Transportation of exotic fission products targeted for the goals of astrophysical r-process

D Simonovski¹, S V Chenmarev²

¹Saint Petersburg State University. 7/9 Universitetskaya Emb., 199034, Saint Petersburg, Russian Federation
²Petersburg Nuclear Physics Institute of National Research Centre “Kurchatov Institute”, 188399 Gatchina, Leningradskaya oblast, Russian Federation

E-mail: simonovski.dimitar@yandex.ru

Abstract. The gas-jet extraction and transportation method of fission products produced by thermal neutron irradiation of the uranium target near the reactor’s active zone has been considered. The gas-jet transportation system has already been successfully implemented, tested and used in the facilities on the TRIGA research reactor in Mainz (Germany). The main goal of this work consisted in the optimization of the transport parameters of the gas-jet system, which were experimentally measured and theoretically modelled and subsequently adopt them for the case of the high flux reactor PIK, which is under commissioning at the PNPI in Gatchina (Russian Federation). As a result of this research a formula for the fission products transport time has been derived. An optimization was suggested by using He as carrier and buffer gas and theoretical calculations have been done, in order to make predictions about the new nuclides, which would be able to be studied under the planned conditions on the PIK reactor. This information can be very helpful in the creation of the setup for production and investigation of exotic nuclides in the on-line regime on high-flux reactors.

1. Introduction

One of the most important current scientific research fields in nuclear physics is tightly bound to the production and investigation of newly unstudied exotic nuclides, which are located far from the stability line in the chart of nuclides. Because of their highly asymmetric ratio of the number of neutrons and protons, they are usually beta-unstable nuclides possessing very short half-lives, which in its own accord is the main reason of their natural absence in our everyday earthly conditions and their unordinary exotic nature.

The most current nuclear physics models suggest the existence of thousands of different nuclei, out of which only approximately 3200 have been actually experimentally studied and detected [1]. Among the great number of yet unstudied nuclides are the neutron rich nuclides, which are composed of a highly asymmetric number ratio of their protons and neutrons. The investigation and experimental measurement of their ground state properties like their mass, spin and half-lives are not only considered of great importance for the fundamental development of the nuclear physics theories and models, but are also crucial for the scientific explanation of the astrophysical nucleo-synthesis processes. Under stellar conditions, the fusion process can contribute to the production of all nuclides that consist of no higher number of protons than iron. For the productions of the remaining nuclides
heavier than iron the slow and rapid neutron-capture processes (s- and r- processes) are supposed to be responsible for. The s-process occurs when a nuclide is irradiated by a high neutron flux and as a result of the capturing one neutron it consequently decays by the β⁻ process before catching the next one. Such a process can synthesize no heavier nuclides than Bi-209. The r-process is the process that is responsible for the nuclear synthesis of much more heavier nuclides. During the r-process a nucleus captures a larger number of neutrons between two consequential decays than during the s-process, which produces as a result more neutron-rich exotic nuclides [2].

In order to study these theoretically assumed astrophysical processes under earthly conditions one needs to first obtain and study the ground state properties of the neutron-rich nuclides, which could be produced by such processes. Because of the short half-lives of the exotic nuclides and the absence of their earthly natural source, they need to be artificially produced in order to be studied. One of the most advanced facilities that are built for the purpose of such investigations is the scientific research reactor. The TRIGA research reactor in the Nuclear chemistry institute on the Johannes Gutenberg University located in Mainz (Germany) is one of the remaining scientific research reactors which is still in use and in full operational mode. The TRIGA-SPEC branch of the apparatus [3] is used for the extraction and very fast transportation of the produced fission products using the gas-jet transportation method and their consequential ionization, whereas the TRIGA-TRAP branch is used for high precision ion mass measurement using Penning traps. A very similar experimental branch is planned on the still upgrading PIK reactor [4] in Gatchina (Russian Federation), where an excitingly higher neutron fluxes are expected to be present as well as a more massive target than the ones used on the TRIGA-Mainz reactor. The new experimental conditions that are going to be present at the PIK reactor are highly promising for the detection and mass measurement of newly unstudied neutron-rich nuclides and for the purposes of even higher optimization of the results to be achieved they demand a new theoretical modeling and study of the already operating experiment at the TRIGA-Mainz reactor, especially the gas-transport method, which assures rapid transport of short-lived fission products.

In this paper is presented a mathematical modeling of the experimental data obtained on the TRIGA-Mainz reactor, an optimization of the gas-jet method and numerical estimations of the expected experimental results on the planned experimental apparatus on the PIK-reactor.

2. The gas-jet method
The gas-jet method has been studied and used since the 1960s in various different scientific facilities [5] and all have their own specifics and differences, but the main scheme of the method which is common in all present cases is going to be discussed in this section. As a reference for this explanation we are going to use the gas-jet method used on the TRIGA-SPEC facility.

Firstly the carrier gas N₂ is flushed out from the gas container threw a capillary tube into an aerosol oven where aerosol particles are produced through a steaming process. The aerosol clusters get mixed into the carrier gas and they are flushed out from the aerosol oven into the target chamber. On one of the target chamber walls a 2 milligram \(^{235}\text{U}\) is mounted and irradiated with high neutron fluxes approximately \(10^{11} \text{n/s/cm}^2\). High energy fission products are then thermalized into the carrier gas and get attached to the aerosol clusters which are all flushed out from the target chamber into a system of two consequentially connected capillary tubes, where by a laminar flow stream they are transported till the end of the capillary tube. In order to separate the gas particles from the aerosol, carrying the fission products at a 1 mm distance from the capillary tube’s exit a skimmer has been mounted. Because of the different angle distributions of the light nitrogen molecules and heavy loaded aerosol particles they get quite successfully separated by the skimmer and only a fraction of the gas molecules pass through the skimmer. After they passed through the skimmer the aerosol particles and the attached fission products are brought into the ion source chamber where the aerosols and fission products get separated by collisions with the ion source walls and were the process of fission products ionization takes place. After the ionization process they are all extracted by an electrostatic electrode system and brought on the separating magnet, which depending on the charge-mass ratio of the ion selects the ones that need
to be studied by positioning an orifice at the end of the magnet and letting pass only those ions with such charge-mass ratio that obtain trajectories in the magnetic field that pass through the orifice.

After the separating magnet the TRIGA-TRAP branch begins with an ion beam buncher, which collects the ions into bunches and passes them into a twofold Penning trap system one of which does the gas ion cooling and one that is used for high precision mass measurement. After the last Penning trap they are sent to the detector and using the ToF method their masses are measured.

3. The transport time measurement
On the TRIGA-SPEC facility the fission products transport time has been experimentally measured by obtaining gamma spectra of the passing activity through a certain point of the capillary tube where the detector was mounted and directed. The experiment [3] was done for different carrier gas volumetric flows ranging from 300 mL/min to 800 mL/min. The transport time experimental results are shown in table 1 columns two and six.

| Volumetric Flow (mL/min) | Transport time \( \Delta t_{\text{ex}} \) (ms) | Transport time \( \Delta t_{\text{th}} \) (ms) | Pressure \( P \) \((-10^5 \text{ Pa})\) |
|--------------------------|----------------------------------|----------------------------------|--------------------------|
| Q                        | \( \Delta t_{\text{ex}} \)       | \( \Delta t_{\text{th}} \)       | \( P \)                  |
| 300                      | 1505(14)                         | 1500                             | 1.00                     |
| 400                      | 1376(23)                         | 1390                             | 1.13                     |
| 500                      | 1263(10)                         | 1290                             | 1.30                     |
| 600                      | 1094(30)                         | 1150                             | 1.38                     |
| 700                      | 1028(9)                          | 1040                             | 1.50                     |
| 800                      | 945(5)                           | 950                              | 1.60                     |

In order to mathematically model the whole experiment and estimate the transport time theoretically the geometry of the target chamber has been taken into consideration and the Navier-Stokes equation was solved for the case of laminar flow for the purpose of obtaining the velocity distribution of the traveling aerosols loaded with fission products. The geometry of the target chamber, which is schematically illustrated on figure 1, is twofold: the first section represents a cylinder with length \( L_{m1} \) and radius \( R_{m1} \) and the second section represents a truncated cone with radii \( R_{m1} \) and \( R_{m2} \) and length \( L_{m2} \). It was estimated that the aerosol speed does acquire the carrier gas speed in the matter of few milliseconds and therefore it was considered that throughout the whole transport process they were moving with the same velocity. Also the stopping range of the fission products in the carrier gas was taken into account, which shortens their transport trajectory. By taking all of these considerations into account, a formula for the fission products transport time was derived, which is shown below

\[
\Delta t_{\text{th}} = \frac{L_{m1} - l_{\text{max}}}{2Q} + \frac{L_{m2}}{6Q} \left( R_{m1}^2 + R_{m1}R_{m2} + R_{m2}^2 \right) + \frac{\pi R_{t1}^2 L_{t1}}{2Q} + \frac{\pi R_{t2}^2 L_{t2}}{2Q},
\]

where \( \Delta t_{\text{th}} \) is the theoretically estimated transport time, \( L_{m1} \) and \( L_{m2} \) are the lengths of the two parts of the target chamber, \( L_{t1} \) and \( L_{t2} \) are the lengths of the two capillary tubes, \( R_{m1} \) and \( R_{m2} \) are the radii of the two parts of the target chamber, \( R_{t1} \) and \( R_{t2} \) are the radii of the two consequently connected capillary tubes after the target chamber, \( Q \) is the volumetric flow of the carrier gas and \( l_{\text{max}} \) is the maximum stopping range of the fission products. The stopping rage was estimated for all of the \(^{235}\text{U}\) fission products and it was concluded that it reaches its maximal value for the case of the fission product \(^{143}\text{Ce}\), which was used in all of the calculations using the formula 1 in order to estimate the fission products transport time.
Figure 1. A schematic illustration of the cross-section of the target chamber and capillary system

If we compare the values obtained theoretically and experimentally from table 1 one can easily conclude that the formula 1 can be successfully used in order to approximately describe the fission products transport time from the point of time of their creation in the target chamber until the moment of their detection.

4. Optimization of the gas-jet method
A theoretical research has been done using all of the noble gasses He, Ar, Ne, Kr, Xe and N\textsubscript{2} in order to optimize the gas-jet method. The main characteristics upon which they were judged and compared were the velocity constrictions given by the Reynolds number limit Re<2300 because of the need of laminar flow through the whole experimental apparatus, the effective thermalization of the fission products and the constrictions they impose upon the geometry of the capillary tubes because of the maximum pressure limit in the target chamber. Using the formula for the Reynolds number constriction and the Heigens-Poiselle formula for the pressure drop in the capillary tubes it was estimated that He is the best candidate to be used out of all the gasses in question. The main reasons for this conclusion were the higher limit in the flow velocity and lower number of imposed constrictions on the geometry of the capillary tubes, which are achieved by using helium, and as a result capillaries with much smaller radii can be used. The only negative side to by using helium is the need for high volumetric flows in order to achieve the needed thermalizing pressure, which is not a problem because of the light constrictions of the He on the volumetric flow. Usually such thermalization problems are solved by putting a layer of thin metal like Al on the surface of the target, which additionally thermalizes the fission products and does not impose any additionally needed constraints on the buffer gas properties. The second best candidate is Ar, which because of its similar properties with the molecular nitrogen in this context can be used as a complete substitute for him [6].

5. Newly available exotic nuclides at the PIK-reactor
Taking into account the previously concluded optimization of the gas-jet method by using He instead of molecular nitrogen and taking into account the new experimental conditions planned for the apparatus at the PIK-reactor, a theoretical investigation was done in order to estimate which nuclides would be available in the case of the PIK-reactor that are not available at the TRIGA-Mainz reactor. The new experimental conditions, which are mainly contributed by the 2g uranium target and the average neutron flux more than $10^{13}$ n/s/cm$^2$ along the channel, by their own accord, exceed the target mass and average neutron flux present at the TRIGA-Mainz reactor. As a result it can be easily be
approximated, that the production rate of fission products at the PIK-reactor would exceed the production rate at the TRIGA reactor by the factor of $10^5$. Such experimental conditions highly increase the probability of the survival rate of the transported short-lived fission products.

Taking into consideration the newly presented experimental conditions in Table 2 are presented some of the newly future available nuclides at the PIK-reactor. The last radioactive isotopes have been presented in a given isotopic chain, which can survive the gas-jet transport process. From Table 2 it is quite easy to conclude that in the case of the experimental conditions of the PIK reactor a presence of more exotic nuclides in a given isotopic chain will be available for study. The question marks in the brackets near the nuclides indicate that their half-lives have not yet been studied and experimentally measured.

### Table 2. Nuclides that can survive the transportation process at the TRIGA-Mainz and PIK reactor

| Z  | TRIGA-Mainz | PIK  |
|----|-------------|------|
| 26 | N$_2$       |      |
| 27 | /           |      |
| 28 | ⁶⁴Ni (680 ms) |      |
| 29 | ⁷⁷Cu (469 ms) |      |
| 32 | ⁸⁶Ge (300 ms) |      |
| 33 | ⁸⁸As (300 ms) |      |
| 34 | ⁹¹Se (270 ms) |      |
| 63 | ¹⁶⁴Eu (2 c)  | ¹⁶⁶Eu (1.24 c) |
| 64 | ¹⁶⁶Gd (4.8 c) | ¹⁶⁶Gd (3 c) |
| 65 | ¹⁶⁷Tb (19 c) | ¹⁶⁹Tb (5.13 c) |
| 66 | ¹⁶⁸Dy (8.7 m) | ¹⁷⁰Dy (55 c) |
| 67 | ¹⁶⁹Ho (4.7 m) | ¹⁷⁰Ho (43 c) |

### 6. Conclusions

In order to successfully implement the gas-jet transport system in the newly planned experimental conditions in the facilities on the PIK-reactor for the purpose of mass measurement of exotic nuclei especially of astrophysical interests a study has been done on the gas-jet transport method based on the already operating one on the TRIGA-reactor in Mainz. The study included experimental measurements of the transport parameters of the gas-jet system and their subsequent mathematical modeling, which makes the task of optimization, transference and implementation of the gas-jet method possible for the new experimental conditions presented by the PITRAP project. As a result from the theoretical modelling of the gas-jet transport process a formula for the transport time has been derived, which was in agreement with the experimentally obtained results for the fission products transport time at the TRIGA-reactor facility. As a result from the optimization of the gas-jet method, which involved all noble gases as candidates for the gas-jet transport method including nitrogen, which is already used in the TRIGA-TRAP project, an improvement is suggested by using helium as a carrier gas, because of its much shorter transport times and fewer experimental constraints implied on the experimental apparatus by using it. Also Ar has been proposed as the second best candidate for the substitution of molecular nitrogen, because of their very similar thermalization properties and constraints imposed on the flow velocity and apparatus geometry. Taking the previously elaborated improvements into consideration and taking into account the planned operating conditions on the PIK-reactor a prediction has been made for the expected survival of yet unstudied short-lived exotic nuclei after the transport process.
Acknowledgements

The authors express a great gratitude to Yu.N. Novikov and A.V. Popov from the PIAF institute (Russia) for their unique interest and undoubtfully helpful critical comments during our work. Also we would like to thank C. Blaum from the Max Planck institute in Heidelberg (Germany), M. Block, K. Dullman, S. Nagy and all the coworkers from the Nuclear chemistry institute in Mainz (Germany) for the welcoming and collaboration on the TRIGA-Mainz reactor. Also the authors express a gratitude to the SPSU University for the support for the internship at the TRIGA reactor in Mainz in Germany.

References

[1] G Audi, A H Wapstra, C Thibault 2003 Nucl. Phys. A 729 337
[2] E M Burbidge, G R Burbidge, W A Fowler and F Hoyle 1957 Rev. Mod. Phys. 29, 580
[3] J Grund at al. (private communication) 2017 Institut für Kernchemie (JGU) in Mainz(Germany)
[4] Yu I Gusev et al. 2015 Atomic Energy (in Russia) 118 334-340.
[5] R Macfarlane, R Gough, N Oakey, D Torgerson 1969 Nucl. Instr. Meth. 73 285
[6] D Simonovski et al. 2017 Atomic Energy (submitted)