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Neutron Radiation Damage from Actinide Decay and Fission in the Hanford Waste Tanks

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Introduction

The absolute neutron flux and neutron damage parameters in the steel walls of Hanford Site Waste Tank 101 SY at Richland, Washington, were determined to ascertain if significant neutron radiation damage had occurred from the storage of radioactive waste. The tank had been in operation for many years. The tank contains actinides of various species and other radioactive material in various concentrations in approximately 1 million gal. of waste sludge. Because the tank obviously is not, and has not been critical, any neutron flux in the tank must be the result of the neutron sources in the tank and their multiplication through fission of the fissionable material present in the tank.

There are six possible sources of neutrons in the tank. They are: (alpha,n) reactions, spontaneous fission, (gamma,n) reactions, cosmic rays, delayed neutrons and the fission process itself. In these calculations, we made conservative estimates of the amounts of radioactive isotopes that result in alpha decay, gamma production, and spontaneous fission in order to calculate the neutron source from these processes. Cosmic rays and delayed neutrons as a neutron source were also considered. A conservative estimate of the neutron sources was necessary because the contents and history of the tank are not well known. This neutron source was used in a neutron transport calculation to determine the multiplication resulting from actinide fission and the subsequent neutron fluxes. The absolute flux was used to determine the radiation damage parameters of helium production, hydrogen production, and displacement per atom in the wall of the tank.

Determination of the (alpha, n) and Spontaneous Fission Sources

An experimental determination of the absolute activity of each of the alpha emitting radioactive isotopes in the tank, or a list of the absolute activity of each of the isotopes subject to spontaneous fission, did not exist. Thus, the TRAC [1] computer code, which gives a calculated estimate of the isotopes and their activity in a tank, was used. We have found that TRAC results are not a reliable source of information, thus this output was used as a starting point for a list of isotopes in the tank and as an indication of the relative magnitudes of the various isotopes. Each isotope's activity in this list was normalized to the activity of the ^{239}Pu in the list and then multiplied by the maximum value of the activity of ^{239}Pu [2] that had been found experimentally (i.e., 2.71×10^{-8} Ci/cm^3). If there was an experimentally determined value [3] of an isotope, and it was larger than that obtained from the normalization, it was replaced with the higher value. The final list of isotopes, their activity, number density, and mass, used in the calculation to determine the (alpha,n) source and spontaneous fission source, is given in Table 1.

We considered three representative sludges [3,4] or slowing down mediums for the (alpha,n) production. The activities from Table 1 and a slowing down medium were the input to the SOURCES [5] code, which calculates the (alpha,n) and spontaneous fission neutron sources. The representative sludge composed of nitrogen, 5.799 a/o; oxygen, 37.285 a/o; chlorine, 0.288 a/o; phosphorus, 0.107 a/o; carbon, 1.702 a/o; hydrogen, 43.773 a/o; sodium 9.765 a/o; aluminum, 1.134 a/o; and potassium, 0.262 a/o resulted in the highest (alpha,n) production of 1302 neutrons/cm^3-s. The spontaneous fission source was 17 neutrons/cm^3-s.

These results are very conservative. Greater than 99% of the (alpha,n) neutrons result from the decay of ^{241}Am. An experimental value [3] for ^{241}Am activity is 0.34 μCi/cm^3. The value used in the calculation is 5.41×10^4 μCi/cm^3. The activity of ^{239}Pu to which the isotopes were normalized, the maximum experimental value found, would lead to a total mass of ^{239}Pu in the tank of 1,897 g. The nominal value of ^{239}Pu in the tank is 910 g. The maximum experimental value [3] of alpha activity in sludge is on the order of 0.7 μCi/cm^3. The total alpha activity in Table 1 is 5.46 x 10^4 μCi/cm^3. Thus, the calculated neutron source is probably several orders of magnitude greater than the actual source.

Determination of the (gamma,n), Cosmic Radiation, and Delayed Neutron Sources

None of the radioactive isotopes listed in the TRAC output noted above, decay with a gamma of sufficient energy to produce a (gamma,n) reaction in deuterium, which has a threshold of 2.2 MeV. With the exception of beryllium, all other thresholds for (gamma,n) production exceed 2.2 MeV. Thus, the only source of gammas of sufficient energy to undergo a (gamma,n) reaction would be the result of a capture gamma or fission. We made the assumption that all neutron sources result in a capture and produce a capture gamma in the range of 7 to 8 MeV. These gammas were used as a source in a ONEDANT [6] transport calculation to ascertain a gamma flux. The MATXS7 [7] 24 gamma energy group cross sections were used in the transport calculation. The transport medium was water. Using this flux and (gamma,n) cross sections from Ref. 8, an estimate of the (gamma,n) neutron source from reactions with deuterium was made. The source was less than 10^{-3} neutrons/cm^3-s. The (gamma,n) reactions were eliminated as a source of neutrons.

To determine the contribution from cosmic rays, the results from Ref. 9 were used. In uranium metal, the source strength is on the order of 10^{-3} neutrons/cm^3-s. Thus, the source in the tank would be considerably smaller than this. Cosmic radiation was eliminated as a source of neutrons.

Delayed neutrons are a result of the decay of certain fission products. About 0.7% of the fission neutrons are delayed. The half life of the longest averaged precursor is 55 min. Delayed neutrons will make a insignificant contribution to the total neutron flux and were eliminated as a source of neutrons.

Transport Calculations

To calculate the absolute fluxes, a transport model was assumed. The transport model was an infinite cylinder of water, 22.86-m in diameter, contained in an iron tank with walls 1-cm thick and surrounded by a 1-m thick representative clay. Vacuum boundary conditions were used. Water was used as the transport medium as opposed to sludge, as the use of water would lead to higher calculated fluxes. ONEDANT was the transport code, and the 69 neutron group MATXS7 library was the cross section set utilized.

The maximum concentration of both plutonium (0.436×10^{-6} g/cm^3) and uranium (1.86×10^{-4} g/cm^3) found in the literature was assumed to be the concentration of the entire volume of the tank. The plutonium was assumed to be all ^{239}Pu and the uranium was assumed to be all ^{235}U. The source strength used in the transport calculation was the 1302 neutrons/cm^3-s from the (alpha,n) reactions and 18 neutrons/cm^3-s from spontaneous fission.

The results indicated 3% multiplication. The absolute 69 group fluxes in the tank wall summed to include three groups: thermal (1.0×10^{-5} to 4 eV), epithermal (4 eV to 0.111 MeV), and fast (0.111 to 10 MeV); and are 0.788×10^4, 0.536×10^4, and 0.137×10^5 n/cm^2-s, respectively. This flux results in a helium production, hydrogen production, and iron displacement rates of 0.147, 2.71, and 8.78×10^{-17} atoms/cm^3-s, respectively. There would be no significant radiation damage to the wall at these rates.

Conclusion

At the beginning of this project, we had a choice of making "best estimate" or conservative calculations. A "best estimate" calculation would give our best estimate of the absolute value of the flux in the wall. An extremely conservative calculation would unequivocally set an upper bound on the magnitude of the flux in the tank wall. Because the values of most of the parameters were not known to any great accuracy, it was felt that conservative calculations would be the most useful. The results of these extremely conservative calculations indicate that the radiation damage in the tank wall is insignificant.

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Table 1. Isotopes, Activities, Number Density and Mass used to determine the (alpha,n) and Spontaneous Fission Source

	Activity	Number density	Mass
Isotope	curie/cm3	Atoms/cm3	grams/cm3
^{210}Pb	0.122×10^{-14}	$0.457 \times 10^{+05}$	0.159×10^{-16}
^{213}Bi	0.541×10^{-10}	$0.791 \times 10^{+04}$	0.280×10^{-17}
^{210}Po	0.108×10^{-14}	$0.690 \times 10^{+03}$	0.241×10^{-18}
^{218}Po	0.146×10^{-06}	$0.146 \times 10^{+07}$	0.527×10^{-15}
^{223}Fr	0.135×10^{-09}	$0.954 \times 10^{+04}$	0.353×10^{-17}
^{223}Ra	0.108×10^{-07}	$0.570 \times 10^{+09}$	0.211×10^{-12}
^{226}Ra	0.135×10^{-13}	$0.364 \times 10^{+08}$	0.137×10^{-13}
^{225}Ac	0.541×10^{-11}	$0.250 \times 10^{+06}$	0.933×10^{-16}
^{227}Ac	0.108×10^{-07}	$0.397 \times 10^{+12}$	0.150×10^{-09}
^{227}Th	0.947×10^{-08}	$0.818 \times 10^{+09}$	0.308×10^{-12}
^{229}Th	0.406×10^{-11}	$0.499 \times 10^{+11}$	0.190×10^{-10}
^{230}Th	0.271×10^{-11}	$0.343 \times 10^{+12}$	0.131×10^{-09}
^{231}Pa	0.271×10^{-07}	$0.149 \times 10^{+16}$	0.573×10^{-06}
^{233}U	0.180×10^{-05}	$0.481 \times 10^{+18}$	0.186×10^{-03}
^{234}U	0.271×10^{-10}	$0.112 \times 10^{+14}$	0.434×10^{-08}
^{235}U	0.189×10^{-15}	$0.224 \times 10^{+12}$	0.876×10^{-10}
^{238}U	0.108×10^{-16}	$0.814 \times 10^{+11}$	0.322×10^{-10}
^{237}Np	0.541×10^{-04}	$0.195 \times 10^{+21}$	0.767×10^{-01}
^{238}Pu	0.217×10^{-05}	$0.320 \times 10^{+15}$	0.126×10^{-06}
^{239}Pu	0.271×10^{-07}	$0.110 \times 10^{+16}$	0.436×10^{-06}
^{240}Pu	0.541×10^{-07}	$0.598 \times 10^{+15}$	0.238×10^{-06}
^{241}Pu	0.244×10^{-06}	$0.590 \times 10^{+13}$	0.236×10^{-08}
^{241}Am	0.541×10^{-01}	$0.394 \times 10^{+20}$	0.158×10^{-01}
^{242}Am	0.812×10^{-04}	$0.250 \times 10^{+12}$	0.101×10^{-09}
^{242m}Am	0.812×10^{-04}	$0.193 \times 10^{+17}$	0.774×10^{-05}
^{243}Am	0.271×10^{-04}	$0.336 \times 10^{+18}$	0.135×10^{-03}
^{242}Cm	0.676×10^{-04}	$0.509 \times 10^{+14}$	0.205×10^{-07}
^{244}Cm	0.122×10^{-03}	$0.371 \times 10^{+16}$	0.150×10^{-05}
^{245}Cm	0.541×10^{-08}	$0.774 \times 10^{+14}$	0.315×10^{-07}