^{178m2}Hf AND ^{237}Pu ISOTOPES PRODUCTION ON THE U-200 CYCLOTRON

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ABSTRACT

The results of producing a high current He ion beam on the U-200 cyclotron for isotope production are presented. The paper presents also the results of experiments on the production of the ^{178m2}Hf isotope in the reaction ^{176}Yb(^{4}He,2n) and of the ^{237}Pu isotope in the reaction ^{235}U(^{4}He,2n). The target assembly is described.

1. U-200 CYCLOTRON

The two meter cyclotron U-200[1)] is a sector focussing machine with four straight sectors for acceleration of heavy ions with the charge to mass ratio within the range $0.14 \leq q/A \leq 0.36$. The maximal energy is determined by K = 145.

The ions with $q/A \leq 0.263$ are extracted from the cyclotron by a stripping carbon foil, installed at the boundary valley - hill[2)]. After stripping the ion motion becomes radially unstable due to the difference of the magnetic fields in the valley and on the hill, and ions are extracted from the vacuum chamber. Depending on the ratio of ion charges before and after stripping and on the position of the stripping foil ions can be extracted from the vacuum chamber on making one, two or more turns in the magnetic field.

At the U-200 cyclotron the one turn extraction of ions with the charge increased by the factor of two is used. A mechanical assembly for moving the stripping foil in the radial and azimuthal directions allows to regulate the energy of the extracted beam within 35% below the maximum. The extracted beam has the horizontal emittance of 30π mm·mrad, the vertical - 10π mm·mrad, the energy spread ~1%.

Last years the mode of long time operation with an intense ($\geq$100 eμA, 36 MeV) He ion beam was investigated for the production of isotopes for fundamental and applied research.

For the production of the He^{1+} ion beam the gas discharge ion source with a directly heated cathode was used[3)]. The head of the source and the diagram of the power supply circuit are shown in Fig.1 and Fig.2.

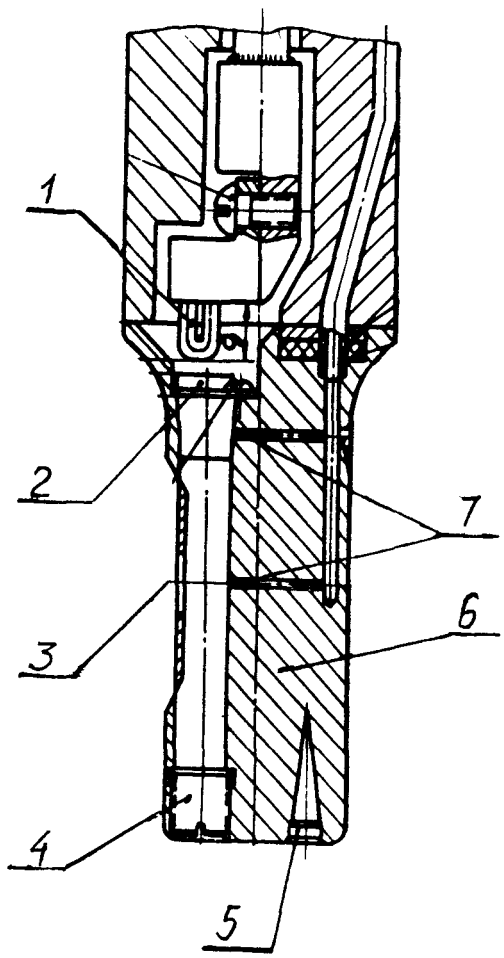

Fig. 1. The head of the ion source:
1-cathode, 2-diaphragm, 3-emission slit,
4-anticathode, 5-cooling, 6-chamber body
(anode), 7-gas inlet.

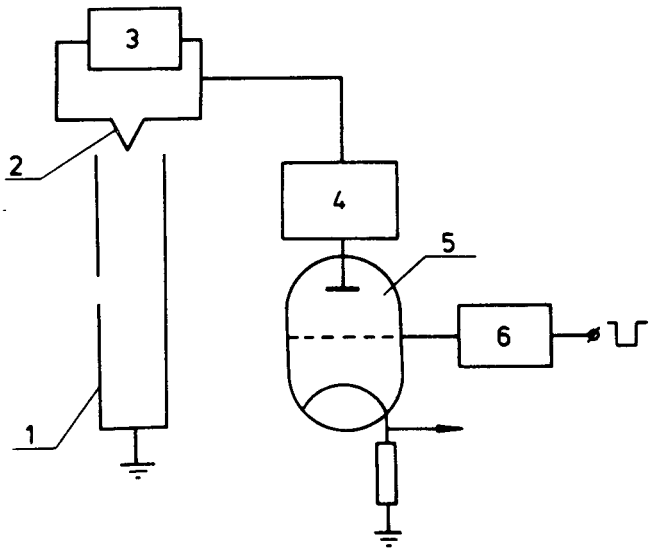

Fig. 2. The power supply circuit of the ion source: 1-discharge chamber (anode), 2-cathode, 3-cathode power supply, 4-discharge power supply, 5-modulating valve, 6-amplifier.

It was necessary to find the optimal from the point of view of source service life and the beam intensity mode of the source operation because the ion source head is activated due to vacuum losses of accelerated ions. The estimations show that at the operating pressure of $\sim 10^{-5}$ Torr the level of vacuum losses is about 50%.

The ion source operated in the pulsed mode with the repetition rate of 150 Hz and the pulse length of 2–3 ms. For the production of 100 eμA of He ions at the physical target one needs the following mode of operation: the discharge current of 2.5–3 A, the disharge voltage of ~200 V, the consumption of He ~2 cm^3/min.

The ion source service life is mainly determined by the sputtering rate of the directly heated cathode. Different cathodes were investigated. Except for the sputtering the cathode is affected by the Lorentz force in the sections where the direction of the current is perpendicular to the direction of the cyclotron magnetic field. The cathodes, prepared from the Ta and W rods with the diameters within the range 2–4 mm were tested. For heating the Ta cathode to the temperature of thermo-emission one needs a lower current that allows to use a larger diameter of the rod, but the service life of Ta cathodes never exceeds 10 hours with the subsequent break-up of the cathode. It seems that it is caused by the loss of the mechanical strength of Ta at the temperatures of termo-emission.

The most optimal one is a W cathode with 2.5 mm diameter. For the ignition of the discharge the heating current 280–290 A is necessary, which should be reduced during the operation of the source because the cathode diameter is decreased due to sputtering. It means that the source service life depends also from the skill of the operator. The average service life of the ion source is about 16 hours, the best result for the time being is 40 hours.

The other systems of the cyclotron showed stable operation during the acceleration of a high current beam.

Since the level of neutron radiation was relatively high the cyclotron operated in the high current mode only in the evening and at night. Figure 3 represents the record of target current during one of the such periods of irradiation for production ^{178m2}Hf isotope (see section 2).

The experience shows that the cyclotron can provide an integral number of He ions of about $\sim 10^{20}$ per month.

Now we are studying the possibilities to improve the vacuum inside the cyclotron and to develop an ion source with an increased service life.

2. LONG LIVED ^{178m2}HF ISOTOPE PRODUCTION

The ^{178m2}Hf isomer has unique properties due to its high spin $I^\pi = 16^+$, relatively low excitation energy $E^* = 2.447$ MeV and half-life $T_{1/2} = 31$ years.

At first the long-lived ^{178m2}Hf isomer was produced the in reaction of thermal neutron radiation capture with a very low yield[4]. The isomeric ratio for this reaction is $5 \cdot 10^{-10}$. The total amount of ^{178m2}Hf $\sim 10^{15}$ nuclei could provide a possibility to measure with a high accuracy the magnetic dipole and the electric quadrupole moments of the isomer, to define the changes of the rms radius of nuclei in the ground and isomeric states by the methods of nuclear orientation and laser spectroscopy. The use of this isomer as a target provides a possibility to define the influence of the isomeric state spin value in the reactions of sythesis and fission, to perform experiments on the Coulomb excitation of ^{178m2}Hf with mass-separation of ^{178}Hf isotopes[5,6].

For the optimal choice of the reaction for the accumulation of a microweight quantity of ^{178m2}Hf the experiments on light and heavy ion interactions with different targets were performed at FLNR, JINR. It turned that the optimal reaction is a ^{176}Yb(^{4}He,2n). The isomeric ratio for this reaction equals 0.05.

During the 1990–1991 the irradiations of enriched ^{176}Yb target (96% enrichment) by the He beam with the energy of 36 MeV and intensity of more than 100 eμA were fulfilled at the U-200 cyclotron. An inclined ($\sin\alpha$=0.2–0.3) water-cooled target was used. The target is situated in a closed metal collector with the entrance hole of 30 mm diameter for the ion beam. The cover of the collector is made of aluminum. Before the entrance of the collector a copper diaphragm for the limitation of the beam size is situated. The target material (Yb_2O_3 powder) was pressed on the 3 mm thickness aluminum substrate. The effective thickness of the target equals 80 mg/cm^2.

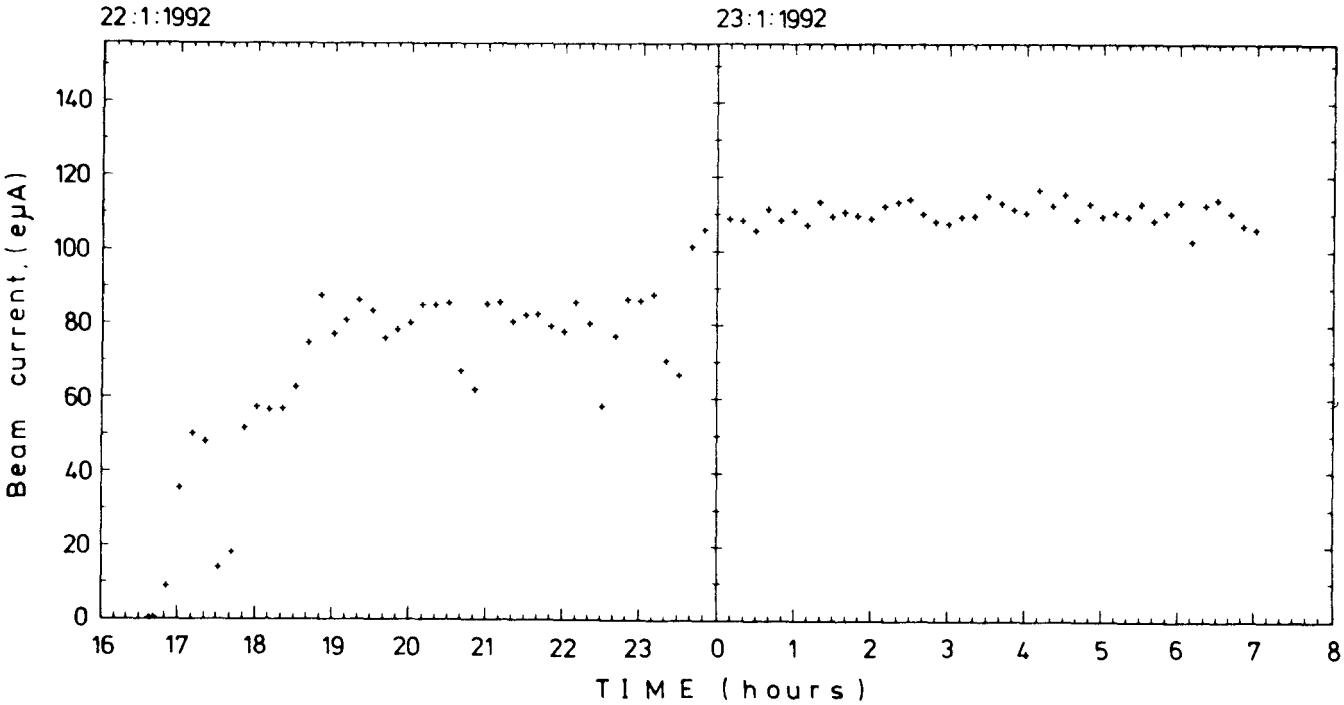

Fig. 3 The record of a target current during irradiation.

During the first series of irradiations of the enriched ^{176}Yb target, about $3 \cdot 10^{14}$ atoms of ^{178m2}Hf were accumulated. The identifications of Hf isotopes and the control of their yield were performed by measuring γ-spectrums with a Ge(Li) detector. The method of chemical isolation of Hf from the irradiated target material is described in reference[6].

The target prepared from the super enriched (99.998%) ^{176}Yb was also irradiated. Such super enrichment was fulfilled at PARIS[6] mass-separator in CSNSM (Orsay) for the reduction of the admixture of Hf isomer yield. About $3 \cdot 10^{14}$ atoms of ^{178m2}Hf were accumulated with a sharp decrease of other Hf isomer yields.

The experience of such irradiations and the results of ^{178m2}Hf isotope yield measurements show, that with a steady operation of the U-200 cyclotron one can expect the yield of 10^{15} atoms of ^{178m2}Hf in a month of irradiation with a current $\geq$100 eμA.

3. PRODUCTION OF THE ^{237}Pu ISOTOPE

Due to its nuclear-physical properties (T=45.2 days, EC=99.99%) ^{237}Pu is the only plutonium isotope which is sutable, according to modern medical concepts, for studying the plutonium metabolism *in vivo*. The content of neughboring isotopes of ^{236}Pu and ^{238}Pu which are the α-emitters in such a sample should not exceed 10^{-7} Bq/Bq.

The most suitable reaction for the production of ^{237}Pu is the ^{235}U(^{4}He,2n) reaction[7]. But it is practically impossible to provide the necessary purity of the sample produced in such a reaction due to covering the excitation function of the reactions ^{235}U(^{4}He,n) and ^{235}U(^{4}He,3n). Therefore it was suggested to make an additional purification of the produced sample of ^{237}Pu at an electromagnetic mass-separator.

The highly enriched (99.99%) ^{235}U isotope was used as the target material. The irradiations were fulfilled at the internal beam of the U-200 cyclotron in the energy range from 22 up to 26 MeV. The yield of ^{237}Pu isotope in this energy range is 0.2 kBq$\cdot\mu$A$^{-1}\cdot$h$^{-1}\cdot$mg^{-1} and the relative content of plutonium isotopes ^{236}Pu/^{237}Pu/^{238}Pu=$2.2 \cdot 10^{-4}/1/1.5 \cdot 10^{-4}$.

On the basis of this results the experiments on the production of the ^{237}Pu sample sufficient for the mass separation and subsequent use in a metabolism study were fulfilled. The target with the thickness of 10 mg/cm^{-2} was irradiated by He ions with the energy of 25 MeV during 20 hours. The activity of the ^{237}Pu sample after radiochemical isolation and purification from the substrate material and products of nuclear reactions is 1.5 MBq. The relative content of the plutonium isotopes ^{236}Pu/^{237}Pu/^{238}Pu=$1.6 \cdot 10^{-4}/1/1.5 \cdot 10^{-4}$.

Such a sample was purified at the mass separator YaSNAPP-2 of the Laboratory of Nuclear Problems, JINR. The yield of ^{237}Pu after separation is about 40%. The relative activity of plutonium isotopes after separation is ^{236}Pu/^{237}Pu/^{238}Pu=$2 \cdot 10^{-7}/1/\leq 3 \cdot 10^{-7}$ Bq/Bq.

The prepared sample is radiochemically and isotopically ultrapure and is practically safe for use in the research of plutonium metabolism *in vivo*.

4. REFERENCES

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