Plasmachemical Synthesis and the Assessment of the Thermal Conductivity of Fuel Compounds "UO_2–MgO"

Igor Shamanin[1, a)], Alexander Karengin[1, b)], Alexey Karengin[1, c)], Ivan Novoselov[1, d)], Andrey Poberezhnikov[1, e)] and Evgeny Alyukov[1, f)]

[1]*Tomsk Polytechnic University, 30 Lenina Avenue, Tomsk, 634050 Russian Federation*

[a)]shiva@tpu.ru
[b)]Corresponding author: karengin@tpu.ru
[c)]karenginaa@tpu.ru
[d)]inovoselov@tpu.ru
[e)]andrewpad@tpu.ru
[f)]john.judo@mail.ru

Abstract. The process of plasma-chemical synthesis from water-organic nitrate solutions of uranium and magnesium of fuel compositions including a matrix of magnesium oxide with a high thermal conductivity and low neutron absorption is investigated. The compositions of the solutions, including acetone (ethanol), as well as the regimes that providing the direct synthesis of "UO_2–MgO" fuel compositions of different concentrations in air plasma are determined. The calculation of their thermal conductivity and comparison with experimental data are presented.

INTRODUCTION

Traditional nuclear fuel (NF) based on uranium dioxide enriched by uranium-235 has a low thermal conductivity and a short operation cycle [1].

For nuclear power engineering, it is promising to use dispersive NF, in which there are no direct contacts between particles of fissile material due to their uniform distribution in a matrix with high thermal conductivity and low neutron absorption. This provides high thermal conductivity and mechanical properties, radiation resistance and strength of the fuel compositions, higher burnup of fissile material and localization of fission products [1-5].

Major disadvantages of applied plasma technologies for synthesis of fuel compounds (FC) from nitric solutions are high energy costs (up to 4.0 MW·h/t) and necessity of additional hydrogen reduction for obtaining compounds of required composition [6-9].

The advantages of plasma-chemical synthesis of FC from water-organic nitrate solutions (WONS) including organic component should include: one-step process, low energy consumption (up to 0.1 MW·h/t), obtaining compounds of required composition without additional hydrogen reduction [10-12].

The purpose of this work is determination of possibility of direct plasma-chemical synthesis of FC "UO_2–MgO" of different compositions in air plasma from WONS and the assessment of the impact of MgO matrix proportion on thermal conductivity of such FC.

The following tasks were defined: determination of the WONS composition and modes for their plasma processing, which ensure the direct plasma-chemical synthesis of FC of required composition; the assessment of the impact of MgO matrix proportion on the thermal conductivity of FC "UO_2–MgO" using different mathematical models, and comparison results with the available experimental data.

CALCULATION OF BURNING PARAMETERS AND COMPOSITION OF WONS

Lower heat value of WONS is determined from formula [13]:

V International Conference for Young Scientists, Post-Graduate Students and Students
AIP Conf. Proc. 2101, 020017-1–020017-6; https://doi.org/10.1063/1.5099609
Published by AIP Publishing. 978-0-7354-1831-8/$30.00

$$Q_l = \frac{(100 - W - A) \cdot Q_{lc}}{100} - \frac{2.5 \cdot W}{100}, \quad (1)$$

where Q_{lc} is lower heat value of combustible component of WONS composition, MJ/kg; W and A are content of water and noncombustible components, wt.%; 2.5 is the value of latent heat of vaporization for water at 0 °C, MJ/kg.

Authors in [10-12] described, that WONS compositions having $Q_l \geq 8.4$ MJ/kg allow to make the plasma processing energy efficient and direct plasma-chemical synthesis of FC of required composition.

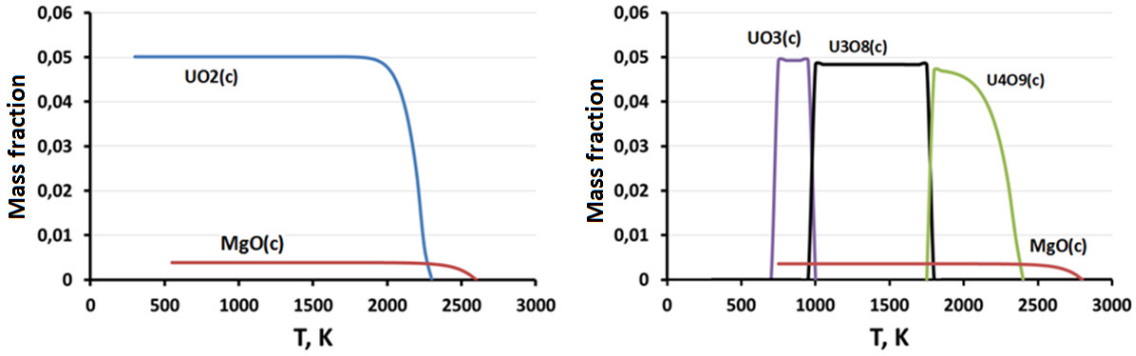
According to the results of the calculations the following WONS compositions ($Q_l \geq 8.4$ MJ/kg) providing synthesis of particular FC were determined:

- WONS-1 (26.1 % H_2O – 34.0 % C_2H_6O – 31.2 % $UO_2(NO_3)_2 \cdot 6H_2O$ – 8.7 % $Mg(NO_3)_2 \cdot 6H_2O$) for obtaining FC-1 "95.0 % UO_2 – 5.0 % MgO";
- WONS-2 (28.1 % H_2O – 29.0 % C_3H_6O – 33.5 % $UO_2(NO_3)_2 \cdot 6H_2O$ – 9.4 % $Mg(NO_3)_2 \cdot 6H_2O$) for obtaining FC-1 "95.0 % UO_2 – 5.0 % MgO";
- WONS-3 (28.4 % H_2O – 34.0 % C_2H_6O – 23.6 % $UO_2(NO_3)_2 \cdot 6H_2O$ – 14.0 % $Mg(NO_3)_2 \cdot 6H_2O$) for obtaining FC-2 "90.0 % UO_2 – 10.0 % MgO";
- WONS-4 (30.6 % H_2O – 29.0 % C_3H_6O – 25.4 % $UO_2(NO_3)_2 \cdot 6H_2O$ – 15.0 % $Mg(NO_3)_2 \cdot 6H_2O$) for obtaining FC-2 "90.0 % UO_2 – 10.0 % MgO";
- WONS-5 (27.8 % H_2O – 34.0 % C_2H_6O – 24.6 % $UO_2(NO_3)_2 \cdot 6H_2O$ – 14.4 % $Mg(NO_3)_2 \cdot 6H_2O$) for obtaining FC-3 "85.0 % UO_2 – 15.0 % MgO";
- WONS-6 (29.9 % H_2O – 29.0 % C_3H_6O – 25.6 % $UO_2(NO_3)_2 \cdot 6H_2O$ – 15.5 % $Mg(NO_3)_2 \cdot 6H_2O$) for obtaining FC-3 "85.0 % UO_2 – 15.0 % MgO".

CALCULATION OF THE PROCESS OF PLASMA-CHEMICAL SYNTHESIS OF FC "UO_2–MgO"

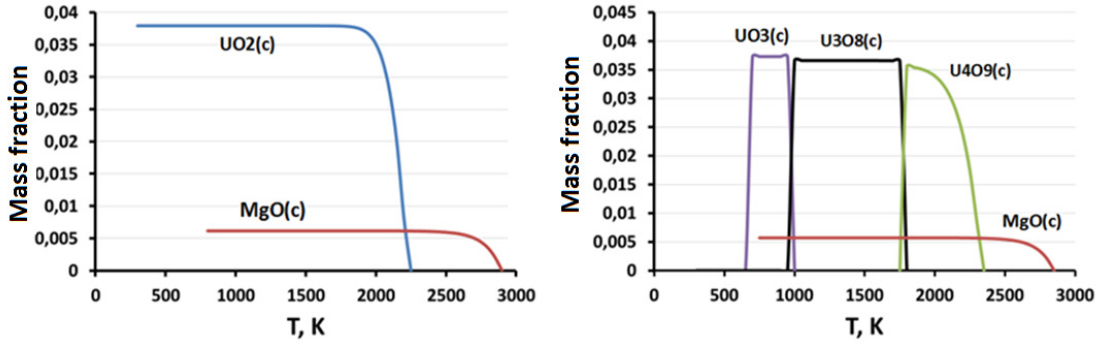
The calculation of the synthesis process of the FC "UO_2–MgO" from the WONS compositions was carried out using the licensed program "TERRA" for the thermodynamic calculation of the phase composition of heterogeneous systems at atmospheric pressure (0.1 MPa), in a wide temperature range (300–4000 K) and mass fractions of air plasma coolant (10–90%).

Figures 1-3 show the effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-1, FC-2 and FC-3 from compositions of WONS -1, WONS -3 and WONS -5 based on ethanol with a mass fraction of air of 72% (a) and 74% (b).



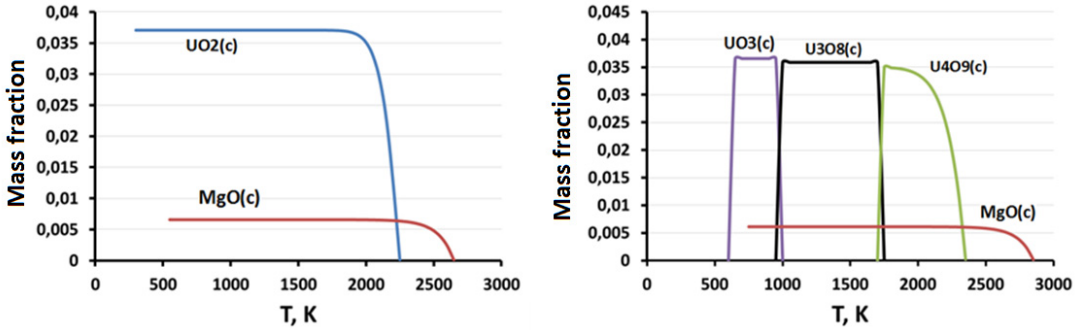
(a) (b)

FIGURE 1. Effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-1 from compositions of WONS -1 with the mass fraction of air 72 % (a) and 74 % (b).



(a) (b)

FIGURE 2. Effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-2 from compositions of WONS -3 with the mass fraction of air 72 % (a) and 74 % (b).

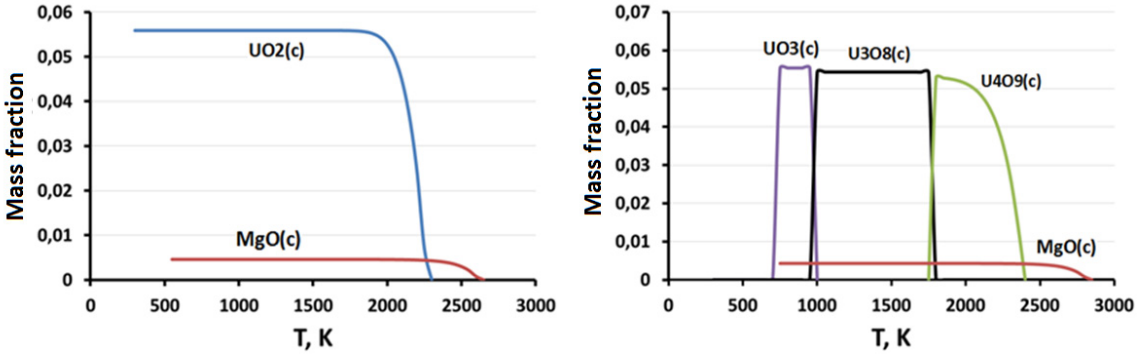


(a) (b)

FIGURE 3. Effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-3 from compositions of WONS -5 with the mass fraction of air 72 % (a) and 74 % (b).

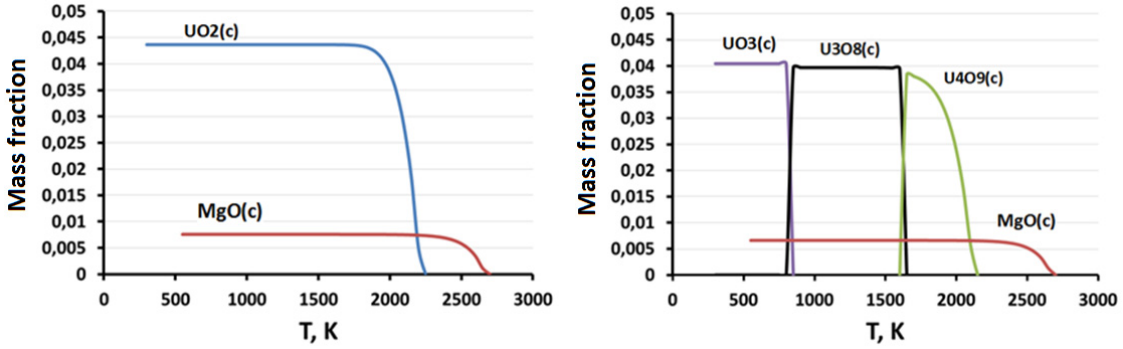
Analysis of composition of condensed products shows that plasma-chemical synthesis of WONS-1, WONS-2 and WONS-3 leads to formation of required FC-1 "95.0 % UO_2 – 5.0 % MgO", FC-2 "90.0 % UO_2 – 10.0 % MgO" and FC-3 "85.0 % UO_2 – 15.0 % MgO" with air mass fraction 72 % and temperatures below 2000 K (Fig. 1a, Fig. 2a, Fig. 3a). Decreasing air mass fraction below 72 % leads to formation of carbon and its further growth. Increasing air mass fraction from 72 % to 74 % (Fig. 1b, Fig. 2b, Fig. 3b) leads to formation of unwanted compositions "U_3O_8–MgO" at 1000–1600 K or "U_4O_9–MgO" at above 1600 K.

Figures 4-6 show the effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-1, FC-2 and FC-3 from compositions of WONS -2, WONS -4 and WONS -6 based on ethanol with a mass fraction of air of 69% (a) and 71% (b).



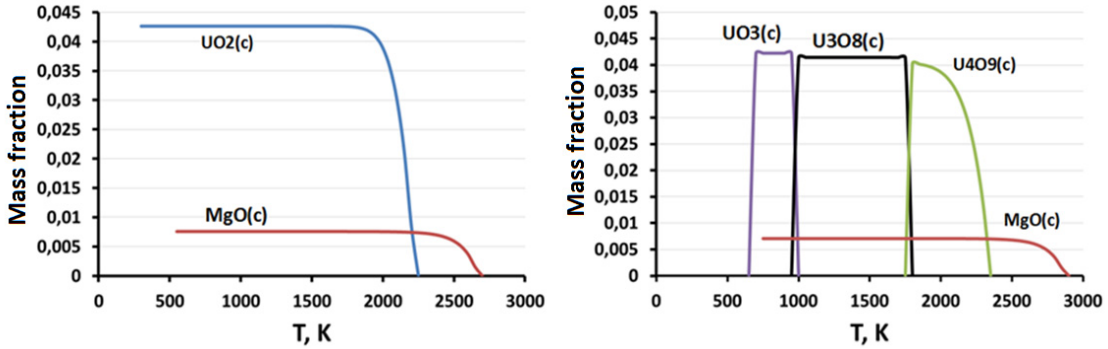
(a) (b)

FIGURE 4. Effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-1 from compositions of WONS -2 with the mass fraction of air 69 % (a) and 71 % (b)).



(a) (b)

FIGURE 5. Effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-2 from compositions of WONS -4 with the mass fraction of air 69 % (a) and 71 % (b).



(a) (b)

FIGURE 6. Effect of temperature on the composition of condensed products of plasma-chemical synthesis of FC-3 from compositions of WONS -6 with the mass fraction of air 69 % (a) and 71 % (b).

From the analysis of composition of condensed products it follows that plasma-chemical synthesis of WONS-2, WONS-4 and WONS-6 leads to formation of required FC-1 "95.0 % UO_2 – 5.0 % MgO", FC-2 "90.0 % UO_2 – 10.0 % MgO" and FC-3 "85.0 % UO_2 – 15.0 % MgO" with air mass fraction 69 % and temperatures below 2000 K (Fig. 4a, Fig. 5a, Fig. 6a). Decreasing air mass fraction below 69 % leads to formation of carbon and its further growth. Increasing air mass fraction from 69 % to 71 % (Fig. 4b, Fig. 5b, Fig. 6b) leads to formation of unwanted compositions "U_3O_8–MgO" at 1000–1600 K or "U_4O_9–MgO" at above 1600 K.

CALCULATION OF THE COEFFICIENTS OF THERMAL CONDUCTIVITY OF FC "UO_2–MgO" AND DISCUSSION OF THE RESULTS

The calculation of the thermal conductivity coefficients λ of FC "UO_2–MgO" was carried out using mathematical models [12], describing the thermal conductivity of the elementary cells of FC containing inclusions (block 2) and matrix (block 1).

Table 1 and Fig. 7 present the experimental data [13-17] and the results of calculations of λ for FC-1, FC-2 and FC-3, obtained using the Lichtenecker generalized conductivity model (Lichte model 2) for mixtures with equal components (inclusions - square), described by equation [13]:

$$\lambda = \lambda_1^{1-m_2} \cdot \lambda_2^{m_2},$$

where λ_1 and λ_2 are the thermal conductivity coefficients of the matrix materials (block 1) and inclusions (block 2), respectively, m_2 is the volume concentration of the material of block 2.

From the analysis of Table 1 and the obtained graphs (Fig. 7) it follows that Lichtenecker generalized conductivity model for mixtures with equal components having square form (Lichte2) most accurately describes the experimental data for thermal conductivity coefficient λ of FC-1, FC-2 and FC-3.

TABLE 1. Effect of temperature on coefficient of thermal conductivity of FC-1, FC-2 and FC-3

FC composition	λ, W/(m·K)					
	600 K	800 K	1000 K	1200 K	1400 K	1600 K
100 % UO_2 (exper.)	5.38	4.10	3.65	3.00	2.15	1.77
"95 % UO_2 – 5 % MgO" (calc.)	6.01	4.60	3.99	3.30	2.37	1.97
"95 % UO_2 – 5 % MgO" (exper.)	5.80	4.60	3.90	3.30	2.90	2.60
"90 % UO_2 – 10 % MgO" (calc.)	6.72	5.05	4.36	3.50	2.62	2.19
"90 % UO_2 – 10 % MgO" (exper.)	6.00	4.85	4.05	3.50	3.00	2.70
"85 % UO_2 – 15 % MgO" (calc.)	7.52	5.59	4.77	3.90	2.88	2.43
"85 % UO_2 – 15 % MgO" (exper.)	6.60	5.30	4.45	3.70	3.20	3.00

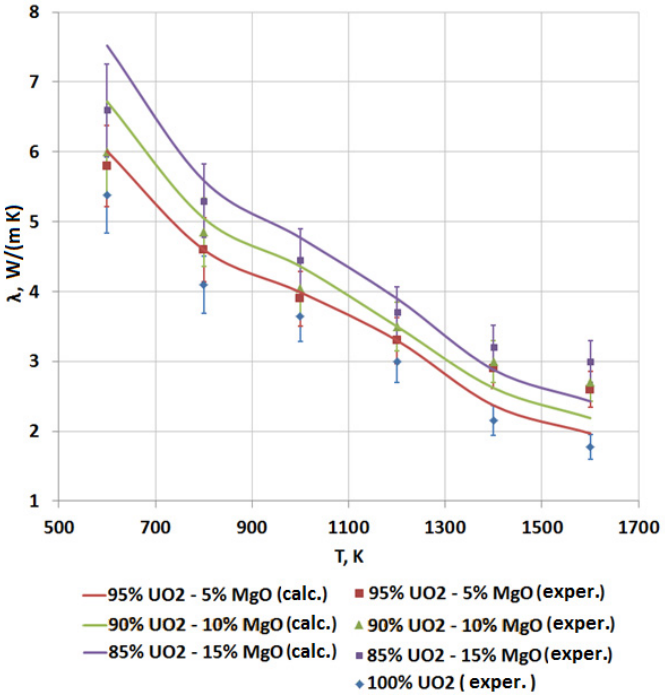


FIGURE 7. Effect of temperature on coefficient of thermal conductivity λ of FC-1, FC-2 and FC-3.

From the analysis and comparison of experimental data, it follows that an increase in the volumetric content of the MgO matrix in UO_2 by 5 %, 10 %, and 15 % results in an increase in the thermal conductivity coefficient of FC by 1.35, 1.50 and, 1.5 times at 1400 K, and by 1.47, 1.53, and 1.70 times at 1600 K respectively. Thus, the Lichte2 model can be used to assess the effect of an increase in the content of MgO matrix on the thermal conductivity of FC "UO_2–MgO" of different composition.

CONCLUSION

As a result of the calculations of optimal compositions of ethanol-based and acetone-based water-organic nitrate solutions as well as the modes (mass phase ratio, temperature) providing the plasma-chemical synthesis of FC-1 "95 % UO_2 – 5 % MgO", FC-2 "90 % UO_2 – 10 % MgO", and FC-3 "85 % UO_2–15 % MgO" were determined.

As a result of the calculations for thermal conductivity coefficients of FC with the use of a number of models it was shown that the Lichtenecker generalized conductivity model for mixtures with equal components having square form (Lichte2) most accurately describes the experimental data and can be used to assess the influence of the content of magnesium oxide on the thermal conductivity of dispersive nuclear fuel in the form of fuel compositions "UO2 – MgO".

ACKNOWLEDGMENT

The project was supported by the Russian Science Foundation (project 18-19-00136).

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