Thermal properties of $(Al,Gd)O_3$ doped uranium dioxide

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Abstract. In this paper were defined thermal characteristics (thermal diffusivity up to 2000 °C, heat capacity up to 1250 ° C and thermal conductivity of up to 1250 °C) of oxide nuclear fuel with additives of burnable neutron absorber in the form of $(Al,Gd)O_3$. Also for comparison samples UO_2 with gadolinium Gd_2O_3 and pure uranium dioxide were studied. It is shown that the thermal diffusivity and thermal conductivity of UO_2 with dopant of gadolinium in the form of (Al, Gd) O_3 is considerably higher than those of Gd_2O_3 –doped uranium dioxide, due to the fact that (Al, Gd)O_3 does not form a solid solution with UO_2.

1. Introduction

In order to achieve high burnup of WWER fuel requires using burnable neutron absorbers (Gd for WWER fuel), to reduce the reactivity in the initial stages of TVEL action. But the injection to the fuel gadolinium in the form of Gd_2O_3 leads to a significant reduction of uranium dioxide thermal conductivity [1,2,3], and as a consequence an increase of the temperature gradient between fuel and cladding. To solve this problem were offer to inject in uranium dioxide gadolinium as a perovskite phase $AlGdO_3$, which improves the thermal conductivity of the fuel without sacrificing Gd amount in it [4].

To carry out the work following uranium dioxide samples were made: R - pure uranium dioxide, G - uranium dioxide with 6.9% Gd_2O_3 and AG - uranium dioxide with 8.9% $AlGdO_3$. The density of the samples obtained are given in table 1.

Table 1. Density of the sample investigated batches

Batch	Theoretical density (TD) (g/cm^3)	Density ρ (g/cm^3)	% of TD
R	10.96	10.56	96
G	10.80	9.78	91
AG	10.41	9.62	92

2. Determination of heat capacity

The heat capacity in the temperature range 35-1215 °C determined by differential scanning calorimetry method in accordance with standards DIN 51007, ASTM E 1269 in an atmosphere of high purity argon. Specimens have a diameter of 6 mm and a thickness of 1.5-1.6 mm. The heat capacity measurements were performed on the synchronous thermal analyzer Netzsch STA 409 CD. As a

reference were used disk of sapphire 6 mm in diameter and 1 mm thick. Relative error of the method is less than 5%. The results are shown in figure 1.

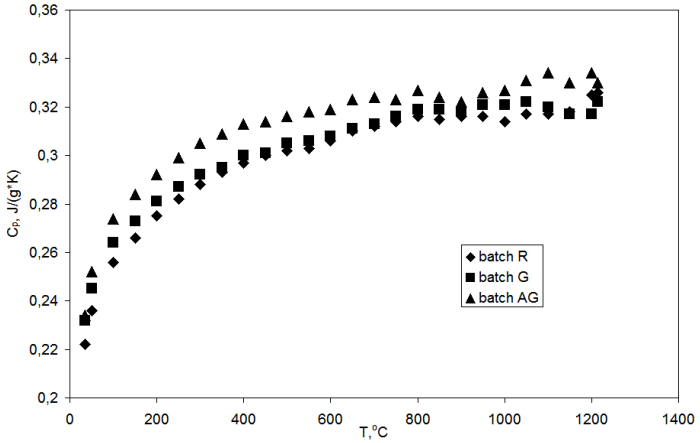

Figure 1. The heat capacity of UO_2 samples of all batches.

It should be noted that because of the small amounts of additives difference of the specific heat values of samples from different batches is small and does not exceed the measurement error.

3. Determination of thermal diffusivity

Measurements were carried out on Netzsch LFA 427 by laser flash method in a high purity argon atmosphere. The samples had form of discs 10 mm in diameter, 2 mm thick.

Uranium dioxide thermal diffusivity values of R, G and AG batches shown in figure 2.

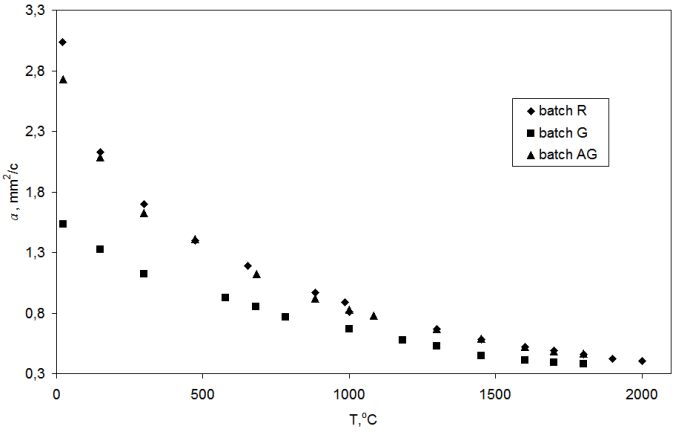

Figure 2. Thermal diffusivity UO_2 samples of all batches.

From these results, it's clear that thermal diffusivity of the samples decreases from 3 to 0.3 mm^2/s while temperature increasing. The thermal diffusivity of the samples with gadolinium additives is lower than that of pure uranium dioxide, but it is clear that a large influence exert form of burnable poison (Gd) is injected into the fuel. In samples of batch G gadolinium injected as Gd_2O_3, and during sintering it dissolves in the matrix and forms a solid solution with UO_2, which as known negatively affect on the thermal conductivity. In the samples of batch AG gadolinium is in the form of a perovskite phase $AlGdO_3$, and does not introduce distortion into UO_2 lattice, resulting in only a slight reduction in thermal conductivity compared to pure uranium dioxide in figure 2.

4. The calculation of the thermal conductivity

From the obtained data of thermal diffusivity, specific heat and density using the expression (1) were calculated temperature dependence of the thermal conductivity.

$$\lambda(t) = a(t) \cdot C_p(t) \cdot \rho(t). \quad (1)$$

Obtained data are recalculated to 95% TD by the expression

$$\rho_p = \rho_{100}(1-p)^{2,5}. \quad (2)$$

The results are shown in figure 3.

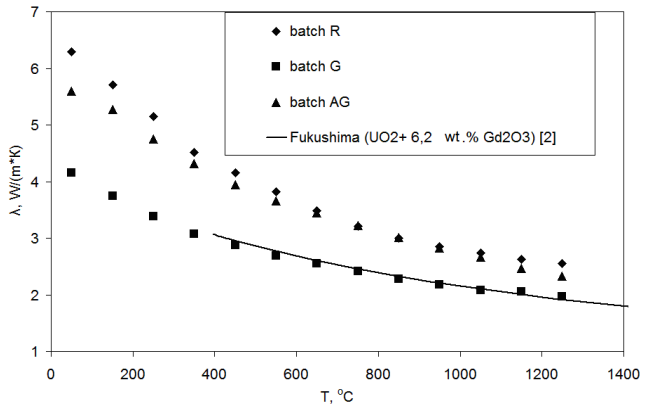

Figure 3. The temperature dependence of the thermal conductivity of samples UO_2.

The thermal conductivity of the samples is in the range 2 - 6.5 W/(m·K). Also we can see that dopants of $AlGdO_3$ much weaker affect on the thermal conductivity UO_2 than Gd_2O_3 one.

5. SEM-study UO_2 samples

The state of gadolinium in the samples G and AG confirmed by SEM (figures 4, 5 and tables 2, 3).

In Figure 4 - the microstructure of the sample AG in secondary electrons, UO_2 looks light gray, $AlGdO_3$ - dark gray. From the distribution map Gd, and gained spectra shows that all gadolinium is in $AlGdO_3$ form. From Figure 5, which shows the microstructure in the secondary electron and gadolinium distribution for sample batch G seen that gadolinium is evenly distributed throughout the matrix UO_2.

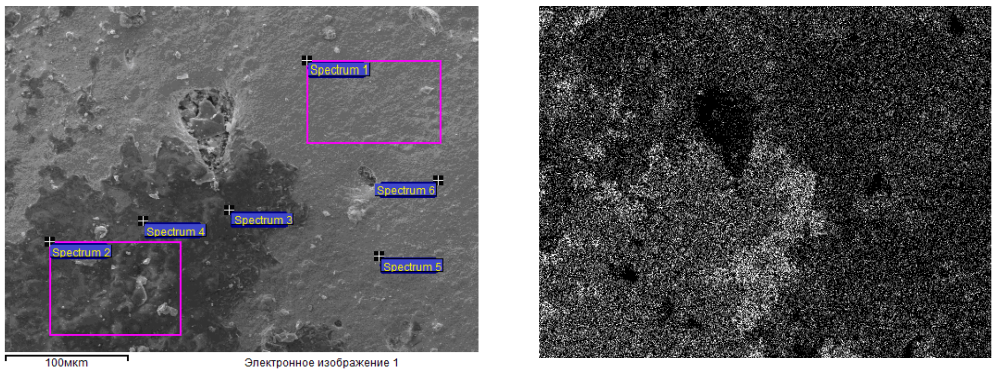

Figure 4. SEM image and distribution map of gadolinium in the sample batch AG.

Table 2. Composition data of the sample batch AG (wt.%)

	O	Al	Si	Gd	U
Spectrum 1	12.42		0.47		87.10
Spectrum 2	32.29	31.82	2.28	12.31	21.30
Spectrum 3	31.27	35.16		12.92	20.65
Spectrum 4	33.86	27.83		8.58	29.72
Spectrum 5	12.73				87.27
Spectrum 6	12.53				87.47

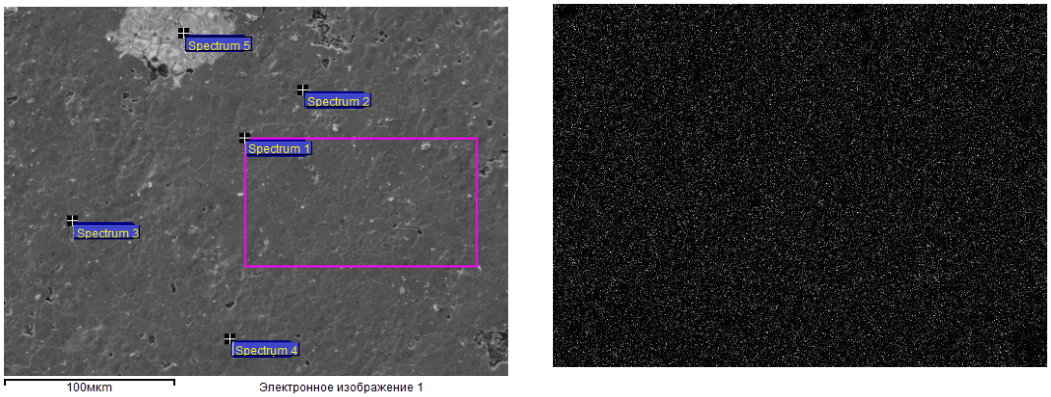

Figure 5. SEM image and distribution map of gadolinium n the sample batch G.

Table 3. Composition data of the sample batch G (wt.%)

	O	Al	Si	Gd	U
Spectrum 1	11.76			4.77	83.48
Spectrum 2	16.94	4.71		6.86	71.49
Spectrum 3	6.49	1.35	0.51	4.90	86.75
Spectrum 4	9.16			5.16	85.69
Spectrum 5	27.30			4.39	68.30

6.Conclusions

In the present work measured basic thermophysical characteristics oxide fuel with additives burnable neutron absorber in the form of a perovskite phase (Al, Gd) O_3 and in the form of a solid solution UO_2 - Gd_2O_3. State of gadolinium in the samples is confirmed by SEM-studies (figures 5, 6).

It is shown that the thermal conductivity of the samples decreases with increasing temperature and is in the range of 2-6.5 W / (m·K), moreover serious influence exert gadolinium state in UO_2: formation of a solid solution UO_2 - Gd_2O_3 causes distortion of UO_2 crystal lattice and as a result, a strong decrease (35%) of thermal conductivity. Gadolinium it the form of perovskite phase does not make distortion into the lattice and thermal conductivity decreased less than 5% in the entire investigated temperature range.

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References

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