ELECTRON LINAC APPLICATION FOR CHARACTERIZATION AND DISPOSAL OF RADIOACTIVE WASTE

N.P. Dikiy, N.A. Dovbnya, S.Yu. Sayenko, V.L. Uvarov
National Science Center "Kharkov Institute of Physics and Technology",
61108, Kharkov, Ukraine
uvarov@kipt.kharkov.ua

Rapid development of nuclear engineering, medicine and radiation technologies is accompanied by increase of the radioactive waste (RAW) including long-lived ones. A RAW handling assumes their element content and activity analysis (characterization), compacting and disposal. This problem is of ultimate importance after shutdown of Chornobyl nuclear power plant Unit 4. The activity of the RAW inside the unit is estimated as much as 20 MCi (mainly on account of the Cs-137). These circumstances ensure a necessity of elaboration of the especial methods for express-analysis of large RAW fluxes. An immobilization of the long-lived radionuclides is entailed in turn with the problem of their localization into stable matrix as well as placing in resistive to radiation containers and geological structures. The report contains an overview of methods elaborated in NSC KIPT for RAW characterization and investigation using the bremsstrahlung of the high-current electron accelerator of radiation stability of the artificial and natural barriers for radionuclide immobilization.
PACS numbers: 29.17.+w, 28.41.Kw

1 INTRODUCTION

Radioactive waste (RAW) management includes a number of procedures. First of all it is RAW characterization i.e. determination of the amount, activity, radionuclide and element content etc. The available methods of RAW characterization are based either on analysis of their inherent radiation (γ-, β-, α-spectrometry) or on the profound radiochemical treatment of the samples with an extraction of the corresponding analyzed fraction for its further spectrometry. The shortcomings of the first group methods are a relatively low accuracy and a restriction to the analysis of only thin RAW layers (particularly β- and α-active ones) as well as a small number of identifiable elements. Second group methods are devoid of these shortcomings. However they are rather labor-consuming, expensive and low operative (a duration of one radiochemical analysis is up to several days). It is known that the activation method based on secondary radiation of electron accelerators is widely used at present for nondestructive express-analysis of samples of ore and different materials [1], fission materials [2] and in other fields.

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

The next RAW management stage is their immobilization and disposal in the steady geological structures. This task calls elaboration of experimental prognostication methods for lasting (up to thousand years) conduct of the disposal environment under complicated radiation and corrosive conditions.

2 ACCELERATOR

2.1. For analysis of large amounts of RAW samples by the γ-activation method and implementation of other concomitant programs it is needed an electron accelerator with a beam power up to 10 kW and a wide range of particle energy regulation.

The complex LU-20 [3] designed at the "Accelerator" R&D Prod. Est. of NSC KIPT satisfies these requirements (see Table 1).

Table 1. Basic parameters of LU-20 Linac

Energy range, MeV	10...30
Pulse duration, μs	4
Maximum repetition rate, Hz	300
Maximum peak current, mA	1000
Maximum average current, μA	1000
Beam scanning frequency, Hz	3
Beam size at the accelerator exit, cm	2x30
Absorbed dose rate (electrons), Gy/h	up to $4 \cdot 10^7$
Absorbed dose rate (braking photons), Gy/h	up to $1 \cdot 10^5$

2.2. A necessary set of radiation forming and diagnostics devices has been developed for ensuring γ–activation analysis (Fig. 1).

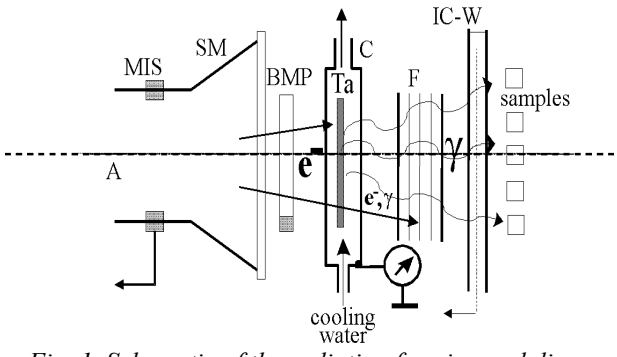

Fig. 1. Schematic of the radiation forming and diagnostics devices.

An electron beam at the accelerator **A** exit is scanned using the electromagnet **SM.** A continuous

beam current monitoring is carried out by the magnetoinductive sensor MIS and linear beam coordination using the beam position monitor **BMP** [4]. A converter assembly **C** consists of a tantalum plate that is placed into an aluminium casing and is cooled by running water. The filter **F** (5 aluminium plates) absorbs the part of the electron beam that passed the converter assembly. A braking photon flux after filter **F** measuring typical dimensions 150×500 mm is controlled by a wide-aperture ionization chamber **IC-W** [5]. The capsules with samples being analyzed are placed just behind the **IC-W**.

In case when the electron energy exceeds 10 MeV an isotropic stream of photoneutrons is emitted from the converter together with braking photons. These neutrons can be used also in the framework of the problem under consideration.

3 RAW ANALYSIS

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

As an example, Fig. 2, 3 show the induced γ-spectrum for two samples of materials of the destroyed ChPP Unit 4: fragments of the reactor concrete shield (Fig. 2) and lava-like fuel-containing mass (LFCM), which was formed in under-reactor premises as a result of the accident (Fig. 3).

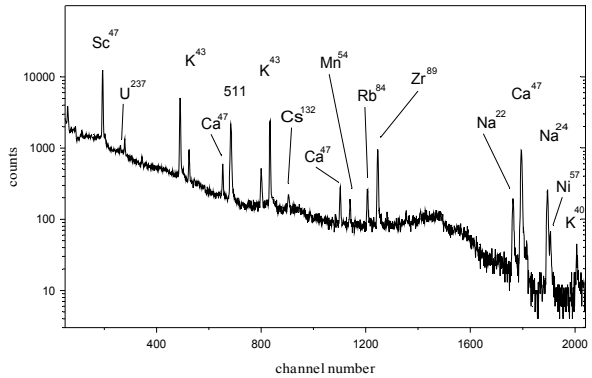

Fig. 2. Induced γ-activity spectrum of concrete sample.

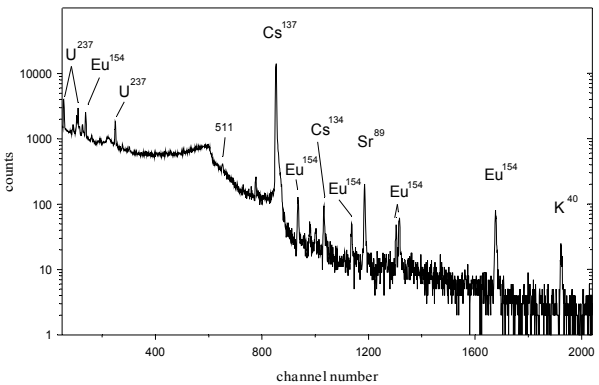

Fig. 3. Spectrum of activated LFCM sample.

The spectrum in Fig. 2 includes the U-237 line. This isotope was generated in the sample under the activation process in the $^{238}U(\gamma,n)^{237}U$ reaction. This example demonstrates the ability of the γ-activation method to analyze the elements the identification of which is impossible by means of a traditional spectrometry methods. The quantitative data obtained concerning the element content in the samples allow to carry out a correlation analysis as well.

Fig. 3 demonstrates also the ability of the γ-activation method in analysis of the samples having their own activity of a different nature. So, apart the lines of γ-radiating nuclides (Eu-154, Cs-137 and Cs-134) there are shown the lines of U-237 and Sr-89. The last result is especially important because the γ-radiating nuclide Sr-89 is created as a consequence of the β-radiating Sr-90 activation. It is known that an analysis of the γ-radiating nuclides is realized technically simpler and for more thick RAW layers (up to 30 cm or so).

The spectrum in Fig. 4 corresponds to the activated U-238 dioxide water solution (with concentration 30 mg U-238/***l***). These data show that the γ-activation method can be used also for the analysis of liquid RAW with identification limit no more then 2 μg U-238 /***l***.

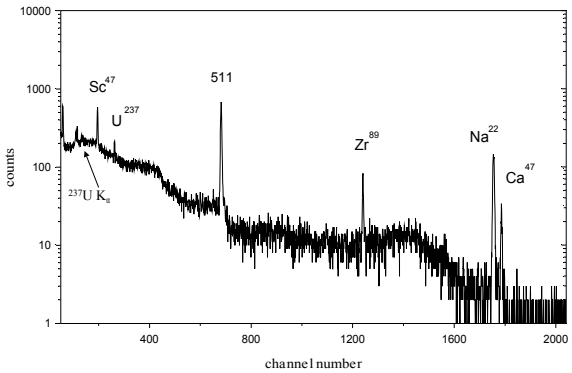

Fig. 4. γ-spectrum of the activated U-238 water solution.

4 INVESTIGATION OF MATERIALS FOR RAW DISPOSAL

It is known that during lasting disposal of the high-level RAW or nuclear spent fuel a situation can arise when the RAW immobilization matrix (including geological structure) will contact with ground water. Thus a structure of "RAW-water-geological barrier" type originates. A radionuclide transport in such a structure determines a reliability of the RAW disposal. Besides, such transport depends on the absorbed dose of radiation from the RAW estimated as much as 10^8 Gy during disposal period.

For research of radionuclide transport processes the granite samples (that is considered as a perspective environment for disposal of long-lived RAW) were selected. 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 a γ-radiating nuclide-tracer which is analogous to actinides in its chemical properties. For this nuclide production under reac-

tion $^{168}Yb(n, \gamma)^{169}Yb$ the pellets of stable $^{168}Yb_2O_3$ were irradiated with photoneutrons. Then the pellet was dissolved in concentrated HCl acid (0.2 ml) and finally the aqueous solution with pH=1.8 was prepared.

The solution obtained (40 ml) together with the sample irradiated up to the given dose value ($3 \cdot 10^6 ... 3 \cdot 10^7$ Gy) were placed into the thermostable flask. The latter was heated with the water steam during 32 hours.

Then each specimen was washed in distillate water during 24 hours and dried out at 60°C in the drying box. Further the layers (2...50 μm) from the 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. Typical spectrum of the material removed of the sample, irradiated with braking photons of LU-20 accelerator up to the dose value 3 10^7 Gy is shown in Fig. 5. Spatial distribution of Yb-169 concentration within depth of the specimen is demonstrated in Fig.6. These results allowed to determine a dose dependence of the radionuclide diffusion as well as to find out its mechanism [7].

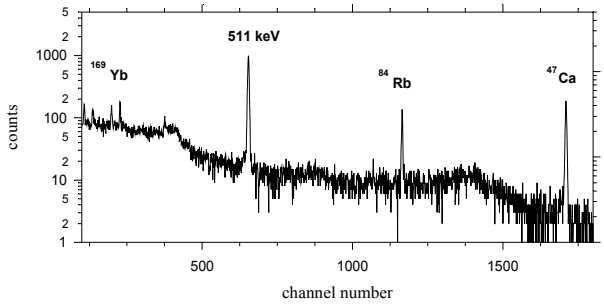

Fig. 5. γ-spectrum of irradiated granite ($3 \cdot 10^7$ Gy).

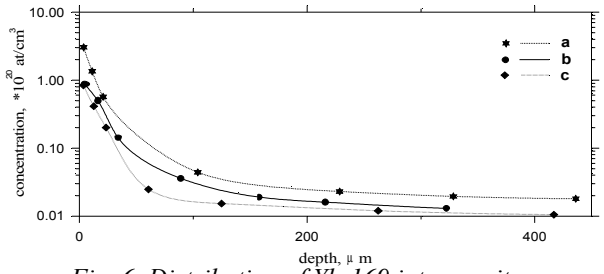

Fig. 6. Distribution of Yb-169 into granite:
***a** – irradiated ($3 \cdot 10^7$ Gy);*
***b** – pristine state (granite with pegmatite structure);*
***c** - pristine state (granite with uniform grain structure).*

5 COMPUTER SIMULATION

A method of the computer simulation based onthe standardized code GEANT (or other like it) can be used for optimization of the sample irradiation conditions, isotope generation modelling as well as for investigation of metrological characteristics of the measuring sensors under their interaction with radiation. Such code allows to calculate these parameters with appropriate accuracy (not less than 10%) considering real composition of the radiation forming systems as well as of the irradiated object [8].

6 CONCLUSIONS

1. High-current electron accelerator with the energy range 10...30 MeV allows to solve effectively different problems of radioactive waste management, in particular, operative analysis of the nuclide and element content of the RAW using the γ-activation method without destruction of samples.

This method provides also an ability of distant-reading analysis under automatic operation. Such facility is important for large amount of samples to be analyzed, for example, when extracting the RAW from Chernobyl power plant unit 4.

2. A powerful (~10 kW) electron accelerator is comparable, by its absorbed dose rate ability, with the Co-60 source having the activity up to 1 MCi. This circumstance as well as a possibility to control the upper limit of the braking photons spectrum allow to use linacs for research of radiation and chemical stability of materials intended for immobilization and disposal of RAW. The radionuclides produced directly at linac can be used as tracers in these investigations.

3. A *l*inac provides the radiation of different intensity and nature (accelerated electrons, bremsstrahlung and photoneutrons) that gives a possibility of radiation test of materials within the wide range of their operation conditions.

Work is supported by STCU under contract N 1580.

REFERENCES

1 Toms M. Elaine. *Photonuclear Activation Analysis with Ge(Li) Detector*. Nav. Res. Lab., USA, Rep. 7554, 1973.

2 T.Gozani et al. Measurement of Prompt and Delayed Neutrons from Photofission // *ANS Trans.* 1968, v. 11, p. 659.

3 A.N.Dovbnya et al. Electron Linacs Based Radiation Facilities of Ukrainian National Science Center "KIPT" // *Bul. of the Amer. Phys. Soc.* 1997, v. 42, N 3, p. 1391.

4 V.L.Uvarov, V.N.Boriskin et al. Calibration of Electron Beam Measuring in Technological Linacs // *Proc. of ICALEPS'99*, Trieste, Italy, 1999.

5 A.A.Butenko, S.P.Karasyov et al. Technological Measuring Channel for Bremsstrahlung Monitoring // *Problems of Atomic Science and Technology. Issue: Nuclear-Physics Research* (35). 1999, v. 4, p. 49.

6 A.Borovoi. Post-Accident Management of Destroyed Fuel from Chernobyl: Technologies Used and Lessons Learned // *IAEA*. 1990, p. 15.

7 N.P.Dikiy, S.Yu.Sayenko, V.L.Uvarov, E.P.-Shevyakova. Application of Nuclear-Physics Methods for Studying the Radionuclide Transport in Granite Rocks // *Problems of Atomic Science and Technology. Issue: Nuclear-Physics Research* (36), 2000, v. 2, p. 54. (in Russian).

8 S.P.Karasyov, S.V.Maryokhin, V.L.Uvarov et al. On Computer Modelling of Primary Transducers in Electron Radiation. Diagnostics // *Proc. of EPAC'98, Stockholm*, June 1998, p. 134.