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COLLECTING NUCLEAR REACTION PRODUCTS
BY ADSORPTION FROM A GAS FLOW
IN A SLIT CHANNEL

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1. Introduction

Thermalization of recoil atoms in gas and transportation of them by the gas flow to radioactive decay detectors is an efficient technique for the study of short-lived nuclei properties [1-5]. As a rule, especially prepared aerosols serving as carriers of adsorbed recoil nuclei are added to the gas flow. However, when the aerosols are deposited on the recoil atoms collector they form rather thick layers of ballast material. At the gas flow rate of 1-10 l/min and the aerosol concentration of 10^{-9} g/cm³ the aerosol deposition rate on the collector is 10-100 $\mu\text{g}/(\text{cm}^2 \cdot \text{min})$ [4]. Due to that it is impossible to use the recoil atoms collector as the detector (or entrance window of the detector) of short range particles. Accordingly, the registration of radioactive decay in a solid angle close to 4π can not be done in a simple design. Besides that the aerosol can interact chemically with the detector substance.

The initial premise for starting the present experiments was the need for a highly efficient and fast registration technique of cluster decay of short-lived nuclei produced at heavy-ion accelerators [6,7]. We have carried out experiments on collecting recoil atoms from the gas flow (without using aerosols) by means of adsorption on the walls of the 1 mm wide slit channel formed by the sensitive surfaces of solid state nuclear track detectors. The charged particles from the decay of the atoms adsorbed on any wall of the slit channel are registered by detectors on both walls. The solid angle for registration of the adsorbed atoms radioactive decay can be $>2\pi$. The corrections for energy losses in the thin gas layer between detectors can be introduced with the sufficient accuracy.

The estimation of the medium mass atoms diffusion coefficient D in argon is $0.1-0.15 \text{ cm}^2/\text{s}$ [8]. Respectively, the time of recoil atoms diffusion T from the flow center in the channel to the wall is $0.01-0.05 \text{ s}$, if one uses for estimation the Einstein- Smoluchowski theory for the mean square displacement of a particle in Brownian motion [9]:

$$\overline{x^2} = 2D \cdot T \quad (1).$$

According to [10] this formula can be used and for diffusion of atoms and molecules.

Taking into account the fact that the recoil atoms diffuse to the wall from an arbitrary point in the channel cross-section, one can speak about the average "life-time" τ of the recoil atoms in the channel

$$\tau = c \cdot h^2 / D \quad (2),$$

where h is the characteristic channel dimension (height, radius), and the dimensionless coefficient $c < 1$ appears as a result of averaging the diffusion time over the channel cross-section of a certain shape (rectangular, circular or other).

For achieving high collecting efficiency it is necessary that the time of the recoil atoms moving along the channel should not be shorter than their "life-time" τ . The set-up parameters were selected with this consideration in mind

2. Experimental set-up

The experiments on collecting nuclear reaction products were carried out on an extracted beam of the heavy ion cyclotron U-400 at the Flerov Laboratory of Nuclear Reactions, JINR.

Fig.1 Schematic drawing of the experimental set-up. a) With a linear slit channel. b) With a disk slit channel. 1- ion beam, 2- target, 3- recoil atoms, 4- solid state nuclear track detectors, 5- gas filled cylinder for thermalization of recoil atoms, 6- teflon capillary.

Fig.1 shows two variants of the experimental set-up. The nuclear reaction products knocked out from the target are slowed down in gas flowing through a cylinder 20 mm in diameter and 30 mm deep. The cylinder depth was selected to exceed the recoil atoms range in argon and nitrogen at atmospheric pressure (10-20 mm for the reactions investigated by us)

The input gas flow is shaped by several openings 2 mm in diameter positioned along the perimeter of the half of the target close to gas inlet. In the variant presented in Fig.1a, the gas with the recoil atoms thermalized in it exits through the slit channel of $1 \times 13 \text{ mm}^2$ in cross section which is positioned perpendicularly to the beam. The channel walls are formed by strips of plastic (polycarbonate, polyethylene-terephthalate) which at the same time serve as recoil atoms collectors and track detectors of charged particles [11].

The variant of the set-up shown in Fig.1b was designed for removing the detector from the irradiation zone in order to decrease the background, caused by secondary radiation produced by the ion beam. From the cylinder the gas is fed through a teflon capillary 2 or 4.5 mm in diameter and 10 or 50 cm in length into the center of the circular slit volume and then flows radially. The volume is restricted by two 60 mm diameter plastic detectors separated by a 1 mm gap.

The working gas (pure argon or nitrogen) is fed through a pressure regulator from a standard gas tank with volume of 40 l and gas pressure up to 150 atm. The gas flow rate is stabilized in the range of 1-10 l/min by an electronic controller equipped with heat sensors. The gas pressure in the recoils thermalizing cylinder exceeds the atmospheric pressure by a few tens of Torr. The gas outlet is connected with air atmosphere through special filters.

3. Experimental Results

The technique was verified using the well-known nuclide ^{149}Tb ($T_{1/2}=4.15$ hour, α -decay branch of 16.7%, $E_{\alpha}=3.97$ MeV). A natural Zr

target of 3 mg/cm^2 in thickness was irradiated with 306 MeV ^{59}Co ions. ^{149}Tb was produced in the nuclear reactions $^{\text{nat}}\text{Zr}(^{59}\text{Co}, \text{xp}+\text{yn})$ either directly or as the result of β^+ -decay of the short-lived isotopes ^{149}Ho and ^{149}Dy .

To determine the total yield of ^{149}Tb a special experiment was performed. The entire internal surface of the cylinder 5 (see Fig. 1a) was covered with thin nickel and aluminium foils. The thickness of aluminium foil covering the target was 0.48 mg/cm^2 . It is much less than the mean range of recoil atoms in Al which is equal to $3.2\text{--}3.6 \text{ mg/cm}^2$ [12]. The cylinder was filled with nitrogen at atmospheric pressure and then the gas flow was stopped.

Fig.2 Distribution of recoil atoms over the inner surface of stopping cylinder in experiment without gas flowing. The values of distances equal to 0 and 3 cm are obtained for α -activity on forward (with target) and back cross-sections of cylinder. $\bar{R}$ - calculated recoils mean range in nitrogen [12].

In this conditions all the recoil atoms knocked out of the target are

thermalized in the gas and then adsorbed on surface of the foils whose α -activity after the irradiation was measured by Si(Au)-detectors. The α -activity distribution over the inner surface of the cylinder in this experiment is shown in Fig.2. The effective cross-section of the ^{149}Tb production over all reaction channels (averaged according to the total target thickness) is $8.5 \cdot 10^{-27} \text{ cm}^2$. All further evaluations of the collecting efficiency of recoil atoms were based on this experimentally measured value.

3.1 Experiments with a linear collector

(variant 1a)

The experimental results of ^{149}Tb atoms adsorption on a linear collector are presented in Fig.3. They show that in a 1 mm wide slit channel $\approx 60\%$ of all recoil atoms can be collected along the length of 20 cm. After irradiation up to 3 hours the 3.97 MeV α -line of ^{149}Tb was registered in the solid angle of $0.6 \cdot 2\pi$ with the energy resolution of 30 KeV, which was entirely determined by the Si(Au)-detector properties.

In experiments searching for ^{114}Ba cluster decay in irradiating a natural Ni target with 318 MeV ^{58}Ni ions [13] we also measured the distribution of I, Te and Sn isotopes along the slit channel length. The α -activity of the nuclear reaction products (in which the main contribution was made by ^{110}I and $^{108,109}\text{Te}$ isotopes [14,15]) was measured by means of the tracks counting in cellulose-nitrate foils which were used as the material for the channel walls. The distribution of ^{109}In and ^{110}Sn isotopes over the collector was measured with a Ge(Li) γ -spectrometer according to the known γ -lines of these isotopes

[16]. No substantial difference in the deposition behaviour of the atoms of various elements which could be attributed to their chemical or physical properties was determined.

Fig.3 Distribution of recoil atoms in a linear slit channel. a)- differential, b)- integral. Straight lines- exponential fit by Eg. (3).

3.2 Experiments with a disk collector

(variant 1b)

The experimental results of collecting ^{149}Tb on the walls of a disk slit channel are presented in Fig.4. On a disk facing the one through which the capillary is passing a somewhat larger amount of recoil atoms is adsorbed. The α -particle tracks distribution over the disk cellulose-nitrate collector measured over a 0.2 mm wide radial strip is presented in Fig.5.

Fig.4 Integral efficiency of recoil atoms collection in a disk slit channel as a function of the collector radius. (o)- transport capillary 4.5 mm in diameter and 10 cm in length. (+)- transport capillary 2 mm in diameter and 10 cm in length.

Fig.5 Radial distribution of α - tracks in the disk slit channel along the strip 0.2 mm in width. Straight line- fit by Eq. (5).

For this set-up design the question about recoil atoms loss due to their adsorption in the capillary is essential. The distribution of

β -active recoil atoms along the length of the capillary 4.5 mm in internal diameter was measured with a β -radiation counter (see Fig.6).

Fig.6 Distribution of β - activity along the capillary 4.5 mm in diameter. (+)- capillary length is 10 cm, (o)- capillary length is 50 cm. Dashed line - fitting by Eq. (4).

A comparison in identical conditions of β -activities of the 10 cm long capillary and the collector with a known number of collected ^{149}Tb recoil atoms allows to make us the conclusion that the capillary of this length adsorbs 25% of all recoil atoms. The capillary 50 cm in length adsorbs about 50% of reaction products. This is consistent with the decrease of the total collecting efficiency from ≈ 60 to $\approx 30\%$ when changing the capillary length from 10 to 50 cm.

In experiments with the disk collector we tested the influence of the electric field on collecting recoil products. The ± 50 V potential in the 1 mm gap between the disks provided the electric field gradient of 1000 V/cm. The ion drift velocity is $2 \cdot 10^3$ cm/s [17]. The gas velocity at the flow rate of 2.8 l/min changes along the radius from 330 to 25 cm/s. Consequently, the deposition of charged recoil products must be taking

place on a ≈ 1 mm length. However, use of an electric field of any sign produced just a slight (up to 20%) redistribution of the activity over the collectors and did not increase the collecting efficiency. Probably, most of the recoil products in the channel have no charge and that is why the adsorption technique is optimal for collecting them.

The dependence of the collecting efficiency on the beam intensity is shown in Fig. 7. The energy release of the ^{59}Co beam in gas contained in the cylinder volume at the intensity of $3 \cdot 10^{11}$ particles/s is 7 W. At the flow rate of 2.8 l/min, without taking into account the heat exchange with the water-cooled walls, the gas can be heated up to 80°C .

Fig. 7 Efficiency of recoil atoms collection as a function of the beam intensity. Disc slit channel, transport capillary 4.5 mm in diameter and 10 cm in length. Ar-flow rate is 2.8 l/min. Nuclear reactions $^{\text{nat}}\text{Zr} + ^{59}\text{Co} (306 \text{ MeV}) \rightarrow ^{149}\text{Tb}$. The line is drawn to guide the eye.

In model experiments when the gas was heated by a 15 W heating capacity spiral, the measurements showed that if all parts of the set-up were water cooled down to 16°C the gas was not heated higher than 30°C in the slit channel entrance. Nevertheless, the pulse character of the beam

(150 pulses per second, the duration of each pulse being 1 ms) can certainly produce pressure and volume perturbations in the gas flow. At high beam intensity or insufficient gas flow rate the portion of the recoil atoms deposited on the surface of the target containing cylinder rises.

The time of recoil atoms collecting can be estimated as follows. At the gas flow rate of 2.8 l/min the complete gas renewal in the target containing cylinder volume (Fig.1) is accomplished in 0.2 s. The area of the initial recoil atoms distribution occupies about 50% of the length of the cylinder and is 15 mm in diameter. The capillary for extracting the recoil atoms is inserted into part of the cylinder which corresponds to the maximum of the distribution of recoil atoms over their ranges in the stopping gas. Consequently, one can expect the recoil atoms extraction time to be reduced to 0.05 s. The transit time in the capillary with the internal diameter of 4.5 mm and length of 10 cm is 0.03 s. Thus, the delay time from the recoil nucleus production to its reaching the recoil atoms collector is < 0.1 s.

4. Model of recoil atoms distribution over the collector

From the definition of recoil atoms life-time in slit channel by equation (2) one can obtain a qualitative description of their adsorption along the channel length. Moving in x-direction together with the carrier gas, the recoil atom is staying in the volume element of the channel $dV = S \cdot dx$ during the time $dT = dV/Q$, (S is the cross-section area, Q is the gas flow volume rate). During the time dT the proportion of recoil atoms leaving the flow is $dN/N = -dT/\tau$. After obvious

transformations and integration we get for the rectangular cross-section

$$N(x)/N_0 = \exp[-2Dbx/(c \cdot Qh)] \quad (3),$$

for the circular cross-section

$$N(x)/N_0 = \exp[-\pi Dx/(c \cdot Q)] \quad (4),$$

for the disk slit channel

$$N(r)/N_0 = \exp[-2\pi D(r^2 - r_0^2)/(c \cdot Qh)] \quad (5),$$

where N_0 is the total flow of recoil atoms entering the channel, $N(x)$ is the flow of recoil atoms at the distance x from the channel entry, $N(r)$ is the flow through a circumference with the radius r in the disk channel, r_0 is the initial (capillary) radius in the disk channel, b is the width of the channel, $2h$ is the gap height, c is the nondimensional coefficient from the formula (2). Obviously, the number of adsorbed recoil atoms is equal to $N_{\text{ads}} = N_0 - N$.

The derived expressions are valid if the mechanism according to which the recoil atoms leave the gas flow and are deposited on the walls is purely diffusive and the gas flow is laminar. At the gas flow rates and geometrical dimensions which we used the Reynolds number is <700 , hence, the gas flow in the channel after a certain transitional section becomes laminar and stable to perturbations [18].

For a more correct result when working out this problem one should take into account the parabolic distribution of the gas flow rate over the channel height, the initial and boundary conditions for the recoil atoms concentration in the channel and solve the corresponding diffusion equations. The solutions can be found in [19]:

for the rectangular cross-section channel

$$N(x)/N_0 = 0.9099 \exp(-1.885\mu) + 0.0531 \exp(-21.43\mu); \quad \mu = 2Dbx/Qh \quad (6),$$

for the circular cross-section channel

$$N(x)/N_0 = 0.819 \exp(-1.828\mu) + 0.0975 \exp(-11.15\mu) + 0.032 \exp(-28.48\mu) + 0.0157 \exp(-53.8\mu); \quad (\mu = 2\pi D x / Q); \quad (7),$$

for the disk slit channel

$$N(x)/N_0 = 0.9104 \exp(-2.8278\mu) + 0.0531 \exp(-32.147\mu) + 0.0153 \exp(-93.475\mu) + 0.0068 \exp(-186.805\mu); \quad (\mu = 4\pi D (r^2 - r_0^2) / 3Qh). \quad (8)$$

A comparison shows, that the solutions obtained from simple consideration coincide with the first harmonic of exact solutions, if in the equation for the life-time (2) we set $c=0.53$ for the linear and disk channels, and $c=0.27$ for the circular channel. The difference in c -values means that τ for channel with circular cross section is two times less than for channel with rectangular cross section when $r=h$ and $h \ll b$. Physically it means that for the circular cross section the diffusion has two-dimensional character and in formula (1) coefficient 4 instead of 2 must be used. In Fig.3 we can see that the recoil atoms distribution over the channel length really has exponential character. However, not just one, but two exponents are observed. Evidently, this is due to the "speed-up" section at the gas flow entrance into the channel, where laminar flow is established [20]. Its length can be estimated [20] using equation (9):

$$L \approx 0.03 \cdot d \cdot Re \quad (9)$$

where d is the transversal size of the channel, Re is the Reynolds number.

5. Conclusion

The experience in collecting recoil atoms by means of the adsorption

technique in the slit channels showed its reliability and reproducibility. An important advantage is that the gas flow is produced simply due to gas pressure in the tank, avoiding the necessity to employ powerful vacuum pumps used in the "helium-jet" technique [1-3].

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Сбор продуктов ядерных реакций методом адсорбции
из газового потока в щелевом канале

Представлены экспериментальные результаты по сбору атомов отдачи в реакциях с тяжелыми ионами. Замедленные в газе до тепловых скоростей атомы отдачи транспортируются газовым потоком в щелевой канал с зазором в 1 мм, образованный чувствительными поверхностями детекторов радиоактивного распада. За счет адсорбции из газового потока в щелевом канале $\geq 50\%$ исходных атомов отдачи могут быть собраны на поверхности детекторов за время ≤ 0.1 сек.

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Collecting Nuclear Reaction Products by Adsorption from
a Gas Flow in a Slit Channel

Experimental results of collecting recoil atoms produced in reactions with heavy ions are presented. The recoil atoms thermalized in gas are transported by the gas flow into a 1 mm wide slit channel formed by the sensitive surfaces of radioactive decay detectors. Due to the adsorption from the gas flow it is possible to collect $\geq 50\%$ of the initial recoil atoms on the detector surfaces within a time interval of ≤ 0.1 s.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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