Summary of professional accomplishments

- 1. Name and surname: Krzysztof Adam Miernik
- 2. Academic degrees
 - a) Master of Science Faculty of Physics, University of Warsaw; master thesis: "Betadecay studies of ¹⁰⁶Sb and ¹⁰⁷Sb; supervisor: dr Zenon Janas, 15.06.2005
 - b) Doctor of Philosophy Faculty of Physics, University of Warsaw; PhD thesis: "Twoproton radioactivity of ⁴⁵Fe; supervisor: dr hab. Zenon Janas, 28.09.2009
- 3. Employment
 - a) Uniwersytet Warszawski Wydział Fizyki, 01.10.2009 30.09.2012
 - b) Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 01.10.2010 30.09.2013
 - c) Uniwersytet Warszawski Wydział Fizyki, 01.10.2013 obecnie
- 4. Scientific achievement as defined by the Act "Ustawa o stopniach naukowych i tytule naukowym oraz o stopniach i tytule w zakresie sztuki (Dz. U. nr 65, poz. 595 ze zm.)"
 - a) title: Beta decays of neutron-rich nuclides and accompanying phenomena
 - b) Series of thematically related publications:

[H1] K. Miernik, K. P. Rykaczewski, R. Grzywacz, C. J. Gross, D. W. Stracener, J. C. Batchelder, N. T. Brewer, L. Cartegni, A. Fijałkowska, J. H. Hamilton, J. K. Hwang, S. V. Ilyushkin, C. Jost, M. Karny, A. Korgul, W. Królas, S. H. Liu, M. Madurga, C. Mazzocchi, A. J. Mendez II, D. Miller, S. W. Padgett, S. V. Paulauskas, A. V. Ramayya, R. Surman, J. A. Winger, M. Wolińska-Cichocka, and E. F. Zganjar; " β -decay study of neutron-rich bromine and krypton isotopes" Phys. Rev. C **88**, 014309 (2013)

[H2] K. Miernik, K. P. Rykaczewski, C. J. Gross, R. Grzywacz, M. Madurga, D. Miller, J. C. Batchelder, I. N. Borzov, N. T. Brewer, C. Jost, A. Korgul, C. Mazzocchi, A. J. Mendez II, Y. Liu, S. V. Paulauskas, D. W. Stracener, J. A. Winger, and M. Wolińska-Cichocka; "Large β -delayed one and two neutron emission rates in the decay of ⁸⁶Ga"; Phys. Rev. Lett. **111**, 132502 (2013)

[H3] K. Miernik; "Phenomenological model of β -delayed neutron emission probability"; Phys. Rev. C **88**, 041301(R) (2013)

[H4] K. Miernik, C. J. Gross, R. Grzywacz, M. Madurga, A. J. Mendez II, K. P. Rykaczewski, D. W. Stracener, and E. F. Zganjar; "*No Evidence of Isomerism for the First Excited State of* ^{93}Rb "; Nuclear Data Sheets **120**, 56 (2014) **[H5]** K. Miernik, K. P. Rykaczewski, C. J. Gross, R. Grzywacz, M. Madurga, D. Miller, J. C. Batchelder, N. T. Brewer, C. U. Jost, K. Kolos, A. Korgul, C. Mazzocchi, A. J. Mendez II, Y. Liu, S. V. Paulauskas, D. W. Stracener, J. A. Winger, M. Wolińska-Cichocka, and E. F. Zganjar; "*Excited states in*⁸²As studied in the decay of ⁸²Ge"; Phys. Rev. C **90**, 034311 (2014)

[H6] K. Miernik; " β -delayed multiple-neutron emission in the effective density model"; Phys. Rev. C **90**, 054306 (2014)

[H7] K. Miernik; "*Beta-delayed energy spectrum calculated in effective density model*"; Acta Phys. Pol. B **46**, 717 (2015)

[H8] K. Miernik; "Mean lifetime measurements in low-statistics experiments"; Acta Phys. Pol. B **46**, 725 (2015)

c) Objectives and results

Introduction The current state-of-the-art nuclear models predict about 8000 possible bound systems of protons and neutrons [1]. Since there is no Coulomb repulsion between neutrons, greater part of nuclei will be those with excess of number of neutrons compared to number of protons. So far we have experimentally detected about 3000 nuclides. However, the neutron drip-line was reached up to the isotopes of oxygen (Z = 8) [2, 3]. For comparison the proton emitters, nuclei which are beyond proton drip-line, are known up to the isotopes of bismuth (Z = 83). Thus, the neutron-rich nuclei constitute a majority of a unknown species.

The best experimentally studied cases, due to availability, are the stable or close to stability nuclides. Most of the theoretical models must in some way rely on experimental data in order to fit the key parameters. It is obvious then, that they are mostly based on the properties of the best known cases. One of the still open questions is suitability and reliability of extrapolations of properties of the stable nuclides into the territory of exotic isotopes. Strong asymmetry in number of neutrons to protons may significantly affect the nuclear structure, beyond the consequences derived from the stable nuclei. Such phenomena are observed for exotic systems and are a result of many different effects. One of the examples is the "island of inversion" located around N = 20 for isotopes of neon, sodium and magnesium, where the well-established magic numbers are replaced by the new ones [4]. A similar effect was also observed in the most neutron-rich isotope of oxygen (24 O) [5]. These and other observed phenomena are due to the specific occupation of single particle states, proton-neutron interactions, effects of interaction with unbound states, or a general evolution of the nuclear structure with a growing ratio of neutrons to protons.

So far there are no nuclear models capable of reliable predictions of such effects. Experimental studies of exotic isotopes are therefore strongly needed for further development of nuclear physics. Experimental data are also used in applications of nuclear physics, and are being supplemented by theoretical models in areas beyond experimental reach.

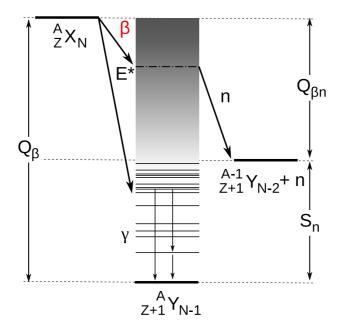


Figure 1: Beta-delayed neutron emission.

One of the most basic methods of experimental studies of exotic nuclei is based on decay spectroscopy. For neutron-rich isotopes the main decay branch is the β^- transmutation. The β decay may, depending on the structure of parent and daughter, lead to excited states. For nuclei close to stability typically they are followed by a γ -rays emission. However, with growth of the β -decay energy (Q_{β}) , the decay may feed states which are beyond the neutron separation energy of the decay daughter (S_n) . Since there is no Coulomb barrier for neutrons, they will be emitted with a large probability even for low energies above the threshold. As a consequence, this decay channel – beta-delayed neutron emission (βn) – will be successfully competing with de-excitation by a γ -rays emission. In the description of this phenomenon often a energy window for neutron emission defined as $Q_{\beta n} = Q_{\beta} - S_n$ is used (Fig. 1). This parameter will be in a natural way connected with the probability of a neutron emission.

From the point of view of a nuclear structure, the β decay can supply information in two ways. The excited states, populated in the decay, emit γ -rays. Energies and connections of transitions into cascades allow us to determine the structure of decay daughter. However, this method requires us to collect a significant number of decays, which is often difficult for exotic nuclei with low production rates. On the other hand, the delayed neutrons emission probability (P_n) and the half-life ($T_{1/2}$) are the two most basic parameters of the β -decay that depend on the integrated quantities of nuclear structure. These values can be determined even in a very low statistics experiments, down to tens of observed decays.

The total beta decay probability on a unit of time (decay constant λ) depends on sum of probabilities of transition to all possible final states in the daughter nuclide. Each transition depends on a squared matrix element, which connects the initial and the final state via transition operator, and on the so-called Fermi function which describes the

phase-space for leptons emitted during the decay. The value of the matrix elements determined in function of the excitation energy is called the beta-strength function – $S_{\beta}(E)$ – and includes the description of the nuclear structure. The Fermi function, or its integral (f(Z, E)) is known and tabularized (e.g. [6]) but the strength function is in general unknown and difficult to calculate theoretically. The half-life and the delayed neutrons emission probability depends on these values in a following way

$$\lambda = \frac{\ln 2}{T_{1/2}} = \kappa \int_0^{Q_\beta} S_\beta(E) f(Z+1, Q_\beta - E) dE,$$
(1)

$$P_n = \frac{\int_{S_n}^{Q_\beta} \frac{\Gamma_n(E)}{\Gamma_{\text{tot}}(E)} S_\beta(E) f(Z+1, Q_\beta - E) dE}{\int_0^{Q_\beta} S_\beta(E) f(Z+1, Q_\beta - E) dE},$$
(2)

where E is the daughter nuclide excitation energy, κ is constant, Γ_n is a width for neutron emission, and Γ is a total state width. The $T_{1/2}$ provides information on a integrated strength function properties, whereas the P_n on the part above the neutron separation energy. Therefore, experimental measurement of these values provides the first insight into the nuclear structure.

The delayed neutron emission was discovered for the first time in 1939 [7]. A sample of uranium nitrate was observed to be emitting neutrons even couple of minutes after the irradiation from a neutron source. In the same year, this phenomenon was interpreted as a delayed neutron emission after beta decays of fission fragments [8]. This discovery had significant consequences for controlling the nuclear reactors. Compared to prompt neutrons, emitted directly during the fission, the delayed neutrons are only a fraction of a total neutron flux. The reactor is operated in a sub-critical state with respect to the prompt neutrons, and the criticality is obtained with the delayed neutrons. The delay in emission allows for mechanical control of the neutron flux. As a result, the delayed neutron emission was studied extensively for applications in nuclear energetics.

The main method used in these experiments is the so-called integration method where a total flux of neutrons from a sample of fissionable material is studied [9, 10]. We should be able to derive the integrated properties from individual nuclides created in a fission. In order to calculate the total delayed neutron flux we need to know, for each isotope, the probability of delayed neutron emission (P_n) , the half-life $(T_{1/2})$ and the total fission yield (Y). In fission of 235 U induced by the thermal neutrons about 1000 different nuclides may be created [11]. In the ENDF/B-VII.1 (2011) database, which is a standard for nuclear energetics, we can find direct fission yields for 844 nuclides, for which 679 $T_{1/2}$ and 153 P_n values are known. Data for other fissionable materials like ²³³U or ²³⁹Pu are similar. From experimental mass measurements and theoretical mass model we can estimate that about 270 ²³⁵U fission fragments may emit delayed neutrons [12] (Fig. 2). This means that the P_n is the least experimentally known value. As a result, the experimental data, supplemented with theoretical models are not able to reproduce the results obtained in the integration experiments. Therefore, for nuclear energetics, a simplified description of delayed neutrons is used, based on the so-called six groups parametrization [9, 12].

Another important information about decays of neutron-rich nuclides that is used in nuclear energetics are the decay schemes. Branching ratios of feeding to excited states in the β -decay and energies and intensities of emitted γ -rays allow us to calculate the decay heat. The decay heat is responsible for about 8% of power of the reactor, however it is a very important source of heat generated after the reactor shutdown. Since many of the fission products have relatively long half-lives, the energy is being released for a period of a few days. One of the key information is the distribution of energy between γ and β radiation. These particles have different penetrabilities and require different type of shielding and result in different cooling time. This issue has not been solved yet, as there are discrepancies between calculations based on individual isotopes and results from integration measurements [13, 14].

The issue of determining the distribution of feeding in the beta decay is also connected with the so-called "reactor anomaly" problem. In experiments studying reactor neutrino oscillations, a deficit of neutrinos was observed in the near detectors where oscillations should not be visible [15]. It might be an indication of a new neutrino flavor, the so-called "sterile" neutrino [15]. However, it might also be due to lack of precision or systematical errors in the decay data for nuclides in the nuclear reactor and the simulated neutrino spectrum [16].

Due to high complexity of nuclear reactor, the microscopic models of its operation are not so far used to simulate or design these devices. However, the less complex problems, e.g. calculations of radiation of spend nuclear fuel are performed in such a way. The constant development of powerful super-computers suggests that at some point it should be possible to simulate the whole reactor, starting from properties of single nuclides. New generations of nuclear reactors may be designed as safer, more efficient and economical devices. In order to perform such calculations, an input from both experimental and theoretical nuclear physics is needed.

Another application of nuclear physics where the neutron-rich isotopes are of a great importance is astrophysics. One of the paths of nucleosynthesis, particularly important for elements heavier than iron, is the so-called r-process [17]. This process takes place in a high temperature and high neutron flux environment, which may appear during the supernovae type-II explosion or the neutron star merger [18]. Beginning from nuclei of iron and nickel, which can be created during the normal burning process in stars heavier than eight masses of the Sun, the sequence of neutron capture reactions and β -decays creates neutron-rich isotopes with growing atomic number (Fig. 2). This process stops at heavy fissionable elements.

The properties of the β -decay have impact on r-process path and final products mass abundances. The nuclear physics data needed in order to simulate the r-process are nuclear masses, cross-sections for neutron capture and photodisintegration, half-lives and delayed neutron emission probabilities [18]. Currently, only a fraction of the rprocess path nuclei are available for experimental studies. Therefore, the nuclear theory models are needed to calculate the properties of nuclides beyond experimental reach. The knowledge of properties of nuclei between r-process path and the stability valley is needed to calculate the final mass abundance formed during the so-called "freeze-out" phase. Delayed neutron emission changes the mass number of the products (beta decay preserves it), but on the other hand it increases the number of free neutrons and gives more time for nucleosynthesis and enhances production of heavier elements. In particular the delayed neutron emission is responsible for smoothing of the mass abundance,

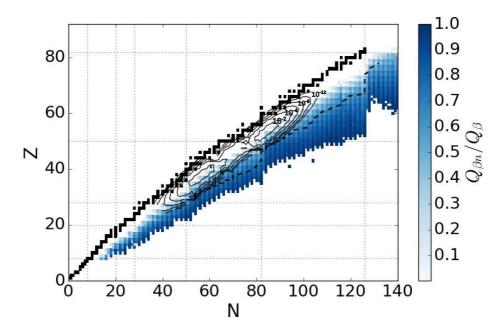


Figure 2: Chart of nuclides with energy window for delayed neutrons diveded by the total decay energy $(Q_{\beta n} / (Q_{\beta})$ shown by intensity of color. Solid countour lines present distribution of ²³⁵U fission fragments. Dashed line show approximate r-process path [19].

creation of the rare-earth peak [20, 18] and the height of the A = 195 peak [20].

Objectives The main goal of this work was to extend the knowledge on decays of neutron-rich nuclides, both by experimental studies and by development of theoretical models. Since it is not possible to finish the studies completely in such a wide field, in a limited time scale, the work was particularly focused on experimental studies of nuclei close to the doubly-magic ⁷⁸Ni (Fig. 3), and on a development of new model of delayed neutron emission.

The region of ⁷⁸Ni was chosen due to a couple of important reasons. The location close to the doubly magic nuclei implies a specific structure of the nuclides, which provides feedback for nuclear theory development. It is of importance also from the point of view of nuclear energetics – one of the maximum of ²³⁵U fission yield is located nearby, and the astrophysics – the r-process starts here (Fig. 3). The history of experimental studies in this area dates back to 1960's. However, there are still many pieces of information missing, also as basic as half-lives or main γ -rays emitted in the beta-decay. Some of the collected data need a critical verification due to large uncertainties. The latter is possible thanks to the development of new detectors and digital electronics, which allows for more thorough data analysis.

Improvements in production of exotic isotopes allowed us to reach to the r-process nuclides in limited areas. This opens a possibility for direct measurements of properties of nuclides needed by astrophysics. One of the interesting phenomena, predicted by the theoretical models, that may occur for the currently most exotic isotopes, is the beta-delayed multi-neutron emission. So far, such decay channels (mainly $\beta 2n$) were

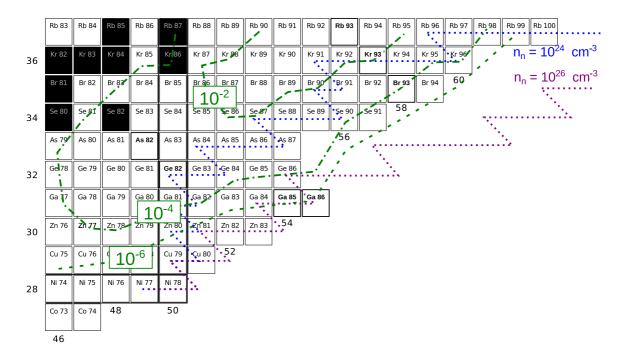


Figure 3: Chart of nuclides showing area of the experimental research described in this work. Dotted lines show approximate r-process paths for two different neutron yields [19]. Dashed lines show contour lines of 235 U fission fragments yields.

observed for light elements, such as ¹¹Li [21] or ¹⁹B [22], that do not take part in the r-process. For heavier elements, only two cases are known ^{98,100}Rb. However, the branching ratio for $\beta 2n$ for these nuclides is extremely low and yields 0.060(9)% [23] and 0.16(8)% [24] respectively. The experimental verification of theoretical predictions of the significant contribution of $\beta 2n$ in the decay of heavier elements may have a important impact on the r-process modeling. Therefore, one of the goals of this work was experimental measurements of the decay of ⁸⁶Ga, which was a candidate for observation of $\beta 2n$ channel.

In parallel to the experimental work, a theoretical model capable of calculations of beta-delayed neutron emission probability for a whole chart of nuclides was developed. New experimental results suggested further improvements of this model, particularly by extending its capabilities of predictions of one, two and three delayed neutrons branching ratios. Most of the currently used models describe the multi-neutron channels by a very simplified method. The goal was to develop and implement a more realistic description of competition of all decay channels (i.e. γ , n, 2n, 3n).

Results

Nuclides important for nuclear energetics Among nuclides important for nuclear energetics decay studies of isotopes 93 Kr and 93 Br were performed. The cumulative fission yield for thermal neutrons and 235 U target for these nuclides is 4.9 and 0.3 mb,

and probability of delayed neutron emission is 1.95(11)% and 68(7)% respectively [11]. This places the chosen nuclides among significant contributors in the environment of the nuclear reactor. The decay of 93 Kr is also important for explanation of a reactor anomaly – it is listed as number 19 for most important contributors from products of fast neutron fission of 238 U. A even more important is the 93 Rb, placed as number 5 contributor in the thermal neutron fission of 235 U [16].

In publications **[H1,H4]** the decay of selected isotopes was studied in Holifield Radioactive Ion Beam Facility (HRIBF) [25, 26] at Oak Ridge National Laboratory (ORNL), Tennessee, USA. The nuclides of interest were produced in a fission reaction induced by a 10 μ A proton beam impinging on a uranium carbide target of 6 g/cm² thickness. The ionized fission products were next extracted from the target by diffusion and effusion, and accelerated to the 40 keV energy. The ions were subsequently mass analyzed by a two-step electromagnetic separator with a preliminary separation with a mass resolution $\Delta m/m \approx 1000$, and a final separation with a mass resolution 10000. The two-step separation process and introduction of a high-resolution magnet allowed us to significantly increase the selectivity of the process compared to the typical mass-separators.

Beam of radioactive ions of mass A = 93 was directed to the decay station equipped with a four high-purity germanium clover detectors for γ -ray detection, two scintillation detectors for β -particles detection, and a moving tape collector. The ions were implanted into a ferromagnetic tape in the middle of the detector setup for a period of a one second, the ion beam was next deflected upstream and a decay of collected activity was observed for a period of one second. After that the tape was moved by 18 inches, in 0.425 s, so the previously irradiated spot was hidden behind a lead shielding. Thanks to that, a background radioactivity from a long-lived decay products was greatly reduced. The cycle was repeated throughout the whole experiment. Both this and other experiments described in this work were based on a digital electronics system where all events are recorded with time-stamps without master triggering system. The spectra, coincidence gates, etc. are built after the experiment by a software program, and the parameters of the analysis may be changed and fine-tuned without loss of data.

In the decay of 93 Kr, an isotope 93 Rb is created. Since 1970, an information on a isomeric character of the first excited state in this nucleus at 253 keV may be found in databases [11, 27]. The reported half-life of this state yields 57 μ s. In our experiment we have proved that the half-life of this state is shorter than 10 ns. This nucleus was studied many times, and with over 200 γ -transistions and 56 excited levels known is one of the best studied β -decays. Nevertheless, wrong information on the isomeric level was copied in all the databases. After our publication [H4] the entry in NNDC database was corrected [11].

The half-life of ⁹³Br of 102(10) ms was measured for the first time in 1988 [28]. At the same time the P_n was reported to be 10(5)%. Later the authors revised the same data and corrected this value to 68(7)%. In out experiment we found a half-life of $T_{1/2} = 152(8)$ ms and $P_n = 53(10)$ %. The difference between half-lives exceeded 3σ for ⁹³Br, while for other isotopes measured at the same time (^{93,94}Kr) we obtained values compatible with literature data. This suggests a systematical error in previous ⁹³Br halflife measurement, but due to lack of details in published material it is not possible to define its source. Apart of correction of half-life, we assigned new γ transitions to this decay, particularly following the delayed neutron emission. It allowed us to determine the P_n for this nucleus with a different than previously used method. New $T_{1/2}$ and P_n values were used in a r-process model [18], and yielded change in final mass abundance on the level of 20% in the mass region A = 92–94.

These experiments are examples of a need of verification of already known cases used in modeling in nuclear engineering and astrophysics. Even though the single isotopes do not have drastic impact on the simulations, the accumulation of many uncertainties and errors may significantly affect the final calculation results. It might be one of the sources of failure in reproducing the integrated delayed neutrons properties starting from data on individual nuclides.

Verification of theoretical models The odd-odd nuclides are in general less well studied, than their even-even or odd-mass neighbors. From the point of view of the-oretical models, the calculations are much more sophisticated in these cases. Even in the simplest shell model approach, the ground state must be described by a residual interaction between a valence proton and a neutron. This interaction leads to a splitting of a multiplet of particles placed on the last filled orbitals. Due to a complex interaction with other particles, often even description of a basic properties of the ground state, such as spin and parity, is difficult. From the experimental point of view, the multiplet splitting leads to the high level density also at low excitation energies, and emergence of phenomena like isomerism due to large spin differences between states. This results in much more complex decay schemes and difficulties in proper interpretation of the results. HERE

These problems make the odd-odd nuclides a perfect playground for nuclear model predictive power tests. Particularly in shell model variants, different interactions are often tuned to selected regions of chart of nuclides. Since the properties of many odd-odd isotopes are unknown they are not taken into account, contrary to the neighboring nuclides. This creates a situation in which the results are interpolated, which should be generally more precise than extrapolation into unknown experimentally territories, and gives an estimation of the lower limit of accuracy of a model.

The odd-odd nuclei are created in β decays of even-even nuclides. The ground state of the even-even nucleus is, in all cases, $J^{\pi} = 0^+$. In case of neutron-rich isotopes only the Gamow-Teller type transitions are possible because the isobaric analog state, which is fed by a Fermi type decay is located above the ground state. The $0^+ \rightarrow 0^+$ transitions are not possible for the Gamow-Teller type decay, therefore the only allowed transitions are $0^+ \rightarrow 1^+$. This allow us to assign, at least partially, spins and parities to the levels seen experimentally in the decay, even in the case of low-statistics experiment when the angular correlation studies are not possible. It is worth mentioning that often the observed states are complementary to those seen by other methods (such as γ -rays emitted during spontaneus fisson of heavy nuclides), where mainly the Yrast states are observed.

In our work **[H5]** ⁸²As was selected as an example of a odd-odd nuclide. The information on the decay of it parent, ⁸²Ge, was rather scarce, with only three γ -rays assigned [29]. At the same time it was known that ⁸²As has an isomeric state, with a unknown excitation energy, that decays via β transition with a half-life close to the one of the ground-state (13.6 and 19.1 s, respectively [30]). The spins and parities of these states were not well estabilised, and different values were proposed in the literature. The ⁸²As, with 33 protons and 49 neutrons, is located close to the doubly-magic ⁷⁸Ni, and should be described quite well by a shell-model.

The experiment was performed at the HRIBF, and a similar production method as described before was used. The average proton beam intensity was increased to 15 μ A, and a resonant laser ionization source was used for extraction of ions of gallium [31]. The implanted gallium ions decayed to the studied nuclides of germanium. Thanks to two-step electromagnetic separation coupled with laser ions source, an isobaric purity of the beam was achieved. A total of 19 γ transitions were assigned to the decay of ⁸²Ge, and spins and parities were determined for 4 levels, including the ground state (2⁻), and the isomeric state (5⁻). The excitation energy of the isomeric state was determined to be 132.1 keV. The experimental results were compared to the theoretical shell model calculations. Two types of interactions were used: realistic (i.e. derived from nucleon-nucleon interactions) represented by N3LO and V18 [32], and effective (i.e. fitted the the known structure of nuclei) JUN45 [33] and JJ44 [34]. Additionally calculations with schematic delta type interaction (SDI) were performed.

The comparison of experimental and theoretical results revealed that both the realistic and the delta interaction is unable to describe the structure of ⁸²As properly, even in a approximate way (Fig. 4). These models indicate ground state of 5⁻, and the lowest 2⁻ state is locate over 1 MeV above, and does not lead to isomerism. The key 1⁺ states needed for ⁸²Ge half-life determination are also misplaced by more than 1 MeV. The results of the effective interaction JJ44 are also of similar accuracy. Only the JUN45 interaction is able to place the states with accuracy better than 0.5 MeV. Closer inspection of the matrix elements between different interactions shows that the largest differences refer to description of interaction of particles at the $g_{9/2}$ orbital. This orbital is particularly important in the case of ⁸²As as it is occupied by 9 neutrons. The verification of shell-model calculations show that the use of the realistic interaction in the description of nuclides beyond experimental reach should be performed with a great care, and, if possible, tested with experimentally known cases.

Beta-delayed two-neutron emission Beta-delayed multi-neutron emission is energetically possible whenever Q_{β} exceeds the two-, three- or more neutron separation energy of the decay daughter. Such a possibility opens for currently most neutron-rich known isotopes. One of the candidates in the ⁷⁸Ni region, predicted by the mass-models, is ⁸⁶Ga. So far it was observed in fragmentation experiments, however, no properties were known, including the half-life.

The experimental results published in **[H2]** were collected during an experiment performed at the HRIBF. The production and extraction was the same as it was described above, with use of the laser ion source. In this case the isobaric purity of the beam was of primary importance, as its intensity was at a very low level of 0.3 ion per second. The detector suite built around the implantation point consisted of 48 neutron counters filled with ³He (600 liters in total), two germanium "Clover" detectors and two β scintilla-

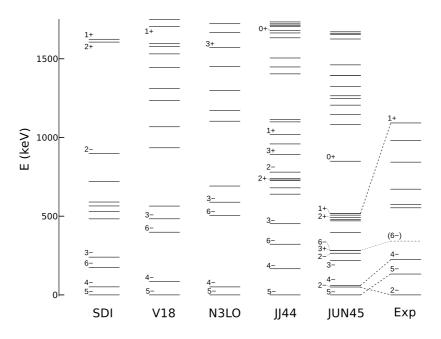


Figure 4: Comparison of experimental and theoretical low-energy structure of ⁸²As obtained with different interactions.

tion counters. Thanks to use of different detectors, all kinds of particles, i.e. γ , β and neutrons, could be detected and correlated with each other. The detector system was complemented, as before, by a moving tape collector. The decay of ⁸⁵Ga was measured before the ⁸⁶Ga, in order to properly interpret the obtained spectra.

The neutron-gated γ spectra shown at Fig. 5 c,d, reveal 624 keV transition for both the decay of ⁸⁵Ga and the ⁸⁶Ga. Since it is correlated with neutron emission in both cases, it must be emitted after two-neutron emission in the case of decay of ⁸⁶Ga. Gamma-transition of such energy is also known from other experiments in the ⁸⁴Ge [35]. At the same time the neutron counters show cases of a simultaneous registration of two neutrons. The ³He counters work on a basis of a neutron capture reaction, so such events can not be a result of single neutron scattering. The hypothesis of random events of two neutrons detection can be rejected based on a statistical analysis. Thanks to the two independent detection methods, it can be therefore proved without any doubt that ⁸⁶Ga emits two delayed neutrons.

In the neutron-gated γ -spectrum (Fig. 5 c) transitions belonging to ⁸⁵Ge (107, 250 and 365 keV) were observed, while in the β -gated spectrum (Fig. 5 a), an unknown before 527 keV line (in anti-coincidence with neutrons) was seen. The latter one was intepreted tentatively as transition from the first excited (2⁺) to the ground state of ⁸⁶Ge. Based on measured energy and Raman systematics [36] the deformation of $\beta_2 = 0.24(2)$, was calculated, which suggests fast onset of the deformation for germanium isotopes. Our measurement indicates that the suggested possibility of appearance of semi-magic number N = 54 [37] above the doubly-magic ⁷⁸Ni can be rejected.

Based on intensities of observed γ -transitions, and on data from neutron-counters, the absolute branching ratios for all three decay branches for ⁸⁶Ga were determined to be

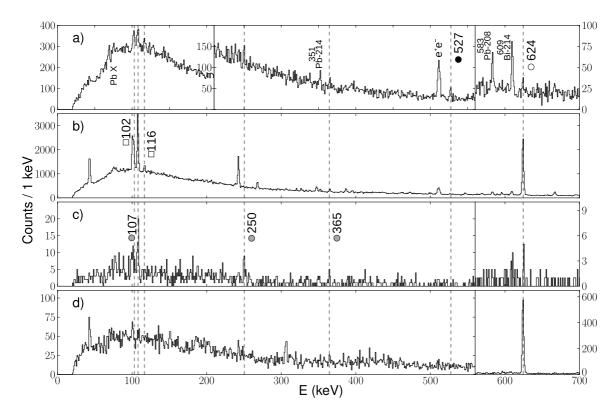


Figure 5: Experimental γ -spectra. Background lines are marked by parent identification, other lines are marked by symbols according to the parent and decay mode: black circles ($\beta 0n^{86}$ Ga \rightarrow^{86} Ge), gray circles (βn^{86} Ga \rightarrow^{85} Ge), open circles ($\beta 2n^{86}$ Ga \rightarrow^{84} Ge), open squares ($\beta 0n^{85}$ Ge \rightarrow^{85} As) (a) β -gated γ -spectrum for ⁸⁶Ga. (b) β -gated γ -spectrum for ⁸⁵Ga. (c) neutron-gated γ -spectrum for ⁸⁶Ga. (d) neutron-gated γ -spectrum for ⁸⁵Ga.

 $\beta 0n \ (20 \pm 10\%), \ \beta n \ (60 \pm 10\%) \ and \ \beta 2n \ (20^{+10}_{-5}\%)$ (Fig. 6). It is worth noticing that an advanced analysis method was used for these calculations, in which delayed neutrons emission from all decay chains starting from ⁸⁶Ga had to be taken into account.

The moving tape collector was operated with a cycle consisting of 2 s of implantation, 1 s of decay with no beam, and 0.7 s tape transport time that removed the previously irradiated spot. The activity build-up and decay periods exceeded many times the unknown previously half-life of ⁸⁶Ga. This fact, together with a low level of statistics, meant that the typical method of grow-in and decay curve fitting could not have been used. A more advanced method, based on a maximum likelihood function was implemented. The likelihood function included the random background events probability and the cycle length. Thanks to this method a half-life of $T_{1/2} = 43^{+21}_{-15}$ ms was found for ⁸⁶Ga.

The obtained value of P_{2n} indicates that this decay branch becomes significant also for heavier isotopes. It confirms the theoretical predictions of importance of this channel for r-process isotopes. This fact should be taken into account both by nucleosynthesis models and by future experiments, that should be able to distinguish this channel from

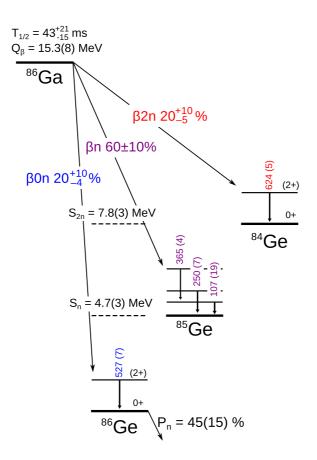


Figure 6: Experimental decay scheme of ⁸⁶Ga. Values in brackets next to gamma line energies are absolute lines intensities normalized to 100 parent decays.

a single-neutron emission.

The production of exotic nuclides, both neutron and proton-rich is, in general, declining when moving out of the valley of stability. Often the new half-lives are determined from a very low number of observed decays. This requires a specific statistical treatment of the data. These methods must also be able to deal with not only short half-lives, but also with significant impact of random events, detectors dead time and limited length of measurement window, that can vary with each individual event. The method of analysis developed for ⁸⁶Ga half-life determination was generalized for other low-statistics experimental cases and published in a separate article **[H8]**.

Phenomenological model of delayed neutron emission From the estimated 8000 possible bound nuclei more than a half meets the delayed neutron emission condition. Currently known emitters are therefore only a small fraction of the total number. Even though the production of exotic nuclides was greatly improved in the recent years, most of the predicted emitters are beyond experimental reach. Many of the r-process nuclides belong to that group. In such a situation an input from theoretical models, particularly the delayed neutron emission probabilities, is needed.

The theory of β decay that is able to calculate the P_n value, should include a description of the parent ground state, excited states in the daughter nuclide, and be able to find probability of transition to all of these states. Models such as a shell-model (e.g. [38]), or Random Phase Approximation (e.g. [39, 40]) attempt to solve these issues on a microscopic level. However, the many-body problem at medium excitation energies represents a major difficulty for these methods. Simultaneous accounting for effects such as a deformation, interaction with continuum, deep-core excitations and many-particle excitation is often beyond their capabilities. In practice, therefore, a phenomenological approach may give better results, however, without full understanding of the problem.

In most of the areas of applications of delayed neutrons the full information on the decay is not needed. Often the P_n value is of primary interest. In order to find this value from Eq. 2, a method of approximating the strength function S_β is needed. In the phenomenological model presented in article **[H3]**, the strength function is modeled with the following equation

$$S_{\beta}(E) \sim \rho(E) = \frac{\exp(a_d \sqrt{E})}{E^{3/2}},\tag{3}$$

which is based on a statistical total level density in a Fermi-gas model. Since not all of the states can participate in the β -decay, the a_d is an effective parameter. It is worth emphasizing that this function does not describe any physical density, and should be treated on a phenomenological level only. In **[H3]** the a_d parameter was found for 159 experimentally known cases, and a systematics of this parameter was created. It was found that it is relatively flat with discontinuities whenever the neutron magic number N = 28, 50, and 82 is passed. It can be described by a following function

$$a_{d}(Z,N) = a_{1}N' + a_{2}Z' + a_{3}\sqrt{N} + \exp(m)$$

$$N' = N - (N_{m}^{i} + 2)$$

$$Z' = Z - Z_{m}^{i}$$
(4)

where N_m^i and Z_m^i is the last closed magic shell, and

$$m = \begin{cases} m_n / \sqrt{N} & N = N_m^i + 2 \text{ or } 3\\ 0 & \text{otherwise.} \end{cases}$$
(5)

Parameters a_1, a_2, a_3, m_n were fitted to the experimental data divided into even-even, odd-odd, and odd-mass nuclides.

The model was compared with experimental data. It as found that among the tested models, including phenomenological Kratz-Herman formula [41], McCutchan et al. formula [42], and microscopic QRPA [40] and Gross-Theory [43] it has lowest χ^2 (Tab. 1 and Fig. 7). The predictive powers were tested by removal of the 41 most neutron-rich isotopes from the dataset. The parameters were refitted, and results for removed isotopes were tested. It was found the χ^2 was the same as before, therefore the short-range extrapolations of the model are valid.

The recent revival of delayed neutron emission studies and their usefulness for applications in nuclear energetics was noticed by the International Atomic Energy Agency

Table 1: Normalized χ^2 (total χ^2 divided by the total number of experimental points) for different models.

Model	χ^2
Miernik	66
McCutchan [42]	78
KHF [41, 42]	109
Gross Theory [43]	415
QRPA [40]	548

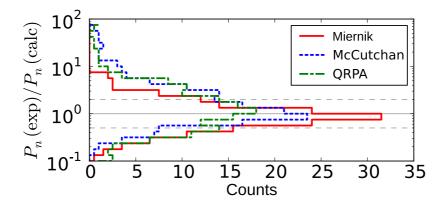


Figure 7: Comparison of distribution of results of different models. Calculated P_n are normalized by the experimental values P_n^{exp} .

(IAEA), which started the Coordinated Research Project aiming at creation of up-todate delayed neutron emission experimental and theoretical results database [44]. The model presented here is included in this database [45].

Equation 2 describes total delayed neutron emission probability, i.e. of one or more neutrons. As it was shown in the case of ⁸⁶Ga it is important to be able to find branching ratio for all decay channels such as βn , $\beta 2n$, etc. So far the main method of calculations of these channels was the so-called "cut-off" approximation. In this method is it assumed that all the feeding into states above the one-neutron separation energy and below two-neutrons separation energy results in the one delayed neutron emission, feeding between two- and three-neutrons separation energy – in two delayed neutrons emission, etc. In reality, it is possible that other channels are active, e.g. emission of a γ -ray or emission of one neutron of larger energy instead of two neutrons of smaller energies (Fig. 8).

In publication **[H6]**, the phenomenological model of delayed neutron emission was extended by calculations of $\beta 2n$ and $\beta 3n$ emission probabilities. The main assumption of the model is the sequential mechanism of emission, which has not been proven experimentally. However, it was confirmed for emission of delayed protons, an analogous process for proton-rich nuclei [46].

In model we assume that only the allowed beta transitions take place, therefore the excited states are described by $I = I_0$, $I_0 \pm 1$ spins, where I_0 is the parent ground state

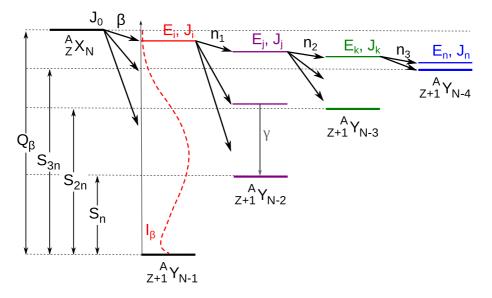


Figure 8: Schematic view of β -delayed many-neutron emission and competing processes.

spin. The relative number of states of each spin is found from

$$w(J_i) = \frac{2J_i + 1}{3(2J_0 + 1)}.$$
(6)

If the spin and excitation energy of the E_i, J_i state are known it is possible to find the neutron transmission factor to the final E_j, J_j state. The nucleus was modeled as a square-well plus centrifugal potential, and transmission factors were found analytically. The density of final states E_j, J_j is found from Gilberta-Camerona statistical formula [47]

$$\rho(U, J_j) = \frac{\exp(2\sqrt{aU})}{a^{1/4}U^{5/4}} (2J_j + 1) \frac{\exp(-(J_j + 1/2)^2/2\sigma^2)}{48\sqrt{2}\sigma^3},$$
(7)

where U is nucleus excitation energy modified by the paring gap, and a is a density parameter. The competition with a γ emission is approximated by an assumption that neutron is emitted if the transmission factor is larger than the threshold, chosen to reflect the experimentally observed properties. The subsequent de-excitation of daughter nuclides are treated in the same manner.

The formula for delayed neutrons emission probabilities obtained in this model is fully analytical, however, it needs numerical treatment of integrals. With this model a P_n , P_{2n} , and P_{3n} values for 1293 neutron-rich nuclei were calculated (Fig. 9).

A significant suppression of probability of emission of two and three neutrons is observed, compared to models based on the cut-off method. It is mainly due to oneneutron emission channel opened also for states above many-neutron separation energy. The largest found P_{2n} values are at the level of approximately 60%, while the cut-off method predicts nuclides for which the P_{2n} reaches 100% [40].

Due to a very limited number of many-delayed neutron emission cases studied experimentally it is not possible to verify the predictions of the model. However, the calculations of number of isotopes that soon will be available for experimental studies were

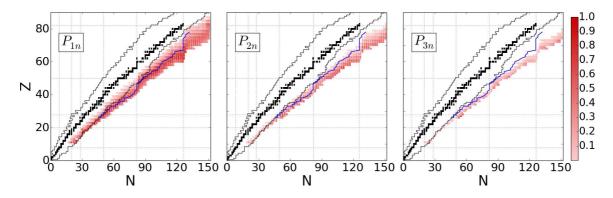


Figure 9: Results of calculations of beta-delayed one-, two-, and three-neutrons emission probability. Dashed line show approximate r-process path [19].

performed. The results of the model were used in planning of an experiment at RIKEN facility at Japan, that should be starting in early 2016.

The delayed neutron emission model described in **[H6]** is capable of calculating the neutron spectra, also in the case of one neutron emission. The experimental data on delayed neutron spectra are limited to about 20 most important cases for nuclear reactors. This allows for an independent verification of the model. It will not be able to reproduce the details of the spectra, as it does not include the description of the nuclear structure. However, the results should be comparable to experimental ones on the statistical level. Such calculations were performed in article **[H7]**, in which the average neutron energy and standard deviation of the spectrum were found for several experimentally cases known (Table 2). The results are on a satisfactory level, taking into account approximations of the model.

Nuclide	$\bar{E_n}^{\exp}$ (MeV)	σ^{exp} (MeV)	$\bar{E_n}^{\mathrm{calc}}$ (MeV)	$\sigma^{ m calc}$ (MeV)
⁸⁵ As	726	9.4	830	10.1
⁸⁷ Br	212	5.6	213	5.9
94 Rb	399	13.7	440	7.5
97 Rb	437	11.8	607	9.7
134 Sn	478	9.4	520	8.4
135 Sb	843	12.0	680	11.0
¹³⁶ Te	381	5.9	325	6.5
¹³⁷ I	538	7.0	604	10.4

Table 2: Average neutron energy $(\overline{E_n})$ and standard deviation (σ) for theoretical and experimental delayed neutron spectra.

Summary and outlook Thanks to the new methods of production and detection the studies of neutron-rich nuclides strongly progressed in recent years. The experimental

results show decay channels that are suppressed for isotopes located closer to the valley of stability. This calls for new solutions by both experimentalist and theoreticians. The studies presented in this work are exploratory and are a starting point for a more detailed work in future.

The measured probability of two delayed neutron emission in the decay of ⁸⁶Ga indicates an important contribution of this decay channel for isotopes near the r-process path. This must be taken into account both by r-process simulations and by future experiments that should be ready to detect this decay mode. It is also worth noticing that the obtained P_{2n} is so far the largest observed value for the whole chart of nuclides. The phenomenological model presents an approach based on a statistical description of nucleus and can be further developed. The results can be useful for planning of future experiments and for nuclear physics applications.

Together with experimental studies on new, unknown isotopes and their decay channels a critical verification of already gathered data is very important. The new experimental methods allow for more detailed or different methods of measurement of already known values, and for detection of possible systematical errors. This was shown in the cases of half-life of ⁹³Br and existence of isomeric state in ⁹³Rb. The nucleus of ⁸²As was so far very little studied in the experiments, even though the information on neighboring isotopes is much more detailed. The comparison of results with theoretical calculations revealed deficiencies in the description of odd-odd isotopes, and can help in future development of the shell-model.

5. Description of other scientific achievements

The references in this part given in round brackets are listed in attachment "Wykaz opublikowanych prac naukowych", part II A. Only the achievements completed after the PhD title was obtained will be discussed.

Neutron-rich nuclei The experimental studies on which the following work is based were conducted together with the team of collaborators. During the experimental campaigns many different nuclei, and different aspects of their decays were studied. In each case, a selected leader was responsible for analysis and preparation of the publication. Nevertheless, the experiment could not be conducted without the help of other team members. Typically, each member has some specific responsibilities. In my case it was taking part in preparation of detectors, development of computer acquisition and analysis codes and participation in data collection. As a team member I am a co-author of number of publications described below.

In publications (24, 34) the main focus was on measurements of unknown half-lives (^{82,83}Zn, ⁸⁵Ga, ⁸⁶Ge) or verification of previous experiments (^{84,85}Ge, ^{84–87}As). These studies are important for nuclear structure theory as well as for nuclear energetics and r-process calculations.

In publications (22, 37, 42, 45, 47, 51, 52) the studies were directed toward the γ -ray emission and β -decay schemes of nuclides $^{71-73}$ Co, 85 Ga, 86 As, and 124,126 Cd. The low-energy excitation schemes were build and compared with nuclear structure models. All measured nuclides are relatively close to the doubly-magic 78 Ni and 132 Sn, which are of particular interest for nuclear theory.

Measurements of β -decay with germanium detectors allow for registration of γ -lines with high precision. However, these detection setup have relatively small efficiency – in the aforementioned experiments is it about 5% for 1.33 MeV γ -rays – which declines with growing energy. This means that germanium detectors setups are not sensitive to de-excitation of levels by many γ -rays of high energy and small intensity. For a complete understanding of decay scheme a complementary measurement with a high-efficiency detectors is needed. One of the solution of this issue, known as "Pandemonium" effect [48] are Total Absorption Spectrometers (TAS). Such detectors are build from large volume of active material and reach high efficiencies. The cost of germanium crystal is large, therefore, usually a much cheaper sodium iodine (NaI) is used. The drawback of such a solution is a poor energy resolution of the NaI detectors. In ORNL I took part in development of modular TAS detector (MTAS), build with 19 NaI crystals of total weight of 1000 kg. This detector reaches detection efficiency of 96–98% in a large range of energies.

Decay studies performed with MTAS detector (30, 41, 44) were focused on most important nuclides for nuclear energetics and decay-heat problem [13]. In all measured cases it was shown that the γ component is underestimated, compared to β component, in previous experimental results that were used in applications in nuclear energetics.

An important part of the experimental studies is construction of a new detectors and devices allowing us to study new isotopes. In publication (28) a development of the neutron spectrometer VANDLE (Versatile Array for Neutron Detection at Low Energies) was presented, in (33) a resonant laser ionization source commissioning experiment, and in (40) a development of 3-Hen neutron detector, that was used in measurements of ⁸⁶Ga.

Proton-rich nuclei The main topic of my PhD was experimental studies of two-proton radioactivity in the case of 45 Fe. This was the first time when an angular distribution of emitted protons was measured, which allowed to get some insight into the mechanism of this newly discovered radioactivity mode. I continue work within this subject with team of collaborators I worked with during the PhD studies. I am a co-author of two theoretical publications connected with two-proton radioactivity (16, 35), and of experimental works aiming at discovery of new two-proton emitters 48 Ni (21, 23) and 59 Ge (49). A still unsolved issue is the connection between the angular correlation pattern and the nuclear structure of the emitter. According to some theoretical models, such a connection may exist. Other suggest that it will be masked by the Coulomb interaction between protons. To answer this question, another experiment with sufficient large statistics (like the 45 Fe measurement) is needed. So far only a few of decays of 48 Ni and 54 Zn were observed.

The studies of two-proton radioactivity were possible due to construction of a detector of a new type – an Optical Time Projection Chamber (OTPC) – which also was part of my PhD thesis. This detector is now used in the measurements of other decay modes with the charged particles emission. Publications in this field are: delayed deuteron emission in the decay of ⁶He (48), delayed tritium emission in the decay of ⁸He (15), delayed protons emission in the decay of ⁴⁸Ni, ⁴⁶Fe, ⁴⁴Cr (43) and ⁴³Cr (17,19). In all cases the production cross-section for isotope of interest or branching ratio for decay-channel is very small. Their selection and detection is possible due to unique capabilities of OTPC, in which the tracks of particles are registered by a CCD camera and a photomultiplier in a event-by-event mode, and fully

reconstructed in 3D. OTPC allows for measurements of delayed particles emission with a low threshold. In a typical experimental setup silicon detectors, sensitive also to β particles, are used. Due to summing of signals from protons and β -particles the detection threshold for protons is about 500–900 keV, depending on the thickness of the detector. In the case of OTPC it is possible to reduce the threshold to about 100–300 keV, depending on the gas mixture used. This means that the OTPC measurements are complementary to the results from silicon detectors, and a more complete picture of charged particles emission in the decay of proton-rich nuclides is obtained.

Super-heavy elements In years 2010–13, I worked at ORNL as a Wigner fellow. One of my main topics I pursued was the studies of super-heavy elements conducted in collaboration with Joint Institute for Nuclear Research (JINR) in Dubna, Russia, and GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany. The ORNL supplied materials for actinide targets used in the super-heavy element research. My task was to broaden the involvement of the ORNL by building a detector setup that could be used in experiments in JINR.

So far in experiments at JINR a position sensitive silicon strip detectors were used. The most modern type of silicon detectors are double sided silicon strip detectors (DSSSD), where strips on both sides allow to precisely determine the position of the decay. The main technique used in super-heavy elements detection is the measurement of chain of correlated α -decays. Thanks to implementation of the DSSSD the correlation of chains should be easier and more unequivocal, compared to the position sensitive detectors. In the latter case a sophisticated and difficult calibration methods are needed to determine the position based on differences in signals detected at both ends of the strip. The new detector setup was coupled with a digital acquisition setup, allowing for detection of decays as fast as 100 ns after the implantation. This is a great development compared to the previously used analogue electronics with a 7 μ s dead time. The development of the acquisition system and analysis codes that were ready to use with other detectors was an important part of my work at ORNL.

The constructed detector setup consists of the so-far largest manufactured DSSSD (128 × 48 strips 1 mm wide) and 7 auxiliary detectors composed in a form of a "box" around the main detector. Together with the acquisition system it was installed in JINR and is used in experiments. As a result of collaboration with JINR I am a co-author of publications (25, 32, 36, 39, 46, 50), in which super-heavy elements were studied. In articles (25, 36) the heaviest isotopes of Z = 117, and Z = 118 were of primary interest. An attempt to create isotope of flerovium (Z = 114) linking the island of heavy-fusion with the nuclear mainland was described in publication (50). A connection by α -chain to nuclides known from other than hot-fusion methods would allow to finally prove the masses and charges of produced elements. Unfortunately, the new isotope of ²⁸⁴Fl is undergoing a spontaneous fission with a half-life of 2.5 ms, and does not emit α -particles.

As a member of collaborative team of ORNL and GSI I am a co-author of publication (38), in which TASCA separator confirmed results from JINR concerning the production of a new element Z = 117 [49]. The ORNL produced target material (249 Bk) for both experiments. This publication, along with (25, 36) were part of material used by International Union of Pure and Applied Chemistry to officially recognize the discovery of elements 115, 117, and 118. In another experiment at GSI at SHIP separator an attempt to create atoms of element Z = 120 was made. The digital acquisition system from ORNL was used for that purpose, as described in (18). Only an upper limit for production of this element in ⁵⁴Cr + ²⁴⁸Cm reaction was found.

Kynh & Kuch

Warsaw, 31 January 2016

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