RAS 5602 72-22-ISFSI - State Exhibit 143 - Rec'd 6/25/02

Calculation of Neutron Dose at Elevated Concrete Temperatures

The following calculations are based on information from:

Kaplan, M.F, 1989. *Concrete Radiation Shielding: Nuclear Physics, Concrete Properties, Design and Construction*. Avon, Great Britain: Longman Scientific and Technical.

In order to calculate the loss of neutron shielding associated with evaporation of water in the concrete, it is necessary to discuss briefly the interaction mechanisms at play. Neutrons are grouped into categories based on their energy levels: thermal neutrons have low energies and can be absorbed or captured, while fast neutrons have higher energy and need to be slowed before capture. Table 1, below, shows the energy distribution of fast neutrons used as the design basis fuel in the Holtec HI-STORM SAR.

Table 1: Neutron Energy Distribution of Design Basis Fuel: From HI-STORM SAR

Lower Energy (MeV)	Upper Energy (MeV)	35,000 MWD/MTU, 5-year cooled		45,000 MWD/MTU, 5-year cooled		45,000 MWD/MTU, 9-year cooled	
		Neutrons/s	% of total	Neutrons/s	% of total	Neutrons/s	% of total
0.1	0.4	7.19E+06	3.8%	1.63E+07	3.8%	1.40E+07	3.8%
0.4	0.9	3.68E+07	19.3%	8.33E+07	19.4%	7.15E+07	19.4%
0.9	1.4	3.37E+07	17.7%	7.63E+07	17.8%	6.55E+07	17.7%
1.4	1.85	2.49E+07	13.1%	5.62E+07	13.1%	4.84E+07	13.1%
1.85	3	4.42E+07	23.2%	9.92E+07	23.1%	8.56E+07	23.2%
3	6.43	3.99E+07	21.0%	9.01E+07	21.0%	7.75E+07	21.0%
6.43	20	3.52E+06	1.9%	7.98E+06	1.9%	6.85E+06	1.9%
Totals		1.90E+08	100.0%	4.29E+08	100.0%	3.69E+08	100.0%

For fast neutrons, the *effective removal cross section* (Σ_R) describes the removal of neutrons by a shielding mechanism. It is used in the following equation:

$I=I_o e^{-\Sigma_r T}$; where T is the thickness of the shielding.

Neutron cross-sections are greatly affected by the neutron energy and the atomic weight of the various chemical elements in the shielding medium. However, according to Kaplan's text, the effective removal cross section is considered to be approximately constant for neutron energies between 2 and 12 MeV (Kaplan, pp 235) which accounts for approximately 50% of the neutron distribution given above. For comparison, we will assume the cross sections are constant throughout the range of energies listed above.

For materials (such as concrete) which are comprised of a variety of elements, the total effective removal cross section can be calculated as the sum of the weighted averages of the individual effective removal cross sections. Kaplan has listed various effective removal cross sections for components commonly found in concrete. In Table 2, below, we list the elemental makeup of "ordinary concrete" used in the Kaplan text and the concrete to be used for the HI-STORM 100 overpack. The HI-STORM concrete data is taken from Table 5.3.2 of the HI-STORM SAR

Table 2: Elemental Makeup of Concrete and Neutron Removal Cross Sections

	Ordinary Concrete			Holtec HI-STORM 100 Overpack Concrete		
Element	g element/cm^3 concrete	Σ_R/ρ (cm^2/g)	Σ_R (cm^{-1})	G element/cm^3 concrete	Σ_R/ρ (cm^2/g)	Σ_R (cm^{-1})
H	0.015	0.598	0.0090	0.0141	0.598	0.0084
O	1.057	0.0346	0.0366	1.175	0.0346	0.0407
Na	0.041	0.0341	0.0014	0.03995	0.0341	0.001362295

Template=SECY-028

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SECY-02

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Mg	0.085	0.0333	0.0028	--		
Al	0.137	0.0292	0.0040	0.1128	0.0292	0.00329376
Si	0.487	0.0295	0.0144	0.74025	0.0295	0.021837375
P	0.002	0.0283	0.0001	--		
S	0.002	0.0275	0.0001	--		
K	0.015	0.0247	0.0004	0.04465	0.0247	0.001102855
Ca	0.295	0.0243	0.0072	0.19505	0.0243	0.004739715
Ti	0.011	0.022	0.0002	--		
Mn	0.003	0.0202	0.0001	--		
Fe	0.178	0.0214	0.0038	0.0282	0.0214	0.00060348
Concrete Properties	**2.328**		**0.0799**	**2.35**		**0.0820**

The HI-STORM 100 cask contains a concrete layer 67.95 cm thick. Therefore, we can estimate the attenuation of neutrons by this shielding by solving the exponential absorption equation:

$I/I_o = e^{-\Sigma_r T}$

For the "ordinary concrete" used in the Kaplan text, $I/I_o = 0.0044$, while for the HI-STORM overpack concrete $I/I_o = 0.0038$. Further, the HI-STORM 100 SAR has specified a neutron dose rate adjacent to the mid-height of the HI-STORM overpack as 1.88 mrem/hour assuming 45,000 MWD/MTU, 5-year cooled MPC-24 fuel. Using this value as I in the above equation we can solve for I_o to estimate the neutron dose rate assuming no shielding.

I_o = 1.88 mrem/hour x $\exp^{.0820 \times 67.95}$ = **495 mrem/hour assuming no neutron shielding by concrete**

Temperature Effects of Neutron Shielding Ability of Concrete

Increased temperature of concrete results in a decrease in the amount of water, which results in an increase in the neutron flux density transmitted through a concrete shield of given thickness. The Kaplan text presents the results of experiments in which the effective removal cross section of concrete were estimated (and experimentally measured) at temperature values of room temperature, 100°C, 200°C, and 300°C. Below, I use these results to estimate the effect on the HI-STORM concrete, assuming that an equivalent loss of hydrogen (by weight %) in the HI-STORM overpack concrete as in the experimental concrete. The same calculations were performed as appear in Table 2 of this report: the relative proportions of the various elements are not included in Table 3 for brevity.

Table 3: Effect of Temperature Increase on Neutron Shielding

		"Ordinary Concrete" used in Kaplan text	HI-STORM 100 Overpack Concrete
Unheated (as cured)	ρ, g/cm^3	2.328	2.35
	H density, g H/cm^3 concrete	0.015	0.0141
	Σ_R (calculated), cm^{-1}	0.0801	0.0820
	Σ_R (measured), cm^{-1}	0.0780	--
100 °C	ρ, g/cm^3	2.258	2.283
	H density, g H/cm^3 concrete	0.007	0.00658
	Σ_R (calculated), cm^{-1}	0.0731	0.0755
	Σ_R (measured), cm^{-1}	0.0735	--
200°C	ρ, g/cm^3	2.238	2.266
	H density, g H/cm^3 concrete	0.005	0.0047

	Σ_R (calculated), cm^{-1}	0.0713	0.0738
	Σ_R (measured), cm^{-1}	0.0724	--
300°C	ρ, g/cm^3	2.227	2.258
	H density, g H/cm^3 concrete	0.004	0.00376
	Σ_R (calculated), cm^{-1}	0.0704	0.0730
	Σ_R (measured), cm^{-1}	0.0702	--
All Water Evaporates	ρ, g/cm^3	2.194	2.242
	H density, g H/cm^3 concrete	0	0
	Σ_R (calculated), cm^{-1}	.0668	.0703
	Σ_R (measured), cm^{-1}	--	--

Previously, we have estimated the unshielded dose rate to be 495 mrem/hour. To estimate what the shielded dose rate is as a function of temperature, we simply repeat the calculation: $I=I_o e^{-\Sigma r T}$ for the varying temperatures. This is done for the HI-STORM 100 cask below.

Table 4: Estimated Dose Rates Due To Neutrons as a Function of Concrete Temperature

Temperature	**Dose Rate Adjacent to Cask Mid-Height (mrem/hour)**
Unheated (as cured)	1.88
100°C	2.94
200°C	3.28
300°C	3.47
All Water Evaporates	4.16

However, the above assumes that thermal neutrons will be attenuated once they are reduced in energy. According to the Kaplan Text, "the concept of an effective removal cross-section is dependent on the presence of hydrogen." (70) If there are insufficient hydrogen atoms to thermalize and consequently contribute to the absorption of a neutron after it has been slowed, then the equations used above may underpredict the amount of radiation emanating from a hydrogen-free shielding material. Usually, concrete contains sufficient hydrogen and is dominated by the collision reactions. When hydrogen is not present, the thermal interactions may dominate from a shielding perspective. If this is the case, the neutron dose rate computed above is likely to be somewhat higher.

For example, the Kaplan text provides values of the thermal diffusion length of a certain type of concrete at two different temperatures: unheated (as cured) and at 100°C). Assuming a shield thickness equal to 67.95 cm, the following I/I_o values for thermal neutrons are presented:

Table 5: Thermal Diffusion Length as a function of Concrete Temperature

Concrete	Temperature	Density (g/cm^3)	L (cm)	I/I_o
0-HW1	Unheated (as cured)	2.33	6.98	5.92×10^{-5}
0-HW2	100°C	2.26	8.97	5.12×10^{-4}

The density of this concrete is very similar to that of the HI-STORM overpack concrete. For our purposes, we assume they are identical in terms of shielding. If we assume a similar loss in hydrogen content (from .015 to .007 g/cm^3 concrete) as a result of heating to 100°C that was witnessed in the "ordinary concrete" discussed in the Kaplan text, we note that a decrease in hydrogen content by approximately 50% leads to a decrease in thermal neutron shielding by an order of magnitude. If we assume a linear relationship between thermal diffusion length and hydrogen content, we can make the following estimates of loss of thermal shielding as a function of temperature (and consequently, hydrogen content).

Table 6: Estimated Thermal Diffusion Length as a Function of Hydrogen Content

Temperature	Density (g/cm^3)	H content (assumed)	L (cm)	I/I_o
Unheated (as cured)	2.33	.015	6.98	5.92×10^{-5}
100°C	2.26	.007	8.97	5.12×10^{-4}
200°C	2.238	.005	9.66	8.80×10^{-4}
300°C	2.227	.004	10.04	1.15×10^{-3}
No Hydrogen Left	2.194	0	11.95	3.39×10^{-3}

Thus, if the concrete in a HI-STORM 100 cask were to lose all of its water, it is estimated that the amount of thermal neutron radiation passing through would be approximately 57.3 times greater than that calculated in the HI-STORM SAR. In terms of radiation dose, assuming proportionality, this would increase the neutron dose to workers to **1.88 mrem/hour x 57.3, or 108 mrem/hour.**