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RADIOACTIVITY OF THE COOLING WATER

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ABSTRACT

The most important source of radioactivity at the exit manifold of the pile will be due to O^{19}, formed by neutron absorption of O^{18}. A recent measurement of Fermi and Weil permits to estimate that it will be safe to stay about 80 minutes daily close to the exit manifolds without any shield. Estimates are given for the radioactivities from other sources -- both in the neighborhood and farther away from the pile.

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1. If the water contains p parts per million of an element with an atomic weight m and slow neutron cross section σ, this substance will absorb about the fraction

$$f = \frac{pV\sigma L \times 10^{-6}}{m\Sigma} = \frac{10^{24}\sigma p}{m} \times .425 \times 10^{-6} \qquad (1)$$

of all neutrons. In the above, $L = 6 \times 10^{23}$ is Loschmidt's number, $V = 3 \times 10^{6}\,cm^{3}$ is the total volume of the water in the pile and Σ is the total cross section of all materials (U, C, etc) in the pile. For a 200 ton U pile $\Sigma \approx 4.25 \times 10^{6}\,cm^{2}$. Since, in a 500,000 kw pile, 3.5×10^{19} neutrons are absorbed per second, the number of new nuclei formed by neutron absorption of the element in question becomes

$$N = 3.5 \times 10^{19} f = \frac{10^{24}\sigma p}{m} \times 1.5 \times 10^{13}\,sec^{-1} \qquad (1a)$$

The nuclei thus formed by neutron absorption may or may not be radioactive. If, in the former case, the radioactivity is connected with the emission of γ-rays, it may present a hazard to the personnel at the water exit end of the pile. This hazard is, of course, removed if the pile is shut down as soon as the water which has been in the pile during operation has left the system but is present as far as the personnel inspecting the water outlet system during operation is concerned. In addition, the radioactivity of the cooling water--from the above and other sources to be discussed below--may make it necessary to shield the outlet pipes etc., for some distance.

The β radiation will present little actual danger. More probably, it may render it difficult to detect failures in the U sheathing or coating.

by overshadowing the fission activity which escapes into the water by failures.

To anticipate the result of the following calculations: the above described danger appears to be small, the greatest amount of γ activity probably arising from O^{19}, formed by neutron absorption of the relatively rare isotope O^{18}, and permitting a daily sojourn of more than an hour in the immediate neighborhood of the exit pipings of the pile during full (500,000 kw) operation.

2. Let us assume that the water stays for 3 seconds in the manifolding at the exit end of the pile and that the half life τ of the radioactive product is longer than this. The total number of disintegrations in these pipings etc., then becomes

$$Q = \frac{3 \ln 2}{\tau} N = 3.1 \times 10^{13} \frac{10^{24}\sigma\rho}{m\tau} \frac{\text{disintegrations}}{\text{sec}} \qquad (2)$$

We shall assume that every disintegration is connected with the emission of a γ ray and that the absorption of this in the pipings etc., can be neglected. The activity at the distance ρ from the plane of the exit pipings, along the axis of the pile then becomes

$$A = \frac{Q}{\pi R^2} \int_0^{R_1} \frac{2\pi r\,dr}{4\pi(r^2+\rho^2)} = \frac{Q}{4\pi R^2} \ln \frac{R^2+\rho^2}{\rho^2} \qquad (3)$$

where R   500 cm is the radius of the pile. The above calculation contains the optimistic assumption that the activity is uniformly distributed in the plane of the pipings. On the other hand, the axis of the pile is the worst position from the point of view of radiation. Inserting for a the value (2), for R - 500 cm, for ρ - 100 cm (3) becomes

$$A = 3.2 \times 10^7 \frac{10^{24}\sigma\rho}{m\tau} = .064 \times \frac{10^{24}\sigma\rho}{m\tau} \frac{\text{r un}}{\text{sec}} \qquad (3a)$$

It is assumed that 1 r unit corresponds to 5×10^8 γ rays per cm^2.

The following will give an enumeration pf possible sources of radioactivity in the cooling water, either from the above or other reasons, as far as we can foresee them at present.

Hydrogen

$p = 1.11 \times 10^5$, $10^{24}\sigma = .39$ for H^1 so that $f \approx 1.8 \times 10^{-2}$. The product from this reaction H^2, is stable so that no radioactivity results from this source. For H^2 itself, $p = 4 \times 10^{-4} \times 1.11 \times 10^5$, $10^{24}\sigma = 1.8 \times 10^{-3}$ so that the activity would represent ($\tau = 10^9$ sec)

$$A = \frac{.064 \times 1.8 \times 10^{-3} \times 44.4}{2 \times 10^7} = 2.5 \times 10^{-12} \frac{r}{sec}$$

which is a negligible activity quite apart from the fact that the H^3 formed emits no γ rays and even its β- ray is extremely soft.

Oxygen

The nuclei formed from O^{16} and O^{17}, viz O^{17} and O^{18} are again stable. O^{18} itself has $p = .89 \times 2 \times 10^3$ and, according to Fermi and Weil's recent measurement, $10^{24}\sigma \approx 3 \times 10^{-4}$, $\tau = 31$ sec. Hence

$$A \approx \frac{.064 \times 3 \times 10^{-4} \times 1.8 \times 10^{3}}{1.8 \times 31} = 6 \times 10^{-5} \frac{r}{sec}$$

It was mentioned before that this is the most serious source of radioactivity to be expected in the cooling water at the exit pipings. If it is connected with a penetrating γ ray, which is very likely, it will make it unadvisable to stay very near to the exit manifolds of the pile for more than about 80 minutes daily. On the other hand, since the radioactivity of O^{19} decreases by a factor of 10^6 in 10 minutes, this radioactivity will become insignificant rather near to the entrance of the water into the river.

There are a few other nuclear reactions which may cause radioactivity in the water. Only one of these

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$$O^{17} + n = C^{14} + He^4 + 1.8 \text{ MeV} \qquad (a)$$

is caused by the neutrons directly (this reaction may be responsible for a non-negligible fraction of the absorption of oxygen). Most of the others are induced by the fast recoil protons produced by the fission neutrons in water. Among these

$$O^{17} + H^1 = F^{17} + n - 5.5 \text{ MeV}$$

may be safely disregarded because F^{17} is produced more abundantly by the

$$O^{16} + H^1 = F^{17} + h\nu \ (3.3 \text{ MeV}) \qquad (b)$$

reaction. Both

$$O^{18} + H^1 = F^{18} + n - 2.2 \text{ MeV}$$

$$O^{17} + H^1 = F^{18} + h\nu \ (5.5 \text{ MeV}) \qquad (c)$$

produce F^{18} but the first one is probably the more abundant source.

Reaction (a) probably has a reasonably high cross section. However, the abundance of O^{17} is even lower (4×10^{-4}) than that of O^{18} and the half life of C^{14} very long ($\tau = 5 \times 10^{7}$ sec). Hence, (a) will cause no serious radioactivity.

Both (b) and (c) require reasonably fast protons. The number of these can be estimated as follows: The number of fission neutrons crossing the water jacket is 3.5×10^{19} per sec. They cross, on the average, .3 cm of water which contains $2 \times 6 \times 10^{23}/18$ H atoms per cm^3. Assuming a cross section of $2 \times 10^{-24} cm^2$ for this, the number of recoil protons per second becomes

$$3.5 \times 10^{19} \times .3 \times .67 \times 10^{23} \times 2 \times 10^{-24} = 14 \times 10^{18} \frac{\text{protons}}{\text{sec}}$$

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Only a small fraction of these protons will be able to participate in reactions (b) and (c) to any considerable extent because of the potential barrier that they have to penetrate. Let us assume that this fraction is .1, representing the protons of 3 MeV and more. Actually, this number cannot be calculated at present because the fission spectrum is not well known. The above appears to be a safe estimate. The proton will remain effective for reactions (b) and (c) only until it loses about 1 MeV energy by collisions. This gives them an effective range of less than 10 cm in air or about 1.3×10^{-2} cm in water. If the cross section of reaction (b) or (c) is σ, the number of reactions will be

$$N = 1.4 \times 10^{17} \times 1.3 \times 10^{-2} \times .33 \times 10^{23} \sigma = .6 \times 10^{38} \sigma$$

in case (b), the number of O^{16} atoms per cm^3 being $.3 \times 10^{23}$. For σ we can assume about $10^{-28} cm^2$ in this case which gives $N = 6 \times 10^9$ F^{17}/sec. This is a strong positron activity which has to be reckoned with when the leak detecting apparatus is designed. However, it is very much smaller than the activity due to O^{19} of which about $N = 5 \times 10^{11}$ are formed in a second.

The situation is similar with respect to reaction (c). The number of the O^{18} nuclei which participate in (c) is 500 times smaller than that of the O^{16} nuclei participating in (b). On the other hand, the cross section for (c) may be $10^{-25} cm^2$ (this is about the highest cross section known for a nuclear reaction of this type) so that, conceivably, $N = 10^{10}$ F^{18} nuclei may be formed per second. This, again, is more important for the design of leak detecting counters than for the total radioactivity in front of the pile. Only if O^{19} should not emit γ rays would (b) become the largest source of radioactivity in pure water. The half life of F^{17} is 64 sec and thus has a larger probability of disintegrating in the front pipings than the F^{18} the half life of which is 6700 sec. Actually, the total radiation is very small, both from (b) and (c) so that, if O^{19} is really free from γ rays, the impurities in the water may contribute fully as much radiation as F^{17} or F^{18} do.

It is, probably unnecessary to mention that, while the radioactivity due to O^{19} can be estimated reasonably accurately, the amount of F^{17} and F^{18} may be twice greater or more than ten times smaller than given above.

Impurities.

The most important impurities in water are, apparently, Fe, SiO_2, Ca, Mg, Na, K, SO_4, NO_3, Cl, Br, CO_3. To these PO_4, may be added which is used as an inhibitor. In a good water source these are present only to the extent of a few parts per million. The radioactive nuclei which may be formed from these elements are Fe^{55} (harmless), Fe^{59}, Si^{31} (no γ, harmless), Ca^{41}, Ca^{45}, Ca^{49}, Mg^{27}, Na^{24}, K^{42} (probably no γ), S^{35} (probably no γ), S^{37} (hardly to be expected to amount to anything), N^{16}, Cl^{36}, Cl^{38}, Br^{80}, Br^{82}, C^{14} (harmless), P^{32} (no γ , harmless). The following table gives N and A for the case that the element in question is present to 1 ppm (i.e., the p assumed in the table is the abundance of the relevant isotope). The table gives, therefore, a sort of "danger coeffecients" for outside the pile (N) and for the exit maniforld (A). However, in the former case, allowance should be made for the life time of the radioactive nucleus. This will decrease the danger both if it is very long and also if it is very short. In the former case, most of the disintegration will occur after the water is well mixed with river and perhaps sea water. In the latter case, most of the disintegration will occur in the neighborhood of the plant, partly already in the conduit to the river where proper precautions can be taken against the effects of it.

Nucleus	10^{-11} N sec	τ	γ energy	A sec/r	ppm in Site X water
Fe^{59}	$\ll 8, \sim .02$	47 days	1 MeV	$\ll 7.8 \times 10^{-10}$	5
Ca^{41}	1.6	8.5 "	1.1 "	10^{-9}	)
Ca^{45}	$\ll 1.6, \sim .03$	180 "	.7 "	$\ll 4.4 \times 10^{-11}$	) 26
Ca^{49}	$< .03$	2.5 hrs.?	.8 "	$< 1.4 \times 10^{-9}$	)
Mg^{27}	$< .17$	10.2 min	.9 "	$< 1.1 \times 10^{-7}$	10^{-3} ?
Na^{24}	2.5	14.8 hrs.	1,2,3 "	2×10^{-8}	5 - x
K^{42}	.18	12.4 ")	probably none	1.7×10^{-9}	x
S^{35}	$\ll 2, \sim .08$	88 days)		$\ll 10^{-10}$	2 1/3
N^{16}	$< 4.3, \times 10^{-4}$	8 sec	probably present positron active	2.3×10^{-8}	1/2
Cl^{36}	110	$\sim$ 300 years		4.8×10^{-12}	) 3
Cl^{38}	.35	37 min	2,2.5 MeV	6.6×10^{-8}	)
Br^{80}	6 6	(4.4 hrs) (18 min)	Soft	1.8×10^{-7} 2.6×10^{-6}	) ?
Br^{82}	1.5	34 hrs.	.65	5.3×10^{-9}	
O^{19}	5	31 sec.	?	6×10^{-5}	

The last column gives the analysis of site X water. It is added only as an illustration of a possible water analysis. The last row gives, for sake of comparison, the actual (not for 1 ppm) radioactivity of O^{19}. The $\ll$ sign means that the corresponding figure has been arrived at by attributing all the neutron absorption of the element to the isotope in question. The $\sim$ means an approximate figure obtained by assuming the same absorption cross section for all the isotopes of the element. The rest of the figures was obtained on the basis of a cross section measurement of one isotope.*

Water saturated with air contains about 20 ppm N_2. The radioactivity of this (if it emits γ rays) is still considerably below the radioactivity of the O^{19}.

*F. Rasetti, Phys. Rev. 58, 869, 1940. Goldhaber and O'Neal, Phys. Rev. 59, 102, 109, 1941. Manley, Haworth and Luebke, Phys. Rev. 59, 109, 1941. Sinma and Yamasaki, Phys. Rev. 59, 402, 1941.

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