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CROSS SECTION MEASUREMENTS, INTEGRAL VALIDATION EXPERIMENTS AND NUCLEAR-MODEL CODE DEVELOPMENTS OF IMPORTANCE TO ACCELERATOR-BASED TRANSMUTATION

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Abstract Present concepts and objectives of accelerator-based transmutation are briefly summarized. The significance of nuclear data for the prediction of neutron fluxes achievable in an extended transmutation blanket and for the estimate of the resulting transmutation rates is outlined, and the status of available data is discussed. The data base for neutron production cross sections of spallation reactions with -100-1500 MeV protons is sufficiently available from previous experiments. Some systematic discrepancies, however, still exist for measured double-differential neutron production cross sections, especially in the higher energy region above -20 MeV. For radiation transport calculations the nuclear data base is reasonably well known in the energy range from thermal to about 20 MeV. Above that energy, experimental cross section data are rather sparse. The important transmutation cross sections are reasonably well known for accelerator-based transmutation concepts employing thermal neutrons, except for some short-lived intermediate chain nuclides. A special problem in present accelerator-based transmutation concepts is radiation damage in the primary target containment and other structural materials. More reliable estimates of the life times for such components require more accurate calculations of displacement and gas production rates in suitably selected materials. While dpa and gas production cross sections below ~20 MeV can be largely derived from existing evaluated nuclear data files, additional values above that energy are often merely extrapolations from low energy results. The role of cross-section systematic and nuclear-model calculations in predicting unknown data is briefly discussed.

(Key words Medium-energy protons: Cross sections for spallation and fission reactions, for neutron and secondary proton production model validation from integral measurements of yields, spectra and reaction rates; nuclear-model code developments for transmutation studies)

1. Introduction

Accelerator-based transmutation of high-level radioactive waste from LWRS has become an interesting alternative to nuclear incineration in thermal and fast fission reactors. Present investigations of accelerator-based transmute concepts cover a wide scope of approaches ranging from incineration of single nuclides, carrying a major fraction of the total toxic inventory, to effective transmutation of all long-lived actinides and fission products. In addition, pure transmutes, hybrid systems of power and burner reactors assisted by a proton accelerator as well as transmutes with additional energy production or fuel breeding from fertile materials are presently under discussion. For utilization concepts of transmutation reactions there are also different approaches: One is the direct incineration in thin and in quasi-infinite primary spallation targets with primary" protons and direct products of the nucleon meson-cascade. Two other possibilities refer to transmutations with secondary neutrons in fields of either fast or thermal high-intensity neutron fluxes. A common feature in all concepts is the usage of proton (deuteron) accelerators for energies between about 600 to 1500 MeV. High-intensity cw beams with currents of a few tens to several hundreds of mA are typical for all applications.

In the present survey the main emphasis is on transmutation aspects only; data and validation needs for additional energy production or fuel breeding are not generally covered. Current concepts and objectives in accelerator-based transmutation technology are briefly outlined in Sect. 2. The significance of nuclear data for the special field is discussed in Sect. 3. Section 4 is devoted to cross section measurements for high-energy protons, neutrons and other important particles involved. The situation for integral validation experiments is discussed in Sect. 5. In Sect. 6 the status of nuclear model codes for the calculation of unknown nuclear data is summarized. Section 7 gives a brief survey of desirable improvements for differential cross sections measurements, integral data testing and nuclear model codes.

2. Transmutation Concepts and Objectives

Many years of accelerator-based transmutation studies have led to an increased understanding of the utilization of accelerators for nuclear transmutation. The virtues and shortcomings of the different concepts have been worked out in some detail. The general possibilities to tackle the problem of high-level waste from LWRS by burning instead of storing the most problematic high-toxicity products from this waste have been outlined. The different approaches for accelerator-driven transmutation technology can be divided into four main categories of concepts:

1. Concepts in which the high-level waste is burned in the primary spallation target itself either by spallation or fission processes. In this case the critical waste can be irradiated in form of purified material concentrated in thin samples, or as a certain class of the most problematic nuclides (e.g. minor actinides) concentrated in a quasi-infinite sample (the whole primary spallation target) This concept has e. g. been studied at the PSI [1].

2. Concepts which make use of the large number of secondary neutrons produced by spallation reactions in heavy element targets. In these cases the transmutation is accomplished in large blankets surrounding the primary neutron target. High-flux fields of fast or thermal neutrons are being considered in current concepts of different groups [2-4].

Tab. L Some important fission products and minor actinides contained in the high-level waste from a PWR * (from Ref. 2).

Fission products

Nuclide	kg/yr	Half-life (yrs)	Cross Section (b)
^{99}Tc	21.5	2.1X10^{5}	20
^{129}I	5.87	1.6x1O'	27
1 37~5	30.9	30	0.25
^{90}Sr	13.5	29	0.9

Minor Actinides

Nuclide	kg/yr	Half-life (yrs)	Atoms X10^{25}
^{237}Np	14.5	2.1X106	3.66
2 4 1 *	16.6	432	4.13
^{243}Am	3.43	7.4X10^{3}	0.73
^{244}Cm	0.58	18.1	0.13

" All amounts are annual production for a PWR running at 3 GW thermal with fuel burnt to 3.3x10^{4}megawatt-days per ton.

3. Concepts using burner reactors assisted by a high-current medium-energy proton accelerators [5-7] which are mainly advanced subcritical fast fission reactors for energy production and transmutation purposes.

4. Concepts for advanced fuel-cycle (e.g. thorium) reactors and burners with low radioactive inventory and minimum production of long-lived high-toxicity actinides and fission products [3,8].

Also the first two concepts consider occasionally additional energy production or fuel breeding, in order to achieve a more favorable overall energy balance, i.e. to operate them more economically. Two more recent considerations of accelerator-based transmutation refer also to the use of intense electron beams and muon-catalized fusion [9,10],.

Regarding the long-lived waste discharge from LW power reactors the most desirable goal is to care for both the most problematic actinides and fission products, simultaneously. Assuming that uranium and plutonium can be efficiently separated chemically, and reentered to the fuel cycle, only the fission products (FP) and the so-called "minor actinides" (MA) need special consideration. In Tab. I some of the important nuclides contained in the high-level waste are listed together with their production rates, half-lifes and thermal cross sections. The most important MAs to be burnt in future transmutes are ^{2}37 Np and ^{241}Am and, possibly, ^{243}Am, ^{244}Cm and ^{245}Cm. For fission products major concern is presently given to ^{99}Tc and ^{12}91 and, with lower priority, to ^{137}Cs and ^{90}Sr. But even in very high thermal neutron fluxes of the order of ~1x10^{16} cm-z S-l, there does not seem to be a reasonable chance for an efficient transmutation of the latter two fission products. This is mainly due to the low thermal neutron cross sections.

3. Significance of Nuclear Data

It is now widely agreed that accelerator-based transmutation involving effective chemical separation methods and partitioning is an attractive possibility to deal with the long-standing problem of waste management for LWR spent fuel. To arrive, however, at a better judgement on the realization and the achievable specifications of such facilities, a large number of new and/or more reliable nuclear data is needed. These range from proton-induced spallation reactions and fission processes over γ- and neutron transport cross sections, cross sections for various transmutation reactions to damage and gas production cross sections for materials damage calculations. The energy region for such data ranges from thermal to the highest energies involved, i e. the energy produced by the applied accelerators (up to ~1500 MeV). This paper will mainly concentrate on the energy range above ~ 20 MeV. For lower energies, there are, at least for neutrons, various evaluated data libraries available or under development from previous work in fission- and fusion-reactor technology. (ENDF/B-VI, JEF-2, EFF, JENDL-3 BROND, CENDL-2, EAF, REAC, FENDL-2 etc.) [11-18]. In this lower energy range we, thus, shall consider only those cases for which additional or more precise data are needed for the special purpose considered here.

For transmutation aspects the major data needs refer to the following four types.

1. Nuclear data for high-energy proton reactions (including high-energy fission) on a large number of target materials. For those cases in which trans-

mutation from direct proton reactions is considered there is a special need of cross sections for the production of direct spallation and fission products on the important MAs and, perhaps, FPs to be burnt. For concepts based on large fluxes of neutrons with either thermal or fast neutron spectra, neutron production cross sections in heavy elements such as Ta, W, Pb, U or the eutectic Pb-Bi must be well known for the determination of the resulting neutron fields. For neutron field characterization double-differential neutron production cross sections are required. Furthermore, elastic and inelastic scattering cross sections for the major spallation target materials must be known, to get a realistic estimate of the neutron spectrum leaking from the surface of massive primary targets.
2. For neutron transport and, eventually, effective neutron moderation a large number of neutron- and γ-transport cross sections need to be known with sufficient accuracy. Such estimates have to consider the effects of scattering, absorption and self-shielding, not only in the solvent and moderator materials but also in the dissolved waste.
3. Depending on the type of the considered neutron field (thermal or fast), a large amout of cross sections for all major transmutation cross sections on the waste nuclides to be burnt and on the nuclides involved in the important reaction chains is needed, for thermal neutron concepts mainly neutron capture, fission cross sections and resonance integrals. A large amount of these data are already known from fission-reactor work. But there is also a clear need for additional thermal data for short-lived nuclides such as ^{238}Np and, possibly, for other short-lived MAs and FPs.
4. For all types of concepts radiation damage in the primary target containment and other nearby structural components is a special problem, since these are exposed to extremely high fluences of high-energy neutrons. Especially in the primary target containment, radiation damage for fluxes of the order of 10^{16} cm-z S-l is much more severe than that in the first wall of future fusion reactors. Thus, for more reliable estimates of the total life-time of such components, a large number of damage and gas production cross sections in the energy region from thermal to several hundred MeV are needed. While for energies below ~ 20 MeV these can be largely derived from existing evaluated nuclear data files [19], additional values above that energy are often merely extrapolations from low-energy results. Far beyond 20 MeV, cross sections are typically obtained from not well established calculations with special versions of the Nucleon-Meson Transport Code, e.g. VNMTC [20].

Summarizing, there is a large number of additional cross sections and nuclear data that are required for more detailed investigations on accelerator-based transmutation concepts. Since there is only little capacity for new cross section measurements in the future, the majority of such results must come from nuclear-model calculations with suitably validated codes. For nuclear-model validations rather different types of integral experiments are needed to perform suitable data testing. In the following two sections the present status of cross section measurements and of integral validation experiments is discussed in some detail.

4. Differential Cross Section Measurements.

4.1 (p,xn') and (p,xp') Cross Sections

Mainly due to the previous interest in high-intensity spallation sources for solid and liquid-state physics, but also in connection with cosmology and space investigations, a larger number of microscopic cross section measurements have been performed for protons and other light charged particles in the energy range from several tens to 200 MeV and in the range from ~ 600 to 3000 MeV [21]. A typical example for a systematic study of differential neutron production cross sections is shown in Fig. 1. Double-differential cross sections for 585 MeV protons and a large number of target elements from C to U have been measured by the KfK

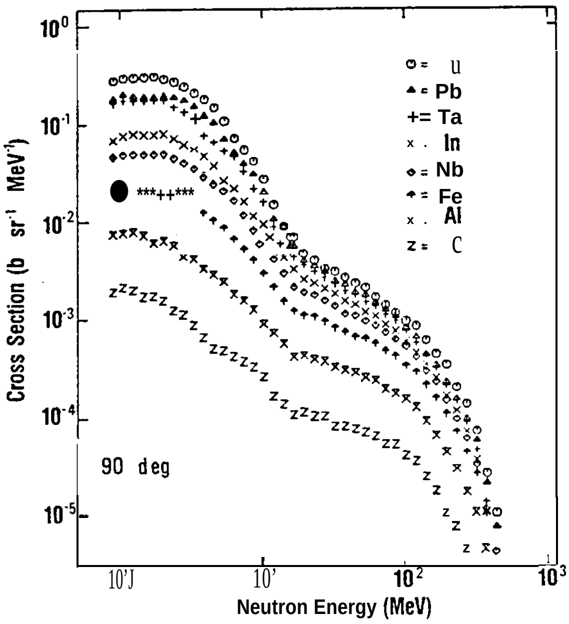


Fig. 1. Double-differential neutron production cross sections for 585 MeV protons on eigth target elements from C to U at 90° laboratory angle. From Ref. 22.

group at the PSI cyclotron [22]. The figure shows the results at 90° emission angle. It can be seen that the spectra consists mainly of two contributions: (a) neutrons from evaporation processes which are characteristic for the Maxwellian-type of spectrum below about twenty MeV and b) neutrons from direct processes produced during the intranuclear cascade and during high-energy nucleon transport creating the broad shoulder around 50 MeV and extending to energies almost as high as the incident proton energy. For very small emission angles a third and fourth contribution comes from (c) quasielastic charge-exchange (CEX) reactions forming a pronounced peak at the high-energy end of the spectrum, and (d) quasifree pion production processes exhibiting a broader peak slightly below the CEX one [23].

Even more extended systematic measurements than at KfK have been performed at Los Alamos [24-29]. Especially in the last few years, double-differential cross section measurements where performed for different proton energies between 100 and 800 MeV. Even though the investigators of neutron production cross sections give rather small uncertainties for their results of the order of ~ 15%, the agreement between such measurements does not seem, in general, to be as good as that. This can be judged from the example shown in Fig. 2. It displays a typical result from a recent comparison of the LANL and KfK results for 597 and 585 MeV, respectively. The diagram shows the measured cross sections at 30° emission angle for Pb. It can be seen that there is a reasonable agreement in the spectrum shape below ~20 MeV. But above that energy the cross sections sections deviate rapidly in absolute values, reaching a factor of about 2 around 100 MeV. For 150° the general observation is similar; in addition, however, there are also major differences close to the cutoff energies.

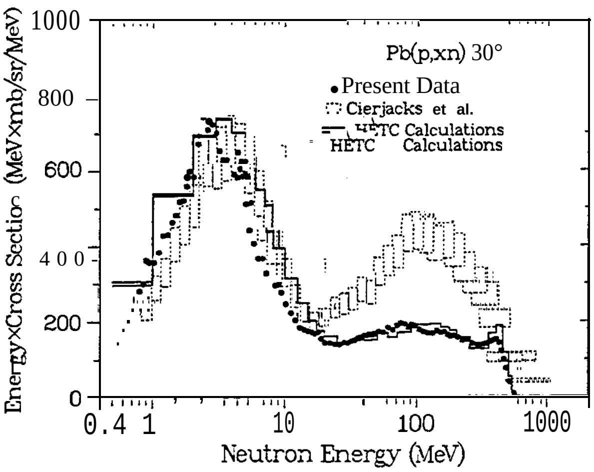


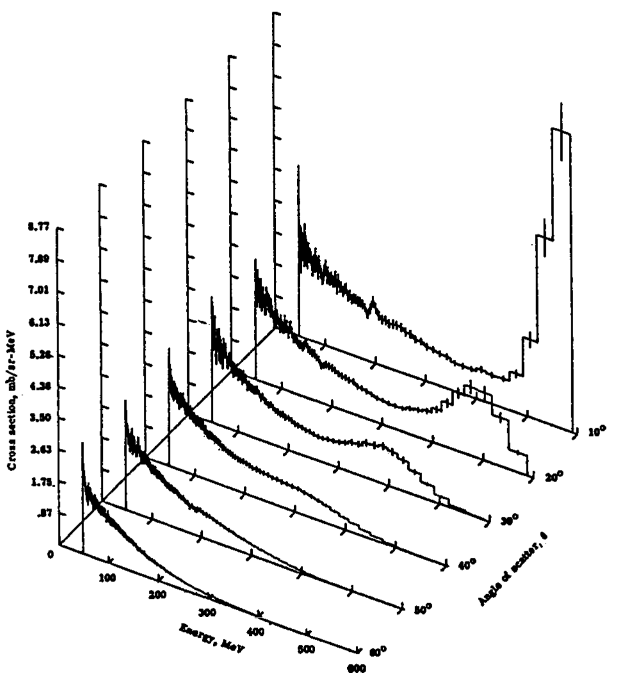
Fig. 2. Comparison of neutron production cross sections for 585 and 597 MeV protons on lead. From Ref. 27.

Similar discrepancies are found for most of the other commonly measured target elements. Thus it becomes clear that there are still large systematic errors in such measurements, which are not properly taken into account. The two critical points in neutron cross section measurements are neutron detection efficiencies and detector thresholds. For all types of thin hydrocarbon scintillation detectors, detection efficiencies are extremely small (a few %) at high energies and close to the detector thresholds where the efficiency rises rapidly from zero to maximum value. Therefore, a careful control of the detector threshold throughout the experiments and a direct measurements of the detector efficiency at high neutron energies is an important prerequisite for precise neutron cross section measurements.

Other maesurements over an extended angular range and for several target elements have been made by Wachter et al. [30]. For protons of 800 MeV Bonner et al. [23] have performed a systematic study of 0° neutron-production cross section for a number of targets between Al and U. Other 0°, 50° and 130° neutron production cross sections for 647-, 740- and 800-MeV protons on Be, C, Al and U have been measured by Madey and Waterman [31] and by Cassapakis et al. [32]. For 120- and 160-MeV protons Scobel et al. [33] measured double-differential (p,xn) cross sections for “Al, ^{90}Zr and ^{208}Pb at 13 angles between 0° and 145°.

For (p,xp′) reactions also a number of cross section measurements have been performed for various energies between 450 and 1500 MeV. For this kind of experiments absolute measurements are more straight-forward than for neutrons, but such measurements typically cover only the higher parts of the energy spectra, because low energy protons are typically stopped, even in rather thin samples.

Proton reaction cross sections for 220 to 570 MeV protons on 8 elements and H_2O, B_4C, NaJ and CH_2 have been measured by Renberg et al. [34] at CERN. A systematic study of double-differential (p,p′) cross sections, similar to that shown for neutrons has been performed by Beck and Powell [35]. A sample result from their measurements is given in Fig. 3. This shows the results for 558-MeV protons incident on Pb and for secondary protons emitted at 10° to 60°. For reasons mentioned above, proton cross sections were only obtained for energies above 50 MeV, so that the evaporation part of the spectra is already cut off. The measured portions of the spectra are very similar to those observed for secondary neutrons. They show the same characteristic shoulders at energies around ~ 100 MeV. For the emission angle of 10° the elastic scattering peak is clearly visible, which is the equivalent to the charge-exchange peak in the neutron data. From theoretical arguments it is expected that the energy-integrated cross sections for both (p,xn′) and (p,xp′) should have practically the same absolute values for thresholds above ~50 MeV, where the effect of the Coulomb barrier is negligible. A test of this assumption by comparison with the KfK neutron data for 585-MeV protons gave a positive result [22]. Energy spectra of secondary charged particles produced by 660-MeV protons on Be, C, Cu and U targets have been measured by Azhgirey et al. [36] at several forward angles ≤30°. At bombarding energies of 800 MeV systematic cross section measurements for protons scattered to forward angles between O and 30° have been performed for various target elements between Li and Pb by Chrien et al. [37].



(a) Protons.

Fig. 3. Measured secondary-proton-production cross sections for 558 MeV protons on Pb for emission angles 10” to 60°. From Beck and Powell [35].

4.2 Spallation and Fission Cross Sections

Concerning spallation and fission-product cross sections several measurements have been made for medium-energy protons. Often, however, these data are not complete. In many cases only certain types of radionuclides (β^--, y-ray-, weak X-ray-emitters) have been investigated. Regarding spallation products, Pate and Poskanzer [38] measured cross sections for the production of isotopes of U, Pa, Th and Ac in the irradiation of ^{238}U, ^{235}U and ^{232}Th with 680 and 1800 MeV protons. In addi-ion, some spallation yields were determined at other bombarding energies ranging up to 6.2 GeV. Activation cross sections for about thirty spallation products from 100-340 MeV protons on ^{23}*U and ^{232}Th targets have been determined by Lindner and Osborne [39]. A rather complete cross section measurement of the spallation products from 3-GeV protons on Ag was carried out by Silberberg and Tsao [40]. Concerning high-energy fission cross sections, the available experimental data available before x1980 have been summarized in two surveys from Takahashi [41] and Atchison [42]. Total fission cross sections for 1-9 GeV protons on ^{209}Bi, ^{235}U, ^{238}U ^{237}Np and ^{239}Pu are available from experiments of Matusevich and Regushevskii [43]. A large number of formation cross sections for fission products from several fissile nuclei bombarded by protons (10-340 MeV) and deuterons (20-190 MeV') are available from the experiments of Stevenson et al. [44]. For proton energies below 200 MeV and at energies of 600, 800, 1200, 1600 and 2600 MeV Michel et al. [21] have mesured reaction cross sections for a large number of target elements ranging from C to Au and an enormous number of radioactive product nuclei decaying by γ-ray emission. Reaction cross sections for a large number of fission and spallation products from bombardment of Pb and depleted U with protons of 600 and 1100 MeV have been determined by Amian et al. [45]. A typical example from this work is given in Fig. 4. This shows the results for 1100-MeV protons on depleted uranium as taken at the SATURNE machine. An obvious observation is that the measured data for fission products are clearly different in absolute raction rates for β^-- (alone) and β^-- plus X-ray (electron capture) emitters, respectively. From comparisons with HETC calculations it can be seen that, at least HETC/KFA- 1 [46], does not well reproduce the measured double-humped fission product distribution, and gives a clear shift to lower masses by about 20 mass units. But due to the simplified treatment in the KFA calculations, a recalculation with more realistic assumptions would be desirable in this case.

Although a large number of special measurements have been performed by several authors, in general, differential cross section and reaction rate results are still rather incomplete in view of the overall needs for transmutation applications.

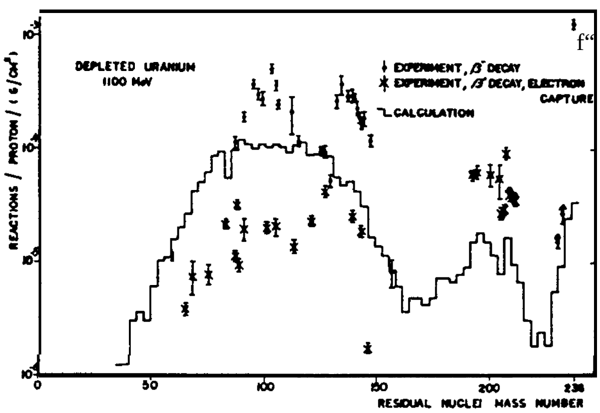


Fig. 4. Measured and calculated isotope production rates versus mass of spallation and fission products. Experimental results are from Amian et al. [45]. Model calculations were performed wit the HETC/KFA-1 code.

4.2 Damage Cross Sections

As mentioned in Sec. 3, the main interest is in displacement and and gas-production cross sections, and relates to life-time considerations for the primary target container and other structural materials. In this context, the main emphasis is on the effects of high-energy neutrons. Total displacement and He-production cross sections below ~ 20 MeV can be largely derived from existing evaluated nuclear data files. For 14.8 MeV Neff et al. [47] have recently made new extensive systematic measurements of He-production cross sections in pure elements, isotopes and alloy steels.

Measurements above 20 MeV are rather rare. At 0.6 and 3 GeV Kruger and Heymann [48] have measured H- and He-production cross sections for C, O and Si. In addition, Kwiatkovski et al. [49] studied mass- , energy- and angular distributions of reaction products formed in collisions of 180 Mev protons with Al target nuclei. An interesting new method for the measurement of element transmutation from irradiations with high-energy protons has been established by the PSI group [50]. This group has demonstrated that a total reflection X-ray fluorescence (TXRF) spectrometer, using monoenergetic synchrotrons radiation as the primary X-ray source, is suitable to measure extremely small concentrations of transmuted elements. So far, only chemical yields from medium-weight-Z metal targets have been investigated. But the method can be also applied to high-Z targets for spallation and transactinide product measurements.

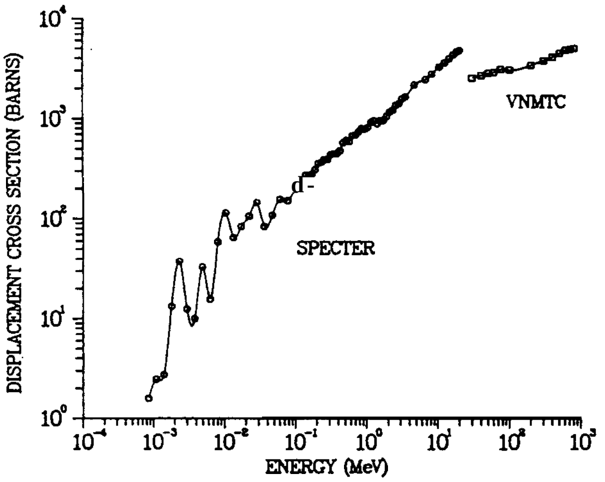


Fig. 5. The damage cross section for neutrons in iron vs neutron energy [20]. The two branches are calculated from ENDF/B-V (left side) and VNMTC (right side) cross section data. It can be seen that the two branches do not match at the intersection point.

From the above description it becomes clear that the majority of damage cross sections above 20 MeV must come from extrapolations of low energy data, or - for largely extended energies - from suitable medium-energy nuclear model calculations. This procedure may be a reasonable approach for helium production cross sections, as shown by Wechsler et al. [20], but does not work suffiently well for displacement cross sections. The latter deficiency is illustrated in Fig. 5 which shows an example from the work of the above authors, who used measured cross sections below 20 MeV and calculated ones above that energy. It can be seen that there is a glaring discontinuity at the intersection point of 20 MeV: While cross section data for high energies were taken from nucleon-meson transport calculations, the corresponding results for low energies were obtained by using well established neutron data from the evaluated neutron data file ENDF/B-V.

5. Integral Validation Experiments

In principle, all types of expedients performed for differential cross section measurements have been also applied similarity to integral validation measurements employing realistic target sizes and materials. While the experimental methods are widely the same, such integral measurements have the advantage that, in contrast to cross sections, also the quality of transport code predictions can be tested simultaneously. On this basis, only the corresponding results that are specific for transmutation applications will be briefly mentioned.

5.1 Neutron and Proton Production

Concernig neutron production, the most general quantity is the total neutron yield, i.e. the number of neutrons produced per incident proton. The pioneering work in this field has been performed by Fraser and Bartholomew in 1966 [51], who bombarded thick elemental targets with protons of energies between 500 and 1500 MeV. A typical result from their work is shown in Fig. 6. It can be seen that the number of neutrons per incident proton increases almost linearily with increasing proton energy. In addition, the integral yields increase rapidly with increasing target mass. These early measurements have been supplemented by experimental results from various laboratories in the meantime for extended proton energies and target masses, so that there is now a rather complete set of total yields. Total neutron yield data are presently reasonably well predicted by existing nuclear model codes. There is, however, a significant difference in total neutron yields from integral thermal and differential fast neutron measurements. From the latter a more pronounced saturation is indicated around ~ 1500 MeV [52].

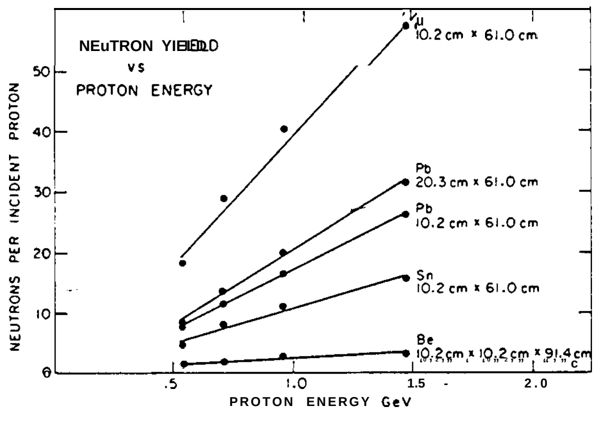


Fig. 6. Total neutron yields obtained from the bombardment of thick targets by intermediate energy protons. Targets of Sn, Pb and U were cylinders; the first number given is the target diameter, the second the target length. The Be target was rectangular (from Ref. 51).

Spectral yields of neutrons penetrating the surfaces of massive spallation targets have been measured in several laboratories. These include measurements for different target diameters and cross sections, different incident proton energies as well as their dependence on emission angle and penetration depths into the target. In the framework of the previous SNQ project the Karlsruhe group measured thick-target yield spectra for depleted-U and Pb targets at 590 and 1100-MeV protons for a large number of penetration depths into the target [52,53]. Various measurements of surface neutron yields from stopping-length and near-stopping-length targets for 100-300 MeV protons were also

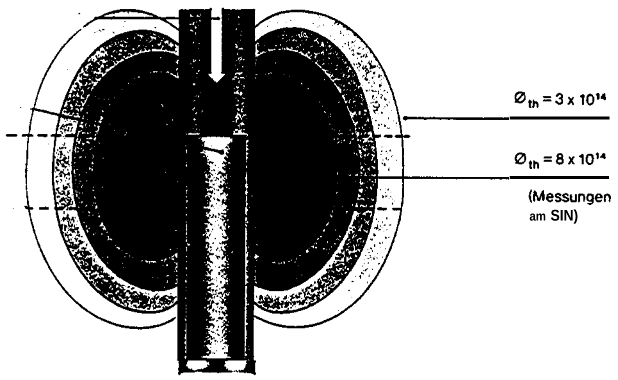


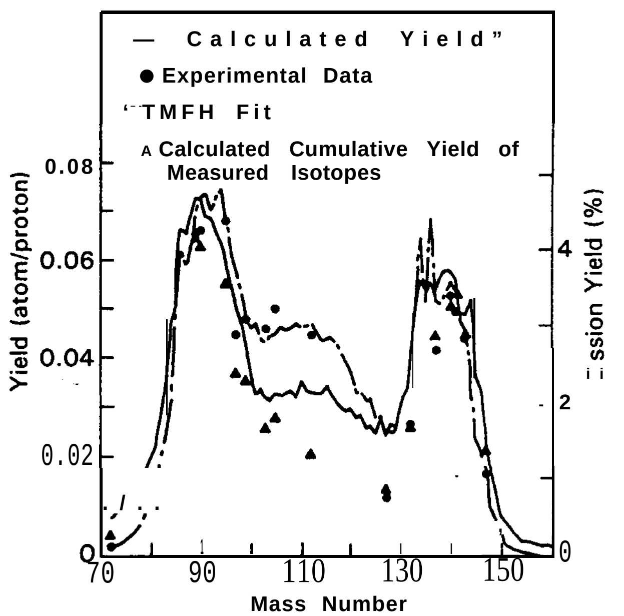
Fig. 7. Flux distribution for a D, O-moderated thick Pb cylindrical target. Measurements with 590 MeV protons where performed at the PSI in a heavy water tank of ~ 4 m³. From Ref. 60.

made by the Los Alamos group [25,26,54]. At 800 MeV Fullwood et al. [55] and Waterman and Madey [56] measured some other thick target yields. At TRIUMF Wachter et al. [571 measured similar cross sections at 450-MeV for some light elements and a few emission angles. The Russian group at St. Petersburg performed a number of measurements for 1 -Gev protons incidents on targets of Al, Fe, Pb and composite U-Al [58]. Neutron spectra at different positions of the target by the activation method, and total neutron yields where derived. Additional measurements for lead and other target with protons of 2 and 2.5 GeV are in progress [59]. A special time-of-flight spectrometer for use at the proton machine in Dubna has been and set up and is operational now. Even more sensitive for validation of presently used high-energy nuclear transport codes are space-dependent flux measurements in moderated target assemblies. A typical result of this kind is that from the SNQ study obtained by Bauer et al. [60]. These authors measured extended flux contours for a target assembly using a thick Pb target inserted into a large cubic D_2O tank. Their result for the middle-plane of the target is given in Fig. 7. Even though less frequent, also a few integral measurements of secondary proton spectrum measurements have been made for thick targets, e.g. [30,57].

The general observations from integral neutron experiments are similar to those found for differential cross section measurements: Neutron yields from different laboratories are often discrepant in absolute values and shapes. Especially for large emission angles, different portions of high-energy neutrons are observed by different groups.

5.2 Spallation and Fission Products

Integral validation experiments have often been made in combination with thin target measurements (see [451 etc.). For 1-GeV protons incident on large diameter and totally absorbing targets of Al, Fe, Pb and composite U-Al, Ivanov et al. [61] measured various yields of radioactive reaction products. Results of such activation measurements were obtained for several penetration depths of protons and different target radii. Axial distributions of fission and spallation products in stopping-length targets of Th and depleted U bombarded by 800-MeV protons have been determined by Gilmore et al. [62]. Ratios of yields were measured for 3 spallation and 16 fission products; integration over the whole target length gave total production yields. A partial result from this work is given in Fig. 8. This shows the fission product yields from Th for the front face foil of the central rod. The mass-yield distribution is double-humped, and exhibits some signs for high-energy fission. The shape of the mass-yield curve is asymmetric, with the heavy peak (around mass 140) lower in magnitude and narrower in width than the light peak (around mass 95). The shape and magnitude of the mass-yield curve can be reasonably well described by the RAL fission model.



Pig. 8. Comparison of measured and calculated fission product yields from the bombardment of Th by 800-MeV protons. The solid lie was calculated using the RAL fission model with B_0 = 8 MeV. From Ref. 62.

6. Nuclear-Model Code Developments

For calculations of the various data needed for accelerator-based transmutation concepts appropriate nuclear model codes are available. An extensive survey of the currently existing codes has been recently finalized by Takahashi [63]. One of the most frequently used type of codes for this purpose is the High-Energy Nucleon-Meson Transport Code (HETC) in its various incarnations. Even though the terminus HETC is often used in a wider sense as a code system for various kinds of broad-scale Monte-Carlo transport calculations, we shall restrict here to its genuine meaning of a nuclear-model co& for the treatment of intermediate-energy nuclear reactions only. A full description of the physics involved and the routines produced for utilization can be found elsewhere [64-69]. Thus, only a very brief characterization is given below.

The fundamental part of all HETC-code versions is the intra-nuclear cascade (INC) model that describes the interaction of nucleons with nuclei. This is done with a two-stage model. The first stage is an intra-nuclear cascade of individual nucleon-nucleon interactions described by the appropriate medium-energy kinematics, relevant conservation laws and 'free' particle nucleon-nucleon and nucleon-meson cross sections. The treatment of intranuclear scattering includes an estimate of Pauli exclusion for prohibiting scattering into occupied states and a consideration that the nucleons of the target nucleus have a Fermi momentum distribution which broadens the range of energies for nucleon-nucleon collisions. The fate of both nucleons (and/or one or two mesons) is followed until the ICN transport shows that the cascade products have either reached the nuclear surface or that all nucleons have fallen below an arbitrary low energy cutoff (often ~15 MeV). This first part of the interaction is typically treated by Monte-Carlo methods using a so-called "Nucleon-Meson Transport Code" (NMTC). The outcome of this stage is a set of escape particles and a residual nucleus left in a more or less highly excited state. The second stage is the treatment of the deexcitation of the residual nuclei by particle and y-ray emission. For this step usually a simple statistical model for a fully equilibrated compound nucles is employed, so that there is typically no consideration of preequilibrium effects.

The original HETC code developed at ORNL has been improved in the past by several groups, and exists now in various different laboratory versions (e.g. KFA, PSI, LANL, JAERI). An important step in the new versions was the introduction of an adequate fission model. Presently the most widely used one is the RAL model developed by Atchison [42]. Modifications have also been made by using improved level density assumptions [70]. At JAERI various modifications of the NMTC computer files have been made (or are in progress) which take particularily care of the needs for nuclear spallation simulations and transmutation analyses [71]. These concern the inclusion of an extended range of nuclides with higher mass numbers (up to A = 250) and a new treatment of high-energy fission. Another improvement made at JAERI is the production of a new algorithm which allows to treat also particle emission from preequilibrium states. This led to an important improvement in the prediction of measured high-energy tails in the neutron-production cross sections: An illustration is given in Fig. 9. which shows an example of the KfK data measured for 585 MeV protons on lead. It can be seen that the inclusion of preequilibrium emission gives a much better agreement between calculations and measurements thanprevious standard calculations.

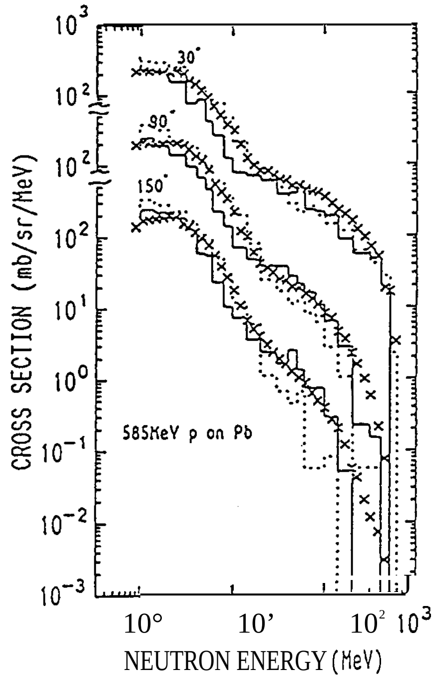


Fig. 9. Comparison of measured and calculated double differential Neutron production cross sections for 585 MeV protons on lead [71]. xxx experimental data [22], . . . HETC standard calculations _ HETC calculations with an exciton model.

Another approach for the treament of the first-step high-energy cascade has been recently described by Blann [72,73,33] (and the advantage has been recognized also by other groups): This author showed that a great simplification is possible over the INC-model approach, if one looks only at the partition in energy that results when there is a nucleon-nucleon scattering process. In this approach the exciton model, which was originally proposed by Griffin [74] can be applied. A hierarchy of configurations following one, two, three etc. N-N scattering events is taken into account. In this way one can use simple statistical formulas, or partial-state densities to calculate the number of (assumed) equally-like energy partitions for each hierarchy based on number of scattering events. The physics of this model and the suitable selection of model paramters are well described elsewhere, and shall not be discussed here further. Instead, it should be stressed only that the pre-equilibrium (PE) model is another interesting alternative to INC, which has already been successfully tested for energies up to -200 MeV [21,33]. Inclusion of relativistic kinematics, direct contributions and inelastic nucleon-nucleon channels are expected to extend the validity of PE models also to energies up to 1 GeV and more [72]. First tests of this option have been made by several groups in different laboratories [72,75,76]. Finally, the two intranuclear cascade codes PHENIX [77] and SITHA [78] should be mentioned, which have been developed at Moscow and Leningrad, and are widely used by the Russian physicists. To my understanding these codes are presently also undergoing various modifications, in order to be most useful for applications in accelerator-based transmutation studies.

7. Future Improvements

In general, there is a substantial amount of cross sections, integral validation experiments and nuclear model code developments available. This material is, however, widely spread around in various laboratory reports, conference papers and journal articles. So a very useful starting point would be a suitable compilation in a CINDA-type index to literature and computer files on microscopic and integral nuclear data for transmutation. Such work has, in principle, already been started at the US National Nuclear Data Center in Brookhaven [79], even though in a more general context of applications.

A second worthwhile task seems to be a limit-d evalution of the most critical microscopic data including integral benchmark experiments, in order to solve or, at least, identify important problems in the existing data base, e.g. for double differential neutron production cross sections and integral yields. For neutron transport data Arthur et al. [80] continue to develope higher energy transport libraries and perform the necessary benchmarking. More generally, it might also be timely for special sensitivity studies on data, for which significant and system-driving uncertainties exist, and identify on this basis urgent validation measurements to solve some of the most urgent problems.

Although there are certainly many other desirable activities for improvements of contemporary transmutation predictions, at least two special ones, stressed in the various comments, I received during the preparation of this paper, should be mentioned. One is certainly the influence of short-lived isotopes involved in thermal neutron transmutation chains. A special nucleus of this kind is clearly ^{238}Np, and there are possibly several others which may have considerable influence on the finally achievable reduction in toxicity and the resulting 'effective' half lifes. Moreover, there is still a need for neutron reaction data on fission products such as ^{137}Cs, for which significant and system-driving uncertainties now exist. Another example may be the data impacting radionuclide production that are important in determiningg the overall mass balance of a transmutation system, i.e. the mass balance of materials transmuted versus wastes (chemical and nuclear) created.

Other areas which might need experimental and theoretical efforts to improve the data base are spallation and fission reaction that are part of the neutron target radionuclide production mechanism. In the PSI concept for ^{237}Np in the primary target; the theoretically achievable reduction of the toxicity depends on the high energy fission model. An additional effect arising from neutron interaction on unstable nuclei is the production of long-lived radionuclides in the primary target, which seems (from present calculations) to be dominated by low energy neutron reaction processes.

A final aspects to be mentioned is nuclear data for the investigation of radiation damage, which is a severe consideration at least for the primary target container. In general, it is an important future task to gather the extensive data information from operating spallation neutron sources, now available or to become available soon.

8. Summary

Even though many other aspects of accelerator-driven transmutation technology have been studied in the past, one of the most urgent goals is nuclear incineration of the high-level waste from LWRS. If uranium and plutonium can be effectively separated chemically, and fed back to the fuel cycle for energy production, the main task is to bum the long-lived toxic isotopes of the minor actinides and of the fission products. The most ambitious approach is to transmute both MAs and FPs in one type of facility. Present concepts for accelerator-transmutation-of-waste (ATW) facilities involving efficient chemical separation methods and portioning look rather promising for this major goal, and there are claims, that such facilities may transmute all long-lived waste discharge from ten 1 GW_e power reactors per year [2]. -

For all transmutation studies a large number of additional, not yet available nuclear data is needed. Although not yet well specified in detail, the general data needs range from high energy proton reaction cross sections, via radiation transport data as well as transmutation reaction cross sections to radiation damage cross sections for the whole range from thermal energies up to more than 1 GeV. Such data are not only needed for protons and neutrons, but also for the various products generated during the nucleon-meson cascade or on the path of long-range particles and cascade products on a large variety of technologically important elements and nuclei. The best situation exists presently for neutron data below ~20 MeV. Extended data libraries are well developed from previous fission- and fusion-reactor programs, and suitable upgrades for some transport cross sections up to ~100 MeV for ATW applications are being made. Present shortcomings belong mainly to special elements and isotopes especially short-lived isotopes such as ^{2}38Np, and possibly others. Concerning energy ranges and angular distributions there is still a need for improvements in the range from 6-15 MeV and from 20 MeV to 1 GeV. For protons and other light charged particles existing measurements and, even more, suitable compilations, are still poor; some of the existing libraries and compilations are obsolete, since work has been terminated 15 to 20 years ago. Fortunately, however, cosmogonic and space-research studies have brought considerable impact on various measurements and code validation experiments, primarily for energies up to ~200 MeV, but occasionally also in the region from 600 to 3000 MeV. For neutron and light-ions reactions, suitable nuclear-model codes for energies up to 200 MeV are readily available, and reasonable starter libraries could, in principle, be produced by quick calculations with these codes. The accuracy of such data is, however, not yet well known, and its specification needs particular data testing. This applies at least for the most critical cross sections determiningg the system- or specification-relevant quantities of transmutation concepts. Regarding radiation damage data, such as displacement, H-, He-production and element-transmutation cross sections, very little information above ~20 MeV is presently available from experiments. Data obtained from model calculations are still discrepant with respect to those derived by extrapolations from low energy data. This applies at least for displacement cross sections in typical structural materials of steel alloys.

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