# ELECTRON ACCELERATORS IN STUDY OF MATERIALS FOR RADIOACTIVE WASTE LONG-TERM DEPOSITION*

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*Abstract*

The increase of radioactive waste (RAW) as the result of development of the atomic energetics and nuclear technologies poses the problem of its reliable deposition. So, the materials and geological structures contacting with a 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. In the report it is shown a possibility of application of the electron accelerator bremsstrahlung in energy range 10...30,MeV to solve a number of tasks for prognostication of the material durability under effect of RAW radiation. Especial target unit has been designed for a simulative irradiation of investigated materials. The unit is provided with the measurement channels for continuous monitoring of dose rate and temperature of the samples. Target geometry was previously optimized by means of computer modelling. The experimental methods of investigation of radiation & chemical durability for the natural and synthetic materials were elaborated as well. These methods are based on the analysis of γ-radiating nuclide-tracers displacement in a system "protective barrier-water" under absorbed dose within a range of interest. A nuclide-tracer production in photonuclear reactions under effect of braking photons both in a separate target and in the volume of investigated sample is demonstrated.

## 1 INTRODUCTION

The main goal of imitation exposure of the materials that are used for the immobilization of radionuclides is a creation of absorbed doze up to $10^8$ Gy in investigated samples during acceptable period of the time (no more than 1 year) at controlable parameters of the radiation effect.

The gamma-ray unit with radionuclide sources (basically, Co-60) 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 provide of the absorbed doze rate (ADR) about 10 Gy/s. Electron accelerator can provide the same conditions with converting its beam to bremsstrahlung (BR). The value of the BR 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.[1]). A possibility of the electron energy and flux control provides an expansion of range of the influence parameters to the sample during its test.

The report deals with the results of development of the electron accelerator based methods and devices for investigation of radiation & chemical durability of the materials which are intended for a radionuclide immobilization.

## 2 COMPUTER SIMULATION

As known, granite is the promising geological environment for radioactive waste disposal [2]. Therefore, the samples from granite (that consists of more than 70% of the $SiO_2$) was chosen as one from objects to study. The preliminary calculations have shown that replacement of the real matrix from granite to pure $SiO_2$ while simulation of radiation effect gives the results varied no more than 3%, but, the calculation time is reduced considerably in this case. Version of the target unit (TU) irradiation geometry analyzed with a method of the computer simulation is presented in Fig.1.

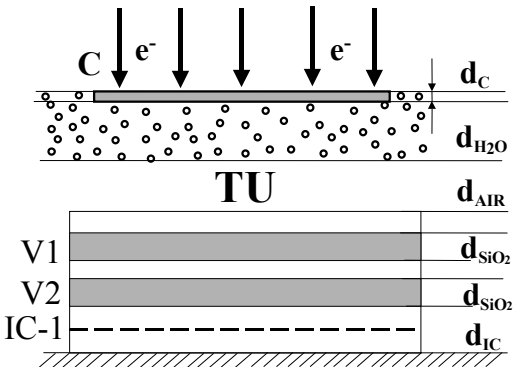


Fig.1: Geometry and parameters of simulation

| $d_C$,mm | $d_{H_2O}$,cm | $d_{air}$,cm | $d_{SiO_2}$,cm | $d_{IC}$,cm |
|---|---|---|---|---|
| 2 | 15 | 5 | 2 | 2 |

Scanned beam is directed to converter of bremsstruhlung **C**, that is a plate of material with high atomic number (tantalum, tungsten) and placed into a tank, cooling by the running water. The thickness of water layer under the plate is chosen so that the part of accelerated electrons passed through the plate is absorbed in this layer. The **TU** is placed under the bottom of the tank symmetrically to the line of beam scaning. The unit consists of the vessels V1 and V2 (when a separate irradiation of the samples in the air and liquid is necessary) as well as of ionization chamber IC-1 for a continuous monitoring of the photon stream passing the

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samples ($SiO_2$). An irregularity of the photon stream density on the surface of TU is defined by distribution of the electron stream density on the surface C, and also by width of TU and typically is less than ±5%.

The calculations were made for three values of accelerated electron energy: 10, 15 and 20 MeV with average beam current 1mA(see Table 1). Analysis of the data obtained shows that the considered version of the TU provides an irradiation of the samples by "pure" braking photon stream in all range of the accelerated electron energy.

Table 1: Results of simulation

| | ELECTRON ENERGY, MeV | | |
|---|---|---|---|
| | **10** | **15** | **20** |
| Electron energy flux, kW | 10 | 15 | 20 |
| Number of absorbed electrons, % in converter<br>in water filter | 99.5<br>- | 92.79<br>6.92 | 49.07<br>50.73 |
| Photon energy flux on TU, kW | 1.07 | 2.76 | 5.08 |
| Average energy of photons, MeV | 0.82 | 1.12 | 1.44 |
| Absorbed power P of photon radiation in $SiO_2$, W | 16.32 | 50.18 | 110.8 |
| Ionization current *I* in IC, μA | 60 | 191 | 419 |
| k=*I*/P, μA/W | 3.64 | 3.80 | 3.78 |

The average photons energy $\bar{E}_\gamma$ along their spectrum occurs near the values for such representative components of the radioactive waste as Cs-137 ($E_\gamma$=0.662 MeV) and Co-60 ($E_\gamma$=1.25MeV), and ADR in $SiO_2$ is ≥10 Gy/s, that satisfies to conditions of the imitation exposure.

Close to linear dependence of ionization current from absorbed power of the braking photons radiation in $SiO_2$ is observed in investigated energy range of accelerated electrons. This result gives a reason to carry out continuous nonintercepting monitoring of the absorbed doze in irradiated samples from ionization charge in IC-1. This range of the electron energy provides also a possibility of the BR absorbed dose accumulation in the samples in the activation mode ($E_\gamma$>10 MeV) for following determination of their isotope composition by gamma-activation method or radionuclide-tracer generation [3], and without activation ($E_\gamma$<10 MeV).

## 3 MEASURING CHANNEL

In the context of the results noted above the measuring channel for the TU (Fig.2.) was designed for on-line monitoring of absorbed doze and ADR of the photon radiation and also for samples temperature measurement during irradiation. The channel provides measurements both in the stand-alone mode and in the remote mode controlling by computer using RS-232 interface.

The channel includes two measuring circuits. The free-air ionization chamber IC-1 is used as primary sensor in the measurement circuit of the braking photon radiation parameters. Thermistor TR-1 is used as a probe of the samples temperature.

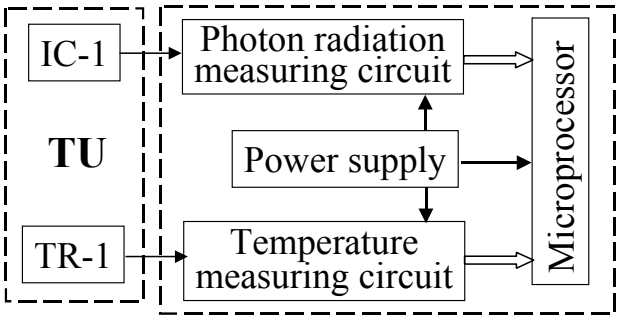


Fig. 2: Block-diagram of measuring channel

The microprocessor module provides:
- conversion of analogue signals from measuring circuits to digit code by the ADC;
- store the results of the measurements and conversion of them to physical units.

## 4 METHOD OF RADIATION & CHEMICAL TEST OF THE MATERIALS

For a preliminary study the granite samples made in the form of parallelepiped with the size 10x10x10.5 mm and mass (2.8-2.9) g were chosen. The samples were irradiated in the bremsstrahlung field of the electron accelerators at two stages: first – at the value of upper boundary of the photon energetic spectrum $E_{\gamma max}$=10 MeV up to the value of the absorbed dose (AD) $1.7 \cdot 10^7$ Gy, after that the samples were activated at $E_{\gamma max}$=23 MeV during 7 days up to the summary value of AD $3.0 \cdot 10^7$ Gy. Then the samples were kept for some days to reduce the level of the 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 the dynamic test on leaching in the plant based on Soxhlet extractor [4] by method described in the regulation [5].

Temperature of the leachant in the cylinder of extractor was 72°C. The leachant was discharged every 1.5, 3.5, 30, 105 and 113 hours. After each discharge of the leachant its activity was measured by Ge(Li) spectrometer with energy resolution 2.8 keV (on the line $E_\gamma$=1333 keV).

## 5 EXPERIMENTAL RESULTS AND DISCUSSION

Gamma-activation analysis of samples has shown that the feature of element composition of the studied granite is the increased content of uranium – 10 μg/g or 3 times exceeding its typical clark value.

The content of Zr and Nb in the sample was also higher than typical values.

On the spectrum (see Fig.3) the lines from photo-nuclear reactions on the matrix and microelements of granite can be definitely see: $^{48}Ca(\gamma,n)^{47}Ca \rightarrow {}^{47}Sc$, $^{23}Na(\gamma,n)^{22}Na$, $^{23}Na(n,\gamma)^{24}Na$, $^{93}Nb(\gamma,n)^{92m}Nb$, $^{85}Rb(\gamma,n)^{84}Rb$, $^{89}Y(\gamma,n)^{88}Y$, $^{133}Cs(\gamma,n)^{132}Cs$, $^{55}Mn(\gamma,n)^{54}Mn$, $^{238}U(\gamma,n)^{237}U$.

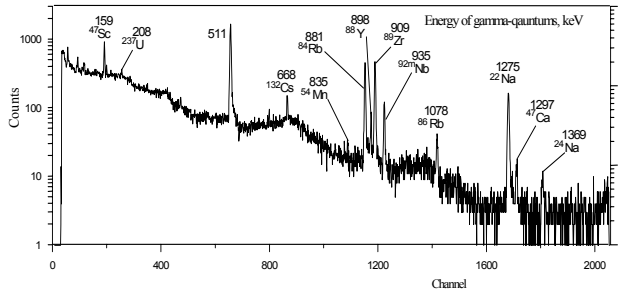


Fig. 3: γ-spectrum of granite sample after activation by bremsstrahlung

Obtained dependencies of diffusion coefficients of different elements on the logarithm of the leaching time (sec.) are given in Fig.4.

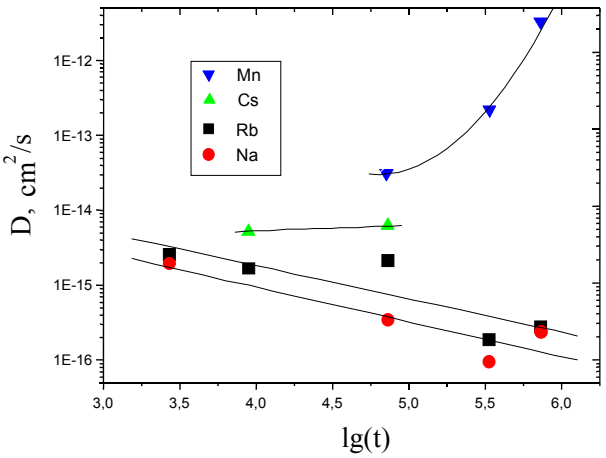


Fig. 4: Coefficients of diffusion of alkaline elements and Mn

Analysis of the leachant spectrums shows that sodium, rubidium and calcium are leached from granite most intensely. There was no noticeable release of uranium and yttrium from these samples.

## 6 SUMMARY

There has been developed the method of investigation of the radiation and chemical stability of non-organic materials that can be used for selection of perspective environments for deep geological disposal of the radioactive waste. The method includes dynamic test on leaching of the samples which underwent the combined irradiation in the bremsstrahlung field of the electron accelerator with the following analysis of gamma-radiation of the leachant and samples.

Sensitivity of the method is by factor of value higher than that of the traditionally used chemical methods of the leachant analysis.

A special target unit has been designed which provides a radiation test of the samples under controllable conditions. The unit is supplied with the PC coupled measuring channel for real-time monitoring of photon radiation parameters and temperature of samples in range

- absorbed doze rate of photon radiation, Gy/s – 1...100;
- absorbed doze, Gy - $10^3$...$10^{10}$;
- average photon energy, MeV - 0.8...1.4;
- samples temperature, T°C - 0...80.

A possibility of radionuclide-tracer production using electron accelerator with following study of their transport in volume of the samples under different absorbed dose values has been demonstrated earlier as well [3].

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