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ACTIVATION CROSS SECTIONS IMPROVEMENTS NEEDED FOR IFE POWER REACTORS DESIGNS

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Uncertainties in the prediction of the neutron induced long-lived activity in the natural elements from H to Bi due to activation cross section uncertainties are estimated assuming as neutron environment those of the HYLIFE-II and Sombrero vessel structures. The latest available activation cross section data are employed. The random variables used in the uncertainty analysis have been the concentration limits (CL´s) corresponding to hands-on recycling, remote recycling and shallow land burial, quantities typically considered in ranking elements under waste management considerations. The CL standard value (CL_{nom}), i.e. without uncertainties, is compared with the 95th percentile CL value (CL95). The results of the analysis are very helpful in assessing the quality of the current activation data for IFE applications, providing a rational basis for programmatic priority assignments for new cross sections measurements or evaluations. The HYLIFE-II results shown that a significant error is estimated in predicting the activation of several elements. The estimated errors in the Sombrero case are much less important.

I. INTRODUCTION

The activation assessment of all chemical elements under waste management considerations was performed [1,2,3,4] to define Low Activation (LA) material specifications for the first structural wall of thick-liquid protected IFE chambers. The effect of cross section uncertainties in the long-lived activity of some important elements was specifically addressed in [3,4], by using a comprehensive first-order Taylor series sensitivity-uncertainty analysis method. This methodology although was found practical for providing the uncertainties of the concentration limits due to the uncertainty of each of the reaction cross sections separately, it was found impractical to deal with the synergetic/global effect of the uncertainties of the complete set of cross sections. To overcome this limitation a Monte Carlo procedure was developed [5] and successfully applied to deal with typical operational scenarios of inertial fusion experimental facilities [6].

In this paper we apply the Monte Carlo method to estimate the uncertainties in the long-lived activity of all the natural elements due to the cross section uncertainties. Most of the results are provided for the neutron environment of the HYLIFE-II [7] (thick-liquid wall concept) vessel structure, but also some are presented for the Sombrero[8] (dry wall concept) design. Elements leading to significant uncertainty in the long-lived activity are identified (section 3). Some of the critical cross sections responsible of this uncertainty are selected, and the effect of their improvement is quantified (section 4).

The latest available activation cross section library (EAF-2003) is employed[9]. Comparison with results obtained using the current IAEA activation cross section library (FENDL/A-2.0)[10] is also presented.

II. PROBLEM DESCRIPTION AND CALCULATIONAL METHOD.

Two different neutron environments corresponding to IFE conceptual power plants are considered. The first is taken from the midplane region of the HYLIFE reactor vessel. The flux intensity is 1.29×10^{15} n/cm^2 s and the average neutron energy is 0.426 MeV. We have assumed a continuous irradiation of 30 years (desirable FSW lifetime). The second scenario is taken from de FSW of the SOMBRERO reactor. The flux intensity is 9.55×10^{15} n/cm^2 s and the average neutron energy is 3.067 MeV. We have assumed a continuous irradiation of 5 yr.

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The neutron flux has been calculated by the TART Monte Carlo transport code[11]. Radionuclide inventory, contact _-dose rate, and the waste disposal ratings (WDR) have been computed with the ACAB code[5]. The nuclear data libraries used for inventory calculation are those from The European Activation File EAF-2003 (EAF_XS, EAF_UN, and EAF_DEC)[9]. Also the FENDL nuclear data (FENDL/A-2.0 and FENDL/D-2.0)[10] have been used for comparison purposes.

In defining CL's for recycling, two options are considered: hands-on recycling is acceptable when the contact dose rate doses not exceed 10 _Sv/h at 100 yr cooling, and remote recycling when the dose rate is kept below 10 mSv/h within 50 yr cooling. The CL's on each of the elements are calculated by assuming the element to be placed in a non-active matrix of iron.

For SLB we have adopted the US class C waste criteria (regulatory guide 10CFR61) using as specific activity limits (SAL's in Ci/m^3) those calculated by Fetter et al [12]. The WDR is defined, as the sum of the ratios between the specific activity of all radionuclides and the corresponding SAL's, and the acceptance rule for SLB is WDR$\leq$ 1. The concentration limit for SLB, i.e., that for which WDR=1, is computed here in wt fraction by assuming the elements to be present in a non-active matrix of a materials with density that of iron (7.87 g cm^{-3}). Limits (in wt fraction) on elements placed in a matrix of different density, $D_{ma,}$ can be obtained by multiplying the limits computed in this paper by the factor 7.87/ D_{ma}. The SLB-concentration limits are calculated for shutdown after operation time.

The uncertainty analysis has been performed using the ACAB code, which uses a method based on a simultaneous random sampling of all the cross sections probability density functions (PDF) involved in a problem. The PDF for each cross section is assumed to be lognormal. This means that $\log(\sigma/\sigma_0)^9$ follows a normal distribution $N(0,\Delta)$ with σ_0 being the best-estimate cross section value contained in the EAF_XS-2003 cross section library and $\Delta = \Delta_{LIM}{}^2/9$ being $\Delta_{LIM}{}^2$the variance included in the EAF_UN-2003 library [8]. The uncertainties values included in the EAF_UN-2003 library $\Delta_{LIM}{}^2$ are defined as tree times the experimental standard deviation, that is $\Delta\sigma_{LIM} = 3\Delta\sigma_{expt}$ (to represent a 99.73% confidence limit). All results presented in this paper have been obtained with a 1000 histories sample size, which was found[6] appropriate for our applications.

III. UNCERTAINTY ANALYSIS: CONCENTRATION LIMITS FOR SLB AND RECYCLING.

Concentration limits without considering uncertainties (using the best estimate cross section) (CL_{nom}) and the percentile 95 of the probabilities distributions of those CL (CL95) for all naturals elements with atomic number between Z=1 to Z=83 have been calculated in an extensive work[13], for two scenarios. Here we present the most significant results referred to major constituents and potential impurities of LA structural materials.

Let the CL95 for SLB be the value defined as the concentration for which the WDR$\leq$ 1 with a probability of 0.95, and let the CL95 for HoR and RR be the concentrations for which the contact dose rate is kept below 10 _Sv/h at 100 yr cooling and 10 mSv/h within 50 yr cooling respectively with a probability of 0.95. Them, the relative error of the CL at a 95% confidence level (E_{95}) is defined as: E_{95}= (CL95-CL_{nom})/ CL_{nom}

Table 1 gives the E_{95} of CL for all natural elements from H to Bi under the neutron environment of the HYLIFE-II vessel structure. A significant error in the prediction of the activation of several elements is shown.

The E95 values under the neutron environment of the Sombrero have been calculated[13]. In this case, most of the E95 values are found it below 50 %. Only "P" for SLB and "Cl, P, Ge, O, Ga, S" for RR and HoR have errors higher than 50%.

Table 1.
E_{95} of the concentrations limit corresponding to SLB, HoR and RR, using EAF–2003.

E_{95}	SLB	RR	HOR
100- 90%	Br, Kr, Lu		
90- 80%	Sm, Gd	Lu	Sn, Lu
80- 70%	Tl, Dy, Ce, Au, Tb, Ag, Er	Ag, Ge	Sb, Ag
70- 60%	Pt, Yb, In, Bi, Ho Tm	P, As, In, Hf, Te	P, Ge, In, Te, As
60- 50%	Hg, Cd, Hf, Rh, Ta, Se, W, Os, Ir, La, Re	Ho, O, Tm, Sb, Yb, S	Ho, O, Tm, S, Yb, Pt, Hf, Ga
50- 40%	Sn, Pd, Pb, Ga, Ge, As, Mn, Sr, Nd, Cr, Rb, Cs	Ga, Dy, Er, Cd, Cl, Tb, Pt, Ta, Re, Ir, W, Os, Pd, Se, Cr, Cs, Ce	Cd, Dy, Ir, Cl, Er, Tb, Ta, Re, Os,Cs, W, Pd, Se, Cr, Au
40- 30%	C, Ar, Co, Sc, F, Fe, Nb, V, Sb, In, Zn, Y, Ti	Rh, Mn, Ru, Zr Au, Co, F Mn Xe Mo La	Ce, Rh, Mn, Ru, Zr, Y, In, Co, Xe, F, Mo La
30- 20%	Te, Xe, Cu, K, P, Na, Ne, Zr	Zn, Tl, Ti, Ar, V, Br, Sc, Nb, Hg, Pb, Nd, Ba, Sn, Bi, Fe, Rb, Y, Ne	Tl, Zn, Nd, Ti, Ar, V, Sc, Nb, Hg, Pb, Br, Bi, Fe, Rb, Ne, Sr
20- 10%	Ca, S, Ru, Mg, Be, O, Al, Si, Mo Ni, Ba, Cl	Ni, Kr, Na, Mg, Ca, Si, Cu, K, Sr, Al	Ni, Kr, Na, Mg, Ba, Si, Cu, K, Al, Ca
10-0%	N		

It is worthwhile to emphasize that the uncertainties in the activation calculations depend of the neutron energy range used. With average neutron energy of 0.426 MeV we obtain bigger uncertainties than in the case of average neutron energy of 3.067 MeV.

III.A. SLB

Table 2 gives, for HYLIFE-II neutron environment, some elements limited for the SLB criterion. They are listed in decreasing order of relative error 95 (E_{95})

It can be seen that potential impurity elements (Nb, Tb, Gd, Dy) with a very restricted CL value, present a large error: Gd (0.87), Dy (0.79), Tb (0.74), Nb (0.35). Therefore if we consider uncertainties in our CL calculations we obtain more restrictive limitations in the use of these elements in the LA structural materials. Elements such as C, Cr, V, which are major constituents of some proposed LA materials show an excellent behavior with and without consider uncertainties.

The effect of use different libraries in activation calculations without consider uncertainties, has been performed in previous work[1]. Here we present a comparison between uncertainties results based on different cross section libraries, EAF-2003 (library of reference) and FENDL-UN/A-2.0

Table 2 shows that there are important differences in the CL values for each library. It is very significant in the CL values of Tb, Cr, Co, Fe, and Mn. Nevertheless if we compare their respective E_{95} values, we can observe that the state of the uncertainties for these elements is similar in both libraries.

We can also observe in Table 2 that elements with identical CL values present differences in CL95. Therefore there are still cross sections data libraries with a significant uncertainty in theirs cross sections due to significant lack of experimental data.

To analyze this effect, we have defined the relative difference value (R) as: $R = (CL95 - CL95_{ref}) / CL95_{ref}$
Important differences are shown on elements such as Gd (R= -0.99), Al (R=0.45) and N (R=0,16). When relative differences are positive, the cross section uncertainties are lower in the EAF-2003 library than in FENDL Negative values indicate smaller uncertainties in FENDL This analysis suggest that an improvement of some cross sections is possible an advisable.

III.B. Recycling

Table 3 shows the E95 values corresponding to RR and HoR for some elements (listed in E^{95} decreasing order).

Potential impurities with a significant error for SLB, have also a significant error for RR and HoR: Dy (0.48), Tb (0.45), Ta (0.49), Nb (0.35).

These results suggest the need to study the effect of cross section improvements on the activations of these elements. The method proposed in this paper is used to priorize the cross section requiring more accuracy.

Table.2 CL and CL95 for SLB (wt fraction) and dominants nuclides (D.N)

		EAF-2003			FENDL		
EL	D.N	CL_{ref}	$CL95_{ref}$	E_{95}	CL	CL95	E_{95}
Gd	Ho166m	1.55E-6	1.34E-7	8.70E-1	1.50E-6	3.17E-7	7.90E-1
Dy	Ho166m	1.77E-5	3.63E-6	7.90E-1	2.27E-5	1.24E-6	4.50E-1
Tb	Ho166m	9.17e-5	2.35e-6	7.40E-1	1.21E-6	7.73E-6	3.60E-1
Bi	Bi208	7.63E-5	2.73E-5	6.42E-1	7.64E-5	2.78E-5	6.35E-1
Hf	Hf182 / Ir192s	3.23E-3	1.43E-3	5.57E-1	2.72E-3	1.44E-3	4.71E-1
Ta	Ir192s	2.78E-3	1.24E-3	5.53E-1	1.90E-3	9.42E-4	5.05E-1
W	Ir192s	3.28E-3	1.53E-3	5.33E-1	2.24E-3	1.24E-3	4.44E-1
Mn	No Limit	3.39E+2	1.89E+2	4.42E-1	2.48E+3	1.71E+3	3.08E-1
Cr	No Limit	1.16E+7	6.57E+6	4.34E-1	6.22E+7	4.15E+7	3.32E-1
C	No Limit	1.65E+1	1.01E+1	3.88E-1	1.65E+1	1.55E+1	0.57E-1
Co	Fe60 / Co60	1.71E-2	1.07E-0	3.71E-1	7.06E-2	5.08E-2	2.80E-1
Fe	No Limit	7.55E+0	4.83E+0	3.60E-1	6.58E+1	4.46E+1	3.21E-1
Nb	Nb94	8.33E-7	5.47E-7	3.50E-1	8.43E-7	5.99E-7	2.80E-1
V	No Limit	9.28E+7	6.16E+7	3.36E-1	9.80E+7	6.40E+7	3.46E-1
Ti	No Limit	9.88E+3	6.88E+3	3.03E-1	8.83E+3	6.26E+3	2.90E-1
Al	Al-26	1.58E-2	1.33E-2	1.60E-1	1.11E-2	7.38E-3	3.34E-1
Si	No Limit	5.28E+1	4.45E+1	1.57E-1	3.80E+1	2.38E+1	3.72E-1
Mo	Tc-99	9.14E-6	7.82E-6	1.44E-1	1.16E-5	1.08E-5	0.67E-1
Ni	Ni59 / Ni63	2.37E-1	2.03E-1	1.41E-1	2.97E-1	2.47E-1	1.68E-1
Cl	Cl36	2.76E-4	2.41E-4	1.25E-1	2.69E-4	2.57E-4	0.46E-1
N	C14	9.80E-4	8.72E-4	1.09E-1	9.80E-4	7.31E-4	2.53E-1

Table. 3. E_{95} for RR and HoR and Dominants Radionuclides

EL	D.N	E95 (RR)	E95 (HoR)
Hf	Ta182 / Ir192	6.18E-1	5.24E-1
Dy	Ho166m	4.82E-1	4.82E-1
Cl	No Limit	4.65E-1	4.75E-1
Tb	Ho166m	4.57E-1	4.57E-1
Ta	Ir192	4.49E-1	4.49E-1
W	Ir192	4.30E-1	4.30E-1
Cr	No Limit	4.18E-1	4.18E-1
Mn	Co60	3.82E-1	3.82E-1
Co	Co60	3.56E-1	3.54E-1
Mo	Nb91 / Nb94	3.38E-1	3.38E-1
Ti	No Limit	2.86E-1	2.85E-1
V	No Limit	2.78E-1	2.77E-1
Nb	Nb94	2.73E-1	2.73E-1
Bi	Bi-207	2.42E-1	2.42E-1
Fe	Co60	2.41E-1	2.41E-1
Ni	Co60	1.98E-1	1.98E-1
Si	No Limit	1.53E-1	1.53E-1
Al	Al26	1.24E-1	1.24E-1

IV. IMPROVEMENTS IN SLB ASSESSMENT WHEN REDUCING SELECTED CROSS-SECTION UNCERTAINTIES.

In previous section, we have demonstrated that an increase of the accuracy on the activation results for some elements is needed. Here, we show (see Table 4) the usefulness of our methodology to rank the cross-section requiring a better knowledge for the HYLIFE neutron environment.

The elements included in Table 4, are of interest in selecting structural materials, from different considerations: potential impurities (Nb, Ag Cd, Gd, Tb, Dy, Bi), elements for LA alloying (W, Ta) and constituents in conventional steels (Mo, Nb).

For each of them, the SLB-dominant nuclides and the reactions with the cross-sections introducing the largest uncertainties in their production are shown.

We have defined as a reasonable cross-section improvement when the standard deviations of cross-section data are below ±5%. This limit is named *acceptable limit*. Then, for each cross-section, σ_j of table 4, the corresponding $CL95_j$ is calculated, assuming that the standard deviation of this cross section is set to the ±5% *acceptable limit* when the uncertainty is higher, and leaving its original value when is smaller. The rest of the cross section uncertainties are leaving unchanged.

The $RE95_j$ index allows us to evaluate the reduction in the WDR uncertainty (CL95 index) when the cross section uncertainty is reduced to the *acceptable li*mits. For each element, the cross-section σ_j inducing the highest improvement when reducing its uncertainty to *acceptable limit* leads to the highest $CL95_j$.

Table 4 shows that uncertainties in some of the cross sections of interest are important. These $RE95_j$ indexes suggest that some cross-sections need further improvement with important effect in reducing activation uncertainties. Values of $RE95_j$ as high as 1.66 have been observed.

V. CONCLUSIONS

Significant errors in the long-lived activity of some elements under the neutron environment of HYLIFE-II have been obtained when considering the effect of the cross section uncertainties.

In performing this analysis the latest activation cross section library (EAF-2003) has been used as library of reference. The results for individual elements have been used to compute the quality of the EAF-2003 for IFE applications.

Table. 4 Uncertainty information for cross section that contribute most to the uncertainty in the SLB of some elements.

Element	Reactions			σ_j	Δ	$CL95_j$	$RE95_j$
Nb → ^{94}Nb	^{93}Nb	(n,γ)	^{94m}Nb	6.96E-01	0.900	5.56E-07	0.03
	^{94}Nb	(n,γ)	^{95}Nb	2.49E+00	0.632	7.33E-07	0.35
Ag → ^{108m}Ag	^{107}Ag	(n,γ)	^{108}Ag	4.04E+00	0.750	1.10E-03	0.98
	^{109}Ag	(n,2n)	^{108m}Ag	7.20E-03	0.200	5.61E-04	0.02
	^{108m}Ag	(nγ)	^{109}Ag	7.24E+00	0.850	6.03E-04	0.09
	^{108m}Ag	(nγ)	^{109m}Ag	6.96E+00	0.850	5.96E-04	0.08
Cd → ^{108m}Ag	^{107}Ag	(n,γ)	^{108}Ag	4.04E+00	0.750	6.78E-03	0.25
	^{107}Ag	(n,γ)	^{108m}Ag	4.32E-02	0.742	5.85E-03	0.08
	^{106}Cd	(nγ)	^{107}Cd	1.48E+00	0.541	5.63E-03	0.04
Gd → ^{166m}Ho	^{165}Ho	(n,γ)	^{166}Ho	1.78E+01	0.279	1.33E-07	0.00
	^{165}Ho	(n,γ)	^{166m}Ho	9.54E-01	0.527	1.38E-07	0.03
Dy → ^{166m}Ho	^{164}Dy	(n,γ)	^{165m}Dy	1.53E+00	1.274	8.06E-06	1.22
	^{165}Ho	(n,γ)	^{166}Ho	1.78E+01	0.279	3.62E-06	0.00
	^{165}Ho	(n,γ)	^{166m}Ho	9.54E-01	0.527	3.53E-06	-0.03
Ta → ^{192n}Ir	^{182}W	(n,γ)	^{183}W	6.02E+00	0.732	1.24E-03	0.00
	^{183}W	(n,γ)	^{184}W	1.16E+01	0.393	1.24E-03	0.00
	^{184}W	(n,γ)	^{185}W	1.61E+00	0.906	1.28E-03	0.03
	^{190}Os	(n,γ)	^{191}Os	5.10E-01	0.808	1.24E-03	0.00
	^{192}Ir	(n,n')	^{192n}Ir	1.85E-02	1.000	1.24E-03	0.00
	^{192n}Ir	(n,γ)	^{193}Ir	4.44E+01	1.140	1.64E-03	0.32
W → ^{192n}Ir	^{184}W	(n,γ)	^{185}W	1.61E+00	0.906	1.56E-03	0.02
	^{190}Os	(n,γ)	^{191}Os	5.10E-01	0.808	1.53E-03	0.00
	^{191}Ir	(n,γ)	^{192n}Ir	3.28E-03	0.611	1.52E-03	-0.01
	^{192}Ir	(n,n')	^{192n}Ir	1.85E-02	1.000	1.51E-03	-0.01
	^{192n}Ir	(n,γ)	^{193}Ir	4.44E+01	1.140	2.17E-03	0.42
Bi → ^{208}Bi	^{209}Bi	(n,2n)	^{208}Bi	2.67E-02	0.100	2.73E-05	0.00
	^{209}Bi	(n,2n)	^{208m}Bi	4.64E-03	0.300	2.74E-05	0.00
	^{208}Bi	(n,γ)	^{209}Bi	4.20E-01	1.013	7.26E-05	1.66

Note: σ_j is the average cross section, Δ is the corresponding standard deviation. $CL95_j$ (in wt) is the CL95 index calculated when reducing the standard deviation of the σ_j to the *acceptable limit*. $RE95_j = (CL95_j - CL95)/CL95$.

Elements considering as potential impurities of some proposed LA materials, Dy, Tb, Ta, Nb present a significant error in theirs CL's for hands-on recycling, remote recycling and sallow land burial due to high uncertainty in some critical cross sections.

The improvement of CL's corresponding to SLB when reducing the uncertainty of these critical cross sections has been demonstrated.

The results show the needed of new cross sections measurements or evaluations.

Large differences are found when comparing EAF-2003 vs FENDL results.

The errors estimated in the SOMBRERO case using EAF-2003 library are less important. This indicates that the significance of the uncertainties cross sections depend of the energy range used. Neutron environment with average neutron energy of 0.426 MeV leads to bigger errors than those obtain with 3.067 MeV.

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