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TITLE: ACCELERATOR TRANSMUTATION STUDIES AT LOS ALAMOS WITH  
LAHET, MCNP, AND CINDER '90

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# MASTER

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## Accelerator Transmutation Studies at Los Alamos with LAHET, MCNP, and CINDER'90

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### Abstract

Versions of the CINDER code have been used over three decades for determination of reactor fuel inventories and aggregate neutron absorption and radioactive decay properties. The CINDER'90 code, an evolving version which requires no predetermined nuclide chain structure, is suitable for a wider range of transmutation problems including those treated with older versions. In recent accelerator transmutation studies, the CINDER'90 code has been linked with the LAHET Code System (LCS) and, for high-energy calculations, with SUPERHET. A description of the nature of these linked calculational tools is given; data requirements for the transmutation studies are described; and, examples of linked calculations are described for some interesting accelerator applications.

### Introduction

The description of the inventory of radionuclides in a radiation environment has been an interesting area of study during the 50-year history of the nuclear reactor industry. Nuclides are produced there as products of various neutron-induced reactions, including fission, and as daughters of the decay of radionuclides; similarly, nuclides are destroyed by the same mechanisms. The system of coupled differential equations describing reactor inventories requires nuclear data for energy-dependent reaction cross sections, fission yields, and radioactive decay constants and branchings. Cross-section data are required for neutrons below 10 MeV, and experience has shown that data were required only for stable and long-lived nuclides. Significant reactions are limited to fission,  $(n,\gamma)$  and — for a few actinides —  $(n,2n)$ . These data and a solution algorithm were first incorporated in the CINDER code,<sup>1</sup> which uses Markovian chains in the solution of the system of equations. The quality of available nuclear data for reactor inventory calculations has significantly improved because of a national nuclear data development effort for the Evaluated Nuclear Data File (ENDF/B),<sup>2</sup> which is currently limited to data describing neutron-induced reactions below 20 MeV.

The application of this methodology to accelerator transmutation requires knowledge of the energy-dependent fluxes of all particles and the energy-dependent cross sections for all particles on all longer-lived nuclides. Particle fluxes are available as a product of the transport calculation performed with LCS,<sup>3</sup> which combines LAHET (the Los Alamos enhanced version of the HETC code<sup>4</sup> developed at the Oak Ridge National Laboratory) with MCNP<sup>5</sup> — which transports neutrons below 20 MeV. However, reaction data to be used with such particle fluxes are sparse for neutrons above 20 MeV and for other particles at nearly all energies.\* LAHET models ion and pion reactions at all energies and neutron reactions above 20 MeV using on-line nuclear reaction models.

A hybrid method of solution has evolved using the results of medium-energy (20 MeV – a few GeV) reactions, modeled during the LAHET transport calculation, with the neutron reactions below 20 MeV calculated in CINDER'90 using evaluated neutron reaction data and neutron fluxes from MCNP.

## Calculation Tools

### LAHET & SUPERHET

Medium-energy Monte-Carlo (M-C) particle transport calculations for incident neutrons and protons were performed in the '60s and '70s using such codes as HETC<sup>4</sup> from the Oak Ridge National Laboratory (ORNL). These continuous-energy M-C codes used reaction cross sections from the simple intranuclear cascade models of Bertini<sup>6</sup> and Chen et al.<sup>7</sup> with residual excitation energy expended with the evaporation of nucleons from the excited system.<sup>8</sup> A high-energy fission model<sup>9</sup> was added as a reaction path competing with intranuclear cascade.

In HETC, ions at all energies above a cutoff and neutrons above the limits of evaluated nuclear data were calculated with these models; neutrons born or scattered to energies within the limits of evaluated neutron data were banked as source particles for subsequent transport calculations with other codes using processed evaluated *multigroup* cross-section data. An interesting history and introduction to HETC and associated codes is given by Dietz.<sup>10</sup>

The LAHET code was initially a Los Alamos National Laboratory (LANL) adaptation of HETC, with the LANL advanced *continuous-energy* M-C code MCNP<sup>5</sup> replacing the older ORNL multigroup M-C code MORSE.<sup>11</sup> An additional intranuclear cascade model has been adapted from the ISABEL code,<sup>12,13</sup> which is derived from VEGAS<sup>7</sup> and permits hydrogen and helium ions and antiprotons as projectiles. An alternate high-energy fission model developed by Atchison<sup>14</sup> has also been incorporated into LAHET.

One shortcoming of intranuclear cascade and evaporation models has been the discrepancy between modeled and measured data at energies below about 100 MeV.<sup>15,16</sup> Improved agreement has been obtained by adding the Fermi breakup model,<sup>17</sup> replacing evaporation to describe the breakup of light nuclei; also, an optional multistage preequilibrium model,<sup>18</sup> based on the GNASH code,<sup>19</sup> is now available as an intermediate stage between intranuclear cascade and evaporation. A library of neutron elastic scattering cross-section data describing neutrons in the range 20 – 100 MeV has been added; such scattering has been shown to be important for light target nuclides<sup>20</sup> but negligible for higher-Z targets.<sup>15</sup>

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\* A relevant review has just been provided: A. J. Koning, "Review of High Energy Data and Model Codes for Accelerator-Based Transmutation," Netherlands Energy Research Foundation (ECN) report ECN-C-93-005 (January 1993).

Few medium-energy experiments have been reported for use in comparisons with calculations.<sup>21</sup> Although the use of evaluated nuclear data in calculations has naturally shown better agreement with measured results,<sup>16,22</sup> calculations with data from the enhanced models alone show reasonable agreement.<sup>23-25</sup> The breadth of such medium-energy calculations, the sparsity of measured basic nuclear data, and the near absence of support for data evaluation efforts combine to lend importance to the development and use of such models.

The upper-energy limit of LCS is that of the intranuclear cascade model — or about 3.5 GeV, due to the model's limitation to single or double pion production in nonelastic reactions; multiple pion production is physically permitted at higher energies. The code's application has been extended to much higher energies using the reaction physics coding of FLUKA,<sup>26-28</sup> which is limited to validity above 5 GeV. In this extended code version called SUPERHET,<sup>29,30</sup> the LAHET or FLUKA reaction physics are alternately selected by a probability distribution varying linearly over the range 2 to 5 GeV; at and outside these limits, only the applicable physics resource is used.

In either configuration of LCS — LAHET or SUPERHET — the products of each reaction tallied are retained on a history file for later examination. Neutrons emerging from reactions with energies of 20 MeV and less are recorded for calculations with HMCNP — a version of MCNP modified to accept these neutrons as source particles for the lower-energy M-C transport. The  $\gamma$ s produced in the LAHET calculation can be followed in the HMCNP calculation, along with the  $\gamma$ s produced in the lower-energy neutron transport. With SUPERHET, the  $\gamma$ s produced in both high-energy and lower-energy M-C calculations are typically banked for subsequent transport with EGS4.<sup>31</sup>

## MCNP

The MCNP code, incorporating the results of over 250 man-years of M-C development at Los Alamos, represents the state of the art in neutral particle M-C radiation transport. The code is rich in detailed neutron and photon physics, transport and reaction data, advanced variance reduction capabilities, and advanced 3-D geometry setup and plotting. The code uses continuous-energy cross sections for neutrons (commonly to 20 MeV) and photons (to 100 MeV); a version using multigroup data is available. Thermal neutron scattering is described by both free-gas and  $S(\alpha, \beta)$  models. The code includes incoherent and coherent photon scattering, fluorescent emission following photoelectric absorption, and absorption in pair production with local emission of annihilation radiation.

Within LCS, a version of the code, named HMCNP, obtains source particles from a source file produced in the medium-energy transport calculation. MCNP finds application in accelerator transmutation studies in transporting the neutrons and photons initiated in higher-energy transport as well as transporting the photons emitted in the radioactive decay of activation products.

## CINDER'90

Calculations of radionuclide inventories in high-current, medium- and high-energy accelerator targets have required the development of a new inventory code (CINDER'90)<sup>1</sup>, evolved from earlier versions of CINDER<sup>1,32-36</sup> and REAC,<sup>37-40</sup> and continuing development of cross-section and decay data. CINDER'90 uses these data with problem-specific spallation product production and neutron flux data calculated with LCS.

The temporal concentrations of nuclides depleted and produced in materials subject to irradiation are described by a large set of coupled differential equations, each nuclide's concentration being determined by a history of gains from neutron absorption reactions (spallation, fission,  $[n, \gamma]$ ,  $[n, 2n]$ ,

etc.) and radioactive decay of parent nuclides, and losses from its own decay and particle absorption. The solution for these nuclide concentrations was simplified in 1962 with the CINDER code, which resolved the complicated nuclide couplings into *linear chains*, each chain representing a unique path from nuclide to nuclide, resulting in small independent sets of differential equations describing the rate of change of *partial concentrations* of nuclides in each chain. The solution of a large set of coupled differential equations is thus reduced to the solution of a number of small sets of differential equations, each characterized by a single generalized form. Because of the linear nature of the chain (a result of the Markov process), the generalized equations may be solved sequentially for the partial concentration of each *linear nuclide* in the chain. Nuclide concentrations are then obtained by summing partial concentrations.

Each chain begins with a nuclide having an initial atom density  $N_1^0$  (atoms/barn-cm) and/or a constant spallation yield production rate  $\bar{Y}_1$  (atoms/s-barn-cm). The partial concentration  $N_n(t)$  for the  $n^{\text{th}}$  linear nuclide in a chain at the end of the time interval  $t$  is given by:

$$N_n(t) = \prod_{k=1}^{n-1} \gamma_k \left\{ \bar{Y}_1 \left[ \frac{1}{\prod_{l=1}^n \beta_l} - \sum_{j=1}^n \frac{e^{-\beta_j t}}{\beta_j \prod_{i=1, i \neq j}^n (\beta_i - \beta_j)} \right] + N_1^0 \left( \sum_{j=1}^n \frac{e^{-\beta_j t}}{\prod_{i=1, i \neq j}^n (\beta_i - \beta_j)} \right) \right\}, \quad (1)$$

where  $\beta_i$  is the total temporal transmutation probability of nuclide  $i$

$$\beta_i = \lambda_i + \phi \sigma_a^i \quad (2)$$

and  $\gamma_i$  is the temporal probability of the  $i^{\text{th}}$  nuclide transmuting (decaying or absorbing) to the  $i+1^{\text{st}}$  nuclide ( $\gamma_i \leq \beta_i$ ). Here  $\phi$  is the energy-integrated neutron flux and  $\sigma_a^i$  is the flux-weighted-average cross section for neutron absorption by nuclide  $i$ . This equation can also be written as

$$N_n(t) = a_n + \sum_{j=1}^n \left( N_1^0 - \frac{\bar{Y}_1}{\beta_j} \right) b_{n,j} e^{-\beta_j t}, \quad (3)$$

where

$$a_1 = \frac{\bar{Y}_1}{\beta_1}, \quad (4)$$

$$a_j = a_{j-1} \gamma_{j-1} / \beta_j, \quad j > 1 \quad (5)$$

$$b_{1,1} = 1, \quad (6)$$

$$b_{n,j} = b_{n-1,j} \gamma_{n-1} / (\beta_n - \beta_j), \quad j < n; n > 1 \quad (7)$$

$$b_{n,n} = - \sum_{j=1}^{n-1} b_{n,j}. \quad (8)$$

The CINDER'90 code differs from earlier CINDER versions in that earlier versions required the development of a library of transmutation chains prior to a calculation. Chains of such libraries

were selected to follow transmutation paths that the user considered necessary and sufficient for the problem. Chains developed for one problem were not necessarily applicable to others. The CINDER'90 code uses a library of basic nuclear data to trace all possible transmutation paths, determining the partial concentration and associated activity of each linear nuclide as well as the integrated transmutation of each linear nuclide during a time increment. A linear nuclide's integrated transmutation, called the *passby*, indicates the sum of subsequent partial concentrations in chains continuing from the nuclide. Each linear nuclide's partial concentration, activity and passby are examined to determine whether a chain should be terminated relative to input significance criteria.

Nuclide concentrations and activities are accumulated from linear nuclide properties as they are calculated. These data are combined in postprocessing with decay and neutron absorption data to obtain density (atoms/barn-cm and kg), activity (curies/cm<sup>3</sup> and curies), decay power (watts/cm<sup>3</sup> and watts), macroscopic neutron absorption (cm<sup>-1</sup>), and decay spectra properties listed by nuclide, element Z, and mass A. Major contributors ( $\geq .1\%$ ) to mass, activity, decay power and macroscopic absorption are also tabulated.

Many applications of these calculations are addressed with the direct utilization of the individual-nuclide and aggregate results — activity inventory, decay power, macroscopic neutron absorption, etc. Some applications require the transport of the decay source  $\gamma$ s to obtain a desired response, such as dose or dose-equivalent rates.

#### **MICROSHIELD/ISOSHLD**

A convenient alternative to the use of conventional radiation transport calculations of the decay source  $\gamma$ s is the use of a shielding code such as MICROSHIELD,<sup>41</sup> a microcomputer adaptation of the venerable ISOSHLD code.<sup>42</sup> This is a point kernel shielding code using exponential attenuation coefficients and build-up factors to calculate the dose-equivalent rate at a point in space near a shielded radioactive source described by regular geometries (i.e., spheres, cylinders, etc.). The source may be selected from several hundred radionuclides for which the code has decay spectral data; alternatively, the source can be described with multigroup values not exceeding 10 MeV. Although the spectral data, energy limit, and geometry selection restrict the application of MICROSHIELD, it is a powerful tool for limited applications of calculated inventory and decay spectra.

### **Nuclear Data Development for CINDER'90**

Transmutation calculation results are, of course, limited in accuracy by appropriate problem definition and by the validity of the nuclear data used in the calculation. These data include the neutron absorption cross sections and decay constants for each nuclide transmutation path, as well as associated branching fractions to ground and isomeric states produced. Additional data describing the energy spectra and toxicity associated with the decay of radionuclides are required. The collection, calculation and evaluation of these data are ongoing efforts currently involving dozens of scientists internationally.<sup>43,44</sup>

#### **ABSORPTION: Transmutation cross sections and branchings**

Earlier transmutation calculations with CINDER'90 used the cross-section data of the REAC codes developed by Mann.<sup>37-40</sup> The early REAC codes and libraries, though containing recognized deficiencies, provided a significant advance in transmutation computation recognized internationally and have continued their own independent evolution.



Deficiencies in earlier REAC-2 cross-section data have been largely overcome for major reactions with data accumulated by Mann and Lessor<sup>45</sup> (Westinghouse Hanford) and by Kopecky<sup>46-51</sup> (ECN, Netherlands) from available evaluations and nuclear model calculations.

Cross-section data for CINDER'90 have thus been taken from a number of sources as listed below:

- Westinghouse Hanford collection of Mann and Lessor,<sup>45</sup> consisting of
  - ENDF/B-VI evaluations,<sup>2</sup>
  - Netherlands Energy Research Foundation (ECN) collection of Kopecky et al. as the European Activation File version 2 (EAF-2),<sup>46</sup>
- Netherlands Energy Research Foundation (ECN) collection of Kopecky et al. in the European Activation File version 3 (EAF-3),<sup>47-51</sup> consisting of data from
  - Evaluated Nuclear Data File, versions IV, V and VI (ENDF/B-IV, -V and -VI),<sup>2</sup>
  - Japanese Evaluated Nuclear Data Library, versions 2 and 3 (JENDL-2, JENDL-3),
  - Joint European Data File, version 1 (JEF-1),
  - Lawrence Livermore National Laboratory (LLNL) evaluations in the Activation Library (ACTL),
  - radiative capture model code MASGAM results,
  - model calculations with the THRESH code,<sup>52</sup> often with ECN low-energy extensions if  $Q \geq 0$  (THRESH-ECN),
  - FISPRO code results,
  - Russian activation file BOSPOR,
  - French data file BRC,
  - European Fusion File EFF-2,
  - IAEA dosimetry file IRDF-90,
  - other data collections identified as IBJ and TAT,
  - estimated data
- results of GNASH code calculations for many common accelerator materials,<sup>53</sup>
- PGY evaluations by P. G. Young and M. B. Chadwick for the most significant reactions of neutrons on Hf, Ta, W and Pb,<sup>54,55</sup>
- values collapsed from the ENDF/B-V based TOAFEW-V library<sup>56</sup> of fission-product and actinide cross sections for use until such ENDF/B-VI data are processed.

Branchings to isomeric and ground-state products for  $E_n = 14.5$  MeV have been treated with systematics by Kopecky. These have been used with partial thermal cross-section and resonance-integral data from the Chart of the Nuclides<sup>57</sup> to produce energy-dependent branching fractions to the ground and isomeric states of some (n, $\gamma$ ) reactions. In the absence of such additional data, the 14.5 MeV branchings have been used at all neutron energies.

A new 63-group neutron energy structure was defined by LaBauve, who processed continuous energy cross-section data into this group structure.

A library of multigroup cross sections has been formed from these data using the following priority of data sources:

- PGY,
- ENDF/B-VI,
- ENDF/B-V, -IV,
- JEF-1,
- JENDL-3, -2,
- ACTL,
- MASGAM,
- GNASH,
- THRESH-ECN,
- IRDF-90,
- TAT,
- THRESH,
- other.

To date, applications and validation efforts have been limited to nuclides of  $Z \leq 84$  because accelerator materials studied have been within this range and because treatments of spontaneous fission and neutron-induced fission have not yet been completed in CINDER'90. The validation of these cross-section data began with emphasis placed on reaction data to which calculated results were most sensitive; such data examinations lead initially to many data corrections using available sources — especially the GNASH studies of Chadwick and Young.<sup>54,55</sup> A systematic comparison of library cross sections with three main data sources was initiated. First, the thermal cross-section and resonance-integral data present on the Chart of the Nuclides<sup>57</sup> for  $(n,\gamma)$ ,  $(n,p)$  and  $(n,\alpha)$  reactions were tabulated and compared with values constructed from the library data. Of 469 reactions with data on the Chart of the Nuclides, some 73 disagreed by more than a factor of two; 53 disagreed by more than a factor of three. Other data sources were found to justify adjustments to the library for 21 of these. The adjustments by which the data from the given data source were scaled uniformly are listed in Table 1. We have also obtained a file of the thermal cross-section and resonance-integral data of Mughabghab et al.<sup>58,59</sup> for use in similar comparisons. On a larger scale, we are currently transferring to Los Alamos the voluminous energy-dependent experimentally-measured neutron cross-section data of the CSISRS on-line data base<sup>60</sup> maintained by the National Nuclear Data Center (NNDC) at the Brookhaven National Laboratory (BNL) for use in more extensive comparisons. Following these validation efforts, our goal is to produce a file of adjusted continuous-energy transmutation cross sections that can be Doppler broadened to any temperature and processed into any user group structure.

Although the experimental data base described above is voluminous, it only covers a fraction of the total data needs. Thus we will be forced to rely, for the foreseeable future, on systematics and theory

to supply most of the needed data. Until now, most of these supplemental data have been produced using the simple systematics code THRESH.<sup>52</sup> Many of these THRESH data may be replaced in the future with data calculated with a planned, automated, user friendly version of GNASH using global optical model potentials.

Table 1  
Cross-Section Adjustments Resulting from  
Comparison with the Chart of the Nuclides

Target	Reaction	Adjustment	Data Source
Ca 43	(n, $\gamma$ )	.6	JENDL
Mn 54	(n, $\gamma$ )	.26	MASGAM
Co 58	(n, $\gamma$ )	11.	JEF
Cu 64	(n, $\gamma$ )	.05	MASGAM
Ge 74	(n, $\gamma$ )	4.	ENDF5
Se 75	(n, $\gamma$ )	20.	MASGAM
Se 82	(n, $\gamma$ )	.088	ENDF5
Kr 86	(n, $\gamma$ )	.048	ENDF6
Rb 86	(n, $\gamma$ )	4.	ENDF5
Zr 90	(n, $\gamma$ )	2.5	ENDF6
Zr 93	(n, $\gamma$ )	.4	ENDF5
Nb 95	(n, $\gamma$ )	4.	ENDF5
Sn113	(n, $\gamma$ )	.35	MASGAM
Sn117	(n, $\gamma$ )	.6	ENDF6
Sb124	(n, $\gamma$ )	2.4	ENDF5
Xe128	(n, $\gamma$ )	.5	ENDF5
Xe136	(n, $\gamma$ )	2.	JEF
Ba132	(n, $\gamma$ )	.4	JENDL
Tm171	(n, $\gamma$ )	.3	MASGAM
Tl205	(n, $\gamma$ )	.15	ACTL
Bi209	(n, $\gamma$ )	.001	JEF

#### DECAY DATA: Decay constants, branchings, $\gamma$ spectra, $\bar{E}$

Decay data used in the CINDER'90 solution of nuclide inventory include the decay constants and branching fractions required to define the transmutation chains. In postprocessing, average decay energies are required to calculate decay power; spectral data are obviously required to calculate aggregate spectra. These data have been taken from the following data sources, listed in order of preference:

- Evaluated Nuclear Data File, version VI (ENDF/B-VI),<sup>2,61-65</sup>
- Master Decay Library (MDL) of Mann et al.,<sup>66</sup>
- Joint European Data File, preliminary version 2.2 (JEF-2.2),<sup>67</sup>
- Browne and Firestone,<sup>68</sup>

- Lederer and Shirley,<sup>69</sup>
- Chart of the Nuclides.<sup>57</sup>

The preliminary JEF-2.2 data have not been completely processed and applied to the extent anticipated following official release.

In applications with LAHET and SUPERHET, the distributions of spallation products typically extend beyond the range of nuclides for which decay data have been evaluated; these nuclides are assumed in CINDER'90 to decay by  $\beta^+$  or  $\beta^-$  decay toward stability with an arbitrarily short half-life of .1 s and with decay energy 5 MeV.

Validation of decay energy data has been limited to examination of  $\sim 185$  major ( $\geq .1\%$ ) decay power contributors in accelerator applications. Table 2 lists 19 nuclides for which decay energies taken from the MDL were replaced with data from other sources or with  $\frac{2}{3}Q$ .

Table 2  
Recent Modifications to Decay Energy Data

Nuclide	Energy, MeV					
	MDL value Lib. value	Q value	Browne & Firestone	Lederer & Shirley	JEF-2.2 Lib. value	New Library value
Be 7	5.74E-07	0.862	0.0498		0.337	0.337
Be 8	9.30E-08	0.092	0.092			0.092
C 15	7.503	9.772	6.493		6.478	6.493
F 23	133.2	8.510		5.117	10.123	5.117
Co 55	6.517	3.451	2.002		2.275	2.002
Co 56	8.621	4.566	3.502		3.538	3.502
Co 58m	5.227	0.025	0.023		0.025	0.023
Co 61	5.768	1.323	0.565		0.554	0.565
Pr138	1.975	4.437	1.986		1.975	1.986
Pm140	3.084	6.070	2.194		3.084	2.194
Pm142	2.237	4.890	1.447		2.237	1.447
Lu166	2.492	5.480	1.864		2.483	1.864
Ta174	1.280	4.000	1.309		1.280	1.280
Os178	6.043	2.240				1.493
Os179	11.19	3.610			2.406	2.407
Ir183	3.180	3.190			3.346	2.127
Ir187	63.05	1.500			1.000	1.000
Pt187	5.120	2.900			1.933	1.933
Au187	22.64	3.720			4.877	2.480

Decay  $\gamma$  spectra have been processed from ENDF/B-VI (for 930 nuclides), the MDL (for 1029 nuclides), and from the tabulated data of Browne & Firestone (for 8 nuclides) into a 25-group structure. Each nuclide's spectrum has also been identified with decay Q value and Z-N even-oddness. The  $\gamma$  spectra processed from ENDF/B-VI have been used to produce average spectra for these nuclides in

five decay Q-value bins and the four even-oddness bins. These average spectra are used as surrogate spectra for nuclides not having evaluated spectra as assigned by the nuclide's Z-N even-oddness and decay Q-value calculated from parent-daughter nuclear mass differences.

Additional spectral data for  $\beta^-$ ,  $\beta^+$ ,  $\nu$  and  $\bar{\nu}$  are obtainable from the processing of ENDF/B-VI, MDL and JEF-2.2. The processing and application of these spectra in CINDER'90 are anticipated in FY94.

## FISSION YIELDS

The path from an actinide to a fission product in CINDER'90 is determined by the fission rate of the actinide and the yield of the fission product. Fission-product yields have been evaluated for sixty fissioning systems in ENDF/B-VI.<sup>61,62,70-72</sup> Evaluations exist for spontaneous fission and/or neutron-induced fission in thermal, fast and high-energy ( $\sim 14$  MeV) spectra. These evaluations have recently been completed. However coding has not yet been added to CINDER'90 to include fission paths because older code versions and libraries have been sufficient to satisfy the existing demand for such calculations. Nevertheless, we expect to add such fission-path coding to CINDER'90 and associated data to the libraries in the current fiscal year.

## Radiological Hazard Data

In a recent revision to 10CFR20, values limiting the release of radionuclides into the environment have been tabulated.<sup>73</sup> These values are derived from an Annual Limit on Intake (ALI) resulting in either:

- a committed effective dose equivalent of 5 rems, or
- a dose equivalent of 50 rems to any individual organ or tissue.

Values are listed for a number of nuclides in each of a number of chemical compositions. Similarly, default values are given for three classifications of nuclides depending on half-life and decay mode — again for different chemical compositions. We have constructed an envelope of maximum values for the nuclides explicitly listed and for the default groups such that limiting aggregate air and water dilution values can be accessed for arbitrary combinations of radionuclides regardless of chemical composition.<sup>74</sup> Each value specifies the maximum activity concentration permitted for environmental release. Separate values are specified for release in air and water in units of microcuries/ml (or curies/m<sup>3</sup>). These data have not yet been applied in CINDER'90 postprocessing.

## Accelerator Applications

The development of CINDER'90 and its data libraries has been driven by accelerator investigations at LANL. Because of the variety of problems studied and the results desired, the code and data have evolved to their present levels. The quantity and quality of calculated results have experienced a similar evolution. The studies by which the code and data development have been driven and to which they have been or will be applied are described in the following sections, ordered more or less in increasing energy.

## Benchmark Experiments

Not surprisingly, the number of experiments performed with detailed documentation permitting precise comparison of calculated accelerator activation is small. In preparation for the Japanese OMEGA project for the accelerator transmutation of nuclear waste, an experiment was performed with 500-MeV protons incident on a large cylindrical Pb target.<sup>75</sup> No calculations have been made of this experiment to date, partly because a revision of this experiment is being considered to eliminate axial streaming.

At LANL's Weapons Neutron Research Facility (WNR), experiments have been made with 800-MeV protons axially incident on long, square cross-section targets of W and Pb. Scoping calculations were made prior to these experiments,<sup>76</sup> but analysis of the experimental results have not been completed; calculations with current libraries for true irradiation conditions have not yet been made.

Earlier high-quality experiments were performed at LANL's WNR to study the transmutation of actinides relative to the conversion of fertile nuclides to fissile nuclides. Earlier comparison of these measured data with calculated data have necessarily excluded lower-energy neutron transmutation.<sup>77</sup> These FERFICON experiments remain to be calculated with CINDER'90 pending the integration of neutron-induced fission and fission-product production into the code and its data libraries.

## LANSCE Tungsten Target

During operation cycles 44 – 58 of the Los Alamos Meson Physics Facility (LAMPF), extending from November 1985 through October 1990, beam was fed to the Los Alamos Neutron Scattering Experiment (LANSCE). At end of operation, some  $\frac{1}{4}$  amp-hour of 800-MeV protons had been delivered to the tungsten target at LANSCE. LAHET calculations modeled the transport of the beam throughout the LANSCE target. A detailed irradiation history was constructed from the operation history and used in CINDER'90 to obtain nuclide inventories at shutdown and a variety of cooling times. These results were used to evaluate the storage and disposal requirement for the target. Following the October 10, 1990 shutdown, the target was removed on April 7, 1991. Upon removal,  $\gamma$  dose rates along the outside surface and within the flux trap of the target were measured. Recent calculations with LAHET using great detail in modeling source components and surrounding shielding were used with CINDER'90 to obtain inventories at removal; calculated  $\gamma$  source spectra were used in MCNP to obtain dose rates at locations corresponding to measurements. These agreed within a factor of two at all locations, differing by 30% for the highest dose location in the flux trap.<sup>78</sup>

## ATW

Initial calculations for the accelerator transmutation of nuclear waste (ATW) program considered targets of liquid flowing lead.<sup>79</sup> These calculations revealed the need for better neutron absorption cross sections and absorption branchings to isomer and ground-state products for high-Z targets.<sup>43,54,55</sup> The cascade of particles induced by the  $\sim 1$  GeV proton beam leads to multiplication within the target and an eventual neutron yield exceeding 10 n/p. These neutrons, moderated by a D<sub>2</sub>O blanket, transmute long-lived fission products and actinides to shorter-lived radionuclides requiring less extensive disposal procedures.

An interesting variant of the ATW uses a subcritical multiplying actinide blanket to produce heat power for electrical production while depleting the long-lived radionuclide inventory.<sup>80,81</sup>

Calculations were made for solid, stopping-length cylindrical targets of Ta, W and Pb with thick

D<sub>2</sub>O blankets to scope neutron yield and activation legacy of high-current targets at 800-MeV. A target design for many applications has evolved from many physics and engineering considerations that employs He-cooled W target elements separated by He-filled leakage regions and surrounded by a cylindrical Pb multiplying region and D<sub>2</sub>O blanket. This design has grown from the experience of Russell in the LANSCE target design.<sup>82</sup>

### APT

The considerable neutron yield of targets evolving from Russell target studies has been applied to the production of tritium required for defense programs, using the huge  ${}^3\text{He}(n,p){}^3\text{H}$  and  ${}^6\text{Li}(n,\alpha)$  thermal cross sections in competition of reactor concepts using the same reaction paths. Concepts studying the accelerator production of tritium (APT) at BNL have used the  ${}^6\text{Li}(n,\alpha){}^3\text{H}$  path in their Spallation-Induced Lithium Conversion (SILC) target studies. Studies at LANL have concentrated on the  ${}^3\text{He}$  path. CINDER'90 has been used to calculate the radionuclide production in each program for designs as they evolve. Comparison calculations have been made at BNL with versions of ORIGEN<sup>83</sup> using 1-group cross-section libraries produced for a range of reactor applications, resulting in inventories bracketing CINDER'90 results.<sup>84</sup> Decay power differences revealed decay energy discrepancies for Be nuclides as shown in Table 2. One-group cross sections produced in CINDER'90 calculations of the Lead target region of the SILC concept are now being used to refine comparisons with results of CINDER'90 and ORIGEN code versions.

### SSC Detectors

Components of the Superconducting Super Collider (SSC) under construction near Dallas, Texas will be subjected to radiation environments different from those experienced at any other facility because of the high energies and variety of particles produced by the colliding beams of 20-TeV protons. The activation of accelerator components, air, and detectors are of concern to many. Indeed, the temporal, spatial particle fluxes in a detector being designed for the SSC must include the emissions from transmutation products. Our first calculations followed high-energy particles produced in such collisions as modeled with the ISAJET code.<sup>85</sup> LCS was extended to higher energies by linking to the FLUKA code<sup>26-28</sup> in SUPERHET29, 30 as described above.

These tools were first used to model the activation of the L\* detector to determine activity densities and decay power of its components.<sup>86-88</sup> This detector concept gave way to what is now called GEM — for gammas, electrons and muons.<sup>89</sup> Calculations for this detector have used an evolved event generator DTUJET.<sup>90</sup> We have modeled the forward calorimeter of the GEM detector in 16-region calculations to obtain spatial activity densities. These have been used in MICROSIELD to estimate both the  $\gamma$  dose-equivalent rates near the forward calorimeter ( $\leq 250$  mrem/h at  $10^8$  events/s for  $10^7$  s operation and 1 day cooling) and the  $\gamma$  source depth that must be considered ( $\leq 2$  cm).<sup>91</sup> These calculations were followed by 41-region calculations of selected regions of the entire detector.

These codes and techniques have also been applied to the detector advanced by the Solenoidal Detector Collaboration (SDC). This detector has been modeled in 58-region calculations to obtain spatial temporal activities, decay powers and decay  $\gamma$  spectra, which have been transported with MCNP to obtain  $\gamma$  dose-equivalent rates near quadrupoles. The activity densities of airborne radionuclides were also calculated for use in evaluating the magnitude of SSC radioactive effluents. SDC calculations have been summarized by Palounek et al.<sup>92</sup>

## Conclusions

The development of the code and data have been driven by the demands of the various applications. The quality and validity of cross-section, decay, fission-yield, and hazard data available for use in CINDER'90 calculations have been greatly advanced by the efforts of a number of international experimentalists and evaluators as indicated in the data references. We gratefully acknowledge the cooperation of F. M. Mann of Westinghouse Hanford, C. Dunford and V. McLane of the NNDC, H. Gruppelaar and J. Kopecky of the Netherlands Energy Research Foundation, and M. Konieczny and C. Nordborg of the OECD Nuclear Energy Agency.

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