Decommissioned Nuclear Reactors Are Hot

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One inescapable fact about decommissioned nuclear reactors; they are extremely radioactive. It's not just the used nuclear fuel that remains radioactive for centuries in dry storage containers. Some metal parts of the reactor itself, called greater than class C waste (GTCC), are as radioactive as nuclear fuel and will have to be disposed of in a deep underground repository along with the used nuclear fuel. Much as Northstar would like to ship all the so-called "low-level waste" from Vermont Yankee to a landfill in Texas, GTCC is not suitable for surface burial. The proposed WCS landfill is not licensed to accept these hot reactor parts; the State of Texas has not received permission from the Nuclear Regulatory Commission to regulate GTCC at the WCS landfill. If Vermont Yankee follows the pattern at other decommissioned reactors, GTCC will be placed in concrete storage containers or casks, much as the used nuclear fuel.

Reactor Waste

For 40 or more years of operation, nuclear utilities leave waste that will remain toxic for centuries. Nuclear fuel is packaged into dry storage canisters that sit in concrete silos (named HI-STORM, Holtec designer), or horizontal concrete boxes (named NUHOMS, Orano TM designer), at 70 or so reactor sites. While the Nuclear Waste Policy Act of 1982, amended 1987, required the Department of Energy (DOE) to construct the Yucca Mountain repository in Nevada, the State strongly objected and the project has stalled. (Disclaimer: the author is an advisor to Nevada on transportation issues.)

While nuclear fuel sits in the reactor for 3 to 4½ years before being placed in a fuel pool, the stainless steel reactor components that hold the fuel within the reactor remain inside the reactor for 40 to 60 years. The stainless steel of these reactor components are activated or become radioactive. Neutron bombardment within the reactor converts non-radioactive cobalt, to radioactive cobalt-60, nickel to radioactive nickel-59, etc. In terms of gamma radiation, used nuclear fuel and activated reactor components are comparable. Gamma rays are electromagnetic radiation, like X-rays, but are more energetic and much more powerful.

In the 1970's, it was commonly understood that shutdown reactors could be sealed for 300 years before being taken apart. But in 1978, four engineering students and I at the State University of New York in Buffalo, in a paper² cautioned that high concentrations of Ni-59 (half-life 75,000 years) meant that reactor parts would be radioactive for tens of thousands of years. This was followed by an analysis of Nb-94 (half-life 20,000 years), in a paper by Cornell Professor Robert Pohl³. An article in Science magazine⁴ credited us with changing the method of safely managing reactor components. In 1978, the NRC classified low-level waste into classes A, B and C, according to the concentration of gamma

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² (Harwood 1976)

³ (Stephen 1978)

⁴ (Science 1982)

emitting and long-lived radionuclides. Classes A, B and C could be buried in a surface landfill, such as the WCS landfill in Texas, where Northstar has partnered with other companies at the WCS facility. At present, under NRC regulations, WCS is not licensed to accept GTCC.

Radioactivity Comparison HBF vs GTCC

In Table 1 below, we compare the total fission products in high burnup fuel (HBF) (60 GWd/MTU) to the activation products in the core shroud of a PWR reactor. Note, we have not included many of the actinides in HBF because we are focused on the radionuclides that are gamma emitters. The fission products in HBF are taken from a PNL report by Roddy⁵; this is the same report used by Holtec in its HI-STORM 100 SAR. The pertinent spreadsheet pages are in the Appendix. As seen, from 10 years on, the core shroud is more radioactive than HBF. The long-lived Ni-59 (half-life 75,000 years) accounts for the long-lived radioactivity in both the core shroud and high burnup fuel. The core shroud also contains the long-lived Nb-94 (half-life 20,000 years), which is a gamma emitter. Reactor internals closest to or part of the reactor core, such as the core shroud, are the most radioactive internal components; other internals have the same activation products, but are less radioactive.

Table 1. Fission and Activation Products (Ci)

<u>Year</u>	<u>1</u>	<u>10</u>	<u>100</u>	<u>500</u>	<u>1000</u>	<u>10000</u>
HBF	6.45E+06	8.05E+05	9.11E+04	132	24.6	17.1
GTCC ^b	$3.33E+06^{a}$	9.13E+05	1.22E+05	9.27E+03	1.98E+03	1.45E+03

^a at reactor shutdown; ^b core shroud GTCC

Direct Gamma Comparison HBF vs GTCC

In Table 2 below, we compare the direct gamma whole body equivalent dose rate on contact (1 cm off surface) from unshielded HBF and the unshielded core shroud GTCC. Note that HBF, 1 year out of reactor is extremely radioactive. A two minute contact gamma dose is likely lethal. The calculations, carried out with Microshield software is at 1 cm from the midpoint of a PWR assembly. At 10 cm from the midpoint, the dose equivalent is even greater. However, note that the gamma dose from the core shroud is more than 10 times greater for the first ten years. From 500 years to 10,000 years the dose rate from GTCC is greater than from HBF due to the presence of Nb-94 and Ni-59.

Table 2. Direct Gamma Dose Equivalent (rem/hr)

Year	1	10	100	500	1000	10000
HBF	2.06E+04	3.0E+03	177	0.0235	0	0
GTCC ^b	$2.44E+05^{a}$	6.03E+04	1.91	0.79	0.776	0.57

^a at reactor shutdown; ^b core shroud GTCC

The calculations in Table 2 for HBF are based on a single PWR HBF assembly. HI-STORM F/W casks contain up to 37 PWR assemblies. A simplifying assumption was also made for the core shroud.

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⁵ (Roddy 1986)

Reactor internals are in many shapes and sizes. To simplify we calculated the gamma dose at 1 cm from a 1m by 1m plate.

Volume Considerations

We differ with the NRC's estimate of the volume of activated metals GTCC that require disposal near surface or in an underground repository. According to the NRC⁶, the total GTCC volume is 880 + 370 = 1250. Based on actual experience, we estimate a volume approximately 3 times larger. At the decommissioned CT Yankee, 3 HI-STORM casks contain GTCC waste. The volume of the HI-STORM 100 canister (diameter 68 inches, length 178 inches) is 10.6 cubic meters. Roughly estimating 3 canisters at approximately 100 reactors, the total volume is approximately 3178 cubic meters, or approximately 3 times greater than NRC estimates.

Summary

Because of the high direct gamma dose rate extending out to 10,000 years, in our judgment, GTCC cannot be buried in a near surface landfill. This conclusion was made by the NRC in 1978 and is just as true today. While this report focuses on the radioactivity and dose rate, it is clear that the declining economics of nuclear power energy is responsible for NRC's move to store GTCC in a surface landfill. While the economics of nuclear power have changed in 39 years, the physics and need to protect the public health and safety remains the same.

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^{6 (}NRC 2019)

References

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Appendix

M Resnikoff, RWMA

Isotopes the Nuclear Industry Overlooked⁷

The problem of what to do with worn-out nuclear power plants has taken on an important new dimension in the past few years, as evidence has come to light that some reactor components may remain radioactive for thousands of years after a plant is shut down. The conventional wisdom had previously been that radiation levels would decline to insignificance after several decades.

The culprits are very long lived isotopes of nickel and niobium, which are formed as the result of bombardment by neutrons. The formation of these isotopes was overlooked by the nuclear industry until the late 1970, when the problem was brought to public attention largely as the result of work by undergraduate students.

Their discovery may have an important impact on regulations governing the decommissioning of nuclear plants. In particular, the Nuclear Regulatory Commission (NRC) may forbid utilities to entomb reactors in concrete and leave them in place-an option that was long considered the cheapest way of dealing with the problem. Instead, the NRC staff is considering requiring that reactors be dismantled relatively soon after they are shut down and that the radioactive waste be shipped to a disposal site (see accompanying story). Components containing the long-lived isotopes may even have to be consigned to a geological repository when one is eventually established.

When a reactor is first shut down, the pressure vessel and other components close to the core are intensely radioactive, largely because of the presence of cobalt-60. This isotope is formed when atoms of cobalt, a constituent of most steels, are hit by neutrons from fission reactions in the reactor fuel. Because cobalt-60 has a half-life of 5.27 years, the radioactivity diminishes relatively quickly. After a century, the amount of cobalt-60 will have dropped by a factor of about one million.

Although it has always been known that isotopes of other elements would be formed by neutron bombardment, it was thought that they would be present in such tiny quantities that they would contribute negligible amounts of radioactivity. Thus, once the cobalt-60 had decayed, the reactor would be relatively harmless. In February 1976, however, Marvin Resnikoff, a physicist then on the staff of the New York Public Interest Research Group, went public with calculations indicating that nickel-59 may pose a long-term radiation problem.

Resnikoff says that he and four undergraduate students realized that nickel-59 may cause difficulties when they looked at data on the dismantling of the Elk River reactor, a small power plant in Minnesota that was shut down in 1968 after only 4 years of operation. Although only trace amounts of nickel-59 were present in Elk River components, Resnikoff calculated that significant quantities would be formed in a large power reactor during 30 years of operation.

Nickel-59 is potentially important because, although it contributes only a tiny fraction of the radiation inventory when a reactor is shut down, it has a half-life of about 80,000 years. It will therefore be around long after cobalt-60 has decayed to insignificance, giving off radiation well above permitted levels.

Resnikoff recalls that he was initially anxious about releasing his calculations because "they went against the whole mindset at the time." The nuclear industry was then saying that if a reactor is entombed for 180 years, it will cool down to a safe level, he pointed out. Nevertheless, he published a press release challenging the industry's plans. Resnikoff says that his calculations were vigorously attacked by the industry, but most studies since then have acknowledged the problem with nickel-59. "It is an example of what happens when you have thousands of engineers all moving in one direction, and a handful of outside critics takes a look at their work," Resnikoff claims.

A year later, a second long-lived isotope, niobium-94 was identified as a potential problem in irradiated reactor components. Again, the discovery came from researchers outside the nuclear industry.

Robert Pohl, a professor of physics at Cornell University, said that he decided, in the light of Resnikoff's findings, to see whether there are any hazardous activation products among trace elements in steel. An undergraduate student, John Stephens, looked through data on radioactive isotopes and flagged niobium-94 as a potential problem. It decays with a half-life of 20,300 years, emitting very energetic gamma rays. A literature search indicated that niobium is added to some steels to inhibit cracking, and that it is a trace constituent in stainless steel. Pohl and Stephens published their findings in *Nuclear Engineering and Design* in 1978.

"Nobody in the nuclear business knew of the problem at the time," says Pohl. It is now generally accepted, however. A 1980 report by Battelle Pacific Northwest Laboratories indicates, for example, that the decay of niobium-94 will dominate the radiation dose rate from irradiated steel about 70 years after a reactor is shut down.

An environmental impact statement on reactor decommissioning, published last year by the NRC, indicates that the dose rate from niobium-94 in reactor components will be about 17,000 rems per year if the reactor is operated for 30 to 40 years. That from nickel-59 will be about 800 rems per year. "These dose levels are substantially above acceptable residual radioactivity levels," the statement notes. Entombing a disused reactor in concrete would thus be acceptable only if the long-lived isotopes were removed or if the integrity of the entombing structure could be maintained for thousands of years, the study concludes.

After the problems with nickel-59 and niobium-94 were discovered, the NRC commissioned a study to see whether any

⁷ (Science 1982) Science, p. 377, January 22, 1982, p. 377.

other potential activation products may cause trouble. "So far, we haven't identified any on the scale of those two," says Donald Calkins, NRC's manager of decommissioning programs.- Colin Norman.

TABLE C.1-3. Radioactivity Levels in Major Activated Reactor Components at Time of Reactor Shutdown

			* *			2		
			,	Core Mid-P	lane Radioactivit	ies (Ci/m³) Lower 5.02 m	linner	lower / \
9		W Charles	Lower 4.72 m of Core Barrel	Thermal Shields	Vessel Inner Cladding	of Vessel Wall	Upper Grid Plant (a)	Grid Plate(a)
Isotope	Half-Life	Shroud	Of Core barrer	31110143			/	
95 _{Nb}	35 day	2.0×10^{3}	7.6 x 10 ⁰	3.5×10^{0}	5.6×10^{-3}	1.7 x 10 ⁻³		
59 _{Fe}	45 day	4.6 x 10 ⁴	4.4×10^3	2.0×10^{3}	1.0×10^{2}	2.7×10^{1}		
58 _{Co}	72 day	1.5 x 10 ⁵	1.0 x 10 ⁴	4.6×10^3	3.3×10^2	6.6 x 10 ⁰		
95 _{Zr}	65 day	1.1×10^{-1}	6.2×10^{-3}	2.9×10^{-3}	2.0×10^{-4}	7.2 x 10 ⁻⁴		
65 _{Zn}	245 day	1.2×10^{2}	1.1×10^{0}	5.0×10^{-1}	6.7×10^{-4}	3.5×10^{-5}		
54 _{Mn}	√300 day	6.8 x 10 ⁴	3.7×10^3	1.7×10^{3}	1.2×10^2	4.7×10^{1}		
55 _{Fe}	2.7 yr	1.3 x 10 ⁶	1.5 x 10 ⁵	6.7×10^4	3.5×10^{3}	7.2×10^2		
		9.6 x 10 ⁵		4.7×10^4	2.5×10^{3}	7.5×10^{1}		
60 _{Co} (b) upper	5.27 yr	9.6 x 10 ⁵ 7	3.1 x 10 ⁴	1.6 x 10 ⁴	8.2 × 10 ²	2.5×10^{1}		
lower		3.2 x 10 ,	3.1 X 10			3.8×10^{0}		
63 _{Ni}	~100 yr	1.2 x 10 ⁵	1.5 × 10 ⁴	6.8 x 10 ³	3.6×10^{2}			
93 _{Mo}	√3000 yr	3.6 x 10 ⁻¹	5.2 x 10 ⁻²	2.4 x 10 ⁻²	1.2 x 10 ⁻³	1.3 x 10 ⁻³		
14 _C	√5,750 yr	1.5 x 10 ²	1.8 x 10 ¹ -	8.3×10^{0}	4.0×10^{-1}	1.9 x 10 ⁻²		
94 _{Nb}	~20,000 yr	5.4×10^{0}	2.6×10^{-1}	1.2 x 10 ⁻¹	9.5×10^{-3}			
59 _{Ni}	√80,000 yr	2 -	1.3 x 10 ²	5.0×10^{1}	3.0×10^{0}	3.2×10^{-2}		2.97 x 10 ⁶
Sum (Ci/m ³)		2.97 x 10 ⁶	3.07 x 10 ⁵	1.45 x 10 ⁵	7.73×10^3	9.04×10^2	2.97 x 10 ⁶ 0.003 x 4.74 ^(c)	0.08 x 4.74(c)
Average/Peak		0.755	0.637	0.778	0.637	0.637	0.003 x 4.74	1.403 x 10 ²
Ci/kg(d)		2.787×10^{2}	2.433 x 10 ¹	1.403 x 10		7.164 x 10 ⁻²	5.254 x 10 ⁰	
UI/Kg		12,312	26,783	10,413	2,074	245,582	4,627	3.946
Weight of Material (kg)		3.431×10^6		1.461 x 10	1.581 x 10 ³	1.759 x 10 ⁴	2.431 x 10 ⁴	5.534 x 10 ⁵
Sum (Ci)(e)					4 826 x 100-	Curies		
TOTAL - Radioactivity					1.786 × 10 ¹⁷	becquerels		

⁽b) Upper and lower bounds were computed using the maximum and minimum levels of ⁵⁹Co contaminant in the materials. All totals were computed using the upper bound values.

⁽c) Activity (Plate Average) = 4.74, Activity (Shroud at Plate Location) Activity (Plate Edge) = 4.74, Activity (Shroud at Axial Midplane) = {0.005, upper plate}

⁽d) Conversion factor assumes stainless steel density of 8.038 x $10^3~{\rm kg/m}^3$ (0.29 lb/in. 3).

⁽e) The number of significant figures carried is for computational accuracy and does not imply precision to four places.

Core									
Shroud	PWR								
			_	Reactor					10000
Isotope	Half-Life	Units	LN2/τ	Shutdown	10 yrs	100 yrs	500 yrs	1000 yrs	yrs
Nb-95	35	day	0.0198042	4.00E+03	1.62E-28	0.00E+00			
						6.24E-			
Fe-59	45	day	0.0154033	9.20E+04	3.52E-20	240			
						7.44E-			
Co-58	72	day	0.009627	3.00E+05	1.65E-10	148			
						2.01E-			
Zr-95	65	day	0.0106638	2.20E-01	2.74E-18	170			
Zn-65	245	day	0.0028292	2.40E+02	7.86E-03	3.41E-43			
Fe-55	300	day	0.0023105	1.36E+05	2.96E+01	3.22E-32			
Co-60									
upper	5.27	yr	0.131527	1.92E+06	5.15E+05	3.73E+00	5.28E-23	1.45E-51	0.00E+00
Co-60									
lower	5.27	yr	0.131527	6.40E+05	1.72E+05	1.24E+00	1.76E-23	4.84E-52	0.00E+00
Ni-63	100	yr	0.0069315	2.40E+05	2.24E+05	1.20E+05	7.50E+03	2.34E+02	1.89E-25
Mo-93	3000	yr	0.000231	7.20E-01	7.18E-01	7.04E-01	6.41E-01	5.71E-01	7.14E-02
C-14	5750	yr	0.0001205	3.00E+02	3.00E+02	2.96E+02	2.82E+02	2.66E+02	8.99E+01
Nb-94	20000	yr	3.466E-05	1.08E+01	1.08E+01	1.08E+01	1.06E+01	1.04E+01	7.64E+00
Ni-59	80000	yr	8.664E-06	1.48E+03	1.48E+03	1.48E+03	1.47E+03	1.47E+03	1.36E+03
			Total	3.33E+06	9.13E+05	1.22E+05	9.27E+03	1.98E+03	1.45E+03

Isoto pe	60 GWd	/MTU	60 GWd/MTU					PWR 60 GWd/M TU					
	W/MTU		Ci/MTU					microCi /cc					
	Time	since	Time	since		Half-lifet	In(2)/	1 yr	10yr	100 yr	500 yr	1000	10000
	discharg	e (yr)	discharg	e (yr)		t (yr)	t					yr	yr
	1	10	1	10									
C-14			2.44E+	2.44E+	yr	5.73E+0	1.21E	6.34E+0	6.33E+	6.26E+	5.97E+	5.62E+	1.89E+
			00	00		3	-04	0	00	00	00	00	00
Mn- 54			4.59E+ 02		day	3.12E+0 2	2.22E -03	1.19E+0 3	1.17E+ 03				
Fe-55			3.24E+ 03	4.76E+ 02	day	3.00E+0 2	2.31E -03	8.42E+0 3	2.20E+ 02				
Co-60	1.47E+	4.50E+	9.54E+	2.92E+	yr	5.27E+0	1.32E	2.48E+0	6.65E+	4.80E-			
	02	01	03	03		0	-01	4	03	02			
Ni-59			6.40E+	6.40E+	yr	7.60E+0	9.12E	1.66E+0	1.66E+	1.66E+	1.66E+	1.65E+	1.52E+
			00	00		4	-06	1	01	01	01	01	01
Ni-63			1.05E+	9.83E+	yr	9.46E+0	7.33E	2.73E+0	2.54E+	1.31E+	7.00E+	1.80E+	4.15E-
			03	02		1	-03	3	03	03	01	00	29
Zn-65			4.78E+		day	2.45E+0	2.83E	1.24E+0	7.99E+				
			01			2	-03	2	01				
Zr-95	1.48E+		2.93E+		day	6.50E+0	1.07E	7.61E+0	2.31E+				
	02		04			1	-02	4	01				
Nb-								0.00E+0					
94								0					
Nb-	3.16E+		6.59E+		day	3.50E+0	1.98E	1.71E+0	3.72E+				
95	02		04			1	-02	5	01				
Ru-			2.84E+		day	3.93E+0	1.77E	7.38E+0	0.00E+				
103			03			1	-02	3	00				

Ru-	2.28E+		3.84E+	7.88E+	yr	1.01E+0	6.88E	9.98E+0	1.03E+	1.36E-			
106	01		05	02	-	0	-01	5	03	24			
Rh-	3.68E+	7.56E+	3.84E+	7.88E+	yr	1.01E+0	6.88E	9.98E+0	1.03E+	1.36E-			
106	03	00	05	02		0	-01	5	03	24			
Cs-	2.66E+	1.29E+	2.62E+	1.27E+	yr	2.06E+0	3.36E	6.81E+0	2.36E+	1.69E-			
134	03	02	05	04		0	-01	5	04	09			
Cs-	1.97E+	1.60E+	1.78E+	1.44E+	yr	2.94E+0	2.36E	4.62E+0	3.65E+	4.39E+	3.55E+	2.73E-	
137	02	02	05	05		1	-02	5	05	04	00	05	
Ba-	6.60E+	5.36E+	1.68E+	1.37E+	yr	3.06E+0	2.27E	4.36E+0	3.48E+	4.53E+	5.23E+	6.26E-	
137m	02	02	05	05		1	-02	5	05	04	00	05	
Ce-	2.84E+		4.29E+	1.42E+	yr	7.78E-01	8.90E	1.11E+0	1.51E+	2.39E-			
144	02		05	02			-01	6	02	33			
Pr-	3.15E+		4.29E+	1.42E+	yr	7.78E-01	8.90E	1.11E+0	1.51E+	2.39E-			
144	03		05	02			-01	6	02	33			
Pr-			5.14E+	1.70E+	yr	7.78E-01	8.90E	1.34E+0	1.81E+	2.84E-			
144m			03	00			-01	4	00	35			
Pm-	3.37E+	3.12E+	9.39E+	8.71E+	yr	2.62E+0	2.64E	2.44E+0	1.74E+	8.19E-			
147	01	00	04	03		0	-01	5	04	07			
Sm-			5.30E+	4.95E+	yr	9.13E+0	7.59E	1.38E+0	1.28E+	6.45E+	3.09E+	6.95E-	1.48E-
151			02	02		1	-03	3	03	02	01	01	30
Eu-	2.09E+	1.01E+	2.33E+	1.13E+	yr	8.62E+0	8.04E	6.05E+0	2.71E+	1.95E+	2.10E-		
154	02	02	04	04		0	-02	4	04	01	13		
Eu-			1.42E+	4.05E+	yr	4.97E+0	1.39E	3.69E+0	9.15E+	3.26E-			
155			04	03		0	-01	4	03	02			
Other	7.25E+	7.00E+	7.55E+	2.29E+		1.78E+0	3.88E	1.96E+0	1.06E+				
-FP	01	00	03	02		0	-01	4	02				
Other			6.47E+	4.16E+		1.41E+0	4.91E	1.68E+0	1.92E+				
-AC			01	01		1	-02	2	01				
TOTA	1.16E+	9.89E+					Total	6.45E+0	8.05E+	9.11E+	1.32E+	2.46E+	1.71E+
L	04	02						6	05	04	02	01	01