppm	170	153	137	26
Mn	ppm	160	218	146	201
Fe	%	4.00	12.4	5.40	3.94
Co	ppm	61	30	43	11
Ni	ppm	165	111	97	22
Cu	ppm	190	82	90	50
Zn	ppm	180	283	111	93
Ga	ppm	76	19	36	22
As	ppm	170	43	40	49
Se	ppm	9.6	7.0	12.1	1.7
Br	ppm	5.1	3.5	5.3	6.7
Rb	ppm	157	130	111	78
Sr	ppm	720	344	821	240
Y	ppm	93	51	63	14
Zr	ppm	240	229	193	160
Nb	ppm	30	25	25	5.1
Mo	ppm	34	104	19	21
Ag	ppm	1.2	<1	<2	0.27
Sb	ppm	9.1	15.2	3.0	1.7
Cs	ppm	19	<8	12	29
Ba	ppm	1180	740	820	820
La	ppm	103	48	70	26
Ce	ppm	191	104	133	44
Sm	ppm	17.4	7.8	10.6	3.4
Eu	ppm	3.6	1.8	2.3	0.84
Tb	ppm	3.0	1.5	1.7	0.53
Yb	ppm	9.1	4.8	6.0	1.6
Lu	ppm	2.1	1.6	1.22	0.47
Hf	ppm	6.9	5.0	5.4	4.8
Ta	ppm	2.5	1.5	1.8	0.58
W	ppm	8.2	4.7	3.4	51
Pb	ppm	94	95	54	78
Th	ppm	28	15.5	19.1	14.4
U	ppm	14	29	9.0	7.2

TABLE 4.21. Concrete Ingredients (Palo Verde) Elemental Concentrations

Element		Concrete	Coarse Aggregate	Sand	Fly Ash	Cement
Na	ppm	18200	20700	20300	14400	1000
Al	%	6.1	7.1	6.8	3.3	2.2
S	%	0.27	<0.2	<0.2	<0.41	1.6
K	%	2.5	2.9	2.9	1.34	0.53
Ca	%	9.2	0.98	0.72	1.14	48.5
Sc	ppm	5.3	5.0	6.0	10.5	4.3
Ti	ppm	2100	2000	1900	1600	1810
V	ppm	34	30	40	42	43
Cr	ppm	47	50	13.2	26	105
Mn	ppm	290	384	225	201	347
Fe	%	1.9	1.86	1.85	3.94	2.12
Co	ppm	5.6	3.98	4.9	11	9.9
Ni	ppm	30	9.7	15	22	62
Cu	ppm	28	15.5	13	50	85
Zn	ppm	90	43	31	93	470
Ga	ppm	13	13	41	22	5.8
As	ppm	2.0	8.6	11.1	49	22
Br	ppm	2.0	0.95	<0.8	6.7	0.73
Rb	ppm	170	109	126	78	16
Sr	ppm	220	131	180	240	460
Y	ppm	96	39	26	14	11
Zn	ppm	160	198	210	160	76
Nb	ppm	9.2	12.5	7.4	5.1	7.1
Sb	ppm	1.4	1.0	1.0	21	1.29
Ba	ppm	885	825	1000	820	210
Sm	ppm	3.9	4.8	3.1	3.4	2.4
Eu	ppm	0.77	0.88	0.79	0.84	0.58
Ta	ppm	0.90	0.92	0.62	0.58	0.63
W	ppm	3.9	2.0	2.5	51	0.72
Pb	ppm	34	15.5	15	78	33
Th	ppm	12	13.8	7.7	14.4	3.2
U	ppm	2.7	3.4	1.7	7.2	1.5

TABLE 4.22. Concrete Ingredients (Enrico Fermi) Elemental Concentrations

Element		Concrete	Sand	Fly Ash	Cement
Na	ppm	5220	7300	2800	2400
Al	%	2.0	2.7	13.8	2.6
S	%	0.42	0.15	0.45	1.1
K	%	0.60	0.82	1.63	0.70
Ca	%	17.5	14.4	2.98	45.3
Sc	ppm	3.0	3.4	26	4.2
Ti	ppm	1030	1180	7040	1210
V	ppm	32	32	216	43
Cr	ppm	27	43	137	67
Mn	ppm	270	420	146	414
Fe	%	1.1	1.36	5.40	1.78
Co	ppm	3.8	4.3	43	5.5
Ni	ppm	14	28	97	36
Cu	ppm	10	16	90	18
Zn	ppm	33	33	111	37
Ga	ppm	5.5	6.3	36	7.1
As	ppm	4.9	4.4	40	6.8
Br	ppm	5.6	4.2	5.3	<0.4
Rb	ppm	22	27	111	22
Sr	ppm	310	170	821	205
Y	ppm	6.7	10	63	15
Zr	ppm	52	69	193	55
Nb	ppm	1.8	2.6	25	3.9
Sb	ppm	0.5	2.2	3.0	0.59
Ba	ppm	706	232	820	266
Sm	ppm	1.7	1.5	10.6	2.5
Eu	ppm	0.34	0.40	2.3	0.51
Ta	ppm	0.24	0.30	1.8	0.35
W	ppm	0.6	1.4	3.4	0.86
Pb	ppm	5.9	9.2	54	<3
Th	ppm	2.1	1.9	19.1	3.3
U	ppm	3.0	1.7	9.0	6.0

TABLE 4.23. Concrete Ingredients (Hartsville) Elemental Concentrations

Element		Concrete	Coarse Aggregate	Sand	Fly Ash	Cement
Na	ppm	510	450	450	3450	1400
Al	%	1.3	0.73	0.75	10.1	2.4
S	%	0.33	0.88	0.12	1.0	0.88
K	%	0.31	0.32	0.29	1.78	0.40
Ca	%	34.7	34.4	36.6	2.10	46.1
Sc	ppm	2.3	1.03	0.97	23	3.5
Ti	ppm	850	510	450	6000	850
V	ppm	37	9.9	8.8	630	41
Cr	ppm	19	7.8	8.4	153	28
Mn	ppm	214	253	237	218	191
Fe	%	1.1	0.36	0.35	12.4	2.0
Co	ppm	3.0	0.98	0.96	30	5.3
Ni	ppm	31	19	19	111	34
Cu	ppm	16	6.4	7.4	82	27
Zn	ppm	33	15.3	15.5	283	56
Ga	ppm	3.7	1.6	1.0	19	5.1
As	ppm	4.6	0.85	0.95	43	6.5
Br	ppm	1.4	0.89	0.86	3.5	0.42
Rb	ppm	13	10.6	9.4	130	12
Sr	ppm	490	426	422	344	789
Y	ppm	6.9	3.9	4.5	51	8.6
Zr	ppm	33	27	25	229	<3
Nb	ppm	2.0	<1	<1	25	3.1
Sb	ppm	0.8	0.068	0.089	15.2	0.48
Ba	ppm	93	49	40	740	159
Sm	ppm	1.4	0.79	0.77	7.8	1.7
Eu	ppm	0.27	0.16	0.16	1.8	0.38
Ta	ppm	0.20	0.11	0.10	1.5	0.26
W	ppm	0.5	0.18	0.16	4.7	0.47
Pb	ppm	5.4	2.8	3.5	95	<3
Th	ppm	1.9	1.0	0.98	15.5	2.5
U	ppm	2.8	0.82	0.90	29	1.9

TABLE 4.24. Weighted Least Squares Fit to Concentrations of Elements in Hartsville Bioshield Concrete

Element	Percent Contribution		
	Aggregate	Fly Ash	Cement
Na	44.4	18.61	37.0
Al	38.5	28.4	33.1
S	29.2	13.3	57.4
K	58.6	19.7	21.7
Ca	74.6	0.2	25.2
Sc	30.6	39.8	29.6
Ti	44.7	32.7	22.6
V	16.2	63.6	20.2
Cr	34.6	34.5	30.9
Mn	79.0	4.0	17.1
Fe	22.4	43.4	34.3
Co	23.9	40.8	35.3
Ni	55.6	17.8	26.7
Cu	38.7	23.5	37.8
Zn	33.7	33.7	32.6
Ga	29.3	30.5	40.1
As	18.8	46.7	34.5
Br	73.9	16.5	9.7
Rb	47.6	36.1	16.3
Sr	64.7	2.9	32.4
Y	46.9	29.1	24.0
Zr	66.6	33.4	0.0
Nb	31.2	42.8	26.0
Sb	8.5	79.3	12.3
Ba	32.5	32.9	34.6
Sm	46.6	25.8	27.6
Eu	44.4	27.4	28.3
Ta	39.7	32.6	27.7
W	29.4	47.4	23.2
Pb	36.8	54.7	8.5
Th	39.2	34.0	26.8
U	30.0	53.0	17.0

Aggregate = 73.4 ± 9.6%
Fly Ash = 4.0 ± 0.5%
Cement = 19.7 ± 4.5%

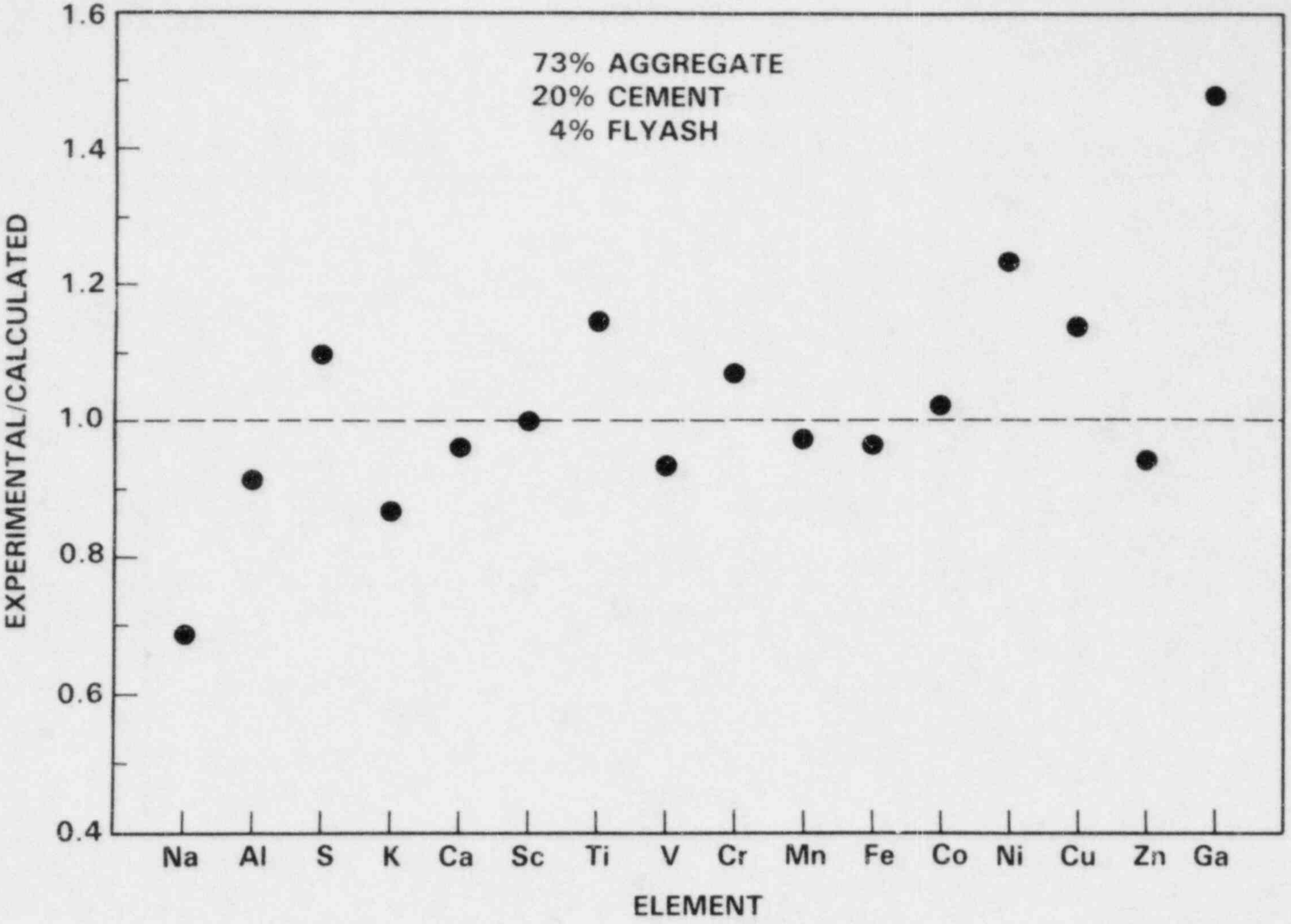

FIGURE 4.2a. Weighted Fit of Three Components to Concentration of 32 Elements for Hartsville Bioshield Concrete

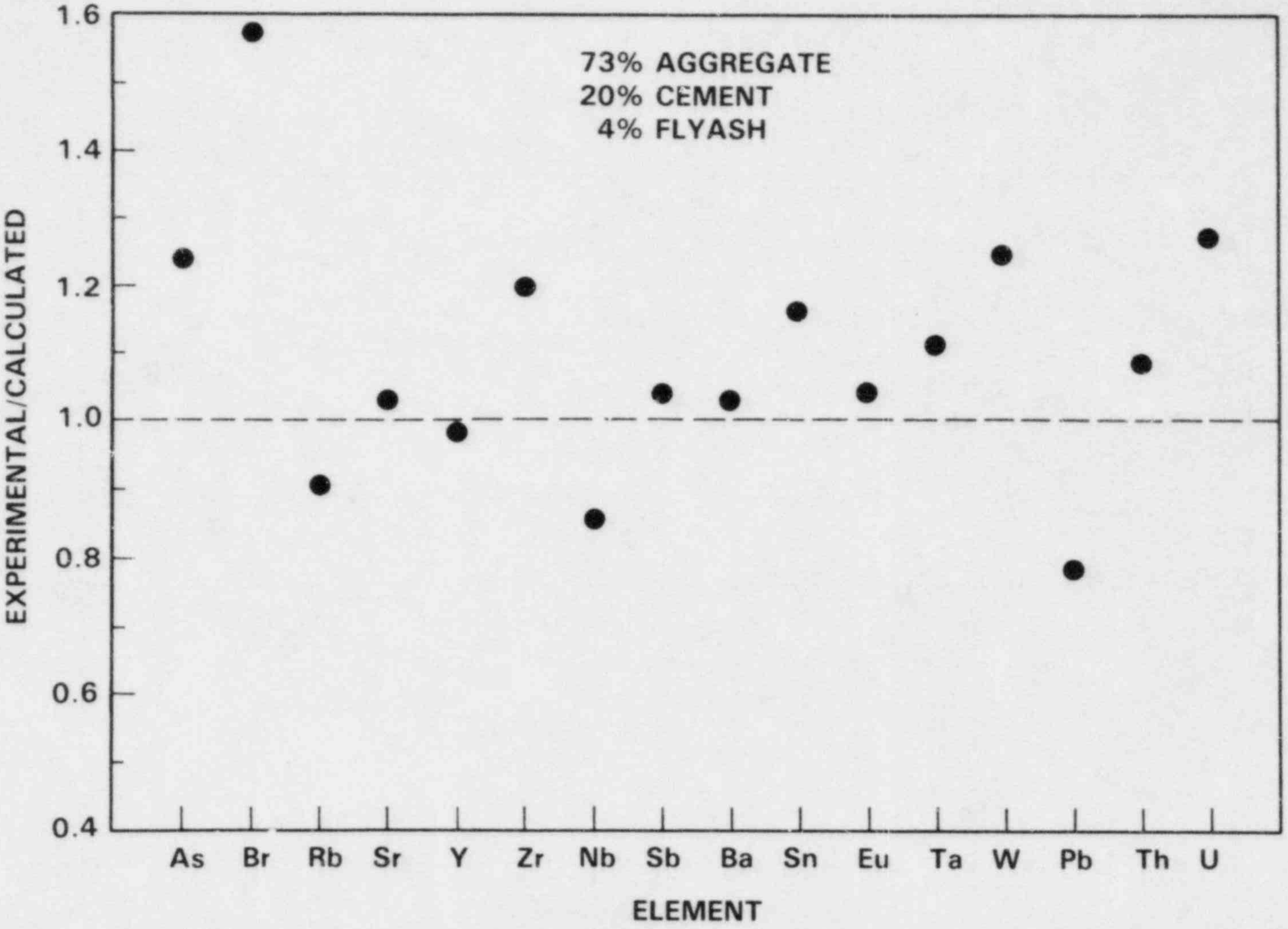

FIGURE 4.2a. (contd)

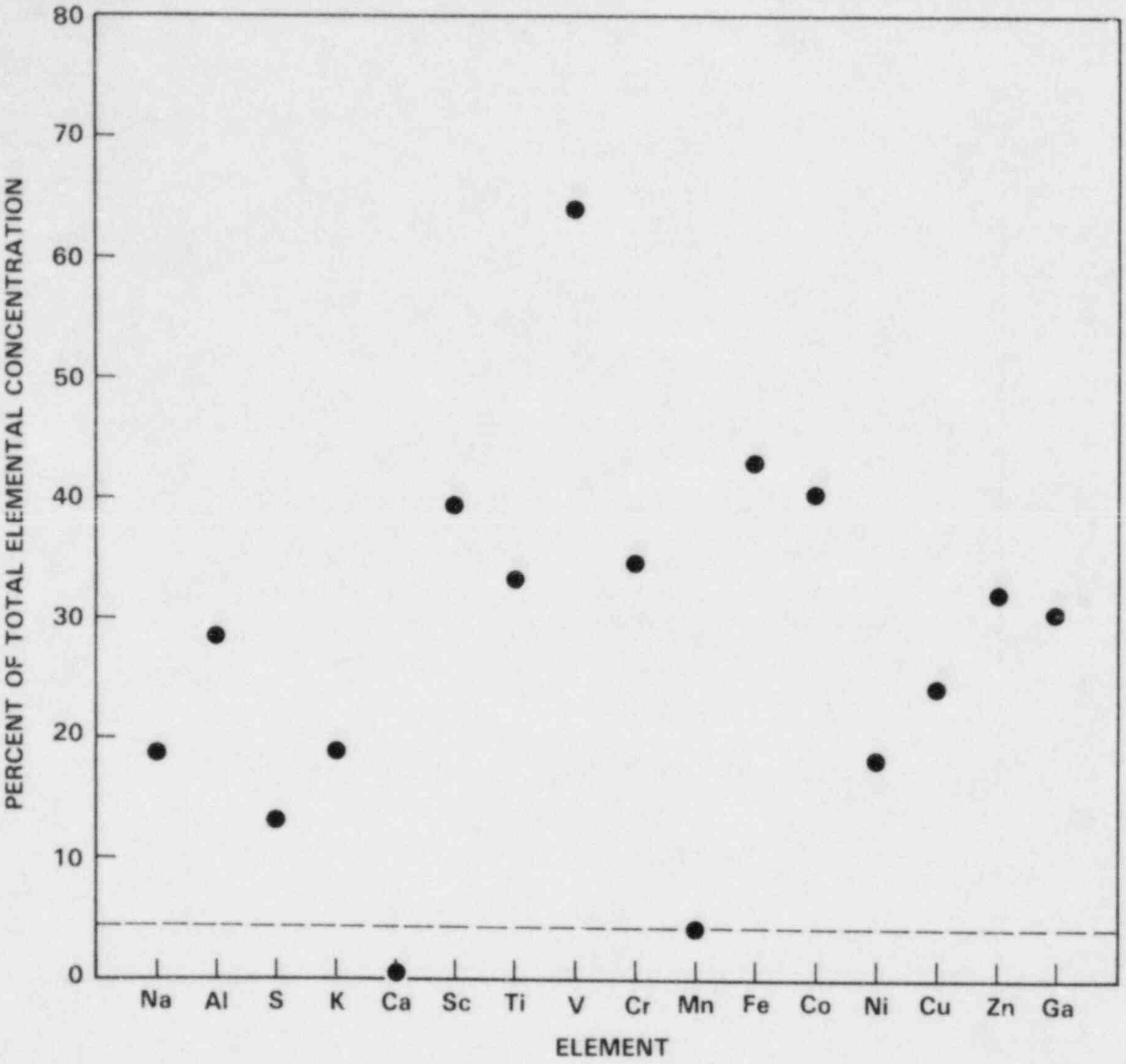

FIGURE 4.3a. Contribution of Fly Ash to Elemental Concentrations of Hartsville Bioshield Concrete

V, Sb, Pb, and U. This is a relatively minor change, falling well within the normal range of variability of those elements anyway. Addition of fly ash to cement does not therefore appear to cause any unusual problems with regard to activation products, provided the proportion added is kept small as was the case with the Hartsville samples.

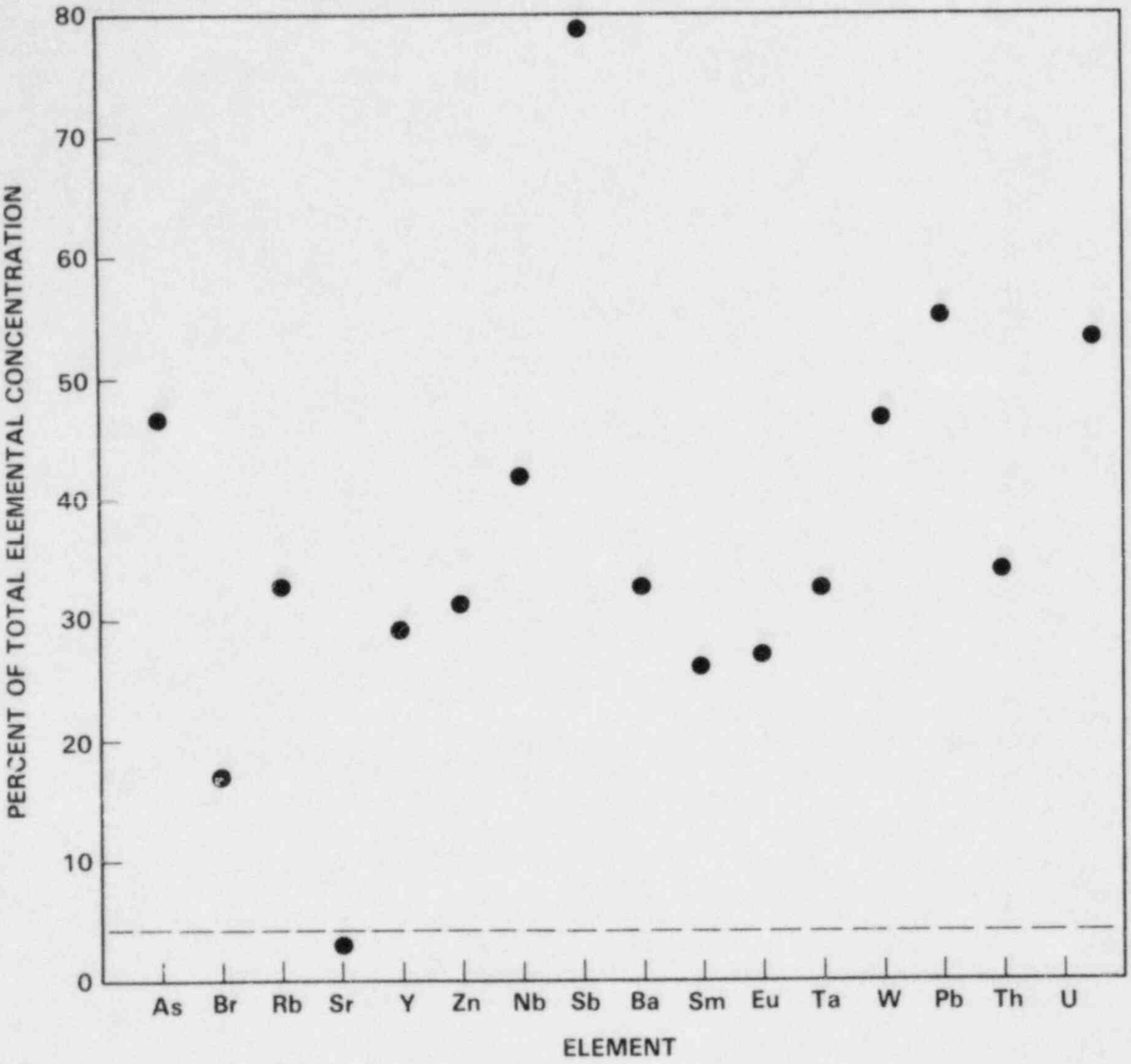

FIGURE 4.3a. (contd)

5.0 ACTIVATION CALCULATIONS

5.1 CALCULATION METHOD

In order to fully determine the expected levels of activation products in the various construction materials previously discussed, a suitable computer model was required. Early in the program it was determined that the available software such as the ORIGEN code used by Smith, Konzek, and Kennedy (1978) in their work was at the same time over-designed for the task and inflexible in its application to long lived activation products for which a limited data base was available. It was thus decided instead to develop a series of simpler codes specifically designed for this purpose. An additional goal was the inclusion of multi-group fast neutron reactions. The program was designed to be run on PDP-11 minicomputers in a fully interactive fashion. This approach proved very successful and was eventually adapted to a VAX/11-780 system in order to obtain adequate computing speed for the large number of cases considered. This family of programs consists of three main programs to generate flux, composition, and nuclear data libraries, a series of isotope selection files structured to match the isotope with the correct cross-section library, and a main program to carry out the activation calculation itself and store the results on a disk file. A full documentation and listings of the family of programs are included in the Appendix.

All nuclear parameters are tabulated in a series of libraries created by code NUCLIB. Five different kinds of libraries can be created. These include N-γ, N-2N, N-P, N-D, and N-α reactions in separate libraries thus allowing flexibility to evaluate the relative importance of those different reaction modes for the same product isotope if desired. Each block of library entries contains information on the type of reaction, the mass of the product isotope, the number of elements in the production chain, the isotopic abundances, half-lives, fission yields, production cross-sections and matching resonance integrals as well as the corresponding burnup cross-sections for each isotope having a significant contribution to the final product isotope. In constructing these production chains, it was necessary to make a subjective decision on the size of the chain based on available cross-section data (or lack of it). For most isotopes, only thermal neutron capture or resonance capture was important. These were contained in the N-γ library. These include ^{3}T, ^{14}C, ^{60}Co, ^{63}Ni, ^{81}Kr, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{108m}Ag, ^{110m}Ag, ^{145}Pm, ^{151}Sm, ^{152}Eu, ^{154}Eu, ^{155}Eu, ^{166m}Ho, ^{178}Hf, ^{233}U, ^{236}U, and ^{239}Pu. The other four libraries contained in addition to the N-γ information, an excitation function for the appropriated fast neutron reaction. These cross-sections were included in 0.5 MeV increments from 0 to 15 MeV. Isotopes tabulated in the N-2N library included ^{55}Fe, ^{59}Ni, ^{65}Zn, ^{79}Se, ^{92}Nb, ^{93}Zn, ^{93}Mo, ^{99}Tc, ^{121}Sn, ^{133}Ba, ^{146}Sm, ^{158}Tb, and ^{205}Pb. The N-P library contained ^{39}Ar, ^{54}Mn, ^{63}Ni, ^{85}Kr, ^{92}Nb,

^{93}Zr, ^{106}Pd, ^{121}Sn, ^{134}Cs, ^{135}Cs, ^{137}Cs, and ^{155}Eu. The only N-D reaction of importance was that producing ^{53}Mn. The N-α library contained only ^{36}Cl, ^{55}Fe, ^{85}Kr, and ^{93}Zr.

A second program, FLUX1 was used to create files containing the reactor conditions including temperature, thermal neutron flux, epithermal flux, and fast flux in thirty energy groups up to 15 MeV. This data was then permanently stored on a disk file. It was thus possible to calculate activation in various parts of the reactor by simply using different flux files for each set of calculations.

A third program, COMPOS, was used to assemble files of composition information on different materials. This program in turn references a master data file containing fifty element names together with their atomic number and atomic weights. Composition files were created for several average representative materials including type 304L stainless steel, carbon steel, concrete, and rebar.

Files created in the above described manner were then available for editing and updating as more information became available. They could be combined in the appropriate order to carry out activation calculations, via code ACTIV. Code ACTIV performed in the following manner. The composition file was first requested and assigned, the flux file was then requested, opened and read. The thermal and epithermal fluxes were read, the epithermal flux converted to flux per unit lethargy and a cadmium ratio computed. The temperature was read and the fast flux input into an array. The remaining part of the calculation was carried out automatically. The first groups of isotopes were selected in sequence, with the matching library file opened, rewound in each case, and the nuclear information input. For each case, the composition file was rewound, the appropriate chemical elements located and input. A second pass was required to obtain the correct data for Th and U in order to compute a fission contribution. Depending upon reaction type, several indices were adjusted to allow selection of the appropriate element and isotope combinations during the actual activation calculation. The activation calculation itself was done by repeated calls to subroutine NCAP. NCAP is basically an isotope production and depletion routine including fission, multiple neutron capture and fast neutron reactions. Thermal cross-sections are temperature-corrected from the 2200 m/s value. Production and depletion is done by a repetitive numerical integration of atoms produced at all points in the chain versus atoms lost to decay or burnup, in 3 day increments for 30 years with results printed for 1 year intervals. This iterative method is reasonably accurate provided there is not a major competition between beta decay and burnup of intermediate products. In that case, beta decay is selected first. This could result in a minor error due to the relatively coarse mesh size. This approach is also rather costly in terms of computation time while using very small amounts of memory, thus making is suitable for use on small low cost computers. Computation speed was quite satisfactory on the VAX system averaging only a few seconds per isotope. This also resulted in very minimal computer costs. In order to verify the accuracy of the method, a comparison was made with a code ORIGEN calculation for several cases with common data bases. Results agreed to within a few percent.

5.2 CROSS-SECTION SELECTION

In this work, a careful review was made of all available information in order to construct the most reliable cross-section data base. This was checked and updated several times before the final calculations were undertaken. Reliable cross-section information for all isotopes of interest, both stable and radioactive, has in the past been rather difficult to obtain. This situation has improved markedly in the last few years particularly for thermal neutron capture cross-sections. Resonance integral information is generally unavailable for isotopes with several isomeric states. The situation is particularly true for fast neutron reactions; however, this is a very minor problem since they contribute a negligible amount to the activation products inventory.

5.2.1 N-γ-Thermal

Several sources of thermal neutron cross-section information were available. These included the Seventh Edition of the Table of Isotopes, (Browne, Dairiki and Doebler 1978), the Twelfth Edition of the G.E. Chart of the Nuclides (Walker, Kirouac and Rourke 1977), and various publications from the Brookhaven National Neutron Cross-Section Center such as Mughabghab, Divadeenam and Holden (1981). In general, satisfactory production cross-section information was available for all important isotopes. Information on burnup cross-sections is relatively scarce and does represent a minor area of uncertainty since burnup can be important, particularly in the high flux region where the shroud is located.

5.2.2 N-γ-Epithermal

Epithermal cross-sections (resonance integrals) were taken from the same source as the thermal cross-sections. In some cases only the total resonance integral was available for several isomeric states. In that case it was arbitrarily assumed that the resonance integral distributed between isomers in the same manner as the thermal cross-sections. In cases where no resonance integral was listed, a 1/V contribution of 0.45 times the thermal cross-section was automatically included in the cross-section library as the default. The resonance integral can be an important contributor to the total activation even for the minimal 1/V case since there are regions of the reactor where the cadmium ratio is less than unity.

5.2.3 Fast Neutron Reactions

Fast neutron reaction data is in many cases not available for the appropriate long-lived isotopes. In order to avoid this difficulty, the computer code THRES2 (Pearlstein 1973, 1975) was obtained from the National Neutron Cross-Section Center to calculate fast neutron excitation functions. THRES2 is a semi-empirical code which uses well-measured excitation functions combined with known nuclear reaction systematics to compute unknown cross-sections for up to 19 different reaction types for product isotopes with atomic numbers in the region $Z = 21$ to $Z = 83$. This code has been demonstrated to be reasonably successful at predicting accurate excitation functions, provided the product nuclei are not too far from stability. A further limitation is the inability of the code to predict independent isomeric yields. This could in principle be done on the basis of spin selection factors, however, that was not necessary for any of the isotopes considered in this work. Only a few isotopes of importance lie outside the atomic number range covered by THRES2. These included ^{39}Ar, ^{36}Cl, ^{14}C, ^{10}Be, and ^{3}T. Appropriate experimental data was available in the open literature for everything except ^{10}Be. Code THRES2 produces cross-section data in 0.5 MeV intervals from 0 to 20 MeV, making it very suitable for use in the activation calculations in this work.

5.3 NEUTRON FLUX INFORMATION

Thermal and epithermal neutron flux information used in this work was taken from the ANISN calculations carried out by Smith, Konzek and Kennedy (1978) and Oak et al. (1980). That information was provided in its original form by the authors of those reports. The data is shown on Figure 5.1 for the PWR case. Neutron thermalization varies considerably with radial distance from the axial centerline of the fuel zone, resulting in a complex situation for nuclear activation. This is illustrated in Figure 5.2. The corresponding situation for a BWR is shown in Figures 5.3 and 5.4. All calculations done in this work were for the case of core axial midplane. An approximate total inventory can be obtained by integrating this data over the axial neutron distribution as given by the ANISN calculation. In order to obtain a fast neutron spectrum the multigroup code DOT results of Gritzner et al. (1977) were used and renormalized such that the integral flux above 1 MeV agreed with that given by ANISN calculations shown in Figures 5.1 and 5.3.

5.4 RESULTS OF ACTIVATION CALCULATIONS

An extensive series of activation calculations were carried out in the manner described above, using the average compositions of the appropriate materials, as reported in Section 4.2. In this discussion, all results are reported for core axial midplane, using volume averaged one-dimensional neutron

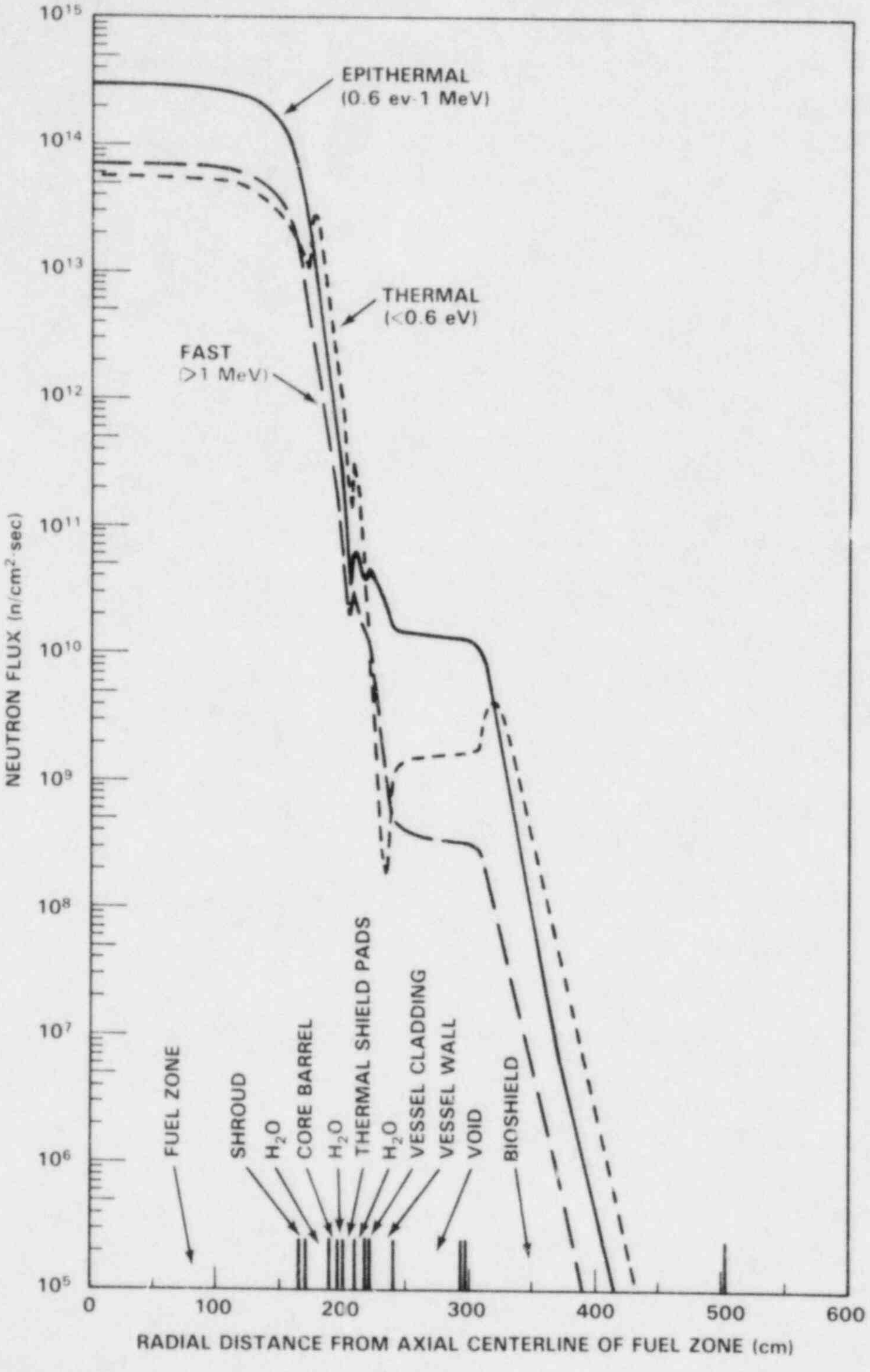

FIGURE 5.1. Radial Three Group Neutron Flux Distribution at Core Axial Midplane, ANISN Calculation for Westinghouse PWR

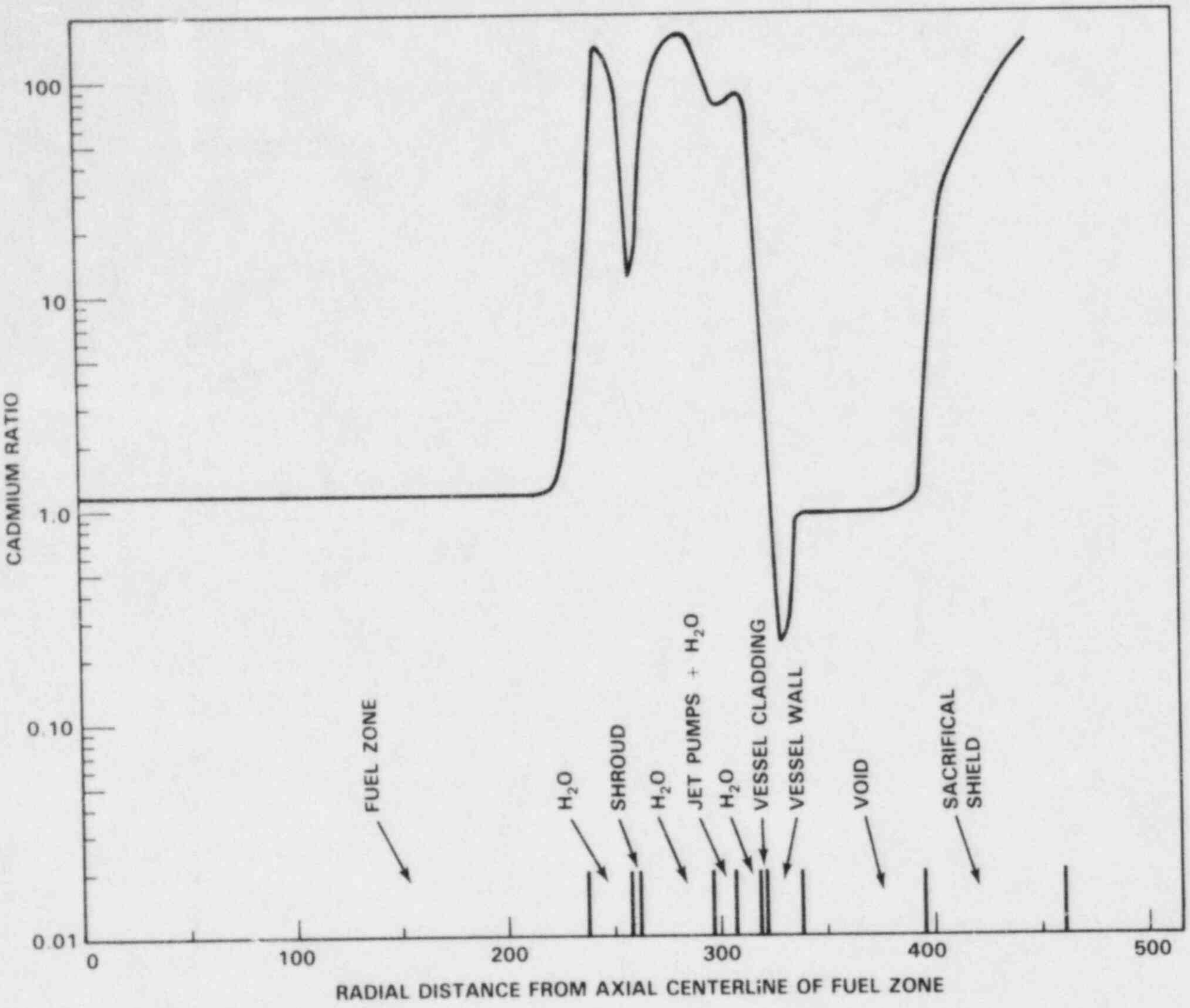

FIGURE 5.2. Cadmium Ratio Radial Distribution at Core Axial Midplane, ANISN Calculation for Westinghouse PWR

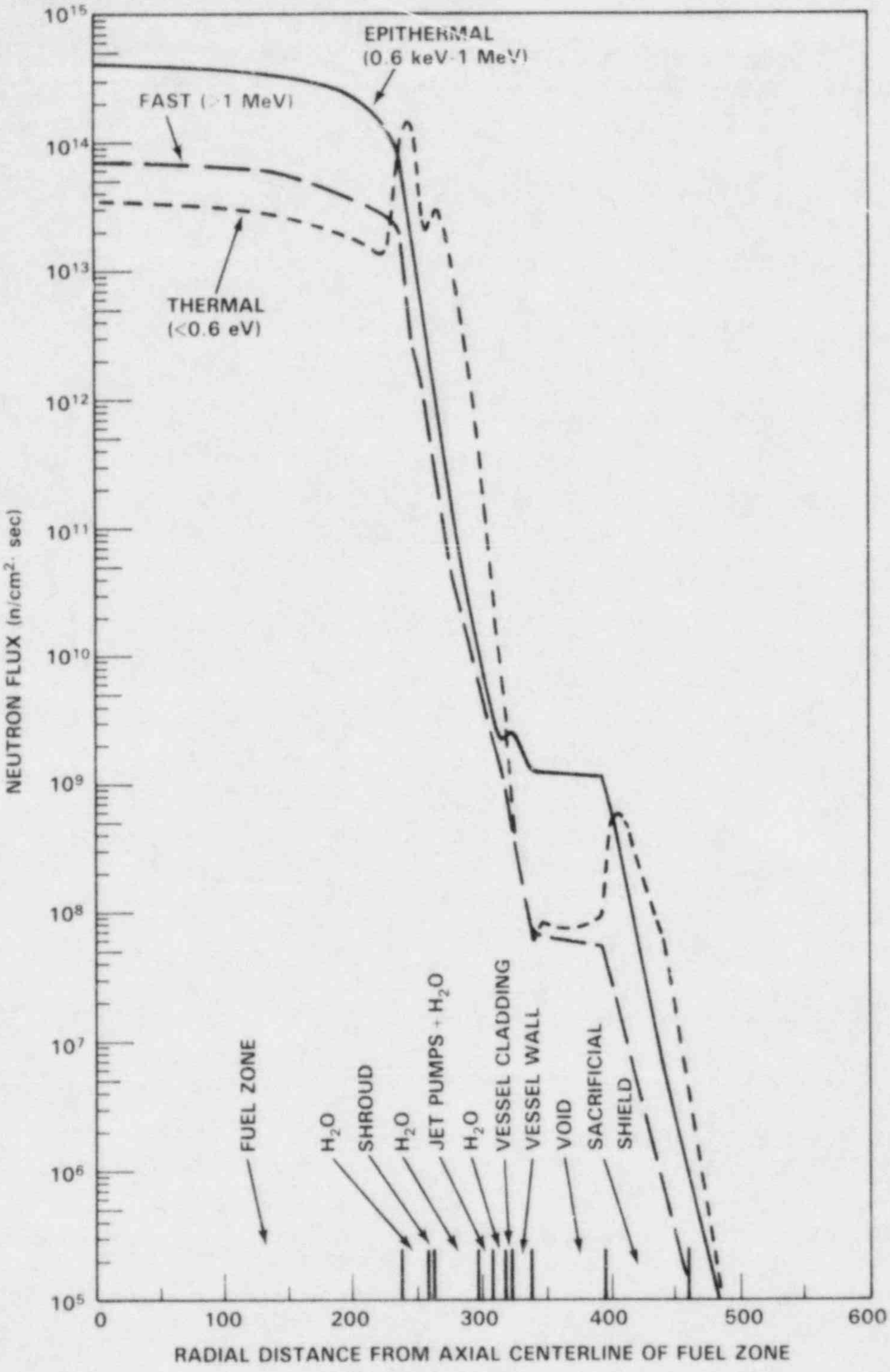

FIGURE 5.3. Radial Three Group Neutron Flux Distribution at Core Axial Midplane, ANISN Calculation GE BWR

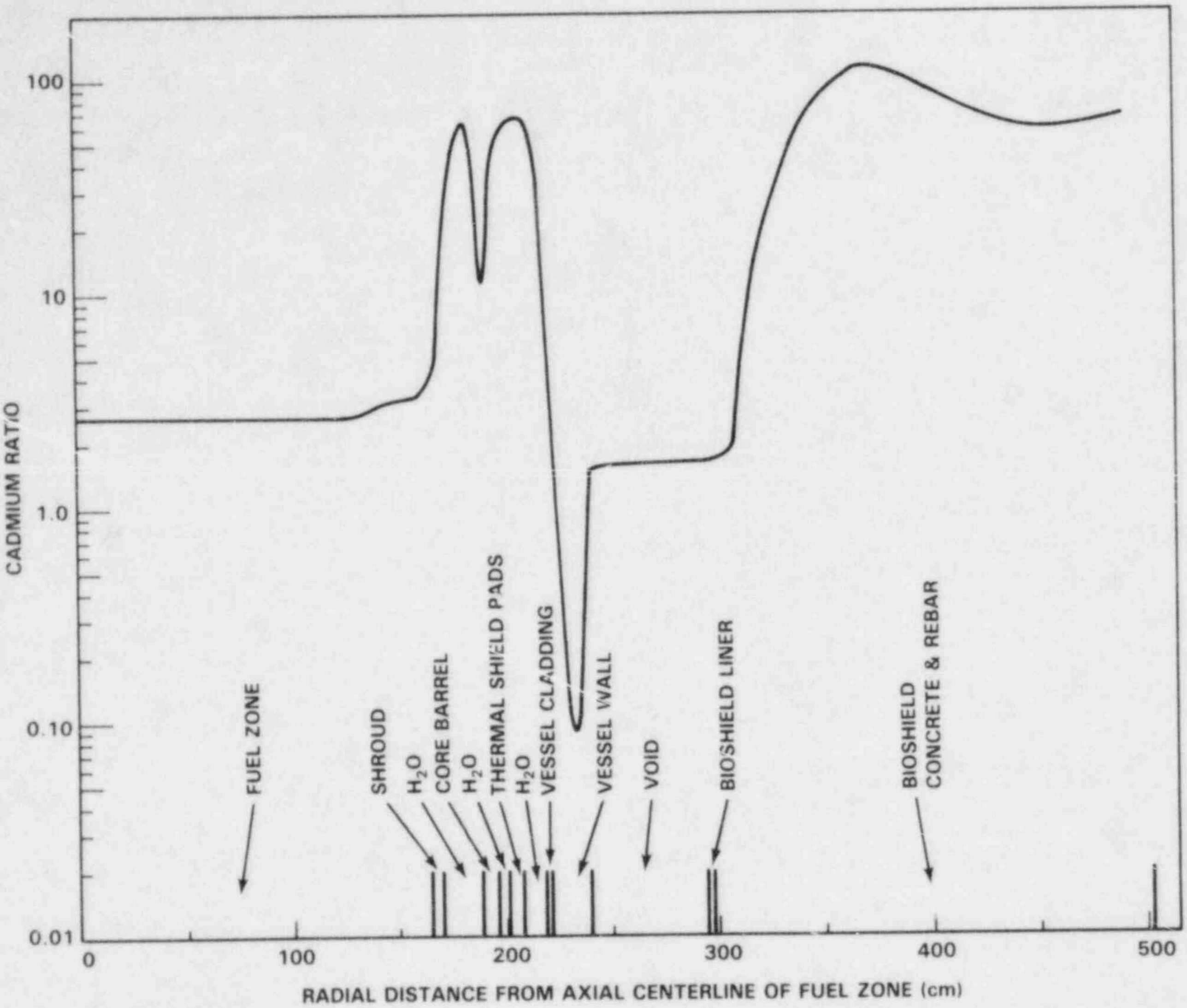

FIGURE 5.4. Cadmium Ratio Radial Distribution at Core Axial Midplane, ANISN Calculation for GE BWR

transport data for the components of interest. Results are reported for the most massive reactor components, both internal and external, exclusive of the fuel itself. Two reference geometries were used, a Westinghouse PWR and a GE BWR.

5.4.1 Internals

Table 5.1 lists the calculated results for the 40 most significant activation products produced in a PWR shroud, core barrel, thermal pads and vessel cladding. This represents the specific activity of each isotope at the time of shutdown after a 40 year operating history at 75% duty factor. Most of the activity is located in the shroud, with a drop off in specific activity of about three orders of magnitude out to the vessel cladding. The ^{60}Co level in the shroud was calculated to be 0.13 Ci/gm based on an average cobalt concentration of 1414 ppm. Smith, Konzek, and Kennedy (1978) calculated 0.12 Ci/gm using an upper bound for the Co content of 1500 ppm. In this work it was found that the Co content of stainless steel can be up to twice that high affecting the ^{60}Co inventory correspondingly. Figure 5.5 shows the specific activity of the most abundant isotopes as a function of time on the decommissioning and dismantling time scale. The situation is heavily dominated by only a few isotopes. Cobalt-60 dominates the total activity from five years to about 20 years; however, it will dominate the gamma dose for 80 to 100 years. After about 100 years, ^{94}Nb becomes the dominant gamma emitter with a small contribution from ^{59}Ni. The nickel isotopes tend to dominate the total activity after about only 20 years. Nickel-63 will be dominant for the first few hundred years, after which the steel contains mainly ^{59}Ni for several hundred thousand years. The latter case is illustrated in Figure 5.6 which can be considered a long-term disposal time scale. Figures 5.7, 5.8, and 5.9 illustrate basically the same situation for the core barrel, thermal pads, and vessel cladding, with decreasing levels of activity with increasing distance from the core. Minor differences in the patterns result from changes in neutron thermalization for different parts of the reactor. This results in a somewhat increased importance for ^{14}C. Another interesting difference on Figures 5.8 and 5.9 is the appearance of minor amounts of ^{152}Eu in the outer portions of the reactor. This is caused by the diminishing importance of burnup of ^{152}Eu as the neutron flux drops off. This still represents a trivial contribution, however, due to the very low abundance of europium in the stainless steel.

Table 5.2 gives the equivalent results for a BWR geometry. Due to the larger size of the core, specific activities are significantly lower in the BWR case, averaging about a factor of four lower in the shroud and a factor of thirteen lower for the vessel cladding. This is a bit deceptive, however, since the larger masses of the BWR components partially offsets the lower fluxes in the production of the total radionuclide inventory.

TABLE 5.1. Activation of PWR Internals (Ci/gm) Type 304L Stainless Steel 30 EFPY at Core Axial Midplane

Isotope	Half Life (y)	Shroud	Core Barrel	Thermal Pads	Vessel Cladding
^{3}T	12.3	1.0 E-5	9.6 E-6	2.2 E-6	3.3 E-7
^{14}C	5730	2.5 E-5	3.0 E-6	4.6 E-7	6.6 E-8
^{36}Cl	3.0 E+5	5.1 E-7	6.4 E-8	1.0 E-8	1.4 E-9
^{39}Ar	269	<1.5 E-7	<8 E-9	<4.2 E-10	6 E-10
^{41}Ca	1.03 E+6	4.7 E-9	5.6 E-10	8.6 E-11	1.2 E-11
^{53}Mn	3.7 E+6	3.2 E-9	2.1 E-10	1.0 E-11	6.9 E-12
^{54}Mn	0.85	6.5 E-3	3.6 E-4	1.8 E-5	1.2 E-5
^{55}Fe	2.7	2.1 E-1	2.4 E-2	3.7 E-3	5.4 E-4
^{59}Ni	8.0 E+4	1.1 E-4	1.9 E-5	3.0 E-6	4.3 E-7
^{60}Co	5.272	1.3 E-1	1.4 E-2	2.0 E-3	3.3 E-4
^{63}Ni	100	1.8 E-2	2.4 E-3	3.8 E-4	5.4 E-5
^{65}Zn	0.67	6.4 E-4	4.5 E-5	5.9 E-6	9.9 E-7
^{79}Se	6.5 E+4	6.1 E-10	4.6 E-11	4.7 E-12	1.2 E-12
^{81}Kr	2.1 E+5	7.6 E-10	3.4 E-12	3.5 E-14	2.1 E-15
^{85}Kr	10.5	<8.5 E-7	1.9 E-8	9.5 E-10	1.3 E-10
^{90}Sr	29	2 E-6	<5 E-9	<2.6 E-9	<3.5 E-10
^{92m}Nb	2.7 E+7	1.2 E-12	6.5 E-14	3.2 E-15	2.0 E-15
^{93}Zr	9.5 E+5	1.1 E-10	3.9 E-12	2.9 E-13	8.6 E-14
^{93}Mo	3500	9.4 E-7	3.9 E-8	2.1 E-9	1.1 E-9
^{94}Nb	2.0 E+4	4.0 E-7	2.9 E-8	3.0 E-9	7.5 E-10
^{99}Tc	2.1 E+5	1.3 E-7	8.2 E-9	4.2 E-10	2.4 E-10
^{108m}Ag	130	<1 E-7	<9 E-9	<1.1 E-9	<2.2 E-10
^{121m}Sn	50	4.8 E-9	3.4 E-10	1.7 E-11	1.1 E-11
^{129}I	1.6 E+7	<6 E-13	<1.4 E-14	<7.9 E-16	<1.1 E-16
^{133}Ba	10.4	<3 E-6	<3 E-7	<4.5 E-8	<7.1 E-9
^{134}Cs	2.06	<7 E-6	<1 E-6	<1.7 E-7	<2.3 E-8
^{135}Cs	2.3 E+6	<4 E-11	<9 E-13	<5.1 E-14	<7.8 E-15
^{137}Cs	30.1	<2 E-6	<5 E-8	<2.7 E-9	<4.2 E-10
^{145}Pm	17.7	8.9 E-10	1.4 E-10	2.2 E-11	3.1 E-12
^{146}Sm	1.0 E+8	1.0 E-16	1.3 E-17	6.7 E-19	4.3 E-19
^{151}Sm	93	4.6 E-9	4.5 E-9	2.9 E-9	5.9 E-10
^{152}Eu	13	0	9.1 E-10	1.7 E-7	7.6 E-8
^{154}Eu	8.6	5.6 E-7	6.0 E-7	6.2 E-8	1.2 E-8
^{155}Eu	4.76	4.1 E-7	1.4 E-7	5.0 E-9	5.2 E-10
^{158}Tb	150	1.9 E-9	1.7 E-10	8.5 E-12	5.6 E-12
^{166m}Ho	00	1.6 E-7	1.3 E-8	1.1 E-9	3.0 E-10
^{178m}Hf	30	4.3 E-8	2.8 E-8	3.1 E-9	1.2 E-9
^{205}Pb	1.4 E+7	1.8 E-12	1.3 E-13	1.3 E-14	3.4 E-15
^{233}U	1.6 E+5	<3.6 E-10	<1.0 E-10	1.1 E-11	3.4 E-12
^{239}Pu	2.4 E+4	<7.0 E-9	<2.3 E-9	1.4 E-10	9.6 E-11

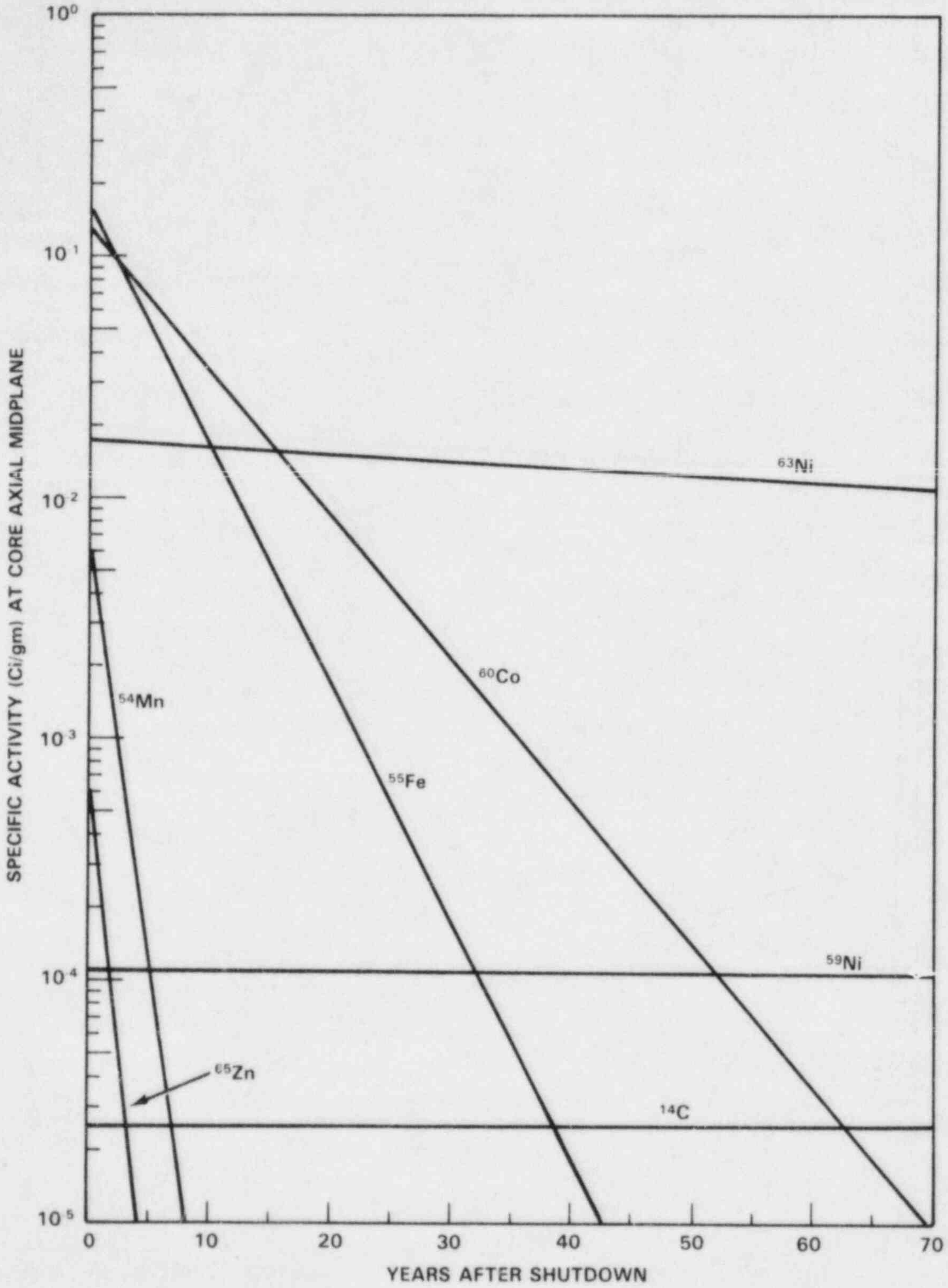

FIGURE 5.5. Activation of PWR Shroud Type 304 Stainless Steel 30 EFPY

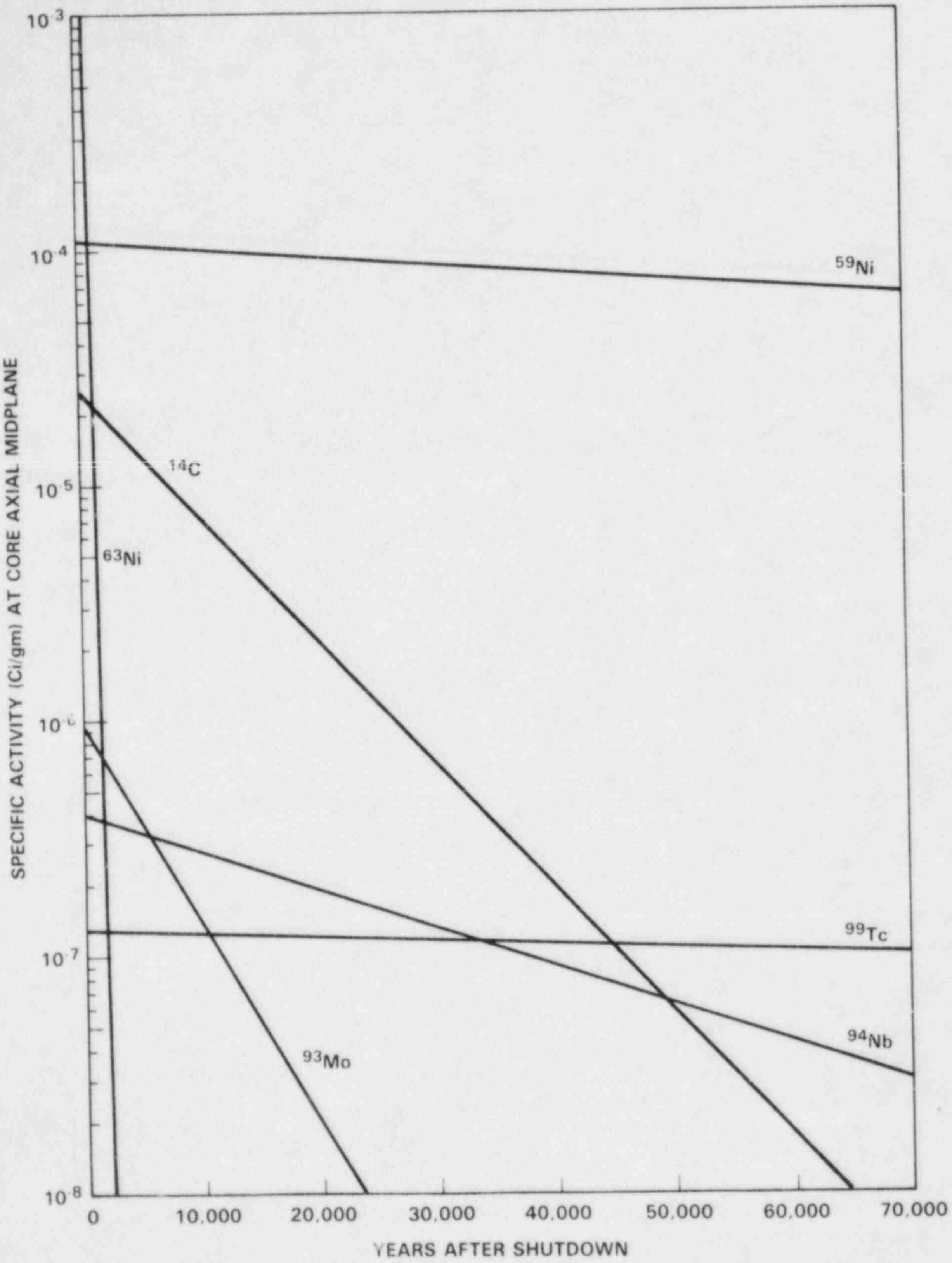

FIGURE 5.6. PWR Shroud Type 304 Stainless Steel 30 EFPY

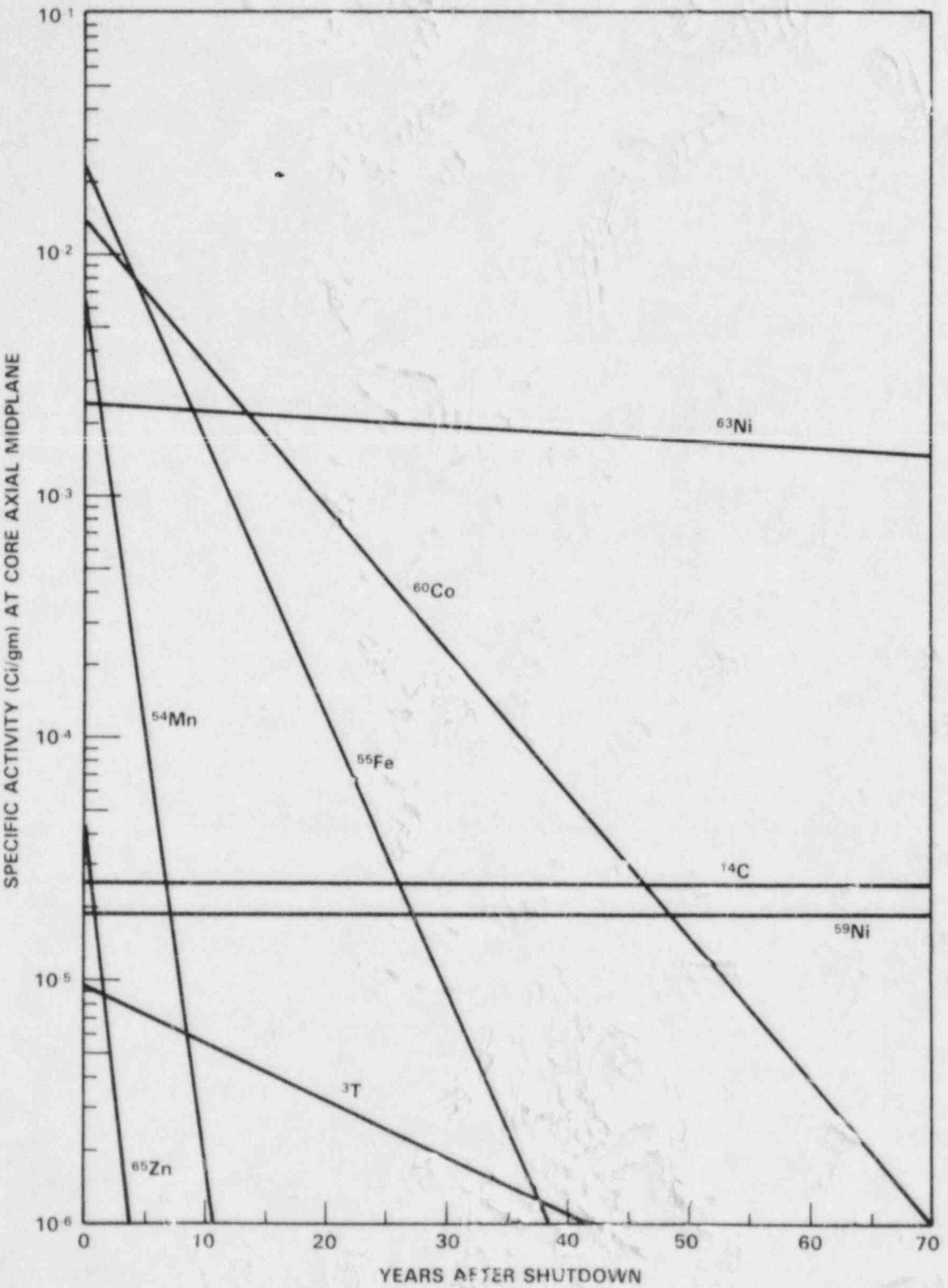

FIGURE 5.7. Activation of PWR Core Barrel Type 304 Stainless Steel 30 EFPY

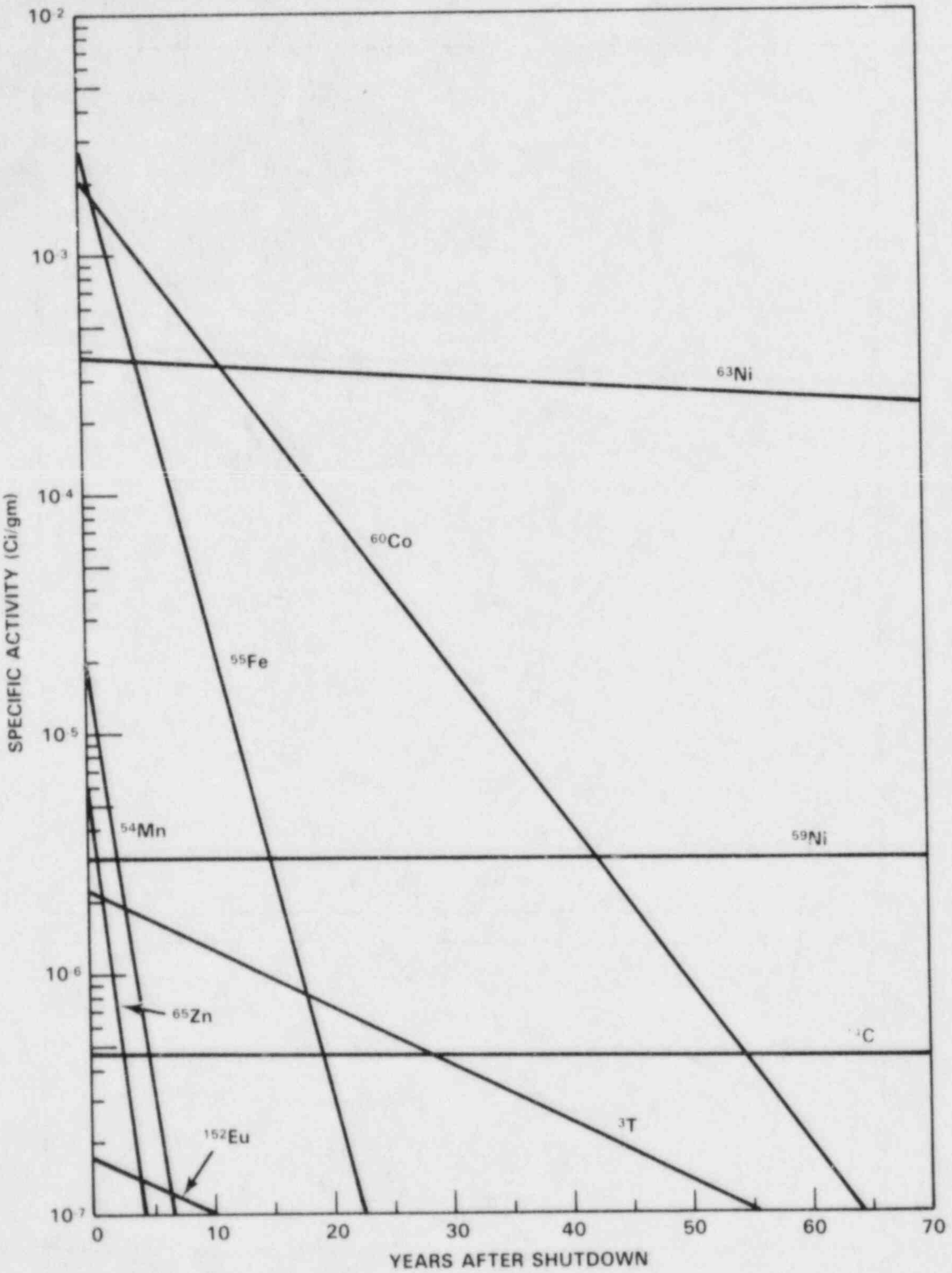

FIGURE 5.8. Activation of PWR Thermal Pads Type 304 Stainless Steel 30 EFPY

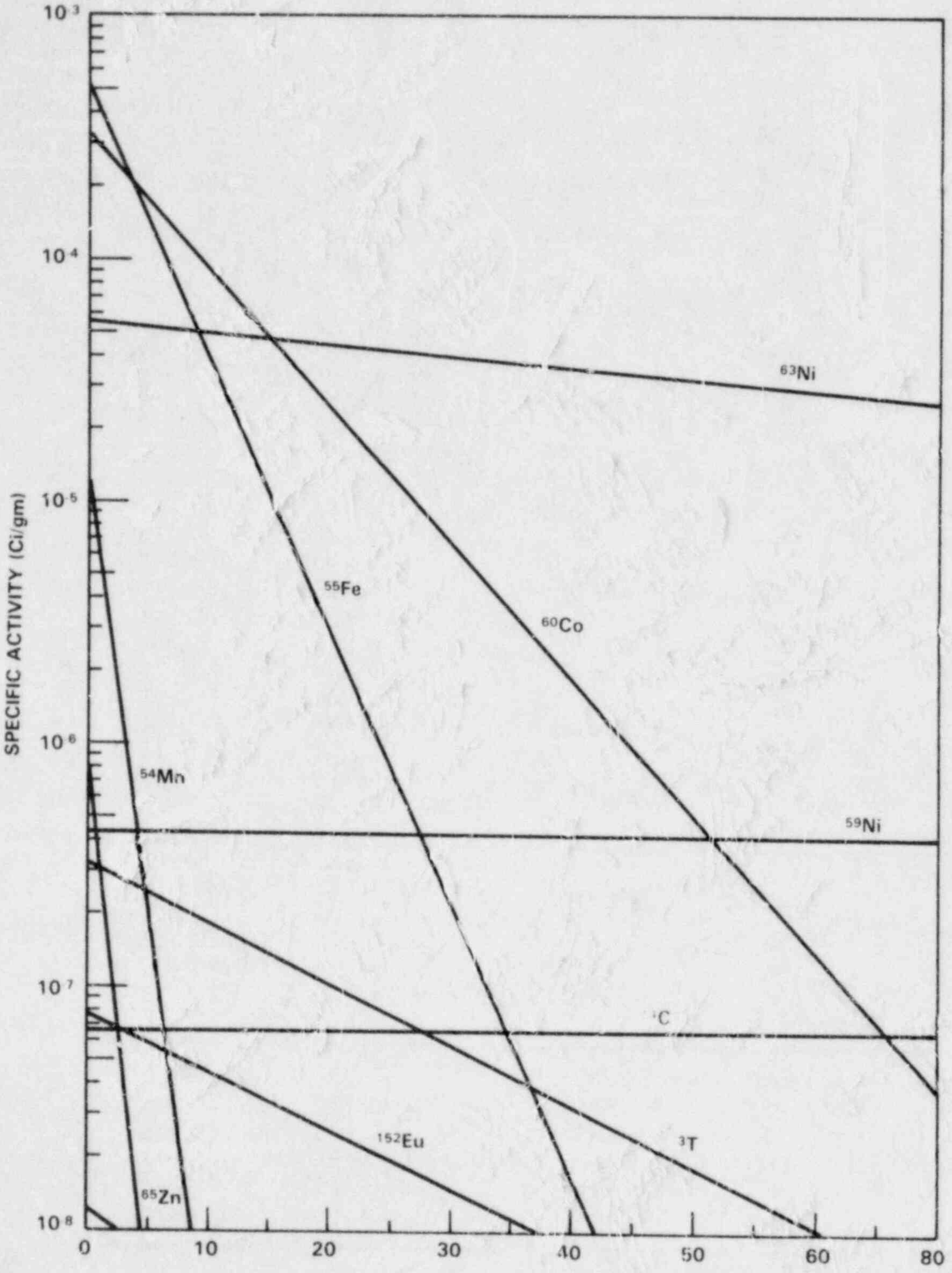

FIGURE 5.9. Activation of PWR Vessel Cladding Type 304 Stainless Steel 30 EFPY

TABLE 5.2. Activation of BWR Internals (Ci/gm) Type 304L Stainless Steel 30 EFPY at Core Axial Midplane

Isotope	Half-Life (Y)	Shroud	Vessel Cladding
^{3}T	1.20E+01	1.30E-05	2.50E-08
^{14}C	5.70E+03	7.30E-06	4.90E-09
^{36}Cl	3.00E+05	1.60E-07	1.00E-10
^{39}Ar	2.70E+02	1.90E-08	5.30E-11
^{41}Ca	1.00E+05	1.40E-09	9.10E-13
^{53}Mn	3.70E+06	4.60E-10	1.40E-12
^{54}Mn	8.50E-01	8.30E-04	2.00E-06
^{55}Fe	2.70E+00	5.90E-02	4.10E-05
^{59}Ni	8.00E+04	4.30E-05	3.20E-08
^{60}Co	5.30E+00	3.20E-02	2.40E-05
^{63}Ni	1.00E+02	5.70E-03	4.00E-06
^{65}Zn	6.70E-01	1.10E-04	7.10E-08
^{79}Se	6.50E+04	1.00E-10	1.70E-13
^{81}Kr	2.10E+05	<1.60E-11	<9.40E-18
^{85}Kr	1.10E+01	<5.80E-08	<9.00E-12
^{90}Sr	2.90E+01	<1.50E-07	<2.50E-11
^{92m}Nb	2.70E+07	4.50E-14	3.80E-16
^{93}Zr	9.50E+05	1.00E-11	1.20E-14
^{93}Mo	3.50E+00	7.70E-08	7.70E-11
^{94}Nb	2.00E+04	6.31E-08	4.90E-11
^{99}Tc	2.10E+05	1.50E-08	1.50E-11
^{108m}Ag	1.30E+02	<2.00E-08	<1.50E-11
^{121m}Sn	5.00E+00	7.60E-10	2.00E-12
^{129}I	1.60E+07	<4.20E-14	<7.60E-18
^{133}Ba	1.00E+01	<7.40E-07	<6.00E-10
^{134}Cs	2.10E+00	<2.40E-06	<1.70E-09
^{135}Cs	2.30E+06	<2.70E-12	<6.40E-16
^{137}Cs	3.00E+01	<1.50E-07	<3.50E-11
^{145}Pm	1.80E+01	<3.10E-10	<2.30E-13
^{146}Sm	1.00E+08	2.90E-17	7.90E-20
^{151}Sm	9.30E+01	3.80E-09	3.10E-11
^{152}Eu	1.30E+01	2.20E-14	6.70E-09
^{154}Eu	8.60E+00	9.10E-07	8.00E-10
^{155}Eu	4.80E+oo	3.60E-07	2.90E-11
^{158}Tb	1.50E+02	3.80E-10	1.10E-12
^{166m}Ho	1.20E+03	2.80E-08	1.90E-11
^{178m}Hf	3.00E+01	<3.70E-08	<7.50E-11
^{205}Pb	1.40E-07	2.90E-13	4.50E-16
^{233}U	1.60E+05	<1.60E-10	<2.20E-13
^{239}Pu	2.40E+04	<2.70E-09	<5.80E-12

5.4.2 Pressure Vessel Wall

Table 5.3 lists calculated activities in pressure vessel walls for both PWR and BWR geometries. The BWR case results in specific activities about a factor of 15 lower as previously discussed. The activation pattern is similar to stainless steel with, however, a diminished importance for the Co and Ni isotopes. One interesting consequence of this is that after about 80 years decay, ^{152}Eu actually dominates the gamma activity. That would only be true for about 20 to 30 years at which point ^{94}Nb becomes the dominant gamma emitting isotope with no further significant decay occurring on a historical time scale. This situation is demonstrated on Figures 5.10 and 5.11. Figure 5.11 appears somewhat more complex than the equivalent case for stainless steel. This is somewhat of an artifact of the plotting method which is confined to four decades of relative importance. Due to the lower abundance of Ni in carbon steel, other isotopes such as ^{14}C and ^{36}Cl become more noteworthy on the long time scale, however, the absolute levels are very low. In the case of ^{14}C in a PWR vessel wall. For example, calculated levels fall more than a factor of 500 below Class A disposal limits.

5.4.3 Bioshield Concrete

Activation levels of PWR bioshield concrete at the time of shutdown are given in Table 5.4 for several depths, in the bioshield. The maximum activation occurs between 10 and 20 cm from the inner surface for most isotopes due to neutron thermalization by the concrete. Activation levels for the decommissioning and disposal time scales respectively are shown on Figures 5.12 and 5.13. These plots are conspicuously more complex than the corresponding plots for steels. This is due to the much more complex mixture of major, minor, and trace elements found in the geological materials from which concrete is fabricated. On the 100 year time scale the total activity in concrete is dominated by tritium. On the long-term disposal scale, the total activity is completely dominated by ^{41}Ca, comprising about 99% of the total. Gamma activity on the 100-year time scale is mainly due to ^{152}Eu with ^{60}Co being of relatively minor importance after the first 20 years. Europium-152 will in fact dominate the gamma activity for several hundred years after which time the material will be essentially nonradioactive with respect to gamma ray emission. The comparable data for a BWR are given in Table 5.5. Activation levels are reduced by about one order of magnitude due to the greater distance from the core center.

5.4.5 Rebar

Activation of rebar was considered independently in this study. Estimates of the relative proportion of rebar to concrete are difficult to obtain. An examination of the blueprints for the WNP No. 1 Reactor project, for example, showed an estimated weight percentage of rebar of about 10%, however, this is very uncertain and varies considerably with location. Table 5.6 lists the calculated results for PWR bioshield rebar. The time dependent levels are illustrated in Figures 5.14 and 5.15. The pattern is similar to that of the vessel wall but at reduced levels. The comparable situation for a BWR is given in Table 5.7. As was the case with the concrete, activation levels are reduced about a factor of 10 relative to a PWR.

TABLE 5.3. Activation of Pressure Vessel Walls (Ci/gm) 30 EFPY at Core Axial Midplane

Isotope	Half-Life (Y)	PWR	BWR
^{3}T	1.20E+01	1.30E-05	2.50E-08
^{14}C	5.70E+03	7.30E-06	4.90E-09
^{36}Cl	3.00E+05	1.60E-07	1.00E-10
^{39}Ar	2.70E+02	1.90E-08	5.30E-11
^{41}Ca	1.00E+05	1.40E-09	9.10E-13
^{53}Mn	3.70E+06	4.60E-10	1.40E-12
^{54}Mn	8.50E-01	8.30E-04	2.00E-06
^{55}Fe	2.70E+00	5.90E-02	4.10E-05
^{59}Ni	8.00E+04	4.30E-05	3.20E-08
^{60}Co	5.30E+00	3.20E-02	2.40E-05
^{63}Ni	1.00E+02	5.70E-03	4.00E-06
^{65}Zn	6.70E-01	1.10E-04	7.10E-08
^{79}Se	6.50E+04	1.00E-10	1.70E-13
^{81}Kr	2.10E+05	<1.60E-11	<9.40E-18
^{85}Kr	1.10E+01	<5.80E-08	<9.00E-12
^{90}Sr	2.90E+01	<1.50E-07	<2.50E-11
^{92m}Nb	2.70E+07	4.50E-14	3.80E-16
^{93}Zr	9.50E+05	1.00E-11	1.20E-14
^{93}Mo	3.50E+00	7.70E-08	7.70E-11
^{94}Nb	2.00E+04	6.31E-08	4.90E-11
^{99g}Tc	2.10E+05	1.50E-08	1.50E-11
^{108m}Ag	1.30E+02	<2.00E-08	<1.50E-11
^{121m}Sn	5.00E+00	<7.60E-18	2.00E-12
^{129}I	1.60E+07	<4.20E-14	<7.60E-18
^{133}Ba	1.00E+01	<7.40E-07	<6.00E-10
^{134}Cs	2.10E+00	<2.40E-06	<1.70E-09
^{135}Cs	2.30E+06	<2.70E-12	<6.40E-16
^{137}Cs	3.00E+01	<1.50E-07	<3.50E-11
^{145}Pm	1.80E+01	<3.10E-10	<2.30E-13
^{146}Sm	1.00E+08	2.90E-17	7.90E-20
^{151}Sm	9.30E+01	3.80E-09	3.10E-11
^{152}Eu	1.30E+01	2.20E-14	6.70E-09
^{154}Eu	8.60E+00	9.10E-07	8.00E-10
^{155}Eu	4.80E+oo	3.60E-07	2.90E-11
^{158}Tb	1.50E+02	3.80E-10	1.10E-12
^{166m}Ho	1.20E+03	2.80E-08	1.90E-11
^{178m}Hf	3.00E+01	<3.70E-08	<7.50E-11
^{205}Pb	1.40E-07	2.90E-13	4.50E-16
^{233}U	1.60E+05	<1.60E-10	<2.20E-13
^{239}Pu	2.40E+04	<2.70E-09	<5.80E-12

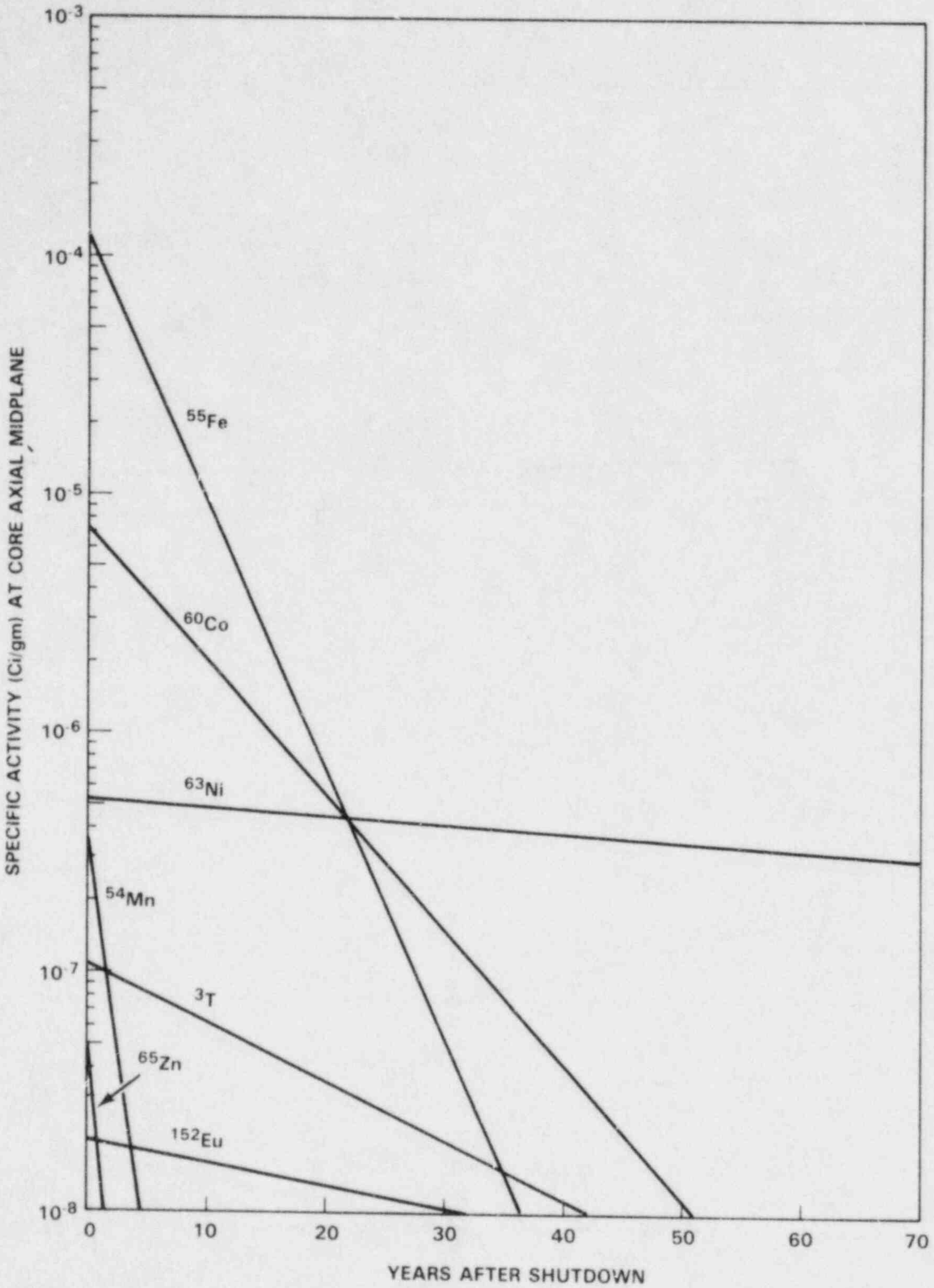

FIGURE 5.10. PWR Vessel Wall 30 EFPY - Decommissioning Time Scale

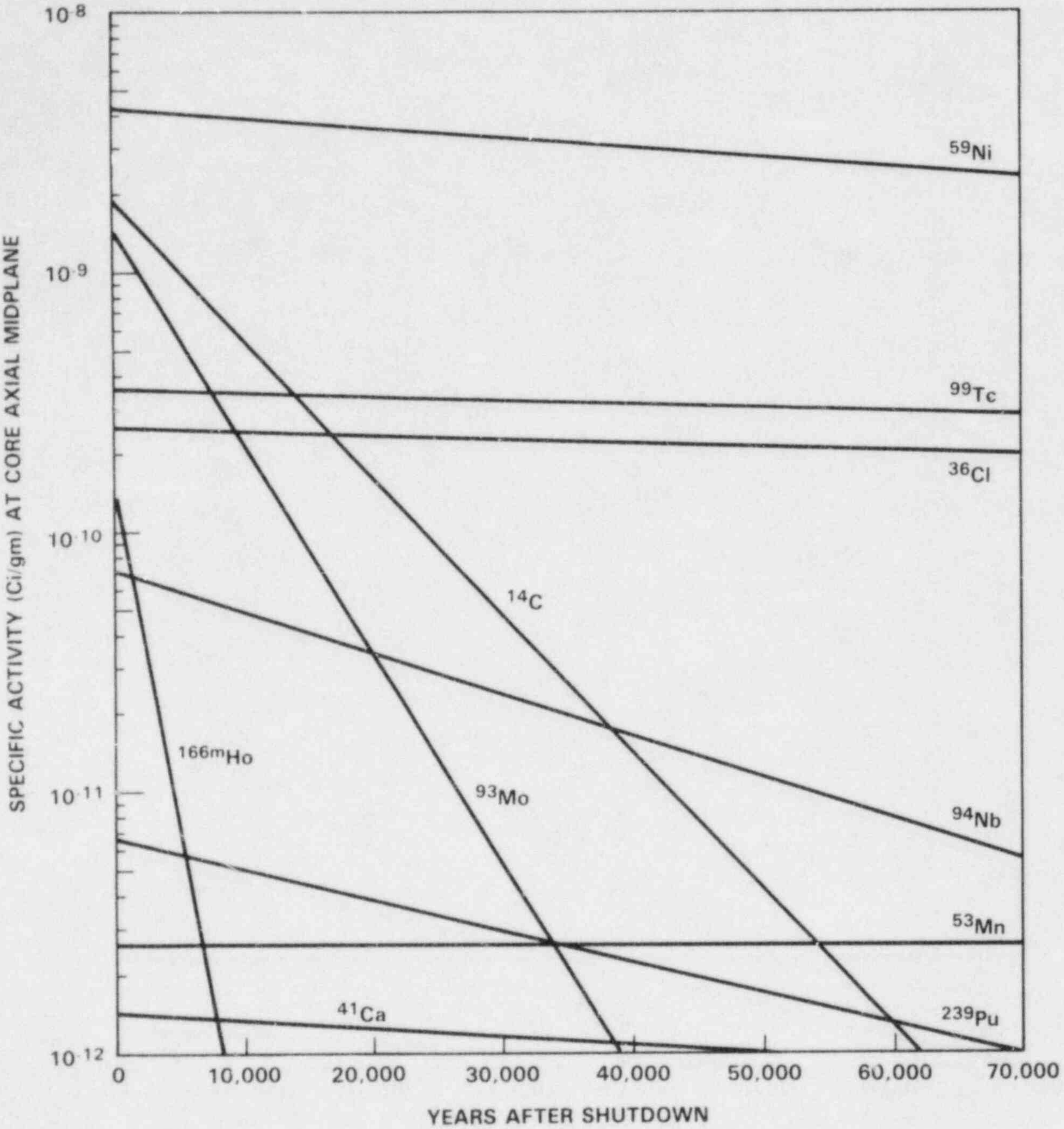

FIGURE 5.11. PWR Vessel Wall 30 EFPY - Long Term Disposal Time Scale

TABLE 5.4. Activation of PWR Bioshield (Ci/gm) Average Concrete 30 EFPY at Core Axial Midplane

Isotope	Half-Life (Y)	Inner Edge	10 cm	24 cm	55 cm
^{3}T	1.20E+01	5.00E-06	8.30E-06	4.40E-06	2.50E-07
^{14}C	5.70E+03	1.70E-09	2.80E-09	1.50E-09	8.30E-11
^{36}Cl	3.00E+05	9.80E-11	1.50E-10	7.90E-11	4.40E-12
^{39}Ar	2.70E+02	2.70E-08	1.20E-08	3.10E-09	1.60E-10
^{41}Ca	1.00E+05	1.20E-08	1.90E-08	1.00E-08	5.60E-10
^{53}Mn	3.70E+06	1.70E-14	7.40E-15	1.90E-15	1.00E-16
^{54}Mn	8.50E-01	2.80E-08	1.20E-08	3.10E-09	1.70E-10
^{55}Fe	2.70E+06	2.90E-06	4.70E-06	2.50E-06	1.40E-07
^{59}Ni	8.00E+04	1.60E-11	2.60E-11	1.40E-11	7.90E-13
^{60}Co	5.30E+00	2.20E-07	3.60E-07	1.70E-07	9.20E-09
^{63}Ni	1.00E+02	2.00E-09	3.30E-09	1.70E-09	9.80E-11
^{65}Zn	6.70E-01	2.20E-08	2.50E-09	1.20E-08	6.40E-10
^{79}Se	6.50E+04	2.30E-15	2.80E-15	1.40E-15	7.90E-17
^{81}Kr	2.10E+05	1.10E-16	5.50E-17	7.60E-18	1.70E-20
^{85}Kr	1.10E+01	4.30E-11	4.20E-11	1.70E-11	9.40E-13
^{90}Sr	2.90E+01	4.50E-11	7.30E-11	3.80E-11	2.10E-12
^{92m}Nb	2.70E+07	1.40E-18	1.20E-19	1.90E-19	1.00E-20
^{93}Zr	9.50E+05	3.00E-14	2.90E-14	1.30E-14	7.00E-16
^{93}Mo	3.50E+00	1.20E-12	5.60E-13	1.40E-13	4.40E-15
^{94}Nb	2.00E+04	7.50E-12	5.30E-12	2.00E-12	9.40E-14
^{99}Tc	2.10E+05	2.80E-13	1.40E-13	3.70E-14	1.30E-15
^{108m}Ag	1.30E+02	<3.60E-12	<3.20E-12	<1.40E-12	<7.30E-14
^{121m}Sn	5.00E+00	<6.50E-13	<5.20E-13	<1.80E-13	<1.20E-14
^{129}I	1.60E+07	1.40E-17	2.20E-17	1.20E-17	6.60E-19
^{133}Ba	1.00E+01	1.30E-09	2.00E-09	1.00E-09	5.70E-11
^{134}Cs	2.10E+00	8.30E-09	1.60E-08	8.70E-09	4.90E-10
^{135}Cs	2.30E+06	9.50E-16	1.40E-15	7.50E-16	4.20E-17
^{137}Cs	3.00E+01	5.10E-11	7.60E-11	4.00E-11	2.20E-12
^{145}Pm	1.80E+01	5.90E-12	9.80E-12	5.20E-12	2.90E-13
^{146}Sm	1.00E+08	3.90E-19	1.70E-19	4.40E-20	2.30E-21
^{151}Sm	9.30E+01	1.40E-09	1.40E-09	6.10E-10	3.10E-11
^{152}Eu	1.30E+01	2.40E-07	3.90E-07	2.10E-07	1.20E-08
^{154}Eu	8.60E+00	5.60E-08	4.80E-08	2.00E-08	1.00E-09
^{155}Eu	4.80E+00	1.70E-09	1.30E-09	5.00E-10	2.40E-11
^{158}Tb	1.50E+02	2.20E-13	9.60E-14	2.50E-14	1.30E-15
^{166m}Ho	1.20E+03	3.30E-11	3.90E-11	1.30E-11	5.60E-13
^{178m}Hf	3.00E+01	3.40E-10	1.80E-10	5.30E-11	2.10E-12
^{205}Pb	1.40E+07	2.80E-16	2.70E-17	1.20E-16	6.60E-18
^{233}U	1.60E+05	2.80E-12	1.20E-11	5.70E-13	2.50E-14
^{239}Pu	2.40E+04	4.00E-11	1.70E-11	3.70E-12	9.30E-14

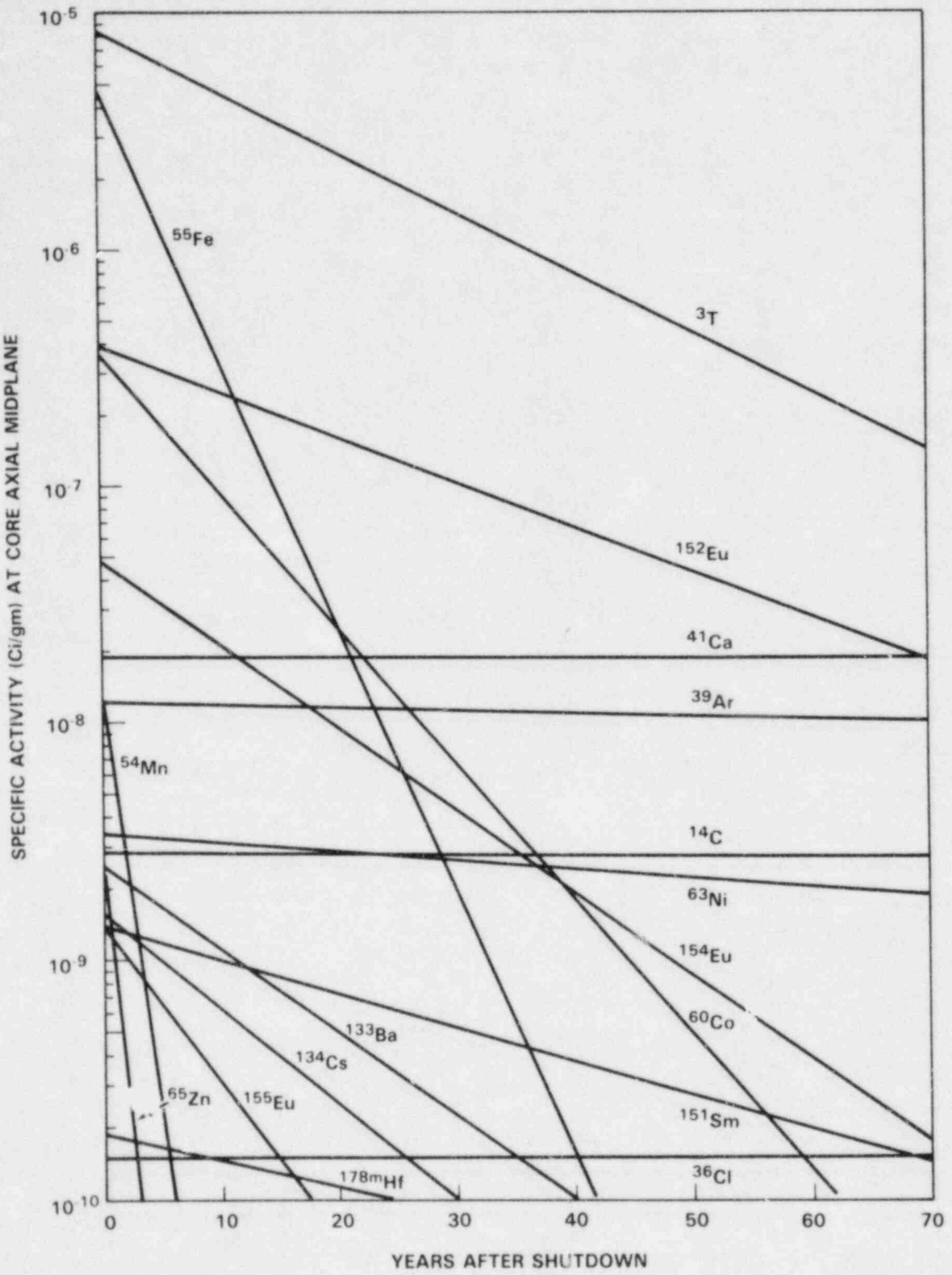

FIGURE 5.12. PWR Bioshield Average Concrete - 10 cm Depth 30 EFPY - Decommissioning Time Scale

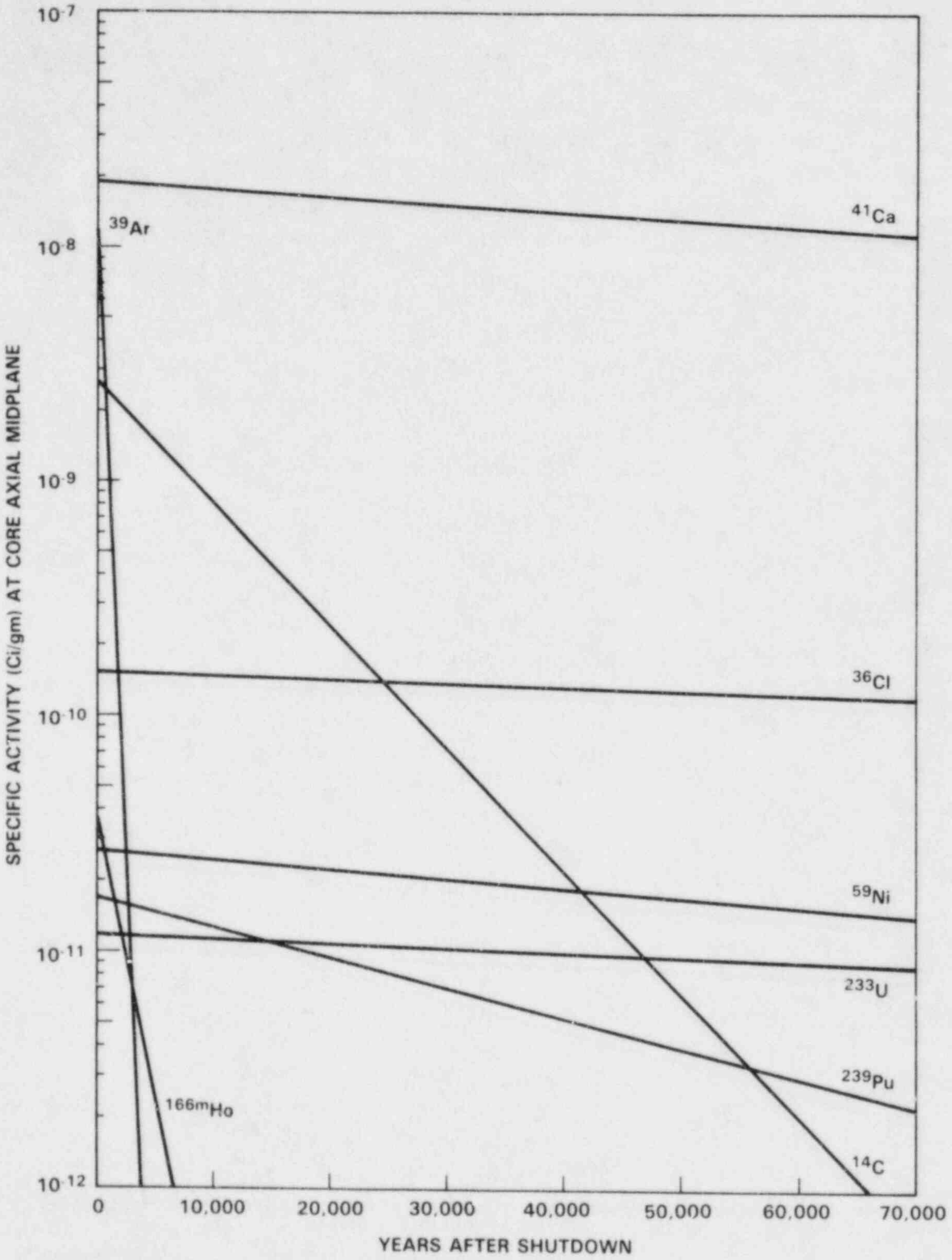

FIGURE 5.13. PWR Bioshield Average Concrete - 10 cm Depth 30 EFPY - Long Term Disposal Time Scale

TABLE 5.5. Activation of BWR Bioshield (Ci/gm) Average Concrete 30 EFPY at Core Axial Midplane

Isotope	Half-Life (Y)	Inner Edge	10 cm	20 cm	30 cm	60 cm
$^{3}\mathrm{T}$	1.20E+01	4.90E-07	8.60E-07	6.50E-07	3.40E-07	1.73E-08
$^{14}\mathrm{C}$	5.70E+03	1.70E-10	2.90E-10	2.20E-10	1.10E-10	5.78E-12
$^{36}\mathrm{Cl}$	3.00E+05	1.10E-11	1.60E-11	1.20E-11	6.00E-12	3.15E-13
$^{39}\mathrm{Ar}$	2.70E+02	4.70E-09	1.90E-09	8.30E-10	3.20E-10	2.65E-11
$^{41}\mathrm{Ca}$	1.00E+05	1.10E-09	2.00E-09	1.50E-09	7.60E-10	3.91E-11
$^{53}\mathrm{Mn}$	3.70E+06	3.80E-15	1.50E-15	6.70E-16	2.50E-16	2.14E-17
$^{54}\mathrm{Mn}$	8.50E-01	4.90E-09	2.00E-09	8.70E-10	3.30E-10	2.76E-11
$^{55}\mathrm{Fe}$	2.70E+00	2.90E0-07	4.90E-07	3.60E-07	1.90E-07	9.66E-09
$^{59}\mathrm{Ni}$	8.00E+04	1.60E-12	2.70E-12	2.00E-12	1.10E-12	5.47E-14
$^{60}\mathrm{Co}$	5.30E+00	2.80E-08	3.50E-08	2.50E-08	1.30E-08	6.41E-10
$^{63}\mathrm{Ni}$	1.00E+02	2.00E-10	3.40E-10	2.50E-10	1.30E-10	6.81E-12
$^{65}\mathrm{Zn}$	6.70E-01	2.00E-09	2.50E-09	1.70E-09	8.70E-10	4.49E-11
$^{79}\mathrm{Se}$	6.50E+04	3.20E-16	3.30E-16	2.20E-16	1.10E-16	6.16E-18
$^{81}\mathrm{Kr}$	2.10E+05	7.20E-19	4.50E-19	1.60E-19	3.40E-20	8.69E-23
$^{85}\mathrm{Kr}$	1.10E+01	6.30E-12	4.60E-12	2.90E-12	1.40E-12	8.06E-14
$^{90}\mathrm{Sr}$	2.90E+01	4.30E-12	7.40E-12	5.60E-12	2.90E-12	1.49E-13
$^{92m}\mathrm{Nb}$	2.70E+07	3.70E-19	2.10E-19	6.50E-20	2.40E-20	2.10E-21
$^{93}\mathrm{Zr}$	9.50E+05	2.90E-15	2.90E-15	2.00E-15	9.70E-16	5.07E-17
$^{93}\mathrm{Mo}$	3.50E+00	9.40E-14	4.50E-14	2.00E-14	7.20E-15	3.56E-16
$^{94}\mathrm{Nb}$	2.00E+04	6.10E-13	4.70E-13	2.90E-13	1.30E-13	6.72E-15
$^{99}\mathrm{Tc}$	2.10E+05	2.20E-14	1.10E-14	5.20E-15	2.00E-15	9.90E-17
$^{108m}\mathrm{Ag}$	1.30E+02	<3.10E-13	<3.10E-13	<2.10E-13	<1.00E-13	<5.13E-15
$^{121m}\mathrm{Sn}$	5.00E+00	<1.20E-13	<7.40E-14	<4.30E-14	<2.00E-14	<1.23E-15
$^{129}\mathrm{I}$	1.60E+07	1.30E-18	2.30E-18	1.70E-18	8.90E-19	4.58E-20
$^{133}\mathrm{Ba}$	1.00E+01	1.30E-10	2.10E-10	1.50E-10	7.80E-11	4.03E-12
$^{134}\mathrm{Cs}$	2.10E+00	8.50E-10	1.70E-09	1.30E-09	6.60E-10	3.41E11
$^{135}\mathrm{Cs}$	2.30E+06	9.90E-17	1.50E-16	1.10E-16	5.70E-17	2.97E-18
$^{137}\mathrm{Cs}$	3.00E+01	5.30E-12	8.00E-12	5.90E-12	3.00E-12	1.58E-13
$^{145}\mathrm{Pm}$	1.80E+01	5.80E-13	1.00E-12	7.60E-13	4.00E-13	2.04E-14
$^{146}\mathrm{Sm}$	1.00E+08	8.20E-20	3.30E-20	1.50E-20	5.50E-21	4.61E-22
$^{151}\mathrm{Sm}$	9.30E+01	1.20E-10	1.30E-10	8.50E-11	4.20E-11	2.14E-12
$^{152}\mathrm{Eu}$	1.30E+01	2.40E-08	4.20E-08	3.20E-08	1.60E-08	8.44E-10
$^{154}\mathrm{Eu}$	8.60E+00	4.70E-09	4.50E-09	3.00E-09	1.40E-09	7.35E-11
$^{155}\mathrm{Eu}$	4.80E+00	1.40E-10	1.20E-10	7.10E-11	3.30E-11	1.59E-12
$^{158}\mathrm{Tb}$	1.50E+02	4.80E-14	2.00E-14	8.60E-15	3.20E-15	2.71E-16
$^{166m}\mathrm{Ho}$	1.20E+03	5.10E-12	3.30E-12	1.80E-12	8.10E-13	4.09E-14
$^{178m}\mathrm{Hf}$	3.00E+01	2.70E-11	1.50E-11	7.60E-12	3.10E-12	1.56E-13
$^{205}\mathrm{Pb}$	1.40E+07	3.40E-17	3.00E-17	2.00E-17	9.50E-18	5.20E-19
$^{233}\mathrm{U}$	1.60E+05	2.20E-13	1.50E-13	8.10E-14	3.60E-14	1.80E-15
$^{239}\mathrm{Pu}$	2.40E+04	3.10E-12	1.30E-12	5.20E-13	1.60E-13	7.81E-15

TABLE 5.6. Activation of PWR Bioshield (Ci/gm) Average Rebar 30 EFPY at Core Axial Midplane

Isotope	Half-Life (Y)	Inner Edge	10 cm	24 cm	55 cm
^{3}T	1.20E+01	2.50E-08	4.10E-08	2.20E-08	1.30E-09
^{14}C	5.70E+03	1.10E-09	1.80E-09	9.50E-10	5.40E-11
^{36}Cl	3.00E+05	6.70E-14	2.90E-14	7.60E-15	4.00E-16
^{39}Ar	2.70E+02	1.40E-10	6.20E-11	1.60E-11	8.70E-13
^{41}Ca	1.00E+05	3.20E-12	5.20E-12	2.70E-12	1.50E-13
^{53}Mn	3.70E+06	4.30E-13	1.90E-13	4.90E-14	2.50E-15
^{54}Mn	8.50E-01	7.00E-07	3.00E-07	7.90E-08	4.20E-09
^{55}Fe	2.70E+00	7.30E-05	1.20E-04	6.20E-05	3.50E-06
^{59}Ni	8.00E+04	4.50E-10	7.40E-10	4.00E-10	2.20E-11
^{60}Co	5.30E+00	3.40E-06	3.70E-06	1.80E-06	9.60E-08
^{63}Ni	1.00E+02	5.50E-08	9.30E-08	4.90E-08	2.80E-09
^{65}Zn	6.70E-01	2.00E-08	2.20E-08	1.10E-08	5.70E-10
^{79}Se	6.50E+04	4.60E-15	5.50E-15	2.80E-15	1.60E-16
^{81}Kr	2.10E+05	<4.50E-17	<2.30E-17	<3.20E-18	<7.00E-21
^{85}Kr	1.10E+01	<2.10E-11	<1.50E-11	<6.30E-12	<3.50E-13
^{90}Sr	2.90E+01	<1.50E-11	<2.40E-11	<1.30E-11	<7.10E-13
^{92m}Nb	2.70E+07	<1.90E-17	<5.80E-18	<1.50E-18	<7.90E-20
^{93}Zr	9.50E+05	4.50E-15	4.80E-15	2.10E-15	1.10E-16
^{93}Mo	3.50E+03	2.50E-11	1.20E-11	2.90E-12	9.60E-14
^{94}Nb	2.00E+04	<7.00E-11	<4.90E-11	<1.80E-11	<8.70E-13
^{99}Tc	2.10E+05	5.70E-12	2.60E-12	6.10E-13	1.80E-14
^{108m}Ag	1.30E+02	7.60E-11	6.80E-11	3.00E-11	1.50E-12
^{121m}Sn	5.00E+00	2.00E-12	8.40E-13	2.20E-13	1.20E-14
^{129}I	1.60E+07	<4.60E-18	<8.00E-18	<3.90E-18	<2.20E-19
^{133}Ba	1.00E+01	<5.40E-10	<8.20E-10	<4.30E-10	<2.40E-11
^{134}Cs	2.10E+00	<3.80E-09	<7.30E-09	<4.00E-09	<2.30E-10
^{135}Cs	2.30E+06	<3.20E-16	<4.80E-16	<2.50E-16	<1.40E-17
^{137}Cs	3.00E+01	<1.70E-11	<2.60E-11	<1.30E-11	<7.40E-13
^{145}Pm	1.80E+01	<1.20E-13	<2.00E-13	<1.10E-13	<5.90E-15
^{146}Sm	1.00E+08	<7.20E-21	<3.40E-21	<8.80E-22	<4.60E-23
^{151}Sm	9.30E+01	<2.80E-11	<2.90E-11	<1.30E-11	<6.30E-13
^{152}Eu	1.30E+01	<3.90E-08	<6.40E-08	<3.50E-08	<2.00E-09
^{154}Eu	8.60E+00	9.10E-09	<7.80E-09	<3.30E-09	<1.70E-10
^{155}Eu	4.80E+00	<4.10E-11	<3.60E-11	<1.20E-11	<4.90E-13
^{158}Tb	1.50E+02	<5.40E-13	<2.30E-13	<6.10E-14	<3.20E-15
^{166m}Ho	1.20E+03	<2.80E-11	<1.70E-11	<5.70E-12	<2.50E-13
^{178m}Hf	3.00E+01	<1.20E-10	<6.70E-11	<1.90E-11	<7.60E-13
^{205}Pb	1.40E+07	<4.20E-16	<4.00E-16	<1.80E-16	<9.80E-18
^{233}U	1.60E+05	<4.80E-13	<2.90E-13	<9.70E-14	<4.30E-15
^{239}Pu	2.40E+04	<1.30E-11	<5.80E-12	<1.20E-12	<3.10E-14

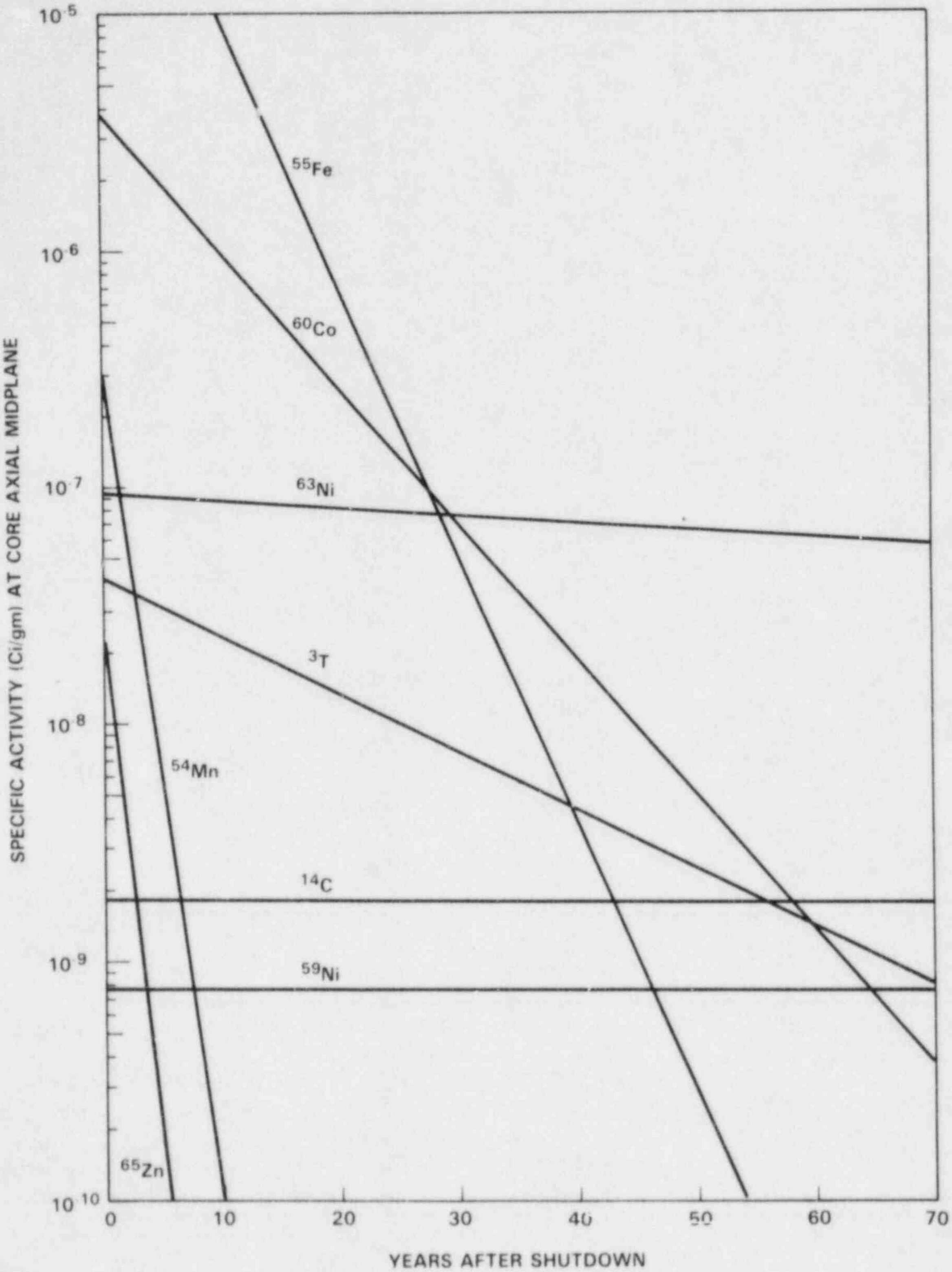

FIGURE 5.14. PWR Bioshield Rebar - 10 cm Depth 30 EFPY - Decommissioning Time Scale

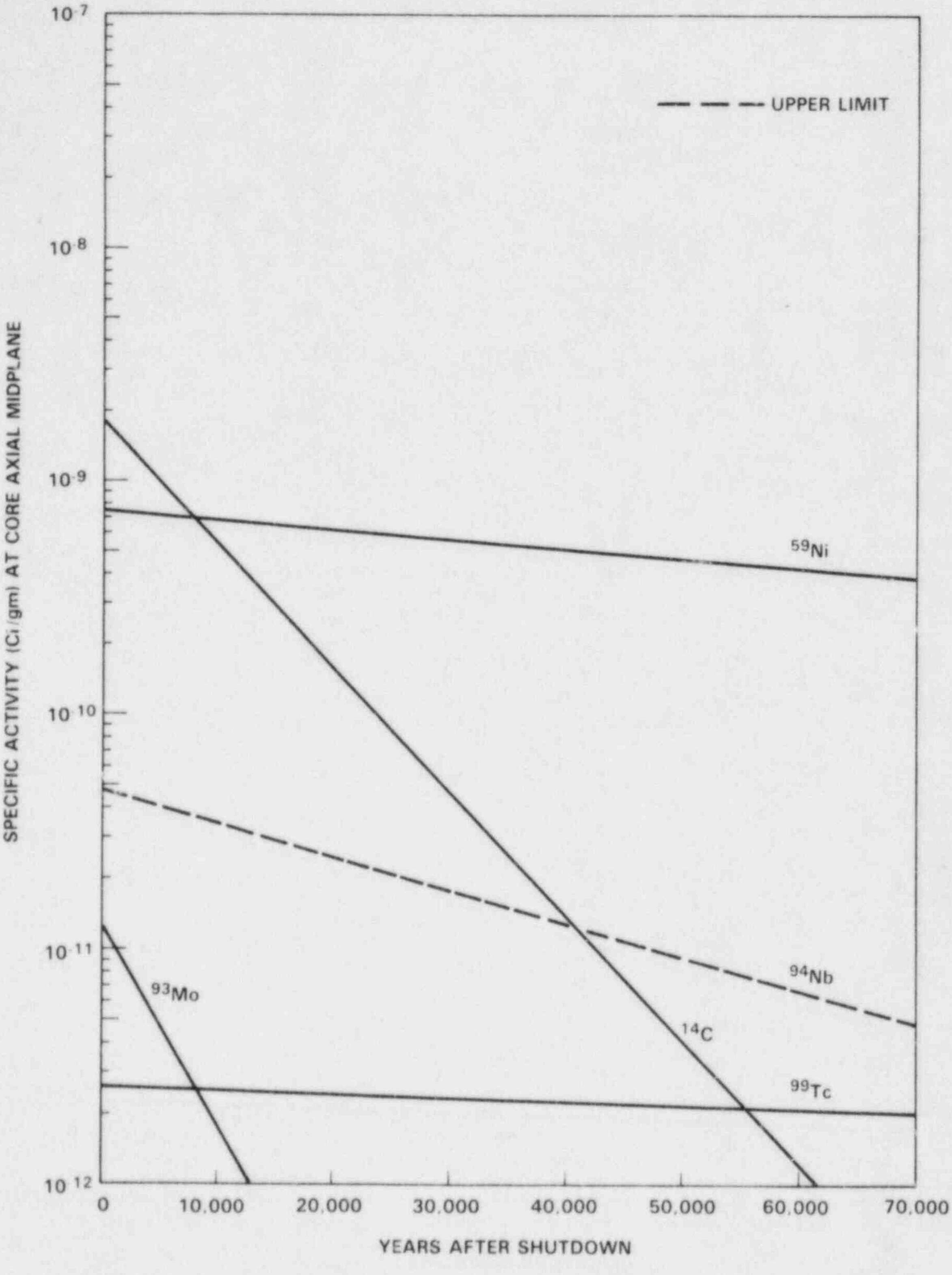

FIGURE 5.15. PWR Bioshield Rebar - 10 cm Depth 30 EFPY - Low Term Disposal Time Scale

ABLE 5.7. Activation of BWR Bioshield (Ci/gm) Average Rebar
30 EFPY at Core Axial Midplane

Isotope	Half-Life (Y)	Inner Edge	10 cm	20 cm	30 cm	60 cm
^{3}T	1.20E+01	2.46E-09	4.31E-09	3.23E-09	1.68E-09	8.60E-11
^{14}C	5.70E+03	1.06E-10	1.85E-10	1.39E-10	7.21E-11	3.70E-12
^{36}Cl	3.00E+05	1.17E-14	4.72E-15	2.07E-15	7.78E-16	6.60E-17
^{39}Ar	2.70E+02	2.49E-11	1.00E-02	4.41E-12	1.66E-12	1.40E-13
^{41}Ca	1.00E+05	3.13E-13	5.36E-13	4.00E-13	2.07E-13	1.10E-14
^{53}Mn	3.70E+06	9.48E-14	3.85E-14	1.68E-14	6.30E-15	5.40E-16
^{54}Mn	8.50E-01	1.23E-07	4.97E-08	2.18E-08	8.20E-09	6.90E-10
^{55}Fe	2.70E+00	7.19E-06	1.22E-05	9.08E-06	4.70E-06	2.40E-07
^{59}Ni	8.00E+04	4.42E-11	7.69E-11	5.75E-11	2.99E-11	1.50E-12
^{60}Co	5.30E+00	2.98E-07	3.71E-07	2.60E-07	1.30E-07	6.70E-09
^{63}Ni	1.00E+02	5.40E-09	9.52E-09	7.13E-09	2.70E-09	1.90E-10
^{65}Zn	6.70E-01	1.77E-09	2.21E-09	1.55E-09	7.80E-10	4.00E-11
^{79}Se	6.50E+04	6.64E-16	6.55E-165	4.43E-16	2.20E-16	1.20E-17
^{81}Kr	2.10E+05	<3.01E-19	<1.86E-19	<6.60E-20	<1.40E-20	<3.60E-23
^{85}Kr	1.10E+01	<3.21E-12	<2.00E-12	<1.20E-12	<5.30E-13	<3.30E-14
^{90}Sr	2.90E+01	<1.43E-12	<2.48E-12	<1.85E-12	<9.60E-13	<5.00E-14
^{92m}Nb	2.70E+07	<2.95E-18	<1.20E-18	<5.20E-19	<2.70E-19	<2.20E-20
^{93}Zr	9.50E+05	<4.30E-16	<5.00E-16	<3.10E-16	<1.50E-16	<8.60E-18
^{93}Mo	3.50E+00	1.99E-12	9.60E-13	4.20E-13	1.50E-13	7.50E-15
^{94}Nb	2.00E+04	<5.69E-12	<4.40E-12	<2.65E-12	<1.20E-12	<6.30E-14
^{99}Tc	2.10E+05	4.46E-13	2.10E-13	8.70E-14	3.00E-14	1.50E-15
^{108m}Ag	1.30E+02	6.47E-12	6.50E-12	4.30E-12	2.10E-12	1.10E-13
^{121m}Sn	5.00E+01	3.90E-13	1.58E-13	6.90E-14	2.60E-14	2.20E-15
^{129}I	1.60E+07	<4.40E-19	<7.70E-19	<5.70E-19	<3.00E-19	<1.50E-20
^{133}Ba	1.00E+01	<5.53E-11	<8.065E-11	<6.30E-11	<3.30E-11	<1.70E-12
^{134}Cs	2.10E+00	<3.93E-10	<7.70E-10	<5.80E-10	<3.10E-10	<1.60E-11
^{135}Cs	2.30E+06	<3.42E-17	<5.10E-17	<3.70E-17	<1.90E-17	1.00E-18
^{137}Cs	3.00E+01	<1.86E-12	<2.70E-12	<2.00E-12	<1.00E-12	<5.30E-14
^{145}Pm	1.80E+01	<1.86E-14	<2.04E-14	<1.50E-14	<7.90E-15	<4.20E-16
^{146}Sm	1.00E+08	<1.64E-21	<6.60E-22	<2.90E-22	<1.10E-22	<9.20E-24
^{151}Sm	9.30E+01	<2.37E-12	<2.60E-12	<1.75E-12	<8.60E-13	<4.40E-14
^{152}Eu	1.30E+01	<3.92E-09	<6.90E-09	<5.20E-09	<2.70E-09	<1.40E-10
^{154}Eu	8.60E+00	<7.74E-10	<7.70E-10	<4.80E-10	<2.40E-10	<1.20E-11
^{155}Eu	4.80E+00	<2.83E-12	<2.40E-12	<1.50E-12	<6.90E-13	<3.40E-14
^{158}Tb	1.50E+02	<1.18E-13	<4.80E-14	<2.10E-14	<7.90E-15	<6.60E-16
^{166m}Ho	1.20E+03	<2.25E-12	<1.50E-12	<8.20E-13	<3.60E-13	<1.80E-14
^{178m}Hf	3.00E+01	<9.81E-12	<5.60E-12	<2.80E-12	<1.13E-12	<5.70E-14
^{205}Pb	1.40E+07	<5.04E-17	<4.44E-17	<2.90E-17	<1.40E-17	<7.70E-19
^{233}U	1.60E+05	<3.85E-14	<2.50E-14	<1.40E-14	<6.10E-15	<3.10E-16
^{239}Pu	2.40E+04	<1.02E-12	<4.44E-13	<1.70E-13	<5.40E-14	<2.60E-15

A comparison between maximum activation levels in the concrete matrix and the associated rebar is given in Table 5.8. For most activation products, the concrete matrix tends to dominate. Exceptions are those isotopes produced from Fe, Co, Ni, Mo, and Ag. At a 10% rebar content, for example, the rebar would contribute about as much ^{60}Co as the concrete, as well as contributing about three times as much ^{63}Ni and ^{59}Ni. It should not represent any unusual problems, particularly if co-disposed with the concrete.

5.4.6 Regulatory Considerations for Waste Disposal

The U.S. Nuclear Regulatory Commission has recently adopted formal guidelines for classification of waste for shallow land disposal. These regulations are given in 10 CFR Part 61. Maximum levels are specified for several long lived radionuclides. The regulation also specifies that indirect metnods may be used to determine the inventory of activation products of interest. The calculation method used in this study combined with chemical analysis and a normalization to at least one isotope such as ^{60}Co could thus be used for waste classification.

Several examples of this approach together with the appropriate regulatory limits from 10 CFR 61.55 are shown in Tables 5.9, 5.10, and 5.11 for comparison with expected maximum activation levels of the relevant isotopes. For all three cases the levels shown are at the point of maximum activation. Table 5.9 shows activity levels for PWR internals. The shroud clearly exceeds the Class C concentrations on the basis of ^{59}Ni, ^{63}Ni, ^{94}Nb, and ^{14}C content. In particular, the ^{63}Ni level is about a factor of 20 above the maximum level classified as Class C waste. Since the nickel content is relatively constant in type 304 stainless steel, chemical variations wil not alter that conclusion. The 100 year half-life of ^{63}Ni prevents large changes due to decay. The core barrel also appears to be marginally above the Class C limit on the basis of the ^{63}Ni content, although there are large uncertainties in the flux calculations. The vessel cladding would be classified as Class B waste based on its ^{63}Ni content. The vessel wall, with its considerably lower nickel content and lower flux, would clearly be a Class A waste. Table 5.10 lists the same type of comparisons for the BWR case. Again, according to this analysis, the shroud would not be classified as generally suitable for near surface disposal based on its ^{59}Ni, ^{63}Ni, and ^{94}Nb content. The cladding and vessel would be Class A waste. It should be noted, however, that the isotope inventories calculated in this fashion pertain only to direct neutron activation. Vessel cladding, for example, may contain a radionuclide inventory dominated by translocation products associated with the surface corrosion film. In that case, the waste classification could be substantially different, but also potentially modifiable by decontamination. The bioshield results for the two geometries is shown on Table 5.11. Even for the highest levels produced at core axial midplane and 10 cm depth, this material should be classified as Class A or B for the PWR,

TABLE 5.8. Comparison of Rebar Versus Concrete Matrix of PWR Bioshield 30 EFPY at Core Axial Midplane 10 cm Deep

Isotope	Half-Life (Y)	Concrete	Rebar	Ratio
$^{3}\mathrm{T}$	1.20E+01	8.30E-06	4.10E-08	200.0
$^{14}\mathrm{C}$	5.70E+03	2.80E-09	1.80E-09	1.6
$^{36}\mathrm{Cl}$	3.00E+05	1.50E-19	2.90E-14	5200.0
$^{39}\mathrm{Ar}$	2.70E+02	1.20E-08	6.20E-11	190.0
$^{41}\mathrm{Ca}$	1.00E+05	1.90E-08	5.20E-12	3700.0
$^{53}\mathrm{Mn}$	3.70E+06	7.40E-15	1.90E-13	0.04
$^{54}\mathrm{Mn}$	8.50E-01	1.20E-08	3.00E-07	0.04
$^{55}\mathrm{Fe}$	2.70E+00	4.70E-06	1.20E-04	0.04
$^{59}\mathrm{Ni}$	8.00E+04	2.60E-11	7.40E-10	0.04
$^{60}\mathrm{Co}$	5.30E+00	3.60E-07	3.70E-06	0.10
$^{63}\mathrm{Ni}$	1.00E+02	3.30E-09	9.30E-08	0.04
$^{65}\mathrm{Zn}$	6.70E-01	2.50E-09	2.20F-08	0.11
$^{79}\mathrm{Se}$	6.50E+04	2.80E-15	5.50E-15	0.51
$^{81}\mathrm{Kr}$	2.10E+05	5.50E-17	2.30E-17	2.40
$^{85}\mathrm{Kr}$	1.10E+01	4.20E-11	<1.50E-11	>2.80
$^{90}\mathrm{Sr}$	2.90E+01	7.30E-11	<2.40E-11	>3.0
$^{92m}\mathrm{Nb}$	2.70E+07	1.20E-19	<5.80E-18	>0.02
$^{93}\mathrm{Zr}$	9.50E+05	2.90E-14	4.80E-15	6.0
$^{93}\mathrm{Mo}$	3.50E+00	5.60E-13	1.20E-11	0.05
$^{94}\mathrm{Nb}$	2.00E+04	5.30E-12	<4.90E-11	>0.11
$^{99}\mathrm{Tc}$	2.10E+05	1.40E-13	<2.60E-12	>0.05
$^{108m}\mathrm{Ag}$	1.30E+02	<3.20E-12	6.80E-11	<0.05
$^{121m}\mathrm{Sn}$	5.00E+00	<5.20E-13	8.40E-13	<0.62
$^{129}\mathrm{I}$	1.60E+07	2.20E-17	<8.00E-18	>2.9
$^{133}\mathrm{Ba}$	1.00E+01	2.00E-09	<8.20E-10	>2.4
$^{134}\mathrm{Cs}$	2.10E+00	1.60E-08	<7.30E-09	>2.2
$^{135}\mathrm{Cs}$	2.30E+06	1.40E-15	<4.80E-16	>2.9
$^{137}\mathrm{Cs}$	3.00E+01	7.60E-11	<2.60E-11	>2.9
$^{145}\mathrm{Pm}$	1.80E+01	9.80E-12	<2.00E-13	>49.0
$^{146}\mathrm{Sm}$	1.00E+08	1.70E-19	<3.40E-21	>50.0
$^{151}\mathrm{Sm}$	9.30E+01	1.40E-09	<2.90E-11	>48.0
$^{152}\mathrm{Eu}$	1.30E+01	3.90E-07	<6.40-08	>6.1
$^{154}\mathrm{Eu}$	8.60E+00	4.80E-08	<7.80E-09	>6.2
$^{155}\mathrm{Eu}$	4.80E+00	1.30E-09	<3.60E-11	>3.61
$^{158}\mathrm{Tb}$	1.50E+02	9.60E-14	<2.30E-13	>0.42
$^{166m}\mathrm{Ho}$	1.20E+03	3.90E-11	<1.70E-11	>2.3
$^{178m}\mathrm{Hf}$	3.00E+01	1.80E-10	<6.70E-11	>2.7
$^{205}\mathrm{Pb}$	1.40E+07	2.70E-17	<4.00E-16	>0.07
$^{233}\mathrm{U}$	1.60E+05	1.20E-11	<2.90E-13	>41.4
$^{239}\mathrm{Pu}$	2.40E+04	1.70E-11	<5.80E-12	>2.93

TABLE 5.9. Classification of PWR Intervals for Low Level Waste Disposal - Core Axial Midplane - 30 EFPY, Activity in Ci/m^3 at Shutdown

Isotope	Shroud(d)	Core(d) Barrel	Thermal(d) Pads	Vessel(d) Cladding	Waste Class(a)		
					A	B	C
^{3}T	78	75	17	2.6	40	(b)	(b)
^{14}C	195	23	3.6	0.5	8	---	80
$T_{1/2}$ < 5 yr(c)	1.6E6	1.9E5	2.9E4	4.2E3	700	(b)	(b)
^{59}Ni	858	148	23	3.4	22	---	220
^{60}Co	1.0E6	1.1E5	1.6E4	2.6E3	700	(b)	(b)
^{63}Ni	1.4E5	1.9E4	3000	420	35	700	7000
^{94}Nb	3.1	0.23	0.023	0.006	0.02	---	0.2
^{99}Tc	1.0	0.064	0.003	0.0018	0.3	---	3

(a) According to definitions established in 10 CFR Part 61.55.

(b) There are no limits established for these radionuclides in Class B or C waste. Practical considerations such as the effect of external radiation and heat generation on transportation, handling and disposal will limit the concentrations for these wastes.

(c) Dominant activation products with half-lives of <5 yr are ^{55}Fe and ^{51}Cr. It is assumed for disposal purposes that the ^{51}Cr will be fully decayed.

(d) A density of 7.8 x 10^6 gm/m^3 is used to convert activities into current units.

and Class A for the BWR. Most of the mass of the bioshield is of considerably lower levels of activation with much of the outer layers non-radioactive.

The above discussion is very general and simply serves as an example of the application of the 10 CFR 61 guidelines to the decommissioning problem with respect to the activated components. In practice, the situation is somewhat more complicated. In the examples given, average chemical compositions and volume averaged fluxes were used. In practice, some chemical analysis will be required to relate measured ^{60}Co levels to the neutron flux in order to normalize the calculation. That should then allow a reasonably accurate estimate of the activation product inventory provided the material is reasonably homogeneous. That is not necessarily the case with certain types of reactor components such as control rods. Control rods pose a particularly difficult problem due to their high neutron opacity. The stainless steel cladding on the control

TABLE 5.10. Classification of BWR Intervals for Low Level Waste Disposal - Core Axial Midplane - 30 EFPY, Activity in Ci/m^3 at Shutdown

Isotope	Shroud$^{(d)}$	Vessel$^{(d)}$ Cladding	Waste Class$^{(a)}$ A	B	C
^{3}T	100	0.2	40	(b)	(b)
^{14}C	57	0.04	8	---	80
$T_{1/2} < 5$ yr$^{(c)}$	4.6E5	320	700	(b)	(b)
^{59}Ni	340	0.25	22	---	220
^{60}Co	2.5E5	190	700	(b)	(b)
^{63}Ni	44500	31	35	700	7000
^{94}Nb	0.49	3.8E-4	0.02	---	0.2
^{99}Tc	0.12	1.2E-4	0.3		3

(a) According to definitions established in 10 CFR Part 61.55.

(b) There are no limits established for these radionuclides in Class B or C waste. Practical considerations such as the effect of external radiation and heat generation on transportation, handling and disposal will limit the concentrations for these wastes.

(c) Dominant activation products with half-lives of <5 yr are ^{55}Fe and ^{51}Cr. It is assumed for disposal purposes that the ^{51}Cr will be fully decayed.

(d) A density of 7.8×10^6 gm/m^3 is used to convert activities into correct units.

rods should contain activity levels similar to that found in the shroud material and from that standpoint are presumably unsuitable for shallow land burial. PWR control rods contain an alloy of silver, cadmium, and indium and therefore may be expected to contain a large inventory of the 130 y ^{108m}Ag; however, that isotope is not considered in 10 CFR 61. BWR control rods contain boron carbide. Some ^{14}C will undoubtedly be present due to neutron capture on ^{13}C, but in all probability the activity in the cladding will predominate with ^{63}Ni being the limiting isotope.

TABLE 5.11. Classification of Bioshield Concrete for Low Level Waste Disposal - Core Axial Midplane - 30 EFPY, 10 cm Depth Activity in Ci/m^3 at Shutdown

Isotope	PWR	BWR	Waste Class$^{(a)}$ A	B	C
^{3}T	25	2.6	40	(b)	(b)
^{14}C	0.008	7.7E-4	0.8	---	8
$T_{1/2}$ < 5 yr$^{(c)}$	14	1.5	700	(b)	(b)
^{60}Co	1.1	0.10	700	(b)	(b)
^{63}Ni	0.011	0.0011	3.5	70	700
^{90}Sr	2.3E-4	2.3E-5	0.04	150	7000
^{99}Tc	3.8E-7	3.8E-8	0.3	---	3
^{129}I	7.6E-11	7.6E-12	0.008	---	0.08
^{137}Cs	2.3E-5	2.3E-6	1	44	4600
Transuranics	3.8E-5	3.8E-6	0.08	---	0.8

(a) According to definitions established in 10 CFR Part 61.55.

(b) There are no limits established for these radionuclides in Class B or C waste. Practical considerations such as the effect of external radiation and heat generation on transportation, handling and disposal will limit the concentrations for these wastes.

(c) Dominant activation products with half-lives of <5 yr are ^{55}Fe and ^{51}Cr. It is assumed for disposal purposes that the ^{51}Cr will be fully decayed.

(d) A density of 3 x 10^{6} gm/m^{3} is used to convert activities into correct units.

5.5 RADIONUCLIDE INVENTORIES

The specific activities for various isotopes given in Section 5.4 can be used to compute estimates of total isotope inventories. Since the neutron fluxes have already been volume averaged in computing the average specific activity, the only additional information required is the axial neutron distribution and the mass of the component included in that distribution. The parameters used are shown in Table 5.12. The PWR data was taken from Smith, Konzek, and Kennedy (1978). For the BWR case the data was taken from Oak et al. (1980). The calculated inventory for PWR internals is given in Table 5.13. It should be noted that this is not really a total inventory at shutdown since some short-lived activities such as ^{51}Cr and ^{59}Fe have not been included. The

TABLE 5.12. Parameters Used for Radionuclide Inventory

	Peak/Average	Mass of Component (kg)
PWR Shroud	0.755	12,312
PWR Core Barrel	0.637	26,783
PWR Thermal Pads	0.778	10,413
PWR Vessel Cladding	0.637	2,074
PWR Vessel Wall	0.637	245,582
BWR Shroud	0.467	30,100
BWR Vessel Cladding	0.467	3 440
BWR Vessel Wall	0.467	122,700

(a) Ratio of maximum activation at core axial midplane to average for entire component.

isotope inventory in the reactor internals is, of course, sharply dependent upon the time scale chosen. For the greater than one year time scale the total is 4.1E6 Ci, more than 80% of which is in the shroud. The majority of the activity on that time scale is ^{55}Fe. For the greater than 5, 100, and 1000 year time scale the dominant activities are ^{60}Co, ^{63}Ni, and ^{59}Ni, respectively. Inventories of long-lived activation products in reactor internals at shutdown are listed in Table 5.13 and 5.14. The comparable data for PWR and BWR vessel walls is given in Table 5.15. In order to calculate an inventory for the bioshield, some information on depth-dependent production rates is needed. This is shown in Figure 5.16 for PWR concrete. Maximum activation occurs at around 10 cm depth for most isotopes due to increasing neutron thermalization up to that point. Some minor differences in the profile do occur depending on the production mechanism. For example, ^{154}Eu which has a very large resonance contribution and ^{39}Ar which is produced entirely by fast neutrons do not exhibit that peaking behavior. In any case it is quite clear from Figure 5.15 that essentially all of the activated concrete is contained within 30 to 50 cm of the inner edge of the bioshield. To calculate a total inventory from Figure 5.15, a 10% rebar component was added and a numerical integration carried out over the outer 75 cm of the bioshield in 5 cm increments for a 502 cm vertical section of the shield. The axial integration factor for the vessel was also included. A similar calculation was carried out for a 451 cm segment of a BWR sacrificial shield. Activity inventories are summarized in Table 5.16. Long-lived activation product inventories of reactor internals are somewhat lower for the BWR than the PWR due to the core design. The activation product inventory for the bioshield is actually about one order of magnitude lower for the BWR due to the considerably reduced neutron fluxes.

TABLE 5.13. Activity Inventory of PWR Internals at Shutdown (Total Ci)

Isotope	Half-Life (Y)	Shroud	Core Barrel	Thermal Pads	Vessel Cladding
^{3}T	1.20E+01	9.30E+01	1.64+02	1.79E+01	4.40E-01
^{14}C	5.70E+03	2.32E+02	5.10E+01	3.70E+00	8.70E-02
^{36}Cl	3.00E+05	4.70E+00	1.10E+00	8.10E-02	1.85E-03
^{39}Ar	2.70E+02	<1.40E+00	<1.40E-01	<3.00E-03	<3.4[illegible]E-04
^{41}Ca	1.00E+05	<4.30E-02	<9.55E-03	<6.97E-04	<1.60E-05
^{53}Mn	3.70E+06	3.00E-02	3.60E-03	8.10E-05	9.00E-06
^{54}Mn	8.50E-01	6.04E+04	6.14E+03	1.46E+02	1.58E+01
^{55}Fe	2.70E+00	1.95E+06	4.09E+05	3.00E+04	7.13E+02
^{59}Ni	8.00E+04	1.02E+03	3.24E+02	2.43E+01	5.70E-01
^{60}Co	5.30E+00	1.21E+06	2.39E+05	1.62E+04	4.36E+02
^{63}Ni	1.00E+02	1.68E+05	4.09E+04	3.08E+03	7.13E+01
^{65}Zn	6.70E-01	5.95E+03	6.78E+02	4.78E+01	1.30E+00
^{79}Se	6.50E+04	5.70E-03	7.85E-04	3.80E-05	2.00E-06
^{81}Kr	2.10E+05	<7.10E-03	<5.80E-05	<2.84E-07	<2.77E-09
^{85}Kr	1.10E+01	<7.90E+00	<3.24E-01	<7.70E-03	<1.72E-04
^{90}Sr	2.90E+01	<1.86E+01	<8.50E-02	<2.10E-02	<4.76E-04
^{92m}Nb	2.70E+07	1.10E-05	1.00E-06	2.59E-08	2.64E-09
^{93}Zr	9.50E+05	1.00E-03	6.70E-05	2.00E-06	1.13E-07
^{93}Mo	3.50E+00	8.70E+00	6.65E-01	1.70E-02	1.50E-03
^{94}Nb	2.00E+04	3.70E+00	4.95E-01	2.40E-02	9.90E-04
^{99}Tc	2.10E+05	1.20E+00	1.40E-01	3.40E-03	3.17E-04
^{108m}Ag	1.30E+02	<9.30E-01	<1.50E-01	<8.90E-03	<2.91E-04
^{121m}Sn	5.00E+00	4.40E-02	5.80E-03	1.38E-04	1.50E-05
^{129}I	1.60E+07	<6.00E-06	<2.39E-07	<6.40E-09	<1.45E-10
^{133}Ba	1.00E+01	<2.70E+01	<5.11E+00	<3.60E-01	<9.40E-04
^{134}Cs	2.10E+00	<6.50E+01	<1.70E+01	<1.37E+00	<3.00E-02
^{135}Cs	2.30E+06	<4.00E-04	<1.50E-05	<4.10E-07	<1.03E-08
^{137}Cs	3.00E+01	<1.90E+01	<8.50E-01	<2.20E-02	<5.55E-04
^{145}Pm	1.80E+01	<8.00E-03	<2.39E-03	<1.78E-04	<4.00E-06
^{146}Sm	1.00E+08	9.30E-10	2.21E-10	5.50E-12	5.70E-13
^{151}Sm	9.30E+01	4.30E-02	7.70E-02	2.30E-02	7.80E-04
^{152}Eu	1.30E+01	0.00E+00	1.55E-02	1.38+00	1.00E-01
^{154}Eu	8.60E+00	5.20E+00	1.02E+01	5.00E-01	1.60E-02
^{155}Eu	4.80E+00	3.80E+00	2.39E+00	4.05E-02	6.90E-04
^{158}Tb	1.50E+02	1.80E-02	2.90E-03	6.90E-04	7.00E-06
^{166m}Ho	1.20E+03	1.50E+))	2.21E-01	8.90E-03	4.00E-04
^{178m}Hf	3.00E+01	<4.00E-01	<4.77E-01	<2.50E-02	<1.60E-03
^{205}Pb	1.40E+07	1.70E-05	2.00E-06	1.05E-07	4.50E-09
^{233}U	1.60E+05	<3.30E-03	<1.70E-03	<8.90E-05	<4.00E-06
^{239}Pu	2.40E+04	<6.50E-02	<3.92E-02	<1.0E-03	<1.27E-04

TABLE 5.14. Activity Inventory of BWR Internals at Shutdown (Total Ci)

Isotope	Half-Life (Y)	Shroud	Vessel Cladding
^{3}T	1.20E+01	1.83E+02	1.40E-02
^{14}C	5.70E+03	1.03E+02	2.80E-03
^{36}Cl	3.00E+05	2.24E+00	5.70E-05
^{39}Ar	2.70E+02	<2.67E-01	<3.00E-05
^{41}Ca	1.00E+05	2.00E-02	5.20E-07
^{53}Mn	3.70E+06	6.50E-03	8.00E-07
^{54}Mn	8.50E-01	1.17E+04	1.14E+00
^{55}Fe	2.70E+00	9.29E+05	2.36E+01
^{59}Ni	8.00E+04	6.04E+02	1.80E-02
^{60}Co	5.30E+00	4.50E+05	1.38E+01
^{63}Ni	1.00E+02	8.0'E+04	2.30E+00
^{65}Zn	6.70E-01	1.55E+03	4.10E-02
^{79}Se	6.50E+04	1.40E-03	9.80E-08
^{81}Kr	2.10E+05	<2.24E-04	<5.40E-12
^{85}Kr	1.10E+01	<8.15E-01	<5.00E-06
^{90}Sr	2.90E+01	<2.11E+00	<1.40E-05
^{92m}Nb	2.70E+07	6.33E-07	2.20E-10
^{93}Zr	9.50E+05	1.41E-04	6.90E-09
^{93}Mo	3.50E+00	1.08E+00	4.40E-05
^{94}Nb	2.00E+04	8.86E-01	2.80E-05
^{99}Tc	2.10E+05	2.10E-01	9.00E-06
^{108m}Ag	1.30E+02	<2.18E-01	<9.00E-06
^{121m}Sn	5.00E+00	1.07E-02	1.00E-06
^{129}I	1.60E+07	<5.90E-07	<4.40E-12
^{133}Ba	1.00E+01	<1.0rE+01	<3.50E-04
^{134}Cs	2.10E+00	<3.37E+01	<9.80E-04
^{135}Cs	2.30E+06	<3.80E-04	<3.67E-10
^{137}Cs	3.00E+01	<2.11E+00	<2.00E-05
^{145}Pm	1.80E+01	<4.40E-03	<1.30E-07
^{146}Sm	1.00E+08	4.07E-10	4.50E-14
^{151}Sm	9.30E+01	5.30E-02	1.80E-05
^{152}Eu	1.30E+01	3.09E-07	3.85E-03
^{154}Eu	8.60E+00	1.28E+01	4.60E-04
^{155}Eu	4.80E+00	5.06E+00	1.70E-05
^{158}Tb	1.50E+02	5.30E-03	6.31E-07
^{166m}Ho	1.20E+03	3.93E-01	1.10E-05
^{178m}Hf	3.00E+01	<5.20E-01	<4.30E-05
^{205}Pb	1.40E+07	4.00E-06	2.58E-10
^{233}U	1.60E+05	<2.25E-03	<1.30E-07
^{239}Pu	2.40E+04	<3.80E-02	<3.00E-06

TABLE 5.15. Activity Inventories of PWR and BWR Vessel Walls at Shutdown (Total Ci)

Isotope	Half-Life (Y)	PWR	BWR
^{3}T	1.20E+01	1.70E+01	6.10E-02
^{14}C	5.70E+03	2.80E-01	1.20E-02
^{36}Cl	3.00E+05	1.70E-02	1.43E-04
^{39}Ar	2.70E+02	<3.70E-02	<1.10E-03
^{41}Ca	1.00E+05	2.20E-04	2.00E-06
^{53}Mn	3.70E+06	4.06E-04	1.00E-05
^{54}Mn	8.50E-01	6.08E+01	1.27E+01
^{55}Fe	2.70E+00	1.87E+04	1.14E+02
^{59}Ni	8.00E+04	6.55E-01	8.00E-02
^{60}Co	5.30E+00	1.17E+03	9.55E+01
^{63}Ni	1.00E+02	7.96E+01	9.55E+00
^{65}Zn	6.70E-01	8.74E+00	2.90E-02
^{79}Se	6.50E+04	7.90E-07	1.00E-06
^{81}Kr	2.10E+05	<2.80E-08	<3.04E-11
^{85}Kr	1.10E+01	<3.00E-02	<2.20E-05
^{90}Sr	2.90E+01	<8.30E-04	<6.10E-05
^{92m}Nb	2.70E+07	1.29E-08	2.90E-09
^{93}Zr	9.50E+05	3.00E-06	8.10E-08
^{93}Mo	3.50E+00	2.30E-01	7.96E-04
^{94}Nb	2.00E+04	1.10E-02	7.20E-05
^{99}Tc	2.10E+05	5.50E-02	1.59E-04
^{108m}Ag	1.30E+02	<1.15E-02	<7.80E-05
^{121m}Sn	5.00E+00	3.90E-04	1.40E-05
^{129}I	1.60E+07	<2.50E-10	1.88E-12
^{133}Ba	1.00E+01	<1.00E-01	<2.20E-03
^{134}Cs	2.10E+00	<2.70E-01	<3.50E-03
^{135}Cs	2.30E+06	<3.60E-08	<2.46E-09
^{137}Cs	3.00E+01	<2.00E-03	<1.38E-04
^{145}Pm	1.80E+01	<1.20E-05	<9.41E-08
^{146}Sm	1.00E+08	3.00E-12	6.20E-13
^{151}Sm	9.30E+01	3.70E-03	1.45E-04
^{152}Eu	1.30E+01	3.12E+00	1.70E-02
^{154}Eu	8.60E+00	1.01E+00	4.50E-03
^{155}Eu	4.80E+00	6.09E-03	1.88E-04
^{158}Tb	1.50E+02	2.20E-04	8.00E-06
^{166m}Ho	1.20E+03	2.00E-02	1.59E-04
^{178m}Hf	3.00E+01	<1.15E-02	<7.09E-04
^{205}Pb	1.40E+07	2.00E-06	3.04E-09
^{233}U	1.60E+05	<5.00E-05	<2.00E-06
^{239}Pu	2.40E+04	<1.10E-03	<6.80E-05

TABLE 5.16. Activity Inventory Summary

Isotope Half-Life	Internals		Vessel		Bioshield	
	PWR	BWR	PWR	PWR	PWR	BWR
>1 year	4.1E6	1.4E6	2.0E4	2020	4.7E3	376
>5 years	1.7E6	5.3E5	1.3E3	14	1.7E3	146
>100 years	2.1E5	8.1E4	81	0.88	3	0.3
>1000 years	2.0E3	770	1.7	0.019	3	0.3

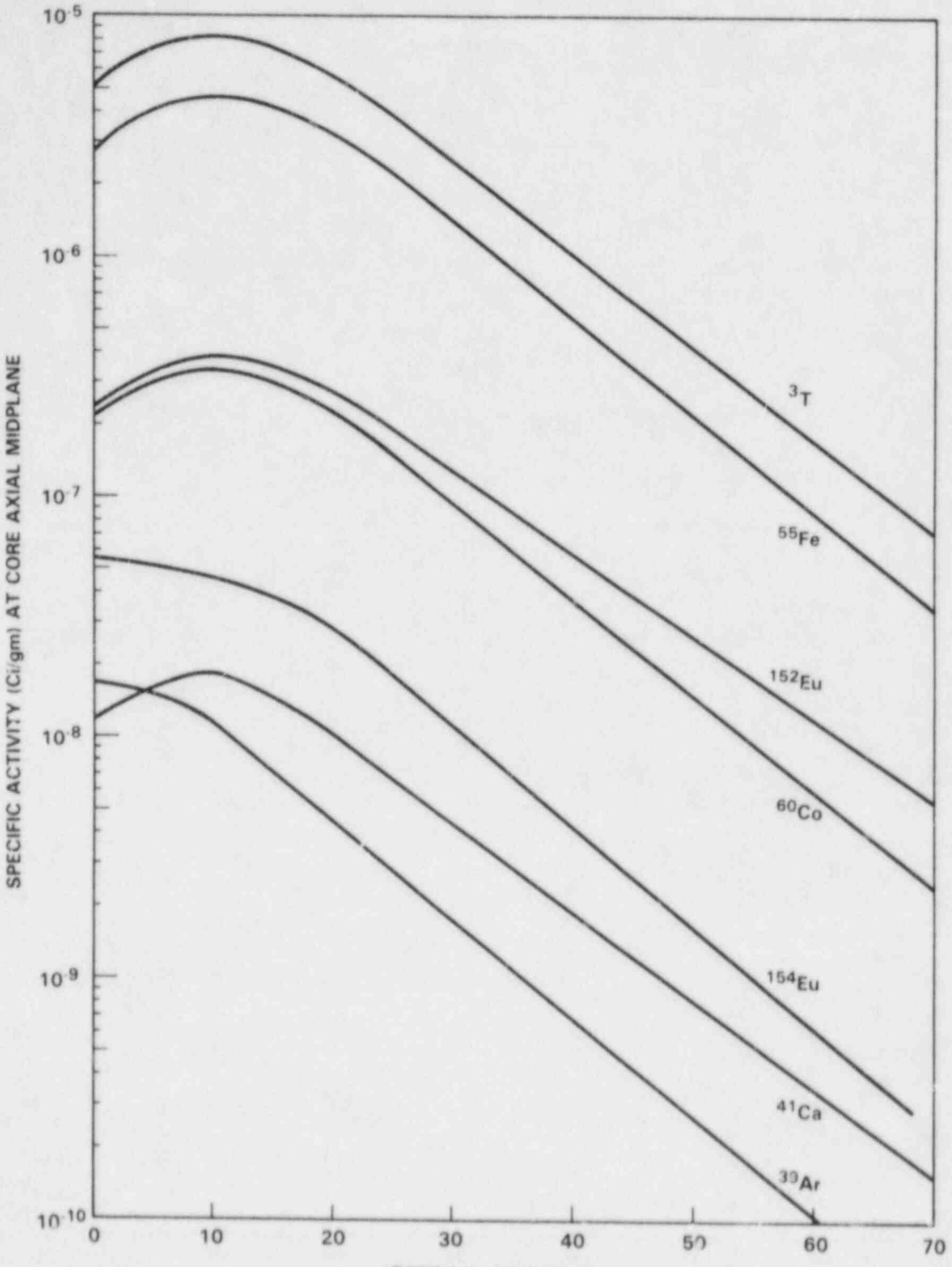

FIGURE 5.16. PWR Bioshield Average Concrete Activity Versus Depth 30 EFPY Operation

6.0 RADIOCHEMICAL ANALYSIS PROGRAM

Throughout the course of this work, samples of activated reactor components were sought to provide verification of the calculation method. Ideal samples were, in general, not available from operating reactors although some useful samples of opportunity were obtained. Several shutdown reactors including Pathfinder, Humboldt Bay, and Indian Point No. 1 were also included in the programs. The terms of the operating license for these reactors prevents any serious sampling effort from taking place at this time for either bioshield or internals samples, however. A major effort was undertaken at Pathfinder to obtain a core through the bioshield in the area adjacent to the reactor core region. This ultimately proved infeasible with the facilities and contamination controls available. Such a task would be relatively simple during an actual dismantling operation. Concrete samples were taken from accessible levels at the Pathfinder, Humboldt Bay and Turkey Point No. 4 reactors; however, they were from areas of highly reduced flux with only a limited radiochemical analysis possible.

6.1 SAMPLES ANALYZED

The following is a brief description of the samples subjected to a limited radiochemical analysis.

6.1.1 Steels

Point Beach No. 1 Fuel Support Structure

Samples of 304L stainless steel and Type 718 Inconel were cut from a fuel support structure originating in the Point Beach No. 1 reactor (a 497 MWe Westinghouse PWR). The fuel bundle (B25) was removed from the reactor on 5/10/74 after two cycles of irradiation running from 11/2/72 to 9/29/72 and 3/3/73 to 4/6/74. The samples were removed from the fuel bundle during a series of nuclear waste vitrification studies at PNL. The samples were extremely radioactive, representing a difficult handling problem even after eight years decay. Drill turnings were taken in a shielded glove box for XRF and radiochemical analysis. The remainder of the sample was archived. XRF analysis of the Inconel sample showed it to be very high in stable niobium (5%) making it useful for the determination of ^{94}Nb. Niobium was in fact detectable in this sample by a simple gamma scan without chemical separation from the ^{60}Co.

Monticello Core Spray Sparger Pin

Support pins from the Monticello Reactor (a 545 MWe GE BWR) were removed during a scheduled maintenance outage. The pins were located on the vessel wall 287 cm above the top of the reactor core. This sample thus corresponds reasonably well to the flux calculations for BWR vessel cladding given in Section 5.4.1, but at a significantly reduced flux level. The period of radiation was from November 1975 to October 1977. One of the pins was shipped to PNL for analysis. The pin is fabricated from type 304L stainless steel. It was cut into several pieces with samples of both ends taken for radiochemical analysis. The sample posed no unusual handling problems, particularly after the corrosion film had been stripped off.

Westinghouse - EPRI Surveillance Capsule Samples

A number of activated vessel steel samples were obtained through a pressure vessel surveillance program run by Westinghouse for EPRI. The samples were provided by Westinghouse at the request of EPRI. The samples consisted of both base metal and welds. All appeared to be very similar in composition and thus a limited selection was analyzed for radiochemistry. The samples had been irradiated in the University of Virginia reactor for periods up to 1 year to simulate PWR conditions. The samples were irradiated as close as possible to the core face. A stainless steel sample of the surveillance capsule itself was also provided. That sample was composed on reactor grade type 304L stainless steel and is thus typical of reactor internals.

6.1.2 Concretes

Pathfinder

A 7.5 cm diameter core of the concrete floor directly beneath the pressure vessel was taken from the Pathfinder reactor (An Allis-Chalmers 58.5 MWe boiling water reactor which had operated for a 42 month period up to September 1967). The core was sliced into 1 cm sections for gamma counting. The lower section of the core was ground in a shatterbox for radiochemical analysis.

Humboldt Bay

Four samples were chipped from the top and bottom of the shield ring in the dry well surrounding the Humboldt bay reactor (a 65 MWe GE BWR which operated from 1963 until July 1, 1976).

Turkey Point No. 4

An extensive sampling progrm was carried out at the Turkey Point Generating Station in October 1981 during an outage for steam generator replacement. During that outage it was possible to gain access to the sump region directly beneath the pressure vessel on Unit 4 about 24 hr after shutdown. The Turkey Point Unit 4 Reactor is a 693 MWe Westinghouse PWR which has been in operation since 1973. In spite of high temperatures and high radiation fields in the sump area, it was possible to obtain two 7.5 cm diameter concrete cores from the sump floor. The samples were sliced into 1 cm sections for gamma counting and were also subjected to radiochemistry after grinding in a shatterbox.

6.2 ANALYTICAL METHODS

6.2.1 Nondestructive Gamma Assay

Concrete core samples were cut into 1 cm slabs using a rock saw with a diamond-impregnated cutting wheel. The individual slices were then repackaged in plastic and mounted directly on the face of a 100 cm^3 Ge(Li) diode for counting. The counter was standardized using a mockup prepared in an identical geometry and matched density. The mockup was made from a mixture of epoxy, powdered iron, and powdered aluminum doped with a mixture of NBS or NBS-traceable liquid radioisotope standards. Standards for ^{60}Co, ^{125}Sb, ^{137}Cs, ^{152,154,155}Eu and ^{207}Bi were included. An efficiency curve was prepared from those peaks using a regression fit.

Counting results were reduced using a standard peak identification, peak area routine with the efficiency curve used for this final quantification. All reported results have been decay corrected to the time of last known irradiation and corrected for a saturation factor corresponding to a 30 year irradiation.

6.2.2 Radiochemical Separations and Countings

Samples of steel and concrete were digested in hot hydrochloric acid. Sample size varied from 10 mg to 50 gm depending upon the specific activity of ^{60}Co in the sample. The solutions were filtered and diluted to constant volume. An aliquot was taken for gamma assay of ^{54}Mn, ^{60}Co, and ^{137}Cs. Aliquots of the remaining sample were subjected to radiochemical separations as described below.

6.2.2.1 ^{55}Fe Determination

Iron isotopes were separated from other radionuclides by anion exchange chromatography. Iron was electroplated onto a copper disc from an ammonium oxalate medium. Radiochemical yields were determined by tracing with ^{59}Fe. Iron-55 was measured using an intrinsic germanium diode with a 5 mil beryllium window to detect the characteristic manganese K X-rays.

6.2.2.2 ^{63}Ni and ^{59}Ni Determination

Nickel isotopes were separated from other radionuclides by precipitating with dimethyl-glyoxime in the presence of chelating agents and hold-back carriers. Nickel was electroplated onto a stainless steel disc from a sulfate medium. Chemical yields were determined by tracer technique using 2.52 hr ^{65}Ni. Nickel-63 was measured using an anti-coincident shielded windowless proportional beta counter. Nickel-59 was measured using an intrinsic germanium diode with a 5 mil beryllium window to detect the characteristic cobalt K X-rays.

6.2.2.3 ^{93}Mo Determination

Molybdenum isotopes were separated from other radionuclides by repeated scavenging with ferric hydroxide, ether extractions, and anion exchange chromatography. Molybdenum was electroplated onto a stainless steel disc from a basic sulfate medium. Radiochemical yields were determined by tracing with ^{99}Mo. Molybdenum-93 was measured using an intrinsic germanium diode with a 5 mil beryllium window to detect the characteristic niobium K X-rays.

6.2.2.4 ^{94}Nb Determination

Niobium was separated from other radionuclides by precipitation of niobic oxide from an acid medium during dissolution of the solid sample. Both niobium carrier and ^{95}Nb were present during dissolution. Niobium-94 was measured by gamma-ray spectrometric techniques and radiochemical yield determined by tracing with ^{95}Nb.

6.2.2.5 ^{99}Tc Determination

Technetium isotopes were separated from other radionuclides by scavenging methods and anion exchange chromatography. Technetium was electroplated from a basic ammonium sulfate medium. Technetium-99 was corrected for radiochemical yield by tracing with ^{95m}Tc. Technetium-99 was determined by beta absorption techniques using a windowless beta proportional counter that was anti-coincident shielded to reduce background.

6.2.2.6 Europium Isotopes

Europium radionuclides were separated along with other rare earth isotopes by precipitating with hydrofluoric acid in an acid medium using rare earth carriers and hold-back carrier. Radiochemical yields were determined by tracer techniques using ^{140}La. Europium-152, ^{154}Eu, and ^{155}Eu were measured by gamma-ray spectrometric techniques with a 100 cm^3 Ge(Li) diode.

6.3 RADIOCHEMICAL ANALYSIS RESULTS

6.3.1 Steels

The results of the radiochemical analysis of stainless steel samples are given in Table 6.1. Three of the samples are type 304L stainless steel, the material most typical of reactor internals, while the fourth is type 718 inconel. The inconel sample is very high in both stable nickel and niobium thus making it an ideal case for studying the production of the long-lived activation products from those elements, i.e., ^{59}Ni, ^{63}Ni, and ^{94}Nb. All of the results given in this section are corrected for decay to the end of irradiation and adjusted to an equivalent of 30 years of irradiation. Calculations were carried out using the previously described computer codes combined with the individual sample compositions as measured by X-ray fluorescence. For the Monticello sample, the flux calculation for BWR vessel cladding was used, while for the other three, conditions typical of a PWR shroud were used. In all cases this procedure resulted in a significant overestimate of the thermal flux. In order to have a point of reference for the actual neutron received by the samples, all calculations were normalized to the measured ^{55}Fe in each sample. It should be noted that this will mainly serve to scale the thermal neutron flux, since ^{55}Fe is produced primarily by thermal neutron capture. Normalizing to ^{55}Fe results in overall agreement to within a factor of two in most cases. In view of the less than ideal sampling conditions, this can be considered to be reasonably good agreement. A few curious anomalies did appear, however. In particular, the ^{94}Nb measurement from the Point Beach inconel sample is considerably higher than expected. The SS304L sample from the same area is also a bit higher in ^{94}Nb than expected. This could suggest that the cross-section used in the calculation is in error, however, a more likely explanation lies in the details of the flux distribution. Those two samples came from the core of the reactor where there is a particularly high epithermal neutron component, and a significantly lower cadmium ratio than in the shroud. Niobium-94 does have a substantial resonance contribution to its production rate. It is worth noting that ^{93}Mo, which is produced almost entirely by resonance capture is also underpredicted in the Point Beach samples and overpredicted in the samples from the University of Virginia and the Monticello reactors. Clearly, better documented samples are required to obtain more accurate results. Agreement between the measured and calculated activities for ^{54}Mn, and ^{63}Ni was rather poor for the Monticello core spray sparger pin sample. The reason for the disagreement is not apparent.

The equivalent results for vessel steels is given in Table 6.2. All four samples were of similar composition irradiated under similar though not identical conditions in a research reactor. Good agreement was obtained for ^{54}Mn, ^{60}Co, and ^{63}Ni. Molybdenum-93 is in all four cases overpredicted by up to a factor of four presumably due to the reasons previously discussed. It was

TABLE 6.1. Irradiated Stainless Steel - Measured Versus Calculated Activity in μCi/gm

	SS-TAP (SS304L)		Monticello (SS304L)		Point Beach (SS304L)		Point Beach (Inconel)	
	Measured$^{(1)}$	Calculated$^{(2)}$	Measured$^{(1)}$	Calculated$^{(3)}$	Measured$^{(1)}$	Calculated$^{(2)}$	Measured$^{(1)}$	Calculated$^{(2)}$
^{54}Mn	640	826	0.020	0.0070	876	662	162	114
^{53}Fe	26000	≡26000	0.145	≡0.145	20800	≡20800	3580	≡3580
^{59}Ni	<22	140		<0.029	25.0	11.9	40.9	42
^{60}Co	8970	15200	0.014	0.083	8310	12300	722	
^{63}Ni	1260	2130	0.049	0.013	1290	1980	4160	6570
^{93}Mo	0.038	0.15	<0.006	2.4E-7	0.15	0.086	1.13	0.84
^{94}Nb	<0.003	0.017	<0.0047	1.4E-7	0.40	0.28	56.8	18.5
^{99}Tc	1.28	0.020						
^{110m}Ag	1.0							
^{151}Sm	<0.6							
^{152}Eu	<0.3				<0.05	0	<0.05	
^{154}Eu	<0.2		<0.00035		0.79	0.034	2.5	
^{155}Eu	<0.04		<0.00028		0.50	0.024	1.2	

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(1) Corrected to end of irradiation and 30 EFPY
(2) Normalized to measured ^{55}Fe. Based on measured composition and flux conditions for PWR shroud
(3) Normalized to measured ^{55}Fe. Based on measured composition and flux conditions for BWR vessel cladding.

TABLE 6.2. Irradiated Vessel Steels - Measured Versus Calculated Activity in μCi/gm

	P-21 (A508)		P-34 (A533)		GP-55 (A533)		P-147 (A533)	
	Measured(1)	Calculated(2)	Measured(1)	Calculated(2)	Measured(1)	Calculated(2)	Measured(1)	Calculated(2)
$^{54}\mathrm{Mn}$	139	1360	1212	1740	673	442	686	518
$^{55}\mathrm{Fe}$	42800	≡42800	54800	≡54800	13900	≡13900	16300	≡16300
$^{59}\mathrm{Ni}$	<1	1.2	<0.8	1.3	<1.1	0.36	<0.5	0.41
$^{60}\mathrm{Co}$	1320	1540	1430	1990	485	506	486	593
$^{63}\mathrm{Ni}$	258	199	155	213	46.2	59	49	67
$^{93}\mathrm{Mo}$	0.11	0.31	0.10	0.39	0.071	0.096	0.058	0.12
$^{94}\mathrm{Nb}$	<0.006	0.007	<0.02	0.0087	<0.015	0.0044	<0.007	0.005
$^{99}\mathrm{Tc}$	6.5	0.043	16.5	0.054	8.6	0.013	1.91	0.016
$^{110m}\mathrm{Ag}$	0.040		0.64		0.19		0.55	
$^{151}\mathrm{Sm}$	<0.1		<0.12		<0.15		<0.1	
$^{152}\mathrm{Eu}$	<0.05		<0.05		<0.04		<0.02	
$^{154}\mathrm{Eu}$	<0.03		<0.03		<0.02		<0.1	
$^{155}\mathrm{Eu}$	<0.003		<0.003		<0.003		<0.006	

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(1) Corrected to end of irradiation and 30 EFPY

(2) Normalized to measured ^{55}Fe. Based on measured composition and flux conditions for PWR shroud

not possible to measure ^{59}Ni or ^{94}Nb in these samples with adequate sensitivity to check the model for those isotopes. All of the samples were rather high in ^{99}Tc. This is evidently some type of contamination since it is clearly too high to be the result of direct activation.

6.3.2 Concretes

Radiochemical analyses were carried out on a series of concrete samples obtained at the Pathfinder, Humboldt Bay, and Turkey Point No. 4 reactors. Data for the concretes was treated in the same manner as previously described for the steels with the exception that ^{152}Eu was used for neutron flux normalization rather than ^{55}Fe. None of the samples available for analysis were really fully satisfactory. Since access to the more highly activated regions of the bioshield was not possible at any of the sites sampled, it was necessary to obtain samples from regions receiving highly attenuated neutron fluxes. In addition to the fact that detection limits become a serious problem experimentally, contamination by translocation products can be dominant for some isotopes, particularly near the surface of the concrete. This is illustrated in Table 6.3 and Figure 6.1 which plots the depth profile of the cobalt and europium isotopes in a concrete core taken directly beneath the reactor vessel at the Pathfinder reactor. The europium is clearly a pure activation product while the ^{60}Co near the surface is obviously a translocated contaminant. The ^{60}Co activity appears to be leveling off near the pure activation level, but does not quite reach it. The core is also seen from Table 6.4 to be contaminated with ^{55}Fe even 8 centimeters beneath the surface. This situation is even more obvious for the data on the Humboldt Bay samples as given in Tables 6.5 and 6.6. While the two europium isotopes show good internal consistency as

TABLE 6.3. Pathfinder Concrete Core PCC-8 Directly Beneath Vessel Activity in pCi/gm Corrected to Shutdown and 30 EFPY

Depth (cm)	^{60}Co	^{63}Ni	^{152}Eu	^{154}Eu
0-1	3200		5.93	1.20
1-2	75.0		5.84	1.14
2-3	39.1		6.37	1.30
3-4	40.2		6.68	1.14
4-5	21.4		6.68	1.09
5-6	21.8		8.46	1.35
6-7	16.5		7.84	1.35
7-8	17.6		8.24	1.40
8-9	14.6	<400	7.84	1.35

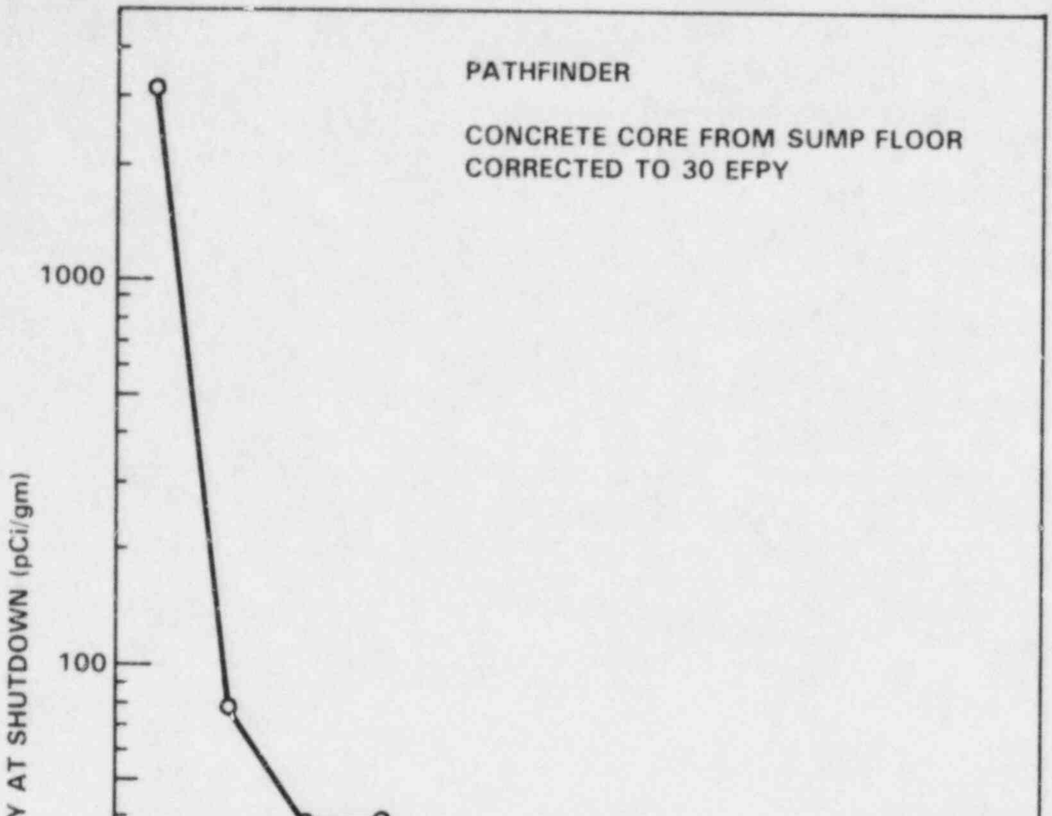

FIGURE 6.1. Depth Profile of Radionuclides in Concrete Core Taken from Sump Floor at Pathfinder

TABLE 6.4. Pathfinder Concrete Core PCC-8 (8-9 cm) Comparison of Measured and Calculated Activities

	Measured (pCi/gm)	Calculated (pCi/gm)	Measured/ Calculated
^{55}Fe	16000	73	220
^{60}Co	14.6	3.7	4.0
^{63}Ni	<400	0.2	
^{152}Eu	7.84	≡7.84	≡1
^{154}Eu	1.35	0.85	1.6

TABLE 6.5. Humboldt Bay Concrete Bioshield Samples Activity at Shutdown and 30 EFPY (pCi/gm)

	^{54}Mn	^{55}Fe	^{60}Co	^{63}Ni	^{125}Sb	^{134}Cs	^{137}Cs	^{152}Eu	^{154}Eu	^{155}Eu
HBRS-22 Bottom Inside Edge	42.7		378		0.45	8.2	199	188	11.6	2.2
HBRS-23 Bottom Inside Edge	25.7		821		1.67	30.1	475	309	18.1	3.61
HBRS-24 Top Outside Edge	39.7	7380	303	55	1.41	51.0	544	63	4.9	0.80
HBRS-25 Top Inside Edge	176	16600	1308	41	<0.7	153	72	293	31.6	5.3

pure activation products, everything else appears to be contamination. The best samples obtained were those taken beneath the pressure vessel at the Turkey Point No. 4 reactor. Since these samples were taken immediately after shutdown it was possible to measure a number of shorter-lived activation products in addition to the rather limited list of long-lived isotopes detectable. Two cores were analyzed with the saturation and decay corrected results given in Tables 6.7 and 6.8. The data for core TP-1 is plotted as a function of depth in Figure 6.2. Most of the isotopes show only shallow penetration of contamination from the surface. A notable exception is ^{137}Cs which exhibits a much less rapid fall off with depth, with considerable contamination present in

TABLE 6.6. Humboldt Bay Concrete from Upper Shield Ring HBRS-25 Comparison of Measured and Calculated Activities

	Measured (pCi/gm)	Calculated (pCi/gm)	Measured/ Calculated
^{54}Mn	176	8.5	21
^{55}Fe	16600	2080	8.0
^{60}Co	1308	233	5.6
^{63}Ni	41	2.9	14
^{134}Cs	153	9	17
^{152}Eu	293	≡293	≡1.0
^{154}Eu	31.6	31.7	1.0
^{155}Eu	5.3	0.7	7.6

even the deepest sample. Table 6.9 gives a comparison with the activation model for the deepest point in core TP-I. Agreement is excellent for most of the isotopes.

6.3.3 Conclusions

Within the framework of the samples available, only a limited verification of the predictive capability was possible. In the steel samples, ^{59}Ni, ^{63}Ni, and ^{93}Mo were found to be present at approximately the expected levels based on a neutron flux normalization. The presence of ^{94}Nb in stainless steel samples was verified at levels somewhat higher than expected. The discrepancy appears to be attributable to epithermal capture in the core region for those particular samples. For the concrete samples, the dominant role of the europium isotopes was verified, with good agreement between measurement and calculation obtained for a number of shorter-lived isotopes. It was not possible to determine ^{41}Ca with the available techniques on the rather low level samples obtainable at this time.

TABLE 6.7. Turkey Point Unit 4 Concrete Core Taken Directly Beneath Vessel Activity at Shutdown and 30 EFPY (pCi/gm)

	0-1	1-2	2-3	3-4	4-5	5-6	6-7
$^{46}\mathrm{Sc}$	1001	1350	1390	1380	1360	1450	1300
$^{51}\mathrm{Cr}$	8140	4830	1835	626	580	440	450
$^{54}\mathrm{Mn}$	76	3.1	3.1	4.0	1.8	<1.8	<1.8
$^{55}\mathrm{Fe}$	12800	14500	14500				18675
$^{58}\mathrm{Co}$	165	19	22	19	17	23	25
$^{59}\mathrm{Fe}$	670	780	820	780	780	810	720
$^{60}\mathrm{Co}$	10100	1700	1400	1420	1430	1540	1320
$^{63}\mathrm{Ni}$							<16
$^{65}\mathrm{Zn}$	630	61	53	54	59	52	50
$^{94}\mathrm{Nb}$	<3	<2	<2	<2	<2	<2	<2
$^{108m}\mathrm{Ag}$	<3	<2	<2	<2	<2	<2	<2
$^{110m}\mathrm{Ag}$	13	<5	<5	<5	<5	<5	<5
$^{124}\mathrm{Sb}$	49	20	19	17	12	8.5	12.5
$^{134}\mathrm{Cs}$	500	188	116	85	80	72	58
$^{137}\mathrm{Cs}$	6550	1610	772	471	353	224	133
$^{141}\mathrm{Ce}$	63	98	85	80	80	89	54
$^{152}\mathrm{Eu}$	2490	4010	4170	4090	3860	4210	3670
$^{154}\mathrm{Eu}$	346	470	470	390	370	440	350
$^{155}\mathrm{Eu}$	<13	<42	<30	<27	<27	<33	<33
$^{166m}\mathrm{Ho}$	<6	<20	<14	<13	<13	<16	<16

TABLE 6.8. Turkey Point Unit 4 Concrete Core Taken Directly Beneath Vessel (TR-J) Activity at Shutdown and 30 EFPY (pCi/gm)

	0-1	1-2	2-3	3-4	4-5	5-6	6-7	7-8
^{46}Sc	1024	1266	1323	1560	1190	2203	1502	1703
^{51}Cr	3667	1390	760	890	580	670	490	850
^{54}Mn	49	<2	2.7	<2	7.6	12.5	8.9	<1.8
^{58}Co	134	19	25	26	12	23	21	36
^{59}Fe	680	740	780	890	2120	4510	2850	950
^{60}Co	8090	1590	1540	1708	3300	6226	4310	1710
^{65}Zn	2420	79	83	83	80	84	84	100
^{94}Nb	<1.7	<0.6	<0.6	<0.6	<0.6	<1.0	<1.0	<0.6
^{108m}Ag	<0.8	<0.6	<0.6	<0.6	<1.1	<0.6	<0.6	<0.6
^{110m}Ag	<10	<8	<5	<7	<7	<9	<7	<5
^{124}Sb	72	31	21	22	12	17	17	22
^{134}Cs	530	115	77	86	53	57	67	67
^{137}Cs	3190	165	80	76	32	28	35	27
^{141}Ce	54	63	67	38	76	103	98	72
^{152}Eu	3041	4440	4080	4870	3400	3730	4370	5280
^{154}Eu	810	1100	950	1170	740	775	980	1100
^{166m}Ho	<13	<20	<23	<20	<23	<33	<10	<15

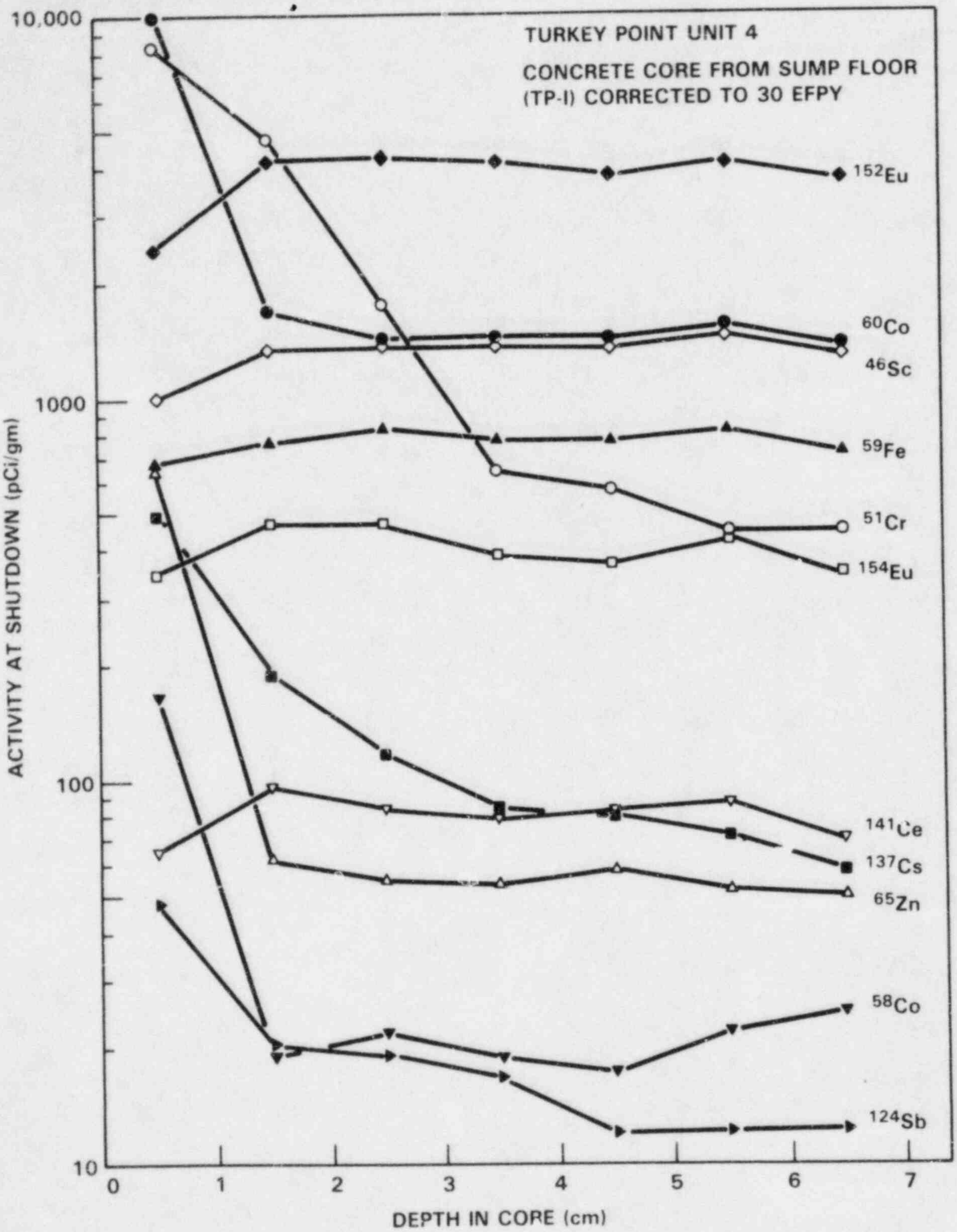

FIGURE 6.2. Depth Profile of Radionuclides in Concrete Core Taken from Sump Floor at Turkey Point No. 4

TABLE 6.9. Turkey Point Unit 4 Concrete Core TP-I Comparison of Measured and Calculated Activities

	Measured(a) (pCi/gm)	Calculated(b) (pCi/gm)	Measured / Calculated
^{46}Sc	1300	1000	1.3
^{51}Cr	450	570	0.8
^{54}Mn	<2	7	<0.3
^{55}Fe	18675	27900	0.7
^{58}Co	25	<3	>8.3
^{59}Fe	720	780	0.9
^{60}Co	1320	1870	0.7
^{63}Ni	<16	<60	
^{65}Zn	50	130	0.4
^{94}Nb	<2	0.11	<18
^{104m}Ag	<2	<0.15	--
^{110m}Ag	<5	<28	--
^{124}Sb	12.5	26	0.5
^{134}Cs	58	295	0.2
^{137}Cs	133	5.7	23
^{141}Ce	54	63	0.9
^{152}Eu	3670	≡3760	≡1.00
^{154}Eu	350	448	0.8
^{155}Eu	<33	18	<1.8
^{166m}Ho	<16	--	--

(a) Activity corrected to reactor shutdown and 30 EFPY. Deepest point in core used.

(b) Calculation based on measured composition of core. PWR flux at 10 cm depth used. All data normalized to measured ^{152}Eu in core.

7.0 EFFECT ON DECOMMISSIONING STRATEGIES

This program has taken a more detailed look at the question of nuclear activation of reactor construction components. A major goal of the program was to determine if there are any significant reaction pathways and product isotopes which could have a major impact on decomissioning and disposal strategies. It is clear from this work that there should be no real surprises provided relatively ordinary materials have been used for construction. The range of neutron activation expected falls within that considered in the Smith, Konzek, and Kennedy (1978) and Oak et al. (1980) assessments and their conclusions remain valid. Certainly, the decommissioning problem could have been somewhat alleviated by careful selection of materials during construction. Cobalt levels in stainless steel were found to vary in the extreme by more than a factor of ten for example. Selection of materials may not, however, be practical due to the large quantities of steel involved. High cobalt steels are also required in some critical reactor applications due to hardness requirements.

8.0 RECOMMENDATIONS FOR FUTURE STUDIES

It is clear from the assessment done in this study that a good predictive capability exists for determining expected levels of activation products present in light water reactors. Neutron capture cross-sections are reliably available for all important isotopes of interest. The neutron flux distribution is, however, complex particularly within the high flux region and there is a resulting large uncertainty in the calculation of activation levels. Furthermore, activatable trace element levels were found to vary over more than an order of magnitude for several key trace elements including cobalt and niobium. It was not possible within the framework of this program to carry out a full verification of the predictive capability since samples were not, in general, available from the appropriate reactor regions. This type of sampling will only realistically be possible during an actual full scale reactor decommissioning experience. Carefully documented samples should be taken at all points in the decommissioning operation. A sampling plan should in fact be included as part of the predecommissioning planning. For many years to come, each project undertaken will represent a rare opportunity to obtain complete information about the neutron activated components of the reactor system. Sampling should include bioshield cores at several locations, vessel wall sections including cladding, and various samples of reactor internals including control rods. All samples, particularly the internals, should be subsampled onsite to reduce them to a manageable radiation level for analysis. These samples should also be fully documented and archived for future use.

It is important that this be done as part of the dismantling operation in order to take full advantage of the handling facility and decontamination controls in place at that time. Analysis of the samples should include both chemical and radiochemical analysis. This is a rather formidable problem for the highly radioactive components. Energy dispersive, X-ray fluorescence analysis was found to be very successful for analyzing even highly radioactive samples. With the addition of wavelength dispersive analysis, a rather complete chemical characterization can be done fully nondestructively. Finally, neutron flux calculations should be carried out for the reactor under study with activity levels predicted in the manner of the present work.

Control rods pose a special problem, since their complex geometries and high neutron opacities render the calculation of activation inventories impractical. A detailed experimental study is suggested. This would involve sectioning the control rods to define production gradients with samples taken for radiochemical analysis. Isotopes analyzed should include ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni, and ^{94}Nb in the cladding. Analysis of PWR control rods should include ^{110m}Ag and ^{108m}Ag. BWR control rods should be assayed for ^{14}C and ^{10}Be. This program need not be carried out in conjunction with a decomissioning operation, since control rods are periodically charged out during routine operations. It will, however, require special handling facilities capable of subsampling highly radioactive materials in complex geometries.

9.0 REFERENCES

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APPENDIX A

CODE DESCRIPTION

APPENDIX A

CODE DESCRIPTION

GENERAL INFORMATION

A group of computer programs was written to calculate expected production rates of long-lived activation products on an isotope by isotope basis. A description of the calculation method is given in Section 5.1. The code is highly interactive, simple to use, and runs on widely available minicomputers. It would be suitable for use by utilities for developing procedures for compliance with 10 CFR 61. A simplified version, including only those isotopes of major importance, would be most useful in that context since many of the isotopes considered here are of negligible importance and are in any case not covered by the regulatory guidelines.

The computer code is written in FORTRAN IV. It is designed for the DEC family of computers, but could easily be adapted to many other systems with minor changes in file management. All of the programs are run interactively with results written to a file, rather than directly to a printer. All of the data generated in this report were run on a VAX 11/780, although the programs were originally developed on a PDP 11/34 using an RSX 11/M operating system with only 32K of memory available for the program. No overlaying was used. Conversion to the VAX was trivial with computing speed greatly improved. This extra speed proved useful, though not essential. The iterative method used in the calculation, while requiring very little memory, is time consuming. The following programs were used:

NUCLIB - interactively creates the nuclear data libraries. Five different types of libraries can be formed including N-gamma only, N-2N, N-P, N-D, and N-alpha.

DATDMP - used to list a nuclear data library of interest. It uses a single subroutine, DATBAS, for most of the formatting and writes the results in an easily readable form on a file named PRINT.LIB.

COMPOS - used to create composition files for each material type of interest. Fifty-three elements are included. The file ELEMENT.DAT is used as the basic template. It contains the element names, atomic numbers and atomic weights. The name of the composition file for output is specified interactively at program initiation.

FLUX1 - used to create a file of neutron flux information and reactor conditions including thermal flux, epithermal flux, fast flux in 0.5 MeV increments from 0.5 to 15 MeV, and reactor temperature. The name of the flux file is specified interactively by the operator at program initiation. An easily readable reformatted version is also automatically written to a file called PRINT.FLX, which can be hard copied as a permanent record. FLXDMP can also be used to list the flux file. It is output on PRINT.FLX.

ACTIV - used for the actual activation calculation using the nuclear data libraries, the composition and flux files and five batch files containing the isotope names for use with each library. Subroutines used include NCAP, DMP, FINAL, and COMBAS. The batch files are created and named using the system editor. The results are written on a file named PRINT.ACT.

Hard copy results from any of the above are obtained simply by using the appropriate system COPY commands.

CODE USE

NUCLIB

The program will first ask "File for nuclear data library?." This will be the output file and must be a legal DEC file name plus extension. For input into ACTIV, the file name must be one of the following:

NUCLIB0.LIB	N-gamma only
NUCLIB1.LIB	N-2N
NUCLIB2.LIB	N-P
NUCLIB3.LIB	N-D
NUCLIB4.LIB	N-alpha.

A different file name can be used initially and renamed or concatenated with a larger piece of the file later. Ultimately, there should only be one version of each on the disk to avoid losing the main libraries during general file purges. The program will next ask for the reaction type with a menu of choices displayed. All entries in that particular library will then be assigned that reaction type automatically. Next, the product element and isotope mass will be requested. The isotope mass is read as an I3 format, so it is necessary to include a leading zero or blank for two digit entries. It will then request the number of elements in the chain. This allows for multiple neutron capture and beta decay considerations. This allows considerable flexibility in assembling the library. Some judgment is required based on an examination of the chart of the nuclides and the available data. For N-γ or N-2N reactions, in most cases, a single element is all that is required. For charge particle reactions, such as N-P, at least two elements are needed. The program can handle up to nine elements in a chain. The atomic number (Z) of the product element is requested. The program will then request the number of isotopes at the Z corresponding to the first element in the chain, and the mass of that isotope. It will then step its way up the chain asking at each point for isotopic abundance, fission yield, half-life, production cross section, production resonance integral, depletion cross section, and depletion resonance integral. This process will continue for each isotope in the chain. In most cases, a simple carriage return (i.e., no data) will insert a zero. For the half-life, a carriage return sets the half-life to 1E31 years, a computational approximation to infinity. This is intended for stable isotopes, and is a somewhat whimsical choice based on the limits on the nucleon lifetime set by a number of proton decay experiments recently. A carriage return for a resonance integral will set it equal to 0.45 times the thermal cross section. That is simply the 1/V value with no resonances and, as such, is the theoretical minimum. Following all of the above entries, the program will then request entries for the high energy excitation function. For the N-gamma only library (reaction type 0), that step is omitted and it loops directly back to the start. A new isotopic entry can be started or a control Z entered to close the file and exit the program. In its present form, ACTIV requires that all five library types exist since it automatically steps through them. This could easily be modified.

Some ingenuity is needed in constructing the libraries. For example, if one wishes to cross compare relative yields by direct neutron capture and N-2N, the same isotope can be placed in both libraries with the direct capture cross sections set to zero in the N-2N library. Tritium and carbon-14 are computed in the N-γ only library because the cross-section data is typically available in that form. Tritium, for example, is simply entered into the library like an N-γ reaction on lithium. A listing of the program follows.

DATDMP

This program produces a listing of any of the nuclear data libraries in a conveniently readable format. It simply requests the file name and produces the result on PRINT.LIB for later inspection or hard copy. A listing of the program and the subroutine DATBAS follows.

COMPOS

This is used to create a composition file for 53 elements. The file ELEMENT.DAT is used to provide element names, atomic numbers, and atomic weights. The program first requests the sample type. This can be up to

72 characters. The file name is requested. This should be in some conveniently identifiable format (i.e., for type 304 stainless steel, a file name SS304.CMP would be appropriate). The program then steps through the elements in sequence displaying each on the screen and requesting its concentration in parts per million. A carriage return will enter a zero and flag it as not measured. An entry will be followed by a request asking whether it is an upper limit (Y or N). Some radioactive elements, such as plutonium, are included since the output file is also used by ACTIV to provide atomic weights. A listing of COMPOS and ELEMENT.DAT follows.

FLUX1

This program creates a a file for a specific set of reactor conditions. It first requests a file name for the output file. A .FLX extension is useful for keeping track of file types. The thermal flux, epithermal flux, and fast flux in 29 groups from 0.5 to 15 MeV are then requested. The temperature is requested followed by a request for reactor conditions. The latter is simply an up to 72 character description of the conditions represented by the fluxes used. A reformatted copy of the data is automatically output to PRINT.FLX. A listing of FLUX1 follows.

FLXDMP

This is an alternative method for listing a flux file in a conveniently readable format. The program simply requests the file name and lists the results on PRINT.FLX. A listing of the program and some sample output follow.

Batch Run Files

Five batch run files are required to provide the names of isotopes to be calculated by the five libraries. They are paired as follows:

BATCH0.LIB - NUCLIB0.LIB
BATCH1.LIB - NUCLIB1.LIB
BATCH2.LIB - NUCLIB2.LIB
BATCH3.LIB - NUCLIB3.LIB
BATCH4.LIB - NUCLIB4.LIB.

The isotope names given in the batch files must match the isotopes entered into the appropriate library. ACTIV will automatically search the NUCLIB0.LIB file for each isotope listed in BATCH0.LIB, etc. If the isotope is not present, a warning will appear on the screen, and it will continue with the next isotope. The files are created using the standard DEC EDITOR. There are no blanks proceeding the element name and it must be spelled correctly. The isotope mass must be in an I3 format with the correct number of leading blanks or zeros. An example of the five batch files follows. Please note that the file name is included only for information. It should not actually be imbedded in the file.

ACTIV

The activation calculation is carried out by a numerical integration method using the files created by NUCLIB, COMPOS, and FLUX1 together with a batch file created by the EDITOR specifying which isotopes to compute. The main program uses repeated calls to subroutine NCAP to determine incremental changes in the number of atoms of each isotope in the chain. Subroutine DMP is used for printing results in 1-year increments. FINAL keeps track of the run time for each isotope and COMBAS produces a listing of the composition file at the end. The program first request the name of the composition file and opens that file. The sample type is read and printed on the screen. It then opens an output file. Results will appear on PRINT.ACT. Next, the name of the flux file is requested, the file opened, and the data read. No further operator input is needed. The program will open the five-batch run files in sequence and compute the results for each isotope. Results are written to file PRINT.ACT. As each isotope is finished, a message will appear on the screen giving the time consumed for that isotope. This is useful for benchmarking and, on slower machines, keeping track of the progress of the calculation. After all computing is done, a listing of the composition file is made on PRINT.ACT, all files closed, and execution terminated. Listings of the ACTIV, NCAP, DMP, FINAL, and COMBAS are found in Appendix B.

APPENDIX B

PROGRAM LISTINGS

```
      PROGRAM NUCLIB
C     CREATES NUCLEAR DATA LIBRARIES FOR ACTIV
C     WRITTEN BY J. C. EVANS/ BATTELLE N. W. LABORATORY
C     DEC VAX 11/780 VERSION
C
C
C
      DIMENSION ELNAME(3),SIGF(29),FILE4(5)
      INTEGER Z,ZZ
 1100 TYPE 51
   51 FORMAT(1X,'FILE FOR NUCLEAR DATA LIBRARY')
      READ(5,52,ERR=1100)FILE4
   52 FORMAT(5A4)
      CALL ASSIGN (1,FILE4)
 1102 TYPE 40
   40 FORMAT(1X,'REACTION TYPE?')
      TYPE 41
   41 FORMAT(1X,'0=N-GAMMA ONLY')
      TYPE 42
   42 FORMAT(1X,'1=N-2N')
      TYPE 43
   43 FORMAT(1X,'2=N-P')
      TYPE 44
   44 FORMAT(1X,'3=N-D')
      TYPE 45
   45 FORMAT(1X,'4=N-ALPHA')
      READ(5,7,ERR=1102)ITYPE
  400 TYPE 1
  1   FORMAT(1X,'PRODUCT ELEMENT (CONTROL Z TO TERMINATE)?')
      READ(5,2,END=999,ERR=400)ELNAME
  2   FORMAT(3A4)
 1101 TYPE 3
  3   FORMAT(1X,'ISOTOPE MASS?')
      READ(5,4,ERR=1101)M
  4   FORMAT(I3)
      WRITE(1,5)ELNAME,M,ITYPE
  5   FORMAT(1X,3A4,I3,10X,I1)
 1103 TYPE 6
  6   FORMAT(1X,'HOW MANY ELEMENTS IN THE CHAIN?(1-9)')
      READ(5,7,ERR=1103)N
  7   FORMAT(I1)
 1104 TYPE 60
  60  FORMAT(1X,' PRODUCT ELEMENT Z=?')
      READ(5,70,ERR=1104)Z
  70  FORMAT(I2)
      WRITE(1,8)N,Z
  8   FORMAT(1X,I1,10X,I2)
      DO 100 I=1,N
      ZZ=Z-N+I
 1105 TYPE 9,ZZ
```

```
  9     FORMAT(1X,'HOW MANY ISOTOPES AT Z=',I2,1X,' (1-9)?')
        READ(5,7,ERR=1105)J
 1106   TYPE 10
  10    FORMAT(1X,'MASS OF FIRST ISOTOPE?')
        READ(5,4,ERR=1106)MM
        WRITE(1,80)J,MM
   80   FORMAT(1X,I1,10X,I3)
        DO 200 K=1,J
        M=MM+K-1
 1107   TYPE 13,M
  13    FORMAT(1X,'ABUNDANCE OF ',1X,I3,' ?')
        READ(5,14,ERR=1107)ABUN
  14    FORMAT(F7.4)
 1108   TYPE 15,M
  15    FORMAT(1X,'FISSION YIELD OF ',I3,' ?')
        READ(5,14,ERR=1108)FYIELD
        WRITE(1,17)ABUN,FYIELD
  17    FORMAT(1X,F7.4,10X,F7.4)
 1109   TYPE 18
  18    FORMAT(1X,'HALF-LIFE OF ISOTOPE IN YEARS(CR IF STABLE)')
        READ(5,12,ERR=1109)THALF
  12    FORMAT(E8.2)
        IF(THALF.EQ.0.)THALF=1.E31
        WRITE(1,31)THALF
  31    FORMAT(1X,E8.2)
 1111   TYPE 20
  20    FORMAT(1X,'PRODUCTION CROSS-SECTION ?')
        READ(5,12,ERR=1111)SIGMAT
 1112   TYPE 21
  21    FORMAT(1X,'PRODUCTION RESONANCE INTEGRAL ?')
        READ(5,12,ERR=1112)SIGMAF
        IF(SIGMAF.EQ.0.)SIGMAF=.45*SIGMAT
 1113   TYPE 11
  11    FORMAT(1X,'DEPLETION CROSS-SECTION ?')
        READ(5,12,ERR=1113)DEPLT
 1114   TYPE 101
 101    FORMAT(1X,'DEPLETION RESONANCE INTEGRAL ?')
        READ(5,12,ERR=1114)DEPLF
        IF(DEPLF.EQ.0.)DEPLF=.45*DEPLT
        WRITE (1,22)SIGMAT,SIGMAF,DEPLT,DEPLF
  22    FORMAT(1X,E8.2,3(10X,E8.2))
 200    CONTINUE
 100    CONTINUE
        IF(ITYPE.EQ.0)GO TO 400
        TYPE 46
   46   FORMAT(1X,'ENTER FAST CROSS-SECTION AT EACH ENERGY')
        DO 600 LL=1,29
        E=(FLOAT(LL)/2.)+.5
 1115   TYPE 47,E
   47   FORMAT(1X,F5.1,1X,'MEV=')
```

```
      READ(5,12,ERR=1115)SIGF(LL)
  600 CONTINUE
      WRITE(1,23)(SIGF(LL),LL=1,10)
      WRITE(1,23)(SIGF(LL),LL=11,20)
      WRITE(1,24)(SIGF(LL),LL=21,29)
   23 FORMAT(10E8.2)
   24 FORMAT(9E8.2)
      GO TO 400
 999  CALL CLOSE(1)
      STOP
      END
```

```
      PROGRAM DATDMP
C     LISTS NUCLEAR DATA LIBRARIES TO A FILE PRINT.LIB
C     WRITTEN BY J. C. EVANS/ BATTELLE N.W. LABORATORIES
C     DEC VAX 11/730 VERSION
C
C
C
      BYTE DUM(12)
      DIMENSION FILE1(5)
      EQUIVALENCE(FILE1,DUM)
      CALL ASSIGN(2,'PRINT.LIB')
      TYPE 1
  1   FORMAT(1X,'FILE FOR NUCLEAR DATA LIBRARY')
      ACCEPT 2,DUM
  2   FORMAT(12A1)
      CALL ASSIGN(3,FILE1)
      CALL DATBAS
      CALL CLOSE(2)
      CALL CLOSE(3)
      STOP  'DATA IS ON PRINT.LIB'
      END
```

```
      SUBROUTINE DATBAS
C     MAIN SUBROUTINE FOR DATDMP
C
      DIMENSION ELNAME(3),SIGF(29)
      INTEGER Z,ZZ
  400 READ(3,1,END=999)ELNAME,M,ITYPE
    1 FORMAT(1X,3A4,I3,10X,I1)
      WRITE(2,2)ELNAME,M
    2 FORMAT(1H1,' NUCLEAR PARAMETERS FOR ',3A4,I3)
      READ(3,3)N,Z
    3 FORMAT(1X,I1,10X,I2)
      WRITE(2,4)N
    4 FORMAT(1H0,I2,1X,'ELEMENTS IN PRODUCTION CHAIN',//)
      DO 100 I=1,N
      ZZ=Z-N+I
      WRITE(2,5)ZZ
    5 FORMAT(2X,'DATA FOR Z=',I2)
      READ(3,6)J,MM
    6 FORMAT(1X,I1,10X,I3)
      WRITE(2,7)
    7 FORMAT(3X,'A',5X,'ABUN',5X,'FYIELD',5X,'THALF(Y)',5X,
     1'SIGMAT',5X,'SIGMAF',7X,'DEPLT',7X,'DEPLF')
      DO 200 K=1,J
      M=MM+K-1
      READ(3,8)ABUN,FYIELD
    8 FORMAT(1X,F7.4,10X,F7.4)
      READ(3,12)THALF
   12 FORMAT(1X,E8.2)
      READ(3,9)SIGMAT,SIGMAF,DEPLT,DEPLF
    9 FORMAT(1X,E8.2,3(10X,E8.2))
      WRITE(2,10)M,ABUN,FYIELD,THALF,SIGMAT,SIGMAF,DEPLT,DEPLF
   10 FORMAT(1X,I3,2X,F7.4,4X,F7.4,5X,1PE8.2,3X,0PF8.2,3X,F8.2,4X,
     1F8.2,4X,F8.2)
  200 CONTINUE
      WRITE(2,11)
   11 FORMAT(1H0)
  100 CONTINUE
      IF(ITYPE.EQ.0)GO TO 400
      IF(ITYPE.EQ.1)WRITE(2,13)
      IF(ITYPE.EQ.2)WRITE(2,14)
      IF(ITYPE.EQ.3)WRITE(2,15)
      IF(ITYPE.EQ.4)WRITE(2,16)
   13 FORMAT(1H0,1X,'CROSS-SECTIONS FOR N-2N REACTION')
   14 FORMAT(1H0,1X,'CROSS-SECTIONS FOR N-P REACTION')
   15 FORMAT(1H0,1X,'CROSS-SECTIONS FOR N-D REACTION')
   16 FORMAT(1H0,1X,'CROSS-SECTIONS FOR N-ALPHA REACTION')
      WRITE(2,17)
   17 FORMAT(1H0,5X,'E(MEV)',10X,'SIGMA')
      READ(3,18)(SIGF(LL),LL=1,10)
   18 FORMAT(10E8.2)
```

```
      READ(3,18)(SIGF(LL),LL=11,20)
      READ(3,19)(SIGF(LL),LL=21,29)
 19   FORMAT(9E8.2)
      DO 500 LL=1,29
      E=(FLOAT(LL)/2.)+.5
      WRITE(2,20)E,SIGF(LL)
 20   FORMAT(7X,F5.2,7X,1PE8.2)
500   CONTINUE
      GO TO 400
999   CONTINUE
      RETURN
      END
```

```
      PROGRAM COMPOS
C     CREATES A COMPOSITION FILE FOR ACTIV
C     WRITTEN BY J. C. EVANS/ BATTELLE N. W. LABORATORIES
C     DEC VAX 11/780 VERSION
C     REQUIRES FILE ELEMENT.DAT
C
C
C
      LOGICAL ELNAME(53,12)
      LOGICAL*1 ANS,YES
      BYTE DUM1(8)
      INTEGER Z(53)
      DIMENSION SAMP(18),CMPOS(5)
      DIMENSION A(53)
      DIMENSION FTNOTE(12),UP(12),UNDON(12)
      EQUIVALENCE(DUM1,CMPOS)
      DATA YES/'Y'/
      DATA BLANK/' '/
      DATA UP/'U','P','P','E','R',' ','L','I','M','I','T',' '/
      DATA UNDON/'N','O','T',' ','M','E','A','S','U','R','E','D'/
      CALL ASSIGN(2,'ELEMENT.DAT')
      DO 100 I=1,53
      READ(2,2) Z(I),A(I),(ELNAME(I,J),J=1,12)
2     FORMAT(1X,I2,5X,F10.3,5X,12A1)
100   CONTINUE
      TYPE 1
1     FORMAT(1X,'SAMPLE TYPE?')
      ACCEPT 20,SAMP
20    FORMAT(18A4)
      TYPE 7
  7   FORMAT(1X,'FILE FOR COMPOSITION INFORMATION?')
      ACCEPT 8,DUM1
  8   FORMAT(8A1)
      CALL ASSIGN(1,CMPOS)
      WRITE(1,21)SAMP
21    FORMAT(1X,18A4)
      TYPE 3
3     FORMAT(1X,'INPUT ELEMENT CONCENTRATIONS IN PPM(E10.3)')
      DO 200 I=1,53
      DO 201 IL=1,12
      FTNOTE(IL)=BLANK
201   CONTINUE
      TYPE 4,(ELNAME(I,J),J=1,12)
4     FORMAT(1X,'CONCENTRATION OF '12A1,1X,'=')
      READ(5,5,END=999)CONC
5     FORMAT(E10.3)
      IF(CONC.EQ.0.)GO TO 502
      GO TO 503
502   DO 203 IL=1,12
203   FTNOTE(IL)=UNDON(IL)
```

```
      GO TO 501
503   CONTINUE
      TYPE 9
  9   FORMAT(1X,'UPPER LIMIT?')
      ACCEPT 10,ANS
 10   FORMAT(A1)
      IF(ANS.EQ.YES)GO TO 500
      GO TO 501
500   DO 202 IL=1,12
      FTNOTE(IL)=UP(IL)
202   CONTINUE
501   CONTINUE
      WRITE(1,6) Z(I),A(I),CONC,(ELNAME(I,J),J=1,12),FTNOTE
6     FORMAT(1X,I2,10X,F10.3,10X,E10.3,10X,12A1,10X,12A1)
200   CONTINUE
999   CALL CLOSE(1)
      STOP
      END
```

```
**** ELEMENT.DAT ****

  1     1.008        HYDROGEN
  3     6.94         LITHIUM
  5     10.811       BORON
  7     14.0067      NITROGEN
 11     22.99        SODIUM
 13     26.982       ALUMINUM
 16     32.064       SULFUR
 17     35.453       CHLORINE
 18     39.948       ARGON
 19     39.102       POTASSIUM
 20     40.08        CALCIUM
 25     54.938       MANGANESE
 26     55.847       IRON
 27     58.9332      COBALT
 28     58.71        NICKEL
 29     63.54        COPPER
 30     65.35        ZINC
 33     74.93        ARSENIC
 34     78.96        SELENIUM
 35     79.904       BROMINE
 36     83.80        KRYPTON
 37     85.468       RUBIDIUM
 38     87.62        STRONTIUM
 39     88.905       YTTRIUM
 40     91.22        ZIRCONIUM
 41     92.906       NIOBIUM
 42     95.94        MOLYBDENUM
 43     99.          TECHNETIUM
 46     106.4        PALLADIUM
 47     107.87       SILVER
 50     118.69       TIN
 51     121.75       ANTIMONY
 53     126.9045     IODINE
 55     132.905      CESIUM
 56     137.33       BARIUM
 62     150.35       SAMARIUM
 63     151.96       EUROPIUM
 65     158.924      TERBIUM
 66     162.50       DYSPROSIUM
 67     164.93       HOLMIUM
 71     174.97       LUTETIUM
 72     178.49       HAFNIUM
 74     183.85       TUNGSTEN
 75     186.2        RHENIUM
 76     190.2        OSMIUM
 77     192.2        IRRIDIUM
 82     207.200      LEAD
 83     208.98       BISMUTH
 90     232.038      THORIUM
 91     233.         PROTOACTINIUM
 92     238.03       URANIUM
 93     239.         NEPTUNIUM
 94     239.         PLUTONIUM
```

```
      PROGRAM FLUX1
C     CREATES FILE OF NEUTRON FLUX
C     AND REACTOR CONDITIONS
C     WRITTEN BY J. C. EVANS/ BATTELLE N. W. LABORATORIES
C     DEC VAX 11/780 VERSION
C     FILE IS NAMED BY USER
C     A PERMANENT COPY OF THE DATA IN A MORE
C     READABLE FORMAT IS WRITTEN TO PRINT.FLX
C
C
C
      DIMENSION FLUX(29),FILE2(5),COND(18)
      TYPE 1
    1 FORMAT(1X,'FILE FOR FLUX INFORMATION?')
      ACCEPT 2,FILE2
    2 FORMAT(5A4)
      CALL ASSIGN(1,FILE2)
      TYPE 12
   12 FORMAT(1X,'THERMAL NEUTRON FLUX?')
      ACCEPT 5,PHI
      WRITE(1,5)PHI
      TYPE 13
   13 FORMAT(1X,'EPITHERMAL FLUX (.4 TO 1MEV)?')
      ACCEPT 5,EPI
      WRITE(1,5)EPI
      TYPE 3
    3 FORMAT(1X,'ENTER FAST FLUX AT EACH ENERGY')
      DO 500 I=1,29
      E=(FLOAT(I)/2.)+.5
  110 TYPE 4,E
    4 FORMAT(1X,F5.1,1X,'MEV=?')
      READ(5,5,ERR=100)FLUX(I)
      GO TO 105
  100 TYPE 20
   20 FORMAT(1X,'INPUT FORMAT ERROR!!!!')
      GO TO 110
  105 CONTINUE
    5 FORMAT(E8.2)
  500 CONTINUE
      WRITE(1,6)(FLUX(I),I=1,10)
      WRITE(1,6)(FLUX(I),I=11,20)
      WRITE(1,7)(FLUX(I),I=21,29)
    6 FORMAT(10E8.2)
    7 FORMAT(9E8.2)
      TYPE 14
   14 FORMAT(1X,'TEMPERATURE (CENTIGRADE)?')
      ACCEPT 15,TEMP
   15 FORMAT(F5.0)
      WRITE(1,15)TEMP
      TYPE 16
```

```
 16   FORMAT(1X,'REACTOR CONDITIONS')
      ACCEPT 17,COND
 17   FORMAT(18A4)
      WRITE(1,17)COND
      CALL CLOSE(1)
      CALL ASSIGN(1,'PRINT.FLX')
      WRITE(1,8)FILE2
  8   FORMAT(5X,'FAST FLUX DATA FOR FILE ',5A4)
      WRITE(1,9)
  9   FORMAT(5X,'FLUX DATA IN UNITS OF NEUTRONS/CM2*SEC*MEV',/)
      WRITE(1,10)
 10   FORMAT(1H0,4X,'ENERGY(MEV)'10X,'FLUX',/)
      DO 600 I=1,29
      E=(FLOAT(I)/2.)+.5
      WRITE(1,11)E,FLUX(I)
 11   FORMAT(11X,F5.2,6X,1PE8.2)
600   CONTINUE
      STOP
      END
```

```
      PROGRAM FLXDMP
C     LISTS FILES CREATED BY FLUX1 ON FILE PRINT.FLX
C     DEC VAX 11/780 VERSION
C
C
C
      DIMENSION FLUX(29),FILE2(5),COND(18)
      TYPE 1
    1 FORMAT(1X,'FILE FOR FLUX INFORMATION?')
      ACCEPT 2,FILE2
    2 FORMAT(5A4)
      CALL ASSIGN(1,FILE2)
      READ(1,5)PHI
      READ(1,5)EPI
    5 FORMAT(E8.2)
      READ(1,6)(FLUX(I),I=1,10)
      READ(1,6)(FLUX(I),I=11,20)
      READ(1,7)(FLUX(I),I=21,29)
    6 FORMAT(10E8.2)
    7 FORMAT(9E8.2)
      READ(1,15)TEMP
   15 FORMAT(F5.0)
      READ(1,17)COND
   17 FORMAT(18A4)
      CALL CLOSE(1)
      CALL ASSIGN(1,'PRINT.FST')
      WRITE(1,8)FILE2
    8 FORMAT(1H1,5X,'FAST FLUX DATA FOR FILE ',5A4)
      WRITE(1,25)COND
   25 FORMAT(1H0,5X,18A4)
      WRITE(1,9)
    9 FORMAT(1H0,5X,'FLUX DATA IN UNITS OF NEUTRONS/CM2*SEC*MEV',/)
      WRITE(1,10)
   10 FORMAT(1H0,5X,'ENERGY(MEV)'10X,'FLUX',/)
      DO 600 I=1,29
      E=(FLOAT(I)/2.)+.5
      WRITE(1,11)E,FLUX(I)
   11 FORMAT(11X,F5.2,6X,1PE8.2)
  600 CONTINUE
      WRITE(1,20)PHI
   20 FORMAT(1H0,'THERMAL FLUX=',1PE8.2,' N/CM2-SEC')
      WRITE(1,21)EPI
   21 FORMAT(1X,'EPITHERMAL FLUX=',1PE8.2,' N/CM2-SEC')
      WRITE(1,22)TEMP
   22 FORMAT(1X,'TEMPERATURE=',F5.0,' DEGREES CENTIGRADE')
      STOP
      END
```

```
**** BATCHO.LIB ****

TRITIUM
  3
CARBON
 14
COBALT
 60
NICKEL
 63
KRYPTON
 81
STRONTIUM
 90
TECHNITIUM
 99
SILVER
110
SAMARIUM
151
EUROPIUM
152
EUROPIUM
154
EUROPIUM
155
HOLMIUM
166
HAFNIUM
178
URANIUM
233
URANIUM
236
PLUTONIUM
239
```

```
**** BATCH1.LIB ****


IRON
 55
NICKEL
 59
ZINC
 65
SELENIUM
 79
NIOBIUM
 92
ZIRCONIUM
 93
MOLYBDENUM
 93
TECHNETIUM
 99
TIN
121
BARIUM
133
SAMARIUM
146
TERBIUM
158
LEAD
205
```

```
**** BATCH2.LIB ****

ARGON
 39
MANGANESE
 54
NICKEL
 63
KRYPTON
 85
NIOBIUM
 92
ZIRCONIUM
 93
PALLADIUM
107
TIN
121
CESIUM
134
CESIUM
135
CESIUM
127
EUROPIUM
155


**** BATCH3.LIB ****

MANGANESE
 53



**** BATCH4.LIB ****

CHLORINE
 36
IRON
 55
KRYPRON
 85
ZIRCONIUM
 93
```

```
      PROGRAM ACTIVE
C     MAIN PROGRAM FOR ACTIVATION CALCULATION
C     WRITTEN BY J. C. EVANS/ BATTELLE N.W. LABORATORIES
C     DEC VAX 11/780 VERSION
C     REQUIRES SUBROUTINES NCAP, DMP, FINAL, AND COMBAS
C     RESULTS ARE WRITTEN TO PRINT.ACT
C
C
C
      DOUBLE PRECISION DELT,TAU(9,9)
      INTEGER Z,ZZ,ZZZ
      BYTE DUMMY(12),BIGDUM(12),DUMDUM(12),DUM(12)
      DIMENSION FILE(5),SAMP(18),ELMENT(3),COND(18),SEARCH(50)
      DIMENSION ISO(9),M(50),UPLIM1(3),UPLIM2(3) UPLIM3(3),UPLIM4(3)
      DIMENSION FILE2(5),FILE4(5),CONC(9),FILE10(5)
      EQUIVALENCE(DUMMY,FILE10)
      EQUIVALENCE(BIGDUM,FILE4)
      EQUIVALENCE(DUMDUM,FILE)
      EQUIVALENCE(DUM,FILE2)
      COMMON/CAP1/ATOMS(9),ABUN(9,9),FYIELD(9,9),SIGMAT(9,9),
     1SIGMAF(9,9),DEPLT(9,9),DEPLF(9,9),PHI,CADR,THALF(9,9),N,JJ(9)
     2,URAN,MM(10),TEMP,THOR
      COMMON/CAP2/ACTIV,RATIO(9),IT,KK,ITYPE,SIGF(29),FLUX(29),JX,IY
      COMMON/CAP10/T1
      CALL ERRSET(63,.TRUE.,.FALSE.,.FALSE.,.FALSE.,15)
      FILE10(1)='BATC'
      FILE10(2)='H0.L'
      FILE10(3)='IB  '
      FILE4(1)='NUCL'
      FILE4(2)='IB0.'
      FILE4(3)='LIB '
      BIGDUM(12)=0
      TYPE 1
    1 FORMAT(1X,'FILE FOR COMPOSITION INFORMATION?')
      ACCEPT 2,DUMDUM
    2 FORMAT(8A1)
C     OPEN COMPOSITION FILE
      CALL ASSIGN(1,FILE)
C     OPEN PRINT FILE
      CALL ASSIGN(2,'PRINT.ACT')
      READ(1,3)SAMP
    3 FORMAT(1X,18A4)
      WRITE(2,4)SAMP
      TYPE 4,SAMP
    4 FORMAT(1H0,18A4)
      CALL CLOSE(1)
      TYPE 80
   80 FORMAT(1X,'FILE FOR FLUX INFORMATION')
      ACCEPT 2222,DUM
 2222 FORMAT(12A1)
```

```
      CALL ASSIGN(3,FILE2)
C     INPUT THERMAL FLUX
      READ(3,6)PHI
    6 FORMAT(E8.2)
      WRITE(2,7)PHI
    7 FORMAT(1H0,'THERMAL NEUTRON FLUX='1X,1PE8.2,1X,'N/CM2*SEC')
C     INPUT EPITHERMAL FLUX
      READ(3,6)EPI
C     CONVERT TO FLUX PER UNIT LETHARGY
      EPIU=EPI/14.27
C     COMPUTE CADMIUM RATIO
      CADR=PHI/EPIU
C     INPUT FAST FLUX
      READ(3,81)(FLUX(LL),LL=1,10)
      READ(3,81)(FLUX(LL),LL=11,20)
      READ(3,82)(FLUX(LL),LL=21,29)
   81 FORMAT(10E8.2)
   82 FORMAT(9E8.2)
C     INPUT TEMPERATURE
      READ(3,33)TEMP
   33 FORMAT(F5.0)
      WRITE(2,10)CADR,TEMP
   10 FORMAT(1H ,'CADMIUM RATIO=',F8.2,10X,'TEMPERATURE=',F5.0,1X,
     1 'DEGREES CENTIGRADE')
      READ(3,19)COND
      CALL CLOSE(3)
   19 FORMAT(18A4)
      WRITE(2,20)COND
   20 FORMAT(1H0,18A4)
      T1=SECNDS(0.)
C     OPEN BATCH RUN FILE
      DO 500 IJK=1,5
      I=1
      IF(IJK.EQ.1)DUMMY(6)='0'
      IF(IJK.EQ.1)BIGDUM(7)='0'
      IF(IJK.EQ.2)DUMMY(6)='1'
      IF(IJK.EQ.2)BIGDUM(7)='1'
      IF(IJK.EQ.3)DUMMY(6)='2'
      IF(IJK.EQ.3)BIGDUM(7)='2'
      IF(IJK.EQ.4)DUMMY(6)='3'
      IF(IJK.EQ.4)BIGDUM(7)='3'
      IF(IJK.EQ.5)DUMMY(6)='4'
      IF(IJK.EQ.5)BIGDUM(7)='4'
      CALL ASSIGN(4,FILE10)
  100 CONTINUE
      READ(4,42,END=110)SEARCH(I)
   42 FORMAT(A4)
      READ(4,14)M(I)
   14 FORMAT(I3)
      I=I+1
```

```
       GO TO 100
 110   I=I-1
       CALL CLOSE(4)
       IF(I.EQ.0)CALL FINAL
       DO 120 J=1,I
       CALL ASSIGN(3,FILE4)
 130   READ(3,15,END=99)ELMENT,MASS,ITYPE
  15   FORMAT(1X,3A4,I3,10X,I1)
       IF(M(J).NE.MASS)GO TO 130
       IF(SEARCH(J).NE.ELMENT(1))GO TO 130
       GO TO 140
  99   TYPE 21,M(J)
  21   FORMAT(1X,'ISOTOPE',1X,'AT MASS',1X,I3,1X,
     1 'NOT FOUND IN LIBRARY')
       CALL FINAL
       GO TO 120
 140   CONTINUE
       READ(3,17)N,Z
  17   FORMAT(1X,I1,10X,I2)
       ZZ=Z-N+1
       K=1
C      OPEN COMPOSITION FILE
       CALL ASSIGN(1,FILE)
       READ(1,3)SAMP
C      FIND FIRST MEMBER OF CHAIN
 150   READ(1,16,END=999)ZZZ,AA,CONC(K),UPLIM1
       IF(ZZZ.NE.ZZ)GO TO 150
       ATOMS(K)=6.023E17*CONC(K)/AA
       IF(ATOMS(K).EQ.0.)ATOMS(K)=1.E5
       IF(N.EQ.1)GO TO 166
C      FIND REST OF CHAIN
       DO 160 K=2,N
       READ(1,16,END=999)ZZZ,AA,CONC(K),UPLIM2
  16   FORMAT(1X,I2,10X,F10.3,10X,E10.3,32X,3A4)
       ATOMS(K)=6.023E17*CONC(K)/AA
       IF(ATOMS(K).EQ.0)ATOMS(K)=1.E5
 160   CONTINUE
 166   CONTINUE
       CALL CLOSE(1)
C      REWIND COMPOSITION FILE TO LOCATE TH AND U
       CALL ASSIGN(1,FILE)
       READ(1,3)READ
 165   READ(1,16,END=999)ZZZ,AA,THOR,UPLIM3
       IF(ZZZ.NE.90)GO TO 165
       THOR=6.023E17*THOR/AA
 168   READ(1,16,END=999)ZZZ,AA,URAN,UPLIM4
       IF(ZZZ.NE.92)GO TO 168
       URAN=6.023E17*URAN/AA
       GO TO 170
 999   TYPE 22
```

```
  22   FORMAT(1X,'COMPOSITION FILE INCOMPLETE')
       GO TO 120
 170   CONTINUE
       DO 180 II=1,N
       READ(3,23)JJ(II),MM(II)
  23   FORMAT(1X,I1,10X,I3)
       DO 190 L=1,JJ(II)
       READ(3,24)ABUN(L,II),FYIELD(L,II)
  24   FORMAT(1X,F7.4,10X,F7.4)
       READ(3,25)THALF(L,II)
  25   FORMAT(1X,E8.2)
       READ(3,26)SIGMAT(L,II),SIGMAF(L,II),DEPLT(L,II),DEPLF(L,II)
  26   FORMAT(1X,E8.2,3(10X,E8.2))
 190   CONTINUE
 180   CONTINUE
       IF(ITYPE.EQ.0)GO TO 400
       READ(3,81)(SIGF(LL),LL=1,10)
       READ(3,81)(SIGF(LL),LL=11,20)
       READ(3,82)(SIGF(LL),LL=21,29)
 400   CONTINUE
       KX=N-1
       KY=N-2
       MNM=N
       IF(ITYPE.EQ.0)IY=N
       IF(ITYPE.EQ.1)IY=N
       IF(ITYPE.EQ.2)IY=N-1
       IF(ITYPE.EQ.3)IY=N-1
       IF(ITYPE.EQ.4)IY=N-2
       IF(ITYPE.EQ.0)MNM=N
       IF(ITYPE.EQ.2)MNM=N-1
       IF(ITYPE.EQ.3)MNM=N-1
       IF(ITYPE.EQ.4)MNM=N-2
       JX=JJ(MNM)
       IF(ITYPE.EQ.1)JX=JJ(N)-1
       IF(ITYPE.EQ.0.OR.ITYPE.EQ.1)GO TO 156
       IF(ITYPE.EQ.2.OR.ITYPE.EQ.3)GO TO 153
       GO TO 154
 153   MS=MM(KX)-1
       DO 155 LX=1,JJ(KX)
       MS=MS+1
       IF(MASS.EQ.MS)LIX=LX
 155   CONTINUE
       GO TO 156
 154   MS=MM(KY)-1
       DO 158 LX=1,JJ(KY)
       MS=MS+1
       IF(MASS.EQ.MS)LIX=LX
 158   CONTINUE
 156   CONTINUE
       HFLIF=THALF(JX,IY)
```

```
      WRITE(2,27)ELMENT,MASS,HFLIF
 27   FORMAT(1H1,3A4,I3,10X,'HALF-LIFE=',1X,1PE8.2,1X,'YEARS')
      WRITE(2,270) CONC(IY),UPLIM1
270   FORMAT(1H0,'DIRECT CAPTURE TARGET ELEMENT CONCENTRATION
    1 =',F9.2,1X,'PPM',10X,3A4)
      IF(ITYPE.EQ.0)WRITE(2,280)
      IF(ITYPE.EQ.1)WRITE(2,281)
      IF(ITYPE.EQ.2)WRITE(2,302)CONC(N),UPLIM2
      IF(ITYPE.EQ.3)WRITE(2,303)CONC(N),UPLIM2
      IF(ITYPE.EQ.4)WRITE(2,304)CONC(N),UPLIM2
280   FORMAT(1H0,'N-GAMMA AND FISSION REACTIONS ONLY')
281   FORMAT(1H0,'N-2N REACTION INCLUDED')
302   FORMAT(1H0,'TARGET FOR N-P REACTION CONCENTRATION
    1 =',F9.2,1X,'PPM',10X,3A4)
303   FORMAT(1H0,'TARGET FOR N-D REACTION CONCENTRATION
    1 =',F9.2,1X,'PPM',10X,3A4)
304   FORMAT(1H0,'TARGET FOR N-ALPHA REACTION CONCENTRATION
    1 =',F9.2,1X,'PPM',10X,3A4)
      WRITE(2,20)COND
      DO 200 JK=1,JJ(N)
200   ISO(JK)=MM(N)+JK-1
      WRITE(2,29)Z
 29   FORMAT(1H0,41X,'BURNUP RATIOS FOR Z=',I2)
      WRITE(2,31)(ISO(JK),JK=1,JJ(N))
 31   FORMAT(1X,'T(YEARS)',10X,'ACTIVITY(CI/GM)',9(5X,I3))
      CALL NCAP
      TYPE 3111,ELMENT,MASS
3111  FORMAT(1H0,'CALCULATION COMPLETE FOR ',3A4,I3)
      CALL FINAL
120   CONTINUE
500   CONTINUE
      CALL ASSIGN(1,FILE)
      CALL COMBAS
      CALL CLOSE(1)
      CALL CLOSE(2)
      STOP  'DATA ON PRINT.ACT;*'
      END
```

```
      SUBROUTINE NCAP
C     ISOTOPE PRODUCTION AND DEPLETION SUBROUTINE FOR ACTIV
C     VAX 11/780 VERSION
C     INCLUDES FISSION PRODUCTS FROM U235-NATURAL ABUNDANCE
C     CALCULATES FISSION PRODUCTS FROM MULTIPLE NEUTRON
C     CAPTURE ON U238 AND TH232
C     FISSION YIELDS FOR U235 USED IN ALL CASES
C     INTEGRATION INCREMENT IS 3 DAYS
C     BETA DECAY IS GIVEN PRIORITY OVER BURNUP
      DOUBLE PRECISION DELT,TAU(9,9),T
      COMMON/CAP1/ATOMS(9),ABUN(9,9),FYIELD(9,9),SIGMAT(9,9),
     1 SIGMAF(9,9),DEPLT(9,9),DEPLF(9,9),PHI,CADR,THALF(9,9),N,JJ(9)
     2 ,URAN,MM(10),TEMP,THOR
      COMMON/CAP2/ACTIV,RATIO(9),IT,KK,ITYPE,SIGF(29),FLUX(29),JX,IY
      DIMENSION START(9),PROD(10,10),MT(9)
      BARN=1.E-24
      DELT=3.15E5
      KK=JJ(N)
      DO 50 L=1,JJ(N)
      START(L)=ATOMS(N)*ABUN(L,N)
      IF(START(L).EQ.0.)START(L)=1.E5
   50 CONTINUE
C     COMPUTE TEMPERATURE CORRECTION TO 2200 M/S CROSS-SECTION
      TCOR=SQRT(.79*(293./(273.+TEMP)))
      UFIVE=URAN*.0072
      UEIGHT=URAN*.993
      UTHREE=0.
      PLUT=0.
      FAST=PHI/CADR
      DO 150 II=1,N
      DO 150 L=1,JJ(II)
      TAU(L,II)=.69315/(THALF(L,II)*3.15E7)
      PROD(L,II)=ATOMS(II)*ABUN(L,II)
      SIGMAF(L,II)=SIGMAF(L,II)*BARN
      SIGMAT(L,II)=SIGMAT(L,II)*BARN*TCOR
      DEPLT(L,II)=DEPLT(L,II)*BARN*TCOR
      DEPLF(L,II)=DEPLF(L,II)*BARN
  150 CONTINUE
      FFAC=0.
      IF(ITYPE.EQ.0)GO TO 152
C     INTEGRATE FLUX TIMES CROSS-SECTION IN FAST REGION
      DO 151 LLX=1,29
  151 FFAC=FFAC+SIGF(LLX)*BARN*FLUX(LLX)
  152 CONTINUE
      DO 300 IIT=1,3000
      IT=IIT
      DO 100 II=1,N
C     CALCULATE FISSION PRODUCT ADDITIONS TO FIRST MEMBERS
C     OF EACH ROW
      PROD(1,II)=PROD(1,II)+UFIVE*(PHI*5.8E-22*TCOR+
```

```
    1 FAST*2.8E-22)*DELT*FYIELD(1,II)
      PROD(1,II)=PROD(1,II)+UTHREE*(PHI*5.3E-22*TCOR+
    1 FAST*7.6E-22)*DELT*FYIELD(1,II)
      PROD(1,II)=PROD(1,II)+PLUT*(PHI*7.4E-22*TCOR+
    1 FAST*3.E-22)*DELT*FYIELD(1,II)
      DO 200 L=2,JJ(II)
C     CALCULATE NEUTRON CAPTURE ADDITIONS+FISSION PRODUCTS
C       FOR REMAINING ISOTOPES
      LL=L-1
      PROD(L,II)=PROD(L,II)+PROD(LL,II)*(PHI*SIGMAT(L,II)+
    1 FAST*SIGMAF(L,II))*DELT
      PROD(L,II)=PROD(L,II)+UFIVE*(PHI*5.8E-22*TCOR+
    1 FAST*2.8E-22)*DELT*FYIELD(L,II)
      PROD(L,II)=PROD(L,II)+UTHREE*(PHI*5.3E-22*TCOR+
    1 FAST*7.6E-22)*DELT*FYIELD(L,II)
      PROD(L,II)=PROD(L,II)+PLUT*(PHI*7.4E-22*TCOR+
    1 FAST*3.E-22)*DELT*FYIELD(L,II)
  200 CONTINUE
C     INCLUDE FAST FLUX CONTRIBUTION
      PROD(JX,IY)=PROD(JX,IY)+PROD(JJ(N),N)*FFAC*DELT
      DO 201 L=1,JJ(II)
      JT=II+1
      IF(JT.GT.N)GO TO 202
      MT(II)=MM(JT)-MM(II)
      KT=L-MT(II)
      IF(KT.LT.1)GO TO 202
C     CALCULATE BETA CHAIN CHANGES
      PROD(KT,JT)=PROD(KT,JT)+PROD(L,II)*(1.-DEXP(-TAU(L,II)*DELT))
  202 CONTINUE
C     CALCULATE DECAY LOSSES
      PROD(L,II)=PROD(L,II)*DEXP(-TAU(L,II)*DELT)
C     CALCULATE BURNUP OF EACH ISOTOPE IN THE CHAIN
      PROD(L,II)=PROD(L,II)-PROD(L,II)*(PHI*DEPLT(L,II)+
    1 FAST*DEPLF(L,II))*DELT
  201 CONTINUE
  100 CONTINUE
      DO 203 L=1,JJ(N)
      RATIO(L)=PROD(L,N)/START(L)
  203 CONTINUE
C     CONVERT ATOMS OF PRODUCT TO CURIES/GM.
      ACTIV=PROD(JX,IY)*TAU(JX,IY)/3.7E10
C     CALCULATE PRODUCTION OF FISSIONABLE ISOTOPES
      UTHREE=UTHREE+THOR*(PHI*7.4E-24*TCOR+
    1 FAST*8.5E-23)*DELT
      PLUT=PLUT+UEIGHT*(PHI*2.7E-24*TCOR+FAST*2.7E-22)*DELT
C     CALCULATE DEPLETION OF FISSIONABLE ISOTOPES
      UTHREE=UTHREE-UTHREE*(PHI*5.8E-22*TCOR+
    1 FAST*9.0E-22)*DELT
      PLUT=PLUT-PLUT*(PHI*1.0E-21*TCOR+FAST*5.E-22)*DELT
      UFIVE=UFIVE-UFIVE*(PHI*6.8E-22*TCOR+FAST*4.2E-22)*
```

```
     1 DELT
C      CALCULATE DEPLETION OF URANIUM AND THORIUM
       THOR=THOR-THOR*(PHI*7.4E-24*TCOR+FAST*8.5E-23)*DELT
       UEIGHT=UEIGHT-UEIGHT*(PHI*2.7E-24*TCOR+FAST*
     1 2.7E-23)*DELT
       CALL DMP
  300  CONTINUE
       WRITE(2,16)
   16  FORMAT(1H0,5X,'ACTIVITY(CI/GM)',10X,'YEARS AFTER SHUTDOWN')
       DO 500 IK=1,7
       IA=IK-1
C      CALCULATE DECAY OF PRODUCT AFTER SHUTDOWN OF REACTOR
       T=10.**IA*3.15E7
       ACTIV=ACTIV*DEXP(-TAU(JX,IY)*T)
       T=T/3.15E7
       WRITE(2,17)ACTIV,T
   17  FORMAT(11X,1PE10.2,20X,1PE10.2)
  500  CONTINUE
       RETURN
       END
```

```
      SUBROUTINE DMP
C     USED BY ACTIV TO LIST RESULTS IN 1 YEAR INCREMENTS
C
      COMMON/CAF2/ACTIV,RATIO(9),IT,KK,ITYPE,SIGF(29),FLUX(29),JX,IY
      T=.01*FLOAT(IT)
      TT=AMOD(T,1.)
      IF(TT.NE.0.)GO TO 10
      WRITE(2,1)T,ACTIV,(RATIO(JK),JK=1,KK)
    1 FORMAT(1X,F8.0,17X,1PE8.2,9(3X,0PF5.3))
   10 CONTINUE
      RETURN
      END
```

```
      SUBROUTINE FINAL
C     USED BY ACTIV TO CALCULATE TIME ELAPSED FOR EACH ISOTOPE
C
      COMMON/CAP10/T1
      CALL CLOSE(3)
      CALL CLOSE(1)
      DELTA=SECNDS(T1)
      DELTA=DELTA/60.
      TYPE 40,DELTA
   40 FORMAT(1X,'ELAPSED TIME ='F8.2,1X,'MINUTES')
      RETURN
      END
```

```
      SUBROUTINE COMBAS
C     USED BY ACTIV TO LIST COMPOSITION DATA USED
C
      DIMENSION SAMP(18),ELNAME(3),UPLIM(3)
      INTEGER Z
      READ(1,3)SAMP
    3 FORMAT(18A4)
      WRITE(2,33)SAMP
   33 FORMAT(1H1,18A4)
      WRITE(2,4)
    4 FORMAT(1H0,/)
      WRITE(2,5)
    5 FORMAT(1X,' Z',19X,'A',11X,'CONC(PPM)',10X,'ELEMENT',/)
      DO 100 I=1,53
      READ(1,6)Z,A,CONC,ELNAME,UPLIM
    6 FORMAT(1X,I2,10X,F10.3,10X,E10.3,10X,3A4,10X,3A4)
      WRITE(2,7)Z,A,CONC,ELNAME,UPLIM
    7 FORMAT(1X,I2,10X,F10.3,10X,F10.3,10X,3A4,10X,3A4)
  100 CONTINUE
      RETURN
      END
```

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Technical Information (5)

NRC FORM 335
(11-81)

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

1. REPORT NUMBER *(Assigned by DDC)*
NUREG/CR-3474
PNL-4824

2. *(Leave blank)*

3. RECIPIENT'S ACCESSION NO.

4. TITLE AND SUBTITLE *(Add Volume No., if appropriate)*
Long-Lived Activation Products in Reactor Materials

5. DATE REPORT COMPLETED
MONTH July | YEAR 1984

7. AUTHOR(S) J. C. Evans, E. L. Lepel, R. W. Sanders, C. L. Wilkerson, W. Silker, C. W. Thomas, K. H. Abel, D. R. Robertson

DATE REPORT ISSUED
MONTH August | YEAR 1984

9. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*

Pacific Northwest Laboratory
P.O. Box 999
Richland, Washington 99352

6. *(Leave blank)*

8. *(Leave blank)*

12. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS *(Include Zip Code)*
Division of Engineering Technology
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555

10. PROJECT/TASK/WORK UNIT NO.

11. FIN NO. B2296

13. TYPE OF REPORT
Final Report

PERIOD COVERED *(Inclusive dates)*
1st Quarter 1978 to Present

15. SUPPLEMENTARY NOTES

14. *(Leave blank)*

16. ABSTRACT *(200 words or less)*

The purpose of this program was to assess the problems posed to reactor decommissioning by long-lived activation products in reactor construction materials. Samples of stainless steel, vessel steel, concrete, and concrete ingredients were analyzed for up to 52 elements in order to develop a data base of activatable major, minor, and trace elements. Large compositional variations were noted for some elements. A thorough evaluation was made of all possible nuclear reactions that could lead to long lived activation products. It was concluded that all major activation products have been satisfactorily accounted for in decommissioning planning studies completed to date. A comparison is made between calculated activation levels and regulatory guidelines for shallow land disposal according to 10 CFR 61. Most of the massive components were found to qualify as either Class A or Class B waste with the exception of PWR and BWR shroud material which clearly exceeds Class C limits. Selected samples of activated steel and concrete were subjected to a limited radiochemical analysis program as a verification of the computer model. Reasonably good agreement with the calculations was obtained where comparison was possible. In particular, the presence of ^{94}Nb in activated stainless steel at or somewhat above expected levels was comfirmed.

17. KEY WORDS AND DOCUMENT ANALYSIS

Nuclear Reactor Decommissioning
Long Lived Activation Products
Low Level Waste Disposal

17a. DESCRIPTORS

17b. IDENTIFIERS/OPEN-ENDED TERMS

18. AVAILABILITY STATEMENT
Unlimited

19. SECURITY CLASS *(This report)*
Unclassified

21. NO. OF PAGES

20. SECURITY CLASS *(This page)*
Unclassified

22. PRICE
$

NRC FORM 335 (11-81)

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555

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