## Why is High Level Nuclear Waste So Dangerous for So Long?

by Gordon Edwards PhD

The high level waste (that is, irradiated nuclear fuel) from nuclear reactors remains extremely radiotoxic for many millions of years -- essentially forever. I have been asked to explain why this is so.

There are hundreds of human-made radioactive materials (radionuclides) within irradiated nuclear fuel. Many of these disappear within the first few years, but even after ten years of "cooling" and rapid decay, there are still hundreds of such radionuclides left. Here is a list of 211 of them, taken from a publication by AECL (Atomic Energy of Canada Ltd): <u>http://www.ccnr.org/hlw\_chart.html</u>

## Chart of 211 Radioactive Poisons in 10-Year Old CANDU Spent Fuel

The following <u>chart</u> identifies 211 radioactive poisons that are present in every ten-year old irradiated CANDU fuel bundle. The list is not complete.

These data, compiled from <u>AECL-9881</u>, refer to the radioactive contents of an irradiated fuel bundle from the Bruce A reactors, ten years after discharge.

The origin (manner of creation) of each radioactive poison is indicated in the chart:

- **F.P. indicates** "Fission Products": these atoms are made from the broken pieces of heavier atoms that were split or fissioned in the reactor core to produce energy [fission products are also created when an atomic bomb explodes].
- F.I.A.P. indicates "Fuel Impurity Activation Products": during fission, non-radioactive impurities in the fuel become radioactive by absorbing neutrons.
- **Z.A.P. indicates** "**Zircaloy Activation Products**": non-radioactive elements in the zirconium sheath also become radioactive by absorbing neutrons.
- "Actinides" refer to some of the heavier decay products of uranium as well as the transuranic (heavier-than-uranium) elements created during fission.

The activity (in "becquerels") of each radioactive poison is only roughly indicated:

- a single yen-sign ¥ indicates the presence of a particular radioactive poison;
- a triple yen-sign ¥ ¥ ¥ indicates over a million becquerels of that radionuclide
  per kg of uranium fuel (for FP, FIAP, and Actinides) or
  - per kg of zirconium alloy (for ZAP).

The list is organized according to the electric charge on the nucleus (the socalled "atomic number"), from the smallest charge (hydrogen-3, also known as "tritium", with a charge of one unit) to the largest charge (californium-252, with a charge of 98). This agrees with the order of the elements in the <u>periodic table</u>.

Within each chemical species, the radioactive varieties (called "isotopes" or "nuclides") are organized according to the mass of the nucleus, indicated by the accompanying number in the following chart, called the "atomic mass number".

Standard Chemical Symbol	Common Name of element	Atomic Mass Number	F.P. Fission Product	F.I.A.P. Activation Product	Z.A.P. Activation Product	Actinide (includes progeny)
H (T)	Hydrogen (Tritium)	3	¥¥¥	¥	¥	
Be	Beryllium	10		¥	¥	
C	Carbon	14		¥¥¥	¥¥¥	
Si	Silicon	32		¥	¥	
Р	Phosphorus	32		¥	¥	
S	Sulphur	35		¥		
CI	Chlorine	36		¥		
Ar	Argon	39		¥	¥	
Ar	Argon	42		¥	¥	
К	Potassium	40		¥		
K	Potassium	42			¥	
Ca	Calcium	41		¥		
Ca	Calcium	45			¥	
Sc	Scandium	46		¥		
V	Vanadium	50			¥	
Mn	Manganese	54		¥	¥¥¥	
Fe	Iron	55		¥¥¥	¥¥¥	
Fe	Iron	59			¥	

Standard	Common	Atomic	F.P.	F.I.A.P.	Z.A.P.	Actinide
Chemical	Name of	Mass	Fission	Activation	Activation	(includes
Symbol	element	Number	Product	Product	Product	, progeny)
Ni	Nickel	59		¥	¥¥¥	
Ni	Nickel	63		¥¥¥	¥¥¥	
Zn	Zinc	65		¥	¥	
Se	Selenium	79	¥¥¥			
Kr	Krypton	81	¥			
Kr	Krypton	85	¥¥¥			
Rb	Rubidium	87	¥			
Sr	Strontium	89	¥		¥	
Sr	Strontium	90	¥¥¥	¥	¥	
	Mitalaan					
Y	Yttrium	90	¥ ¥ ¥	¥	¥	
Y	Yttrium	91	¥		¥	
Zr	Zirconium	93	¥¥¥	¥	¥¥¥	
Zr	Zirconium	95	¥	¥	¥	
Nb	Niobium	92			¥	
Nb	Niobium	93m	¥¥¥	¥	¥¥¥	
Nb	Niobium	94	¥	¥	¥¥¥	
Nb	Niobium	95	¥	¥	¥	
Nb	Niobium	95m	¥		¥	
Мо	Molvbdenum	93		¥	¥	
Тс	Technetium	99	¥¥¥	¥	¥	
Ru	Ruthenium	103	¥			
Ru	Ruthenium	106	¥¥¥			
Rh	Rhodium	103m	¥			
Rh	Rhodium	106	¥¥¥			
Pd	Palladium	107	¥¥¥			
Ag	Silver	108	¥	¥	¥	
Ag	Silver	108m	¥	¥¥¥	¥	
Ag	Silver	109m	¥	¥	¥	
Ag	Silver	110	¥	¥	¥	
Ag	Silver	110m	¥	¥	¥	

Standard	Common	Atomic	F.P.	F.I.A.P.	Z.A.P.	Actinide
Chemical	Name of	Mass	Fission	Activation	Activation	(includes
Symbol	element	Number	Product	Product	Product	, progeny)
In	Indium	113m			¥	
In	Indium	114	¥	¥	¥	
In	Indium	114m			¥	
In	Indium	115			¥	
Sn	Tin	113			¥	
Sn	Tin	117m	¥	¥	¥	
Sn	Tin	119m	¥¥¥		¥¥¥	
Sn	Tin	121m	¥		¥¥¥	
Sn	Tin	123	¥		¥	
Sn	Tin	125	¥¥¥		¥	
Sn	Tin	126				
Sb	Antimony	124	¥		¥	
Sb	Antimony	125	¥¥¥		¥¥¥	
Sb	Antimony	126	¥		¥	
Sb	Antimony	126m	¥¥¥			
	····· <b>,</b>					
Те	Tellurium	123	¥		¥	
Те	Tellurium	123m	¥		¥	
Те	Tellurium	125m	¥¥¥		¥¥¥	
Те	Tellurium	127	¥		¥	
Те	Tellurium	127m	¥		¥	
I	lodine	129	¥		¥	
Cs	Cesium	134	¥			
Cs	Cesium	135	¥¥¥			
Cs	Cesium	137	¥¥¥			
Ba	Barium	137m	¥¥¥			
	Lanthonum	129	· · · · · · · · · · · · · · · · · · ·		I	1
La	Lannahum	130				
Ce	Cerium	142	¥			
Ce	Cerium	144	¥¥¥			
Pr	Praseodymium	144	¥¥¥			
Pr	Praseodymium	144m	¥¥¥			
1	I	I	1	1	I	L

Standard	Common	Atomic	F.P.	F.I.A.P.	Z.A.P.	Actinide
Chemical	Name of	Mass	Fission	Activation	Activation	(includes
Symbol	element	Number	Product	Product	Product	, progeny)
Nd	Neodymium	144	¥			
Dm	Drom othium					
Pm	Prometnium	147	+++			
Sm	Samarium	147	¥			
Sm	Samarium	148	¥	¥		
Sm	Samarium	149	¥			
Sm	Samarium	151	¥¥¥			
Eu	Europium	152	¥¥¥	¥		
Eu	Europium	154	¥¥¥	¥		
Eu	Europium	155	¥¥¥	¥		
Gd	Gadolinium	152	¥	¥		
Gd	Gadolinium	153	¥	¥		
Tb	Terbium	157		¥		
Tb	Terbium	160		¥		
Dy	Dysprosium	159		¥		
Но	Holmium	166m	¥	¥		
Tm	Thulium	170		¥		
Tm	Thulium	171		¥		
		4.50				 
Lu	Lutetium	1/6			¥	
Lu	Lutetium	1/6			¥ V	
LU	Lutetium	1/6			Ť	
L F	Hafnium	175		I	· · · · · · · · · · · · · · · · · · ·	I
	Hafnium	1/0			T V	
LIF	Hafnium	182			Y Y	
	mannum	102				
Та	Tantalum	180			¥	
Та	Tantalum	182			¥	
W	Tungsten	181	-		¥	
W	Tungsten	185			¥	
W	Tungsten	188			¥	

	-		·			
Standard	Common	Atomic	<b>F.P.</b>	F.I.A.P.	Z.A.P.	Actinide
Chemical	Name of	Mass	Fission	Activation	Activation	(includes
Symbol	element	Number	Product	Product	Product	progeny)
		_				
Re	Rhenium	187			¥	
Re	Rhenium	188			¥	
		_	 	-	 	
Os	Osmium	194			¥	
 				-		 
Ir	Iridium	192			¥ V	
Ir	Iriaium	192m			¥	
Ir	Iridium	194			¥ V	
Ir	Iriaium	194m			Ŧ	
D+	Plotinum	102	-	-		-
FL	Flatiliulii	193			T	
 ТI	Thallium	206	-	-	¥	
ті	Thallium	207				¥
TI	Thallium	208				¥
ті	Thallium	209				¥
						· ·
Pb	Lead	204			¥	
Pb	Lead	205			¥	
Pb	Lead	209				¥
Pb	Lead	210				¥
Pb	Lead	211				¥
Pb	Lead	212				¥
Pb	Lead	214				¥
Bi	Bismuth	208	- -	-	¥	-
Bi	Bismuth	210			¥	¥
Bi	Bismuth	210m				¥
Bi	Bismuth	211				¥
Bi	Bismuth	212				¥
Bi	Bismuth	213				¥
Bi	Bismuth	214				
Ро	Polonium	210			¥	¥
Ро	Polonium	211				¥
Ро	Polonium	212				¥
Ро	Polonium	213				¥
Ро	Polonium	214				¥
Ро	Polonium	215				¥
Ро	Polonium	216				¥
Ро	Polonium	218				¥

Standard	Common	Atomic	F.P.	F.I.A.P.	Z.A.P.	Actinide
Chemical	Name of	Mass	Fission	Activation	Activation	(includes
Symbol	element	Number	Product	Product	Product	progeny)
At	Astatine	217				¥
Bn	Badon	219				¥
Bn	Radon	220				¥
Rn	Radon	222				¥
Fr	Francium	221				¥
Fr	Francium	221				¥
Ra	Radium	223				¥
Ra	Radium	224				¥
Ra	Radium	225				¥
На	Radium	226				¥
На	Radium	228				¥
Ac	Actinium	225				¥
Ac	Actinium	227				¥
Ac	Actinium	228				¥
Th	Thorium	227				¥
Th	Thorium	228				¥
Th	Thorium	229				¥
Th	Thorium	230				¥
Th	Thorium	231				¥
Th	Thorium	232				¥
Th	Thorium	234				¥¥¥
Ра	Protactinium	231				¥
Ра	Protactinium	233				¥¥¥
Ра	Protactinium	234				¥
Ра	Protactinium	234m				¥¥¥
U	Uranium	232				¥
U	Uranium	233				¥
U	Uranium	234				¥¥¥
U	Uranium	235				¥
U	Uranium	236				¥¥¥
U	Uranium	237				¥¥¥
U	Uranium	238				¥¥¥
U	Uranium	240				¥
Np	Neptunium	237				¥¥¥
Np	Neptunium	238				¥
Np	Neptunium	239				¥¥¥
Np	Neptunium	240				¥
Np	Neptunium	240m				¥

Standard	Common	Atomic	F.P.	F.I.A.P.	Z.A.P.	Actinide
Chemical	Name of	Mass	Fission	Activation	Activation	(includes
Symbol	element	Number	Product	Product	Product	progeny)
Pu	Plutonium	236				¥
Pu	Plutonium	238				¥¥¥
Pu	Plutonium	239				¥¥¥
Pu	Plutonium	240				¥¥¥
Pu	Plutonium	241				¥¥¥
Pu	Plutonium	242				¥¥¥
Pu	Plutonium	243				¥
Pu	Plutonium	244				¥
Am	Americium	241				¥¥¥
Am	Americium	242				¥¥¥
Am	Americium	242m				¥¥¥
Am	Americium	243				¥¥¥
Am	Americium	245				¥
Cm	Curium	242				¥¥¥
Cm	Curium	243				¥¥¥
Cm	Curium	244				¥¥¥
Cm	Curium	245				¥
Cm	Curium	246				¥
Cm	Curium	247				¥
Cm	Curium	248				¥
Cm	Curium	250				¥
Bk	Berkelium	240				¥
	Berkelium	243				T Y
	Derkenum	200				<b>T</b>
Cf	Californium	249				¥
Cf	Californium	250				¥
Cf	Californium	251				¥
Cf	Californium	252				¥

AECL-9881. J. C. Tait, I. C. Gould, and G. B. Wilkin. Derivation of Initial Radionuclide Inventories for the Safety Assessment of the Disposal of Used CANDU Fuel. AECL Whiteshell Nuclear Research Establishment, August, 1989.

This list is by no means complete.

You will notice that the radioactive waste materials in irradiated nuclear fuel are classified into 4 categories by AECL. Those categories are: Fission Products, Activation Products (2 types abbreviated as FIAP and ZAP), and Actinides.

**"Fission products" (FP)** are atoms formed from the broken pieces of heavier atoms that have been fissioned ("split") in the core of the reactor, along with their radioactive decay products. These fragments are generally about 1/3 or 2/3 the "size" (mass) of the original atoms of uranium or plutonium from which they were produced by fission. For example, the fission of uranium-235 gives rise to strontium-90 and cesium-137; the first is roughly 1/3 and the second roughly 2/3 of the mass of the original uranium atom, with its 235 atomic mass units.

"Activation Products" (AP) are radioactive atoms created when a nonradioactive atom absorbs a neutron and becomes unstable. Thus non-radioactive cobalt-59 becomes intensely radioactive cobalt-60, and non-radioactive iron-54 becomes radioactive iron-55. These activation products are the main reason why metal components and other debris from the core area of a nuclear reactor cannot be recycled but must be treated as radioactive waste. The fuel itself also contains troublesome activation products from impurities in the fuel (FIAP) and impurities in the zirconium fuel cladding (ZAP).

"Actinides" have radioactive atoms with an atomic number higher than that of Actinium. The term includes some naturally occurring radioactive materials (e.g. uranium and thorium) as well as human-made radioactive materials like neptunium, plutonium, americium, and curium, all of them heaver than uranium. Human-made actinides are created when a naturally-occurring actinide such as uranium-238 or thorium-232 absorbs one or several neutrons, followed by one or more "beta decays" by which they are transmuted into atoms of higher atomic number than uranium's 92 (the highest naturally-occurring atomic number).

Those man-made actinides that are beyond uranium in the periodic table are often called transuranic elements (TRU). The graphic reproduced below, taken from a 1978 publication of the US Geological Survey, shows the radiotoxicity of selected radionuclides in irradiated nuclear fuel over a period from a thousand years to ten million years after discharge, compared with the radiotoxicity of the associated mill tailings (i.e. the leftovers from mining the uranium needed to produce the same amount of energy as the nuclear fuel did) over the same time period: <u>http://ccnr.org/usgs.html</u>.

## How Toxic is Nuclear Waste Over 10 Million Years?

The following chart is taken from a very interesting circular published by the US Geological Survey on the subject of High Level Radioactive Wastes (HLW). In order to compare the toxicity of HLW with (for example) uranium mill tailings, it is necessary to have a crude measure of the toxicity of radioactive materials in general. This is provided by using drinking water standards, which specify -- for each radioactive substance -- the maximum concentration that is permissible in drinking water. Thus a crude measure of toxicity can be obtained by calculating the amount of water needed to dilute a given quantity of radioactive material to the maximum permissible level of radioactive pollution that is legally permitted for drinking water. When this is done, it can be seen that -- after the first 1000 years or so -- uranium mill tailings are in fact more hazardous than the HLW.

## Figure 1: Ingestion hazard of selected radionuclides in high-level waste during ten million years. \*

The radiotoxic hazard measure is obtained by dividing the number of curies present for a given nuclide by the number of curies allowed by the maximum permissible amount of that nuclide in a cubic meter of drinking water. Data are normalized for one metric ton of light-water reactor fuel. The nuclide curves are plotted from data in table 2 of <u>Hamstra (1975)</u>; the curve for uranium mill tailings was derived from figure 2 of <u>Hamstra (1975)</u> and figure 7 of <u>Pigford & Choi (1976)</u>.

from Geologic Disposal of High-Level Radioactive Wastes -- Earth-Science Perspectives, U.S. Geological Survey, <u>Circular 779</u> by J.D. Bredehoeft et al (U.S. Gov't Printing Office, 1978)

> $^{239}$ Pu = Plutonium-239  $^{226}$ Ra = Radium-226  $^{237}$ Np = Neptunium-237  $^{229}$ Th = Thorium-229  $^{233}$ U = Uranium-233



The text of the USGS publication begins by asking the question, how much water would theoretically be required to dilute all the high level waste expected to be on hand in the USA by the end of the 20th century, to existing drinking water standards? (In other words, to achieve the maximum permissible degree of radioactive pollution of drinking water....) The answer: If you add up all the fresh water in the world, including not only all lakes and rivers and glaciers and ground water, but also all the soil moisture (which far exceeds the sum total of all the other sources), and then double that grand total, you will have about the right amount of water to do the required dilution.

The USGS points out that this calculation is only intended to emphasize why it is so important to keep this material out of the environment to an unprecedented degree of perfection. In my own words, 99% containment is nowhere near good enough. **Even 99.99% containment would represent an unacceptable target**. The USGS points out explicitly that even after 1 million years, the amount of water needed for the aforesaid dilution would be comparable to the volume of the entire Great Lakes water basin.

The next graphic (below) comes from the 1978 Report "A Race Against Time: Interim Report on Nuclear Power in Ontario" published by the Ontario Royal Commission on Electric Power Planning. The chart illustrates the radiotoxicity of just one year's worth of irradiated fuel discharged from a single CANDU reactor. The horizontal time scale covers a period of ten million years after the irradiated nuclear fuel is removed from the reactor: The vertical scale is a crude measure of radiotoxicity, based on how much water would be needed to dilute the waste down to the maximum degree of radioactive contamination allowed for drinking water -- the same measure that USGS was using in the previous graphic. Volume of water (cubic metres)



from "A Race Against Time", Queen's Printer, Toronto, 1978

Note that for the first 500-1000 years, the radiotoxicity (blue line) is dominated by the Fission Products (red line). In particular the inventory of cesium-137 and strontium-90, each with a 30-year half-life, plays a very important role for the first few centuries. But after that time, the toxicity is dominated by the actinides. From 1000 to 100,000 years, this is mainly due to the transuranic elements such as plutonium. But after 100,000 years, the toxicity actually increases rather than continuing to decrease. This is due to the ingrowth of radioactive decay products that are more radiotoxic than their parents.

For example, plutonium-239 has a half-life of 24,000 years, but its immediate radioactive decay product is uranium-235, which has a half-life of 700 million years -- so in fact plutonium-239 remains dangerous not just for hundreds of thousands of years, but for hundreds of millions of years. And by the way, while plutonium-239 is an excellent nuclear explosive material, so is uranium-235, its immediate radioactive decay product!

One thing that many people do not know is that MOST of the irradiated fuel is still U-238 (naturally-occurring depleted uranium) with a half-life of 4.5 BILLION years. While it may be true that U-238 is not highly radiotoxic, its decay products are EXTREMELY radiotoxic, especially the radium, radon and polonium isotopes.

After about a million years, the irradiated fuel is more radiotoxic than a 98% grade natural uranium ore deposit, which has never been seen on the face of the earth. At Cigar Lake in North Saskatchewan, uranium ore reaches an "extremely high" grade of over 20 %. Miners have to use robots to extract it because the radiation levels are far too high for humans. Well, after a million years, irradiated nuclear fuel is five times more radiotoxic than the Cigar Lake ore, based on the deadly uranium decay products alone!

You will notice in the color chart above that the buildup of RADIUM in the irradiated fuel is what adds so much to the toxicity after 100,000 to 1,000,000 years. It is important to realize that, as toxic as radium is, the polonium isotopes which accompany it are far more toxic. According to the Los Alamos Nuclear Laboratory's web site, polonium-210 is 250 BILLION times more toxic than cyanide (on a gram by gram basis).

The PENETRATING radiation from irradiated nuclear fuel diminishes greatly over the first 500-1000 years. It is primarily gamma radiation from the relatively short-lived fission products and activation products.

Thus the irradiated nuclear fuel becomes more approachable after a few centuries and can even be "handled safely" for short periods of time after several centuries have passed -- a fact that nuclear proponents often use to suggest that the wastes aren't very radioactive (and by implication, not very dangerous) after 500 years.

But this is grossly misleading, because the radiotoxicity of the irradiated fuel is still extraordinarily high as an ingestion or inhalation risk. This stuff is extremely poisonous even though the penetrating radiation has dropped off significantly.

The long-term radiotoxicity is mainly due to the fact that most actinides have extremely long half-lives and are -- for the most part -- emitters of a non-penetrating but deadly form of atomic radiation called alpha radiation.

While alpha radiation is generally harmless outside the body, it is extremely dangerous when it comes in close contact with living cells. Throughout the 20th century, the most

dangerous naturally-occurring radioactive materials by far were all alpha emitters -- this includes radium, radon, and the polonium isotopes, as well as uranium and thorium.

It is a well established scientific fact that non-penetrating alpha radiation is much more biologically damaging than an equivalent amount of energy from gamma radiation -- it is about 20 times more damaging per unit of radiation energy deposited in tissue, and about 200 times more damaging per radioactive disintegration. Thus an irradiated nuclear fuel bundle remains one of the most dangerous objects on Earth forever.

Besides, the penetrating radiation isn't gone forever -- it returns with a vengeance after a few more millennia. By the time a million years have gone by, the high level waste has once again become unapproachable -- as the daughter products of the actinides include many strong gamma emitters.

Gordon Edwards, email communication, August 14, 2012