PRODUCTION OF NEUTRON-RICH NUCLEI IN LOW-ENERGY MULTINUCLEON TRANSFER REACTIONS*

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Multinucleon transfer processes in low-energy heavy ion collisions open new field of research in nuclear physics, namely, production and studying properties of not-yet-explored heavy neutron-rich nuclei. Beams of very heavy U-like ions are needed to produce new long-living isotopes of transfermium and superheavy elements. Beams of medium-mass ions can be used for the production of neutron-rich nuclei located along the neutron closed shell $N = 126$ (the last waiting point) having the largest impact on the astrophysical r-process. Low-energy multinucleon transfer reactions is a very effective tool also for the production and spectroscopic study of light exotic nuclei. The corresponding cross sections are found to be 2 orders of magnitude larger as compared with high energy fragmentation reactions.

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1. Motivation

Upper part of the present-day nuclear map consists mainly of proton-rich nuclei approaching the proton drip line (see Fig. 1). Very successful epoch of ^{48}Ca induced synthesis of new superheavy (SH) elements is over. The heaviest available target material, californium, was used to discover element 118 [1]. The formation cross sections of SH elements in fusion reactions of ^{48}Ca with actinide targets were found to be rather high (few picobarns)

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having almost equal values for elements from 112 to 118. Such behavior and the cross sections themselves were predicted in [2] and explained by increase of the fission barriers of synthesized SH nuclei owing to increase of the shell correction [4, 5] near the proton and neutron closed shells forming the island of stability ($Z \sim 110 \div 120$ and $N \sim 184$). To synthesize more heavier elements one needs to use heavier than ^{48}Ca projectiles. Our estimations show that the corresponding cross sections are at least 20 times lower as compared with ^{48}Ca synthesis [3], which significantly impedes the experiments.

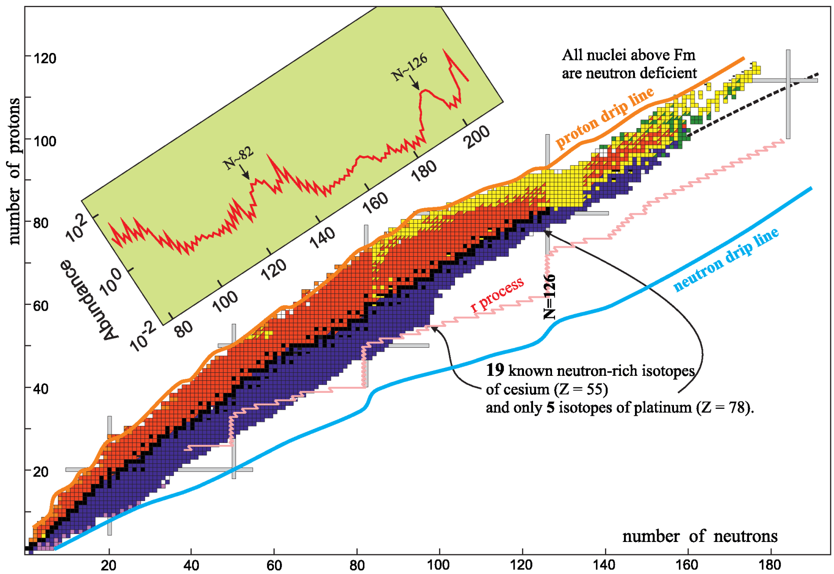

Fig. 1. Nuclear map as it looks today. Upper-right area of the nuclear map (near and to the right of the beta-stability line) is not yet explored.

At the same time neutron enriched isotopes of heavy elements were not synthesized and studied so far. This unexplored area of heavy neutron enriched nuclides (also those located along the neutron closed shell $N = 126$ to the right-hand side of the stability line) is extremely important for nuclear astrophysics investigations and, in particular, for the understanding of the r-process of astrophysical nucleogenesis.

Light and medium-mass neutron-rich nuclei are successfully produced in high-energy fragmentation processes and in fission reactions, correspondingly. Great progress here was done lately and dozens of new nuclei have been synthesized and studied, mainly at the laboratories of NSCL MSU [6], RIKEN [7] and GSI [8]. Evidently these reactions cannot be used for the production of heavy neutron enriched nuclei.

There are only three methods for the production of heavy elements: (1) fusion reactions, (2) a sequence of neutron capture and β^- decay processes, and (3) multi-nucleon transfer reactions. Because of the bending of the stability line toward the neutron axis, in fusion reactions only proton-rich isotopes of heavy elements can be produced. That is the main reason for the impossibility of reaching the island of stability ($Z \sim 110 \div 120$ and $N \sim 184$) in the superheavy mass region by fusion reactions with stable projectiles. The use of neutron-rich radioactive projectiles cannot help to solve this problem due to low intensities of such beams and extremely low production cross sections. Because of that we also have almost no information about neutron-rich isotopes of heavy elements located in the whole north-east part of the nuclear map: for example, there are 19 known neutron-rich isotopes of cesium ($Z = 55$) and only 5 of platinum ($Z = 78$).

The neutron capture process is an alternative (the oldest and natural) method for the production of new heavy elements. Strong neutron fluxes might be provided by nuclear reactors and nuclear explosions under laboratory conditions and by supernova explosions in nature. The "fermium gap", consisting of the short-living isotopes $^{258-260}$Fm located on the β-stability line and having very short half-lives for spontaneous fission, impedes the formation of nuclei with $Z > 100$ by the weak neutron fluxes realized in existing nuclear reactors. Theoretical models predict also another region of short-living nuclei located at $Z = 104 \div 108$ and $A \sim 275$. In nuclear and supernova explosions (fast neutron capture) these gaps may be bypassed if the total neutron fluence is high enough. Note that elements 99 and 100 (einsteinium and fermium) were first discovered in debris of the test thermonuclear explosion "Mike" [9].

The resulting charge number of the synthesized nuclei might be increased by sequential neutron flux exposure if two or several nuclear explosions were generated in close proximity to each other [10]. The same process of multiple neutron exposures might be also realized in pulsed nuclear reactors. Here, the pulse duration is much longer than in nuclear explosions (up to several milliseconds). However, the neutron fluence usually does not exceed 10^{16} n/cm^2 in existing nuclear reactors ($n_0 \sim 10^{19}$ $n/\text{cm}^2\text{s}$ during one millisecond pulse). Thus, the time of neutron capture $\tau_n = (n_0\sigma_{n\gamma})^{-1} \sim 10^5$ s, and only the nearest long living isotopes ($A + 1$ or $A + 2$) of irradiated elements can be formed during the pulse. Multi-pulse irradiation here corresponds to the "slow" neutron capture process, in which the isotopes of new elements with larger charge numbers are situated close to the line of stability and finally reach the fermium gap where the process stops. In this case, the probability for formation of heavy elements with $Z > 100$ is negligibly small independent of the number of pulses and total time of irradiation. The situation may change if one could be able to increase somehow the intensity of the pulsed reactor. The neutron fluence of one pulse and frequency of pulses should be high enough to bypass both gaps of short living nuclei on the way to the island of stability. The specifications of the high-intensity pulsed reactors of the next generation depends strongly on properties of heavy neutron-rich nuclei located to the right of these gaps. Using theoretical estimations for the decay properties of these nuclei (which are not yet synthesized) we have found that an increase of the neutron fluence in the individual pulse by about three orders of magnitude as compared with existing pulsed reactors could be quite sufficient to bypass both gaps [10].

Thus, for the moment there is only one method for the production of heavy neutron-rich nuclei, namely, multinucleon transfer reactions in collisions of heavy ions. Before the practical use of this method, the following questions have to be answered. What are the cross sections for production of new neutron-rich isotopes of heavy elements? What is the optimal combination of colliding nuclei for production of a given nucleus? What is the optimal beam energy? How to separate a given transfer reaction product from other "garbage" produced in such kind of reactions?

2. Damped collisions of heavy ions

Damped collisions of heavy ions and, in particular, multinucleon transfer processes were studied intensively during many years (see, for example, recent review papers [11, 12] and appropriate references therein). Transport models (Focker–Planck [13] and master equations [14] for the corresponding distribution function, and the Langevin equations [15]) were proposed many years ago for the description of heavy ion damped collisions. However, those time only qualitative understanding of these reactions was achieved. Quite recently it has become possible to describe quantitatively all the features of heavy ion deep inelastic scattering and related quasi-fission processes (energy, angular, mass and charge distributions of reaction products) [16].

Calculations performed within the microscopic time-dependent Schrödinger equations [17] have clearly demonstrated that at low collision energies of heavy ions nucleons do not "suddenly jump" from one nucleus to another. Instead of that, the wave functions of valence nucleons occupy the two-center molecular states spreading gradually over volumes of both nuclei. The same adiabatic low-energy collision dynamics of heavy ions was found also within the TDHF calculations [18, 19]. This means the following: (1) Any perturbation model based on a calculation of the sudden overlapping of single-particle wave functions of transferred nucleons (in donor and acceptor nuclei, respectively) is not applicable for description of multinucleon transfers in low-energy heavy-ion damped collisions. (2) The so-called DNS model assuming independent nucleon motion in two isolated mean fields is absolutely contrary to physics. (3) One-dimensional potential energy $V(R)$ has no meaning at $R \le R_{\text{contact}}$ as well as any speculations on the depth of potential pocket of $V(R)$. In low-energy collisions of heavy ions at $R \le R_{\text{contact}}$ the multi-dimensional potential energy (dependent on the shape parameters of the united nuclear system) must be used. The two center shell model looks most appropriate for calculation of this potential energy.

It was shown in Refs. [16] that the Langevin-type dynamical equations of motion (using this two center adiabatic driving potential) can be successfully applied for simultaneous description of strongly coupled multinucleon transfer, quasi-fission and fusion-fission reaction channels of low-energy heavy ion collisions. The distance between the nuclear centers $\boldsymbol{R}$ (corresponding to the elongation of a mono-nucleus when it is formed), dynamic spheroidal-type surface deformations of both fragments δ_1 and δ_2, their rotation angles, and the neutron and proton asymmetries, η_N and η_Z are the most relevant degrees of freedom for the description of damped collisions of heavy ions jointly with fusion-fission dynamics. For all the variables, with the exception of the neutron and proton transfers, we use the usual Langevin equations of motion with the inertia parameters, μ_R and μ_δ, calculated within the Werner–Wheeler approach

$$\frac{dq_i}{dt} = \frac{p_i}{\mu_i}, \qquad \frac{dp_i}{dt} = \frac{\partial V_{\text{eff}}}{\partial q_i} - \gamma_i \frac{p_i}{\mu_i} + \sqrt{\gamma_i T}\,\mathit{\Gamma}_i(t)\,.$$

Here, q_i is one of the collective variables, p_i is the corresponding conjugate momentum, V_{eff} includes the centrifugal potential, $T = \sqrt{E^*/a}$ is the local nuclear temperature, $E^* = E_{\text{c.m.}} - V_{\text{eff}}(q_i;t) - E_{\text{kin}}$ is the excitation energy, γ_i is the appropriate friction coefficient, and $\mathit{\Gamma}_i(t)$ is the normalized random variable with Gaussian distribution. The quantities γ_i, E^* and T depend on the coordinates and, thus, on time.

Nucleon exchange (nucleon rearrangement) is described by the inertialess Langevin-type equations of motion derived from the master equations for the corresponding distribution functions [16]

$$\begin{aligned}
\frac{d\eta_N}{dt} &= \frac{2}{N_{CN}} D_N^{(1)} + \frac{2}{N_{CN}} \sqrt{D_N^{(2)}}\,\mathit{\Gamma}_N(t)\,, \\
\frac{d\eta_Z}{dt} &= \frac{2}{Z_{CN}} D_Z^{(1)} + \frac{2}{Z_{CN}} \sqrt{D_Z^{(2)}}\,\mathit{\Gamma}_Z(t)\,.
\end{aligned}$$

Here, $D^{(1)}$, $D^{(2)}$ are the transport coefficients. We assume that sequential nucleon transfers play a main role in mass rearrangement. Other details as well as discussion on the values of nuclear viscosity and nucleon transfer rate can be found in [16].

For the moment this approach is the only one which reproduces quite properly all the regularities of heavy ion deep inelastic scattering and quasi-fission processes. As an example, in Fig. 2 experimental and theoretical energy-mass distributions of reaction fragments are shown formed in collisions of ^{86}Kr with ^{166}Er at 464 MeV center-of-mass energy.

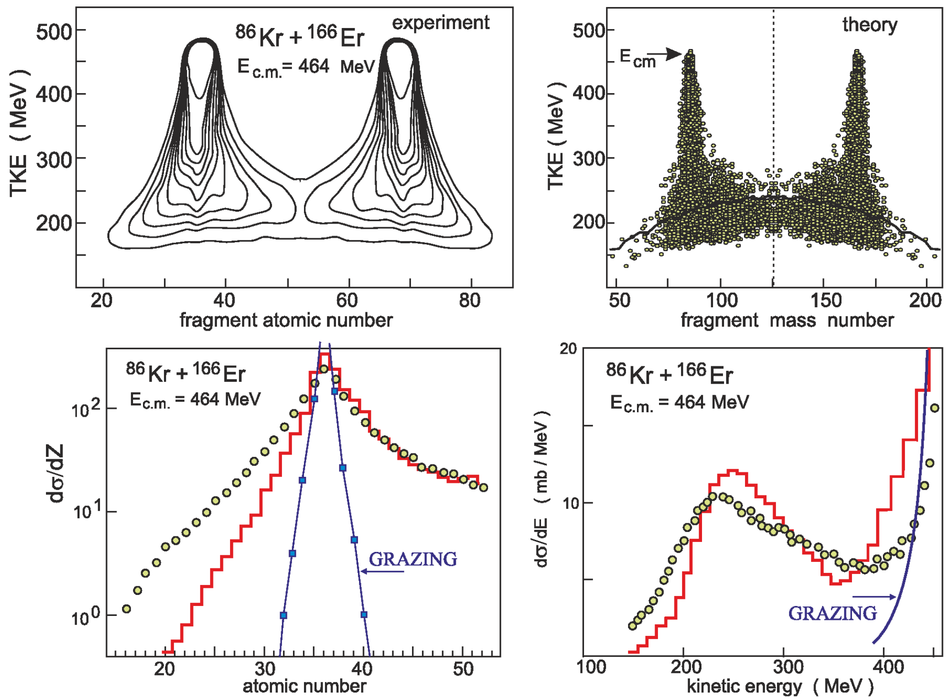

Fig. 2. Charge, mass and energy distributions of reaction fragments in collisions of ^{86}Kr with ^{166}Er at $E_{\mathrm{c.m.}} = 464$ MeV [20]. The histograms indicate the calculations performed within the model described above whereas the curves show the GRAZING calculations.

Some misunderstanding (or terminology) should be clarified here. The well known GRAZING code [21] (which is used very often for description of nucleon transfers) describes quite well few nucleon transfer reactions in grazing collisions. However, this code is not designed for the description of *multinucleon* transfer processes. It gives too narrow mass distributions of reaction fragments because the damped reaction channels with large kinetic energy loss are not included in the model (see Fig. 2).

3. Nuclei located along the closed neutron shell $N = 126$

Near barrier collisions of ^{136}Xe and ^{192}Os with ^{208}Pb target were predicted to be quite promising for the production of new nuclei with $N \sim 126$ [22]. This area of the nuclear map (being the last waiting point) has largest impact on astrophysical r-process. Our predictions of the production cross sections are not yet confirmed experimentally (because of difficulty of isotopic separation of heavy transfer reaction products) but all other reaction regularities (mass, energy and angular distributions) were found to be in good agreement with the predictions [23].

Low-energy collisions of stable neutron-rich isotopes of elements located below lead (such as ^{192}Os or ^{198}Pt) with available actinide targets look even more favorable for the production and study of new neutron-rich nuclei located along the closed neutron shell $N = 126$. Distribution of primary reaction fragments in (N, Z) plane is shown in Fig. 3 for the case of low-energy collisions of ^{198}Pt with ^{238}U at $E_{\rm cm} = 700$ MeV. As can be seen a lot of new isotopes in the region of the closed neutron shell $N = 126$ can be synthesized in this reaction with cross sections higher than 1 μb.

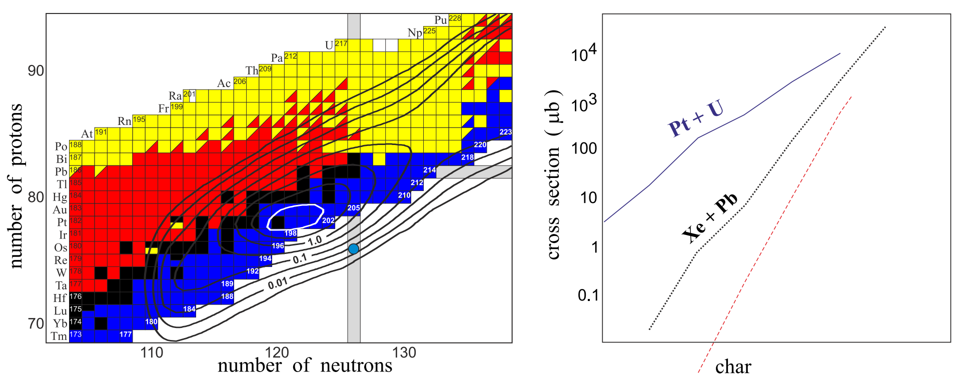

Fig. 3. (Left) Landscape of the cross sections for the production of isotopes around $N = 126$ in collisions of ^{198}Pt with ^{238}U at $E_{\rm cm} = 700$ MeV. (Right) Cross sections for the production of nuclei with $N = 126$ in low-energy collisions of ^{198}Pt+^{238}U and ^{136}Xe+^{208}Pb and in high-energy proton removal reaction [24].

4. Neutron-rich transfermium and superheavy nuclei

The use of actinide beams and actinide targets gives us a possibility to produce new neutron enriched isotopes of transfermium elements located along the stability line and to the right of it, that is in the unexplored area of the nuclear map (see Fig. 1). Properties of neutron-rich fermium isotopes with $A > 260$ are extremely interesting for several reasons. First, as mentioned above, all known isotopes of fermium (and of more heavy elements) are located to the left-hand side of the beta-stability line (see Fig. 1). Second, the well known “fermium gap” ($^{258-260}$Fm isotopes with very short half-lives for spontaneous fission) impedes formation of nuclei with $Z > 100$ by the weak neutron fluxes realized in existing nuclear reactors. It is extremely interesting to know what is the first β^--decayed fermium isotope and how long is its half-life. This is important not only for the reactor but also for the explosive nucleosynthesis in which this fermium gap might be bypassed [10]. As can be seen from Fig. 4 neutron-rich fermium isotopes can be produced in low-energy transfer reactions with cross sections of about 0.1 μb, that is quite sufficient to be produced at available accelerators.

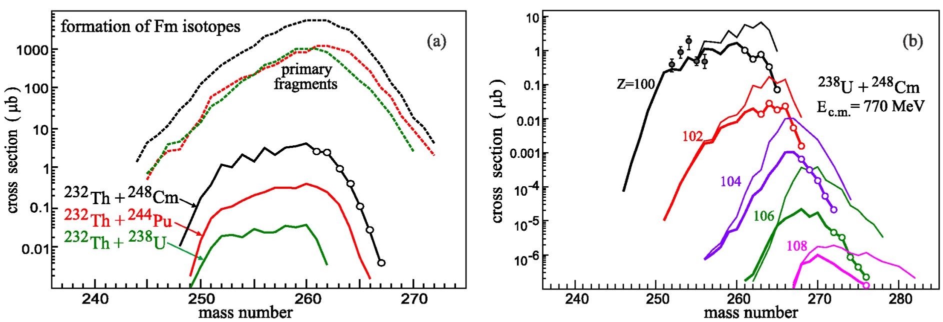

Fig. 4. (a) Isotopic yields of fermium nuclei in collisions of ^{232}Th with ^{238}U, ^{244}Pu and ^{248}Cm at $E_{\rm cm}$ = 715, 730 and 750 MeV, correspondingly. Open circles indicate new isotopes. (b) Cross sections for the production of neutron-rich superheavy nuclei in collisions of ^{238}U with ^{248}Cm target at $E_{\rm cm}$ = 770 MeV. Thin curves are obtained with the nucleon transfer rate parameter multiplied by factor 2.

The yields of neutron enriched long living isotopes of superheavy elements in transfer reactions might be significantly enhanced owing to the shell effects leading to the so-called “inverse quasi-fission” phenomena [25]. In Fig. 4 the results of our calculations are shown for the formation of survived isotopes of some transfermium elements in reaction ^{238}U+^{248}Cm at 770 MeV center-of-mass energy. The obtained results are rather optimistic. New neutron-rich isotopes of transfermium elements with Z = 100–104 (located already at the stability line and beyond it) can be produced with the cross sections of several hundreds of picobarn. The cross sections for the production of new neutron-rich isotopes of seaborgium and hassium ($Z = 106, 108$) are also higher than 1 picobarn. Predicted cross sections depend on the values of neutron and proton transfer rates which are not yet determined very accurately (see Refs. [13, 14] and [16]).

5. Low-energy nucleon transfers vs. high-energy fragmentations

Being inspired by good agreement of the model with available experimental data on damped collisions of heavy ions, we studied also the multinucleon transfer reactions in low-energy collisions of light heavy ions with heavy targets [26]. The results of these calculations (shown in Fig. 5) demonstrate that the cross sections for the production of quite exotic light neutron-rich nuclei produced in the low-energy multinucleon transfer reactions are higher by about 2 orders of magnitude as compared with high energy fragmentation reactions. Thus, the low energy damped collisions of light heavy ions with heavy targets look very promising (and quite competitive to the high-energy fragmentation reactions) also for the production and study of light exotic nuclei.

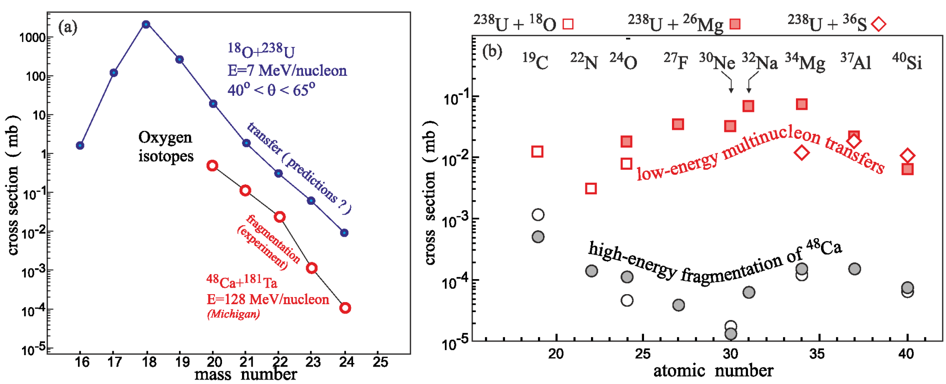

Fig. 5. (a) Cross sections for the production of neutron-rich oxygen isotopes in low-energy collisions of ^{18}O with ^{238}U target and in fragmentation process [27]. (b) Formation of light neutron-rich nuclei in low-energy collisions of ^{18}O, ^{26}Mg and ^{36}S with ^{238}U target and in fragmentation of 128 A MeV ^{48}Ca on ^{181}Ta target [27] (filled circles) and 345 A MeV ^{48}Ca on ^{9}Be [7] (open circles).

6. Laser separation of heavy reaction products

In contrast with fusion reactions it is more difficult to separate a given product of multinucleon transfer reaction (having rather broad angular and velocity distributions) from all the other reaction fragments. Heavy nuclei with $Z > 70$ formed in the multinucleon transfer reactions cannot be separated and studied at available setups created quite recently just for studying the products of deep inelastic scattering (such as VAMOS, PRISMA and others). These fragment separators (as well as other setups) cannot distinguish heavy nuclei with $Z > 70$ by their atomic numbers. However, during the last several years a combined method of separation is intensively studied based on stopping nuclei in gas and subsequent resonance laser ionization of them [28]. One of such setups (named GaLS: in **Ga**s cell **L**aser ionization and **S**eparation) is currently created at the Flerov Laboratory (JINR, Dubna). First experiments at this setup aimed on the production and studying properties of new neutron-rich heavy nuclei are planned to start in the beginning of 2016.

7. Summary

The use of multinucleon transfer reactions in low-energy collisions of heavy ions opens new field of research in nuclear physics: synthesis and studying properties of heavy neutron-rich nuclei. These nuclei can be produced neither in fusion reactions nor in fragmentation processes.

^{136}Xe+^{208}Pb, ^{192}Os+^{208}Pb and ^{198}Pt+^{238}U are the most appropriate reactions for the production of neutron-rich nuclei located along the closed neutron shell $N = 126$ having the largest impact on the astrophysical r-process. The use of actinide beams and actinide targets allows one to produce new neutron enriched (and longer living) isotopes of transfermium and superheavy elements located along the stability line and to the right of it. Shell effects (inverse quasi-fission mechanism) might significantly enhance the cross sections for the production of trans-target nuclei. The low energy damped collisions of light heavy ions with heavy targets look also very promising and quite competitive to the fragmentation reactions for the production and study of light exotic nuclei. Separators of new type based on selective laser ionization of heavy reaction products have to be designed and installed for this new field of research.

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