INVESTIGATION OF CHERNOBYL 4-th UNIT MATERIALS BY GAMMA ACTIVATION METHOD

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Isotope and element content the samples of Chornobyl 4-th wrecking unit materials (concrete fragments and lava-like materials) were investigated by γ-activation method using bremsstrahlung of the electron accelerator. The concentration of a number of nuclides (U-238, Cs-137, Sr-90, Ni-58, Zr-90 etc.) and their depth distribution into concrete were determined as well as the corresponding correlation ratio. The comparison of the obtained data with the structure-phase analysis results was carried out.
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1. INTRODUCTION

Detailed study of materials of the Chernobyl 4-th accidental unit, in the first place, of concrete and lava-like fuel containing mass (LFCM) allows restoring a scenario of the wreck development as well as to determine a mechanism of the radionuclide transfer. The latter is of sufficient practical importance because it allows a prognostication of durability of the reactor radiation shield as a confinement for the radioactive waste (RAW) in case of emergency. Investigation of concrete is actual also because it is a main part of RAW of the 4-th unit. LFCM, in the turn, is the main source of radionuclide pollution of the unit constructions during postaccidental period.

2. ELEMENTARY ANALYSIS

2.1. CONCRETE

The samples of concrete were choosed from the constructions situated at different distance from the reactor trunk, i.e. in the conditions that are significantly different in radiation, temperature and other kind of effects.

Two concrete cores (Fig. 1) of about 40 mm thick and 70 mm radius have been obtained by test boring: the first (K1) from internal wall of the unit building (one side coloured with a blue paint) and the second – from biological shield nearly the reactor surface.

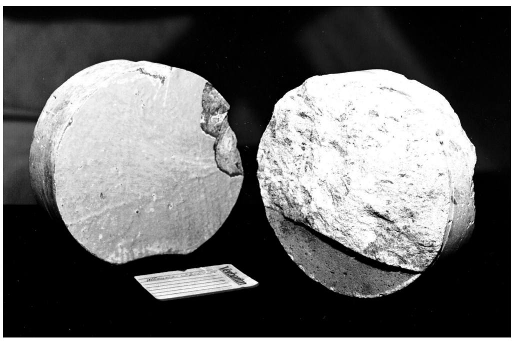

Fig. 1. *Concrete cores (pristine state, K1 from the left)*

The samples of 2 to 15 g weights were separated from the cores using diamond disk and numerated as shown in Fig. 2. During cores treatment it has been find out that the concrete is much crumbling. Thus, there was no possibility to obtain the homogenous samples of right form. For a tentative determination of the core elementary composition the samples were irradiated (activated) with braking photons on LU-20 electron linac [1]. The accelerator has been equipped with specially designed system for formation and monitoring of bremsstrahlung field [2,3], which provides an activation of the samples under the controllable conditions.

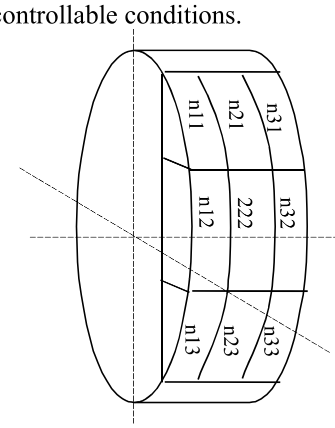

Fig. 2. *Numeration of samples (n=1,2 for first and second core correspondingly)*

The parameters of bremsstrahlung under activation were

- high limit of braking photons energy E_γ, MeV - 20.2;
- absorbed dose rate, Gy/h - $5.8\cdot10^4$.

Induced γ activity of the samples was measured by means of Ge(Li) detector of 50 cm^3 volume having energy resolution of 2.8 keV along 1333 keV line [4]. The spectrums thus obtained (see, for instance, Fig. 3) allow to determine an isotope composition of the samples. In particular, the line E_γ=208 keV observed in the spectrum, corresponds to ^{237}U which has been obtained in $^{238}U(\gamma,n)^{237}U$ reaction.

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The carried out measurements to be have shown that ^{238}U content in the K2 is twice larger than in the K1. Therefore, 11 samples of 2 mm thick were prepared along K2 axis using thin diamond disk for more detailed study of the isotope concentration in this core. The samples have been placed into aluminum capsules and activated for 82 hours. At the same time the activation of the paint sample from the K1 was carried out.

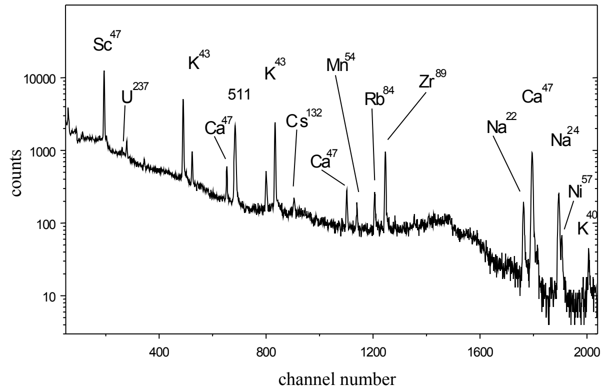

***Fig. 3**. Gamma spectrum of activated concrete sample*

Analysis of spectrometric study of the samples has shown that mean content of ^{238}U is 0.0034 at.p.c., Zr – 0.060 at.p.c. and Mn – 0.46 at.p.c. The correlation with the coefficient of 0.78 between ^{238}U and Zr content is observed also. The overall estimated uncertainty of measurements not exceeds ±15 %. A concentration of U and Zr differs in the various samples several times. Content of Zr in the concrete is more than 10 times larger in comparison with U one. Concentration of Ce is practically invariable (~0.010 at.p.c. with the uncertainty ± 22%).

Gamma spectrum of induced activity of the paint sample is presented in Fig. 4. The lines corresponding to Ti and Zn (main components of the paint) are shown distinctly. So, ^{47}Sc is produced under reaction $^{48}Ti(\gamma,p)^{47}Sc$. Besides, there is the line E_{γ}=909 keV corresponding to $^{89}Sr(T_{1/2}$=50.5 days), which content is up to 0.0075 at.p.c.

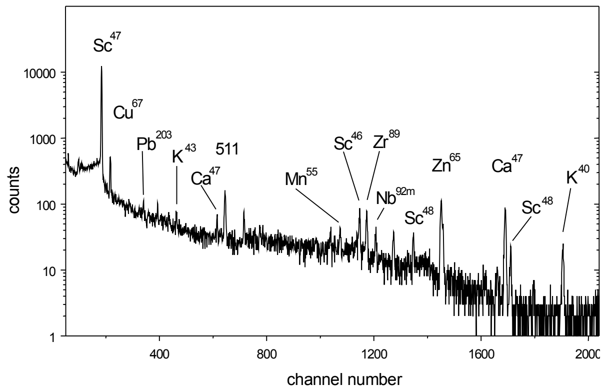

***Fig. 4.** Gamma spectrum of paint sample activated by LU-20*

2.2.LFCM

The investigated LFCM samples were the grains of light brown color of about 1 mg weigh. Their own radiation was within limits allowed by Ukrainian radiation security regulations (NRBU-97).

To determine an isotope composition, the samples of LFCM have been activated under bremsstrahlung field of accelerator together with the concrete ones. Thus, γ radiation of the samples after activation was caused by the sum of induced and proper activity. Their ratio was about 7.

Gamma spectrums of the samples (Fig. 5) are practically identical.

So, together with intense line of $^{137}Cs(E_{\gamma}$=662 keV) there are lines of ^{235}U fission fragments – ^{134}Cs and $^{154,155}Eu$, produced during 4-th unit exploitation. The radiation of ^{237}U as well as ^{89}Zr and ^{99}Mo is most intensive for induced activity. The mean content of U in the studied LFCM samples is about 15 w.p.c.

3. STRUCTURAL-PHASE ANALYSIS

Crystalooptic (petrography) analysis of the samples was carried out using polarizable microscope with the magnification of 350-800.

3.1. CONCRETE

The samples for crystallooptic analysis have been taken from same layers of the cores as for activation one. The fulfilled study has shown that they consist of rather friable concrete mass with a flocculent cement mass. Against its background there are subrounded, rounded and rare isomorphic grains of minerals, which constitute the clastic fraction of concrete filler. The material of the cement is notable for high porosity and widely branched system of the cracks. The rounded and shit pores are of 0.2-0.5 to 1-2 mm diameter and the crack pores of up to 0.5 mm width.

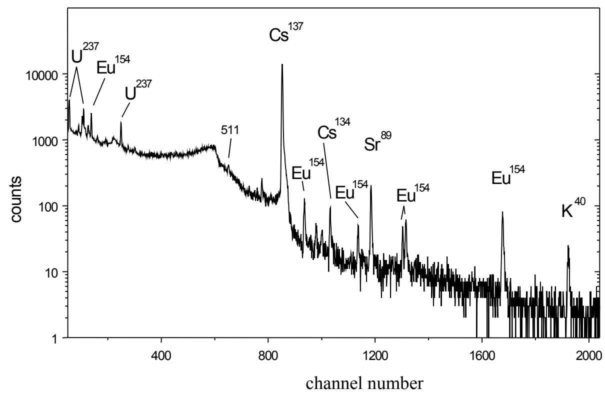

***Fig. 5.** Gamma spectrum of LFCM sample after activation during 135 hours*

As a feature of the K2 samples is a presence of inclusions with right spherical form as well as of dropping aggregates gethered into annular formations (Fig.6). The color of the spheres is black with characteristic vitreous lustre. They are of 0.014-0.030 to 0.40 mm diameter. Some spheres are surrounded with a thin (of 0.02-0.03 mm thick) envelope of semi-transparent glass-like substance having more light color. The behaviour of the spheres under || and ⊥ polarizers testifies to belonging at least a part of them to uranium oxide (pitchblende-II).

3.2.LFCM

A basic mass (BM) of the LFCM fragments consists of glass-like transparent colorless matrix with the isotropic structure and index of refraction N=1.556±

0.003. The BM is varied in the different fragments up to 60 vol.p.c. max value. There is seen a presence of globules against a glass background (N=1.535±0.001) with a concentration up to 20 vol.p.c. that is typical for fusion of heterogeneous badly intermingled mass. There are simultaneously well made out gas or gas-liquid inclusions measuring in range 0.002-0.020 mm with concentration of 3-5 to 20 vol.p.c.

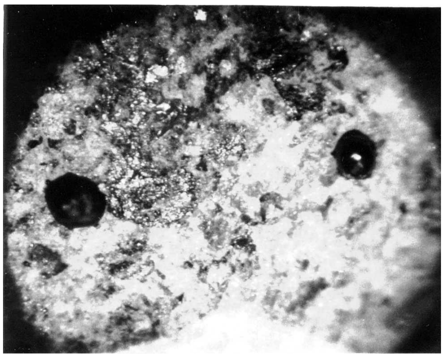

Fig. 6. *Fragment of concrete surface with spherical inclusions (x300)*

The next LFCM phase is the uranium oxide, which is presented with a great number of small grains measuring from 0.015 to 0.030 mm. The uranium oxide content in the different fragments is varied reaching of 30...50 vol.p.c. in some of them.

A feature of the studied samples is the development of secondary mineral new formations. They are represented by two phases:

- carbonaceous partly hydrogenated one of small-grained or clot-dispersive structure with the grains of 0.003 to 0.010 mm;
- hydrooxided one in the form of elongate-fibrous stretched prismatic colorless transparent crystals gathered sometimes in the zonary edging at periphery of the glass fragments.

The size of grains is of 0.06x0.020 to 0.010x0.060 mm, N_m=1.530...1.535(±0.003).

The content of each new formation phase not exceeds of 15...20 vol.p.c. and they are distributed sporadically. There was observed in rare cases (2-3 grains) a presence of rounded slightly zoned fine-dispersive aggregates with isotropic structure and $N>N_{glass}$ but less than 1.870±0.001 which are intensively colored in the light orange and yellow-red tones. These features allow identifying them as hydroxide modifications of the uraniite [5].

4. DISCUSSION

1. As it follows from γ activation analysis, the ^{238}U content in the investigated concrete samples (especially, in the K2 core of biological shield of the reactor) is several times more than typical natural concentration. It could be explained by migration of uranium into concrete during 1986 accident when a large amount of active formations of U have got into environment as a result of high temperature in the reactor zone. A big amount of cracks and pores in the concrete find out by structural-phase analysis favours the transfer of U as well.

2. A structure-phase state of the concrete is determined by the last in its effect postradiation period of existence of the mineral component with a gradual decay of the pristine crystalline structures, a break of chemical bonds in the complex alumosilicates of the cement BM and with deformation of the concrete fragment part. There have been realized the new formations with more simple phases: carbonates, sulfosalts and silicate amorphous materials that are distinctly fixed in the infrared absorption spectrums.

3.The spectrum-mineralogical analysis of the paint samples demonstrates a presence of the intensivesuperimposed mineral formation at the border between the paint and concrete. It is naturally to connect with this region a concentration of ^{90}Sr find out under activation analysis of the paint.

4. The glass-like component of the LFCM samples is characterized by mainly calci-alumosilicate composition with a small admixture of MgO, Na_2O and ZrO_2.

Taking into account these facts, one could suppose that the find out new formatted phases with carbonate and hydrosilicate composition are the products of natural reactions of hydration and carbonization, which proceed under atmospheric conditions (i.e. in the presence of sufficient supplying with water and season changing of the temperature).

The identified inclusions of hydrosilicate modifications of the uraniite (orange aggregates) are analogous in their nature to formations, which have appeared on surface of the LFCM accumulation [6].

5. The carried out study has shown also that γ activation method with use of high-current electron accelerator provides daily more than 100 analyses of radionuclide and element composition of the samples of up to 1 kg weight without their breach. Such operativeness solves the problem of characterization of large mass of the RAW to be evoked from the 4-th unit before deposition of them.

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