# NUCLIDE PRODUCTION AT INTERMEDIATE ENERGIES

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## Abstract

For any modelling of nuclide production at intermediate energies detailed calculations of the intra- and internuclear cascades are necessary considering reaction modes such as direct, equilibrium and preequilibrium reactions, spallation, fission and fragmentation. Depending on target thickness secondary particles play the dominant role in nuclide production. Reliable calculations of production rates in thick targets today can only be performed by combining calculated spectra of primary and secondary particles with integral thin-target cross sections of the underlying nuclear reactions. The available nuclear reaction models do not satisfy the accuracy needs of applications when predicting production cross sections. Therfore, comprehensive measurements of thin-target cross sections as well as integral thick-target experiments have to be performed to meet the requirements of applications such as accelerator based waste transmutation or energy amplification. Improvements of theoretical models are urgently needed.

## Introduction

Production of residual nuclides by nucleon- and light-nuclei (LN) -induced reactions at intermediate energies (200 MeV/A $\leq$ E $\leq$ 10 GeV/A) is an important issue for a wide range of applications in a variety of scientific and technical fields (1). It is a first priority topic when discussing the feasibility of accelerator based nuclear waste transmutation or of accelerator driven energy amplification systems. Accelerator nuclear waste transmutation (2,3) as well as accelerator driven energy amplification (4) foresee scenarios in which high-current accelerators with energies between 800 and 1600 MeV are used as intense neutron sources. Depending on the design, waste transmutation and energy amplification occur by both the spallation process or by intense low-energy neutron fields.

The feasibility of these concepts depends on a variety of scientific and technical problems, nuclide production at intermediate energies being one of them. Reliable modelling of radioactive and stable nuclides is essential to describe material damage, radioactive inventones of spallation targets, activation of accelerator components, shields and ambient air and, last not least, to estimate the necessary storage times for materials containing transmuted and activated matter. For a survey on scientific and technological aspects see e.g. Fukahori (5), Nakahara (6), Cierjacks (7). The data needs for feasibility studies and development of accelerator-based transmutation systems were discussed recently in some detail by Koning (8) and Mizumoto (9).

## Reaction Mechanisms

The particular features of nuclide production at intermediate energies are closely connected to the phenomena of intra- and internuclear cascades. Due to their high energies the primary particles impinging on a nucleus have wave lengths which are small compared to the distance of nucleons in the target nucleus. As a consequence the initial phase of a reaction will consist of a cascade of nucleon-nucleon interactions occuring on short (10-20 - $10^{-22}$s) time scales.

By this intranuclear cascade of nucleon-nucleon interactions the energy of the primary particle is dissipated to many degrees of freedom involving hadronic and mesonic states. When the emission of fast knock-on particles (p, n, $\pi$, $\mu$, LN) and the parallel dissipation of excitation energy on the remaining constituents of the nucleus have lowered the mean energy of nucleons, a second slow step of the reaction provides further deexcitation of the residual system by statistical equilibrium processes including evaporation of nucleons and LNs, fission and fragmentation.

This scenario, originally proposed by Serber (10), still gives a good overall description of intermediate energy nuclear reactions, though the details are much more complicated with respect to the reaction mechanisms involved as well as to the transition between the two steps which certainly include preequilibrium phenomena.

A necessary consequence of the emission of fast cascade secondaries which themselves have energies high enough to initiate intranuclear cascade processes, the reaction proceeds in extended matter via an internuclear cascade making it necessary to include hadronic and mesonic transport phenomena as having their influences on nuclide production. The hadronic and mesonic cascades are accompanied by leptonic and electromagnetic ones, which, however, are of minor importance for nuclide production except in deep shields.

Because of the differing contributions of primary and secondary particle contributions to nuclide production, it is convenient to distinguish production scenarios according to the target thicknesses in terms of the interaction lengths $\mu$. Targets with a thickness d can be categorized as thin targets (d $< 0.1\mu$), thick targets (O. 1 $\mu \leq$ d c 10 $\mu$) and extended targets (10 $\mu \leq$ d). Taking the approximate nonelastic cross section of protons $\sigma$ =49.9A$^{3}$ mb (1 1), one calculates interaction length $\mu$ of 84., 127. and 196. g/cm$^2$ for O, Fe and Bi, respectively.

Depending on the target thicknesses, primary and secondary particles contribute with different importance to nuclide production. Primary particles dominate in thin targets, but secondary protons, neutrons and LN may already significantly contribute. In thick targets, the primaries contribute non negligibly, but secondary particles, in particular neutrons, make up the bulk of nuclide production. In extended targets secondary neutrons dominate, until at very deep shielding (d >> 10 $\mu$) muon-induced reactions take over.

According to the involvement of secondary particle production and transport, all possible reaction mechanisms occuring from thresholds to the energy of the primaries have to be taken into account. This comprises practically all types of low- and medium-energy nuclear reactions.

## Modelling of Primary and Secondary Particle Fields

Any modelling of primary and secondary particle fields must consider transport and attenuation of primary particles and both, production and transport, of secondary ones. According to the stochastic character of the intra- and internuclear cascades all models (except for some semi-empirical approaches) use Monte Carlo techniques.

There is a number of codes available for these calculations (12- *16),* most of which are descendants of the High Energy Transport (HET) code of Armstrong and Alsmiller (17). The actual versions contain some theoretical refinements as well as inclusion in code systems which allow submission of results to other codes describing different aspects of the intra- and internuclear cascades such as electromagnetic and leptonic cascades and low-energy neutron transport. The quality of the models involved for describing the production of secondary particles was investigated in some detail. For a recent discussion see Filges et al. (18).

These codes can be used to calculate spectra of secondary protons and neutrons for wide varieties of target geometries. Histones of neutrons usually are followed only to 14.7 MeV. At these energies, neutron transport codes such as MORSE (19) and MCNP (20) take over to calculate the further production and transport down to thermal energies. Typical spectra ($E > 1$ MeV) as calculated by HET/MORSE within the HERMES (12) code system for a thick iron target are exemplarily shown in Fig. 1.

The spectra depend on depth and size of the irradiated body, on type and energy of the primaries and on the bulk chemistry of the irradiated specimen. The proton spectra typically consist of a peak of primary particles, determined in energy and shape by electronic stopping and straggling, and a continuum of secondary protons. The latter exhibits a steep increase towards lower energies followed by a broad maximum at about 100 MeV, which is caused by the fact that electronic stopping becomes the dominant process for removal of protons from the spectrum.

In contrast, the spectra of secondary neutrons, starting at the energies of the primary particles, show a continuous increase with decreasing energies. If there is an extended target, this increase continues down to thermal energies. In thin targets no significant thermal neutron fields can be produced since leakage dominates the moderation process and the spectra break down with decreasing energies below a more or less prominent evaporation peak. With increasing thickness of the target low-energy and thermal fluxes built up, strongly influenced by the moderation and absorption capabilities of the target material. Significant thermal neutron fields are to be expected in targets with $d > 3\mu$.

The available codes have, generally, some drawback with respect to secondary particle production. Complex particles such as $^2$H, $^3$H, $^3$He and $^4$He are produced only by evaporation; that means that there is presently no chance for a reliable description of fast secondary LN fields. The importance of these particles in secondary particle production has been demonstrated by Koch (21). For residual nuclide production there are some indications of significant contributions in special cases (22).

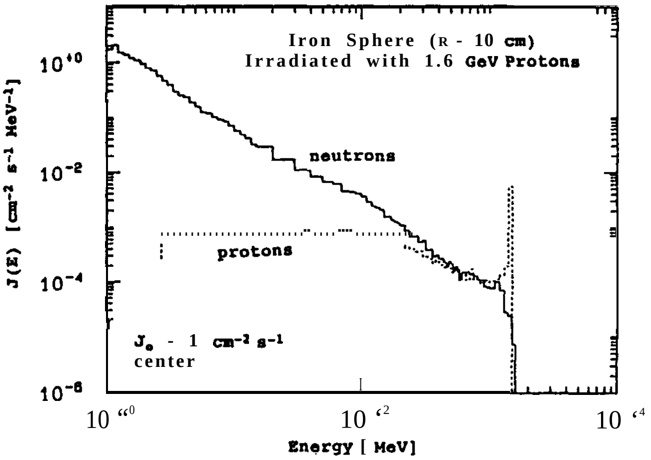

Fig. 1: Spectra of total (primary and secondary) protons and of secondary neutrons in the center of an iron sphere (R = 10 cm) irradiated isotropically with 1.6 GeV protons,

Monte Carlo codes are particularly optimized for calculation of particle spectra though they can also be used to calculate residual nuclide production cross sections. However, the inclusion of transport in calculations of residual nuclide production rates decreases the attainable statistical accuracy, so that the direct calculation of nuclide production in complex systems considering transport and production would require inadequate computing times. Moreover, the capabilities of the models to predict production cross sections are inadequate. Today, only a combination of calculated spectra with experimental and theoretical integral production cross sections allows for a high accuracy modelling of nuclide production.

## Experimental Thin-Target Cross sections

Among all the data needed, integral thin-target cross sections for proton- and neutron-induced reactions from thresholds up to 1.6 GeV are of first priority for a quantitative description of residual nuclide production in the technological application. Cross sections for nuclide production by reactions of secondary light nuclides ($A \leq 4$) are of secondary importance.

During the last three decades intensive investigations of proton-induced reactions have been performed. But the available cross sections are far from being sufficient. The existing data base is incomplete concerning target elements as well as product nuclides and often suffers from severe lack of quality (23). Systematic measurements of production cross sections have been performed by the author and various collaborators. The experiments were performed with accelerators at IKP/KFA Jülich, University Louvain La Neuve, IPN/Orsay, PSI/Villigen, TSL/University of Uppsala, CERN, LANL and LNS (experiments LNS 169, LNS 172, LNS232). These investigations cover now p-induced reactions for energies from thresholds up to 2600

**MeV** for more than 600 **target/product** combinations; (24-32) and references therein.

All these experiments were performed in order to understand the interactions of solar and galactic cosmic ray particles with extraterrestrial matter (33) and the choice of target materials was according to the data needs of **cosmo**physics and -chemistry (23). The target elements investigated were C, N, O, Na, Mg, Al, Si, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Y, **Zr**, Nb, Rh, Ba, and Au. The now available set of excitation functions presently is the largest consistent data set for residual **nuclide** production by proton-induced reactions. Fig. 2 gives a typical example.

As a matter of fact, the data needs for accelerator based waste transmutation and accelerator driven energy amplification are widely the same as for the **modelling** of galactic cosmic ray interactions with terrestrial and **extraterrestrial** matter. The latter were outlined in detail earlier by the author (23). The **cosmochemically** relevant target elements cover those elements which are of importance when calculating activation of accelerator parts, ambient air and concrete shields. However, a number of medium and heavy target elements have to be added to the element coverage in accelerator based energy technology.

For elements, which are not relevant in gee- and cosmophysics, the situation is much worse. Though there are quite a number of measurements to be found in the relevant compilations (8), they are neither comprehensive nor reliable. One of the best investigated cases is the target element Au which is useful only for reasons of nuclear systematic. In order to improve this **situation,** a systematic investigations of other heavy target elements was initiated recently at LNS (experiment LNS 275) for energies between 200 **MeV** and 2.6 **GeV** and other accelerators for lower energies by our collaboration.

With **respect** to **long-term storage** of transmuted waste or of **activated accelerator components** long-lived nuclides such as $^{10}$**Be**, C, $^{26}$**Al**, $^{36}$**Cl**, $^{41}$**Ca**, $^{53}$**Mn**, $^{59}$**Ni**, $^{129}$**I** are essential. Though these **nuclides** are widely irrelevant with respect to committed doses, they are of political concern. In geological repositories some of them could give first evidence of a break-down of retentive barriers. Since these **nuclides** are also important cosmogonic **nuclides,** they were investigated in our earlier work by AMS (26-29). Up to now, detailed excitation functions are available for **cosmochemically** relevant target elements ($Z < 29$) for $^{10}$**Be**, $^{26}$**Al**, $^{36}$**Cl**. Investigations of $^{14}$**C**, $^{41}$**Ca**, $^{53}$Mn, $^{59}$Ni, and $^{129}$I are in progress. **For** all these **nuclides** no data exist for the production from heavy target elements.

For neutron-induced reactions the situation is much worse. In spite of the fact that secondary neutrons are dominating the production of residual **nuclides** in **thick**-targets, there is an extreme lack of experimental data. Most available data are for energies equal to or below 14.7 MeV, just a minority of investigations went up to 30 MeV. Evaluated data files exist only up to 20 MeV, mainly for target elements relevant for fission and fusion reactor technology. Above 30 MeV, there is nearly a complete lack of integral production cross sections, just some measurements being recently reported (34,35).

Therefore, one has to rely mostly on cross sections calculated by nuclear reaction models as e.g. the statistical model according to **Weißkopf** and Ewing (36) in combination with **Blann's** hybrid model of **preequilibrium** reactions (37) or **Intra-Nuclear-Cascade-Evaporation** models as used in HETC ( 17). This emphasizes the importance of the predictive capabilities of model codes. It adds considerable uncertainties to calculations of **nuclide** production at medium energies and makes validations of the **calculational** methods necessary.

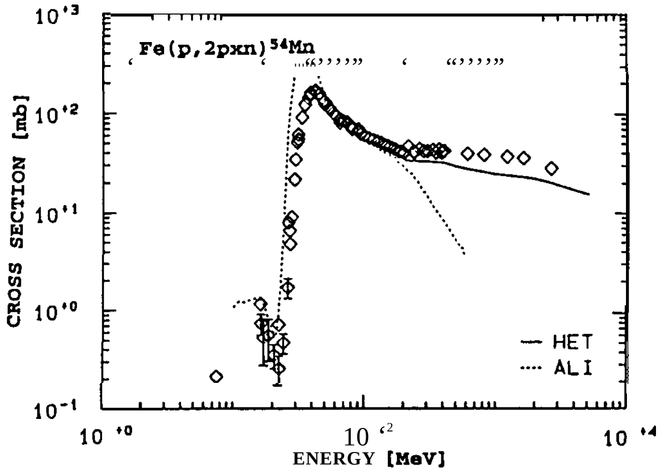

Fig. 2: Cross sections for the reaction **Fe(p,2pxn)$^{54}$Mn** as measured by our collaboration (22,25,30,42). Theoretical excitation functions were calculated by the ALICE (38) and **HET/HERMES** (12,17) codes.

## Theoretical Estimates of Production Cross Sections

For theoretical predictions of production cross sections direct and **preequilibrium** reactions, **spallation,** fragmentation, fission and final reaction steps in statistical equilibrium have to be considered. Presently, there is no model available which covers all these aspects simultaneously. The situation is characterized, on the one hand, by attempts to extend the applicability of codes up to 1 GeV, which were developed for energies below 200. On the other hand, one tries to improve **INC/E** models, originating from medium- and high-energy physics, with respect to the inclusion of further reaction modes (e.g. fission or **preequilibrium** decay) and to extend their applicability to lower energies. For a survey on the existent models and codes see Koning (39) and **Nakahara** (40).

One of the most frequently used "low-energy" models is the hybrid model of **preequilibrium** reactions (37), e.g. in the form of the code ALICE LIVERMORE 82 (41). It has proved to be very efficient for ***a priori*** calculations of production cross sections up to 200 **MeV** (24,25). Recent versions of this code have an extended applicability up to 900 **MeV** (38), though there is no change in the physics considered by the model.

Our own experience from experiment LNS 232 (42) shows, however, that these calculations have a tendency to underestimate significantly the experimental data at energies above 200 MeV, e.g. Fig. 2. The discrepancies become larger with increasing energies. Several **improve**-

ments with respect to the reaction cross sections (43) used and the applied optical model parameters have been made by our group, but no adequate description of the measured excitation functions for energies above 200 MeV could be obtained (Fig. 2). Moreover, the application of ALICE presently is restricted for proton-induced reactions to differences between target and product atomic and mass *numbers* of 7 and 22, respectively.

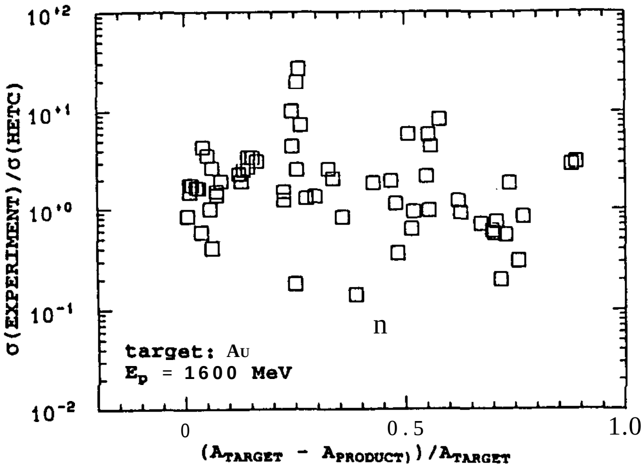

Fig. 3: Ratios of experimental cross sections for the production of residual nuclides from Au at 1.6 GeV from our work (44) and theoretical ones, calculated by the HET code within the HERMES code system.

Therefore, the predictive power of the Monte Carlo codes is of particular importance. We have used the results from our experiments to perform a systematic survey on the capabilities of HET within the HERMES system (12). The results can be summarized as follows. For energies above 200 MeV HET describes the shapes of the excitation functions fairly well, e.g. Fig. 2. In general, however, the absolute magnitude of the cross sections is only reproduced with an average uncertainty of about a factor of two, as is demonstrated for the target element Au in Fig. 3. One draw-back of the code is that fragmentation is not considered, resulting in underestimates of fragmentation products by several orders of magnitude (Fig. 4). This underestimate can be avoided when including this reaction mode in the calculations, as shown by results obtained by the ISABEL code (16) in combination with the statistical multifragmentation model (SMM) (45). Further, preequilibrium decay has to be considered in all codes in order to allow an adequate description of the nuclear phenomena at energies below 200 MeV.

Another “high-energy” approach to describe residual nuclide production is the application of semi-empirical formulas (48-5 1). A detailed analysis of the capabilities of these formulas on the basis of our new data showed, however, that they are by no means sufficient to describe medium- and high-energy cross sections with an accuracy sufficient for application (22,23,44). Also for these formulas uncertainties of a factor of two have to be taken into account, in particular for product nuclides near to the line of stability. A further problem of the semi-empirical formulas is that they assume equal cross sections for proton- and neutron-induced reactions at medium energies, a presumption which must be disgarded on the basis of our experience (22).

In summary, there is presently no model or code is capable to predict residual nuclide production within the accuracy of better than 30 %, which according to Nakahara (6) is needed for the technological application.

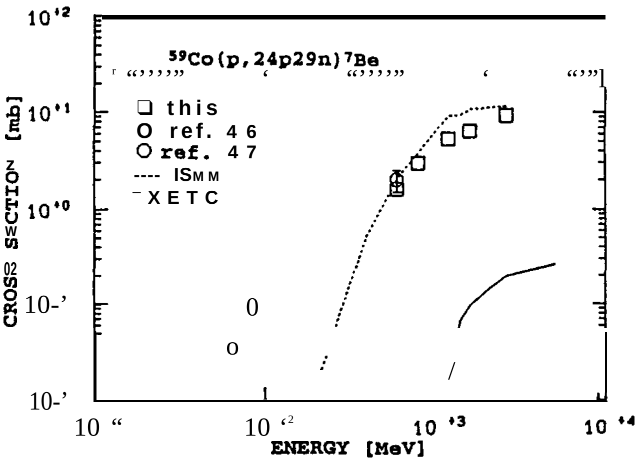

Fig. 4: Cross sections for the production of ‘Be from Co. The experimental data are from our group and from (46,47) Calculations were performed by HET (17) in the HERMES (12) system and by ISABEL (16) in combination with SMM (45).

## Modelling Radionuclide Production in Thick Targets

In spite of the shortcomings of nuclear model codes and of the shortage of thin-target cross sections, there is a way to derive reliable production rates in thick and extended targets irradiated with medium-energy protons by combination of calculated spectra of primary and secondary particle fields with experimental and theoretical integral production cross sections. For such calculations, high-quality thin-target cross sections for proton-induced reactions and realistic thick-target experiments, in which significant secondary particle fields contribute to nuclide production, have to be available.

This method, which also offers solutions for the problem of nuclide production in accelerator based energy systems, has been successfully applied for the modelling of the interactions of galactic protons with meteorites and lunar surface materials. Besides the experimental investigation of the cosmophysically relevant proton-cross sections, during recent years a series of thick-target simulation experiments has been performed (52). In these thick-target experiments for all relevant cosmogonic nuclides production rates were measured, which allow to validate and to improve model calculations of nuclide production.

In an *a priori* approach production rates were calculated by folding the depth dependent particle spectra with thin-target cross sections, using for p-induced reactions our experimental cross sections and for n-induced reactions theoretical ones calculated by various reaction models (38,12). The results of the *a priori* calculations are moderatly good. There are many products for which experiments and *a priori* calculations agree within the limits of experimental errors. Mostly, an agreement within 30 % is obtained, if the excitation functions for the proton-induced production are well known. In some cases, however, the experimental data are underestimated by about a factor of two, a failure which was attributed to a lack of quality in the theoretical neutron cross sections.

A detailed analysis of the experimental depth profiles inside the thick-targets of the siumlation experiments allowed, however, to improve the cross sections of neutron-induced reactions by fitting methods (22,53). Using the improved neutron cross sections, production rates of cosmogonic nuclides in meteoroids and lunar surface materials can be calculated with uncertainties better than 5 %, production rate ratios better than 3'70.

## Conclusion

- Systematic measurements of thin-target cross sections of proton- and light-nuclei-induced reactions are needed for a reliable modelling of nuclide production in technological applications.
- Long-lived nuclides which determine finally the storage times in waste depositories have to be included in the measurements.
- Investigations of radionuclide production by fast (E > 20 MeV) neutrons are urgently needed.
- Integral experiments have to be performed which allow to validate and improve model calculations of radionuclide production.
- Theoretical models and codes have to be improved or newly developed which are capable to predict unknown cross sections at intermediate energies.

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