DEVELOPMENT OF NEW TRENDS IN APPLIED NUCLEAR PHYSICS WITH THE USE OF HIGH-ENERGY BRAKING RADIATION

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A review is given about investigation in nuclear medicine, atomic energetics (Chernobyl problem including), geology etc. that carried out in the NSC KIPT mainly during last decade on the basis of home made electron linacs.

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INTRODUCTION

History of creation of the electron linacs in the NSC KIPT for investigation in nuclear physics is inseparably linked with the endeavour to apply these facilities in different branches of the science & technology. First of all, it were some investigations in nuclear medicine & biology [1,2], dosimetry [3] etc. During last decade owing to commissioning a number of accelerators [4, 5] some new trends have been developed with the use of high-power braking radiation: elaboration of safe for ecology ("soft") technologies of isotopes production (mainly medical ones), characterization of radioactive waste (RAW) and radiation testing of perspective materials and structures for immobilization (disposal) of long-lived RAW, gamma-activation analysis etc. The short survey of some obtained results is given in the paper.

1. ELABORATION OF SOFT TECHNOLOGIES FOR ISOTOPES PRODUCTION

1.1. Nowadays the basic methods for isotope production include nuclear reactions under effect of heavy particles (mainly neutrons and protons) generated in the reactors and accelerators. Although the cross-sections of such reactions are essentially higher than photonuclear ones, however the heavy charged particle interacting with the target material looses rapidly its energy and leaves the resonance region. So, the efficiency of the isotope production (isotope nuclei generation rate per unit of the beam power) on the heavy particle accelerators is not very high.

In case of reactors a great amount of the RAW accompanying the useful isotope production constitutes a problem. For example, while generating 1Ci of ^{99}Mo (parent-isotope for ^{99m}Tc - one of the most widely used nuclide for medical diagnostics) on a reactor is up to 50 Ci of long-lived waste produced parallelly.

Thus, taking into account a continuous growth of the medical isotopes utilization, the elaboration of secure technologies for their manufacturing is a problem of extreme importance.

For production of some medical & biophysical isotopes it is possible to use a braking radiation of the electron accelerator. In this case the generation efficiency is significantly higher than one by using heavy charged particles and neutrons in spite of the relatively low specific activity of produced isotope (≤1Ci/g) [6]. Besides, isotope manufacturing using an electron accelerator is accompanied by much less amount of RAW comparison with another known technologies.

Characteristics of some isotopes, which are used in nuclear medicine and promising for manufacturing on electron accelerator, are shown in Table 1.

1.2. The effective production of isotopes is possible only by providing a large particle flux. Therefore the different versions of design of the converter assembly were investigated as the first stage of technology elaboration. As the criterions of optimisation there were chosen the maximum of the conversion coefficient in the range of braking photons energy corresponding to photonuclear reactions, capability of effective heat rejection, and also maximum absorption of primary electrons in the converter to decrease a heat load on the target. To solve this problem the computer simulation was carried out in 2D-geometry relative to the axis of the electron beam on the basis of GEANT package supplemented with the data on cross-sections of corresponding photonuclear reactions [7].

Taking into account the obtained outcomes we have fulfilled the calculations for generation of different isotopes with the converter unit consisted of two tantalum plates separated by 3 mm water layers. It was supposed as well that the infinite layer with thickness of 10 mm from a given material of natural isotope composition is placed as a target 10 mm apart behind the converter.

Results of the calculations made in such geometry are listed in Table 2. It presents the data for a relative yield of generated isotope $\mathbf{Y_1}$ (recalculated per 1 primary electron), activity of the isotope **A**, produced per one operating day for a beam current 1 mA, and also a beam power **P** absorbed in the target at the optimum regime of irradiation (E=25 MeV).

As one can see from the obtained outcomes it is possible an effective production different radionuclides using high-current electron accelerator. The indispensable conditions of such manufacturing are beam parameters control on the converter & target setup and the ensuring of its continuous cooling.

1.3. The experimental study of regularities of the ^{99}Mo/^{99m}Tc generation using the targets of different phase composition was carried out [8] and experimental samples of ^{99m}Tc as well as ^{57}Co-sources for calibration of gamma-chambers were produced and tested [9-11]. A possibility of production of other isotopes using electron accelerators was investigated also [12,13].

Table 1. *Characteristics of some isotopes for medical & biophysical application*

Isotope	Half-life period, days	Basic decay type	Energy of radiation , MeV	Application	Required activity of isotope
^{32}P	14.36	e^-	1.71	β-radiometry of neoformation, spermatogenesis, immunoenzyme analyze	up to 300 kBq/kg
^{33}P	25.34	e^-	0.248		
^{57}Co	270.9	γ	0.122	calibration of γ-chambers	up to 10^2MBq
^{99m}Tc	0.25	γ	0.140	isotope scanning	10^2... 10^3 MBq
^{181}W	121.2	γ	0.06	enhistotherapy	- « -
^{185}W	75.1	e^-	0.429	- « -	- « -

Table 2. *Data of computer analysis for isotope production*

Isotope	Reaction	E, MeV						P, kW
		15		20		25		
		Y_1	A, MBq	Y_1	A, MBq	Y_1	A, MBq	
^{32}P	$^{33}S(\gamma,p)\,^{32}P$	^{32}P	1.14	$4.37*10^{-8}$	19.0	$1.41*10^{-7}$	61.4	2.19
^{33}P	$^{34}S(\gamma,p)\,^{33}P$	$4.55*10^{-9}$	1.12	$1.60*10^{-7}$	39.4	$6.44*10^{-7}$	15.9	2.19
^{57}Co	$^{58}Ni(\gamma,p)\,^{57}Co$ $^{58}Ni(\gamma,n) \rightarrow {}^{57}Ni \rightarrow {}^{57}Co$	$1.38*10^{-6}$	31.8	$3.32*10^{-5}$	$7.65*10^2$	$9.4*10^{-5}$	$2.16*10^3$	4.98
^{99}Mo	$^{100}Mo(\gamma,n)^{99}Mo$	$7.92*10^{-5}$	$1.73*10^3$	$4.28*10^{-4}$	$9.36*10^4$	$8.54*10^{-4}$	$1.87*10^5$	5.18
^{181}W	$^{182}W(\gamma,n)\,^{181}W$	$1.04*10^{-4}$	$5.36*10^3$	$5.03*10^{-4}$	$2.59*10^4$	$8.86*10^{-4}$	$4.57*10^4$	7.60
^{185}W	$^{186}W(\gamma,n)\,^{185}W$	$4.08*10^{-5}$	$3.4*10^3$	$1.50*10^{-4}$	$1.24*10^4$	$2.48*10^{-4}$	$2.06*10^4$	7.60

2. ELECTRON LINACS IN RADIOACTIVE WASTE PROBLEM

2.1. RAW CHARACTERIZATION

Development of nuclear technologies is accompanied by the growth of RAW including long-lived ones. The waste inside the Chernobyl 4-th unit is of particular importance because their amount is estimated as much as 20MCi. Under the circumstances the problem operative characterization of the waste (i.e. determination of their amount, activity, radionuclide and element composition etc.) is urgent.

This section presents an overview of the RAW characterization methods elaborated in the NSC KIPT and based on γ-activation analysis using braking radiation of high-current electron linac.

Taking into account that a RAW sample activated by high-energy braking photons emits a radiation that is caused both by its inherent activity and one initiated as a result of photonuclear reactions, thus an analysis of such radiation gives quantitative information about radionuclide and element composition of the specimen without its dissection.

For the analysis of large amount of the RAW samples by means of γ-activation method and implementation of other concomitant programs it is needed an electron accelerator with the beam power up to 10 kW and a wide range of particle energy regulation. The complex LU-20 designed in "Accelerator" R&D Prod. Est. of NSC KIPT satisfies these requirements [5]. The necessary set of devices for formation & diagnostics of radiation under γ-activation analysis has been developed.

A studied RAW sample is irradiated as a rule together with a specimen of the standard isotope content. Concentration of this isotope in the sample is determined by means of comparison of induced γ-activity of each sample along the lines corresponding to given isotope (taking into account their mass).

So, there were investigated the samples of lava-like fuel-containing mass (LFCM), which was formed in underreactor premises of the wrecked 4-th Unit of Chernobyl station. The obtained spectrums include the lines of ^{237}U which was generated under activation process in $^{238}U(\gamma,n)^{237}U$ reaction. This example demonstrates the ability of the γ-activation method to analyse the elements which identification is impossible by means of traditional spectrometry methods. Thus obtained quantitative data concerning the element content in the samples of RAW allows carrying out a correlation analysis as well.

It was demonstrated also the ability of γ-activation method in analysis of the samples having their own activity of different nature. So, apart the lines of γ-radiating nuclides (^{154}Eu, ^{137}Cs and ^{134}Cs) there were observed the lines of ^{89}Sr. The last result is especially important because γ-radiating nuclide ^{89}Sr is created as a consequence of the β-radiating ^{90}Sr transmutation [14].

2.2. RAW DISPOSAL

2.2.1. The next RAW handling stage is immobilization and disposal. The materials and geological structures contacting with the RAW have to keep their protective properties with respect to radionuclide displacement under absorbed dose up to 10^8 Gy during 1000 years and more. The conducted investigations showed a possibility of application of the electron accelerators braking radiation in energy range 10...30 MeV to solve a number of tasks for prognostication of the material durability under effect of RAW radiation. So, the main goal of imitation exposure of the materials that are used for immobilization of radionuclides is a creation of absorbed doze up to 10^8 Gy in investigated samples during

acceptable period (as a rule no more than 1 year) at controlable parameters of irradiation.

The gamma-ray unit with radionuclide sources (basically, ^{60}Co) is a traditional radiation source for testing in doze range up to 10^8 Gy. The advantage of such tests is stability of influence conditions to the sample. Therewith, the setup with activity up to 1 MCi is needed to reach the absorbed doze rate (ADR) about 10 Gy/s. Electron accelerator can provide the same parameters under converting its beam to braking photons. So, the ADR in a sample about 10 Gy/s for electrons with energy 10 MeV is reached by converting a beam with power 10 kW, that corresponds to parameters of modern industrial accelerators (e.g. [5]). A possibility of the electron energy and flux control provides also an expansion of range of the influence parameters to the sample under its test.

2.2.2. It is known also that during lasting disposal of the high-level RAW or nuclear spent fuel can arise a situation when the RAW immobilization matrix (including geological structure) will contact with ground water. Thus originates a structure of "RAW-water-geological barrier" type. A radionuclide transport in such structure determines a reliability of the RAW disposal. Such transport depends besides all on absorbed dose of radiation from the RAW.

For a research of radionuclide transport processes the granite specimens (which is considered as a perspective environment for disposal of long-lived RAW) were choosed. A piece of granite was cut into the specimens in the form of blocks with the size of 10x10 mm in cross-section and 30 mm in thickness. Each block was covered with epoxy except for 10x10 surface.

Isotope Yb-169 was used as γ-radiating nuclide-tracer, which is analogous to actinides in its chemical properties. For this nuclide production under reaction $^{168}Yb(n, \gamma)^{169}Yb$ the pellets of stable $^{168}Yb_2O_3$ were irradiated by photoneutrons. Then the pellet was dissolved in concentrated HCl acid (0.2 ml) and finally the aqueous solution with pH=1.8 was prepared.

Obtained solution (40 ml) together with specimen irradiated up to given dose ($3\cdot10^6...3\cdot10^7$ Gy) was placed into thermostable flask. The latter was being heated by water steam during 32 hours.

Then each specimen was being washed in distillate water during 24 hours and dried out at 60°C in the drying box. Further the layers (2...50 μm) from free surface of the specimen were removed by means of precision grinding. Material of the removed layers was used for γ-spectrometry with the Ge(Li)-detector. These results allowed determining a dose dependence of the radionuclide diffusion as well as to find out its mechanism [15].

2.2.3. For the study of radiation & chemical durability of granite its samples were irradiated with braking radiation at two stages: first – at the value of upper boundary of the braking photons energetic spectrum $E_{\gamma\ max}$=10 MeV up to the absorbed dose (AD) of $1,7\cdot10^7$ Gy, after that the samples were activated at $E_{\gamma\ max}$=23 MeV during 7 days up to the total AD of $3,0\cdot10^7$ Gy. Then the samples were kept for some days to reduce their induced radioactivity, ground up into granules with the size less than 0.83 mm which allowed to increase their surface area from 6,2 to 59 cm^2 and underwent dynamic test on leaching in the plant based on Soxhlet extractor. Analysis of the leachant γ-spectrums showed that sodium, rubidium and calcium are leached from granite most intensely. There was no noticeable release of uranium and yttrium from these samples [16].

3. OTHER APPLICATIONS AND METROLOGICAL MAINTENANCE

The characteristic γ-radiation of the samples irradiated by high-energy braking photons can be used also for operative determination of their element (isotope) composition. So, we have showed a possibility of its application in the analysis of the rare and noble metals [17-19] as well as in biophysics [20].

The radiation facilities of the NSC KIPT [5] are enable the testing of different materials, devices and construction elements within a wide range of the radiation parameters and dose values. So, during last period there was tested a number of the fission reactor elements and materials [21,22], magnetics [23], as well as semiconductor detectors of γ-radiation [24].

The analytical methods and technologies using braking photons irradiation [25] demand continuous monitoring of the radiation parameters. To provide a certification of the accelerators and technologies as well as a metrological maintenance of the radiation treatment a number of the working standards and technological measurement channels were developed [26, 27]. The latters are based on the sensors that non-disturb a radiation field (Rogovski coils of different modification [28], radiation – acoustic line [29], thin-wall ionization chambers [30] etc.). Most of them were previously investigated by means of computer analysis using GEANT code [31].

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