NUCLEAR DATA EVALUATION FOR ACCELERATOR-BASED TRANSMUTATION OF RADIOACTIVE WASTE

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ABSTRACT

The necessity of high-energy nuclear data libraries for accelerator-based transmutation research is argued. After a brief sketch of the high-energy nuclear reaction chain in an incineration system, the most important data. needs for accelerator-based transmutation are categorized by reaction type and material. The present status of the collection of experimental data for high energies is investigated by scanning data bases for measured cross sections. Some relevant nuclear model codes are mentioned. A survey is given of the evaluation work that has been done so far, and a proposal for three high-energy data libraries is presented.

INTRODUCTION

The transmutation of actinides and long-lived fission products is now being recognized as an interesting option to diminish the radioactive waste problem, because it might lead to a situation in which it would only be necessary to store relatively short-lived waste in geological repositories. In particular, the incineration of waste components by means of accelerator-based transmutation with a hybrid system of an accelerator and a subcritical reactor has received growing attention during recent years [1-6]. The main idea behind the accelerator method is the possibility to generate a sufficiently intense neutron spectrum (resulting from ~ 1.5 GeV proton bombardment of a spallation target), yielding an effective transmutation of unwanted isotopes into non-radioactive and short-lived reaction products. This and other arguments [.5] (mainly concerning safety problems) have led to an increasing popularity of the hybrid system concept.

In this contribution, we will discuss the nuclear data aspects of accelerator-based transmutation research (a more comprehensive exposition of the subject can be found in two Nuclear Energy Agency (OECD) reports [7, 8]). The high-energy protons that are produced by the accelerator initiate a large variety of nuclear reactions in the accelerator shielding, the spallation target and the reactor. Besides the incident proton beam, secondary particles will induce further reactions and this process continues until therm alization has taken place. Clearly, the successful development. of an accelerator-based incineration system requires knowledge of a wide range of nuclear data from thermal energies to about *1500* MeV. Therefore, an inventory has been made of the available experimental data, systematics, theories, nuclear model codes and evaluations that are relevant to accelerator-based transmutation [7]. In this paper, we summarize the most important results. Since neutron-induced reactions below 20 MeV have already been subject, to extensive evaluations (mainly for reactor purposes), we have focussed on the energy region 20-1500 MeV. The primary reason for this study is the proposed extension of the current evaluated nuclear data files to energies above 20 MeV, *so* that they can be used in accelerator-based transmutation calculations.

THE IMPORTANCE OF HIGH-ENERGY DATA LIBRARIES

Nowadays, the analysis of nuclear applications of high-energy particle processes are almost invariably performed with high-energy transport codes, in particular the various versions of HETC [5, 6]. Such codes are based on classical models incorporating an *intra*nuclear cascade followed by evaporation. These high-energy transport codes have an integrated character in the sense that they internally generate the nuclear data which are subsequently supplied to the transport part of the code: the *inter*nuclear cascade. Hence, in the current nuclear data situation, an overall calculation of the transmutation process (from the $\sim$ 1.5 GeV incident proton beam down to the thermalized particles in the shielding) is restricted by the use of evaluated data files and their associated processing and transport codes for neutron-induced reactions below 20 MeV and the (integrated) high-energy transport codes for all other reactions and energies. This situation is unsatisfactory since the high-energy transport codes may produce unreliable results for incident particle energies below about 100 MeV (a more systematic evaluation of this statement is currently under investigation in two NEA-Data Bank benchmark exercises [9- 11]. In other words, it is not sufficient to describe the region from 20 to 100 MeV with a single (intranuclear cascade + evaporation) model. Instead, a variety of nuclear models is required, whereby each model has its own appropriate purpose (e.g. optical model and coupled-channels calculations for elastic scattering and reactions to discrete states, pre-equilibrium and equilibrium models for reactions to the continuum, an adequate fission model for the high-energy region, etc.). If the diversity of nuclear models needed to create a reliable data set becomes large, the best strategy is to create evaluated data files for the relevant materials. When needed, certain sections of the data. file can be adjusted with up-to-date nuclear data. from experiments, systems.tics and model codes. In this way, the closest possible connection between transport calculations and the available nuclear data. is guaranteed. Therefore, as a complement to the high-energy transport codes, it is strongly recommenced to subject the 20-100 MeV region to similar evaluation routines as the 0-20 MeV region.

HIGH-ENERGY REACTIONS

To elucidate the indispensability of nuclear data files that go beyond 20 MeV, it is instructive to give an outline of the bulk properties of the total reaction chain in an incineration system. Above all, it is crucial to distinguish between a thin target (a single nucleon-nucleus interaction) and a thick target (internuclear or transport processes). In fig. 1 both the *intra*nuclear processes (solid arrows) and the inter-nuclear processes (dotted arrows) are depicted.

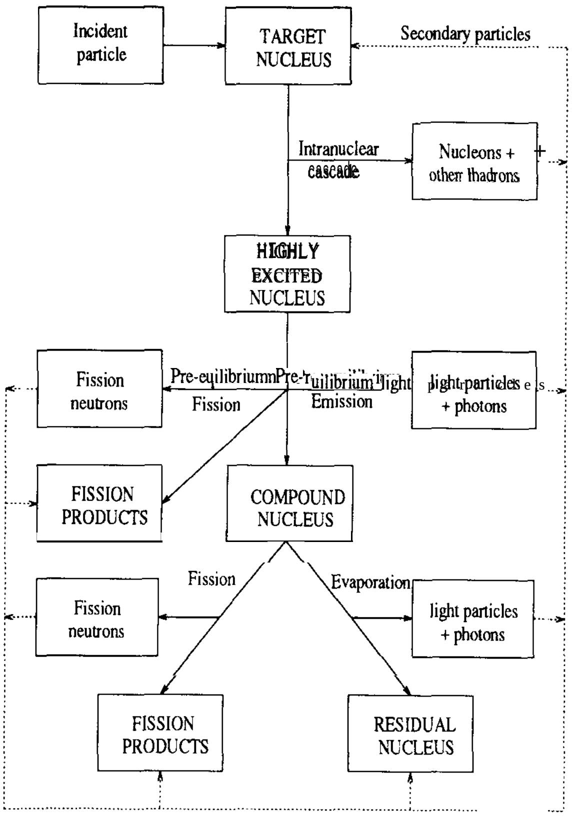

Figure 1: High-energy nuclear reaction. The solid arrows refer to an intranuclear cascade process (thin target; single proton-nucleus reaction), whereas the clotted arrows represent, the internuclear cascade process (thick target).

In the first stage of the reaction, which is an entirely direct process, the incident proton will cause the knock-out of a few high-energy nucleons (this can be envisaged as a classical "billiard-ball" mechanism). Also other hadrons will be formed (such as π- and Ii-mesons). This intranuclear cascade stage has ended when a residual nucleus with an excitation energy of several tens of MeV to about 100 MeV is left. This excitation energy is still so high that a lot of energy may be accumulated on one or a few nucleons, implying the occurrence of direct-like processes. This time however, the emission of these nucleons can no longer be described by classical, direct mechanisms (because the wavelength of the fast nucleon is no longer much shorter than the size of the nucleus). Instead, the reaction process has now reached the stage where the equilibration of the excited nucleus starts to take place but where it is still possible for a fast nucleon to be emitted after a few collisions. This so-called precompound or pre-equilibrium emission can be described by the classical exciton or hybrid models [12], or (preferentially) by quantum-mechanical multi-step reaction models [13]. Also high-energy fission is possible in this stage. Finally, in the third stage, a compound nucleus is left that evaporates low-energy particles (mostly neutrons). This is called the compound or evaporation stage. In this stage also competition with fission occurs.

The high-energy secondary particles (intranuclear cascade emission from the first nucleus) that encounter a new target nucleus (which can also be a (fission) product from a reaction that has already taken place) will proreed through all stages again and will cause new particles (mainly nucleons) to be emitted from this nucleus. The low-energy secondary particles will skip the intranuclear cascade and pre-equilibrium stage and will at once form a compound nucleus with the new target nucleus, from which subsequent evaporation takes place. At the end of the complete (thick target) reaction process, it will turn out that the secondary (tertiary, etc.) high-energy particles have provided the main contribution to the total neutron yield. With these multiple collision mechanisms, the large difference in the average number of outgoing neutrons for an incident 1.5 GeV proton on a thin target (-S-15) and a thick target (~30-40) can be explained. These considerations concerning neutron production hold for the spallation target, the shielding of the accelerator (since they are both subject to the 1.5 GeV proton bombardment) and to a lesser extent for the reactor part surrounding the target.

NUCLEAR DATA NEEDS

For the transmutation of waste with intense neutron fluxes, detailed knowledge of double-differential neutron production cross sections for the heavy target elements is essential. Apart from nuclear data relevant to the transmutation of the radioactive waste and to the neutron production by the heavy target elements, nuclear data are needed for the materials used in the construction and shielding of the accelerator, the target system and the surrounding blanket of the subcritical reactor. Such data include neutron and gamma-ray emission cross sections as well as activation cross sections for materials such as concrete, lead, stainless steel and for constituents of air and coolant. Besides activation also heating, material damage and gas production calculations should be performed. The nuclear data needs for accelerator-based transmutation can be categorized in the following classes:

1. Proton-induced data.

Besides the primary proton beam, also secondary protons with lower energy than the bombarding energy are involved. Needed data include total, elastic, inelastic, spallation cross sections, fission cross sections and production cross sections for outgoing light particles (p, n, d, t, $\tau, \alpha, \gamma, \pi^+$, n-o, π^-) and fission products. In particular, total neutron and proton production cross sections are relevant. Energy-angle distributions are mainly required for outgoing high-energy neutrons anti protons.

2. Neutron-induced data..

The high-energy neutrons which have been knocked out by the initial intranuclear cascade will induce secondary nuclear reactions in the actinides, in the shielding and in the spallation target. For these neutrons, the saline data as needed for proton-induced reactions are required. For low-energy neutrons (emitted in the final evaporation stage), existing evaluations can be used. The additional neutron-induced data that are required are therefore mainly those for energies between 20 and 1500 MeV.

3. Other nuclear data.

Fundamental nuclear structure and reaction data are needed as input for the various high-energy transport codes that simulate the spallation process. These data do not only include nucleon-nucleon and pion-nucleon cross sections , but also level-density parameters, the fission-barrier height, the nucleon-density distribution, the nuclear radius and the mass formula. Existing intranuclear cascade codes may be based on old nuclear information, and could be upgraded when a modern data set of these quantities is available.

4. Reactions induced by other particles.

The outgoing particles other than protons and neutrons that are formed during the spallation reactions will cause further interactions with the target system and shielding. Although their contribution may not be negligible, these processes probably have a lower priority than those of classes 1 and ‘2. We have categorized the relevant materials as follows:

1. Actinides: ^{238}U, ^{237}Np , ^{238}Pu, ^{239}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, ^{245}Cm.
2. Target materials: Ta, W, Pb, Bi.
3. Structural, shielding, coolant and other materials (e.g. stainless steel, concrete, air): H, C, N, O, Na, Mg, Al, Ar, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr.

As mentioned, the establishment of priority criteria. within each group of materials is required, but we shall not address to that issue here. We can add here, however, that the results of the current model code benchmarks [9- 11] which involve lead and tungsten, may give an indication about the most promising target materials. The choice of target material depends not only on the magnitude of the total neutron production, but also on the product yields of unwanted (radioactive) nuclides.

EXPERIMENTAL DATA

The present status of available experimental cross sections at high-energies (20-1500 MeV) has been established by scanning two nuclear reaction data bases, NSR and EXFOR, for relevant measurements. The NSR data base contains bibliographic information and EXFOR contains bibliographic information plus the numerical data. We have retrieved measured cross section references from these data bases and automatically processed them into tables. In fig. 2 the results are shown for some important target materials (the tables for the complete periodic system and more reaction types can be found in [7]). Measurements at a particular incident energy in the NSR data base are represented by boxes with vertical dashes and occurence in the EXFOR data base is displayed by boxes with horizontal dashes. See [7] for a more complete description of the tables. It is observed that there is a large difference in coverage between the bibliographic compilation (NSR) and the numerical data compilation (EXFOR). The tables display that there is a large treasure of experimental data, most of which have not been compiled in computer readable format,. It is also evident that many of the needs for transmutation studies are not covered by experimental data.

MODEL CODES

From the point of view of reaction mechanisms, the model codes that are used to predict the various cross sections above 20 MeV fall apart in two categories: the intranuclear cascade regime above 100 MeV and the region between 20 and 100 MeV. In another contribution to this conference [1 1], a number of intermediate energy codes will be compared with each other and with experimental data (with incident energies ranging from 25 to 1600 MeV). In [7], short descriptions of several intermediate-energy codes are given. Here, we will restrict ourselves to mentioning a few exponents of the aforementioned energy regions.

For energies between 100 and 1500 MeV, the most commonly used code is HETC, see [.5, 6] for a review. HETC, which exists in various forms. is based on Monte-Carlo intranuclear cascade followed by evaporation [14]. During the last, decade, various extended and modified versions of HETC have been developed. Two important improvements are the inclusion of high-energy fission [15] and pre-equilibrium/equilibrium emission. A HETC versions that includes both features is LAHET [16].

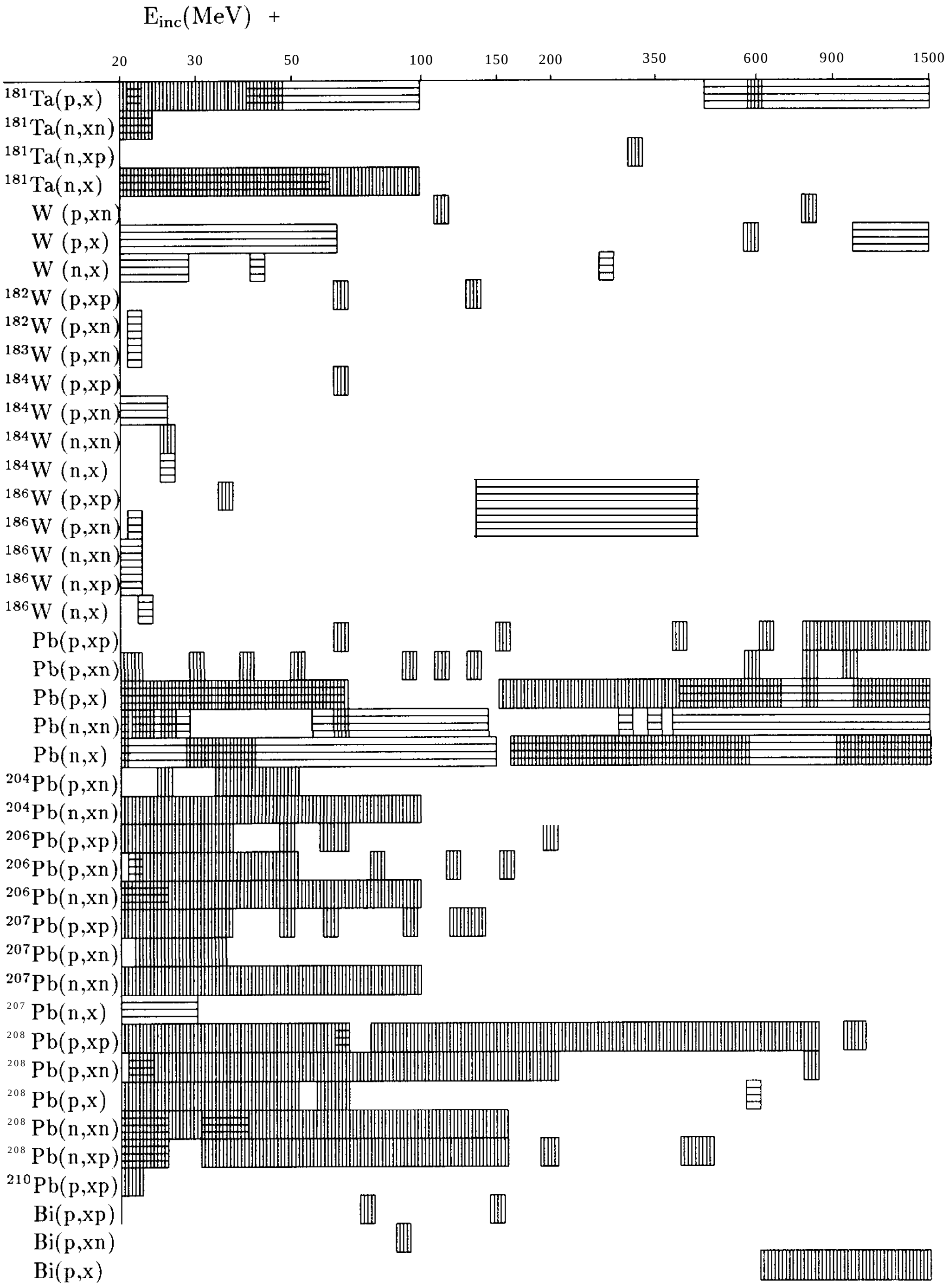

Figure 2: Retrieval of NSR (vertical lines) and EXFOR (horizontal lines) database for some possible target materials.

For the region between 20 and 100 MeV, we mention here the code GNASH [17], developed at the Los Alamos National Laboratory, which incorporates pre-equilibrium (Exciton model) and equilibrium (Hauser-Feshbach model) emission, as well as fission. A special version has been developed for the energy range above 20 MeV.

The code EXIFON [18] is a relatively fast computer code developed at the Technical University of Dresden, which predicts energy spectra and activation cross sections up to 100 MeV according to a statistical multi-step model.

Finally, the code ALICE [19, 20], developed at Lawrence Livermore National Laboratory, predicts pre-equilibrium, equilibrium and fission cross sections for energies up to several hundreds of MeV.

SYSTEMATIC

As the incident energy increases above several tens of MeV, many cross sections will display a smooth behaviour as a function of both incident and outgoing energy. Instead of resonances and other fluctuating quantum-mechanical phenomena that characterize the region of incident energies below 20 MeV, simple dependencies of the cross sections on the mass number and energy are expected to occur. On the other hand, the number of open channels becomes so enormous that it may be impracticable to measure all relevant cross sections or even to calculate them by nuclear models. These two aspects of high-energy reactions suggest that simple phenomenological parametrization of cross sections can or must be used as evaluation tools for high-energy nuclear data. Several investigators have systematically scanned the available experimental data in order to construct empirical formulae for the various types of cross sections. This has resulted in a valuable collection of easily implementable parametrizations for, among others, the elastic, nonelastic and total cross section, the (p,xn) cross section (very important for the first stage of the transmutation process), angular distributions, neutron yields and spallation cross sections. Empirical formulae for these quantities are listed in [7], together with their references.

THE STRUCTURE OF HIGH-ENERGY DATA LIBRARIES

Until recently, the effort of evaluators was almost completely restricted to the construction of data files for neutron-induced reactions below 20 MeV. Since a few years, it is recognized that high-energy nuclear applications, among which accelerator-based transmutation, can benefit by the availability of evaluated nuclear data libraries for incident energies up to 100 MeV or higher. A list of existing evaluations that exceed 20 MeV has been presented in [7]. Of these evaluations, the most recent ones are those by the Los Alamos National Laboratory [24, 25], for about 20 nuclides and energies up to 100 MeV, and those by the Brookhaven National Laboratory [21, 22], for ^{56}Fe, ^{208}Pb and ^{209}Bi and energies up to 1000 MeV.

In [8], a proposal is presented for the construction of three high-energy data libraries for accelerator-based transmutation and a detailed outline is given of the structure of each library (i.e. in terms of END F-6 nomenclature). Here, we restrict ourselves to the most important characteristics.

1. Transport data library to 100 MeV.

We assign the highest priority to this library, which can be used as a complement to the high-energy (> 100 MeV) data and transport calculations by HETC. It is recommended to construct a proton and neutron "starter" library which contains ENDF-6 formatted data for all relevant nuclides and for the most important reactions (see section on data needs). For the construction of a 100 MeV data, file , procedures different from those used in the low-energy files have to be employed since it is no longer possible to store each possible reaction in a section with a separate MT-number. However, for accelerator-based transmutation calculations, detailed low-energy neutron data will probably be as important as they are in normal reactor calculations. Therefore, we recommend to retain the original specification as obtained from low-energy evaluations for neutrons. For energies above '20 MeV, the detailed information concerning each individual excited state of the target nucleus is less important than for low-energy neutrons and the data should be lumped as much as possible in MT5 (which comprises all nonelastic processes that

Table 1: Directory of proposed proton data file to 100 MeV.

MF	MT	Description
1	451	General information
3	2	Elastic cross section
3	5	(p,anything) cross section
3	18	Total fission cross section
6	2	Elastic angular distribution
6	5	(p,anything) energy-angle distribution
6	18	Total fission neutron energy-angle dist.
8	454	Independent fission product yields
8	459	Cumulative fission product yields

are not explicitly considered in other MT numbers). The particle, photon *and* product yields and the required energy-angle distributions should be stored in MF6. For protons, a more uniform approach can be followed (because there is no connection with existing low-energy files). As an example, a proposed directory of a proton file to 100 MeV is presented in table 1. Note that almost all data are lumped in MT5, apart from fission, which is kept separated.

2. Reference data library to 1500 MeV.

The purpose of this library is to compare the data with HETC calculations or the direct use of the data in high-energy transport codes (in this way, the nuclear data part would be entirely decoupled from the transport part, just as for reactor calculations). The advantage is that more specialized nuclear model codes and experimental data can be used for certain sections of a datafile. Furthermore, instead of performing comprehensive Monte Carlo calculations (with a code like HETC) repeatedly for a certain material, it could perhaps be more economical to read the data. from an evaluated file. If the contributions from hadrous other than nucleons (such as π- and K-mesons) are important, extension of the ENDF-6 format is required. Perhaps it is even necessary to introduce separate libraries for *incident* hadrons (e.g. a pion library). If necessary, the part below 20 MeV could be added from existing libraries.

3. Activation data library to 100 MeV.

The purpose of an activation library is the calculation of the activation/transmutation of the target and its use in the reactor part of the hybrid system. This library should contain a very large number of nuclides with only activation and transmutation cross sections and without energy-angle distributions. Therefore, it is practical to separate it from the aforementioned files. In activation files below 20 MeV [23], reaction cross sections are stored by using MF3 in combination with many MT-numbers (one for each combination of outgoing particles). For an activation file to 100 MeV, this file structure cannot be adopted, since the number of possible different reaction channels becomes so large that a specification in terms of different MT-numbers is impossible. Therefore, for the extension to a high-energy activation file, we propose to construct a file consisting of MF3/MT5 for the absolute value of the total inelastic cross section and MF6/MT5 for all possible product nuclide yields.

CONCLUSIONS

A review is given of the available experimental data, theories, model codes and systematic that are relevant to accelerator- based transmutation of radioactive waste. It appears that there is a number of interesting tools for nuclear data evaluation beyond 20 MeV. It is important to validate these tools by proper benchmarking, such as the current NEA model code intercomparison [9-11]. This could be a good starting point for an "International Evaluated Nuclear Data File for Transmutation". Provisionally, such a library should emphasize the energy range up to about 100 MeV. For this library. the current ENDF-6 format is appropriate for the representation of data [8]. Above 100 MeV, high-energy transport codes based on the intranuclear cascade model can be used and the need for externally supplied data may be less urgent. For future reference libraries in this energy region. the ENDF-6 format must be extended in order to take into account pions and other hadrons.

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