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THE PROCESS OF FRAGMENTATION OF MODEL CORIUM DURING INTERACTION WITH THE COOLANT

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This paper presents the results of a study of the granulometric parameters, elemental and phase compositions of a solidified model melt of a nuclear reactor core (corium) obtained in out-of-pile conditions. The out-of-pile experiments are conducted in the EAGLE test-bench of the IAE NNC RK, which allows obtaining a melt of the burden from the components of the core and implementing its interaction with the coolant. To obtain the model corium of a fast neutron reactor, aluminum oxide (Al_2O_3) was used, and sodium was used as a coolant.
The results of the studies revealed the presence of three modifications of aluminum oxide Al_2O_3 in the phase composition of the solidified simulator. The main one is the α-modification (corundum), which is also the basic component of the initial burden. The appearance and regular distribution of other crystalline modifications of aluminum oxide is definitely a sign associated with the features of the processes occurring during the interaction between model corium and the coolant. Such features may be characteristic and extend to the actual melt of the reactor core, but may also be specific to the simulator used.

Keywords: *model corium, sodium coolant, fragmentation, melt, experimental modeling.*

INTRODUCTION

Ensuring the safe operation of nuclear reactors is one of the most important tasks of the nuclear power industry. A large amount of work, including both theoretical and experimental studies, is carried out to address issues related to the safety of power reactors [1–9]. The most informative data on the possible consequences of severe accidents in power reactors of various types associated with the cessation of core cooling or an unauthorized increase in reactivity followed by melting of the core can be obtained by conducting experimental studies. Such studies make it possible to refine existing or develop new calculation methods designed to determine the parameters of severe accidents when substantiating designs for nuclear power plant reactor installations and analyzing the safety of existing installations [10–14].

In relation to fast neutron reactors with liquid metal coolant, studies are being conducted on the final stage of a severe accident, aimed at eliminating the possibility of the formation of a repeated critical configuration of the fuel during its melting in the core (re-criticality) and the efficiency of its cooling. The studies are focused on studying the processes of interaction of the fuel melt with the coolant and structural materials [15].

One of the phenomena accompanying the interaction of the melt with the coolant is the fragmentation of the melt flow with the formation of solidifying particles or fragments. The study of this process is important, and its parameters should be taken into account in approaches to localizing and minimizing the consequences of a severe accident with the reactor core melting down.

This paper presents the results of the implementation of the process of simulating a severe accident in a fast neutron reactor; shows the results of the interaction of a model reactor core melt with a sodium coolant; describes a methodology for materials science research that allows identifying systematic features of corium fragmentation during its cooling.

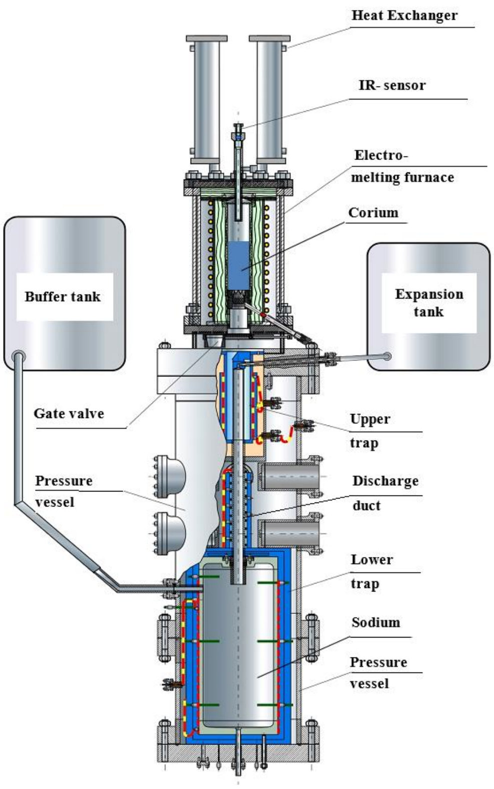
METHODOLOGY OF EXPERIMENTAL RESEARCH

Research on the interaction in corium-coolant systems was conducted in the EAGLE test-bench, Figure 1a presents it's the basic diagram, using the experimental device shown in Figure 1b.

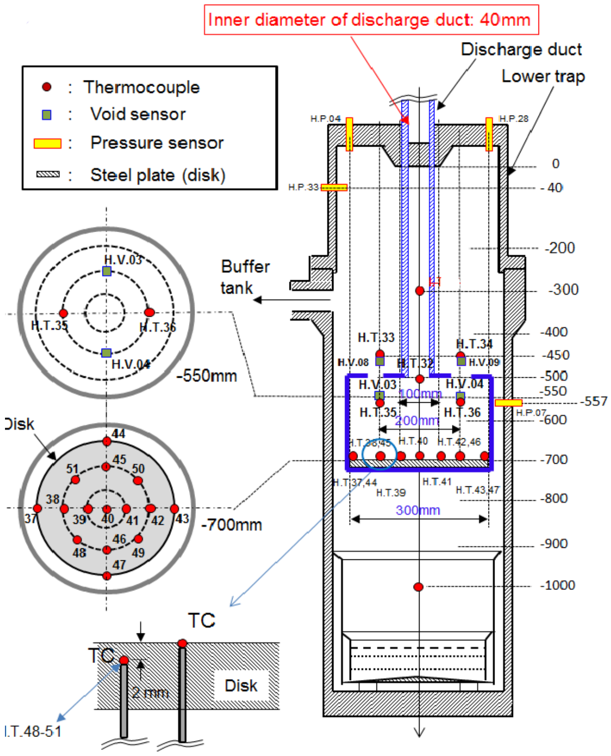
The EAGLE test-bench is designed to conduct out-of-pile experiments to simulate and study processes that occur during the interaction of a fast nuclear power reactor melt simulator with its structural elements and coolant.

The EAGLE test-bench includes an electric induction melting furnace (EMF) designed to produce corium melt and an experimental device that houses the test discharge pipe and melt trap (lower trap). The EMF can produce up to 26 kg of melt containing uranium dioxide and stainless steel or up to 15 kg of aluminum oxide melt. During the experiments, liquid sodium is located in the discharge pipe and/or lower trap.

The simulation of the process of overheating and melting of the core of a fast neutron reactor consists of the following: sintered aluminum oxide (fuel simulator) weighing about 15 kg is loaded into the crucible, the charge is heated to a melting temperature of ~2100 °C, after which the plug of the electric melting furnace (EMF) is chipped off;the melt is discharged into the upper melt trap, simulating the internal cavity of the reactor core fuel assembly (FA); it melts through the thin-walled section of the drain pipe (with a steel wall thickness of 1–3 mm) and moves along the thick-walled section of the discharge pipe (with an aluminum wall thickness of 3–5 mm) into the lower melt trap, simulating the lower plenum of the reactor, where it interacts with the sodium coolant. The sodium in the volume of the lower melt trap before the melt is discharged is in a liquid state at a temperature of about 400 °C and a pressure of 0.35 MPa.



a) EAGLE test-bench diagram



b) Diagram of the experimental device placed in the lower melt trap of the EAGLE stand

Figure 1. EAGLE Test-Bench

An experimental device, which includes an experimental container and a basket, is installed on special rods in the internal cavity of the lower melt trap. At the end of the experiment, liquid sodium is drained through the drainage system into the sodium tank. The sodium residues, together with fragments of the solidified melt, remain in the materials and on the units of the installation, located in the lower trap case.

As a result of conducting such an experiment, at various levels of the bench, the material of the solidified corium simulator, the products of its interaction with the coolant and structural materials remains.

After the experiment, the material of the solidified corium simulator in the area of the lower melt trap is still impregnated with metallic sodium. The material is cleaned of sodium by treatment with a superheated steam-gas mixture (nitrogen, water vapor). For the final removal of water-soluble sodium compounds after steam treatment, the material is washed in water.

Samples of the obtained materials are subject to research, including granulometric analysis, determination of elemental and phase composition.

Research Results

As a result of treating the products of interaction between the model corium and sodium coolant with a superheated steam-gas mixture, the sodium oxidized and formed a fragmented dry conglomerate with corium, which had a grayish-white color of various shades.

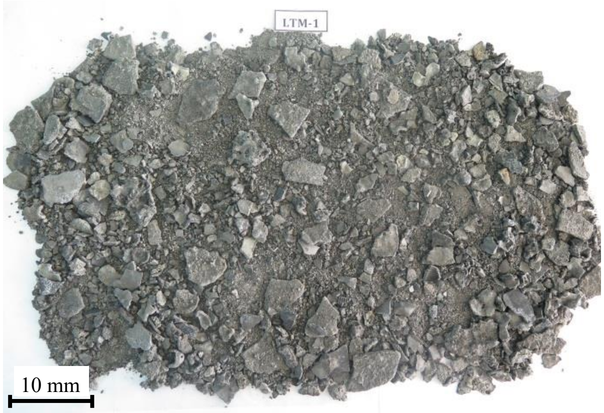
When first poured with water, the material reacted vigorously with water, releasing gas and heat, which indicated the presence of traces of metallic sodium or its oxides. Thus, primary concentrated solutions were formed, which received the corresponding designations NC-1 – from the experimental container and NC-2 – from the bottom trap basket. The density of the solutions after the first pouring was 1.18 g/cm^3 and 1.26 g/cm^3 respectively.

The material was treated with water until it was completely washed free of sodium. After each stage of treatment, the corium simulator was weighed, Table 1 provides the results of mass determination.

Table 1. Results of determining the masses of fragmented corium simulator from the lower trap at different stages of preparation

Corium designation	Corium mass after dry steam treatment, g	Corium mass after washing with water, g	Corium mass after drying, g
NC-1 (from the experimental container)	5650	3440	3060
NC-2 (from the lower trap basket)	10680	2379	2074

Figure 2 shows the view of the corium simulator extracted from the lower trap of the EAGLE test-bench after entire cleaning procedures.



From the experimental capacity – NC-1

From the lower trap basket – NC-2

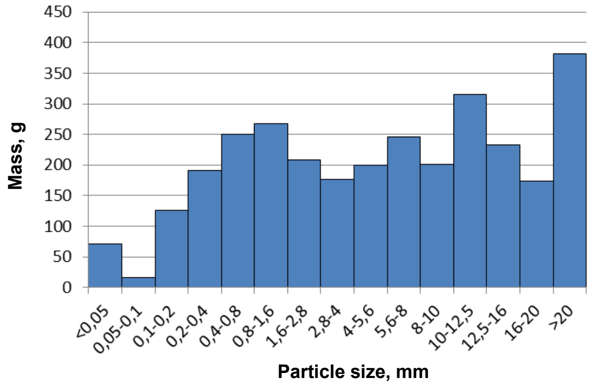
Figure 2. View of the corium simulator after entire washing procedures

Granulometric analysis of the corium simulator consisted of separation of particles by size fractions by sieving on a set of sieves (fractionation), weighing the resulting particle fractions and studying the resulting distribution of particle mass by fractions. The diagrams (Figure 3) show the fractionation results.

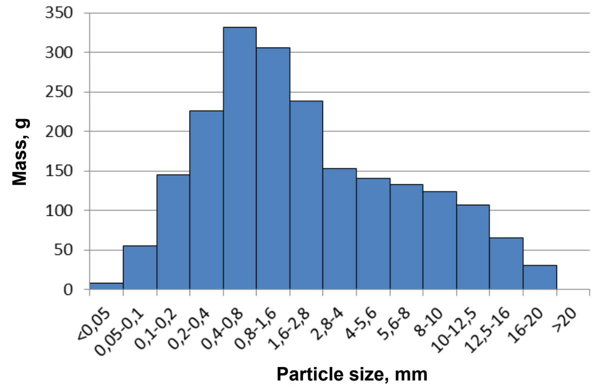
The diagram showing the size distribution of fragmented corium from the lower trap (Figure 3b) shows a clear peak, the apex of which corresponds to the size of the fraction "0.4 mm÷0.8 mm". No such peak is observed in the size distribution of fragmented corium from the experimental container (Figure 3a). The largest share is made up of particles with a size of > 20 mm. The smallest mass from the experimental container has the fraction "0.05 mm÷0.1 mm", and from the basket – "< 0.05 mm". The average weighted particle size in the corium from the experimental container is 8.9 mm, and from the lower trap – 3.4 mm.

The particle shape characteristics were also investigated. Photographs of particles of some fractions are presented in Figures 4 and 5.

The characteristics of the shape and appearance of the particles from the experimental container and the basket of the lower trap turned out to be similar to each other. In the fractions with a particle size of more than "> 5.6 mm", the main share is made up of plate-shaped particles. As a rule, the opposite surfaces of the particles have different colors: one of the surfaces is darker, the other is lighter. Basically, one of the surfaces of the plate-shaped particles is smooth, the other is relief. In the fractions of > 1.6 mm, there are particles with both a smooth surface and a developed one. In the fractions of "200 μm÷1.6 mm" the main share of particles has a developed surface. In the fraction "50 μm÷100 μm" a significant proportion of particles have an elongated shape. In the fraction < 50 μm there are rounded particles, some particles are translucent. No obvious presence of metal particles was detected in the fractions.



a) From the experimental container



b) From the basket

Figure 3. Size distribution of fragmented simulator of corium from the lower trap

For the analysis, a representative portion of corium was taken from each fraction and ground into powder to a particle size of < 50 μm. A thin, even layer of powder was fixed to the microscope sample holder using carbon tape. The area of the analyzed region was ~ 16 mm^2.

The analysis of the atomic content of the elements was carried out on a Supermini 200 X-ray fluorescence wave dispersive spectrometer with a range of determined elements from Al to U. The error in the analysis does not exceed 10% relative to the determined values. The results of quantitative elemental analysis of the samples are presented in Tables 2 and 3.

a) fraction “8 mm – 10 mm” b) fraction “4 mm – 5.6 mm” c) fraction “1.6 mm – 2.8 mm”

Figure 4. View of corium particles from the experimental container

a) fraction “8mm – 10 mm” b) fraction “4 mm – 5.6 mm" c) fraction “1.6 mm – 2.8 mm”

Figure 5. External view of the corium from the lower trap basket

Table 2. Results of elemental analysis of fractionated corium simulator samples from the experimental container

Fraction	Content of elements, wt.%									
	O	Na	Mg	Al	P	K	Ca	Ti	Cr	Fe
5.6–8 mm	41.52	1.64	—	55.28	0.46	—	0.29	0.11	0.14	0.57
2.8–4 mm	31.88	1.32	—	65.11	—	—	0.44	0.22	0.25	0.78
0.8–1.6 mm	47.00	0.88	0.71	50.64	0.13	—	0.18	0.05	0.06	0.34
200–400 μm	47.71	0.66	—	50.70	—	0.11	0.15	0.12	0.17	0.39
50–100 μm	50.31	0.86	0.18	47.79	0.08	0.10	0.23	0.06	0.10	0.29
less than 50 μm	39.46	0.49	—	56.57	0.32	—	1.34	0.12	0.38	1.32

Table 3. Results of elemental analysis of fractionated simulator samples of corium from the lower trap basket

Fraction	Content of elements, wt. %										
	O	Na	Mg	Al	P	K	Ca	Ti	Cr	Fe	Ni
5.6–8 mm	49.29	1.56	0.15	47.95	0.09	0.10	0.22	0.09	0.14	0.42	—
2.8–4 mm	44.20	1.42	0.82	51.98	0.27	—	0.37	0.08	0.19	0.67	—
0.8–1.6 mm	39.26	0.81	2.03	56.89	—	—	0.23	0.10	0.21	0.46	—
200–400 μm	45.91	0.11	—	52.85	—	0.18	0.22	0.08	0.16	0.49	—
50–100 μm	45.53	0.54	0.69	52.02	—	0.23	0.34	0.05	0.16	0.45	—
less than 50 μm	46.19	0.61	0.64	48.90	0.06	0.14	1.37	—	0.36	1.34	0.39

As shown by the results of elemental analysis, the main compound in the corium collected from the lower trap is aluminum oxide. The ratio of Al and O concentrations in most samples is very close to the ratio for the stoichiometric composition of Al_2O_3, which indicates satisfactory accuracy of the estimates obtained in the application of the semi-quantitative method.

In addition, in the elemental composition of the corium samples, both from the lower melt trap and from the experimental container, in addition to impurity elements, stainless steel components such as Cr, Fe, Ni were found. The highest concentration of stainless steel components is observed in the fraction "< 0.05 mm". This fraction also has the highest concentration of calcium. The fraction of particles "< 0.05 mm" also contains a large amount of suspended material, presumably a product of chemical precipitation of compounds in an alkaline solution. Probably, this is the reason for the increased content of such elements as Ca, Fe, Cr, Ni.

The sodium content in the sample material, which increases with increasing particle size, can be explained by both incomplete removal of sodium from the particles during the washing process and the presence of water-insoluble sodium-aluminum compounds formed as a result of the chemical interaction of corium with sodium during the experiment.

The phase analysis of the samples was carried out using an Empyrean X-ray diffractometer (Panalytical) in copper radiation. The identification of phase components was carried out using crystallographic data from the PDF-2 database, the HighScore spectrometric measurement results processing program, OriginPro-7.5.

The phase composition of the samples of fragments of solidified corium simulator from the basket of the lower melt trap after washing from soluble sodium compounds is represented mainly by aluminum oxide in various crystalline modifications: α-Al_2O_3, γ-Al_2O_3 and β-Al_2O_3.

The crystalline modification in most corium samples is α-Al_2O_3 (corundum), which is the main stable low-temperature phase.

β-phase aluminum oxide is a compound of aluminum and alkali metal oxides and is identified in diffraction patterns as a phase with a hexagonal crystal lattice of the P63/mmc space group. The presence of the β-Al_2O_3 crystalline modification (up to 10÷15 wt.%) in the phase composition is typical for samples of all fractions and increases with increasing particle size. The tendency for the amount of β-Al_2O_3 to increase in the composition of the material coincides with the increase in the Na content in the elemental composition of the samples, which indirectly confirms the expected chemical composition of the phase $Na_2O \times 11Al_2O_3$.

Aluminum oxide of the γ-phase is satisfactorily identified in diffraction patterns as a crystalline phase with a cubic lattice of the Fd3m space group of the spinel type. In fact, the γ-Al_2O_3 modification is a high-temperature crystalline modification of aluminum oxide, and its presence can presumably be explained by quenching of the high-temperature phase in the surface layer of the solidifying melt. However, there are data confirming the nature of the formation of this phase from aluminum hydroxide. With a decrease in particle size, the proportion of γ-Al_2O_3 in the phase composition of corium increases significantly.

Semi-quantitative assessments based on corundum numbers, along with a decrease in the intensity of the α-Al_2O_3 modification peaks by almost 2 times, suggest a high content of this phase in the fraction of particles smaller than 50 µm, which, according to various estimates, is 30–60 wt.%. In the material of fractions with particle sizes larger than 1 mm, the content of the phase is low and is on the verge of detection.

Samples of fraction "10 mm÷12.5 mm", prepared from particles with characteristic external features, showed a virtually identical phase composition, characteristic of the material of large fraction particles. Some differences were revealed in the structural parameters of β-Al_2O_3, which has characteristic shifts of individual reference lines on the diffraction patterns of the samples. A detailed study revealed the presence of shifted lines in the diffraction patterns of other samples with a high content of β-Al_2O_3which suggests the existence of two modifications β1 and β2 with different structural parameters.

DISCUSSION

Based on the research results, the following can be highlighted:

− The largest mass fraction in the corium remaining in the experimental container consists of fairly large particles, > 20 mm in size, which have a predominantly lamellar shape and α-modification of Al_2O_3 in the phase composition;

− The ratio of aluminum and oxygen concentrations in a significant portion of the corium samples from the experimental container corresponds to the stoichiometric composition of Al_2O_3, which indicates satisfactory accuracy of the estimates obtained in the application of the semi-quantitative method. The small difference in the ratio of Al and O concentrations was presumably influenced by the process of washing the samples in water and keeping the corium in an aqueous solution to get rid of sodium;

− It was found that the phase composition contains three crystalline modifications of aluminum oxide, the main one being the α-modification (corundum). β-Al_2O_3 and γ-Al_2O_3 were identified as additional phases. The presence of the crystalline modification β-Al_2O_3 (up to 10÷15 wt.%) in the phase composition is typical for samples of all fractions and increases with increasing particle size. The tendency for the amount of β-Al_2O_3 to increase in the composition of the material coincides with the increase in the Na content in the elemental composition of the samples, which indirectly confirms the expected chemical composition of the phase $Na_2O \times 11Al_2O_3$. The γ-Al_2O_3 modification is a high-temperature crystalline modification of aluminum oxide, and its presence can be explained by quenching of the high-temperature phase in the surface layer of the solidifying melt.

The study confirmed the possibility of using a corium simulator made of aluminum oxide to describe the corium/coolant interaction process. Data were obtained on the granulometric composition of the products of the corium simulator interaction, data characterizing the shape of the particles, allowing one to judge the nature of the process occurring at the final stage of a severe accident at the reactor. The obtained data can be used to design systems and devices for localizing the consequences of a severe accident at the reactor.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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МОДЕЛЬДІК КОРИУМНЫҢ ЖЫЛУТАСЫМАЛДАҒЫШПЕН ӨЗАРА ӘРЕКЕТТЕСУІН ФРАГМЕНТТЕУ ПРОЦЕСІ

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Бұл жұмыста реактордан тыс жағдайларда алынған ядролық реактордың (кориум) белсенді аймағының қатқан модельдік балқымасының түйіршікөлшемдік параметрлерін, элементтік және фазалық құрамдарын зерттеу нәтижелері ұсынылған. Реактордан тыс эксперименттер ҚР ҰЯО РМК АЭИ «EAGLE» стендінде жүргізіледі, ол белсенді аймақ компоненттерінен шихта балқымасын алуға және оның жылутасымалдағышпен өзара әрекеттесуін іске асыруға мүмкіндік береді. Шапшаң нейтрондардағы реактордың модельдік кориумын алу үшін натрий жылутасымалдағыш ретінде алюминий оксиді (Al_2O_3) пайдаланылды.

Зерттеу нәтижелері фазалық құрамында Al_2O_3 алюминий тотығының үш модификациясының қатқан имитаторын анықтады. Негізгісі α-модификациясы (корунд), ол сондай-ақ шығыс шихтаның базалық компоненті болып саналады. Алюминий тотығының басқа кристалдық модификацияларының пайда болуы және заңды таралуы модельдік кориумның жылутасымалдағышпен өзара әрекеттесуі кезінде туындайтын процестердің жүру ерекшелігімен байланысты белгілер екені анық. Осындай ерекшеліктер реактордың белсенді аймағының балқымасына тән және сонда қолданылуы мүмкін, бірақ қолданылатын имитатор үшін төлсипатты болуы мүмкін.

Түйін сөздер: *модельдік кориум, натрий жылутасымалдағышы, фрагменттеу, балқыма, эксперименттік модельдеу.*

ПРОЦЕСС ФРАГМЕНТАЦИИ МОДЕЛЬНОГО КОРИУМА ПРИ ВЗАИМОДЕЙСТВИИ С ТЕПЛОНОСИТЕЛЕМ

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В настоящей работе представлены результаты исследования гранулометрических параметров, элементного и фазового составов затвердевшего модельного расплава активной зоны ядерного реактора (кориума), полученного во внереакторных условиях. Внереакторные эксперименты проводятся на стенде «EAGLE» ИАЭ НЯЦ РК, который позволяет получить расплав шихты из компонентов активной зоны и реализовать его взаимодействие с теплоносителем. Для получения модельного кориума реактора на быстрых нейтронах был использована оксид алюминия (Al_2O_3), в качестве теплоносителя – натрий.

Результаты исследований выявили присутствие в фазовом составе затвердевшего имитатора трех модификаций окиси алюминия Al_2O_3. Основной является α-модификация (корунд), которая также является базовым компонентом исходной шихты. Появление и закономерное распределение других кристаллических модификаций окиси алюминия определенно является признаком, связанным с особенностями протекания процессов, возникающих при взаимодействии модельного кориума с теплоносителем. Подобные особенности могут оказаться характерными и распространяться на собственно расплав активной зоны реактора, но могут оказаться и специфичными для применяемого имитатора.

Ключевые слова: *модельный кориум, натриевый теплоноситель, фрагментация, расплав, экспериментальное моделирование.*