

Neutron- Induced Activation of Materials for the First Wall of Conceptual Fusion Reactors

by

Amjad Khursheed

*A thesis submitted for the degree of Doctor of Philosophy of the University of
London*

Centre for Fusion Studies
Imperial College of Science and Technology
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Abstract

Activation properties of structural materials for the first wall of a deuterium-tritium fuelled tokamak fusion reactor are predicted from inventory calculations performed with the newly developed fusion activation code FISPACT and the substantially upgraded UK fusion activation database UKACT1, in neutron irradiation conditions derived from the outer wall position of the Culham DEMO reactor design. The materials examined include all stable elements with atomic numbers in the range 3-83 and 7 candidate compositions for the first wall, the latter comprising of 2 commercially available steels, 2 low-activation steels, 2 vanadium-based alloys and silicon carbide.

An analysis of the sensitivity of the inventories of long-lived products with respect to the cross-section database is made, allowing the principal reaction production channels to be identified and approximate errors to be assigned to computed nuclide concentrations. The adequacy of the cross-section database is discussed, in respect to both the critical cross-section data highlighted by the sensitivity analysis and the overall requirements for fusion activation calculations, and a set of criteria for the inclusion of cross-section data is formulated.

Potential exposure scenarios involving the long-lived activation products from candidate first wall materials are considered, with the emphasis on post-disposal environmental impact.

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To the

Songs my mother would half-sing
In the kitchen,
While the eye of God dozed peacefully
In the cupboard under the sink.

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*And (I will) pluck when time and times are done,
The silver apples of the moon
The golden apples of the sun.*

- W.B. Yeats : The Song of Wandering Aengus (1899)

*White coat and purple coat
Can each be worn in turn
In white a man will freeze
In purple burn.*

- Dannie Abse : White Coat, Purple Coat (1989)

INTRODUCTION

As we approach the end of the 20th Century, the outlines of the energy debate appear to be hardening - if the body of the argument has not yet acquired transparency, then it has certainly achieved translucence with the emergence of environmental issues as one of the key factors ; their importance enhanced by recent reports ¹ which suggest that the "greenhouse effect" is now a reality. A direct consequence is that the potential advantages of any new energy generation scheme, with the emphasis on nuclear fusion here, must be balanced against the likely environmental consequences at an early stage in its development.

The attractions of a system of power generation built upon nuclear fusion are obvious and have been widely disseminated ; a readily available source of fuel, the absence of radioactive products from the fusion reaction, a weight for weight superiority in energy yield over fission. These advantages are confirmed if some potentially useful fusion reactions are examined ;

- | | | |
|----|---|--------------------|
| 1. | $D + D \longrightarrow T + H$ | $+ 4\text{MeV}$ |
| 2. | $D + D \longrightarrow \text{He}^3 + n$ | $+ 3.25\text{MeV}$ |
| 3. | $D + T \longrightarrow \alpha + n$ | $+ 18 \text{ MeV}$ |

It has become increasingly apparent, however, that fusion reactors based upon reaction (3), the reaction adopted by most conceptual reactor designs, would not be entirely free of radioactive products. An

intrinsic feature of these designs will be the activation of structural components adjacent to the plasma chamber by the energetic neutrons produced by the deuterium-tritium reaction. As the part of the structure closest to the source of activation, and also the structural component requiring the most frequent replacement, the reactor first wall will contain the major part of the reactor's radioactive inventory. Activation of the first wall is therefore the most obvious place to start a study of fusion reactor activation.

Throughout this thesis, therefore, the discussion will be restricted to activation in fusion reactors based upon the D-T fuel cycle. It should be pointed out here that in the long term, when plasma confinement technologies for plasmas are well-established, it may be that designs based upon reactions whose products are all charged particles - thus confining the energy released by the reaction to the plasma and removing the need for a lithium blanket - will be preferred.

0.1 Activation mechanisms

First wall activation will result in the main from interactions with the primary flux of 14MeV neutrons emanating from the plasma, and a scattered flux with energies in a range between 0 and 14MeV. The types of interactions which can be expected to occur between neutrons and host nuclei have been summarised in figure 0.1, taken from Fermi². With the exception of the scattering reaction (n,n') and a small number of direct reactions of the type $N^{14}(n,p)C^{14}$ - the reaction responsible for the production of atmospheric C^{14} - the reactions proceed by the formation of a compound nucleus. At energies below the thresholds for particle emission, the dominant interaction is therefore radiative capture (n,γ) . At energies above a few MeV particle emission

becomes increasingly important, such that it is the dominant class of interaction for many first wall nuclei. The number of particle emission permutations permissible at neutron energies up to 14MeV is large, the most important of which are shown on a Z/A field in figure 0.2.

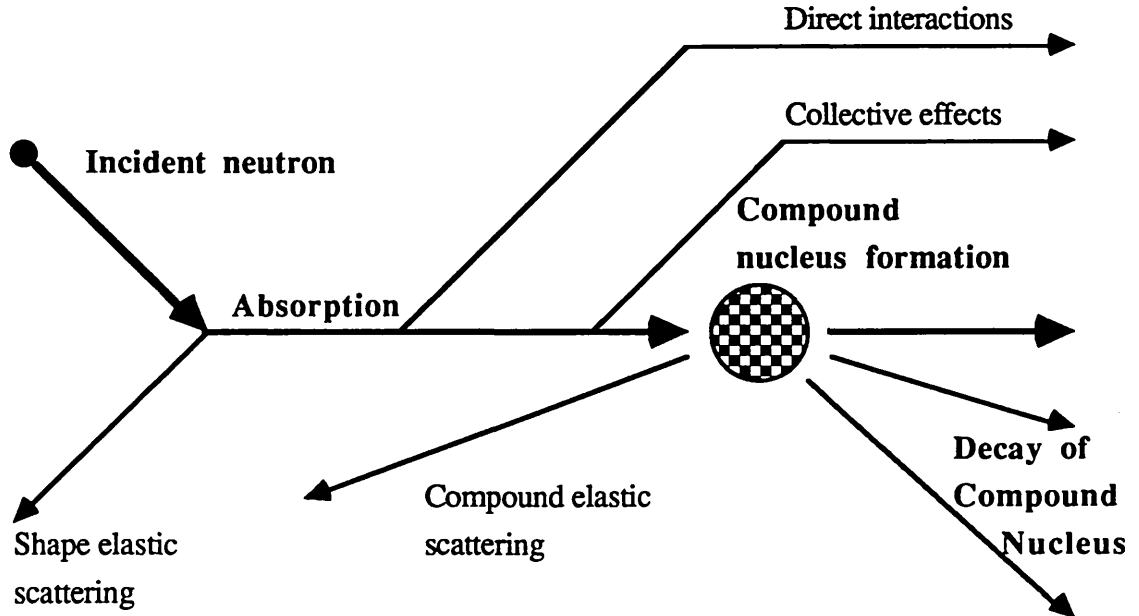


Figure 0.1 : Neutron interactions in the first wall ²

Neutron-induced reactions, though the most probable type, are not the only processes which could result in first wall activation. There will also be activation from secondary charged particle reactions and reactions caused by runaway electrons. The former would occur after neutron-induced reactions of the (n,p), (n, α) type etc., and could be significant in blanket regions where there are large amounts of lithium and beryllium. The latter can lead to a high energy tail in the neutron energy spectrum, which might introduce a new range of interactions. It is unlikely that any radically new reactions will take place at energies up to 20MeV however, and both these sources of

activation will probably be fringe phenomena in the first wall.

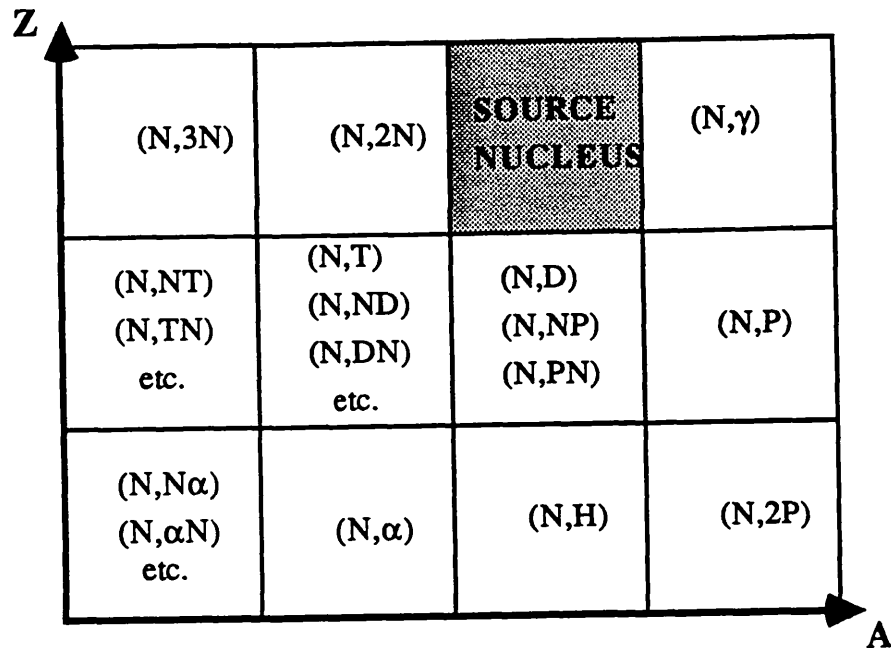


Figure 0.2 : Particle emission permutations in the energy range upto 14Mev

At the high fluences received by a first wall over its lifetime, it is overwhelmingly probable that some nuclei will undergo successive reactions, transmuting the daughter nuclei considerable distances away from the parent in a Z/A field. The presence of these activation chains greatly increases radionuclide inventories and increases the chance of hazardous species being produced. Their inclusion is essential, therefore, to the success of any model of first wall activation

0.2 Activation and reactor design

Current designs for conceptual fusion reactors have engineering

requirements which would push existing technologies to their very limits, in some cases further. The activation behaviour of a material is one, and by no means the most fundamental, of many properties which would determine the suitability of that material for first wall construction. Thus most fusion reactor designs have not hitherto incorporated features which would minimise activation, and there is considerable scope for reducing levels of activation by altering design parameters. Strategies which could reduce activation are

- i) the selection of materials with intrinsically low levels of activation for critical structural components
- ii) repositioning and shielding of highly activated components where this may be done without an impairment of their performances. A candidate for this approach would be the superconducting coils (already an issue because of radiation-induced resistivity) ³, as the inner limbs are in close proximity to the plasma, with a minimum of shielding.
- iii) adjusting the operating conditions of the components with respect to the neutron wall loading and the component lifetime.
- iv) recycling and reuse of activated material. This approach would be particularly suited to high cost components made from rare materials - such as vanadium-based first wall alloys, materials which are very pure, or materials which have been isotopically tailored.

The strategy which appears to offer the greatest reductions in the radioactive inventory at the lowest cost to important design parameters, is i). Programmes exist both in Europe, Japan and the United States to develop first wall materials with intrinsically low levels of activation, and much of the previous work on fusion activation has been orientated in this direction.

0.3 Review of previous activation calculations

The first serious attempts to assess the activation in conceptual fusion reactor designs were centred at Brookhaven in the United States in the early seventies. This early work concentrated on specific first wall materials, particularly those based upon niobium and molybdenum, materials which were then thought to have good structural properties. Subsequent work has shown these elements to possess relatively unfavourable activation properties.

In common with other early assessments of activation in a fusion reactor context, many assumptions made about the nature of the activation in this study were derived from experience with fission reactors. Some of these have now been shown to be incorrect or misleading. Thus the first phase of the Brookhaven work ended with a restricted database with data for only 39 reactions ⁴, as it was assumed that multiple reactions would not be important in the fusion first wall activation case. It is now clear that chains of multiple reactions are in fact extremely important in typical fusion reactor irradiation conditions, an example being the production of Ir192n from W186 by 6 successive capture reactions.

This early work has been thoroughly reviewed by Jarvis ⁵, whose work at UKAEA Harwell played a large part in laying down the foundations for UK participation in the field of fusion activation. The

rationale underpinning his work then, as it still remains in current work, was to extend the database and widen the range for candidate materials while retaining an essentially sound calculational method derived from fission applications. Important contributions were also made to the assessment of long-term gamma doses in handling/reuse scenarios and methods of disposal were discussed. As this work has become a standard for much of the subsequent work in the UK, including the present study, a closer examination of some of the assumptions and important aspects is warranted.

An initial set of activation calculations was conducted for five candidate structural materials ⁵, with neutron energy spectra obtained for the Culham Conceptual Tokamak Mark IIA ⁶ (CCTR), using the ORIGEN inventory code and the UKCTRIII fusion activation database. The materials compositions were in some cases complex, and it was concluded that a more profitable course would be to look at the activation properties of elements separately : activation properties of compositions could then be obtained by summing the contributions for individual elements, thereby providing knowledge of the origins of particular activation products, knowledge which could be employed to optimise low-activation compositions.

A second, and more complete, set of calculations ⁷ followed for what were then considered to be the main structural elements. This second set of calculations contained some points of interest. Firstly, the database UKCTR-IIIA ⁸ had been upgraded, and contained 100 group cross-section data for 1477 reactions and 410 target nuclei which were stable or had half-lives in excess of 10 days. The patchy coverage and low quality of much of these data, however, merited mention. Secondly, the same neutron spectra as for the first set of calculations were

employed at a cumulative wall-loading of 20MWyr/m^2 , made up of a single 2 year burn at a power loading of 10 MW/m^2 . It was argued with respect to these irradiation conditions that

a) activation scaled with fluence (irrespective of the combination of flux and irradiation time)

b) the neutron energy spectrum is largely insensitive to reactor design

There are now considerable doubts as to the validity of the widespread application of these rules (see Chapter I), and it is evident that they may lead to some misleading assumptions. Thirdly, the principal index for radiological hazard was taken to be surface gamma doserate in the infinite slab approximation after 30 and 100 year cooling periods - the timescales relevant to reuse and disposal respectively. Potentially significant approximations were made in the doserate calculations by the assumptions that all α particles, all β particles and γ rays with energies less than 200keV would be removed by shielding, and that the gamma attenuation coefficients were those for the element under consideration. The concept of "Maximum Permissible Concentration", derived from the gamma doserates as the percentage fraction of the first wall which could be made from a certain element if some limit on doserate could not be exceeded, was applied to each of the elements examined with the limit of $25\mu\text{Sv/hr}$ - the so called "hands on" criterion. According to this limit most of the elements included in the study were acceptable at concentrations of 100% (see *table 1.1*), notably from the viewpoint of low-activation materials V, Cr, Mn, Fe, Ta, W and Pb. The following elements were restricted however Al, Si, Ti, Ni, Cu, Nb, Mo, Ag and Cd, some to levels of 1ppm or below.

In a third and final phase ⁹, the calculation of gamma doserates

was refined by the inclusion of a treatment for bremsstrahlung emitters, estimates were made for activation levels in minority and impurity elements which did not appear in the earlier calculations, and the existing regulations and guidelines for the disposal of activated material were discussed. This phase was in the nature of a supplement to the main body of work, dealing with topics which were less self-contained than the elemental activation calculations. The effects of applying the bremsstrahlung correction appeared to be restricted to a few relatively unimportant elements, and did not significantly alter the long-term doserates for any of the common structural elements. With the wider range of elements now under consideration for first wall elements however, the bremsstrahlung correction may play a significant role, particularly for light element inventories (i.e. lighter than Na), in which there are no long-lived gamma-emitters. The estimates for minority and impurity elements, as in the Brookhaven work, were invalidated by the assumption that multiple reactions are rare - activation calculations for these elements should be performed with a complete database.

Further refinements to the basic Jarvis model were added in the calculations of Giancarli ¹⁰, including the use of a neutronics model based upon a Culham DEMO conceptual reactor design - stemming from the realisation that the breeding blanket in the CCTR design would not achieve tritium self-sufficiency - and a version of the UKCTR-IIIA database strengthened by THRESH ^{11,12} cross-section determinations for missing reactions and by the inclusion of some long-lived metastable species. In addition, the irradiation conditions were changed to the somewhat more realistic ones of a single burn of length 2.5 years at a first wall power density of 5MW/m², and gamma doserates were calculated under the assumption that each element

was present at a 1ppm level in an iron matrix with the gamma attenuation coefficients of iron.

As in Jarvis' work, the study was centred on the long-term activation properties of first wall and blanket materials, and standardised the presentation of elemental doserates to the Maximum Permissible Concentrations after a 100 year cooling period. Significant differences resulted for K, Ca, Co, Ni, Cu, Ta, W and Pb, the Giancarli figures being in all cases more restrictive. A factor singled out for mention was the softer neutron energy spectra employed by Giancarli, which substantially increased single group capture cross-sections. In individual cases, other factors, such as the different irradiation conditions, or the altered database (notably for Ta and W), also come into play to differing degrees.

Outside the UK, sets of fusion activation calculations aimed at long-term gamma doserates from the first wall and blanket, have been performed in the United States ¹³ and in Italy ¹⁴, both with activation databases based on REAC ¹⁵. Taking the results from the latter as being representative of both sets of results, it is noted that major differences occur with Giancarli in the MPC's for Sc, Ti, Fe, Sr, In, Sn, Sb, Te, Xe, Lu, Hf, Ta, W and Pb. The disparities can in most cases be ascribed to improved cross-section data in the REAC data library, but this does not appear to be the case for Ti and Fe. A closer examination of these two critical constituents of low-activation materials is called for.

Adopting a similar treatment for candidate first wall compositions as for pure elements, Giancarli examined the activation properties of a range of steels, vanadium alloys and silicon carbide, the results of which are discussed fully in Chapter V, in relation to results from the

present study.

The radionuclide inventories given by these activation calculations have been taken as the basis for studies on the environmental impact of fusion waste ¹⁶ and the hazards associated with potential recycling scenarios. The former concludes, in support of the earlier discussion by Jarvis ⁹, that shallow land burial is not a viable option for any of the candidate wall materials examined. The study on potential recycling scenarios was confined to the inventories produced by low-activation steels and concludes, amongst details of the chemistry and mechanics of recycling, that the 25 μ Sv/hr "hands on" limit on gamma dose rate is unduly pessimistic, and that 25mSv/hr would be more realistic with the handling procedures likely to be adopted.

Recent work in Italy ¹⁷ and the United States ¹⁸ has modelled post-disposal environmental impact and used the resulting doses, in combination with existing waste disposal guidelines in Europe and the United States, to provide MPC's which have a more precise basis than those based entirely on surface gamma dose rate and the "hands on" criterion. This may be seen as part of a trend : as activation calculations achieve higher degrees of accuracy, the burden of future research in fusion activation will move more towards detailed dosimetric assessments, over a range of timescales and hazard scenarios.

0.4 Aims of the present work

The programme to assess activation in fusion reactors and develop low-activation materials is essentially a programme with very long-term aims. The goals of such a programme are liable to shift with changes in design, new technology or the political climate. The work

described below is therefore but one stage in a long process and will attempt to tackle the most pressing deficiencies of previous work while striving to take in new developments which have occurred in the intervening time.

The specific aims are

- 1) to assess the long-term activation properties of all the stable elements under the neutron irradiation conditions likely to be found in the first wall of a fusion power reactor based upon the D-T fuel cycle.
- 2) to assess the dependence of activation properties on irradiation conditions , neutron energy spectra and neutron wall-loadings
- 3) to assess the adequacy of the newly formed UK fusion activation database for the task outlined in 1).
- 4) to perform a sensitivity analysis on the newly formed activation database in order that
 - i) critical cross-section data, and hence reactions pathways, may be identified
 - ii) estimates can be made of errors in nuclide concentrations
- 5) to assess the activation properties of a number of candidate first wall compositions and the radiological implications of potential exposure scenarios.

CHAPTER I

Pure Element Activation Calculations

*There is shadow under this red rock,
(Come in under the shadow of this red rock),
And I will show you something different from either
Your shadow at morning striding behind you
Or your shadow at evening rising to meet you ;
I will show you fear in a handful of dust.*

- The Burial of the Dead

Introduction

The problems associated with realistically modelling fusion activation chains in the fusion reactor first wall, are partly problems in the manipulation and the interpretation of large volumes of data. While the need for large nuclear data libraries appears to be an irreducible feature of fusion activation calculations, interpretation of the radionuclide inventories produced from sometimes complex material compositions can be simplified greatly by considering activation of the constituent elements separately. As noted in the review of previous work in the Introduction, this procedure has the advantage of revealing information about the relative activation properties of elements which can be used to improve materials compositions. The approach taken here, therefore, is to consider first the activation properties of individual elements, and later (Chapter V) consider the collective properties of these elements as they occur in candidate first wall compositions.

To make the study as wide-ranging as possible, all stable elements

with atomic numbers between 3 and 83 are examined. Previous studies have concentrated on well-known structural materials ^{7,10,14} : it was felt that widening the scope of the investigation would also enable the roles of potential impurities and novel alloying additions in low-activation materials to be assessed.

1.1 The Activation Calculation

The method for modelling activation in fusion reactors is well established, being based on a procedure developed for the analytically similar problem of fuel burn-up in fission reactors : a set of differential equations describing the time-dependent concentrations of nuclides is solved by a standard inventory code, employing a constant flux obtained from a preceding neutronics calculation. This procedure has been found to be satisfactory in the past within the limits imposed by the other errors present in the analysis, and it has consequently been adopted here, albeit with minor variations which shall be explained.

1.1.1 The inventory equations

Consider a fusion reactor material which contains n nuclides at concentrations N_i at time t . Then the time differential of concentration N_i is equal to the sum of the rates of depletion and production of nuclide i .

$$\frac{dN_i}{dt} = -\left(\lambda_i + \int \sigma_i(E) \phi(E) dE\right) N_i + \sum_{j=1}^n \left(b_{ji} \lambda_j + \int \sigma_{ji}(E) \phi(E) dE\right) N_j + T_i \quad (1.1)$$

where λ_i is the decay constant associated with nuclide i

σ_i is the total absorption cross-section at energy E for nuclide i

ϕ is the neutron flux in unit energy interval

λ_i is the decay constant associated with the decay of i

b_{ji} is the branching ratio for the decay of nuclide j to i

σ_{ji} is the cross-section at energy E for the transmutation of nuclide j to i

T_i is a source term for other production channels (e.g. fission)

If the neutron energy spectrum is known in the 100 group GAM-II⁸ energy structure, as were the spectra used in this study, the expression (1.1) is modified to

$$\frac{dN_i}{dt} = - \left(\lambda_i + \sum_{k=1}^{100} \sigma_{ik} \phi_k \right) N_i + \sum_{j=1}^{100} \left(b_{ji} \lambda_j + \sum_{k=1}^{100} \sigma_{jik} \phi_k \right) N_j + T_i \quad (1.2)$$

A further simplification can be made if the cross-section data has previously been collapsed into single energy group :

$$\frac{dN_i}{dt} = - \left(\lambda_i + \sigma_i \phi \right) N_i + \sum_{j=1}^n \left(b_{ji} \lambda_j + \sigma_{ji} \phi \right) N_j + T_i \quad (1.3)$$

where the single group cross-section σ_s and flux ϕ are defined as

$$\sigma_s = \frac{\sum_{k=1}^{100} \sigma_{sk} \phi_k}{\sum_{k=1}^{100} \phi_k}$$

$$\phi = \sum_{k=1}^{100} \phi_k$$

In the parlance of numerical analysis, the set of equations (1.3) can be categorised as a stiff set of ordinary value, first order differential equations. In general, classical methods employing first order Taylor expansions are not suitable for this type of equation, as they require large numbers of time steps, and exponential forms are to be preferred. The particular form of the expansion adopted by FISPACT, the dedicated fusion activation code developed in the course of this study ¹⁹, was taken directly from the FISPIN6 inventory code, and is based upon the method of Sidell ²⁰ :

$$N_i(t+h) = N_i(t) + \frac{(e^{\lambda_i h} - 1)}{\lambda_i} \left(\frac{dn_i}{dt} \right)_t$$

where λ_i represents $(\lambda_i + \sigma_i \phi)$.

The solution has been tested against alternatives, such as that utilised by the matrix inversion code ORIGEN ²¹, and has been found to compare favourably, the computational errors being overshadowed by errors in respective databases ²².

1.1.2 First wall irradiation conditions

The irradiation conditions adopted in these calculations have been derived for an outer first wall position in the Culham DEMO conceptual tokamak design ²³. They assume a single continuous burn of length 2.5 years, the estimated lifetime of a first wall component.

The wall power loading, however, has been increased from 2.6MW/m² to 5MW/m², as it was felt that the original value was lower than would be expected in a power reactor. This departure from design is justified by the need to provide typical first wall conditions to assess the activation properties of a range of candidate first wall materials, rather than assess the merits of a particular fusion reactor design.

The cumulative wall-loading is therefore 12.5MWyr/m². It is relevant to point out here, contrary to what was recently the prevalent view ⁷, that the activation effects of irradiations which have arrived at the same cumulative wall-loading by different combinations of power and irradiation time, can be different if the activation chains contain nuclei with half-lives short with respect to the irradiation period (see Chapter III). An example of such an activation chain is the production of Ar42 from titanium via the unstable Ca45 isotope ($t_{1/2} = 165$ days). Thus the approach here has been to simulate the actual irradiation conditions as closely as possible.

The representation of what in practise is likely to be a pulsed irradiation cycle by a single continuous burn was felt to be a safe approximation : the only nuclides likely to be affected are those target nuclei with half-lives less than the length of the off-time in each pulse (~ 50 seconds), the roles of which will probably be marginal in a study of long-lived products (see Chapter III).

The irradiation was followed by a cooling period of 100 years, chosen in accordance with the study's brief of examining the long-lived activation properties of first wall materials, particularly with regard to post-disposal environmental impact. The choice of this decay timescale inevitably restricts the scope of the findings, but useful conclusions may yet be drawn for other timescales if suitable care is taken.

1.1.3 The neutron energy spectrum

The first wall neutron energy spectrum employed in these calculations was evaluated by means of a 1-D cylindrical transport model (ANISN) for a Culham DEMO solid breeder tokamak design ¹⁰. Although first wall spectra from other sources are not available, comparisons with 3-D Monte Carlo models in the blanket region ²³ suggest that the model is satisfactorily accurate for the purpose of activation calculations.

The main features of the spectrum are shown in figure 1.1 ; a thermally broadened spike of primary fusion neutrons at about 14MeV, a backscattering peak which maximising at about 1MeV, and a gradual decline with decreasing energy thereafter. It has been stated by previous workers that the neutron energy spectrum is insensitive to reactor design ⁷. That this is by no means certain, can be appreciated by comparing the power normalised (to a wall-loading of 1MW/m²) first wall spectra corresponding to the DEMO, CCTR and NET ²⁴ designs, as shown in figure 1.2. The differences are most noticeable below 100 keV, in the thermal and resonance regions, where the water-bearing NET design exhibits greater thermalisation than the DEMO design. In contrast, the CCTR spectrum tapers off more rapidly than DEMO in this energy region, probably owing to poor neutron multiplication in the blanket. These differences may have potentially significant consequences for capture reaction rates and will be investigated further.

1.1.4 Evaluation of hazard indices

From the nuclide concentrations obtained from the inventory

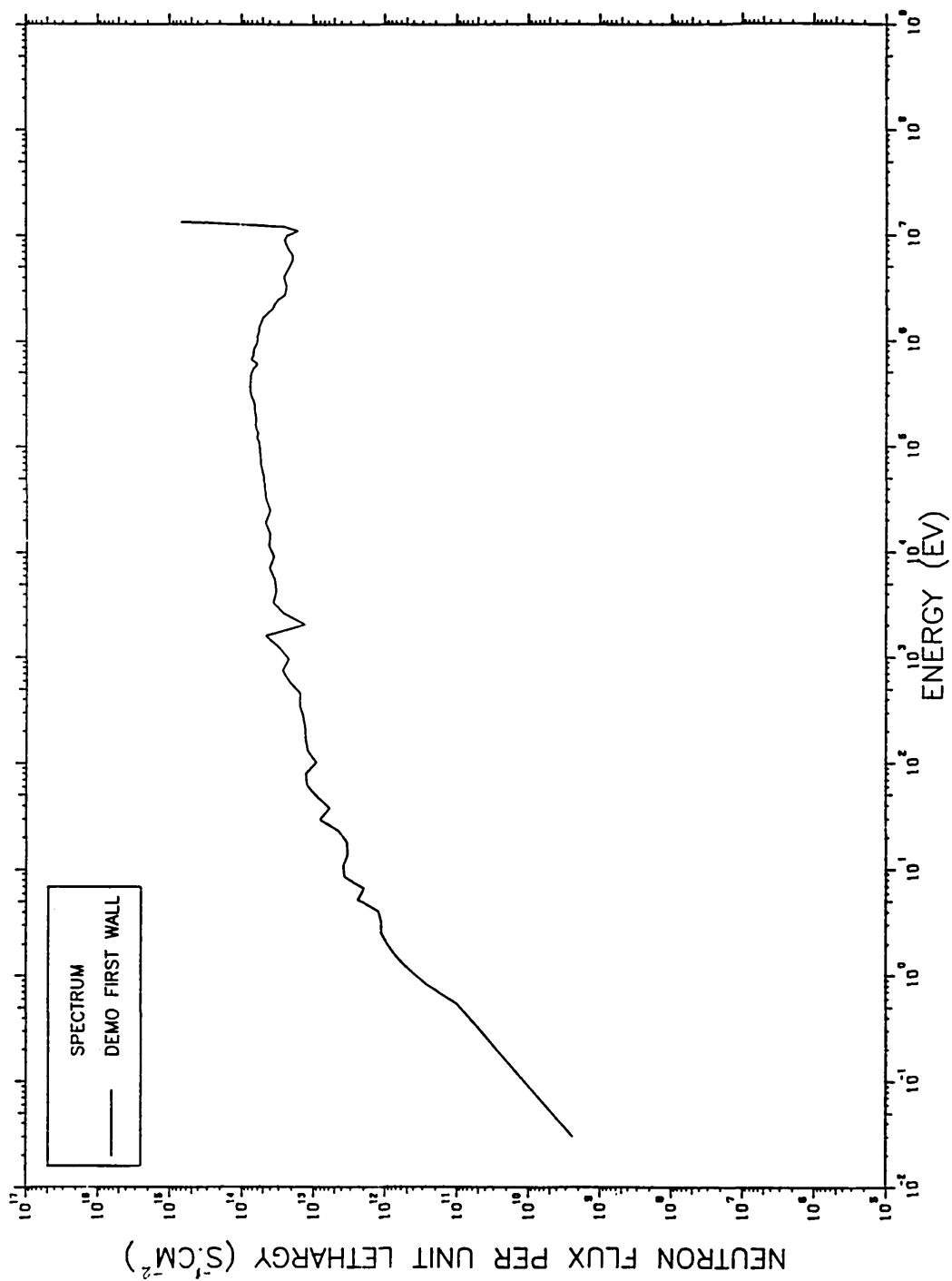


Figure 1.1 : First wall neutron energy spectra for the Culham DEMO conceptual reactor design

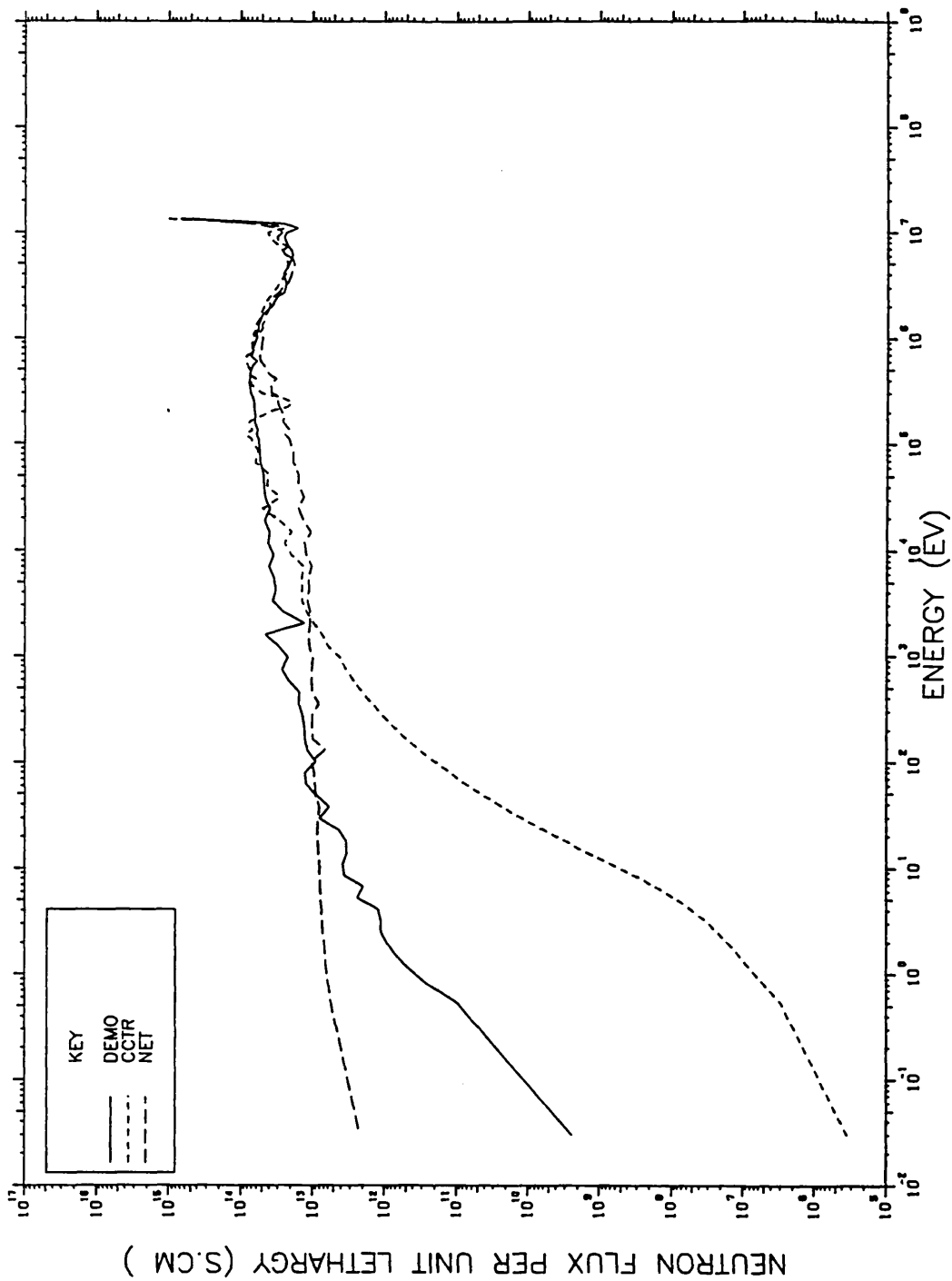


Figure 1.2 : Comparison between first wall neutron energy spectra for the DEMO, CCTR and NET reactor designs

calculation, the following four hazard indices have been evaluated for each inventory nuclide ; activity, surface gamma dose rate, specific potential ingestion dose and specific potential inhalation dose.

Activity, defined simply as $N_i \lambda_i$, is the most fundamental of these indices, since the others are obtained from it by the application of the appropriate factors. Used as the sole index for hazard, however, it can be misleading as it conveys no information about the potential biological hazards.

Specific ingestion and inhalation doses for inventory nuclides are obtained directly from the activity

$$D_{\text{ing}} = N_i \lambda_i d_{\text{ing}}$$

$$D_{\text{inh}} = N_i \lambda_i d_{\text{inh}}$$

where d_{ing} and d_{inh} are committed effective dose equivalents (CEDE) for the ingestion and inhalation per unit activity of a nuclide, obtained from NRPB datafiles ¹⁹. It is important to note that these doses are somewhat idealised : in physical terms they correspond to the doses resulting from the intake of 1kg of first wall material, not a very probable event. In practice the quantities of ingested and inhaled material would be determined by the pathways which would link the subject to the activated material. Caution must be exercised in their interpretation too because CEDE's are not available for some short-lived and uncommon isotopes, with the consequence that the ingestion and inhalation doses from inventories containing these isotopes may be greatly underestimated.

The advantage of using surface gamma dose rate as a hazard index

is that it is independent of irradiation geometry. It can be shown ²⁶ that the gamma flux at the surface of an infinite sheet of thickness h is given by

$$\phi = \frac{S_v}{2\mu_s} \left[1 - E_2(\mu_s h) \right] \quad (1.4)$$

where $\mu_s = \mu_a / \mu_m$, the quotient of the attenuation coefficients of the material and air, and S_v is the rate of gamma emission ($\text{MeV kg}^{-1} \text{ s}^{-1}$). For large $\mu_s h$, the function E_2 tends to zero, hence the flux at the surface of an infinite slab of infinite thickness is

$$\phi = \frac{S_v}{2\mu_s} \quad (1.5)$$

That the first wall of a fusion reactor may be taken to be of infinite thickness can be demonstrated by taking as a typical first wall a sheet of 1cm thick steel. Substituting the appropriate attenuation coefficients at 100keV (where the difference between the two expressions should be greatest) and $h = 1\text{cm}$ into (1.4) and (1.5), and a value for the integral E_2 obtained from tables ²⁶, it is shown that the difference between the two expressions amounts only to 1.8%. Furthermore, the approximation is made safe because E_2 varies more slowly as h becomes small.

Expression (1.5) has therefore been adopted to form the basis of an expression for surface gamma dose rate. The full expression for the

doserate, defined as the absorbed dose in air, and including a build-up factor $B = 2$ to treat Compton-scattered secondaries, is

$$D = 5.76 \times 10^{-10} \mu_a S_v \frac{B}{2 \mu_m}$$

It has been noted by previous workers ^{9,10} that a significant contribution to gamma dose from inventories including energetic beta emitters can come from bremsstrahlung X-rays. The formulation adopted by FISPACT to treat this bremsstrahlung contribution is essentially the same as that used by Jarvis ⁹, and is derived from an empirical relationship for the production of X-rays by the bombardment of a heavy target with a monoenergetic beam of electrons²⁷. This states that the number of photons with energy E produced in energy interval dE is given by

$$dN = a Z \frac{(E_0 - E)}{E} dE \quad (1.6)$$

where E_0 is the energy of the electron beam in keV

$$a = 2.76 \times 10^{-6}$$

Z is atomic number of target material

By straightforward integration it can be shown that the number of photons produced in energy interval ϵ_1 to ϵ_2 is therefore

$$N = a Z (\epsilon_1 - \epsilon_2) \left[\frac{E_0}{\epsilon_1 - \epsilon_2} \ln \left(\frac{\epsilon_1}{\epsilon_2} \right) - 1 \right]$$

(1.7)

In this way the bremsstrahlung flux in each of the 24 gamma emission energy groups is evaluated and the resulting surface gamma dose rate calculated as before.

It is clear that expression (1.7) is only an approximate representation of bremsstrahlung flux which might with some effort be improved upon. One possible improvement may be to treat the beta particle source as a continuum with respect to energy rather than a monoenergetic beam with energy E_0 , as is implicit in the adopted treatment. Evaluations of the bremsstrahlung dose from the three beta emitters Be10 ($\langle E_\beta \rangle = 203$ keV), Cl36 ($\langle E_\beta \rangle = 246$ keV) and Y90 ($\langle E_\beta \rangle = 695$ keV) with the binned energy distributions given in Browne and Firestone ²⁸ (Appendix I), suggest that representation of the energy continuum by a monoenergetic source with average energy $\langle E_\beta \rangle$ systematically underestimates the dose, roughly by a factor of 2.

In line with previous workers in the the U.K., low energy photons (with the cut-off set at 100keV here) have not been included in the treatment of surface gamma dose rate on grounds of computational convenience, with the justification that they would be removed by shielding in controlled handling/recycling scenarios.

1.1.5 Data libraries

The combined cross-section and decay data library used in these activation calculations, UKACT1 ²⁹, represents a considerable advance in the activation database, both in size and quality. Its antecedents can be traced to the REAC library ¹⁵, from which it now differs substantially, the current version containing data for 625 target nuclei and 8719 reactions. The rationale behind the development of the

database and the impact its use has for the activation calculations are discussed fully in Chapters II and IV.

1.1.6 Results and Discussion

The overall pattern of pure element activation may be discerned most clearly from figure 1.3, which shows specific activity for each element following the 100 year cooling period. A striking feature is an apparent periodicity of activity with Z number. It is precisely this apparently periodic nature of the activation, with regions of highly activated elements interspersed with "low-activation windows", which opens the ways for the development of low-activation compositions.

The peaks are associated with the generation of certain long-lived activation products. Superimposed upon the overall pattern of peaks and enhancing it further is the clustering of long-lived nuclei about certain Z numbers : for example, the elements Cr, V, Ti and Sc have only one marginally important long-lived isotope, $Ti44$, whereas Nb alone has three, $Nb91$, $Nb93m$ and $Nb94$.

It is possible to see how the peaks are formed by considering an example pertinent to the development of low-activation vanadium alloys, the peak at $Z = 18$, corresponding to the isotopes $Ar42$ ($t_{1/2}=33\text{years}$) and $Ar39$ ($t_{1/2}=269\text{years}$). The isotopes are produced in progressively decreasing quantities as the atomic numbers of the source element increasingly differ from that of Argon. The boundary of the peak is defined at the Z number where they are no longer the dominant nuclides in the radioactive inventory. In the case of $Ar42$, this happens in the inventory $Z = 25$ (Mn), where the dominant nuclide becomes $Mn53$ ($t_{1/2}=3.74 \times 10^6 \text{years}$). It is noted that at low atomic numbers, where the principal activation chains proceed by reactions

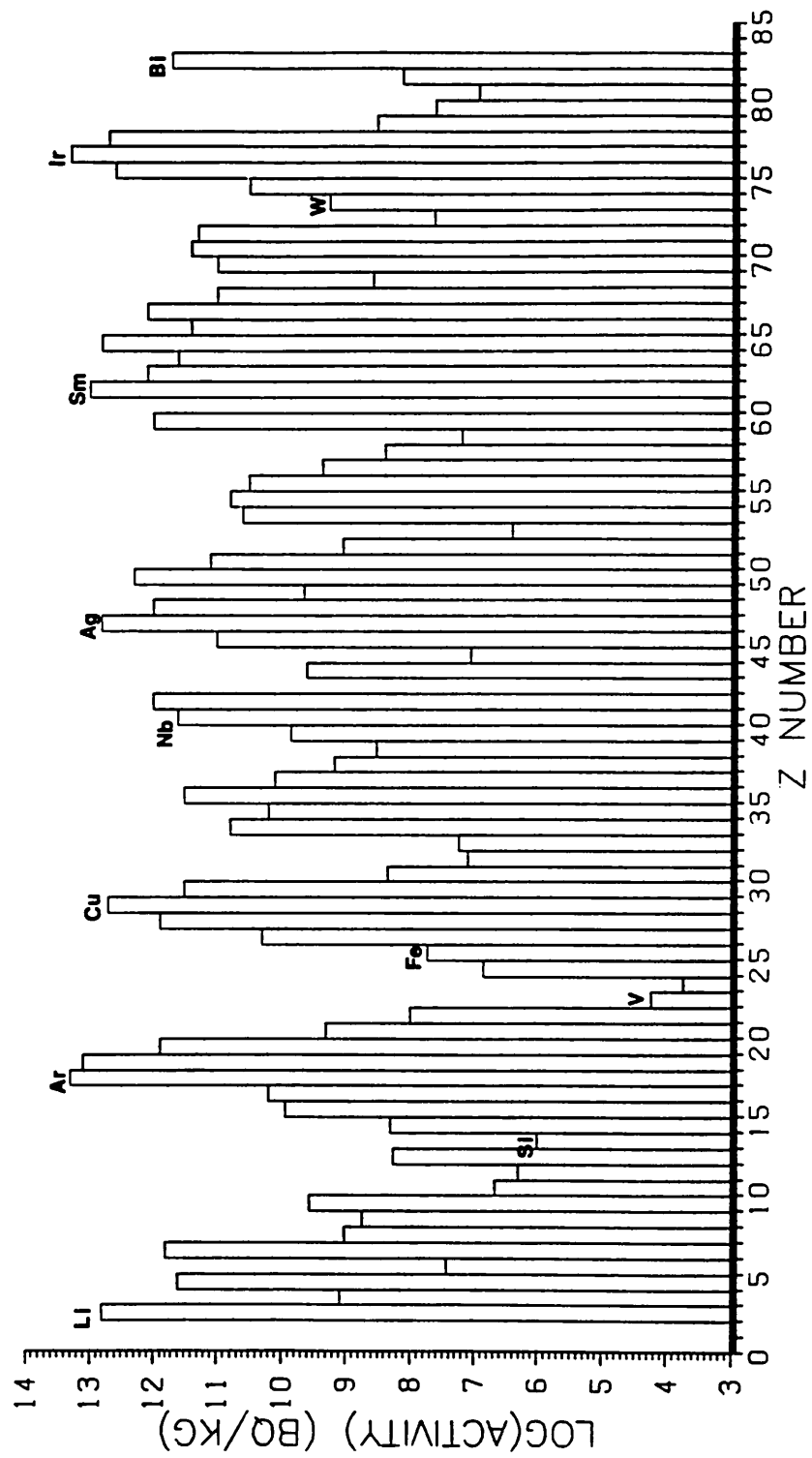


Figure 1.3 : Histogram of specific activities (Bq/Kg) of pure elements in the first wall after a 100 year decay period

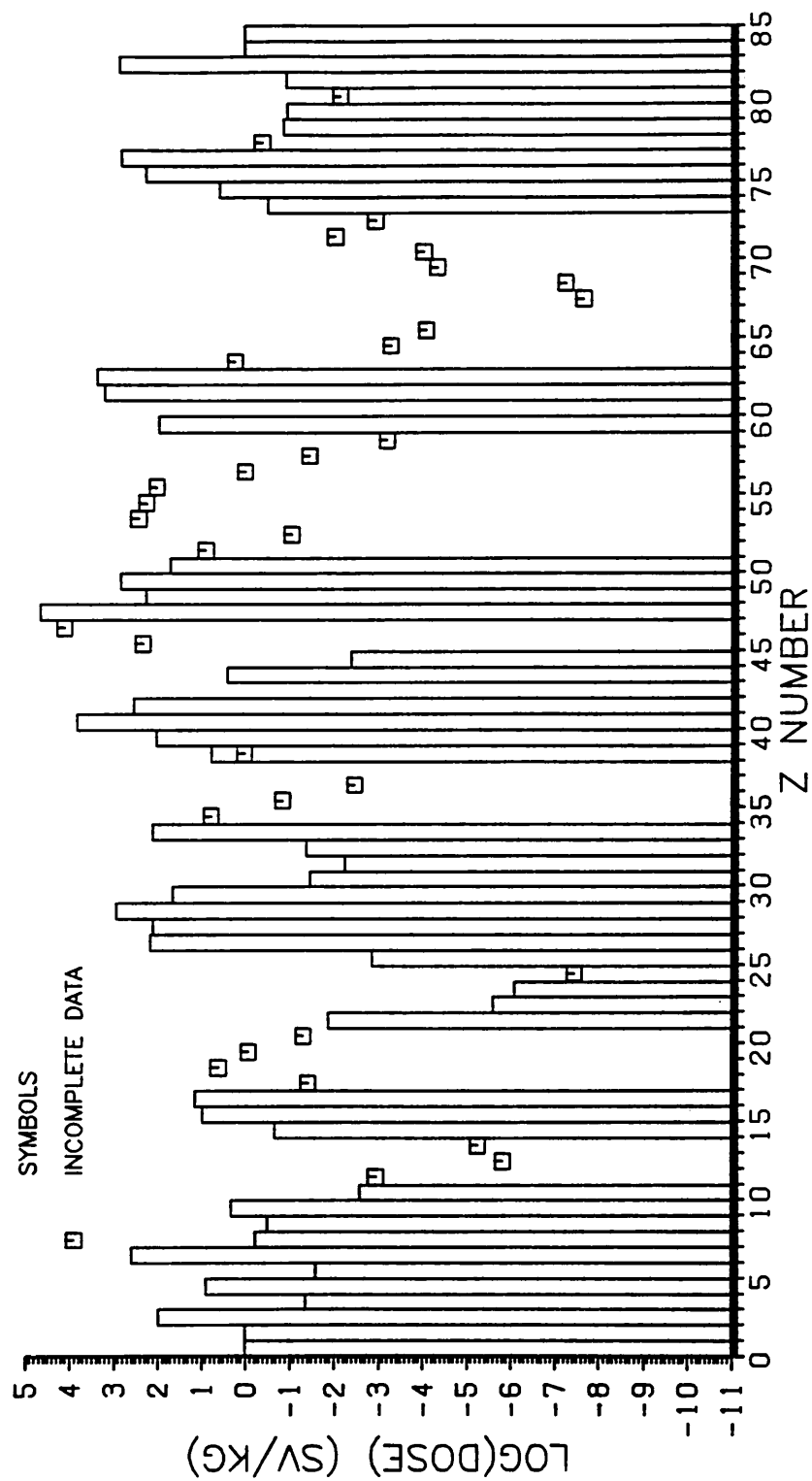


Figure 1.4 : Histogram of the potential ingestion doses (Sv/kg) for pure elements in the first wall after a 100 year decay period

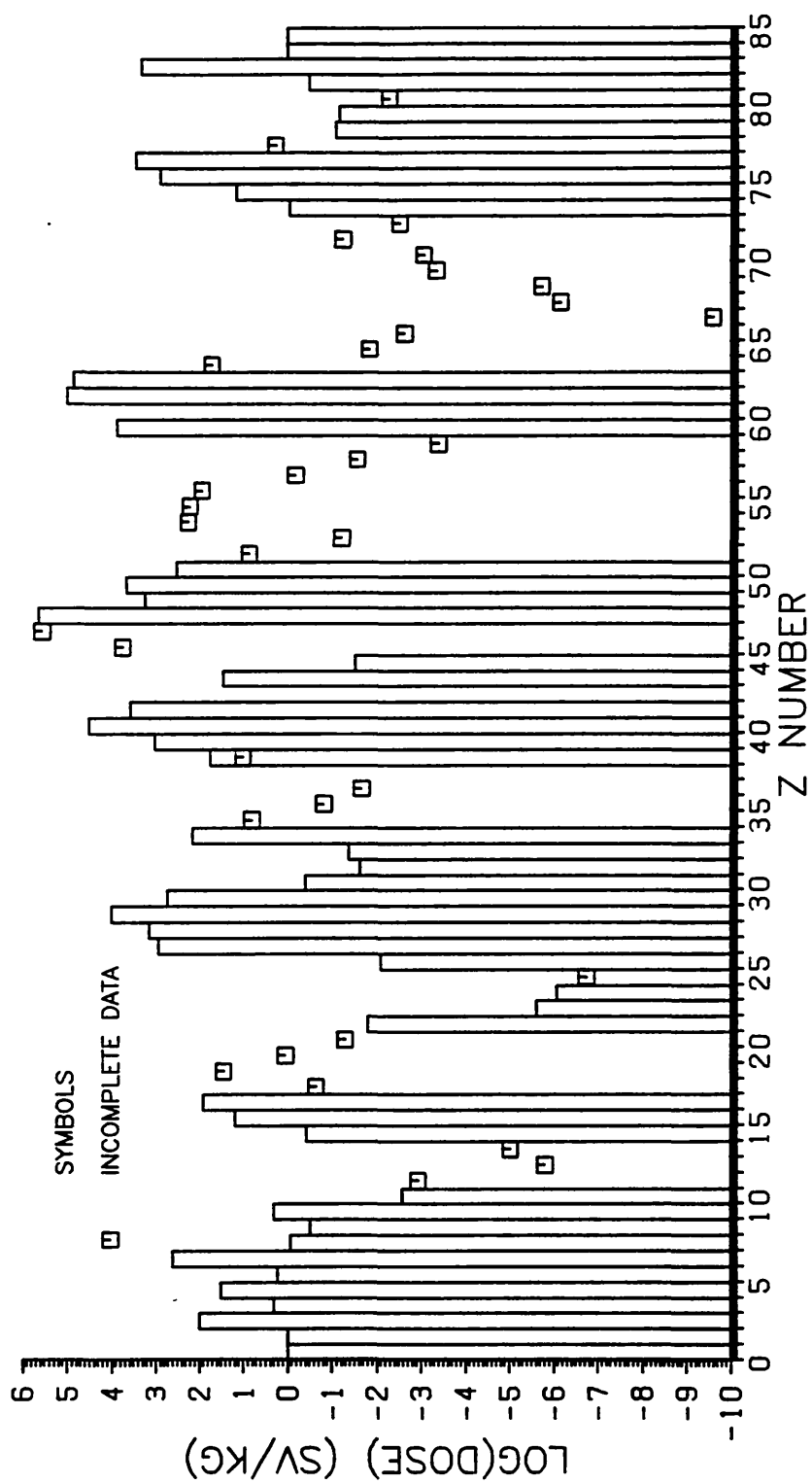


Figure 1.5 : Histogram of potential inhalation doses (Sv/kg) for pure elements in the first wall after a 100 year decay period

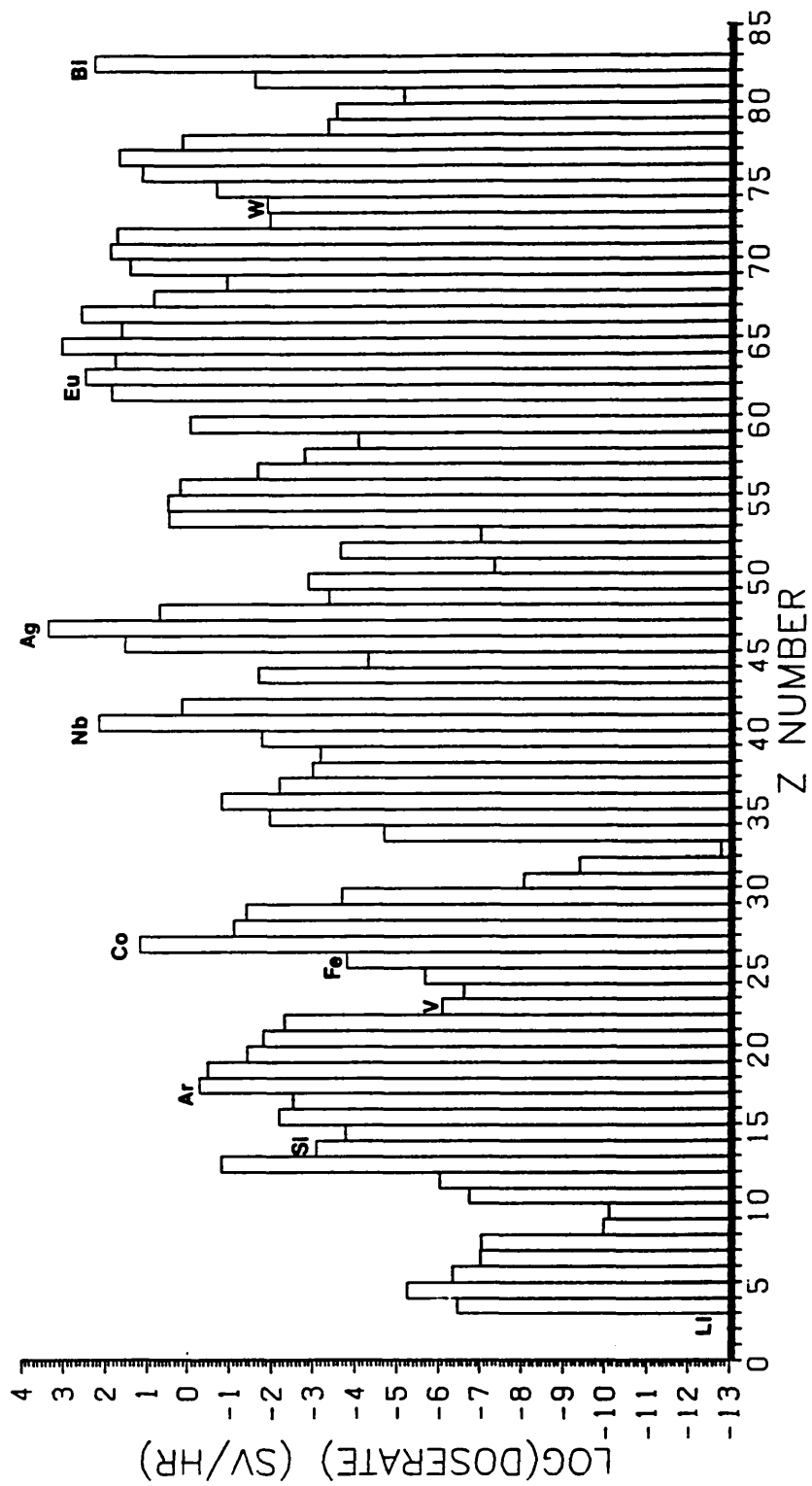


Figure 1.6 : Histogram of surface gamma dose rate (Sv/hr) for pure elements after a 100 year decay period

involving particle emission, the peaks are asymmetric, falling more rapidly on the low Z side. In contrast at high atomic numbers, where the principal chains proceed by capture reactions, the asymmetry is reversed.

The equivalent histograms showing potential ingestion and inhalation doses plotted against atomic number, figures 1.4 and 1.5, exhibit the same overall characteristics as the plot for activity. The differences are differences of scale rather than differences in essential characteristics. Spurious differences occur where CEDE data are unavailable for some inventory nuclides ; the inventories affected have been marked accordingly.

The histogram for surface gamma dose rate, figure 1.6, differs more substantially from the corresponding histogram for specific activity : some peaks in activity are removed or greatly reduced, since not all radioisotopes emit gamma rays above 100keV. This applies particularly to the elements lighter than Na, for which there are no long-lived gamma-emitting isotopes, the peak at $Z = 5$, which appears to be an exception, being associated with the bremsstrahlung emitting nuclide Be10 ($t_{1/2}=1.62 \times 10^6$ years).

Surface gamma dose rate is perhaps the most useful of the four hazard indices, as it can be applied more or less directly to handling and post-disposal intrusion scenarios. In order to facilitate a comparison with the activation levels found by previous workers, the elemental surface gamma dose rates predicted here have been expressed as maximum permissible fractions in table 1.1, beside those of Jarvis ⁷, Giancarli ¹⁰ and Ponti ¹⁴. In this context, the maximum permissible concentration (MPC) is defined as the percentage weight fraction of an element in an iron-based first wall which alone would

Table 1.1 : Maximum permissible concentrations of first wall elements according to 25 μ Sv/h limit on surface gamma dose rate.

Element	Present	Giancarli ¹⁰	Jarvis ⁷	Ponti ¹⁴
Li	100%	-	-	-
Be	100%	-	-	-
B	100%	-	-	-
C	100%	-	-	-
N	100%	-	-	-
O	100%	-	-	-
F	100%	-	-	-
Ne	100%	-	-	-
Na	100%	-	-	-
Mg	100%	-	-	-
Al	170ppm	46ppm	30ppm	100-500ppm
Si	6.0%	71%	20%	20%
P	15%	100%	100%	100%
S	0.38%	100%	100%	100%
Cl	0.85%	0.86%	0.1-1.0%	5%
Ar	480ppm	27ppm	-	50ppm
K	76ppm	30ppm	40%	100ppm
Ca	670ppm	0.4%	5%	0.1-1.0%
Sc	0.16%	0.07%	0.03%	2%
Ti	0.51%	0.26%	0.2%	5-10%
V	100%	100%	100%	100%
Cr	100%	100%	100%	100%
Mn	100%	100%	100%	100%
Fe	16%	19%	100%	100%
Co	1.6ppm	2.5ppm	0.1%	5%
Ni	0.03%	0.04%	1%	0.1%
Cu	0.06%	0.08%	1%	0.1%
Zn	12%	16%	5%	50%
Ga	100%	100%	100%	100%
Ge	100%	100%	100%	100%
As	100%	100%	100%	100%
Se	100%	100%	100%	100%
Br	0.2%	2.8%	100%	5%
Kr	166ppm	96ppm	-	0.1%
Rb	0.4%	1.4%	-	1%
Sr	2.5%	100%	-	5%
Y	3.7%	100%	100%	100%
Zr	0.14%	0.6%	0.3%	1-5%

Element	Present	Giancarli ¹⁰	Jarvis ⁷	Ponti ¹⁴
Nb	0.18ppm	0.2ppm	1.0ppm	1.0ppm
Mo	18ppm	120ppm	100ppm	50ppm
Ru	0.12%	1.6%	-	1-5%
Rh	51%	89%	-	100%
Pd	7.4ppm	5.0ppm	-	10-50ppm
Ag	0.011ppm	0.008ppm	0.01ppm	0.01ppm
Cd	5.0ppm	3.7ppm	10ppm	20ppm
In	5.7%	5.1%	1%	100%
Sn	1.9%	0.6%	1%	10-50%
Sb	100%	100%	100%	0.1%
Te	11%	100%	100%	5%
I	100%	100%	100%	100%
Xe	8.6ppm	9.3%	-	20%
Cs	85ppm	21ppm	-	100-500ppm
Ba	16ppm	17ppm	-	10-50ppm
La	0.12%	21%	-	1-5%
Ce	1.5%	19%	-	5%
Pr	29%	100%	-	100%
Nd	29ppm	61ppm	-	0.1-0.5%
Sm	0.33ppm	6ppm	-	10ppm
Eu	0.80ppm	0.005ppm	-	0.01ppm
Gd	4.1ppm	20ppm	-	1ppm
Tb	0.023ppm	0.005ppm	-	0.01ppm
Dy	0.62ppm	7ppm	-	10-50ppm
Ho	0.067%	0.03ppm	-	0.05ppm
Er	3.7ppm	33ppm	-	1-10ppm
Tm	0.022%	0.006%	-	0.01%
Yb	0.97ppm	0.5%	-	0.5%
Lu	0.33ppm	0.13%	-	1ppm
Hf	0.46ppm	5.0ppm	-	1ppm
Ta	0.23%	0.9%	100%	0.005%
W	0.19%	7.1%	100%	0.5%
Re	0.012%	0.07%	-	0.05%
Os	1.9ppm	0.13ppm	-	1-5ppm
Ir	0.51ppm	0.025ppm	-	0.05%
Pt	18ppm	81ppm	-	50ppm
Au	5.7%	100%	-	100%
Hg	9.0%	100%	-	100%
Tl	100%	100%	100%	100%
Pb	0.099%	0.08%	100%	1-5%
Bi	0.14ppm	0.11ppm	-	1ppm

give rise to a surface gamma dose rate of $25\mu\text{Sv/h}^{10}$. Although somewhat artificial and against the trend for dosimetric assessments for specific hazard scenarios, MPC's nevertheless provide a rough and comparative guide as to the contributions of long-term gamma emissions from pure elements.

It is clear from a cursory examination of table 1.1 that the updated database employed in the present calculations has resulted in significant alterations to radionuclide inventories. The closest match occurs with the results reported in CLM-R275, which employed identical irradiation conditions, with the ORIGEN inventory code and a modified form of the UKCTR-IIIA database. A full discussion of reaction channels to important inventory nuclides and the critical cross-section data involved, follows in Chapter IV, but it is noted here that the maximum permissible concentrations of the common structural elements - Mn, Fe, Ni, Nb and Bi - have not changed significantly, by less than a factor of two in all of these cases, probably because the cross-sections in the UKCTR database for these elements are founded upon relatively well-established determinations.

For less common elements, including minor structural elements, impurities in steels, novel elements for low-activation materials and non-metallic elements, the maximum permissible concentrations have changed significantly in some cases. In this respect it is again noted that the lack of restrictions for the elements from Li to Mg (not treated by previous workers) is misleading, since these elements produce no long-lived gamma-emitting nuclides, but do have long-lived beta-emitting nuclides, notably Be^{10} and C^{14} , which would represent potential ingestion and inhalation hazards : a maximum permissible acceptability based upon specific ingestion or inhalation dose would

undoubtedly limit the levels of Be and N in a first wall.

Of especial interest are changes in the predicted activation behaviour of elements in the low-activation windows which have been identified as being potentially suitable for alloy development. These will be dealt with individually.

1.1.6.1 Si and Al

The gamma doserates for Si and Al have increased by a factor of 12 and decreased by a factor of 4 relative to Giancarli's figures respectively. This means that Si would be allowed at a concentration up to about 6%, and Al at an impurity level of up to about 170ppm. The potential ingestion and inhalation doses are uncertain, since CEDE data are unavailable for Al26.

1.1.6.2 Ti, V, Cr, Mn and Fe

The concentration limit of Ti has approximately doubled to 0.51%, which places it among elements which are marginally acceptable as minor constituents. Interestingly, approximately 10% of the gamma doserate arises from bremsstrahlung emissions. It is unlikely that further improvements to the cross-section database will alter the maximum permissible concentration by very much ³⁰ (say a factor of 2), but the picture may change dramatically when more becomes known about the behaviour of the main long-lived radionuclide products Ar42 and Ar39 in potential hazard scenarios. Unavailability of CEDE's for these nuclides is responsible for the uncertainties in potential inhalation and ingestion doses. If, as seems likely in recycling scenarios, the argon isotopes are removed, the prospects for Ti improve appreciably, as the dominant inventory nuclide will then become Ti44, with substantial reductions in all four hazard indices.

Also in the same low-activation window as Ti are V, Cr, Mn and

most importantly, Fe - the intrinsically low activation of Fe underpinning the concept of low-activation steels. In line with CLM-R275, the activation properties of V, Cr and Mn appear to be exceptionally good with respect to all four hazard indices, and their permissible concentrations are unrestricted.

1.1.6.3 Mo

The acceptability of Mo has been reduced by approximately a factor of 7 to 18ppm. This would make it a potentially troublesome impurity in compositions which fall within the 25 μ Sv/hr limit, as the currently estimated impurity level in steels is 50ppm (see Chapter V). The potential ingestion and inhalation doses from Mo are also relatively high.

1.1.6.4 Ta and W

The acceptabilities of Ta and W have decreased significantly from the figures given by CLM-R275 to 0.23% and 0.19% respectively. There are good reasons to suspect that the doserates are overestimated in the predictions from UKACT1, however (see Chapter IV). Calculated values for potential ingestion and inhalation doses are uncertain due to the absence of hazard data for Hf178n and Ir192n.

Other low-activation windows open at Z numbers for which materials properties are either unsuitable or little known : it may be possible that some of these materials will be incorporated in the next generation of first wall compositions or for other plasma chamber applications. The most prominent of these windows in the surface gamma doserate histogram are found about Z = 10 (Mg), Z = 33 (As), Z = 53 (I) and Z = 81 (Tl).

For the bulk of the other elements, where differences with respect to calculations performed with the UKCTR-III database (Giancarli ¹⁰ and Jarvis ⁷) occur, the overall effect of the improved database appears

to be an increase in activation levels originating mostly from the opening up of new reaction pathways. This is particularly noticeable at large atomic numbers where the capture reaction has been found to be more important than hitherto believed and the treatment of isomeric states has been improved. Other changes have resulted from the adoption of improved systematics and the treatment of bremsstrahlung isotopes.

With regard to the Ponti ¹⁴ data, the marked disparities are unexpected, since the database employed by him is, like UKACT1, based upon the REAC database. It is, however, significant that major differences appear for the elements Fe, Co and Ti, for which sound data exist. This would point to the calculational method rather than the database as the source of the disparities. The major disparities are in fact traceable to the differences in the irradiation conditions employed, which in the case of Ponti's calculations were those appropriate for the first wall in the NET ²⁴ design, and later scaled up to a cumulative wall-loading of 10MWyrs/m². It is relevant to note here that a simple scaling-up procedure further assumes that activation inventory concentrations vary linearly with fluence, whereas a simplified activation chain model (ChapterIII) shows that activation varies with the n^{th} power of fluence where n is the length of the activation chain. The effect of assuming a linear relationship would be to underestimate inventory concentration and underestimate doserates, which appears to be the case for Ponti's treatment of Ti and Fe. Other source of differences could be the inclusion of X-rays with energy of less than 100keV by Ponti and the treatment of bremsstrahlung. Differences for elements for which capture cross-sections are large, such as Eu, arise from a reevaluation of some

capture cross-sections in the UKACT1 database.

1.2 First Wall Reaction Rates

It is important to remember that the figures given for pure element activation in section 1.1.6 have been calculated for a specific site (the outer first wall position) in one particular fusion reactor design (the Culham conceptual DEMO tokamak ²³). The question which then arises is : to what extent are these findings applicable to other parts of the tokamak and to other tokamak designs ? In an attempt to shed some light on this matter, and in the process understand which parts of the neutron energy spectrum are responsible for the reactions, energy-dependant reaction rates of threshold and capture reactions were evaluated over the fusion energy range for power-normalised NET, DEMO and CCTR first wall spectra. The rates were evaluated in a pointwise manner in the GAM-II energy structure, and expressed in units of (lethargy⁻¹ s⁻¹ cm⁻²). So that a comparison might be made between the the typical reaction rates on heavy and light nuclei, the rates were evaluated for both Co59 and W186 nuclei.

It can be seen from figure 1.2 that the CCTR, DEMO and NET first wall neutron energy spectra are quite similar in the energy range for threshold reactions (approximately 1 - 14MeV), and consequently the rates of threshold type reactions should be similar. This is confirmed by figures 1.7 and 1.8, showing the rates of Co59(n,p) and W186(n,2n) reactions for the three spectra. The greatest spread occurs in the trough between the 14MeV spike and the backscattering peak, where the DEMO spectrum is lower than the others by about a factor of 2. In the highest yielding 14MeV energy group, the difference is less. It can

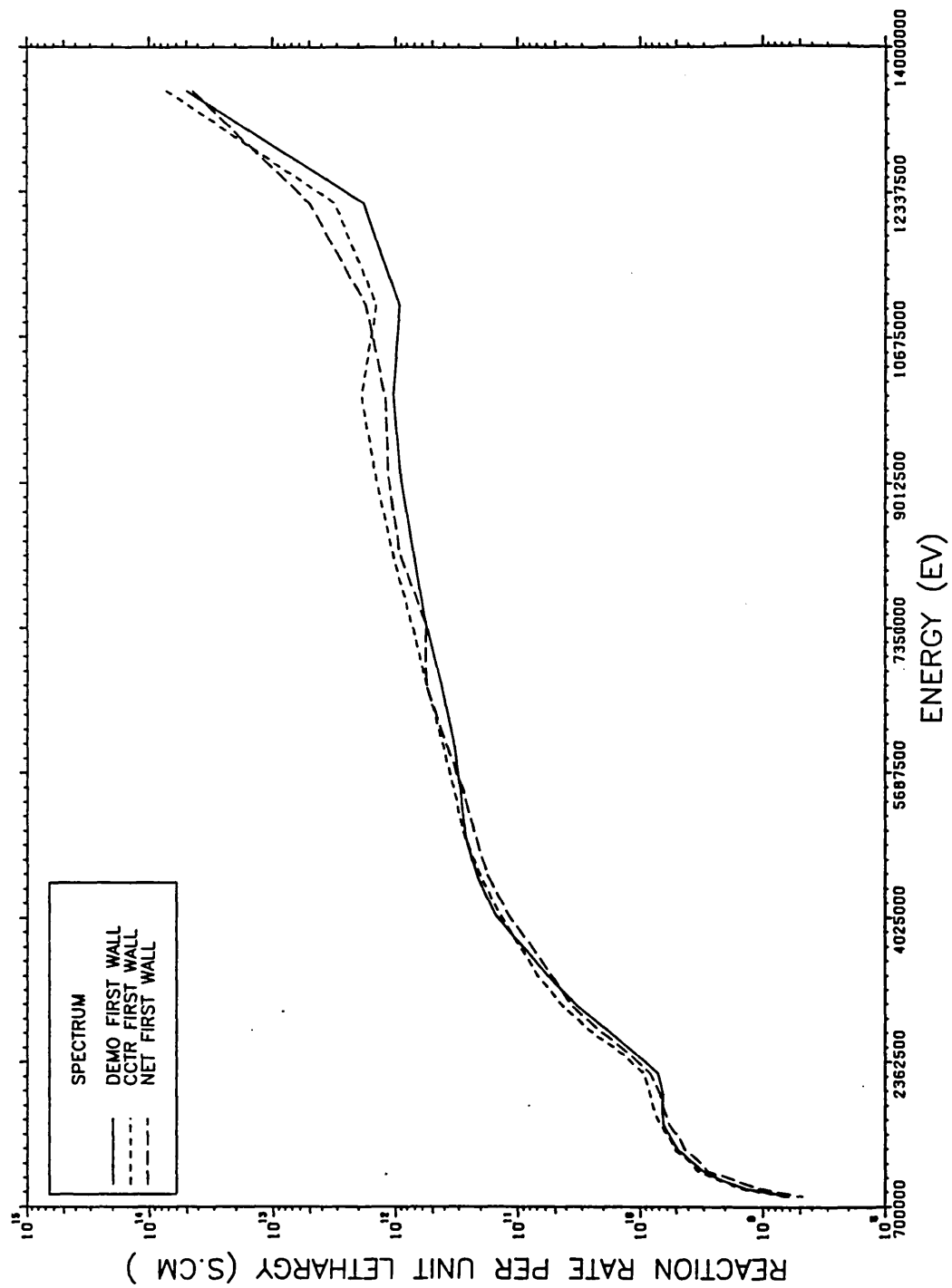


Figure 1.7 : Reaction rate per unit lethargy for the Co59(n,p) reaction

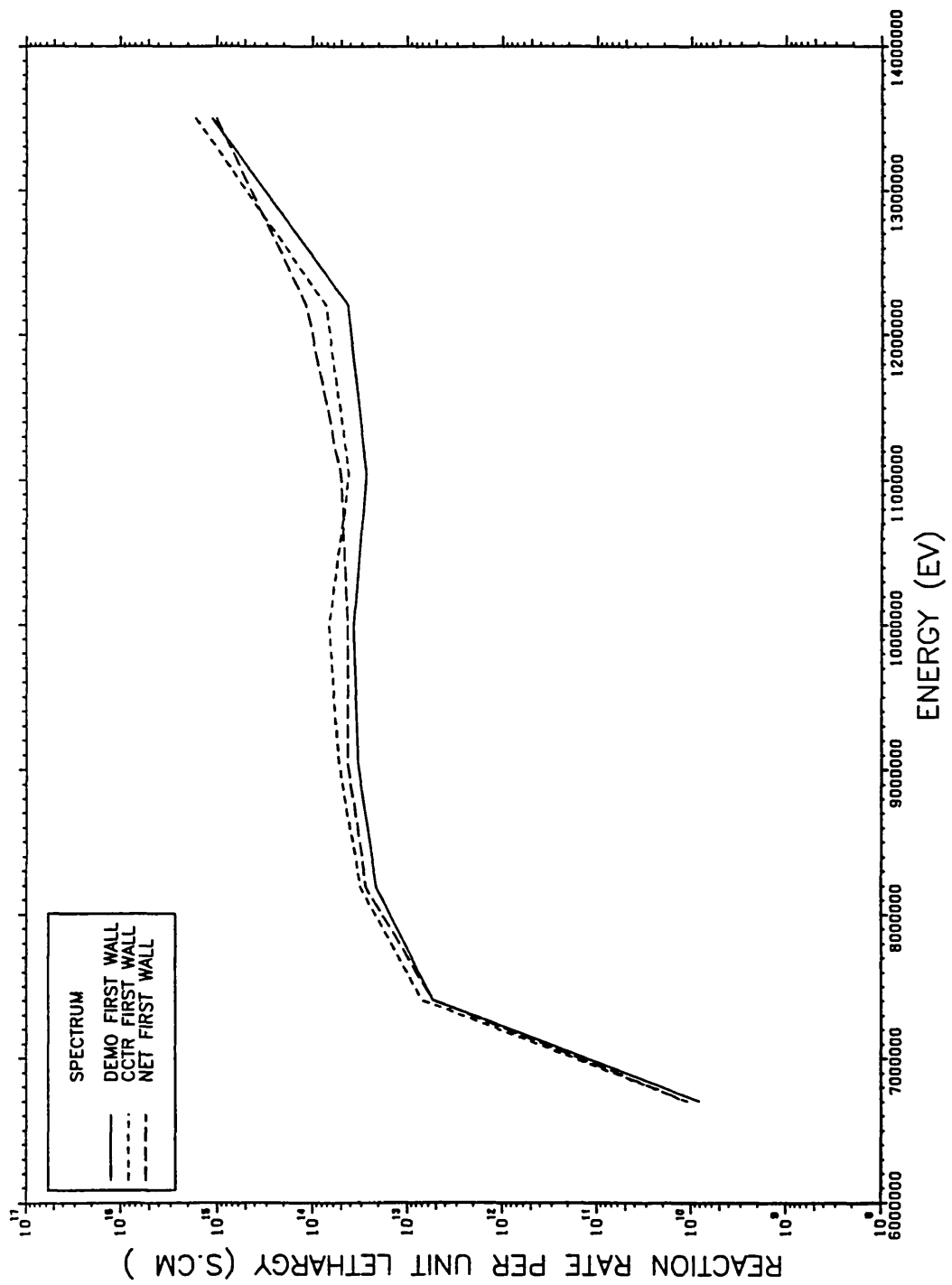


Figure 1.8 : Reaction rate per unit lethargy for W186(n2n) reaction

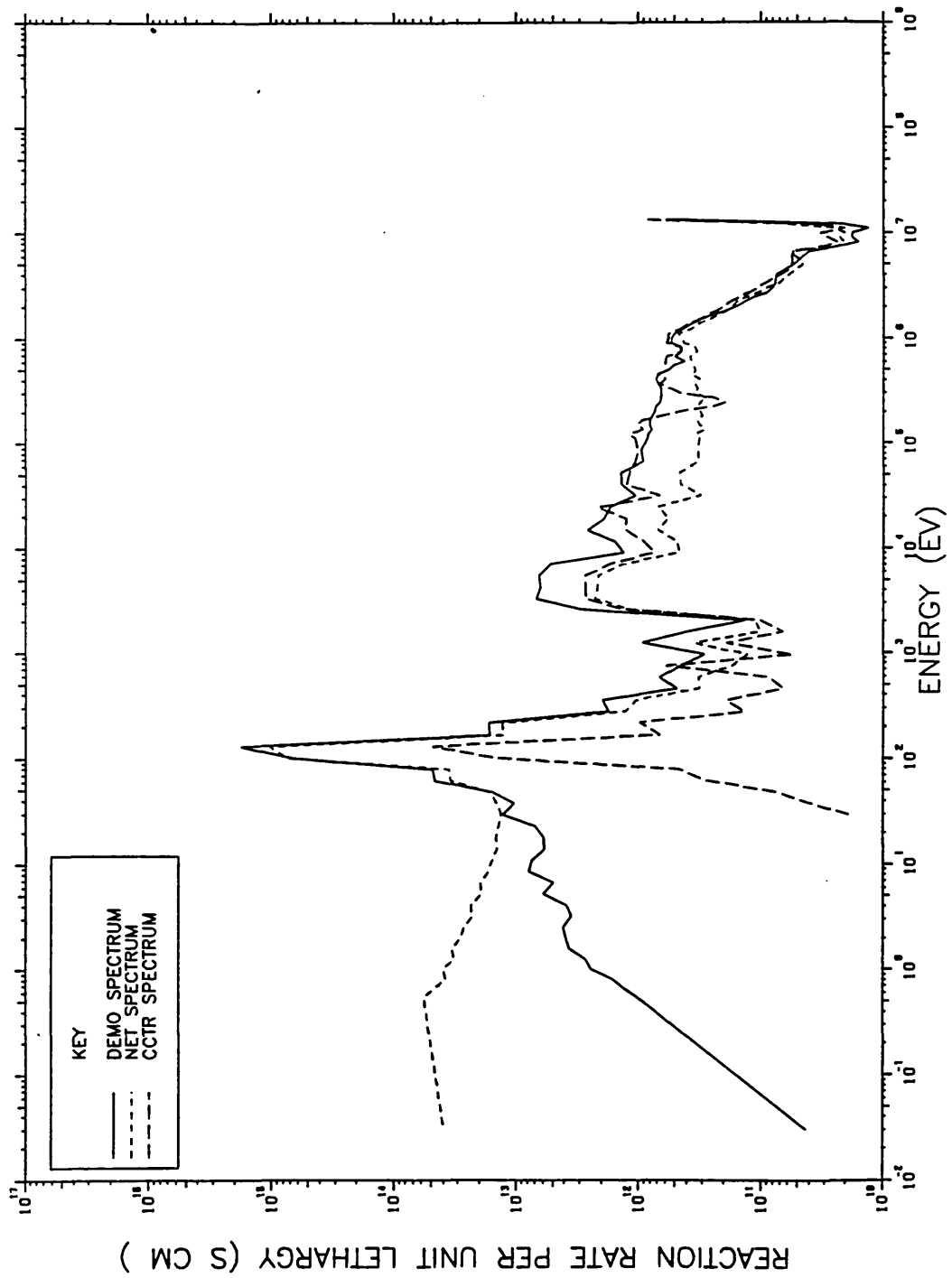


Figure 1.9 : Reaction rate per unit lethargy of $\text{Co}59(n,\gamma)$ reaction

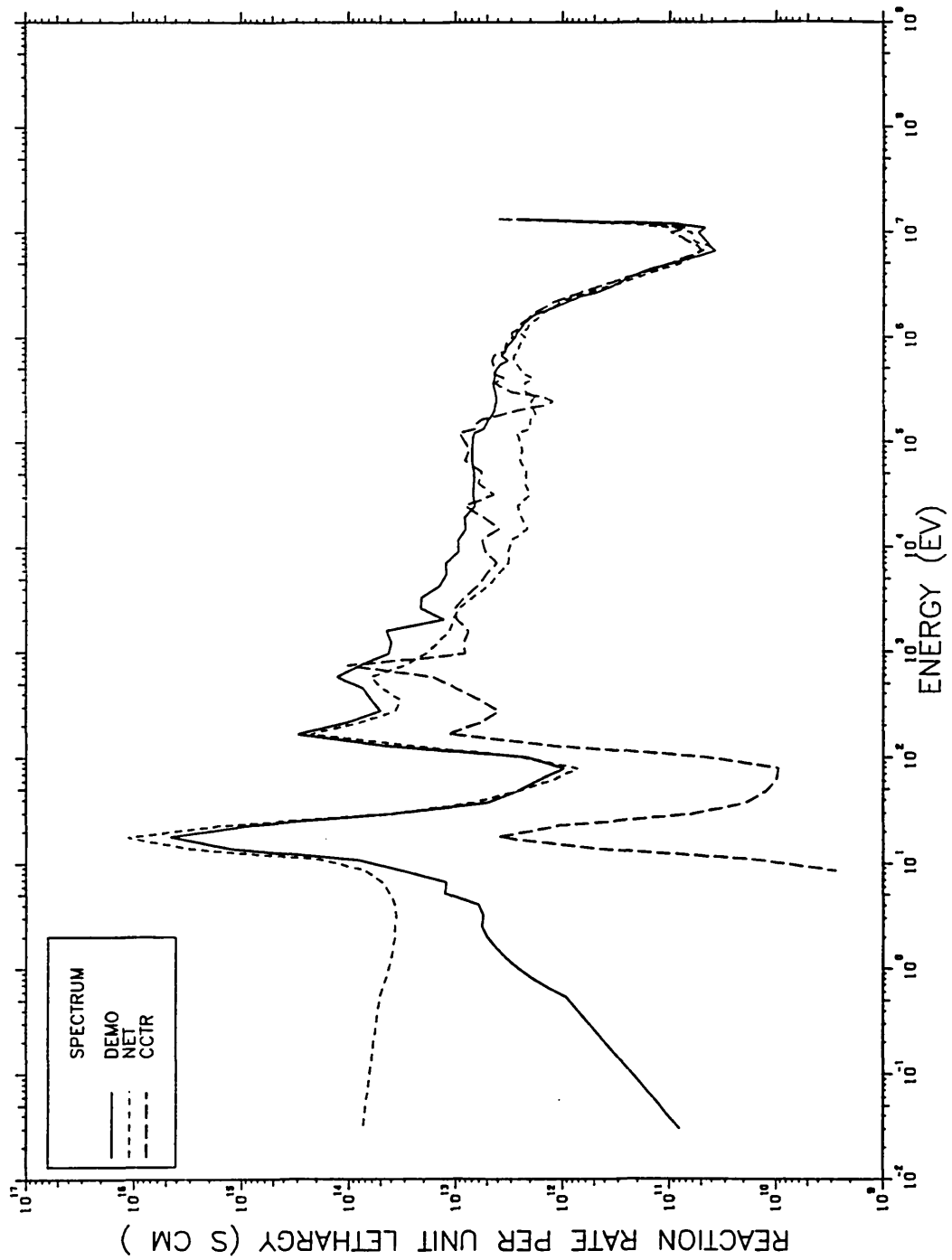


Figure 1.10 : Reaction rate per unit lethargy of $W186(n,\gamma)$ reaction

therefore be concluded that first wall activation rates by threshold reactions are independent of reactor design if the wall power loading is the same.

It can be seen from figures 1.9 and 1.10 that the rates of the capture reactions on Co59 and W186 peak in the resonance region and, unlike the rate for a threshold type reaction, the role of neutrons with energy greater than 1MeV appears to be insignificant. For the two nuclei examined here the effect of the contribution from the thermal group is small too, but it is felt that given the broad range of cross-sections for the thermal group, that it may be significant for a small number of nuclei with very large thermal cross-sections. The important parts of the three first wall neutron energy spectra are therefore the resonance and thermal regions, and it is interesting to note that the three spectra are similar at energies above about 1keV, but below this they begin to diverge, with first the hard CCTR spectrum falling away rapidly and later, at around 10eV, the DEMO spectrum falling away from the NET spectrum. The effect this has on the rates of the capture reactions on Co59 and W186, as can be seen in figures 1.9 and 1.10, is to reduce the total reaction rate in the CCTR design with respect to DEMO and NET, which have similar rates. If, however, the target nuclei were to have resonances below 10eV or very large thermal cross-sections, the reaction rate in a NET first wall could be substantially greater than that for a DEMO first wall. Fortunately such nuclei are rare, leading to the conclusion that activation rates in a NET first wall will be similar to a DEMO first wall, all other things being equal, and the findings of this study with regard to material activation properties can be applied to other conventional tokamak designs, if a necessarily cautious approach is taken.

1.3 Conclusion

There are considerable variations between the long-term activation properties of elements. The level of activation varies with atomic number in an apparently periodic fashion, the peaks in activation corresponding to the presence of particular long-lived nuclei. In between peaks are low-activation windows which may be suitable for first wall materials development, the ones occurring about Si ($Z = 14$), Cr ($Z = 24$) and W ($Z = 74$) being particularly suitable for exploitation in low-activation first wall alloys.

Improvement of the activation database has resulted in some significant changes to the predicted inventories of some elements. The changes are restricted, however, mainly to minor constituents in candidate first wall materials and impurities. Notable exceptions are reductions in the MPC's for Si, Mo, Ta and W, and increases for Al and Ti relative to predictions from the UKCTR-III database.

Gamma doserates for the elements lying between Li and Mg, elements not treated by the previous workers considered here, were found to be low, with the main source of gamma radiation being bremsstrahlung. Consequently MPC's for these elements were 100%. In contrast, the potential specific ingestion and inhalation doses from these elements can be high, and would almost certainly be significant in some potential hazard scenarios. For most of the other stable elements, potential specific ingestion and inhalation dose levels generally concur with gamma doserates.

On the whole, the level of activation is higher than that given by Giancarli, owing to the inclusion in the database of new reaction pathways to long-lived products - especially capture reactions on heavy

nuclei, some of which result in high burn-up rates (e.g. in excess of 90% in Eu). Where activation levels have been reduced, the principal cause is the renormalisation of cross-sections according to improved systematics.

Finally, the rates of threshold reactions in a first wall appear to be design-independent at standardised wall power loading, whereas the rates of capture reactions are mildly sensitive. Rates of reactions of the latter type peak in the resonance energy region, with the thermal region possibly becoming marginally significant for nuclei with large thermal capture cross-sections.

CHAPTER II

The Sensitivity Method

*When Lil's husband got demobbed, I said
 I didn't mince my words, I said to her myself,
 HURRY UP PLEASE ITS TIME
 Now Albert's coming back, make yourself a bit smart.
 He'll want to know what you done with that money he
 gave you
 To get yourself some teeth. He did, I was there.
 You have them all out, Lil, and get a nice set,
 He said, I swear, I can't to look at you.*

- A Game of Chess

2.1 Introduction

An analysis of the sensitivity of calculated nuclide concentrations to cross-section data would be valuable in a number of ways. Firstly, the items of cross-section data which are important in a calculation may be identified. From them the routes by which nuclides are produced may be delineated. Later cross-section determination efforts can then be concentrated on a small number of critical reactions. Secondly, a treatment of the propagation of standard errors from data becomes possible, and the accuracy of inventory concentrations estimated.

The "sensitivity" of nuclide concentration N_i to an item of data x_j can be expressed quantitatively by the sensitivity coefficient S_{ij} , defined as

$$S_{ij} = (\partial N_i / \partial x_j) (N_i / x_j)^{-1}$$

Clearly if the functional form of $N_i(x_j)$ is known, S_{ij} can be obtained directly by differentiation. In the fusion activation case considered here, where the N_i are obtained by a numerical method, the sensitivity coefficients must also be evaluated by numerical means, the method employed here being that of James ³¹.

It is observed that the set of simultaneous inventory equations set up in Chapter I

$$\frac{dN_i}{dt} = -\left(\lambda_i + \sigma_i \phi\right)N_i + \sum_{j=1}^n \left(b_{ji}\lambda_i + \sigma_{ji}\phi\right)N_j + T_i$$

can, in matrix form, be represented by

$$dN/dt = AN + T \quad (2.1)$$

If this were differentiated with respect to data item x and the order of differentiation changed, a set of simultaneous equation solvable for $\partial N/\partial x$ would be obtained :

$$d(\partial N/\partial x)/dt = -A(\partial N/\partial x) + (\partial A/\partial x)N + \partial T/\partial x \quad (2.2)$$

If a new source term T^* is now defined as

$$T^* = (\partial A/\partial x)N + \partial T/\partial x \quad (2.3)$$

then it is seen that this set of equations for $\partial N/\partial x$ has a similar form to the inventory equations with $\partial N/\partial x$ substituted for N and T^* for T :

$$d(\partial N/\partial x)/dt = -A(\partial N/\partial x) + T^*$$

(2.4)

Like the inventory equations, they too are a first order, stiff set of equations. If a new set of boundary conditions are employed, therefore, the method of solution used for the inventory equations may be used again here, a great advantage in practical terms since an inventory code may be extended to evaluate sensitivity coefficients simply by making appropriate substitutions in the numerical integration. The boundary conditions employed for the evaluation of sensitivity coefficients with respect to cross-sections σ_j are

$$\partial N_i/\partial \sigma_j = 0 \text{ at } t = 0$$

2.2 The source term

The functional form of the source term T depends upon the data item x , and may be obtained by evaluating (2.3). Thus for cross-section data, T takes the following forms

$$\begin{aligned} x = \sigma_i & \quad T^* = -\phi N_i \\ x = \sigma_{ji} & \quad T^* = \phi N_j \\ x = \sigma_k \text{ where } \sigma_k \neq \sigma_i, \sigma_{ji} & \quad T^* = 0 \end{aligned}$$

Similarly, T^* may be obtained for decay constants :

$$x = \lambda_i \quad T^* = -N_i$$

$$x = \lambda_{ji} \quad T^* = b_{ij}N_j$$

Source terms can also be obtained for the other data used in activation calculations, including branching ratios, hazard data and fluxes. In the fusion first wall case, however, there is little interest in setting up these since errors in the cross-section data dominate ^{7,10}. Errors introduced by fission branching ratios and flux data are more important in the more widespread application of the inventory equations to the burn-up of actinides, and the forms of the source terms for these and other data have been obtained by James ³¹.

The discussion has thus far been restricted to the types of nuclear data which are present in the activation data libraries. Yet it has been noted by Jarvis ⁵ and others that the result of an activation calculation depends also upon the database used in the preceding neutron transport calculation, working through the 100 group neutron energy spectrum obtained from the latter. It is not difficult to see how the "sensitivity" of the activation calculation to the transport data might be formulated. First, consider the inventory equations expressed in the 100 group form :

$$\frac{dN_i}{dt} = - \left(\lambda_i + \sum_{k=1}^{100} \sigma_{ik} \phi_k \right) N_i + \sum_{j=1}^{100} \left(b_{ji} \lambda_j + \sum_{k=1}^{100} \sigma_{jik} \phi_k \right) N_j + T_i$$

Let y represent a data item used in the neutronics calculation. Then differentiation with respect to y gives

$$\frac{dN_i}{dt dy} = - \left(\lambda_i + \sum_{k=1}^{100} \sigma_{ik} \phi_k \right) \frac{dN_i}{dy} + \sum_{j=1}^n \left(\lambda_{ji} + \sum_{k=1}^{100} \sigma_{jik} \phi_k \right) \frac{dN_j}{dy} + T_i \quad (2.5)$$

where $b_{ij} \lambda_j$ has been replaced by λ_{ji} , and it has been assumed that

$$\partial \lambda / \partial y = \partial \sigma / \partial y = 0$$

Then evaluation of the source term T , where

$$T_i = - \sum_{k=1}^{100} \sigma_{ik} \frac{\partial \phi_k}{\partial y} N_i + \sum_{j=1}^n \sum_{k=1}^{100} \sigma_{jik} \frac{\partial \phi_k}{\partial y} N_j + \frac{\partial T_i}{\partial y} \quad (2.6)$$

and substitution into (2.5) allows the $\partial N_i / \partial y$ to be calculated in the same manner as the $\partial N_i / \partial x$. It is assumed that the $\partial \phi_k / \partial y$ have been obtained by a previous sensitivity analysis conducted on the neutronics calculation. Though not carried out in this study, the application of this procedure would a suitable subject for a supplementary study.

2.3 The Computational Method

The above sensitivity method has been incorporated in the fusion activation code FISPACT, the general features of which have been discussed in the previous chapter. The implementation of the method was the subject of some experimentation : whereas the setting up of terms and their substitution into the integration subroutine was straight forward, the treatment of the source term proved problematic.

The difficulty lies in the dependance (demonstrated above) of the

source term on nuclide concentrations N_i and N_{ji} , as the inventory concentrations of all nuclides other than those at the head of activation chains change significantly over the irradiation period, the rate of change increasing with the length of the chain. Initially, the approach employed by James' Winfrith code FISPSENS was tried. This used a separate integrating routine for the sensitivity calculation which split the irradiation period into 3 subintervals. Constant source terms were taken over each interval, the concentrations interpolated from their values at the beginning and end of the irradiation period. It was found, however, that the interpolated values were not sufficiently accurate and consequently errors in the solution grew out of control with succeeding time steps.

The eventual approach recognises the essential similarity of the situation to a fuel burn-up calculation, where the source term has a flux-dependence which varies over the irradiation period. FISPIN6 performs burn-up calculations by splitting the irradiation period into a number of subintervals over which flux may be considered to be constant, the full integration performed for each subinterval. Likewise, in the present version of FISPACT the irradiation period is split into a user specified number of subintervals for which linked inventory and sensitivity inventories are performed. The source term in the sensitivity calculation is kept constant over each time subinterval, and is obtained from the concentrations given by the preceding inventory calculation. The overall scheme is shown in figure 2.1.

It is noted that each call of the integrating routine gives the set of differential sensitivities of all nuclide concentration with respect to data item x (that is, $\partial N_i / \partial x$, $i = 1, n$ where n is approximately 1300 for UKACT1). A separate call must therefore be made for each data item.

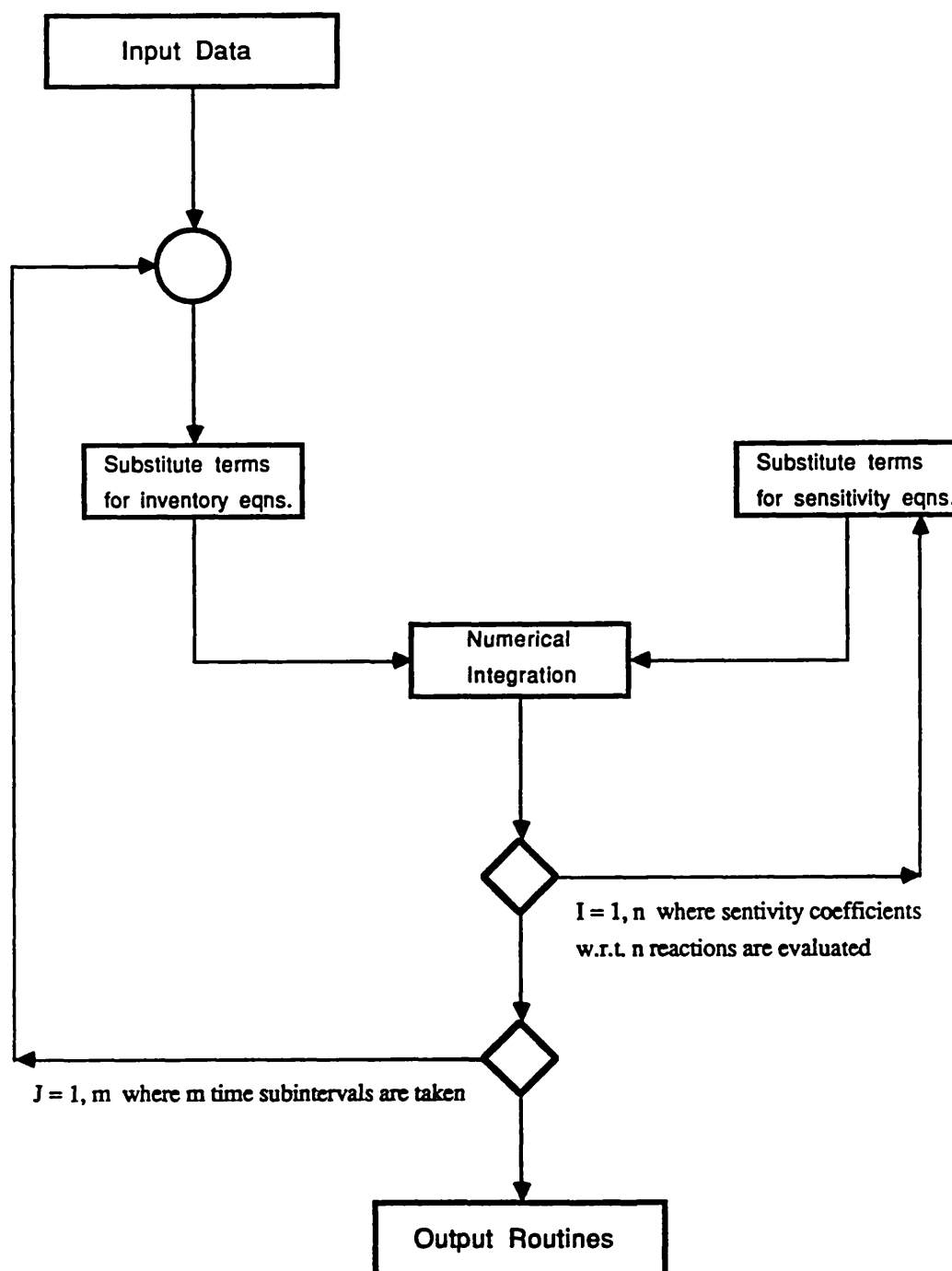


Figure 2.1 : Computational scheme employed in sensitivity module

Since the number of nuclides in the library is large , it is necessary to restrict calculations to those sensitivity equations involving user-specified data items. A direct limit is placed on the number of sets of sensitivity equations which may be calculated at one time by the size of the array which stores the differential sensitivities at the end of each time subinterval. At present the size of this array is 20,000 elements, allowing approximately 15 sets of equations to be solved simultaneously in calculations involving the full UKACT1 library.

2.4 Delineation of Reaction Production Channels

An unfortunate feature of most inventory codes which employ a simultaneous solution is that they provide an inventory of the nuclides at the end of the period of irradiation without any knowledge of the routes taken in their formation. Knowledge of the production channels of troublesome nuclides in the inventories of structural materials is necessary for improvements to be made to the cross-section database, which in turn will eventually lead to a higher accuracy for the inventory calculation. The sensitivity coefficients S_{ij} , of nuclide concentration with respect to cross-section data, provide a ready measure of the relative importances of cross-sections in relation to the production of a given nuclide. This may be appreciated if the physical meaning of the sensitivity coefficient is considered. The differential component $(\partial N_i / \partial \sigma_j)_t$ is obviously a time dependant quantity, related to the change in concentration N_i brought about by a change in the cross-section σ_j over the time interval 0 to t .

For small σ_j , $(\partial N_i / \partial \sigma_j)$ is a slowly varying function of σ_j , and to a first order approximation may be considered independent. Under this condition, $(\partial N_i / \partial \sigma_j) \sigma_j$ is then the change in concentration N_i which

N.B. The sensitivity coefficients in figure 2.2 are the sensitivities of the concentration of the end nuclide B with respect to the cross-sections for each step. That is

$$S_1 = \left(\frac{\partial N_B}{\partial \sigma_1} \right) \left(\frac{\sigma_1}{N_B} \right)$$

$$S_2 = \left(\frac{\partial N_B}{\partial \sigma_2} \right) \left(\frac{\sigma_2}{N_B} \right)$$

.....etc.

would be brought about by omitting the reaction which has cross-section σ_j from the inventory calculation. The sensitivity coefficient $(\partial N_i / \partial \sigma_j)(\sigma_j / N_i)$ therefore may be interpreted as the fraction of nuclide i which is transmuted by the reaction whose cross-section is σ_j . From this it follows that for a set of linear, independent activation chains, starting and finishing from the same nuclides, in the small cross-section assumption, the sum of the sensitivity coefficients for each reaction channel is unity. That is, say for the final reactions in the chains,

$$\sum_j (\partial N_i / \partial \sigma_{ji}) \approx 1$$

This can be seen more clearly by considering the idealised set of 3 activation chains effectively transmuting nuclide A to nuclide B below

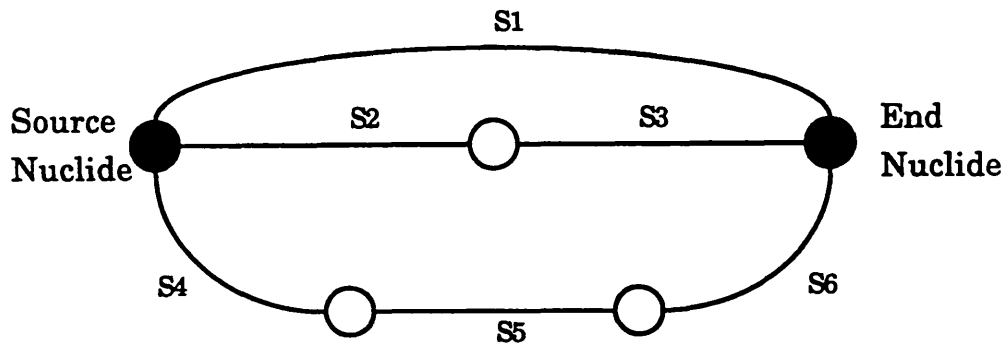


Figure 2.2 : Idealised example showing competing activation chains

where the nodes represent stages (i.e. intermediary nuclides) in each chain and the S's are the sensitivity coefficients associated with each

step.

It follows from the discussion that the following simple relationships exist between the sensitivity coefficients

- 1) $S_2 = S_3$
- 2) $S_4 = S_5 = S_6$
- 3) $S_1 + S_2 + S_4 \approx 1$

In practise, however, the interpretation of sensitivity coefficients is made more complicated by linkages between the activation chains, which would nullify 1) and 2). In general, relationship 3) still holds, except in cases of high burn-up (equivalent to the small cross-section assumption).

The effect of burn-up can be demonstrated by considering the simple example of a chain consisting of 2 nuclides, N_1 and N_2 , linked by a reaction with cross-section σ . The time dependence of concentrations N_1 and N_2 are obtained analytically in the textbook ² manner :

$$\frac{dN_1}{dt} = -\sigma \phi N_1$$

$$\frac{dN_2}{dt} = \sigma \phi N_1$$

$$N_1 = N_{10} e^{-\sigma \phi t}$$

$$N_2 = \sigma \phi N_{10} \int_0^t e^{-\sigma \phi \tau} d\tau$$

Now assume that at $t = 0$ $N_1 = N_{10}$, $N_2 = 0$

$$N_2 = N_{10} (1 - e^{-\sigma \phi t})$$

If the definition of the sensitivity coefficient is now applied to concentration N_2

$$S(N_2, \sigma) = \frac{dN_2}{d\sigma} \frac{\sigma}{N_2}$$

Substituting for the derivative

$$\frac{dN_2}{d\sigma} = N_{10} \phi t e^{-\sigma \phi t}$$

one obtains the following expression

$$S(N_2, \sigma) = \frac{x e^{-x}}{1 - e^{-x}}$$

where $x = \sigma \phi t$. If only low fluences and small cross-sections are considered, the exponential terms can be expanded to give the

approximation

$$S(N_2, \sigma) \approx \frac{x(1-x)}{1-(1-x)}$$

$$S(N_2, \sigma) \approx 1 - \sigma \phi t$$

Thus it can be seen that the sensitivity coefficient is a function of σ and fluence, which becomes independent of these variables as they tend to zero. Similarly, the argument can be extended to granddaughter and great granddaughter nuclides, with similar conclusions. An example of the effects of burn-up, taken from the activation of tungsten, is given in figure 2.3, showing the variation in the Os188 concentration with the cross-section for Re187(n, γ). It is seen that at small σ , the differential sensitivity (simply the first derivative of the curve shown) varies slowly with σ . As σ increases beyond a few barn, the variation becomes increasingly more significant.

If cross-sections or fluences are large, therefore, relationship 3) no longer holds. The values of the coefficients nonetheless provide an indication of the relative importance of competing production channels.

The procedure used to delineate production channels and identify critical cross-sections can be demonstrated by taking the activation of titanium as an example. The main source of gamma dose rate in the inventory produced from titanium after a 100 year cooling period is Ar42. In Chapter IV the sensitivity coefficients of reactions leading to its production are evaluated, and the dominant production channels are shown to be

(cont. p70)

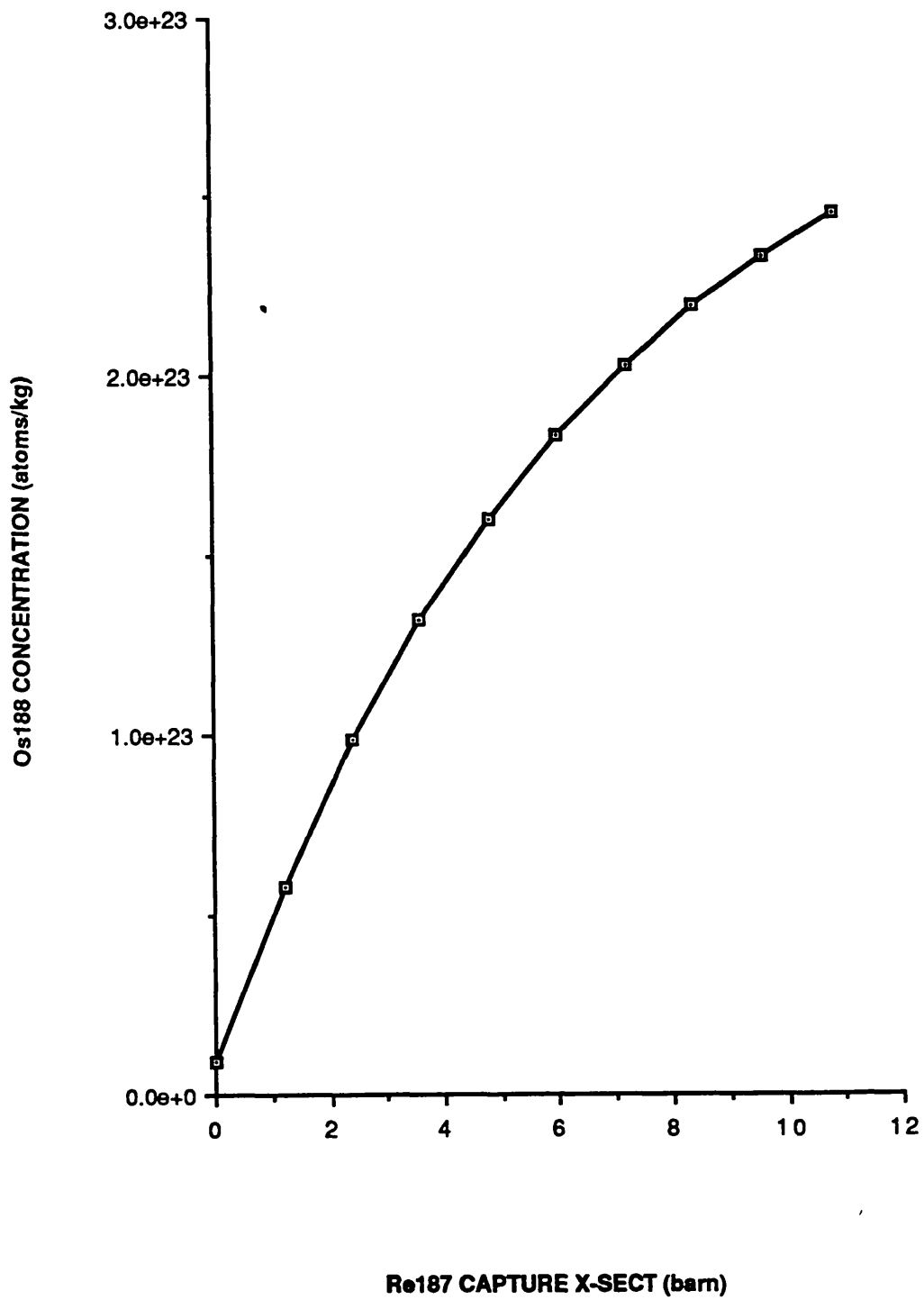


Figure 2.3 : Variation in the Os^{188} concentration with respect to the cross-section for $\text{Re}^{188}(\text{n},\gamma)$

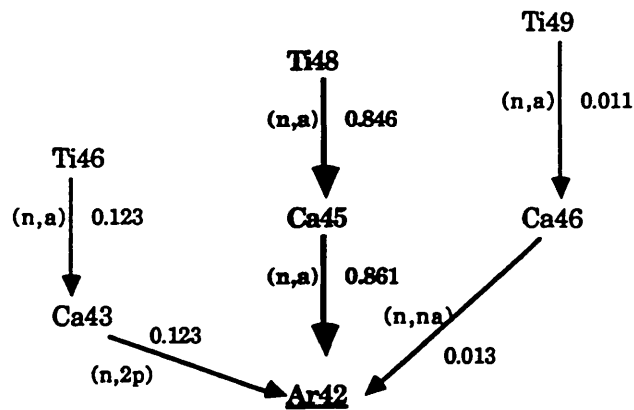


Figure 2.4 : Dominant production channels to Ar 42 from titanium

The case is similar to the idealised example given in figure 2.2 , save for the fact that the transmutation chains start from different source nuclei. This does not alter rule 3) however : it is seen that the sensitivity coefficients from equivalent stages of each chain add up to unity. It is important to stress, when looking at the sensitivity coefficients displayed in figure 2.4, that they do not express the absolute quantities of material transmuted in each step, but rather the fractional contributions to the nuclide N_i , and that only selected reactions have been shown. It is quite possible, therefore, for the coefficients for the coefficients of successive stages to alter as one proceeds down a chain.

The analysis can be taken further if the origins of the cross-sections for reactions shown in figure 2.4 are examined : it is found that the $\text{Ti48}(n,\alpha)$ cross-section has been experimentally measured and is known with some degree of certainty, whilst the $\text{Ca45}(n,\alpha)$ cross-section has been obtained from a nuclear model, as Ca45 is unstable. The value of the latter cross-section is therefore the most significant unknown in the calculation, and this item of data can therefore be

considered to be a "critical" cross-section.

2.5 The Propagation of Errors

In the standard treatment of medium and small errors, the perturbation propagated in nuclide concentration ΔN_i , by perturbations in items of data, $\Delta \sigma_j$, is given by

$$\Delta N_i = \sum_j (\partial N_i / \partial \sigma_j) \Delta \sigma_j \quad (2.7)$$

If sensitivity coefficient S_{ij} is then defined as

$$S_{ij} = (\partial N_i / \partial \sigma_j) (\sigma_j / N_i)$$

the perturbation in nuclide concentration becomes

$$\Delta N_i = \sum_j S_{ij} (N_i / \sigma_j) \Delta \sigma_j \quad (2.8)$$

It can be shown from this ³² that the variance in N_i is given by

$$\text{Var}(N_i) / N_i^2 = \sum_j S_{ij}^2 \text{Var}(\sigma_j) / \sigma_j^2 \quad (2.9)$$

Thus if the sensitivity coefficients are known and errors have been assigned to cross-sections, the resultant errors in nuclide concentration may be obtained. Likewise the errors in quantities which are functions of nuclide concentrations, such as doserates D , can be obtained by using expression (2.9) again with the appropriate substitutions

$$\text{Var}(D)/D^2 = \sum_i (\partial D/\partial N_i)^2 (N_i/D)^2 \text{Var}(N_i) / N_i^2 \quad (2.10)$$

2.6 Verification and Accuracy of Sensitivity Module

Ideally, it would be desirable to verify the computational method by testing it against analytical solutions to the same problems. Analytical solutions are, however, applicable only to simple chains which contain a small number of cross-sections. They do not realistically represent the type of complex activation chains which would be found in the fusion first wall. A study of the sensitivity equations shows that in any time sub-interval, the differential coefficient $\partial N_i/\partial \sigma_{ji}$ is a function of all other differential coefficients $\partial N_j/\partial \sigma_{ji}$. Therefore, though it may be possible to make such tests for simple activation chains, their actual value would be doubtful, since the accuracy of the computational method is clearly dependent on the number and complexity of the chains, and the particular data parameters required are defined by the size and nature of the UKACT1 library.

Instead, the more laborious method of Sowerby et al ³³ has been used to provide a standard. This, in essence, obtains differential sensitivities by changing a single cross-section σ_j , by amount $\Delta\sigma_j$, and measuring the resultant change in concentration N_i , ΔN_i . The sensitivity coefficient is then

$$S_{ij} = (\Delta N_i/\Delta\sigma_j)(\sigma_j/N_i) \quad \text{as } \Delta\sigma_j \rightarrow 0$$

For small σ_j , where $(\Delta N_i/\Delta\sigma_j)$ is a slowly varying function of σ_j , a single value of $\Delta\sigma_j$ may suffice, in the manner of Sowerby et al ³³. For large σ_j ,

however, the dependance is more marked and it is necessary to take the limit as $\Delta\sigma_j$ tends to 0 (see figure 2.2 for a typical example).

The procedure followed here was to evaluate N_i at 5 evenly spaced values of σ on each side of σ_j , from which were obtained 10 values for $(\Delta N_i / \Delta \sigma_j)$, $I_1, I_2, \dots, I_{10}$, by taking

$$I_1 = (N_{i1} - N_i) / (\sigma_1 - \sigma_j)$$

where (N_{i1}, σ_1) are the values of N_i and σ_1 at the first point, (N_{i2}, σ_2) at the second etc. . The value of the differential sensitivity at $\Delta\sigma_j = 0$ was then interpolated from the 10 values of I and $(\sigma - \sigma_j)$ by a routine in the NAG library ³⁴, which employed Aitken's method of successive linear interpolations ³⁵. The accuracy of this method can be estimated by comparing the result with previous interpolates ³⁴. Applied here, the interpolated differential sensitivity coefficients were shown to have a 4 figure accuracy or better. Figures of less than 0.01% for the percentage accuracy ψ of the FISPACT evaluated differential sensitivity coefficients, defined as

$$\psi = 100 (D_{ij} - D_0) / D_0$$

where D_{ij} is the FISPACT calculated differential sensitivity coefficient and D_0 is the interpolated value, must therefore be regarded with suspicion.

The tests were conducted on two activation chains of quite different characters. Chain 1, shown in fig 2.4, is typical of longer chains found in low and medium Z elements, characterised by a movement down and to the left in a Z/A field - propagated by successive (n, α) reactions -

and low burn-up rates, typically less than a few percent. Chain 2, shown on page 131, is typical of the type of chain which predominates among heavy Z nuclei, consisting of successive capture reactions alternating with β^- decay steps. An important aspect of this type of chain is that capture cross-sections can be large (> 1 barn) for some heavy nuclei, causing substantial levels of burn-up (tens of percent). A notable feature, too, is the greater frequency of long-lived (> 1 day) isomeric states in heavy elements, creating "loops" in activation chains which rejoin the main chain by decay to the ground state (this becomes clearer when one looks at the pictorial representation of activation chains in Chapter IV).

It is to be expected that the accuracy of the sensitivity calculation depends largely on how quickly the source term varies and the number of time sub-intervals selected in the integration τ . Factors determining the former are the position of the cross-section in the chain (m) and the position of nuclide i (n). In an activation chain made up entirely of reactions with low burn-up and stable nuclei, it can be shown (see Chapter 3.) that the concentration of the n th nuclide varies with the n th power of time. Hence it can be expected that accuracy decreases with chain length - hence the importance of taking long test chains. Further, it is noted that a long chain can be broken into smaller chains of length 1, 2, 3, ..., n , by taking the n th nuclide as the chain endpoint. Chain 1 can therefore be considered to be 1, 2 or 3 steps long, depending on whether $Ti49$, $Ca45$ or $Ar42$ are taken as the endpoints.

In tables 2.1 and 2.2 the accuracy of the sensitivity coefficients, relative to the interpolated standards, is presented with respect to the position of the x -sect (m), the position of the nuclide concentration (n) and the number of time subintervals taken for the two chains (τ).

Table 2.1 : Percentage Accuracy of Differential Sensitivity Coefficients for Chain 1

n m Level		1	2	3
1	2	0.000	-5.253E-2	-6.419E-2
	5	0.000	-2.189E-2	-1.646E-2
	10	0.000	-1.751E-2	-9.875E-3
	20	0.000	1.642E-2	-8.229E-3
	50	0.000	1.751E-2	-8.229E-3
2	2		1.165	15.822
	5		0.2755	2.650
	10		0.1459	0.7507
	20		0.1114	0.2753
	50		0.1028	0.1418
3	2			12.664
	5			3.470
	10			0.5209
	20			0.1442
	50			3.826E-2

Table 2.2 : Percentage Accuracy of Differential Sensitivity Coefficients for Chain 2

n m Level		1	2	3	4	5	6
1	2	6.993	0.3739	-1.692	-3.695	-5.119	-6.894
	5	1.288	9.639E-2	-0.2725	-0.7959	-0.7886	-1.143
	10	0.4909	4.966E-2	8.737E-2	-0.3805	-0.1383	-0.2263
	20	0.2911	3.505E-2	-4.254E-2	-0.2755	2.421E-2	6.998E-3
	50	0.2368	3.213E-2	-2.989E-2	-0.2493	6.917	7.232E-2
2	2		-9.341	-9.415	-5.544	2.778	16.593
	5		-1.812	-1.663	-1.081	0.5775	3.126
	10		-0.8146	-0.4702	-0.3183	0.3407	0.9366
	20		-0.5692	-0.1706	-0.1184	0.2887	0.3662
	50		-0.5013	-8.532E-4	-6.662E-4	0.2714	0.2043
3	2			-6.401	17.678	41.788	84.762
	5			-1.028	2.963	6.378	14.732
	10			-0.1980	0.7233	0.9868	3.673
	20			1.212E-2	0.1594	0.3739	0.9256
	50			7.071E-2	0.000	-0.7570	0.1532
4	2				-1.727	43.283	98.539
	5				-0.2310	7.012	14.706
	10				7.702E-2	1.596	3.712
	20				0.1594	0.3739	0.9256
	50				0.1815	-0.1204	0.2406
5	2					12.947	98.399
	5					3.988	16.431
	10					1.464	4.997
	20					0.4533	2.189
	50					0.1011	1.406

2.7 Discussion and Conclusion

The success of a numerical scheme depends on whether the amount of effort expended in its use achieves an acceptable level of accuracy in the context of the problem. The above computational solution to the sensitivity equations is certainly convenient to use. As to whether its accuracy is acceptable, it is necessary to refer back to the level of accuracy required by the applications of the sensitivity coefficients ; to the delineation of reaction channels and the propagation of errors.

The first of these roughly grades cross-sections by the size of their sensitivity coefficients as being insignificant in the production of the end nuclide N_i ($S_{ij} < 0.01$), of minor importance ($0.15 > S_{ij} > 0.01$), or major importance ($S_{ij} > 0.15$). This is an inherently approximate procedure, and it makes very little difference if its sensitivity coefficient is uncertain to 10%. In the case of the second application, it is important that the uncertainty in the coefficients be smaller than the errors attached to cross-section data. Since these vary from a few percent (for the very best experimental determinations) to hundreds of percent (for determinations obtained from models), the required accuracy does not have to be particularly high. An accuracy of 5% may therefore be considered to be adequate in most situations. It can be seen from tables 2.1 and 2.2 that this degree of accuracy can be reached for nearly all the sensitivity coefficients provided by the two chains if 5 or more time subintervals are taken.

In accordance with expectations the precision of the calculation appears to be influenced by

a) the number of time subintervals taken in the integration

b) the length of activation chains

c) the position of cross-sections in chains

though the variations of b) and c) with τ are by no means regular.

Both a) and c) are linked to the rate at which the source term varies over the time subinterval. For nearly all the coefficients verified, the accuracy appears to be limited in a direct way by a). This may be demonstrated if the sensitivity coefficients with respect to a certain cross-section, say the $W186(n,\gamma)$ cross-section ($m = 1$), are calculated at different numbers of time subintervals τ , and accuracy ψ plotted against τ , as in figure 2.5. This shows ψ to be a smooth function of τ , converging towards a limit as τ tends to infinity, the value at large τ defined by the other errors in the computation. In general, 5 to 10 subintervals appear to be enough to give an accuracy of better than 5%.

The effects of b) and c) on accuracy can be seen more clearly in figures 2.6 and 2.7 : the overall trend is for accuracy to be lost as m and n increase, but irregularities may occur for individual coefficients. Unless the combination of chain length and cross-section position are particularly exacting (and the example chains taken here do represent extremes), or irradiation conditions particularly severe, a value of 5 for τ will suffice for most purposes.

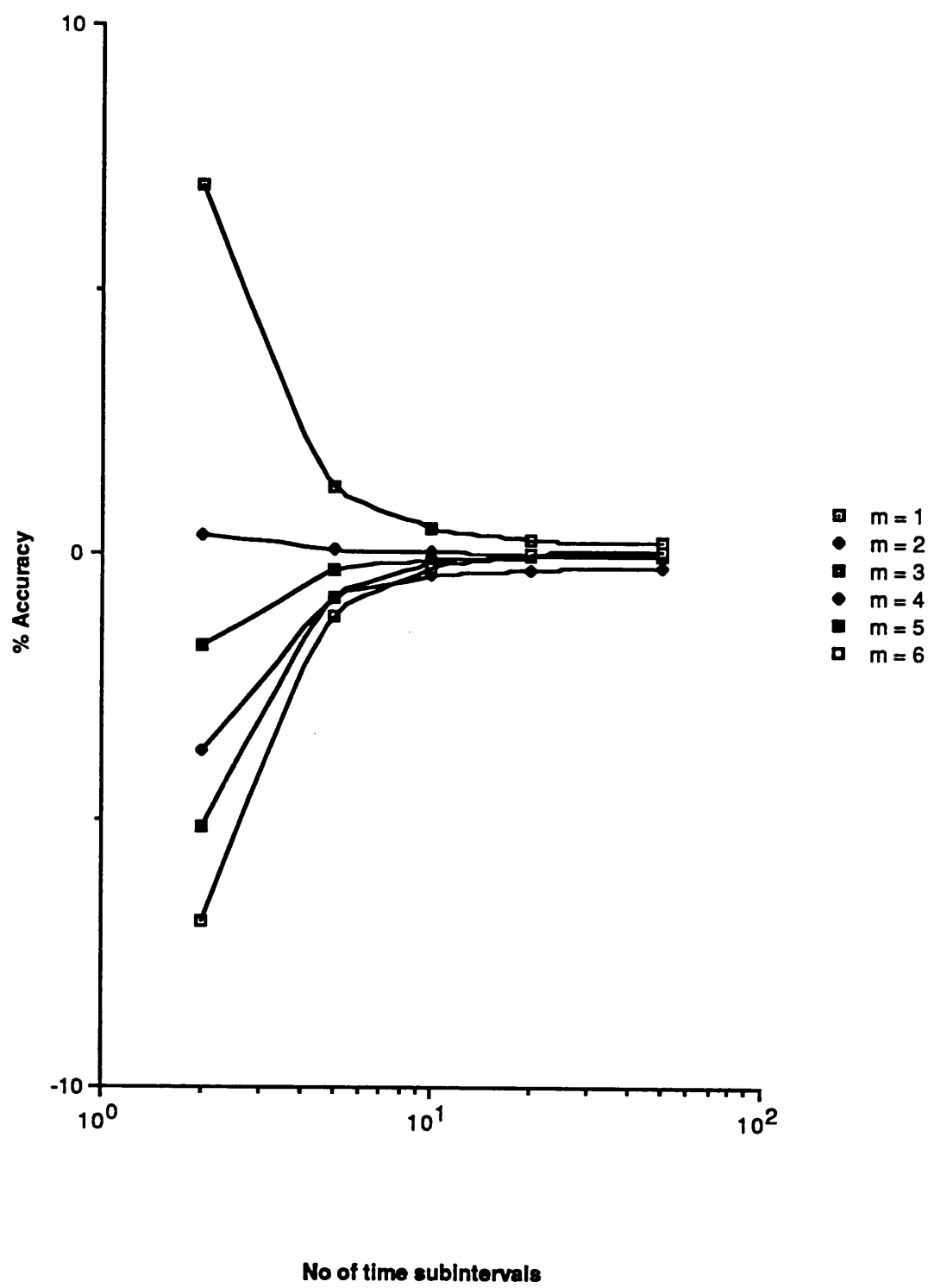


Figure 2.5 : Accuracy of computed sensitivity coefficients with respect to the number of time subintervals

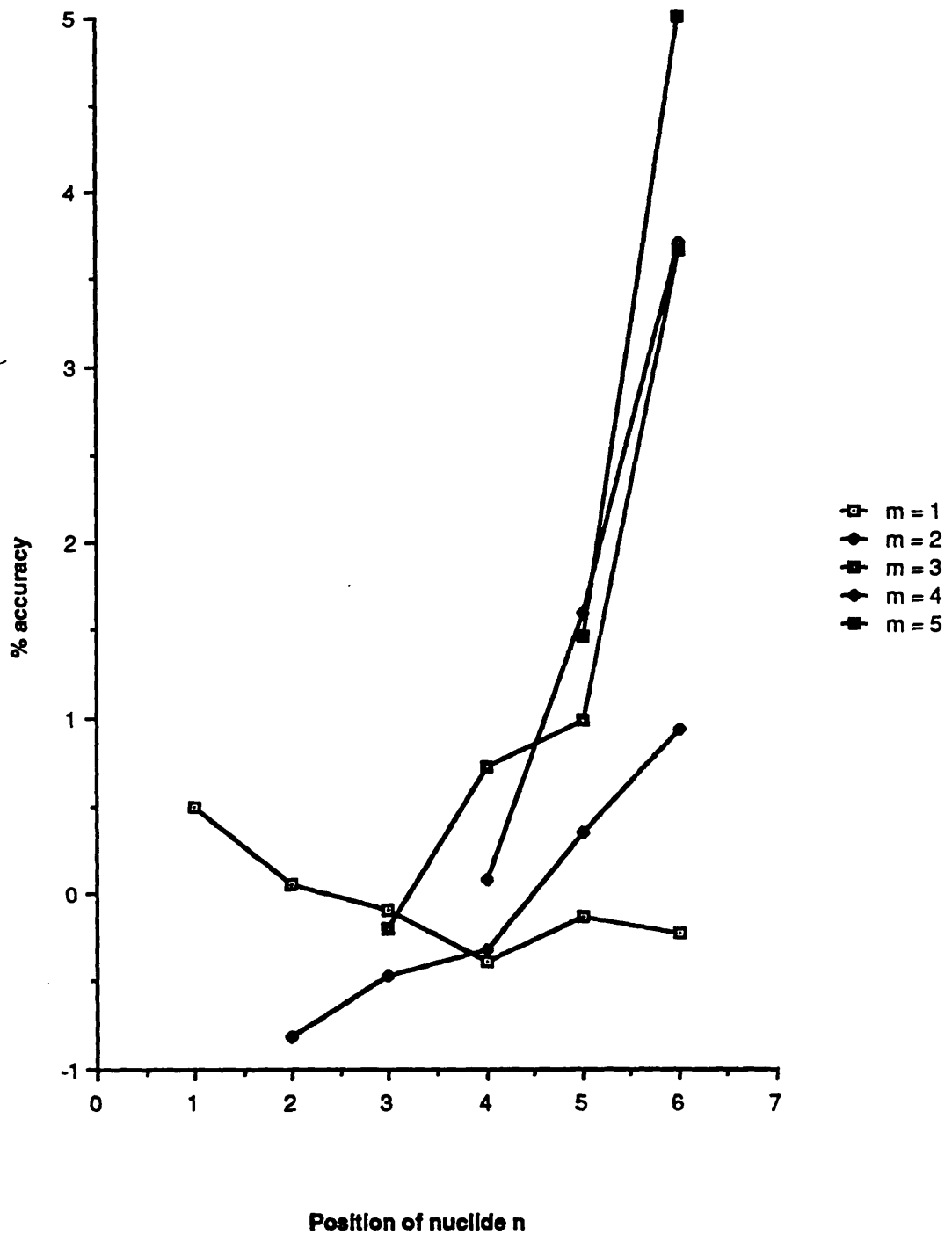


Figure 2.6 : Accuracy of computed sensitivity coefficients with respect to the length of the activation chain n

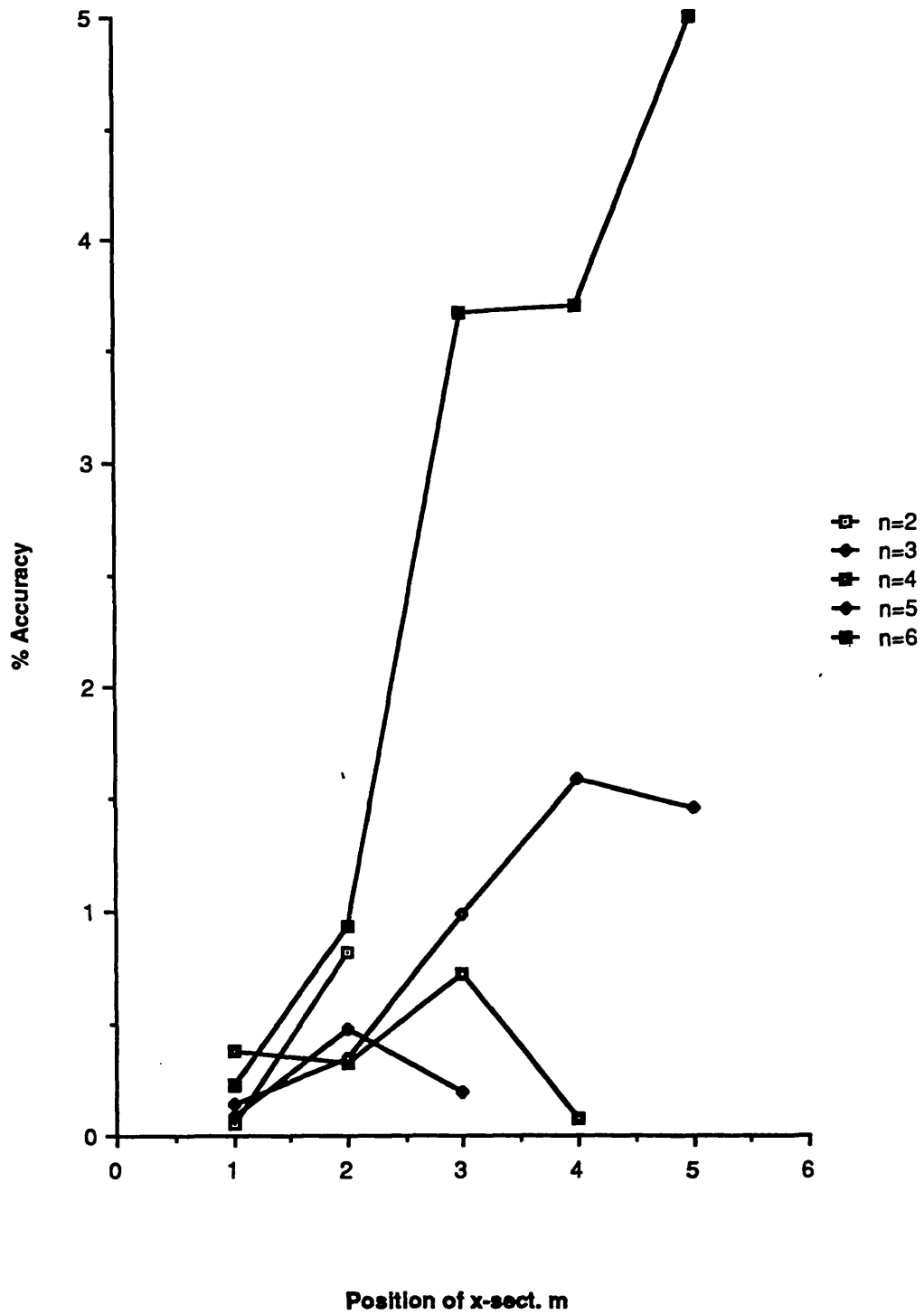


Figure 2.7 : Accuracy of the computed sensitivity coefficients with respect to the position of the cross-section in the activation chain m

CHAPTER III

Adequacy of the Activation Database

Who is the third who walks always beside you ?

When I count, there are only you and I together

But when I look ahead up the white road

There is always another one walking beside you

Gliding wrapt in a brown mantle, hooded

I do not know whether a man or a woman

-But who is that on the other side of you ?

- What the Thunder said

3.1 Introduction

The current philosophy in fusion activation calculations, and the one on which the work reported here is based, is to seek improvements in the accuracy of such calculations by improving the database rather than the method of calculation. The database used here, UKACT1, is the latest and most comprehensive in a line of UK fusion activation databases, and represents a several-fold expansion of the previous UKCTR-III database. It is still, however, in some respects interim : progress towards a satisfactory database is seen as an evolutionary process, proceeding by alternate steps of data determination and sensitivity studies which measure the effect of the new data and test its adequacy. The process of improvement may be considered to be at an end when the sizes of errors introduced into the calculations by uncertainties in the data are of the same order as the errors introduced by the computation and modelling.

It is pertinent to ask at this stage in the database's development, what a satisfactory database will look like, and to establish criteria for the inclusion and treatment of data in such a database.

It is possible to categorise the errors resulting from inadequacies in the cross-section database into two distinct types ;

- a) errors introduced by the exclusion of cross-section data for reactions
- b) errors introduced by inaccuracies in included cross-section data

It is important to realise that each of these types of error are propagated in different ways. The effect on activation calculations of type b) errors is to alter the concentrations of the inventory products. Providing that errors in the cross-sections and their sensitivity coefficients can be obtained, the resulting errors in the nuclide concentrations can be calculated in the manner described in Chapter II.

Errors of type a) differ in that, as well as carrying errors to nuclide concentrations, they may exclude pathways to products which may not appear in the nuclide inventory otherwise. Unlike type b) errors, they cannot be treated in a formal error analysis, since their sizes are unknown. Clearly, it is necessary to have a set of criteria for the inclusion of data in the database which is sensitive to the consequences of possible omission : for only when it can be shown that further expansions of the database will not significantly change the results of calculations can the process of expansion come to an end.

The sizes and propagation of errors is treated in Chapter IV: the rest of this chapter will deal specifically with establishing criteria for including cross-sections in the database.

3.2 Formulation of Data Inclusion Criteria

3.2.1 Chain propagation

The multiple reactions undergone by nuclei in the high fluxes found in the first wall of a fusion reactor may be thought to constitute a set of independent activation chains. Of course this is merely a notional convenience, since a large number of daughter nuclei can be produced from given target nuclei - providing linkage between the chains.

The chains may be propagated by two mechanisms ; transmutation and decay. The amount of material passed on by a transmutation is dependent on a number of factors, as will be demonstrated below, but is usually (in the irradiation conditions considered here) small compared to concentration of the parent. Propagation of a chain by decay depends on the size of the decay constant : the quantity of material passed on can vary from nothing to nearly all of the parent. Decay can therefore be a more efficient mode of propagation than transmutation. Commonly, long activation chains contain a mixture of the two.

As noted in the Introduction, broadly speaking there are two types of activation chains ;

- a) those proceeding by successive (n,γ) reactions and β^- decays, characterised by a movement up and to the right in a Z/A field, giving products with higher atomic numbers than the source nuclei.
- b) those proceeding by successive threshold reactions of the (n,p) , (n,d) , (n,α) , $(n,2n)$ variety, with intervening β^+ (or sometimes β^-) decay steps, giving products with lower atomic numbers than the source nuclei.

3.2.2 Chain Model - No Burn-up

It is possible to trace the concentrations of materials transmuted down an activation chain by exact solutions, as demonstrated in

section 2.4, but the expressions are extremely cumbersome, and do not lend themselves to formulating an easily applied set of inclusion criteria. Alternatively, if one starts with the premise that burn-up is small and that the change in the concentration in the parent is negligible over the irradiation period, then one may obtain an approximate expression for concentration of successive generations of daughters as follows

$$dN_1 = \sigma_1 \phi N_0 dt$$

By simple integration

$$N_1 = \sigma_1 \phi N_0 T \quad (3.1)$$

where T is the length of irradiation.

Similarly, the concentration of the granddaughter is given by

$$\begin{aligned} dN_2 &= \sigma_2 \phi N_1 dt \\ &= \sigma_1 \sigma_2 \phi^2 N_0 T dt \end{aligned}$$

$$\Rightarrow N_2 = \sigma_1 \sigma_2 (\phi T)^2 N_0 \quad (3.2)$$

Then by iteration the general form for the concentration of the n^{th} nuclide in the chain is

$$N_n = \sigma_1 \dots \sigma_n (\phi T)^n N_0 \quad (3.3)$$

Thus it is seen that in a simple activation chain consisting of

transmutations only, the concentration of the n^{th} daughter varies with the n^{th} power of fluence.

In practise, however, many chains contain steps involving transmutations from unstable nuclei. These may be treated in the following way :

$t_{1/2} > T$: the nuclide is treated as stable. It can be reasoned that the error produced by this approximation will be less than a factor of 2 by considering the worst case - a source nuclide with half-life of T : the concentration of this nuclide will fall from its initial value to half over the irradiation period. Assuming the simple linearity of equation 3.1, the amount of material transmuted will therefore be greater than half of the amount which would be transmuted from a stable parent.

$t_{1/2} < T$: the amount of transmuted material in the i^{th} step is then

$$N_i = (\sigma_i \phi) N_{i-1} / \lambda_i \quad (3.4)$$

Therefore for a chain consisting of $m + n$ steps, m of which are transmutations from unstable nuclei, n from stable nuclei, the concentration of the $(m + n)^{\text{th}}$ nuclide is

$$N_{m+n} = (\sigma_1 \dots \sigma_m \phi)^m (\sigma_1 \dots \sigma_n \phi T)^n N_0 / (\lambda_1 \dots \lambda_m) \quad (3.5)$$

It is noted that the introduction of decay steps brings in a variation with flux, in addition to the variation with fluence remarked upon earlier.

If the cross-sections are approximately of the same size, then the

expression may be reduced to

$$N_{m+n} = (\sigma \phi)^m (\sigma \phi T)^n N_0 / \prod \lambda_i \quad i = 1, \dots, m \quad (3.6)$$

Likewise, the equivalent form for a chain consisting entirely of transmutations from stable nuclei is

$$N_n = (\sigma \phi T)^n N_0 \quad (3.7)$$

3.2.3 Chain Endpoints

As a steadily shrinking quantity of material is transmuted by successive reactions down an activation chain, there comes a point when the quantity of material may be considered to be so small as to be radiologically insignificant. At this point the chain may be deemed to have reached its endpoint, and cross-section data for reactions further down the chain are not required by the database. It can be seen from equation (3.3) that in the idealised case of a chain made up entirely of stable nuclei, the attainment of a previously set minimum concentration level depends on the number of reactions which have taken place on the nucleus n , the fluence ϕT , the size of the cross-sections σ and the concentration of the source nuclei N_0 . For given irradiation conditions and reaction type therefore, it is possible to state approximately how many steps are necessary for a chain reach an endpoint.

Before this can be done, however, "insignificance" must be given a precise meaning, provided by the context of the activation calculation. If T_c is the cooling time and an insignificant concentration is defined, with some justification from component handling / recycling

scenarios, as the concentration at which an isotope with the gamma attenuation coefficients of iron, emitting a 1MeV photon with half-life of T_c , gives a surface gamma dose rate of $25\mu\text{Sv/hr}$, then at cooling times of 1 day, 30 years and 100 years, that concentration would be

T_1	N_{crit} (atoms/Tonne)
1 day	$7.5\text{E}+12$
30 yrs	$8.2\text{E}+16$
100 yrs	$2.7\text{E}+17$

It can be seen that the chain endpoint, according to this limit on gamma dose rate, is strongly dependent on cooling time, and hence data inclusion criteria for operational or component replacement scenarios are stiffer than for, say, disposal. The last figure, N_{crit} for a one hundred year cooling period, is the one which will be adopted for the rest of this argument, since it is most relevant to the timescales considered in this study.

Taking 0.05 barn to be a typical cross-section size for a threshold reaction of the type (n,2n) or (n,p) in nickel, figure 3.1 shows how many transmutations are necessary before the chain endpoint is reached at a number of different cumulative wall-loadings (at 5MW/m^2 power) in the range 2.5 to 50MWy/m^2 , as predicted by expression 3.7.

Superimposed on these curves is one showing successive (n,2n) reactions from Ni^{64} under a wall-loading of 12.5MWy/m^2 , on nickel isotopes which are either stable or long-lived. Accepting the non-uniformity of cross-sections sizes, the curve shares the same general characteristics, departing from the exponential noticeably in the $n=3$ step because of the high cross-section for this step, but finishing at a value close to the idealised line for 12.5MWy/m^2 . It falls below N_{crit} in

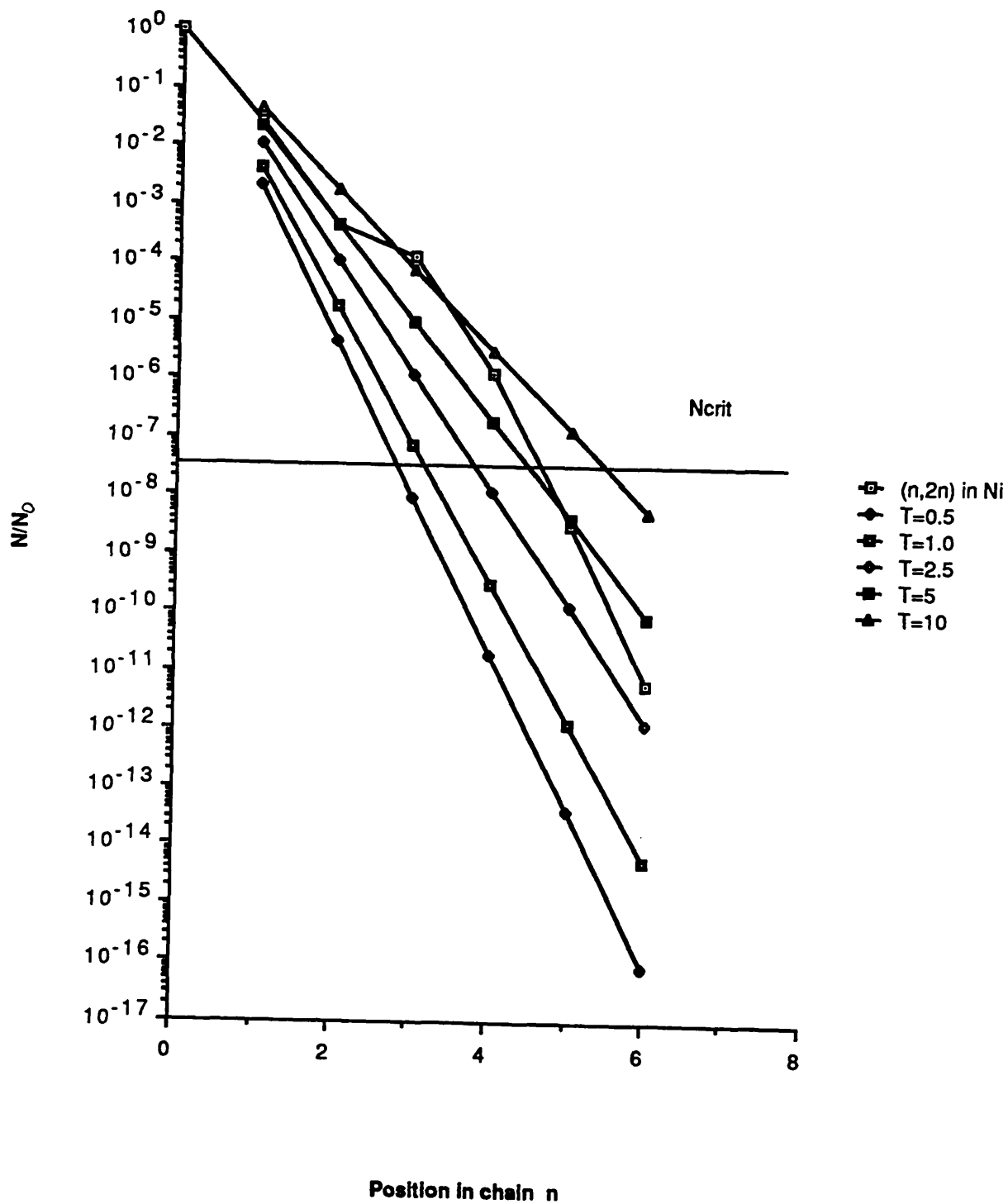


Figure 3.1 : No. of transmutations needed for chain endpoint to be reached for different wall-loadings

the $n=5$ step.

An indication of the effect of wall-loading on how quickly the chain concentration falls below N_{crit} can be obtained by considering the idealised lines. The effect of increasing the wall-loading by a factor of 5 is to increase the chain length by approximately 1 step. Since the wall-loading of a fusion reactor is well-defined, restricted at the upper level by radiation damage and erosion, at the lower level by machine efficiency, most reactor designs employing loadings in the range 5 and 20 MWy/m² ³, it is unlikely to change from 12.5MWy/m² by much more than a factor of 2, it can be concluded that criteria for the inclusion of cross-sections are not sensitive to wall-loading for activation calculations with long timescales.

Of greater significance for the formulation of inclusion criteria is the range of cross-section sizes associated with the different reactions possible in the first wall, and the variation of cross-section size with Z number. Table 3.1 presents examples of some typical effective cross-sections for common reactions on 4 nuclei of differing atomic numbers, as obtained from UKACT1 and a DEMO first wall spectrum.

Table 3.1 : Typical cross-section sizes for common first wall reactions °

	(n,p)	(n, α)	(n,2n)	(n, γ)
C13	1.55E-3	1.14E-2	4.39E-2	1.81E-5
Fe56	1.56E-2	4.90E-3	5.61E-2	1.68E-2
Cs133	7.78E-4	1.19E-6	2.20E-1	3.07E+0
Bi209	8.40E-5	6.37E-5	8.22E-4	1.17E-2

The figures illustrate the following trends ;

- i) at low Z numbers (e.g. C), the largest cross-sections are for reactions involving the emission of charged particles and neutrons. Capture reactions occur with low probability.
- ii) at intermediate Z number (e.g. Fe), cross-sections for neutron-induced reactions involving particle emission are roughly equal to those for capture reactions.
- iii) at high Z numbers, the dominant reactions are capture reactions, followed by reactions involving neutron emission, in turn followed by reactions involving charged particle emission.

It should be added, however, that these trends are dependent on the neutron energy spectrum. It has been shown in Chapter 1 that this can vary significantly between reactor designs, especially in the thermal and resonance region. The trends might be different, therefore, for another reactor design, or another part of the DEMO machine.

It is obvious from expression (3.7) that the size of the average cross-section affects the propagation of material along an activation chain in much the same way as fluence, the concentration at the n th step varying with the n th power of the cross-section. Figure 3.2 is similar to figure 3.1, but shows how quickly chain concentration falls below N_{crit} for a number of idealised average cross-section values at a wall-loading of 12.5MW/m^2 . It is noticeable that at cross-sections greater than about 10^{-1} barn, such as are found for some $(n,2n)$ and (n,γ) reactions, the curve tends towards the horizontal, allowing a large number of reactions to take place before N_{crit} is reached. At the other extreme, reactions with cross-sections smaller than about 10^{-7} barn drop below N_{crit} in just one step.

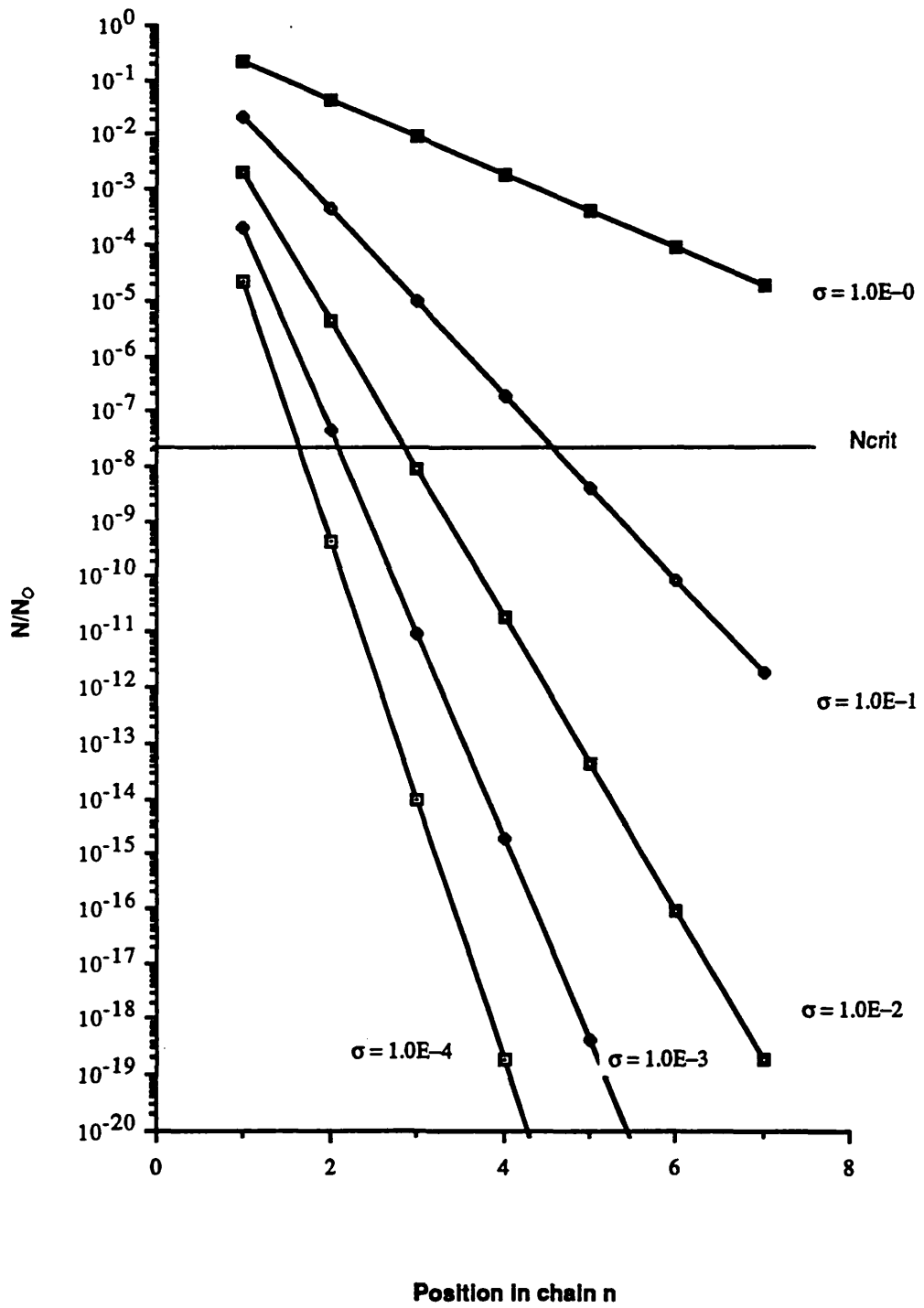


Figure 3.2 : Number of transmutations needed to bring chain concentration down to endpoint for different idealised cross-sections

3.2.4 Reactions on short-lived species

On examination of the database, it is found that the class of reactions for which the application of inclusion criteria is most needed is reactions on unstable nuclei. Below a certain half-life, the equilibrium concentration of a decaying nuclide would be sufficiently low for the amount of material transmuted from it by a single reaction to be less than N_{crit} . The value of this limit on the half-life of target nuclei is one of the principal factors defining the size of the database, successive updates of the database having adopted successively lower values, until the present value of 1 day was reached in UKACT1. The correctness of this adopted limit may be tested by applying the simple model of chain propagation. From expression 3.5 it is easy to obtain

$$N_{m+n}/N_0 = (\sigma \phi)^m (\sigma \phi T)^n \prod t_{1/2_i} \quad i = 1 \dots m \quad (3.8)$$

Taking the nuclei to be identical, and $n=0$, then

$$N_m/N_0 = (\sigma \phi)^m (t_{1/2})^m \quad (3.9)$$

The implications this relationship has for defining chain endpoints for reactions on unstable nuclei may be appreciated from figure 3.3, showing N_m/N_0 plotted against $t_{1/2}$ for $m=1,2,3$ at a flux of $2.68\text{E}+15$ neutrons/cm²s and a cross-section size of 10^{-2} barn. Immediately apparent is that the chain concentration drops below N_{crit} in one transmutation on a nuclide with half-life of about 10^3 seconds, two transmutations on nuclides with half-lives of about 5×10^6 seconds and three transmutations on nuclides with half-lives of about 10^8 seconds.

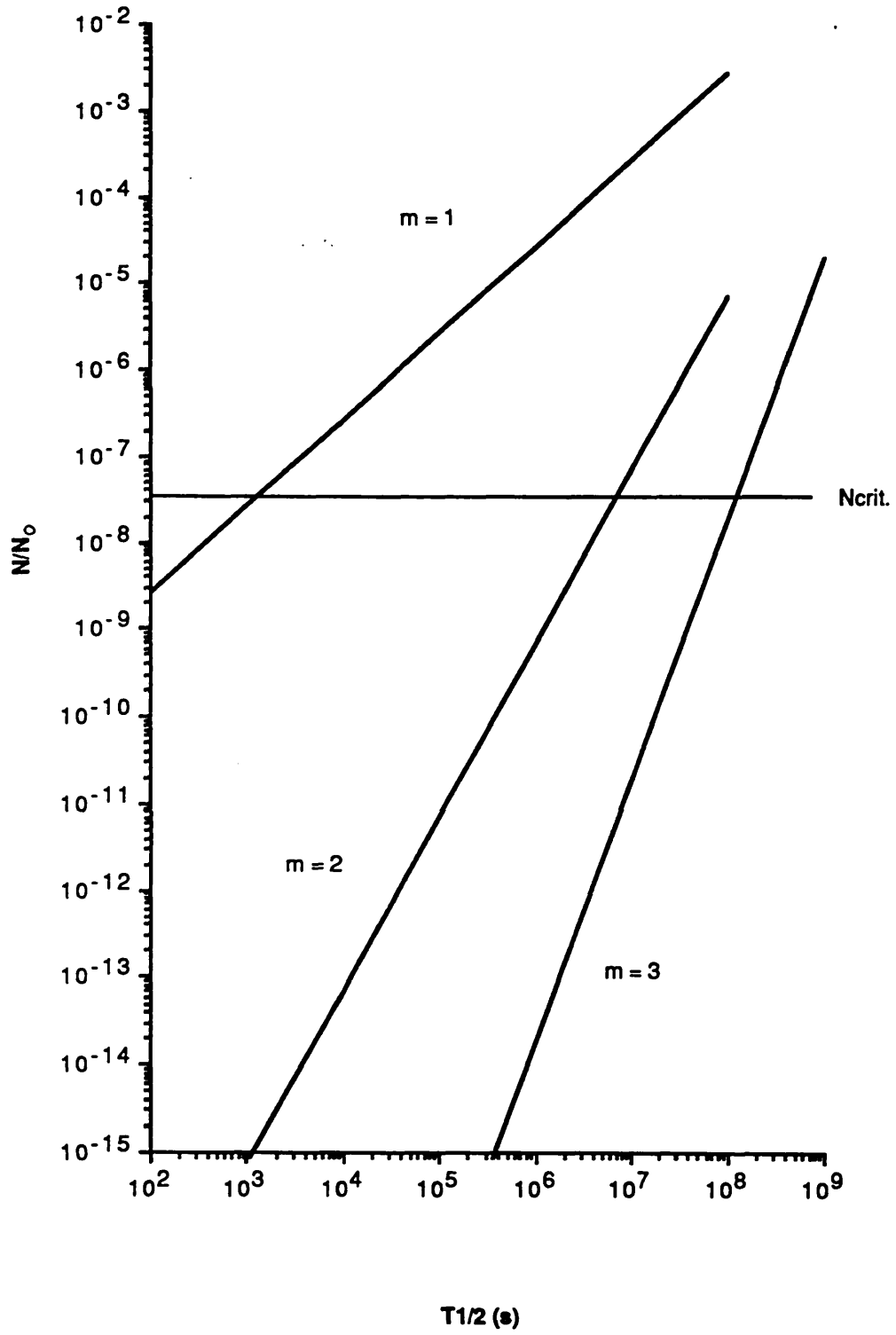


Figure 3.3 : N_m/N_0 plotted against $t_{1/2}$ for $m = 1, 2, 3$

It can be seen however, from table 3.1, that the size of cross-sections, especially for $(n,2n)$ and (n,γ) reactions, can often exceed 10^{-2} barn. The effect of cross-section size on N/N_0 can be gathered from figure 3.4, in which expression 3.9 is evaluated for a one step chain (i.e. $m=1$) for a number of different cross-section sizes. The conclusion to be drawn from this figure is that extreme caution is required when considering transmutations on short-lived nuclei, as a reaction with a cross-section of 1 barn, on a nuclide with half-life as short as 10 seconds, can still transmute a significant concentration of material.

These conclusions are watered down somewhat when one considers the real situations in which short-lived nuclides would be produced : in UKACT1 they usually result from "lateral" activation chains (in a Z/A field), starting from a source nuclide close to the line of stability, moving to nuclides of progressively shorter half-lives as the chains move further away from the line of stability. The most common situation is a chain consisting of 2 stages ; a transmutation from a stable source nuclide to a short-lived nuclide, which in turn is transmuted to the long-lived species of interest. In this case, the values of N/N_0 in figure 3.4 would be reduced by a factor of $\sigma \phi T$, and for the limiting case of a large cross-sections of the order of 1 barn, nuclides with half-lives of about 50 seconds would be transmuted to a significant degree.

For longer chains, including the frequently encountered chain which consists of 3 stages ; a transmutation from a stable source nuclide to a slowly decaying daughter (half-life of the order of days), which in turn is transmuted to a shorter lived granddaughter (half-life of the order of hours or minutes), which is then transmuted to the long-lived species of interest, it is best to return to expression 3.8 .

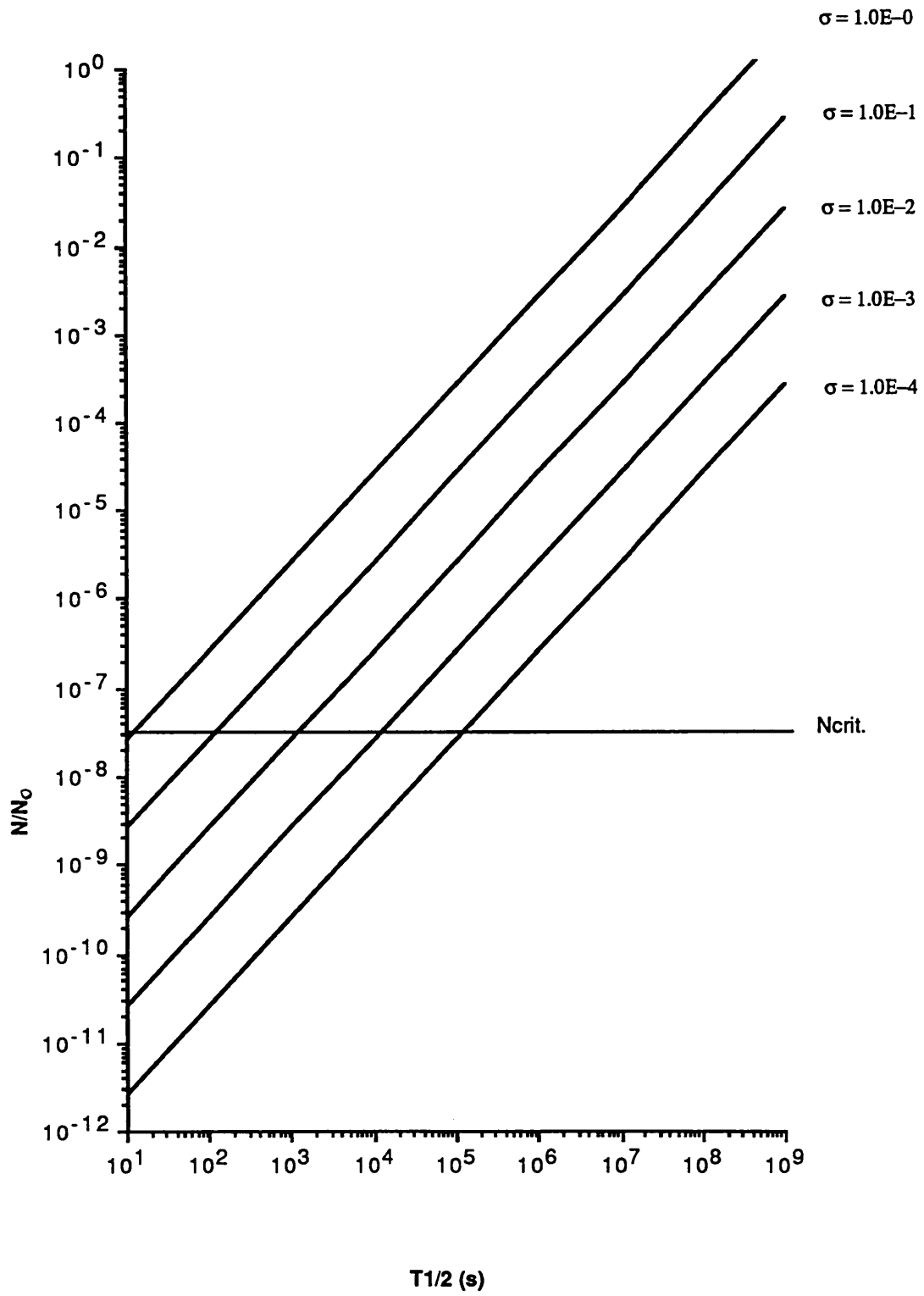


Figure 3.4 : N/N_0 plotted against idealised cross-section size

3.3 Reactions in the Fusion Reactor First Wall

The above arguments, though approximate, will be used as a rough guide in the following discussion on the inclusion of cross-section data for specific classes of first wall reactions. Taking the same nuclei as table 3.1, table 3.2 gives the number of successive reactions which would bring the chain concentration down to N_{crit} for the irradiation conditions and definitions already set up, if the cross-sections are taken as being typical of nuclei of a similar Z number. Given in table 3.3, is the half-life of a daughter nuclide which would yield an N_{crit} quantity of a granddaughter, in a chain whose parent is a stable source nuclide.

Table 3.2 : Number of reactions required to bring chain concentration down to N_{crit} .

	(n,p)	(n, α)	(n,2n)	(n, γ)
C13	3	3	4	2
Fe56	4	3	4	4
Cs133	2	2	6	41
Bi209	2	2	3	3

Table 3.3 : Half-life (s) of second nuclide in a 2 stage chain which would result in N_{crit} being transmuted.

	(n,p)	(n, α)	(n,2n)	(n, γ)
C13	1.84E+7	3.40E+5	2.30E+4	1.35E+11
Fe56	1.82E+5	1.84E+6	1.41E+4	1.56E+5
Cs133	7.31E+7	3.12E+13	9.14E+2	4.70E+0
Bi209	6.27E+9	1.09E+10	6.55E+7	3.23E+5

3.3.1 Capture Reactions

Potentially the largest yielding type of reaction in the first wall, and likely to dominate in other parts of the structure where the neutron energy spectrum is moderated to a higher degree, capture reactions provide a limiting case for data inclusion criteria. The size of a single group cross-section depends on the irregular occurrence of resonances, which cannot adequately be treated by nuclear models, and which can vary markedly between neighbouring nuclei. The example of Cs133 shows that the size of the cross-sections for these reaction can be such that in the first wall irradiation conditions considered here, activation chains can be propagated almost indefinitely by capture reactions, being terminated only by nuclei with low capture yields. However, in practise a chain 41 reactions long would be an impossibility, owing to the wide range of cross-sections sizes in a naturally occurring chain, and the absence of burn-up in the above argument. An example of a long capture chain which has already been mentioned, is the production of Ir192n by 6 successive captures and a number of β^- decays from W186.

Table 3.3 suggests that in the region of the periodic table where

capture reactions can be large, the criterion of including all reactions on nuclei with half-lives of greater than 1 day, could lead to significant omissions.

3.3.2 Threshold Reactions

The most probable reactions at neutron energies of greater than about 1MeV are reactions characterised by particle emission, and by excitation curves which exhibit conspicuous energy thresholds. A large number of particle emission permutations are possible, the most probable of which can occur with probabilities approaching those for capture reactions.

A lower limit for the inclusion of reactions can be provided by considering the size of a cross-section transmuting N_{crit} in one step from the source nuclide. It was shown in section 3.2.2 that this is approximately 10^{-7} barn.

Activation chains consisting of (n,p) and (n, α) reactions typically reach endpoints after 3 or 4 steps in light materials, 2 in heavy materials. An example of a long chain of this type is the production of Ar42, the main source of external dose in the Cr inventory, from Cr51 by 3 α -emitting reactions. The 1 day criterion applied to the inclusion of reactions on short-lived nuclei, deals adequately with the examples of (n,p) and (n,a) reactions taken in table 3.3.

Activation chains consisting of (n,2n) reactions tend to be slightly longer than chains made of (n,p) and (n,a) reactions, and do not exhibit the same shortening at large Z numbers. Like the capture cross-sections, the (n,2n) cross-sections peak in region of medium heavy nuclei, such as Eu (Z=63). The 1 day inclusion criterion does not appear to be adequate for large (n,2n) cross-sections, but is probably

satisfactory for the other types of threshold reactions.

3.3.3 Secondary Charged Particle Reactions

It has been suggested that a possibly significant class of reactions in the first wall and blanket is the secondary reactions involving charged particles produced by primary neutron-induced reactions. Though this looks unlikely on the surface - the sizes of the cross-sections for these reactions being less than the primary reactions by a factor which takes into account the probability of the second reaction taking place, they merit examination because their movement in a Z/A field may open up new reaction pathways : for like successive capture reactions, they result in a movement up and to the right, producing heavier nuclei, but with fewer steps than neutron - induced reactions. In table 3.4 are listed various charged particle reactions and the combination of neutron induced reactions which would be equivalent.

Table 3.4 : Neutron-induced equivalents to charged particle reactions

Charged particle reaction	Neutron-induced equivalent
(d,p), (t,d)	(n, γ)
(t,p)	2(n, γ)
(α ,p)	3(n, γ)
(d,2p)	(n,p)
(t,2p)	(n,p)(n, γ)
(α ,2p)	2(n, γ)

It is clear that the inclusion of charged particle reactions which are equivalent to common neutron-induced reactions with large cross-

sections, such as (n,g) and (n,p) reactions, would not alter inventories by much, since they would be swamped by the primary reactions. The reactions which could be significant are those which are equivalent to a multiple number of neutron-induced reactions. These would require sizable fluxes of tritons and α particles, which are probably only to be found in light elements.

If the α -induced reactions in carbon are taken as an example, and the assumptions made in the simple chain model maintained, the rate of the secondary reaction is

$$dN_c / dt = S_c \sigma \phi N_0$$

$$\Rightarrow N_c / N_0 = S_c \sigma \phi T$$

where S_c is the probability of the second reaction taking place. If N_{crit}/N_0 is taken to be $2.7E-8$ as before, and the $C13(n,\alpha)$ primary reaction is considered, then S_c must be greater than 10^{-5} to warrant inclusion of the reaction in the cross-section library.

Whether S_c achieves this degree of probability will be strongly dependent on the energy spectrum of the charged particle flux and the excitation functions of the secondary reactions, as given by the following relationship

$$S_c = \int \Sigma(E) \phi(E) dE / \int \phi(E) dE \quad \text{where} \quad (3.10)$$

$$\phi(E) = \int \sigma(E_n) \phi(E_n) dE_n \quad (3.11)$$

Without having to evaluate S_c , however, several things are clear ;

- i) at all masses except for the very light, the thresholds for charged particle reactions occur at energies of above several MeV e.g. about 10MeV for the $\text{Ni60}(\alpha, n)$ reaction ³⁶. Thresholds for α -induced reactions have noticeably higher threshold energies than for p, d and t-induced reactions.
- ii) the primary, neutron-induced reactions, which produce the charged particle flux, also have thresholds of at least several MeV. The charged particle energy spectrum resulting from a 14MeV neutron flux will therefore peak at energies below the thresholds for secondary reactions. In consequence the effective single group cross-sections will be smaller than those for primary reactions.
- iii) The Q-values of charged particles reactions on light nuclei can be low, and it is in this class of nuclei, if any, that charged particle reactions may be significant. A previous study of these reactions on alternative NET blanket designs ³⁷, in which S_c was evaluated in the form expressed above is inconclusive however, and a fuller study on the reaction rates in the first wall and blanket is necessary before they can be safely excluded from the database.

3.4 Conclusions

The most salient points to emerge from this discussion on cross-section data inclusion criteria may be summarised as follows ;

- i) the endpoint of an activation chain is defined as the point where the concentration of material passes below a level which can be considered significant according to predetermined radiological criteria. The main criterion taken here is derived from the nuclide concentration N_{crit} which would give the "hands on" limit for surface gamma dose rate

after a 100 years cooling period.

ii) inclusion criteria are sensitive to the limit on nuclide concentration which corresponds to radiological insignificance.

iii) in activation chains characterised by stable nuclei and low burn-up rates, the chain concentration varies with n^{th} power of fluence, where n is the number of steps in the chain.

iv) activation chains with cross-sections of a barn or greater, such as occur for some capture reactions, will be propagated indefinitely until a nucleus with a low cross-section appears in the chain.

v) reactions with single group cross-sections below about 10^{-7} barn can be ignored, according to the criteria adopted to define chain endpoints here.

vi) the 1 day inclusion limit on target nuclei is inadequate for capture and $(n,2n)$ reactions with large cross-sections.

CHAPTER IV

A Sensitivity Analysis of the UKACT1 Activation Database

Gentile or Jew

*Oh you who turn the wheel and look to windward,
Consider Phlebas, who was once handsome and tall as
you.*

- Death by Water

4.1 Introduction

In this chapter, the sensitivity method described in Chapter II is applied to the upgraded activation database UKACT1. The evaluated sensitivity coefficients are employed in both of the roles which were formulated in that earlier chapter ; namely to delineate reactions channels to important inventory nuclides, and to treat the propagation of errors arising from the database.

Given the large size of the database, the set of sensitivity coefficients is enormous (approximately = No. of nuclides x No. of items of data = 1314 x 8000 for the cross-section data alone). Evaluation of every coefficient is neither feasible from considerations of computing time and data manipulation, nor desirable from the viewpoint of how useful the data would be. It is therefore important to carefully define the scope of the analysis.

The analysis described here will be restricted to cross-section data relevant to the long-lived activation products from elements which are potential constituents of first wall compositions. Nearly all of the elements examined are present in the candidate first wall

Table 4.1 : Elements treated in the sensitivity analysis and their main long-lived radionuclide products

Element	Main Long-lived Products
C	Be10, C14
N	Be10, C14
Al	C14, Al26, Na22 ?
Si	Al26, Si32
K	Cl36, Ar39
Ca	Cl36, A39, Ca41, Ar42
Ti	Ar42, Ti44
V	Ar42, Ti44
Cr	Ar42, Ti44
Mn	Mn53, Fe55, Co60
Fe	Mn53, Fe55, Co60
Co	Co60, Ni59, Ni63
Ni	Co60, Ni59, Ni63
Cu	Co60, Ni63
Zr	Sr90, Zr93, Nb94
Nb	Nb94
Mo	Nb91, Nb93m, Nb94, Tc99
Ag	Ag108m, Cd113m
Sn	Ag108m, Cd113m, Sn121m, Sn126
Ce	Cs137
Nd	Sm151, Eu152, Eu154
Sm	Sm151, Eu152, Eu154
Eu	Sm151, Eu152, Eu154, Gd150
Tb	Tb158
Ho	Ho166m
Ta	Hf178n, Hf182, Re186m
W	Hf178n, Re186m, Ir192n
Ir	Ir192n, Pt193
Pb	Pb205, Bi207, Po210
Bi	Bi207

compositions discussed in Chapter V, but some additional common structural elements, such as Cu (used in the magnets) and Pb (used in the blanket), have also been included.

The other important factor which will define the the scope of the sensitivity study is the selection of the radiologically dominant nuclides in each of the elemental activation inventories. Here it is necessary to reach a compromise between calculating redundant data in an over-exhaustive approach or running the risk of possibly missing out nuclides which could play significant roles in some of the potential hazard scenarios. The eventual approach was to include in the analysis those nuclides which give rise to greater than 1% of the total surface gamma dose rate, total specific ingestion dose, or total specific inhalation dose after a cooling period of 100 years. It should be added that where inhalation and ingestion hazard data were unavailable for inventory nuclides, omissions may have occurred for some marginally important beta-emitters.

The selected inventory nuclides and their source elements, selected on the basis of the set of activation calculations described in Chapter 1, have been tabulated in table 4.1.

It should be noted that the list of nuclides in table 4.1, though covering perhaps the majority of radiologically significant nuclides, is by no means comprehensive : it is unique to the selection procedure outlined above. If, for example, the selection procedure was altered to suit a shorter timescale, say that for the operational hazard to personnel in the reactor hall, a range of nuclides with half-lives shorter than those in table 4.1 will come into play. Other lists selected on the basis of specific hazard scenarios may therefore also depart substantially from table 4.1, with nuclides brought into

reckoning by virtue of the range of mobility coefficients, shielding considerations etc..

It should be noted here, too, that the findings of the sensitivity analysis will not be wholly independent of the irradiation conditions and neutron energy spectra employed in the calculations, as the relative importance of competing reactions pathways varies with these factors. Given the essential similarities between the neutron energy spectra from most current tokamak designs, as discussed in Chapter I, it is expected that the nature of the dependence will be weak, however, and the conclusions from this study on the DEMO design will, say, also have relevance to the NET design.

4.2 Delineation of Production Channels

4.2.1 Introduction

The sensitivity coefficients $(\partial N_i / \partial \sigma_j)(\sigma_j / N_i)$ associated with the important nuclides tabulated in table 4.1 have been evaluated by the computational method described in chapter II. Coefficients of greater than 0.01 are presented in table 4.2. The reactions given here, can be regarded as the most important ones for the nuclides under consideration. Also given are the sources of the cross-section data in the activation library, and estimates of the errors attached to the single group cross-sections (see section 4.3).

The same information is presented in a more digestible, graphical form in figures 4.1 - 4.17, where dominant production channels and their corresponding sensitivity coefficients have been put in bold type.

In interpreting this sensitivity data as a straightforward representation of the respective yields offered by competing

Table 4.2 : Critical reactions, their sensitivity coefficients, the sources of the cross-section data in UKACT1 and the fractional errors adopted for error analysis.

Wall	Product	Reaction	Sens. Coeff.	Source	Error
C	Be10	C13 (n, α)Be10	+0.998	T-GA64A	0.47
"	C 14	C13 (n, γ)C14	1.000	ACTL	0.30
N	Be10	B11 (n, d)Be10	0.156	T-ENDF	0.30
"	"	C13 (n, α)Be10	0.794	T-GA64A	0.47
"	"	C14 (n, $n\alpha$)Be10	0.044	T-R	0.75
"	"	N14 (n,2n)N13	0.038	T-ENDF/QA81	0.47
"	"	N14 (n,p)C14	0.046	T-ENDF/QA81	0.42
"	"	N14 (n,d)C13	0.751	T-ENDF	0.77
"	"	N14 (n, α)B11	0.154	T-ENDF	0.47
N	C 14	N14 (n,p)C14	0.995	T-ENDF/QA81	0.42
Al	C 14	O17 (n, α)C14	0.987	T-ENDF	0.47
"	"	O18 (n, $n\alpha$)C14	0.018	T-R	0.75
"	"	Ne20 (n, α)O17	0.964	T-GA64	0.47
"	"	Ne21 (n, $n\alpha$)O17	0.033	T-P	0.75
"	"	Ne21 (n, α)O18	0.019	T-P	0.47
"	"	Na23 (n, α)F20	0.188	T-ENDF/PE84	0.47
"	"	Mg24 (n, $n\alpha$)Ne20	0.767	T-P	0.75
"	"	Mg24 (n, α)Ne21	0.051	T-ACTL/GA64	0.47
"	"	Al27 (n, α)Na23	0.185	T-QA82	0.47
"	"	Al27 (n, α)Na24	0.406	T-ENDF/QA81	0.47
"	Al26	Al27 (n,2n)Al26	1.000	T-SM84	0.30
Si	Al26	Al27 (n,2n)Al26	1.000	T-SM84	0.30
"	"	Si28 (n,d)Al27	0.972	T-HE82	0.77
"	Si32	Si31 (n, γ)Si32	0.562	ACTL	0.30
"	Si32	Si30 (n, γ)Si31	1.000	ACTL	0.30
"	Si32	P31 (n, γ)P32	0.410	Est.-N13	0.30
"	Si32	P32 (n,p)Si32	0.409	T-P	0.42
K	Cl36	K39 (n, α)Cl36	0.997	T-ACTL	0.47
"	Ar39	K39 (n,p)Ar39	0.991	T-ACTL/QA81	0.42
Ca	Cl36	Ar36 (n,p)Cl36	0.080	T-ACTL/RCN2	0.42
"	"	Ar37 (n,d)Cl36	0.138	T-P	0.77
"	"	K39 (n, α)Cl36	0.633	T-ACTL	0.47
"	"	K40 (n, $n\alpha$)Cl36	0.044	T-P	0.75
"	"	Ca40 (n, $n\alpha$)Ar36	0.081	T-ACTL	0.75
"	"	Ca40 (n,p)K40	0.038	T-ACTL	0.094
"	"	Ca40 (n,d)K39	0.630	T-R	0.77
"	"	Ca40 (n, α)Ar37	0.238	T-ACTL/QA81	0.47
Ca	Ar39	K39 (n,p)Ar39	0.082	T-ACTL/QA81	0.42
"	"	K40 (n,d)Ar39	0.096	T-ACTL/QA81	0.75

"	"	Ca40 (n,p)K40	0.090	T-ACTL	0.094
"	"	Ca40 (n,d)K39	0.078	T-R	0.77
"	"	Ca40 (n,2p)Ar39	0.799	T-R	0.75
"	"	Ca42 (n, α)Ar39	0.019	T-P	0.42
"	Ar42	Ca43 (n,2p)Ar42	0.982	T-P	0.75
"	Ca41	Ca40 (n, γ)Ca41	0.965	ACTL	0.30
"	"	Ca42 (n,2n)Ca41	0.034	T-P	0.30
Ti	Ar42	Ca43 (n,2p)Ar42	0.123	T-P	0.75
"	"	Ca45 (n, α)Ar42	0.861	T-P	0.47
"	"	Ca46 (n, α)Ar42	0.013	T-P	0.75
"	"	Ti46 (n, α)Ca43	0.123	ACTL	0.47
"	"	Ti48 (n, α)Ca45	0.846	T-ENDF/QA82	0.261
"	"	Ti49 (n, α)Ca46	0.011	T-ACTL	0.47
V	Ar42	Ca45 (n, α)Ar42	0.976	T-P	0.47
"	"	Ca46 (n, α)Ar42	0.025	T-P	0.75
"	"	Ti48 (n, α)Ca45	0.972	T-ENDF/QA82	0.261
"	"	Ti49 (n, α)Ca46	0.020	T-ACTL	0.47
"	"	V51 (n, α)Sc48	0.970		0.047
"	Ti44	Ti45 (n,2n)Ti44	1.000	T-H	0.30
"	"	Ti46 (n,2n)Ti45	1.000	T-ACTL	0.30
"	"	Ti47 (n,2n)Ti46	0.816	T-ACTL	0.30
"	"	V50 (n, α)Sc47	0.091	T-ACTL	0.47
Cr	Ar42	Ca43 (n,2p)Ar42	0.013	T-P	0.75
"	"	Ca45 (n, α)Ar42	0.121	T-P	0.47
"	"	Ca46 (n, α)Ar42	0.870	T-P	0.75
"	"	Ti46 (n, α)Ca43	0.010	ACTL	0.47
"	"	Ti48 (n, α)Ca45	0.072	T-ENDF/QA82	0.261
"	"	Ti49 (n,2n)Ti48	0.030	T-ACTL	0.30
"	"	Ti49 (n, α)Ca46	0.897	T-ACTL	0.47
"	"	Cr50 (n, α)Ti47	0.028	T-AP	0.222
"	"	Cr52 (n, α)Ti48	0.019	T-AP	0.75
"	"	Cr52 (n, α)Ti49	0.794	T-AP	0.242
"	Ti44	Ti45 (n,2n)Ti44	1.000	T-H	0.30
"	"	Ti46 (n,2n)Ti45	1.000	T-ACTL	0.30
"	"	Ti47 (n,2n)Ti46	0.259	T-ACTL	0.30
"	"	Cr50 (n, α)Ti46	0.629	T-AP	0.75
"	"	Cr50 (n, α)Ti47	0.283	T-AP	0.222
Mn	Mn53	Mn54 (n,2n)Mn53	0.992	T-P	0.30
"	"	Mn55 (n,2n)Mn54	0.992	T-ENDF	0.30
"	"	Mn55 (n, γ)Mn56	-0.023	ENDF	0.30
"	Fe55	Mn55 (n, γ)Mn56	0.979	ENDF	0.30
"	"	Fe56 (n,2n)Fe55	0.995	LANL/QA81	0.077
"	Co60	Mn55 (n, γ)Mn56	0.990	ENDF	0.30
"	"	Fe56 (n, γ)Fe57	1.020	ACTL	0.30
"	"	Fe57 (n, γ)Fe58	1.030	ACTL	0.30
"	"	Fe58 (n, γ)Fe59	1.030	ENDF	0.30

"	"	Co59 (n, γ)Co60	0.664	ENDF	0.30
"	"	Co59 (n, γ)Co60m	0.290	ENDF	0.30
Fe	Mn53	Fe54 (n,d)Mn53	0.981	T-P	0.77
Fe	Fe55	Fe54 (n, γ)Fe55	0.040	ENDF	0.30
"	"	Fe56 (n,2n)Fe55	0.954	LANL/QA81	0.30
"	Co60	Fe58 (n, γ)Fe59	0.998	ENDF	0.30
"	"	Co59 (n, γ)Co60	0.615	ENDF/ACTL	0.30
"	"	Co59 (n, γ)Co60m	0.269	ENDF/ACTL	0.30
Co	Co60	Co59 (n, γ)Co60	0.564	ENDF/ACTL	0.30
"	"	Co59 (n, γ)Co60m	0.247	ENDF/ACTL	0.30
"	"	Co60 (n, γ)Co61	-0.014	ACTL	0.30
"	Ni59	Co59 (n, γ)Co60	0.624	ENDF/ACTL	0.30
"	"	Co59 (n, γ)Co60m	0.280	ENDF/ACTL	0.30
"	"	Ni60 (n,2n)Ni59	1.000	T-ENDF	0.240
"	Ni63	Co59 (n, γ)Co60	0.644	ENDF/ACTL	0.30
"	"	Co59 (n, γ)Co60m	0.282	ENDF/ACTL	0.30
"	"	Co60 (n, γ)Co61	0.989	ACTL	0.30
"	"	Ni60 (n, γ)Ni61	0.012	ACTL	0.30
"	"	Ni61 (n, γ)Ni62	1.020	ACTL	0.30
"	"	Ni62 (n, γ)Ni63	1.010	ENDF	0.30
Ni	Co60	Ni60 (n,p)Co60	0.110	T-ACTL/QA81	0.384
"	"	Ni60 (n,p)Co60m	0.611	T-ACTL/QA81	0.384
"	Ni59	Ni58 (n, γ)Ni59	0.611	ENDF	0.30
"	"	Ni60 (n,2n)Ni59	0.386	T-ENDF	0.240
"	Ni63	Ni62 (n, γ)Ni63	0.665	ENDF	0.30
"	"	Ni64 (n,2n)Ni63	0.325	T-ACTL	0.067
Cu	Co60	Cu63 (n, α)Co60	0.346	T-ACTL/QA81	0.051
"	"	Cu63 (N, α)Co60m	0.610	T-ACTL/QA81	0.051
"	Ni63	Cu63 (n,p)Ni63	0.932	T-ACTL/QA81	0.25
Zr	Sr90	Zr91 (n,2p)Sr90	0.069	T-R	0.75
"	"	Zr92 (n, γ)Zr93	0.022	ACTL	0.30
"	"	Zr93 (n, α)Sr90	0.228	T-ACTL	0.47
"	"	Zr94 (n,2n)Zr93	0.190	T-ENDF	0.30
"	"	Zr94 (n,n α)Sr90	0.697	T-ACTL	0.75
"	Zr93	Zr92 (n, γ)Zr93	0.098	ACTL	0.30
"	"	Zr94 (n,2n)Zr93	0.884	T-ENDF	0.30
"	Nb94	Zr94 (n, γ)Zr95	0.205	ACTL	0.30
"	"	Zr96 (n,2n)Zr95	0.779	T-ENDF/QA81	0.30
"	"	Nb95 (n,2n)Nb94	0.395	T-P	0.30
"	"	Nb95 (n,2n)Nb94m	0.394	T-P	0.30
"	"	Mo95 (n,d)Nb94	0.103	T-QA81	0.77
"	"	Mo95 (n,d)Nb94m	0.102	T-QA81	0.30
"	"	Nb93 (n, γ)Nb94	0.481	ACTL	0.30
"	"	Nb93 (n, γ)Nb94m	0.471	Est-Nb93	0.30

Nb	Nb94	Nb93 (n,γ)Nb94	0.481	ACTL	0.30
"	"	Nb93 (n,γ)Nb94m	0.471	Est-Nb93	0.30
Mo	Tc99	Mo98 (n,γ)Mo99	0.704	ACTL	0.30
"	"	Mo100 (n,2n)Mo99	0.276	T-ACTL/QA81	0.30
"	Nb94	Mo94 (n,p)Nb94	0.417	T-ACTL	*
"	"	Mo94 (n,p)Nb94m	0.417	Est-Mo94	
"	"	Nb94 (n,γ)Nb95	-0.137	ACTL	
"	Mo93	Mo92 (n,γ)Mo93	0.646	ACTL	
"	"	Mo94 (n,2n)Mo93	0.332	T-ACTL	
"	"	Mo93 (n,γ)Mo94	-0.100	ACTL	
"	Nb91	Mo92 (n,d)Nb91	0.781	T-ACTL	
"	"	Mo92 (n,2n)Mo91	0.159	T-ACTL/QA81	
"	"	Mo92 (n,d)Nb91m	0.033	T-ACTL	
Ag	Ag108m	Ag107 (n,γ)Ag108m	0.910	ACTL	0.30
"	"	Ag109 (n,2n)Ag108m	0.071	T-ACTL	0.30
"	"	Ag109 (n,γ)Ag110	-0.042	ACTL	0.30
Ag	Cd113m	Ag109 (n,γ)Ag110	0.627	ACTL	0.30
"	"	Ag109 (n,γ)Ag110m	0.108	ACTL	0.30
"	"	Ag110m (n,γ)Ag111	0.046	ACTL	0.30
"	"	Ag110m (n,γ)Ag111m	0.046	ACTL	0.30
"	"	Cd110 (n,γ)Cd111	0.427	JEF1/NJOY	0.30
"	"	Cd110 (n,γ)Cd111m	0.427	JEF1/NJOY	0.30
"	"	Cd111 (n,γ)Cd112	0.956	JEF1/NJOY	0.30
"	"	Cd112 (n,γ)Cd113m	0.985	JEF1/NJOY	0.30
"	"	Cd113 (n,n')Cd113m	0.009	T-H	0.75
Sn	Ag108m	Cd108 (n,p)Ag108m	0.077	T-P	0.30
"	"	Cd109 (n,d)Ag108m	0.044	T-H	0.30
"	"	Sn112 (n,α)Cd109	0.812	T-P	0.47
"	"	Cd109 ?			
"	Cd113m	Cd113 (n,n')Cd113m	0.019	T-H	0.75
"	"	Sn116 (n,α)Cd113m	0.790	T-P	0.30
"	Sn121m	Sn120 (n,γ)Sn121m	0.661	JEF1/NJOY	0.30
"	"	Sn122 (n,2n)Sn121m	0.332	T-QA81	0.30
"	Sn126	Sn124 (n,γ)Sn125	0.992	JEF1/NJOY	0.30
"	"	Sn125 (n,γ)Sn126	0.961	JEF1/NJOY	0.30
Ce	Cs137	Cs136 (n,γ)Cs137	0.307	JEF1/NJOY	0.30
"	"	Ba137 (n,p)Cs137	0.566	T-P	0.42
"	"	La138 (n,2p)Cs137	0.011	T-R	0.77
"	"	La139 (n,h)Cs137	0.103	T-P	0.52
"	"	La139 (n,α)Cs136	0.305	T-QA84	0.47
"	"	Ce140 (n,2n)Ce139	0.206	T-QA81	0.30
"	"	Ce140 (n,2n)Ce139m	0.204	T-QA81	0.30
"	"	Ce140 (n,α)Ba137	0.283	T-QA84	0.430
"	"	Ce140 (n,α)Ba137m	0.283	Est-Ce140	0.30
Nd	Sm151	Nd148 (n,γ)Nd149	0.189	JEF1/NJOY	0.30

"	"	Nd150 (n,2n)Nd149	0.088	T-QA81	0.30
"	"	Nd150 (n,γ)Nd151	0.596	JEF1/NJOY	0.30
"	"	Sm149 (n,γ)Sm150	0.134	JEF1/NJOY	0.30
"	"	Sm150 (n,γ)Sm151	0.285	JEF1/NJOY	0.30
"	"	Sm151 (n,γ)Sm152	-0.829	JEF1/NJOY	0.30
"	Eu152	Nd148 (n,γ)Nd149	0.088	JEF1/NJOY	0.30
"	"	Nd150 (n,2n)Nd149	0.036	T-QA81	0.30
"	"	Nd150 (n,γ)Nd151	0.812	JEF1/NJOY	0.30
"	"	Sm149 (n,γ)Sm150	0.084	JEF1/NJOY	0.30
"	"	Sm150 (n,γ)Sm151	0.140	JEF1/NJOY	0.30
"	"	Sm151 (n,γ)Sm152	0.080	JEF1/NJOY	0.30
"	"	Sm152 (n,γ)Sm153	0.337	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152	0.191	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152m	-0.102	JEF1/NJOY	0.30
"	"	Eu152 (n,γ)Eu153	-0.631	JEF1/NJOY	0.30
"	"	Eu153 (n,γ)Eu154	-0.322	JEF1/NJOY	0.30
"	Eu154	Nd148 (n,γ)Nd149	0.074	T-QA81	0.30
"	"	Nd150 (n,2n)Nd149	0.291	JEF1/NJOY	0.30
Nd	Eu154	Nd150 (n,γ)Nd151	0.841	JEF1/NJOY	0.30
"	"	Sm149 (n,γ)Sm150	0.073	JEF1/NJOY	0.30
"	"	Sm150 (n,γ)Sm151	0.199	JEF1/NJOY	0.30
"	"	Sm151 (n,γ)Sm152	0.415	JEF1/NJOY	0.30
"	"	Sm152 (n,γ)Sm153	0.491	JEF1/NJOY	0.30
"	"	Eu153 (n,γ)Eu154	0.541	JEF1/NJOY	0.30
"	"	Eu154 (n,γ)Eu155	-0.485	JEF1/NJOY	0.30
Sm	Eu152	Sm150 (n,γ)Sm151	0.165	JEF1/NJOY	0.30
"	"	Sm151 (n,γ)Sm152	0.017	JEF1/NJOY	0.30
"	"	Sm152 (n,γ)Sm153	0.058	JEF1/NJOY	0.30
"	"	Sm154 (n,2n)Sm153	0.039	T-QA81	0.30
"	"	Eu151 (n,γ)Eu152	0.046	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152m	-0.025	JEF1/NJOY	0.30
"	"	Eu152 (n,γ)Eu153	-0.944	JEF1/NJOY	0.30
"	"	Eu153 (n,2n)Eu152	0.923	T-QA81	0.30
"	"	Eu153 (n,γ)Eu154	-0.792	JEF1/NJOY	0.30
"	Eu154	Sm149 (n,γ)Sm150	0.035	JEF1/NJOY	0.30
"	"	Sm150 (n,γ)Sm151	0.111	JEF1/NJOY	0.30
"	"	Sm151 (n,γ)Sm152	0.059	JEF1/NJOY	0.30
"	"	Sm152 (n,γ)Sm153	0.102	JEF1/NJOY	0.30
"	"	Sm154 (n,2n)Sm153	0.039	JEF1/NJOY	0.30
"	"	Eu153 (n,γ)Eu154	0.199	JEF1/NJOY	0.30
"	"	Eu154 (n,γ)Eu155	-0.820	JEF1/NJOY	0.30
"	"	Eu155 (n,2n)Eu154	0.014	T-R	0.30
Eu"	Sm151	Sm150 (n,γ)Sm151	0.193	JEF1/NJOY	0.30
"	"	Sm151 (n,γ)Sm152	-1.050	JEF1/NJOY	0.30
"	"	Sm152 (n,2n)Sm151	0.697	T-FR80	0.30
"	"	Eu151 (n,2n)Eu150	0.220	T-QA81	0.30
"	"	Eu151 (n,γ)Eu152	-0.430	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152m	0.111	JEF1/NJOY	0.30

"	"	Eu152 (n,γ)Eu153	-0.074	JEF1/NJOY	0.30
"	"	Eu153 (n,2n)Eu152m	0.025	JEF1/NJOY	0.30
"	"	Eu153 (n,γ)Eu154	-0.029	JEF1/NJOY	0.30
"	Eu152	Eu151 (n,2n)Eu150	-0.019	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152m	-0.952	JEF1/NJOY	0.30
"	"	Eu152 (n,γ)Eu153	-1.820	JEF1/NJOY	0.30
"	"	Eu153 (n,2n)Eu152	0.073	JEF1/NJOY	0.30
"	"	Eu153 (n,γ)Eu154	-0.082	JEF1/NJOY	0.30
"	"	Gd152 (n,2n)Gd151	0.018	T-QA81	0.30
"	Eu154	Eu151 (n,γ)Eu152	0.149	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152m	-0.068	JEF1/NJOY	0.30
"	"	Eu152 (n,γ)Eu153	0.045	JEF1/NJOY	0.30
"	"	Eu153 (n,γ)Eu154	-0.015	JEF1/NJOY	0.30
"	"	Eu154 (n,γ)Eu155	1.000	JEF1/NJOY	0.30
"	Gd150	Eu151 (n,2n)Eu150	0.992	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152	-0.524	JEF1/NJOY	0.30
"	"	Eu151 (n,γ)Eu152m	-0.524	JEF1/NJOY	0.30
"	"	Gd152 (n,2n)Gd151	0.012	JEF1/NJOY	0.30
Tb	Tb158	Tb158 (n,γ)Tb159	-0.476	JEF1/NJOY	0.30
"	"	Tb159 (n,2n)Tb158	0.738	T-QA81	0.30
"	"	Tb159 (n,2n)Tb158m	0.247	T-QA81	0.30
"	"	Tb159 (n,γ)Tb160	-0.561	JEF1/NJOY	0.30
Ho	Ho166m	Ho165 (n,2n)Ho164	-0.010	T-ACTL/QA81	0.30
"	"	Ho165 (n,γ)Ho166	-0.635	ACTL	0.30
"	"	Ho165 (n,γ)Ho166m	0.968	ACTL	0.30
"	"	Ho166m (n,γ)Ho167	-0.551	REAC	0.30
Ta	Hf178n	Hf177 (n,γ)Hf178n	0.213	ACTL	0.30
"	"	Hf178 (n,n')Hf178n	0.480	T-H	0.75
"	"	Hf178n (n,γ)Hf179	-0.207	JEF1/NJOY	0.30
"	"	Hf179 (n,2n)Hf178n	0.280	T-H	0.75
"	"	Ta180 (n,2n)Ta179	0.074	T-R	0.30
"	"	Ta181 (n,nα)Lu177m	0.249	T-ACTL	0.75
"	"	Ta181 (n,γ)Ta182	-0.264	ACTL	0.30
"	Hf182	Hf181 (n,γ)Hf182	0.505	REAC	0.30
"	"	Hf181 (n,γ)Hf182m	0.224	REAC	0.30
"	"	Hf182 (n,γ)Hf183	-0.024	JEF1/NJOY	0.30
"	Re186m	Ta181 (n,γ)Ta182	0.829	ACTL	0.30
"	"	Ta182 (n,γ)Ta183	0.500	JEF1/NJOY	0.30
"	"	W182 (n,γ)W183	0.171	ACTL	0.30
"	"	W182 (n,γ)W183m	0.171	Est-W182	0.30
"	"	W183 (n,γ)W184	0.899	ACTL	0.30
"	"	W184 (n,γ)W185	0.531	ACTL	0.30
"	"	W184 (n,γ)W185m	0.531	Est-W184	0.30
"	"	Re185 (n,γ)Re186m	0.961	Est-Re185	0.30
"	"	Re186m (n,γ)Re187	-0.218	REAC	0.30
W	Hf178n	Hf178 (n,n')Hf178n	0.357	T-H	0.30

"	"	Hf178n(n,γ)Hf179	-0.244	JEF1/NJOY	0.30
"	"	Hf179(n,2n)Hf178n	0.272	T-H	0.75
"	"	W181(n,α)Hf178	0.011	T-P	0.47
"	"	W181(n,α)Hf178n	0.029	T-P	0.75
"	"	W182(n,nα)Hf178	0.121	T-H	0.75
"	"	W182(n,nα)Hf178n	0.261	T-H	0.75
"	"	W182(n,α)Hf179	0.051	T-ACTL	0.47
"	Re186m	W184(n,γ)W185	0.092	ACTL	0.30
"	"	W184(n,γ)W185m	0.092	Est-W184	0.30
"	"	W186(n,2n)W185	0.041	T-ENDF/QA81	0.30
"	"	W186(n,2n)W185m	0.016	Est-W186	0.30
"	"	W186(n,γ)W187	0.508	ACTL	0.30
"	"	Re185(n,γ)Re186m	0.248	Est-Re185	0.30
"	"	Re187(n,2n)Re186	0.723	T-P	0.30
"	"	Re187(n,γ)Re188	-0.280	ACTL	0.30
"	Ir192n	W186(n,γ)W187	0.880	ACTL	0.30
"	"	Re187(n,γ)Re188	0.833	ACTL	0.30
"	"	Os188(n,γ)Os189	0.510	Est-W182	0.75
"	"	Os188(n,γ)Os189m	0.510	Est-W182	0.75
"	"	Os189(n,γ)Os190	0.990	Est-W183	0.75
"	"	Os190(n,γ)Os191	0.519	Est-W182	0.75
"	"	Os190(n,γ)Os191m	0.519	Est-W182	0.75
"	"	Ir191(n,γ)Ir192n	0.690	Est-Ta181	0.75
Ir	Ir192n	Ir191(n,γ)Ir192	0.018	Est-Ta181	0.75
"	"	Ir191(n,γ)Ir192n	0.488	Est-Ta181	0.75
"	"	Ir192(n,n')Ir192n	0.451	T-H	0.75
"	"	Ir192n(n,γ)Ir193	-0.039	ACTL	0.30
"	"	Ir192n(n,γ)Ir193m	-0.039	ACTL	0.30
"	"	Ir193(n,2n)Ir192	0.021	T-P	0.30
"	"	Ir193(n,2n)Ir192n	0.052	T-H	0.30
"	"	Ir193(n,γ)Ir194	-0.041	Est-Ta181	0.75
"	Pt193	Ir191(n,γ)Ir192	0.272	Est-Ta181	0.75
"	"	Ir193(n,2n)Ir192	0.039	T-P	0.30
"	"	Ir193(n,γ)Ir194	0.074	Est-Ta181	0.30
"	"	Pt192(n,γ)Pt193	0.333	Est-W182	0.75
"	"	Pt192(n,γ)Pt193m	0.333	Est-W182	0.75
"	"	Pt193(n,γ)Pt194	-0.282	Est-W183	0.75
"	"	Pt194(n,2n)Pt193	0.075	T-R	0.30
"	"	Pt194(n,2n)Pt193m	0.075	T-R	0.30
Pb	Pb205	Pb206(n,2n)Pb205	0.968	T-ACTL	0.30
"	"	Pb208(n,γ)Pb209	1.000	ACTL	0.30
"	"	Pb209(n,γ)Pb210	0.949	ACTL	0.30
"	Bi207	Pb208(n,γ)Pb209	1.000	ACTL	0.30
"	"	Bi208(n,2n)Bi207	0.970	T-R	0.30
"	"	Bi209(n,2n)Bi208	0.983	T-FR80	0.30
"	Po210	Pb208(n,γ)Pb209	1.000	ACTL	0.30
"	"	Bi209(n,2n)Bi208	-0.256	T-FR80	0.30
"	"	Bi209(n,γ)Bi210	0.994	JEF1/NJOY	0.30

Bi	Bi207	Bi207 (n,2n)Bi206	-0.022	T-R	0.30
"	"	Bi208 (n,2n)Bi207	0.962	T-R	0.30
"	"	Bi209 (n,2n)Bi208	0.975	T-R	0.30

Data Sources : this information has been obtained mostly from notes in the GREAC* library file.

T-P THRESH determination normalised to Petten systematics

T-H THRESH determination normalised to Harwell systematics

T-R THRESH determination normalised to systematics used for the original REAC library

T-ACTL THRESH determination normalised to an ACTL file

T-ENDF THRESH determination normalised to an ENDF file

ACTL An evaluated ACTL determination

ENDF An evaluated ENDF determination

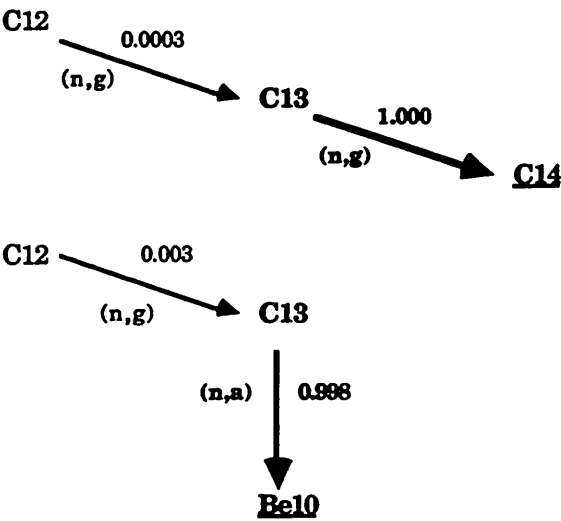
JEF An evaluated Joint European Fusion file determination

JEF/NJOY Harwell fit to capture cross-section with NJOY

For certain experimental determinations the reference code employed in the GREAC library file is given.

*REAC ECN3; H. Gruppelaar, H.A.J. Van der Kamp, J. Kopecky and D. Nierop; ECN-207 (May 1988)

Carbon Z=6



Nitrogen Z=7

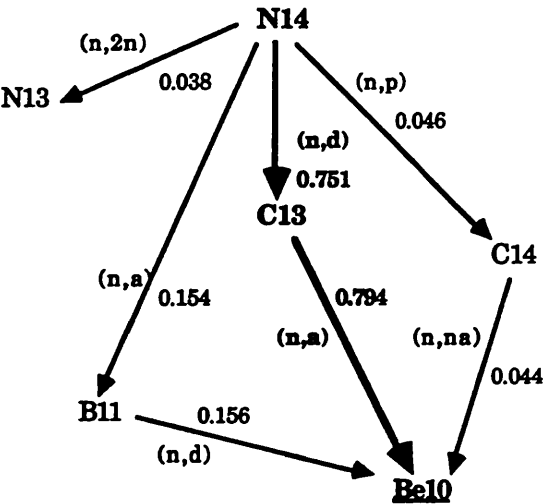


Figure 4.1 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

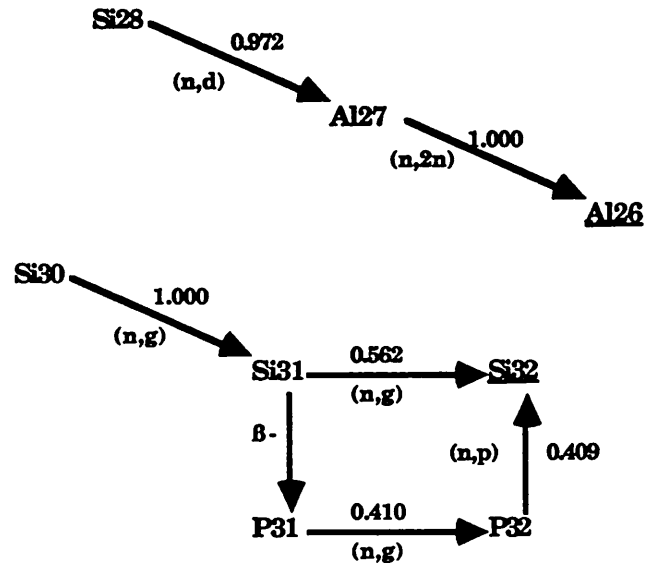
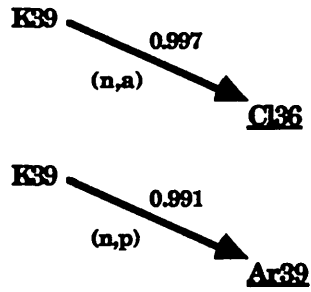
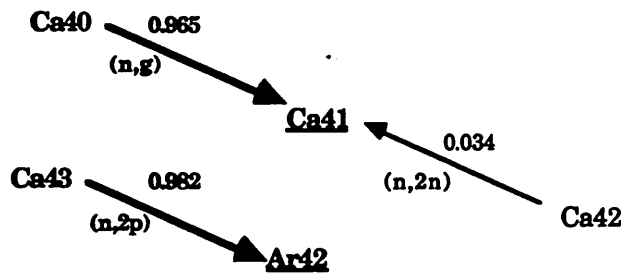
Silicon Z=14**Potassium Z=19****Calcium Z=20**

Figure 4.3 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

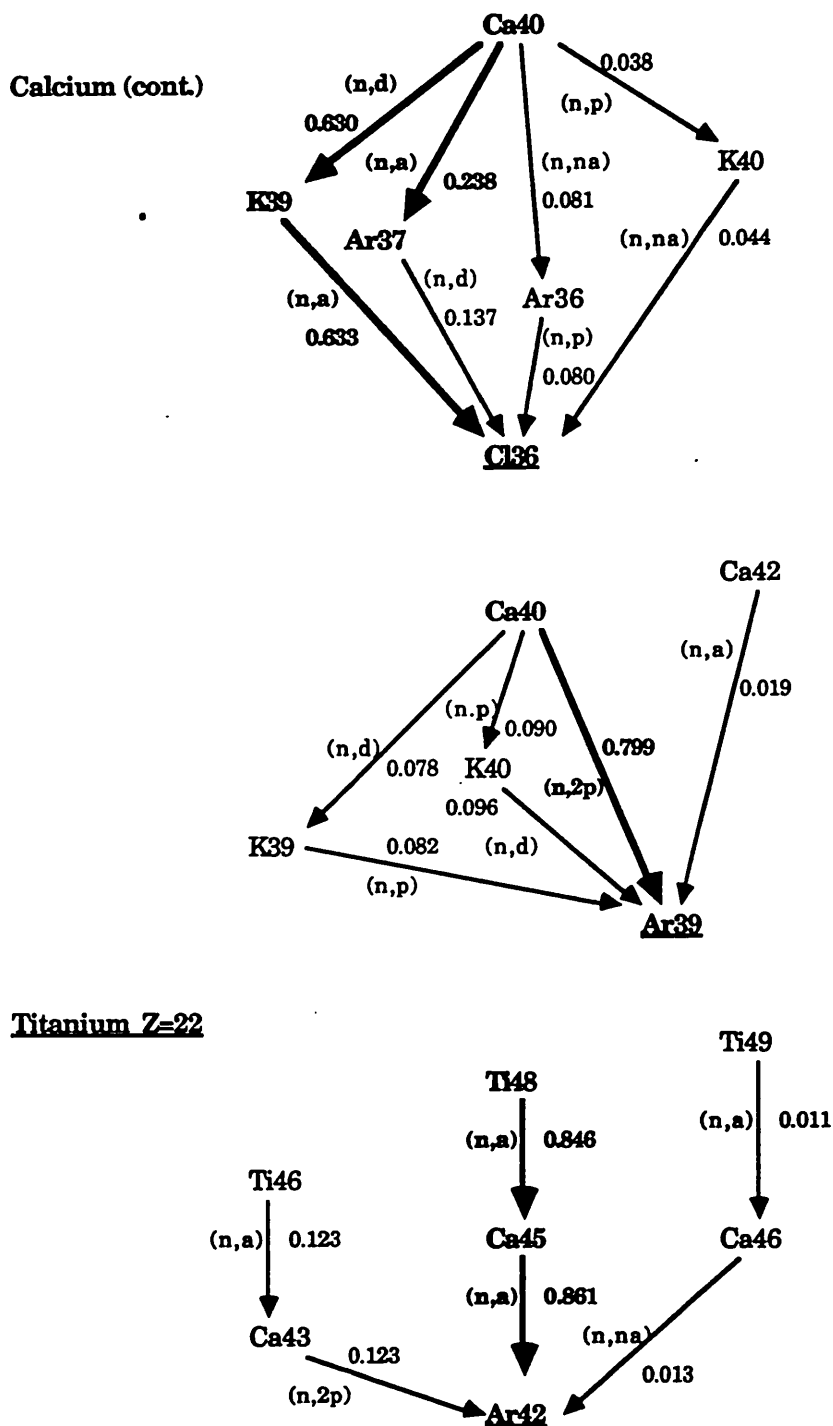


Figure 4.4 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

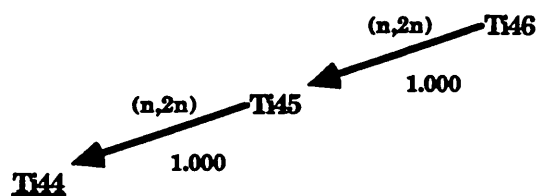
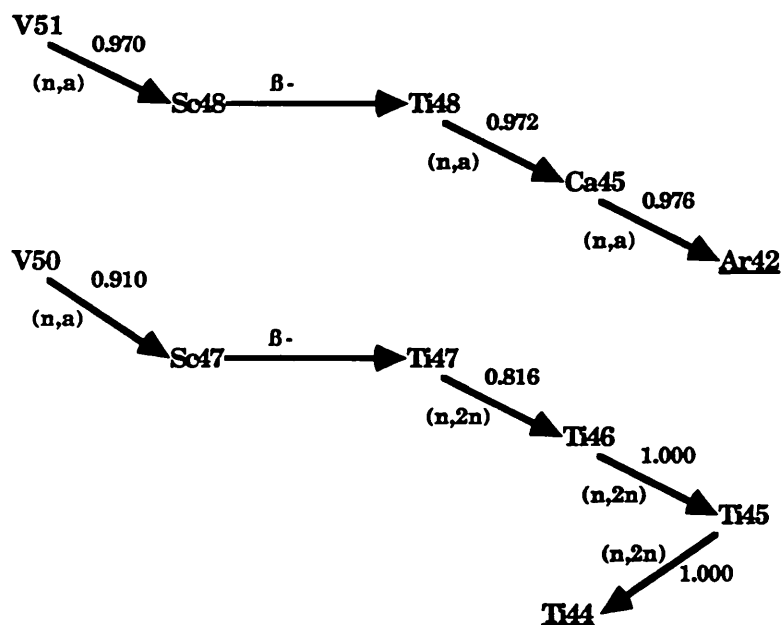
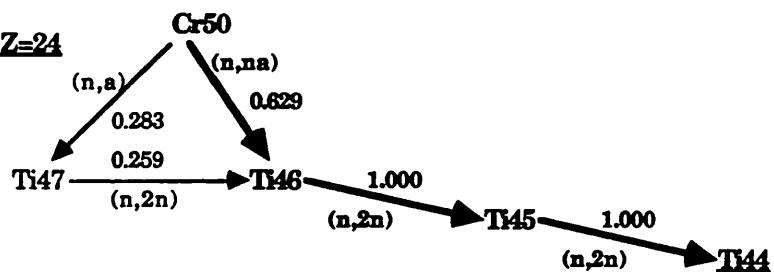
Titanium (cont.)**Vanadium Z=23****Chromium Z=24**

Figure 4.5 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

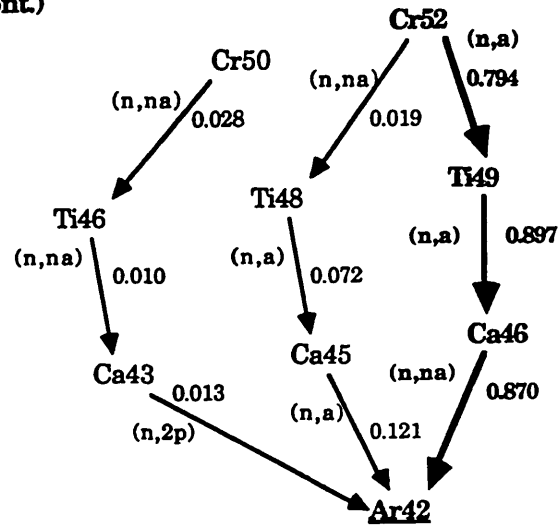
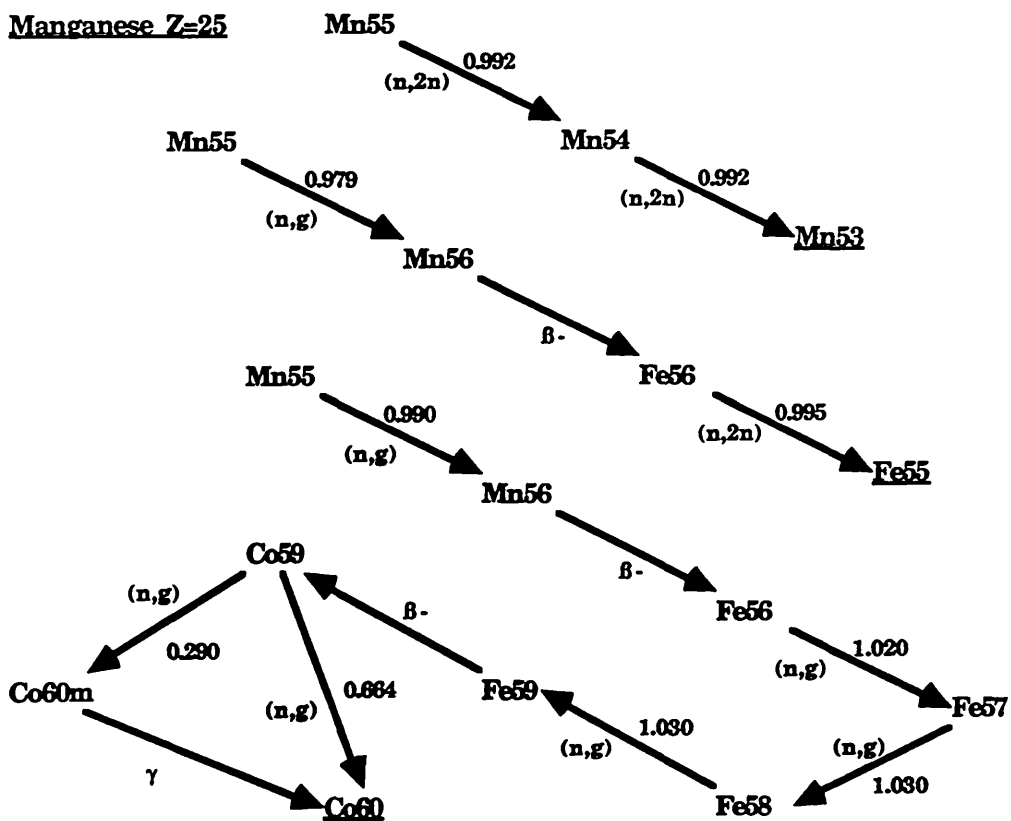
Chromium (cont.)**Manganese Z=25**

Figure 4.6 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

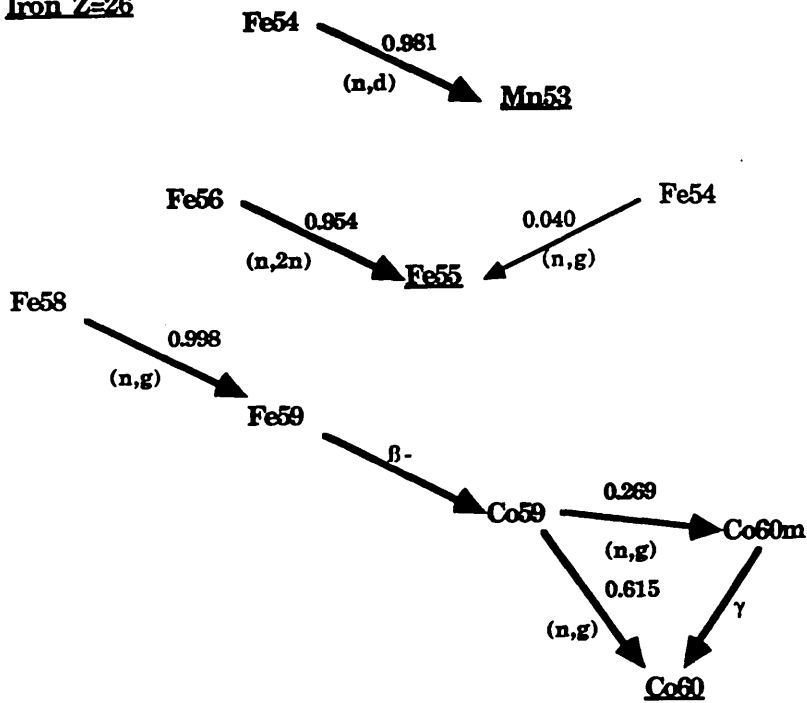
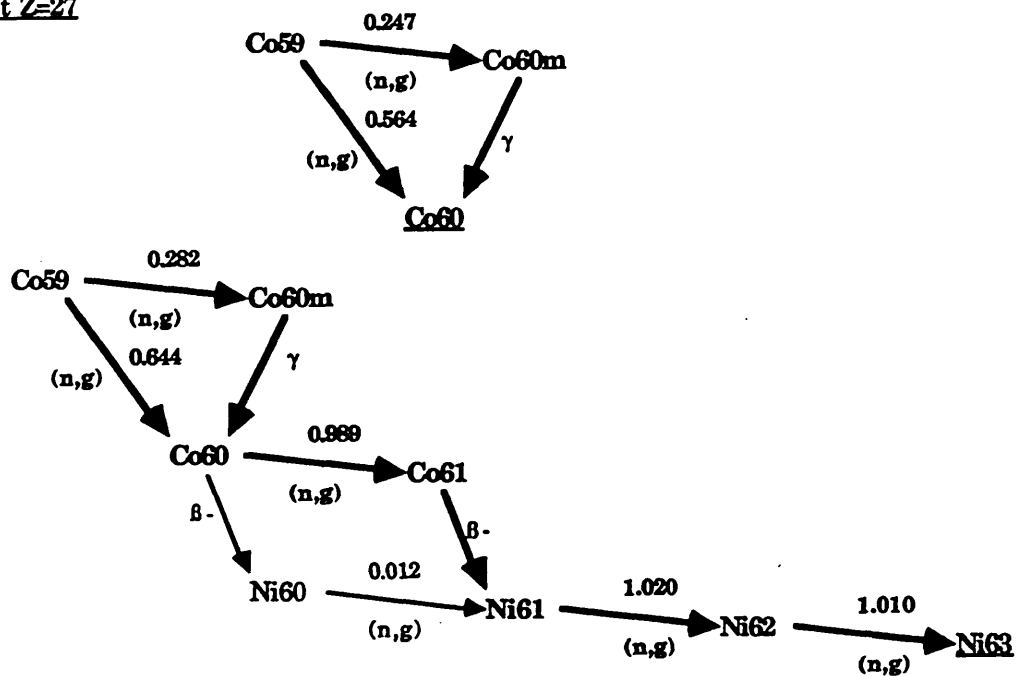
Iron Z=26Cobalt Z=27

Figure 4.7 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

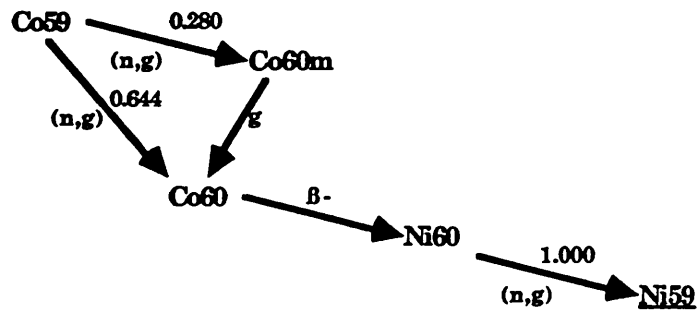
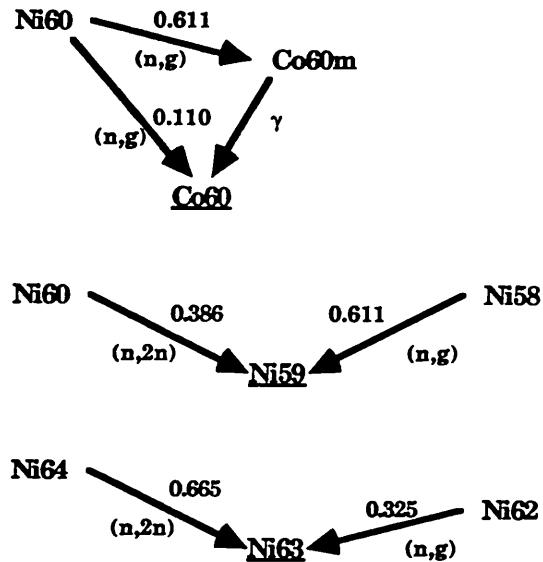
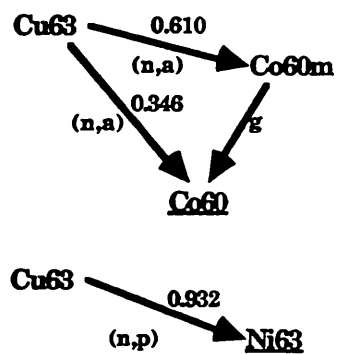
Cobalt (cont.)**Nickel Z=28****Copper Z=29**

Figure 4.8 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

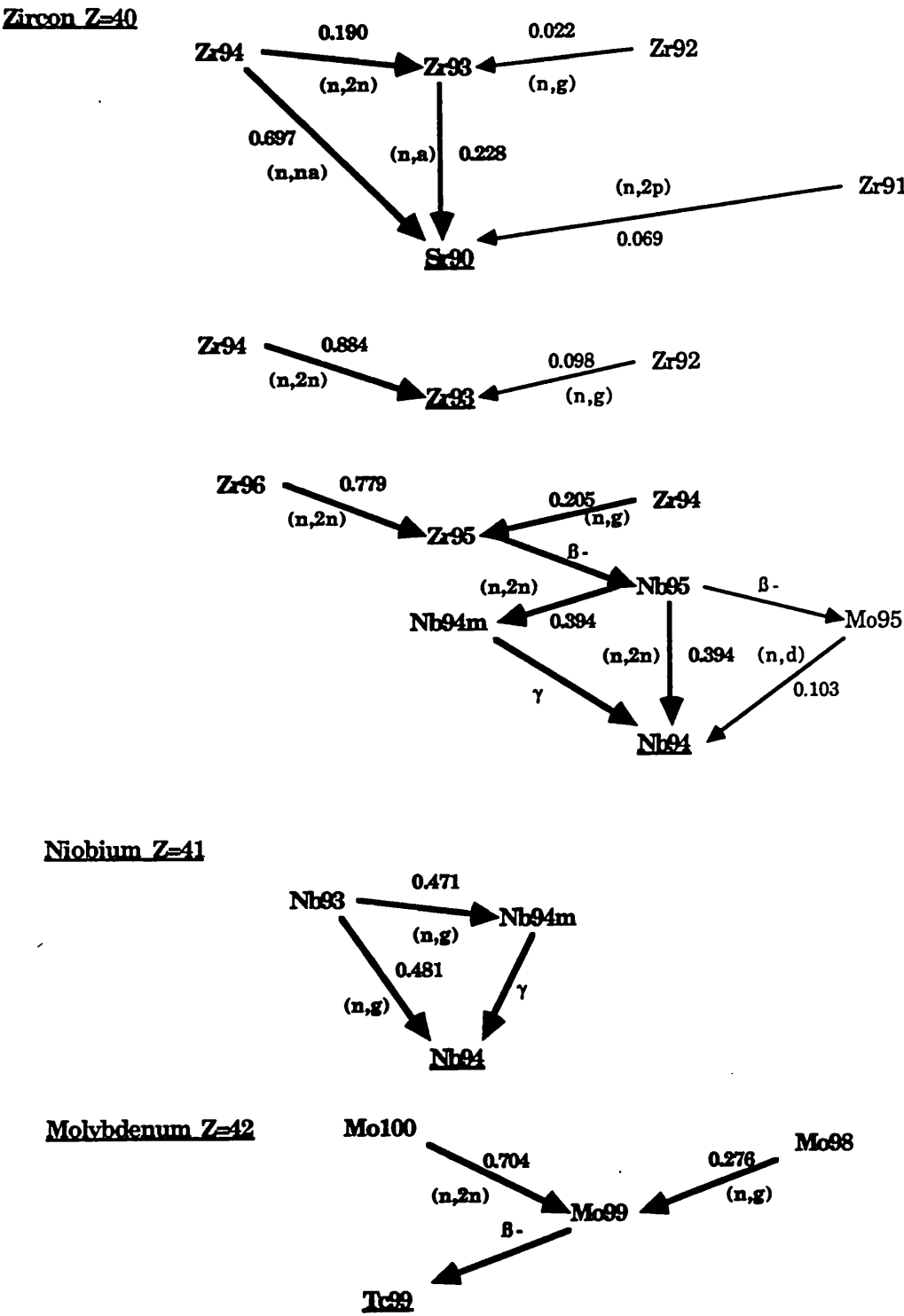


Figure 4.9 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

Molybdenum (cont.)

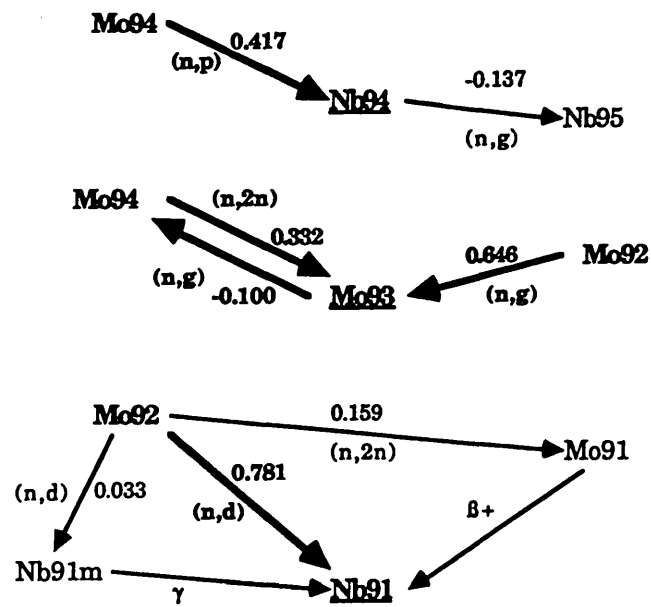
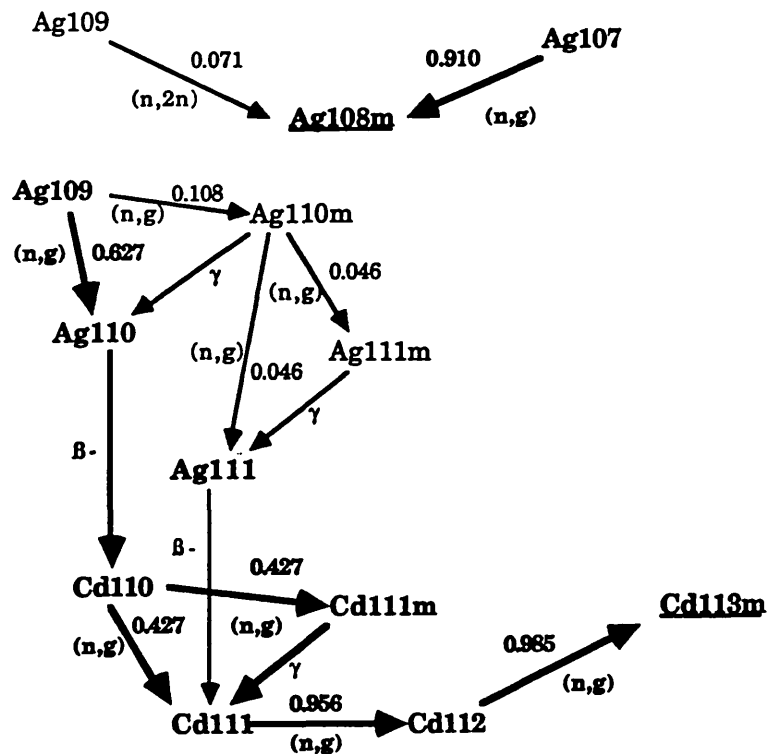
Silver Z=47

Figure 4.10 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

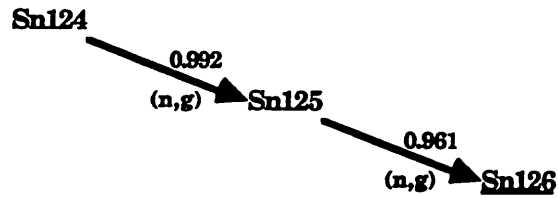
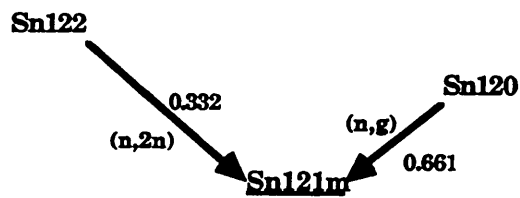
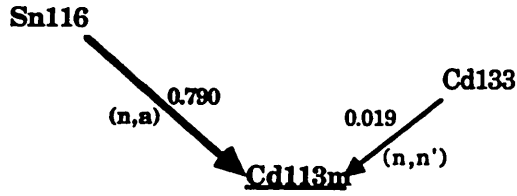
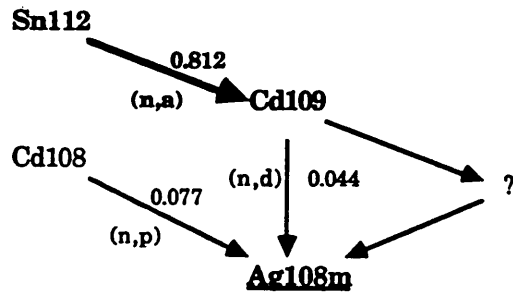
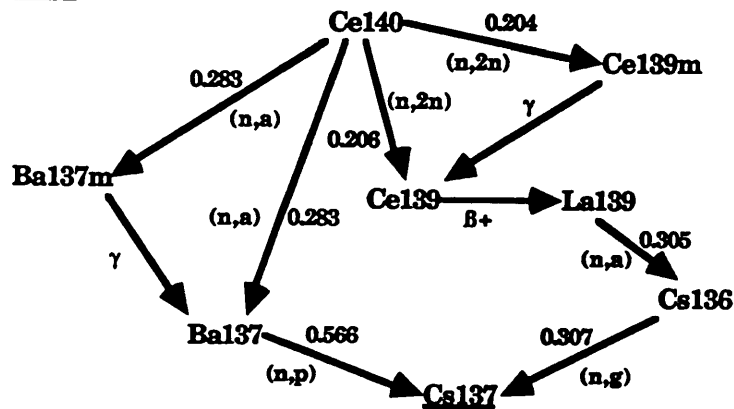
Tin Z=50**Cerium Z=58**

Figure 4.11 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

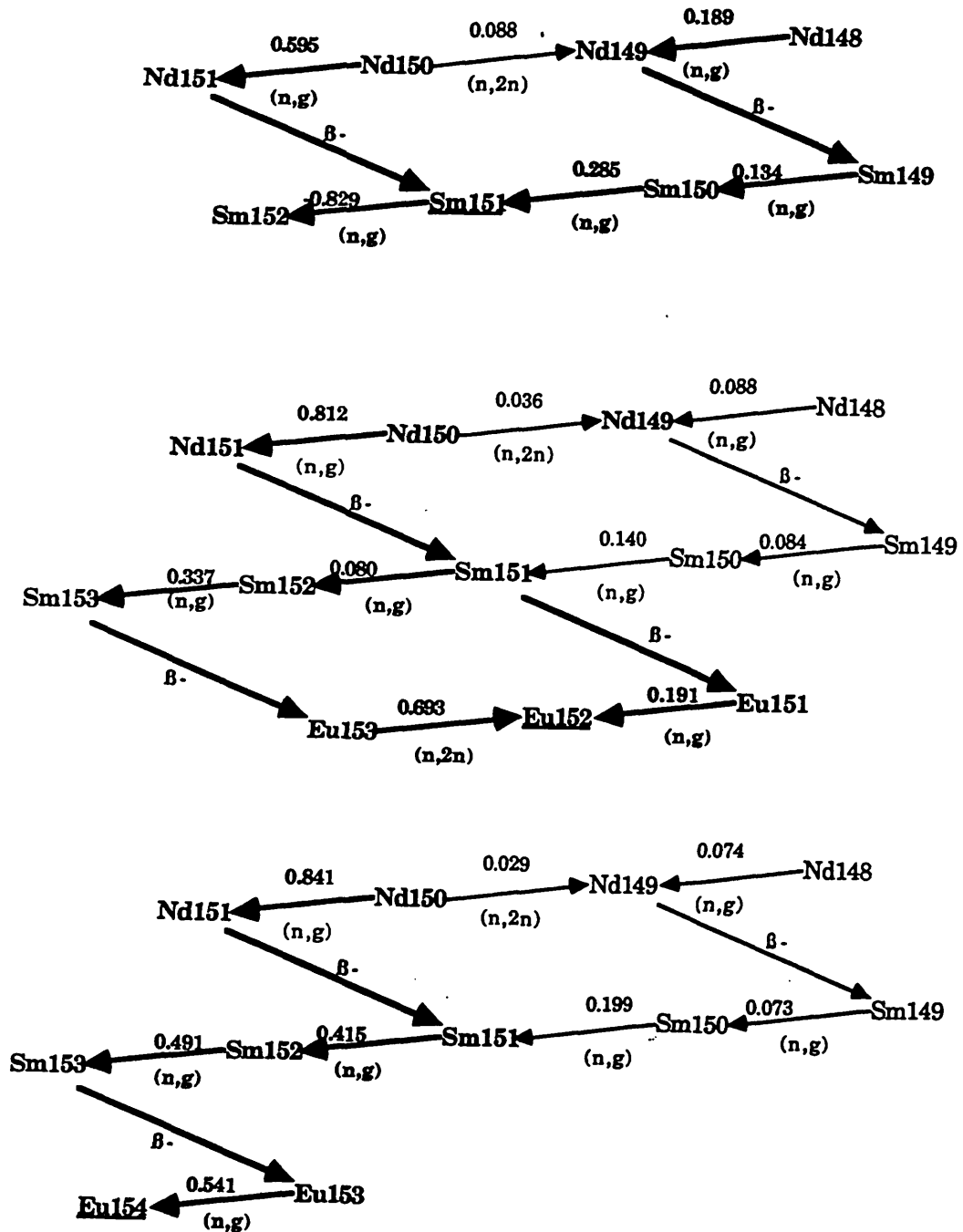
Niodymium Z=60

Figure 4.12 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

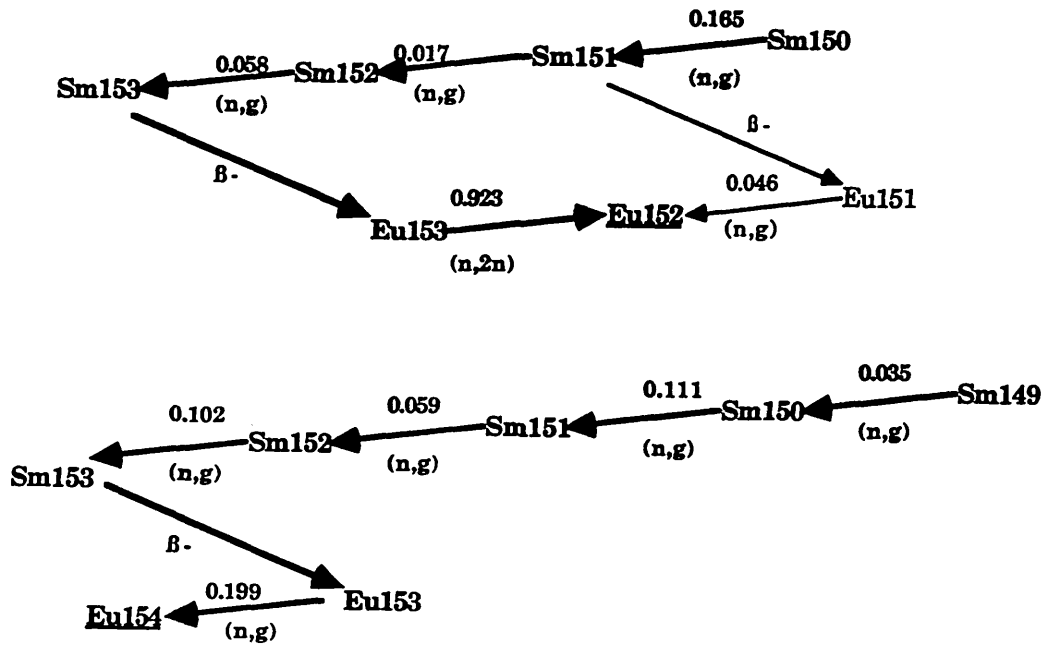
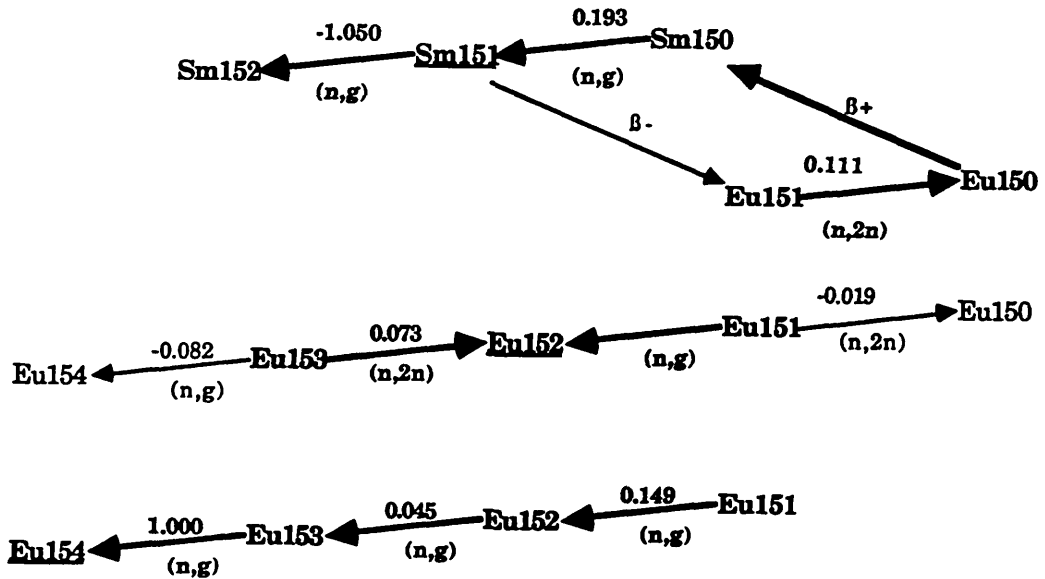
Samarium $Z=62$ Europium $Z=63$ 

Figure 4.13 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

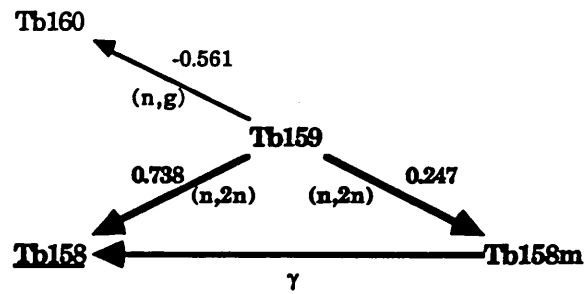
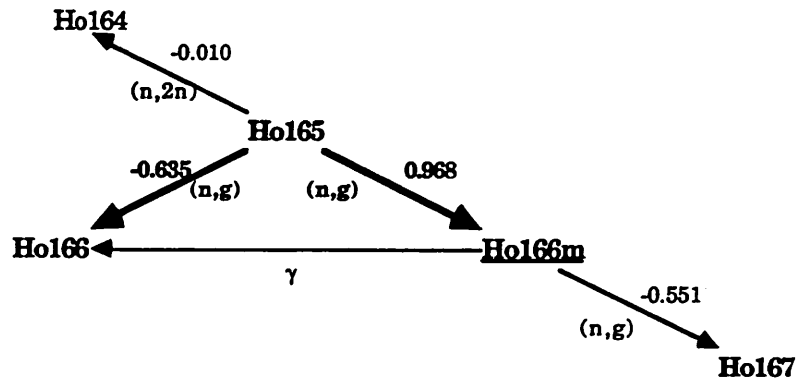
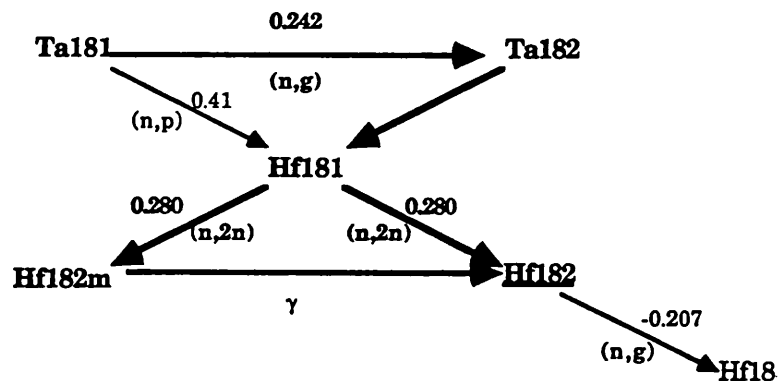
Terbium Z=65**Holmium Z=67****Tantalum Z=73**

Figure 4.14 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

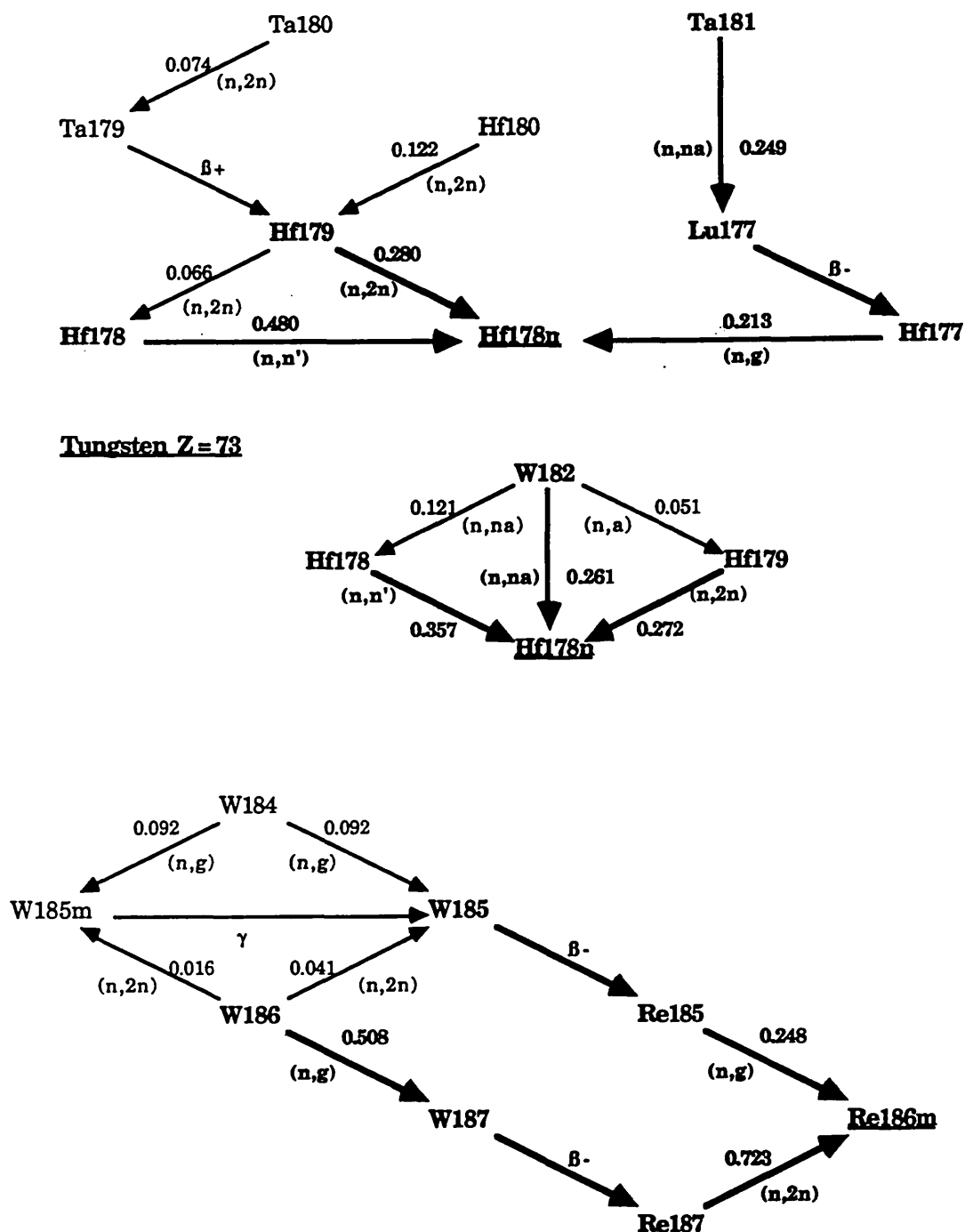


Figure 4.15 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

Tungsten (cont.)

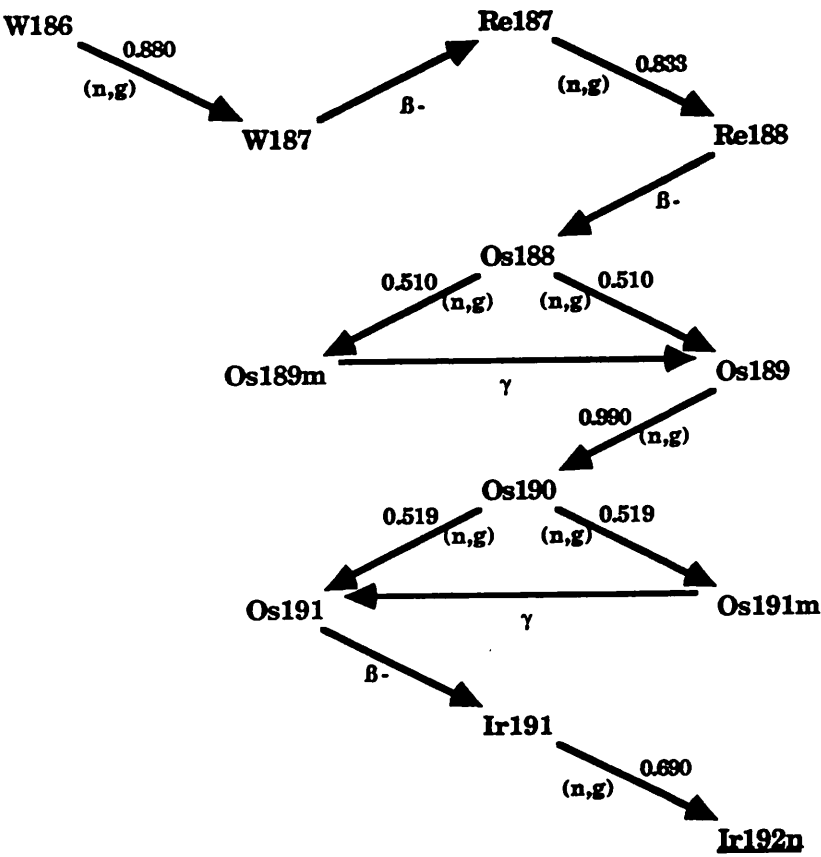


Figure 4.16 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

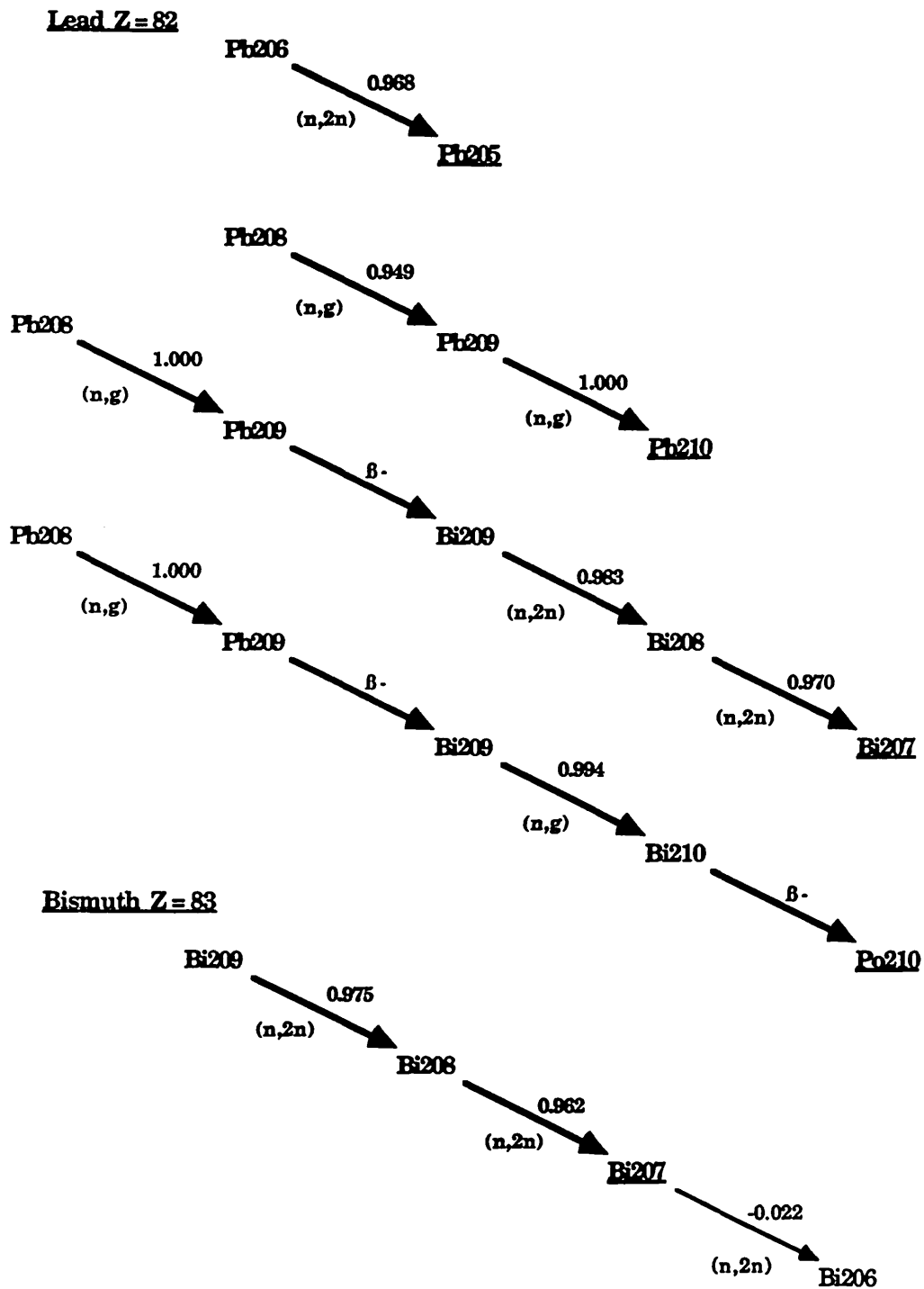


Figure 4.17 : Principal reaction pathways to long-lived radionuclides from selected source elements, and the sensitivity coefficients associated with the reaction cross-sections. Dominant pathways are in bold and the chain endpoint is underlined.

production channels, the caveat concerning burn-up expressed in Chapter II must be recalled : though a valid interpretation for the majority of sensitivity coefficients, there are a small number of reactions for which caution must be adopted. For these reactions, the sensitivity coefficients at equivalent stages of competing channels do not add up to 1. This is immediately apparent for the cases of Nd, Sm and Eu, which involve reactions with large capture cross-sections.

4.2.2 Data Sources

The plethora of experimental and theoretical methods of nuclear data evaluation employed in the preparation of the UKACT1 database constitutes a sizable body of knowledge, a detailed description of which is clearly outside the scope of this thesis. Some remarks about the principle methods employed, however, are necessary to provide a context for a discussion on the sensitivity analysis.

4.2.2.1 THRESH Evaluations

The bulk of the cross-sections of the threshold type in UKACT1 have been obtained from the semi-empirical model of nuclear reactions THRESH ¹². In essence, this fits an exponential curve, the shape of which is obtained from the statistical model of nuclear interactions ¹¹, to points (usually at 14MeV) which have been obtained either from evaluated datafiles or systematics. The quality of the generated cross-sections depends very much on the quality of the datapoints used for the fit and can vary enormously, ranging from fits normalised to a single systematic at 14MeV, to fits normalised to a set of experimental data. The sizes of the errors in the 14MeV cross-section resulting from both poor and good fits may

be gauged from the study of systematics undertaken by Forrest³⁸. It is shown, for example, that the best systematic devised for (n, α) cross-sections at 14MeV has associated with it an error factor of 1.58, corresponding to a standard deviation of 47% in the 14MeV cross-section (see Forrest³⁸ for an explanation of this). The corresponding figures for lower yielding reactions with charged particle products, such as (n,n α) and (n,d) are higher. Experimental determinations at 14MeV, however, can usually hope to carry errors in the range 5-20%, with well-measured cross-sections, such as that for Co59(n, α), below 5%. It can be shown (see section 4.3 and Appendix II) that the error at 14 MeV is, to a first order approximation, a good estimate of the error in the single group cross-section.

For a small number of well-measured reactions such as Fe(n,2n), comprehensive experimental evaluations have been performed, and the excitation functions have been obtained directly from the evaluated datafiles. Unfortunately these reactions are the exception rather than the rule.

4.2.2.2 Capture reaction evaluations

The excitation functions of capture reactions are quite different to those of the neutron-induced threshold type reactions treated by THRESH, being a more direct reflection of nuclear structure. This means that the scope for determinations from models is more limited, as no nuclear reaction model has yet been constructed which can accurately predict the shape of the excitation function in the all-important resonance energy range. The determinations present in UKACT1 generally fall into 3 types ;

a) determinations of the kind available from the evaluated datafiles such as ENDF and ACTL : thanks to the fission programme, a large number of thermal and resonance integral measurements were performed.

b) determinations obtained from processing codes such as NJOY which fit the overall shape of excitation function without the resonances, giving the $1/E$ variation at low energies and the characteristic trend in the intermediate and MeV range. Bearing in mind the conclusion in Chapter 1 which states that the single group capture cross-section is dominated for most nuclei by the contribution from resonances, and that the roles of cross-sections at high and thermal energies are usually marginal, these cross-sections should be regarded with suspicion if they do not employ experimental points in the resonance region.

c) estimations : usually taken from the nearest nucleus with a similar configuration of neutrons and protons (i.e. odd-odd, even-even or odd-even) . Since even the capture cross-sections of isotopes of the same element can vary greatly, it is not unusual for cross-sections obtained in this way to be orders of magnitude removed from the real values.

4.2.3 Discussion of important reaction channels

4.2.3.1 Carbon and Nitrogen

Both of these elements are potential alloying additions in candidate first wall materials. As well as being a candidate first wall material in its own right, carbon would be present as a major constituent in SiC and a minor constituent in some steels. Nitrogen

is being considered as a minor constituent in low-activation martensitic steels.

In the absence of strong gamma-emitters in the inventories of light elements, the long-term hazard from both of these elements would arise from the twin β -emitters, Be10 ($t_{1/2} = 1.62\text{E}+6$ years) and C14 ($t_{1/2} = 5730$ years). The production channels from carbon to these species are dominated by single (n, γ) and (n,p) reactions from C13 respectively. Both of the data for these reactions in UKACT1 have an experimental source, allowing some confidence to be attached to the evaluated concentrations. The (n, α) data were generated from THRESH and normalised to experimental points, the (n, γ) data originating from the ACTL data library.

Production of C14 from nitrogen is equally straightforward, in excess of 99% being transmuted directly by N14(n,p), the data for which is a THRESH determination normalised to ENDF points. Production of Be10 is more indirect, proceeding principally by (n,d) followed by (n, α) reactions on N14 (~ 75%), but also the secondary and tertiary routes of N14(n, α)(n,d) (~ 15%) and N14(n,p)(n, α) (~ 4%). It is noted that cross-section data employed for all of the reactions included here have been normalised to experimental measurements, with the exception of the C14(n, α) reaction, which was normalised to the systematics used in the original REAC library. The yield of the production route via C14 is so low, however, that it is doubtful that even a large error (say a factor of 5) would have much impact on the final concentration of Be10.

In conclusion, the UKACT1 data for the production of C14 and Be10 from carbon and nitrogen appears to be good and the calculated concentrations can be stated with some confidence. Possible

unknowns are the effect of secondary charged particles and the size of the $C14(n,\alpha)$ cross-section.

4.2.3.2 Aluminium and Silicon

The principal long-lived gamma emitter in the radionuclide inventories of aluminium and silicon is $Al26$ ($t_{1/2} = 7.23E+5$ years). The principal contributors to total specific ingestion and inhalation doses are $C14$ ($t_{1/2} = 5730$ years) and $P32$, the latter a short-lived daughter of $Si32$ ($t_{1/2} = 104$ years). $Al26$ would probably be a significant contributor to internal dose too, but ingestion and inhalation hazard data are not available. Over a timescale of tens of years after irradiation, $Na22$ ($t_{1/2} = 2.60$ years) is also a significant gamma-emitter.

As was noted in Chapter 1, the surface gamma dose rate from aluminium has fallen significantly in the present predictions in comparison with the earlier figures of Giancarli ¹⁰ and Jarvis ⁷, figures obtained from calculations with the UKCTR-111 database. The principal cause is a lowering, in accordance with recent measurements at 14MeV by Smithers et al ³⁹, of the single group cross-section for the $Al27(n,2n)Al26$ reaction, the dominant channel for the production of $Al26$ in the aluminium and silicon inventories. Uncertainties remain over the correct value of this cross-section, however, and another set of recent determinations ⁴⁰ reduces the cross-section further. The difficulties in obtaining accurate experimental measurements arise from the proximity of the energy threshold of the reaction to the thermally broadened distribution of primary fusion neutrons at 14MeV.

In contrast to aluminium, the surface gamma dose rate from

silicon has risen significantly in comparison with previous calculations, as a consequence of a modified treatment of the $\text{Si}^{28}(\text{n},\text{d})$ cross-section, an increase which more than compensates for the lowering of the $\text{Al}^{27}(\text{n},2\text{n})$ cross-section. Though normalised to experimental data, the data for both of these reactions should be regarded with some caution, and may change with further experimental determinations.

It is seen that the production of Si^{32} (of note because of its β^- -emitting daughter P^{32}) from silicon proceeds from Si^{31} ($t_{1/2} = 2.60$ years) via two almost equal production channels. The slightly larger route involves a capture on Si^{31} , demonstrating that nuclides with half-lives of a few hours can be transmuted in significant concentrations. The capture cross-section data used for this channel appears derives from the evaluated ACTL library, and is therefore probably sound. The data used for the other channel is less reliable, the $\text{P}^{31}(\text{n},\gamma)$ cross-section having been taken from an experimental determination on a similar nuclide, and the $\text{P}^{32}(\text{n},\text{p})$ cross-section obtained from a THRESH fit to systematics at 14MeV.

Production of C^{14} from aluminium proceeds by combinations of 4 successive reactions of the (n,α) or $(\text{n},\text{n}\alpha)$ type, the predominant combination being $\text{Al}^{27}(\text{n},\alpha)(\text{n},\text{n}\alpha)(\text{n},\alpha)(\text{n},\alpha)$. The cross-section data employed for these reactions are THRESH determinations, generally normalised to experimental data, with the following exceptions ; $\text{O}^{18}(\text{n},\text{n}\alpha)$, $\text{Ne}^{21}(\text{n},\text{n}\alpha)$, $\text{Ne}^{21}(\text{n},\alpha)$ and $\text{Mg}^{24}(\text{n},\text{n}\alpha)$. As only the last reaction is in the dominant production channel, it is clearly more critical than the others.

A comparison of the time-dependent gamma dose rate from SiC with earlier calculations ¹⁰ shows that in the immediate post-irradiation period the present calculations predict a lower rate,

presumably from a lower Na22 concentration. Production of this nuclide from aluminium appears to proceed by the competing routes $\text{Al}^{27}(\text{n},2\text{n})(\text{n},\text{n}\alpha)$ (~ 60%) and $\text{Al}^{27}(\text{n},\text{n}\alpha)(\text{n},2\text{n})$ (~ 40%).

4.2.3.3 Potassium and Calcium

The materials properties of potassium and calcium exclude them from consideration as candidate elements for first wall construction. They have been identified, however, as potential impurities in steels, at concentrations estimated to be approximately 2ppm in the compositions considered in Chapter V. On the basis of surface dose rate, they appear to be less troublesome than impurities such as silver and niobium.

Significant contributions to internal and external doses (by bremsstrahlung) from both inventories arise from the energetic β emitting isotopes Cl^{36} ($t_{1/2} = 3.01\text{E}+5$ years) and Ar^{39} ($t_{1/2} = 269$ years). The calcium inventory also contains the gamma-emitting Ar^{42} ($t_{1/2} = 32.9$ years) and the β emitting Ca^{41} ($t_{1/2} = 1.03\text{E}+5$ years) at significant levels.

Potassium appears to present the greater potential long-term hazard of the two elements. It is noted that the activation pathways are simple, the critical reactions being $\text{K}^{39}(\text{n},\alpha)$ and $\text{K}^{39}(\text{n},\text{p})$, the cross-section data for which appears to be satisfactory, having been normalised against the evaluated ACTL file. It can be concluded therefore that inventory concentrations are therefore unlikely to change much from the values calculated here. However, gamma dose rate may be altered by a more rigorous treatment of the bremsstrahlung contribution.

4.2.3.4 Titanium, Vanadium and Chromium

These elements, possessing excellent low-activation properties, are potentially among the most promising for low-activation alloy development^{30,41}. Their residual activity is limited by the argon isotopes Ar39 and Ar42, isotopes whose likely behaviour in first wall operating conditions and subsequent handling/recycling scenarios is subject to considerable uncertainty. If the argon isotopes were to be removed from the activated material, as they would be in some recycling scenarios, the dominant gamma-emitter would become Ti44 ($t_{1/2} = 47.3$ years).

The most highly activated element of the three is titanium, as it is closest in atomic number to Ar42. A detailed account of the activation of titanium, and the significance of this for its potential role in low-activation alloys, has been given elsewhere³⁰, but here it is noted that 3 production channels to Ar42 have been identified, all of which involve 2 stages, the highest yielding ($\sim 85\%$) channel being $\text{Ti48}(n,\alpha)(n,\alpha)$. It is noted that whereas the cross-section data for first stages appears to be sound (THRESH normalised to ACTL and ENDF files), the data for the second stages is weak, and relies on systematics. The critical item of data appears to be the cross-section for the $\text{Ca45}(n,\alpha)$ reaction, the experimental evaluation of which must be a matter of high priority for the development of low-activation vanadium alloys. The Ar42 concentration, already reduced by approximately a factor of 2 from previous calculations by a renormalisation of the data for $\text{Ca45}(n,\alpha)$ reaction, may therefore be subject to further adjustments.

The production of Ar42 from vanadium proceeds almost entirely ($\sim 97\%$) via the $\text{V51}(n,\alpha)(n,\alpha)(n,\alpha)$ channel. As for titanium, the data

for the first 2 stages is sound and the weakest link in the activation chain is the cross-section data for the $\text{Ca}45(n,\alpha)$ reaction.

In chromium, $\text{Ar}42$ is produced mostly (~ 80%) by $\text{Cr}52(n,\alpha)(n,\alpha)(n,n\alpha)$. Again the data for the first 2 stages is sound, and the critical data is the THRESH fit to $(n,n\alpha)$ systematics for the $\text{Ca}46(n,n\alpha)$ reaction.

With regard to the production of $\text{Ti}44$, the sensitivity analysis confirms what would seem to be intuitively clear : that production proceeds by 2 successive $(n,2n)$ reactions on $\text{Ti}46$. It is noted that the cross-section for the second of these is THRESH generated, fitted to systematics.

4.2.3.5 Manganese, Iron, Nickel, Cobalt and Copper

The above elements can be grouped together in as much as they are present in steel compositions, the first three as major constituents, the other two as impurities. Another feature which they have in common is that, at cooling times of up to a few hundred years, their radionuclide inventories would all contain significant quantities of the gamma-emitting $\text{Co}60$ ($t_{1/2} = 5.27$ years) . In the inventories of manganese and iron, substantial contributions to external and internal dose would also be made by $\text{Fe}55$ ($t_{1/2} = 2.73$ years) and $\text{Mn}53$ ($t_{1/2} = 3.74\text{E}+6$ years). The inventories of cobalt, nickel and copper would contain substantial amounts of the gamma-emitting $\text{Ni}59$ ($t_{1/2} = 7.51\text{E}+4$ years) and the beta-emitting $\text{Ni}63$ ($t_{1/2} = 100$ years), though in the case of copper the gamma dose from the former is swamped by that from $\text{Co}60$.

Taking the production of $\text{Mn}53$ in manganese first, it is clear that by far the most important channel (> 99%) proceeds by 2 successive $(n,2n)$ reactions on $\text{Mn}55$. The cross-section data for these reactions

appears to be sound, having been normalised against ACTL and ENDF files. The production of the same nuclide in iron is by $\text{Fe54}(n,d)$ ($>98\%$). The cross-section data for this reaction, however, is normalised to the (n,d) systematic, the standard deviation for which is approximately 77%.

Cross-section data for the production of Fe55 in manganese and iron appears to be good, the route being $\text{Mn55}(n,\gamma)(\beta^-)(n,2n)$ in the former and principally $\text{Fe56}(n,2n)$ ($>95\%$) in the latter.

The production of Co60 in manganese proceeds by 5 successive captures and 2 β^- decays from Mn55. The channel is the same in iron, but the starting point is Fe58, from which point 2 reactions and 1 decay are required. It is noted that the cross-section data for the $\text{Fe58}(n,\gamma)$ reaction in UKACT1, obtained from ACTL and ENDF files, is different from the data in the original REAC library, which was an estimate derived from a similar nuclei. It is possible that this may explain the discrepancy between the calculated doserates from iron in the calculations described here and those performed by Ponti¹⁴ with the GREAC library. With respect to the sensitivity coefficients of cross-section data for the $\text{Co59}(n,\gamma)\text{Co60}$ and $\text{Co59}(n,\gamma)\text{Co60m}$ reactions, it is observed that the sum of the two coefficients is less than 1, even though no other obvious production channels exist. The reason for this is that the $\text{Co59}(n,\gamma)$ reaction has a large cross-section in the resonance region, such that there is significant burn-up of Co59, taking the sensitivity coefficient out of the region of linearity. The same effect is noticed in the production of Co60 in cobalt and nickel, but not in copper, where the important reaction is $\text{Cu63}(n,\alpha)$.

Nickel is one of the elements which has been minimised or

replaced in low activation steel compositions, owing to the potentially high long-term doses from Ni59 and Ni63. The sensitivity coefficients show that these nuclides are produced in nickel by (n, γ) or (n,2n) reactions on neighbouring stable isotopes, with the capture route slightly favoured in both cases. Recently, experimental evaluations⁴² of the cross-sections at 14 MeV of the multiplication reactions Ni60(n,2n) and Ni64(n,2n), and the principal reaction from copper Cu63(n,p), have become available. The data is in agreement with the data in the UKACT1 library for the Ni64(n,2n) reaction at 14MeV, but suggests that the corresponding UKACT1 values for the Cu63(n,p) reaction are overestimated by a factor of 2.5, and the figures for Ni60(n,2n) overestimated by as much as a factor of 3. The effect of adopting the lower cross-sections would be marginal in nickel, since the Ni60(n,2n) route to Ni59 is of secondary importance, but would be greater with regard to the long-term internal dose from copper.

To summarise, the surface gamma doserates calculated for the elements discussed here have not changed greatly in comparison with previous calculations¹⁰. In the past, the activation database for these elements has been more thoroughly studied than for most of the other elements under consideration for the first wall, and though adjustments to certain dubious cross-sections are required, the production rates of long-lived nuclides are unlikely to depart markedly from present predictions.

4.2.3.6 Zirconium, Niobium and Molybdenum

Zirconium has been included in the candidate first wall compositions discussed in Chapter V as a potential impurity in steels at a level of 30ppm. Niobium and molybdenum are included as

minor constituents in steels, and as impurities in vanadium-based alloys. All three elements produce significant quantities of the gamma-emitting isotope Nb94 ($t_{1/2} = 7.51\text{E}+4$ years), which has been shown to be a major contributor to gamma dose from some conventional steels in handling and post-disposal environmental impact scenarios. Also examined in the case of zircon, are production routes to the beta emitters Sr90 ($t_{1/2} = 7.51\text{E}+4$ years) and Zr93 ($t_{1/2} = 7.51\text{E}+4$ years) which are the significant contributors to the the long-term specific ingestion and inhalation doses. In niobium, only routes to Nb94 have been examined, as this is by far the largest source of potential internal and external hazard. In molybdenum, the total surface gamma dose rate is comprised of several more or less equal contributions, and the routes to Tc99 ($t_{1/2} = 7.51\text{E}+4$ years), Mo93 ($t_{1/2} = 7.51\text{E}+4$ years) and Nb91 ($t_{1/2} = 7.51\text{E}+4$ years) have also been delineated.

The production of Nb94 in zircon proceeds mainly by 2 successive (n,2n) reactions on Zr96. It is noted, however, that the cross-section data for the Nb95(n,2n) reaction has been normalised against 14MeV systematics, and a substantial reduction in the cross-section could make the alternative Zr94(n, γ) production channel the most important one. Similarly, the importance of the Mo95(n,d) channel would be enhanced by such a reduction. Data for the principle production channels to Sr90 and Zr93 are taken from, or normalised against ENDF and ACTL datafiles, with the exception of the Zr91(n,2p)Sr90 cross-section.

The principle routes to Nb94 from molybdenum are Mo94(n,p)Nb94 and Mo94(n,p)Nb94m(IT)Nb94, the data for the former being obtained from a THRESH fit to ACTL data, the data for the latter estimated from the former. Comparison with recent

experimental determinations ⁴² show that the cross-section for the ground state reaction is sound, but the cross-section for the reaction to the metastable state is overestimated by factor of approximately 2. The present Nb94 concentration is therefore overestimated by approximately 25%. However, taking account of the above reservations, the concentration of Nb94 in the molybdenum inventory is known to good degree of certainty.

The ^{case} is somewhat similar to the production of Nb94 from niobium : the main channels are Nb93(n,γ)Nb94 and Nb93(n,γ)Nb94m(IT)Nb94, the former owing its origins to the ACTL file, the latter an estimate of the former, taking a branching ratio of 0.5. Since yields of reactions to metastable states are uncertain, the uncertainty in the Nb94 concentration will be largely determined by the uncertainty in cross-section of the reaction to the Nb94 isomeric state . Delineation of productions channels to Tc99, Mo93 and Nb91 was straightforward, and largely involves data derived from the ACTL file, with no data which appears to be obviously weak. A minor complication is presented by the large cross-section for the Mo93(n,g) back reaction, which results in a sizable negative sensitivity coefficient.

4.2.3.7 Silver

Potentially a troublesome impurity in first wall steels and alloys, the long-term radionuclide inventory of silver is dominated by the metastable species Ag108m ($t_{1/2} = 127$ years), and the corresponding ground state Ag108 ($t_{1/2} = 2.37$ minutes). Two production channels have been found, the primary being Ag107(n,γ)Ag108m, which has a yield of approximately 91%, the secondary channel being

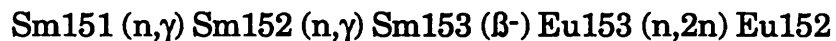
Ag109(n,2n)Ag108m which yields approximately 7% of the nuclide. Significant too, may be the Ag109(n, γ) reaction, which depletes the quantity of material which is transmuted by the secondary route. The cross-section data for these reactions originates from the ACTL datafile.

In some hazard scenarios Cd113m, produced by 4 successive captures from Ag109, may provide a significant internal dose.

4.2.3.8 Neodymium, Samarium and Europium

These elements have been identified as potential impurities in steels, at levels of 2, 0.5 and 0.2ppm respectively. A common feature of the activation of all three is the presence of long activation chains propagated by capture reactions which often have effective cross-sections of greater than a barn. They therefore provide an extreme test for the interpretation of sensitivity coefficients.

If the production of Eu152 from Sm150 is taken as an example which displays the essential problems in interpreting the sensitivity coefficients for these elements (see figure 4.13), it is seen that sensitivity coefficients along the activation chain



are not equal, ranging through 0.017, 0.058 to 0.923. A close examination of the alternative channels to Eu152 from the nuclides in this chain suggests that the bulk of the Eu152 is indeed transmuted by the aforementioned chain. How are these sensitivity coefficients to be interpreted then ? The apparent anomaly can be accounted for by the sizes of the cross-sections for the reactions ; 18.3,

13.8 and 0.131 barn respectively. In the irradiation conditions considered here, the burn-up of a stable parent nuclide which results from a reaction with cross-section 20 barn is 98.54 % . Consequently the differential sensitivity is to be found in the region of non-linearity identified in figure 2.3 , and the sum of the sensitivity coefficients of competing chains is not unity. The effect of the high burn-up rate on Sm151 is to reduce the sensitivity (in its general sense) of the Eu152 concentration to variations in the Sm151(n, γ) cross-section, and hence reduce the sensitivity coefficient . Conversely, the relatively low burn-up (2.7%) caused by the Eu153(n,2n) reaction results in a sensitivity coefficient which may be interpreted as the fraction of Eu152 which is produced via that channel, as in the rest of the analysis.

4.2.3.9 Tantalum and Tungsten

The strategy of minimising the concentrations of the highly activated elements niobium and molybdenum in low activation steels, has thrown the spotlight on tantalum and tungsten as possible alloying replacements. Earlier studies conducted by Jarvis with the UKCTR-III database ⁷ suggested that their long-term activation properties compare favourably with the aforementioned elements, allowing unrestricted permissible concentrations if the end of lifetime "hands on" criterion (i.e. a gamma dose rate limit of 25 μ Sv/hr) were to be adopted. The later calculations of Giancarli ¹⁰ and Ponti ¹⁴, utilising libraries which included more reactions for these elements - particularly for reactions to the isomeric states of Hf178, Re186 and Ir192 - severely reduced their elemental acceptability. As noted in Chapter 1, the figures presented here significantly reduce the elemental acceptabilities of tantalum and

tungsten further, but as the sensitivity analysis will show, the database is still poor in this area and probably overestimates the long-lived activity.

Calculations with the UKACT1 database predict that the long-lived radionuclide inventory produced from tantalum will be dominated by the gamma-emitting Hf178n ($t_{1/2} = 31.1$ years). Present at levels which would give specific gamma and potential ingestion/inhalation doses 3 or 4 magnitudes below those from this nuclide, Hf182 ($t_{1/2} = 9.02\text{E}+6$ years) and Re186m ($t_{1/2} = 2.0\text{E}+5$ years) could become significant in some scenarios.

As the figure for the production of Hf178n shows, the production channels from tantalum are varied and indirect. The important reactions in the last stage of the principal activation chains have been identified, but there remain gaps in the earlier stages, notably for the production of the Hf178 ground state, from which the primary production channel appears to proceed by inelastic scatter Hf178(n,n')Hf178n. Considering the cross-section data for the important last stage reactions to the metastable state first, the data for which is probably the most uncertain, it is noted that the cross-sections for 2 of the 3 reactions in question, Hf177(n, γ) and Hf179(n,2n) have been obtained by applying a fixed branching ratio (0.01) to the data for the ground state reaction. It is noted, too, that the n metastable state in Hf178 has a nuclear spin of 16+ and is considerably more energetic (~ 2.5 MeV) than the ground state ; bearing in mind the systematic correlation (admittedly weak) between nuclear spin and isomer ratio ³⁸, the present branching ratio of 0.01 may be judged to be an overestimate.

If the ground state reactions are now considered, the data for the

Hf177(n, γ) is probably sound, being derived from the evaluated cross-section datafile JEF1, likewise the data for the Hf179(n,2n) is a THRESH fit to the ACTL file. The data for the Hf178(n,n') reaction, however, are more uncertain, since the method employed by THRESH to generate neutron inelastic scattering cross-sections does not appear to be to employ systematics ¹², resulting in errors that have not been quantified. If it is assumed that the cross-section data for reactions in the preceding stages of the three main activation chains have been normalised against evaluated datafiles, it may be concluded that potential reductions in the Hf178n concentration resulting from lower branching ratios will be limited by the Hf178(n,n') channel, and the uncertainty associated with the generation of (n,n') cross-sections.

The production routes to Hf178n from tungsten exhibit similarities to the tantalum case, with the Hf178(n,n') and Hf179(n,2n) routes again being important, but with a third channel proceeding by W182(n, α), the cross-section data for which has been normalised to a systematic. The overall profile of the activation inventory produced from tungsten, however, is different in that the largest contributor to long-term gamma dose rate is Ir192n ($t_{1/2} = 2.0\text{E}+5$ years), produced primarily by 6 successive captures from W186. Significantly for the Ir192n concentration, the cross-section data for last reactions in the chain has been estimated from similar nuclides (W182, W183 and Ta181). Bearing in mind the size of the variation of the overall capture cross-section between even neighbouring isotopes (e.g. often > 100 times), the calculated level of Ir192n must be regarded with extreme suspicion. A comparison of available cross-section data for tungsten and osmium ⁴³, suggests

that the cross-sections in question have been overestimated.

Likewise, it can be concluded that the $\text{Ir191}(n,\gamma)\text{Ir192m}$ cross-section (employing a branching ratio of 0.01) is also probably an overestimate (nuclear spin of n metastable being $9+$) if a similar argument to that used for Hf178m is used again. It is quite possible, therefore, that the reduction in the Ir192m concentration which would be brought about by substituting more realistic cross-section data in the calculation could relegate this isotope to a position of secondary importance.

The cross-section data available for the production of Re186m is more complete. The uncertainties associated with estimating cross-sections for reactions to the metastable state however, are similar. Cross-section data for the reactions in question are estimates obtained from the ground state reactions with the application a fixed branching ratio (0.5), the primary route proceeding by $\text{Re187}(n,2n)$, the data for which has been fit to a systematic, and a secondary terminating by $\text{Re185}(n,\gamma)$, the data for which derives from the ACTL datafile.

To conclude this discussion, the activation database for tantalum and tungsten is weaker than for most other candidate first wall elements, particularly with respect to the production of the long-lived metastable states which dominate the inventories of both elements. The levels of activity and dose rate presented here are therefore only rough estimates, and are liable to change. In this context, significance should not be attached to the small increases in the gamma dose rates which the present calculations predict in comparison to earlier work.

4.2.4 Conclusions to production channel analysis

It has been shown by the extended discussion in the subsections

in 4.2.3 that in the majority of cases sensitivity coefficients provide a straightforward means of pathway delineation. Complications in interpretation can arise where capture reactions with large cross-sections are involved, as demonstrated by the production of Co60 from Co59 and reactions in Nd, Sm and Eu. In these circumstances, a careful examination of nuclide concentrations, cross-sections sizes and all production channels is necessary to avoid misinterpretation of the coefficients. Apart from the given examples, problems in the interpretation are infrequent and of secondary importance.

It can be seen from table 4.2 that for the timescale and the elements covered by the sensitivity analysis, the critical items of cross-section data in the UKACT1 database derive largely from evaluated datafiles. It should be noted, however, that this is not a cast iron guarantee of goodness, as these files often combine data from a number of disparate theoretical and experimental sources, and that short of studying individual datapoints in the files, the accuracies of single group cross-sections are known only within broad terms.

Nevertheless, at this stage in the development of the database, of paramount interest must be those critical items of data which are derived from systematics or estimations. For convenience they have been summarised in table 4.3.

Table 4.3 : Summary of critical reactions employing data obtained from systematics and estimates.

C14(n,n α)	Nb93(n, γ)Nb94m
Al27(n,2n)	Hf178(n,n')
Si28(n,d)	Hf177(n, γ)Hf178n
P32(n,p)	Hf179(n,2n)Hf178n
Mg24(n,n α)	W182(n,n α)Hf178n
Ca45(n, α)	Re187(n,2n)Re186m
Ca46(n,n α)	Re185(n, γ)Re186m
Fe54(n,d)	Os189(n, γ)
Nb95(n,2n)	Os190(n, γ)
Zr91(n,2p)	Os191(n, γ)Ir192n

4.3 Propagation of Errors

The sensitivity coefficients listed in table 4.2 can now be employed to estimate standard errors in important nuclide concentrations, in the manner described in Chapter II. Before this can take place, however, it is necessary to assign errors to the single group cross-sections used in inventory calculations.

4.3.1 Errors in Data

For reactions of the threshold kind, it is assumed that the error in the 14MeV cross-section, the point at which most THRESH fits are normalised, provides a good approximation to the single group error. The validity of this assumption is demonstrated in Appendix II by a rigorous covariance treatment of the errors in the Co59(n, α) reaction cross-section. Where models have been normalised to experimental measurements which include error estimations, estimations of the

error in the 14MeV cross-section have been adopted if available (in practise the estimations in reference 38). Otherwise errors have been assigned on the basis of the standard deviation of data used to form a systematic, as explained by Forrest ³⁸.

The estimation of single group errors associated with capture reactions is a more complex problem, since the errors are a consequence of fitting evaluated curves to cross-sections in erratic resonance regions. The professional evaluators of capture cross-sections ⁴⁴ advise, however, that it would be reasonable to assume that single group cross-sections obtained from evaluated cross-section files are usually known to better than 30% accuracy. The errors attached to cross-sections estimated from similar nuclides will naturally be greater; a figure of 75% (probably optimistic) has been assigned in these cases, as it has been for reactions to metastable states with unknown branching ratios.

4.3.2 Errors in Calculated Quantities

Standard errors in the concentrations of the main contributors listed in table 4.1 have been calculated with the error assignation scheme described above. From them the resulting errors in gamma doserate after 100 years were evaluated for each element in the manner described by expression (2.10)

$$\text{Var}(D)/D^2 = \sum_i (\partial D/\partial N_i)^2 (N_i/D)^2 \text{Var}(N_i) / N_i^2 \quad (2.10)$$

The fractional error in the doserate has been taken as

Fract. error = $\sqrt{\frac{\text{Var(D)}}{D^2}}$

the symbols being as defined in Chapter II.

These fractional errors in the nuclide concentrations and doserates are given in table 4.4.

Table 4.4 : Estimations of the standard fractional errors attached to the evaluated concentrations of important inventory nuclides, and the resultant errors in the elemental gamma doserate after a 100 year cooling period.

Element	Long-lived Products	Error in Conc.	Error in Total Gamma Doserate
C	Be10	0.47	0.47
	C14	0.30	
N	Be10	0.85	0.85
	C14	0.42	
Al	C14	2.12	0.30
	Al26	0.30	
	Na22		
Si	Al26	1.05	1.05
	Si32	0.76	
K	Cl36		
	Ar39		
Ca	Cl36	1.13	0.55
	Ar39	0.79	
	Ca41	0.31	
	Ar42	0.74	

Ti	Ar42	0.79	0.79
	Ti44		
V	Ar42	0.79	0.79
	Ti44	0.89	
Cr	Ar42	1.39	1.39
	Ti44	1.21	
Mn	Mn53	0.60	0.60
	Fe55	0.37	
	Co60	1.51	
Fe	Mn53	0.77	0.51
	Fe55	0.09	
	Co60	0.56	
Co	Co60	0.25	0.25
	Ni59	0.51	
	Ni63	1.18	
Ni	Co60	0.28	0.26
	Ni59	0.28	
	Ni63	0.22	
Cu	Co60	0.05	0.05
	Ni63	0.05	
Zr	Sr90	0.74	0.52
	Zr93	0.29	
	Nb94	0.59	
Nb	Nb94	0.29	0.29
Mo	Nb91	0.22	0.14
	Nb93m	0.27	
	Nb94	0.10	
	Tc99	0.33	
Ag	Ag108m	0.31	0.31
	Cd113m	1.09	

Sn	Ag108m	0.42	0.43
	Cd113m	0.38	
	Sn121m	0.90	
	Sn126	0.59	
Ce	Cs137	0.90	0.90
	Ba133	0.41	
Nd	Sm151	0.64	0.81
	Eu152	0.85	
	Eu154	0.92	
Sm	Sm151	0.53	0.39
	Eu152	0.92	
	Eu154	0.42	
Eu	Sm151	0.85	0.49
	Eu152	0.91	
	Eu154	0.41	
	Gd150	0.57	
Tb	Tb158	0.61	0.61
Ho	Ho166m	0.65	0.65
Ta	Hf178n	0.69	0.69
	Hf182		
	Re186m		
W	Hf178n	0.75	1.64
	Re186m	0.60	
	Ir192n	1.64	
Ir	Ir192n	0.34	0.30
	Pt193	0.44	
Pb	Pb205	0.29	0.89
	Pb210	0.58	
	Bi207	0.89	
	Po210	0.61	

Bi	Bi207	0.65	0.65
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4.3.3 Discussion and Conclusion

The error estimates presented in table 4.4 effectively provide a quantitative summary of the extended discussion on the quality of the activation database in section 4.2.

The approximate nature of the treatment, with most errors in the database assigned from systematics, should not be forgotten, however, nor the precise meaning of the standard error in this context. This point is particularly pertinent to cross-sections carrying large errors. In the treatment of standard errors, the error in a calculated quantity is based upon the summation of the small (i.e. $\sim < 15\%$) perturbations in the calculated function which would result from independent small perturbations in the variables ⁴⁵. An error calculated in this way is in fact the variance $[< \Delta N^2 >]^{-1/2}$, and unlike the error factor ³⁸, does not carry information about the shape of the error distribution about the calculated value, the shape of which is asymmetric, falling away more rapidly for negative errors than for positive. It is quite possible therefore, for the quoted standard fractional error in a nuclide concentration to exceed 1, though a physical interpretation of this is not clear. One possible interpretation is to use the standard error as a rough guide to the upper error limit (which it will underestimate slightly). The lower limit can then be obtained by assuming that the ratio of the lower limit to actual value is the same as the ratio of the actual value to the upper limit. As the fractional error tends to zero, and the

asymmetrical distribution tends towards a symmetrical distribution, positive and negative errors become equal.

The effect of relying on systematics to provide estimates of the errors associated with single group cross-sections, rather than individual datafiles (where relevant), is to compress the range of errors across the group of elements considered in the study : thus it is probable that the error associated with the production of Co60 in iron has been overestimated by assuming that the Fe58(n, γ) and Co59(n, γ) cross-section are known only to a 30% accuracy, whereas the error associated with the production of Ir192n from tungsten has been underestimated by assuming that capture cross-sections in osmium taken from "similar" nuclides in tungsten are known to 75% accuracy.

In order to set the errors presented here in their proper perspective, the question which must be asked is : how do they alter our perception of the adequacy of the database with respect to application of the calculated inventories to low-activation material development and dosimetric assessments of potential hazard scenarios ? If the development of low-activation materials is taken first, the answer must be that the sizes of the present error bars allows for significantly misleading conclusions to be made. For example, with regards to the use of Ta and W in low activation steels, it is reasonable to suppose that a factor 5 alteration in a hazard parameter may be influential in determining elemental selection. As far as present dosimetric assessments are concerned, there exist large errors in modelling hazard scenarios ^{16,18,46} which could overshadow the errors in nuclide concentrations given in table 4.4. The current emphasis in dosimetric assessments would be in

refining the dosimetry calculation, and it would be unlikely therefore that the errors in nuclide concentration would significantly alter the analysis of results from such a calculation at present.

Finally, it is to be hoped that at some future date a more complete analysis would also consider errors arising from the neutron transport and dose rate calculations.

CHAPTER V

Candidate Compositions for the Reactor First Wall

Ganga was sunken, and the limp leaves

Waited for rain, while far distant

Black clouds gathered over Himavant.

The jungle crouched, humped in silence.

Then spoke the Thunder

DA

datta

- What the Thunder Said

5.1 Introduction

The selection of materials for the first wall of a fusion reactor is subject to a large number of constraints, deriving from both structural and physics requirements. At the current early stage of fusion development the minimisation of activation inventories is but one of these constraints and not necessarily the first in order of priority. It should be borne in mind in the following discussion, therefore, that a necessary prerequisite for any candidate low-activation material is that it can operate within these other constraints.

Strategies orientated towards the development of low-activation first wall materials can be divided into two streams ;

i) the evolutionary approach : the taking of tried and tested structural materials and making adjustments to their compositions so as to

reduce activation to an acceptable level without impairing their established materials properties. In practise, this applies mainly to steel making technologies, but would also encompass specialist alloys such as those used in the aerospace industry etc., where fabrication techniques are well established.

ii) the garden of Eden approach : the taking of materials with the best activation properties and developing a substantially new fabrication technology which would meet the structural and physical needs demanded of the first wall.

The advantage of i) is that it would provide results quickly, the disadvantage that the reductions in activity may be limited. In contrast, ii) is the more far sighted approach, giving potentially greater gains, but taking longer to achieve them.

The candidate first wall materials examined here have, through the weight of previous work on low-activation materials in the UK, become regarded as reference standards, and can be said to be a mixture of both the evolutionary and garden of Eden approaches. They are not, as the large number of low activation windows in figure 1.1 shows, the only types of low activation materials which may be suitable for the first wall, but are merely those for which a discussion has already been initiated.

5.2 Optimisation of Culham Candidate Compositions

The percentage weight compositions of the 7 materials considered here, and the estimated levels of impurities, are given in table 5.1 . Of the seven, 2 are commercially available steels ; the austenitic SS316 and the martensitic FV448, and 2 are low activation steels, the

austenitic OPTSTAB and the martensitic LA12Ta, which may be regarded as the elementally substituted analogues of the commercial alloys. As noted in Chapter I, the residual activity from the Fe bulk material is low, the long-lived activity in steels being arising from minority elements and impurities. The strategy used to optimise the compositions of low activation steels ⁴⁷, therefore, has been to replace highly activated minority elements, such as Nb, Mo and Ni, with their chemical equivalents. At best, it can be hoped that activation in steels would be governed by contributions from Fe.

It goes without saying that the effectiveness of this approach depends upon the correct identification of the main elemental contributors to the dose in the first place, and would be limited to the timescale and particular methodology employed to calculate the dose. To this end, bar charts showing the relative elemental contributions to the 100 year gamma doserates from each of the candidate compositions are shown in figures 5.1 - 5.7 .

On the basis of figures 5.1 and 5.2, the following general conclusions may be made with respect to elemental contributions to the surface gamma doserate from the 2 commercial steels ; the doserate from FV448 is dominated by the minor constituents Nb and Mo, the doserate from SS316 is shared more evenly between Mo, Nb and Ni, with some impurities in the threshold of significance. Thus in the Culham low activation austenitic steel OPTSTAB, Mn and N replace Ni, W replaces Mo, and Ta replaces Nb. The result, as can be seen in figure 5.4, is that elemental contributions, with exception of the impurity Ag, have been brought down to the level of Fe. The emphasis for further optimisation appears to have now shifted from selecting minor alloying additions (although the roles of Ta and

Table 5.1 : Assumed compositions of candidate first wall materials
(with estimated levels of impurities in steels)

	FV448	SS316	LA12Ta	OPTST.	SiC	VCrTi	VTiSi
C	1.5E-1	3.0E-2	1.6E-1	7.0E-2	3.0E+1	-	-
N	6.0E-2	1.5E-2	4.2E-2	3.0E-1	-	-	-
Al	1.0E-3	1.0E-3	1.0E-3	1.0E-3	-	-	-
Si	4.0E-1	5.0E-1	3.0E-2	2.0E-1	7.0E+1	-	1.0E-0
K	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Ca	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Ti	-	-	-	-	-	5.0E-0	3.0E-0
V	2.5E-1	-	2.5E-1	-	-	8.0E+1	8.1E+1
Cr	1.1E+1	1.8E+1	9.0E-0	1.5E+1	-	1.5E+1	1.5E+1
Mn	7.5E-1	1.8E-0	8.0E-1	1.6E+1	-	-	-
Fe	8.6E+1	6.6E+1	8.8E+1	6.5E+1	-	-	-
Co	1.0E-3	1.0E-3	1.0E-3	1.0E-3	-	-	-
Ni	7.5E-1	1.2E+1	4.0E-4	4.0E-4	-	-	-
Cu	2.0E-3	2.0E-3	2.0E-3	2.0E-3	-	-	-
Zr	3.0E-3	3.0E-3	3.0E-3	3.0E-3	-	-	-
Nb	3.0E-1	2.0E-3	2.0E-5	2.0E-5	-	-	-
Mo	6.5E-1	2.4E-0	5.0E-3	5.0E-3	-	-	-
Ag	5.0E-5	5.0E-5	5.0E-5	5.0E-5	-	-	-
Sn	1.0E-3	1.0E-3	1.0E-3	1.0E-3	-	-	-
Ba	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Ce	4.0E-4	4.0E-4	4.0E-4	4.0E-4	-	-	-
Nd	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Sm	5.0E-5	5.0E-5	5.0E-5	5.0E-5	-	-	-

Eu	2.0E-6	2.0E-6	2.0E-6	2.0E-6	-	-	-
Tb	5.0E-5	5.0E-5	5.0E-5	5.0E-5	-	-	-
Ho	1.0E-6	1.0E-6	1.0E-6	1.0E-6	-	-	-
Ta	1.0E-3	1.0E-3	1.2E+1	1.0E-3	-	-	-
W	1.0E-3	1.0E-3	8.5E-1	1.2E-0	-	-	-
Ir	1.0E-5	1.0E-5	1.0E-5	1.0E-5	-	-	-
Bi	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-

W have not been fully resolved yet) to the control of impurities. The same conclusion seems also to apply to the Culham low activation martensitic steel LA12Ta. The size of the potential reduction to be achieved by further improvements, as provided by an estimate based upon the doserate from the Fe bulk material, is approximately an order of magnitude.

In the corresponding figures for the vanadium alloys (5.5 and 5.6), it is seen that the gamma doserate after 100 years from these materials is limited by the concentration of the Ti addition. Replacement of this element in VCrTi type compositions would appear to offer the greatest reduction in doserate. The contribution to the gamma doserate from a 1% Si addition does not appear to be significant in the intermediate timescale considered here, but after a few hundred years the contribution from Al26 becomes increasingly dominant.

5.3 Activation Calculations for Candidate First Wall Compositions

Activation calculations identical in methodology to those performed for pure elements in Chapter I, have been performed for the

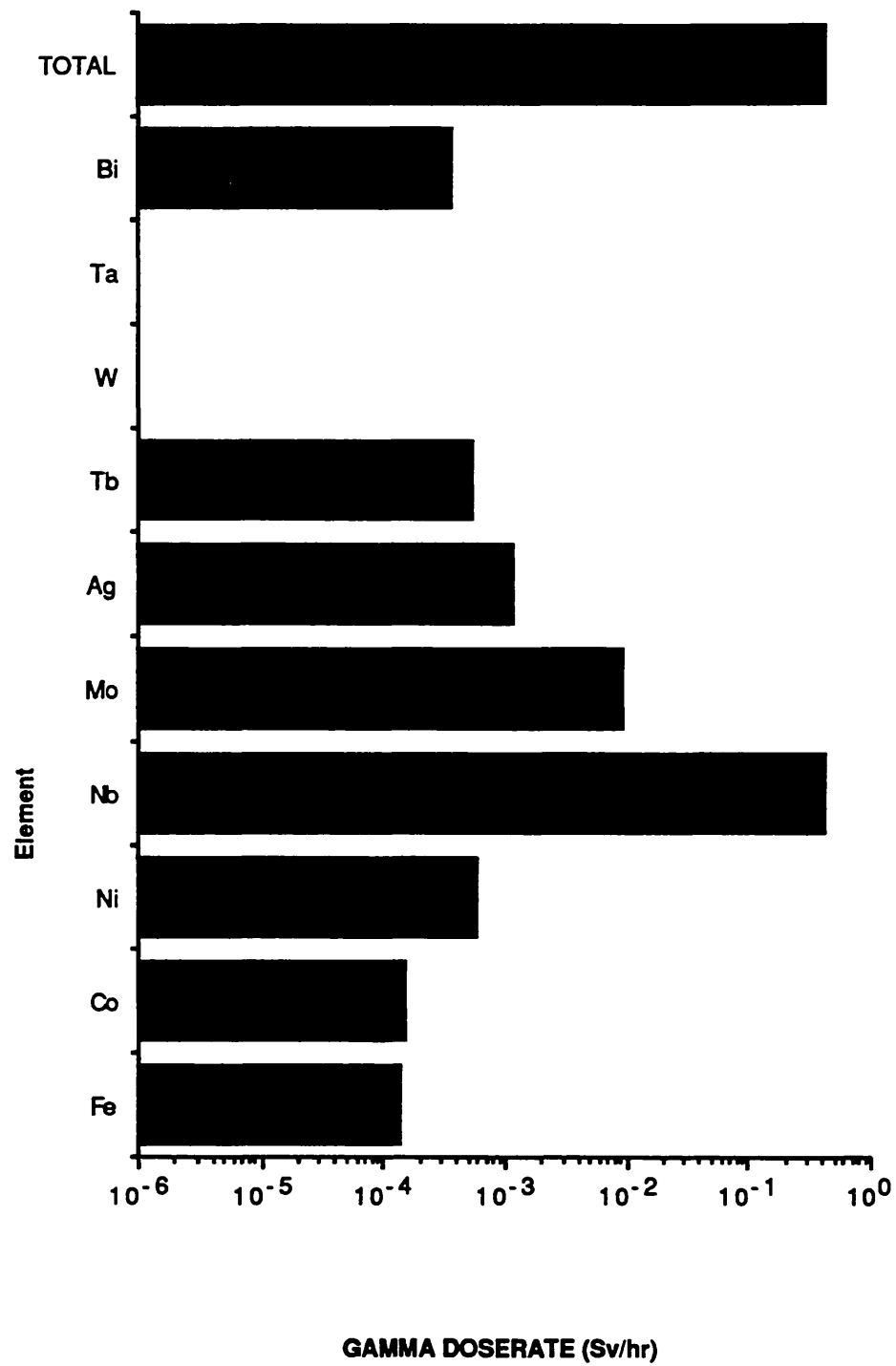


Figure 5.1 : Elemental contributions to the 100 year gamma doserate from FV448

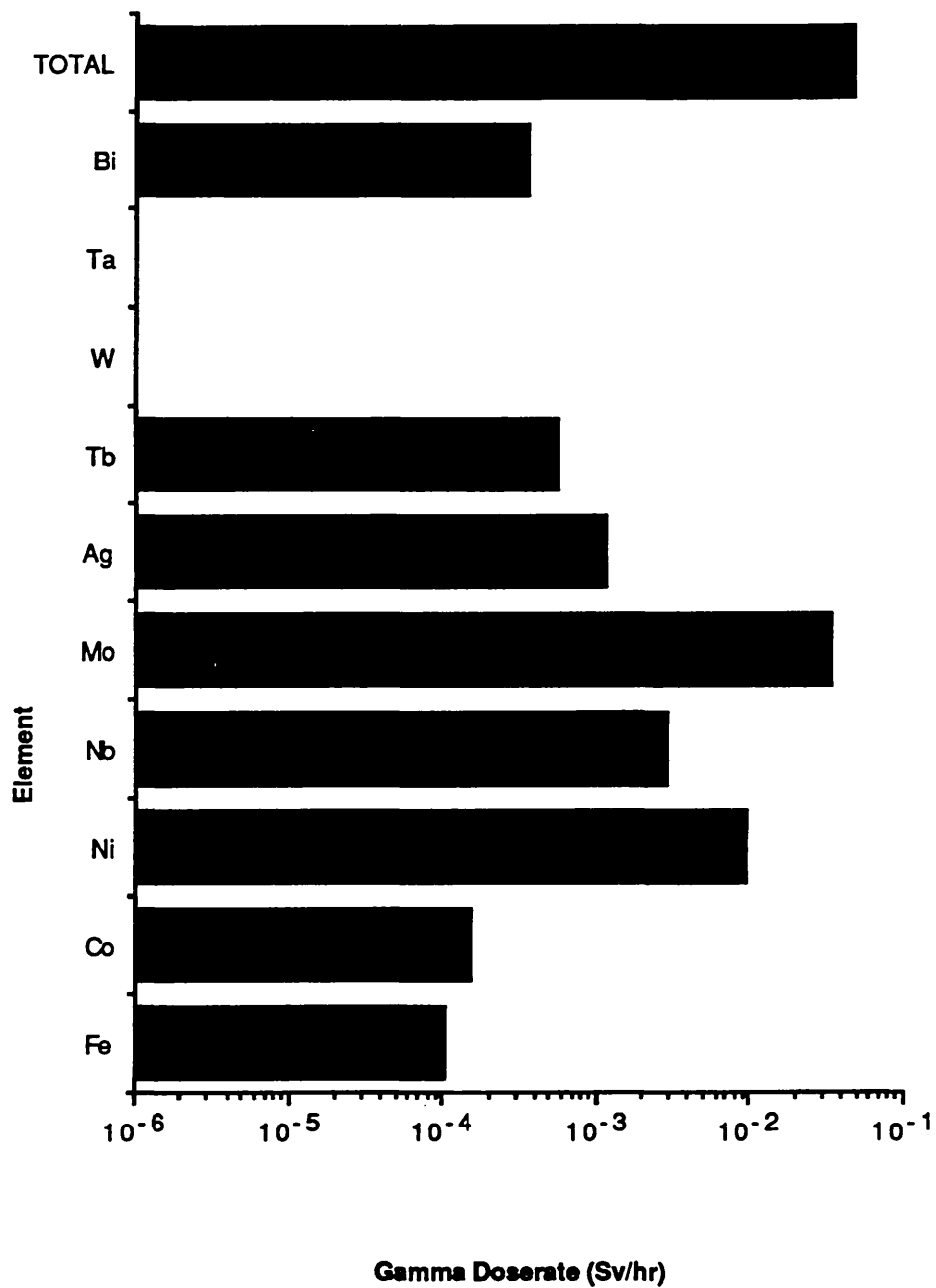


Figure 5.2 : Elemental contributions to the 100 year gamma doserate from SS316

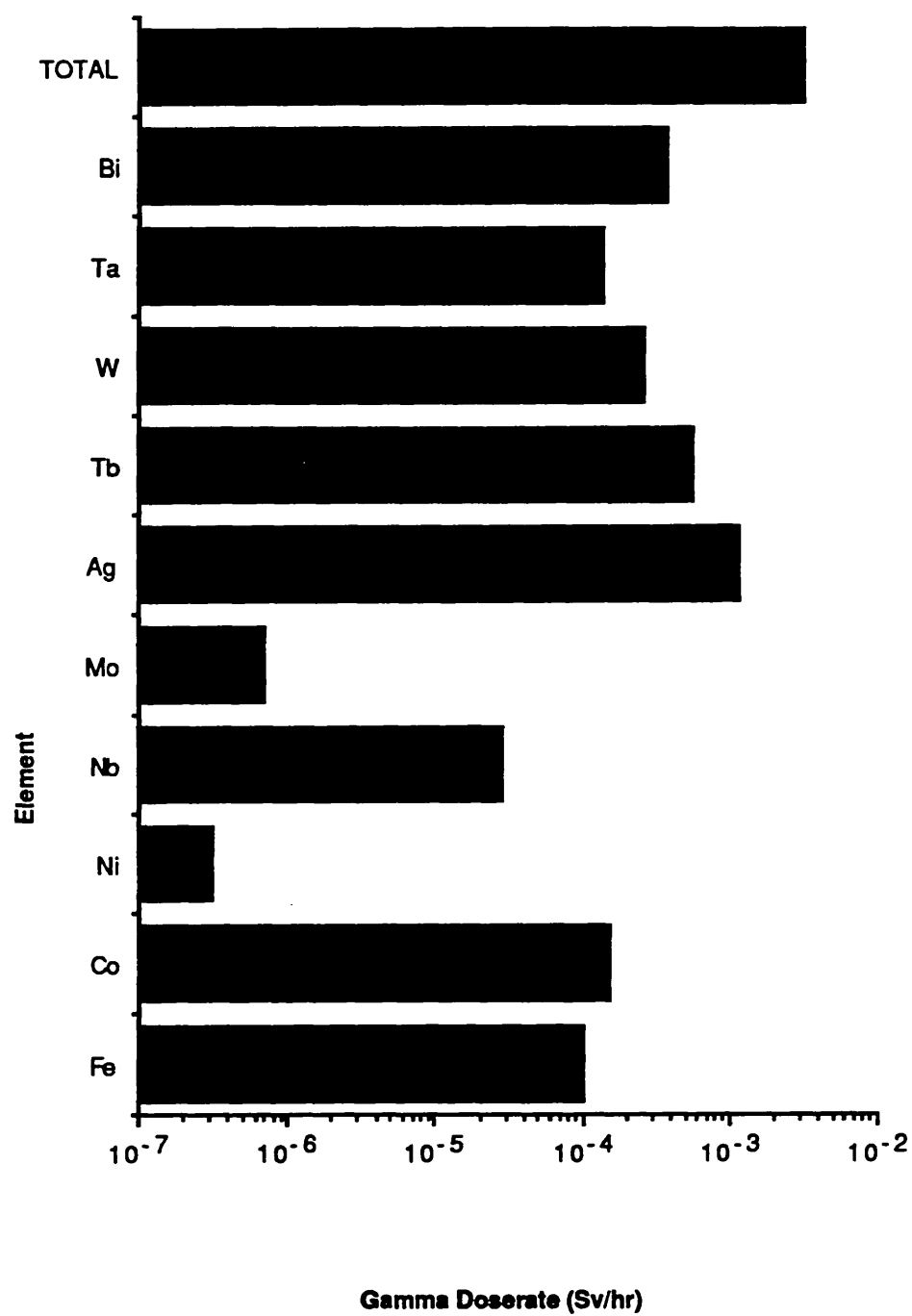


Figure 5.3 : Elemental contributions to the 100 year gamma doserate from LA12Ta

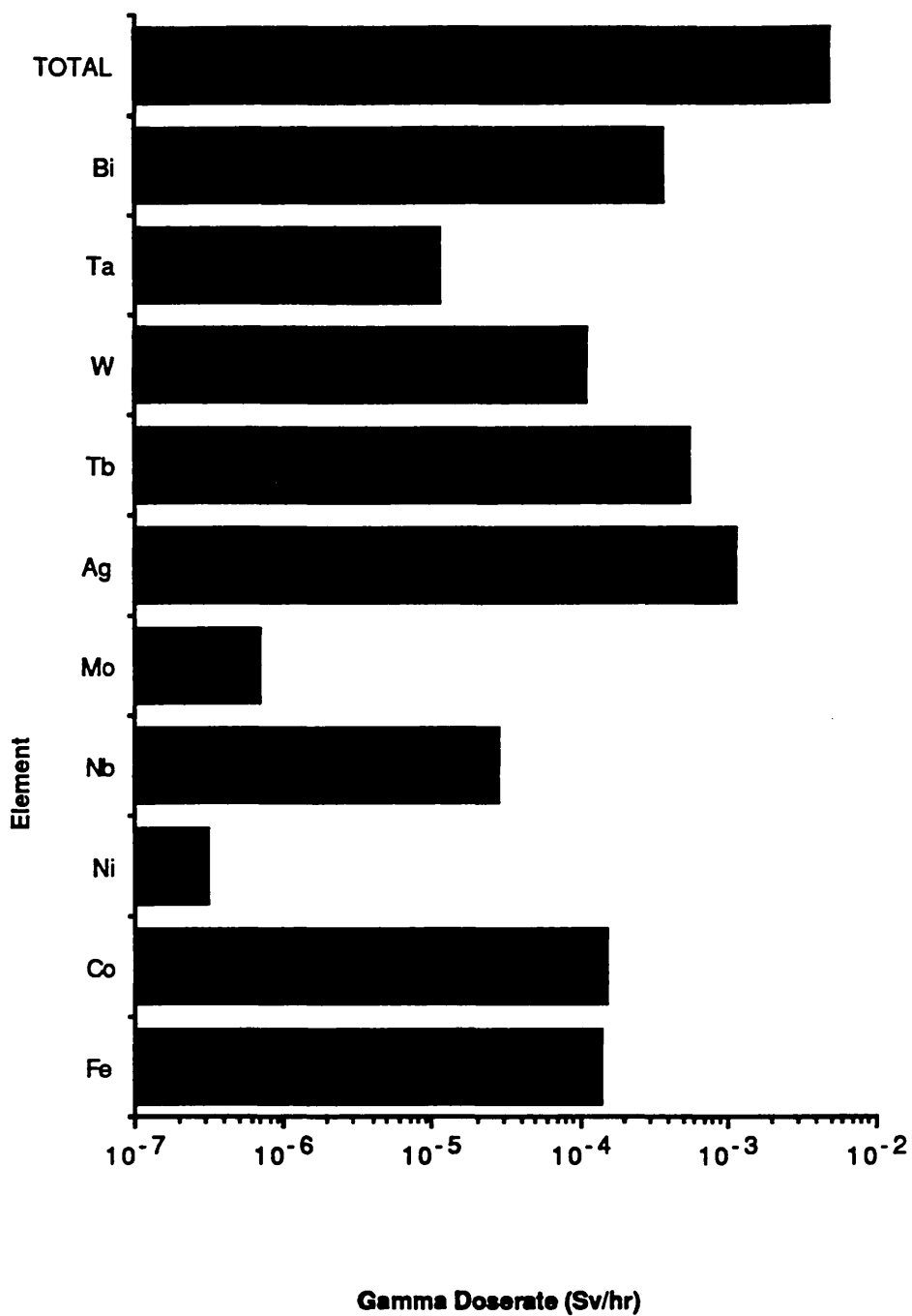


Figure 5.4 : Elemental contributions to the 100 year gamma doserate from OPTSTAB

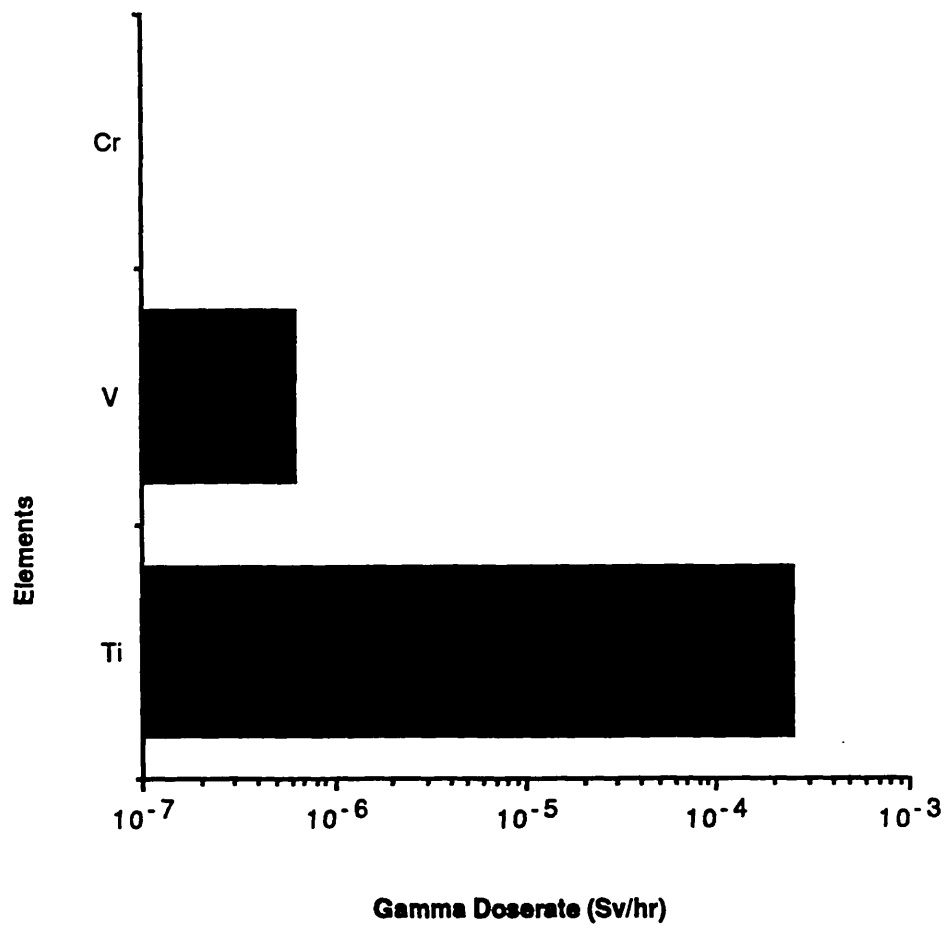


Figure 5.5 : Elemental contributions to the 100 year gamma doserate from V-15Cr-5Ti

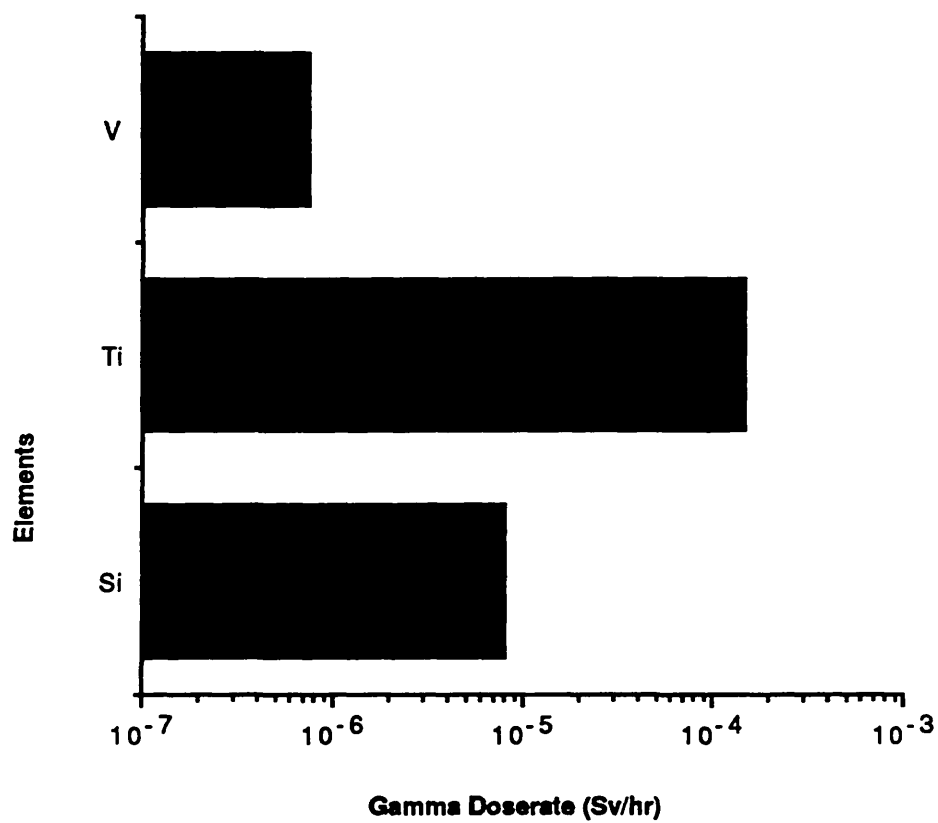


Figure 5.6 : Elemental contributions to the 100 year gamma doserate from V-3Ti-1Si

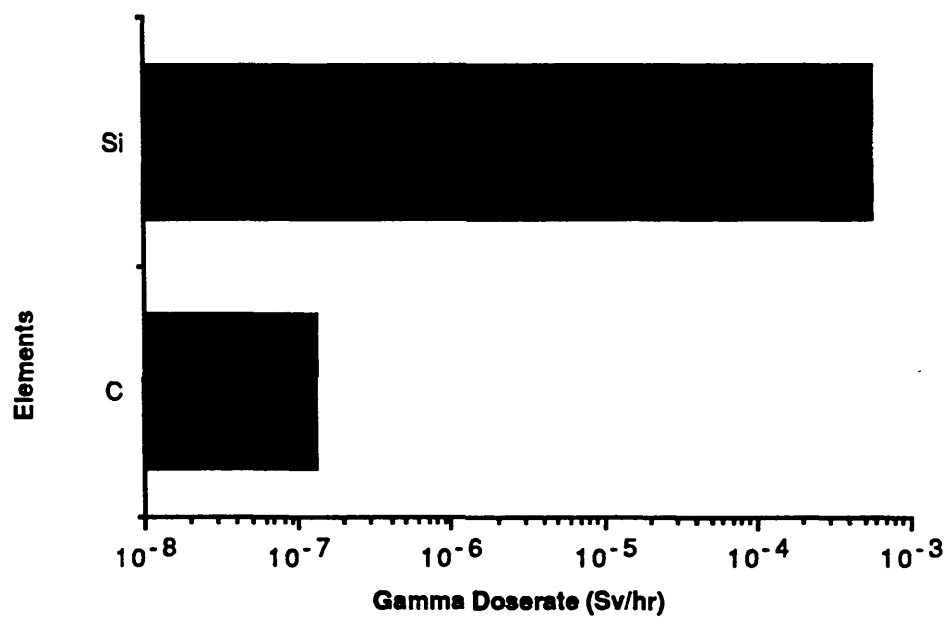


Figure 5.7 : Elemental contributions to the 100 year gamma dose rate from SiC

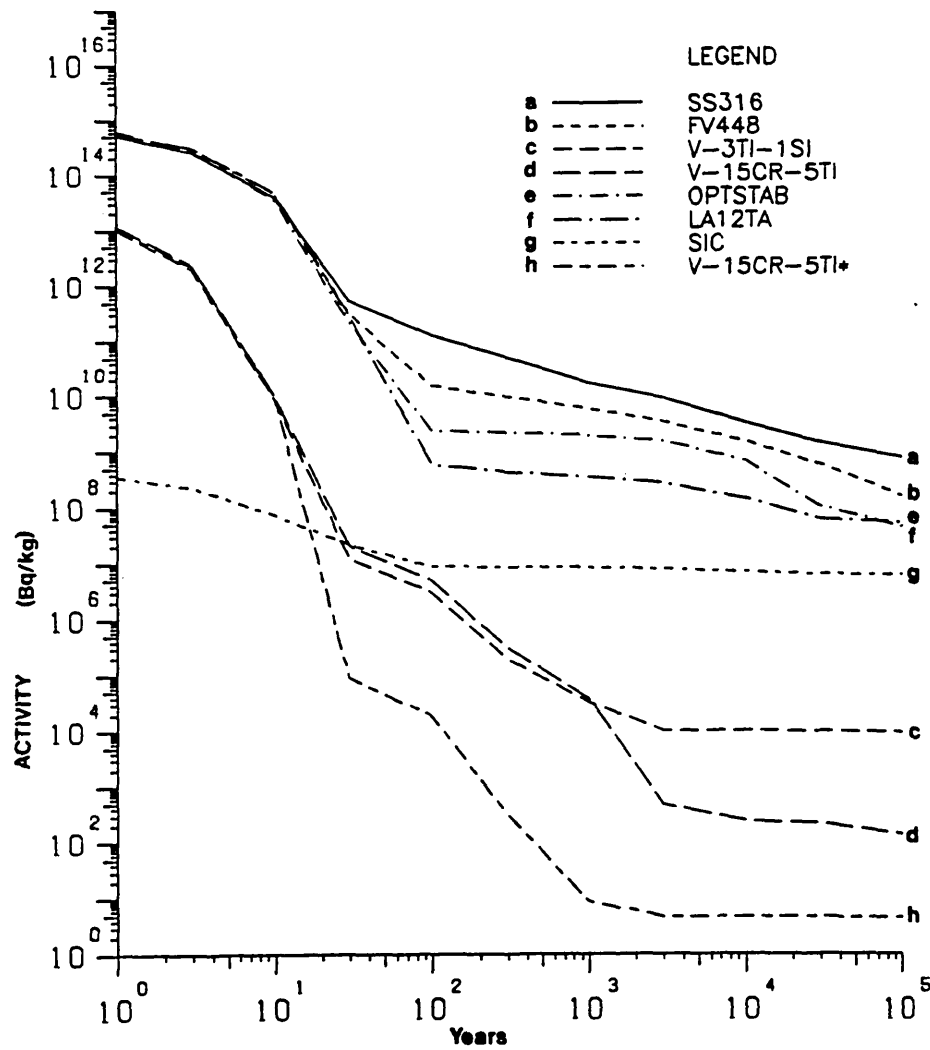


Figure 5.8 : Time dependence of specific activity (Bq/kg) from Culham candidate first wall compositions

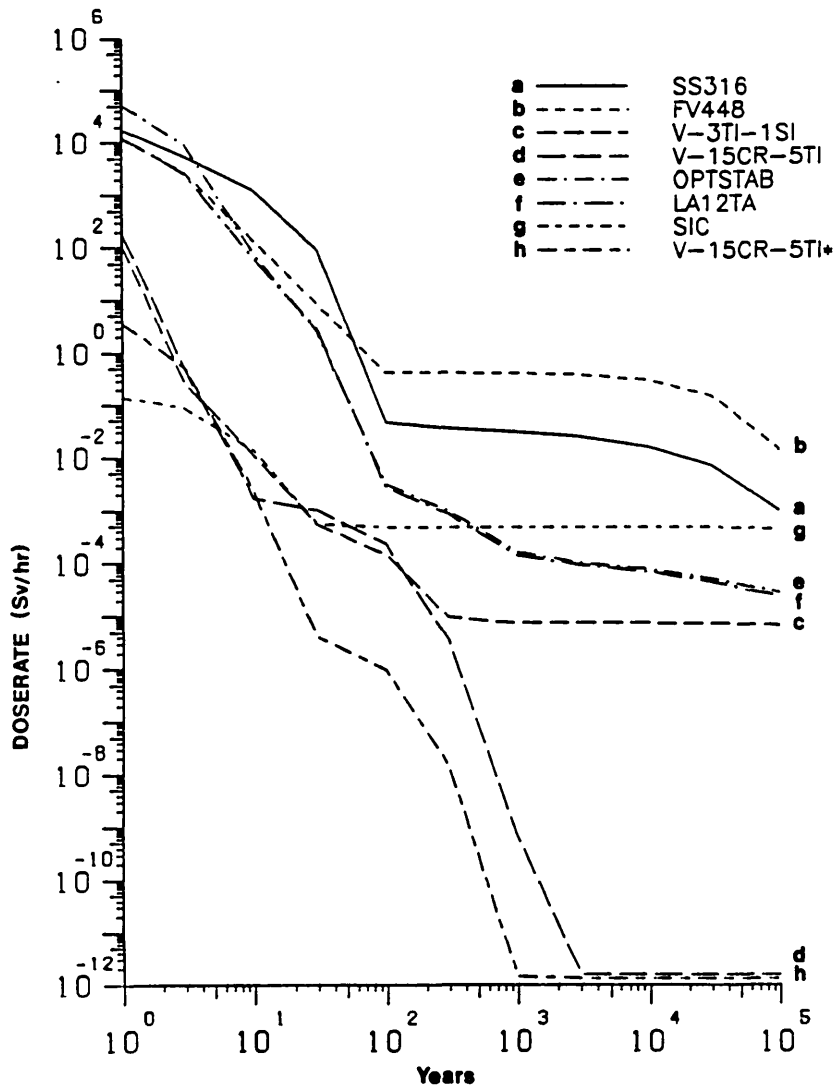


Figure 5.9 : Time dependence of surface gamma doserate (Sv/hr) from Culham candidate first wall compositions

7 candidate first wall materials. In addition to a cooling period of 100 years, a number of other cooling periods upto 100,000 years were considered, allowing the time dependence of activity and dose rate to be studied, the results presented in figures 5.8 and 5.9 . It must be remembered while looking at these figures that impurities have not been included in the vanadium alloys and SiC.

If the steels are taken first, it is seen that the overall features of the time-dependant specific activities and gamma dose rates have not changed substantially in comparison with the study by Giancarli ¹⁰ ; the dose rate reductions given by the low activation compositions are small, but they increase after about 30 years, such that after 1000 years they give dose rates which are approximately 2 or 3 orders of magnitude lower than those for SS316 and FV448 respectively.

The pure vanadium alloys give substantially lower dose rates than the steels at all times, particularly at long times, when the advantage is very substantial indeed. It is observed that the V-Ti-Si alloy becomes less attractive than the V-Cr-Ti alloy after about 100 years, due to the presence of Al26 in the Si inventory. An isotopically tailored version of the V-Cr-Ti alloy, in which the Ti component is present only as Ti50, shows some advantage over the 10 to 1000 year time interval, the period in which the dominant radio nuclide is Ar42 ($t_{1/2} = 33\text{yrs}$).

With regard to SiC, the obvious features of the dose rate curve are unaltered in comparison with Giancarli's results ¹⁰ ; a rapid fall in dose rate following removal from the reactor, followed by a high long-term dose rate, resulting from the long-lived species Al26. However, the altered dose rate for Si discussed in previous chapters increases the level of this tail by about an order of magnitude, and the

short-term dose is higher too, presumably from an increase in the Na22 concentration. The prospects for SiC suffer further when activation of the C component is examined : though the gamma doserate would be low, the potential ingestion dose from C14, produced by C13(n, γ), would be high in some exposure scenarios, since C is selectively absorbed by the biosphere.

It must be stressed again that potential impurities have not been considered for the vanadium alloys and SiC. Preliminary determination suggest that Nb, Mo and Ag would be present in vanadium ⁴⁸. Ideally it would be necessary to keep the levels of these elements down to about 0.005ppm, 0.5ppm and 0.0004ppm respectively, if the low intrinsic doserate of the vanadium host material is not to be compromised. It should be added, however, that since the limiting factor in vanadium alloys may be the long-term dose from the activation products of the titanium component, namely Ar42(K42), some relaxation of the allowable impurity levels may be permitted.

5.4 Potential Exposure Scenarios

5.4.1 Introduction

It is clear that the indices of specific activity and surface gamma doserate, useful as they are in providing a ready comparison between alternative wall materials, are somewhat limited in their usefulness. The suitability of a material for the first wall (on activation grounds) must ultimately be judged in reference to radiation exposure scenarios, and the application of dose limits provided by legislature or international guidelines. The potential doses received in these exposure scenarios will depend on physical details of the scenarios, such as shielding conditions, routes for the intake of radioactive

material, the timescale of the scenario etc. A natural result of improving the accuracy of the activation calculation, would be to try to improve the method for calculating doses.

Scenarios which could result in significant doses are

- 1) dispersion of radioactive material in the torus hall in the course of normal operation, in the form of dust or corrosion products in coolants
- 2) end of lifetime component handling
- 3) component recycling
- 4) accidental releases of activated material
- 5) permanent disposal of activated material

Whereas methodologies for the dosimetric assessment of scenarios 1) to 4) are still in their infancy, the timescales for 1), 3) and 4) do not match the 100 year cooling period upon which this study has been standardised. An acceptable first order approximation to 2) is given by surface gamma dose rate. The study has concentrated, therefore on the post-disposal environmental impact scenario, a scenario for which a provisional methodology already exists ¹⁶.

5.4.2 Post-Disposal Environmental Impact Scenarios

Maximum annual doses to members of the public, arising from the emplacement of 1000 tonnes of the candidate first wall compositions in a variety of repositories, have been calculated according to the models described in NRPB-R210 ¹⁶. It will be sufficient here to provide a brief outline of the repository designs and dosimetric models. The repository designs considered were

- a) a simple trench for shallow burial
- b) a well-engineered trench for shallow burial

- c) a deep geological repository in clay
- d) a deep geological repository in shale

In addition, the original study also examined deep ocean disposal. However, this option has lost favour in the intervening time, and has not been included here. The present study is restricted, therefore, to land-based disposal options.

For all four repository options the radiological impact of a "normal evolution" scenario, in which radionuclides were transported to the surface by ground water, where they were ingested with drinking water, was determined. For the two shallow burial options the radiological impact of an "intrusion" scenario, based upon the commencement of building work after a 125 year interval was determined. The exposure pathways considered in this scenario were ingestion, inhalation and external gamma, the latter assumed to be from a uniform concentration of radioactive material.

The maximum annual doses were calculated first for unit concentrations of long-lived nuclides, as shown in table 5.2 . It is relevant to note in this respect that the present study increased the number of radionuclides included in this stage, principally to consider the impact from vanadium alloys and SiC for the first time, but also to treat some previously updated nuclides in the updated steel inventories. The doses given by the unit concentrations of long-lived nuclides were then adjusted according to their respective concentrations in the inventories of pure elements and candidate first wall materials to give the maximum annual doses associated with the disposal of 1000 tonnes of material quoted in tables 5.3 and 5.4.

It is necessary, before a discussion can be developed on the

significances of the maximum annual doses which arise in the various disposal scenarios, to establish limits for what constitutes an acceptable dose. The obvious limit to take is the ICRP guideline⁴⁹ which states that a member of the public should not receive an annual dose in excess of 5mSv. However, with the large uncertainties that are present in the dose calculation, this limit may be to err on the side of complacency. A more cautious dose limit is provided by the principles for land disposal of waste issued by the UK Authorising Departments⁵⁰, which states "the appropriate target applicable to a single repository at any time is a risk to an individual in a year equivalent to that associated with a dose of 0.1mSv ; about one chance in a million". The latter, 0.1mSv limit, was taken as the standard in the original NRPB study¹⁶, and is also adopted here as the principal dose limit.

If the two standard steels are taken first, it is seen that using the updated inventories and including a wider range of nuclides does indeed modify the maximum annual doses. Most notable are increases to the doses for normal evolution of the shallow burial options, thought to be connected to an enlarged Tc99 inventory (produced from the Mo component by Mo100[n,2n] and Mo98[n,γ]), increases in the doses for building excavation of the simple trench, and a decrease in the doses for building excavation of the engineered trench. It is interesting to note that this last difference is against the trend set in NRPB-R210¹⁶ in that the engineered trench gives a lower dose than the simple trench. This conclusion may be the result of misreading of the methodology for the building excavation scenario outlined in NRPB-R210¹⁶, but the fact that in table 5.2 a unit concentration of the dominant radionuclide Nb94 gives a lower dose for the engineered option than for the simple

trench, suggests that the engineered trench should give rise to the lower dose. These modifications do not, however, alter the general conclusions drawn from the original report : that the doses for the shallow burial options exceed the 0.1mSv limit, narrowly in the case of the normal evolution scenarios, by a substantial margin in the intrusion scenarios, and that under the present guidelines the only viable option for these materials would appear to be deep geological burial. The correctness of this conclusion is, of course, subject to the validity of the many assumptions made in what is still after all a provisional dosimetric methodology, and may be overturned by altering the factors of ground water flow patterns, repository designs, impurity levels in materials etc.

Turning now to the low activation steels, there appears to be little advantage over conventional steels in normal evolution scenarios : maximum annual doses are similar to those for the two commercial compositions. Indeed, the use of N as an alloying addition in OPTSTAB leads to a somewhat higher dose. The picture differs somewhat from the earlier study with respect to the intrusion scenarios, however, with the doses from intrusion being of the same order as doses from normal evolution, the drop apparently resulting from reduced levels of Tb158 in the updated inventories. The doses for the shallow burial options are nevertheless still an order of magnitude or more greater than the 0.1mSv limit. Given, however, that that optimisation of the compositions of low activation steels has yet to be achieved, the shallow burial options cannot be ruled out completely at this stage.

With respect to the deep geological burial options for the low activation steels, it is seen that the present results give significantly lower doses than the original study (from $3.1\text{E-}7$ Sv/y to $2.8\text{E-}9$ for LA12Ta, and from $2.1\text{E-}7$ to $6.3\text{E-}9$ for OPTSTAB in clay), presumably

owing to new cross-section data employed in the UKACT1 database for reactions leading to Re186m from W (see Chapter IV). Deep geological burial remains the only option which would give doses well below the 0.1mSv limit for these materials.

It is important to remember again, while examining the maximum annual doses quoted in table 5.4 for the vanadium alloys and SiC, that impurities, which will almost certainly have a deleterious effect, have not been included in the treatment. This in part explains the size of the advantage vanadium alloys appear offer over steels. Another factor to consider is the omission of argon products, the dominant radionuclide in the Ti inventory after 100 years, in the treatment, since there is uncertainty over their behaviour in the environment and data giving biological hazard from ingestion/inhalation is lacking. In the case of the gamma-emitting Ar42, the omission can be partly justified by a half-life which is short in comparison with the timescale of the normal evolution scenarios (not, however, with respect to the intrusion scenarios). Of the two compositions investigated, both give rise to maximum annual doses below the 0.1mSv limit for all the scenarios examined, but the alloy containing a Si addition fares the least well, owing to its inventory of Al26. The figures, as they stand, suggest that it may be possible to employ shallow burial disposal options for vanadium alloys. Unlike the low activation steels, however, no further optimisation of the compositions is possible, and the figures can only get worse, as the effects of impurities and argon products are added.

The Al26 inventory is also responsible for the surprisingly high (in the light of previous claims for the material ¹⁰) dose given by intrusion scenarios following the disposal of SiC, which exceeds the

0.1mSv limit for both the simple trench and engineered trench designs. Since the scope for elemental tailoring is small, this high intrusion dose may effectively exclude shallow burial options for SiC.

In conclusion, the findings of the present study agree with the previous study ¹⁶ insofar as they suggest that shallow land burial would not be a suitable disposal option for a fusion reactor first wall constructed from FV448 and SS316 steels. They do not rule out the possibility, however, of employing this option for some low activation steels and vanadium alloys. The latter exhibit the most favourable activation properties with respect to post-disposal environmental impact of the materials examined in the study, but are likely to become less attractive when the treatment is expanded to include argon activation products and the effect of impurities in the vanadium bulk material.

Table 5.1 : Assumed compositions of candidate first wall materials
(with estimated levels of impurities in steels)

	FV448	SS316	LA12Ta	OPTST.	SiC	VCrTi	VTiSi
C	1.5E-1	3.0E-2	1.6E-1	7.0E-2	3.0E+1	-	-
N	6.0E-2	1.5E-2	4.2E-2	3.0E-1	-	-	-
Al	1.0E-3	1.0E-3	1.0E-3	1.0E-3	-	-	-
Si	4.0E-1	5.0E-1	3.0E-2	2.0E-1	7.0E+1	-	1.0E-0
K	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Ca	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Ti	-	-	-	-	-	5.0E-0	3.0E-0
V	2.5E-1	-	2.5E-1	-	-	8.0E+1	8.1E+1
Cr	1.1E+1	1.8E+1	9.0E-0	1.5E+1	-	1.5E+1	1.5E+1
Mn	7.5E-1	1.8E-0	8.0E-1	1.6E+1	-	-	-
Fe	8.6E+1	6.6E+1	8.8E+1	6.5E+1	-	-	-
Co	1.0E-3	1.0E-3	1.0E-3	1.0E-3	-	-	-
Ni	7.5E-1	1.2E+1	4.0E-4	4.0E-4	-	-	-
Cu	2.0E-3	2.0E-3	2.0E-3	2.0E-3	-	-	-
Zr	3.0E-3	3.0E-3	3.0E-3	3.0E-3	-	-	-
Nb	3.0E-1	2.0E-3	2.0E-5	2.0E-5	-	-	-
Mo	6.5E-1	2.4E-0	5.0E-3	5.0E-3	-	-	-
Ag	5.0E-5	5.0E-5	5.0E-5	5.0E-5	-	-	-
Sn	1.0E-3	1.0E-3	1.0E-3	1.0E-3	-	-	-
Ba	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Ce	4.0E-4	4.0E-4	4.0E-4	4.0E-4	-	-	-
Nd	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-
Sm	5.0E-5	5.0E-5	5.0E-5	5.0E-5	-	-	-
Eu	2.0E-6	2.0E-6	2.0E-6	2.0E-6	-	-	-
Tb	5.0E-5	5.0E-5	5.0E-5	5.0E-5	-	-	-
Ho	1.0E-6	1.0E-6	1.0E-6	1.0E-6	-	-	-
Ta	1.0E-3	1.0E-3	1.2E+1	1.0E-3	-	-	-
W	1.0E-3	1.0E-3	8.5E-1	1.2E-0	-	-	-
Ir	1.0E-5	1.0E-5	1.0E-5	1.0E-5	-	-	-
Bi	2.0E-4	2.0E-4	2.0E-4	2.0E-4	-	-	-

Table 5.2 : Maximum annual dose (Sv/y) from unit concentration (1TBq/tonne) of long-lived radionuclides.

	Normal Evolution				Building Exc.	
	Simple Trench	Eng. Trench	Deep Clay	Deep Shale	Simple Trench	Eng. Trench
H3					1.2E-06	4.8E-08
Be10	1.1E-08	1.1E-08	4.9E-14	7.8E-12	6.3E-00	2.9E-01
C14	1.3E-06	2.6E-08	8.3E-20	5.8E-16	4.3E-02	1.6E-03
Al26	8.5E-06	1.7E-07	1.8E-09	1.5E-08	1.3E+03	6.2E-02
Si32				1.5E+01	6.9E-01	
Ca41	1.6E-08	1.2E-08	1.9E-16	1.8E-13	3.1E-02	1.3E-03
Ti44				1.8E+02	8.4E+01	
Mn53	7.2E-12	7.1E-12	2.3E-25	2.8E-21		
Ni59	3.9E-09	2.2E-09	5.4E-15	1.8E-12	5.3E-02	2.3E-03
Mo93	1.6E-08	2.9E-09	3.6E-29	2.3E-24	5.0E-01	2.2E-02
Zr93	1.5E-09	1.5E-09	3.3E-17	2.9E-14		
Nb94	6.3E-25	5.3E-25			8.0E-02	3.5E+02
Tc99	8.6E-07	1.7E-08	7.3E-10	6.8E-09		
Ag108m					4.1E+02	1.8E+02
Cd113M					4.5E+02	2.0E-03
Pm145					1.1E-02	3.7E-03
Eu152					7.7E-01	3.4E-01
Eu154					3.2E-02	1.4E-02
Tb158					2.1E+02	9.5E+01
Ho166m					8.2E+02	3.7E+02
Ho178n					7.4E+01	3.3E+01
Hf182	1.5E-08	1.5E-08	6.3E-13	3.6E-11	8.3E+02	3.4E+02
Re186m	2.1E-06	4.1E-08	1.7E-09	1.7E-08	1.7E+01	7.3E-00
Bi207					7.9E+01	3.5E+01

Table 5.3 : Maximum annual dose (Sv/y) resulting from disposal of 1000 tonne of pure elements

	Normal Evolution				Building Exc.	
	Simple Trench	Eng. Trench	Deep Clay	Deep Shale	Simple Trench	Eng. Trench
C	2.0E-07	8.0E-09	1.8E-14	2.8E-12	4.4E-04	7.8E-06
N	1.8E-02	3.6E-04	4.5E-15	8.6E-12	0.11	1.6E-03
Si	1.7E-07	3.4E-09	3.6E-14	3.0E-10	4.9E-03	9.1E-04
Ti	1.6E-12	1.2E-12	1.9E-20	1.8E-17	7.3E-06	1.3E-06
V	3.1E-17	2.3E-17	3.6E-25	3.5E-22	1.5E-11	2.8E-12
Cr	1.3E-17	1.1E-08	1.1E-25	1.0E-22	5.9E-11	1.1E-11
Mn	1.0E-12	1.0E-12	3.3E-25	5.1E-22	5.8E-15	9.8E-18
Fe	8.0E-12	7.9E-12	1.6E-19	5.2E-17	1.5E-10	1.7E-11
Ni	9.8E-07	5.5E-07	1.4E-15	4.5E-10	7.8E-11	1.3E-03
Nb	1.1E-06	1.0E-06	2.7E-13	4.2E-10	1.1E+03	1.9E+02
Mo	2.0E-04	2.3E-05	9.6E-08	9.0E-07	5.3E-00	0.84
Ta	4.9E-08	1.0E-09	3.9E-11	3.9E-10	1.2E-02	2.1E-03
W	7.4E-06	1.4E-07	6.0E-09	6.0E-08	1.2E-02	2.0E-03

Table 5.4 : Maximum annual doses (Sv/y) resulting from the disposal of 1000 tonne of Culham candidate first wall materials

	Normal Evolution				Building Exc.	
	Simple Trench	Eng. Trench	Deep Clay	Deep Shale	Simple Trench	Eng. Trench
FV448		6.0E-04	1.6E-05	3.1E-08	2.9E-07	3.3
0.56						
SS316	3.7E-04	2.6E-05	1.1E-07	1.1E-06	0.17	2.5E-02
LA12Ta	3.8E-04	7.7E-06	2.8E-09	2.8E-08	8.5E-03	1.5E-03
OPTSTAB	2.7E-03	5.4E-05	6.3E-09	6.3E-08	8.9E-03	1.5E-03
V-15Cr-5Ti	4.0E-12	3.0E-12	4.8E-20	4.5E-17	3.6E-07	6.6E-08
V-3Ti-1Si	8.4E-08	1.7E-08	1.8E-11	1.5E-10	4.9E-05	9.1E-06
SiC	9.0E-06	2.4E-07	1.2E-09	1.0E-08	3.6E-03	6.4E-04

APPENDIX I

Returning to the treatment given to the bremsstrahlung correction in Chapter I, it can be shown from the study of X-ray production in tubes with thick targets ²⁷, that the number of photons dN produced in the photon energy interval dE , from an incident beam of monoenergetic electrons of energy E_0 , is given by the empirical relationship

$$dN = a Z \frac{(E_0 - E)}{E} dE$$

where a is a constant and Z the atomic number of target. If this expression is now modified by considering the incident electron beam to contain an energy continuum, then we obtain

$$dN = a Z \frac{(E_0 - E)}{E} f(E_0) dE dE_0$$

where $f(E_0)$ is the fraction of the electron flux in unit energy interval at energy E_0 . Therefore the photon flux in the i th photon energy interval ϵ_{i-1} to ϵ_i is

$$dN_i = a Z \int_0^{E_d} \int_{\epsilon_{i-1}}^{\epsilon_i} \frac{(E_0 - E)}{E} f(E_0) dE dE_0$$

where E_d is the beta endpoint energy. As shown in Chapter I, the inner integral can be resolved easily, giving

$$dN_i = a Z \int_0^{E_d} \left[E_0 \ln \left(\frac{\epsilon_i}{\epsilon_{i-1}} \right) - \epsilon_i + \epsilon_{i-1} \right] f(E_0) dE_0$$

If it is now assumed that information relating to the beta energy spectrum is available in an energy bin form, then the integral can be approximated by a summation over the beta energy bins thus

$$dN_i = a Z \sum_j \left[\ln \left(\frac{\epsilon_i}{\epsilon_{i-1}} \right) - \epsilon_i + \epsilon_{i-1} \right] E_j f_j \quad (A1.1)$$

where E_j is the mean energy of the j th beta energy bin, and f_j is the fraction of the total beta flux in that bin.

The total gamma dose rate can now be obtained simply from the combined N_i by an expression of the form

$$D = C \sum_i N_i E_i$$

(A1.2)

where C is a constant and E_i the mean energy of the i th photon group.

A comparison between a bremsstrahlung correction which assumes the electron beam to be monoenergetic, and a correction which assumes it to contain an energy continuum, can now be made. Tables A1.1 to A1.3 show the sizes of the respective corrections according to both methods for Be10, Cl36 and Y90 in terms of the group photon fluxes resulting from each electron energy bin, N_{ij} , and the dose proportional summation over i in expression A1.2.

Table A1.1 : Bremsstrahlung correction for Be10

Monoenergetic		Binnned distribution	
$\langle E_{\beta} \rangle = 203\text{keV}$		Bin 1	Bin 2
		(200)	(370)
100-200keV	40.70	21.25	35.64
200-300	0.022	0.0	11.77
300-400	0.0	0.0	2.07
$\sum N_i E_i = 6110.6$		$\sum N_i E_i = 12200.5$	

Table A1.2 : Bremsstrahlung correction for Cl36

Monoenergetic		Binnned distribution		
<E _β > = 246keV		Bin 1	Bin 2	Bin 3
		(200)	(450)	(871)
100-200keV	70.51	16.80	73.75	5.541
200-300	4.925		28.69	2.785
300-400			10.25	1.038
400-500			1.045	1.038
500-600				0.647
600-700				0.377
Σ N _i E _i = 11807.7		Σ N _i E _i = 27987.7		

Table A1.3 : Bremsstrahlung correction for Y90

Monoenergetic		Binnned distribution			
$\langle E_\beta \rangle = 695\text{keV}$		Bin 1	Bin 2	Bin 3	Bin 4
		(200)	(450)	(950)	(1505)
100-200keV	381.7	4.790	55.73	290.4	51.86
200-300	181.8		21.69	148.3	28.06
300-400	99.94		7.747	90.11	18.31
400-500	55.08		0.7896	58.23	12.97
500-600	26.71			38.07	9.592
600-700	7.153			24.15	7.260
700-800				13.96	5.553
800-900				6.180	4.249
900-1000				0.709	3.221
$\Sigma N_i E_i = 181809.$		$\Sigma N_i E_i = 256846.$			

It would appear from these results that even with the coarse representations of the beta decay spectra obtained from Browne and Firestone ²⁸, the bremsstrahlung correction is systematically underestimated roughly by a factor of 2 by representing the beta flux as a monoenergetic stream with average energy. The size of the discrepancy appears to mount, also, as the beta endpoint energy tends towards the 100keV cut-off value applied to gamma doserates here .

APPENDIX II

It is assumed in Chapter IV that the error associated with a single group fusion cross-section for a reaction of the threshold variety can be approximated by taking the error associated with the cross-section at 14MeV. That this is a reasonable assumption will be demonstrated for a reaction for which comprehensive experimental data exists in the fusion energy range : namely the Co59(n, α) reaction.

A rigorous treatment of the propagation of errors to a single group cross-section obtained by collapsing the multi-group cross-sections provided by an evaluated data file such as ENDF/B-V, must take into account the covariances between pointwise cross-section data in such a file, some of which derive from common measurements.

The variance in a quantity R which employs the n items of data x_i in its evaluation, is given by

$$\frac{\text{Var}(R)}{R^2} = \sum_i^n S_i^2 \frac{\text{Var}(x_i)}{R^2} + \sum_i^n \sum_j^n S_i S_j \frac{\text{Cov}(x_i, x_j)}{R^2}$$

(A2.1)

where $\text{Var}(R)$ is the variance in R, $\text{Cov}(x_i, x_j)$ is the covariance of x_i and x_j , and S_i is the differential sensitivity, defined as

$$S_i = \frac{\partial R}{\partial x_i}$$

(A2.2)

It is conventional, however, for evaluated cross-section files to present covariances in the form of a correlation matrix, whose members are related to covariances in the following way

$$\rho_{ij} = \frac{\text{Cov}(x_i, x_j)}{\text{S.D.}(x_i) \text{S.D.}(x_j)} \quad (\text{A2.3})$$

where S.D.(x_i) is the standard deviation of x_i . Expression A2.1 is therefore modified to

$$\frac{\text{Var}(R)}{R^2} = \sum_i^n S_i^2 \frac{\text{Var}(x_i)}{R^2} + \sum_i^n \sum_j^n S_i S_j \frac{\rho_{ij} \text{S.D.}(x_i) \text{S.D.}(x_j)}{R^2} \quad (\text{A2.4})$$

If one now lets R be the single group cross-section σ , and the x_i be multi-group cross-sections σ_i , defined thus

$$\sigma = \frac{\sum_k^{100} \sigma_k \phi_k}{\sum_k^{100} \phi_k} \quad (\text{A2.5})$$

where the ϕ_k are the group neutron fluxes, then the S_i can be evaluated by direct substitution into expression (A2.2) to give

$$\frac{\partial \sigma}{\partial \sigma_i} = \frac{\phi_i}{\sum_i^{100} \phi_i} \quad (\text{A2.6})$$

Substitution into expression (A2.1) now gives the variance of the single group cross-section to be

$$\frac{\text{Var}(\sigma)}{\sigma^2} = \sum_i^n \frac{\phi_i^2}{\left(\sum_k^n \sigma_k \phi_k\right)^2} \text{Var}(\sigma_i) + \sum_i^n \sum_j^n \frac{\phi_i \phi_j}{\left(\sum_k^n \sigma_k \phi_k\right)^2} \rho_{ij} \text{S.D.}(\sigma_i) \text{S.D.}(\sigma_j) \quad (\text{A2.7})$$

This expression has been evaluated from the 22 energy group cross-section data shown in table A2.1, which is derived from measurements taken by Meadows *et al*⁵¹ of the Co59(n,α) cross-section. The corresponding 22 group DEMO first wall neutron energy spectrum was obtained in an approximate fashion from the 100 group GAM-II spectrum employed in the activation calculations by linear interpolation, and is given in table A2.2. The correlation matrix employed in the calculation, also obtained from Meadows *et al*⁵¹, is given in figure A2.3.

The resulting standard deviation in the single group cross-section, expressed as a percentage of the cross-section, was found to be 4.904%. This is significantly more than the error associated with the cross-section for group 22, suggesting that the errors associated with the cross-sections over the rest of the fusion neutron energy spectrum do contribute significantly to the error for the single group cross-section. It is concluded from an examination of the summations in expression A2.7, however, that the single group variance is largely the result of terms for which $i = 22$, the principal contribution coming from the $i = 22, j = 22$ term. To a first order approximation, the level of approximation appropriate for the sensitivity analysis described in Chapter IV, the total error in the 14MeV (group 22) cross-section (3.9%) can represent the error associated with the single group Co59(n,α) cross-section (4.9%).

The validity of this approximation may be undermined, however, for reactions whose excitation functions depart radically from those generated by models, and more comparisons of the type made above will be required before a definitive statement can be made.

Table A2.1 : Cross-section evaluation for Co59(n, α) from Meadows *et al* ⁵¹

Group	Energy (MeV)	X-section (mb)	Total error (%)
1	4.0	0.00074	20.8
2	4.5	0.0089	16.0
3	5.0	0.096	11.5
4	5.5	0.38	7.6
5	5.75	0.66	5.7
6	6.25	1.41	4.7
7	6.75	2.65	4.7
8	7.25	4.31	4.3
9	7.75	6.53	3.6
10	8.25	8.97	3.5
11	8.75	11.39	3.5
12	9.25	12.71	3.9
13	9.75	14.54	3.8
14	10.25	16.35	5.0
15	10.75	18.52	5.6
16	11.25	20.38	5.9
17	11.75	22.51	5.9
18	12.25	25.05	5.9
19	12.75	26.88	5.9
20	13.25	28.43	5.7
21	13.75	29.64	5.1
22	14.25	30.13	3.9

Table A2.2 : 22-group neutron fluxes appropriate for the DEMO first wall

Group	Energy Range (MeV)	Flux (n s ⁻¹ cm ⁻²)
1	3.75 - 4.25	3.22E+12
2	4.25 - 4.75	2.65E+12
3	4.75 - 5.25	2.22E+12
4	5.25 - 5.625	1.38E+12
5	5.625 - 6.0	1.25E+12
6	6.0 - 6.5	1.54E+12
7	6.5 - 7.0	1.47E+12
8	7.0 - 7.5	1.54E+12
9	7.5 - 8.0	1.52E+12
10	8.0 - 8.5	1.43E+12
11	8.5 - 9.0	1.44E+12
12	9.0 - 9.5	1.36E+12
13	9.5 - 10.0	1.21E+12
14	10.0 - 10.5	1.15E+12
15	10.5 - 11.0	0.77E+12
16	11.0 - 11.5	0.738E+12
17	11.5 - 12.0	1.09E+12
18	12.0 - 12.5	1.04E+12
19	12.5 - 13.0	1.00E+12
20	13.0 - 13.5	0.962E+12
21	13.5 - 14.0	0.927E+12
22	14.0 - 14.5	65.5E+12

Table A2.3 : Correlation matrix from Meadows *et al* ⁵¹.

1.
0.10 1.
0.14 0.18 1.
0.21 0.27 0.37 1.
0.27 0.36 0.50 0.75 1.
0.16 0.21 0.30 0.45 0.60 1.
0.11 0.14 0.20 0.30 0.40 0.64 1.
0.09 0.12 0.17 0.26 0.34 0.60 0.82 1.
0.13 0.16 0.23 0.35 0.46 0.54 0.62 0.68 1.
0.12 0.16 0.22 0.33 0.44 0.54 0.47 0.54 0.70 1.
0.10 0.14 0.19 0.29 0.38 0.49 0.45 0.45 0.60 0.70 1.
0.08 0.11 0.15 0.23 0.31 0.40 0.40 0.42 0.47 0.59 0.79 1.
0.08 0.10 0.14 0.22 0.29 0.37 0.36 0.40 0.48 0.48 0.59 0.68 1.
0.05 0.06 0.09 0.14 0.18 0.22 0.20 0.21 0.30 0.31 0.23 0.33 0.57
1.
0.04 0.05 0.07 0.11 0.14 0.16 0.16 0.16 0.19 0.22 0.15 0.12 0.36
0.71 1.
0.04 0.05 0.06 0.10 0.13 0.13 0.13 0.13 0.14 0.15 0.13 0.12 0.17
0.50 0.75 1.
0.03 0.04 0.06 0.09 0.12 0.13 0.12 0.12 0.14 0.13 0.13 0.12 0.13
0.19 0.48 0.73 1.
0.03 0.04 0.06 0.09 0.12 0.13 0.12 0.12 0.14 0.13 0.13 0.12 0.13
0.12 0.19 0.46 0.73 1.
0.03 0.04 0.06 0.09 0.12 0.13 0.12 0.12 0.14 0.13 0.13 0.12 0.13
0.12 0.13 0.19 0.46 0.73 1.
0.03 0.04 0.06 0.09 0.12 0.12 0.11 0.12 0.13 0.13 0.12 0.13 0.12
0.12 0.12 0.13 0.18 0.46 0.73 1.
0.02 0.03 0.04 0.07 0.09 0.09 0.09 0.09 0.10 0.10 0.09 0.09 0.09
0.09 0.09 0.10 0.10 0.16 0.46 0.74 1.
0.02 0.02 0.03 0.05 0.06 0.06 0.06 0.06 0.07 0.07 0.07 0.06 0.07
0.06 0.06 0.07 0.07 0.07 0.14 0.48 0.66 1.

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