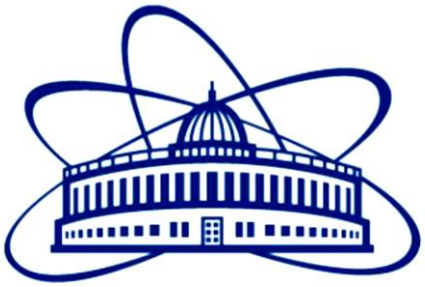

JOINT INSTITUTE FOR NUCLEAR RESEARCH
Flerov Laboratory of Nuclear Reactions

FINAL REPORT ON THE INTEREST PROGRAMME

Optimization of the solid ISOL method for volatile reaction products of heavy ion beam reactions

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Abstract

The search for the predicted Island of Stability and the study of heavy neutron-rich nuclei near neutron shells N=126 and N=152 have become cutting-edge research lines in modern nuclear physics. The MASHA mass-separator at the Flerov Laboratory of Nuclear Reactions (FLNR) at JINR is a facility with extensive expertise determining mass-to-charge ratios and detecting superheavy reaction products. Many studies have been recently developed on the analysis of the production parameters (separation efficiency and absolute cross-sections) of super-heavy Hg and Rn nuclei by complete fusion and multinucleon transfer reactions, due to the similarity of its chemical properties respect to that expected for heavier elements such as Copernicium (Z=112) and Flerovium (Z=114). However, previous tests show that these elements are highly volatile, making the optimization of the ISOL method inevitable to perform such experiments. A detailed analysis of the results obtained after these modifications is presented as part of this report, based fundamentally on the analysis of the heat map graphs and the emission spectra of alpha particles corresponding to the radioisotopes of interest. Finally, the results obtained are compared with those reported in previous references.

Summary

1. Introduction

The search for the predicted Island of Stability and the study of heavy neutron-rich nuclei near neutron shells N=126 and N=152 have become fundamental research lines in modern nuclear physics [1], enabling a better understanding of the nuclear structure and reaction mechanisms that take place in systems like the stars. To reach real conclusions regarding the fundamentals of nuclear physics behind these problems, it requires carrying out a large number of precise measurements, including mass-to-charge ratios, reaction cross sections, energy spectra, half-lives, decay modes, and properties. Kinematic separators are widely used worldwide for separating short-lived radioactive nuclei [2], using In-Flight Separation (IFS) or Isotope Separator On-Line (ISOL) techniques to obtain exotic isotopes, far from stability with short half-lives. More specifically, ISOL method involves trapping isotopes produced in nuclear reactions in a solid matrix, selectively releasing these isotopes, and then analyzing them using various detection techniques. This method is well-suited for volatile products that would otherwise be challenging to study due to their short half-lives or low production cross-sections. However, based on the fact that half-lives of the majority of superheavy nuclei are too small, traditional kinematic separators need to be complemented with mass analyzers to make accurate determination of their masses possible [5].

The mass-separator MASHA (Mass Analyzer of Super Heavy Atoms), located at the Flerov Laboratory of Nuclear Reactions (FLNR) at JINR, is a key facility that allows for the precise on-line determination of mass-to-charge ratios of the superheavy reaction products, as well as the simultaneous detection of their corresponding α −decays, spontaneous fission, and other mechanisms [2,3]. This facility, known for its high-intensity heavy ion beams, offers a controlled environment for these experiments, thus allowing researchers to efficiently isolate and study volatile isotopes produced during these reactions.

The optimization of the ISOL method for volatile reaction products at the MASHA facility is critical for enhancing the sensitivity, selectivity, and efficiency of isotope identification. By fine-tuning the parameters of the ISOL setup, such as the target material, temperature, and extraction mechanisms, researchers can improve the overall performance of the system.

This report is made up of a total of three sections. The setup of MASHA spectrometer is described in the first one. The results of data analysis is indicated in the second section, as well as its corresponding comparison with those presented in the different references. The project's primary outcomes are outlined in the final section of the conclusions

2. MASHA spectrometer

Figure 1 depicts a general schematic representation of the MASHA facility, which includes beam delivery from the U-400M cyclotron, primary beam diagnostics, block "target-hot catcher" unit, ECR ion source,[1] 2-stage separator and mass analyzer, intermediate detector, focal plane detector (FPD), secondary beam diagnostics, vacuum, computer control, and other auxiliary systems [2].

[1] Electron Cyclotron Resonance.

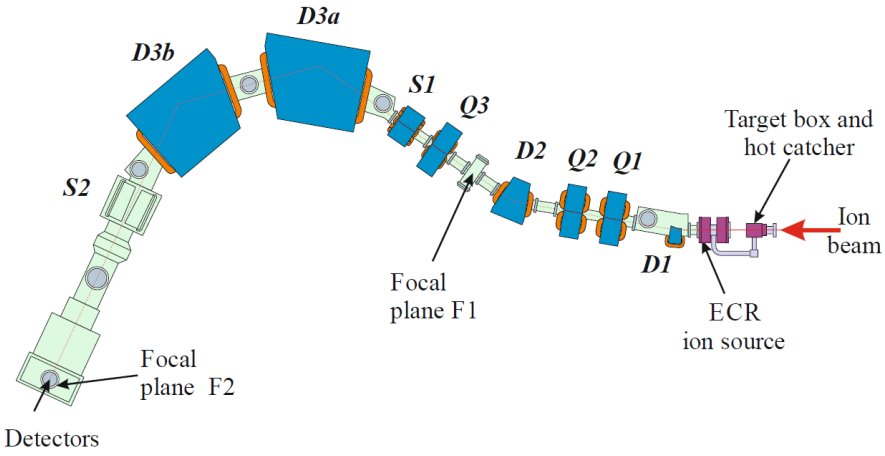

Fig. 1: Current diagram of the MASHA mass separator where the names of the different components of the facility are indicated. To identify the parts of the analyzer system, they are represented the four dipole magnets (D1, D2, D3a, D3b), three quadrupole lenses (Q1, Q2, Q3), two sextupole lenses (S1, S2) and a focal plane detector system. Image taken from reference [5].

As part of this process, atoms produced in nuclear reactions and emitted from the target pass through the separating foil before being halted in the hot catcher unit. The production and implantation process occurs in the so-called *target+hot catcher* block, whose current configuration layout is shown in **Fig. 2**. Among the current features of this part of the system should be mentioned that it count with one rotating targets, which improves efficiency and heat distribution compared to stationary targets [1]. Regarding to the modern separating foil, this consists in a thin graphite foil, which is much more suitable for these applications compared to those of titanium [1]. The above reason is due to the higher thermal conductivity of these materials, which allows them to withstand high temperatures for a much longer time. Besides, the correct selection of the hot catcher is crucial for optimized ISOL setups, especially for short-lived superheavy nuclei with low production cross-sections. The properties of this component of the system dictate separation time and efficiency, with porous graphite being the best recommendation to achieve these improvements [5]. On the other hand, the heater is made of poly-graphene heated from a tantalum band by direct current, reaching a typical temperature range of $1800 - 2000K$ [3]. All previous modifications enhance the efficiency and performance of the *target + catcher system* in the mass-separator setup.

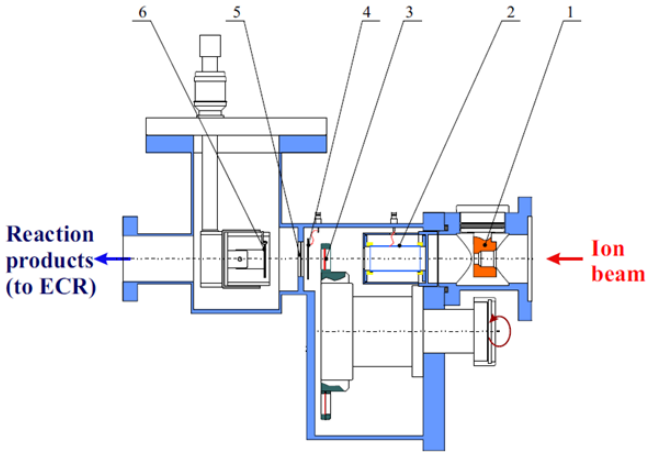

Fig. 2: Schematic overview of the *target+hot catcher* system. Here: (1) split collimator; (2) pick-up sensor connected to beam flux analyzer; (3) target on the wheel; (4) electron emission beam monitor; (5) separating foil; (6) hot catcher. Image taken from reference [1].

Due to thermodiffusion, nuclear reaction products drift in the form of atoms, from the graphite to the surface of the hot catcher, and after passing through a vacuum tube, they reach the ECR ion source [3]. For a detailed description of the characteristics of this source, reference [6] can be consulted. In this region, the incoming atoms are ionized up to a charge state $Q = +1$, and then extracted with an energy in a range of $15-40\,keV$ by three-electrode electrostatic lens [2,4]. With this ion source it is possible to get beam currents composed almost entirely of singly-charged ions. Noble gases, like krypton and xenon, are traditionally chosen for ECR ion source optimization due to their maximum ionization potentials, registering even in these cases about $90\,\%$ of the ionization efficiency [1]. Optimizations of this component of the system have been carried out in recent years for the analysis of certain elements, which main results have been reported in references [1,5]. In this sense, to study volatile elements such as Hg, it must be taken into account that it present high adhesion on steel and when the atom goes inside the chamber, it will suffer multiple collisions with the walls of the source before ionizing, decreasing the efficiency of the process in this case. As a solution to this unwanted phenomenon, it was decided to coat the walls of the source chamber with titanium nitride (TiN), as well as the walls of catcher and transportation line between, since this is a chemical inert compound close to ceramic. In reference [1] it is mentioned that the result of this modification allowed us to observe an increase in ionization efficiency and release time.

The results of a more recent update of the source are presented in reference [5], where a new optimization of the system is carried out by coating the inner surfaces of the vacuum pipelines and chambers with special chemical inert glass-enamel. The importance of this improvement, which was proposed in reference [1], is to allow the detection not only of volatile elements (radon or mercury) but also of nearby elements (At, Fr, Ac) [5].

After the ECR ion source, the superheavy elements (atoms in the mass range $(A \approx 290 \pm 8)$ has to be separated from total ion beam formed, since in addition to these, there will also be a certain component of ^{4}He (ion source gas), ^{48}Ca ions (primary beam), target fission fragments $(A = 70-150)$ and target-like atoms $(A = 200-250)$ [2]. To proceed with the separation magneto-optical mass-to-charge ratio analyzer are used. This process consists of two stages: a first one that involves rough separation of ions, extending from the ion source to the intermediate horizontal focusF_1, and a second one that allowing next separation and precise mass analysis of ions [2].

The separator setup offers advantages for superheavy nuclei separation, such as increased target thickness compared to kinematic separators, which limit target thickness to $0.2-0.3\,mg/cm^2$. This allows simultaneous synthesis of two or even three isotopes. Additionally, if a nuclide have half-time longer than the separation time, mass identification can be done by registering its own decay, or decay of its daughter nuclides. In contrast, only daughter nuclides, resulting from decays of parents with $Z = 112$, are registered and their masses are determined [2].

Finally, mention that in addition to the previous components, the facility count with multiple systems dedicated to control, beam diagnostic, and detection-measurement. Some characteristics of these systems can be found in references [1-4].

3. Data analysis

This section describes the data analysis procedure for the main results obtained from Ar primary beam test experiments to produce super-heavy Hg and Rn isotopes. In the case of mercury isotopes, they were produced as evaporation residua in the complete fusion reaction ^{40}Ar+^{144}Sm to test and tune the mass-separator MASHA. The interest in studying this element is due to the analogy of some of its chemical properties, with respect to Copernicium (Z=112) and Flerovium (Z=114) [1,3]. Summarizing the results of these measurements, we must mention that bidimensional matrix of particle energy vs. strip number was measured with good separation by mass and energy, as shown in **Fig. 3**. The identification of the respective masses is carried out by adjusting the field generated by D3a-D3b dipole analyzer magnets (see **Fig. 1**), which results can be observed in the heat map due to the direct dependence between the strip number and the corresponding X −coordinate of the focal plane. It is evident from the graph that isotopes are efficiently separated by both mass and α −decay energy.

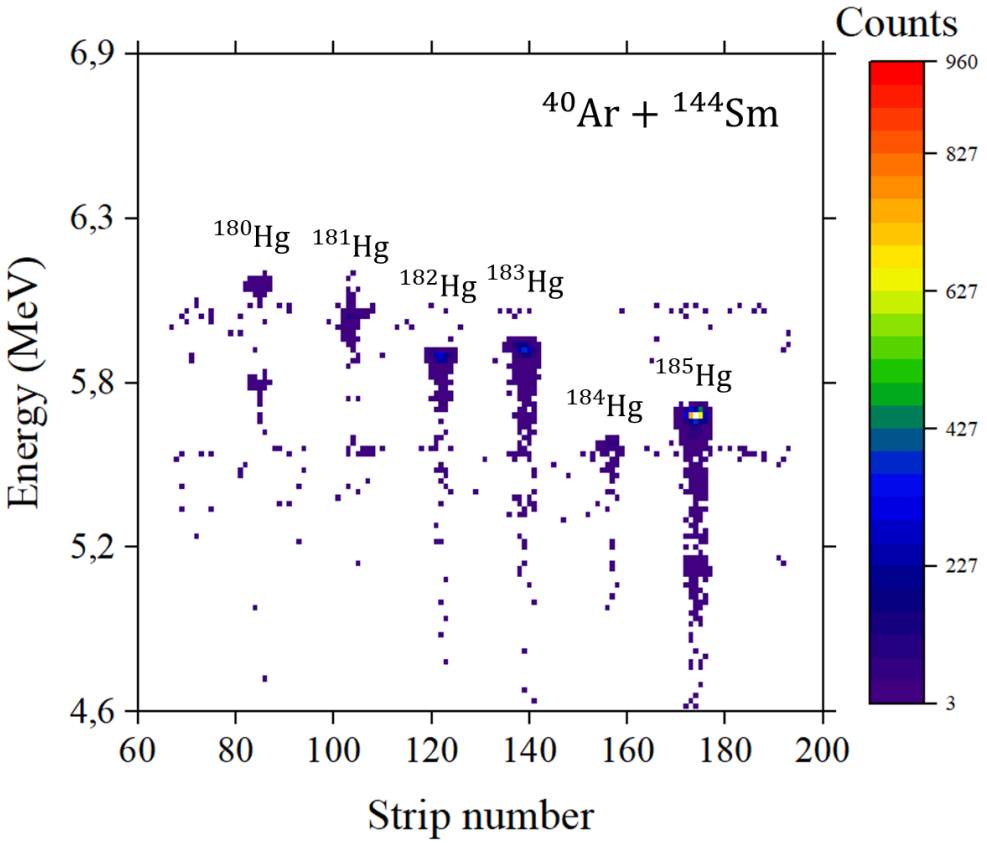

Fig. 3: Bidimensional graph of energy of α −particles vs. strip number (energy-position) registered from decays of Hg isotopes with mass numbers from 180 to 185.

A quantitative analysis of the efficiency of separation of the mass-spectrometer MASHA for residual mercury isotopes ($^{180\text{-}182}$Hg) of the xn −evaporation channels of complete fusion reaction ^{40}Ar+^{144}Sm→ $^{184\text{-}xn}$ Hg+xn is performed in the reference [7]. A complementary study was presented in [8], where the absolute cross sections of complete fusion reactions with ions ^{36,40}Ar and ^{40}Ca that lead to the formation of the

^{184}Hg compound nucleus, were determined. Both studies conclude that, some dependence on this efficiency with nuclide half-lives was observed, reporting the best separation for the ^{181}Hg (1,11%) isotope and the worst for ^{182}Hg (0,81%). This result is contrary to the one obtained for the absolute cross sections of these two isotopes, at least in the energy range between the $E_{lab} = 150 - 175\ MeV$.

Likewise, important results can be inferred through the analysis of the α −particle energy spectrum, which can be shown in full in **Annex 1**. From the analysis of **Fig. 4**, which shows the energy peaks of the α −particles registered from decays of the mother nuclei ^{180}Hg ($E_\alpha = 6111\ keV$) and its daughter nuclei ^{176}Pt ($E_\alpha = 5746\ keV$), can be verified that these values agree with the tabulated parameters with uncertainty around $7 - 8\ keV$, each of these peaks had the full width at half maximum (FWHM) in the range of $49 - 58\ keV$.

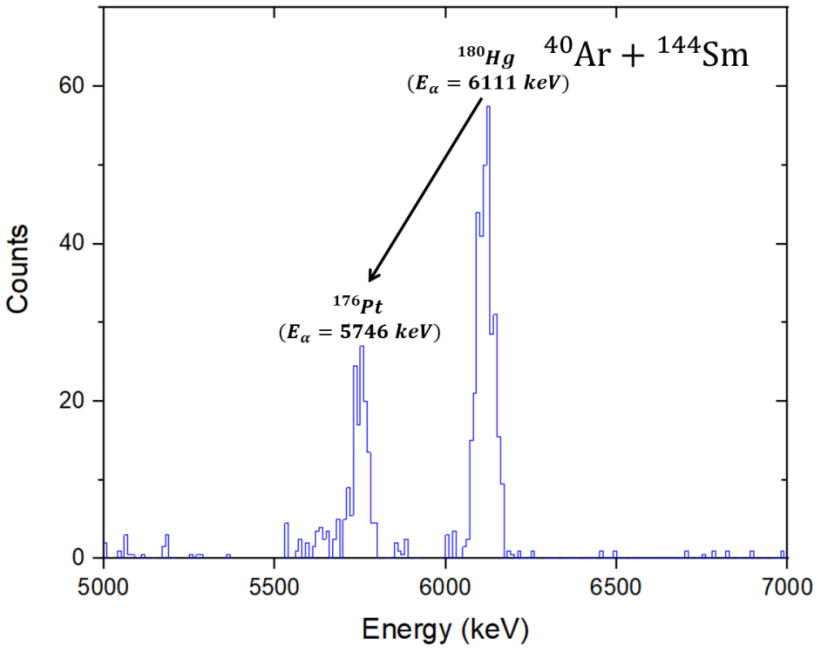

Fig. 4: Energy spectrum of the α −particles from decays of ^{180}Hg.

The corresponding yields for the reaction ^{40}Ar+^{166}Er were also measured, as well as analysed the efficiency of the separation of the $^{201\text{-}205}$Rn nucleus. From the analysis of the heat map shown in **Fig. 5**, the high separation efficiency can be inferred. It is also worth mentioning that there was no problem identifying the characteristic emission peaks of the α −particles emitted by the Rn isotopes resulting from this reaction. Regarding the statistics and processing α −peaks, it should be mentioned that average values of FWHM and uncertainty of the energy peaks, were equals to $55\ keV$ and $\pm 15.25 keV$ respectively.

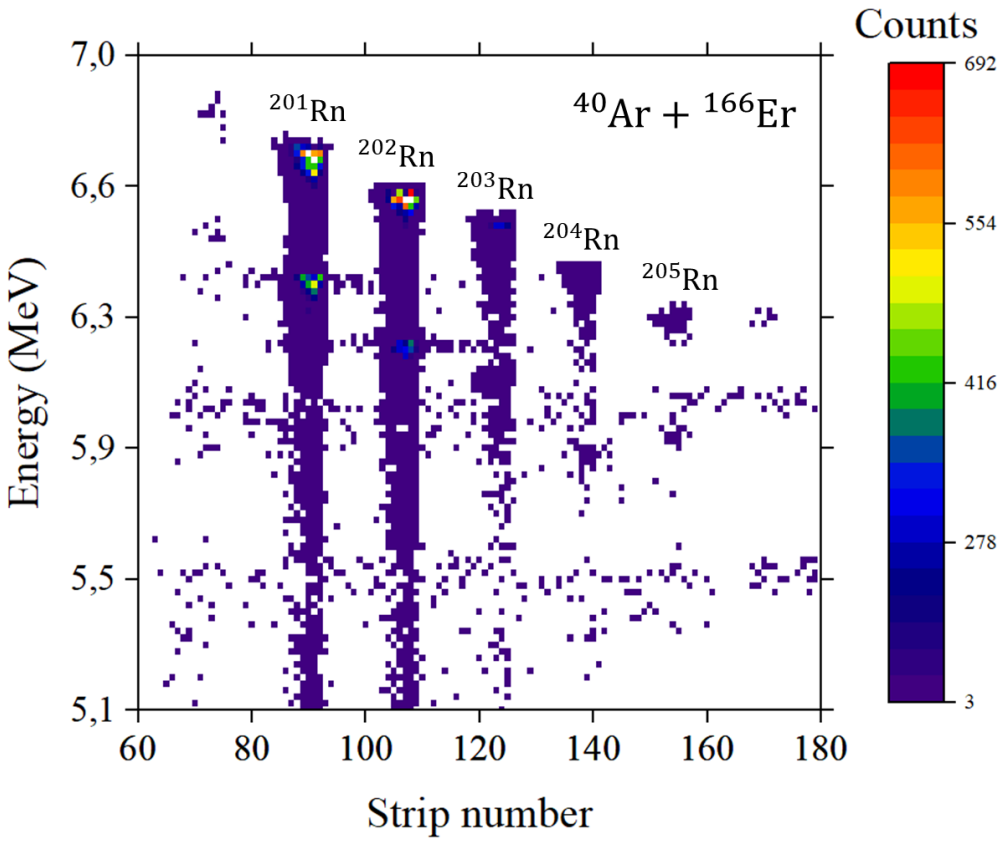

Fig. 5: The heat map for $^{201\text{-}205}$Radon isotopes measured in the reaction ^{40}Ar+^{166}Er.

In this case, as seen in **Fig. 6**, the only problem was registered in the detection of the energy spectrum corresponding to the ^{205}Rn, in which a possible characteristic peak of ^{205}At was also observed, indicating the possibility of certain problem regarding the purity of the beam.

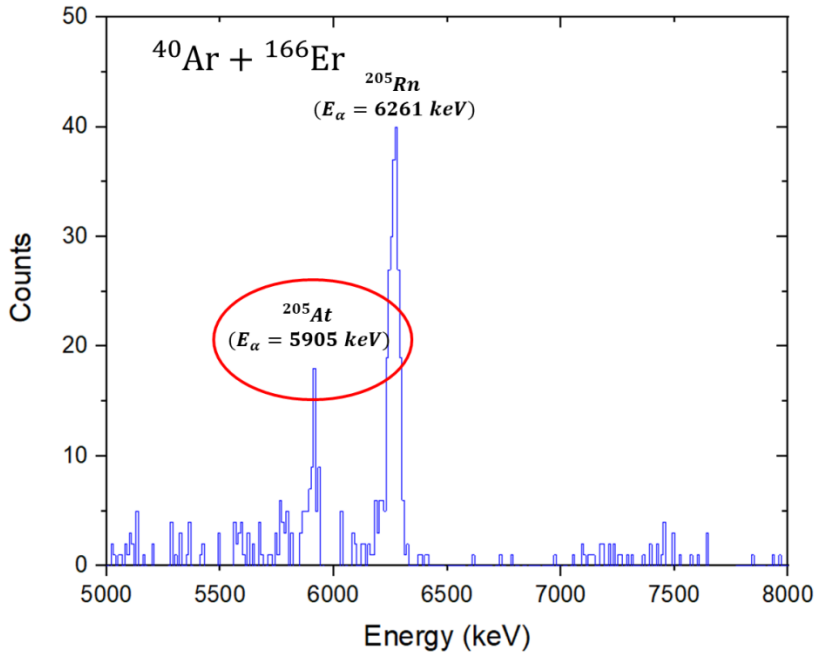

Fig.6: Energy spectrum of the α −particles from decays of ^{205}Rn nucleus.

Finally, test experiments of Rn isotope production in multinucleon transfer reaction $^{48}Ca+^{242}Pu$ were conducted to study the effect of the nuclear shell structure in those nuclei with amounts of neutrons equal to N=126 and N=152 respectively. Fortunately, since the cross-sections of these reactions are quite high, it is not very difficult to obtain a good statistic in general.

Based on the fact that measured average separation time of the separator is $1.8 \pm 0.3\ s$ [4], it can be understood that in the heat map of **Fig. 7,** only the accurate identification of the ^{212}Rn and ^{219}Rn isotopes was possible, whose half-life are 23.9 minutes and 3.96 seconds, respectively. The production and separation of ^{218}Rn could also be witnessed with some difficulty due to its $T_{1/2}$ = 35 ms. Intermediate radon isotopes are not detectable under these experimental conditions since their half-life are even lowest than those of ^{218}Rn, so they quickly decay before passing into the ion source. The above results are in accordance with those reported in the reference [1].

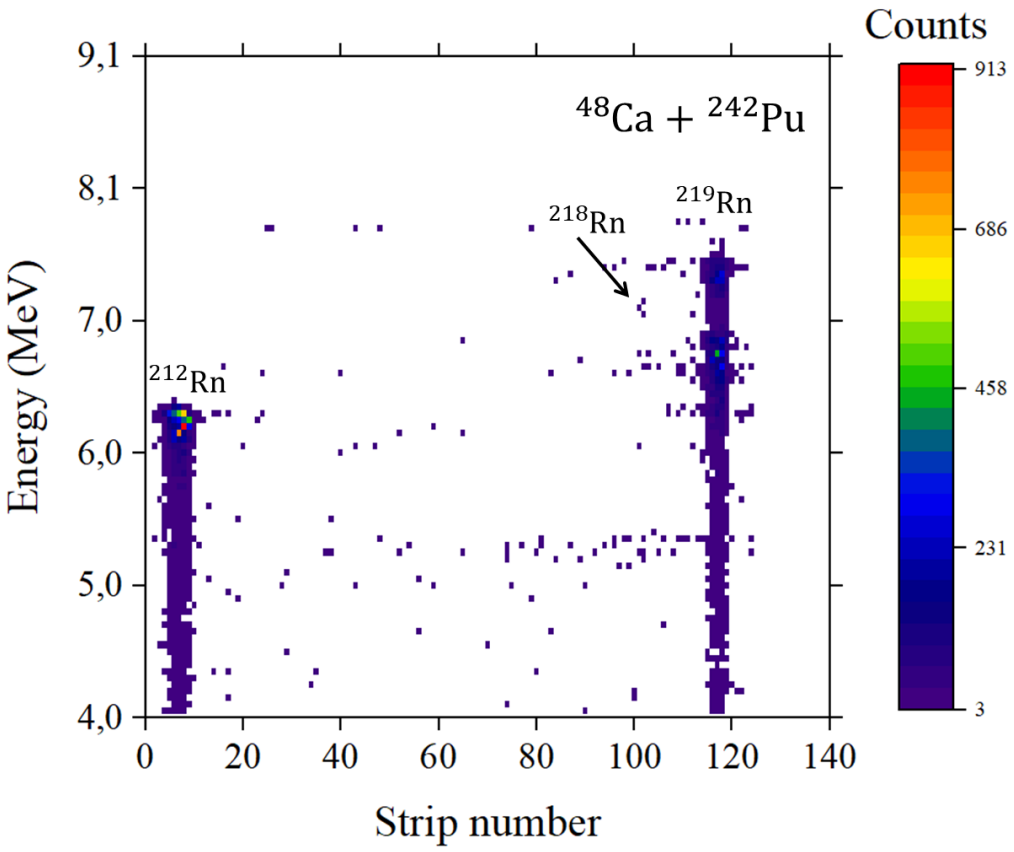

Fig. 7: Bidimensional graph of energy of α −particles vs. strip number (energy-position) registered from decays of Rn isotopes with mass numbers equal to 212 to 219.

By examining the energy spectra of α −particles alpha particles emitted by the radon isotopes produced in this reaction, some of the previously raised results might be confirmed. In that sense, the detection of the characteristic alpha emission peaks corresponding to the ^{212}RN and ^{219}RN spectra was easily performed, obtaining a notable yield. The challenge at this moment was identifying the peaks in the ^{218}RN isotope's spectrum, which was particularly challenging due to its extremely low yield.

Fig. 8 is presented to facilitate the comparison of the yield recorded for the long-lived isotope ^{219}Rn and that of the very short-lived ^{218}Rn. All of the above makes evident the selection criterion of the most viewable radon isotopes for carrying out such studies, such as those with half-lives greater than 35 ms.

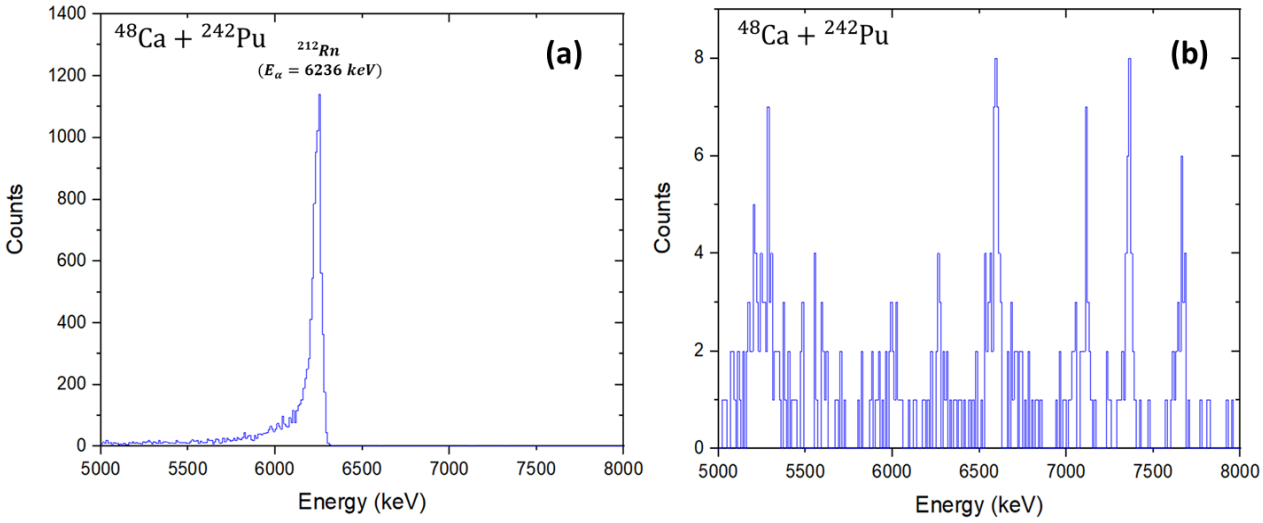

Fig.8: Energy spectrum of the α −particles from decays of: **(a)** ^{212}Rn and **(b)** ^{218}Rn isotopes.

4. Conclusion

As a summary, the results achieved in this project can be summarized in the following ideas:

- The experiments carried out have demonstrated that the complete fusion reaction ^{40}Ar+^{144}Sm is effective in producing mercury isotopes, and the mass spectrometer MASHA has shown good separation capabilities for the detection of the masses and energies of the α −particles emitted by the residual products of this reaction.
- The reaction ^{40}Ar+^{166}Er has allowed for good separation efficiency of radon isotopes, indicating a robust and reliable separation process. In this sense, only one challenge has been identified and is associated to the purity of the beam in the detection of the isotope ^{205}Rn, suggesting a need for improvements in beam quality to avoid interferences.
- In general, the analysis of the alpha particle energy spectrum has confirmed the accuracy of the measurements, with values consistent with tabulated parameters and a Full Width at Half Maximum (FWHM) within the expected range.
- The test experiments for the production of Rn isotopes through the multinucleon transfer reaction ^{48}Ca+^{242}Pu has shown certain correlation between the separation efficiency of $^{212\text{-}219}$Rn isotopes and their half-lives, with the separation and detection of the isotopes ^{212}Rn y ^{219}Rn being the most efficient due to their relatively high cross sections and half-lives.

5. Acknowledgments

I would like to express my sincere gratitude to Mr. Viacheslav Vedeneev for nice and helpful supervision. His guidance has been invaluable. To Professor Oscár Díaz Rizo for his responses to occasional technical queries. To the JINR University Centre, thank you for providing such a wonderful opportunity to learn in these different fields. It has been an enriching experience that I will always cherish. And to all the coordinators of the INTEREST program, your support has been instrumental.

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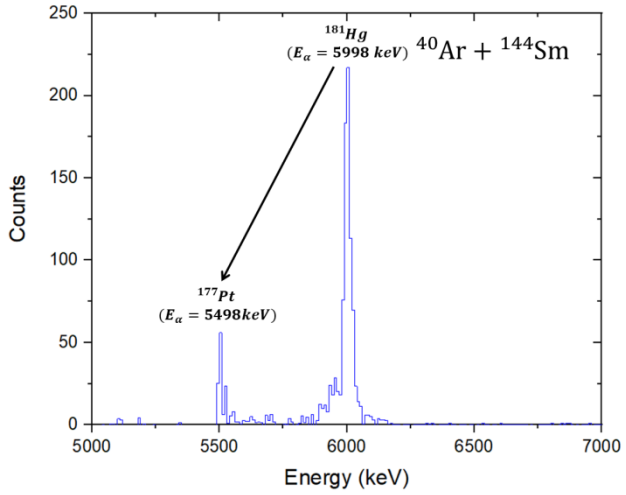
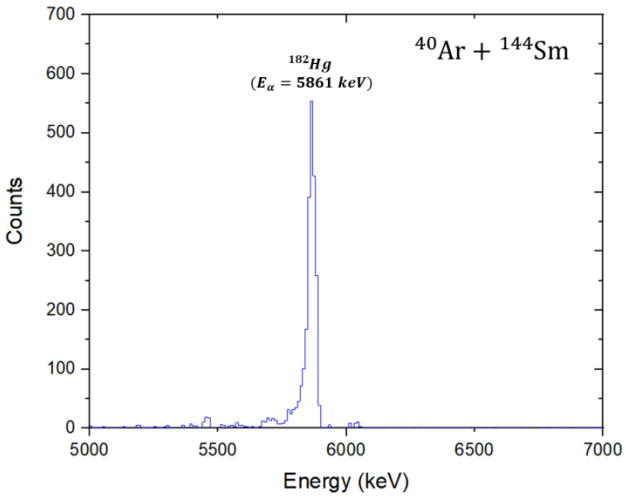
A Appendix

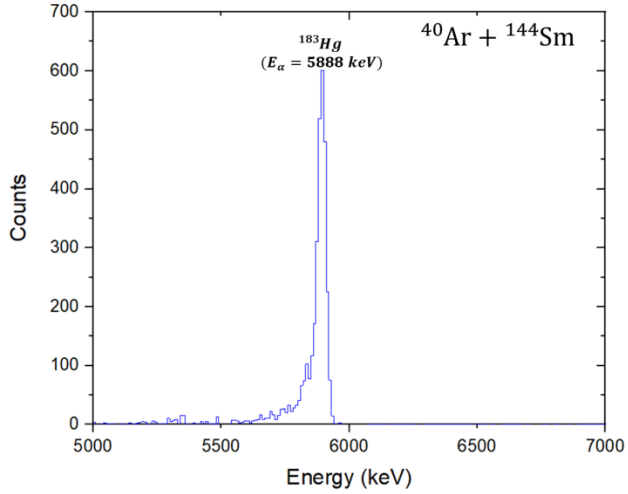
Energy spectrums registered of the α −particles from decays of $^{181\text{-}185}$Hg.



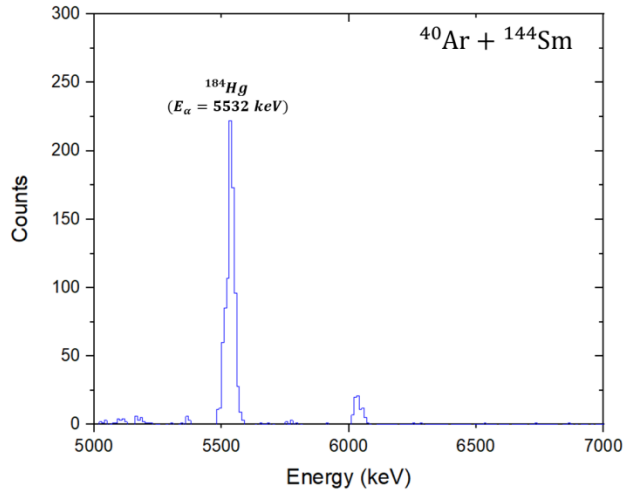
(a) ^{181}Hg and daughter nucleus ^{177}Hg.



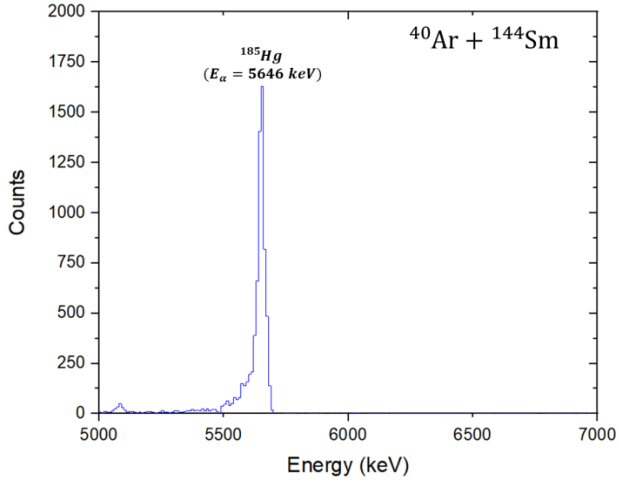
(b) ^{182}Hg.



(c) ^{183}Hg.



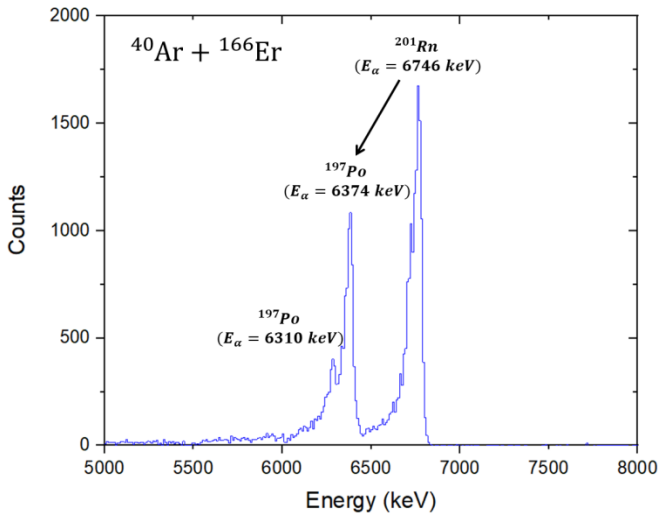
(d) ^{184}Hg



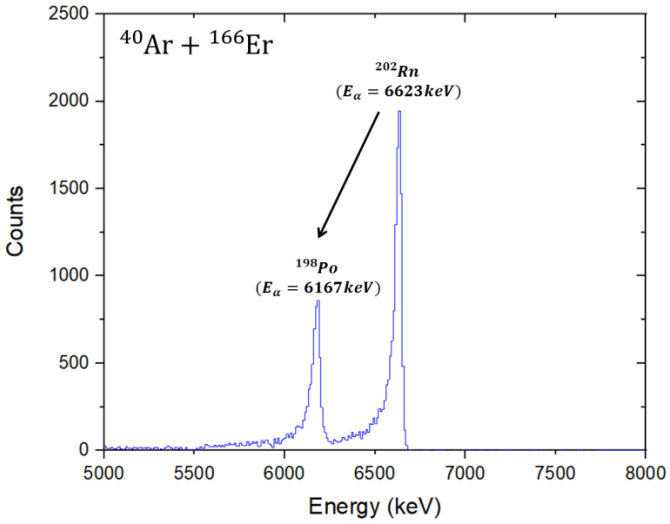
(e) ^{185}Hg.

B Appendix

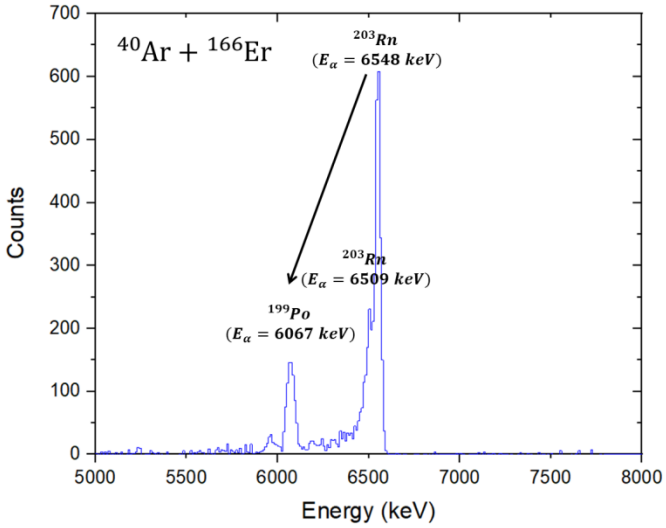
Energy spectrums registered of the α −particles from decays of $^{201\text{-}205}$Rn.



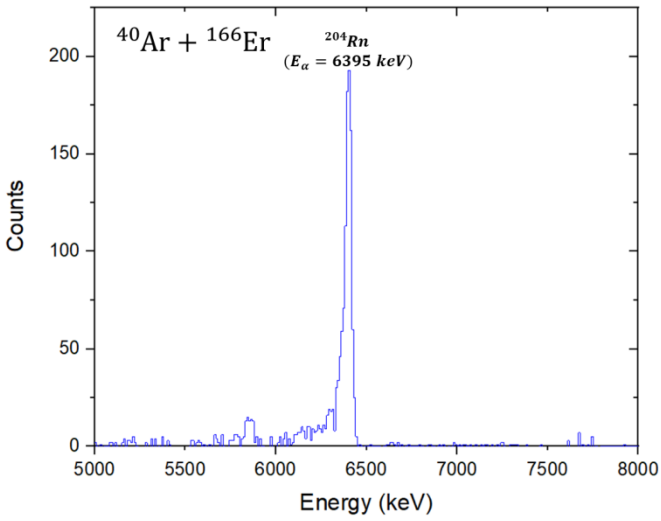
(a) ^{201}Rn and its daughter nuclei ^{197}Po.



(b) ^{202}Rn and its daughter nuclei ^{198}Po.



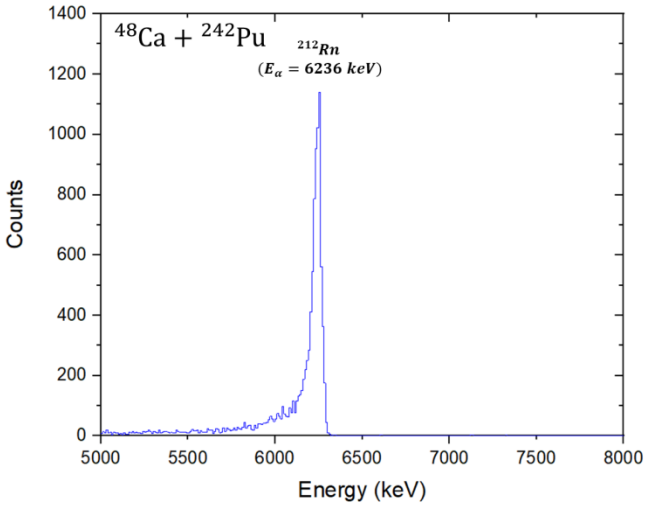
(c) ^{203}Rn and its daughter nuclei ^{199}Po.



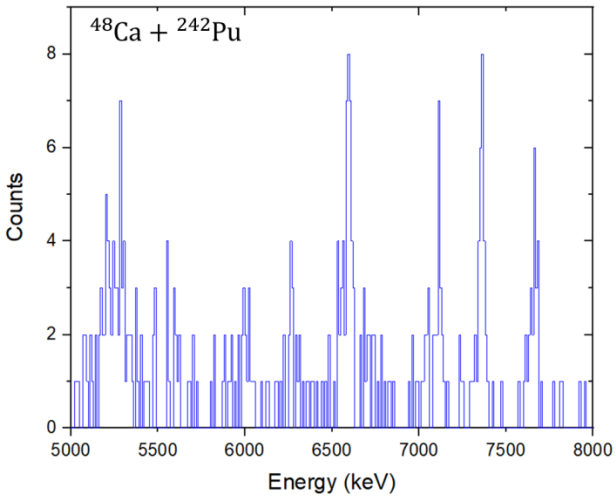
(d) ^{203}Rn and its daughter nuclei ^{199}Po.

C Appendix

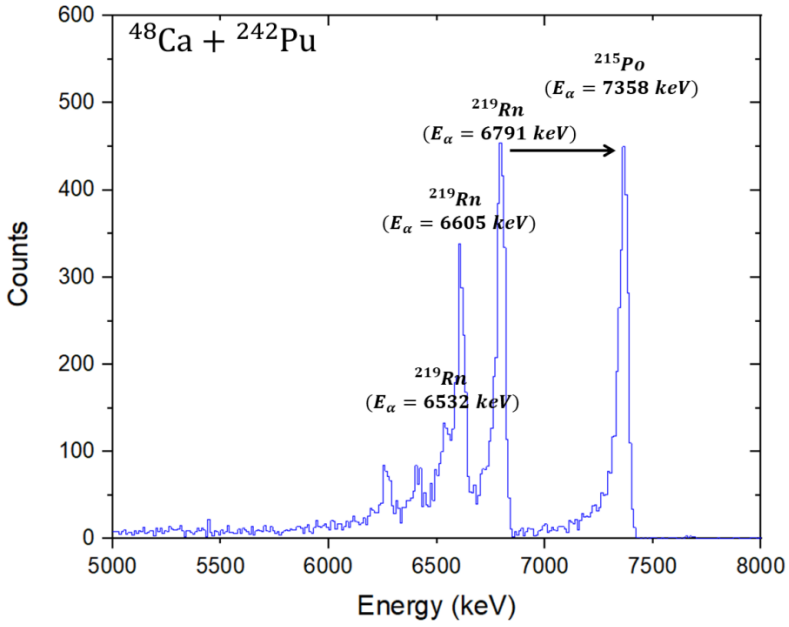
Energy spectrums registered of the α −particles from decays of ^{212,218,219}Rn.



(a) ^{212}Rn.



(b) ^{218}Rn.



(c) ^{219}Rn and its daughter nuclei ^{215}Po.