A STUDY OF GRAPHITE FROM RBMK-TYPE REACTORS

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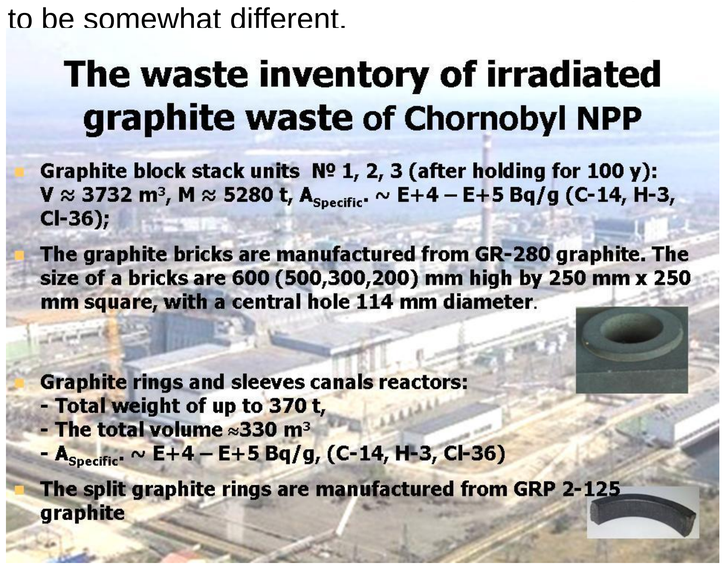
Introduction

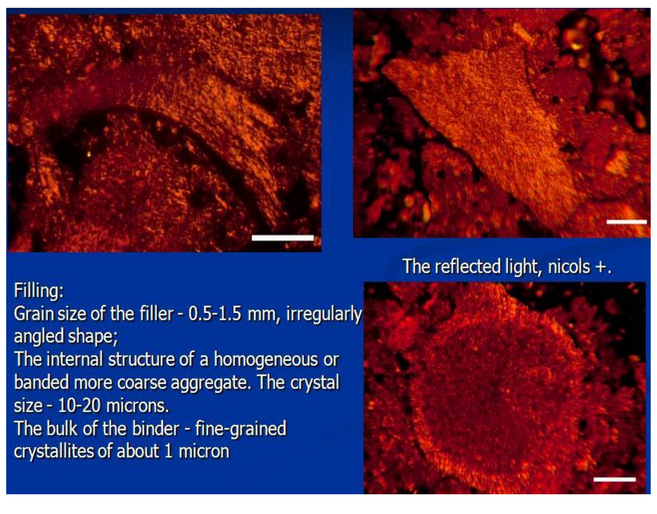
Institutions of the National Academy of Science of Ukraine are working out the various options for using graphite from uranium-graphite reactors during their decommissioning. The morphology of graphite is determined by two main components - the filler and the binder. The binder has a finely crystalline homogeneous structure, a filler based on microcrystallites with a more or less perfect crystal structure. Microcrystallites form various types of formation - grains with different degrees of texture. Since each grade of graphite has unique structure and texture, its irradiation behavior can be expected to be somewhat different.



The research was carried out to investigate the structure and morphology of the surface layer of the graphite stack GR-280 and fine graphite composite GR-2-125 the split rings from technological channels. They are anisotropic materials with densities of 1.71 g/cm^3 and 1.85 g/cm^3, respectively. Microstructural characterization of the filler and binder materials is performed. Using optical microscopy, the macro-scale features of the filler particles and macro-porosity were characterized.

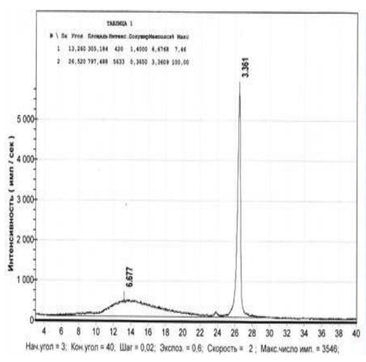
Microstructural characterization of graphite

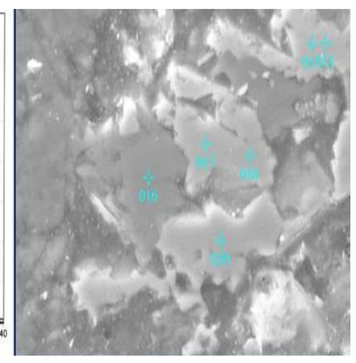
Studies of graphite microstructure have been made using XRD, scanning electron microscopy and energy dispersive X-ray analysis to obtain information of the graphite's polycrystalline features. X-ray powder diffraction data was collected and the line profiles were analysed using the Warren model, estimates were made of the distance between planes and the magnitude of the coherent scattering.



Low angle scattering of X-ray can provide information about the defects and the inter-crystalline pores in non-irradiated graphite. In the case of graphite there are established specific reflections for calculating the unit cell parameters: for the **a**-spacing the (100), (110) and (112) reflections are used; for the **c**-direction, the d-spacing (the distance between consecutive basal layers) is calculated from the (002) and (004) reflections.

By using the Scherrer equation, an average of the out-of-plane crystallite size (coherence length in the **c**-direction) - Lc, can be estimated from the broadening of (002) peak, while the in-place crystallite size (coherent length in the **a**-direction) – La from the broadening of (100). The instrumental broadening was measured from the line profile of a standard silicon specimen. The reflex broadening of the diffraction peak (002) corresponds to the size of the coherent scattering CS ≈ 35nm and ε_{00l}=0.0045. The shape of this diffraction peak indicates the presence of two fractions with different dispersion and crystalline properties (crystallite size, micro-strain) of the sample. These crystallites are relatively small and have a random orientation. The crystallites observed in GR-2-125 to have a high degree of crystalline alignment. The results of experiments were presented at the XX International conference on physics of radiation phenomena and radiation material science.





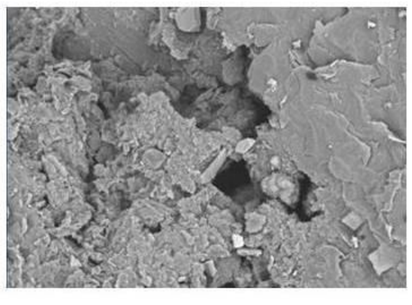
Chemical decontamination of graphite

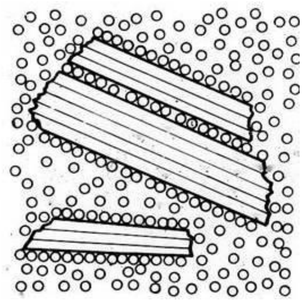
Chemical treatments have been investigated in order to decontaminate irradiated graphite. Experimental studies have shown that the $A_{specific}$ of ^{14}C for graphite rings is $10^{+4} \div 10^{+5}$Bq/g, and is determined primarily by radionuclides: ^{14}C (55%), ^{3}H (4%), ^{36}Cl (5.5%).

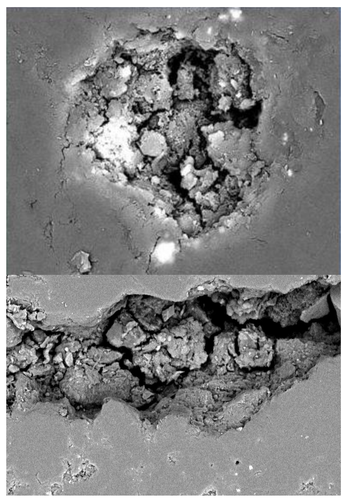
A wide variety of strongly oxidizing agents and chemicals for intercalation has been used for chemical decontamination of graphite. Powder sample graphite was dispersed in solvents with ultrasonic treatment for destruction of a binder phase. Ultrasonic exposure results in dispersions of polycrystalline graphite that are more suitable for an intercalation process, with further disruption under repeated ultrasonic treatment for "exfoliation". It was shown that interlayer spacing (d_{00l}) increases as result of intercalation of the treated graphite.

For characterization of graphite intercalation were used: optical microscopy, SEM and EDAX; XRD analysis

The binder is composed of small randomly oriented crystallites whereas the crystallites in the filler particles are much larger and well aligned with their [001] typically parallel to the particle's radial direction. This preferential oxidation is due to the much larger quantity of exposed reacting surface area on the smaller randomly oriented crystallites. Preferential oxidation of the binder phase has also been observed by Contescu *et al*. Two mechanisms have been identified in the chemical decontamination of graphite; the destruction of the binding material and the selective removal of the surface layer of graphite. The GRP–2–125 graphite total porosity of the sample was P = 0.158± 0.02 from the density measurement by the gravimetric method.







Intercalation of graphite is directly related to the microstructure and also with characteristics such as density and anisotropy, the size of crystallites, the size of the micro-strain in the structure, the ratio of crystalline and amorphous components, the size and total pore volume. As shown of J.G. Castle, a molecular chain of linked layers can pass through different crystallites, being fixed in each intersection. The presence of oriented crystallites in "crosslinked" polymers affects their properties in certain directions.

Chemical intercalation reaction and hydrolysis has a significant impact on the structural characteristics (the amount of broken C-C bonds) of the graphite. Surface morphology exfoliated structure showing a numerous and complicated open pore. The preliminary results of leaching experiments show that the ^{14}C leached from graphite without treatment is less compared to graphite samples after treatment with an oxidizing/intercalated agent. The surface area significantly changes during the course of the reaction as well.

Graphite encapsulation

We studied the encapsulation of graphite (pieces and dust) in clay-cement and organics-cement matrices. Chemical interactions at the interface between cement materials and clays may change the porosity and the pore structure, and affect the diffusive transport of water and C-14 nuclides. We study leaching processes and their impact on physical properties, especially on transport characteristics for pore water.

Graphite encapsulation into different grout matrixes

Sample	Matrix mix	Rate of capillary suction (grams/hour)	Total Porosity (%)
15% graphite load	1Admixture +10% clay +70% OPC	2,09	5,6
	2Admixture+10% clay+70% OPC	1,92	4,8
25% graphite load	1Admixture+15% clay +70% OPC	1,67	5,4
	2Admixture +15% clay+70% OPC	1,41	4,7
30% graphite load	1Admixture +30% clay +70% OPC	1,30	3,2
	2Admixture +30% clay+ 70% OPC	0,96	2,6

Leaching experiments were made to detect ^{14}C release from graphite after concrete encapsulation. The results show that the ^{14}C quantity in the leachant from the encapsulation graphite after more than five years is less than lower detectable limit of the analytical technique.

Acknowledgements

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