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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Wave - 8

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1. Abstract

In the experiments dealing with synthesis of superheavy nuclei, scientists primarily use radiochemistry and decay chain analysis to identify basic nuclides. However, due to the short half-lives of most superheavy nuclei (ranging from 100 microseconds to 10 milliseconds), kinematic separators are commonly employed to separate these nuclei with high reliability and efficiency. These separators do not provide information on the masses of the nuclei, though. To overcome this limitation, the MASHA mass-spectrometer was developed at the FLNR JINR in Dubna, Russia **[1].** It combines the ISOL method for creating and separating radioactive nuclei with traditional mass analysis techniques to enable the identification of the masses of newly synthesized nuclides across a broad mass range. This allows scientists to investigate the α-decays or spontaneous fission of these superheavy nuclei.

2. Introduction

The growth in the synthesis of the new nuclides have prompted for the development for the new methods for identifying synthesized nuclides have been developed using classical mass spectrometric techniques. However, unlike classical mass spectrometry, the masses of newly synthesized nuclides must be measured online, i.e., directly during their synthesis on accelerated heavy ion beams. The Isotope Separation On-Line (ISOL) method is a highly effective way to separate reaction products, specifically super heavy isotopes, so that they can be studied, and their masses determined. The ISOL system involves multiple steps, including production, thermalization, ionization, extraction, mass separation, cooling, charge-state breeding, and acceleration. However, for this method to be successful, several factors must be taken into consideration.

- Firstly, it is essential to ensure that the efficiency of separation is high. The production rate of very exotic nuclei is usually marginal, so any manipulation of the reaction products, such as ionization, purification, acceleration, and transport to the detection system, must be done with great efficiency to avoid losing these "precious" nuclei.
- Secondly, the selectivity of the separation process is crucial. In nuclear reactions, unwanted, more stable nuclei are typically produced more abundantly. Additionally, ISOL systems often generate beams of isotopes from the target material itself or from other components of the target-ion source system. Therefore, the separation process should effectively distinguish between the desired and undesired species.
- To minimize losses caused by radioactive decay between production and arrival at the experimental set-up, the time of separation must be kept short when dealing with short-lived exotic nuclei. Additionally, careful consideration must be given to the choice of catcher material in ISOL, which can include solid, liquid, or gas catchers, with each type having specific applications. For our particular application involving carbon nanomaterial, a solid catcher proved to be the most efficient and speedy option. This approach was implemented using the MASHA setup.**[2]** The Mass Analyzer of Super Heavy Atoms (MASHA) has been created by FLNR, JINR. This mass analyzer possesses unique capabilities, including the ability to measure the masses of synthesized superheavy element isotopes and simultaneously detect their α decays and/or spontaneous fission.

3. MASHA Arrangement and Essential Setup

The MASHA setup consists of following setup:

1. Target box:

After the recoil nuclei exit the target, they are implanted into a heated catcher maintained at a temperature range of approximately 1800-2000K. The target itself is a rotating wheel consisting of six cassettes with two sectors each, providing better efficiency and heat distribution compared to a stationary target. The thickness of the target is determined by the range of the recoil nuclei in the working layer, which depends on the kinetic energy of the heavy atom produced from the fusion reaction.

The hot catcher material is made of thermally expanded graphite with a porous polygraphene structure, having a 75% porosity and a density of 1 g/cm3, and shaped as a 30 mm diameter disk with a thickness of 0.6 mm. The hot catcher operates at a delivery time of 1.8±0.3 s to transport nuclides to the ion source (ECR), with the separation time determined using the beam interruption method.

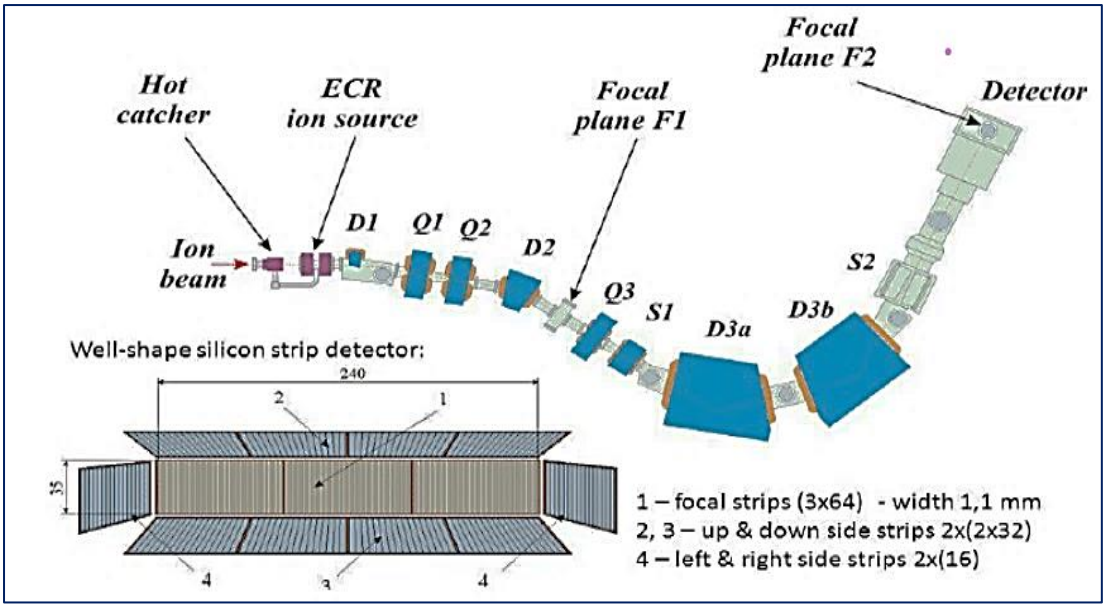


Fig. 1

MASHA schematic diagram

2. Ion source:

The setup uses an ECR ion source with a 2.45 GHz frequency to ionize atoms of nuclear reaction products. The source achieves almost 100% singly ionized atoms, and the ionization efficiency of noble gases is as high as 90%. A hot catcher is used to inject products of complete fusion reactions into the ECR source. The primary beam of heavy ions passes through a diagnostic system composed of a split-type aperture of the electrostatic induction sensor to control the beam position relative to the ion guide. Nuclear reaction products escape from the target, pass through the separating foil, and are stopped in the graphite absorber, which is heated to a temperature of 1500–2000 K. The products diffuse from the graphite absorber to the vacuum volume of the hot catcher and reach the ECR source.

3. Detection and Control System:

A special strip detector and well-type silicon detector are used to measure low direct currents and detect decays of nuclear reaction products, respectively, in the mass spectrometer. The strip detector is an exact copy of the frontal part of the silicon detector and has 192 strips with a pitch of 1.25 mm. A multichannel electronic module is developed for the strip detector to measure low currents. The well-type of silicon detector is installed in the focal plane of the mass spectrometer and covers a 240 × 35 mm area with 192 strips.

Four side detectors are also installed to increase the detection efficiency. Both detectors have a dead layer thickness of less than 50 nm, and the signals from each strip are read out independently using preamplifiers and shaping amplifiers. Two separate data acquisition programs are used for each detector.

4. The mass separator and DAQ in focal plane:

In this setup, a magnetic-optical analyser functions as the mass separator , wherein the ions are separated based on their magnetic rigidity in a constant magnetic field. The mass determination of super heavy atoms is achieved with a high degree of accuracy, with a precision of Δm=0.25-0.30 e.m.u. Detectors are positioned in the focal plane of the magnetic analyzer to detect the position and decay of the separated atoms. The use of well-type position sensitive strip detectors, comprising focal, side, and lateral crystals, enables the registration and determination of the masses and decay energies of both evaporation residues and their daughter decay products, with a greater geometric efficiency. However, to ensure accurate registration of the atoms, it is crucial to eliminate any alpha-particle background resulting from the decay of target-like nuclei, particularly those produced in deep-inelastic collisions or quasi-fission, such as the light isotopes of actinide elements (Th and U), which are around 40-60 e.m.u. away from the mass of the superheavy atom. These nuclei can be effectively separated at the intermediate focal plane.**[4]**

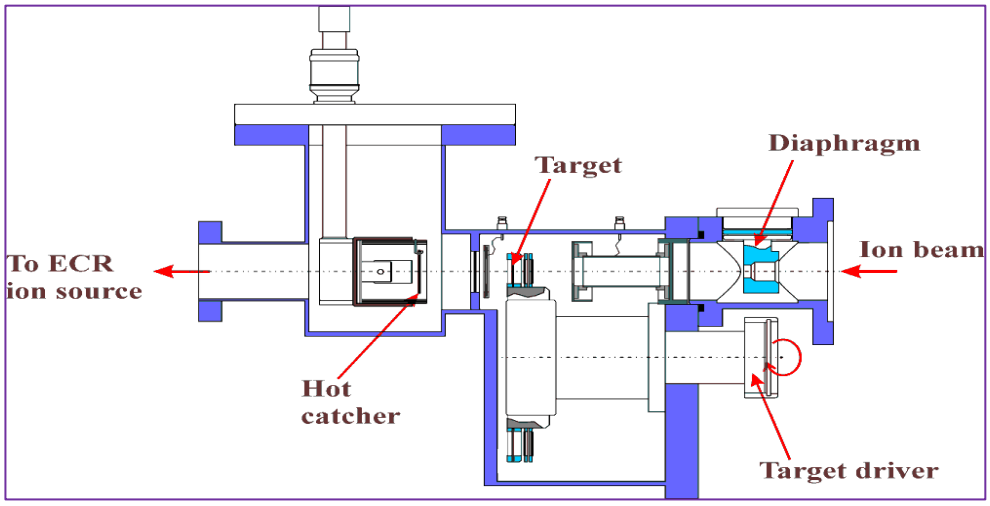


Fig 2.
Target assembly and hot catcher

- **Improvements in MASHA setup [3]:**

The main units of the mass-spectrometer MASHA were modernized, including the hot catcher, ECR ion source, vacuum chambers, hot transport system, and an additional strip detector for separation efficiency control. The hot catcher design now includes a poly-graphene heater and thin graphene foil or carbon nanotube paper catcher to prevent heating and radiation destruction. The ECR ion source has a ceramic coating, allowing for operation at high temperatures, and glass-enamel coating for detecting non-volatile elements close to volatile ones.

Improvement in Ion source setup:
The walls of the source chamber, catcher, and transportation line are covered with titanium nitride to increase ionization efficiency and outgoing time. The optimization of the ECR ion source parameters, such as power, frequency of the microwave generator, and pressure of the buffer gas, is crucial for effective use of the ion source.

Graphite stopper improvement:
To improve the stability of the separation efficiency of the MASHA setup during experiments with high beam intensity, an additional graphene foil of 0.6 mg/cm2 thickness was added 2.5 mm ahead of the main heater at the beam axis. The graphene foil heats up from the main heater through radiation and takes some heating load caused by the beam. It also serves as a separator for low-energy reaction products that stop inside the thin foil, while the beam passes through it and gives almost its entire energy to the graphite heater. This improvement prevents the high beam intensity from corrupting the "Hot Catcher" and decreasing its separation efficiency by more than five times in just a few days.

Improvement in Control System:
Development, realizing and testing of a new controlling system for upgraded MASHA setup based on modular WAGO-I/O-SYSTEM was done. The ion source, hot catcher and target box was replaced with new ones based on modular WAGO-I/O-SYSTEM and integrated with mass-separator into the one common controlling system. The new software, written in LabVIEW, was developed and tested for the controlling system.

4. ISOL Method

The Isotope Separation OnLine (ISOL) method is used for determining short-lived isotopes by cooling and stopping reaction products and analyzing them magneto-optically and electrostatically. The ISOL method is applied in mass analysis and enables separation from the primary beam in an online mode. An experiment was conducted on the U-400M heavy ion beam at MASHA facility, FLNR, JINR to study the application of new carbon nanomaterials and determine their radiation resistance for the ISOL method. Previous research with thermally expanded graphite heat catcher showed incompatibility with high-intensity beams, but

improvements were made to enable synthesizing new products at beam intensities of up to 0.5 pμA and higher for the SHE factory perspective.

5. RESULTS:

Given: Experimental Data of 3 reactions

$^{40}Ar + {}^{148}Sm \longrightarrow {}^{188-xn}Hg + xn$

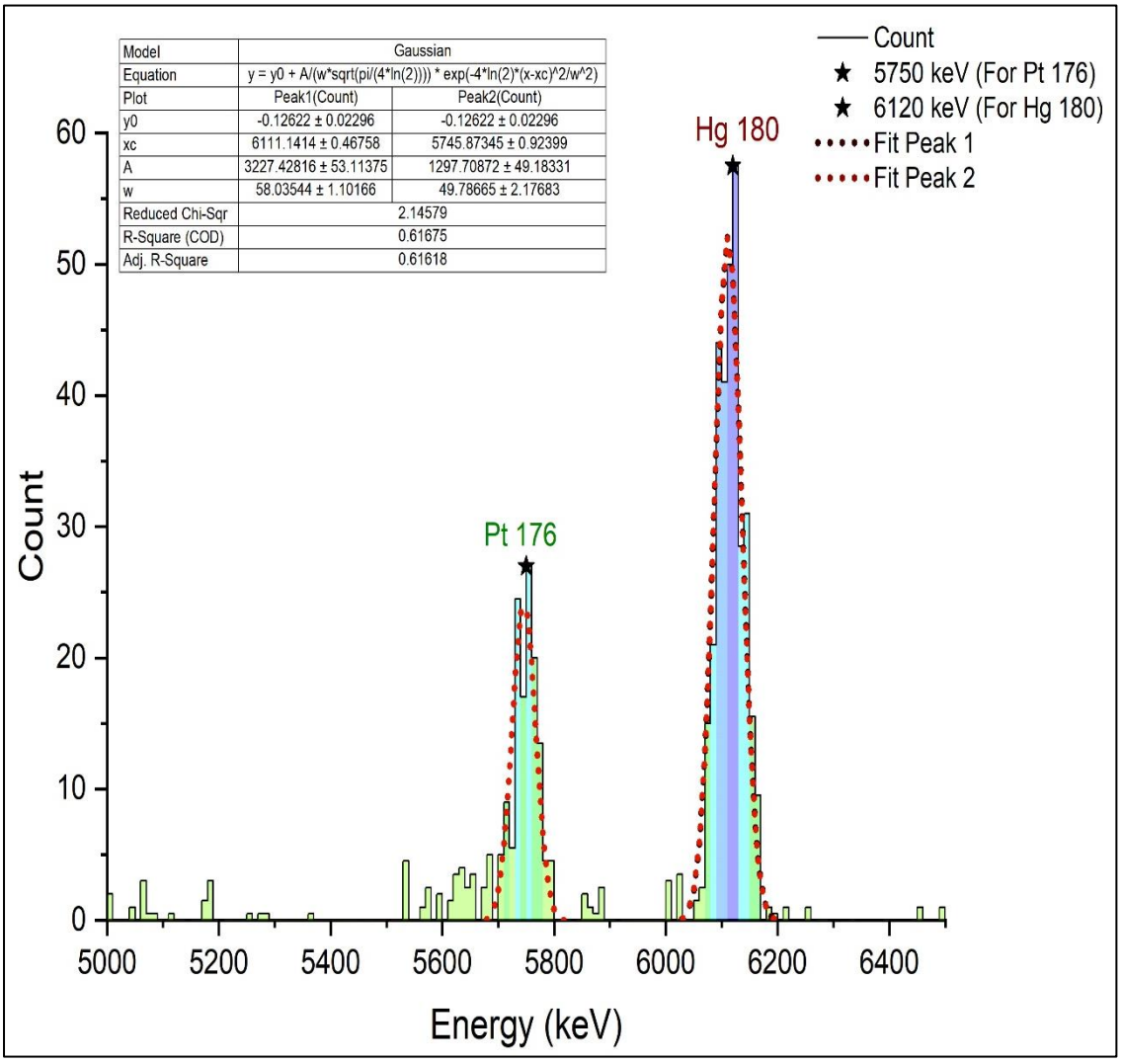
$^{40}Ar + {}^{166}Er \longrightarrow {}^{206-xn}Rn + xn$

$^{48}Ca + {}^{242}Pu \longrightarrow {}^{21x}Rn$

Task : To analyse the data and plot the histogram using Origin Pro software and perform the peak analysis and understand the alpha decay energy of the reactions and draw their contour heat maps.

1. Histograms of Hg

i. Hg^{180} : $^{40}Ar + {}^{148}Sm \longrightarrow {}^{188-xn}Hg + xn$



Hg^{180} :
Half Life: 2.58 sec

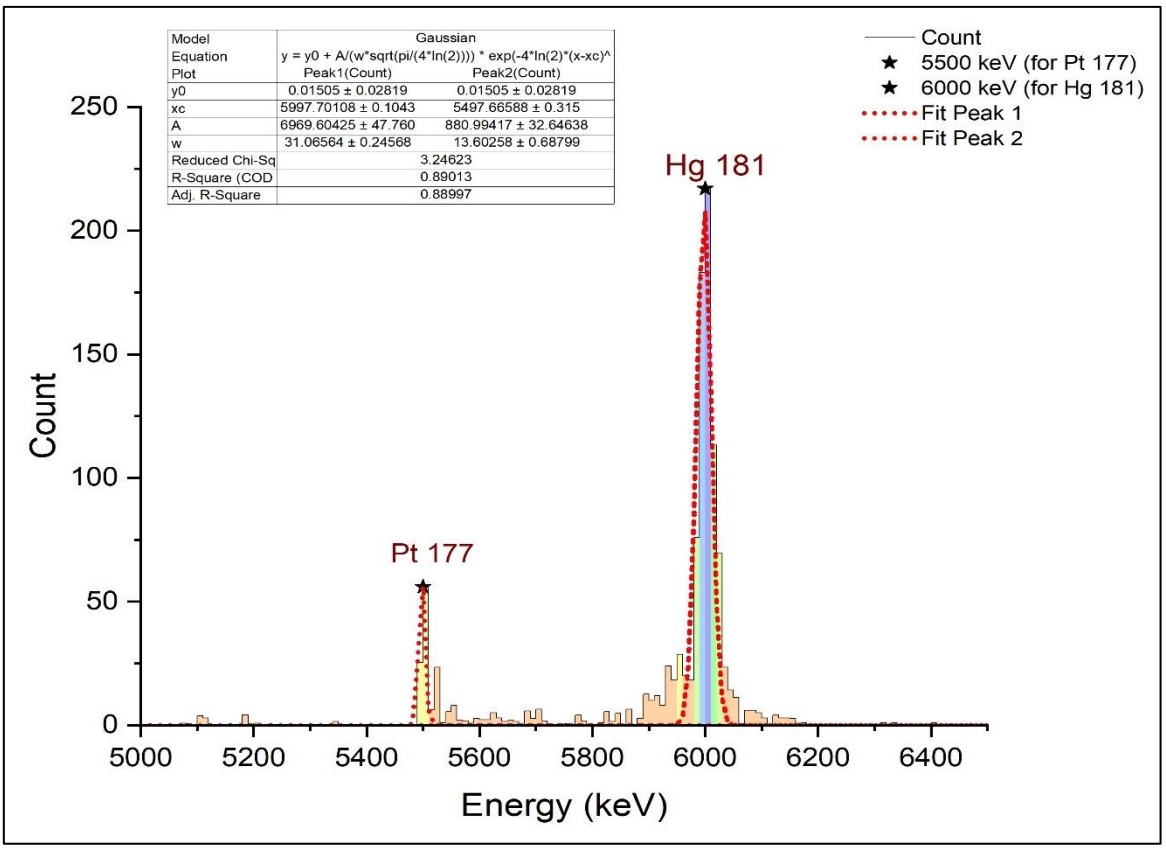
Hg^{180} decays by 48% at 6120keV α-Decay energy level

Pt^{176} :
Half life: 6.35sec

Pt^{176} decays by 40% at 5750keV α-Decay energy Level

Note: Half-life and α-Decay energy level obtained from Nuclides Chart

ii. Hg^{181} :



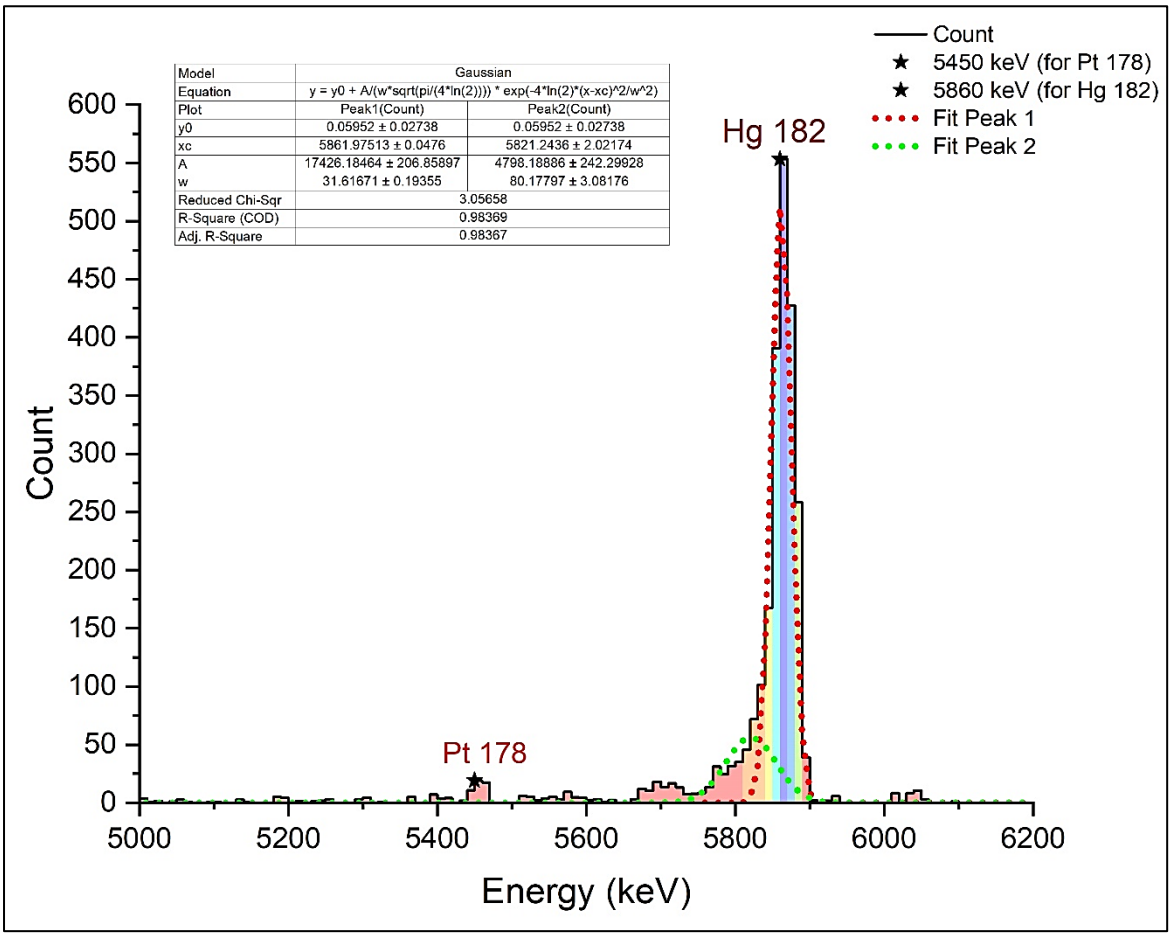
Hg^{181} :
Half Life: 3.5 sec

Hg^{181} decays by 30% at 6000keV α-Decay energy level

Pt^{177} :
Half life: 11 sec

Pt^{177} decays by 5.6% at 5500keV α-Decay energy Level

iii. Hg^{182} :



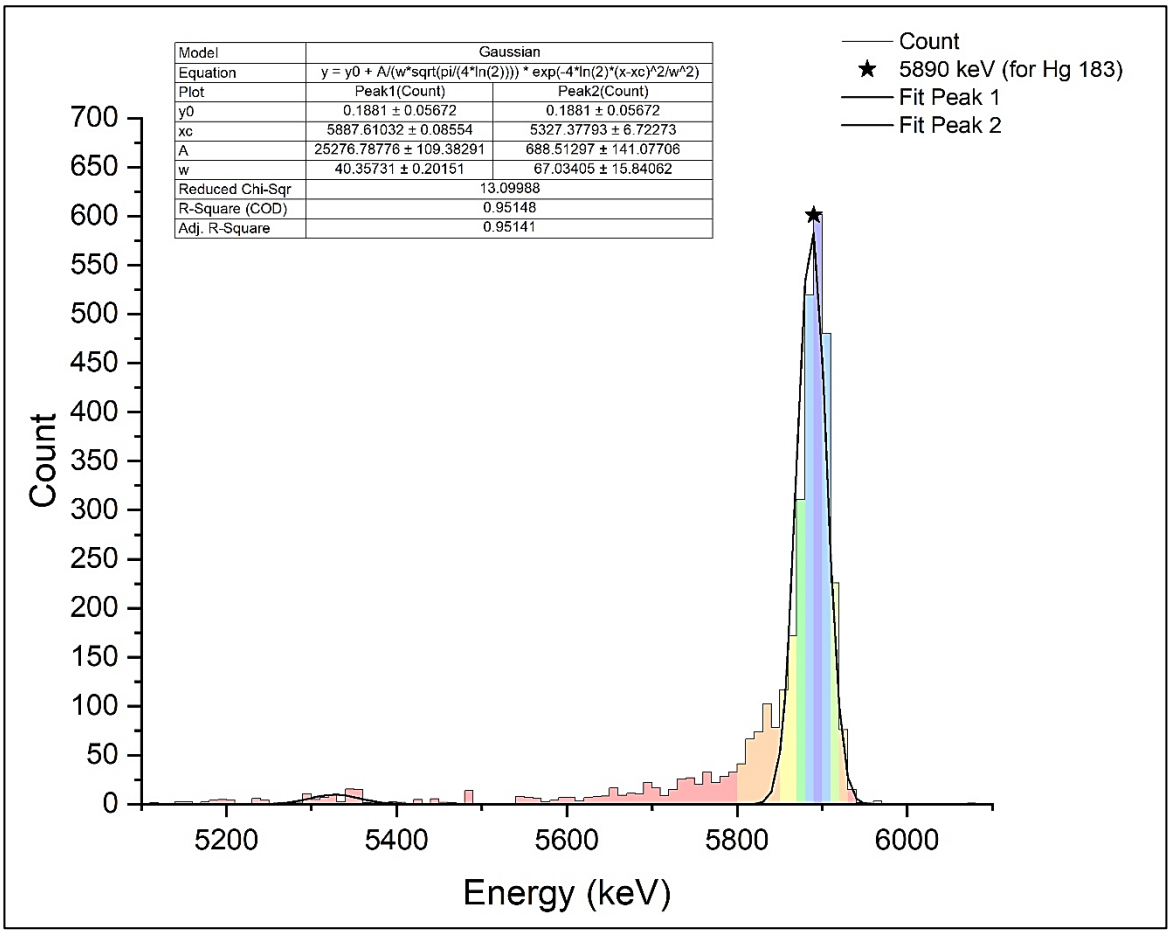
Hg^{182} :
Half Life: 10.835 sec

Hg^{182} decays by 15.2% at 5860 keV α-Decay energy level

Pt^{178} :
Half life: 21.1sec

Pt^{178} decays by 4.6% at 5450keV α-Decay energy Level.

iv. Hg^{183} :



Model	Gaussian	
Equation	y = y0 + A/(w*sqrt(pi/(4*ln(2)))) * exp(-4*ln(2)*(x-xc)^2/w^2)	
Plot	Peak1(Count)	Peak2(Count)
y0	0.1881 ± 0.05672	0.1881 ± 0.05672
xc	5887.61032 ± 0.08554	5327.37793 ± 6.72273
A	25276.78776 ± 109.38291	688.51297 ± 141.07706
w	40.35731 ± 0.20151	67.03405 ± 15.84062
Reduced Chi-Sqr	13.09988	
R-Square (COD)	0.95148	
Adj. R-Square	0.95141	

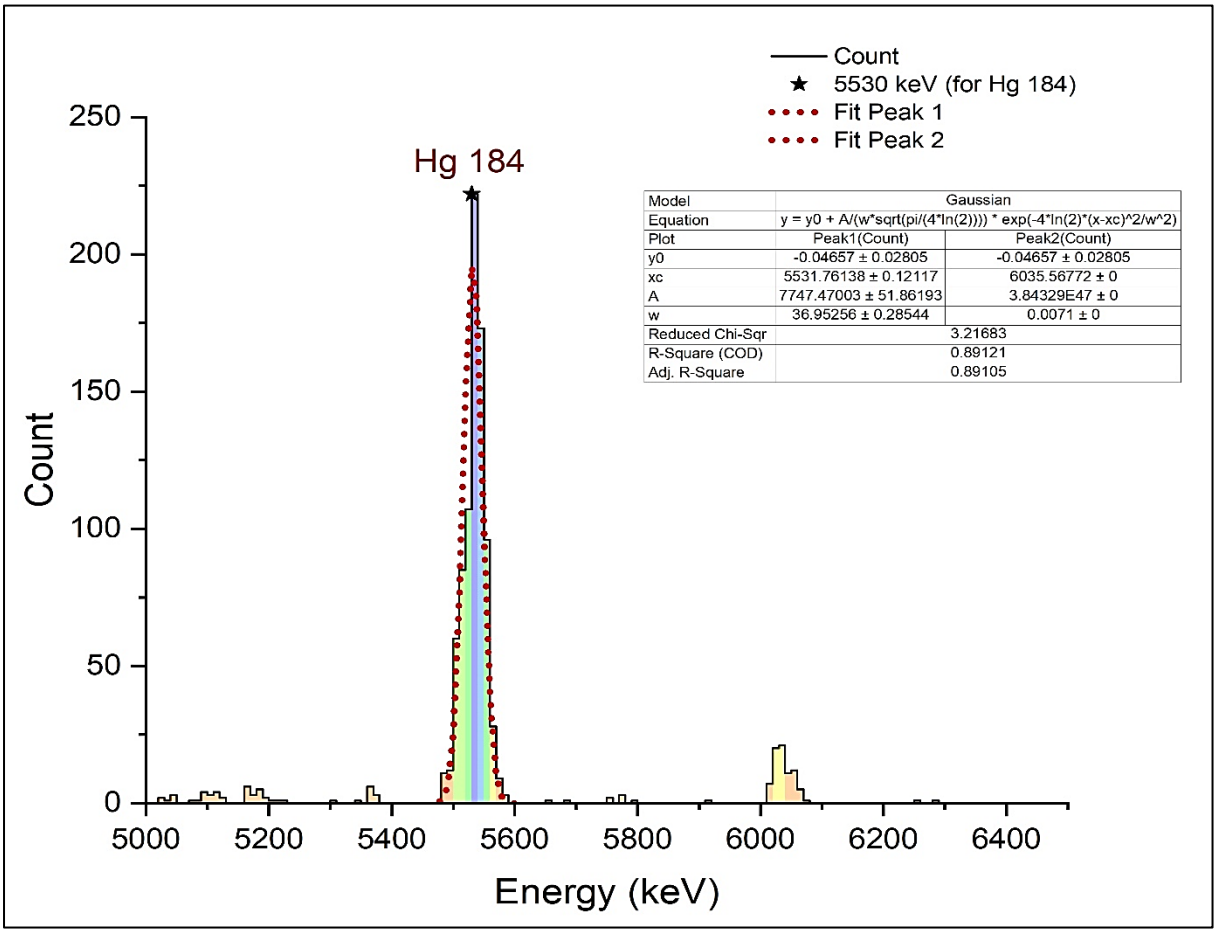
Hg^{183} :
Half Life: 9.4 sec

Hg^{183} decays by 11.7% at 5890 keV α-Decay energy level

Pt^{179} :
Half life: 21.1 sec

Pt^{179} decays by 0.24% at 5200keV α-Decay energy Level.

v. Hg^{184} :



Model	Gaussian	
Equation	y = y0 + A/(w*sqrt(pi/(4*ln(2)))) * exp(-4*ln(2)*(x-xc)^2/w^2)	
Plot	Peak1(Count)	Peak2(Count)
y0	-0.04657 ± 0.02805	-0.04657 ± 0.02805
xc	5531.76138 ± 0.12117	6035.56772 ± 0
A	7747.47003 ± 51.86193	3.84329E47 ± 0
w	36.95256 ± 0.28544	0.0071 ± 0
Reduced Chi-Sqr	3.21683	
R-Square (COD)	0.89121	
Adj. R-Square	0.89105	

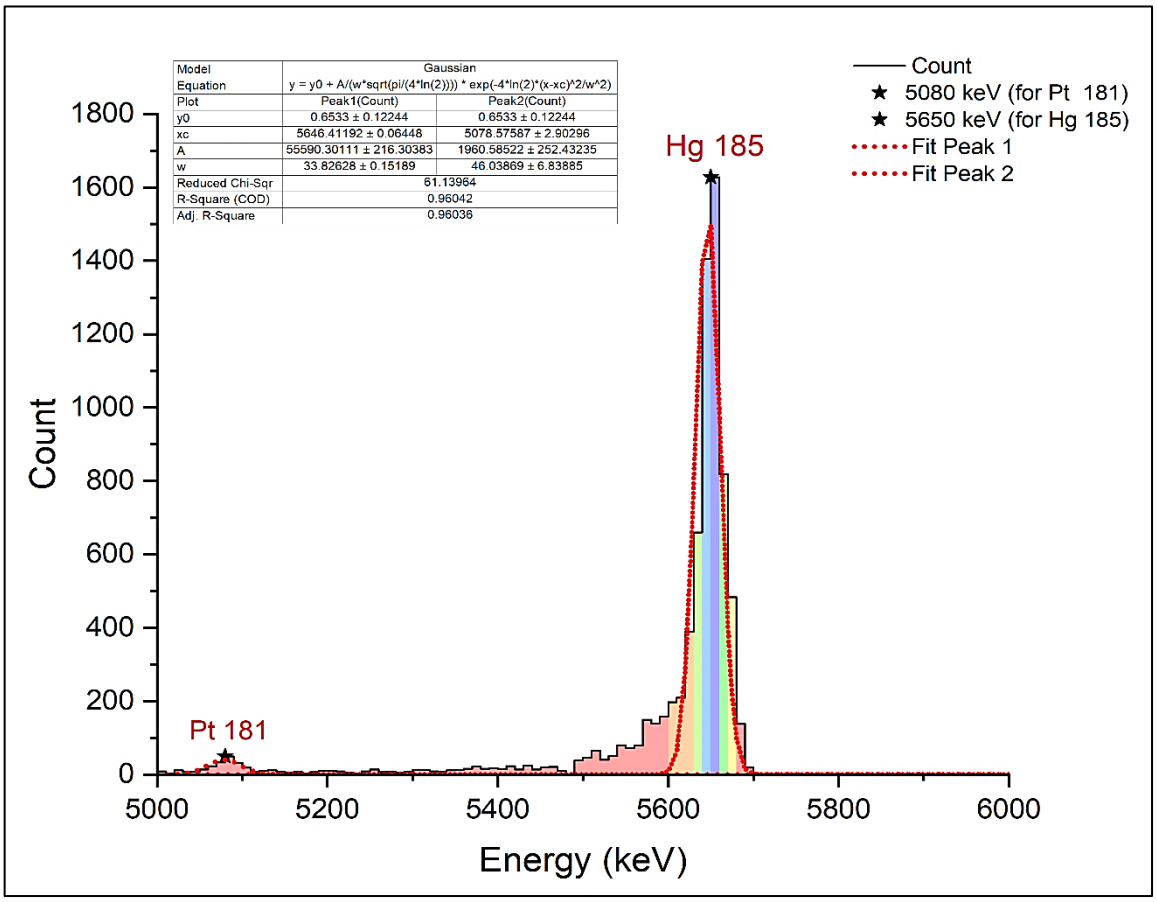
Hg^{184} :
Half Life: 30.9 sec

Hg^{184} decays by 1.26% at 5530 keV α-Decay energy Level

Pt^{180} :
Half life: 56sec

Pt^{180} decays by 0.3% at 5140keV α-Decay energy Level.

vi. Hg^{185} :



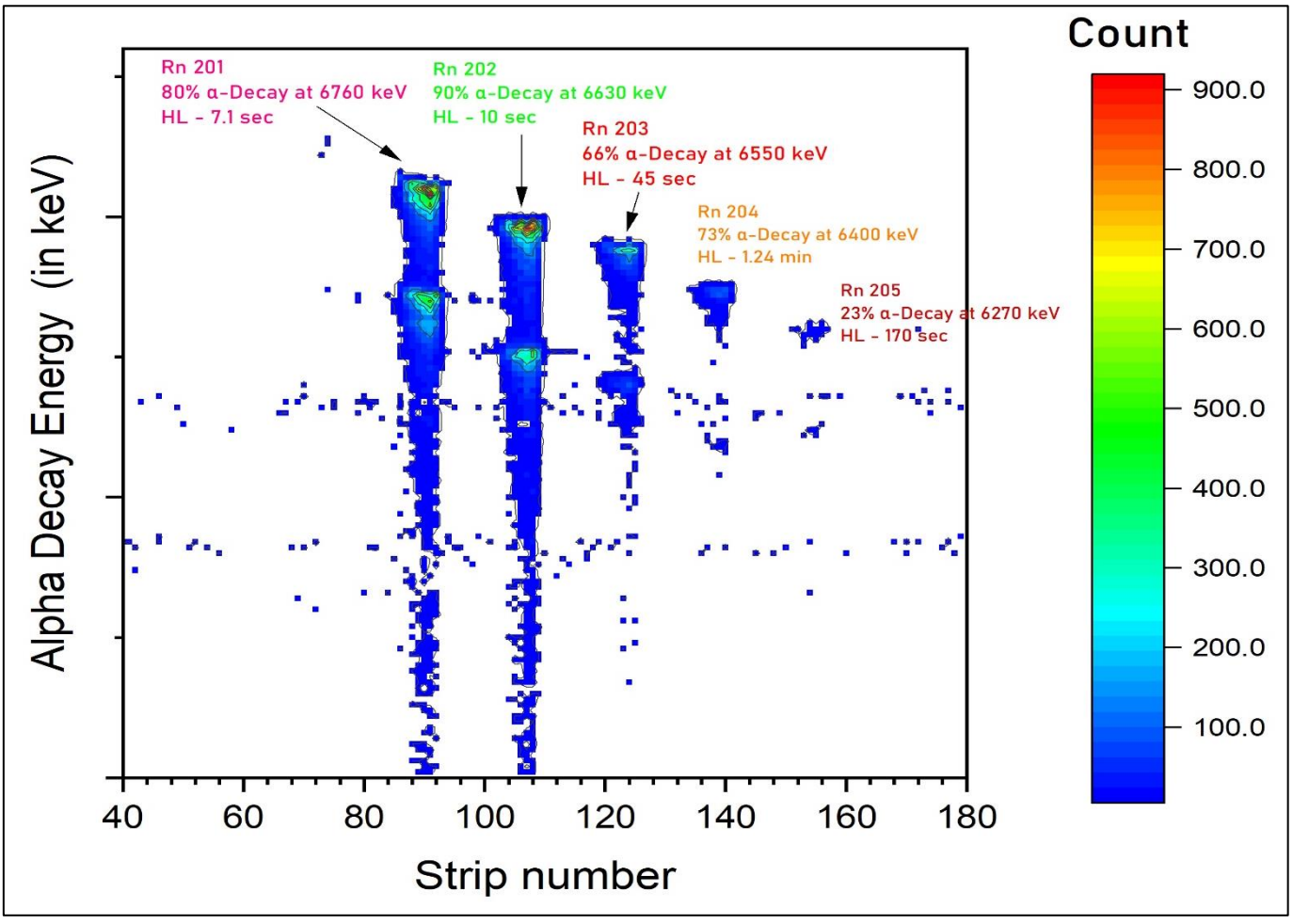
Hg ^{185} :
Half Life: 49.1 sec

Hg^{185} decays by 16% at 5650 keV α-Decay energy Level

Pt^{181} :
Half life: 52sec

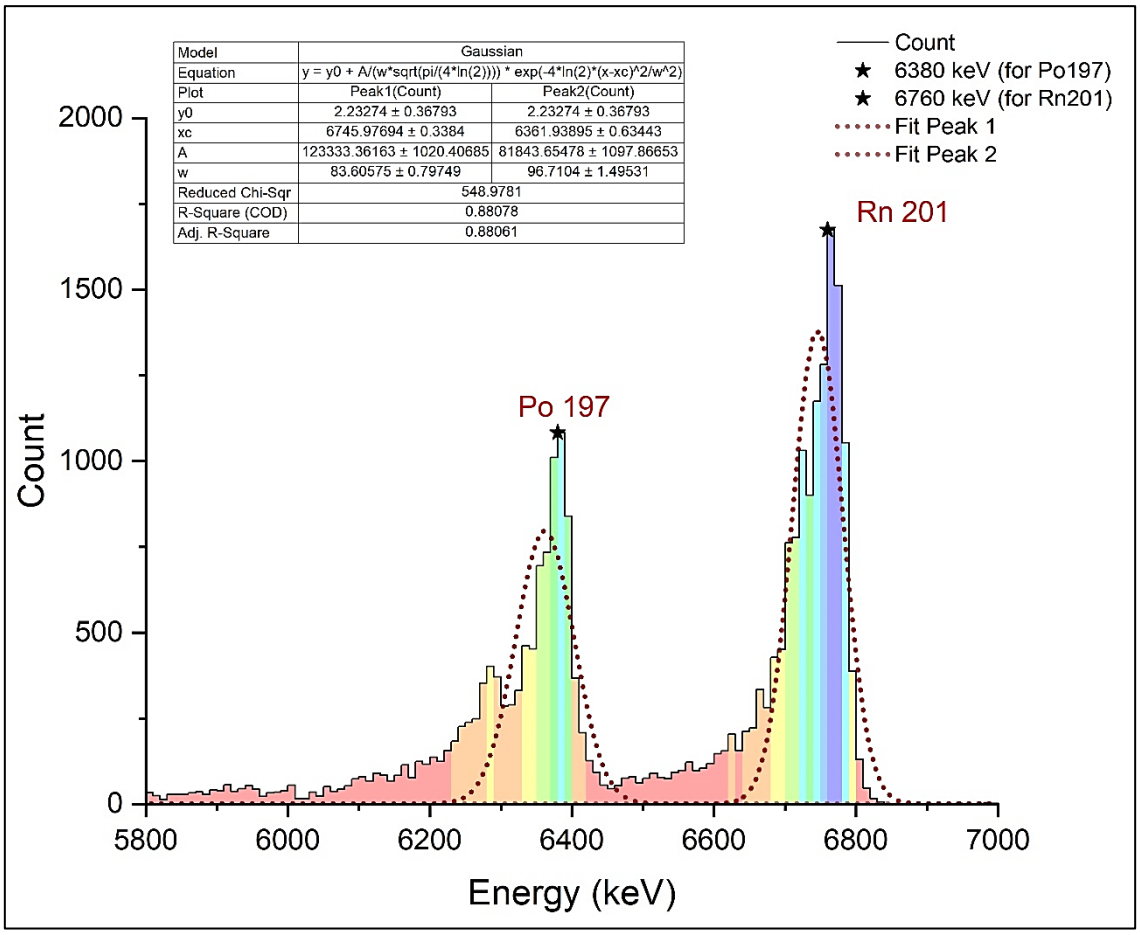
Pt^{181} decays by 0.074% at 5080 keV α-Decay energy Level.

➢ Heat Map for Mercury isotopes ($Hg^{180-185}$ isotopes)



2. Histograms of $Rn^{201 - 205}$

i. Rn^{201} :

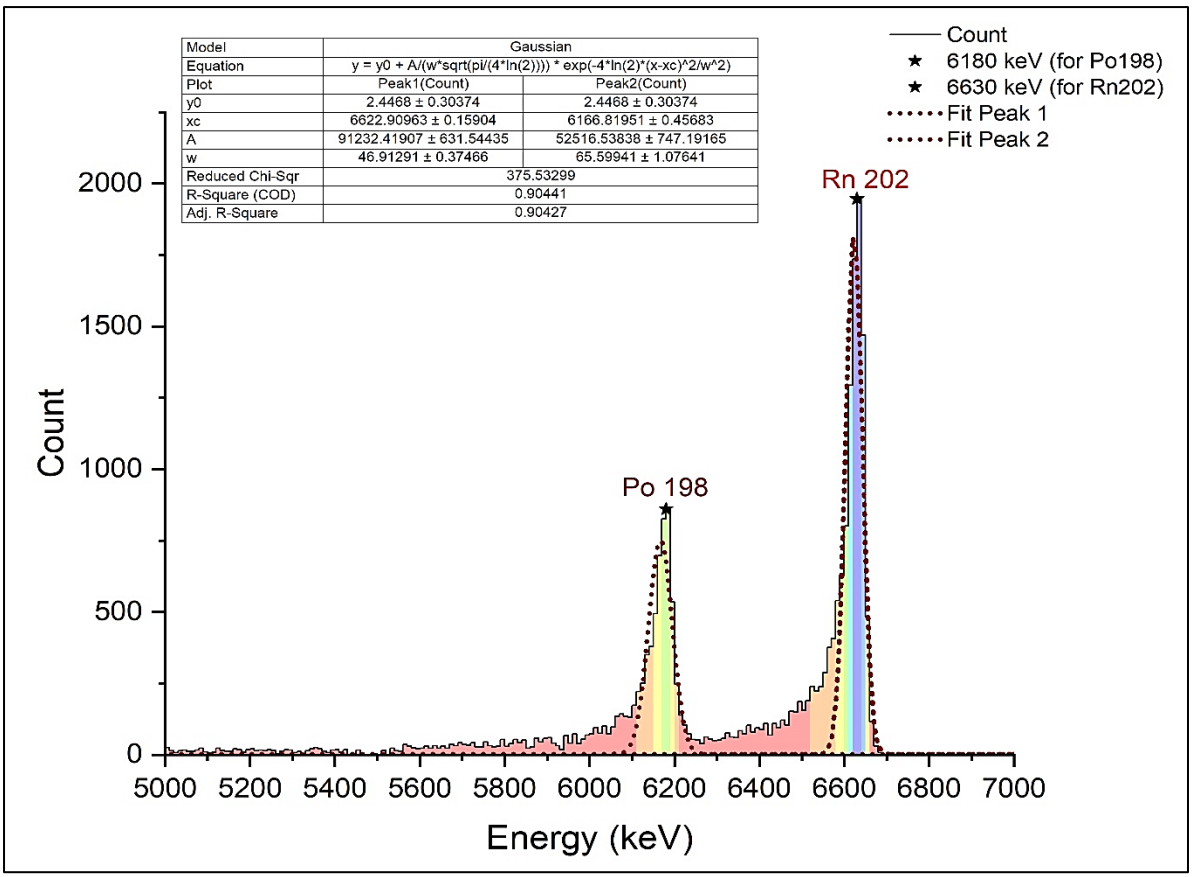


Rn^{201} :
Half Life: 7.1 sec

Rn^{201} decays by 80% at 6760keV α-Decay energy level

Po^{197} :
Half life: 53.6 sec
Po^{197} decays by 44% at 6380keV α-Decay energy Level
Also,
Half life: 25.8 sec
Po^{197} decays by 84% at 6380keV α-Decay energy Level

ii. Rn^{202} :

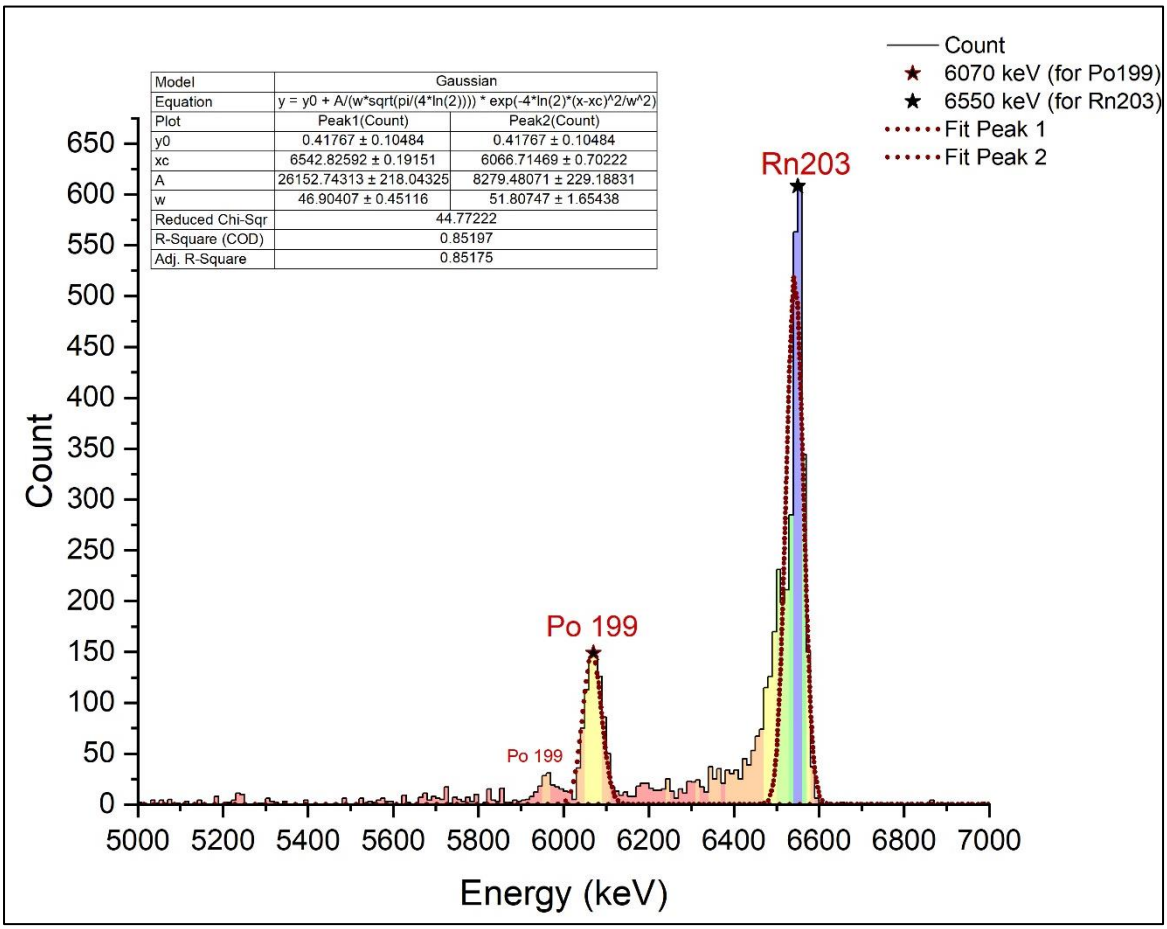


Rn^{202} :
Half Life: 10 sec

Rn^{201} decays by 90% at 6630keV α-Decay energy level

Po^{198} :
Half life: 1.77 min
Po^{198} decays by 57% at 6180keV α-Decay energy Level

iii. Rn^{203} :

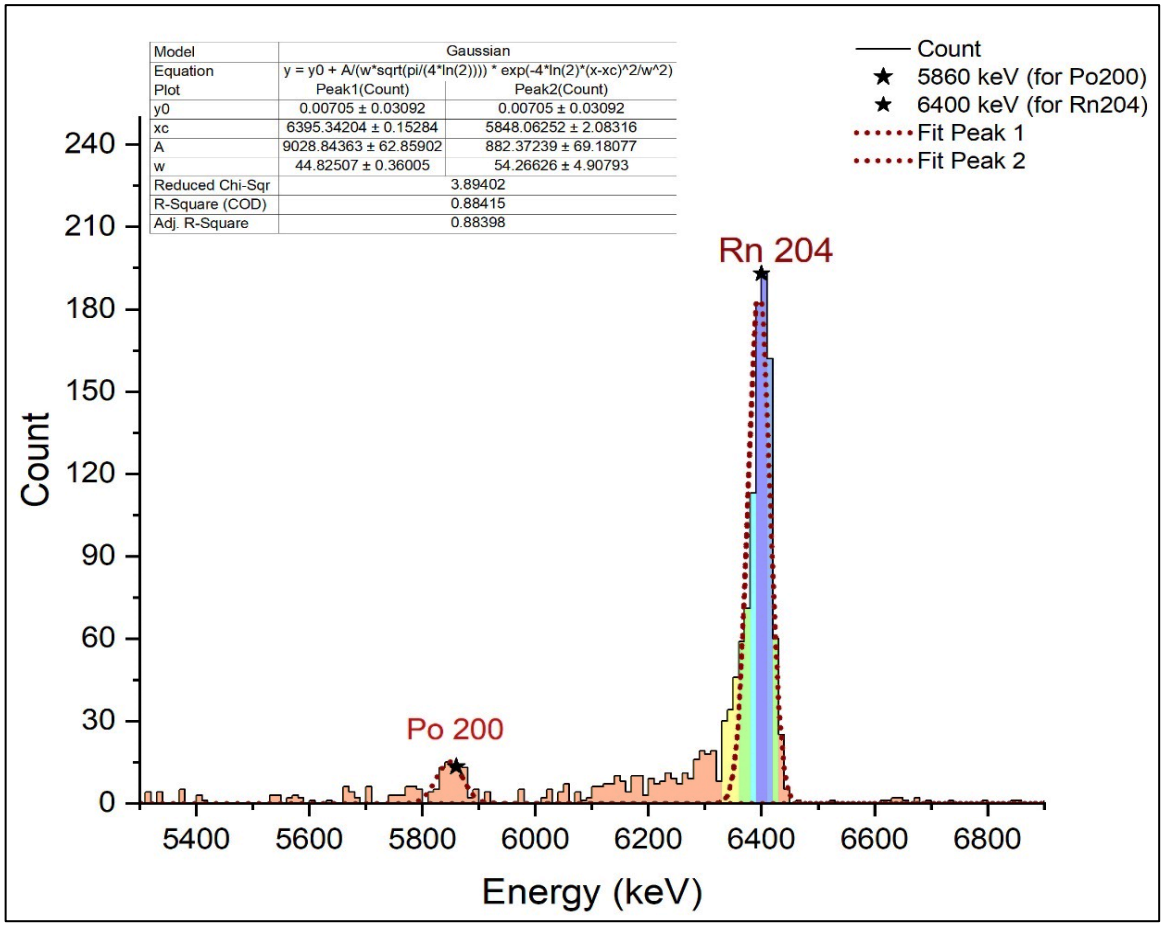


Rn ^{203} :
Half Life: 45 sec

Rn^{203} decays by 66% at 6550keV α-Decay energy level

Po^{199} :
Half life: 5.48 min
Po^{199} decays by 12% at 6070keV α-Decay energy Level

iv. Rn^{204} :



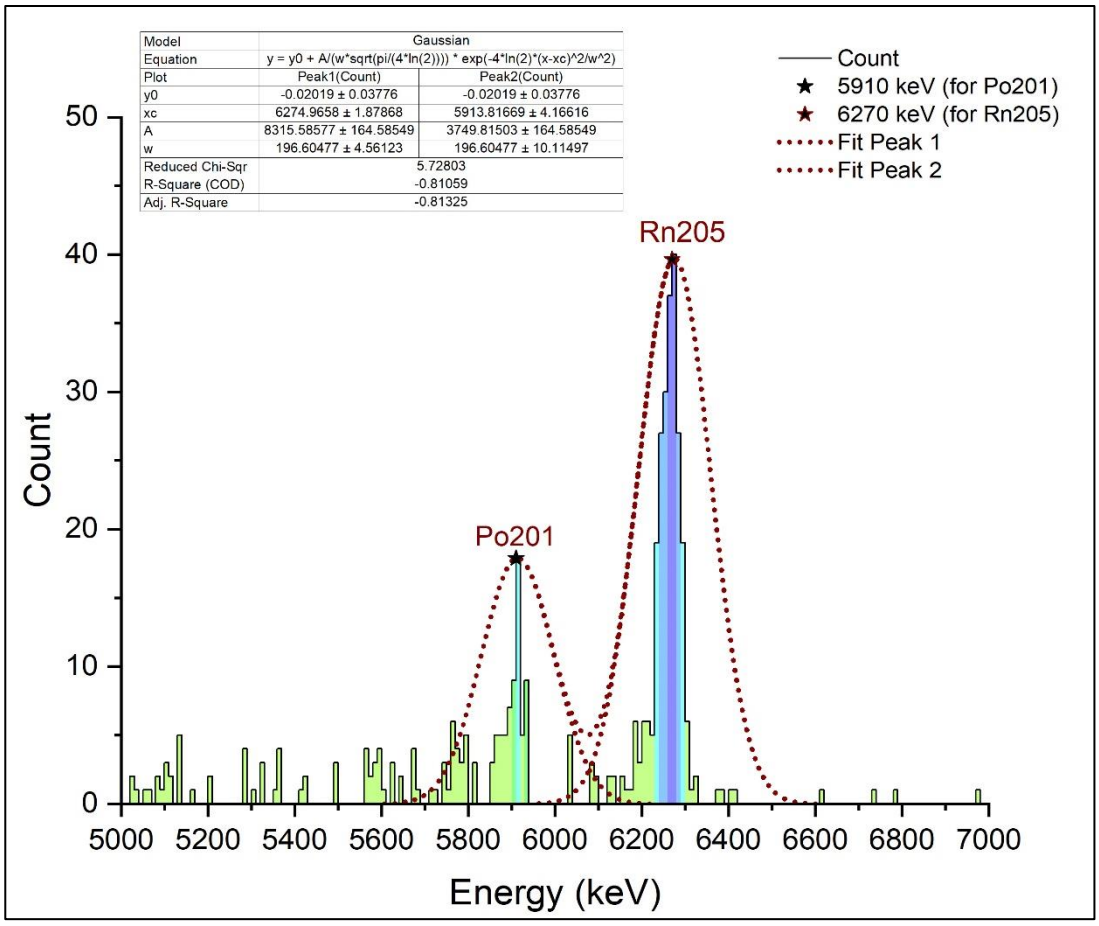
Rn ^{204} :
Half Life: 1.24 min

Rn^{204} decays by 73% at 6400keV α-Decay energy level

Po^{200} :
Half life: 11.5 min

Po^{200} decays by 11% at 5860keV α-Decay energy Level

v. Rn^{205}:



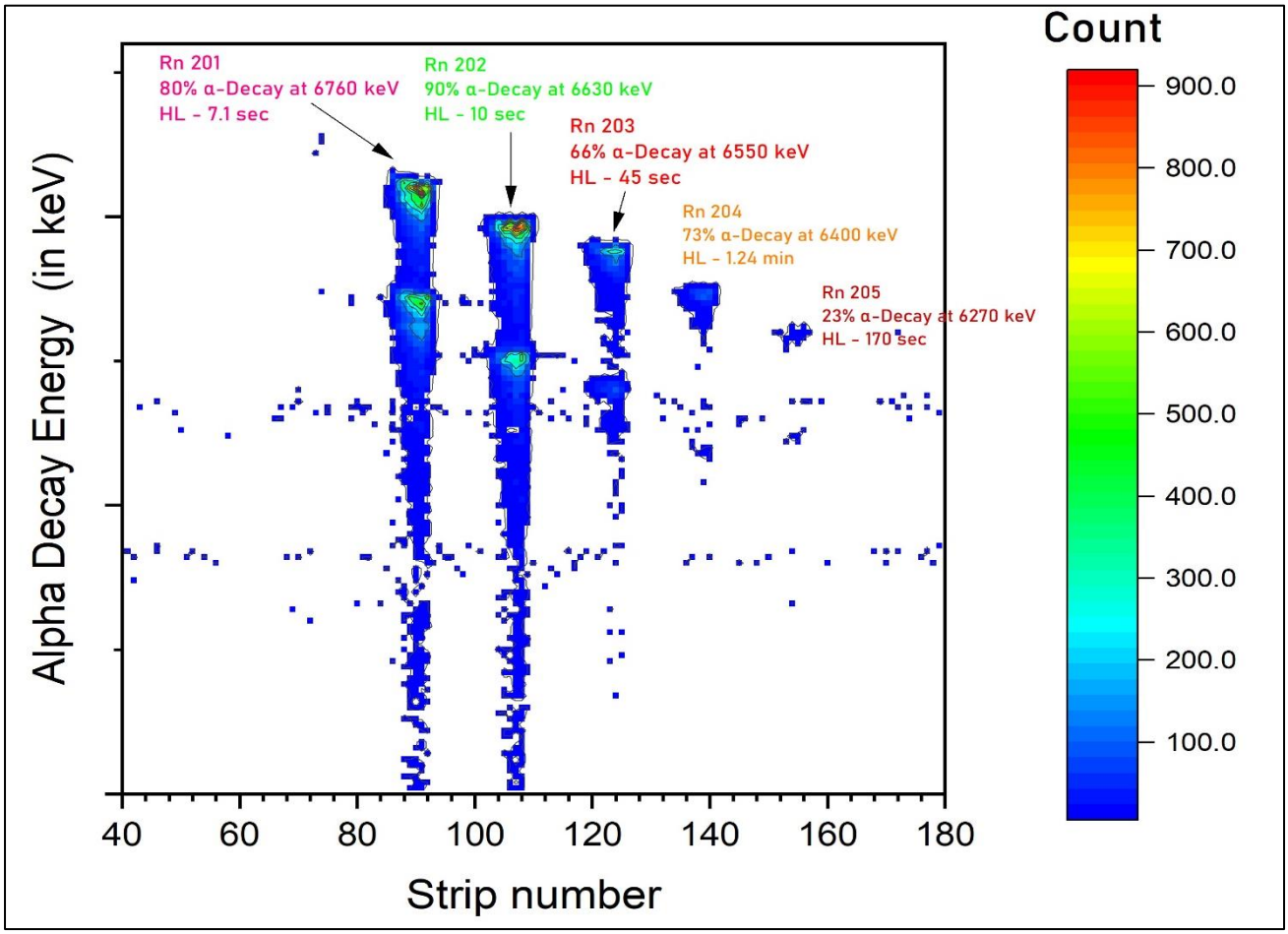
Rn^{205} :
Half Life: 170 sec

Rn^{205} decays by 23% at 6270keV α-Decay energy level

Po^{201} :
Half life: 15.3 min

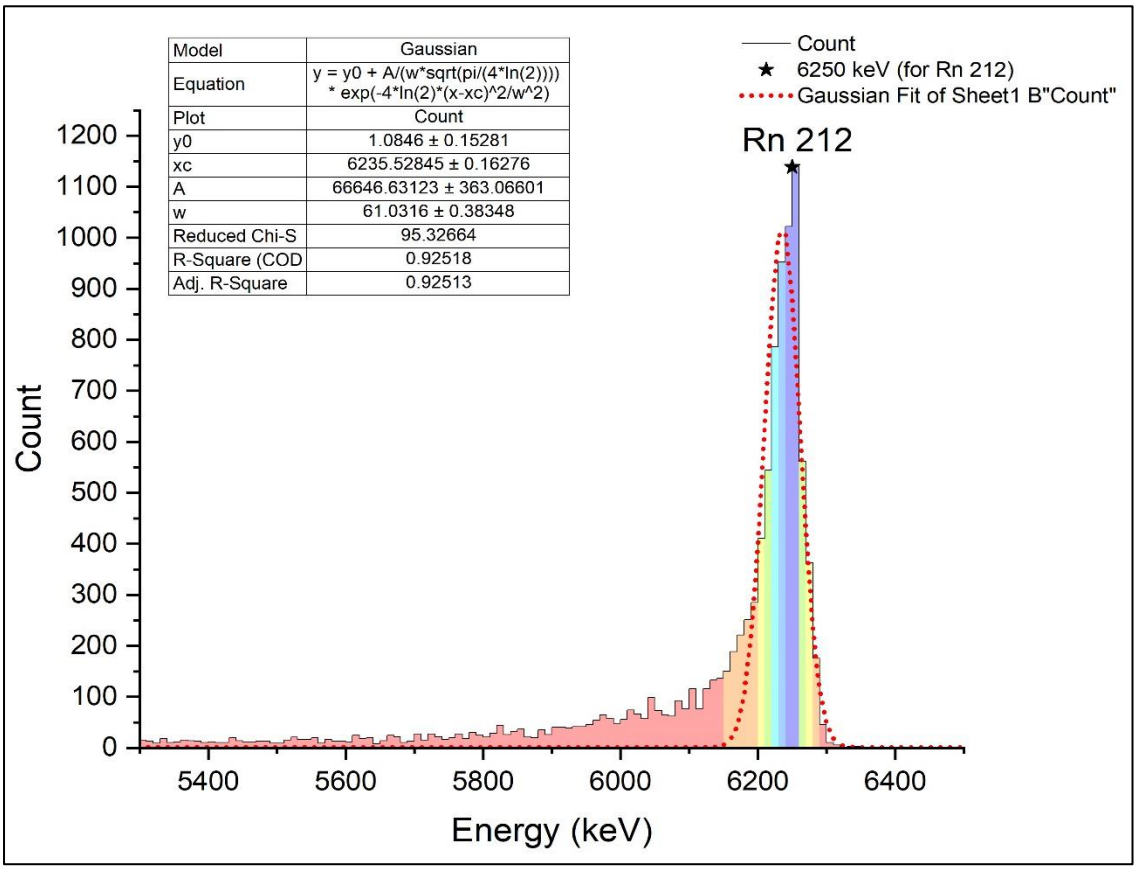
Po^{201} decays by 1.6% at 5910keV α-Decay energy Level

➢ Heat Map for Radon isotopes ($Rn^{201-205}$ isotopes) :



3. Histograms of $Rn^{212-218-219}$

i. Rn^{212}:



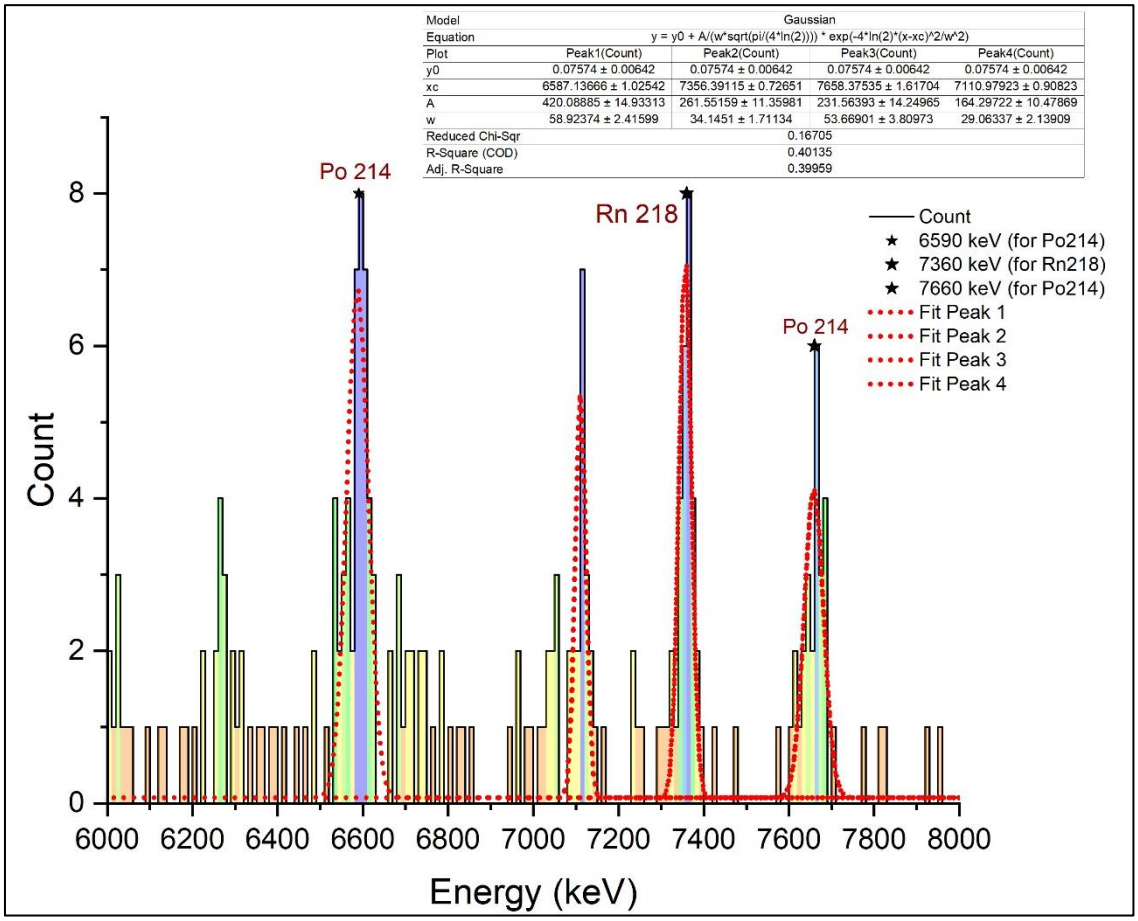
Rn^{212} :
Half Life: 23.9 min

Rn^{212} decays by 100% at 6250keV α-Decay energy level

Po^{208} :
Half life: 2.898 years

Po^{208} decays by 99.99% at 5120keV α-Decay energy Level

ii. Rn^{218} :

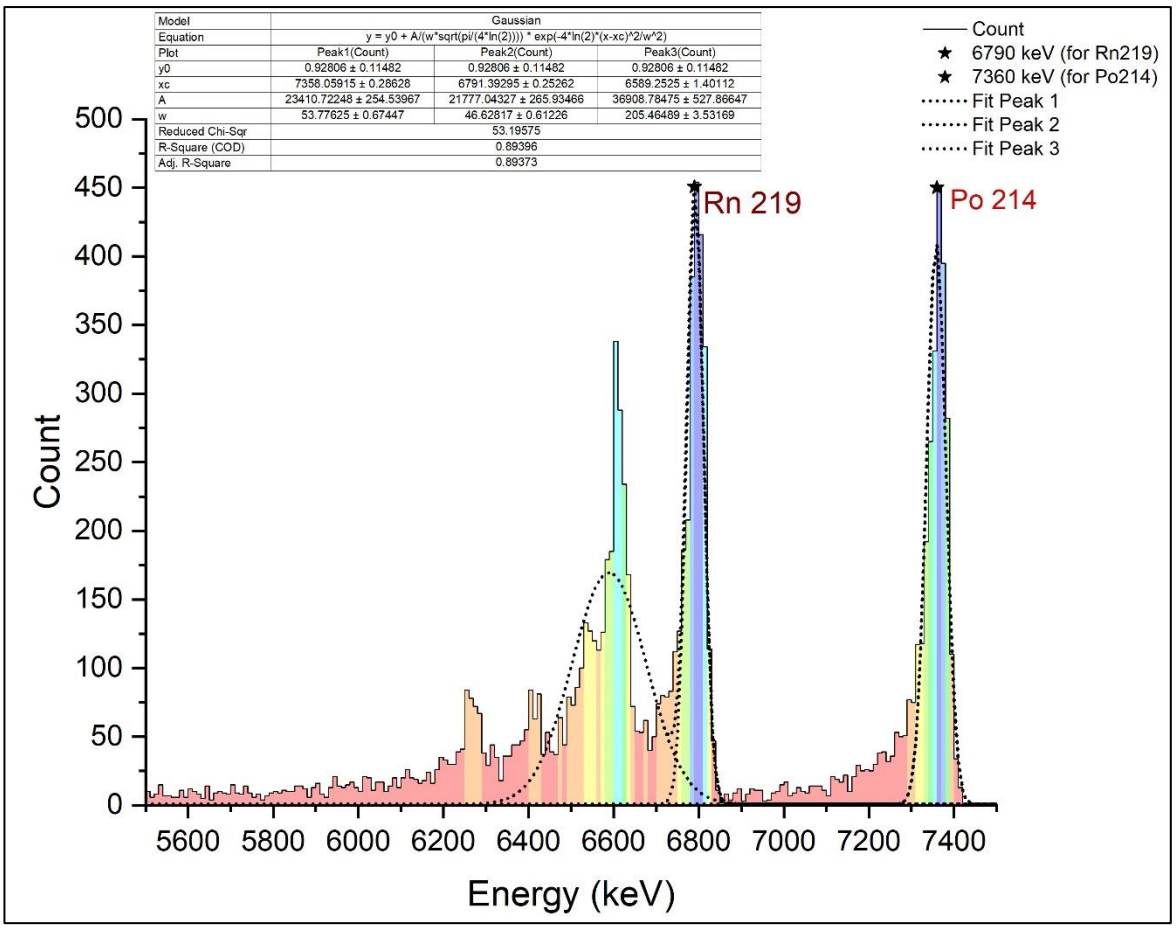


Rn^{218} :
Half Life: 35 msec

Rn^{218} decays by 100% at 7110keV α-Decay energy level

Po^{214} :
Half life: 164.5 μsec
Po^{214} decays by 100% at 7660keV α-Decay energy Level

iii. Rn^{219} :

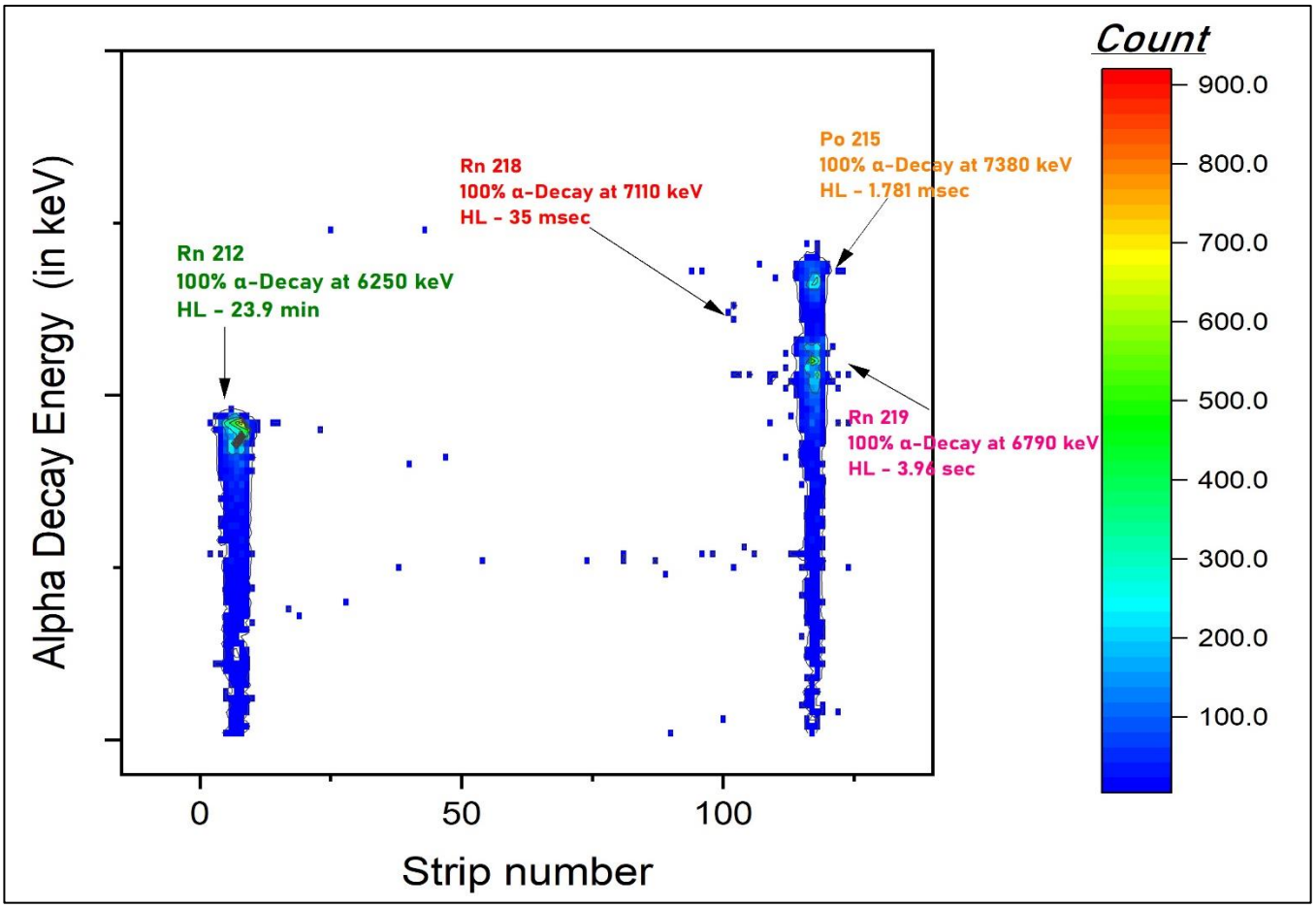


Rn ^{219} :
Half Life: 3.96 sec

Rn^{218} decays by 100% at 6790 keV α-Decay energy level

Po^{215} :
Half life: 1.781 msec
Po^{215} decays by 100% at 7380keV α-Decay energy Level

➢ Heat Map for Radon isotopes ($Rn^{212-218-219}$ isotopes)



6. Conclusion

The study of superheavy elements can provide valuable insights into nuclear reactions and the potential existence of the "island of stability." The ISOL method, coupled with post-acceleration, enables researchers to transport and analyse these nuclei under controlled conditions. The MASHA setup is continually improving to enhance separation efficiency and collect more data on these atoms. Recent experiments have demonstrated the effectiveness of using new nanomaterials, such as graphene foil and carbon nanotube paper sheets, which have shown promising results in improving separation efficiency and reducing separation time, thereby providing exciting opportunities for the analysis of short-lived isotopes.

The MASHA setup, which is constantly being enhanced, includes several key components. Mercury isotopes created through a complete fusion reaction involving 40Ar and 148Sm, while Rn isotopes produced through a fusion reaction involving 40Ar and 166Er, as well as a fusion evaporation reaction of 48Ca and 242Pu, were analysed. Experimental measurements of the alpha decay energies of the isotopes and their daughter nuclei closely match the theoretically predicted values, with small variations.

7. Acknowledgement

I would like to express my gratitude to the JINR institute and INTEREST team for organizing this online program and selecting me to participate in this project. I am particularly grateful to Mr. Vedeneev Viacheslav Yurievich, who has been a friendly and supportive project supervisor. He has shown great patience in explaining complex concepts, and providing me with all the necessary information related to the project. I would also like to extend my thanks and appreciation to my project colleague Mr. Gurucharan S, who have been a constant source of support throughout this project.

8. References

[1] C. Moskowitz, "Superheavy Element 117 Points to Fabled "Island of Stability" on Periodic Table," *Scientific American*, 2014.

[2] P. V. Duppen, "Isotope Separation On line and Post Acceleration," *Springer-Verlag Berlin Heidelberg,* pp. 37-77, 2006.

[3] V. Yu. Vedeneev1 · A. M. Rodin1 · L. Krupa1,3 · A. V. Belozerov1 ·..., "The current status of the MASHA setup," *Springer International Publishing Switzerland ,* 2017.

[4] Y. T. Oganessian, "On-Line Mass Separator of SuperHeavy Atoms," *Nuclear Instrument and Methods A,* pp. 33-46, 2002.

[5] Heinz W Gäggeler. Mendeleev's principle against Einstein's relativity: news from the chemistry of superheavy elements. Russian Chemical Reviews, 78(12):1139, 2009