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Study of Corium Thermophysical Properties of Light Water Reactor with Different Oxidation Degrees of Zirconium.

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Abstract

The work hereby studies phase composition and microstructure condition of corium prototype (received in out-of-pile experiment) of light water power reactor with different oxidation degree; defines its thermo physical properties (specific heat capacity, temperature conductivity and heat conduction) in the range from room temperature to about ~450 ^{o}C. The obtained data on temperature dependences of thermo physical properties (TPP) of corium can be used for conduction of temperature field's computations in the modeling of processes on molten corium retention in the reactor vessel.

Keywords: light water reactor, reactor core, fuel and structural reactor materials, reactor materials, corium, thermo physical properties of corium.

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INTRODUCTION

As is known, hypothetical severe accident occurring in a nuclear power plant is accompanying with the failure of a reactor vessel followed by the yield of nuclear fuel melt into containment that is the last security barrier. Corium is a nuclear molten fuel changing its composition due to meltdown (during its moving) of structural elements, equipment, etc. The properties of the corium can vary over a wide range [1, 2, 3, 4, 5]. Data on its properties including thermophysical ones may be extremely useful on searching and analyzing ways of enhancement safety using NPP and exclusion of severe accidents.

To date TPP of many structural and fuel materials of nuclear reactor are known in the range of their working temperatures [6, 7, 8, 9, 10, 11], but the information about TPP of corium prototype is completely absent (even model corium, i.e. the corium that obtained in reactor tests).

Since melts of structural and fuel materials of reactor core (corium) cannot be observe as some mixture with beforehand known compositions and structures of ingredients, so it is not possible to apply the theoretical estimations of their thermo physical properties of "rules of additiveness", especially these rules are significantly disrupted on phase transitions [12]. Therefore results of experimental determination of TPP samples of any corium (both model and prototype) are of great interest.

The aim of this work is to experimentally determine TPPs of corium prototype of light water reactor that are received from melting mixture materials, contained UO_2, Zr and ZrO_2 in the graphite crucible of high frequency heating installation at the Institute of Atomic Energy NNC RK.

EXPERIMENTAL

To receive data on thermo physical properties of solidified core melt materials we conducted tests on producing corium prototype ingots of defined composition. Melting of corium components was produced by induction heating on the test bench VCG-135 in graphite (R4340) crucible with preliminary coated ZrC on its inner surface. ZrC coating limits carbonization of burden and molten corium components at their interaction with the carbon of graphite crucible.

Scheme of burden loading into crucible, layout diagram of TC-1 thermocouple for measuring melt surface temperature with IMPAC brightness pyrometer and pyrometer of spectral relation of MIKRON 770s are shown in Fig.1a; outward of crucible with coated carbide is shown in Fig.1b; and outward of crucible filled with prepared burden in form of powders and plates are illustrates in Fig.1c. The parameters of conducted tests on producing corium with defined composition are listed in Table. 1.

a – Experimental crucible assembly scheme

b– crucible with protectivecoating from ZrC

c – crucible cover with a hole for pyrometer measuring and the crucible filled with burden

Figure 1: Experimental crucible assembly

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Table 1: Conditions of three tests on receiving corium prototype ingots

Parameter	Meaning		
	№1	№2	№3
Loading of crucible, g			
Granulate UO_2	61,28	89,37	87,18
plates of metallic Zr	27,17	14,07	8,23
powder ZrO_2	27,17	16,99	23,21
Correlation U/Zr	0,47	1,13	1,16
Degree of zirconium oxidation, %	42,54	47,20	67,60
Pressure in process chamber, MPa	0,1	0,1	0,1
Maximal temperature, °C	2475	2450	2420
Color temperature at the melting moment, °C	2180	2280	2280
Mass of received ingot, g	118,0	120,0	118,5

As a result of the conducted tests, the compact ingots of corium prototype were received and outward of them are presented in Fig. 2

ingot №1 ingot №2 ingot№3

Figure 2: Outward of corium ingots

Measuring of thermo physical properties of corium was carried out on thin disk samples (fig. 3) prepared to the conditions of laboratory setting of the thermo physical measuring of UTFI-2.

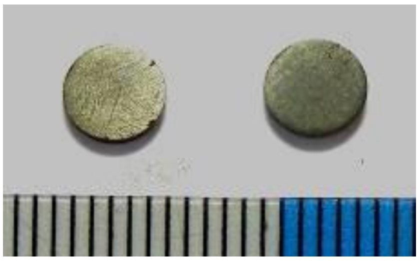

Figure 3: Outward of samples

Preparation of disk samples to measuring of TPP included next basic operations: undercut from corium ingots some flat fragment in thick no less than 8 mm; drilling out work piece of samples (cylinders in about high 8 mm) with core drill with an abrasive tip; undercut of two identical disks from every work piece with a supply on polishing of surfaces (material that remaining in a holder was used in further for preparation of powder samples for X-ray phase analysis); final polishing of samples in the special mandrel to the set thickness and set roughness of surfaces.

X-ray phase analysis of samples was executed on DRON- 3 diffractometer with using of Cu-radiation ($\lambda_{K\alpha1}$ = 0,154056 nm). The state of surface samples structure was studied on the optical microscope OLIMPUS BX41M at magnifications from ×20 to ×650.

At determination of corium samples thermo physical properties on setting of UTFI- 2 we used methodology [13], based on the well-known method of thermal flash. Essence of the methodology realized on UTFI-2 consists in heating of one of flat sample ends with short-term affecting of thermal impulse and registration of temporal dependence of temperature on the opposite sample end.

RESULTS AND DISCUSSION

At the beginning of implementation of research quality and quantitative X-ray phase analysis on powder samples was conducted, then the analysis of structure state on the disk samples surfaces was implemented, after that measuring of TPP on these samples are conducted.

X-ray phase analysis of corium samples

The main components of corium phase composition of all tests, on results of conducted analysis are (see Table 2) a row of solid solutions (U, Zr) O_2 with the face-centered cubic (FCC) lattice of uranium dioxide and phase α- Zr (O, U) on the basis of α-modification of metallic zirconium. Stoichiometric composition of sosoloids (U, Zr)O_2 was determined by evaluation way by linear interpolation of values of grates periods of FCC-phase of UO_2 and Zr O_2 (equal according to 0,547 and 0,511 nm) to the period of grate sosoloid, determined on a diffractogram.

Table 2: Data of phase and element analysis of gross composition of ingot materials

Oxidation degree Zr, % (мол.)	Phase composition (indicative formulaic composition of the main phases and the parameters of their crystal lattices)	Ultimate composition % (mass.)		
		U	Zr	U/Zr
42,5 (Test №1)	$(U_{\sim0,7}Zr_{\sim0,3})O_2$ … $(U_{\sim0,5}Zr_{\sim0,5})O_2$ (a = 0,538 … 0,530 nm); α-Zr(O) (a = 0,330 ± 0,005 nm, c = 0,530 ± 0,005 nm)	52,5	47,5	1,11 ± 0,05
47,2 (Test №2)	$(U_{\sim0,9}Zr_{\sim0,1})O_2$ … $(U_{\sim0,6}Zr_{\sim04})O_2$ (0,544 … 0,536 nm); α-Zr(O) (a = 0,330 ± 0,005 nm, c = 0,530 ± 0,005 nm)	74,6	25,4	3,0 ± 0,15
67,6 (Test №3)	$(U_{\sim0,8}Zr_{\sim0,2})O_2$ … $(U_{\sim0,5}Zr_{\sim0,5})O_2$ (0,542 … 0,530 nm); α-Zr(O) (a = 0,330 ± 0,005 nm, c = 0,530 ± 0,005nm)	75,5	24,5	3,1 ± 0,10

It is found out that within the ingot the stoichiometric composition and correlation of phases differ in the samples selected from different places on a height that indicates discontinuity of ingot crystallization. For samples in upper part of ingots enhancable content of zirconium in the sosoloid (U, Zr)O_2 and element composition is typical, and for samples in the bottom part of ingots the lowered content is typical. However gross composition of ingot concerning uranium and zirconium content is practically identical to initial composition of the burden.

State of corium sample structure

In the study of the microstructure on the surfaces of corium disk samples we determined quantitative content of structural components, corresponding to specific phases and pores in the material samples (all in one) (Fig.4 some of the characteristic images of the surfaces of corium samples from test #1 are shown as an example).

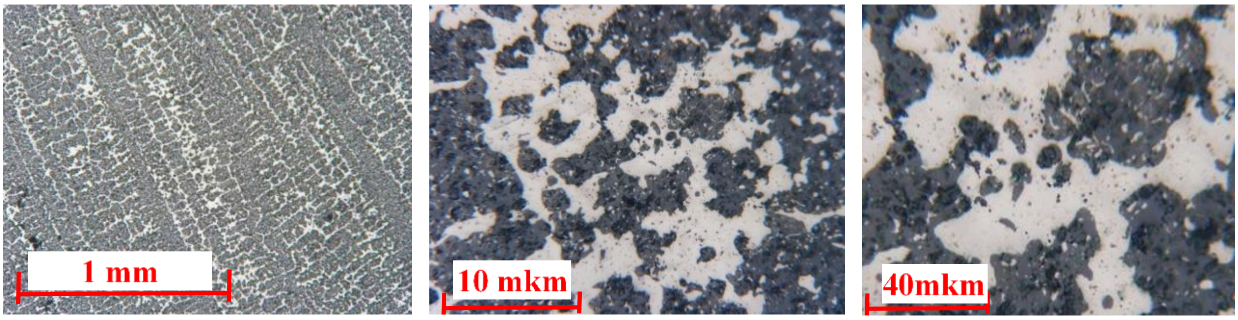

Figure 4: Characteristic microstructure of corium from test №1

Thermo physical properties of corium samples at a room temperature

At determination of thermo-physical properties of six disk samples of corium prototype from tests №1, №2 and №3 great attention was paid on of the precision of measuring results. In particular, results of determination of coefficient of thermal diffusity a, heat conductivity λ and specific heat capacity of C_p corium (and uranium dioxide) at a room temperature, presented in Table 3, were received on the basis of at least 9 measurements.

Table 3: Results of determination TPP of corium at a room temperature

Number of test	Denotation of sampling (kern)	ρ, g/cm^3	a, $10^{-6}\ m^2 \cdot s^{-1}$	C_p, $J \cdot kg^{-1} \cdot K^{-1}$	λ, $W \cdot m^{-1} \cdot K^{-1}$
№1	1-1	8,5 ± 0,1	1,55 ± 0,15	349 ± 20	4,4 ± 0,5
	1-2	8,3 ± 0,2	1,4 ± 0,1	350 ± 10	4,0 ± 0,1
№2	2-1	8,8 ± 0,1	1,35 ± 0,01	303 ± 5	3,6 ± 0,2
	2-2	9,2 ± 0,1	1,36 ± 0,01	301 ± 2	3,8 ± 0,15
№3	3-1	8,5 ± 0,1	1,01 ± 0,02	337±2	2,9 ± 0,15
	3-2	8,8 ± 0,1	0,97 ± 0,01	320 ± 3	2,75 ± 0,08
Pill UO_2	–	10,3 ± 0,1	2,84 ± 0,05	244 ± 1	6,90 ± 0,05

The error of TPP measuring (root-mean-square errors at 95-percent confidence probability are listed in the Table) is conditioned mainly by variation of results of measuring, arising up during the repeated setting of disk samples in the measuring assembling of setting of UTFI- 2. The influence of this factor turned out to be very significant, it is necessary to take into account when analyzing the results of the determination TPP corium.

The analysis of determination results of TPP corium samples at room temperature showed significant differences from TPP of uranium dioxide pellets. The values of specific heat capacity of all samples of corium are higher, than value for the samples of uranium dioxide the values of coefficient of heat conductivity and diffusivity appeared substantially lower, in spite of being in the structure of material metallic zirconium heat conductivity of that is much higher, than at uranium dioxide.

The last circumstance can be explained as by the substantial decline of thermal conductivity composing the structure of corium phases, especially metallic zirconium steady state by oxygen, so by the presence of significant porosity of corium.

A receipt on the whole of nonconflicting results of measuring of TPP corium samples at a room temperature allowed beginning study temperature dependence TPP of these samples.

Temperature dependences of thermo physical properties of corium sample.

Before realization of works on determination of temperature dependence of TPP of corium prototype measuring of thermo physical properties was executed on calibration disk samples from stainless steel in the range from a room temperatures to 600 °C in an inert environment. According to results of the calibration measurement calculation coefficients of effective energy impulse were defined

$$Q_{eff} = Q_{meas} \cdot k \cdot (1 - \alpha(T - T_o)),$$

where Q_{meas} – is electric measureable energy of impulse, k and α – experimentally determined constants.

Received constants were used for computing effective pulse energy on calculating coefficients TPP disc samples of corium prototype of light water reactor.

Measurement of temperature dependences of the TPP corium samples of №1 test were completed in the range of temperature to ~ 600 ° C under argon medium. The results of measurements of the TPP corium on №1 test are shown in Fig. 5.

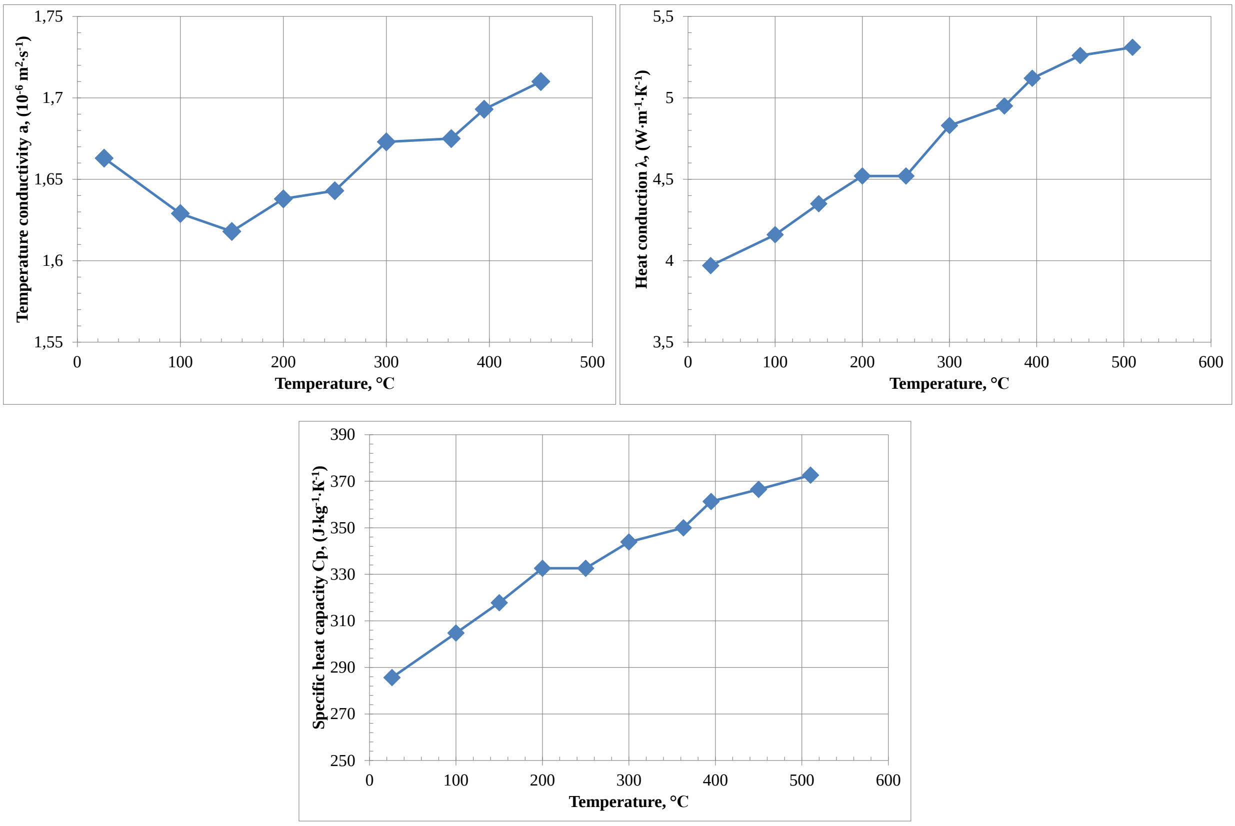

Figure 5: Temperature dependence of the corium prototype TPP in experiment №1

CONCLUSION

- It is shown that by research of microstructure of corium, determinations of its phase composition and compactness in the initial state, it is possible to watch conformities, joining the indicated descriptions of corium with its thermo physical properties.
- It is found out that quantified factor of declining the heat conductivity of corium compared to heat conductivity of uranium dioxide it should be considered substantial decline of heat conductivity composing its phases of variable composition compared to initial phases on the basis of that formed, and also forming of high porosity in solidified corium structure.
- The data on TPP of solidified melt corium prototype material in the range of temperature from 50 to 400 in argon medium indicated steady tendency to an increase of all its thermo physical characteristics with increasing temperature measurements.

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