The Importance of Using the Mixed Neutron Flux in Activation Analysis of D-^{3}He Fueled Reactors

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THE IMPORTANCE OF USING THE MIXED NEUTRON FLUX IN ACTIVATION ANALYSIS OF D-^{3}He FUELED REACTORS

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Abstract

The D-D and D-T secondary reactions in D-^{3}He reactors provide the neutron source term for most of the radioactivity produced in the structure of the reactor. Radionuclides are produced as a result of neutron interactions with their parent nuclides. The amount of activity produced by any radionuclide depends on the number of its parent atoms present at any given time. One approach to account for the activity induced by both neutron sources in any activation analysis is to add their individual contributions. Performing two separate calculations for the D-D and D-T neutron flux components and adding their contributions yields conservative results due to underestimating the destruction of the parent atoms. The overestimation is more pronounced for short and intermediate lived nuclides, long operation time, large neutron flux and large destruction cross section for the parent atoms. In the steel first wall of a typical D-^{3}He reactor, adding the individual contributions of the two neutron sources results in overestimating the activities produced by most of the radioactive isotopes of Ag, Lu, Ta, W and Re. After 30 years of reactor operation, the activity of ^{187}W, which is a major source of safety concern in case of an accident, is more than an order of magnitude higher than its value if the mixed neutron flux is used. The activity of ^{188}Re, which is an important source of offsite dose in case of accidental release, is overestimated by more than a factor of two.

Introduction

In a D-^{3}He reactor the reaction between the deuterium and helium is the principal reaction in the plasma. However, because deuterium is present, a D-D reaction occurs. This reaction has the two following branches:

$$\mathrm{D + D \longrightarrow T\ (1.01\ MeV) + p\ (3.03\ MeV)} \quad (1)$$

$$\mathrm{D + D \longrightarrow {}^3He\ (0.82\ MeV) + n\ (2.45\ MeV)}\ . \quad (2)$$

In the first reaction tritium is produced and in the second reaction 2.45 MeV neutrons are produced. The 2.45 MeV neutrons can activate the reactor structure. The deuterium can interact with the tritium formed by the first branch of the D-D reaction via:

$$\mathrm{D + T \longrightarrow {}^4He\ (3.52\ MeV) + n\ (14.07\ MeV)}\ . \quad (3)$$

Just like the 2.45 MeV neutrons, the 14.07 MeV neutrons interact with the reactor structure and induce radioactivity. These two neutron sources represent the source term for most of the radioactivity produced in D-^{3}He reactors. Any D-^{3}He reactor activation analysis has to account for the radioactivity induced by both neutron sources. It is common to define the 2.45 MeV and the 14.07 MeV neutrons as D-D and D-T neutrons, respectively.

One of the approaches to account for the two neutron sources in activation analysis is to conduct two sets of activity calculations using each of the two neutron sources separately. Upon completing the two separate sets of calculations, one would add the results to calculate the total radioactivity induced in the reactor. In this paper, we show that such a practice results in an overestimation of the radioactivity induced by many of the short and intermediate-lived radionuclides. This overestimation is caused by the fact that both D-D and D-T neutrons try to react with the same parent atoms. Hence performing two separate calculations in which the two neutron source components are used rather than the mixed neutron source result in underestimating the destruction of the parent atoms.

Calculational Procedure

Activation analysis was conducted using the DKR-ICF [1] code with activation cross sections taken from the ACTL [2] library and for 30 full power years (FPY) of operation. Full reactor life operation is used since the structural material in a D-^{3}He reactor is expected to last for the whole reactor lifetime. The neutron transmutation data are in 46 energy group structure. The decay and gamma source data are taken from the table of isotopes with the gamma source data being in 21 group structure format. The neutron flux used in the activation analysis is generated by the one-dimensional discrete ordinates neutron-transport code ONEDANT [3] along with the ENDF/B-V cross section data files. The ONEDANT code was used to generate the neutron flux for each of the neutron source components (D-D and D-T) and the flux of the mixed neutron source. Using neutron fluxes which are similar to those expected in the first wall of a typical D-^{3}He reactor, the DKR-ICF code was used to calculate the radioactivity induced in a target which is made of one of the naturally occurring elements in the periodic table. All elements' parents with atomic number $Z \leq 84$ were considered in this paper.

Another detailed analysis was performed to examine the impact of using the correct mixed neutron source rather than the two neutron source components on the accuracy of the results of an activation analysis performed for a selected D-^{3}He reactor design. The design selected is similar to the ARIES-III design. Radioactivity calculations were conducted for the low activation ferritic steel (modified HT-9) first wall and shield. The activation results were utilized in an offsite dose calculation performed according to the worst case weather conditions used in the FUSCRAC3 code [4]. The elemental composition of the modified HT-9 used in the calculations was taken from the BCSS [5] study.

Results

Radioactive nuclides are produced due to neutron interaction with their parent atoms. In turn the daughter radionuclides may be lost by either another nuclear reaction or by decay. The simple rate equation governing a single chain of activity calculations during reactor operation can be written as:

$$N_p(t) = N_{po} e^{-\sigma_p \phi t} \quad (4)$$

$$N_d(t) = \frac{N_{po}\sigma_{pd}\phi}{(\beta_d - \sigma_p \phi)} \left(e^{-\sigma_p \phi t} - e^{-\beta_d t}\right) \quad (5)$$

where N_{po} is the initial number density of the parent atoms. $N_p(t)$ and $N_d(t)$ are the number densities of the parent and daughter atoms at any time t during reactor operation, respectively. σ_p is the total reaction cross section for the parent atom and σ_{pd} is the parent atom reaction cross section that leads to the formation of the daughter radionuclide. β_d is the daughter radionuclide destruction constant (destruction rate plus decay constant). ϕ is the neutron flux.

From the nonlinearity of the rate equation, it is clear that performing two separate activity calculations for the D-D and D-T neutron source components and adding the results yields conservative results due to underestimating the destruction of the parent atoms. The overestimation ratio depends on the half-life daughter radionuclide, the time of operation, the magnitude of the neutron flux and the destruction cross section of the parent atoms.

Table I. Activity Overestimation Ratios for Some Naturally Occurring Elements.

Element	Daughter	Half-life	Ratio
H	H-3	12.3 y	2.6
He	H-3	12.3 y	1.9
Li	H-3	12.3 y	2.0
B	Be-10	1.6e6 y	2.3
V	V-49	337 d	1.5
Co	Mn-56	2.578 h	1.43
	Fe-59	44.51 d	1.42
	Co-58	70.88 d	1.42
	Co-58m	9.1 h	1.43
	Co-60	5.271 y	1.22
	Co-60m	10.47 m	1.3
Mo	Nb-95	34.97 d	1.4
	Nb-95m	3.61 d	1.55
Ag	Ag-106	24 m	1.5
	Ag-106m	8.5 d	1.5
	Ag-108	2.39 m	1.34
	Ag-108m	130 y	1.2
	Ag-110	24.6 s	16.8
	Ag-110m	249.8 d	15.0
In	Ag-108	2.39 m	2.9
	Ag-110	24.6 s	3.1
	Cd-115	2.228 d	8.5e+3
	In-112	14.4 m	2.9
	In-114	1.198 m	2.22
	In-114m	49.51 d	2.21
	In-116	14.1 s	3.1e+3
	In-116m	54.2 m	3.1e+3
Sb	In-116	14.1 s	1.8
	In-118	5 s	3.4
	Sn-121	1.128 d	1.8
	Sn-123	129.2 d	3.35
Ho	Ho-163	4.57e3 y	2.1
	Ho-164	29 m	9.2
	Ho-164m	38 m	9.2
	Ho-166	1.117 d	5.5
	Ho-166m	1.2e3 y	1.7
Lu	Lu-173	1.37 y	16.5
	Lu-174	3.3 y	8.2
	Lu-174m	142 d	20.0
	Lu-176m	3.66 h	11.1
	Lu-177	6.68 d	3.8e+6
	Lu-177m	160.7 d	2.2e+6
Ta	Lu-177	6.68 d	6.6
	Hf-181	42.4 d	6.6
	Ta-179	1.8 y	5.5
	Ta-180	1.2e15 y	1.9
	Ta-180m	8.15 h	6.6
	Ta-182	114.43 d	4.1
	Ta-182m	15.8 m	4.2
W	Hf-181	42.4 d	1.1
	Hf-183	1.07 h	1.2e+3
	Ta-179	1.8 y	34.0
	Ta-182	114.43 d	9.8
	Ta-183	5.1 d	2.9
	Ta-184	8.7 h	1.1
	Ta-185	49 m	115.1
	Ta-186	10.5 m	115.2
	W-179	38 m	55.2
	W-181	121.2 d	18.9
	W-185	74.8 d	1.1
	W-187	23.9 h	49.6
Re	W-185	74.8 d	283.7
	W-187	23.9 h	3.41
	Re-184	38 d	284.7
	Re-184m	165 d	260.2
	Re-186	3.777 d	27.8
	Re-188	16.94 h	2.5
	Re-188m	18.6 m	2.5
Au	Ir-194	19.3 h	45.7
	Pt-197	18.3 h	45.6
	Au-195	186.12 d	41.2
	Au-196	6.18 d	45.5
	Au-196m	9.7 h	45.7
	Au-198	2.694 d	21.6

In Table I, a list is given for the daughter radionuclides of some naturally occurring elements and for which the overestimation ratios were significant. As shown in the table, the overestimation can be as high as six orders of magnitude for isotopes like ^{177}Lu and ^{177m}Lu. The lutetium isotopes have half-lives on the order of several days. It is clear that the overestimation ratios are particularly small for long-lived radionuclides. The reason is that the second exponential term in Equation 5 determined by the daughter nuclide destruction constant starts to decrease the impact of the parent atom destruction cross section in the first exponential term. The overestimation ratio is also apparent for several radionuclides which are of special interest to reactor safety analysts. ^{187}W($T_{1/2} = 23.9$ hr), which is a major source of concern in case of a loss of coolant accident (LOCA) due to its high decay heat, has an overestimation ratio of about 50. In case of radioactive inventory, ^{187}W was also found to be the major source of the offsite doses produced from the tungsten coating of divertors or steel structures with high tungsten content. Figure 1 shows the overestimation ratio of the ^{187}W activity induced in the first wall of a typical D-^{3}He reactor as a function of operation time. One should note that all the activity values in the figure are normalized to the value of the ^{187}W activity induced by only the D-D component of the neutron source.

It is interesting to notice that the overestimation ratio increases with the reactor operation time. After one year of operation, the ^{187}W activity is only overestimated by a factor of 1.1. Another important radioactive nuclide is ^{188}Re. ^{188}Re($T_{1/2} = 16.94$ hr) is of special importance due to its high volatility rate [6]. After 30 years of reactor operation, ^{188}Re activity would be overestimated by a factor of 2.5.

A Case Study: ARIES-III Reactor

Activation analysis for the ARIES-III D-^{3}He reactor was conducted. Time of operation was taken to be 30 full power years (FPY). The low activation ferritic steel modified HT-9 was used as a structural material in the first wall and shield of the reactor. To show the dependence of the overestimation ratio on the destruction cross section of the parent atom, we

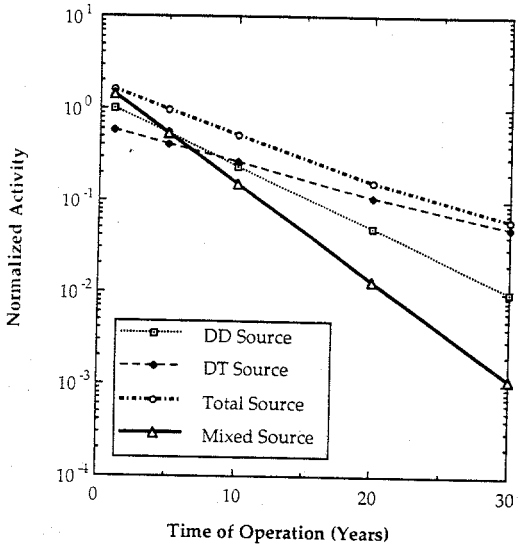

Figure 1. Activity of ^{187}W in the first wall.

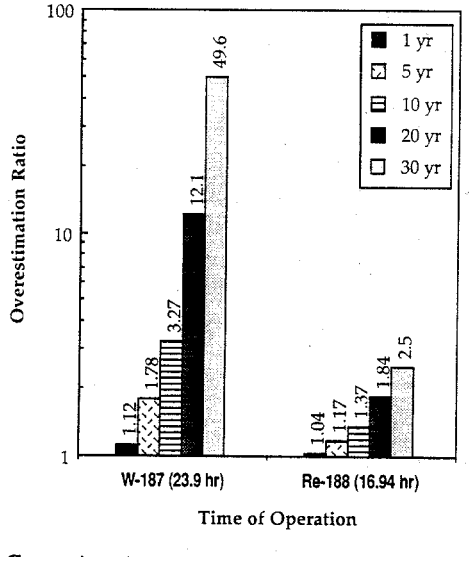

Figure 2. Comparison between the activity induced in the first wall by radioisotopes with comparable half-lives.

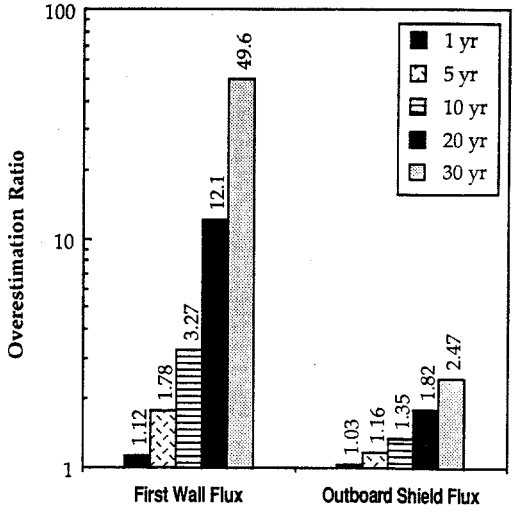

Figure 3. Overestimation of ^{187}W activity at different flux levels.

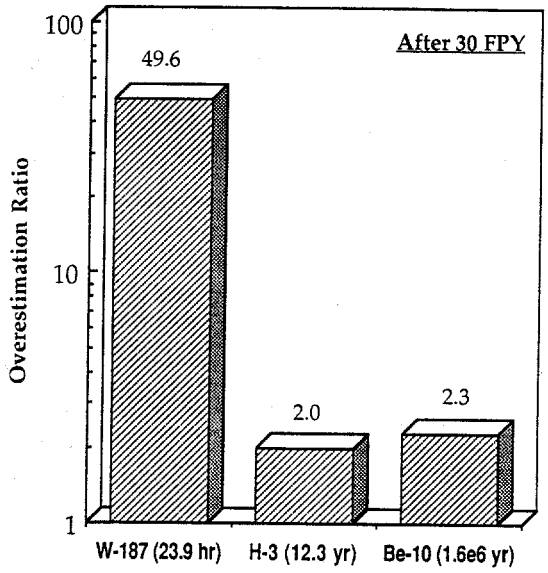

Figure 4. Comparison between the activity induced in the first wall by radioisotopes with comparable production rates.

produced Figure 2 which shows the overestimation ratio as a function of operation time for ^{187}W and ^{188}Re. The two radionuclides have comparable half-lives and are produced in the reactor first wall and hence exposed to the same neutron flux. The major difference between the two isotopes is that ^{186}W, which is the parent nuclide for ^{187}W, has a destruction cross section which is four times that of ^{187}Re which is the parent nuclide of ^{188}Re. As shown in the figure, the overestimation ratio of ^{187}W is higher than that for ^{188}Re for all times of operation. However, one should note that the difference in the overestimation ratio increases as the reactor operation time increases. After 30 FPY, the overestimation ratio for ^{187}W is about 20 times that for ^{188}Re.

Figure 3 shows the impact of the neutron flux on the overestimation ratio. At reactor shutdown, the overestimation of ^{187}W activity generated in the reactor first wall is about twenty times its value at the back of the outboard shield. In this analysis, the first wall neutron flux is four times that at the back of the outboard shield. Finally we examined the impact of the radionuclide half-life on the overestimation ratio. As shown in Figure 4, we selected the three isotopes ^{187}W($T_{1/2} = 23.9$ hr), ^{3}H($T_{1/2} = 12.3$ yr) and ^{10}Be($T_{1/2} = 1.6 \times 10^{r6}$ yr) to illustrate the half-life effect. ^{187}W, ^{3}H and ^{10}Be are produced from ^{186}W, ^{6}Li and ^{10}B, respectively. The reason for selecting these three parent nuclides is that they have comparable destruction cross sections. The overestimation is particularly significant for isotopes with short or intermediate half-life. Its significance diminishes for long-lived nuclides. In fact both ^{3}H and ^{10}Be (long-lived nuclides) which have six orders of magnitude difference in the half-life have the same overestimation ratios.

The values in Table I were produced using the hard first wall neutron spectrum. The overestimation ratios in the table present the worst case values. The ratios are expected to decrease as the neutron spectrum gets softer as a result of slowing down in the reactor shield. We calculated the total activity and decay heat generated in the reactor first wall and shield at reactor shutdown. Figure 5 shows the activity induced by each of the neutron source components in addition to the activity induced if the mixed source is used in the calculations. At shutdown, the overestimation ratio is about 40%. It is interesting to note here that the ^{188}Re activity is overestimated by only 20% compared to 250% in the first wall alone. At the same time, the ^{187}W activity is overestimated by a factor of only 2.65 compared to 49.6 in the first wall. The decay heat results are shown in Figure 6. The absence of the ^{188}Re contribution is due to the lack of its decay data in the decay library used in this analysis.

We calculated the whole body early offsite dose following the acci-

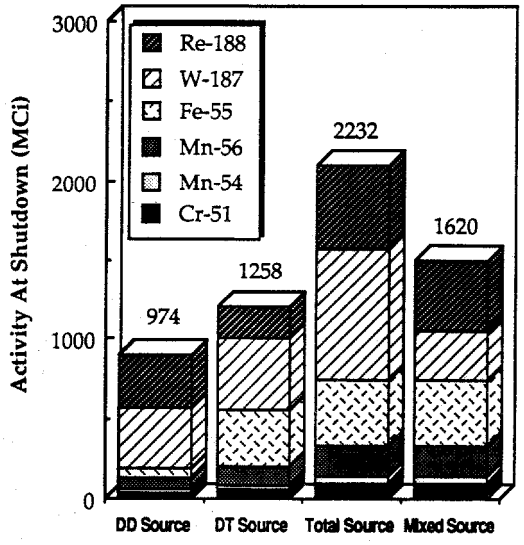

Figure 5. Total activity in the reactor structure.

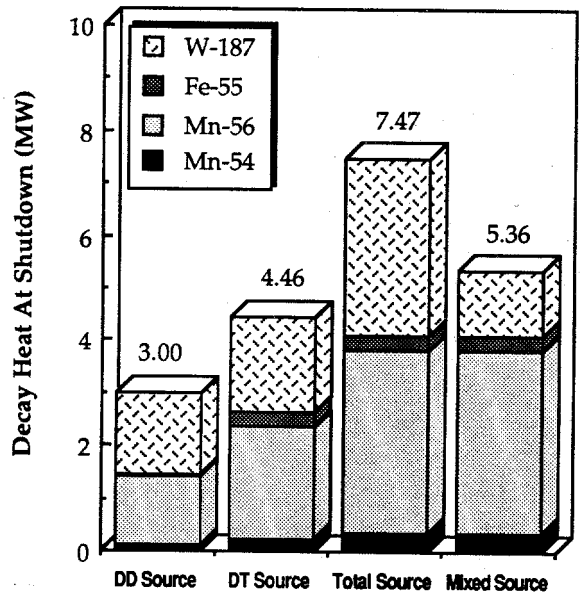

Figure 6. Total decay heat in the reactor structure.

dental release of the reactor activation products. In order to see the impact of the overestimation of the dose if one uses the two neutron source components rather than the mixed source, we calculated the dose produced by the release of 100% of the radioactive inventory. The results shown in Figure 7 show that the overestimation in the calculated dose is about 27%. The dose is dominated by ^{54}Mn, ^{188}Re and ^{187}W. As one would expect the overestimation ratios for the doses produced by ^{187}W and ^{188}Re are the same as these calculated for their induced activities.

Conclusions

The mixed neutron source should be always used in any activation and safety analysis performed for D-^{3}He reactors. Performing any activation analysis by using the two components of the neutron source (D-D and D-T) separately leads to overestimating the induced activity. The overestimation is controlled by the destruction rate of the parent atoms, magnitude of the neutron flux, reactor operation time and the radionuclide half-life. The overestimation was found to be more pronounced for short and intermediate lived nuclides. In a typical D-^{3}He reactor, the overestimation in the calculated activity and decay heat was as high as 40% after 30 years of reactor operation. In the meantime, the offsite dose was overestimated by 27%.

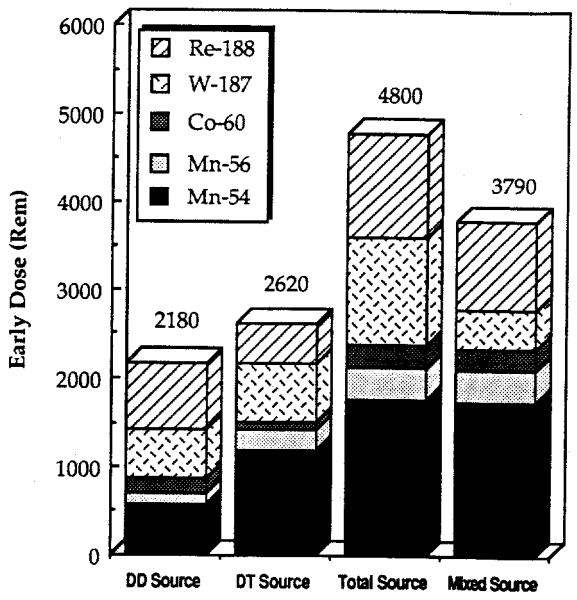

Figure 7. Whole body early dose at 1 km from the reactor.

Acknowledgement

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References

[1] D. L. Henderson and O. Yasar, "DKR-ICF: A Radioactivity and Dose Rate Calculation Code Package," UWFDM-714, Vol. 1, University of Wisconsin, April 1987.

[2] M. A. Gardner and R. J. Howerton, "ACTL: Evaluated Neutron Activation Cross Section Library - Evaluation Techniques and Reaction Index," UCRL-50400, Vol. 18, Lawrence Livermore National Laboratory, 1978.

[3] R. O'Dell et al., "User's Manual for ONEDANT: A Code Package for One-Dimensional, Diffusion-Accelerated, Neutral Particle Transport," LA-9184-M, Los Alamos National Laboratory, 1982.

[4] Lisa J. Porter, "Upgrade of a Fusion Accident Analysis Code and Its Application to a Comparative Study of Seven Fusion Reactor Designs," PFC/RR-89-10, Massachusetts Institute of Technology, June 1989.

[5] D. Smith et al., "Blanket Comparison and Selection Study," ANL/FPP/TM-122, Argonne National Laboratory, 1984.

[6] G. R. Smolik et al., "Volatility of Copper and Tungsten Alloys for Fusion Reactor Applications," Proceedings of the IEEE 13th Symposium on Fusion Engineering, 2-6 October 1989, Knoxville, TN., pp. 670-673 (1989).