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INFLUENCE OF GADOLINIUM ON (U, Gd)O_2 FUEL PELLETS PROPERTIES

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An analysis and systematization of published data on the influence of gadolinium content on the properties of (U,Gd)O_2 fuel pellets, including melting temperature, thermophysical, and mechanical properties, are presented. Comparative burnup calculations of ^{155}Gd and ^{157}Gd were performed for WWER-1000 operating conditions for fuel containing 5 and 8 wt.% Gd_2O_3. Taking into account the influence of Gd_2O_3 on the fuel thermal conductivity and the radial power distribution, fuel center temperatures were calculated for conventional UO_2 and (U, Gd)O_2 fuels at linear heat generation rates ranging from 120 to 360 W/cm. The calculations demonstrate the competing effects of radial power redistribution and the reduction in thermal conductivity on the temperature evolution of gadolinium-bearing fuel during the fuel cycle.

INTRODUCTION

Uranium-gadolinium fuel has found widespread application for improving the technical and economic performance of nuclear power units equipped with WWER, PWR, and BWR reactors, particularly in connection with the transition to longer fuel cycles and higher fuel enrichment levels [1]. Gadolinium has seven stable isotopes: Gd^{152}, Gd^{154}, Gd^{155}, Gd^{156}, Gd^{157}, Gd^{158}, and Gd^{160}. Among them, only ^{155}Gd i ^{157}Gd possess extremely high neutron absorption cross-sections of 60,900 and 254,000 barns, respectively [1]. The unique neutronic properties result in a significant reduction of the fuel assembly multiplication factor (k∞) at the beginning of the fuel cycle. After some time, corresponding to a fuel burnup of approximately 10…15 (MW·d)/kgU, depending on the gadolinium content, these isotopes are almost completely depleted, and the k∞ of the fuel assembly becomes very close to that of an assembly without gadolinium (Fig. 1) [2].

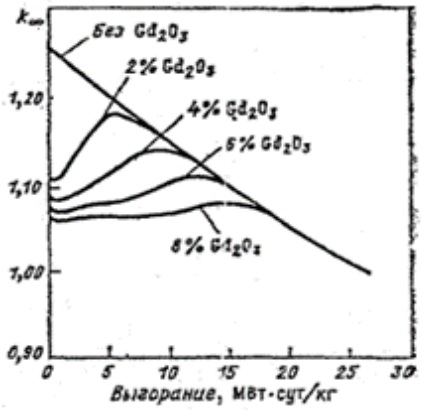


Fig. 1. Change in reactor reactivity during fuel burnup for different Gd_2O_3 contents in fuel pellets. The initial ^{235}U enrichment is 3.7% [2]

In the current fuel loading pattern of WWER-1000 power units in Ukraine, the number of fuel assemblies containing gadolinium is 163, while the number of gadolinium-bearing fuel rods per assembly is six. The fuel composition is UO_2 + Gd_2O_3 (3…5 wt.% Gd_2O_3). The pellet fabrication technology does not fundamentally differ from that used for conventional UO_2 fuel pellets: Gd_2O_3 powder is added to UO_2 powder and thoroughly mixed. This is followed by pellet pressing, sintering, and final machining to the required dimensions [3]. During sintering, a solid solution of (U, Gd)O_2 is formed [1, 3]. Current research and development trends focus on increasing the Gd_2O_3 content to 7…12 wt.% [3]. Technological concepts aimed at improving pellet thermal conductivity through the use of matrix-type fuel compositions, such as Gd_2O_3 or (U, Gd)O_2 dispersed within a UO_2 matrix [4, 5].

Investigation of the thermophysical and mechanical characteristics of UO_2 and UO_2–Gd_2O_3 fuel pellets is of considerable importance for nuclear fuel engineering. In particular, Young's modulus determines the resistance of the fuel to elastic deformation under thermomechanical loading during fuel – cladding interaction. The creep rate, in turn, characterizes the ability of the fuel material to undergo long-term plastic deformation under high temperatures and irradiation, directly influencing stress relaxation, dimensional changes of fuel pellets, and the overall integrity of the fuel rod. The temperature distribution within fuel pellets depends on several factors, primarily the heat generation rate and thermal conductivity. In addition, the radial power profile has a significant effect. In UO_2+Gd_2O_3 fuel pellets, the maximum heat generation is shifted toward the peripheral regions of the pellet. This behavior is caused by a substantial reduction in the neutron flux in the pellet center due to neutron absorption in the outer layers of the pellet [6].

The aim is to analyze and systematize available data regarding the influence of gadolinium on the physical, thermophysical, and mechanical properties of (U,Gd)O_2 fuel pellets, as well as to perform temperature calculations for linear heat generation rates in the range of 120…360 W/cm, taking into account the radial power distribution profile.

1. PHYSICAL PROPERTIES OF UO_2-Gd_2O_3 FUEL PELLETS

1.1. PHASE DIAGRAM OF THE UO_2-GD_2O_3 SYSTEM AND MELTING TEMPERATURE

The UO_2-Gd_2O_3 system is one of the most important systems in nuclear materials science, forming the basis for the fabrication technology and performance characteristics of fuel used in WWER, PWR, and BWR reactors.

The UO_2-Gd_2O_3 system contains the following phases: fluorite-type face-centered cubic (FCC) solid solutions, $F(U_{1-x}Gd_xO_{2\pm x})$, monoclinic Gd_2O_3 (B-type), and cubic Gd_2O_3 (C-type). A minimum melting temperature of approximately 2500 K is observed at a $GdO_{1.5}$ concentration of about 90% (Fig. 2) [1, 7–9]. The system is characterized by a wide compositional range of the fluorite solid solution $F(U_{1-x}Gd_xO_{2\pm x})$, extending up to approximately 70 mol.% $GdO_{1.5}$.

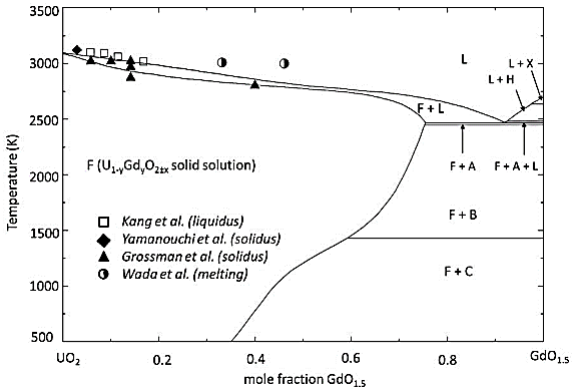


Fig. 2. Phase diagram for UO_2-Gd_2O_3 system [1, 10]

The principal model describing the formation of the $U_{1-x}Gd_xO_{2\pm x}$ solid solution assumes that U^{4+} ions are substituted by Gd^{3+} ions, resulting in the formation of one oxygen vacancy for every two U^{4+} ions replaced by Gd^{3+} ions. According to Kröger-Vink notation, the defect structure of the oxide solid solution with oxygen-vacancy charge compensation can be represented as:

$$Gd_2O_3 \rightarrow 2\,Gd'_U + V_{\ddot{O}} + 3\,O_O^x,$$

where Gd'_U – ion Gd^{3+} denotes a Gd^{3+} ion occupying a uranium lattice site and therefore possessing one less positive charge than U^{4+}; $V_{\ddot{O}}$ – oxygen vacancy; O_O^x – denotes an oxygen ion occupying a regular oxygen lattice site.

The melting temperature of pure UO_2 is approximately 2850 °C [1]. The addition of Gd_2O_3 results in a decrease in the melting temperature of the $U_{1-x}Gd_xO_{2\pm x}$ solid solution (Table 1) [10].

Table 1

Melting temperature of UO_2-Gd_2O_3 [10]

Composition	Solidus	Liquidus
UO_2-4 wt.%Gd_2O_3	2670±25	2820±20
UO_2-6 wt.%Gd_2O_3	2620±25	2818±20
UO_2-8 wt.%Gd_2O_3	2537±25	2783±20
UO_2-12 wt.%Gd_2O_3	2461±25	2739±20

The lattice parameters of $(U,Gd)O_2$ solid solutions are presented in Table 2 [1, 10–13].

Table 2

Lattice parameter of UO_2-Gd_2O_3 (X, wt.%)

a, nm	Preparing
0.5474…0.190 X	Mechanical blending, sintering in H_2, 1700 °C, 4 h [12]
0.5470…0.162 X	Mechanical blending, sintering in Ar, 1700 °C, 4 h [12]
0.54704…0.237 X	Chemical deposition, sintering in H_2, 1600 °C, 2 h [13]
0.54725…0.185 X	Mechanical blending, sintering in H_2, 1800 °C, 2 h [14]
0.54725…0.185 X	Mechanical blending, sintering in H_2, 1700 °C, 4 h [14]

The lattice parameter of $(U, Gd)O_2$ solid solutions decreases with increasing Gd_2O_3 content at a rate of approximately 0.0002 nm per mol.% Gd_2O_3, regardless of the pellet fabrication method (mechanical blending or co-precipitation). Studies [11, 12] have shown that this linear decrease in lattice parameter is maintained up to the composition $(U_{0.5}Gd_{0.5})O_2$. The correlation was obtained by linear regression of experimental data (Fig. 3). The resulting relationship can be expressed as: a=0.54636…0.01592 X, where a – lattice parameter (nm), X – molar content of Gd_2O_3.

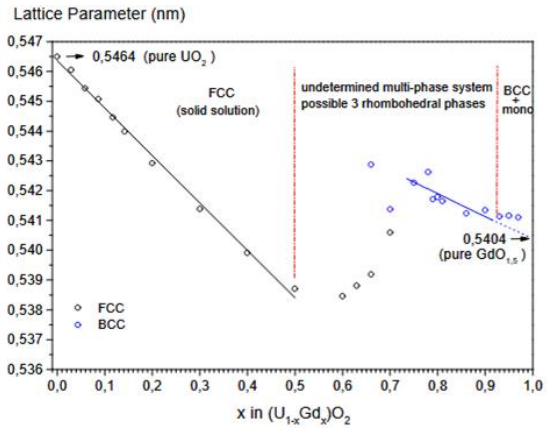


Fig. 3. Lattice parameter as a function of dissolved gadolinium content [11, 12]

1.2. THEORETICAL DENSITY

Numerous studies have shown that the theoretical density of UO_2-Gd_2O_3 solid solutions decreases with increasing Gd_2O_3 content (X, wt.%) (Table 3).

Table 3

Theoretical density of UO_2-Gd_2O_3 pellets

ρ_0, g/cm^3	Ref.
10.96…0.031 X	[15]
10.96…0.033 X	[13]
10.96…0.04 X	[1, 17]

1.3. THERMAL CONDUCTIVITY

Experimental investigations have demonstrated that increasing the gadolinium content leads to a reduction in the thermal conductivity of fuel pellets [16–19] (Fig. 4).

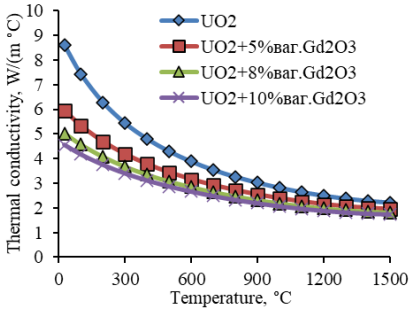


Fig. 4. Thermal conductivity for UO_2+Gd_2O_3 [6]

Reference [6] presents a correlation describing the thermal conductivity of UO_2-Gd_2O_3 fuel as a function of gadolinium content, temperature, pellet porosity, and fuel burnup.

$$\lambda = \left(\frac{1}{a + a_1 bu + a_2 Gd + b_1 bu\,T_p + b_2 Gd\,T_p + bT} + \frac{c}{T^2} e^{\frac{d}{T}} \right) (1-P)^{2.5},$$

where bu – burnup (MW·d/kgU); Gd – Gd_2O_3 content (wt.%); T – temperature, ºC; P – pellets porosity, a, b, c, d – constants:

$a = 0.0375$ $b = 2.165\times10^{-4}$

$a_1 = 0.38360\times10^{-2}$ $b_1 = -0.90849\times10^{-6}$

$a_2 = 0.84476\times10^{-2}$ $b_2 = 0.22149\times10^{-5}$

$c = 4.715\times10^{9}$

$d = 16361.$

1.4. THERMAL EXPANSION

The thermal expansion of UO_2 and UO_2–Gd_2O_3 fuel pellets can be described by the following expression:

$$dL/L_0 = A + BT + CT^2.$$

Table 4 summarizes the values of the constants A, B, and C used in the linear thermal expansion equation (dL/L_0) for uranium – gadolinium fuel pellets fabricated by mechanical blending followed by sintering in a hydrogen atmosphere at 1720 or 1750 °C. The investigated compositions contained 0, 5, 8, and 10 wt.% Gd_2O_3.

Table 4

Constants in the thermal expansion equation [1, 18]

Content Gd_2O_3, wt.%	A, 10^{-3}	B, 10^{-6}	C, 10^{-9}
0	-2.238	7.165	2.095
5	-2.314	7.358	2.156
8	-2.391	7.433	2.278
10	-2.284	7.162	2.430

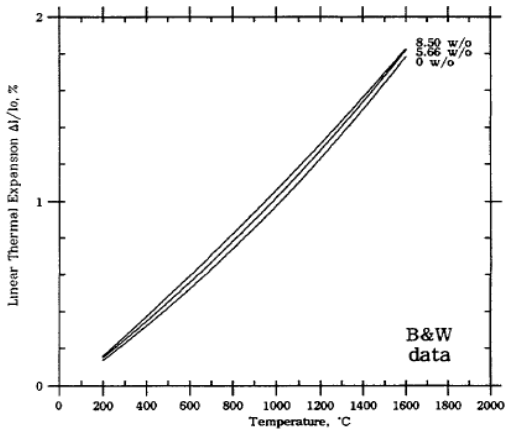


Fig. 5. Thermal expansion of UO_2+Gd_2O_3 pellets [1, 18]

The results indicate that the coefficient of linear thermal expansion increases with increasing Gd_2O_3 content (Fig. 5) [1, 18].

At 1973 K, the linear thermal expansion of UO_2-5 wt.% Gd_2O_3, UO_2-8 wt.% Gd_2O_3, and UO_2-10 wt.%Gd_2O_3 was approximately 2.7, 5.4, and 6.2% respectively, than that of pure UO_2 pellets. TECDOC-1496 provides an empirical correlation for calculating the thermal expansion of fuel containing up to 10 wt.% Gd_2O_3 [19].

$$L = L_{273}(a + bT + cT^2)$$

$$a = 9.9866\times10^{-1} \qquad b = 7.2512\times10^{-6}$$

$$c = 2.0463\times10^{-13}\,g^2 + 3.4846\times10^{-11}\,g + 2.0653\times10^{-9}$$

g: Gd_2O_3 weight percent ,

where L та L_{273} – sample height at T, K and 298 K respectively. In contrast, the authors of Ref. [20] reported that thermal expansion decreases with increasing gadolinium oxide content.

1.5. MECHANICAL CHARACTERISTICS

The results of high-temperature creep investigations of UO_2 and (U,Gd)O_2 pellets with a density of approximately 95% of theoretical density and an average grain size of approximately 10 μm are reported in Refs. [1, 21–23]. Selected results of high-temperature creep testing of (U,Gd)O_2 pellets are presented in Figs. 6 and 7.

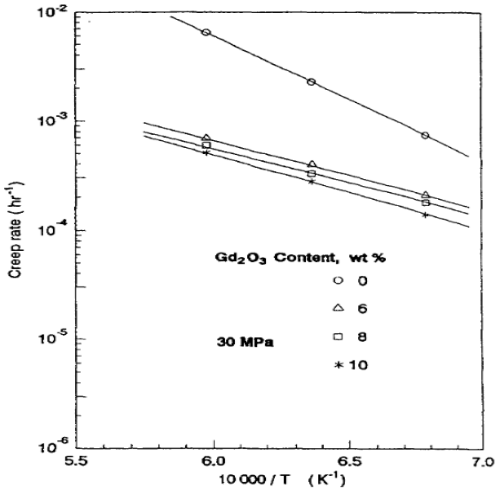


Fig. 6. Temperature dependence of the creep rate of UO_2 and (U,Gd)O_2 fuel pellets [1, 21]

The studies revealed a tendency toward a slight decrease in the creep rate of (U, Gd)O_2 pellets with increasing Gd_2O_3 content. The dependence of the creep rate of (U, Gd)O_2 pellets on the gadolinium oxide concentration X (wt.%) can be expressed as follows:

$$\log \dot{\varepsilon} = \log \dot{\varepsilon}(UO_2) - 0.2X \text{ [21]},$$

where $\dot{\varepsilon}$ is the creep rate and X is the Gd_2O_3 concentration in wt.%.

Correlations describing the dependence of Young's modulus of UO_2 fuel pellets on pellet porosity, fuel burnup, and temperature are presented in Ref. [23].

$$E = 223.7(1 - 2.6\,p)\left[1 - 1.394\cdot10^{-4}(T - 20)\right]\{1 - 0.1506\,[1 - \exp(-0.035\,bu)]\},$$

where p – porosity; T – temperature, °C; bu – burnup, GW d/t h.a.

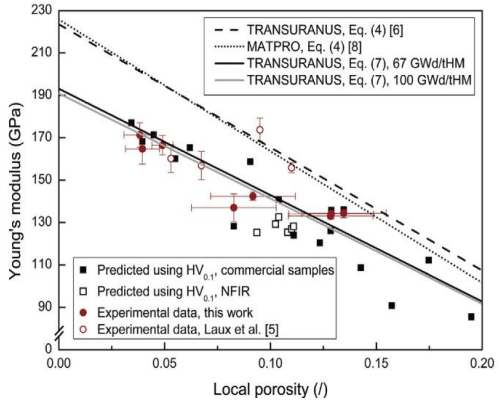


Fig. 7. Young's modulus of UO_2 pellets as a function of porosity [23]

Fig. 8 shows the results of a comparative analysis of the Young's modulus for UO_2 and UO_2-Gd_2O_3 pellets with varying Gd_2O_3 contents [1, 22]. Dependence of the Young's modulus and Poisson's ratio in UO_2-Gd_2O_3 pellets:

$$E/E_o = 1 - 5.686\cdot10^{-3}\,C_{Gd},$$

$$\nu/\nu_0 = 1 - 6.59\cdot10^{-3}\,C_{Gd}.$$

E_0 and ν_0 are the Young's modulus and Poisson's ratio of pure UO_2, C_{Gd} is the Gd_2O_3 content (wt.%).

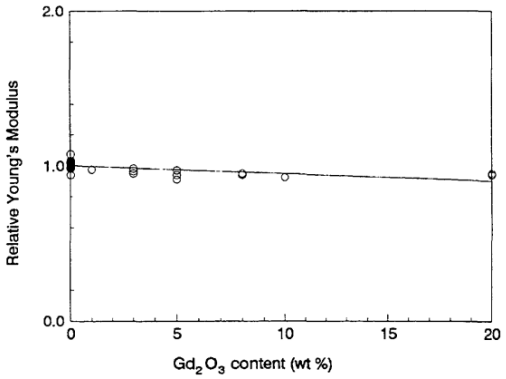


Fig. 8. Young's modulus of UO_2-Gd_2O_3 pellets [1, 22]

2. GADOLINIUM ISOTOPES BURNUP

Burnup calculations of the ^{155}Gd and ^{157}Gd isotopes under WWER-1000 reactor operating conditions were performed using the MCNPX code. The initial fuel enrichment was assumed to be 3.6 wt.% ^{235}U. The initial concentrations of gadolinium oxide were 5 wt.% Gd_2O_3 and 8 wt.% Gd_2O_3.

The calculations showed a very similar trend in the decrease of ^{155}Gd and ^{157}Gd concentrations with increasing fuel burnup for both initial Gd_2O_3 concentrations. At a burnup of approximately 7000 (MW·d)/tU, almost all ^{157}Gd is depleted (Fig. 9). At a burnup of approximately 13000 (MW·d)/tU, almost all ^{155}Gd is depleted.

Fig. 10 presents the radial distribution of gadolinium isotope concentrations at various fuel burnup levels (BU) for initial concentration 5 wt.% Gd_2O_3.

Due to these high absorption cross sections, $(U,Gd)O_2$ "burns" gadolinium practically at the fuel surface, while the interior of the fuel is completely shielded from thermal neutrons. This effect leads to a gradual shift over time of the interface between burned-out and unburned gadolinium from the surface of the fuel pellet toward its center (see Fig. 9).

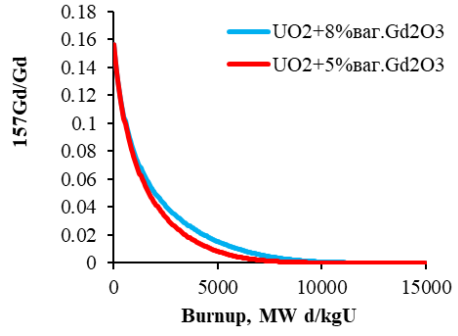


Fig. 9. Burnup of gadolinium isotopes ^{157}Gd (a) and ^{155}Gd (b)

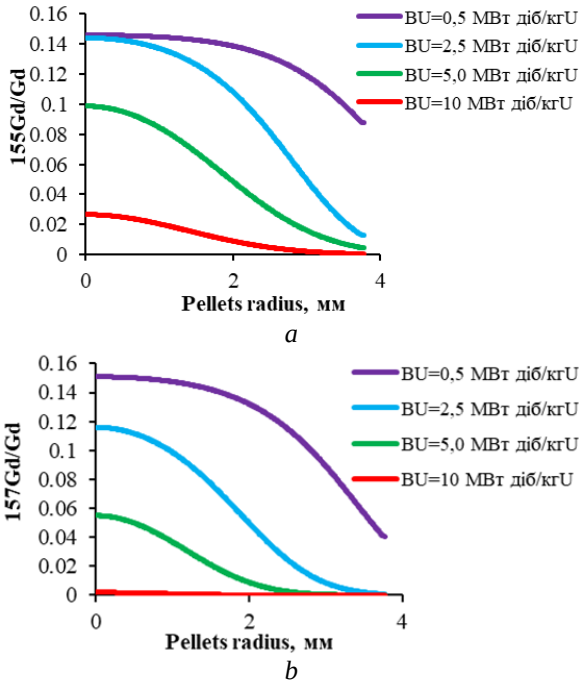


Fig. 10. Radial distribution of ^{155}Gd (a) and ^{157}Gd (b) concentrations in fuel pellets

3. FUEL CENTER TEMPERATURE

Comparative calculations of temperature distributions in UO_2 and UO_2-Gd_2O_3 fuel pellets were performed using the HT_FuelRod code, which was developed for thermal analyses of fuel rods in WWER and PWR reactors. The code employs the finite element method for solving the heat conduction equation [24].

The following assumptions were adopted in the calculations:

1. Fuel pellets were assumed to be coaxially aligned. Thermal expansion, swelling, and densification of the pellets were neglected.
2. Fuel cladding deformation was not considered.

The input parameters used in the calculations were:

1. Linear heat rate in the range {120, 180, 240, 300, 360} W/cm.
2. Coolant temperature: 310 °C.
3. Pellet inner diameter: 0 mm.
4. Pellet outer diameter: 7.55 mm.
5. Cladding inner diameter: 7.77 mm.
6. Cladding outer diameter: 9.10 mm.
7. Heat transfer coefficient from the fuel rod cladding to the coolant: 0.05 W/(mm²·°C).
8. Reactor coolant flow rate: 82,000 m³/h.
9. Fuel rod pitch: 1.275 cm.
10. Thermal conductivity model accounting for the Gd_2O_3 content (Equation 2).
11. Fuel pellet porosity: 0.05 (5%).
12. Gd_2O_3 content: {0, 5, 8} wt.%.
13. The dimensionless parameter b characterizes the radial non-uniformity of heat generation within the fuel pellets and may vary from 0 (uniform distribution) to infinity (highly non-uniform distribution). The parameter b defines the heat generation profile F(r) through a quadratic dependence:

Unlike conventional UO_2 fuel, where the radial heat generation profile is relatively uniform, UO_2-Gd_2O_3 fuel exhibits a significantly non-uniform distribution of local volumetric heat generation [6]. The primary reason for

this behavior is the exceptionally large thermal neutron absorption cross-sections of the gadolinium isotopes ^{155}Gd and ^{157}Gd. Due to neutron absorption in the peripheral region of the pellets, the fission rate in the central region decreases, resulting in a shift of heat generation toward the pellet periphery (Fig. 11) [6].

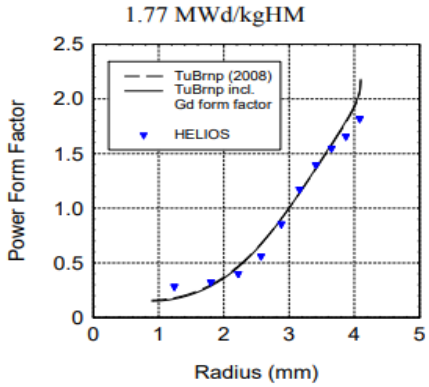


Fig. 11. Radial power distribution profile in UO_2-Gd_2O_3 [6]

As fuel burnup increases, gadolinium isotopes are gradually depleted. Consequently, the spatial heat generation profile evolves with time, becoming more uniform and approaching the distribution characteristic of conventional UO_2 fuel.

Heat generation profiles F(r) were calculated for two values of the parameter b:

b = 9.5, corresponding to a strongly non-uniform radial heat generation profile representative of fresh UO_2-Gd_2O_3 fuel at the beginning of life, and

b = 0.026, corresponding to an almost uniform radial heat generation profile representative of the state after nearly complete depletion of ^{155}Gd and ^{157}Gd, when the heat generation profiles of UO_2 and UO_2-Gd_2O_3 fuels become practically identical (Fig. 12).

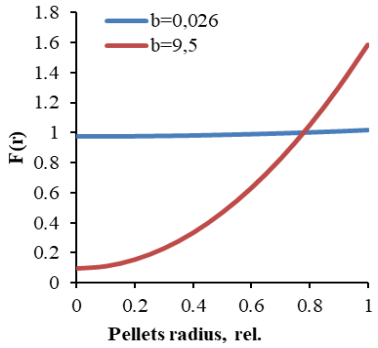


Fig. 12. Radial power distribution profile

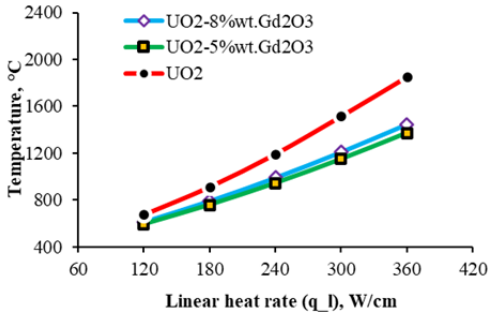


Fig. 13. The relationship between the fuel center temperature and heat release (for UO_2 b = 0.026, for UO_2+Gd_2O_3, b = 9.5)

Calculations have shown that in gadolinium-containing fuel pellets, where the energy release profile is significantly non-uniform across the radius at the start of the fuel cycle (parameter b = 9.5), the temperature at the center of the pellets is slightly lower than the temperature in UO_2 pellets with more uniform energy release (parameter b = 0.026) (Fig. 13).

Once the energy release profiles in UO_2 and UO_2+Gd_2O_3 pellets become identical – which occurs after the burn-up of the gadolinium isotopes ^{155}Gd and ^{157}Gd – the temperature at the center of the pellets containing Gd will be higher than that of the UO_2 pellets. This is due to the lower thermal conductivity of UO_2+Gd_2O_3fuel pellets (Fig. 14).

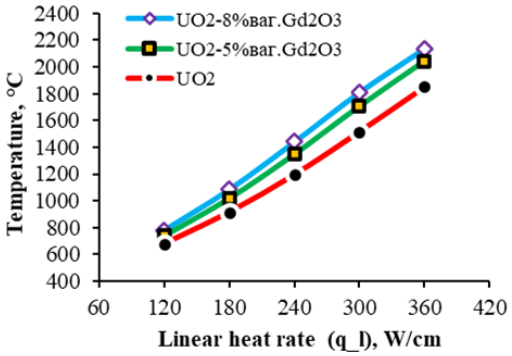


Fig. 14. The relationship between the fuel center temperature and heat release (for UO_2 and UO_2+Gd_2O_3, b = 0.026)

CONCLUSIONS

1. A comprehensive analysis and systematization of published data on the influence of gadolinium content on the most important properties of fuel pellets (physical, thermophysical, and mechanical) were carried out to establish a database of the properties and characteristics of UO_2–Gd_2O_3 fuel materials.

2. Burnup calculations of gadolinium isotopes were performed for UO_2+5 wt.% Gd_2O_3 and UO_2+8 wt.% Gd_2O_3 fuel compositions. The calculations demonstrated a similar depletion behavior of ^{155}Gd and ^{157}Gd for both fuel compositions, indicating comparable radial power distribution profiles for these initial gadolinium concentrations.

3. Temperature calculations at the center of fuel pellets with different initial Gd_2O_3 contents were performed. The results show that, at the beginning of the fuel cycle, the effect of the non-uniform radial power distribution in UO_2-Gd_2O_3 fuel has a greater influence on the pellet centerline temperature than the reduction in thermal conductivity caused by the addition of gadolinium. At the beginning of the fuel cycle, the centerline temperature of Gd-containing fuel pellets may therefore be lower than that of conventional UO_2 fuel due to the predominant heat generation in the outer regions of the pellet. After the complete depletion of ^{155}Gd and ^{157}Gd, the radial power distribution in UO_2-Gd_2O_3 fuel approaches that of conventional UO_2 fuel. Under these conditions, the lower thermal conductivity of the gadolinium-bearing fuel becomes the dominant factor, resulting in a higher pellet centerline temperature compared with conventional UO_2 fuel.

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ВПЛИВ ГАДОЛІНІЮ НА ВЛАСТИВОСІ ПАЛИВНИХ ТАБЛЕТОК (U, Gd)O_2

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Представлено аналіз та систематизацію опублікованих даних щодо впливу вмісту гадолінію на властивості паливних таблеток (U, Gd)O_2, зокрема на температуру плавлення, термофізичні та механічні властивості. Було проведено порівняльні розрахунки вигоряння ізотопів ^{155}Gd та ^{157}Gd для умов експлуатації реактора ВВЕР-1000 для палива, що містить 5 та 8 мас.% Gd_2O_3. З урахуванням впливу Gd_2O_3 на теплопровідність палива та радіальний розподіл потужності було розраховано температури по осі таблетки для традиційного палива UO_2 та палива (U, Gd)O_2 при лінійних показниках теплоутворення в діапазоні 120…360 Вт/см. Розрахунки демонструють конкуруючі ефекти радіального перерозподілу потужності та зниження теплопровідності на динаміку температури палива, що містить гадоліній, протягом паливного циклу.