Transmutation analysis of realistic low-activation steels for magnetic fusion reactors and IFMIF

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Abstract

A comprehensive transmutation study has been performed for steels considered in the selection of structural materials for magnetic and inertial fusion reactors for the international fusion materials irradiation facility (IFMIF) neutron irradiation environment, as well as in the international thermonuclear experimental reactor (ITER) and a fusion demonstration reactor (DEMO) for comparison purposes. An element by element transmutation approach is used in the study, addressing the generation of: (1) H and He and (2) solid transmutants. The IEAF 2001 activation library and the activation code ACAB were applied in the IFMIF transmutation analysis, after proving the applicability of ACAB for transmutation calculations in intermediate energy systems.

1. Introduction

The reduced activation (RA) materials based on modified ferritic/martensitic steels (Fe Cr W V Ta), V Cr Ti alloys and SiC composites are under development by the international fusion materials community [1]. However, there is a global consensus that the qualification of materials in a suitable test environment is inevitable for design, construction and safe operation of DEMO fusion reactors. In this sense, an appropriate fusion materials irradiation facility has been proposed: the international fusion materials irradiation facility (IFMIF). In exploring whether fusion-relevant irradiation damage conditions can be realised in IFMIF, evaluation of both displacement damage and solid/gaseous transmutants is necessary.

Our concern here is to perform a comprehensive transmutation study of some low-activation steels for irradiation simulations in the high flux test module (HFTM) of the IFMIF neutron source. Also, we intend to show that an updated version

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of the ACAB activation code is able to deal properly with this problem using the IEAF-2001 activation library. Calculations for the first wall of international thermonuclear experimental reactor (ITER) and DEMO magnetic fusion reactor are also accomplished and results are compared with those of the IFMIF simulation facility.

In earlier work, different inventory computational codes, previously developed for fusion applications, were shown to be reliable for IFMIF applications with appropriate modifications [2,3]. Following the same trend, this capability has been incorporated into the ACAB code [4]. Applications to transmutation of different steels and some particular elements have been performed [3]. Here, the analysis is extended to all the potential elements expected to be present in steels, and the study is carried out by means of an element-by-element evaluation. This approach allows us to apply the transmutation results for elements in a straightforward way to study transmutation of any material consisting of some of the considered elements, as well as to analyse the transmutation performance of different materials.

In this transmutation study, we have selected some proposed candidate steels for the first structural wall [5]: (i) several existing 300 series stainless steels (SS304, ITER316 and primary candidate alloy PCA) and (ii) some RA ferritic steels (F82H-IEA and EUROFER 97, and the ODS low-activation ferritic steel LAF-3) intended for use in magnetic and inertial fusion systems. The concentration of alloying and impurity elements can also be found in the literature [5]. For the element-by-element study, we have classified the constituent elements into three groups: (i) typical elements intended for reduced activation ferritic steels (RAFS) as well as most of the ones intended for SS (B, C, N, O, Si, P, S, Ti, V, Cr, Mn, Fe, Y, Ta, and W), (ii) some of the impurity elements in RAFS as well as some of the elements intended for SS and Cr Mo steels (Al, Co, Ni, Cu, Nb, and Mo) and, (iii) impurity elements common to all the steels (Zn, As, Se, Zr, Ru, Rh, Pd, Ag, Cd, Sn, Sb, Te, La, Ce, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Os, Ir, Pt, Au, Pb, and Bi).

2. Computational methodology and validation process

Transmutation calculations have been performed for fusion neutron energy spectra using the ACAB radionuclide generation/depletion code [4]. The nuclear data used are those from the European Activation File EAF-2003 (EAF XS and EAF DECAY) [6].

For IFMIF, transmutation calculations have been conducted with an updated version of the ACAB code that can handle the numerous reaction channels for neutron energies over 20 MeV. The updated ACAB code does not rely on a table of fixed reaction types, requiring only a list of resultant isotopes and cross-sections for the production of each. In IFMIF, the neutron source spectrum is extended up to ~55 MeV, with ~20% of the neutrons having energies above 15 MeV. The activation cross-section library processed for IFMIF is the Intermediate Energy Activation File IEAF-2001 [7]. This library contains the neutron activation cross-section files for 679 nuclides, including stable and isomeric states up to 150 MeV. We have used the group-wise IEAF-2001 library in GENDF format with 256 neutron energy group structure. To be consistent with IEAF-2001, we have processed a complete decay library based on JEFF3.1 Decay Library (3849 radionuclides) with an extra number of 135 radionuclides from other decay sources.

In order to validate our updated ACAB code for IFMIF calculations, we have tested our predictions with a set of integral activation experiments [8] having a neutron spectrum very close to IFMIF. In Table 1, we present the comparison of activation

Table 1
Calculation to experiment ratios for the activity inventories induced in the SS 316 intermediate energy activation experiment [8]

Isotope	Decay time (h)	C/E	Isotope	Decay time (h)	C/E
Sc 46	49.68	2.03	Ni 56	49.68	1.60
Sc 48	49.68	1.56	Ni 57	49.68	1.30
V 48	49.68	2.64	Y 87m	49.68	0.40
Cr 48	49.68	4.45	Y 87g	49.68	0.48
Cr 49	1.66	0.82	Y 88	3600	1.00
Cr 51	49.68	1.02	Zr 86	49.68	0.03
Mn 52g	49.68	3.32	Zr 88	700	1.00
Mn 54	49.68	1.09	Zr 97	49.68	0.02
Mn 56	25.93	1.22	Nb 90g	49.68	0.45
Fe 52	25.93	1.22	Nb 92m	49.68	1.45
Fe 59	49.68	1.10	Nb 95g	49.68	1.58
Co 55	49.68	2.57	Nb 95m	49.68	1.18
Co 56	49.68	2.81	Nb 96	49.68	1.83
Co 57	49.68	1.03	Mo 90	49.68	1.48
Co 58g	49.68	1.19	Mo 93m	49.68	3.48
Co 60	49.68	1.21	Mo 99	49.68	1.26
Co 61	1.66	2.26	Tc 99m	49.68	1.26

predictions for a sample of SS316. This steel has been irradiated for 7525 s with a neutron flux of 4.10×10^{11} n/cm^2 s. We have adopted the average neutron flux in a 45-group energy structure at the centre position of the irradiated steel sample reported in Ref. [8]. The calculation-to-experiment ratios (C/E) obtained by ACAB/G-IEAF-2001 are shown in Table 1. For the radionuclides most important to the total activity and contact dose rate (^{56}Mn, ^{54}Mn, ^{57}Ni, ^{58}Co and ^{60}Co), the predictions have demonstrated a reasonable agreement: C/E ratios are between 0.7/1.3. For the rest of the isotopes, larger deviations of C/E from unity were found: C/E ratios are between 0.02/4.45. These discrepancies can arise from: (i) the activation cross-section library, (ii) the fact of neglecting the sequential charged-particle reactions, and (iii) the uncertainties of the sample's initial composition.

Our results are in general in good agreement with those obtained by ALARA and FISPACT codes [2,3]. However, discrepancies up to 30% have been found for a few isotopes (^{52}Fe, ^{87m}Y). Both the different decay data libraries and the adopted neutron spectrum can explain these differences.

3. Transmutation element-by-element: gaseous and solid transmutants

An extensive evaluation for three different neutron environments has been performed to assess the importance of the element-by-element transmutation in fusion materials. We have considered

Table 2
H and He production (appm) from the major minor steel constituents and some impurities after one year of irradiation time in the HFTM/IFMIF, DEMO and ITER

	Element	HFTM/IFMIF		DEMO		ITER	
		H	He	H	He	H	He
Typical intended elements in RAFS as well as most of the intended ones in SS	Cr	1169	255	1312	230	552	97
	C	507	4289	2	7230	1	3105
	Mn	819	201	569	175	239	74
	P	2037	840	1828	1004	785	440
	S	5159	2418	5328	2344	2275	973
	V	699	285	744	106	324	44
	B	1085	1905	852	53057	343	13613
	N	1365	2221	1751	1605	660	655
	W	240	31	26	6	10	3
	Ta	241	18	33	3	13	1
	Si	2424	1395	2387	1627	1027	712
	Ti	865	316	849	224	360	94
	Fe	1551	301	1602	312	679	133
	Y	838	62	863	42	373	17
	O	794	1642	410	1435	174	640
Some of the impurity elements in RAFS as well as some of the intended elements in SS and Cr Mo steels	Cu	1886	321	1947	314	851	134
	Ni	5699	999	6067	752	2477	322
	Mo	1089	190	1196	90	504	38
	Nb	674	117	364	74	153	31
	Al	1733	732	2602	927	1098	400
	Co	1297	273	2309	237	803	101
Impurity elements common to all the steels	As	721	168	473	90	203	37
	Sn	430	62	56	13	23	5
	Zr	650	64	476	57	200	24
	Sb	331	26	72	15	30	6
	Cd	375	54	146	21	61	9
	Bi	229	40	10	145	4	36
	Zn	2575	506	3097	564	1326	247
	Se	367	84	195	45	83	19
	Ag	776	127	463	60	195	25
	Tb	237	31	37	14	15	6
	Pb	199	44	7	4	3	2

Calculations performed with ACAB/IEAF 2001.

the IFMIF environment (5.86×10^{14} n/cm^2 s, $\langle E \rangle \sim$ 6.95 MeV and wall loading 6.4 MW/m^2) in comparison with DEMO (13.0×10^{14} n/cm^2 s, $\langle E \rangle \sim$ 3.13 MeV and wall loading 3.5 MW/m^2) and ITER (3.90×10^{14} n/cm^2 s, $\langle E \rangle \sim 3.92$ MeV and wall loading 1.2 MW/m^2) [2,3].

Firstly, the H and He gas production is analysed. H and He are generated by nuclear reactions (n,xH) or (n,xHe) on all nuclei. Their generation depends on both the neutron flux level and the neutron spectrum. In Table 2, we present the H and He production values for the initial major minor steel constituents and some impurities. For most of the important ones, we have demonstrated that the generation of gaseous transmutants increases linearly as a function of irradiation time.

Secondly, the evolution of solid transmutants is investigated. In Table 3, we illustrate the most important element-by-element transmutation processes in IFMIF after one year of irradiation time: the depletion of the initial Z element and the generation of new elements are given. Regarding major steel constituents, it is seen that the transmutation of W will increase the level of Ta and Re, with approximately the same ratio. The Cr will be transmuted mainly into V, and the Mn into Cr. The Ni will be transmuted into Co by (n,xH) reactions and into Fe by (n,xHe) reactions, 37% and 60%, respectively, and Mo will be transmuted into Nb (52%), Zr and Ru. We have extended such element-by-element calculations to different irradiation times, and we observed that the evolution for most of the initial elements increase/decrease nonlinearly with irradiation time.

4. Transmutation in steels: application to EUROFER

The element-by-element transmutation results presented in Tables 2 and 3 can be used to predict the transmutation performance of any material irradiated in IFMIF, DEMO and ITER. Fig. 1 shows the total production of H and He for the selected steels under the different scenarios after one year of irradiation. These gaseous transmutant concentrations can be comparable to some initial steel constituents. The higher levels of H and He production are in PCA steel as a consequence of Ni concentration by (n,xH) and (n,xHe) reactions. Regarding IFMIF, the contribution for H generation is due to Cr (8.3%), Fe (47.5%) and Ni (41.5%). For He, the main generation is also due to Cr (9.2%), Fe

Table 3
Transmutants (appm) of the initial steel constituents after 1 year of irradiation time in HFTM/IFMIF

Z	Initial element	Transmutants (appm)				
		$Z-2$	$Z-1$	Z	$Z+1$	$Z+2$
5	B	1326	458	1885	0	0
6	C	482	347	1695	0	0
7	N	1780	1422	3372	0	0
8	O	1333	684	2019	0	0
13	Al	262	1758	2016	8	0
14	Si	1349	1095	2425	0	0
15	P	202	1677	1903	14	0
16	S	2490	2471	4952	0	0
22	Ti	264	316	556	0	0
23	V	4	694	761	31	0
24	Cr	293	1995	2331	0	0
25	Mn	81	1657	1825	44	0
26	Fe	430	1153	1669	0	0
27	Co	145	3255	3425	5	0
28	Ni	3802	2339	6238	1	0
29	Cu	255	4232	4959	440	0
30	Zn	1341	2034	3420	30	0
33	As	100	3299	5101	1689	0
34	Se	112	289	839	416	6
39	Y	35	3288	3543	121	0
40	Zr	223	2027	2409	23	129
41	Nb	83	2280	2515	1	0
42	Mo	341	1009	1912	516	18
44	Ru	431	484	1964	955	72
45	Rh	200	3498	5109	1312	0
46	Pd	150	834	2227	1058	31
47	Ag	52	3776	8938	2219	0
48	Cd	106	351	1251	665	63
50	Sn	102	95	651	410	2
51	Sb	1	4437	7481	3019	0
52	Te	47	160	3454	3215	28
57	La	40	578	969	84	0
58	Ce	97	3215	4048	705	21
62	Sm	137	309	2372	1867	13
63	Eu	17	1782	3735	1922	0
64	Gd	20	306	2089	1496	206
65	Tb	17	1013	2454	1438	0
66	Dy	25	433	836	340	1
67	Ho	4	3672	8545	4823	0
68	Er	81	3092	4555	1210	9
69	Tm	94	7420	8388	859	0
70	Yb	57	646	2299	1273	31
71	Lu	14	2080	2934	841	0
72	Hf	33	712	1630	123	0
73	Ta	4	4451	5709	1890	0
74	W	108	1611	3205	1430	1
76	Os	48	206	3285	2674	5
77	Ir	6	3593	9065	3738	0
78	Pt	161	339	1296	566	8
79	Au	27	9138	10663	1459	0
82	Pb	41	442	585	8	0
83	Bi	15	953	1006	15	0

Calculations performed with ACAB/IEAF 2001.

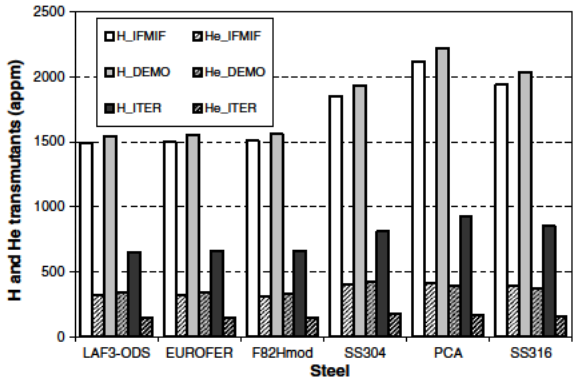

Fig. 1. H and He (appm) production after one year of irradiation time in HFTM/IFMIF, DEMO and ITER for different steels. Calculations performed with ACAB/IEAF 2001.

(46.7%) and Ni (36.8%). For the rest of the steels, iron produces the most important levels of H and He transmutants (~92% and ~84%, respectively for EUROFER). Similar contributions are predicted in DEMO and ITER. The hard neutron spectrum in IFMIF and the high neutron level in DEMO lead to higher H and He transmutation rates in comparison with ITER. We have also calculated the generation of H and He for DEMO and ITER with IEAF-2001 and EAF-2003 libraries. IEAF-2001 always over predicts the total generation of H and He, ~10% and ~3%, respectively, for EUROFER.

Table 4 shows the main transmutants for EUROFER in IFMIF, ITER and DEMO for different irradiation times. The most important transmutation effects are produced in IFMIF. The majority of steel constituents will not change under irradiation (Fe, Cr, C, ...) while some major and minor intended elements will (B +7.3%, Ti +26%, V +10%, Mn +25%, Ta +2%, W −0.3%). New impurities generated are also important (Re 4.7 appm, Hf 1.3 appm, Be 2.3 appm, Mg 1.7 appm). In DEMO and ITER, we observed significant generation of new elements such as Re and Os from W, and the depletion of B.

Table 4
Transmutation (appm) of EUROFER constituents under different irradiation times 1, 3 and 5 full power years (fpy) in the HFTM/IFMIF and one year in DEMO and ITER

EUROFER initial composition		IFMIF (appm)			DEMO (appm)	ITER (appm)
Element	(appm)	1 fpy	3 fpy	5 fpy	1 fpy	1 fpy
H		1500	4493	7477	1556	659
He		319	961	1609	342	147
Li		0.3	0.9	1.5	2.9	0.8
Be		2.3	7.1	11.8	3.0	1.3
B	51	3.7	11.1	18.4	1.5	0.2
C	4860	6.1	18.3	30.4	9.5	4.3
N	1191	3.8	11.3	18.8	3.7	1.4
O	347	0.70	2.11	3.50	0.53	0.24
Mg		1.7	5.1	8.4	2.2	0.9
Al	206	0.69	2.04	3.37	0.003	0.01
Si	990	2.0	6.2	10.3	1.9	0.8
P	90	0.04	0.10	0.16	0.14	0.05
S	87	0.43	1.26	2.08	0.48	0.20
i	116	30	103	183	29	12
V	2183	221	687	1149	235	94
Cr	96236	164	737	1420	73	35
Mn	4048	1014	3582	6738	921	385
Fe	885880	1478	5174	9526	1282	568
Co	47	0.03	0.10	0.28	2.28	0.51
Ni	47	0.11	0.33	0.53	0.20	0.03
Cu	44	0.22	0.65	1.07	0.49	0.16
Nb	6	0.01	0.04	0.06	0.01	0.005
Mo	29	0.06	0.16	0.27	0.13	0.04
Hf		1.3	4.8	9.0	1.3	0.6
Ta	215	4	17	30	17	3
W	3327	10	39	68	129	33
Re		5	17	29	132	34
Os		0.005	0.060	0.165	13.7	0.8

Calculations performed with ACAB/IEAF 2001.

5. Conclusions

The updated version of ACAB code is able to deal with transmutation calculations in the IFMIF facility using the IEAF-2001 intermediate energy library. An integral experimental activation benchmark has been used to validate our system. The calculated results are in quantitative agreement with the experimental data similar to that obtained with other computational approaches already validated for IFMIF applications.

The element-by-element analysis has been demonstrated as a helpful tool to easily analyse the transmutation performance of irradiated fusion materials. The contribution of each source element to the generation of any transmutant product is obtained in a straightforward way.

IEAF-2001 over predicts the generation of gaseous transmutants, H and He, for ITER and DEMO compared to EAF-2003, and they have shown a linear behaviour with the irradiation time.

To complete this study, the impact of the activation cross-section uncertainties on the IFMIF transmutation calculations will be estimated. The sensitivity/uncertainty Monte Carlo methodologies implemented in ACAB will make use of the recent EAF-2005 uncertainty library to perform this job. In addition, a more complete benchmarked calculation of ACAB against the already validated computational methodologies used for activation and transmutation calculations of IFMIF will be performed.

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References

[1] E.E. Bloom, S.J. Zinkle, F.W. Wiffen, J. Nucl. Mater. 329 333 (2004) 12.
[2] U. Fischer, S. Simakov, U.v. Mollendorff, P. Pereslavtsev, P. Wilson, Fusion Eng. Des. 69 (2003) 485.
[3] U. Fischer, S.P. Simakov, P.P.H. Wilson, J. Nucl. Mater. 329 333 (2004) 228.
[4] J. Sanz, ACAB Activation Code for Fusion Applications: User's Manual V5.0, Lawrence Livermore National Labora tory UCRL MA 143238, February, 2000.
[5] J. Sanz, O. Cabellos, S. Reyes, Fusion Eng. Des. 75 79 (2005) 1157.
[6] R.A. Forrest, J. Kopecky, J. Ch. Sublet, The European Activation File: EAF 2003 cross section library, EURATOM/ UKAEA Fusion, UKAEA FUS 486, December, 2002.
[7] U. Fischer, D. Leichtle, U.v. Mollendorff, et al., Intermediate Energy Activation File IEAF 2001, NEA data bank pro gramme library, package NEA 1656/01, 2001.
[8] U. von Moellendorf, F. Maekawa, et al., Forschungszentrum Karlsruhe, Bericht FZKA 6764, October, 2002.