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**RADIONUCLIDES IN UNITED STATES
COMMERCIAL NUCLEAR POWER REACTORS**

N. C. Dyer¹
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ABSTRACT

In the next ten to twenty years, many of the commercial nuclear power reactors in the United States will be reaching their projected lifetime of forty years. As these power plants are decommissioned, it seems prudent to consider the recycling of structural materials such as stainless steel. Some of these materials and components have become radioactive through either nuclear activation of the elements within the components or surface contamination with radioactivity from the operational activities.

In order to understand the problems associated with recycling stainless steel from decommissioned nuclear power reactors, it is necessary to have information on the radionuclides expected on or in the contaminated materials. A study has been conducted of radionuclide contamination information that is available for commercial nuclear power reactors in the United States. There are two types of nuclear power reactors in commercial use in the United States, pressurized water reactors (PWRs) and boiling water reactors (BWRs). Before presenting radionuclide activities information, a brief discussion will be given on the major components and operational differences for the PWRs and BWRs.

Radionuclide contamination information is presented from 11 PWRs and over 8 BWRs. These data include both the radionuclides within the circulating reactor coolant water as well as radionuclide contamination on and within component parts. A comparison between reactor coolant activities on a letdown valve indicates that the reactor coolant activities provide an indication of the expected radionuclide content for recycled reactor components. The highly volatile radionuclides, such as tritium and noble gases, will not be considered since they will be removed from the metal components at the recycling start. For the PWR reactor coolant, about 90% of the radionuclide activities are the radiocesium fission products with the other 10% being iron and cobalt activation products, Fe-55 and Co-60. For the BWR reactor coolant, about 91% of the activities are from Sb-125 which is an activation product of tin from the condenser tubing. The other 9% of the BWR reactor coolant activities is mainly from the activation products, Fe-55 and Co-60. For the samples of reactor component parts, between 86% to as much as 99% of the radionuclide activities are the activation products, Fe-55, Co-60, and Ni-63. This distribution of radionuclides for reactor components appears to be the same whether the activities are contamination on the part surfaces or are contamination distributed within the part materials.

In summary, for all the samples reviewed, the radionuclides that provided 1% or more of the total activities were: Mn-54, Fe-55, Co-60, Ni-63, Zn-65, Sb-125, Cs-134, and Cs-137. It appears that these would be the main radionuclides of concern for the recycling of material and stainless steel components from commercial nuclear power reactors in the United States.

1.0 INTRODUCTION

Many of the nuclear power plants in the United States have been operating for several years, some twenty to thirty years or more. As these plants reach their forty-year projected life span, it is reasonable to consider the recycling of materials used in their construction, such as stainless steel and other structural materials. In order to assess the radiological problems associated with the recycling of the stainless steel, an understanding of the radioactive contamination expected is very necessary. This report will review several studies on the amounts and types of radionuclide activities found in nuclear power plant systems and materials. In addition, a short summary of the major components will be presented for the two kinds of plants, pressurized water reactor (PWR) and boiling water reactor (BWR), used in the United States. A listing will be presented of the radionuclides expected in stainless steel components that may be recycled from nuclear power plants.

2.0 COMMERCIAL NUCLEAR POWER REACTORS

In the United States, there are two types of commercial nuclear power reactors: pressurized water reactors (PWRs) and boiling water reactors (BWRs). Until recently there was one high-temperature gas-cooled reactor (Fort St. Vrain in Colorado), a joint project between a utility and the federal government, but this project has been stopped. In other parts of the world heavy water (deuterium oxide) reactors (Canada), gas-cooled reactors (United Kingdom), and graphite reactors (previous Soviet Union) have been used to produce power, both electrical and heat. Other than Canada, which is still building and selling heavy water reactors, the rest of the world is changing to light water reactors such as PWRs and BWRs. The water moderated and cooled power reactors have a safety advantage over the graphite moderated and water cooled power reactors (similar to the Chernobyl design). A removal of the water from a water moderated and cooled power reactor will cause an automatic shutdown, while the same occurrence for a graphite moderated and water cooled power reactor will cause a temporary increase in power level. The federal government, through the United States Atomic Energy Commission and the United States Department of Energy, has operated and is still operating liquid metal power reactors, graphite moderated power reactors, and heavy water power reactors as well as PWRs and BWRs. The United States Navy uses PWRs as the propulsion source for both submarines and surface ships.

2.1 PRESSURIZED WATER REACTORS

The PWRs used by the utilities in the United States for electrical production have been designed and built by three manufacturers: Westinghouse, Combustion Engineering, and Babcock & Wilcox. Figure 1 presents a simplified schematic diagram for systems of a PWR. The nuclear fuel in the core area of the reactor vessel produces the heat energy. The heat energy is transferred to steam generators by the reactor or primary water system. Figure 1 shows a system with two steam generators or two loops but many reactors,

FIGURE 1. Pressurized Water Reactor

especially Westinghouse units, have three or four loops. The primary water is pressurized and therefore it does not boil even though the water temperature is around 300 degrees Celsius. The pressure level of the primary water is controlled around 2000 pounds per square inch by the pressurizer shown on one of the primary loops. The reactor vessel and primary water components are made of very thick stainless steel in order to contain this operating pressure.

The primary water passes through the nuclear core in the reactor vessel and in this manner becomes contaminated with radioactive nuclides. Figure 1 has represented this by shading of water and system components that come in contact with primary water. The nuclear fuel in the core is contained in ceramic pellets that are sealed in metal rods made of zircaloy. This provides two barriers between the radioactive fission products in the fuel and the primary water, however, the potential for leaks is present and it does occur. As metal and other material dissolved in the primary water pass through the reactor core, the large quantity of neutrons from the fission process causes some of these dissolved elements to be activated or to become radioactive. These fuel failures and activations of dissolved elements are the ways in which the primary water becomes contaminated with radioactivity. This radioactivity in the primary water will contaminate the reactor components through which it flows.

The steam generator of a PWR transfers the heat from the primary water to the secondary water. The secondary water is flashed to steam to power the steam turbine that turns the electrical generator. This secondary steam/water is condensed back to water by the condenser and reused. The steam jet air ejector on the steam line before the turbine removes any noncondensable gases from the steam. Figure 1 shows a u-tube steam generator in which the primary water enters at the bottom of the steam generator, flows up through and back down the unit to exit at the bottom. This is the type of steam generator used by both Westinghouse and Combustion Engineering. The Babcock & Wilcox steam generator is a once through unit in which the primary water enters at one end and exits from the other. The once through steam generators have a smaller secondary water inventory than the u-tube type. The once through type will become dry sooner if the secondary feedwater is stopped. Because of this the once through steam generator will lose its heat removal capability about ten to twenty minutes faster than a u-tube steam generator. Three Mile Island #2 was a Babcock & Wilcox reactor.

As is shown in Figure 1, the radioactively contaminated primary water does not contaminate the major reactor components beyond the steam generators. There is a small side stream of primary water which is directed to the chemical volume and control system (CVCS). This CVCS provides chemical and radioactive cleanup of the primary water through demineralizers and evaporators. By the CVCS the primary water is reduced in both pressure and temperature before being processed, and for this reason the CVCS is often referred to as the letdown system. The water processed through the CVCS is returned to the primary water loops by the charging pumps. Note that the primary water processed through

the CVCS is brought through the containment boundary or out of the containment building but the primary water providing the heat transfer to the steam generators does not pass through the containment boundary.

For controlling the fission process in the reactor core, control rods are mounted in the top or head to the reactor vessel. These control rods will slow or stop the fission process depending upon how far they are inserted into the reactor core. Since the control rods are mounted on the top of the reactor vessel, they are designed to drop by gravity completely into the core if any emergency occurs. For a pressurized water reactor, the power level control for the fission process is not controlled by the control rods but by the element, boron, which is dissolved in the primary water. This dissolved boron provides a chemical shim or nuclear shim which controls the fission process and thereby the power level of the reactor. The CVCS controls the boron shim in the primary water as well as providing cleanup of the primary water.

Not shown on Figure 1 are other systems or components of PWRs which include the radioactive waste processing system, the spent fuel storage system, the component cooling water system, the condenser cooling water system, the service water system, and the fire protection water system. As indicated previously, all water within the reactor systems is reused as much as possible because of the great expense involved with the initial water purification and treatment. Except for the radioactive waste processing and spent fuel storage systems, these other systems and components are usually not contaminated with radioactivity. The major contaminated components for a pressurized water reactor are those associated with the primary water system. Also, all systems or components are doubled or have backup trains and units so that maintenance and safety concerns are satisfied.

2.2 BOILING WATER REACTORS

The BWR is the other (than PWR) type of commercial nuclear power reactor used by electrical utilities in the United States. As can be seen from the schematic diagram of Figure 2, the BWR is much simpler than the PWR. For the BWR, the reactor water system, or the water removing heat energy from the nuclear core, is at or near atmospheric pressure. Therefore, the reactor water is boiling in the reactor vessel and is the water which becomes steam and provides energy to the turbine which turns the electrical generator. After the turbine, the reactor steam/water is cooled in the condenser and provides feedwater for the reactor water system.

As discussed in the Section 2.1 on PWRs, the reactor water will become contaminated with radioactive nuclides through close contact with the reactor core and the nuclear fuel. This occurs from both the dissolving of radioactive fission products that leak from the fuel and the activation of dissolved elements by the fission process neutrons. Therefore, the reactor water is highly contaminated with radioactive material and causes those systems and components it contacts to be highly contaminated. Figure 2 shows that most of the components associated with a BWR come in contact with the radioactive reactor water. That

FIGURE 2. Boiling Water Reactor

is, the reactor water piping, the steam turbine, the steam condenser, the reactor pump, and the reactor water cleanup system all become radioactively contaminated components (note the shading).

The steam jet air ejector on the reactor water steam line removes the noncondensable gases. For the BWR this includes most of the fission product noble gases as well as the activation gases and some of the tritium. There is a small side stream for the reactor water shown on Figure 2. This side stream provides cleanup and removal of radioactive and chemical contaminants in the reactor water. This side stream and the steam jet air ejector provide most of the removal of radioactivity from the reactor water in a BWR.

As with the PWR, the BWR has other component systems such as the radioactive water treatment system, the spent fuel storage system, the condenser cooling water system, the service water system, the component cooling water system, and the fire protection water system. Except for the radioactive waste treatment and the spent fuel storage systems, these other systems and components are usually not contaminated with radioactivity. In addition, there are generally two of every system or component so that maintenance and safety concerns are satisfied.

For a BWR, both the stoppage and the power level of the fission process are controlled by control rods that are mounted in the top or head of the reactor vessel. Since the control rods are used to terminate the nuclear reaction in the core, these rods are designed to drop by gravity if any emergency occurs. Unlike the PWR, the BWR does not have a chemical or boron shim in the reactor water as a controlling mechanism for reactor power level.

In the United States, all currently operating BWRs were designed and made by General Electric. In the early years of nuclear power, Allis-Chalmers designed and made BWRs. The only Allis-Chalmers unit that operated for any period of time was the La Crosse Boiling Water Reactor in Wisconsin which has ceased power operations.

3.0 RADIONUCLIDES IN NUCLEAR POWER REACTORS

3.1 EXPECTED RADIONUCLIDES

For the consideration of radionuclides that would be of interest in the recycling of materials from nuclear power reactors, it is important to consider how soon after reactor shutdown that these materials would be available. It would be reasonable to consider that it would take at least 1000 days (2.7 years) before any nuclear reactor material would enter the recycling process. Therefore, it would seem reasonable to limit consideration to those radionuclides with half-lives longer than 50 days. A radionuclide with a half-life of 50 days would be reduced by over a million times (decay factor of $9.5E-7$) in a period of 1000 days.

Table 1 presents a listing of radionuclides found in nuclear power reactors that have half-lives longer than 50 days. Included in Table 1 for each radionuclide is the half-life in days as reported by Kocher, et al.⁽¹⁾ and the mode of production expected for that radionuclide. In addition, the radionuclides are divided by their nuclear emissions, that is, gamma emitters, non-gamma emitters, or transuranics. The gamma emitters will be the major contributor to any external radiation dose or dose-rate from the reactor material during recycling or use of the recycled material. The non-gamma or low-energy gamma emitters will have a very small contribution to the external radiation dose but may be a major contributor to the internal radiation dose. The internal radiation dose only becomes a factor when radionuclides get inside a person's body through the breathing or air pathway and the eating or food pathway. This requires that radionuclides be released into the working area or general environment. The transuranic nuclides are the alpha emitters and are only of importance to the internal radiation dose. However, alpha emitters cause a much higher internal dose per radioactivity amount than for a beta or gamma emitter.

3.2 REACTOR COOLANT RADIONUCLIDES IN NUCLEAR POWER PLANTS

A review of the literature on radioactivity in nuclear power plants has found several studies that were performed at operating nuclear power plants. The objective of these studies was to provide a qualitative and quantitative measurement of radionuclides in the liquid and gaseous streams associated with commercial nuclear power reactors. The final objective was to develop a radionuclide source term model that could be used in the licensing process.

Table 2 presents a summary of reactor coolant activities that were measured in 8 PWRs and 7 BWRs. The average activities for PWRs are from two reports by Mandler et al.⁽²⁻³⁾ The first report⁽²⁾ provided data on the Westinghouse reactors: Zion #1 and #2 plus Turkey Point #3 and #4, on the Combustion Engineering reactor: Fort Calhoun, and the Babcock & Wilcox reactor: Rancho Seco. The second⁽³⁾ report provided data on the Westinghouse reactors: Prairie Island #1 and #2. The average activities for the BWRs are from two reports, by Pelletier and by Duce et al.⁽⁴⁻⁵⁾ The Pelletier report⁽⁴⁾ provided data on the reactors: Oyster Creek, Millstone Point #1, Dresden #2 and #3, plus Monticello. The other report⁽⁵⁾ provided data on the reactors: Brunswick #1 and #2. All the BWR plants were General Electric units.

The average reactor coolant activities were calculated as they would be after a 1000 day decay period to simulate the expected delay before recovery and processing of the recycled material. The total activities as well as the fraction of the total activities for each radionuclide were calculated for both the PWR and BWR activities. The fraction of the total activity was determined without including the H-3 or tritium activity in the total because of the high volatility of this radionuclide. The fraction of the total activity will provide an importance ranking for each radionuclide in that a large fraction will reflect the need to consider that radionuclide.

TABLE 1. Radionuclides Found in Nuclear Power Reactors
(Limited to half-lives longer than 50 days)

Nuclide	Half-life (days)	Mode of Production
<i>For Gamma Emitters:</i>		
Mn-54	3.1E+2	Activation of iron
Co-57	2.7E+2	Activation of nickel
Co-58	7.1E+1	Activation of nickel
Co-60	1.9E+3	Activation of cobalt
Zn-65	2.4E+2	Activation of zinc
Kr-85	3.9E+3	Fission/Fuel
Zr-95	6.4E+1	Fission/Fuel and Activation of Zr-94
Nb-94	7.4E+6	Fission/Fuel then Activation of Nb-93
Ru-106 & D	3.7E+2	Fission/Fuel
Ag-108m	4.6E+4	Activation of silver
Ag-110m	2.5E+2	Activation of silver
Sb-124	6.0E+1	Activation of antimony
Sb-125	1.0E+3	Activation of tin
Cs-134	7.5E+2	Fission/Fuel then Activation of Cs-133
Cs-137	1.1E+4	Fission/Fuel
Ce-144 & D	2.8E+2	Fission/Fuel
Eu-152	5.0E+3	Fission then
Eu-154	3.2E+3	Fission then
Eu-155	1.8E+3	Fission then
<i>For Non-Gamma or Low-Energy Gamma Emitters:</i>		
H-3	4.5E+3	Activation of deuterium or lithium
C-14	2.1E+6	Activation of nitrogen
S-35	8.7E+1	Activation of sulfur or chlorine
Ca-45	1.6E+2	Activation of calcium
Fe-55	9.9E+2	Activation of iron
Ni-59	2.7E+7	Activation of nickel
Ni-63	3.7E+4	Activation of nickel
Sr-89	5.1E+1	Fission/Fuel

TABLE 1. (continued)

Nuclide	Half-life (days)	Mode of Production
Sr-90 & D	1.0E+4	Fission/Fuel
Y-91	5.9E+1	Fission/Fuel
Nb-93m	5.3E+3	Fission/Fuel
Mo-93	1.3E+6	Activation of molybdenum
Tc-99	7.8E+7	Fission/Fuel
I-129	5.7E+9	Fission/Fuel
Pm-147	9.6E+2	Fission/Fuel
<i>For Transuranics:</i>		
Pu-238	3.2E+4	Fission/Fuel
Pu-239	8.8E+6	Fission/Fuel
Pu-240	2.4E+6	Fission/Fuel
Pu-241	5.3E+3	Fission/Fuel
Am-241	1.6E+5	Fission/Fuel
Cm-242	1.6E+2	Fission/Fuel
Cm-244	6.6E+3	Fission/Fuel

TABLE 2. Reactor Coolant Activities

For Pressurized Water Reactors:				For Boiling Water Reactors:		
Nuclide	Activity Average 8 PWRs ($\mu\text{Ci/g}$)	Activity Decayed 1000 days ($\mu\text{Ci/g}$)	Fraction of Total w/o H-3	Activity Average 7 BWRs ($\mu\text{Ci/g}$)	Activity Decayed 1000 days ($\mu\text{Ci/g}$)	Fraction of Total w/o H-3
<i>Total (w/o H-3)=9.76E-3($\mu\text{Ci/g}$)</i>				<i>Total (w/o H-3)=9.36E-3($\mu\text{Ci/g}$)</i>		
H-3	2.0E-1	1.7E-1	---	3.1E-3	2.7E-3	---
C-14	4.1E-5	4.1E-5	4.2E-3	<1E-6	<1E-6	
S-35	1.8E-5	6.2E-9	6.4E-7	*		
Ca-45	1.1E-4	1.5E-6	1.5E-4	<4E-7	<5E-9	
Mn-54	5.4E-4	5.8E-5	5.9E-3	9.4E-5	1.0E-5	1.1E-3
Fe-55	1.5E-3	7.5E-4	7.7E-2	1.2E-3	6.0E-4	6.4E-2
Co-57	3.8E-4	2.9E-5	3.0E-3	*		
Co-58	4.9E-3	2.8E-7	2.9E-5	2.4E-4	1.4E-8	1.5E-6
Co-60	3.8E-4	2.6E-4	2.7E-2	2.4E-4	1.7E-4	1.8E-2
Ni-63	8.2E-5	8.1E-5	8.3E-3	2.4E-6	2.4E-6	2.6E-4
Zn-65	1.7E-4	9.5E-6	9.7E-4	1.5E-4	8.4E-6	9.0E-4
Sr-89	1.4E-4	1.8E-10	1.8E-8	8.2E-5	1.0E-10	1.1E-8
Sr-90 & D	1.8E-6	1.7E-6	1.7E-4	3.0E-6	2.8E-6	3.0E-4
Y-91	5.9E-6	4.7E-11	4.8E-9	4.2E-5	3.3E-10	3.5E-8
Zr-95	2.1E-4	4.2E-9	4.3E-7	1.2E-5	2.4E-10	3.7E-6
Ru-106 & D	2.9E-5	4.5E-6	4.6E-4	*		
Ag-110m	4.2E-5	2.6E-6	2.7E-4	1.2E-5	7.5E-7	8.0E-5
Sb-124	4.4E-5	4.2E-10	4.3E-8	1.3E-6	1.3E-11	1.4E-9
Sb-125	6.9E-6	3.5E-6	3.6E-4	1.7E-2	8.5E-3	9.1E-1
Cs-134	5.6E-3	2.2E-3	2.3E-1	2.6E-5	1.0E-5	1.1E-3
Cs-137	6.7E-3	6.3E-3	6.5E-1	5.7E-5	5.4E-5	5.8E-3
Ce-144 & D	1.2E-4	1.0E-5	1.0E-3	<3E-5	<3E-6	
Pm-147	8E-7	4E-7	4.1E-5	*		
Eu-152	<8E-5	<7E-5		<2E-5	<2E-5	
Eu-154	2.1E-6	1.7E-6	1.7E-4	*		
Eu-155	3E-6	2E-6	2.1E-4	*		

*: Nuclide not reported.

A comparison between the relative radionuclide fractions for the two reactor types is interesting. For the metal activation nuclides from iron, cobalt and zinc, the fractions are about the same for the PWRs and the BWRs. However, for the activation nuclides from nickel and tin, there appear to be significant differences. The nickel activation nuclides (Co-58 and Ni-63) seem to be about an order of magnitude higher in the PWRs while the tin activation nuclide (Sb-125) is about three orders of magnitude higher in the BWRs. The Sb-125 activity in the BWRs is probably due to dissolved tin in the reactor water from the condenser tubes. The radiocesiums (Cs-134 and Cs-137) are about two orders of magnitude higher in the PWRs than in the BWRs. Since radiocesium is a measure of fuel failures, there may be more failed fuel in the PWRs. However, the radiostrontiums (Sr-89 and Sr-90) are about the same activity level in the two reactor types. This fact does not support higher failed fuel in the PWRs. The volatility of the radiocesiums and their removal by the BWR steam jet air ejector may be a reason for lower radiocesium levels in BWR reactor coolant. The two orders of magnitude lower H-3 activity for the BWR reactor coolant supports this volatility and steam jet air ejector removal argument for radiocesium removal.

A question that can be asked is: How well does the circulating fluid activities represent the activities deposited on piping walls and system surfaces? That is, are reactor coolant activities representative of the contamination activities on or in system components? As a means of assessing this, Table 3 presents a comparison between radioactivities in reactor coolant and on the surface of a letdown valve for Rancho Seco, a pressurized water reactor. The letdown valve is in the reactor coolant cleanup system. These valve data are presented in Section 3.3.2. The Rancho Seco reactor coolant activities are from Reference 2 and were sampled in the late 1978 to early 1979 time period while the letdown valve sample was collected in June 1983. A lot of factors could have changed the reactor coolant activities at Rancho Seco in the time between 1979 and 1983. Nevertheless, a comparison shows that the radiocesium activities are much higher in the reactor coolant while the Co-60, Ni-63, and Sb-125 activities are higher on the valve sample. Even with these differences, the reactor coolant activities provide an indication of the expected radionuclide content for recycled reactor metal material.

3.3 RADIONUCLIDES ON NUCLEAR POWER PLANT COMPONENTS

The following sections will present radionuclide measurement data on components and parts taken from contaminated systems of nuclear power plants

3.3.1 Shippingport Nuclear Power Plant Samples

Tables 4 and 5 present radionuclide activities reported by Pacific Northwest Laboratory (PNL) for samples taken from the Shippingport Nuclear Power Plant during the Plant's decommissioning process.⁽⁶⁾ Shippingport was a small (72 MWe), four-loop, PWR that was built as a demonstration project in the mid-1950's. The Plant was a joint project of the federal government and Duquesne Light Company of Pennsylvania. Shippingport began

**TABLE 3. Reactor Coolant Compared to Valve Sample
Rancho Seco Nuclear Power Plant**

Reactor Coolant Activity			
Nuclide	Decayed 1000 days ($\mu\text{Ci/g}$)	Fraction of Total	Letdown Valve Fraction of Total
<i>Total = 1.722 $\mu\text{Ci/g}$</i>		<i>Total = 0.252 $\mu\text{Ci/cm}^2$</i>	
Mn-54	7.4E-5	4.3E-3	1.2E-2
Fe-55	4.5E-3	2.61E-1	2.90E-1
Co-57	3.6E-6	2.1E-4	7.1E-4
Co-58	1.4E-6	8.1E-5	1.1E-5
Co-60	6.3E-4	3.7E-2	3.65E-1
Ni-63	2.7E-4	1.6E-2	2.98E-1
Zn-65	< 1E-6		6.7E-5
Sr-90 & D	3.2E-6	1.9E-6	
Zr-95	2.7E-4	1.6E-4	
Ru-106 & D	< 2E-5		1.8E-3
Ag-110m	5.9E-6	3.4E-4	5.2E-3
Sb-125	< 2E-5		1.3E-2
Cs-134	3.1E-3	1.80E-1	9.5E-4
Cs-137	8.6E-3	4.99E-1	1.0E-2
Ce-144 & D	3.7E-5	2.1E-3	9.9E-5

**TABLE 4. Samples From Reactor Coolant Loop
Components Shippingport Nuclear Power Plant
(Reactor ceased operations on 10/82)**

Nuclide	Activity on Surface	Activity	Fraction of Total
	as of 9/88 ($\mu\text{Ci}/\text{cm}^2$)	as of 6/85 1180 days before ($\mu\text{Ci}/\text{cm}^2$)	

Average for Five Samples from Reactor Coolant Loop Components
Total Activity = 1.296 $\mu\text{Ci}/\text{cm}^2$

C-14	<2.9E-7	<2.9E-7	
Mn-54	1.7E-4	2.4E-3	1.9E-3
Fe-55	2.4E-1	5.5E-1	4.24E-1
Co-60	2.7E-1	4.2E-1	3.24E-1
Ni-59	1.4E-3	1.4E-3	1.1E-3
Ni-63	3.1E-1	3.2E-1	2.47E-1
Sr-90	<4.2E-7	<4.6E-7	
Nb-93m	2.1E-3	2.5E-3	1.9E-3
Nb-94	1.7E-5	1.7E-5	1.3E-5
Tc-99	1.1E-4	1.1E-4	8.5E-5
I-129	<1.6E-8	<1.6E-8	
Pu-238	3.4E-8	3.5E-8	2.7E-8
Pu-239/Pu-240	1.4E-8	1.4E-8	1.1E-8
Am-241	1.7E-8	1.7E-8	1.3E-8
Cm-244	1.2E-8	1.4E-8	1.1E-8

B-Loop Primary Coolant Check Valve (Highest Activity of Five)
Total Activity = 6.276 $\mu\text{Ci}/\text{cm}^2$

C-14	<5.9E-8	<5.9E-8	
Mn-54	4.9E-4	6.9E-3	1.1E-3
Fe-55	1.18	2.70	4.30E-1
Co-60	1.30	2.00	3.19E-1
Ni-59	6.76E-3	6.8E-3	1.1E-3
Ni-63	1.52	1.55	2.47E-1
Sr-90	<4.2E-7	<4.6E-7	
Nb-93m	1.00E-2	1.2E-2	1.9E-3
Nb-94	8.40E-5	8.4E-5	1.3E-5
Tc-99	5.59E-4	5.6E-4	8.9E-5
I-129	<1.6E-8	<1.6E-8	
Pu-238	1.17E-7	1.2E-7	1.9E-8
Pu-239/Pu-240	4.73E-8	4.7E-8	7.5E-9
Am-241	5.09E-8	5.1E-8	8.1E-9
Cm-244	2.32E-8	2.6E-8	4.1E-9

TABLE 5. Radioactivity in Fuel Bundle Hardware
Shippingport Nuclear Power Plant
(Reactor ceased operations on 10/82)

Nuclide	Bulk Activity as of 11/28/88 ($\mu\text{Ci/g}$)	Activity as of 6/85 1240 days before ($\mu\text{Ci/g}$)	Fraction of Total
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Type 348 Stainless Steel Plenum Sleeve from Blanket Rod
Total Activity = $3.85\text{E-}3 \mu\text{Ci/g}$

Mn-54	9.19E-7	1.5E-5	3.9E-3
Fe-55	*		
Co-60	2.34E-3	3.7E-3	9.61E-1
Ni-59	<1.1E-9	<1.1E-9	
Ni-63	1.16E-4	1.2E-4	3.1E-2
Nb-93m	1.91E-6	2.2E-6	5.7E-4
Nb-94	9.64E-9	9.6E-9	2.5E-6
Mo-93	1.77E-8	1.8E-8	4.7E-6
Sb-125	4.41E-6	1.0E-5	2.6E-3
Cs-137	2.33E-7	2.5E-7	6.5E-5

Type 348 Stainless Steel Plenum Sleeve from Reflector Rod
Total Activity = $2.04\text{E-}3 \mu\text{Ci/g}$

Mn-54	1.19E-6	1.9E-5	9.3E-3
Fe-55	3.99E-4	9.5E-4	4.66E-1
Co-60	4.55E-4	7.2E-4	3.53E-1
Ni-59	1.01E-6	1.0E-6	4.9E-4
Ni-63	3.38E-4	3.5E-4	1.72E-1
Nb-93m	2.78E-6	3.3E-6	1.6E-3
Nb-94	4.49E-8	4.5E-8	2.2E-5
Mo-93	6.04E-9	6.0E-9	2.9E-6
Sb-125	*		
Cs-137	*		

* : Nuclide not reported

operations in December 1957 and was finally shut down on October 1, 1982. The complete dismantlement of Shippingport was begun in 1985 with the restoration of the site to unrestricted use in 1989.

Shippingport was operated by Duquesne under supervision of the United States Department of Energy, Division of Naval Reactors. Besides producing electrical power for Duquesne, Shippingport was a test unit for nuclear fuel development. The final reactor core was a test and demonstration light water breeder core. That is, the plant was operated as a test of nuclear fuel which was designed to produce more fuel than was burned in addition to using light water as the moderator and coolant.

The report on Shippingport presents surface radionuclide activities for five samples taken from components in the reactor coolant loops.⁽⁶⁾ Table 4 presents the average activities for these five samples plus the activities for the sample (B-loop primary coolant check valve) with the highest contamination level. The radionuclide activities, which were reported as of September 1988, have been calculated to a time of 1180 days before sample collection or June 1985. This radionuclide decay correction to June 1985 or calculation was performed so that the sample activities would reflect a time of about 1000 days after the reactor shutdown which occurred on October 1, 1982.

A review of the fraction of the total for the radionuclide activities presented in Table 4 shows that almost all the radioactivity, about 99% is contributed by the activation nuclides, Fe-55, Co-60, and Ni-63. There is very little contribution from the fission product radionuclides, Sr-90, Tc-99, and the transuranics. The Shippingport reactor did not have any significant fuel failures during its operating history and this is the reason for the lack of fission product radionuclide activity.

Table 5 presents the radionuclide activities for two stainless steel samples that were taken from the hardware holding the fuel bundles together for the Shippingport reactor. These particular metal samples were in the reactor core and, therefore, were exposed to the neutron fields directly. The activities for these samples were calculated backward in time by 1240 days, to June 1985, or 1000 days after the reactor shutdown. As with the reactor loop component data, the main contributor, about 99%, to the radionuclide activities for these stainless steel samples were the activation nuclides, Fe-55, Co-60, and Ni-63. The fission product radionuclides, represented by Cs-137, are a very, very small fraction of the total. The activation radionuclide, Sb-125, shows a fractional amount of 0.26% for one of these samples.

The stainless steel samples of Table 5 provide information on what may be expected from bulk radionuclide contamination or activation. That is, components that have been in the reactor core area and exposed to the associated neutrons will have activation radionuclides distributed throughout the material rather than just surface activity. Since the surface activities may probably be chemically bound as an oxide or salt, these surface

activities may be removed with the flux in the melting process. Contrary to this, the bulk activities will probably be within the alloy, thereby, tending to stay in the melt and becoming part of the ingot or recycled material.

3.3.2 Rancho Seco Nuclear Power Plant Samples

A report by PNL presents radionuclide measurement data on samples from the Rancho Seco Nuclear Power Plant.⁽⁷⁾ Rancho Seco was a PWR of the Babcock & Wilcox design that was operated by Sacramento Municipal Utility District of California until the late 1980's when it was closed for economic reasons.⁽⁷⁾ The plant, a 933 MWe producer, went critical in September 1974 and started commercial operation in 1975.

Tables 6 and 7 present the radionuclide activity data for letdown cooler piping and valve samples taken from Rancho Seco in June 1983. This was several years prior to the plant's economic shutdown. In a PWR, the letdown system is another name for the CVCS or the chemical and radionuclide cleanup system for the primary coolant water. The radionuclide activities have been calculated or decayed to a time 1000 days later. This will approximate the activities when recycle processing would be done. A review of Tables 6 and 7 shows that, again, most of the radioactivity, about 96%, is from the activation nuclides, Fe-55, Co-60, and Ni-63. However, the fraction of the total for the transuranic radionuclides is higher by about three orders of magnitude for this Rancho Seco sample as compared to the Shippingport sample data presented in Table 4. This indicates that Rancho Seco did have fuel failures before June 1983 as was indicated in the reactor coolant data presented in Table 3. An additional indicator of failed fuel for Rancho Seco is the Ru-106 and Cs-137 activity fractions shown for the valve sample. It is interesting to note that the activation products, Sb-125 and Ag-110m, are present in these Rancho Seco samples.

3.3.3 Trojan Nuclear Power Plant Sample

From personal communication, radionuclide activity information has been obtained for a steam generator tube sample from the Trojan Nuclear Power Plant.⁽⁸⁾ Trojan was a Westinghouse PWR with a power level of about 1100 MWe. Portland General Electric Company of Oregon ran Trojan with operations starting in December 1975. The plant was closed in January 1993 for economic reasons. Table 8 presents the radionuclide data for this steam generator tube sample. Since this tube sample was obtained and evaluated before the reactor was shut down, the activities have been decayed for 1000 days. Again for this Trojan sample, most of the radionuclide activities, about 99%, are from the activation nuclides, Fe-55, Co-60, and Ni-63. During the early 1980's, Trojan experienced major fuel failures. This is reflected in the higher levels of the transuranics plus Sr-90 and Cs-137 than were seen in the Shippingport samples (Tables 4 and 5).

**TABLE 6. Letdown Cooler Piping Sample
Rancho Seco Nuclear Power Plant**

Nuclide	Activity on Pipe Surface ($\mu\text{Ci}/\text{cm}^2$)	Activity Decayed 1000 days ($\mu\text{Ci}/\text{cm}^2$)	Fraction of Total
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Total Activity = 1.611E-1 $\mu\text{Ci}/\text{cm}^2$

Mn-54	7.1E-3	7.6E-4	4.7E-3
Fe-55	1.00E-1	5.0E-2	3.10E-1
Co-57	6.1E-4	4.7E-5	2.9E-4
Co-58	1.59E-1	9.2E-6	5.7E-5
Co-60	2.27E-2	1.6E-2	9.9E-2
Ni-59	5.1E-4	5.1E-4	3.2E-3
Ni-63	8.98E-2	8.8E-2	5.46E-1
Zn-65	4.5E-4	2.5E-5	1.6E-4
Sr-90 & D	<4E-5	<4E-5	
Zr-95	<1E-4	<2E-9	
Nb-94	<2E-5	<2E-5	
Tc-99	<2E-5	<2E-5	
Ru-106 & D	<8E-4	<1E-4	
Ag-108m	<2E-5	<2E-5	
Ag-110m	1.88E-2	1.5E-3	7.4E-3
Sb-125	9.0E-3	4.5E-3	2.8E-2
I-129	<3E-8	<3E-8	
Cs-134	<5E-5	<2E-5	
Cs-137	<3E-5	<3E-5	
Ce-144 & D	<7E-5	<6E-6	
Pu-238	1.5E-6	1.5E-6	9.3E-6
Pu-239/Pu-240	8.2E-7	8.2E-7	5.1E-6
Am-241	1.9E-6	1.9E-6	1.2E-5
Cm-242	7.2E-6	9.5E-8	5.9E-7
Cm-244	1.8E-6	1.6E-6	9.9E-6

**TABLE 7. Letdown Cooler Valve Sample
Rancho Seco Nuclear Power Plant**

Nuclide	Activity on Valve Surface ($\mu\text{Ci}/\text{cm}^2$)	Activity Decayed 1000 days ($\mu\text{Ci}/\text{cm}^2$)	Fraction of Total
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Total Activity = 2.520E-1 $\mu\text{Ci}/\text{cm}^2$

Mn-54	2.87E-2	3.1E-3	1.2E-2
Fe-55	1.47E-1	7.3E-2	2.90E-1
Co-57	2.35E-3	1.8E-4	7.1E-4
Co-58	4.81E-2	2.8E-6	1.1E-5
Co-60	1.32E-1	9.2E-2	3.65E-1
Ni-59	6.9E-4	6.9E-4	2.7E-3
Ni-63	7.62E-2	7.5E-2	2.98E-1
Zn-65	3.0E-4	1.7E-5	6.7E-5
Sr-90 & D	<2E-5	<2E-5	
Zr-95	<2E-4	<4E-9	
Nb-94	<5E-5	<5E-5	
Tc-99	<2E-6	<2E-6	
Ru-106 & D	3.0E-3	4.6E-4	1.8E-3
Ag-108m	<4E-5	<4E-5	
Ag-110m	2.1E-2	1.3E-3	5.2E-3
Sb-125	6.8E-3	3.4E-3	1.3E-2
I-129	<5E-8	<5E-8	
Cs-134	6.0E-4	2.4E-4	9.5E-4
Cs-137	2.74E-3	2.6E-3	1.0E-2
Ce-144 & D	3.0E-4	2.5E-5	9.9E-5
Pu-238	1.6E-6	1.6E-6	6.3E-6
Pu-239/Pu240	8.4E-7	8.4E-7	3.3E-6
Am-241	1.4E-6	1.4E-6	5.6E-6
Cm-242	9.4E-6	1.2E-7	4.8E-7
Cm-244	1.7E-6	1.5E-6	6.0E-6

**TABLE 8. Steam Generator Tube Sample
Trojan Nuclear Power Plant**

Nuclide	Activity on Tube Surface ($\mu\text{Ci}/\text{cm}^2$)	Activity Decayed 1000 days ($\mu\text{Ci}/\text{cm}^2$)	Fraction of Total
<i>Total Activity = 77.7 $\mu\text{Ci}/\text{cm}^2$ (w/o H-3)</i>			
H-3	1E-2	8.6E-3	---
C-14	9.55E-5	9.6E-5	1.2E-6
Mn-54	3.4E-1	3.6E-2	4.6E-4
Fe-55	7.8E+1	3.9E+1	5.02E-1
Co-58	2.2E-1	1.3E-5	1.7E-7
Co-60	1.98E+1	1.4E+1	1.80E-1
Ni-63	2.49E+1	2.4E+1	3.09E-1
Sr-89	1.22E-4	1.5E-10	1.9E-12
Sr-90 & D	5.3E-2	5.0E-2	6.4E-4
Zr-95	4.8E-3	9.5E-8	1.2E-9
Tc-99	2.6E-4	2.6E-4	3.4E-6
Ru-106 & D	7.8E-1	1.2E-1	1.5E-3
Sb-125	2.8E-1	1.4E-1	1.8E-3
I-129	2.7E-4	2.7E-4	3.5E-6
Cs-137	3.3E-2	3.1E-2	4.0E-4
Ce-144 & D	4.1E-1	3.5E-2	4.5E-4
Pu-238	2.4E-3	2.4E-3	3.1E-5
Pu-239/Pu-240	2.2E-3	2.2E-3	2.8E-5
Pu-241	3.1E-1	2.7E-1	3.5E-3
Cm-242	8.6E-3	1.1E-4	1.4E-6

3.3.4 Steam Generator Manway Insert Samples

An Electric Power Research Institute (EPRI) report presents radionuclide activity measurements on steam generator manway inserts from two nuclear power plants, Robert Emmett Ginna and Turkey Point #3.(9) Both Ginna and Turkey Point #3 are Westinghouse PWRs with u-tube steam generators. Ginna is a 490 MWe, two loop reactor which is operated by Rochester Gas and Electric Company of New York. Significant power operations were started in 1970 at Ginna. Turkey Point #3 is a 666 MWe, three loop reactor that started significant power operations in 1972. Turkey Point #3 is operated by Florida Power and Light Company of Florida.

A steam generator manway insert is a stainless steel disk placed inside the manway access covers in both the hot and cold legs of the primary coolant side. The concept is to collect the radionuclide deposits on the insert rather than contaminating the manway covers which must be removed for maintenance on the primary side of the steam generator. These inserts are usually not reused after a repair removal so the activities only represent the accumulations since the previous steam generator repair. The measured inserts were 3/4-inch thick by 24-inch diameter disks of 304 type stainless steel. Only a 16-inch diameter area of the disks was exposed to reactor coolant.

Table 9 presents the radionuclide activities for the Ginna and Turkey Point #3 inserts. The Ginna samples were taken in March 1975 and the activities represent the average for the two inserts from the hot and cold legs of loop A. The Turkey Point #3 samples were taken in October 1975 and the activities represent the average of the six inserts from the hot and cold legs of all three loops. The measurements on the inserts were performed with gamma-sensitive instruments only, so the pure beta-emitting radionuclides, such as Fe-55 and Ni-63, were not determined. A review of the data shows that most (about 95%) of the activity is from the activation nuclide, Co-60. However, for both Ginna and Turkey Point #3, about 3% of the total activity was from the fission product radionuclide, Cs-137. If the non-measured Fe-55 and Ni-63 radionuclides are assumed to be present at the activity level of Co-60, then the fraction of Cs-137 would be about 1% which is about the same fractional level as for Rancho Seco (Table 7).

3.3.5 Corrosion Products on Boiling Water Reactor Piping

An EPRI report presents corrosion product radioactivity data on reactor coolant piping for BWRs.(10) The data are provided for two different demineralizers in the plant's reactor water cleanup system. Although these data only address corrosion or activation product radionuclides, this information is useful for providing activities on BWR components.

Table 10 presents the BWR coolant piping surface activities for activation nuclides. The activities are presented separately for reactors with a deep-bed demineralizer

TABLE 9. Radioactivity on Steam Generator Manway Inserts

Nuclide	Average Surface Activity ($\mu\text{Ci}/\text{cm}^2$)	Activity Decayed 1000 days ($\mu\text{Ci}/\text{cm}^2$)	Fraction of Total
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For Ginna Nuclear Power Plant - Samples taken 3/75

Total Activity = $9.171 \mu\text{Ci}/\text{cm}^2$

Mn-54	1.8	1.9E-1	2.1E-2
Co-58	1.31E+1	7.5E-4	8.2E-5
Co-60	1.26E+1	8.7	9.49E-1
Zr-95	1.7	3.4E-5	3.7E-6
Cs-137	3.0E-1	2.8E-1	3.1E-2

For Turkey Point #3 Nuclear Power Plant - Samples taken 10/75

Total Activity = $6.822 \mu\text{Ci}/\text{cm}^2$

Mn-54	1.2	1.3E-1	1.9E-2
Co-58	2.81E+1	1.6E-3	2.3E-4
Co-60	9.3	6.5	9.53E-1
Zr-95	1.6	3.2E-5	4.7E-6
Cs-137	2.0E-1	1.9E-1	2.8E-2

**TABLE 10. Corrosion Product Radioactivity
on Boiling Water Reactor Coolant Piping**

Nuclide	Surface Activity ($\mu\text{Ci}/\text{cm}^2$)	Activity Decayed 1000 days ($\mu\text{Ci}/\text{cm}^2$)	Fraction of Total
<i>For Deep-Bed Demineralizer Plant:</i> <i>Total Activity = 3.02 $\mu\text{Ci}/\text{cm}^2$</i>			
Mn-54	2.0	2.1E-1	7.0E-2
Co-58	2.0	1.2E-4	4.0E-5
Co-60	4.0	2.8	9.27E-1
Zn-65	0.2	1.1E-2	3.6E-3
<i>For Powdered-Resin Demineralizer Plant:</i> <i>Total Activity = 0.567 $\mu\text{Ci}/\text{cm}^2$</i>			
Mn-54	0.2	2.1E-2	3.7E-2
Co-58	0.5	2.9E-5	5.1E-5
Co-60	0.7	4.9E-1	8.64E-1
Zn-65	1.0	5.6E-2	9.9E-2

or a powdered-resin demineralizer in the reactor water cleanup system. For both demineralizer types the nuclide, Co-60, contributes the major fraction of between 86% and 93%. The radiozinc activity, Zn-65, shows about a 30 times higher fraction for the powdered-resin demineralizer than for the deep-bed demineralizer. Since these data are for gamma-emitting radionuclides only, the pure beta-emitting radionuclides, Fe-55 and Ni-63, are not included. From the BWR coolant data Table 2), the Fe-55 fraction is about 3 times the Co-60 fraction while the Ni-63 fraction is insignificant. If a proportional amount of Fe-55 is added to the data of Table 10, then the Zn-65 fraction for the deep-bed demineralizer is about 0.096%. For the powdered-resin demineralizer, it would be about 2.7%. This addition of the pure beta-emitting radionuclides does not significantly change these corrosion product nuclide fractions on BWR coolant piping.

4.0 SUMMARY FOR RADIONUCLIDES IN NUCLEAR POWER REACTORS

An inspection has been made of the fractional radionuclide data presented on the previous tables. For the different sample groups of reactor coolant and nuclear reactor components, an itemized list has been developed which identifies the radionuclides with fractional amounts of 1% or greater, less than 1% but 0.1% or greater, and less than 0.1% but 0.01% or greater. This listing is presented in Table 11. At the bottom of Table 11 is a complete listing of all the radionuclides with fractional activity values in the three groupings for any of the nuclear reactor samples.

For the reactor components, all activities refer to surface contaminants except for "Bulk" Shippingport. The "Bulk" Shippingport data were determined from stainless steel samples taken from the reactor core area and thereby represent activities within the metal. The surface activities may tend to be removed with the flux melting process while bulk activities may not. Further experimentation will be needed to clarify this statement. A comparison for the two Shippingport sample groups shown in Table 11 shows that there is not a difference in the significant radionuclides for bulk or surface activities.

As was pointed out when each of the radionuclide data from the samples was presented, from 90% to 99% of the total activities are from the activation nuclides Fe-55, Co-60, and Ni-63. Since only Co-60 is a gamma emitter, the external radiation dose will be determined by the Co-60 activity. However, if any processing of nuclear reactor components occurs which may result in the pure beta emitters and transuranics being taken into the body, then the internal radiation dose from these radionuclides must be considered.

**TABLE 11. Significant Radionuclides
for Nuclear Reactor Components
(Limited to half-lives longer than 50 days)**

<i>Sample Type</i>	<i>Radionuclides Found</i>
<i>Fractions of 1% or greater:</i>	
PWR Coolant	Fe-55, Co-60, Cs-134, and Cs-137.
BWR Coolant	Fe-55, Co-60, and Sb-125.
Trojan	Fe-55, Co-60, and Ni-63.
Rancho Seco	Mn-54, Fe-55, Co-60, Ni-63, Sb-125, and Cs-137.
Shippingport	Fe-55, Co-60, and Ni-63.
"Bulk" Shippingport	Fe-55, Co-60, and Ni-63.
Ginna	Mn-54, Co-60, and Cs-137.
Turkey Point #3	Mn-54, Co-60, and Cs-137.
BWR Pipe (Deep)	Mn-54 and Co-60.
BWR Pipe (Powd)	Mn-54, Co-60, and Zn-65.
<i>Fractions less than 1% but 0.1% or greater:</i>	
PWR Coolant	C-14, Mn-54, Co-57, Ni-63, and Ce-144.
BWR Coolant	Mn-54, Cs-134, and Cs-137.
Trojan	Ru-106, Sb-125, and Pu-241.
Rancho Seco	Co-57, Ni-59, Ru-106, and Ag-110m.
Shippingport	Mn-54, Ni-59, and Nb-93m.
"Bulk" Shippingport	Mn-54, Nb-93m, and Sb-125.
Ginna	None.
Turkey Point #3	None.
BWR Pipe (Deep)	Zn-65.
BWR Pipe (Powd)	None.

TABLE 11. (continued)

Fractions less than 0.1% but 0.01% or greater:

PWR Coolant	Ca-45, Zn-65, Sr-90, Ru-106, Ag-110m, Sb-125, Eu-154, and Eu-155.
BWR Coolant	Ni-63, Zn-65, and Sr-90.
Trojan	Mn-54, Sr-90, Cs-137, and Ce-144.
Rancho Seco	Zn-65 and Cs-134.
Shippingport	None.
"Bulk" Shippingport	Ni-59.
Ginna	None.
Turkey Point #3	Co-58
BWR Pipe (Deep)	None.
BWR Pipe (Powd)	None.

SUMMARY - Significant radionuclides or those with fractional Activities of:

<i>1% or greater</i>	<i>1% to 0.1%</i>	<i>0.1% to 0.01%</i>
Mn-54	C-14	Ca-45
Fe-55	Co-57	Co-58
Co-60	Ni-59	Sr-90
Ni-63	Nb-93m	Eu-154
Zn-65	Ru-106	Eu-155
Sb-125	Ag-110m	
Cs-134	Ce-144	
Cs-137	Pu-241	

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