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# NUCLIDE CONTROL OF STRUCTURAL MATERIALS TESTED IN ELECTRON IRRADIATION TEST FACILITY

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Samples of different alloys (Hastelloy type) held in the container assembly (CA) were irradiated at the Electron Irradiation Test Facility (EITF) in the melt of zirconium and sodium fluorides at the temperature 650°C. The CA consisted of 16 individual containers made of a carbon-carbon (C-C) composite material (placed under covering shell made of stainless steel). The irradiation was done with electron beam at the energy in the spectrum maximum = 9.6 MeV at Linac-10 high-current accelerator that belongs to "Accelerator R&D Complex" affiliated with NSC KIPT. In order to secure the radiation-safe levels of the activity in further work with the samples, the focus of attention was paid to the analysis of γ-spectra in the remnant activities of the materials under study: Hastelloy type samples and fluoride salts after the irradiation. The dynamics was studied of decreasing of the activity levels of EITF components, sample assemblies in the containers being full with fluoride salts and individual samples. Recommendations are issued and measures taken to provide for further radiation-safe work with the irradiated materials.

## INRTODUCTION

The radiation resistance of the materials of molten salt atomic reactors (MSR) is a very acute problem in atomic power production. One of the methods of enhancing the radiation safety of MSRs is simulation with the aid of linear accelerator (linac) of the conditions, in which the reactor structural materials are operable. The study on corrosion resistance of the structural reactor materials in the conditions that are close to those in the reactor blanket calls for a prolonged irradiation of the samples, simulating concomitantly the influence of the aggressive ambience. With these aims in mind, the EITF was built to carry out prolonged radiation tests on samples of different alloys (Hastelloy type) in the zirconium and sodium fluoride salt melts at the temperature 650°C [1]. Irradiation of the CA samples placed in 16 individual containers that were made of C-C composite material (under covering protective shell made of stainless steel) was done with 9.6 MeV electron beam at Linac-10 high-current electron linac at Accelerator R&D Complex affiliated with NSC KIPT.

## RESEARCH RESULTS

Further studies on Hastelloy samples include microscopy, X-ray structural analysis, rupture strength tests and other related work. All of those operations demand extraction of the plates from the containers, their cleaning from solidified salt, cutting the plates to obtain samples of the needed size. The above operations must be done under strict surveillance of the dosimetric control service, as works with the open radiation sources. To provide for radiation-safe levels of the remnant activity, while working further with the samples, the focus of attention was paid to the analysis of the characteristics of the remnant radioactivity of the research materials: Hastelloy type samples and fluoride salts after the irradiation. The measurement was taken of the remnant activity level of the components of the EITF, sample assembly in the containers that were filled up with fluoride salts and individual samples. With the view of continuing work with the carbon containers, full with Hastelloy samples and fluoride salts, the main attention was drawn to the remnant radioactivity level of the CAs. Table gives the levels of the remnant radioactivity (in terms of micro Svt/h) in each container.

**Activity of containers**

| Nomber container | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 |
|---|---|---|---|---|---|---|---|---|
| Activity | 10,5 | 2,8 | 27,4 | 33,1 | 26,4 | 25,8 | 13,7 | 14 |
| Nomber | 9 | 10 | 11 | 12 | 13 | 14 | 15 | 16 |
| Activity | 2,7 | 13,6 | 33,3 | 30,2 | 37 | 32,8 | 13,6 | 16 |

A major difference from others of the remnant activities of the containers 1, 2 and 9 has to do with evaporation of their contents in the course of the irradiation, which is accounted for by peculiar features of the chemical composition of the salts exactly in those containers.

To identify the irradiation-produced nuclides and to predict the container "cooling-off" times as well as to determine the safe amounts of salts and Hastelloy samples, the measurement was taken of the spectra of γ-radiation of the full containers and individual samplings of the salts as well as of the de-salted Hastelloy plates.

By way of the γ-spetcrometric studies we found the spectral content of the gamma-radiation and half-lives of the respective daughter nuclides produced as a results of the photonuclear reactions (γ,n) and (γ,p) It was demonstrated that the main contribution to the remnant activity of Hastelloy is made by the isotope **$^{57}$Co** (with the half-life of 270 days), which is yielded as a result of the reaction $^{58}$Ni(γ,p)$^{57}$Co, while the remnant activity of the zirconium and sodium salts is accounted for mainly by the nuclide $^{95}$Zr with the half-life of 64 days, which is yielded from the reaction $^{96}$Zr(γ,n)$^{95}$Zr. As an exam-

ple, Fig. 1,a gives the γ-spectrum of fluoride salts from the container 1, as measured after 12 days upon completion of the irradiation, Fig. 1,b showing the spectrum of Hastelloy plate with the coatings of Nb and Nb-O.

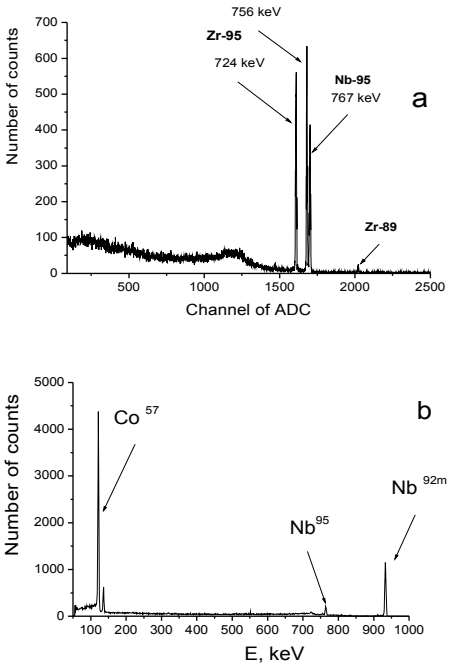


*Fig. 1. γ-spectrum of fluoride salts (a) and spectra of Hastelloy plate with coatings of Nb and Nb-O (b)*

While doing the study on the high-energy portion ($E_\gamma$>900 keV) of γ-radiation from fluoride salts taken from the containers that held alloy samples of the Hastelloy type with the coatings Nb and Nb-O, our finding was such that there was a great amount of the nuclide $^{92m}$Nb, resulting from the reaction $^{93}$Nb(γ,n)$^{92m}$Nb, which was indicative about stimulated dissolution of those coatings in the fluoride salt melts in the course of the irradiation. Fig. 2 (a,b,c) shows variation vs. time of the spectra produced from not-disassembled container #8 after 6, 160 and 240 days, respectively, upon completion of the irradiation.

From the above data it follows that the principal radiating nuclides in the mixture of the salts $ZrF_4$ and NaF are $^{95}$Zr, $^{95}$Nb and $^{89}$Zr.

The longest-lived nuclide is $\beta^-$ active $^{95}$Zr with the half-life 63.9 days yielded from the reaction (γ,n) from $^{96}$Zr; there is an additional channel for appearance of $^{95}$Zr due to the capture of the neutron by $^{94}$Zr, from the reaction (n,γ). During decay of $^{95}$Zr there appears $\beta^-$ active $^{95}$Nb with the half-life 35 days. Fig. 2 clearly demonstrates the process of the "pumping" of γ-lines from $^{95}$Zr (724.2 keV and 756.7 keV) into the line **$^{95}$Nb** with the energy 765.8 keV during the decay of the nuclide $^{95}$Zr following the scheme: $^{95}Zr \xrightarrow{\beta^-} {}^{95}Nb$. The above process causes the "stretching" vs. time of the fall-off of the activity of the irradiated containers on account of the presence of the nuclide $^{95}$Zr. The nuclide $^{89}$Zr has $T_{½}$ =78 h, being radiation-safe to handle. The examination of the spectra of irradiated Hastelloy plates indicated that the main nuclide, determining the further dose load on the service personnel, was the long-lived **Co$^{57}$**. This nuclide was yielded from Ni, as basic ingredient of Hastelloy. The threshold of the reaction (γ,p) was 8.2 MeV.

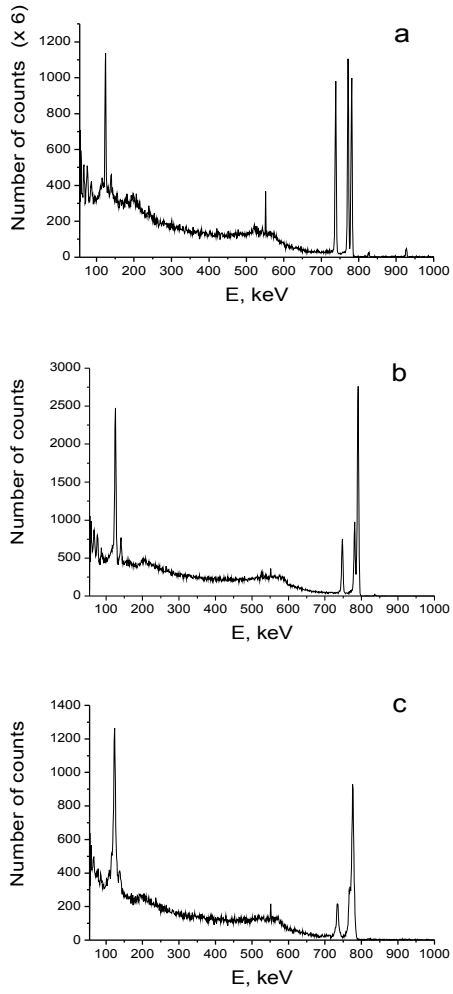


*Fig. 2. Variation vs. time of γ-spectra radiating from the container #8 6 days (a), 160 days (b) and 240 days (c), respectively, after completion of the irradiation*

Other nuclides, such as $^{99}$Mo, $^{99m}$ Tc and $^{92м}$Nb, appearing in the irradiated samples presented no radiation hazard. As the container exposure time ran out, which was determined by lowering of the activity of the nuclide $^{95}$Zr, the above nuclides had decayed, since the longest-lived one from them, $^{92м}$Nb has the half-life 10.8 days.

## CONCLUSIONS

By analyzing the obtained information, one can arrive at the following conclusions and propose a method of work with the samples in keeping with all the norms of the radiation safety, while handling the samples of Hastelloy and fluoride salts after their irradiation:

1. The major sources of γ-radiation are concentrated in Hastelloy plates and solidified salt crusts around

them. Once their content is removed, the carbon containers are safe to handle.

2. Bearing in mind that any further studies on Hastelloy samples are feasible only after their extraction out of the salts, the exposure is needed for whole containers to "cool off" in the course of 1 to 2 half-lives of $^{95}$Zr.
3. Since the studies on the samples will be done requiring immediate contact of the researcher with the material (polishing, etching, TEM microscopy etc.), the cutting of large plates (24x28 mm) into smaller ones should be done together with very careful cleaning of the samples from the residuals of zirconium salts.
4. Handling of smaller-size samples (~3x28 mm) provides the only possibility to bring down the exposure doses to the minimum level, since the long half life of $^{57}$Co (270 days) precludes from lowering of the remnant activity by way of the depositary.
5. At the future irradiation of the materials at the linac, the incident electron energy will come down by 1.5…2 MeV, which shall lower considerably the amount of γ-quanta with the energy above the reaction threshold for formation of $^{95}$Zr and $^{57}$Co (threshold 7.8 and 8.2 MeV). The implementation of the methods of decreasing the partial contribution of the channel (n,γ) to the reaction of formation of $^{95}$Zr will call for additional studies.
6. While handling the irradiated samples, the organizational efforts were reduced to the following: the procedures were refined of sample extraction out of the containers, their desalting, cutting up into fragments of the assigned size and packaging; a special depositary was made to store the radioactive Hastelloy samples and fluoride salts in, providing for strict control over safe-keeping and operative retrieval of the sample needed; the order was worked out to exercise control over the entire mass of radioactive samples and their delivery to destination of further research.

On the basis of research done to date, a controlled process has been refined of irradiated sample preparation for further comprehensive materials studies.

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## РАДИОНУКЛИДНЫЙ КОНТРОЛЬ ПРИ РАДИАЦИОННЫХ ИСПЫТАНИЯХ КОНСТРУКЦИОННЫХ МАТЕРИАЛОВ В УСЛОВИЯХ СОЛЕВОГО РЕАКТОРА

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На стенде для радиационных испытаний проведено облучение сборки образцов различных сплавов (типа хастеллой) в расплаве фторидов циркония и натрия при температуре 650'С, помещенных в 16 отдельных контейнеров из углерод-углеродного композита (в общем защитном кожухе из нержавеющей стали). Облучение проведено пучком электронов с энергией в максимуме спектра = 9,6 МэВ на сильноточном линейном ускорителе электронов ЛУ-10 НИК "Ускоритель" ННЦ ХФТИ. С целью обеспечения безопасных уровней активности при дальнейшей работе с образцами особое внимание уделено анализу характеристик остаточной активности исследуемых материалов – образцов сплава типа хастеллой и фторидных солей после облучения. Измерены уровни активности элементов стенда, сборки образцов в контейнерах, заполненных фторидными солями, и отдельных образцов. Даны рекомендации и приняты меры, обеспечивающие безопасность работы с облученными материалами.

## РАДІОНУКЛІДНИЙ КОНТРОЛЬ ПРИ РАДІАЦІЙНИХ ВИПРОБУВАННЯХ КОНСТРУКЦІЙНИХ МАТЕРІАЛІВ В УМОВАХ СОЛЬОВОГО РЕАКТОРА

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На стенді для радіаційних випробувань проведено опромінення електронами збірки зразків різних сплавів (типу хастелой) в розплаві фторидів цирконію та натрию при температурі 650'С. Збірку складено з 16 окремих контейнерів з вуглець-вуглецевого композиту (в загальній захистній оболонці з нержавіючої сталі). Опромінення проведено пучком електронів з енергією в максимумі спектра близько 9,6 МеВ на сильнострумовому лінійному прискорювачі електронів ЛП-10 НДК «Прискорювач» ННЦ ХФТІ. Для вибору безпечних рівней активності при подальших роботах з зразками особиву увагу приділено аналізу γ-спектрів залишкової активності досліджуваних материалів – зразків сплаву типу хастелой та фторидних солей після опромінення. Досліджено динаміку спаду рівней активності елементів стенду, збірки зразків в контейнерах, які заповнені фторидними солями, та окремих зразків. Дано рекомендації та вжито заходи, що гарантують безпеку при подальших дослідженнях опромінених матеріалів.