Aspects of Calculational Needs for Spallation Facilities

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Abstract

The role of calculations in the design of spallation facilities is considered in general terms: what needs to be calculated, what needs to be taken into consideration and how the present imprecise knowledge of the uncertainty of such calculations may be circumvented.

1 Introduction

Spallation facilities involve the bombardment of some material assembly (usually large) with a beam of particles (normally protons) of energy up to a few GeV. These facilities exploit one or more aspects of the particle cascade initiated by the incident beam to do some particular job. The present most common use is to provide a 'thermal' neutron source for condensed matter studies but they are also considered for use in tritium breeding, as neutron sources for damage studies, in fertile to fissile breeding and for the transmutation of active waste. Despite the diverse goals, the same physical processes are involved in all of them and basically they only differ in the selection of materials and geometric configuration that enhances the planned use and the relative importance of the nuclear physics processes; very similar technical problems have to be solved if a reliable operational device is going to be built. This means that experience from all such existing facilities is applicable and can provide relevant information on practical and calculational matters. Further relevant information comes from the meson factories (TRIUMF, PSI, LAMPF), where beam powers ranging up to the region of 1 MW are handled as a matter of routine. Providing target stations and beam dumps also involves the solution of similar technical problems.

Despite the existence of operating spallation facilities and their intrinsic similarity, new facilities will still involve solutions of both 'old' and 'new' technical problems and require considerable calculational effort to bring to fruition.

The paper will make some rather general comments about calculations for the design of spallation facilities - what needs to be calculated and why, calculational tools and errors. Descriptions of neutron sources etc. will not be given as these may be found, together with discussion of detailed problems, in the Proceedings of the ICANS (International **C**ollaboration on **A**dvanced **N**eutron **S**ources) meetings[1-10].

2 The Requirements from Calculations

Calculations are required at three main stages:

(i) to demonstrate feasibility,

(ii) to optimize the materials and their geometric configuration,

(iii) to assist in the engineering design at the realisation phase.

The first two stages are facility specific and will seek out and then optimize the choice of materials and geometry that enhance the particular aspect of the particle cascade to be exploited. Somewhere within this stage the nuclear physics aspects of the cascade important to the concept are identified and the theoretical treatment (or perhaps the whole concept) verified by experiment (e.g. the IEND comparisons apropros waste transmutation, the SNQ study in Germany[11]). Good examples of configuration optimization are the tailoring of moderators in pulsed neutron sources (see the ICANS Proceedings and reference 12) and the use of enriched uranium as target material at Argonne National Laboratory's IPNS[13] facility as a means of getting round low beam power.

At the engineering design stage, calculations (and within the scope of calculations comes extrapolation of measurement and/or experience from other facilities) are required to give estimates of power densities, activation and radiation damage in the inner part of the facility and the specification of a range of shielding requirements. Calculations also provide essential information for safety analysis of the plant and may also be required to assess decommissioning waste. Power density, specific activation and damage will vary by many orders of magnitude through the system and will follow roughly similar distributions. A range of values of about 7 orders of magnitude is significant for power density, 12 to 13 for specific activation and 2 to 3 for radiation damage.

A further very important job is to identify and help reconcile engineering requirements that run contrary to the goal of the facility (i.e. ensure that engineering realities don't jeopardize the reason for building

Table-I

The Performance of SINQ with several target systems.

	Liquid Pb (ideal)	Liquid Pb (1st Design)	Tungsten Plates	Tantalum Plates	Zircaloy Plates
Neutron Production	10.87	10.46	10.04	10.46	5.53
Loss by Escape	.33	0.29	0.19	0.16	0.14
Loss by Absorption:					
Inner region	2.00	3.22	5.43	6.60	1.01
Moderator	1.58	1.09	0.77	0.61	0.77
Outer region	6.87	5.80	3.65	3.11	3.59
Useful Flux	$1.30 \cdot 10^{14}$	$0.85 \cdot 10^{14}$	$0.60 \cdot 10^{14}$	$0.45 \cdot 10^{14}$	$0.59 \cdot 10^{14}$
$\frac{Fluence}{OuterCapture}$	0.00312	0.00234	0.00264	0.00232	0.00263

Flux is in units of /cm^2/sec at 1 mA
Other values are number/proton onto target

the source). As an example of this, the estimated performance of PSI's neutron source, SINQ, with a variety of targets is shown in Table-I: the first practical design for the liquid Pb target involved the use of steel with consequent performance degradation because of neutron absorption; tungsten and tantalum are attractive materials for a solid target but equally poor performance comes from Zircaloy (further details on this problem may be found in reference 14).

Activation, either directly or indirectly, has the major impact on the engineering design of a spallation facility. Handling active components is, at best, a very time consuming occupation. To obtain a viable plant availability, component layout and material choices have to be a compromise between the needs of active handling and reliability and the needs of the facility's goal.

The problems of activation can be summarized in terms of three half life domains:-

(i) Short: these nuclides both reach equilibrium and decay quickly to give, in effect, contributions to the prompt dose-rates and power. Of major importance are estimates for the cooling fluids; these are required to quantify dose-rates outside pipe runs and plant houses (to allow specification of shielding) and dose-rates (hence damage) to circuit components (valves, pumps, sensors, resin beds, etc).

(ii) Medium (half life up to a few years): these nuclides are the major concern for active handling as they build up significantly during operation but decay rather slowly:-

The time for the dose-rate to decay to acceptable levels is the deciding factor for the amount of hands-on maintenance possible.

They determine the shielding requirements for transport flasks for active components.

They contribute to the activation in cooling circuits. Likely levels of contamination (tritium, ^{7}Be, active corrosion products) are required so that the circuits may be properly handled.

They are a major influence on the hazard rating of the facility and hence the criterion for the specification of containment.

(iii) Very long: these determine the disposal route for the active components both during running and at decommisioning.

Power density values throughout the system are essential information for the engineering design of the various components as is also assessment of radiation damage (which is directly linked to energy deposition). Components will need cooling. In the inner regions (points close to where the primary incident beam enters the material of the system), (i) providing cooling is a major engineering problem particularly as coolant can adversely affect facility performance (e.g. spallation neutron source targets) and (ii) as power density (and damage rates) have to be limited it sets the maximum practicable current density for the incident beam: cooling leads to temperature gradients (as volume heating is involved these go as the square of thickness), hence stresses. The stresses are cyclic (from pulsing of the accelerator - macroscopic and/or reliability) and have to be within (or near) the elastic region: radiation damage alters the mechanical properties. Also, any cooling circuit directly removing power deposited by nuclear particles will result in the coolant being activated and also potential problems from radiolysis. Cooling should be restricted to regions where it is really required (reliability, complexity, cost etc.); this means that power density values need to be extended far enough out so that this region can be positively identified.

3 Calculational Requirements

The basic requirement is a 'tool kit' of codes which covers, in the most effective manner, all aspects of the nuclear physics of the particle cascade initiated by medium energy nucleons in complex multi-media assemblies. The codes tend to be kept separate on practical grounds (they all tend to be individually large) and because tracking all particles of every cascade is not normally necessary. They are linked in some manner to allow produced but non-transportable particles in one to be passed to another. Each code element is given limited testing (their capability normally vastly exceeds data for checking) but, even having knowledge of individual accuracies, still leaves the major open question of how residual errors propagate through the complete calculation.

The code package which has been used for SINQ is shown in Fig. 1; this is the PSI version of the HETC package and uses Monte-Carlo models that cover most aspects of the cascades initiated by protons of energy < 3.5 GeV in complex geometries. Packages that do similar jobs are LAHET[15] and HERMES[16]. The structure is modular and the specific codes may be substituted by any others that do an equivalent job. The essential elements (shown as double lined boxes in Fig. 1) are:-

A geometry package.

The geometries being studied are complex and, to avoid unnecessary duplication of difficult tasks, all codes should use the same package. Although highly simplified geometric models can be adequate at early stages, eventually everything has to be confirmed with as accurate a representation as possible. It is probably this feature alone that forces the use of Monte-Carlo. An essential feature of the geometry package is some means of checking that the data specified does actually represent the system being modelled.

A cross-section based neutron transport code.

Neutrons are the most numerous and/or significant particles in the cascade. The majority will enter the energy range of cross-section libraries and are preferably transported using these rather than theoretical models. Presently the energy range from thermal up to 20 MeV is well covered[17]. The choice of codes is wide but needs to be able to handle complex multi-media geometries: O5R_PSI is the author's present preference, as knowing the code gives the advantage that it may be adapted to the problem rather than the other way round. There will be a suite of programmes to process cross-sections (retrieval from the library, resonance unfolding, doppler broadening, translating them into the form required by the neutronics code).

In terms of the package, the neutron source handled by this part is produced by the 'high energy' code. The transport itself will yield a gamma source. In some cases an auxiliary neutron source might come from (γ,n) reactions (e.g. high energy γs come from π^0 decay).

The same code will normally also handle thermal neutron transport. Because of the long residence time of thermal neutrons in the SINQ system, this is handled as a separate step in the calculational sequence.

A 'high energy' code

This uses theoretical models to handle the parts of the cascade with neutrons above the energy range

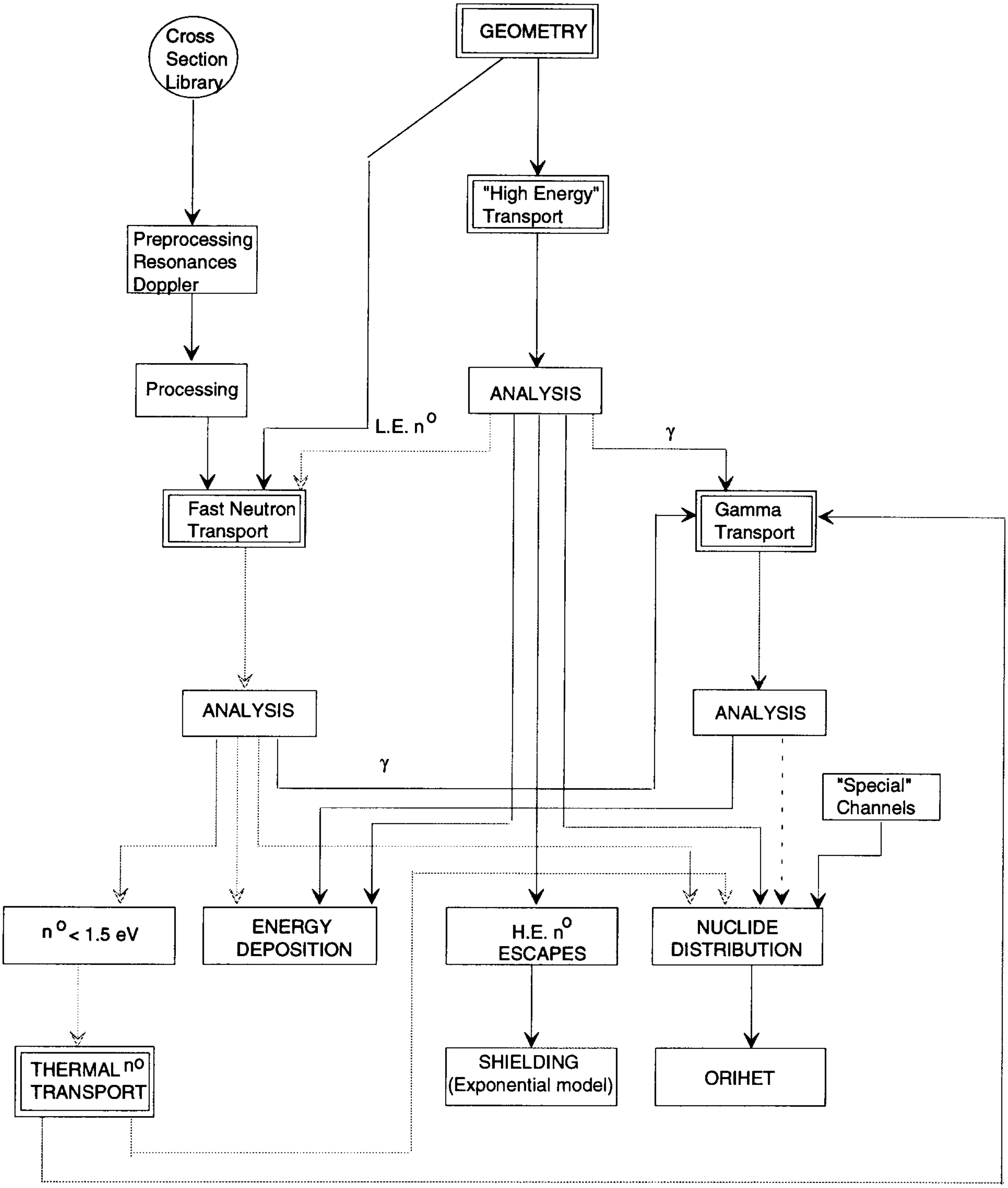

Figure 1: The PSI version of the HETC code package.

of cross-section data and charged particle transport; at high enough energies pions are produced. These codes are built round (subroutine) versions of particle nucleus interaction programmes (Bertini[18] for HETC[19], Chen et al.[20,21,22], Barachenkov et al.[23]); HETC is used at PSI. The essential physical processes are slowing down and coulomb scattering of charged particles, and elastic and ineleastic particle nucleus interactions. The high energy nucleon induced cascade results in the production of secondary nucleons, charged pions & muons (pion decay), neutral pions, fast neutrons, gammas and light ions. Neutral pions decay to high energy gamma pairs and together with nuclear gammas are passed to the appropriate code. Protons are followed to a minimum energy of 15 MeV at which they are assumed to be slowed to rest, neutrons are transported to a user specified minimum energy and then passed to the fast neutron code. Light ions are not transported but can be extracted for use as the source for a separate code if required.

A Gamma Transport Code.

For SINQ, gamma contributions in relation to source performance are not too important and point kernel integrations are adequate. Some detailed work on gamma escape probabilities from targets was made using the EGS[24] system.

Results Analysis.

In the main, the various elements of the code package describe the particle cascade in terms of events. These events are then analysed to yield the results required either with in code routines or with off line codes using a 'history' file output by the transport code. The 'standard' results required are for heating, radiation damage and activation. Activation estimates will require solution of the Bateman equations for the buildup and decay, as only production rates will come from the cascade calculation; depending on the system being studied, auxiliary nuclide production rates may also be required (e.g. α-particle induced reactions).

Particle fields for assessment of prompt dose-rates are also required. Although such a package may be applied to shielding design, this is better handled by other methods (S_n, Discrete Ordinates, point kernel, etc.).

4 The Problem of Errors

The calculations employ a set of codes that use a variety of approximate models for treating individual aspects of the nuclear physics of the interaction of particles with material. The answers are usually macroscopic quantities (e.g. heating rates, activation) which are averages over a wide range of events. A formal assignment of uncertainty to the answers is stymied by the difficulty of assessing how errors in the individual models propagate through the calculation to them. Despite the number of running spallation facilities (which seem to perform much as calculation predicted) there is no actual quantification of how good calculations really are. Qualitative/semi-quantitative assessments on the HETC package may be found in references 25 and 26. Although no formally assessed uncertainties can be given, there are things that can be done to minimize the likelihood of serious error.

The physics that should be being treated in the calculations has to be identified and the relevant parts of the code package checked as well as possible. During the actual calculations, help comes from (i) collecting results in a way that the relative importance of the various contributions can be seen, (ii) checking that the calculation at least conserves charge, mass and energy, (iii) looking for consistency. Operating neutron sources have documented calculational results; although they may not be directly compared (or comparable) with reality, such values can serve as a check-by-inference on a case by case basis.

The results from the calculations of the performance of SINQ with various targets shown in Table-I give a first illustration of some of these points. The first requirement is that neutron production and loss balance. A further check is that the flux in the moderator at points somewhat remote from the targets will be related to the neutrons escaping from the outer surface of the moderator by the thermal neutron transport characteristics of D_2O and the geometry only; the entries under 'fluence/outer capture' are the undisturbed fluxes per proton at the radius of a beam tube divided by the number of captures at the periphery of the tank (per proton). The average value is 0.00261 ± 0.0003 which indicates self consistency in the calculations. Nobody goes into an expensive project like SINQ without good evidence that it can reach the expected performance: comparison of thermal flux measurements in models of SINQ[27] with values from these calculations[26] show very reasonable agreement.

To illustrate further some of the points made, some comments on energy deposition and activation are given in the next two subsections.

4.1 Energy Deposition.

Calculated gross distributions of prompt power (decay power is not included) in components of a neutron source with 570 MeV and 3490 MeV protons onto the target are shown in Table-II. The contribution to the heating has been broken down into three components: from the high energy part of the cascade, from the fast neutrons (mainly produced in the inner part of the system) and from gammas. In both cases, the calculations accounted for all the incident proton energy (94% and 86% of this appears as prompt power, the remaining is binding energy). The high energy contribution dominates in the inner region and gammas at the outer. As these results are for a thermal neutron source, fast neutrons give a significant contribution to the power in the moderator material (D_2O) during slowing down and the gamma source from thermal captures is significant.

More important are power densities: in the same calculation, the contributions to the peak power density obtained were:-

Contributor	keV/g/proton 570 MeV	keV/g/proton 3490 MeV
$\frac{dE}{dX}$	27 (79%)	45 (38%)
Ion Recoils	5 (15%)	54 (46%)
Gammas	2 (5%)	18 (15%)
Totals	34	117

It is useful to work with power density per unit mass as this tends to vary rather slowly with nuclear composition. At 570 MeV ionization loss dominates and as this comes from the Bethe-Bloch formula, which is good at the percent level, the calculated value ought to be rather reliable (other calculational considerations excepted e.g. has coulomb scattering spread the beam too little or too much?). At the higher proton energy, similar magnitude contributions come from $\frac{dE}{dX}$ and ion recoils: the total number of ion recoils has risen due to the increase of secondary interactions, so now the uncertainty in this and the amount of energy going into ion recoils couple to the energy density. Calculated distributions of energy in the interaction of protons with Pb are shown in Fig. 2. The quite good agreement between measurement and prediction for neutron beam intensities gives a strong indication that the number of evaporated neutrons (and hence at least part of this energy distribution) is about right.

A further need for breaking down the energy deposition into components comes when trying to compare calculated values with available data. Heating rates in various samples located near to a spallation target were measured at TRIUMF[28]. Recent comparison with values deduced from calculation[29] gave good agreement (see Table-III Note: the estimated contribution from thermal neutron capture in the sample[28] was not included in the comparison and these values, which can be very important for specific elements, are included in the last line of the table under 'absorption'). The calculated contributions from the various components of the particle field show that, when translating measured values to different situations, the mix of the irradiating particle field needs to be taken into account and also that considerable caution should be exercised if large changes in the relative importance of the contributions are involved.

TABLE-II

Calculated energy deposition in components of a neutron source (all values are in units of MeV per proton incident at the target).

(a) 570 MeV Protons

	Gamma Source		Deposited Energy			
	E* & π^0	Capture	High Energy	Fast Neutron	Gamma	Total
Target	12.27	14.69	394.62	18.57	23.20	436.39
C.C.	0.073	5.78	0.494	0.52	0.39	1.40
D_2O	2.015	6.12	22.89	18.61	23.79	65.28
D_2O Tank wall	0.031	8.48	0.204	0.025	2.09	2.32
H_2O	0.089	13.52	1.024	-	6.24	7.26
H_2O Tank Wall	0.016	0.001	0.069	-	0.89	0.96
Shield	-	-	16.1	-	6.94	23.04
Totals	14.95	48.60	435.37	37.72	63.54	536.7

(b) 3490 MeV Protons

	Gamma Source		Deposited Energy			
	E* & π^0	Capture	High Energy	Fast Neutron	Gamma	Total
Target	372.86	124.22	1332.2	163.7	371.90	1867.82
C.C.	1.26	40.12	4.53	3.78	6.14	14.45
D_2O	40.45	43.17	259.53	154.37	272.53	686.42
D_2O Tank wall	0.62	63.34	3.83	0.31	17.01	21.16
H_2O	2.24	101.42	17.70	-	50.62	68.32
H_2O Tank Wall	0.13	0.005	0.94	-	7.42	8.36
Shield	-	-	285.0	-	64.22	349.22
	417.57	372.28	1903.72	322.18	789.84	3015.74

TABLE-III

Comparison of heating rates measured at TRIUMF[28] with values from scaling SINQ calculation results. Values are given in mW/g for 10 μA proton current

Sample	D_2O	H_2O	Be	C	Al	Fe	Cu	Zr	W	Pb	Bi
Density (g.cm^{-3})	1.1	1.0	1.85	1.62	2.7	7.86	8.93	6.51	19.3	11.3	9.75
A	2	1	9	12	27	56	63.5	91.2	184	207	209
Contribution	Heating Rates										
Fast n^0	1.74	1.96	0.70	0.56	0.27	0.13	0.12	0.08	0.04	0.04	0.04
γ-rays	1.04	1.04	1.04	1.04	1.04	1.04	1.04	1.04	1.04	1.04	1.04
H.E.	1.08	1.52	0.57	0.40	0.17	0.09	0.09	0.07	0.04	0.04	0.04
Calculated Total	3.86	4.52	2.31	2.04	1.48	1.26	1.25	1.19	1.12	1.12	1.12
Measured	5.46	5.77	2.58	2.36	1.67	1.26	1.25	1.20	1.29	1.11	1.14
$\frac{Measured}{Calculated}$	1.41	1.28	1.12	1.16	1.13	1.00	1.00	1.09	1.15	0.99	1.02
Absorption	0	0	0	0	0.03	0.52	0.67	0	0.98	0	0

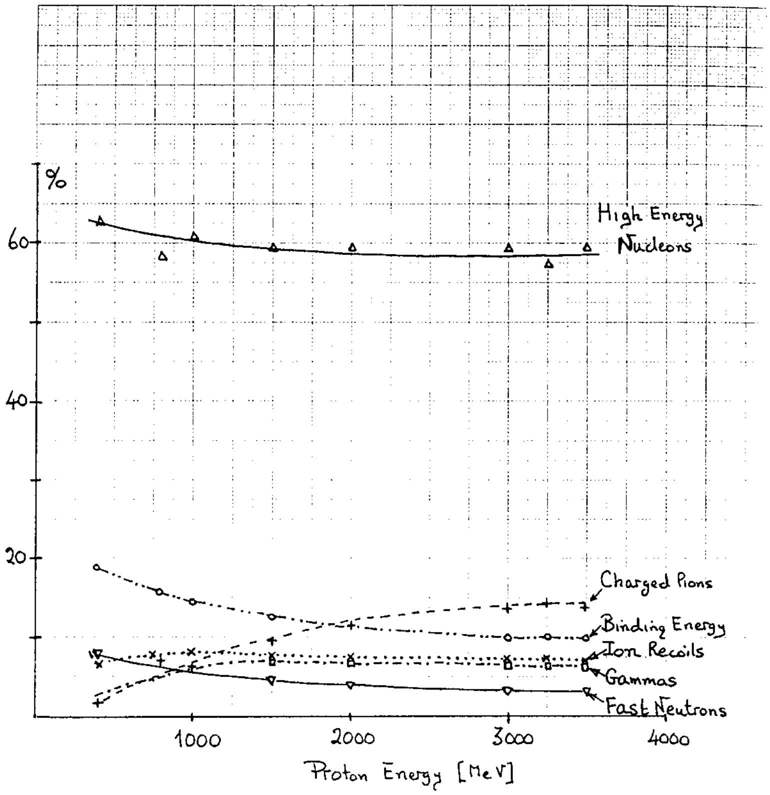

Figure 2: The percentage of the energy of the incident proton taken via particular channels in interactions with Pb as a function of incident energy.

Table-IV

The Distribution of Nuclide by Mass and Half Life

		Time Range (Years)								
A_{min}	A_{max}	< 0.01	0.01 0.1	0.1 1.0	1.0 10	10 100	100 500	500 1000	>1000	Sum
0	20	28	0	1	0	2	0	0	2	33
20	40	108	3	1	1	0	2	0	2	117
40	60	76	5	8	1	2	0	0	4	96
60	80	133	3	4	1	0	1	0	2	144
80	100	212	7	10	0	3	0	1	8	241
100	120	243	7	8	4	1	1	0	2	266
120	140	238	17	8	2	3	0	0	5	273
140	160	214	13	11	4	5	2	0	7	256
160	180	187	9	6	5	1	0	0	2	210
180	200	193	10	12	1	1	2	0	2	221
200	220	184	5	2	2	2	1	0	4	200
220	240	85	13	1	4	3	0	0	13	119
240	260	24	3	1	0	3	2	0	5	38
Totals		1925	95	73	25	26	11	1	58	2214

4.2 Activation

Activation calculations are subject to a rather wide range of uncertainties. There are a set that come from the estimation of the nuclide production rates and give uncertainty to the nuclide inventory: (a) the error in the mean and the spread of the charge and mass of the product nuclides in any unique particle + energy + target-nucleus calculation, (b) the error in the type and energy spectrum of the irradiating field, (c) the uncertainties in the nuclear composition of the materials. There are also uncertainties related to the build-up/decay calculation and use of the results: in the nuclide decay data, details of the decay spectra and the, often ill-defined, solid angle between the activated components and the person/object being subjected to the decay radiation.

In making activation estimates in support of the engineering design, practical considerations in combination with experience help:

- Gamma attenuation lengths are quite short (20 to 30 cm^2/g) and hence quite generous tolerances can be swallowed
- There is a substantial amount of measured data on the activation characteristics of common materials of construction at the working facilities and surveys giving useful information on materials in specific radiation fields[30].
- A high activation D_2O coolant circuit is operated at the ISIS facility[33].

Some quantitative feel for the uncertainties in inventory estimates can be obtained by looking at available information in a rather general way. The majority of nuclides have a short half life. The results of an analysis of decay data in terms of half life and mass is given in Table-IV. The comparative sparseness of the longer half life nuclides means that estimating the more persistent activation will be more susceptable to uncertainty. The nuclides with half life greater than 10 years and their precursors are shown on a (Z-N) plot in Fig. 3.

Correlation formulae (within their applicable regions) are very useful for obtaining rapid estimates for nuclide production rates (Rudstam[31], Silberberg & Tsao[32]). Values, in good agreement with those from correlations, have been obtained using HETC and, because these are based on fits to experimental data, this agreement is indirect evidence that the code is doing quite a good job.

The errors in the nuclide production rates will be caused by the calculational models ejecting the wrong number of nucleons (as we are talking of an average over many interactions, this need not be integer). To obtain some feel for the orders of magnitude, we can consider the effect to cause a shift in Z and N of the nuclide distribution: the spallation product correlation of Rudstam[31] gives a good starting point. For a nucleus (Z_t, A_t) bombarded by a particle of energy E, the production cross-section to a product (Z_p, A_p) is given by

$$\sigma(Z_p, A_p) = f(A_t, E)e^{P(A_t - A_p)}e^{-R|Z_p - Z_{max}|^{\frac{3}{2}}}$$

The pre-exponential function, $f(A_t, E)$, involves the mass of the target nucleus, the energy and type of the incident particle only. The parameters P, R, and Z_{max} (the most probable value of Z_p) are functions of E and mass of the product nuclide only. Using the appropriate values for the constants, the ratios of the production cross-sections for masses $A_p - 1$ and A_p (at the appropriate Z_{max}) for nucleon bombardment are:

Energy (MeV)	200	400	800	1600	3200
Ratio	0.6	0.73	0.82	0.88	0.95

In the charge direction the relevant parameters are the most probable charge, Z_{max}, the most stable charge, Z_{stab}, and the width parameter R for a given product's mass (A_p):

A_p	Z_{stab}	Z_{max}	R
50	22.6	23.4	2.03
100	42.8	44.8	1.49
150	61.5	63.4	1.24
200	78.9	82.0	1.09

The greater range of possible product nuclides at 'high' energy gives a slower rate of variation and estimates for particular masses should not be too sensitive to incorrect loss of 'A' in the calculation. At lower energies (or in thick targets bombarded by high energy particles, where the secondary interactions in the region of a few 100 MeV can dominate) a shift of 1 mass unit would lead to errors in the production cross-section of 50 to 60%.

The products are displaced to the neutron deficient side of the line of stability by a few charge units and with a comparatively narrow spread. Both the spread and the displacement reduce as the product mass becomes lighter (either because the target nucleus is light or, at the extremes of the distribution, from high energy interactions in heavy nuclei). In terms of estimates for long half life products (which are normally close to the stables), the production rates are likely to be more sensitive to mass rather than charge errors. Charge displacement due to the ejection of too little Z will still lead to the product being on the appropriate chain (Fig. 3) and a fairly severe excess charge emission would be required to push the product of the chain in the other direction.

The comparison of predicted nuclide production cross-sections with measurement should be rather a good test of the quality of the handling of the complete interaction by the high energy code. The information in Fig. 3 suggests that some of the long half life nuclides from high energy bombardment of fairly heavy elements (where fission should have an appropriately small effect) could be good candidates (e.g. $1.57 \cdot 10^7$ year ^{129}I $(\beta^-\gamma)$, $6.5 \cdot 10^6$ year ^{107}Pd (β^-), $1.5 \cdot 10^6$ year ^{93}Zr (β^-), 10.76 year ^{85}Kr $(\beta^-\gamma)$, $6.5 \cdot 10^4$ year ^{79}Se (β^-), 100 year ^{63}Ni (β^-), 269 year ^{39}Ar (β^-)); as they are protected by stable isotopes and normally on β^- chains there is a reduced possibility of interference (error compensation) from multiple production channels.

The material composition uncertainty is of particular concern in regions with significant thermal fluxes. Some thermal capture cross-sections to activation significant nuclides have large values (^{59}Co is a well known example) and so relatively small quantities (perhaps at the trace element level) can give important contributions to the inventory. Further potential material related errors can come from the restricted number of nuclei allowed by the codes and the Monte-Carlo. For the transport calculation, simplification to the main components is usually more than adequate but for the nuclide inventory the effect of all component isotopes needs to be considered. The rather weak influence of the target nucleus on the displacement and spread of the products gives an obvious way round the problem for spallation products.

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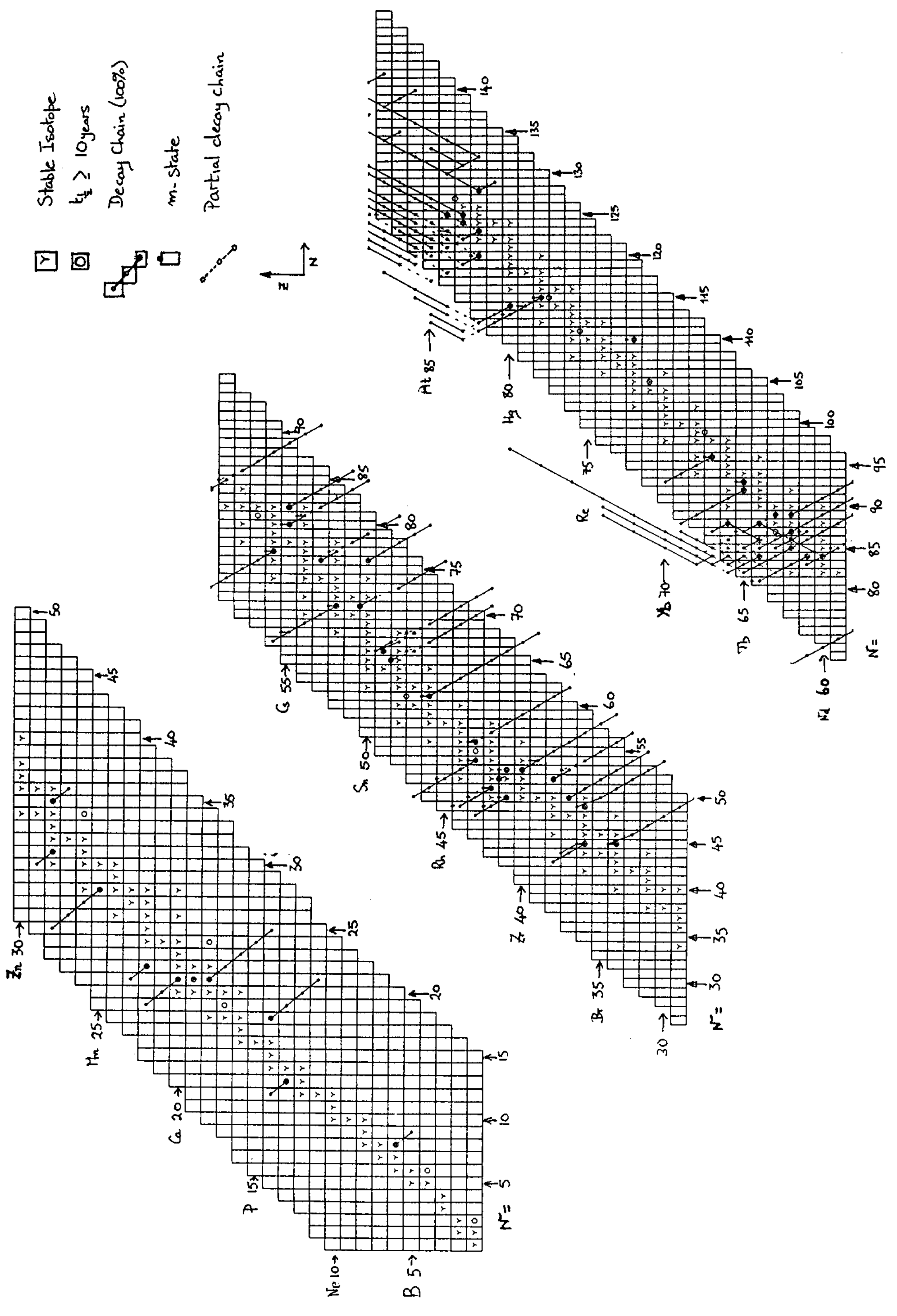

Figure 3: The distribution of nuclides with half life greater than 10 years and their (known) precursors.

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