Analysis of Experimental Data on Gas Release and Swelling of UN Fuel Irradiated in BR-10 Reactor

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Abstract. Uranium mononitride fuel was used in the fourth and fifth BR-10 reactor core loadings. The total number of irradiated fuel pins was equal to 1250 (660 and 590 fuel pins in 4-th and 5-th fuel loadings respectively). Most fuel pins have been irradiated up to design burn-up (8 at%) without cladding failure. In addition to driver subassemblies (SA), some experimental SAs have been irradiated in BR-10 reactor. The post irradiation examination (PIE) of 8 driver and 3 experimental SAs was done in the IPPE hotlab. The paper presents the results of study of fission gas release and nitride fuel swelling. These two phenomena have a significant impact on cladding stress strain state and therefore on the fuel life time. Substantial fission gas release from BR-10 nitride fuel starts at a burn-up of more than 3 at%. Both irradiation temperature increase and fuel density decrease lead to gas release rate increase. $N^{14}(n,\alpha)B^{11}$ nuclear reaction in nitride fuel causes the formation of quite big amounts of helium. This fact should be taken into account in computer codes used for nitride irradiation behavior modeling. The paper presents the measured nitride swelling rate values at the temperature range from 760 to 1200 ^{o}C. Increase of fuel temperature leads to increase of nitride swelling rate.

Key Words: Uranium mononitride fuel, fission gas release, swelling.

1. Introduction

According to the Federal Target Program "Nuclear Power Technologies of the New Generation in 2010-2015 and until 2020" the commissioning of lead-cooled BREST-OD-300 fast reactor is planned [1]. The technical design of the reactor core involves the use of mixed uranium-plutonium nitride. One of the main tasks of the reactor project is the development and proof of operability of fuel pins with such fuel [2].

An important source of the information on the (UPu)N pins performance is the operation experience of the BR-10 reactor. From 1983 to 2002 in the fourth and fifth fuel reactor loadings the UN fuel has been used. The total number of irradiated fuel pins was equal to 1250 (660 in the fourth and 590 in the fifth fuel loadings). Most of the fuel pins reached the goal burn-up (8 at%) without integrity loss [3]. In the fourth core loading two cases of the cladding gas failure were observed. In the fifth core loading one case of the fuel pin integrity loss occurred at the burn-up of 6.3 at %. In other 23 cases the fuel pins failed after exceeding the goal burn-up. In one case a fuel-coolant contact was fixed. These results suggest a fairly reliable operation of fuel pins with UN fuel up to 8 at%.

In addition to driver subassemblies (SA) several experimental subassemblies with nitride fuel were irradiated in the BR-10 reactor. Eight driver and three experimental SAs were examined in the IPPE hotlab. Some data obtained on the performance of driver SAs with nitride fuel has been presented previously [4,5].

To verify the fuel calculation codes, in particular, the DRAKON code, a representative set of experimental data is required. The first stage of the DRAKON code verification was done using the data of post-irradiation examination of fuel pins with mixed uranium-plutonium nitride fuel irradiated in the BOR-60 reactor within the framework of BORA-BORA experiment [6, 7]. To provide the code with data required for the next stage of verification, an analysis of all available BR-10 reactor data on nitride fuel was carried out. This article presents the investigation results on the gas release and fuel swelling in driver and experimental SAs of BR-10 reactor. These two phenomena determine primarily the level of stresses in fuel pin claddings and hence their life-time.

2. The design and operation conditions of fuel pins

In the IPPE hotlab 8 driver SAs and 3 experimental SAs (K-1, K-3, UN-2) with UN fuel irradiated in the BR-10 reactor core were examined (Table 1). Uranium mononitride in driver SAs and experimental K-3 SA was produced by the method of carbothermal reduction of uranium dioxide in the pure nitrogen environment. The density of fuel pellets and its chemical composition are shown in Table 2. The pellets in fuel pins of UN-2 and K-1 subassemblies were made of metallic uranium.

TABLE I: IRRADIATION CONDITIONS OF SUBASSEMBLIES WITH UN FUEL.

SA Number	Time of irradiation	Maximum burnup, at %.	Maximum fluence, 10^{26} n/m^2 (E>0)	Maximum dose, dpa
UN-2	22.06.76–22.11.78	4.3	3.10	18.6
K-3	05.11.74–13.11.79	7.6	4.38	26.5
K-1	04.01.74–03.11.79	7.6	4.54	27.5
N-017	12.05.83–12.10.85	3.4	2.00	11.0
N-019	12.05.83–03.02.87	4.8	3.50	19.0
N-009	24.05.83–31.08.89	5.7	4.31	20.2
N-118	12.05.83–30.10.89	5.8	4.55	25.0
N-095	10.05.83–22.03.88	6.3	5.07	28.0
N-021	12.05.83–10.08.89	7.6	5.90	33.0
N-094	10.05.83–22.07.89	8.2	6.55	36.0
AB-60	24.07.90–24.04.99	8.4	6.25	34.0

Fuel pins with UN fuel had claddings of 8.4 mm in diameter with wall thickness of 0.4 mm and length of 595 mm made of 16Cr -15Ni-3Mo-B (EI-847) austenitic stainless steel. The core length was equal to 400 mm (Fig. 1). Above the fuel column a reflector was mounted in the form of the cylinder of 7.3 mm in diameter and 80 mm long made of the EI-847 steel. In the upper gas plenum of 100.4 mm in height a spring was located. To the lower and upper cladding ends an end tip and a plug respectively were welded by argon-arc. The fuel pin total length was equal to 615 mm. Before the final sealing-in the pin internal volume was evacuated and then was filled with helium of 0.5 MPa pressure. Fuel pins are spaced with the step of 150 mm by the wire of the elliptical cross-section 1.3 mm×0.4 mm made of the EI-847 steel.

TABLE 2: CHARACTERISTICS OF UN FUEL IN DRIVER SA AND IN EXPERIMENTAL K-3 SA

Parameter	Specifications	Driver SA	K-3 SA
Chemical composition, mass. %			
uranium	≥ 93.5	93.5−94.3	94.4−94.5
nitrogen	5.0 ± 0.5	4.5−5.2	4.7−4.8
carbon	≤ 0.7	0.05−0,5	0.3
oxygen	-	0.2−0.94	0.4−0.6
tungsten	≤ 0.5	≤ 0.5	-
Density, g/cm^3	≥ 11.5	12.8−13.4	12.1±0,1 12.5±0,1

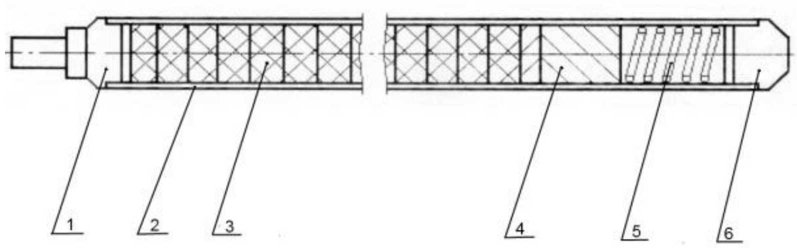

FIG. 1. The BR-10 driver fuel pin design: 1 – lower tip; 2 -cladding; 3 – UN fuel; 4 - reflector; 5 - spring; 6 - plug.

The design of the fuel pins in driver and experimental subassemblies was slightly different (Table. 3). The fuel temperature in the experimental K-1 and K-3 subassemblies was significantly higher as compared to others (Table 4). The fuel temperature was calculated by the DRAKON code.

TABLE 3: CHARACTERISTICS OF UN FUEL PINS OF DRIVER AND EXPERIMENTAL SUBASSEMBLIES

<table>
<tr><td rowspan="3">Parameter</td><td rowspan="3">Driver SA</td><td rowspan="3">К-1, К-3</td><td colspan="2">UN-2</td></tr>
<tr><td colspan="2">bounding</td></tr>
<tr><td>sodium</td><td>gas</td></tr>
<tr><td>Diameter and wall thickness of cladding, mm</td><td>8.4×0.4</td><td>8.3×0.4</td><td colspan="2">8.65×0.32</td></tr>
<tr><td>Length of fuel column, mm</td><td>400</td><td>320</td><td colspan="2">320</td></tr>
<tr><td>Diameter of fuel pellets, mm</td><td>7.4 ± 0.03</td><td>6.94–7.01</td><td colspan="2">7.82–7.85 – driver
6.9–7.28 – control</td></tr>
<tr><td>Pellet density, % of theoretical</td><td>89.4-93.6</td><td>89.5 – К-1
91.55 – К-3</td><td colspan="2">91–96 − driver
80–98 – control</td></tr>
<tr><td>Mean radial “fuel-cladding” gap, mm</td><td>0.1</td><td>0.26</td><td colspan="2">0.08 − driver
0.36–0.55 – control</td></tr>
<tr><td>Gap environment (gas pressure in pin, MPa)</td><td>helium
(0.5)</td><td>helium
(0.1)</td><td>sodium</td><td>argon-air mixture</td></tr>
<tr><td>Length of gas plenum, mm</td><td>100,4</td><td>80</td><td>~ 110</td><td>~ 60</td></tr>
<tr><td>Fuel pin gas volume, cm^3</td><td>5.2</td><td>5.3</td><td>5.5</td><td>-</td></tr>
</table>

TABLE 4: FUEL PIN OPERATION CONDITIONS IN DRIVER AND EXPERIMENTAL SUBASSEMBLIES

Parameter	Driver SA	UN-2	К-1, К-3
Sodium inlet temperature, °C	325–350	350	360
Maximum sodium heating, ºC	165	178	120
Maximum linear power, W/cm	450	500	400
Maximum fuel temperature at BOL, ºC	1200	840 (sodium) 1260 (gas)	1630−1640
Average fuel temperature at BOL, ºC	1015	640 (sodium) 1050 (gas)	1393−1398

3. The amount and chemical composition of plenum gas

The amount of gas fission products in the pin was determined by puncturing of the cladding in a preliminary evacuated device analytical volume and then by measuring the pressure of the gas released. An accuracy of gas amount measurements was ≤ 1.5%. The gas composition was studied by the radiochromotography method with an accuracy of ± 8%. All examined fuel pins have been tight with the exception of one from AB-60 SA and two from UN-2 SA.

It is seen from Fig. 2 that at the fuel burn-up less than 3 at % the gas amount under pin cladding varies insignificantly and is determined only by the helium release. At the fuel burn-up of (3-4) at% the volumetric part of helium in the pin was equal to (43-50) %, at the higher burn-up the volumetric helium part decreased up to (10-30) % due to krypton and xenon release.

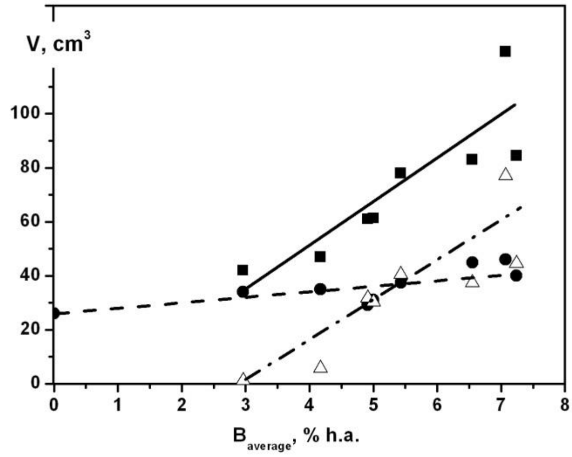

FIG. 2. The gas amount in driver fuel pins of the BR-10 reactor versus average nitride burn-up: ■ - total gas amount; Δ - krypton and xenon; • - helium.

The amount of Kr and Xe released into the nitride fuel pins of 10 SAs after irradiation in the BR-10 versus average fuel burn-up is shown in Fig. 3. One can see that at burn-up of 3 at% the gaseous fission products release is negligible. In the investigated N-017 SA with minimal burn-up value only 1.1 cm^3 of the gas in average was released from the fuel, that is equal to 0.8% of the total amount calculated.

Comparing the total release of krypton and xenon in driver and experimental subassemblies one could note the following: for gas-bonded fuel pins of the experimental UN-2 subassembly this release is twice higher as compared to sodium-bonded pins gas release that is due to higher fuel temperature in the gas-bonded pin. At the average fuel temperature of 1015°C the Kr and Xe release under the cladding in driver subassemblies is 1.7−3 times lower as compared to the experimental K-1 and K-3 subassemblies irradiated at average temperature of 1395°C, that appear to be also due to the higher temperature of fuel in the pin of the experimental subassemblies.

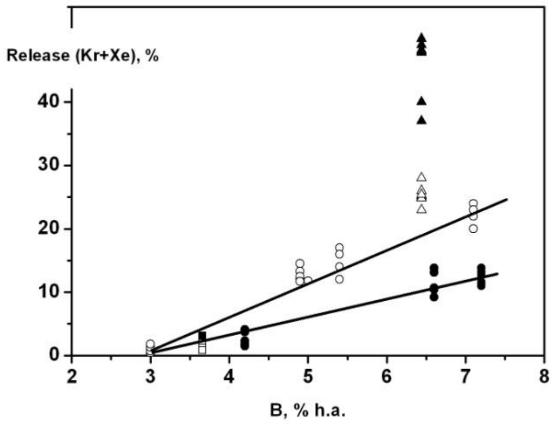

FIG. 3. The total amount of krypton and xenon released from uranium nitride as function of burn-up in: driver N-017, N-019, N-021 subassemblies (●); N-009, N-118, N-095, N-094 (○); experimental K-1 (Δ), K-3 subassemblies (▲), UN-2 with fuel-cladding gap filled by sodium (□) and gas (■)

The release of Kr and Xe in the K-3 SA was nearly twice higher as compared with pins of K-1 SA, despite of similar irradiation temperatures and fuel burn-ups. One could think that this difference is due to the technological peculiarities of manufacturing fuel pellets. The pellets in fuel pins of K-1 SA were made of metallic materials, but the fuel pins of K-3 SA were fabricated using the carbothermic reduction of UO_2. Unfortunately, the lack of data on the of fuel pellet production for fuel pins of subassembly K-1 does not allow such a clear conclusion.

Fig. 4 shows the burn-up dependence of gaseous fission products release from the nitride fuel irradiated in the BR-10 reactor together with the data obtained in other research reactors [8]. It is seen that the gas release rate increases with increasing of nitride irradiation temperature, the incubation period is reduced from 3 at% at the irradiation temperature of 1015ºC to 0.3 at% at the temperature of 1400ºC.

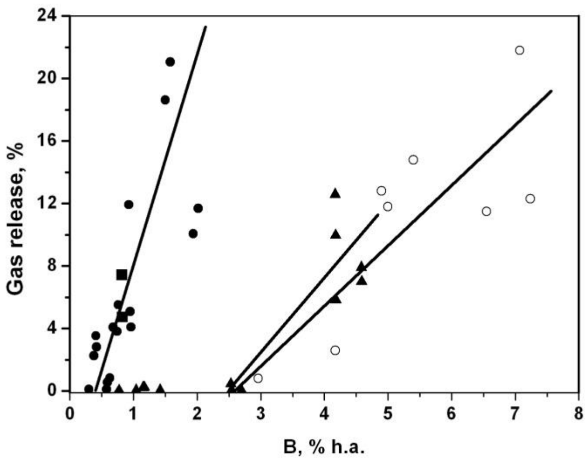

FIG. 4. The burn-up dependence of gas release from nitride fuel irradiated at temperature of 1015°C in BR-10 reactor(○), at 1190°C in test reactor according to the ORNL'-4461 USA Program (▲), at 1400°C in test reactor according to the PWAC-488 Program (●), at 1680°C in EBR-II reactor (■)

4. The fuel swelling

The nitride fuel density before and after irradiation was determined by hydrostatic weighing with an accuracy of ± 0.5 %. The samples were weighed in air and in the working fluid (CCl_4) after soaking for two hours. As a rule, for each subassembly the fuel density was measured in samples cut from three sections along the fuel pin height (at top, bottom and core mid-plane). The density of the control samples manufactured by the same technology as the fuel of investigated pins was also measured. Using data on the density change, the swelling and the swelling rate were calculated.

It is seen from Fig. 5, that the UN swelling increases linearly with burn-up, the swelling rates of fuel pellets in driver and experimental subassemblies differ significantly, that may be caused by differences in both fuel pin irradiation conditions and its fabrication technology. So, the swelling rate of fuel pellets from the central part of the fuel column in driver subassemblies (T_{irr} = 1200°C) is equal to 1.65 % per 1 at% and in the samples from the upper and lower parts of fuel column (T_{irr} = 760-900ºC) is equal to 1.35% per 1 at%. The swelling rate of the fuel pellets in the gas-bonded pins of the experimental UN-2 SA (T_{irr} = 1260°C) is more than two times higher than the swelling rate of fuel pellets in the sodium-bonded ones (T_{irr} = 840°C). The fuel swelling rate in the experimental K-1 SA is 2 % per 1 at%, in K-3 SA is 2.6 % per 1 at% despite the fuel temperatures in these subassemblies were approximately the same. Perhaps, this difference is due to the technological peculiarities of the fuel manufacture. Fuel pellets in K-3 SA were produced by carbothermic reduction of uranium dioxide and in K-1 SA by hydrogenation-nitriding of metallic uranium.

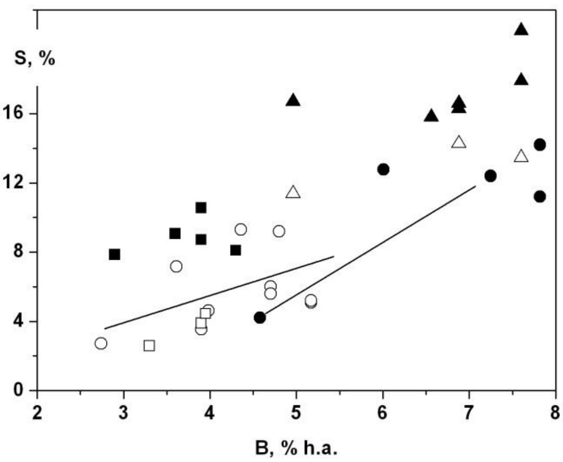

FIG. 5. The burn-up dependence of nitride fuel swelling in driver and experimental subassemblies of BR-10 reactor: ○ - upper and lower cross-sections of driver fuel pins; ● - central cross-section of driver fuel pins; Δ - fuel pins of K-1 SA; ▲ - fuel pins of K-3 SA, □ – sodium-bonded pins of UN-2 SA; (■) – gas-bonded pins of UN-2 SA

Fig. 6 shows the burn-up dependence of UN swelling in fuel pins of BR-10 driver subassemblies (T_{irr} = 760-1200ºC) together with data on UN at higher temperatures of 1190-1680ºC [8]. It is seen that UN swelling rate increases with irradiation temperature increase.

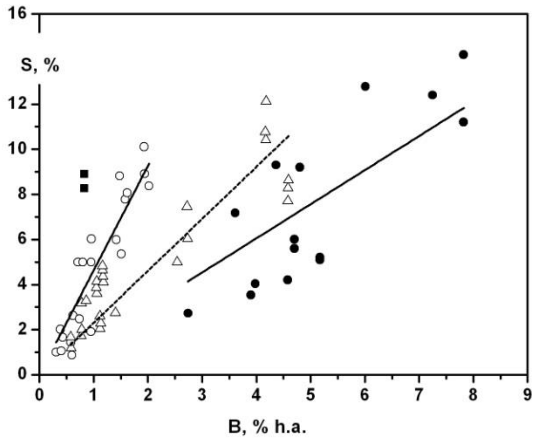

FIG. 6. The burn-up dependence of nitride fuel swelling after irradiation at temperatures of 760-1200°C in BR-10 reactor (○), at 1190°C in test reactor according to the ORNL- 4461 USA Program (▲), at 1400°C in test reactor according to the PWAC-488 Program (●), at 1680°C in EBR-II reactor (■)

Fig. 7 shows dependence of UN swelling rate on the irradiation temperature in the range of 720-1680°C. It is seen from Fig. 7 that with the fuel temperature increase the swelling rate also increases. In studies of the nitride fuel irradiated in EBR-II reactor and in several research reactors with an intermediate neutron spectrum it was established that with fuel temperature increase from 1200 to 1680°C the fuel swelling rate increased from 2.3 % to 10.3% per 1 at% [8].

In Fig. 7 the dashed line shows the temperature dependence of the swelling rate of nitride fuel irradiated in BR-10, EBR-II reactors and other research reactors. This dependence can be described by the following equation

$$S / B = 3.5 - 0.0066T + 4.69 \times 10^{-6} T^2$$

where S/B is the swelling per burn-up of 1 at %; T is the temperature in °C.

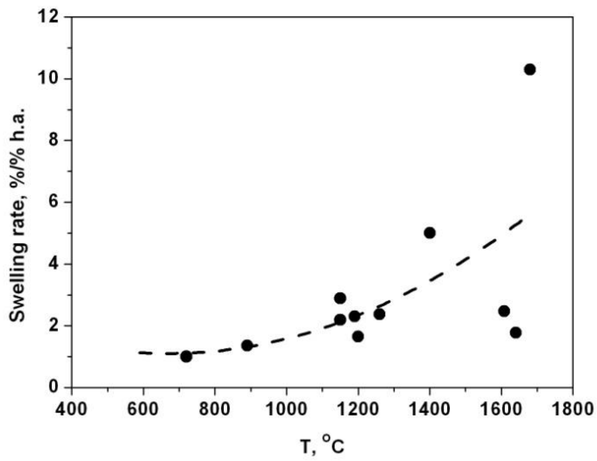

FIG. 7. The temperature dependence of UN swelling rate

5. Discussion

It is interesting to compare the results of this work with the data on the gas release from uranium oxide fuel, as well as with the data for mixed nitride. Fig. 8 presents the data on the output of gaseous fission products in the driver nitride subassemblies of the BR-10 reactor together with similar data for uranium dioxide [9]. It is seen that at the same temperature the release of gaseous fission products from dioxide starts much earlier, and the maximum release value is much higher.

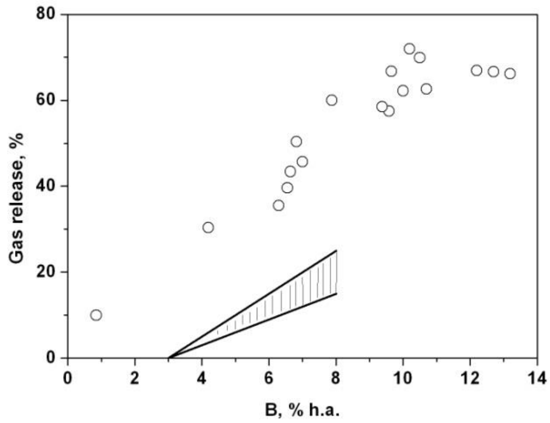

FIG. 8. *The release of krypton and xenon as a function of burn-up:* o *–for uranium dioxide [9] ||| - for uranium mononitride in driver subassemblies of the BR-10 reactor*

A comparison of the BR-10 data with ones for the release of gaseous fission products from $U_{0.55}Pu_{0.45}N$ and $U_{0.4}Pu_{0.6}N$ after irradiation in the BOR-60 reactor demonstrates their quite good agreement [7].

In general, the results of this study confirm the earlier findings indicating that gas release rate from nitride increases with irradiation temperature increase and pellet density decrease [9, 10]. In this work, this conclusion follows from the comparison of fuel pins of UN-2 SA with gas and sodium bounding, as well as from the comparison of driver subassemblies and experimental K-3 SA. In spite of this, for K-3 SA the difference is particularly large since in this case just two factors are important: high temperature and low pellet density.

It is seen from Fig. 2 that the volume fraction of helium in driver fuel pins of the BR-10 reactor is very high. During the manufacture these fuel pins were filled with helium at pressure of 0.5 MPa, i.e. at the normal pressure fuel pin contains 26 cm^3 of helium. Depending on the fuel burn-up the fission of U^{235} results in formation of additional 1.4–3.6 cm^3 of helium. The rest amount of helium from 6.6 to 16.5 cm^3 is generated by $N^{14}(n,\alpha)B^{11}$ reaction. At burn-up of 7 at % helium share is more than 20 % of the total fission gas output. An increased release of helium from nitride fuel is confirmed also by mixed nitride fuel irradiations in BOR-60 and Phenix reactors [7, 11].

6. Conclusion

A significant release of Kr and Xe from the nitride fuel in the BR-10 reactor starts at burn-ups ≥ 3 at %. Both an increase of the irradiation temperature and a decrease of the fuel pellet density results in increase of gas release rate. The $N^{14}(n,\alpha)B^{11}$ nuclear reaction results to appreciable amounts of helium production in nitride fuel that should be taken into account in computer codes used for nitride irradiation behavior modeling. Nitride swelling rates obtained in this work are in the range from 1.35 to 1.65% per 1 at% of burn-up at irradiation temperatures in the range of (760-1200)°C. At higher irradiation temperature the swelling rate is higher.

7. References

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