INVENTORIES FOR ACTIVE-WASTE FROM ACCELERATOR FACILITIES

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Over the last few years, PSI has been developing a practical system for providing a nuclide inventory for waste removed from the accelerator complex. The end result of this work is a computer code system, PWWMBS, that combines a calculation system for nuclide inventories, a database of (accelerator facility specific) operational information and a bookkeeping system which makes it rather simple to specify the required parameters for a large fraction of the waste (Fig. 1).

INTRODUCTION

Several factors make it quite a difficult task to obtain an accurate nuclide inventory for waste from accelerator facilities; the diversity of particle fields producing the activation, the large variety and volume of materials involved, the tendency to use (and re-use) “standard” components bought from industry and that it is continually developing. A practical system for obtaining nuclide inventories has to accept or find some way round the realities; balancing accuracy with degree of elaboration.

A detailed analysis of the PSI accelerator complex as a radioactive waste producer and of elemental activation in terms of its significance to repository safety has lead to the development of a calculation scheme which, subject to confirmation by the validation work, should produce a complete inventory for all the waste (operational plus de-commissioning) with an accuracy of better than a factor of ten. This calculation scheme has been built into a computer code system, PWWMBS.

RADIATION SAFETY

A major consideration is radiation dose to the personnel involved in waste conditioning, particularly where this has a bearing on accuracy in determining the nuclide inventory. This problem has been studied by a working party involving the HSK (the Swiss federal nuclear safety inspectorate), NAGRA (the organisation with the responsibility to develop a waste repository for Switzerland) and PSI. The outcome was summarised in four principles given in the following order of priority:

1. The minimisation of the dose accumulated by personnel handling the waste
2. To provide a radioactive nuclide inventory sufficient for the needs of the safety of the repository; this emphasises completeness rather than having high accuracy.
3. Ensure rapid throughput of waste; from removal from the accelerator complex to packaging for repository storage.
4. Efficient utilisation of the radioactive waste repository.

An almost inescapable consequence of these principles is that reliance has to be put on calculated inventories; the alternative of taking and analysing samples for all the waste requires too much handling. However, a well designed experimental programme of sample analysis has to be made so as to validate the calculations.

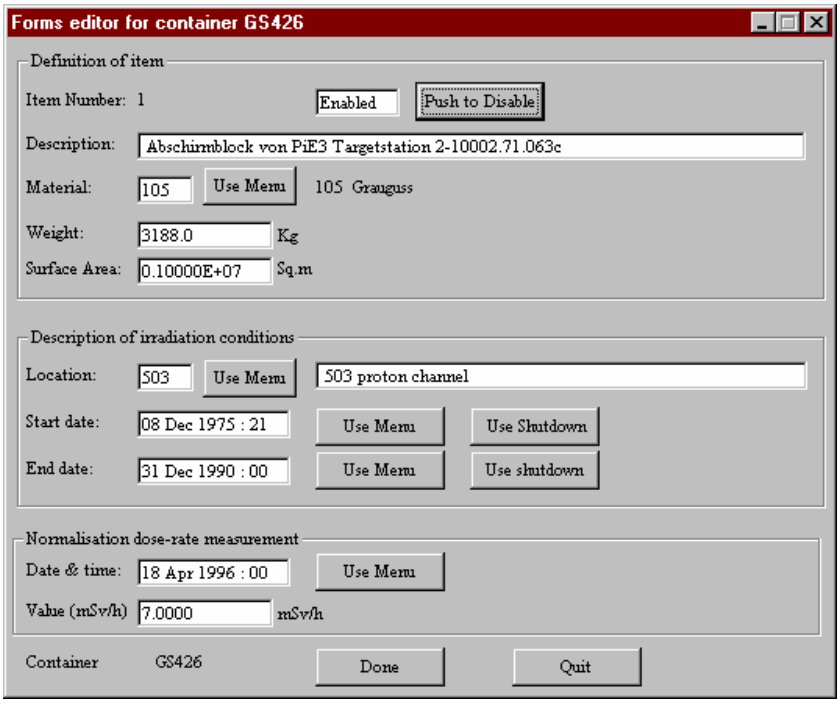

Fig. 1:

The input screen for specifying an item of waste for the Windows™ version of the PWWMBS system. The required information is

1. A descriptive string
2. The material (chosen from a pre-defined list)
3. The weight
4. The surface area of the face nearest the irradiation source.
5. Where the irradiation took place (selected from a pre-defined list).
6. The date of the start & end of the irradiation
7. The date & time of the measurement of the dose-rate for normalisation and its value.

ACTIVATION IN AN ACCELERATOR COMPLEX

All the accelerated particles will be lost somewhere. For a high-beam-power accelerator facility these are limited to a number of designed loss points (target stations, collimators, beam-splitters, beam dumps, etc.), and in order to have a maintainable system, very low loss rates at other points (e.g. the loss rate in the PSI proton channel is generally of the order of one or two nA/m).

The "lost" protons interact with nuclei thereby initiating particle cascades that are propagated through and characterised by the surrounding material. Medium energy particle reactions involve only a small energy transfer to the struck nucleus that is also roughly independent of the incident particle's energy. This means that a large part of the incident kinetic energy leaves the interaction as secondary medium energy particles.

The interaction probability for the primary protons is typically of the order of 80 % (the numerical values in this section are for 590 MeV protons onto a large block of Copper) and the approximate number of interactions caused by these secondary particles (per primary proton) is 2.1 by neutrons, 0.19 by secondary protons, 0.006 by positive pions and 0.024 by negative pions; secondary interactions are dominated by those induced by neutrons.

Together with medium energy secondary particles there are also evaporation products (protons, neutrons, alpha-particles, tritons, etc.). On average 6.5 fast neutrons are produced; these will be moderated and eventually captured but during the process will make about 2 inelastic interactions and produce 0.36 further fast neutrons. The relatively high rate of triton ejection (0.1 per proton) makes tritium a significant concern for accelerator waste.

On average there will be about 3.3 medium energy interactions, 2 fast neutron interactions and 9 neutrons absorbed for each proton lost. These ($\approx$ 15) interactions lead to about 9 unstable products, corresponding to an equilibrium activity of 55 PBq for a 1 mA proton beam loss. A significant part of this inventory is short lived and the value falls to $\approx$ 2 PBq after 3 years decay and $\approx$ 1 PBq after 30 years. There will be some variation with material; for Iron a value of 6 PBq and for concrete 2 PBq (both for 30 years activation and 3 years decay) are obtained. For the same irradiation sequence, the spallation target for SINQ has about 0.1 PBq. This indicates that somewhat over 95 % of the activation will be produced outside the volume of the actual proton beam loss, i.e. in shielding.

REPOSITORY REQUIREMENTS

The main safety assessment for a radioactive waste repository will be based on the nuclide inventory of its contents at closure. As these are collected over a long time period, each waste package (or group of packages) has to satisfy the criterion

$$\frac{1}{W}\sum_{i=1}^{N}\frac{A_i}{ACL_i} \le 1.0 \qquad (1)$$

where there are N nuclides with activity A_i and the ACL_i are the corresponding activation concentration limits. W is the total mass of the waste in the package. The ACL values[1] provide an absolute reference scale for assessing complete nuclide inventories and also are crucial to the problem of material contamination. Values for some of the critical nuclides for accelerator waste are shown in Table 1.

	$t_{1/2}$ (years)	ACL Bq/g	ACL Wtppm
^{209}**Po**	102	120.	0.00019
^{194}**Hg**	440.	450.	0.0029
^{208}**Po**	2.898	12'000.	0.0055
^{42}**Ar**	32.9	$6.0\ 10^5$	0.063
^{90}**Sr**	28.84	$3.4\ 10^5$	0.067
^{207}**Bi**	31.55	$1.4\ 10^5$	0.069
^{108m}**Ag**	418.	$2.1\ 10^4$	0.072
^{3}**H**	12.33	$5.8\ 10^7$	0.16
^{22}**Na**	2.602	$4.5\ 10^7$	0.19
^{210}**Pb**	22.3	$5.9\ 10^5$	0.21
^{231}**Pa**	32'760.	970.	0.55
^{204}**Tl**	3.78	$2.5\ 10^7$	1.5
^{93}**Mo**	4000.	$1.1\ 10^5$	3.09
^{133}**Ba**	10.5	$5.6\ 10^7$	5.92
^{14}**C**	5730.	$1.7\ 10^7$	10.3
^{239}**Pu**	24'110.	24'000.	10.5
^{233}**U**	159'200.	9'400.	26.4
^{116m}**Ho**	1'200.	$2.7\ 10^6$	40.7
^{36}**Cl**	301'000.	$9.6\ 10^5$	786.

Table 1: ACL values for nuclides significant for repositories. Values are given in Bq/g and also Wtppm (= $7.6\ 10^{-11} \times$ ACL $\times\ t_{1/2} \times$ A) which gives a better feeling for the potential seriousness as (no burn up) the Wtppm of a product with a cross-section σ (barns) produced by a flux ϕ in a year is $3.1\ 0^{-11} \times \sigma \times \phi$

NUCLIDE INVENTORY CALCULATIONS

Techniques for calculating radiation effects for spallation sources, including activation, are readily available and can handle systems with a volume of up to a few cubic metres. Such techniques, with some refinements, can be used to estimate activation in high beam-loss regions of the accelerator complex; the main improvements are to the numerical integration scheme and to allow a good representation of the material composition. The calculation normally delivers absolute nuclide production rates within a selected volume.

[1]Declaration and activation concentration limits are issued by NAGRA [1] and the latter set maximum acceptable specific-activation values for all safety relevant nuclides for a specific repository design.

Such methods, being based round Monte-Carlo, are effectively impractical for very large systems; e.g. biological shielding. However, normal shielding consists of large blocks of mainly Iron, concrete and heavy concrete and these blocks will generate a neutron spectrum with shape characterised by the material (the shielding equilibrium spectrum). This shape is entirely predictable by using $S_{n,}$, discrete ordinates or other standard calculation techniques. Nuclide production rates may be calculated for some arbitrary total intensity by folding the spectrum with suitable cross-sections. The nuclide build-up/decay calculation will also give an estimate for the expected gamma dose rate. This may be compared with a measured value to obtain the actual intensity and hence an absolute nuclide inventory.

THE PSIMECX LIBRARY

The nuclide production cross-sections for folding with the neutron spectra come from the PSIMECX library [2]. This contains calculated values for neutrons in the energy range 1 to 800 MeV. The current version has values for all the elements from B to Bi excepting the noble gasses, Tc and Pm and also for ^{232}Th and ^{238}U. The library is in two main parts, the 75 stable isotopes of the 21 elements C, O, Na, Mg, Al, Si, P, S, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Mo, Sb, Pb plus ^{232}Th and ^{238}U which are the principal constituents of normal materials of construction; the rest of the elements are treated by using appropriate scaling laws from these isotopes.

The calculations have been made using the ALICE [3] and MECC [4] codes.

ACTIVATION OF ELEMENTS

Study of the activation of the elements [5] in terms of significance to repository safety is a necessary preliminary to considering materials. This has been done by examining the nuclide inventories for all elements in the PSIMECX library using 18 spectra that typify the range expected in the PSI complex under the largest realistic irradiation expected (30 years at 10^{11} /cm^2/sec) in terms of activation concentrations and declaration limits after 10 years decay.

There are two concerns: (i) which elements can be activated to give an ACL index above one (15 have been found) and (ii) which elements give noticeable contributions when present as traces (18 elements require < 100 Wtppm)? Due to class overlap there are only 19 elements in all; these are shown in Table 2.

	Natural abund. (Wtppm)	ACL index	Wtppm for a declarable nuclide
$_{92}$U	2.7	563	0.0062
$_{90}$Th	9.6	1350	0.064
$_{67}$Ho	1.3	6.3	0.3
$_{63}$Eu	2.0	36.	0.0073
$_{47}$Ag	0.075	2500.	0.095
$_{27}$Co	25.	0.037	0.014
$_{3}$Li	20.	3470.	0.015
$_{83}$Bi	0.0085	191	1.4
$_{82}$Pb	14.	181	2.5
$_{81}$Tl	0.85	241	1.9
$_{80}$Hg	0.085	351.	1.3
$_{55}$Cs	3.0	0.038	1.4
$_{41}$Nb	20.	0.090	9.1
$_{17}$Cl	145.	2.5	4.2
$_{7}$N	19.	10.	1.2
$_{79}$Au	0.004	1.7	120.
$_{66}$Dy	5.2	1.4	40.
$_{58}$Ce	66.5	0.12	96.
$_{42}$Mo	1.2	11.	34.

Table 2: Results from analysing the activation of all relevant elements irradiated in maximum intensity spectra typical of the PSI complex. All elements that had an ACL index above 1 and all which produce a declarable nuclide for an amount less than 100 Wtppm are included.

MATERIALS AND MATERIAL COMPOSITION

Materials play a dual role: they become the active waste but those in large volumes also shape the irradiation spectra. The majority of the material in the complex is shielding in large blocks.

The most difficult problem for characterising radioactive waste is how to take material impurities into account. Materials of construction mainly come from industry and have compositions based on industrial norms. The composition may be considered in three categories: (i) The main components (e.g. Fe, Ni, Cr, etc. for steel) which have amounts that are probably fairly well known and don't vary too much between batches, (ii) trace elements (e.g. Mo, W & Co in steel, Ag in Copper) which are known to be present although their amount may vary widely within any specific material class and (iii) impurities.

Impurities are effectively unknown amounts of any of the elements not appearing under the first two categories. Industrial materials mainly are involved, so there is scope for comparatively large amounts of contamination. Obtaining some sort of quantification is necessary otherwise any claim to completeness for the calculated inventory is compromised; this has been obtained by comparing the activation of elements with that of typical materials. The majority of the PSI waste will be from 10 material classes which are given a

representative composition (e.g. one to represent all stainless-steels, etc.). Their activation by the same 18 spectra is summarised in Table 3. The average activation index is of the order of 0.02 (ignoring Lead and plastic).

The Wtppm of each element that has an ACL index of 0.002 has been calculated [5] (this would increase the maximum ACL index by 10 %). Selecting only elements that require a concentration of less than 0.01 wt%, then only the following 9 elements with the stated Wtppm remain:

U	Th	Bi	Pb	Tl	Hg	Eu	Ag	Li
4	1	10	11	8	6	56	1	0.6

As a likely agency for the introduction of impurities is some natural product (rock, soil, slag, earth, etc.), then comparison with the natural abundance of the elements (Table 2) gives a meaningful scale of reference, particularly when the critical concentration are significantly below the natural abundance (e.g. Th and Li). These 9 elements, and particularly Li & Th, are the most important for having a realistic upper limit to their amount in the material compositions. However, as between 90 and 99 % of the material composition is known, it can be expected that typically only 1 to 10 % of the composition can possibly contain such contamination giving a reduction of a factor of 10 to 100.

THE PWWMBS SYSTEM

The PWWMBS code is a nuclide inventory calculating system based round packages of waste. Data for each waste package is held in an individual computer file as a set of parameters describing individual items of waste. For the majority of waste items only a very few parameters are required (Fig. 1).

The majority of the parameters needed for the actual calculation (material compositions, neutron spectra, nuclide production cross-sections, decay data, accelerator operation schedules and intensities) are contained in a data base. Various utilities to examine and maintain this data-base are included in the package.

For components irradiated by secondary neutrons, the calculation of production rates is carried out internally together with calculation of a normalisation factor based on the measured dose-rate on a specific date. The code delivers the final nuclide inventory for the container broken down into contributions by the individual materials and limited to those above pre-set specific activities.

In the case of components directly irradiated in the proton beam, the results of a suitable transport calculation are passed to PWWMBS as a file containing information about the irradiation schedule and the production rates for the nuclides.

The code has been written in FORTRAN and is available as a DOS version using a commercial window emulation package or as a version integrated into the Windows™ system. Its code should be reasonably easy to adapt for use at other accelerator laboratories.

REFERENCES

[1] M. Hugi, S. Teichmann, *Zulässige Aktivitätskonzentrationen und Deklarationslimiten für radioaktive Abfälle des PSI-West (Beschleunigerareal)*, NAGRA Aktennotiz 99-150 (1999).

[2] F. Atchison, PSI Reports 98-09, 98-10, 98-11 & 98-12 (1998).

[3] M. Blann, *Workshop on computation and analysis of data relevant to nuclear energy & safety*, Trieste 1992, page 512, World Scientific, Singapore (1993) and Private Communication (1995).

[4] H.W. Bertini, Phys. Rev. 188, 174 (1969).

[5] F. Atchison, *Activation concentration limits and the estimation of the contribution of material impurities to nuclide inventories*, PSI Internal Report AN-96-01-15 (2001).

	ACL index		Nuclides contributing 10 % or more to the ACL index							
	Average	Maximum	^{3}H	^{14}C	^{22}Na	^{36}Cl	^{42}Ar	^{93}Mo	^{108m}Ag	^{194}Hg
Stainless steel	0.088	0.13	✓					✓	✓	
Shielding steel	0.0083	0.048	✓	✓				✓	✓	
Mild-steel	0.0076	0.039	✓	✓			✓	✓	✓	✓
Aluminium	0.023	0.073	✓		✓				✓	✓
Copper	0.019	0.060	✓						✓	
Brass	0.025	0.12	✓						✓	✓
Lead	20.	180.							✓	✓
Concrete	0.025	0.081	✓	✓			✓		✓	✓
Heavy concrete	0.0055	0.032	✓	✓			✓	✓	✓	
Plastic	0.70	3.7				✓				✓

Table 3: Expected upper limit activation of the standard PSI materials averaged over 18 spectra in terms of the activation concentration index (see equation 1). The parameters for the critical nuclides may be seen in Table 1.