



JOINT INSTITUTE FOR NUCLEAR RESEARCH  
Flerov laboratory of nuclear reactions

# FINAL REPORT ON THE SUMMER STUDENT PROGRAM

*Investigation of platinum isotopes  
production in photonuclear reactions at  
microtron MT-25*

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## Abstract

In this work will be shown method for separation of platinum isotopes which have been produced in photonuclear reaction on microtron MT-25. As a target is used mixture of cisplatin and cryptomelane. Finding rapid and efficient method for separating isotopes of platinum formed by the  $(\gamma, n)$  reaction presents a difficult problem. Platinum-195m was isolated from target using radiochemical techniques for separation of isotopes, and efficiency of separation was checked using gamma detectors.

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## 1. Introduction

Radioisotopes find wide-ranging applications in various fields, including industry, research, agriculture and medicine. Production of radioisotopes, radiolabelled compounds, radiation sources and other products based on radioisotopes constitute important activities of several national nuclear programmes. These often support several areas of national economic significance such as health care services and industrial quality control. Two major sources of artificial radioisotopes are accelerators and reactors.

To facilitate the study of the properties of platinum activities produced in the synchrotron by (y,n) reactions, an attempt was made to find complexes of these elements which would undergo Szilard-Chalmers type reactions.

### 1.1 Szilard-Chalmers effect

In 1934 L. Szilard and T. A. Chalmers discovered that bond breaking could occur for atoms following nuclear reaction or radioactive decay even though the recoil energy in the initial process is not sufficient to overcome the bonding energy. In the case of thermal neutron capture the processes involved in the emission of the  $\gamma$ -ray, which removes the nuclear excitation energy, impart recoil energy to the atom to break most chemical bonds. If, after rupture of the bonds, the product atoms exist in a chemical state different and separable from that of the target atoms, the former may be isolated from the large mass of inactive target. This provides a means of obtaining high specific activities in reactions where target and product are isotopic.

This process is known as the Szilard-Chalmers reaction and was discovered when, following the irradiation of ethyl iodide with thermal neutrons, it was found that radioactive iodide could be extracted from the ethyl iodide with water. Moreover, when iodide carrier and silver ions were added to this aqueous phase, the radioactive iodide precipitated as silver iodide. The obvious interpretation of these results is that the neutron irradiation of the ethyl iodide, which caused the formation of  $^{128}\text{I}$ , ruptured the bonding of this atom to the ethyl group. The bond energy of iodine to carbon in  $\text{C}_2\text{H}_5\text{I}$  is about 2 eV. Since this exceeds the recoil energies of neutron capture, the bond breakage must have resulted from the  $\gamma$ -emission which followed neutron capture and not the capture process itself.

Isomeric transitions which proceed by emission of  $\gamma$ -rays may not provide sufficient recoil energy to break covalent bonds. However, in these cases or very low energy isomeric transitions, the extent of internal conversion is large. This results in vacancies in the inner electron orbitals. When electrons in outer orbitals move to fill the vacancies, the difference in electron binding energies is sufficient to cause some ionization, resulting in relatively high charge states for the atom, leading to bond rupture.

Compounds which are well suited as targets for Szilard-Chalmers reactions are organic halides and inorganic ions of the type  $\text{MnO}_4^-$ ,  $\text{PO}_4^{3-}$ ,  $\text{ClO}_4^-$ ,  $\text{IO}_3^-$ ,  $\text{IO}_4^-$ ,  $\text{BrO}_3^-$ , etc.

## 1.2 Accelerator production of isotopes

Invention of the cyclotron by Ernest Lawrence in 1931 made it possible to produce radioactive isotopes of a number of biologically important elements. The cyclotron is most widely used accelerator for producing radionuclide. Nowadays exist a wide variety of cyclotrons, from microtrons with energy less than 3 MeV to large cyclotrons with energy in excess of 500 MeV.

The production of radionuclides with an accelerator demands that particle beams be delivered with two specific characteristics. The beam must have sufficient energy to bring about the required nuclear reactions, and there must be sufficient beam current to give practical yields.



**Figure1.** Microtron MT 25 in JINR, Dubna

### **1.3 Radiopharmaceuticals**

Radiopharmaceuticals are medicinal formulations containing radioisotopes which are safe for administration in humans for diagnosis or for therapy. Although radiotracers were tried as a therapeutic medicine immediately after the discovery of radioactivity, the first significant applications came much later with the availability of cyclotrons for acceleration of particles to produce radioisotopes. Subsequently, nuclear reactors realised the ability to prepare larger quantities of radioisotopes. Radioiodine (iodine-131), for example, was first introduced in 1946 for the treatment of thyroid cancer, and remains the most efficacious method for the treatment of hyperthyroidism and thyroid cancer.

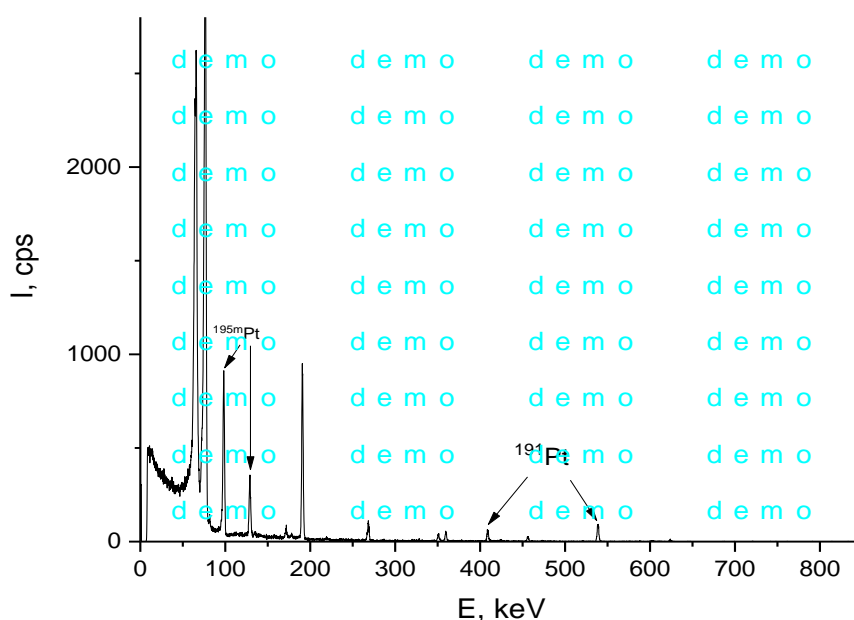
One of the major goals for setting up nuclear research reactors was for the preparation of radioisotopes. Among the several applications of radioisotopes, medical applications were considered to be of the highest priority. Most of the medium flux and high flux research reactors now are routinely used to produce radioisotopes for medical, and also industrial, applications. The most commonly used reactor produced isotopes in medical applications are molybdenum-99 (for production of technetium-99m), iodine-131, phosphorus-32, chromium-51, strontium-89, samarium-153, rhenium-186 and lutetium-177.

Currently there are over 100 radiopharmaceuticals developed using either reactor or cyclotron produced radioisotopes and which are used for the diagnosis of several common diseases and the therapy of a few selected diseases, including cancer. Radiopharmaceuticals production involves handling of large quantities of radioactive substances and chemical processing.

## 2. Experimental part

Platinum radioisotopes have been produced by irradiation of cisplatin at microtron MT-25. To prepare targets for the experiments, first two samples of cisplatin and cryptomelane mixture in the ratio of 1:2 and 1:6 correspondingly were prepared. These targets were placed in a plastic round holder and have been irradiated for 2 days.

After irradiation material was taken from foils and transferred to the glasses. First cisplatin was measured for 300 seconds on HPGe  $\gamma$ -detector as a reference point for radioactivity and quantity estimation.



Then other two glasses were measured on HPGe  $\gamma$ -detector for 300 seconds. After that they were washed with 20 ml of hot water two times, evaporated, dissolved in 5 ml of a mixture of 1 M HCl and 1,5 % H<sub>2</sub>O<sub>2</sub>, separated from manganese on CIX in 0,1 M HCl and measured for 300 seconds again after complete evaporation. The results are shown as follows.

Weight, mg	Target 1	Target 2	Target 3
<b>Cisplatin</b>	16.32	8	22.07
<b>Cryptomelane</b>	36.26	46.78	-

**Table 1.** Activities at EOB (end of bombardment), Bq

	$^{191}\text{Pt}$	$^{195\text{m}}\text{Pt}$	$^{197}\text{Pt}$
<b>Target 1</b>	4674.8	17761.4	78986.5
<b>Target 2</b>	2464.8	8189.0	34572.4

**Table 2.** Activities after separation, Bq

	$^{191}\text{Pt}$	$^{195\text{m}}\text{Pt}$	$^{197}\text{Pt}$
<b>Target 1</b>	2386.1	9998.8	35708.0
<b>Target 2</b>	1296.8	5752.7	11873.2

**Results:**

1. A method for simultaneous production of medically relevant  $^{191,193\text{m},195\text{m}}\text{Pt}$  isotopes with high specific activity was presented.
2. Radiochemical yield was found to be 56 % for the ratio of cisplatinium to cryptomelane 1:2 and 70 % for the ratio 1:6 for  $^{195\text{m}}\text{Pt}$ .



## **Acknowledgment**

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