ESTIMATIONS OF HIGH-TEMPERATURE DIFFUSION IN CHERNOBYL'S CONCRETE CONSTRUCTION

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The content of isotopes Ti, Ni, Zr, Cs and U in the cores, which was taken from the concrete construction of ChAPS, was studied by the gamma-activation analysis. It has been developed a numerical model to analyze heat and mass transfer in concrete at high temperatures accompanying incident of 1986. Comparison of the measured isotope distributions with the simulation data has enabled to estimate some attendant circumstances of the incident.

PACS: 28.41.Te, 02.60.Cb, 07.05.TP, 68.35.Fx, 44.35.+c

1. INTRODUCTION

The major scientific and technical problem of atomic engineering is reliable isolation of radioactive and toxic materials and substances from possible penetrate into an environment. Therefore there are actual the researches, able to give the helpful information for a material choice and development of building designs of the future atomic power stations or storehouses for radioactive waste disposal. In this connection, there is especial interest to studying the samples taken from elements of an environment of reactor ChAPS, undergone to extreme influence as a result of accident 1986.

2. DESCRIPTION OF SAMPLES AND MEASUREMENTS

The samples were taken in 2000 from a concrete wall of the reactor technological channel, which was filled by the fused magma after a time after explosion of a reactor were investigated. Our interest in the present work is concentrated on studying of traces of magma effect on the channel walls while it has not flowed out through the burnt bottom of the channel after 83 hours [3]. The position of test selection was located on 0.5 m above a level of magma. Researched samples represented section of a core 5 cm long received by consecutive drilling the concrete wall in a direction outside to its axis. The upper crumbled core about 2.5 cm long adjoining to the surface was inaccessible to measurements.

The selected samples were irradiated with bremsstrahlung radiation at the electron accelerator [1,2,4] which had the top energy spectrum border 20.2 MeV; the magnitude of absorbed in samples doze was $2 \cdot 10^6$ Gr. There was used the reactions: $^{49}Ti(\gamma,p)^{48}Sc$, $^{238}U(\gamma,n)^{237}U$, $^{90}Zr(\gamma,n)^{89}Zr$, $^{58}Ni(\gamma,n)^{57}Ni$. The gamma-radiation of the activated isotopes was measured by means of spectrometer with Ge(Li) detector. The cesium content profile was estimated at isotope ^{137}Cs activity.

3. MODEL FEATURES

For simulation of dynamic characteristics of diffusion of researched isotopes the numerical model using a finite difference method [5] and taking into account series of concrete important features, exhibiting under the influence of high temperature has been developed.

Concrete represents the complex porous multiphase system, which at a local level is in thermodynamic balance with body interstice, filled by liquid water and a gas phase. Heat and mass transfer processes in concrete structures at high temperatures are accompanied by a number of the accompanying nonlinear phenomena. Along with heat conductivity, such phenomena also include propagation of vaporized and liquid water, the pressure caused by gradients, capillary effects, just as the latent heat of the phase transitions caused by change water (evaporation, desorption in pores, dehydration) and solid phase. Furthermore with change of porous structure at high temperature permeability of concrete sharply grows.

The model consists of the four basic balance equations. The thermal balance equation for whole medium includes heat effects of the phase change and dehydration process. The balance of weight of dry air includes the diffusion and pressure forced flows. The mass balances of the liquid water and of the vapor have been summarized together to eliminate the source term concerning to phase changes (evaporation or condensation). The diffusion equation was accepted in the form, which is not taking into account possible mutual influence of isotopes, participating in process. The carried out simulations have been executed within the framework of one-dimensional model and in the assumption of medium uniformity. The full model allows simulating by means of the finite difference method the evolution of temperature, moisture content and impurities distribution in view of phase transitions in a concrete matrix, which commonly can be described by means of experiments.

4. DISCUSSION OF RESULTS

The measured dependences of the isotope content on core depth are shown in Fig. 1. The concentration level indicates that these isotopes cannot represent components of concrete in the starting state. The liquid sprays of raging magma likely got on the channel walls and have penetrated inside under the action of high temperature. There is content reduction in the top core parts is appreciably for majority of elements. It can be consequence

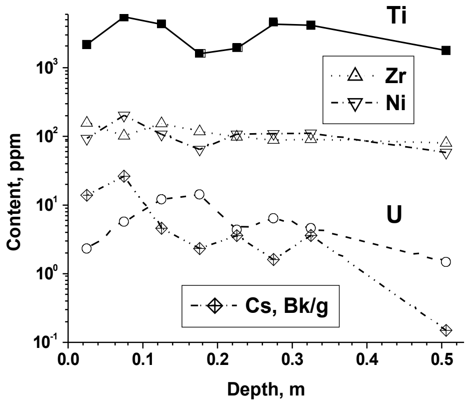

Fig. 1. *Measured isotope content on depth of the core taken from the concrete wall*

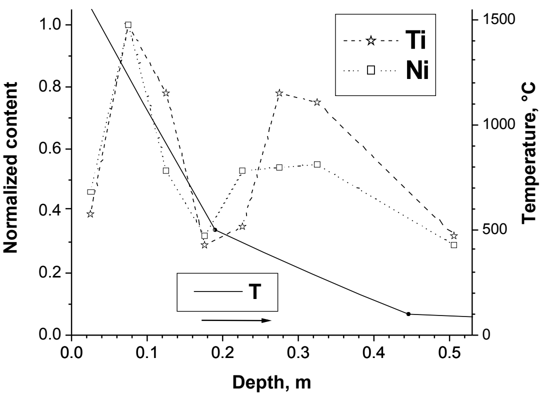

Fig. 2. *Normalized penetration profile Ti and Ni combined with simulated temperature distribution in concrete*

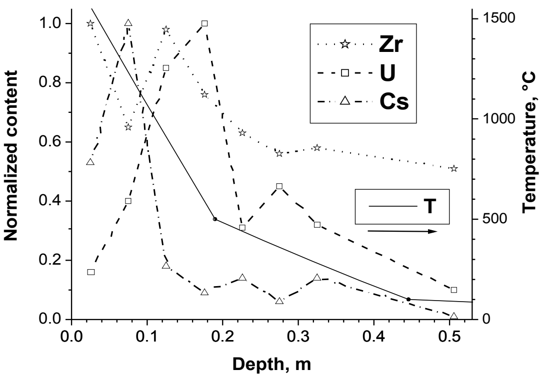

Fig. 3. *Normalized penetration profile Zr, U and Cs combined with simulated temperature distribution in concrete*

of element leaching from surficial region as a result of probable penetration of moisture into the channel till sampling time.

For thermo diffusion process simulation in the presented work the model in iterative process has been used to determine parameters, describing in the best way experimentally measured element contents. At that, taking into account that accessible physical characteristics of concrete in such extreme conditions carry a significant share of uncertainty, it was justified some simplifications in model [6].

The magma temperature magnitude in the channel remained a constant and held out, by different estimations [3], from 1700°C up to 2400°C, owing to its big weight. Therefore we accepted temperature of a concrete surface not smaller, than 1700°C.

At calculation of temperature evolution in concrete, it in the general way is necessary to take into account a number of the complex phase transformations occurred in concrete. For example, hydration (near 95°C) and dehydration (at 120°C) processes removal chemically bound water from concrete crystalline hydrates at temperatures 300…400°C, rising compression and expansion deformations in cement paste and aggregate at 500…800°C, recrystallization of calcium hydro aluminates to corresponding aluminates are higher than 800°C and the beginning of concrete physical destruction, in general, demand power expenses [7]. In our calculations we have combined these processes as two main phase transitions: connected to water evaporation and to phase changes of concrete solid components.

Fig. 2 shows the diagram of the simulated concrete temperature profile, imposed on graph of the normalized contents Ti and Ni, which, obviously, represent captured by liquid melt components of constructional materials. On the temperature diagram the breaks corresponding to the main phase transitions are visible.

From Fig. 2 one can see that penetration profiles for Ti and Ni reveal the similar behavior and break up on a background of temperature distribution to two obvious parts: the determined by high-temperature diffusion area and a zone, apparently, connected with active participation of diffusing metal ions in concrete crystalline hydrate reorganization.

Fig. 3 shows the similar diagram combinations for isotopes Zr, U and Cs, representing components actually reactor materials. One can see, that Cs penetration has quite habitual character when it is isomorphic replaced in crystal components of solid medium. Apparent absence of connection of its content with areas of phase changes can account for its gaseous state (boiling temperature 951.6 K) and consequently could not be appreciably involved to interactions with participation of a liquid phase. It is worthy of notice negative correlation of Cs and Zr contents in high temperature region, connected, probably, with their competitive participation in diffusion processes.

The profile characters of penetration into concrete matrix for Zr and, especially, for U shows their raised migration in the high temperature region, which probably connected with calcium hydroaluminate recrystallization zone in cement paste. One also can see some signs of increase of U ion mobility in the average temperatures zone.

In attempts of more adequate simulation of experimentally measured penetration profiles of isotopes of interest, various kinds of boundary conditions have been tested. However, we did not manage to reveal substantial improvement of fitting quality in the assumption of variable concentration or presence mass exchange at surface, owing probably to the limited experimental data volume. Therefore we have confined within constant element concentration at the concrete surface.

Effective diffusion constants in the high temperature zone and surface contents evaluated within the framework of iterative fitting of accepted model to received experimental data are resulted in the table.

Diffusion constants and surface contents estimates evaluated at data in the high temperature region

Isotope	*Diffusion constant, 10^{-8} m^2/s*	*Surface content, %*
^{49}Ti	*1,8*	*0.9*
^{58}Ni	*1,1*	*0.04*
^{90}Zr	*3*	*0.02%*
^{137}Cs	0,6	110 Bk/g
^{238}U	*8*	*0.018*

Though, owing to high fitting errors, one can only speak about estimations of the received values, it does not cause doubts a high factor of diffusion for U and a big Ti content at the concrete surface.

5. CONCLUSIONS

The technique for thermo diffusion parameter definition in a concrete matrix is offered by comparison of simulated dynamic diffusion process characteristics with results of experiment.

The received estimations should be considered, as the initial stage in necessary studying consequences of accident. Nevertheless, it is necessary to note the catastrophic U leaching, in comparison with other isotopes, in region, where permeability of concrete is sharply increased owing to destruction of its structure at high temperature influence. In view of U properties are similar to other actinium series elements, its behavior features impose special requirements of characteristics of the concrete used in construction of nuclear objects.

The problem in computing sense is complex and demands the further regular calculations on the basis of accumulation of an experimental material and expansion of databases with use of powerful algorithms and computer equipment.

The work is executed within the framework of Program ЯМРТ-2010, Project X866.

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ИССЛЕДОВАНИЕ ВЫСОКОТЕМПЕРАТУРНОЙ ДИФФУЗИИ В ОБРАЗЦАХ БЕТОННОЙ КОНСТРУКЦИИ ЧАЭС

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Методом гамма-активационного анализа измерялось содержание изотопов Ti, Ni, Zr, Cs и U в кернах, отобранных из бетонной конструкции ЧАЭС. Разработана численная модель для анализа тепло- и массопереноса в бетоне при высоких температурах, сопровождавших катастрофу 1986 года. Сравнение измеренных изотопных распределений с результатами модельных расчетов дало возможность сделать оценки некоторых характеристик происшедшего инцидента.

ДОСЛІДЖЕННЯ ВИСОКОТЕМПЕРАТУРНОЇ ДИФУЗІЇ У ЗРАЗКАХ БЕТОННОЇ КОНСТРУКЦІЇ ЧАЕС

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Методом гама-активаційного аналізу вимірювався зміст ізотопів Ti, Ni, Zr, Cs та U у кернах, відібраних з бетонної конструкції ЧАЕС. Розроблено числову модель для аналізу переносу тепла та маси у бетоні при високих температурах, що супроводжували катастрофу 1986 року. Порівняння виміряних ізотопних

розподілів з результатами модельних розрахунків дало можливість зробити оцінки деяких характеристик інциденту, що стався.