

SECTION 2  
**THERMAL AND FAST REACTOR MATERIALS**  
**CARBON-GRAPHITE MATERIALS IN NUCLEAR-POWER**  
**ENGINEERING**  
**(REVIEW)**

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The review presents the data concerning carbon-graphite materials and their extensive application as structural materials for making various devices and elements of nuclear reactors. Carbon-graphite materials are widely used in more than 100 power nuclear reactors due to their availability, satisfactory moderating properties and low neutron capture cross-section. In more detail the problems involving the use of carbon-graphite materials as moderators and neutron reflectors in the nuclear reactors are considered. Also, different-type reactor stacks are reviewed. Particular attention is devoted to the physical and mechanical properties of carbon-graphite materials, as well as, to the changes in the graphite radiation dimensional stability and resistance as a result of graphite-neutron flux interaction

PACS 62.19:621039.52

### INTRODUCTION

Continuous increase of power production and consumption establishes high requirements to developing new methods and upgrading old methods for electrical energy generation. At the same time a period of hydrocarbon fuel deficit is coming. Thermal power station and heat-electric generating plants turn to coal instead of petroleum residue (masut) and natural gas that leads to the significant environmental contamination and to the rise in the cost of energy production.

In this connection, nuclear power plants get great advantage over thermal power stations. Since the latter half of the fifties in the XX century the nuclear power engineering was rapidly developing and its role in the common energy balance of industrially advanced countries was constantly increasing.

It is expected that in the Ukraine the quantity of electricity generated at NPP's should increase and reach more than 50% of the total produced electric power.

At present the NPP part in the national power production is 47.1% [1]. During the last decades the nuclear power production at NPP is more advantageous economically than at thermal power stations and makes 0.4 euro percent/(kW-h) [2], in addition, the CO<sub>2</sub> discharge into the atmosphere decreases.

### USE OF CARBON-GRAPHITE MATERIALS IN THE REACTOR CORE STRUCTURES

Graphite is widely used due to its availability, satisfactory moderating properties and low neutron capture cross-section. In pure graphite the neutron capture cross section is  $0.032 \cdot 10^{-24}$  cm<sup>2</sup> [3]. The energy of neutrons generated by nuclear fuel fission decreases as a result of elastic collision between neutrons and moderator atoms. A fast neutron loses 0.158 of its energy in each collision with carbon atoms. Therefore, for moderation of the fast neutron to the “thermal” (slow) neutron velocity  $\sim 110$  collisions with carbon nuclei are sufficient [4].

The neutron energy loss caused by elastic collision strongly depends on the graphite density. Usually for the nuclear reactor stacks one uses graphite blocks of 1.65...1.75 g/cm<sup>3</sup> density [5].

In the world practice the most commonly used are reactors of several types: channel-type graphite-water boiling water- and superheated vapor reactor, tank-type water-cooled unboiling-water- and boiling water reactors, low-temperature gas-cooled graphite-moderator carbonic dioxide-cooled reactors, high-temperature helium-cooled graphite-moderated reactor, fast neutron sodium-cooled reactors, swimming-pool type water-moderated water-cooled nuclear reactors [6].

All the above reactors, except swimming-pool reactors are used in NPP, and a water-moderated water-cooled unboiling-water reactor is used also in marine nuclear power plants.

In our review we stop only at designs of uranium-graphite reactors the main part of which is large-tonnage graphite stack. Despite a numerous variety of stacking designs there is a series of common principles. The stack is mounted in the form of a pile of graphite blocks by fitting them to each other without significant clearances between them. At the same time, the stacking design should provide a necessary mobility of parts to avoid the structural failure as a consequence of thermal or radiation deformation. The prepared construction is wholly enclosed in the sealed case preventing graphite against oxidation in the in-reactor high-temperature conditions.

To decrease the effect of radiation-induced change in the graphite block dimensions – swelling at low temperatures and contraction at temperatures above 300°C – in some constructions the graphite staking parts e.g. end-plugs mounted together with a system of fuel elements and cooling pipes, are periodically changed [7].

The stack main elements are graphite blocks in the form of a prism with holes. The material and the design of graphite blocks are chosen depending on the graphite

stack temperature, coolant type and design characteristics of the process channels into which the fuel elements are installed.

In the reactor construction practice there are two types of the process channel hole position in the stacking - horizontal and vertical.

In the first experimental reactors F-1 (USSR) [7], CP-1 (USA) [8, 9], BEPO (Great Britain) [8], and in the commercial reactors N 1 and 2 (Windscale, Great Britain) G-1, G-2, G-3 (France) [8], NPR (USA) [10] the graphite channels for fuel elements are made horizontally.

The stacking of the first NPP reactor F-1 consists of horizontally stacked 100x100x600 mm graphite blocks. The lattice spacing is 200 mm [7]. The stacking mass is 450 t.

In the Brookhaven reactor BHL the stacking has a cube shape with an edge of 7.6 m. It is mounted of different-type blocks the main dimensions of which are 102x102x1140 mm. The stacking mass is 730 t [8, 9].

In the G-type reactors (France) the staking looks as a horizontally mounted octagonal prism assembled of vertical blocks [8, 11].

Table 1 presents the main characteristics of the core of foreign graphite-moderated nuclear reactors [12].

Table 1

NPP with a graphite moderator and carbon dioxide as a coolant [12, v. 4, p. 325 and v. 7, p. 327]

Country	USA		France			Italy	Japan	Germany		
	Hanford	Big-Bottom	Marcoule		Chinon			Latina	Tokai-Mura	Julich
Reactor name	N(NPR,HPR)	HTGR-1	G-1	G-2, G-3	EDF-1	EDF-2	EDF-3	Latina (Simea)	Tokai-Mura	AVR
Starting year	1966	1967	1956	1959, 1960	1963	1965	1966	1963	1965	1967
Reactor power MWt:										
Thermal power	4000	115	38	2x200	300	800	1560	705	575	49
Electric power	790	40	2	2x35	70	200	480	200	154	15
Coolant	H <sub>2</sub> O	He	CO <sub>2</sub>	CO <sub>2</sub>	CO <sub>2</sub>	CO <sub>2</sub>	CO <sub>2</sub>	CO <sub>2</sub>	CO <sub>2</sub>	He
Stacking shape	-	Cylindrical	Octagonal prism		Cylindrical			24-face prism	Cylindrical	Cylindrical
Cell arrangement	Horizontal	Vertical	Horizontal		Vertical			Vertical		
Stacking sizes, mm:										
Width (diameter)	1000x1000	2800	8225	7850	8300	12200	14000	12700	11720	3000
Country	USA		France			Italy	Japan	Germany		
Height (length)	11700	2300	8400	8450	9000	7200	9000	7870	6630	3000
Graphite mass, t	1800	-	1200	1200	1050	1700	2530	2100	870	-
Number of cells in the stacking	1004	-	1337	1200	1148	2304	3264	2930	2052	
Lattice type	prism	triangle	square	square	square	hexagon	hexagon	square	triangle	
pitch, mm	203,228	90	200	296-center 224	225	225	203	236		
Graphite block size, mm	-	927	200x200x1500-	200x200x1500	-	-	-	-	-	
Graphite temp. (max), °C		mean	170	400	350	450	-	390	410	900

As is seen from the table, most of the current uranium-graphite reactors have a vertical arrangement of process channels. Such a design facilitates the defueling process. Moreover, graphite in the vertical stacking is influenced by the proper mass action, as the large-tonnage uranium loading is suspended on the upper reactor elements or is supported by the lower under-reactor supports. The stresses produced in the lower graphite blocks by the whole column mass do not exceed 2...4 kg/cm<sup>2</sup>.

By fitting graphite blocks inside the column a vertical channel is formed in which the different-purpose process tubes are installed.

The longitudinal thermal expansion of the column occurs freely without interaction with neighboring columns. For the thermal and radiation expansion of blocks in the transverse direction a clearance between the columns is provided [12].

## STRUCTURE OF GRAPHITE STACKS IN URANIUM-GRAPHITE REACTORS

**The first NPP.** In the first NPP [13] the reactor graphite stack of 300 mm diameter and 4500 mm height is mounted of blocks of two types. The core of 1500 mm diameter and 1700 mm height is piled of vertically standing hexagonal blocks of 600 mm height and the "turnkey" size of 173 mm. The central holes, containing in their inside 128 fuel channels and 22 channels of the safety and control system (SCS), have diameter of 65 mm. The channels form a triangular lattice with a 120 mm pitch.

Each of the process channels having graphite end-plugs comprise fuel elements in the stainless steel shell.

The reflector consists of horizontal blocks mounted on 24 vertical stand pipes through which the reflector cooling water circulates.

The graphite stack temperature in the center of the reactor is 800 °C. The maximum graphite temperature depends on the reactor power. The graphite stack

temperature as a function of the reactor power is shown in Fig. 1 [11]. The difference of temperatures in the center and in the periphery is ~ 17% [14].

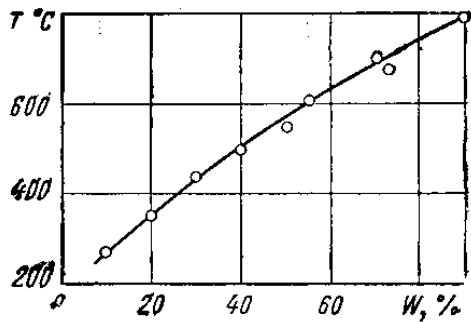


Fig. 1. The graphite stack temperature as a function of the First NPP reactor power  $W$

The graphite stack of the Beloyarsk-1 NPP (BNPP). The graphite stack of BNPP Unit 1 and Unit 2 has a cylindrical shape of 9.6 m diameter, 9m height [15]. The total number of vertical holes is 1134, 998 of them are work holes and 100 are provided for control and safety rods. Fuel channels form a square lattice with a pitch of 200 mm.

The core region of 7.2 m diameter and 7 m height is surrounded with a 0.8 m thick graphite reflector. In the upper stack part mounted are an additional graphite layer of 1 m thickness and a cast-iron layer of 0,5 m playing a role of an upper biological reactor shielding. The working reactor fuel channels are designed by analogy with the First NPP reactor. The outer cylinder of graphite plugs is 75 mm diameter. In the pipes a water or steam-water mixture is circulating. On the outside the stack is enclosed in the sealed carbon-steel case which is filled and continuously feed with pure nitrogen to protect graphite against burn up. Maximum temperature of the graphite stack is 725 °C.

**RBMK reactor.** RBMK-1000 is a high-power channel-type boiling-water graphite-water reactor with direct supply of saturated steam into the turbine [6]. The reactor is a graphite-water stack enclosed in the case and supported by the weld-fabricated metal constructions which together with the case form a sealed cavity filled with helium-nitrogen mixture. The graphite stack has a cylindrical shape and is assembled of columns consisting of separate blocks (250x250 mm) with axial cylindrical holes. In the holes the process channels and SCS channels take place. For the graphite stack one uses graphite GR-280.

The high-power channel type reactor (RBMK) is schematically represented in Fig. 2 [16].

Control system FA and SCS; FA and SCS, concrete; metallic case; graphite stack; 1-st circuit; 2-nd circuit; Nitrogen blow system of the reactor graphite stack  $P = 80$  atm.

To provide the heat transfer conditions and to compensate the hole diameter decrease in the block, resulting from the radiation-thermal graphite shrinkage and channel tube diameter increase in consequence of the creep [17], between the blocks and process channels placed were split graphite rings of 20 mm height as elastic heat-transfer elements. They are arranged along

the channel height closely to each other: one of them is tightly adjacent to the channel wall, and other – to the inner surface of the hole in the graphite block.

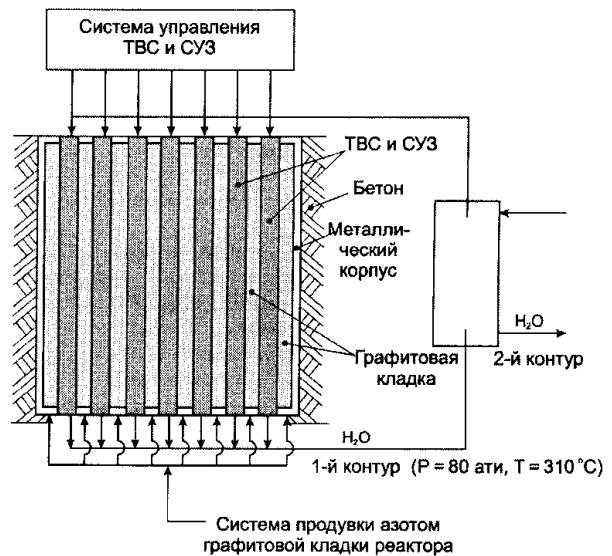


Fig. 2. Schematic representation of a high-power channel-type reactor (RBMK)

The radial clearances, created by the graphite rings, permit a free radiation block shrinkage and tube diameter increase. Such contact rings are placed in the zone of maximum heat flows.

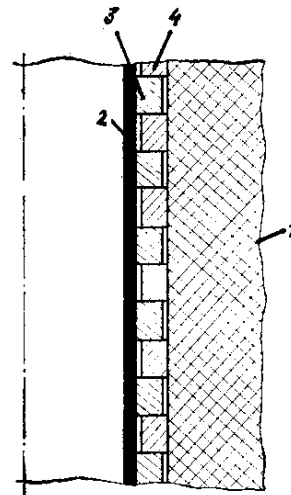


Fig. 3. Arrangement of graphite rings in the RBMK cell: 1 – block; 2 – zirconium tube; 3, 4 – split graphite rings

Fig. 3 shows the arrangement of graphite rings in the RBMK cell. The reactor of such type was operating previously at Chernobyl NPP. Tank-type low-temperature gas-cooled graphite-moderator reactors provide a basis for nuclear industry of Great Britain and France.

The core of this-type reactors is assembled in the stainless steel or ferroconcrete tank mounted on the support columns in the concrete cavity. In the tank there is a plate on which the graphite stack is mounted of vertical columns having holes in the center. Each column is stacked of separate blocks. The stack height is 8...10 m, diameter is 14...16 m. the fuel elements are loaded into the stack holes. The refueling is performed

through the holes in the closure head or in the bottom of the reactor by means of refueling machine. The coolant gas is carbonic acid.

**HTGR reactor.** High-temperature gas-cooled graphite-moderator reactor is one of the promising IV generation reactors [18]. In the tank-type reactor graphite is used as a moderator, reflector and base structural material, and helium is applied as a coolant. Thus, much higher temperatures and, consequently, a high efficiency (40%) are reached.

HTGR is presented in Fig. 4 [16].  
 Spherical fuel element feeding system  
 Absorber spherical pellet feeding system  
 Graphite stack  
 Vessel  
 Spherical fuel elements  
 Absorber spherical pellets  
 2-nd circuit  
 1-st circuit

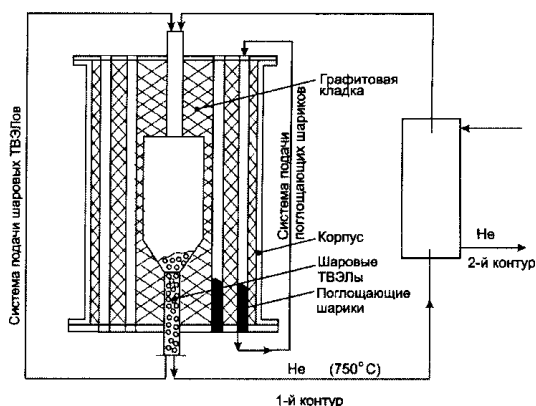


Fig. 4. High-temperature gas-cooled graphite-moderator reactor

Reactors of such type have been built and are operating experimentally in some countries: „Dragon” Great Britain), „Peach-Bottom” and „Fort St.Vrain” (USA), AVR and THTR-300 (Germany) [18]. Developing of high-temperature gas-cooled graphite-moderator reactors in Western Europe has been recommenced in the late 90-th on the base of the international cooperative work by the IAEA Framework Project.

In 2007 to create a coordinated partnership for development of the HTGR technology in Europe the HTR-Technology network (HTR-TN) was created; to advance the R&D works for a next-generation commercial HTGR reactor the plan was worked out. The works were based, in main, on the HTGR technology developed in 1970-1990. The joined project RAFAEL was aimed to the R&D fulfillment in the field of physics of reactor core, nuclear fuel, materials, safety and integration of systems. In the project 34 partner were participating, the project budget was 20 millions euro [19].

A special attention was given to the engineering problems: studying the behavior of coated fuel particle shells at high temperatures, developing carbon materials, selecting heat-resistant and radiation-resistant metal materials for the reactor vessel.

In the USA the development of high-temperature gas-cooled graphite-moderator reactors is implemented

by three projects: GT-MHR, NGNP (Next Generation Nuclear Plant) for the energotechnological complexes and VHTR in the framework of the international project GIF ((Generation IV International Forum) [19].

The project NGNP for generation of electricity, thermal energy and hydrogen is developed in the framework of the US National Program on the Next Generation Nuclear Plant. The NGNP project is funded by the US Department of Energy (DOE), the project manager is Idaho National Laboratory. The reactor is intended to be operated at high temperatures. To realize successfully the project goals it is necessary to solve a number of problems: to develop the structural materials and the fuel which can provide the rise in helium temperature at the reactor exit from 850 to 1000 °C and higher temperatures; to create the fuel enabling the operation with a high-density metal (HM) burn-up fraction of 150...200 GW. day-t, to eliminate a significant temperature gradient in the reactor core and to prevent the hot particle penetration into the coolant.

### APPLICATION OF CARBON-GRAPHITE MATERIALS IN FUEL ELEMENT STRUCTURES

The fuel element is a structure designed to seat the nuclear fuel in a fixed position into the core, to generate the thermal energy in the core and to transfer it into the coolant.

Fuel elements comprise: fuel kernel, cladding, annular and spacing components (Fig. 5) (6). Fig. 5 shows the fuel rods with uranium dioxide fuel kernel. In the fuel kernel the nuclear fission of heavy elements occurs. Depending on the fuel structure the fuel elements can be with homogeneous (monolithic) and dispersion fuel kernels. Fuel kerns can be made of metal (uranium, plutonium, thorium and their alloys) and ceramic materials (oxides, nitrides, carbides and other uranium, thorium, plutonium compounds).

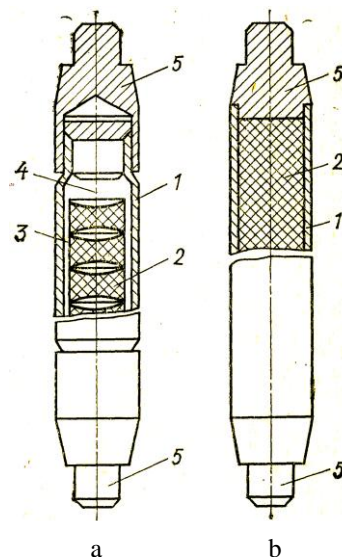


Fig. 5. Fuel rods with uranium dioxide:  
 (a) container type and (b) monolithic/solid type:  
 1 – cladding; 2 – fuel kernel; 3 – gas gap;  
 4 – gaseous fission product collector; 5 – end plugs

Dispersion fuel kernels, composed of fission material particles, are distributed in the continuous matrix made of metals, alloys, ceramic materials, graphite and other non-fissionable materials.

The main advantage of the dispersion fuel is its high radiation resistance with a high burnup [6].

The significant disadvantage of the dispersion fuel is a relatively low specific content of fission materials in the kernel volume. This disadvantage can be eliminated by increased fuel enrichment with fission materials.

In the dispersion fuel elements one uses kernels having a complex structure in which each of fuel particles is enclosed in the protective shell (metallic, graphite or ceramic).

Protective films increase the resistance of dispersion kernels by seating fission fragments in a small volume near the fuel particles. Besides, the coatings promote a uniform particle distribution in the matrix material. Coatings on the fuel particles are obtained by different deposition methods. For the most part in the modern means of production the chemical gas-phase deposition is used. In England the patent application for production of nuclear fuel particles with carbon coating was applied in 1959. The first industrial firms which started the manufacturing of such coatings were “Mynnesota mining and manufacturing” and “Newcamer material and equipment” [19].

At present more than 20 firms and laboratories in the USA and 14 enterprises in other countries are actively develop the nuclear fuel in the form of coated particles [19]. To fabricate fuel elements for the high-temperature gas-cooled reactor the manufacturers in Peach-Bottom have used the carbon-coated particles of compound uranium and thorium dicarbide dispersed in the graphite matrix. The fuel elements of this reactor are operating at temperatures below 1500 °C. Such fuel is used in the reactors „Dragon” and „BBC-Krug” in Europe and “INTEREX” in the USA [19].

Most often the carbon coatings are obtained by the chemical gas-phase deposition of carbon in the boiling layer. Initial gases for carbon deposition by the gas-phase methods are: methane, acetylene, propane, natural gas. The coating deposition process is usually conducted at 1400 °C and higher temperatures.

One of the most promising UO<sub>2</sub>-based fuel elements with carbon coating are produced by “Union Carbide”. The coating is composed of the 20 μm internal layered carbon film and 80 μm external layer of columnar carbon. Such coatings retain well the fission products at high temperatures and high degrees of burn-up [20].

In the high-temperature gas-cooled graphite-moderator reactors two shapes of dispersion fuel elements are used – spherical and block pellets. In the block constructions the microfuel is distributed in cylindrical pellets, which are used for prismatic fuel elements

The refueling is carried out in the shut-down reactor (the USA, Japan and joined Russian-American Project GT-MGR).

In HTGR different fuel cycles can be applied: using pure uranium (with slightly enriched uranium) and with thorium, plutonium and actinides being formed in the irradiated nuclear fuel of other reactors. Results

obtained by the European Organization of Nuclear Research (CERN) show that the heavy isotope transmutation allows one to decrease the spent fuel (SF) activity of the four-generation HTGR by a factor of 10000 as compared to the ZWR SF [18].

Other nuclear fuel design, namely spherical fuel elements, is used in the high-temperature gas-cooled graphite-moderated reactors. In the gas-cooled reactors VGR-50 and VG-400 the core consists of a free charging of spherical uranium-graphite fuel elements where the fuel is in the form of fuel microspheres (MF) [21, 22].

The fission product retention efficiency in such fuel elements depends on the choice of parameters and techniques of manufacturing fuel microspheres and fuel elements based on them.

In standard constructions of MF one uses for fuel the uranium dioxide microspheres having the density of 10 g/cm<sup>3</sup> and more and the nonsymmetry ratio less than 1.05.

Spherical fuel elements of 60 mm in diameter consist of a fuel kernel in which microfuel elements are uniformly dispersed in a graphite shell. The content of U-235 in the fuel elements for VGR is 50...0.5 g, in the fuel elements for VG-400 it is 0.4 g. The microfuel kernel volume filling is 4...15% depending on the required uranium charge and kernel dimensions [21–23].

To manufacture spherical fuel elements the fuel microspheres were mixed with a matrix compound on the base of reactor graphite powder and coal-tar pitch. Then the kernel was pressed under pressure to 10 MPa. Further the kernel was covered by the unfueled shell of the same matrix material built-up by the two-side scheme (Fig. 6) under pressure of 30 MPa and subjected to the thermal heating at 1200...2000 °C.

The spherical fuel element, in which the fuel is contained in the spherical layer, enables the maximum temperature decrease in the operating fuel element by about 150 °C.

Owing to the low volume filling of the kernel with microfuel particles the strength and thermophysical properties of the fuel element are determined by the properties of matrix graphite.

In our review we mention in more detail the technique of manufacturing the spherical fuel elements developed at NSC KIPT. By this technique, instead of pressing we apply the procedure of green article molding followed by subsequent impregnation with pyrocarbon obtained by gas-phase deposition [24, 25].

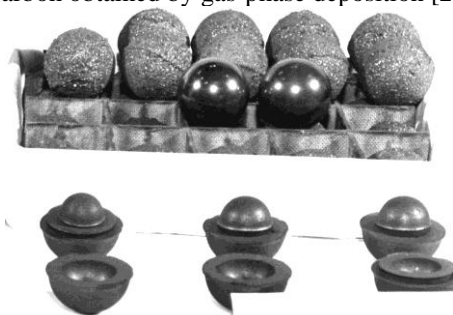


Fig. 6. Two-side scheme of the upper shell formation on the spherical fuel element [23]

The technique of manufacturing spherical fuel elements includes three main stages [25, 25]:

- making of fuel microspheres (FM);
- making of microfuel elements (MF);
- making of spherical fuel elements (SF).

According to the NSC KIPT developments the fuel microspheres are fabricated by mechanical spheroidization of fuel green articles prepared on the base of plasticized blends. The method consists in making round the cylindrical articles to obtain perfect microspheres.

The heat treatment of prepared microspheres is conducted in two stages. In the first stage the vacuum annealing at  $\sim 300\text{ }^{\circ}\text{C}$  is carried out to remove the plasticizer. In the second stage the fuel microspheres are finally sintered in vacuum or in the inert gas medium at  $1450\text{...}2000\text{ }^{\circ}\text{C}$ .

By the NSC KIPT technique, instead of the inner and outer dense pyrocarbon (PyC) layers, the combined (PyC+SiC) coatings of a density  $\geq 2.4\text{ g/cm}^3$  are used (Fig. 7).

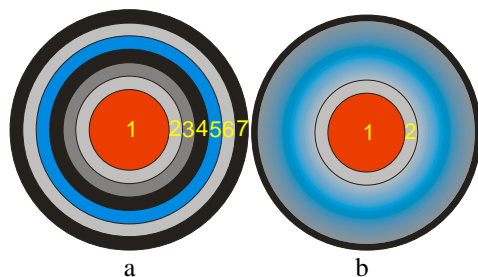


Fig. 7. Structure of spherical microfuel elements:

- a* – with multilayer coating; *b* – solid structure:
- 1 – core,  $500\text{ }\mu\text{m}$ :  $\text{UO}_2$ ,  $\text{UO}_2\text{-Al}_2\text{O}_3\text{-SiO}_2$ , UN, UCN,  $\text{ThO}_2$ ,  $(\text{UTh})\text{O}_2$  and others
  - 2 – PyC buffer layer,  $1.1\text{ g/cm}^3$ ,  $28\text{...}50\text{ }\mu\text{m}$ ;
  - 3 – PyC transition layer,  $1.5\text{ g/cm}^3$ ,  $14\text{...}50\text{ }\mu\text{m}$ ;
  - 4 – PyC dense layer,  $1.8\text{...}1.9\text{ g/cm}^3$ ,  $60\text{ }\mu\text{m}$ ;
  - 5 – SiC,  $3.2\text{ g/cm}^3$ ,  $80\text{ }\mu\text{m}$ ;
  - 6 – PyC buffer layer,  $1.1\text{ g/cm}^3$ ,  $3\text{...}10\text{ }\mu\text{m}$ ;
  - 7 – PyC dense layer,  $1.8\text{...}1.9\text{ g/cm}^3$ ,  $35\text{...}100\text{ }\mu\text{m}$

Such an alternative allows one to decrease gaseous and solid fission-product yield from the microfuel elements by a factor of 100. With a combined protective layer the thickness of a buffer layer can be decreased almost twice. In foreign countries the existing

techniques of manufacturing spherical fuel elements are based on the use of pressing processes [21, 22, 27].

Under high pressures the protective coatings of microfuel elements can be damaged that adversely affects the fission product retention.

The NSC KIPT technology of manufacturing spherical fuel elements by the method of graphite mixture binding with gas-phase pyrocarbon [24, 25, 28] eliminates the above-mentioned disadvantages.

The technology of manufacturing spherical fuel elements by the method of volume gas-phase impregnation of porous media includes the following procedures: preparation of graphite powder having a required granulometric composition; preparation of molding powder for spherical particle cladding; preparation of fuel blend for kernels; molding of fuel kernel blanks; forming of two hemispherical unfueled cans; making of a fuel element blocks (green articles); fuel element blocks impregnation with pyrocarbon, mechanical treatment of spherical fuel elements, thermal treatment and examination of a finished product.

A basic initial material for pyrocarbon-bound fuel elements is graphite powder, having a required nuclear purity, and microfuel particles coated with pyrocarbon layer and silicon-carbide layer. The molding mixture for shells of spherical fuel particles is prepared by mixing the graphite powder with plasticizer. After introducing into the molding blend a required quantity of microfuel particles and subsequent thorough mixing of the blend the initial material for forming the fuel kernels is obtained.

Forming of assembled blanks of shells and kernels for spherical fuel elements is performed in the steel dies. The procedure of blank forming to a rather low density with subsequent pyrocarbon binding to a density of  $1.75\text{...}1.90\text{ g/cm}^3$  and pyrocarbon binding of spherical fuel particles is performed in the special matrix which is placed into the pyrolysis reactor. The pyrolysis process is conducted in the natural gas flow at temperature of  $900\text{...}1100\text{ }^{\circ}\text{C}$ . Duration of the pyrolysis process to density of  $1.75\text{ g/cm}^3$  is  $\sim 70\text{ h}$ .

Table 2 presents the main characteristics of GSP and THTR-300 spherical fuel elements [29]. A similar technique was applied for manufacturing absorber elements (AE) [24] with the use of  $\text{B}_4\text{C}$  powder or other neutron-absorbing material instead of uranium dioxide.

Table 2

Main characteristics of GSP and THTR-300 spherical fuel elements [29]

Characteristic	Fuel elements GSP	Fuel elements THTR-300
Graphite matrix density, $\text{g/cm}^3$	1.75...1.95	1.72
Graphite matrix strength, MPa: compression	100	44.7/45.7
bending	45	20.4/18.6
Dynamic modulus of graphite elasticity, $\text{MPa}\cdot 10^4$	1.0	0.99/1.03
Thermal conductivity at 290 K, W/m·K: without additional heat treatment	50	–
With additional heat treatment	70	67/37
CTLE (290...1270 K), $\cdot 10^{-6}\text{ K}^{-1}$	5.0	3.59/3.92
Dynamic strength (average number of spheres falling on the cladding from the altitude without disruption )	>3000	750
Abrasive wear, $\text{mg/cm}^2\cdot\text{g}$	1...3	3
Degree of anisotropy	1.03...1.05	1.08...1.10

Note: For THTR-300 fuel elements the value in the numerator indicates the direction parallel to the molding axis, in the denominator – perpendicular to the molding axis.

In NSC KIPT a new type of matrix graphite has been developed. Its structure presents a pyrocarbon matrix enveloping the powder filler particles [30].

The main characteristic of the high-quality graphite bound with pyrocarbon (GSP) are investigated in [30, 31]. A principal difference between GSP graphite and graphite of other types is that the binding material in GSP graphite a high-strength low-temperature pyrocarbon is used. A characteristic feature of GSP graphite is a pronounced cellular matrix, the individual elements of which consist of powder filler particles coated with pyrocarbon films deposited on their surface.

In the point of intersection the pyrocarbon deposits coalesce and form a continuous many-dimensional spatial matrix enveloping all the powder filler particles.

By powdering GSP graphite and subjecting the powder obtained to the repeated gas-phase densification it is possible to increase in it the pyrocarbon component content. The compression strength of GSP graphite increases from 120 to 350 MPa when the pyrocarbon content in it increases from 30 to 90 mas.%.

Annealing of specimens at 1600 °C during 8 hours does not lead to any apparent changes.

Many of the investigations on the strength and heat-transfer properties of GSP graphite are reported in [30]. It has been shown that the strength of GSP material increases with its density increasing. The strength characteristics of GSP graphite depends also on the type of powder filler and, substantially, on the pyrocarbon content.

Refs. [32–36] present the investigation results on the corrosion resistance and integrity of microspherical fuel elements (MFE) with silicon carbide and pyrocarbon coatings as applied to the operating condition of NPP light-water reactors.

A main advantage of using MFE, directly surrounded with a light-water coolant, unlike the usual fuel pellets enclosed in the zirconium alloy shell, is a larger (more than twice) specific heat-transfer surface. This provides a short time of heat-exchange lag ( $\tau \sim 0.03$  s), fast heating and coolant evaporation in case of emergency operation that permit rapidly compensate the induced reactivity [32]. With a large specific surface the restrictions caused by the heat-exchange crisis are almost absent and the removal of the decay heat is provided due to the natural coolant circulation.

In the preliminary tested design projects MFE are placed in the fuel assembly casings made from the austenitic stainless steel. Therefore the core does not contain zirconium and the intensity of explosive hydrogen formation (zirconium-steam reaction) sharply decreases. A sphere-like shape of MFE allows one to conduct the continuous (periodic) refueling without reactors shutdown.

Authors give generalized experimental data on the corrosion resistance and integrity of protective coating of silicon carbide (SiC) and pyrocarbon (PyC) for unirradiated MFE.

The results were obtained during life tests using coolant parameters close to the normal operating regime of the light-water reactors (PWR, BWR) and steam-superheat reactor, as well as, under conditions imitating different emergency conditions [33–36].

From the results of the life tests imitating the typical operating conditions for light-water reactors it has been established that the protective coatings of unirradiated MFE possess a high corrosion resistance in water and in high-pressure steam at temperatures 350 and 550 °C.

Experiments confirmed a possibility of long-term operation of MFE with outer protective SiC and PyC coatings in the light-water reactors at coolant temperatures to 600...650 °C respectively. According to the conclusion of the study the temperature of 1200 °C can be considered as a design limit for MFE use in the reactors with light-water cooling.

Unique properties of carbon-carbon composite materials enable to recommend them for manufacturing spent fuel storage casks [37].

One of the effective methods used for reprocessing and conditioning of solid combustible radioactive waste (RAW) is their burning.

The burning of solid RAW allows one to decrease their volume by a factor of 20...100, and the mass by a factor 10...20. As a result of solid RAW burning there ash is formed, which contains about 90% of all radioactive nuclides. A main problem consists in the ash volatility that complicates its long-term storage. To solve this problem ash should be cemented, vitrified and incorporated into the glass-ceramic matrix [38].

An alternative for the above-mentioned methods of RAW immobilization can be the method developed and experimentally checked at NSC KIPT, namely, the method of RAW immobilization in the pyrocarbon matrix [39]. Also, by applying this method, it is possible to combine the process of solid RAW binding with pyrocarbon and the formation of outer hermetic enclosure of carbon-carbon composite.

So, by reasoning from the above-said about carbon-graphite material use in the nuclear engineering, a conclusion can be made that a need in these materials is continuously growing (hundreds of thousand tons). As a result in the industrial countries a new commercial production – reactor graphite manufacturing is developed. For graphite manufacturing one uses coke of suitable microstructure, as a filler, and coal tar pitch, as a binder.

Cokes for reactor graphite can be produced as by-products from the petroleum or from products of condensed coal carbonization.

The process of graphite manufacturing consists in the filler preparation (stages of calcination, crushing, milling, granulation and mixing of the prepared powder filler with a pitch). Then the mixture is formed by molding or pressing into blocks often referred as “green articles”. Further the blocks are backed, impregnated with the coal tar pitch and after re-backing are graphitized. Care must be taken that graphite be pure and without impurities of different elements (in particular, boron) [40].

Table 3 presents the properties of commercial graphite marks in comparison with foreign analogs [41]. The materials under consideration are referred to the class of medium-grained graphite of a density 1.7...1.8 g/cm<sup>3</sup>. This density of graphite grades manufactured by the extrusion pressing is reached after one or several impregnations of backed “green” blocks with pitch.

Table 3

Physical properties of different reactor graphite grades [41]

Grade	d, g/cm <sup>3</sup>	σ, MPa			E, GPa	λ, W/(m·K)	ρ, m·K <sup>2</sup> Ohm/m	CTLE, 10 <sup>-6</sup> K <sup>-1</sup>
		σ <sub>p</sub>	σ <sub>n</sub>	σ <sub>c</sub>				
RBMK	1.68	7.6/6.0	19/14	34/33	6.5/5.0	103/89	10/13	3.8/4.2
EGP-6	1.69		11.7/5.5	28/25	11.2/5.2	80/75	5.9/10.4	3.7/4.9
RZAN	1.68	14/9.2		42/38	11/7.5			2.9/3.9
PGA	1.74	10/6	15/11	32/35	12/5.6	140	6	1.3/3.3
VPG	1.84	16/8	35/20	60/58	12/9	160/115	8/10	5.0/5.4
ATR-2E	1.8	12.5	23/9	57/57	9.3/8.4	179/163		4.4/4.9
H-327	1.78	13/8	21/15	32/28	-/4.5	150/140	5.6/11	1.3/3.1
H-451	1.76	16/14	28/116	56/54	9/8	135/125		3.6/4.6
IG-110	1.78	25	3.9	80	9.4/10	124/128	11.5/-	4.2/4.2
MNG-1	1.73	16	24	55	7.9	120	11	4.8
MNG-2	1.67	35	70	75		90	13	5.1
APBy	1.69	14.5	26	61	6.8/7.8	51/61	16/16	5.3/4.1
GR-1	1.73	15/15	30/25	70/84	8/10	86/90	15/14	5.2/5.4
AG-1500	1.73		41	60	8/0	80/90	18/10.5	4.0/4.3
KPG	1.8	14	30/27	64	9.0/11	140/140	9-12/13	6.8/6.8
MPG	1.8	32	55	100	11	95/95	11.7/10.7	7.3/6.6
SPP	1.56		17	49	8.3	80	13.5	5.8
ER	1.60		20/8	28/25	7.5/17.5	50/170	25/55	5.5/1.2
GSP-50	1.72	33	70	205	17	86	21	5.8
GSP-90	1.79		95	352	21	90	32	5.5
PPEA	1.85	22/19	34/28	82/-	12/11	140/130	9/10	5.0/5.3
SZWXX	1.83	90/16	24/21	64	11/10	165/146	7/9	4.6/5.1
DM332PU	1.81	34	43	91	12	117	11	4.6
CSF	1.68		17/14	52/46	11/5	255/132	4.5/7.7	1.8/3.8

Note: Numerator indicates the direction parallel to the block height, denominator indicates the perpendicular direction. For isotropic graphite grades the averaged value is shown. Thermal conductivity (λ) is given for room temperature. Coefficient of thermal linear expansion (CTLE) is given for the temperature range of 250...500 °C.

Graphite VPG for RBMK hard contact rings, being recommended also for the external reflector and supporting columns [42], has the density, thermal conductivity, CTLE properties closed to the foreign high-density doubly pitch-impregnated reactor graphite grades [33]: German coke pitch-based ATP-2E graphite; American H-451 graphite with petroleum coke-filler; French UCAR graphite. It should be noted that CTLE (being important for the radiation resistance) of VPG graphite exceeds that of ATR and H-451 graphite grades.

The properties of APB graphite produced in the industrial conditions concede the properties of Japan reactor JG-11 and JG-110 graphite grades [43]. However, after the powder of this graphite has been

bound with low-temperature GSP pyrographite its CTLE was significantly increased and the strength was unhanced by several times [24].

In 80-th the production of the fine-grained MPG-type graphite grades (MPG-6, 7, 8) has been realized commercially. The MPG-type graphite grades, despite a high strength, could not be applied as stacking elements because of the insufficient thermal conductivity and limited overall dimensions of blanks.

At present, new technologies are developed for manufacturing graphite having a wide range of properties necessary for advanced reactor designs [43].

The properties of the new commercial graphite grades are presented in Table 4.

Table 4

Properties of new graphite grades based on composite fillers [43]

Graphite grades based on composite fillers	Graphite properties					
	d <sub>x</sub> g/cm <sup>3</sup>	σ, MPa		E, GPa	λ, W/(m·K)	CTLE, 10 <sup>-6</sup> K <sup>-1</sup>
		Bending	compression			
ChKG- 3 (Ø275x260 mm)	1.82	30.2	52.9	10.0	116	4.2
ChKG - 4 (Ø205x260 mm)	1.83	26.4	58.6	11.3	137	4.1
GRCh (Ø125x1200 mm)	1.89	30.5	83.1	12.4	122	5.0
GRCh (430x570x1300 mm)	1.79	19.2	47.3	8.6	152	4.4
GRCH - G(Ø260x230 mm)	1.89	23.7	65.1	11.0	164	4.2



## NEUTRON IRRADIATION INFLUENCE ON PHYSICAL-CHEMICAL PROPERTIES OF REACTOR GRAPHITE

Specific graphite properties, namely, low neutron-absorption cross-section, high moderating power, high thermal properties and high strength provided its wide application in the nuclear engineering. However, under in-reactor irradiation the graphite properties are substantially changing due to the fast-neutron displacement of carbon atoms from the crystalline lattice sites.

Radiation-induced changes of the graphite properties are deteriorated by the fast neutron field nonuniformity and considerable temperature gradients within the limits of a single graphite block.

Therefore, to study the behavior of graphite properties under irradiation in the wide range of temperatures and at fast neutron fluences of  $10^{22}$  cm<sup>-2</sup> and higher is an urgent problem.

The main characteristics of the structural graphite material are: stability of linear dimensions, strength, creep, elastic modulus, coefficients of thermal expansion and heat conductivity and graphite oxidation resistance.

As a result of graphite irradiation with neutrons its heat conductivity sharply decreases [44, 7]. It has been noted, that the maximum rate of  $\lambda$  decreasing is observed in the initial stage of irradiation. The data given in [45] show that under irradiation with a fluence of  $2 \cdot 10^{21}$  n/cm<sup>2</sup> at 300 °C the thermal conductivity of reactor graphite is lower by a factor of 10 than that of unirradiated material while at 600...700 °C it is lower only by 50%.

The irradiation-induced change of the graphite thermal conductivity may be explained by the photon mean free path decrease due to the radiation defect scattering.

In graphite having small-size crystallites and, consequently, a low thermal conductivity in the initial state, the relative change of the thermal conductivity under irradiation is less because of the wide photon scattering at fine crystallite boundaries in the initial graphite and, on the other side, due to a lighter defect diffusion onto the crystallite surface [45].

The heat capacity of irradiated graphite increases as a result of disordering arising in its structure. According to the experimental data [3] this increase tends to the constant value at temperature of ~ 300 K.

A part of displaced atoms which appear in graphite during low-temperature irradiation retain the energy excess as compared to the equilibrium atoms. Thus, in the irradiated graphite its heat content increases. The internal energy of the system increases too. As the temperature increasing exceeds the irradiation temperature the latent or stored energy releases.

The stored energy at a level of irradiation ~  $10^{20}$  n/cm<sup>2</sup> increases almost linearly [46].

After the fluence becomes higher than  $10^{21}$  n/cm<sup>2</sup> the accumulation of the stored energy stabilizes and its value reaches 500...600 cal/g. The rate of stored energy accumulation in graphite sharply decreases with irradiation temperature increasing. For example, the

stored energy level of the irradiation at 750 °C is only 9 cal/g [46]. Besides, the stored energy release occurs at temperature above 1000 °C that does not represent a danger for the stacking operation up to the indicated temperature.

The stored energy value depends on the graphite crystal perfection. As is shown in [28] for the three-dimensional ordered CSF-grade graphite the stored energy value under irradiation is higher than that of the polymer graphite having a lesser ordered structure.

A feature of the stored energy release process is an existence of discrete energy levels on reaching of which in irradiated carbon materials the energy release occur.

As is noted in ref. [47] for graphite irradiated at -193 °C the maximum energy release was observed at temperatures: -63, -53, 27...37, 107...137 and 167...177 °C. Such a discreteness evidences on the presence in the irradiated graphite of different-type defects or on the sequential transformation of primary defects into more complex defects as the annealing temperature increases.

There is a linear dependence between the temperature of starting stored energy release and the irradiation temperature [48]. The difference between them does not exceed 550 °C for graphite irradiated at 30 °C and decreases to 25 °C with irradiation temperature increasing to 160 °C.

The low-temperature neutron irradiation significantly improves the strength characteristics of graphite materials [49]. The radiation hardening is stabilized with damage accumulation that is explained by the equilibrium between produced point defects and their failure.

The dependences of the ultimate compression strength change on the fluence were obtained using the samples of GMZ and KPG graphite irradiated in the wide temperature and fluence range [50, 51]. From the studies it follows that the relative strength quickly changes with fluence increasing to  $(2-5) \cdot 10^{20}$  n/cm<sup>2</sup> and after that the hardening process stabilization begins. The radiation hardening effect decreases with irradiation temperature rising.

One of the important properties of the carbon material, in particular, being in operation during long time, is its creep resistance under loading. In graphite, tested without irradiation at medium temperatures (to 150 °C) the creep is almost absent, however, under irradiation effect the creep in graphite significantly increases [52, 53].

Graphite, like uranium, possesses a pronounced radiation growth power. For this material a notable effect of creep acceleration is characteristic under in-reactor irradiation.

The results on the creep deformation measured with constant load and temperature show that the radiation creep of graphite has two stages: primary creep and secondary (steady-state) creep. The third state corresponds to the accelerated stage. It arises after some "critical stress" is reached.

The dimensional changes of graphite blocks under irradiation are among the main factors limiting the graphite stacking life. This is explained by the fact that upon reaching some neutron fluence the linear

dimensions are changing. Their value and sign are determined by the fluence and density of the neutron flux, irradiation temperature, graphite anisotropy, degree of graphite perfection, level of stresses arising in graphite green articles subjected to the thermal treatment etc.

For massive graphite stacking elements the radiation-thermal effect has some features related with the spatial neutron flux and temperature distribution throughout the stacking and in the individual blocks. Radiation effects are strongly dependent on the temperature, therefore in the central part of the reactor stacking, the temperature of which is comparatively high, the radiation damage accumulation is much less than at the stacking periphery where the temperature is lower.

Across the cell the graphite properties are changing differently. Maximum changes are observed in the layers contacting with uranium charged channels where the fast neutron flux is higher, spectrum is harder, and the temperature is minimal. At the periphery or near absorber rods, where the neutron flux is unsymmetric and nonuniform, the radiation damages in the block will be also unsymmetric and nonuniform. As a result, in peripheric regions a radiation graphite growth takes place and these parts of stacking become wider. In the central reactor part, where the temperature is higher, the process of graphite shrinkage occurs [3, 54, 55].

Such graphite shape change, nonuniform throughout the stacking, causes the curving of channels and construction on the whole.

The presence of internal stresses, related with nonuniform swelling, led to the change of the block shape and longitudinal cracking in it. The cracks were formed in the blocks located in the maximum swelling region at the peripheral stacking part [11].

During the First NPP reactor operation the fluence in the core center was  $0.5 \cdot 10^{22}$  n/cm<sup>2</sup>. As a consequence, the changes in the longitudinal inner holes in the blocks have been happened: the reflector diameter was increases and, simultaneously, the core became narrow with a maximum value in the core center [51].

In 1971 in the course of complete reconstruction the reactor defueling has been performed and the stack condition was completely investigated. All the cells were examined, their diameters were measured, and the physical properties of graphite, taken from the different reactor points, were studied.

Upon the results of this examination a conclusion was made that the graphite stacking can be used in the protective medium at temperatures to 700 °C during 20 years [56].

In the course of the Beloyarsk NPP reactor cladding condition checking the graphite block holes diameters were measured and there was not found visible changes in reactors. In the cells, where the fuel elements have been damaged, the channels were destroyed because of cracks and splits [56].

Basing on the results obtained for the radiation-induced GR-200 reactor graphite form change in the stacks of commercial reactors, a substantiation of the safe reactor operation prolongation to 45 years was made [45, 37].

The samples of GR-280 and GRP-2 graphite developed for RBMK reactors have been tested. During further operation the test results confirmed the guarantees given for graphite of these grades and proved the substantiation of their working life prolongation for 15 more years [15]. It is expected that the application of GR-1 graphite will provide the operation of stacking blocks of the promising reactor MKOR-1500 with a 50-year service life which can replace RBMK reactors after their decommissioning [59].

## CONCLUSIONS

The review has shown that graphite materials are used in more than 100 power reactors, plutonium production reactors and research reactors. A wide use of graphite in the reactors is explained by its availability, good moderating properties and low neutron-capture cross-section. Pure graphite has an effective neutron capture cross-section of  $0.032 \cdot 10^{-24}$  cm<sup>2</sup>. The neutron capture cross-section, which is strongly dependent of the graphite purity, can be decreased to remove from graphite the impurities such as boron, vanadium, rare earth elements etc., having a high neutron capture cross-section. The moderating property of graphite is due to its low atomic weight. Graphite is inferior, by the relative moderation value, to the heavy water but is close to beryllium and exceeds the simple water.

The graphite density is very important for its moderating properties. The higher density the higher its macroscopic moderating power. Usually, graphite blocks for the nuclear reactor stacking have a density of 1.65...1.75 g/cm<sup>3</sup>. By increasing the graphite density and its degree of purity it is possible to decrease slightly the reactor dimensions. In recent years one attaches much importance to carbon-graphite materials with decreased gas permeability, especially to the graphite use in high-temperature gas-cooled reactors.

The use of carbon-graphite material with improved properties obtained by the current technology will enable the development of the most advanced and highly productive nuclear facilities with a prolonged service life.

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Article received 26.01.2015

## УГЛЕРОД-ГРАФИТОВЫЕ МАТЕРИАЛЫ В ЯДЕРНОЙ ЭНЕРГЕТИКЕ (ОБЗОР)

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Освещаются вопросы, связанные с использованием углеграфитовых материалов в качестве замедлителей и отражателей нейтронов в более 100 энергетических ядерных реакторах. Рассмотрены различные типы кладок ядерных реакторов. Проанализированы физические и механические свойства углеграфитовых материалов и изменения радиационной размерной стабильности и стойкости графита в результате взаимодействия его с нейтронным потоком.

## **ВУГЛЕЦЬ-ГРАФІТОВІ МАТЕРІАЛИ В ЯДЕРНІЙ ЕНЕРГЕТИЦІ (ОГЛЯД)**

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Висвітлюються питання, пов'язані з використанням вуглеграфітових матеріалів в якості сповільнювачів і відбивачів нейтронів у більш ніж 100 енергетичних ядерних реакторах. Розглянуті різноманітні типи кладок ядерних реакторів. Проаналізовані фізичні і механічні властивості вуглеграфітових матеріалів та зміни радіаційної розмірної стабільності і стійкості графіту в результаті взаємодії його з нейтронним потоком.