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NUCLEI
Experiment

Investigation of Neutron Generation upon Irradiation of Deuterated Crystalline Structures with an Electron Beam

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Abstract—The possibility of neutron generation by irradiating deuterated crystalline structures with an electron beam with an energy of 20–40 keV was studied. As targets, the deuterated crystalline structures of palladium and textured CVD diamond were used. Measurements of neutron emission are presented, which were carried out by three independent methods—scintillation detectors, counters based on He-3, and track detectors CR-39.The average neutron flux during irradiation was estimated as $1-10\ \mathrm{s}^{-1}$ in 4π sr.

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INTRODUCTION

The results of experiments carried out at ion accelerators to measure the cross sections of nuclear fusion reactions showed significant effects of increasing the yield at low energies of colliding ions and using solid targets (in comparison with a gas target and theoretical calculations) [1–6]. It is assumed that an increase in the yield of nuclear fusion reactions is caused by the screening of colliding ions by their surrounding electrons in a solid target, which leads to an increase in the probability of overcoming the Coulomb barrier and their fusion. In this case, the target material plays an essential role. The greatest amplification effect was obtained on materials containing platinum group metals (Pd, Pt). The possibility of initiating nuclear fusion reactions in solid-state deuterated targets by various external radiation (X-rays, ion and electron beams) has also been reported [7–11]. Thus, in [10], the products of nuclear DD fusion were observed (protons and neutrons) under the action of X-ray quanta with energies of 20–30 keV on deuterated crystal structures of palladium, titanium, and CVD diamond. In [11], using an electron beam with an energy of 450 keV up to 3.0 MeV, generating secondary gamma quanta incident on the ErD_3 and TiD_2 targets, initiated nuclear fusion reactions in them. With a total number of deuterium atoms in the irradiated target of 5×10^{24}, a beam energy of 2.9 MeV, and an accelerator current of 15 mA, the neutron flux from DD synthesis was measured and found to be $(1.5 \pm 0.3) \times 10^3\ \mathrm{s}^{-1}$ into a solid angle of 4π sr. Also, together with neutrons with an energy of 2.45 MeV, neutrons with energies of ∼4 and ∼5 MeV were detected.

DD-reactions go through two main channels (1) and (2). In this work, we investigated the possibility of generating neutrons (products of nuclear DD fusion reaction (1)) in crystalline deuterated targets by an electron beam with an energy of 20–40 keV. This approach presupposes the combined action of both electrons and bremsstrahlung X-ray quanta on the target:

$$d + d \to n\ (2.45\ \mathrm{MeV}) +^3 \mathrm{He}\ (0.8\ \mathrm{MeV}), \quad (1)$$

$$d + d \to p\ (3\ \mathrm{MeV}) + T\ (1\ \mathrm{MeV}). \quad (2)$$

EXPERIMENTAL TECHNIQUE

The layout of the detectors and the target in the vacuum chamber of the electron accelerator is shown in Fig. 1. The research was carried out on the basis of the NRU "BelSU" electron gun with the following parameters: accelerating voltage—10–100 kV; beam current—1–500 μA; emission current—up to 0.8 mA; beam divergence on the target—0.5°; beam current stability—no worse than 1%; the spread in the electron beam energy is no more than 0.1%. The target was placed on a duralumin holder cooled with water, which could move across the electron beam. The electron beam was formed by a focusing system. The beam diameter on the target was 3 or 6 mm. The CR-39 track detector was located at a distance of about 2 cm from the target outside the beam range.

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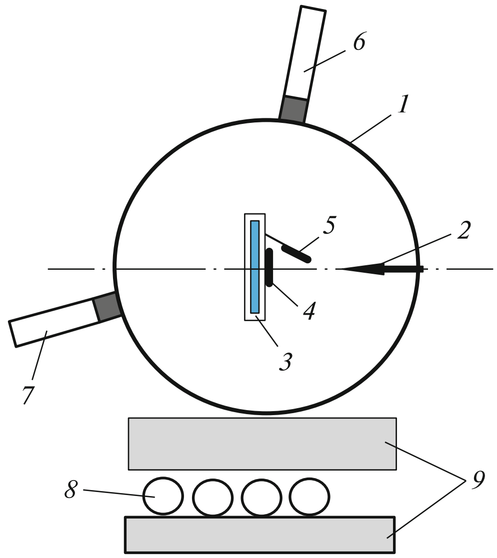

Fig. 1. Layout of the target and detectors. *1*—wall of the vacuum chamber, *2*—electron beam, *3*—target holder, *4*—target, *5*—CR-39 track detector, *6* and *7*—scintillation detectors with photomultiplier, *8*—He-3 counters, *9*—polyethylene neutron moderators.

A neutron detector based on four SNM-18 counters filled with He-3 gas was located at a distance of about 40 cm from the target. It was equipped with polyethylene-based neutron moderators (10-cm thick in front of the detector, and 8 cm behind the detector). Two scintillation detectors based on p-terphenyl were located near the vacuum chamber at a distance of about 17 cm from the point of incidence of the electron beam on the target. One detector was positioned along the electron beam, the other across the beam. The detectors worked independently.

Three independent neutron detection techniques were used. The CR-39 track detector made it possible to detect both charged particles emitted from the target and neutrons (through recoil protons and decay reactions). The coverage of the detectors (44-μm Al) made it possible to avoid the irradiation of the detector with scattered electrons. The procedure for calibrating the track detector is described in detail in [12]. The efficiency of detecting fast neutrons from recoil protons (with energies above 400 keV) with a Cf-252 source is measured as $\sim 6 \times 10^{-5}$. The tracks of recoil protons from the back of the detector were taken into account, since there are also tracks from the protons of the DD reaction (2) on the side facing the target. The presence of tracks from decays of nuclei into two particles or three particles gives an indication of the presence of neutrons with energies greater than 3 MeV and more than 10 MeV, respectively. A neutron detector based on four SNM-18 counters filled with He-3 gas was intended to register thermal neutrons.

Scintillation detectors based on a p-terphenyl crystal (diameter 2.5 cm, height 2.5 cm) were equipped with a Hamamatsu R6094 photomultiplier. The technique of separating signals from neutrons and gamma quanta by the shape of the scintillation detector pulses is described in [13, 14].

Deuterated targets made of palladium and textured CVD diamond were used in the experiment. Targets made of copper, aluminum (target holder substrate), or those saturated with hydrogen under similar conditions were used as background targets. Pd/PdO: D_x samples with dimensions of 2.5×1 cm^2 and 2×4 cm^2 were prepared by thermal oxidation of Pd foil (99.95% purity, 50- and 40-μm thick). As a result, a PdO oxide film $\sim$50-nm thick is formed on the foil surface. Also used were palladium samples without oxide film 2.5×1 cm^2 in size and 500-μm thick. Then, the samples were saturated with deuterium by electrolysis in a 0.3M LiOD solution in D_2O with a Pt anode at a current density $j =$ 20 mA/cm^2. Deuterium penetrated the entire volume of the sample. After saturation with deuterium to the degree $x = \mathrm{D/Pd} \sim 0.4$–$0.6$, the samples were washed in heavy water and cooled with liquid nitrogen to a temperature of $T = 77$ K (cooling the sample to the temperature of liquid nitrogen is necessary to slow down the release of deuterium in order to study the effect of ionizing radiation on the desorption process). Measurements of the sample mass increment showed that up to $\sim 5 \times 10^{21}$ deuterium atoms entered the CVD diamond samples during the electrolysis. Then the target was installed in the holder opposite to the fixed CR-39 detectors and placed into the vacuum chamber of the accelerator for further studies. The time spent on mounting the target did not exceed 40 minutes.

The fabrication of a textured CVD diamond structure is described in detail in [15]. The target used had a diameter of 18 mm and a thickness of 400 μm. The deuterated structure of textured CVD diamond was saturated by electrolysis in a 0.3 M LiOD solution in D_2O with a Pt anode. The samples were used as a cathode. Then the samples were washed in heavy water and cooled with liquid nitrogen to a temperature of $T = 77$ K (cooling the sample to the temperature of liquid nitrogen is necessary to slow down the release of deuterium). Measurements of the sample mass increment showed that up to $\sim 8 \times 10^{20}$ deuterium atoms entered the CVD diamond samples during the electrolysis. The control of the amount of deuterium released from the samples after irradiation was carried out by weighing them.

The exposure time for one target was usually 60–160 minutes. Then the cycle of target saturation and

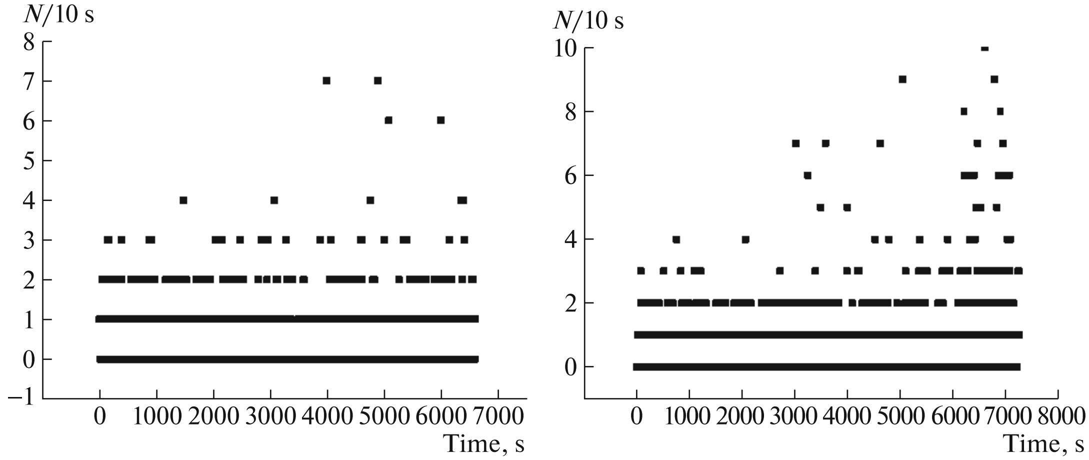

Fig. 2. Counting of a neutron detector based on four He-3 counters for 10 s. The targets are Pd/PdO: H_x (*a*), Pd/PdO: D_x (*b*).

irradiation was repeated. The summary results of the same type of irradiation regimes are presented.

EXPERIMENTAL RESULTS AND DISCUSSION

In the test mode, measurements were carried out on an electron beam in the energy range 20–40 keV and beam currents of 100–170 μA. We used Pd/PdO: D_x targets 2.5–1 cm^2 in size and 50-μm thick. A Pd/PdO: H_x target (saturated with hydrogen under similar conditions) was used as a background target. The readings of the detector based on four He-3 counters for this series of measurements are shown in Fig. 2. The target was moved with a step of 3 mm every 15 minutes. In this case, the beam operation mode was changed—energy 20–40 keV, current 100–170 μA. The background count of the detector (with the beam turned off) was $\langle n\rangle = (0.69 \pm 0.03)/10$ s. The detector data based on He-3 counters are given in Table 1. The average detector count for the entire exposure time and the percentage of counts that exceed 3σ from the average background are given.

Table 1. Detector data based on He-3 counters

Sample	Average count of He-3 detector, s^{-1}	Percentage of exceeding the average background level $\langle n\rangle + 3\sigma$, %
Pd/PdO: D_x	0.10 ± 0.004	4.3
Pd/PdO: H_x	0.081 ± 0.003	1.2
Background without beam	0.069 ± 0.003	0.4

It can be seen from Table 1 that the average count of a detector based on He-3 counters under irradiation of a deuterated palladium sample is about 20% higher than for a hydrogen-saturated sample, and about 30% higher than the natural background. Also, the percentage of counts above the average background value for a deuterated sample is much higher than for a hydrogen-saturated sample.

When measuring neutron emission with two scintillation detectors, we used PdD_x targets of various thicknesses and deuterated CVD diamond. Electron beam parameters: current 150 μA at an energy of 30 keV and a current of 100 μA at an energy of 40 keV. The detectors were located at a distance of 17 cm from the target. The target holder was moved at a speed of 20 μm/s or 10 μm/s. The beam diameter on the target was 3 or 6 mm. The movement was required to reduce the radiation load on the targets. Background measurements were carried out both without an electron beam at different times of the day and when the beam was exposed to pure copper and an aluminum holder. Scintillation detector data are shown in Table 2. The detectors were calibrated using gamma sources, therefore in such detectors in energy spectra the unit of energy is used as the unit keVee, meaning keV in electronic equivalent. The energy range 100–1500 keVee is divided into two sub-ranges. When processing the data, we chose two thresholds for recording recoil protons: lower and upper. The lower threshold is 100 keVee. Below this level, it is difficult to reliably separate neutrons and gamma quanta. The upper threshold is 750 keVee to isolate neutrons related to the DD fusion reaction. The selected range (100–750 keVee) roughly corresponds to the energy range of recoil protons from ≈600 keV to ≈2.8 MeV, which correspond to neutrons with an energy of 2.45 MeV—the products of

Table 2. Summarized results of two scintillation detectors for similar measurement sessions

Target (thickness μm)	Beam. Energy (keV)/current (μA)	Sum measurement time, s	Number of neutrons (sum of readings of two detectors)		Average neutron count by two detectors n, s^{-1} 10^{-3}		n_n (0.6–2.8 MeV) Bg—Day background without beam s^{-1} in 4π sr
			100–750 keVee	750–1500 keVee	100–750 keVee	750–1500 keVee	
Day background without beam	–	112560	199	19	1.77 ± 0.13	0.17 ± 0.04	–
Cu(100)	30/150*	9600	15	0	1.56 ± 0.40	0	0
Cu(100)	40/100*	10800	19	0	1.76 ± 0.40	0	0
CVD + D(400) + converter Ti(55)	30/150*	13200	20	0	1.52 ± 0.34	0	0
CVD + D(400)	30/150*	6000	10	0	1.67 ± 0.53	0	0
PdD_x(500)	30/150*	35040	76	7	2.17 ± 0.25	0.20 ± 0.08	0.96 ± 0.52
PdD_x(500)	40/100*	37200	83	8	2.23 ± 0.24	0.22 ± 0.08	1.12 ± 0.50
PdD_x(40 + 50)	30/150*	18000	34	3	1.89 ± 0.32	0.17 ± 0.10	0.29 ± 0.64
PdD_x(40 + 50)	40/100*	24000	55	4	2.29 ± 0.31	0.17 ± 0.08	1.26 ± 0.62

*—beam diameter—6 mm.

the DD reaction. The upper threshold is 1500 keVee and is due to the dynamic range of scintillation detectors. The range 750–1500 keVee corresponds to recoil protons from ≈2.8 MeV to ≈4.3 MeV. The upper threshold is 1500 keVee and is due to the dynamic range of scintillation detectors. When calculating the neutron yield from the target, the following were taken into account: the background, the geometric factor and the detection efficiency of fast neutrons (2.45 MeV) by scintillation detectors. In this case, neutron scattering in the chamber wall and the lead caps of the detectors was not taken into account. The detection efficiency of fast neutrons with an energy of 2.5 MeV with a scintillation detector was measured with an ING-07D generator.

A CVD diamond target covered with 55-μm Ti from the direct action of the electron beam did not show any excess over the background. The uncoated CVD diamond target showed no excess over the average background, with the PdD_x target showing the maximum excess over background. The average neutron flux can be estimated at the level of ~ 1 s^{-1} in 4π sr. In this case, the maximum amount of deuterium in the sample can be estimated as 4.3×10^{21}. Then the neutron flux can be estimated as $\sim 3 \times 10^{-22}$ s^{-1} per D atom. Comparison with the neutron flux from DD fusion obtained in [11], 1.5×10^3 s^{-1} in 4π sr at the total amount of irradiated deuterium 5×10^{24} atoms gives 3×10^{-22} s^{-1} in 4π sr per atom D. This value is comparable to the neutron flux per deuteron obtained in our work. In [11], bremsstrahlung from electrons was used to stimulate nuclear fusion, while in our work we used the combined effect of both electrons directly and their bremsstrahlung on a deuterated target. It should be noted that the spectrum of X-ray quanta (and electrons) in [11] was much harder (<3 MeV), and the number of irradiated deuterium atoms is about 10^3 times greater than in our work.

The authors of article [11] believe that the main factor leading to the intensification of the DD reaction, in their case, was the elastic scattering of neutrons from the decay of deuterium nuclei under the action of hard X-rays. In our work, the effect was achieved at a much lower electron energy (and X-ray quanta E <40 keV), which does not lead to the decay of the deuterium nucleus into a proton and a neutron (the reaction threshold is 2.2 MeV). Thus, we have shown that the direct action of electrons on a deuterated target is also effective for initiating the DD reaction.

The track detectors were located on the target

Table 3. Estimates of neutron fluxes using the CR-39 detector

Detector index	Target, thickness, μm	Number of deuterium atoms before irradiation (escaped from the sample)	Irradiation time, s	Irradiation conditions Energy (keV)/current (μA)	Number of proton tracks on the back of the detector, cm^{-2}	The number of decays of nuclei into 2 particles, cm^{-2}	The number of decays of nuclei into 3 particles, cm^{-2}	Neutron flux n_n (>0.4 MeV) s^{-1} in 4π sr
B4-1	PdD$_x$(500)	4.1×10^{21} (8.0×10^{20})	26400	30/150*	42	7	2	13 ± 4
B4-2	CVD + D(400) + Ti(55)	7.5×10^{20} (7.5×10^{20})	13200	30/150*	26	2	0	On the background level
B4-3	CVD + D(400)	7.5×10^{20} (7.5×10^{20})	6000	30/150*	18	3	0	On the background level
B4-4	PdD$_x$(500)	4.3×10^{21} (9.4×10^{20})	37200	40/100*	66	11	1	18 ± 5.5
B4-5	PdD$_x$(40 + 50)	1.8×10^{21} (1.2×10^{21})	27600	40/100*	56	11	1	20 ± 5
B4-6	PdD$_x$(40 + 50)	1.7×10^{21} (1.6×10^{21})	24000	30/150*	40	12	1	13 ± 4
B3-1	PdD$_x$(40)	1.1×10^{21} (1.1×10^{21})	48000	30/150**	60	10	2	9.4 ± 3.6
B3-2	CVD + D(400)	7.5×10^{20} (7.5×10^{20})	52000	30/150**	32	10	1	On the background level
B3-Bg1	Cu(100), Al	–	18600	30/150**	22	1	0	0
B4-Bg2	Cu(100)	–	10800	30/150*	18	3	0	0
Bg0	Detector without irradiation	–	–	–	15	1	0	0

*—beam diameter—6 mm, **—beam diameter—3 mm.

holder at a distance of about 2 cm. The coating of the detectors (44-μm Al) made it possible to avoid the illumination of the detector by scattered electrons. The detection of nuclear decays into two (n(^{16}O, ^{13}C)α reaction with a threshold of ∼3 MeV) and on three particles (n(^{12}C, 3α)n' reaction with a threshold of ∼10 MeV) gives an indication of the presence in the spectrum of neutrons with energies greater than 3 and 10 MeV, respectively (see. Fig. 3 and Table 3). We recall that in [11], along with neutrons with an energy of 2.45 MeV, neutrons with energies of ∼4 and ∼5 MeV were also detected.

Table 3 shows that the estimates of the average neutron flux during irradiation of deuterated targets are at the level of ∼10 s^{-1} (or ∼10^{-20} s^{-1} per atom of mobile deuterium) in 4π sr. This is higher than the estimates of neutron fluxes obtained by other detectors (based on He-3 counters and scintillation detectors). The scatter in the estimates obtained by different methods can be explained by the ambiguity of the calibration of the detectors with neutrons of different energies (the efficiency of neutron detection strongly depends on their energy). Accurate quantitative es-

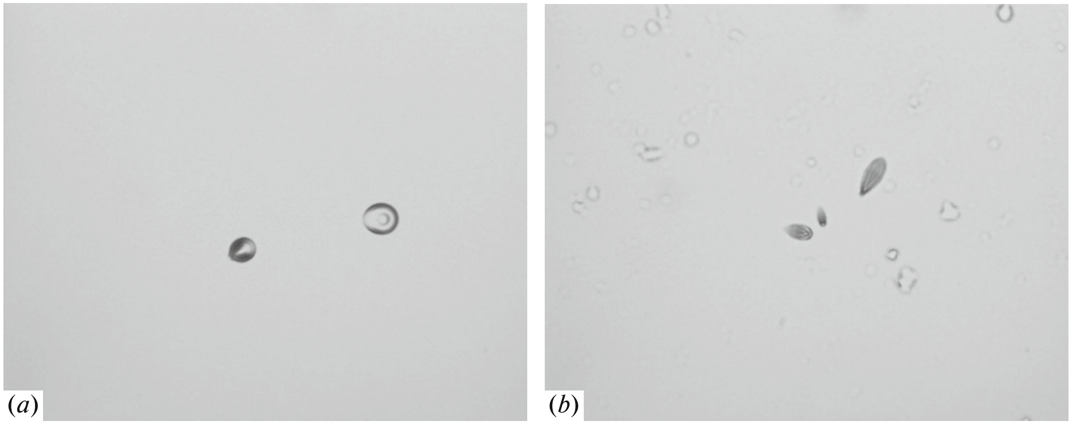

Fig. 3. Photomicrographs of nuclear decays into two (a) and three particles (b) on track detectors CR-39. Image size 120×90 μm.

timation of neutron fluxes requires an additional joint calibration of all detectors using a DD generator.

CONCLUSION

Thus, it has been shown by three independent methods that the action of an electron beam in the energy range of 20–40 keV initiates DD-fusion reactions in deuterated crystal structures with a neutron yield. Indications were obtained that, along with neutrons with energies of 2.5 MeV, there are neutrons with energies greater than 3 MeV.

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