The Impact of Reactor Operating Time on Activation Levels for Safety Analyses

H.Y. Khater and M.E. Sawan

March 1992

UWFDM-888

Prepared for the 10th Topical Meeting on the Technology of Fusion Energy, 7–12 June 1992, Boston MA.

FUSION TECHNOLOGY INSTITUTE

UNIVERSITY OF WISCONSIN

MADISON WISCONSIN

The Impact of Reactor Operating Time on Activation Levels for Safety Analyses

H.Y. Khater and M.E. Sawan

Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

March 1992

UWFDM-888

Prepared for the 10th Topical Meeting on the Technology of Fusion Energy, 7–12 June 1992, Boston MA.

THE IMPACT OF REACTOR OPERATING TIME ON ACTIVATION LEVELS FOR SAFETY ANALYSES

H. Y. KHATER and M. E. SAWAN
Fusion Technology Institute
University of Wisconsin-Madison
Madison, Wisconsin 53706-1687
(608) 263-2167

ABSTRACT

Radioactivity induced in a typical fusion power reactor was calculated for all elements with atomic number Z $\leq$ 84 and for different irradiation times. It was shown that the shutdown activity varies widely with the duration of the irradiation time. In general, the activity induced by radionuclides with half-lives that are significantly longer than the period of irradiation increases with increasing the irradiation time. On the other hand, the level of activity generated by any radionuclide with a half-life which is significantly shorter than the reactor lifetime reaches a peak early during irradiation and then may start to drop to a lower value before the end of irradiation. The severity of this peaking is determined by the destruction rate of the parent element. The activities generated by long-lived nuclides (important for waste management) in any fusion reactor with a lifetime on the order of 30 years reach their peak values at end-of-life. In the mean time, using the activity and decay heat values generated by short and intermediate-lived radionuclides at the end of reactor life to represent the worst case values used in safety analyses related to a loss of coolant accident (LOCA) and accidental release of radioactive inventory might lead to a substantial underestimation of the results.

I. INTRODUCTION

The future public acceptance of fusion power reactors will be definitely influenced by their safety and environmental characteristics. The amount of radioactivity present in any reactor at the end of its life will determine the level of radioactive waste that has to be disposed. Disposal of the radioactive waste as a low level waste (LLW) is one of the goals envisioned by the U.S. fusion community in any reactor design. In the mean time, safety analysts are equally concerned with the potential release of the radioactive inventory of any fusion reactor during any possible accident. In any safety analysis, researchers are usually concerned with postulating the worst possible accident under the worst possible conditions. There is no doubt that a worst accident scenario will have to involve the release of the highest amount of radioactivity to the environment. The two main factors that control such a process are the amount of radioactivity present in the reactor at the onset of the accident and the amount of energy available to release it. In most loss of coolant accidents (LOCA), the decay heat generated in the reactor structure represents the major source of energy available to mobilize its radioactive inventory. Keeping in mind the need to quantify the highest possible levels of activity and decay heat generated in any fusion reactor during an accident, it has been common to use the radioactivity values calculated at the end of the reactor life as worst case results.

In this paper we show that assuming that the highest radioactivity generated in any fusion reactor occurs at its end-of-life is only accurate if one is mainly concerned with identifying the level of radioactive waste generated during the reactor life-time. Most of the radionuclides which are of concern to waste management according to the current U.S. Nuclear Regulatory Commission (NRC) 10CFR61[1] regulation are those with long half-lives. In fact the 10CFR61 document only gives limits to the allowed specific activity of radioactive nuclides with half- lives $\geq$ 5 years. On the other hand, safety analysts can not assume that the end-of-life radioactivity generated by radionuclides with short or intermediate half-lives represents the worst case values especially for a typical fusion reactor with as long as a 30 year lifetime. A similar situation has been identified for ITER with a relatively less pronounced effect due to the lower flux and shorter reactor lifetime compared to power reactors[2]. In this paper we quantify the severity of this problem for all elements with atomic mass Z $\leq$ 84.

II. CALCULATIONAL PROCEDURE

Radioactivity calculations were conducted using the DKR-ICF[3] code with activation cross sections taken from the ACTL library and for irradiation times up to 30 years. The neutron flux used in the activation analysis is generated by the one-dimensional discrete ordinates neutron-transport code ONEDANT[4] along with the ENDF/B-V cross section data files. Using a neutron flux similar to the first wall flux of a typical D-T reactor, the DKR-ICF code was used to calculate the radioactivity induced in several target elements. All naturally occurring elements (parents) in the periodic table with atomic mass Z $\leq$ 84 were considered in this paper. The radioactivity calculations were performed for seven different operation

times. Shutdown activities were calculated for 1 day, 1 week, 1 month, 6 months, 1 year, 10 years and 30 years of irradiation.

Another detailed analysis was performed to examine the impact of accident occurrence during reactor operation on the results of the safety analysis of a selected D-^{3}He reactor design. The design selected is similar to the ARIES-III design.[5] Radioactivity calculations were conducted for the low activation ferritic steel (modified HT-9) first wall and shield of the reactor. Finally, the activation results were utilized in an off-site dose calculation performed according to the worst case weather conditions using the FUSCRAC3 code.[6]

III. THE IMPACT OF IRRADIATION TIME ON THE LEVEL OF INDUCED ACTIVITY

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:

$$N_p(t) = N_{p\circ} e^{-\sigma_p \phi t} \quad (1)$$

$$N_d(t) = \frac{N_{p\circ}\sigma_{pd}\phi}{(\beta_d - \sigma_p\phi)} (e^{-\sigma_p\phi t} - e^{-\beta_d t}), \quad (2)$$

where $N_{p\circ}$ is the initial number density of the parent atom, $N_p(t)$ and $N_d(t)$ are the number densities of the parent and daughter atoms at any time during reactor operation, respectively, σ_p is the parent atom total reaction cross section 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 and t is the irradiation time.

From the nonlinearity of the rate equation, it is clear that the level of radioactivity generated by any radionuclide at the end of irradiation is controlled by the destruction rate of its parent atom. Assuming that longer irradiation times yield higher shutdown activity might not be correct especially if the parent atom has a high destruction cross section. This behavior is more pronounced for radionuclides with short or intermediate half-lives. The peak radioactivity for such radionuclides is usually observed within the first year of irradiation and the level of activity starts to drop with an increase of irradiation time. Using the reactor end-of-life activity in worst case safety analysis of any fusion reactor could result in underestimating the radiological hazard associated with each of the radionuclides present in the reactor at the onset of the accident. In addition to the daughter radionuclide half-life, the level of underestimation is also dependent on the reactor lifetime, the destruction cross section of the parent atoms and the magnitude of the neutron flux. One would expect to observe this behavior more clearly in the reactor first wall and the first few layers of its shield than in layers at the back of the shield which are exposed to a much lower neutron flux.

In Table I, a list is given for the daughter radionuclides of some naturally occurring elements for which the underestimation ratios ≥ 1.4. The underestimation ratios presented in the table are the ratios between the peak activities to the end-of-life activities (30 years). T_{max} is the time during operation at which the radionuclide activity reaches its maximum value. For reactors with a lifetime of 30 years, the underestimation can be as high as seventeen 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 underestimation ratios are particularly small for long-lived radionuclides. The reason is that the second exponential term in Eq. 2 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 underestimation 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 the case of a loss of coolant accident (LOCA) due to its high decay heat, has an underestimation ratio of about 10^7. ^{187}W is also considered to be the major source of off-site doses produced during accidents involving tungsten coated conventional divertors. However, this should not be a major source of concern as divertors are assumed to be replaced every 1-3 years of operation, a period of time at which ^{187}W activity is expected to still be at its peak value. Another important radioactive nuclide is ^{188}Re($T_{1/2} = 16.94$ hr) which is of a special importance due to its high volatility rate. After 30 years of reactor operation, ^{188}Re activity would be underestimated by a factor of 78.

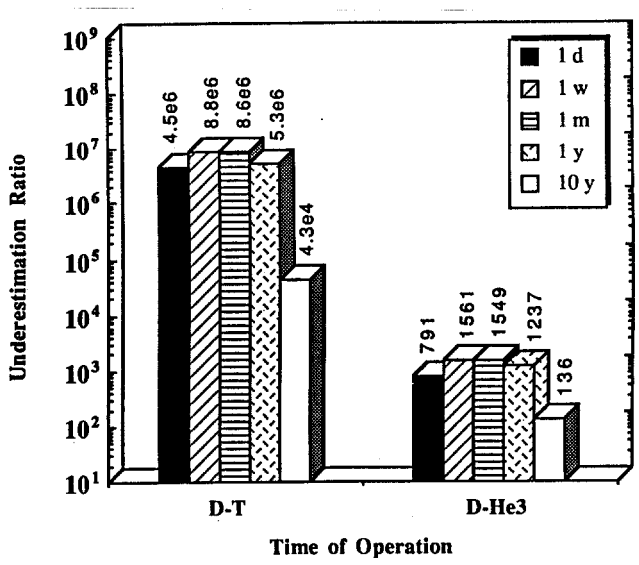

Fig. 1. Underestimation of the ^{187}W activity induced in the first wall of D-T and D-^{3}He reactors.

It is important to note that the underestimation ratios presented in Table I are produced using a high first wall neutron flux which represents the worst possible nuclear environment. The neutron flux used is similar to a typical D-T first wall flux. In general, for a fixed operation time the underestimation ratio will drop significantly with any decrease in the energy integrated reaction rate ($\sigma_p\phi$). Figure 1 shows the impact of $\sigma_p\phi$ on the underestimation ratio of ^{187}W by comparing the results

Table I.

Activity Underestimation Ratios for Some Naturally Occurring Elements

Element	Daughter	Half-life	T_{max}	Ratio
H	H-3	12.3 y	1 y	3.74
Li	H-3	12.3 y	10 y	1.44
Cl	S-35	87.2 d	1 y	1.51
	Cl-34m	32.2 m	1 d	1.80
V	V-49	337 d	10 y	2.35
Mn	V-52	3.76 m	1 d	1.42
	Cr-55	3.497 m	1 d	1.42
	Mn-54	312.2 d	10 y	1.26
	Mn-56	2.578 h	1 w	1.42
Co	Mn-56	2.578 h	1 w	3.61
	Fe-59	44.51 d	1 y	3.42
	Co-58	70.88 d	1 y	3.36
	Co-58m	9.1 h	1 w	3.61
	Co-60	5.271 y	10 y	1.49
	Co-60m	10.47 m	1 d	3.61
Zn	Cu-67	2.58 d	1 m	1.55
Nb	Y-90	2.67 d	1 m	1.39
	Nb-92m	10.13 d	6 m	1.38
	Nb-94m	6.26 m	1 d	1.39
Mo	Nb-95	34.97 d	1 y	3.31
	Nb-95m	3.61 d	1 m	5.05
	Nb-96	23.4 h	1 m	1.55
	Nb-97m	58.1 s	1 d	1.48
Ag	Ag-106	24 m	1 d	4.12
	Ag-106m	8.5 d	6 m	4.02
	Ag-108	2.39 m	1 d	4.19
	Ag-110	24.6 s	1 d	4.01e+04
	Ag-110m	249.8 d	1 y	1.46e+04
In	Ag-108	2.39 m	1 d	3.24e+01
	Ag-110	24.6 s	1 d	1.93e+02
	Cd-115	2.228 d	1 m	1.29e+11
	In-112	14.4 m	1 d	3.24e+01
	In-114	1.198 m	6 m	7.31e+01
	In-114m	49.51 d	6 m	5.67e+01
	In-116	14.1 s	1 d	1.38e+11
	In-116m	54.2 m	1 d	1.38e+11
Sn	Cd-109	462 d	10 y	1.50
	In-110	1.15 h	1 d	1.85
	In-111	2.805 d	1 m	1.84
	In-112	14.4 m	1 d	1.85
	In-112m	20.8 m	1 d	1.85
	In-117m	1.94 h	1 d	1.41
	Sn-111	35 m	1 d	1.85
Sb	In-116	14.1 s	1 d	7.35
	In-118	5 s	1 d	7.26e+01
	Sn-121	1.128 d	1 m	7.31
	Sn-123	129.2 d	1 y	5.27e+01
Ho	Ho-164	29 m	1 d	1.44e+03
	Ho-164m	38 m	1 d	1.44e+03
	Ho-166	1.117 d	1 w	1.42e+03

Table I. (Continued)

Element	Daughter	Half-life	T_{max}	Ratio
Lu	Lu-173	1.37 y	1 y	3.97e+03
	Lu-174	3.3 y	10 y	3.89e+01
	Lu-174m	142 d	1 y	1.98e+04
	Lu-176m	3.66 h	1 w	3.89e+04
	Lu-177	6.68 d	1 m	8.84e+17
	Lu-177m	160.7 d	6 m	4.74e+16
Hf	Hf-181	42.4 d	1 y	1.95
Ta	Lu-177	6.68 d	1 m	4.45e+02
	Hf-181	42.4 d	6 m	4.03e+02
	Ta-179	1.8 y	1 y	7.52e+01
	Ta-180m	8.15 h	1 w	4.71e+02
	Ta-182	114.43 d	1 y	3.33e+02
	Ta-182m	15.8 m	1 d	4.73e+02
W	Hf-181	42.4 d	1 y	1.61
	Hf-183	1.07 h	1 d	9.06e+06
	Ta-179	1.8 y	1 y	8.60e+03
	Ta-182	114.43 d	1 y	4.90e+02
	Ta-183	5.1 d	1 m	3.99e+01
	Ta-184	8.7 h	1 w	1.63
	Ta-185	49 m	1 d	9.06e+06
	Ta-186	10.5 m	1 d	9.05e+06
	W-179	38 m	1 d	5.09e+05
	W-181	121.2 d	1 y	1.75e+03
	W-185	74.8 d	1 y	2.23
	W-187	23.9 h	1 w	8.91e+06
Re	W-185	74.8 d	6 m	1.25e+07
	W-187	23.9 h	1 w	7.76e+01
	Re-184	38 d	6 m	1.30e+07
	Re-184m	165 d	1 y	7.59e+06
	Re-186	3.777 d	1 m	4.39e+03
	Re-188	16.94 h	1 w	7.81e+01
	Re-188m	18.6 m	1 d	7.84e+01
Au	Ir-194	19.3 h	1 w	1.17e+05
	Pt-197	18.3 h	1 w	1.17e+05
	Au-195	186.12 d	1 y	5.02e+04
	Au-196	6.18 d	1 m	1.10e+05
	Au-196m	9.7 h	1 w	1.17e+05
	Au-198	2.694 d	1 m	1.14e+05
Hg	Hg-203	46.61 d	1 y	1.42
Tl	Hg-203	46.61 d	1 y	1.83
	Tl-202	12.23 d	6 m	1.86

obtained using typical D-T and D-^{3}He first wall fluxes. The underestimation of ^{187}W activity generated in the first wall of a D-T reactor is more than three orders of magnitude higher than its value in the first wall of the D-^{3}He reactor ARIES-III.

Comparing the underestimation ratios of ^{187}W and ^{188}Re shows the dependence of the underestimation ratio on the parent atom destruction cross sections. Even though the two radionuclides have comparable half-lives and were both produced using the same neutron flux, their underestimation ratios differ by five orders of magnitude. The major difference between the two isotopes is that, ^{186}W which is the parent atom of ^{187}W, has a destruction rate which is about 500 times that of ^{187}Re

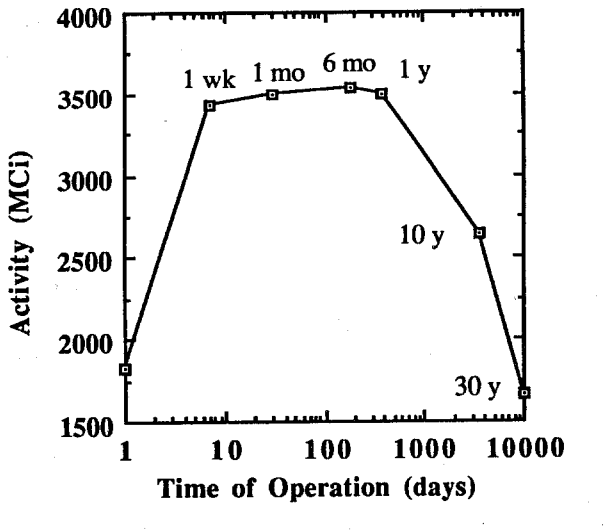

Fig. 2. ARIES-III shutdown activity as a function of operation time.

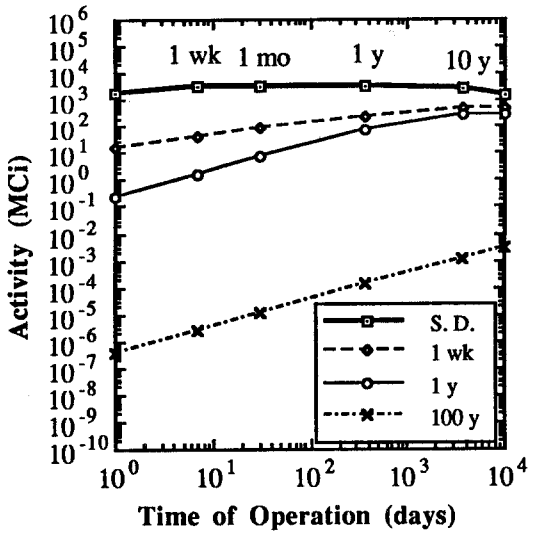

Fig. 3. Activity in ARIES-III after shutdown as a function of operation time.

which is the parent atom of ^{188}Re. Another parameter examined is the impact of the radionuclide half-life on the underestimation ratio. Both parents of ^{188}Re($T_{1/2}$ = 16.94 hr) and ^{3}H($T_{1/2}$ = 12.3 yr), ^{187}Re and ^{6}Li, respectively, have comparable destruction rates. However, ^{188}Re has an underestimation ratio of 78 compared to only a ratio of 1.44 for ^{3}H. This clearly shows that the underestimation is particularly significant for isotopes with short or intermediate half-lives and that its significance diminishes for long-lived nuclides. For instance, ^{94}Nb ($T_{1/2}$ = 2 × 10^4 yr), which is a major source of concern for waste management, has a ratio of one.

IV. A CASE STUDY: ARIES-III REACTOR

An activation analysis for the ARIES-III D-^{3}He reactor was conducted at several operation times. The low activation ferritic steel modified HT-9 was used as a structural material in the first wall and shield of the reactor. As mentioned in the previous section, the values in Table I were produced using the first wall neutron flux in a D-T reactor. Hence, the underestimation ratios in the table present the worst case values. For a fixed operation time, the ratios are expected to decrease as the neutron flux gets lower through the reactor shield. In addition, spectrum softening in the shield will impact the ratios (up or down) depending on the type of reaction producing the radionuclide of interest. We calculated the total first wall and shield activity and decay heat generated in ARIES-III at shutdown and for different operation times. Figure 2 shows the shutdown activity generated in the reactor structure during different operation times. The activity induced in the reactor after 6 months of operation is more than twice its value after 30 years of operation. It is interesting to notice here that after 30 years of operation, the total ^{187}W activity is only underestimated by a factor of 7 compared to a factor of over 1500 in the first wall alone. This larger difference in the underestimation ratio clearly shows the impact of decreasing the neutron flux throughout the rest of the reactor shield.

Figure 3 shows the level of activity present in the reactor during the first 10 years following its shutdown after different operation times. As shown in the figure, the impact of operation time on the level of activity in the reactor starts to disappear after a week from shutdown. ^{54}Mn ($T_{1/2}$ = 312.2 day), ^{55}Fe ($T_{1/2}$ = 2.7 yr) and ^{185}W ($T_{1/2}$ = 75.1 day) are the dominant sources of radioactivity during this period of time. An examination of the level of activity at 100 years following shutdown shows a linear increase with the increase of operation time. As discussed before, this is due to the fact that the reactor activity at this period of time is solely dominated by nuclides with long half-lives. ^{63}Ni ($T_{1/2}$ = 100 yr), ^{14}C ($T_{1/2}$ = 5730 yr) and ^{53}Mn ($T_{1/2}$ = 3.8 × 10^6 yr) dominate the long-term activity in ARIES-III. Similarly, the decay heat generated in the reactor during the first week of operation is about three times its value after 30 years. As with the activity, the underestimation ratio can not be detected after a week from the reactor shutdown regardless of the duration of its operation.

As one final step in the analysis we calculated the whole body early off-site dose following the accidental release of the reactor activation products. In order to see the impact of the underestimation of the dose if one assumes several operation times, we calculated the dose produced by the release of the radioactive inventory. Since the full mobilization of the radioactive inventory in ARIES-III is highly unlikely, the amount of radioactivity released to the environment in an accident would be determined by the highest temperature of the shield. We calculated the whole body (WB) early off-site dose using steel experimental volatility rates[7] at 1200°C as a conservative assumption. During a one hour LOCA, the maximum WB early

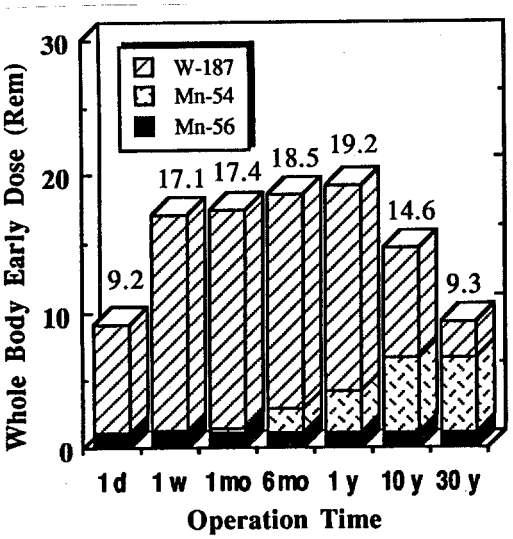

Fig. 4. Whole body early dose at 1 km from ARIES-III as a function of operation time.

dose at the reactor site boundary (1 km) is 19.2 rem and occurs after only one year of operation (Fig. 4). Using the shutdown activity at the end of the reactor life (30 years) to calculate the off-site dose result in a WB early dose of only 9.3 rem. ^{187}W, ^{54}Mn and ^{56}Mn dominate the off-site dose regardless of the time of operation used. The activity of ^{56}Mn reaches its maximum level within the first day of operation and remains at the same level throughout the reactor life. On the other hand, ^{54}Mn starts contributing significantly to the calculated dose after one month of operation and results in more than 60% of the off-site dose after 30 years. The dose produced by ^{187}W reaches its peak level within the first week of operation and the level is sustained during the first year. The off-site dose caused by the release of ^{187}W if an accident occurred during the first year of operation is more than six times the ^{187}W dose produced if the accident occurred at the end of the reactor life.

Based on the above analysis it is clear that the radioactivity related parameters calculated at reactor end-of-life can be used to represent the worst conditions when analyzing waste management. On the other hand, safety and environmental consequences resulting from LOCA and accidental radioactive release should be determined using the activation result with the appropriate irradiation time that corresponds to the worst case with peak radioactivity related parameters.

V. SUMMARY

The accident with worst radiological consequences involving fusion reactors would most probably occur sometime during the reactor operation rather than at the end of its lifetime (30 years). Using the radioactive inventory present in any fusion reactor at the end of its life in any safety analysis might lead to a substantial underestimation of the accident radiological consequences. Radioactivity calculations were performed for all elements with atomic mass Z $\leq$ 84 and for different irradiation times using a typical first wall neutron flux of a D-T fueled fusion reactor. For a typical power reactor life-time of 30 years, the levels of activity generated by most of the radioactive daughters of Ag, In, Sb, Ho, Ta,W, Re and Au reached their peak values within the first year of irradiation. In several cases, it was found that the peak activity could be several orders of magnitude higher than the reactor end-of-life activity. The activity of ^{187}W reaches its peak value after one week of irradiation. ^{187}W sustains the same level of activity within the first year of irradiation which is about seven orders of magnitude higher than its value after 30 years. The end-of-life activity of ^{188}Re is a factor of eighty lower than its value after only one week of irradiation. In general, the underestimation was found to be more pronounced for short and intermediate-lived nuclides. In a typical D-^{3}He reactor (ARIES-III), the radioactivity calculated after one week and up to the first year of operation was two to three times its value after 30 years of operation. In the same time the calculated value of the whole body early off-site dose after one year of operation was more than twice its value after 30 years.

ACKNOWLEDGEMENT

Support for this work was provided by the U.S. Department of Energy.

REFERENCES

1. Nuclear Regulatory Commission, 10CFR part 61, "Licensing Requirements for Land Disposal of Radioactive Waste," Federal Register, FR 47, 57446 (1982).

2. H. ATTAYA, "LOCA Analysis for Manganese-Stabilized Steel," Fusion Technology, Vol. 19(3), Part 2A, pp. 1331-1336 (1991).

3. 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).

4. 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).

5. F. NAJMABADI et al., "The ARIES-III Design," Proceedings of the IEEE 14th Symposium on Fusion Engineering, September 30 - October 3, 1991, San Diego, CA.

6. L. 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).

7. S. J. PIET et al., "Initial Experimental Investigation of the Elemental Volatility from Steel Alloys for Fusion Safety Applications," EGG-FSP-8459, Idaho National Engineering Laboratory (April 1989).