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CALCULATED RADIONUCLIDE INVENTORIES
FOR STRUCTURAL CORE COMPONENTS IN
THE ADVANCED TEST REACTOR

by

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ABSTRACT

Reactor core physics and depletion/activation calculations were performed to evaluate radionuclide inventories in a wide variety of structural core components from the Advanced Test Reactor. All components were located in or near the active core environment and received significant neutron exposure resulting in the production of radionuclides from neutron activation.

The calculated radionuclide inventory estimates are strongly dependent on the Beginning-of-Life (BOL) elemental constituent and impurity concentrations in the component materials. Therefore, significant effort has been devoted to the determination of best-estimate concentrations for each of the component materials. Materials include stainless steel-304,-347,-348, aluminum-6061, Inconel-600, Inconel-X750, natural hafnium metal, and beryllium metal.

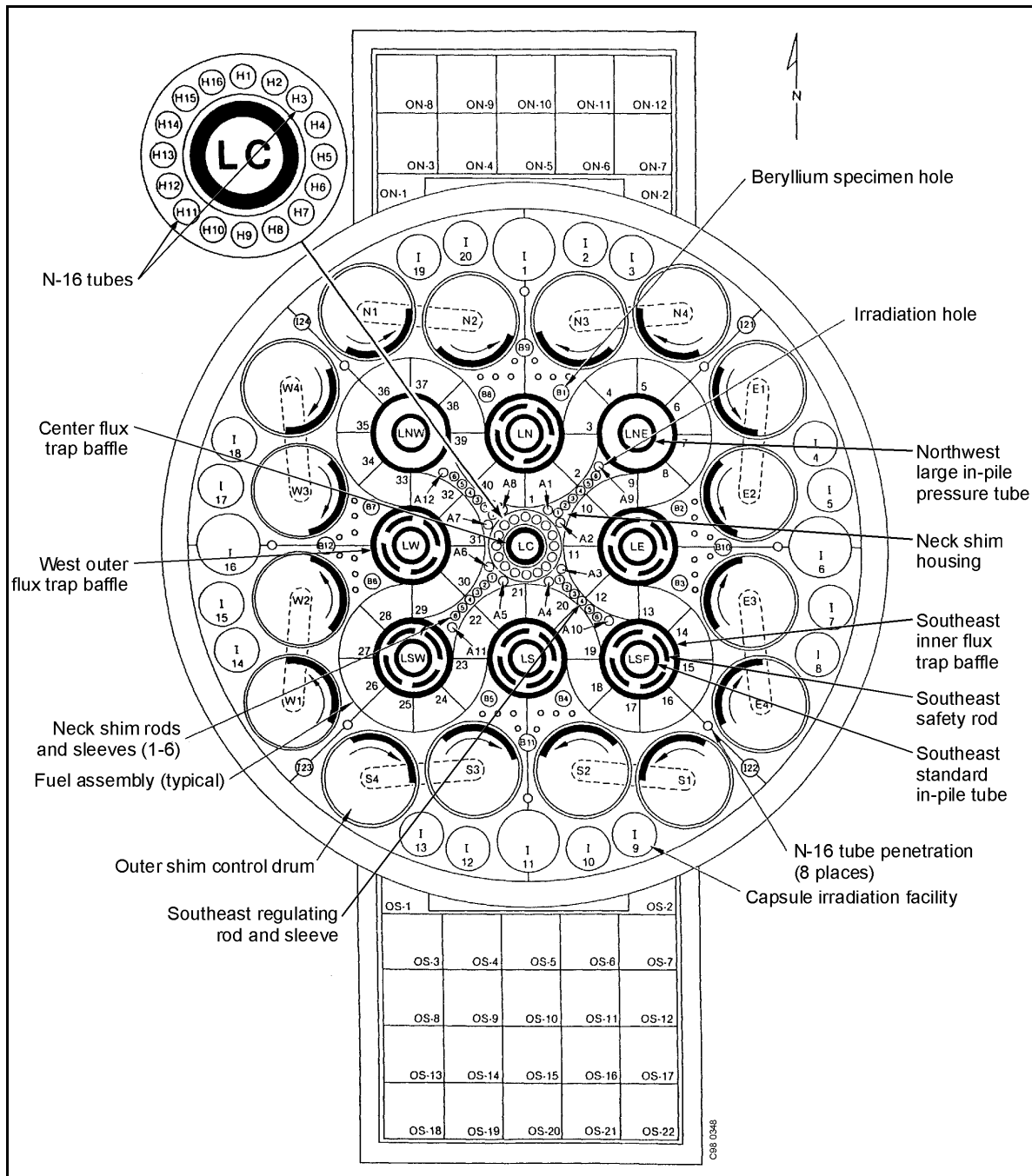
Specific radioactive isotopes of interest include ^{14}C , ^{59}Ni , ^{63}Ni , ^{60}Co , ^{99}Tc , ^{90}Sr , and ^{94}Nb . Radionuclide inventory estimates are presented for a select number of components representing a variety of materials.

INTRODUCTION

Located at the Idaho National Engineering and Environmental Laboratory (INEEL), the Advanced Test Reactor (ATR) is a 250 MW_{th} light water reactor designed specifically to study the effects of intense radiation on reactor fuels and materials. For more than 30 years, the primary role of the ATR has been to serve experimental investigations for the development of advanced nuclear fuels. A variety of test facilities, versatile control systems, and intense neutron flux levels allow for accelerated irradiation tests for a wide range of test specimens.

The ATR core is a complex reactor design with nine individual large volume irradiation flux traps in a 3x3 array within a serpentine driver core configuration as shown in Figure 1. The serpentine driver core is composed of 40 highly-enriched, 4-ft long U-Al_x plate-type fuel elements. The driver core elements are light water cooled and beryllium reflected. In the beryllium reflector, hafnium absorber drums coupled with hafnium shim rods located in the central shim housing control the local power levels in each quadrant of the core.

Currently, the ATR operates at about half the maximum design power rating, or 125 MW_{th} total core power. At this power level, a maximum unperturbed thermal neutron flux of 4.4×10^{14} n/cm²/sec ($E=0.025$ eV), and a fast flux of 3.0×10^{14} n/cm²/sec ($E>1.0$ MeV) can be produced in the flux traps. These fluxes are maintained with relatively constant axial flux profiles over operating power cycles of about 42 days (7-14 day refueling outage between cycles). The axial flux profile is a truncated cosine distribution with a 1.4-1.5 ratio between midplane peak and the axial average flux.



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In order to maintain core component integrity in the high neutron flux environment, periodic component change-outs are required after components reach lifetime fluence accumulation levels. Since initial core criticality in 1968, there have been three major Core Internal Changeouts (CICs): [1] April-August 1977, [2] February-May 1986, and [3] February-August 1994. During each CIC, a variety of structural components are removed from the core.

Until now, the radionuclide inventories in the irradiated stockpile of expended components had not been adequately characterized for disposal. More accurate radionuclide inventory estimates were recently required for these components in order to characterize and classify the components for Department of Energy (DOE) storage and transportation requirements and future component dispositions at the INEEL Radioactive Waste Management Complex (RWMC). The RWMC is the intended site for the ultimate receipt and disposal of those components containing radionuclide concentrations that do not exceed the RWMC's waste disposal acceptance criteria.

Although the current focus at the RWMC recently has been the estimation of the total ^{14}C from disposed reactor components currently at the RWMC, six other radionuclides are also of interest and have been calculated as well, namely: ^{59}Ni , ^{63}Ni , ^{60}Co , ^{99}Tc , ^{90}Sr , and ^{94}Nb . These radionuclides (including ^{14}C) are listed in the Code of Federal Regulations(1) and are the primary radionuclides that regulate acceptable levels of concentrations and inventories at low-level waste disposal facilities. The impact of the six additional radionuclides at the RWMC will be considered in future studies.

Radionuclide estimates in each of the components presented a calculational challenge from the standpoint of the number of variables involved. Computer model variables include component type, length, material composition, component location relative to the nuclear active core region, CIC removal date, decay time, irradiation flux intensity, and neutron cross sections. Some simplification of the calculational process was required to manage the number of computations.

COMPONENT EXPOSURE HISTORY

Since 1968, detailed power histories of the ATR operation have been maintained and were available for the calculations here. These detailed exposure histories list every single ATR power cycle, the cycle duration in days, and the core power levels during the cycle on a daily basis. In any given year, the ATR experiences approximately 6 to 7 separate power cycles followed by a 7-14 day shutdown period for refueling, test specimen changeout, and maintenance.

The detailed power histories were simplified for calculational purposes. All of the individual cycle exposures between CICs were summed to a total core exposure. The total exposures were then divided by the total number of days between CICs in order to develop constant, time-average exposures for the reactor operational time between CICs. These averaged exposures were in turn used to normalize calculated irradiation flux levels in the core.

Table I summarizes the core irradiation history in terms of total core exposure (Megawatt-days), operational time durations between CICs, and the CIC schedules and durations. The decay time given in Table I is the amount of time from the beginning of a CIC to the arbitrary date of January 1, 1998. This is basically the amount of ex-core time for components removed from the start of a particular CIC to January 1, 1998 and is the total time used in the calculations to decay the component radionuclides after removal from the core.

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Table I. ATR Irradiation History and CIC Schedules.

Irradiation Period	CIC No.	Time Period	Core Power or CIC Duration (Days)	Start/End Power Cycles	No. of Power Cycles	Decay Time ^a (Days)	Accumulated Core Exposure (MWD)
1		01-FEB-68 to 11-APR-77	3,357	Initial Criticality to 34C1	115		294,974
	1	12-APR-77 to 08-AUG-77	118			7,570	
2		09-AUG-77 to 02-FEB-86	3,086	35A7 to 72A1	120		305,246
	2	03-FEB-86 to 17-MAY-86	103			4,351	
3		18-MAY-86 to 27-FEB-94	2,843	73A9 to 102B1	90		267,247
	3	28-FEB-94 to 08-AUG-94	161			1,404	

a. Decay time is measured from the end of a particular CIC to January 1, 1998.

STRUCTURAL COMPONENTS

A wide variety of reactor structural components were changed out during the three major ATR CICs. The following list identifies those components and their material compositions that were analytically evaluated for radionuclide inventories:

1. Center Flux Trap Baffle (aluminum mid-section)
2. Outer Flux Trap Baffle (aluminum mid-section)
3. Outer Flux Trap Baffle (Inconel top adaptor)
4. Outer Flux Trap Baffle (stainless steel bottom adaptor)
5. Inner Flux Trap Baffle (aluminum mid-section)
6. Inner Flux Trap Baffle (Inconel top adaptor)
7. Inner Flux Trap Baffle (stainless steel bottom adaptor)
8. Regulating Control Rod (hafnium)
9. Regulating Control Rod Sleeve (Inconel)
10. Neck & Regulating Rod Followers (aluminum)
11. Neck Shim Rod (hafnium)

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12. Neck Shim Rod Sleeve (aluminum)
13. Safety Rod Absorbers (hafnium)
14. Safety Rod Filler Plates (hafnium)
15. Safety Rod Assembly (aluminum)
16. Lower Neck Shim Housing (aluminum)
17. Upper Neck Shim Housing (aluminum)
18. N-16 Tube Assembly in Be Reflector (aluminum)
19. N-16 Tube Assembly in H3/H11 Holes (aluminum)
20. Beryllium Reflector Block (beryllium)
21. Beryllium Control Drum (beryllium)
22. Hafnium Control Drum Shim (hafnium)

Each of these components have specific core locations and are either in the active core region or are just above or below the active core (within 1.5 feet).

The ATR components are constructed of the following eight different materials:

1. Stainless steel 304,
2. Stainless steel 347,
3. Stainless steel 348,
4. Aluminum-6061,
5. Inconel-600,
6. Inconel-X750,
7. Natural hafnium metal, and
8. Beryllium metal.

The elemental constituents and impurities in each of these materials were vital in estimating the radionuclide inventories in each of the components.

MATERIAL COMPOSITIONS

A substantial amount of research was devoted to the development of elemental compositions of these nine structural component materials. Table II gives the best-estimate concentrations of both major elemental constituents and impurities. Concentrations are compiled from a variety of sources: (a) American Society for Testing and Materials (ASTM) handbooks, (b) INEEL Material Specification Requirements for ATR component vendors, (c) vendor-supplied data based on chemical measurements, (d) special chemical test measurements to determine nitrogen, niobium, and nickel concentrations, and (e) other published data.

The best-estimate concentration values given in Table II are intended to represent the average or typical concentrations rather than overly-conservative maximum values. However, some maximum or minimum values are used and prefixed with greater than or less than symbols where best-estimate values are not available. These best-estimate values may be different than concentrations in other similar materials, but are representative of ATR reactor-grade materials. In addition to the precursor concentrations of the seven radionuclides of interest other known precursors of radiologically significant nuclides are also included for completeness.

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Table II. Best-Estimate compositions (wt %) for the ATR structural materials.

Atomic Number	Alloy Elements	2.70 g/cm ³	1.85 g/cm ³	13.3 g/cm ³	8.30 g/cm ³	8.47 g/cm ³	8.02 g/cm ³	8.02 g/cm ³
		Al-6061	Be	Hf	Inconel X-750	Inconel 600	SS304	SS347 or SS348 ^a
1	H	0.000001		≤ 0.0025			0.0007	0.0007
3	Li	0.0005	0.04				0.000013	0.000013
4	Be		97.8 ^b					
5	B	0.02					0.0005	0.0005
6	C	0.02	0.15	≤ 0.015	0.055	0.08	0.07	0.064
7	N	0.0005	0.04	≤ 0.01	0.0054	0.0055	0.047	0.036
8	O	0.05	1.28	≤ 0.04			0.015	0.015
11	Na	≥ 0.00002					0.0037	0.0037
12	Mg	0.90	0.08					
13	Al	97.7 ^b	0.18	≤ 0.01	0.75	1.0	0.01	0.01
14	Si	0.65	0.12		≥ 0.04	0.2	0.60	0.64
15	P	0.001	0.00011	0.01	0.0045	0.0052	0.03	0.025
16	S	0.002	0.04	0.01	0.0005	0.015	0.020	0.019
20	Ca	0.0011	0.04				0.0019	0.0019
22	Ti	0.02		0.01	2.66	2.75	0.05	0.01
23	V	0.02					0.05	0.04
24	Cr	0.05	0.012	0.002	15.37	15.5	18.8	18.0
25	Mn	0.03	0.0074	0.01	0.08	0.5	1.41	1.64
26	Fe	0.20	0.164	≤ 0.025	7.97	8.0	69.0 ^b	67.8 ^b
27	Co	≤ 0.05	0.0016	0.0005	≥ 0.02	≤ 1.0	0.17	0.10
28	Ni	0.007 ^c	0.0283	0.0025	72.0 ^b	76.0 ^b	9.23	10.0
29	Cu	0.25		≥ 0.01	0.03	0.2	0.25	0.14
30	Zn	0.02					0.01	0.01
31	Ga	≤ 0.05					0.045	0.045
33	As						0.01	0.01
34	Se						0.02	0.02
38	Sr	0.00001					0.00002	0.00002
40	Zr	0.02		≤ 4.5			0.002	0.002
41	Nb	≤ 0.0008 ^c	0.000022	0.01	0.97	1.2	0.012	0.87
42	Mo	0.0001	0.001	0.002	0.05	0.04	0.37	0.38
48	Cd	≤ 0.05	0.04					
50	Sn	0.0024					0.01	0.01
51	Sb	0.01					0.01	0.01
72	Hf	≤ 0.05	0.000004	95.3 ^b			0.0002	
73	Ta	≤ 0.05	0.001	0.02	0.01			≤ 0.10
74	W			0.015			0.052	
82	Pb	≤ 0.02					0.002	0.002
92	U			0.001			0.0002	
Total =		100.29	100.02	100.01	100.02	106.50	100.30	100.00

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- Both steels have nearly identical compositions except that Co and Ta are restricted in SS348.
- Values are approximate. These elements represent the balance of the alloy composition.
- Chemical assays of two ATR aluminum alloy 6061 samples showed an average nickel concentration of 70 ppm and detected no niobium in the aluminum alloy.

RADIONUCLIDE PRODUCTION PATHWAYS

Major production pathways leading to our seven radionuclides of interest are shown in Table III. Column 1 lists the component material isotopes; Column 2 lists the nuclear reaction; Column 3 gives the radionuclide produced from the reaction (one of the seven radionuclides of interest). All other potential pathways are relatively minor. The final activation product or fission product concentrations are in general directly proportional to the initial component material isotopic concentrations.

Table III. Major production pathways leading to the seven radionuclides.

Component Material Radionuclide	Nuclear Production Reaction	Radioactive Nuclide
^{14}N	(n,p)	^{14}C
^{17}O	(n, α)	^{14}C
^{13}C	(n, γ)	^{14}C
^{58}Ni	(n, γ)	^{59}Ni
^{62}Ni	(n, γ)	^{63}Ni
^{63}Cu	(n,p)	^{63}Ni
^{66}Zn	(n, α)	^{63}Ni
^{93}Nb	(n, γ)	^{94}Nb
^{94}Mo	(n,p)	^{94}Nb
^{98}Mo	(n, γ)	^{99}Tc
Uranium	(n,fission)	^{99}Tc
^{59}Co	(n, γ)	^{60}Co
^{60}Ni	(n,p)	^{60}Co
^{63}Cu	(n, α)	^{60}Co
$^{60}\text{Co}^*$	(n,n')	^{60}Co
^{88}Sr , ^{89}Sr	2(n, γ)	^{90}Sr
^{93}Zr	(n, α)	^{90}Sr
^{93}Nb , ^{93}Zr	(n,p), (n, α)	^{90}Sr
Uranium	(n,fission)	^{90}Sr

ANALYTICAL METHODOLOGY

The component radionuclide inventory estimates are calculated using a combination of two well-known reactor physics computer codes: the Monte Carlo N-Particle (MCNP4A) neutron transport code(2) and the ORIGEN2 isotopic generation and depletion code(3). A third computer code called MOCUP(4) (MCNP-ORIGEN2-Coupled Utility Programs) was also used to manipulate MCNP and ORIGEN2 input and output data.

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A fully explicit, full-core, three-dimensional MCNP model of the entire ATR core(5) was utilized in order to calculate cell-averaged neutron fluxes, nuclear reaction rates, and neutron cross sections in the components of interest. All model representations are based on actual component vendor specification drawings. Calculated fluxes were normalized to the ATR operating power history to provide component irradiation fluxes for the depletion calculation.

The ORIGEN2 code was used to perform the time-dependent isotopic flux irradiation and depletion/activation calculations. For each component inventory calculation, ORIGEN2 requires the following input data: component isotopic mass, irradiation flux intensity, and neutron reaction cross sections. The reaction cross sections are derived from the MCNP calculation and include six particle reactions: (n, γ), (n,2n), (n,3n), (n,p), (n, α), and (n,fission). ORIGEN2 output automatically provides a full inventory of all generated radionuclides in terms of activity level (curies).

RESULTS

Calculated activity levels of the seven radionuclides are given in Table IV for selected ATR structural components. In addition, the component material, mass, and CIC in which the component was removed from the core are also given. The activity levels have been decay-corrected to January 1, 1998.

CONCLUSIONS

Of particular concern was the calculated ^{14}C inventory and its concentration in the ATR structural components. The largest ^{14}C contributor came from the beryllium reflector blocks (5.38 Ci/block). The second and third most important ^{14}C contributors are the stainless steel (SS348) standard in-pile pressure tubes (1.56 Ci/tube) and the upper neck shim housing (0.353 Ci/housing). Despite being located in a high neutron flux region in the active core, the aluminum and hafnium components contained relatively low concentrations of ^{14}C due to their inherently low levels of dissolved nitrogen and oxygen concentrations.

Both the Inconel-600 large in-pile and the stainless steel (SS348) standard in-pile pressure tubes also contained significant concentrations of ^{59}Ni , ^{63}Ni , ^{60}Co , and ^{94}Nb . The hafnium control rods were the major contributor of ^{99}Tc and ^{90}Sr due primarily from the fission of the uranium (^{235}U) impurity.

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Table IV. Selected Single Component Activity Levels (curies).

Component Description	Primary Material	Mass (grams)	CIC Group	¹⁴ C	⁵⁹ Ni	⁶³ Ni	⁶⁰ Co	⁹⁴ Nb	⁹⁹ Tc	⁹⁰ Sr
Baffle (mid-section)	Al6061	10,902	1	1.42E-02	2.50E-03	9.17E-01	1.43E+02	7.52E-04	9.10E-08	2.68E-07
Baffle (mid-section)	Al6061	10,902	2	1.46E-02	2.50E-03	1.00E+00	4.85E+02	7.66E-04	9.22E-08	3.58E-07
Baffle (mid-section)	Al6061	10,902	3	1.29E-02	2.50E-03	9.92E-01	1.39E+03	7.13E-04	8.75E-08	3.37E-07
Baffle (top adaptor)	X750	5,683	1	1.49E-03	8.29E-01	1.00E+02	1.70E+00	2.01E-02	1.18E-06	2.96E-11
Baffle (top adaptor)	X750	5,683	2	1.54E-03	8.56E-01	1.11E+02	5.84E+00	2.08E-02	1.21E-06	3.93E-11
Baffle (top adaptor)	X750	5,683	3	1.35E-03	7.55E-01	1.04E+02	1.53E+01	1.82E-02	1.07E-06	3.67E-11
Baffle (bottom adaptor)	SS304	10,919	1	8.74E-02	4.40E-01	5.56E+01	4.73E+01	7.81E-04	2.04E-05	1.88E-04
Baffle (bottom adaptor)	SS304	10,919	2	9.06E-02	4.54E-01	6.16E+01	1.63E+02	8.08E-04	2.11E-05	2.46E-04
Baffle (bottom adaptor)	SS304	10,919	3	7.94E-02	4.03E-01	5.78E+01	4.27E+02	7.10E-04	1.86E-05	2.45E-04
Regulating Control Rod	Hafnium	713	1	6.91E-03	2.01E-05	7.30E-03	8.44E-02	6.31E-04	1.01E-06	2.72E-03
Regulating Control Rod	Hafnium	713	2	7.15E-03	2.03E-05	8.05E-03	2.88E-01	6.44E-04	1.01E-06	3.45E-03
Regulating Control Rod	Hafnium	713	3	6.28E-03	1.95E-05	7.67E-03	8.06E-01	5.96E-04	1.02E-06	3.90E-03
Control Rod Sleeve	X750	534	1	2.76E-03	5.78E-01	1.36E+02	7.73E+00	3.92E-02	2.36E-06	4.73E-09
Control Rod Sleeve	X750	534	2	3.98E-03	5.97E-01	1.85E+02	4.67E+01	4.83E-02	2.73E-06	1.20E-08
Control Rod Sleeve	X750	534	3	3.50E-03	5.94E-01	1.83E+02	1.15E+02	4.51E-02	2.61E-06	1.14E-08
Upper Neck Shim Housing	Al6061	146,636	1	3.42E-01	2.87E-02	1.77E+01	2.11E+03	1.34E-02	1.82E-06	2.08E-05
Upper Neck Shim Housing	Al6061	146,636	2	3.53E-01	2.85E-02	1.91E+01	7.13E+03	1.34E-02	1.85E-06	2.78E-05
Upper Neck Shim Housing	Al6061	146,636	3	3.12E-01	2.94E-02	1.94E+01	2.19E+04	1.32E-02	1.75E-06	2.64E-05
Outer Shim Control Drum	Be	58,090	2	4.97E+00	2.87E-02	7.71E+00	9.97E+01	7.90E-05	2.04E-05	3.67E-11
Outer Shim Control Drum	Be	58,090	3	4.97E+00	2.87E-02	8.20E+00	2.94E+02	7.90E-05	2.04E-05	4.46E-11
Reflector Block	Be	83,805	2	5.38E+00	4.34E-02	9.96E+00	1.35E+02	1.04E-04	3.11E-05	NA
Reflector Block	Be	83,805	3	5.38E+00	4.34E-02	1.06E+02	3.98E+02	1.04E-04	3.11E-05	NA
Standard In-Pile Tube	SS348	27,309	1	1.51E+00	4.34E+00	1.01E+02	5.94E+02	1.53E+00	7.22E-04	2.36E-02
Standard In-Pile Tube	SS348	27,309	2	1.56E+00	4.37E+00	1.11E+02	2.03E+03	1.56E+00	7.35E-04	3.03E-02
Standard In-Pile Tube	SS348	27,309	3	1.37E+00	4.26E+00	1.08E+02	5.60E+03	1.42E+00	6.86E-04	3.27E-02
Large In-Pile Tube	600	67,647	1	2.67E-01	5.77E+01	1.03E+04	1.05E+04	3.95E+00	1.76E-04	1.66E-07
Large In-Pile Tube	600	67,647	2	2.76E-01	5.86E+01	1.13E+04	3.58E+04	4.06E+00	1.80E-04	2.21E-07
Large In-Pile Tube	600	67,647	3	2.42E-01	5.49E+01	1.08E+04	9.70E+04	3.66E+00	1.66E-04	2.07E-07

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