CHARACTERISATION OF DIFFERENT TARGET – ION SOURCE UNITS FOR THE PRODUCTION OF RADIOACTIVE ION BEAMS BY THE ISOL METHOD

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Articolul descrie în amănunțime una dintre metodele cel mai folosite pentru studierea proprietăților diferitelor combinații dintre ținta fisionabilă și sursa de ioni folosite în producerea de fascicule radioactive prin metoda ISOL. Sunt detaliate tehnicile folosite pentru măsurarea randamentelor de producție pentru izotopii diferitelor elemente chimice, precum și a proprietăților ansamblului țintă – sursă de ioni din punct de vedere al rapidității de emisie a acestora.

The paper describes in detail one of the methods often used to investigate the properties of various fission target – ion source assemblies for the radioactive ion beams production by the ISOL method. The techniques used for the yield measurements of the isotopes of various chemical elements, as well as for the release time properties of the target – ion source assembly measurements, are detailed.

Keywords: radioactive ion beams; nuclear spectroscopy.

1. Introduction

Physics of radioactive nuclear beams is one of the main frontiers of the nuclear science today. The opportunities offered by beams of exotic nuclei for research in fields as diverse as nuclear structure, nuclear reactions, astrophysics, atomic, material and applied science, are both timely and exciting. The enormous worldwide activity in the construction of different types of radioactive beam facilities reflects the strong scientific interest in the physics that can be probed with such beams. The realization of new accelerator complexes also offers important technical challenges.

Radioactive nuclear beams (RNBs) provide many new directions and means for scientific research and practical applications. These require high-intensity and high-quality beams over wide energy and isotope ranges. Study of reactions relevant to nucleosynthesis need energies below 1 A·MeV, while investigation of nuclear density distributions requires beam energies of several hundred A·MeV or higher. Nuclei at the limit of existence (i.e. near the drip lines)

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have expected half-lives in the millisecond range, so that their fast production and delivering have to be carried out. At the present time, there is no single method for the production of RNBs that fulfills all these requirements. Instead, there are two methods which can be used: *i)* the Isotope Separation On Line or ISOL method, in which a production accelerator or a nuclear reactor yields a production beam of charged particles or neutrons, which is sent on a thick target, the radioactive species thereby produced are transported by a transfer tube to an ion source, and the resulting ions are separated by an isotope/isobar separator, postaccelerated and sent to the experimental area, and *ii)* the In-flight method, sometimes called the Fragmentation method, in which a primary stable heavy-ion beam, produced by a heavy-ion accelerator, is broken into fragments in a thin production target, then a fragment separator selects the radioactive fragments of interest and sends them to the experimental area. The ISOL method provides high-quality beams from low up to, in principle, high energies. However, it has a limitation for the acceleration of short lived isotopes due to the finite release time of radioactive nuclei from the production target and transfer time to the ion source, the present practical limit being of the order of 10 to 100 milliseconds. The In-flight method provides the fastest separation time, of the order of 100 nanoseconds, i.e. the flight time of the radioactive nuclei in the fragment separator. Therefore, not only drip-line nuclei but also many isomers can be produced by this method. However, the quality of the beams is limited, and in particular a low-energy beam of high quality is difficult to obtain at all. This problem can be circumvented if one applies accumulation and cooling techniques, but the cooling process takes time, thereby limiting the usable lifetime above about one second with the present techniques. Therefore, at the present moment, one needs the two types of facilities as complementary means to produce RNBs, and further developments in both need continuous efforts.

2. The ISOL method for the production of radioactive nuclear beams

ISOL is the historically first developed method for making radioactive beams. It makes use of spallation, fission and fragmentation reactions in thick targets driven by light particles from a first accelerator or a nuclear reactor. The radioactivity produced is brought to rest in the target and then has to be separated and transformed into an ion beam in order to be post-accelerated in a second machine. This method has in the past 40 years been used successfully at many on-line mass separators to produce low-energy radioactive ion beams. Techniques for the transfer of the nuclear reaction products into an ion beam have been optimized with respect to the individual physical and chemical properties of 70% of the chemical elements. In Figure 1 the major ingredients of an ISOL – RIB facility are shown. It consists of four closely matched parts: the nuclear target, the transfer

line, the ion source, and the mass-separator injector system plus the post-accelerator. Of these, the first three, which often are integrated in a target and ion-source unit, play the crucial role.

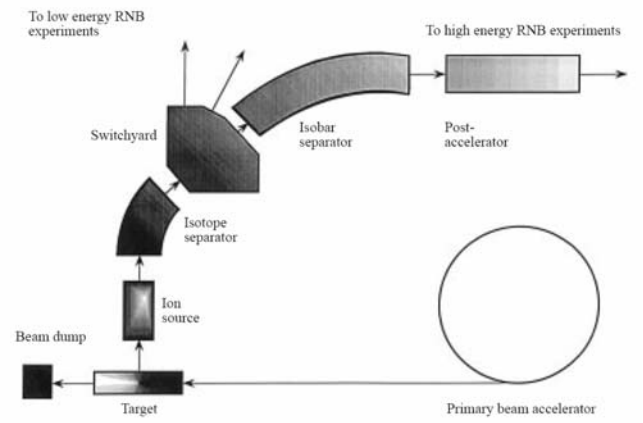

Fig. 1: Schematic drawing of an ISOL-type RIB facility.

The radioactive nuclei formed in the thick target are brought to rest and then have to be separated from the bulk and converted into an ion beam. This operation can be seen as composed from three distinct processes, each of them having its own efficiency: ε_1 – high-temperature thermal diffusion to, and release from, the target surface and transfer by diffusion/effusion through the transfer line from the target to the ion source; ε_2 – ionization; and ε_3 – decay during the entire process. It was quickly realized that these three efficiencies play a more important role in determining the resulting secondary-beam intensities than the usual three factors (σ – the cross section of the production reaction, Φ – the primary beam intensity, N – the thickness of the production target) which determine only the production rate in the target. In fact, the efficiencies that can be obtained for a given product element strongly depend on the properties of the refractory target materials, the primary beam and its time structure, and the type of ion sources which could be adapted to the environment of a particular driver beam. The basic parameters that determine the efficiencies of these new radiochemical separation methods, which for a given product element take place in the target and ion-source unit, are the temperature, diffusion constant, desorption enthalpy and ionization potential [1]. In order to properly choose the target material and the construction materials, it is essential to know the values of ε_1, ε_2 and ε_3. They can be rarely calculated with sufficient precision for a given nuclide. The values of ε_1 and ε_2 can be determined off-line or on-line with various methods to a good precision. The determination of ε_3, whose value depends on the half-life, can only be done by an on-line determination of the delay function, i.e. the probability that an atom formed at time 0 is extracted from the ion source at time T [2].

Depending on the element, values for ε_1 and ε_2 in the range 10 – 90% are not unusual, while ε_3 have values smaller than 10^{-3} for half-lives of the order of milliseconds, increasing with increasing half-life up to about 1 for half-lives greater than 100 seconds.

Another important parameter is the lifetime of the target and ion-source unit. The current techniques allow operation of these units for periods from a few days up to several months before they have to be replaced. During this period sintering, migration or chemical dissociation of the target material, in conjunction with the deleterious effects of the driver beam on the surrounding mechanical structures, eventually causes a failure. For this reason much effort has been put into finding the most economical ways to produce such consumable units.

During the last years at the existing ISOL facilities there has been a steady trend of developing fast uranium carbide targets for on-line production of rare nuclides. Growing interest in the development of uranium carbide targets has been stimulated by the study of exotic neutron-rich nuclei far from stability. In addition, these targets enable the production of a wide region of neutron-deficient heavy nuclei by means of fragmentation reactions with energetic charged particles.

Presently, an increasing demand for targets containing large amounts of ^{238}U has been additionally enhanced by the new projects concerning ISOL facilities of the next generation [3-5], where two-step reactions [6] will be used to produce intense neutron-rich ion beams. As the driver beams of these next-generation ISOL facilities will be about two to three orders of magnitude more intense than those at the presently working installations, a serious problem arises for the replacement of the highly radioactive target assembly after its possible breakdown. Therefore the new target units must be able to work efficiently for several months without failures. It is very important to know whether the target keeps its working characteristics (yields and release efficiency for the produced species) during a long period or not. Also, for a more efficient use of the production beam, the density of the target material containing ^{238}U should be as high as possible.

3. Methods for characterization of target – ion source units

The observables that establish the performance of a target + ion-source unit are the efficiency of separation and the release time of the products. Efficiency is defined as the ratio of the number of ions leaving the source per time unit to the rate of corresponding nuclei created in the target. Release is described by a distribution; in a loose way, the release time is the time span during which a pre-defined fraction of the stable atoms created at time $t = 0$ in the target have left it. In several models this distribution can be characterized by a single constant, but

actually it can be different. It is a problem to find a set of parameters to accurately reproduce this distribution.

The yields and the delay times, in function of target temperatures, obtained in testing different target – ion source units, have to be reported, to demonstrate the reproducibility of the relevant values. The methods used for the experimental measurements do not depend on the production method (fission induced by the primary beam of charged particles or by the fast secondary neutrons from a converter). The atoms of the produced radioactive nuclei are thermally released from the target, ionized in the ion-source, mass-separated and then implanted on a movable tape. Identification of the mass-separated nuclides and counting of their activities are performed by standard decay spectroscopy techniques. A tape transport system, working in the "start-stop" mode, transports the collected activity in front of α, β and γ-radiation detectors. Yield measurements are carried out with constant primary beam intensity to allow for equilibrium to be reached in the target. In contrast, delays between production and delivery onto the tape (release times) are obtained from the analysis of the so-called release curves, i.e. by collecting the radioactive beam during a series of short time intervals after switching off the primary beam and bringing the collected sources one-by-one in front of the detector. Gamma-spectroscopy is normally used to calculate the number of incoming ions. In addition, beta-counting is used for the shortest-lived activities, too weakly produced to be studied by γ-spectroscopy. In this case, the detector is placed close to the implantation station.

3a. Yield measurements.

Yields are number of ions delivered per time unit, and in fact usually they mean the ion current collected and available for experiments. The yield of the target and ion source is attenuated by the efficiency for beam transport in the separator magnet and further beam line. This factor is absorbed in the overall efficiency. For ease of comparison between different target and ion-source units, all yield values should be corrected for the transmission in the beam transport system, thus representing the yield at the exit of the ion source. Also, very often one use the so-called normalized yields, i.e. scaled for a certain target thickness (1 g/cm^2) and for a certain intensity of the incident primary beam (0.1 μA).

The yield is measured in a steady state when the accelerator beam has been on target for a time long enough to reach saturation (output of the target is constant). The requirement that the accelerator current and the performance of the target + ion source must be stable during the measurement cycles is normally satisfied since the measurement can be carried out within few minutes, at least for the strongly produced activities. The ions are collected on a tape during the collection time t_c, then transported with the tape (t_t) and counted by γ (or sometimes α) spectroscopy during another time interval equal to t_c, during which a new sample is collected. The transport time t_t could be of the order of 1-2 seconds,

but it should be as short as possible (even below 1 s) allowing this way the short-lived species to be counted more efficiently. The alternative method is to count directly during collection, in which case one can access very short-lived activities (half-lives of a few milliseconds).

In the most simple case the nucleus of interest has no isomer and its β-decay parent is short-lived (the opposite is very rare). Decays in the target will reach the saturation, so that one measures a current corresponding to the sum of productions cumulated over all the more neutron-rich isobars. There is also a chance that the mother nucleus shall escape from the target before its beta decay, which reduces the extra contribution. This complicates the evaluation of the number of nuclei in the target and, consequently, of the efficiency.

- *yields by gamma spectroscopy*

In the simple and, fortunately most common, case of a nucleus having no isomer, the simple formula found in many papers can be used:

$$A = \varepsilon(\gamma) \cdot b(\gamma) \cdot p \cdot f(\lambda, t_c, t_t) \tag{1}$$

with A the peak area for a γ-ray, $\varepsilon(\gamma)$ the efficiency to detect the peak associated with the gamma ray, $b(\gamma)$ the probability of the γ-ray to be emitted in a decay of the nucleus (also called branching), p the production rate of nuclei, i.e. the ion current to the tape, and

$$f(\lambda, t_c, t_t) = \frac{1}{\lambda}\left(1 - e^{-\lambda t_c}\right)^2 e^{-\lambda t_t} \tag{2}$$

a function dictated by the time constants of the measurement cycle and the half-life.

Usually for the γ-ray detection high purity Ge detectors are used, having the front face placed close to the tape in the off-beam position The energy range covered is typically from 100 keV to about 2 MeV. The peak efficiency depends on the energy of the detected radiation, on the Ge crystal size and on the detection geometry. It should be carefully determined before the measurements using calibration sources of known activity. For example, a typical value for the 1.3 MeV energy is $\varepsilon = 1.2\%$.

Nuclear decay lifetimes are usually well known and their errors even if fairly large have little impact on the final error. Gamma-ray branchings (probability of a γ-ray to be emitted per decay) are supposed to be well known quantities tabulated in compilations or public databases, as, for example, [7]. Unfortunately, for exotic nuclei they become less trustable or even are not quoted in literature. Yield measurements are in fact very sensitive to wrong γ-branchings and it happens to be severe mistakes even for nuclei quite close to stability. For instance, the normalization for branchings of γ-rays in ^{92}Rb decay listed in [7] is too high by an order of magnitude, as it was demonstrated in [8]. There is a suspicion that many other values could be possibly wrong by factors of 2-3 times, this being an issue to be investigated in future. It is especially of interest in

connection with the seemingly odd-even effects in the population of isotopes of a certain element and indirect measurements of release times via efficiency. The branching problem does not exist with β-counting, except for the very exotic nuclei emitting β-delayed neutrons (which seldom have a large neutron branching) or the later mentioned isomers causing electron transitions. The problem in these measurements is a lack of selectivity, but progress has been made recently.

Yield measurements are not always as simple as depicted above. In case the nucleus of interest has an isomer the γ-rays usually cannot be classified as belonging to one or the other decay uniquely. Instead they are populated with different probabilities in each decay mode. One also needs to consider that during collection and measurement the isomer may decay to the ground state. Observed peak areas are a superposition of 2 contributions. Accordingly, at least two γ-peaks are needed to solve a system of 2 equations with 2 unknown production rates, but it is recommended to use more peaks and apply the χ^2 method. This method, recently implemented [9], naturally contains the case without isomer, in which case the solution of the matrix equation reduces to the simple formula for the function *f*. The calculation should be expanded to a ground state and 2 isomers, leading to a 3 by 3 system. The occurrence of 2 isomers in a nucleus is quite rare, but some indium isotopes must be treated this way and there are many other cases near Sn isotopes of special physical interest for RIB facilities. The solution for dimension up to 3 has been worked out and it is probably not necessary to go beyond since a higher-dimension problem can be decoupled in sub-systems of smaller dimension owing to decay selection rules.

- *yields by beta counting*

For the most exotic isotopes the γ-measurement fails due to possibly unknown γ-branchings and/or the low efficiency of Ge detectors, and the decreasing peak-to-background ratio. The alternative is beta counting. The β-branching is 100% in absence of isomers, so this avoids the problems caused by the often inaccurate γ-ray branching. Usually one uses a Si(Li) detector placed next to the collection point to avoid the delay due to transport. Its intrinsic efficiency for the decays of nuclei of interest which have β end-point energies of several MeV is close to 100%, independently of the details of the decay scheme. There is also a geometric efficiency in the measurement position, of about 20%, representing the fraction of the 4π solid angle covered by the detector. Unfortunately, the energy signals of continuous β spectra do not contain a direct signature of the decaying nuclei. The detector triggers also on β-particles from daughter decays (isobars and, although weakly, of A-1 daughters if there is β-delayed neutron emission) as well as occasionally from unwanted molecular ions and long-lived activities being daughters of nuclei collected beside the tape during

previous measurements. The measurement method in this case is accordingly modified with respect to the previous one. After a short beam pulse the decay of the collected activity is followed as function of time without moving the tape. The different half-lives of the collected species allow to extract the number of beta-particles of interest that have decayed and to compute back their production rate. The tape is moved only at the end of the cycle to remove long-lived activities and to keep the background low. The β method is obviously best suited when the half-lives of isobars are very different.

3b. Release time measurements

The release function represents the probability that an atom created at time $t = 0$ shall leave the target at time t. One identifies t with the time of arrival of the ion on the collecting tape since flight through the separator magnet and beam line is fast on the scale of release processes. The release function would be the observable only in the ideal case in which a short beam pulse of sufficient intensity (δ-function like) would be available and the huge rate at the beginning of counting could be processed by the acquisition system. In practice, in order to get better counting statistics one performs an irradiation, followed by a decay period without accelerator primary beam. This integrates the release function, which becomes the so-called release curve. It is safer to concentrate the analysis on the decay part, since beam fluctuation could affect the measurement during the irradiation. The irradiation is normally chosen long enough to reach saturation of the ion current. This simplifies the calculations and decreases the impact of the beam fluctuations.

The diffusion is the process of migration inside the target grain (which could have diameters from 20 μm to 200 μm), while effusion is the process of moving and sticking with the target material and the target container inner surface during the random walk of the atoms in the space between grains and in the target container volume towards the ion source. Each of these two processes is described by a characteristic function: *D(t)*, and *E(t)*, respectively. For sequential diffusion and effusion the release function is a convolution:

$$R(t) = \int_0^t D(t')E(t-t')dt' \tag{3}$$

Indeed, *D(t')dt'* atoms produced at *t=0* cross the boundary of the grain at *t'* and have a time *(t-t')* left for effusion. Detailed theoretical treatments of these processes exist in literature, see for instance [10, 11].

The ion current impinging on the tape represents the flux except for a scaling factor. It is measured indirectly by γ or β spectroscopy. Measurement cycles as described for yield measurements are performed, but with a much shorter collection time t_c and with a different beam structure. Here, short means that t_c is much less than lifetime and release time constants to be measured, which ensures that the ion current can be regarded as a constant during collection. A

number of these short "tape cycles" is recorded with accelerator primary beam on, then is followed by other "tape cycles" without beam. Together they form a measurement cycle that is repeated until enough statistics is collected. In order to reach saturation, the irradiation must have been long enough, i.e. has been a few times the time constant roughly defined by the sum $\lambda + \mu_R$ with μ_R a rough estimate of the unknown release constant.

The mechanical transport prevents the use of the method if the relevant times are much under the second. However, there are many advantages in this method. The activity curve measured and displayed during the acquisition very closely follows the ion current modulated by the decay factor $e^{-\lambda t}$. It can be interpreted during the measurement and, if needed, the settings of the cycle can be modified for better sensitivity. The counting rate remains low owing to the short collection time. Long-lived nuclei can be used to scan a long range of the release time axis without overloading the acquisition system.

An alternative method consists of collecting nuclei on a standing tape during a cycle with an irradiation and decay part. It allows to measure curves for very short-lived nuclei. This is an advantage since the more exotic isobars have low production cross-sections and do not perturb the activity curve too much. A drawback is that the counts represent the integral of the disintegrations of collected nuclei, i.e. are the fold of the integral of the ion current with radioactivity. The release constants are determined by fits based on the deviation of the activity curve with respect to the simple behavior resulting from the nuclear lifetime. This is far less interactive and transparent. Another limitation is that the use of short-lived activities prevents access to the long tails of the release functions, which means that only the fast part of the total function is observable.

4. Examples of practical applications

The above described methods are largely used in many experiments aiming to investigate the best choices for the design of the next generation ISOL-type radioactive beam facilities. They can give answers concerning the comparison between proton- and neutron-induced fission in uranium carbide targets, the most suitable target material (from the point of view of density, grain size, manufacturing methods), the behavior of big mass targets compared with the small mass targets previously used, as well as the influence of different types of ion sources on the emissive performances. Intensive R&D has been done, and is still in progress, in many laboratories around the world. In the following, experimental data from some experiments performed in collaborations by the author at various research facilities will try to illustrate the applicability of the methods for the above mentioned purposes.

- studying the fission induced by secondary neutrons

An experiment at IPN Orsay (France) was performed [12,13] to investigate the radioactive ions production following the fission induced by the secondary neutrons produced in the interaction of a primary deuteron beam with a ^{12}C target. The fast neutron flux inducing the fission of ^{238}U is generated by the break-up of a 1 μA deuteron beam in a 3 mm thick graphite converter in direct contact with $^{238}UC_x$ target. The neutron mean energy was 10 MeV with an energy spread of about 10 MeV.

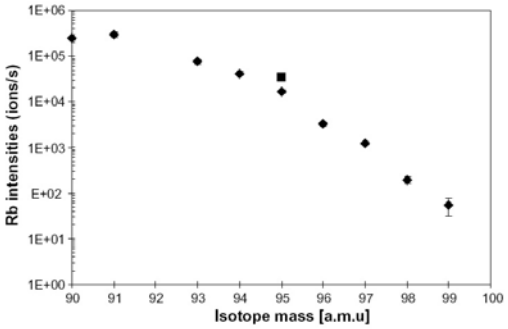

Fig. 2: Yields of neutron-rich Rb isotopes produced with a $^{238}UC_x$/graphite target and a W ionizer at about 2050 °C. The square spot is a measurement of ^{95}Rb carried out with the target at 2200 °C and the ionizer at 2500 °C

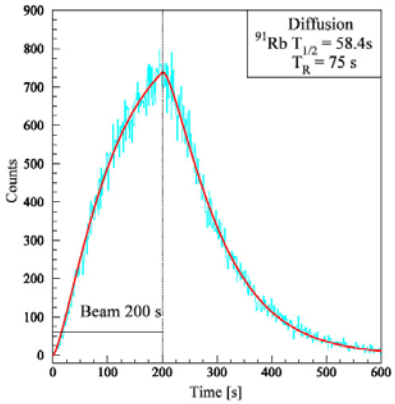

Fig. 3: Fit of the experimental release curve of ^{91}Rb obtained with the lowest χ^2 considering the release process being fully controlled by the diffusion process.

In the same experiment, a release time measurement was performed for the same chemical element, being measured for the ^{91}Rb isotope the experimental release curve presented in Fig. 3.

- studying the fission induced by protons and comparison with that induced by neutrons

An experiment performed at PNPI Gatchina (Russia), where the radioactive nuclei are mainly produced by proton-induced fission, was intended to compare the yields of radioactive nuclei produced this way with those obtained by the neutron-induced fission. The experimental details can be found in ref. [14]. The target material consisted of High Density Power (HDP) uranium carbide, obtained from the fragmentation of High Density Rod (HDR) uranium carbide (UC_2) having a density of 11.2 g/cm^3. The HDP target had a volume of 1.4 cm^3 (8 mm diameter, 28 mm length) and contained about 10 grams of uranium with an effective density of 7.36 g/cm^3 (corresponding to a target thickness of about 20 g/cm^2) and the average grain size of the target material was about 10-20 μm. The target material was introduced into a graphite container, which was housed inside an external tungsten container. The ion source employed was a typical surface-ionization one, made in tungsten. Both target and ion source were operated in the temperature interval of 2100 – 2400 °C by ohmic heating, independently.

A proton beam of 1 GeV energy and intensity of about 50 nA delivered by the synchrocyclotron of PNPI was transported over a distance of 60 m to the target area. There, it impinged directly on the UC_2 target or was deflected by a magnet to impinge on a tantalum converter, located below the target, to generate fast neutrons.

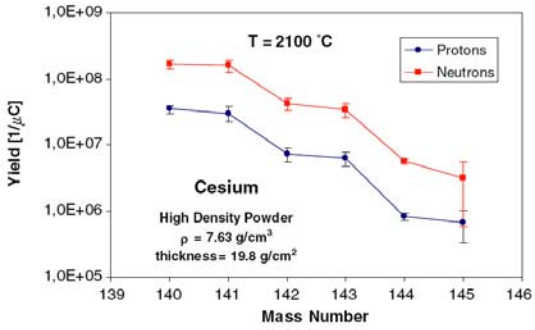

Fig. 4: Yields of neutron-rich Cs isotopes, comparison between the production rates for proton- and neutron-induced fission. The production rates for neutron-induced fission are corrected for the total neutron flux generated in the whole solid angle. The solid lines are only to guide the eye.

Fig. 4 shows the measured experimental yields of the neutron-rich Cs isotopes, in ions per μC, determined at the exit of the ion source. Simulations performed using the MCNPx Monte Carlo code [15] showed that the neutron production rate from the tantalum converter is 5.1 n/p and that only 4% of them are impinging into the uranium carbide target. Hence, the production yields generated by neutrons should be corrected supposing the neutrons produced in the

whole solid angle impinging into the target, to better compare with the yields from proton-induced fission.

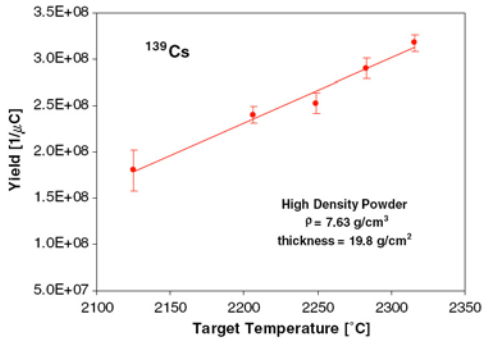

Fig. 5: Production intensity in function of temperature, for ^{139}Cs. The yields are produced by direct fission induced by protons on the target. The solid line is only to guide the eye and give the trend of data.

The production intensities were measured also as a function of temperature by monitoring specific γ-ray activities. In Fig. 5 are presented the results for the Cs isotopes; as expected, the intensities increase with the temperature. The increase in ionization efficiency in this temperature region may be considered negligible, so the rate of increase reflects mainly the decrease of the hold-up time.

- *comparing target materials with different densities*

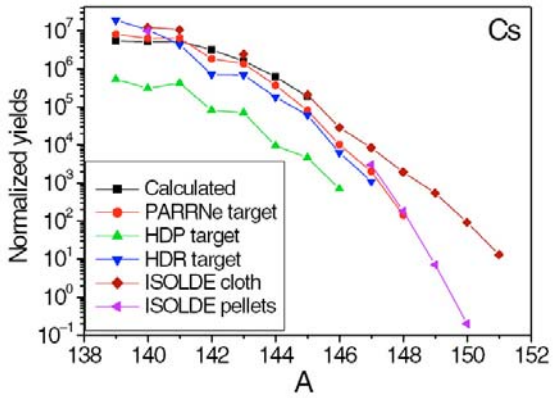

Fig. 6: The yields of Cs isotopes released from a high density rod, high density and low density pellet targets at 2100 °C. The yields from ISOLDE UC_x pellet target are presented. The yields calculated on the basis of measured cross-sections are shown as well.

To compare the different materials for the fission target, many experiments were performed at PNPI (Gatchina, Russia, using the same experimental setup described above [16-18]. Fig. 6 shows the yields of the Cs isotopes, normalized to a target thickness of 1 g/cm^2 and 0.1 μA proton beam

current, obtained from targets with different densities and material structure, as follows: high density rod (HDR) – uranium density of 11 g/cm^3, target thickness 6.7 g/cm^2, 6 mm length, 11 mm diameter, grain size of 200 μm; high density UC powder (HDP), prepared by the method of target metallurgy – uranium density of 12 g/cm^3, target thickness 6.3 g/cm^2, 5.25 mm length, 11.2 mm diameter, composed of 3 pellets each of (1.6 – 1.9) mm thickness, grain size of about 20 μm; low density target (LDT, also referred as PARRNe) – uranium density of 2.3 g/cm^3, 12 mm length, 11 mm diameter, composed of 8 pellets, each of (1.2 – 1.6) mm thickness with the grain size of about 20 μm. For comparison, are also presented the same yields taken from literature for 2 types of standard targets used at ISOLDE, as well as the yields calculated using the experimentally known production cross-sections.

5. Conclusions

The methods described in this paper are simple and efficient tools to investigate various target–ion source combinations for the RIB production using the ISOL method. They use very simple gamma or beta spectroscopy procedures, being in the same time easy to be performed even by non-expert researchers. Their results are of big importance for the nuclear physics community.

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