PRODUCTION CROSS SECTIONS OF RADIONUCLIDES IN PROTON-INDUCED REACTIONS

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Abstract

The use of an intense high-energetic proton beam impinging on sets of different targets is one of the key options for the production of a large variety of exotic beams. Recently performed experiments at GSI in inverse kinematics have shown that the outgoing products of proton-induced fragmentation and high-energy fission reacions cover broad regions of the chart of the nuclides. The calculations of the production cross sections for proton reactions directly with the target were performed with the ABRABLA-code developed at GSI.

1. INTRODUCTION

The production of beams of rare unstable isotopes using their release in high-energy fragmentation and fission reactions is presently discussed within the framework of the proposed future radioactive nuclear-beam facilities.

The physical goals of the experiments with radioactive nuclear beams include investigations on a large set of isotopes ranging from the most-neutron-rich to the neutron-deficient side of the chart of nuclides. The use of an intense high-energetic proton beam impinging on different targets is one of the possible options for the production of the exotic nuclides required by the different experiments. While the advantages of the use of the proton-induced high-energy fission and fragmentation are well known, the complete surveys on isotope production for these reactions are limited to few cases only. Full isotopic production cross sections were measured at GSI Darmstadt by applying the inverse-kinematics technique for the cases of proton-induced reactions using ^{238}U, ^{208}Pb [1], ^{197}Au [2,3], ^{136}Xe, and ^{56}Fe projectiles with projectile energies in the 500 – 1000 A MeV range.

Studies on proton-induced reactions on a larger set of target materials were performed at the ISOLDE facility at CERN [4]. The measurements proved the ability of the ISOLDE facility for the production and extraction of very rare isotopes, for which production cross sections even in the picobarn range are expected. The extracted amount of isotopes depends not only on the primary production cross sections but also on their chemical properties; consequently the extraction is highly selective - in many cases great part of the produced isotopes of certain elements are extracted with small losses while in other cases only a small fraction of the produced nuclides is extracted with high losses. Others could even not be observed.

The study of all possible scenarios on the isotope production can be completed by the application of nuclear reaction codes for the cases where no data exist. The simulation of proton-induced high-energy fission and fragmentation reactions is a rather complex task, since all involved reaction mechanisms over a broad energy range e.g. intranuclear cascade, particle evaporation, have to be considered. The verification of the nuclear-reaction codes is done by comparing their results with known experimental data.

2. EXPERIMENTAL STUDIES ON PROTON-INDUCED REACTIONS WITH THE FRS

The identification of the large amount of different nuclides produced in high-energy fission and fragmentation is a challenge, which has to be met in an experiment on reaction studies and cross sections measurements. Only a very small amount of the outgoing products have long beta-decay half-lives. As a consequence, the fast identification of the reaction products is crucial in order to avoid losses or population of daughter isotopes by subsequent radioactive decays. This requirement of fast identification is difficult to be met in experiments performed in direct kinematics by using a high-energetic proton beam impinged on a fixed target, since a fast release of the residues from the target material is not expected.

In the inverse-kinematics technique for the studies of proton-induced fission and fragmentation reactions, the nuclei used as a target in the experiments performed in direct kinematics are accelerated and impinge on a liquid-hydrogen target.

Due to the high initial velocity of the projectiles (0.88c for 1000 A MeV beam energy), the reaction products from fragmentation as well as from fission reactions are focused in a narrow cone in the beam direction. After leaving the target, the identification of the residues can be done using a high-resolution magnetic spectrometer.

The FRS spectrometer [5] at GSI offers a precise measurement of the magnetic rigidities of the reaction residues. Combined with the velocity measurement by the time-of-flight technique and the charge measurement of the fragments using ionization chambers, the FRS provides the full isotopic identification of the reaction products within less than 100 nsec after the reaction in the target. A scheme of the setup is shown on Fig.1.

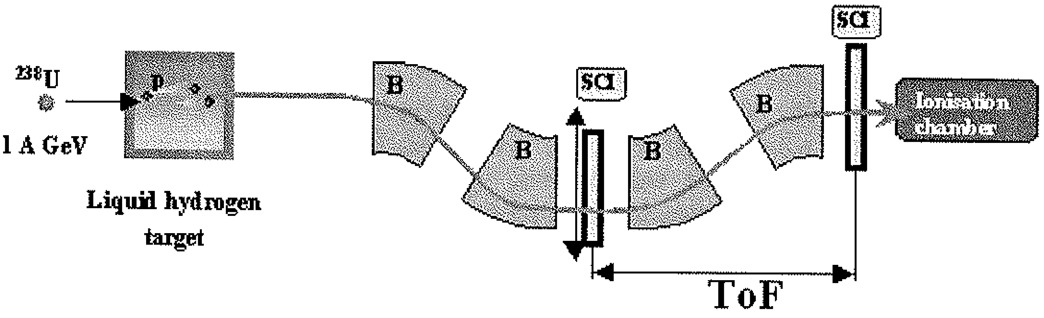


Fig. 1. Setup for reaction studies in inverse kinematics with the FRS specrometer.

Once the residues were identified in mass and charge, a fine measurement of their velocities using the measured magnetic rigidities becomes feasible. In this way, the longitudinal momentum distributions of the residues have been measured with high accuracy. Since the velocity distributions of the outgoing products are different for fission and fragmentation reactions, the FRS allows the determination of the reaction mechanism in which the different isotopes were formed. While the momenta of the fragmentation residues are described by three-dimensional Gaussian distributions, the momenta of the fission fragments populate sphere-like distributions in the three-dimensional momentum space. This difference is clearly observed in the measured velocity distributions shown in Fig. 2. The double-humped structure for the fission fragments results from the limited angular acceptance of the FRS spectrometer combined with their spherical momentum distribution – the fragments are detected only if they have left the target under an angle smaller than the maximum one accepted by the spectrometer, measured relative to the initial beam direction [6]. Based on the measured velocity spectra, the clear separation between the products created in fragmentation and in fission was done. The determination of the amount of residues created in fragmentation or fission processes is an important experimental information for the validation of the reaction simulation codes.

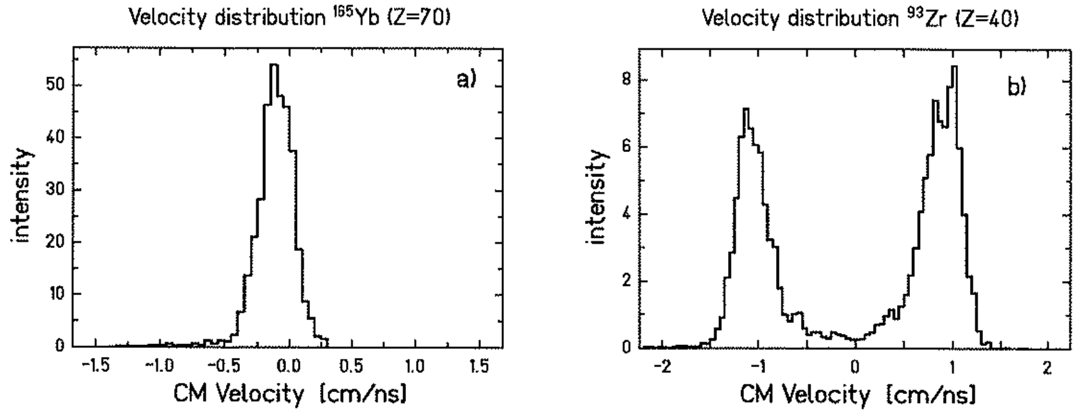


Fig. 2. Measured velocity distributions of residues produced in fragmentation (a) and fission (b) reactions [1].

Up to now the experiments on cross section measurements at the FRS covered different parts of the chart of nuclides, starting from the heaviest projectiles available as ^{238}U, ^{208}Pb, ^{197}Au and continued to the middle heavy projectiles as ^{136}Xe and ^{56}Fe with different targets, covering the high-energy and electromagnetic-induced fission and fragmentation reactions.

The experiments on cross section measurements in electromagnetic fission and proton-induced reactions on ^{238}U performed at GSI are of special importance for future planed ISOL-type facilities since presently two different methods for the production of exotic nuclei based on high- and low-energy fission reactions are discussed. The converter method of production is based on the neutron-induced [7] or electromagnetic fission of ^{238}U [8] from excitation energies around 10 to 30 MeV, while the direct method is using a high-energy proton beam impinging on a ^{238}U-containing target.

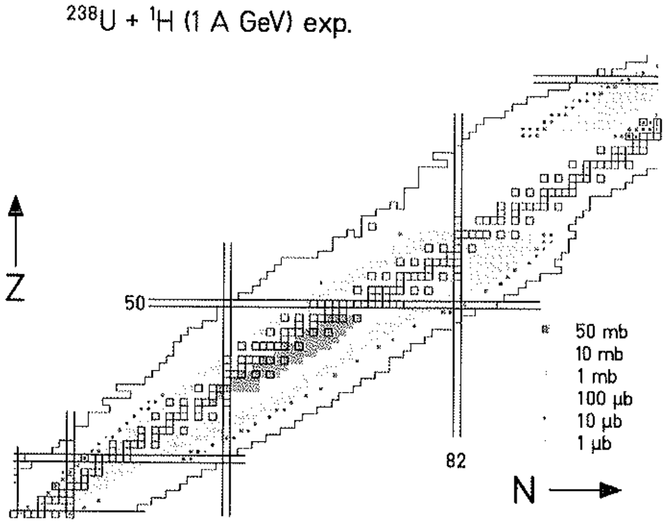
The full isotopic distributions of electromagnetic-induced fission of ^{238}U have been measured at the FRS using a ^{238}U beam, which impinged on a lead target [9]. While previously only the mass distributions of the fission products were known, the experiment on the FRS enabled also the comprehensive measurement of the isotopic cross sections.

While the low-energy fission of uranium has been extensively studied during the last decades, the distributions of products in high-energy fission reactions were poorly known. In the experiments on proton-induced fission of ^{238}U performed at GSI the complete set of production cross sections from those of very light till heavy residues close to the ^{238}U projectile were measured.

The cross section distributions of nuclei produced in high-energy fission show a broad, rather regular pattern, which covers a much wider mass range compared to the distribution of nuclides produced in low-energy fission. While in the low-energy fission case the production of residues is concentrated in two narrow regions around the mass numbers A=100 and A=140 on the neutron-rich side of the chart of the nuclides, the production of nuclides in proton-induced high-energy-fission reactions even extends down to very light residues like the isotopes of potassium. The comparison between the measured distributions of the production cross sections are shown in Fig. 3.

An interesting option for the production of extremely neutron rich isotopes is offered by the two-step reaction mechanism based on fission followed by the cold fragmentation process [10]. After their extraction from the target-ion-source the neutron rich fragments produced in low-energy fission are reaccelerated and impinge on a light target. Extremely neutron-rich isotopes are produced in cold fragmentation reactions, e.g. in the pure proton-removal channels of the fragmentation reaction of the reaccelerated fission fragments with the light target nuclei.

This option is applicable if a postaccelerator, which is capable for delivering beams with energies of E = 100 A MeV and higher is used. The production of neutron-rich isotopes by cold fragmentation is currently part of a dedicated experimental program at the FRS.



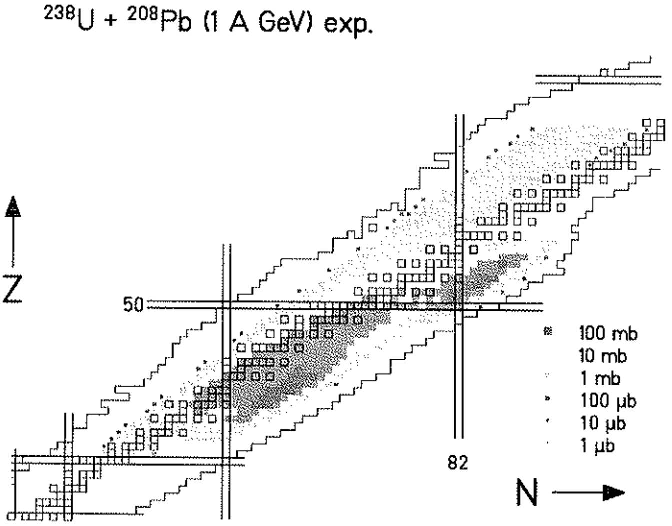


Fig. 3. Measured production cross sections of nuclides in ^{238}U + ^{1}H (1 A GeV) [11] and ^{238}U + ^{208}Pb (1 A GeV) [9] reactions.

3. YIELDS MEASURED AT ISOLDE

Extensive studies on the release and yield measurements of nuclei produced in proton-induced reactions using a large set of different targets and ion-sources have been done at the ISOLDE facility at CERN [4]. As previously described, the yields of isotopes extracted from the ISOLDE target-ion-source do not depend only on the primary production cross sections, but also on the losses caused by the intermediate processes like diffusion and radioactive decays, which take place after the isotopes were produced in the target. The improvements in order to minimize the losses and to make the target-ion-source more efficient are subject of an interdisciplinary research due to the extreme conditions, related to the high temperatures and deposited power, under which the target-ion-source is operated.
The choice of the proper target form, like composite porous block, powder or thin foils, and the ion-source depends on the specific properties of the nuclides to be extracted- e.g. their atomic number, their beta-decay half-lifes, and their production cross sections [12,13].

Using the high-intensity proton-beam, very weakly produced isotopes have been observed. Data measured at FRS in a survey experiment can be renormalized in order to reproduce the ISOLDE yields for the isotopes where both measurements overlap. Following this procedure, the cross section data of nuclides detected in a FRS experiment can be combined with the yields of nuclides, which have been measured at ISOLDE but not observed with the FRS. Using this procedure estimations of the production cross sections even of very weak channels become feasible, an example is shown on Fig. 4 [14].

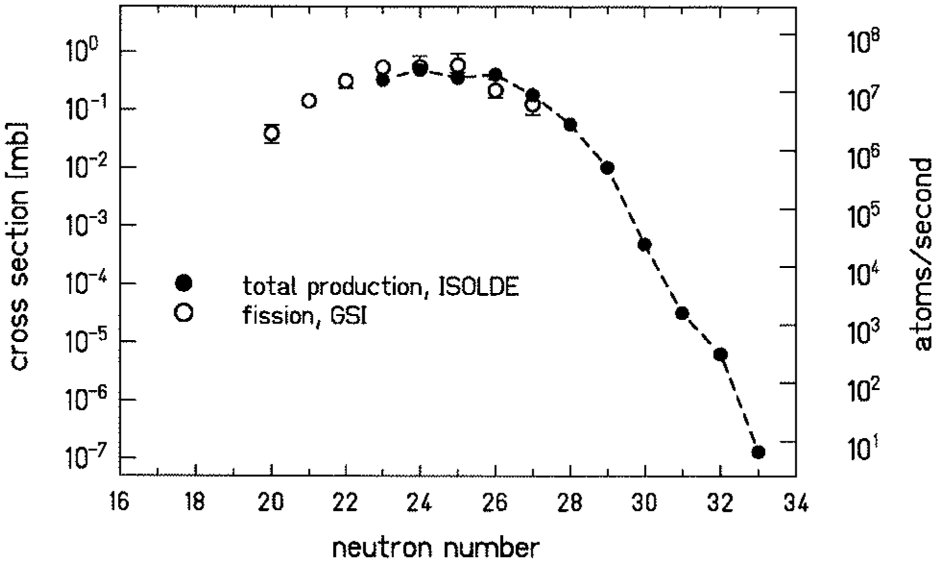


Fig. 4. Production cross sections of potassium isotopes in the reaction $^{238}U + {}^{1}H$ (1 A GeV) compared to the yields measured at ISOLDE.

The production mechanism of isotopes using fissile targets was previously not clear for all cases, e.g. whether light residues are produced by fragmentation or fission. The experiments at the FRS, especially the studies on reaction kinematics have shown, that even very light products down to potassium can be produced by fission [14].

4. THE NUCLEAR REACTION CODE

The calculations of production cross sections for proton-induced reactions have been performed using the ABRABLA code, developed at GSI [15,16,17]. The calculations are based on statistical models of nuclear fission and fragmentation reactions. Fission dynamics is explicitly taken into account by applying a time dependent model of fission probability.

The proton-nucleus collision in the GeV-energy range consists of the initial stage of the abrasion of a few nucleons from the target nucleus. The remaining nucleus, called prefragment, is highly excited, due to the large amount of hole-states and nucleon-nucleon scatterings occurred during the abrasion. For light- to middle-mass-prefragments the following deexcitation proceeds mainly by the emission of protons, neutrons and light clusters that lead to the final nuclei produced in the reaction. The broad distribution of excitation energies which is achievable in the abrasion determines the large number of possible outgoing channels produced in the evaporation stage: the enhanced number of evaporated nucleons from a highly excited prefragment allows the production of light residues even with heavier targets. On the other hand, the isotopic distributions for most elements are shifted to the neutron-deficient side, located between the proton drip-line and the beta-stability line. This feature is also known as the universal evaporation corridor observed in fragmentation reactions of nuclei with low fissility. Since the neutron evaporation is favored in the initial steps of the ablation, the following imbalance in proton and neutron number of the remaining prefragment will equilibrate the emission probabilities for neutrons and protons, which explains the formation of the evaporation corridor stated above.

In proton collisions on heavier nuclei as lead and gold, the introduced excitation energy could be enough for overcoming the fission barrier of the prefragment, so fission will be observed in addition to evaporation. Further increasing the mass of the target nucleus makes fission the main deexcitation channel, since the excitation energy introduced by the abrasion even of a very limited number of nucleons will be enough for overcoming the lower fission barriers of the prefragments in this case.

In the case of proton-induced reactions on fissile nuclei like ^{238}U, fission plays the major role in the formation of the outgoing channels of the reaction. The high excitation energies introduced in the abrasion step of the reaction gradually change the distribution of the fission residues compared to those, known from the low-energy neutron-induced and electromagnetic fission. Fission of prefragments close to ^{238}U with excitation energy of a few MeV above the fission barrier is strongly affected by shell effects. Mostly neutron shells at $N \approx 82$, 88 make asymmetric fission, with fragment distributions peaked around $A \approx 130$ and $A \approx 100$, the most probable deexcitation mechanism. As described by Ignatyuk et al. [18], the influence of the shells will vanish with increasing excitation energy of the fissioning prefragment. In this case, according to the statistical model, fission will proceed preferentially by the symmetric channel, which shows a Gaussian-like distribution of products in mass and charge, peaked at mass symmetry. The assumptions made above are in agreement with data taken in previous experiments performed at GSI; the measured production cross sections were used for confirming and adjusting the parameters included in the ABRABLA code. An important result of the used statistical fission model is the description of the dependence of the widths of the isotopic distributions on the excitation energy of the prefragments. Following the statistical model, one expects, that the variety of produced isotopes should increase as a function of the excitation energy. This result is not surprising, since extremely asymmetric splits in the symmetric-fission-channel would require enough energy to overcome the increased fission barriers in that case.

On the other hand, the abrasion of a large number of nucleons will lead to the production of less fissile prefragments, which will deexcite following the ablation scheme as mentioned above.

In the dynamical description of fission, the evolution of the nuclear shape to the saddle point of the macroscopic nuclear potential, is considered as a collective motion along the nuclear deformation coordinate. The energy of the collective motion is obtained by transfer from the single particle excitation energy in dissipative processes.

An important result of the dynamical treatment is that the fission width, e.g. the probability that fission occurs once the nucleus has been excited, is time-dependent.

Considering the time dependence of the fission probability is necessary since a considerable part from the initial excitation energy can be removed by particle evaporation before the nucleus has arrived at the saddle-point configuration and thus the fission probability is reduced.

In the code the time dependence of the fission width is explicitly taken into account by including an analytical solution of the Fokker-Planck equation with parameters based on the results of previous experiments on cross sections measurements in fission reactions.

5. RESULTS OF THE CALCULATIONS

The calculations of the production cross sections were performed for proton-induced reactions on mid-mass nonfissile target nuclei up to heavy fissile nuclei like ^{232}Th and ^{238}U. The large set of chosen target nuclei enables the theoretical study of the overall behavior of the isotope production as a function of the target mass. The reaction code was verified by comparing the results with the experimental data on production cross sections measured in the reaction ^{238}U (1 A GeV) on proton [11]. The results and a comparison with experimental data are shown in Fig. 5.

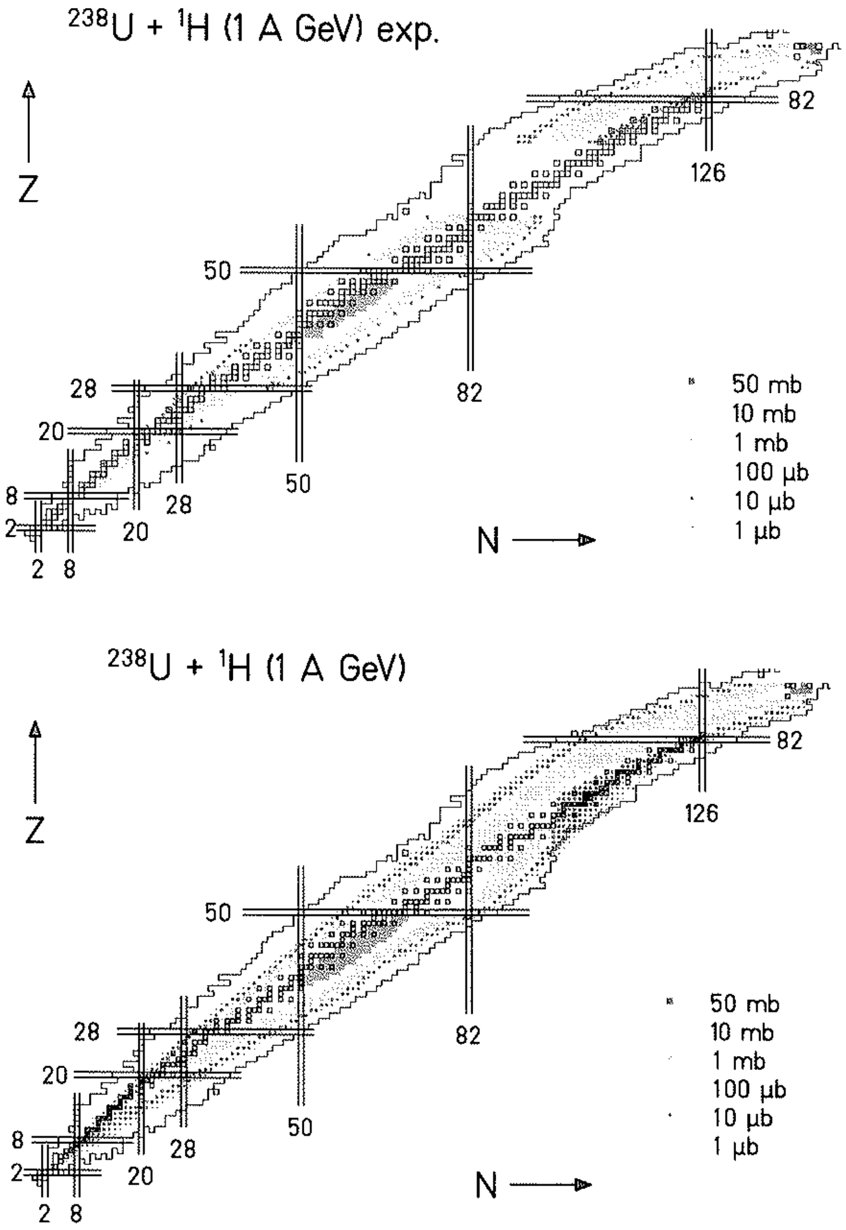


Fig. 5. Experimental and calculated cross sections for the production of nuclides in the reaction ^{238}U (1 A GeV) on proton.

Examples of optimizing the yields of Ni Br and Sn isotopes by choosing different target materials are shown in Fig. 6.

While when using ^{232}Th targets the production of neutron-rich nuclei is preferential, the outgoing channels of the fragmentation reactions on Sr, Nb and La targets consist of mostly neutron-deficient nuclei. Comparing the cross sections of Ni isotopes produced with Nb and Sr targets, the smooth variation of the production cross sections becomes visible: slight changes of the mass and the atomic number of the target nuclei introduce small changes of the production cross sections of a certain isotope. A smooth variation is expected also for the production cross section distribution of nuclides produced with a certain target.

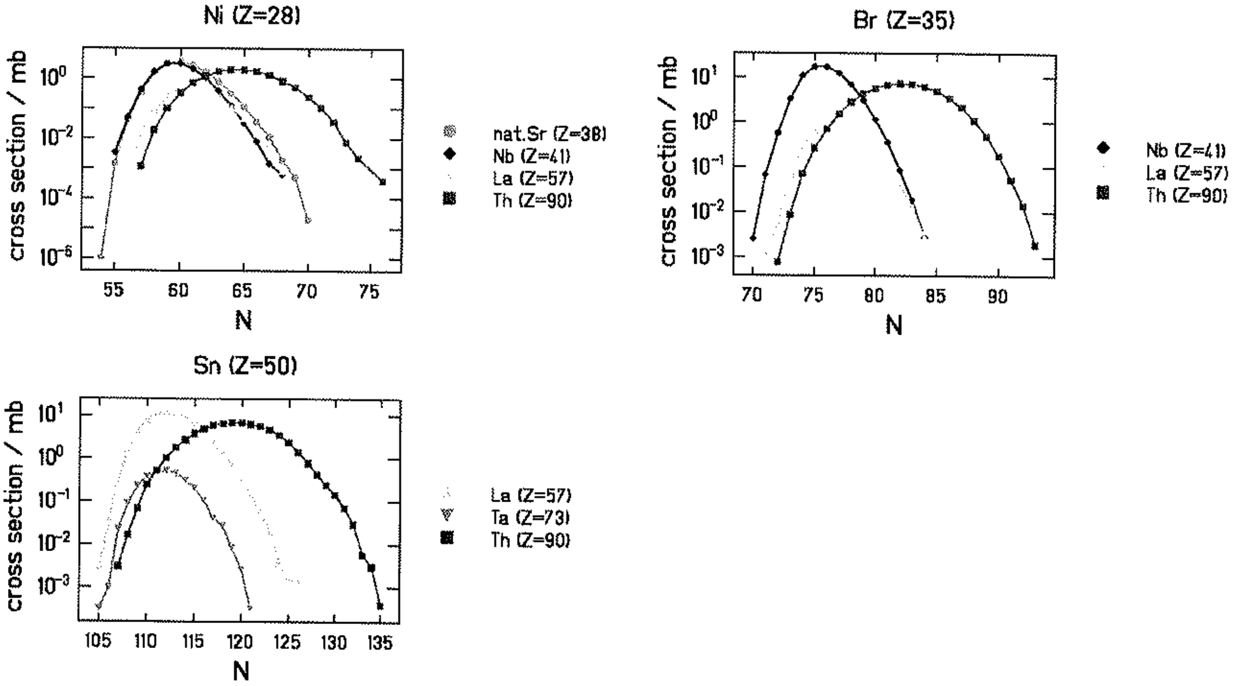


Fig. 6. Calculated production cross sections of Ni, Br and Sn isotopes in reactions of 1 GeV protons with Sr, Nb, La and Th targets.

A similar regular behavior of the yields of nuclei produced using the ISOL technique is not observed. Measurements have shown, that depending on the target composition the yields of isotopes of certain elements can be strongly reduced, or they are not extracted even if they are produced in the target as shown on Fig. 7. The selectivity of the ISOL technique with respect to the atomic number of the extracted isotopes can be explained by element-dependent conditions for the diffusion process out of the target. The ion-source can be constructed in such a way, that it provides high element selectivity, what is crucial for the efficient separation of the outgoing products and for avoiding pollutants in the extracted secondary beam. Since the time needed for the extraction of the produced nuclei out of the target can be comparable or even exceeds their beta-decay half-lives, decay losses are also expected.

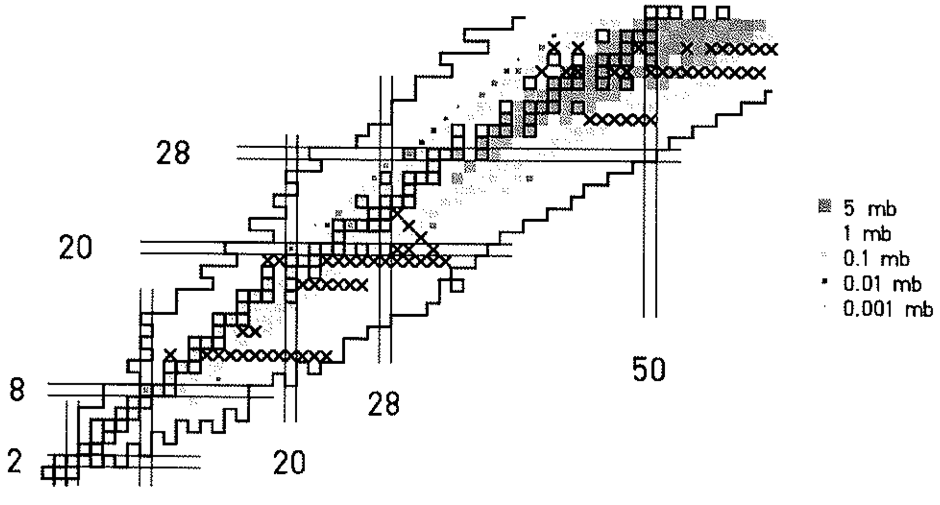


Fig. 7. Measured production cross sections of light fission residues in the reaction $^{238}U + ^{1}H$ (1 A GeV) [14]. The crosses indicate nuclides produced at ISOLDE [4].

Based on the theoretically calculated production cross sections the estimation of the primary production rates of nuclei becomes feasible. The primary production rates can be compared to the measured ISOLDE yields by calculating the release-to-production ratio (RPR) as defined in eq. (1) as the ratio of the measured yield of a certain isotope to the expected yield using the calculated production cross section.

$$RPR_{(N,Z)} = \frac{Y^{ISOLDE}_{(N,Z)}}{Y^{Calc}_{(N,Z)}} \qquad (1)$$

One would expect that the RPR value does not exceed 1. On Fig. 8, the release-to-production ratios of Br isotopes produced in the ISOLDE facility using the 600 MeV proton beam are shown. After comparing the release-to-production ratios with the beta-decay half-lives of the Br isotopes, the decrease of the release-to-production ratios due to beta-decays becomes clearly visible- the RPR values are systematically higher for nuclei with longer halflives, on the other hand, the RPR is lowered for nuclides with shorter halflives.

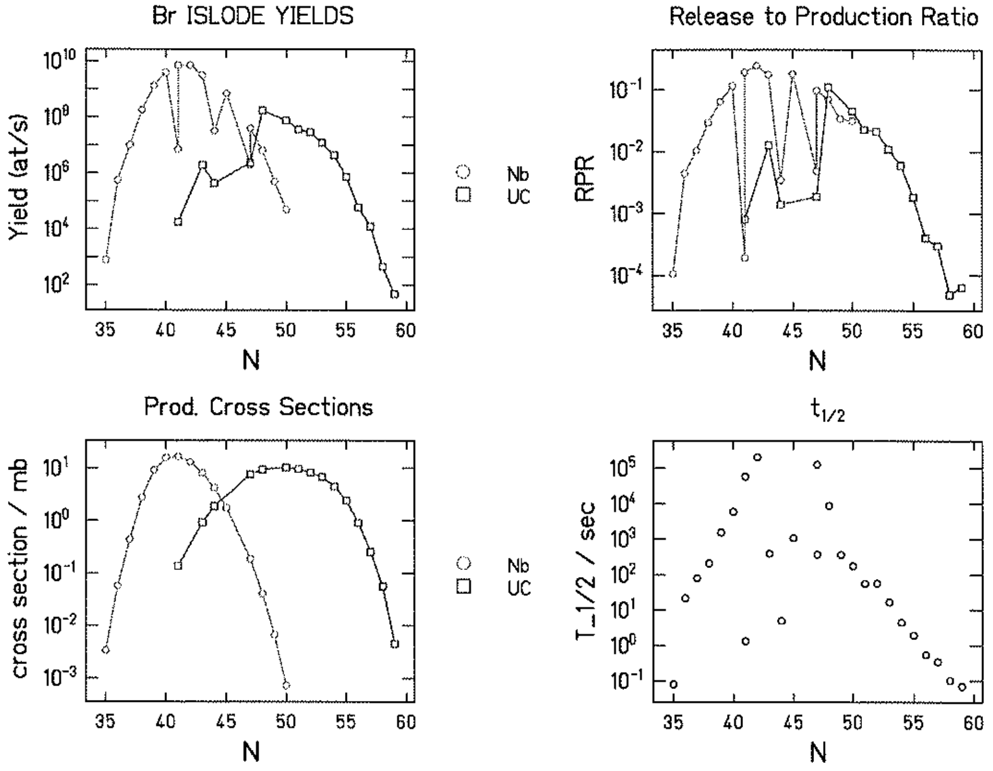


Fig. 8. Yields of Br isotopes measured at ISOLDE using Nb and UC targets [4] compared to calculated production cross sections, release to production ratios and beta-decay half-lifes.

If the beta-decay half-lives become long enough compared to the time needed for diffusion and extraction, the reduction of the yields due to radioactive decays should become negligible. The development of well suited models for the prediction of losses introduced by the different processes, which take place in the target-ion-source is a rather complex task since the evolution in time of the diffusion, the efficiency of the ion-source but also the geometry and material properties of the target have to be considered in the calculations. Since the RPR-values can be interpret as a normalization of the measured yields with respect to the primary production cross sections, they depend only on the properties of the extracted isotopes and their interplay with the used target-ion-source, but not on the mechanism of the reaction in which they have been created.

So far the RPR-values can be used in design studies, optimization procedures and estimations of the maximum achievable intensities of the extracted nuclides.

6. CONCLUSION

The proton-induced reactions are a promising option for the production of a large variety of radionuclides. The experimental data on the production cross sections of radionuclides in proton-induced fission and fragmentation have been collected in experiments at GSI, performed in inverse kinematics. The FRS spectrometer provided the important information on reaction kinematics, which gave the possibility for the clear determination of the reaction mechanisms in which the nuclides are produced. The reaction simulation code ABRABLA, developed at GSI, gives the possibility for the calculation of production cross sections of nuclides in reactions for which no data exist. The code is based on statistical models of fission and fragmentation reactions and on the results of the GSI experiments on fission and fragmentation reactions. Combining the results of the simulations with the measured yields at already existing facilities like ISOLDE at CERN, the estimation of the overall efficiencies of the nowadays used target-ion-sources respecting the extracted amount of nuclides becomes feasible. The access to the nuclides produced in fission and fragmentation reactions for the production of radioactive beams depend largely on the further improvements of the target-ion-source technologies.

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