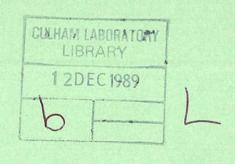
Concentrations of radiologically-important tramp elements in standard-grade stainless steels with particular relevance to activation of the first wall of NET

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Abstract

The dose equivalent rate of activated components determines the optimum time for decommissioning and the procedures adopted for the management of radioactive structural wastes from nuclear In standard-grade stainless steels the elements cobalt, nickel, niobium, molybdenum, silver and possibly some rare earth metals are the only ones likely to contribute appreciably to the  $\gamma$  dose rate at a cooling time around 100y. Up to about 80y after shutdown, 60Co generated from iron, nickel and cobalt dominates the dose rate whilst at longer times 94Nb from niobium and molybdenum and 108 mAg from silver present as an impurity usually become dominant. Activation calculations have been performed for the irradiation regime anticipated for the first wall of the Next European Torus, in order to determine the concentration limits on cobalt, niobium and silver in a type 316L steel such that these potential impurity elements do not appreciably enhance the overall dose rate of the steel. A review of existing information on the impurity contents of stainless steels indicates that it should not be difficult to attain the required limits in a nuclear-grade 316L steel.

\*

## 1. Introduction

The presence of certain tramp elements giving rise to long-lived activation products in structural materials such as stainless steels can have major consequences for dismantling and the management of waste from nuclear reactors. The external  $\gamma$  dose rate from radionuclides generated in the elements Ni, Co, Mo, Nb, Ag, Eu and Sm, for example, may determine the radiation exposure associated with the handling of activated or contaminated components and hence the optimum cooling time required before decommissioning [1]. Of the elements named above, nickel, molybdenum and niobium and, exceptionally, cobalt are employed as alloying additions in stainless steels whilst the others are present usually only as residuals.

Activation of major fission reactor components has not hitherto been regarded as a matter of major concern, since most of the radioactivity is associated with the fuel cycle and it is uncertain to what extent it would be economically justified to decrease the residual radioactivity through the use of reduced-activation structural materials, as has been proposed for future fusion reactors. With a clearer understanding of the operations involved in decommissioning, however, increasing consideration is being given to the possibility of reducing the radiological consequences of reactor decommissioning and the amount of radioactive waste for disposal, through detailed attention to the specification of the elemental constitution of materials used in reactor construction, taking particular care to avoid elements giving rise to long-lived activation products [2,3]. Design studies for the Sizewell B pressurised-water reactor have shown two areas to be especially important in this respect. The first concerns the selection of raw materials for construction of the concrete biological shield, so as to minimise the amount of waste that must be regarded as being radioactive. The second is the specification of some tramp element concentrations in the steel pressure vessel and internal reactor structures, in order to minimise the exposure of personnel during decommissioning.

The activation of fission reactor steels has been studied by various investigators and the compositional analyses reported provide some useful information on the incidence of some of the elements for which few other analytical data are available. When examining information on steels produced some time in the past, however, it should be borne in mind that

steelmaking technology has made rapid advances in the last two decades and that some purity specifications considered unattainable in the past are now routinely achievable. Given sufficient demand it is feasible to produce steels economically to fine compositional limits and thus in future it may be increasingly worthwhile to specify purity limits for nuclear grade steels so as to maximise the radiological benefit relative to the increased material costs. Accompanying the improvements in steelmaking processes, advances in analytical capability have taken place, which now make it possible to measure quickly and cheaply the concentrations of essentially all elements, with limits of detection in the region of 1 ppb [4].

The present paper begins with a brief review of information on the concentrations of tramp element in steels, mostly derived from experience with fission reactor materials, then applies the results of some activation calculations to the formulation of a specification for the allowable tramp element concentrations in the type 316L steel currently proposed for the construction of the Next European Torus (NET).

# 2. Earlier observations on impurities

Investigations on the tramp element concentrations in reactor steels were first made by Stephens and Pohl [5], who drew attention to the dominant contributions from the long-lived radionuclides  $^{94}\mathrm{Nb}$  (t $_{\chi}$  = 20 300y) and <sup>5 9</sup>Ni (t $_{1/2}$  = 75 000y) to the  $\gamma$  dose rates remaining after about 100y, when the contribution from 60Co has subsided. Other potential sources of  $\gamma$ radiation, particularly silver, europium and holmium were identified but Stephens and Pohl also drew considered to be relatively unimportant. attention to the accumulation of 14C as an activation product of nitrogen Although this radionuclide makes negligible in BWR and PWR steels. contribution to the external dose rate it has implications for waste disposal on account of its long half life ( $t_{\chi}$  = 5730y) and high mobility in the biosphere. Their analytical information is rather limited and they give only the results shown in Table 1 for samples of a type 304 stainless steel.

Table 1

Concentrations of radiologically-potent impurity elements in type 304 steel as reported by Stephens and Pohl [5]							
Nb	Мо	N	Eu	Sm	Но		
160 ± 20	0.26%	~400	0.41	< 0.01	<0.01 ppm		

A decommissioning study performed by Smith, Konzek and Kennedy [6] includes an evaluation of the radioactivity inventory of a PWR based on the material composition for type 304 steel given in Table 2. Woollam [7] and Woollam

Table 2

Composition in mass per cent of ASTM 304 steel assumed by Smith, Konzec and Kennedy [6]								
Cr	Ni	Со	Nb	С	Mn	Мо	Si	Fe
17.5	10.1	0.15	0.016	0.08	2.0	0.05	1.0	bal

and Pugh [8] have performed a similar evaluation for a Magnox reactor. In the type of reactor examined by Smith et al the neutron flux is lower for the stainless steel components, which have a total mass of 200t, than for the mild steel components, with a mass of 3000t, and hence the latter make the major contribution to the dose rate. Although these authors measured the concentrations of Co, Ni, Nb, Ag, Eu and Ho in mild steel, their results are not presented here since the residual element concentrations in mild steels and stainless steels are likely to differ significantly.

Boothby and Williams [9] have reported the results of analyses for Co, Nb, Ag and Eu in type 304 reactor-grade steels and have discussed possible ways of reducing the concentrations of these elements. Table 3 summarises their observations on four different batches of 304 steel, designated A-D. Niobium concentrations are expressed only as upper limits on account of spectral interference from chromium. Whereas three of the alloys contained less than 25 ppm Nb, one of them contained 85 ppm. The silver concentration was below 0.1 ppm in most cases but reached 0.8 ppm in one sample. Boothby and Williams estimate that the use of selected, high-purity starting materials, notably sponge iron and high-purity nickel

and chromium, would enable levels of below 50 ppm Co, 5 ppm Nb and 0.5 ppm Ag to be obtained in a type 304 steel.

Table 3

J	Residual element analyses of different heats of reactor grade type 304 steels reported by Boothby and Williams [9].  Concentrations of Co, Nb are in ppm; those of Ag, Eu in ppb.						
Element	Techniques*	Α	В	С	D		
Co, ppm	NAA ICP	140±15 170±10	$320 \pm 30$ $350 \pm 20$	$190 \pm 20$ $190 \pm 10$	$200 \pm 20$ $200 \pm 10$		
Nb, ppm	ICP	<20	<85	<20	<25		
Ag, ppb	RNA AA	90±10 200±100	<10 <100	$800 \pm 80$ $700 \pm 100$	40±5 <100		
Eu, ppb	RNA	$0.3 \pm 0.1$	$0.3 \pm 0.1$	$0.2 \pm 0.1$	< 0.2		

<sup>\*</sup>NAA, neutron activation analysis by direct  $\gamma$ -ray spectrometry

The concentrations of Mo, Ni, Nb, Co, Ag, Eu, Sm and Ho in various reactor steels were measured by Goddard, MacMahon and Peselli as part of a decommissioning study for the Garigliano reactor [10]. The materials investigated are listed in Table 4. Samples 1A-D represent various CrNiNb18/9 steels; sample 2 is vessel basis material; samples 3A and 3B are type 336 steels as used for the Garigliano reactor vessel; samples 4A, 4B and 5 are various types of AGR steel; sample 6 is a reinforcement steel; samples 7A and 7B are type AISI 347 niobium-stabilised steels, and sample 8 is stock 18/8 stainless steel. Results of the analyses are presented in Samples 3A and 3B were subjected to more detailed analyses performed independently by Nucleco and Imperial College, results from which are shown in Table 6. These workers conclude that it should be possible to attain a niobium content below 10 ppm through selection of the steelmaking materials but that special measures may be necessary to reduce the silver content much below 1 ppm.

The records of analyses of several types of stainless steels by the Chemical Analysis Group at Harwell have provided some useful information. The author is indebted to Dr J S Hislop for making these data available. Table 7 shows results for 6 samples of type 321 steel. Some analyses for

RNA, neutron activation analysis with radiochemical separation

ICP, inductively-coupled plasma optical emission spectrometry on acid digest

AA, flameless atomic absorption on solid samples

Table 4

	Steel samples analysed by Goddard, MacMahon and Peselli [10], with concentrations in mass per cent of selected elements taken from specifications								
No.	Steel sample	Mn	Cr	Мо	Ni	Nb	Co		
1A	X10 CrNiNb 18/9	1.61	17.9	-	10.6	0.43	0.11		
1B	X10 CrNiNb 18/9	1.53	18.0	-	10.5	0.62	0.18		
1C	X10 CrNiNb 18/9	1.76	18.6	-	10.6	0.86	0.18		
1D	X10 CrNiNb 18/9	1.53	18.2	0.11	10.1	0.33	0.05		
2	vessel basis	-	0.38	-	0.84	-	0.023		
ЗА	Garigliano PV								
3B	Garigliano PV					AV			
4A	AGR shield	0.70	0.05	0.02	0.10	-	0.011		
4B	AGR fuel tie	< 0.20	15-18	2.5-4.0	42-45	·-	< 0.25		
5	AGR support strut	0.8-1.4	-	-	-	-	< 0.03		
6	reinforcement	0.75	0.01	-	0.06	-	-		
7A	AISI 347	1.41	19.3		9.28	0.61			
7B	AISI 347	1.17	18.0	-	9.12	0.38	-		
8	CrNiNb 18/8	1.60	17.6	0.36	10.6	0.92	-		

Table 5

	Results of analyses reported by Goddard, MacMahon and Peselli for the most important elements for $\gamma$ activation. Except where otherwise indicated the method used is neutron activation analysis.								
No.	Ni, %	Co, %	Nb, ppm*	Ag, ppm+	Eu, ppb	Sm, ppb			
1A	10.3±0.1	0.107±0.007	3535	2.4±0.1	<33	<35			
1B	10.8 ± 0.1	0.184±0.007	4968	1.7±0.1	nm	nm			
1C	11.1 ± 0.1	0.379±0.009	7544	1.1 ± 0.1	<28	<39			
1D	9.38±0.1	0.053±0.008	2819	0.3±0.1	nm	nm			
2	0.76±0.03	0.021 ± 0.001	9.0	0.7±0.1	<41	<31			
ЗА	$0.75 \pm 0.03$	0.0187±0.007	6.0	1.4±0.1	15 ± 24	43±42			
3B	$0.73 \pm 0.03$	0.0178±0.0007	6.0	0.7±0.1	nm	nm			
4A	$0.27 \pm 0.009$	0.0096±0.0008	6.0	1.6±0.1	< 16	<7			
4B	$37.3 \pm 0.3$	0.205 ± 0.017	41.0	$0.7 \pm 0.1$	nm	nm			
5	$0.287 \pm 0.008$	0.0191 ± 0.0016	6.0	$0.9 \pm 0.1$	<12	<11			
6	0.047 ± 0.015	0.0253±0.0042	5.0	$0.3 \pm 0.1$	nm	nm			
7A	8.92±0.11	0.0148±0.007	5306	$0.6 \pm 0.1$	nm	nm			
7B	$8.85 \pm 0.1$	0.134±0.006	3302	$0.5 \pm 0.1$	nm	nm			
8	$10.0 \pm 0.1$	0.159±0.007	7710	4.5 ± 0.1	nm	nm			

nm = not measured

\*measured by ICP, detection limit~5ppm

\*measured by atomic absorption

Table 6

Comparison of analyses by Nucleco and Imperial College of Garigliano reactor vessel steels 3A and 3B. Activation analysis was employed except where otherwise stated and results are expressed in ppm.

Element	S	teel 3A	S	teel 3B
	Nucleco	Imperial College	Nucleco	Imperial College
Ni	7795±390	$7520 \pm 260$	$7853 \pm 380$	$7320 \pm 260$
	175±9	187±7	166±8	178 ± 7
Co	180±36*	nm -	180±36*	nm
	1.1 ± 0.11	$1.33 \pm 0.11$	1.17±0.12	<detection limit<="" td=""></detection>
Ag	1.3±0.26*	1.4±0.1*	0.7±0.14*	0.7±0.1*
Мо	5800 ± 406	5770±160	7200 ± 400	$6970 \pm 170$
	238 ± 47	<183	201 ± 32	<223
Nb	nm	6.0+	nm	6.0+
	nm	<11**	nm	<8**
Eu	< 0.004	$0.043 \pm 42$	< 0.004	nm
Sm	<0.004	nm	< 0.004	0.015±24

nm = not measured

\*measured by atomic absorption

\*measured by ICP

\*\*measured by SIMS

type 347 steel, which contains niobium at about 1% to prevent carbide precipitation at elevated temperatures, are presented in Table 8. Cobalt and niobium contents measured in samples of type 316 steel over a number of years are shown in Table 9, together with the mean and standard deviation of the cobalt concentration over the 14 samples represented.

### 3. More recent analyses

Samples from two casts of 316L type steel have recently been analysed by glow-discharge mass spectrometry as part of the Culham research programme on low-activation materials. The first material comprised a sample of the 316L reference cast prepared in 1981 by Creusot-Loire for the European fusion materials research and development programme, in accordance with the specification CN 13003 I issued by JRC Ispra. The main-element analysis provided by the manufacturer is given in Table 10. The second sample consisted of commercial grade 316L, cast number M6354, produced in 1987 by British Steel Stainless.

Table 7

Impurity element concentrations measured by Chemical Analysis Group, Harwell for various samples of type 321 steel							
	Co ppm	Nb ppm	Ni %	Ag ppb	Eu ppt		
Sample A	260	< 50	9.0	60	200		
Sample B	500	<100	11.6	700	<300		
Sample C	1500	< 100	11.3	-	-		
Sample D	1100	< 40	. 8.6	٠.	•		
Sample E	2300	< 40	8.8	-	-		
Sample F (7 specimens)*	200-1000	<100	8.2-11.6	-	-		
Highest/lowest	12	<b>.</b>	1.4	12	-		
Expected	1000	-	8-13		-:		

<sup>\*</sup>these seven specimens were taken from different components probably made from different batches of steel

Table 8

	ement concentra arwell for various				
,	Co ppm	Nb %	Ni %	Ag ppb	Eu ppt
Sample 1	1300	1.0	11.1	4000	<200
Sample 2	300	0.75	9.7	-	
Sample 3 (3 specimens)*	900-3200	0.7-1.0	9.1-9.7	, -	:-
Highest/lowest	11	1.4	1.2	-	
Expected		0.5-1.0	9-12		•

<sup>\*</sup>these three specimens were taken from different components probably made from different batches of steel.

Table 9

Cobalt and niobium concentrations measured by Chemical Analysis Group, Harwell in different samples of type 316 steel					
Co, %	Nb, %	Date of analysis			
0.32	< 0.005	27-07-85			
0.27	< 0.01	14-08-81			
0.02	< 0.01	14-08-81			
0.05	< 0.01	18-05-82			
0.17	< 0.005	18-05-82			
< 0.01	< 0.01	15-03-84			
< 0.01	< 0.01	11-06-84			
0.17	< 0.01	14-11-84			
< 0.01	0.01	16-12-84			
0.17	-	24-06-85			
0.48	0.01	22-05-80			
0.28	0.01	23-12-80			
0.17	< 0.01	16-09-86			
mean 0.16 std. dev. 0.14	all below 0.01				

Analyses of these materials were performed by Loughborough Consultants Ltd and were specifically aimed at quantification of the elements relevant to long-term  $\gamma$  activation in fusion reactor first wall materials. The results are presented in Table 11. For comparison purposes the table includes values of the concentrations permitted according to the 'hands-on'  $\gamma$  dose rate limit, namely 25  $\mu Svh^{-1}$  at 100y cooling time, calculated for power reactor irradiation conditions, i.e. a neutron wall loading of 5MWm $^{-2}$  for 2.5y, using the data library UKACTl and inventory code FISPACT [11].

Table 10

	Composition of the 316L reference cast quoted by the manufacturer, Creusot-Loire							
Element	Mass percent	Element	Mass percent					
С	0.024	Со	0.17					
Cr	17.44	S	0.001					
Ni	12.38	Р	0.026					
Mn	1.82	Ta	0.01					
Cu	0.20	N	0.06					
Мо	2.3	В	8 ppm					
Si	0.46	Fe	balance					

Table 11

Results of trace element analysis by glow discharge mass spectrometry on two samples of type 316L stainless steel. Concentrations are expressed in ppm by mass except where indicated otherwise.

Element	Element Eurocast 316L		Hands-on limit at 100 years
Al	470	14	184
Co	1600	2400	1.7
Nb	77	160	0.18
Мо	2.8%	2.3%	31
Pd	0.9	0.26	0.72
Ag	2.6	1.3	0.011
Sm	0.019	< 0.006	0.33
Eu	< 0.003	< 0.004	0.074
Gd	0.023	< 0.005	0.39
Tb	0.002	< 0.001	0.021
Dy	0.019	< 0.005	0.54
Но	0.002	< 0.001	0.057
Er	< 0.005	< 0.006	3.1
Yb	< 0.009	< 0.009	0.63
Lu	< 0.002	< 0.002	0.22
Hf	0.041	< 0.004	0.30
lr	0.084	0.081	0.32
Bi	0.13	< 0.006	0.13

It is clear that for both steel samples the rare earth elements are present only at negligible concentrations. The main impurities affecting the long term dose rate are niobium, silver and cobalt, all of which are present at concentrations much higher than the notional hands-on limit. For each of these elements the concentration varies by only a factor 2 between the two samples. It is notable that the European reference material contains a relatively high aluminium content compared with the commercial steel.

# 4. Activation of NET first wall/blanket structures

In the types of stainless steels considered in the preceding section the radiologically-potent rare earth metals Sm, Eu, Gd, Tb, Dy, Ho, Er, Yb and Lu do not appear to be present at concentrations likely to cause

appreciable activation. This finding is likely to be valid for all standard-grade stainless steels, with the possible exception of those in which additions of mixed rare earth metals, e.g. Misch metal, are employed for inclusion shape-modification. The latter possibility could, if necessary, easily be avoided through the use of selected additions of radiologically-benign elements such as cerium. Moreover, the increasing use of vacuum melting and vacuum arc refining for the manufacture of high-performance steels is likely to diminish further the residual concentrations of the rare earth elements, on account of their relatively high volatility [4].

The elements most likely to determine the  $\gamma$  dose rate of a type 316L steel at the time of the NET reactor decommissioning are the alloying addition molybdenum, present at a level around 2.5%, and the tramp elements cobalt, niobium and silver. In order to quantify the relative contribution made by each of the above elements to the overall dose rate, activation calculations have been performed for 316L steel under the irradiation conditions anticipated for the Technology phase of NET. These computations were performed with the aid of the UKACT1 library and FISPACT code [12], using the outboard first wall neutron flux and spectrum corresponding to the NET shielding blanket design [13], with an integral power loading of 0.8  $MWym^{-2}$ . The neutron energy spectrum employed for these calculations is Two sets of calculations were performed for the depicted in Fig.Al. Eurocast 316L steel, taking the composition as given in Table 10, but applying different irradiation conditions. In the first case, 7 years of continuous irradiation at a constant power loading of 0.114 MWm-2 were assumed, whilst in the second case irradiation at a wall loading of 1.0  $MWm^{-2}$  was assumed to occur in 7 phases of 1000h duration, equally distributed over a period of 7y with cooling between the phases of Both cases correspond to the same integral wall loading of operation. 0.8 MWym-2 and were included to test whether the predictions are sensitive to the simulated irradiation regime employed. Differences in the predicted activation behaviour were discernible only up to about ly after the end of the irradiation period. Whilst no appreciable difference would be expected in the inventories of long-lived species such as 94Nb and 108mAg, it is noteworthy that no difference between continuous and irradiation conditions appeared even for the relatively short-lived 60Co isotope.

The cobalt present at 0.17% in the Eurocast 316L steel is not an intended constituent but represents a tramp element. The presence of cobalt is not in any way detrimental to the properties of most steels and usually no attempt is made to limit the cobalt content. An exception applies in the case of steels that are destined for nuclear applications in which activation can occur, for which a low cobalt content may be specified. In order to distinguish the contributions made by the cobalt component, activation calculations were also performed for a 'base steel' having the same composition as the Eurocast 316L but containing no cobalt prior to irradiation.

Figures 1 and 2 show, respectively, the post-irradiation time dependence of the specific activity and contact  $\gamma$  dose rate predicted for the 316L steel both with and without its cobalt content of 0.17%. For cooling times between a few years and about 80 years the dose rate is almost entirely due to 6°Co in both cases. This isotope, with half-life of 5.3y, is generated from cobalt via the following reactions, the amount produced being directly proportional to the fluence [11]:

$$^{59}$$
Co $(n,\gamma)^{60}$ Co (68%)

$$^{59}\text{Co}(n,\gamma)^{60}\text{mCo}(IT)^{60}\text{Co}$$
 (30%)

It is also generated via several different multistep pathways from iron, the most important ones at low fluences being the following:

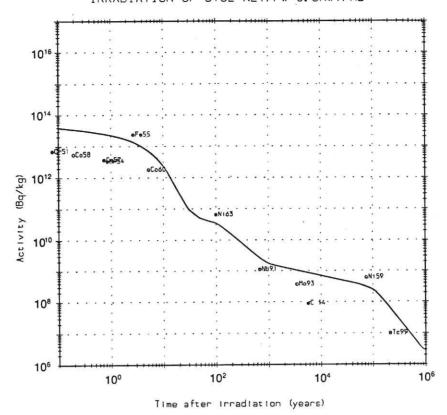
$$^{58}$$
Fe $(n,\gamma)^{59}$ Fe $(\beta^{-})^{59}$ Co $(n,\gamma)^{60}$ Co (69%)

$$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}(\beta^{-})^{59}\text{Co}(n,\gamma)^{60}\text{mCo}(IT)^{60}\text{Co}$$
 (30%)

The production of 6°Co is thus an intrinsic feature of ferrous alloys, irrespective of any other alloying elements or impurities they may contain. Another major constituent of type 316 steels is nickel, which also gives rise to the generation of 6°Co through a large number of competing, mostly multistage, reactions such as

60Ni(n.p)60Co

# IRRADIATION OF 316L NET/FW 0.8MWY/M2



### IRRADIATION OF 316L NET/FW 0.8MWY/M2

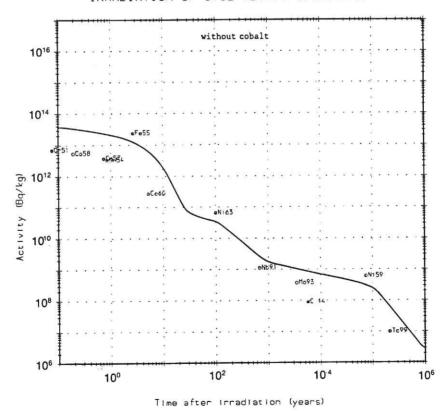
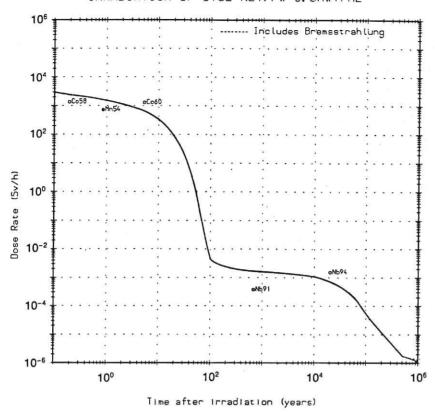


Fig. 1 Specific activity versus cooling time for 316L steel having the composition given in Table 10, with and without the cobalt content of 0.17%. Continuous irradiation for 7y at a first wall power loading of 0.114MW m<sup>-2</sup> in the NET shielding blanket design is assumed.

# IRRADIATION OF 316L NET/FW 0.8MWY/M2



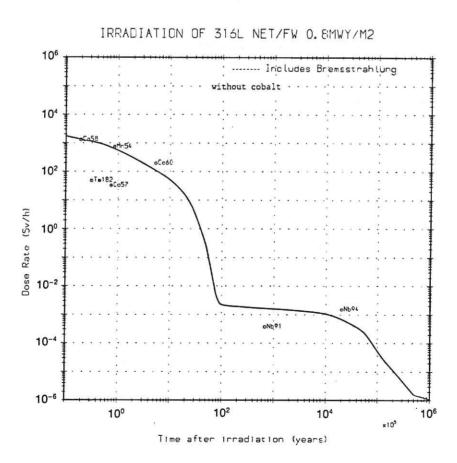


Fig. 2 Contact  $\gamma$  dose rate (E $_{\gamma}$ >0.1 MeV) versus cooling time for 316L steel with and without the 0.17% cobalt content.

From a comparison of the curves in Fig.2 it is evident that the 0.17% cobalt present essentially as an impurity in the steel increases the dose rate by a substantial factor, roughly an order of magnitude for cooling times of 10-100y. From radiological considerations it would clearly be advantageous to postpone decommissioning or the handling of discarded steel components until the activity of 60Co has subsided at about 80y. It will be noted that, because the dose rate contribution from 60Co is falling very rapidly after 80y, the required delay time is only weakly dependent on the amount of 60Co present at shutdown and is thus insensitive to the concentration of cobalt impurity initially present in the steel.

After 100y the dose rate is dominated by the radionuclide 94Nb and decays only marginally over the following 104y. In the present case the 94Nb is derived entirely from molybdenum, mainly through the reactions

94Mo(n,p)94Nb

95Mo(n,d)94Nb

95Mo(n,d)94MNb(IT)94Nb

# 5. The effect of cobalt, niobium and silver impurities

In order to determine the contributions from the potential impurities cobalt, niobium and silver to the overall  $\gamma$  dose rate of the 316L steel, activation calculations under the standard NET irradiation conditions (section 4) were performed for 1 ppm by mass of each of these elements present individually in an inactive 51Fe matrix. This isotope is a fictitious 'pseudo-nuclide' which is stable and undergoes no neutroninduced reactions, so that the matrix is unaffected by the irradiation. By means of this artifice the impurity is represented as being embedded in a non-activating medium that correctly models the properties of steel, such that the absorption of  $\gamma$  radiation and the bremsstrahlung correction are properly represented. Although molybdenum is a specified constituent of 316 steel, calculations were also made for 1 ppm of this element in order to permit comparison of the generation rates of 94Nb in molybdenum and niobium. Dose rate curves for the individual elements are shown in Figs.3 and 4, whilst activity and dose rate values at cooling times of 50 and 100y are tabulated in Table 12.

Table 12

Specific activity and contact  $\gamma$  dose rate values at cooling times of 50 y and 100 y for Co, Nb, Ag and Mo when each is present at 1 ppm by mass in an inactive iron matrix and for 316L steel with and without 0.17% cobalt.

	50y cooling time		100y cooling time			
Material	Activity Bqkg <sup>-1</sup>	Dose rate Svh <sup>-1</sup>	Activity Bqkg <sup>-1</sup>	Dose rate Svh <sup>-1</sup>	Principal radionuclides	
1 ppm cobalt 1 ppm niobium 1 ppm silver 1 ppm molybdenum Eurocast 316L Base steel (no Co)	1.24 E6 4.66 E4 1.29 E6 7.57 E4 5.03 E10 4.82 E10	8.76 E-4 7.15 E-6 4.31 E-4 8.03 E-8 1.77 E0 2.70 E-1	1.74 E3 2.23 E4 9.82 E5 7.46 E4 3.45 E10 3.45 E10	1.22 E-6 7.14 E-6 3.28 E-4 7.94 E-8 4.29 E-3 2.21 E-3	<sup>60</sup> Co <sup>94</sup> Nb, <sup>93m</sup> Nb, <sup>92</sup> Nb, <sup>91</sup> Nb <sup>108m</sup> Ag, <sup>108</sup> Ag <sup>94</sup> Nb, <sup>91</sup> Nb, <sup>93m</sup> Nb, <sup>93</sup> Mo <sup>60</sup> Co, <sup>94</sup> Nb, <sup>91</sup> Nb	

The cobalt contents of standard grade 316 steels are generally in the region of 0.15%. Most of the cobalt is introduced through the ferronickel addition, in which the cobalt content is usually about 3% of the nickel content. The cobalt concentration can therefore be much reduced by the use of high-purity nickel in place of ferronickel and by selecting low-cobalt grades of other ingredients such as ferrochrome. In this way a cobalt content of around 50 ppm might be achievable at reasonable cost. In the fusion neutron spectrum the troublesome radioisotope 6°Co is also produced by reactions on nickel and iron, hence there is no virtue in reducing the cobalt content in a steel below the level at which nickel and iron dominate the generation of 6°Co.

The data in Table 12 indicate that, under NET first wall irradiation conditions, the production of 94Nb is about 100 times greater in niobium than in molybdenum. Since type 316 steel contains about 2.5% molybdenum, equal amounts of 94Nb would be generated from these two parent elements if the niobium concentration were (2.5/100)% or 250 ppm, and there would be little incentive to reduce the niobium concentration much below this value. The analytical data presented earlier in the paper indicate that there should be no difficulty in ensuring that the niobium impurity concentration is below this level.

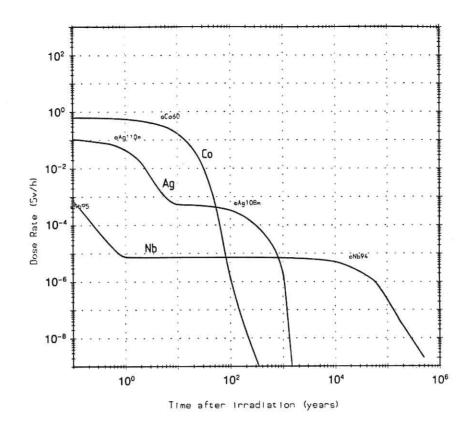


Fig. 3 Contact  $\gamma$  dose rate versus cooling time for 1 ppm by mass of the elements cobalt, niobium and silver present individually in a non-activating iron matrix.

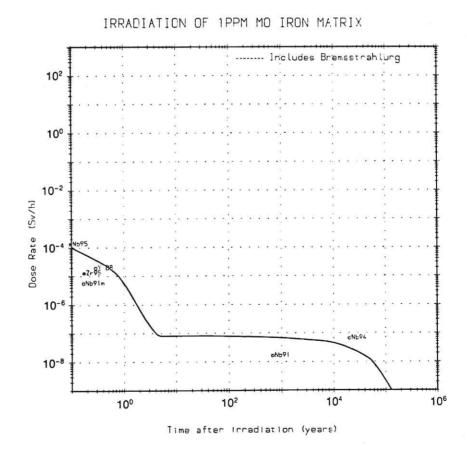


Fig. 4 Contact  $\gamma$  dose rate versus cooling time for 1 ppm by mass of molybdenum in a non-activating iron matrix.

The permitted concentrations of tramp elements can be specified more quantitatively if, for example, some limitation is placed on the extent to which these elements are allowed to increase the overall dose rate above that of the base steel composition. For instance, it might be stipulated that any individual tramp element should not contribute an increase of more than 10% to the dose rate of the base steel at any given cooling time. Such a criterion may seem excessively conservative since a 10% increase in dose rate is not usually significant, though it would allow some margin for the additive effect of several tramp elements.

Figure 5 shows the permitted concentrations of cobalt, niobium and silver calculated by application of the above criterion at different times after the irradiation period. The curve for cobalt shows that, for cooling times of less than about 80y, the concentration of cobalt as an impurity should be limited to about 30 ppm in order to avoid a significant increase in dose rate. Much higher concentrations would be allowable if the dose rate at longer times were the determining factor. In contrast, the elements niobium and silver which lead to the relatively long-lived activation products  $^94$ Nb and  $^{10.8}$ MAg ( $t_{\frac{12}{2}}=127y$ ) are not strongly restricted for short cooling times but are limited to about 30 ppm and 0.7 ppm, respectively, for cooling times around 100y.

For these longer cooling times, of around 100y, silver is the most critical impurity and the concentration in 316L steel should not exceed about 1 ppm. The analytical data presented earlier indicate that the silver content of stainless steels is highly variable but is frequently in the region of 1 ppm. In the commercial production of stainless steels, recycled scrap normally accounts for about half of the input material and probably represents the principal source of the silver impurity. By avoiding the use of scrap and by selecting steelmaking materials that have low silver contents [14] it should be possible to meet the above limitation on silver concentration in a type 316L steel.

It must be emphasised that the concentration limits referred to above are arbitrary insofar as they are based on the requirement that any impurity element should not contribute more than 10% to the overall contact  $\gamma$  dose rate. They can, however, be simply scaled according to the degree of dose rate enhancement considered acceptable and they should also take into account the possible contributions from all significant impurities.

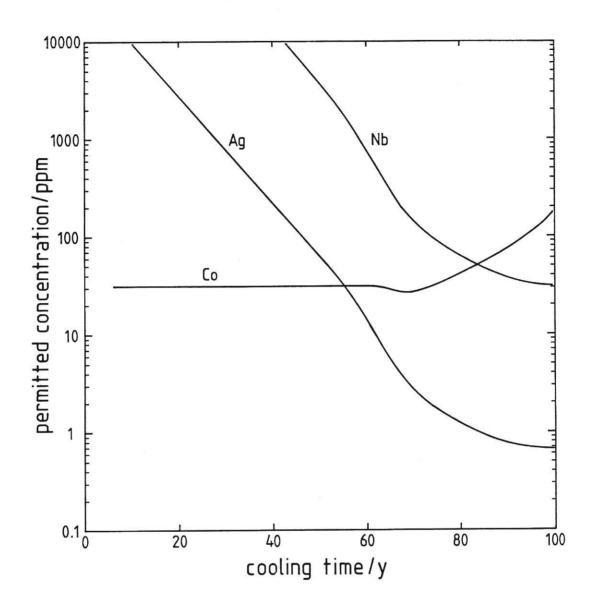


Fig. 5 Permitted concentrations of cobalt, niobium and silver assuming that each element increases the overall contact dose rate of base 316L steel by no more than 10% at a given cooling time.

# 6. Conclusions

In general, the  $\gamma$  dose rates from discarded reactor components made from standard-grade stainless steels will be made up of contributions from radionuclides derived from intended constituents such as iron, nickel and molybdenum and, in addition, contributions from specific impurity elements such as cobalt, niobium, silver and possibly some rare earth metals. The relative contributions of the intended constituents and radiologically-potent impurities must be established by means of activation calculations appropriate to the neutron fluxes, energy spectra and irradiation programme of the reactor concerned. Such a calculation has been performed for a fusion reactor first wall of type 316L steel under irradiation conditions expected during the Technology phase of the Next European Torus.

With this information a compositional specification has been devised such that the residual elements do not appreciably increase the overall dose equivalent rate at a given cooling time and without applying unnecessarily stringent limitations on their permitted concentrations. The results show that there is no need to restrict cobalt and niobium levels to below about 50 ppm and silver below 1 ppm. It should be possible to achieve this degree of purity in a 316L type steel by avoiding the use of scrap metal in the melt and by careful selection of raw steelmaking materials.

# 7. References

- 1. P B Woollam, The radiological consequences of delaying the final dismantling of a Magnox nuclear power station, CEGB Report TPRD/B/0871/R86 (1986).
- 2. P B Woollam, The potential for reducing the radiological consequences of reactor decommissioning through selection of construction materials for activated components, CEGB Report TPRD/B/0514/R84 (1984).
- 3. A R Gregory, J D Regan and P B Woollam, Designing reactors to aid decommissioning, ENC '86 Vol.4, p.397 (1986).
- 4. G J Butterworth, Low activation structural materials for fusion, Fusion Engineering and Design 11 (1989) 231.
- 5. J J Stephens and R O Pohl, Trace elements in reactor steels:

  Implications for decommissioning, Nuclear Engineering and Design 47

  (1978) 125.
- R I Smith, G J Konzec and W E Kennedy, Technology, safety and costs for decommissioning a reference pressurised water reactor, NUREG/CR-0130 (Battelle Pacific Northwest Laboratory, 1978).
- 7. P B Woollam, The radioactive inventory of a decommissioned Magnox power station: I. Measurements of neutron induced activity in samples from the reactor island structure, CEGB Report RD/B/N4231 (1978); II. A summary of neutron induced activation, waste disposal and dose equivalent rates for the reactor island structure, CEGB Report RD/B/R4350 (1978).
- 8. P B Woollam and I G Pugh, The radioactive inventory of a decommissioned Magnox power station, CEGB Report RD/B/R4350 (1978) and in Decommissioning of Nuclear Facilities, IAEA, Vienna, 1979, p.353.
- 9. R M Boothby and T M Williams, The control of cobalt content in reactor grade steels, UKAEA Report AERE R 11107 (1983).

- 10. A J H Goddard, T D MacMahon and M Peselli, Activation of steel components, in 'Decommissioning of Nuclear Power Plants', Proceedings of a European Conference held in Luxembourg, 22-24 May 1984, EUR 9474.
- 11. R A Forrest and D A J Endacott, Activation data for some elements relevant to fusion reactors, UKAEA Report AERE R 13402 (1989).
- 12. R A Forrest, M G Sowerby, B H Patrick and D A J Endacott, The data library UKACT1 and the inventory code FISPACT, Proc. Int. Conf. on Nuclear Data for Science and Technology, Mito, Japan, 30 May-3 June 1988, p.1061.
- 13. K A Verschuur and Ph Cornelissen, Specifications for activation calculations on shielding blanket and divertor concepts to be performed at JRC Ispra, NET/87/NE-S-038 (1987).
- 14. O Gautsch, Impurity levels in steels, 1987 Programme Progress Report Fusion Technology and Safety (JRC Ispra), EUR 11697, p.95.

# NET (outboard)

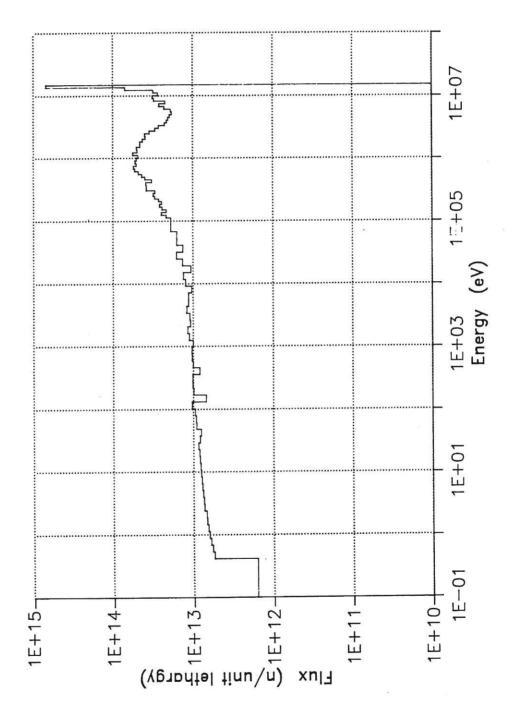
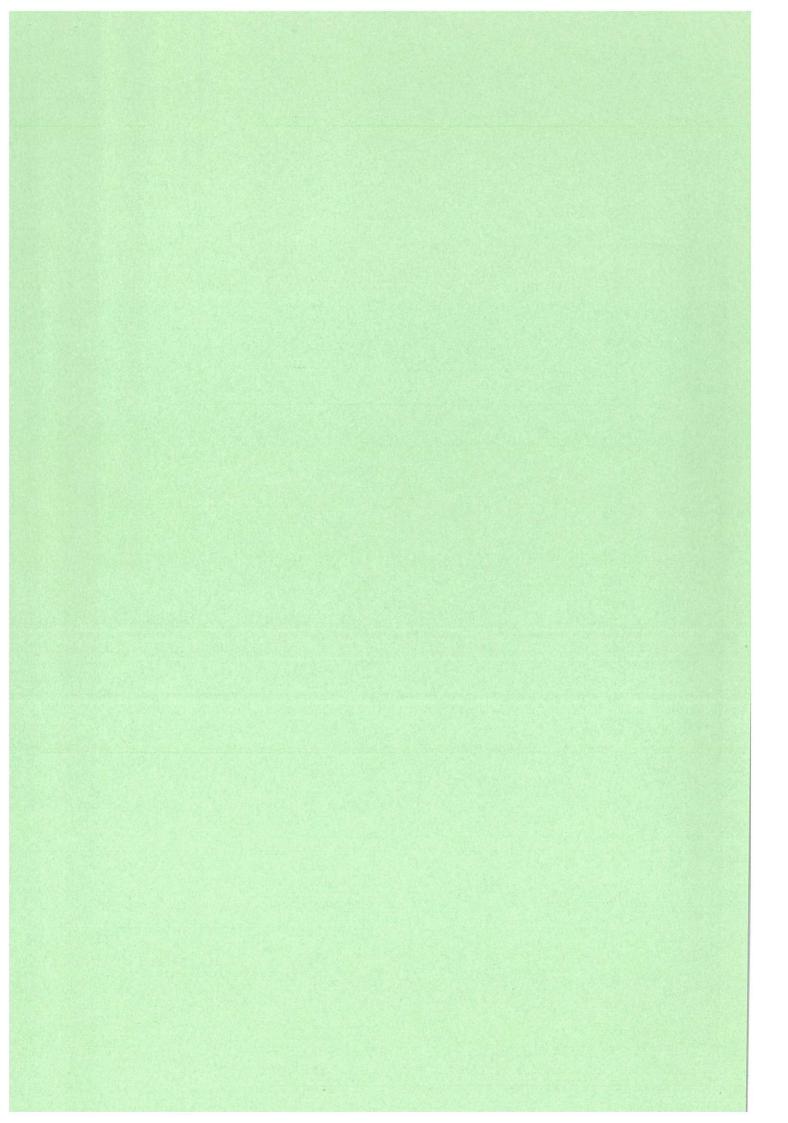


Fig. A1 Neutron energy spectrum assumed for the activation calculations.



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