Search for double K-capture of 124Xe with the large volume copper proportional counter

Yu.M. Gavrilyuk¹, A.M. Gangapshev¹, V.V. Kazalov^{1,*}, V.V. Kuzminov¹, S.I. Panasenko², A.D. Petrenko², S.S. Ratkevich², D.A. Tekueva¹, and S.P. Yakimenko¹

¹Institute for Nuclear Research, RAS, Moscow, Russia; kazalov@inr.ru

²V.N.Karazin Kharkiv National University, Kharkiv, Ukraine

Abstract During several years at the Baksan Neutrino Observatory INR RAS is carrying out the experiment for searching of 2K (2ν)-capture in ¹²⁴Xe. This isotope has several advantages: 1) it has the largest kinetic energy of transition among candidates of nuclei for which predicted the existence of *ECEC*; 2) since xenon is the noble gas, then it could be easily usen as a system isotope-detecting medium in a gas detector. To search for 2K-capture in ¹²⁴Xe the large volume copper proportional counter (CPC) is used.

In our work, we present the results of the simulation, with the Geant4 package, of CPC background from the decays of ²³⁸U and ²³²Th nuclei in the construction materials of the CPC case, as well as in elements of the low-background shield. The influence of neutrons produced in the rock of the underground laboratory from the decay of ²³⁸U and ²³²Th, where the experimental setup is located, on the production of the isotope ¹²⁵I in the working gas of the detector, upon capture of thermalized neutrons by the ¹²⁴Xe isotope is considered. The ¹²⁵I isotope can have a significant influence on the background of the experiment since the total energy release in its decay belongs to the same energy region as ROI of 2K-capture in ¹²⁴Xe.

Keywords: Double beta-decay, 2k-Capture, Proportional Counter, Low-Background Experiments

1. Introduction

A unique state is formed in the daughter atom in the case of the capture of two electrons from the *K*-shell. This state represents a neutral atom with the inflated shell, exposing two vacancies in the *K*-shell. In order to detect such a process, we have to keep in mind that for Z > 30, where *K*-fluorescence yields are large, the dominant decay of double *K* vacancy states happens through the sequential emission of two characteristic fluorescence quanta.

The primary contribution (76.7%) to the ECEC process in ¹²⁴Xe is produced by the capture of two electrons from the *K*-shell [1]. The result of the ¹²⁴Xe (2v, $2e_K$) reaction is a neutral ¹²⁴Te atom with a "lifted" shell, which leaves both *K*-shell vacancies exposed. The residual excitation of the atomic shell in daughter isotope ¹²⁴Te^{**} relaxes via the emission of Auger electrons (e_A , e_A), a single characteristic quantum and an Auger electron (K, e_A), or two characteristic quanta and low-energy Auger electrons (K, K, e_A). In actual experiments, the almost simultaneous emission of two characteristic fluorescence quanta produced in the filling of two vacancies offers considerable advantages in terms of detection of such events.

The detection of the relaxation response of atomic processes after the capture of atomic electrons in a gas medium offers several significant advantages over liquid and solid-state

detectors. At the same time, the impact of the background depends strongly on the energy resolution, which gives the event detection in a gas medium another advantage over detection in liquid. In addition, the interaction of radiation in gas provides an opportunity to determine the topological signature of a rare event, which was demonstrated in our earlier studies [2, 3].

In contrast, extremely small primary ionization tracks typical of interactions in liquid make it difficult to identify the specific features of the signal from low-energy primary particles.

The search technique in our experiment is based to the registration triple coincidences of "shaked" electrons and two fluorescence photons produced in the process of filling of a double *K*-shell vacancy in daughter atoms. A proportional counter filled with gas containing the studied isotope is used as a detector. Useful events have a unique feature set in such a detector. Their total signal comprises three partial pulses with known amplitudes formed as a result of absorption of two characteristic photons and a cascade of low-energy Auger electrons within the working volume. The selection of events with such features from the entire set of digitized pulses allows one to raise the effect/background ratio up to several thousand times. Thus, in our case, the main contribution of the background in the energy region of interest stems from three-point events in the detector. The main contribution to such events can result from the processes of double ionization of the *K*-shell or the emission of a low-energy gamma-quantum with simultaneous registration of the relaxation products of the daughter atomic shell.

2. Setup description

The model of the experimental setup "2K-CAPTURE" was created with the help of the Monte-Carlo *Geant4* package [4] for studying the detector response to decays of radionuclides from U/Th families. The same model was used for simulate neutron transport and scattering. Fig. 1 shows a cross section of the setup as implemented in the simulation. All the most relevant geometrical features are included, such as the multilayer shield with their real shapes and positions. The experimental setup consists of a copper proportional counter (CPC) with a casing made of M1-grade copper (the inner surfaces of casing and flanges were covered additionally with 1.5 and 3 mm layers of M0k copper correspondingly) and passive shield. Shield consists of 18 cm of copper, 15 cm of lead and 8 cm of borated polyethylene.



Fig1. Simulated geometry for the installing "2K-CAPTURE" with a calibration channel in Monte-Carlo Geant4 package.

Additional constructive parts located on the detector flanges, which have high voltage inputs designed to supply high voltage to the anode of the CPC, as well as the pickup of the

signal through a high-voltage separating capacitor, were not taken into account in the model. In the model, the body of the CPC is presented as a 710-mm-long copper cylinder with a working length of 595 mm and an inner diameter of 137 mm, closed on both sides with flanges, Fig.2.



Fig2. Model of copper proportional counter. A darker color indicates the operating area of the detector.

The following structural parts were taken into account in the internal volume of the CPC: a) the anode wire made from gold-plated tungsten with a diameter of 10μ m thick is stretched along the cylinder axis; b) the anode wire passes through the copper tubes along the edges of the detector, which in turn is inserted into the Teflon insulators. The gas volume of the detector is represented by two logical volumes: the full volume and the fiducial volume of the detector, which is part of the full volume, and is located between the copper tubes in which the anode wire is included. The same volume is a sensitive area of the detector; in Fig.2 it is represented in a darker color.

The CPC is filled with xenon gas to a pressure of 4.8 atm. Xenon has a certain isotopic composition that was taken into account when creating the model. The isotopic composition of gas is presented in Table 1.

Isotope	124	126	128	129	130	131	132	134	136
Content, %	20.311	27.12	33.44	18.812	0.071	0.057	0.026	0.088	0.0806

Table 1. The isotope composition of working gas of xenon.

As mentioned above, the CPC is surrounded by multi-layer low-background shield. For example, copper shield also has several volumes. Therefore, to simplify the simulation: a) copper layer around the detector housing located between the counter flanges; b) copper layers on the ends of the detector; c) a layer of copper that surrounds the detector and copper portions on the ends of the detector, in this volume calibrated holes are made, and through it the Teflon calibration channel.

3. Simulation results

The following libraries were used for modeling and calculations: G4DecayPhysics – for modeling decays of unstable particles indicated in *PhysicsList*, as well as for all unstable particles that may occur during the simulation; G4RadioactiveDecayPhysics [5] - allows to simulate radioactive decays of various isotopes, based on ENSDF data [6]; the *G4EmPenelopePhysics* library was used to simulate electromagnetic processes, and *G4EmLivermorePhysics* (for test) was also considered; The

G4HadronPhysicsQGSP_BIC_HP library was used to simulate the interaction of neutrons with matter, this library was chosen based on [7]. The radioactive source (nuclear decay, alpha, beta and gamma radiation) is specified in a mac-file using the *General Particle Source* library. The models we created on the basis of two libraries allowed us to obtain a good agreement between the calculated and experimental response functions from an external calibration gamma source in most of the analyzed cases.

Fig. 3 shows a comparison of the measured and simulated spectra from an external ¹⁰⁹Cd-source. The source is placed above the detector in the calibration channel, which is located inside the low-background shield, Fig.4. To reduce the absorption of gamma-quanta in the shield substance, three calibration holes are made above the center of the detector and along the edges of the sensitive zone of the detector. In standard mode, calibration is carried out in the center hole. The same was done in the model.



Fig3. A comparison of experimental and simulated spectra of CPC obtained from 109Cd-source. Red and green histograms are the Geant4 simulations obtained with the Penelope and Livermore model respectively. Black histogram are experimental data from 109Cd-source.



Fig 4. Calibration channel and calibration holes.

As noted in the introduction, the purpose of our experiment is the search for rare events from 2v2K capture using registration a time correlation (coincidence) between the two K-fluorescence quantum that are emitted after the decay of double K-vacancy states and Auger electrons. The magnitude of the probable effect is estimated on the basis of the results of comparative analysis of background data of the proportional counter of the proportional counter filled with samples of pure xenon with different concentrations of the studied isotope. The ultimate sensitivity of the setup to the effect of interest depends primarily on the intrinsic detector background and the quality of the methods used to separate the desired signal with a specified set of characteristics. This requires an extra small gamma-background. In the same

time, a double K-shell photoionization of the atom can create the "hollow atom" by absorbing a single photon and releasing both K electrons. Relaxation of the excited electron shell of a daughter atom can mimic the useful signal.

As noted in the introduction, the purpose of our experiment is the search for rare events from 2v2K capture using registration a time correlation (coincidence) between the two *K*-fluorescence quantum that are emitted after the decay of double *K*-vacancy states and Auger electrons. The magnitude of the probable effect is estimated on the basis of the results of comparative analysis of background data of the proportional counter of the proportional counter filled with samples of pure xenon with different concentrations of the studied isotope. The ultimate sensitivity of the setup to the effect of interest depends primarily on the intrinsic detector background and the quality of the methods used to separate the desired signal with a specified set of characteristics. This requires an extra small gamma-background. In the same time, a double *K*-shell photoionization of the atom can create the "hollow atom" by absorbing a single photon and releasing both *K* electrons. Relaxation of the excited electron shell of a daughter atom can mimic the useful signal.

3.1. Gamma-background simulation.

Gamma ray background from the structural elements of the shell of the detector as well as in the elements of the shield (copper, lead) is almost exclusively coming from the uranium and thorium decay series as well as from decay of ⁴⁰K. External gamma backgrounds originate from these radionuclides in the surrounding rock of deep underground laboratories. Therefore, our purpose was to estimate the response of the CPC to the gamma background from surrounding material. The activity of natural radioactive isotopes content in different source materials of the low-background shield was measured by the ultra-low background germanium gamma-spectrometer [8]. The simulation included the calculation of decays of members of uranium-radium and actinium natural radioactivity chains contained in the structural elements of the detector shell as well as in the elements of the shield. Fig. 5 shows the background spectrum accumulated for one year of measurement (curve 1) using CPC and Monte-Carlo simulation of gamma-spectrum (curve 0). As can be seen in the figure, the simulated and experimental spectrum of all events with energies above 30 keV coincide quite well. The difference between the spectra at low energies can be related to the fact that at the ends of the detector the proportional amplification changes to the ionization mode, which was not taken into account in this simulation. Also edge effects at the ends were not considered.



Fig 5. Spectrum of the background of CPC filled with xenon: (1) - all events, (2) - two-point events, (3) - three-point events, (0) - Monte-Carlo simulations spectrum of low-energy gamma-quantum for interactions in the detector.

The same figure shows the spectra of two- and three-point events, curve 2 and 3 respectively. Events that are associated with a photon-absorption are distributed among the groups of oneand two-point events in accordance with the probabilities of the Compton process, photo absorption, and the probability for the characteristic radiation yield upon the photo-absorption induced filling of the K-vacancy in the target atom. Two-point events take place if, for example, a photon undergoes the Compton scattering and it is absorbed in the counters volume, a photon undergoes photoeffect and fluorescence quantum are emitted in the process of the vacancy filling or an electron creates the bremsstrahlung. In all these cases, secondary photon is absorbed at some distance from its origin and creates second cluster of ionization. If these two clusters are located at a different distance from the anode, the detected signal will be made of two pulses, with the time difference corresponding to the moments of entering the gas amplification region by two groups of primary electrons. Three-point events can appear if all secondary radiation is absorbed inside the counter, as in, Compton scattering of a photon, followed by the photoeffect and release of the fluorescence quantum by filling the vacancy in the atomic shell, photoeffect followed by the K-shell ionization, by the photoelectron in another atom and release of two fluorescence quantum. Three-point events, which are the main goal of our study, have to be formed as a result of absorption into the fiducial volume of the counter of two characteristic photons and the Auger electrons generated by the double K-shell vacancy production in the investigated xenon samples. All more-than-three-point events are considered as multipoint events. They can appear, for example, as a result of Compton scattering of the photon by the K electron, followed by the K shell photoeffect and release of two K fluorescence quantum. Events can move around within multipoint groups because of possible overlaps of the pulses of separate components of multipoint events. A detailed description of the selection of multipoint events and their components can be found, for example, in [9].

3.2. Neutron background simulation.

The ¹²⁵I may be one of the most critical background sources in the energy region of interest for the search the 2v2K capture in ¹²⁴Xe. Since the energy and type of emission of its decay products are similar to the expected effect of 2v2K capture. In our detector, this isotope may form after the decay of ¹²⁵Xe. It is formed when a thermal neutron is captured by the ¹²⁴Xe with a sufficiently large probability ($\sigma_{th} = 165\pm11$ bn [10]). This can make to the impossibility to separate the desired signal from 2*K*-capture with a significant flux of background thermal neutrons. The half-life of ¹²⁵Xe is about 17 h, after which it decays into an atom ¹²⁵I. Its half-life is 59.49 days and it decays by electron capture (100%) to the first excited state of the daughter ¹²⁵Te which then emits a 35.46 keV gamma ray photon in dropping to its ground state in 1.49×10⁻⁹ seconds. Simultaneous registration of the 35.46 keV gamma-line, characteristic *K* fluorescence quanta, and Auger electrons in working volume of CPC can generate a three-point event with a signature that simulates an event from a 2v2K capture of ¹²⁴Xe.

Therefore, we decided to test the possible number of neutrons that can "penetrate" into the working volume of the detector, thermalize and interact with the ¹²⁴Xe. Several possible sources of neutrons were considered in the simulation calculations. The first of them is neutron production in (α – n)-reactions during the decay of the U/Th series and their daughter nuclei (which undergo α -decay) in the detector case, as well as in the shield elements. These calculations showed that we have zero effect. The next stage included the calculation of the penetration of neutrons produced in the walls of the low-background underground laboratory and evenly distributed in the air.

In the calculations, we focused on the work in which measurements were made of neutrons of different energy ranges, in the same laboratory where the installation for searching for 2*K*-capture in ¹²⁴Xe is located. In the first work, the measurements of the thermal neutron flux were carried out on the neutron background measurements with the [ZnS(Ag)+6LiF] scintillation detector [11]. As a result of the measurements, an estimate was obtained for the thermal neutron flux at the level $(2.6\pm0.4) \times 10^{-5} \text{ cm}^{-2}\text{s}^{-1}$. In the second work [12], measurements of neutrons with energies of ~ 700 keV were carried out. The value of the neutron flux with such energy of $5.3 \times 10^{-7} - 1.8 \times 10^{-7} \text{ cm}^{-2}\text{s}^{-1}$ was obtained.

These values were incorporated into the design model of our installation. The neutron layer surrounding the shield from all sides was selected with a thickness of 100 mm. Monte-Carlo simulation of spectra of neutron passing through the material of passive shield and scattered in the CPC allowed us to obtain the following results. The thermal neutrons (in reality neutrons with energies below 0.5 eV were taken) do not penetrate through the low-background shield into the detector, and therefore do not contribute to the production of the ¹²⁵Xe. Neutrons with an energy of ~ 700 keV can penetrate into the detector through low-background shield. We have ~ 2.5 thermalized neutrons in the detector when normalizing the simulation results for a year of measurements. We will have 0.02 atoms of the ¹²⁵I in the detector in one year of measurements, taking into account the interaction cross-section and the number of ¹²⁴Xe atoms. This will give us no more than one atom of the ¹²⁵I in our detector in 4-5 years of measurements.

4. Conclusions

The reducing of the background is crucial when searching for the 2v2K capture in ^{124}Xe because it determines the sensitivity when the total amount of ^{124}Xe is equal to fixed. To estimate the radioactive contamination of the experimental setup "2*K*-*CAPTURE*" has been applied the *Geant4-based* Monte-Carlo simulation of the energy spectra from background sources. The energy distributions of the detector response obtained in the *Geant4* MC simulation were compared with measurements from a calibration source, which resulted in a good description of the data. Detector response functions of thermal neutrons penetrating into the working volume of the CPC have also been calculated.

References

- [1] Doi M, Kotani T 1992 Prog. Theor. Phys. (Kyoto) 87 1207.
- [2] Ratkevich S S et al. Phys. Rev. C 96 6:065502.
- [3] Gavrilyuk Y M et al. 2015 Physics of Atomic Nuclei 78 13:1563.
- [4] Geant4: a toolkit for the simulation of the passage of particles through matter. CERN 2011 https://geant4.web.cern.ch/.
- [5] Hauf Steffen et al. 2013 Nucl. Sci., IEEE Trans. 60 4 2984.
- [6] Evaluated Nuclear Structure Data File https://www.nndc.bnl.gov/ensdf/.
- [7] Deiev O S 2013 Problems of Atomic Science and Technology **3** 236.
- [8] Gavrilyuk Yu M et al. 2011 Preprint INR RAN (Inst. Nucl. Phys. RAS) Moscow No.1301/2011.

- [9] Gavrilyuk Yu M et al. 2010 Instrum. Exp. Tech. 53 57.
- [10] Mughabghab S F "Atlas of Neutron Resonances" 5th Ed. National Nuclear Data Center Brookhaven National Laboratory Upton, USA https://www.nndc.bnl.gov/atlas/.
- [11] Kuzminov V V et al. 2017 Nucl. Inst. Meth. Phys. Res. A 841 156.
- [12] Abdurashitov J N et al. 2000 Physics of Atomic Nuclei, 63 7 1276.