The measurement of impurity of RBMK-1500 graphite

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Radiological characteristics of RBMK-1500 graphite are crucial in the choice of irradiated graphite treatment technology. The trace impurities (<0.01% wt) during 20-30 years of irradiation in the neutron flux are transmuted into the long-lived nuclides (e.g. ^{14}C, ^{36}Cl, isotopes of Np, Pu, Am, Cm) [1]. Several samples of RBMK-1500 graphite manufactured for different construction elements (stack column, sleeve, bushing) were analysed with instrumental neutron activation analysis (INAA) method (LVR-15 experimental reactor of the Research Centre Řež, Ltd.) and with prompt gamma activation analysis (PGAA) method (Heinz Maier-Leibnitz Zentrum) in stack and sleeve samples in order to obtain the missing information on impurity distribution in the RBMK-1500 graphite. Also the inter-comparison with previously obtained results with INAA & GDMS (CEA Saclay, France) [1] and ICP-MS (FI, Lithuania) [2] in graphite sleeve have been performed.

Graphite constructions in the RBMK-1500 reactor

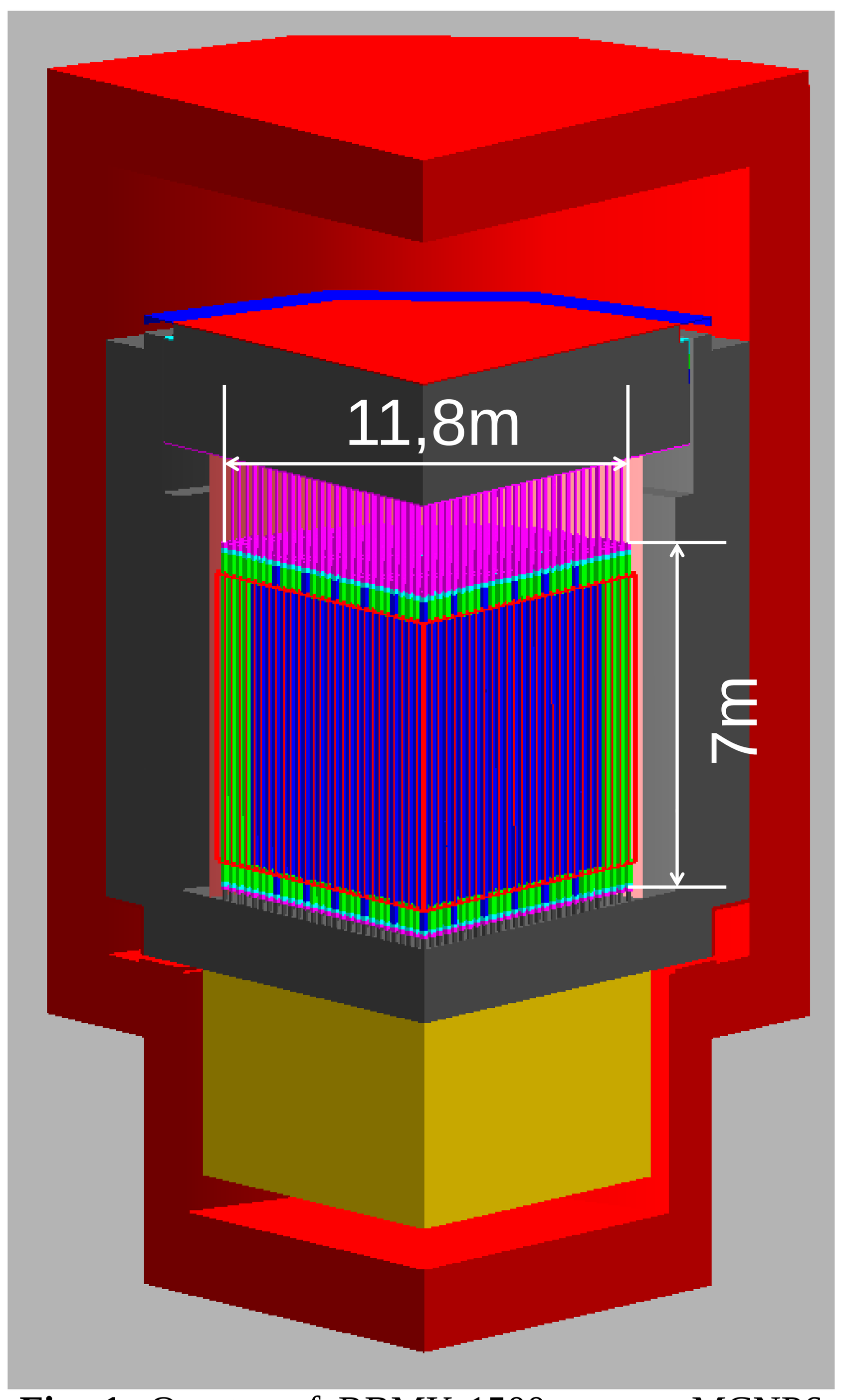


Fig. 1. Quarter of RBMK-1500 reactor MCNP6 model including fuel rods, graphite (core zone, top, bottom, side reflectors), metal plates, cooling tubes, biological shielding (water, concrete, serpentine) and reactor vault.

Two RBMK-1500 reactors were built in the Ignalina nuclear power plant (INPP). Unit 1 operated during 1984 and 2004, Unit 2 – in 1987-2009.

Graphite in the RBMK type reactor is the most massive technological element: used as a moderator and reflector in both reactors constitute ~3600 t, it will contribute largely to the total volume of radioactive waste, especially if it will be treated as a long-lived waste requiring the geological disposal.

Looking for solution to radioactive waste characterization the experimentally validated 3D model (MCNP6) was developed for description of radiological characteristics of different parts of nuclear reactor [3-4].

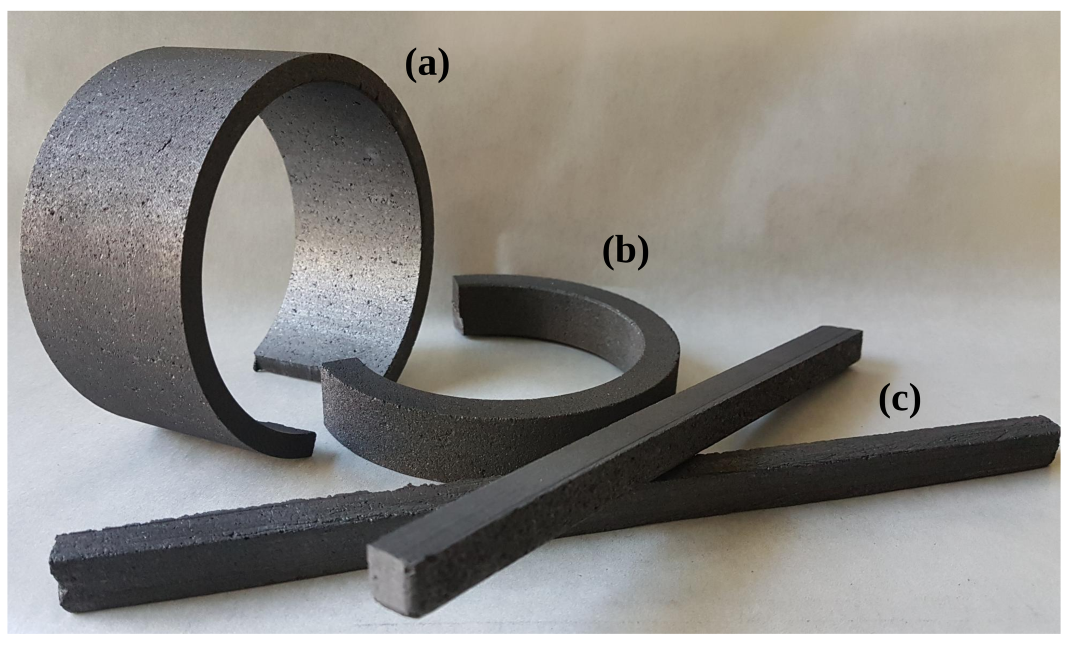


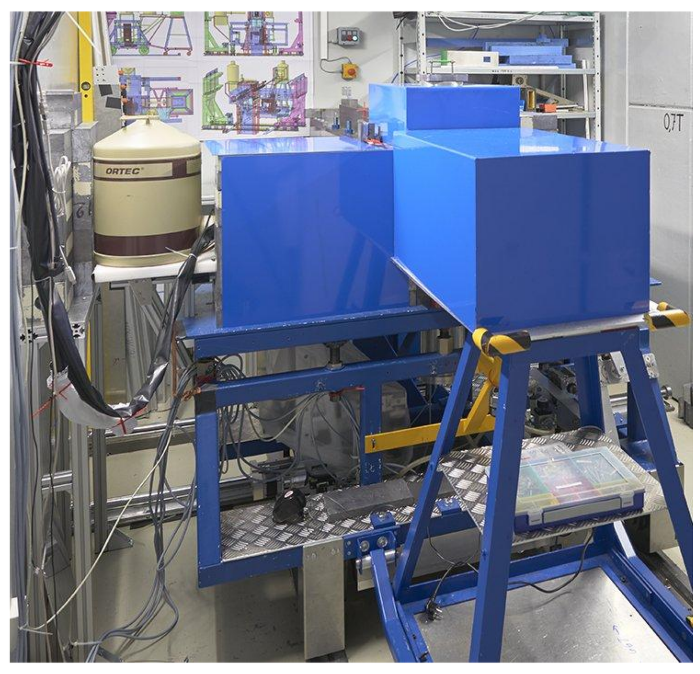
Fig. 2. Graphite of RBMK-1500 reactor constructions: **(a)** bushing; **(b)** sleeve; **(c)** stack fragment**.**

Experimental methods

Graphite samples were analyzed with instrumental neutron activation analysis (INAA) using both short- and long-time irradiations in the LVR-15 experimental reactor (Research Centre Řež, Ltd.), at a thermal neutron flux of $3{\cdot}10^{13}$ cm^{-2} s^{-1}. The induced radionuclides were measured with high efficiency, high resolution coaxial HPGe detectors after several decay times to achieve detection limits of elements determined as low as possible. Altogether 45 elements were determined.

http://cvrez.cz/en/infrastructure/research-reactor-lvr-15/

Other measurements were performed at Heinz Maier-Leibnitz Zentrum by prompt gamma activation analysis. For irradiation thermal neutron flux of $2{\cdot}10^{10}$ cm^{-2} s^{-1} was used. For gamma detection Compton-suppressed spectrometer was used [5]. 16 elements were determined by this method. It was also expected to obtain N impurity concentration by PGAA, as it is impossible to get it by other methods, but due to interference it was not measured.



http://www.mlz-garching.de/pgaa

Currently the 15 ppm concentration of N is used in the calcualtions according to the experimental results of irradiated graphite [6], but independent evaluation of initial concentration would be of great importance.

Results

The impurities of RBMK-1500 graphite stack and bushing samples were analyzed for the first time. 9 graphite samples from different RBMK-1500 graphite construction parts, namely stack, sleeve and bushings were analyzed using INAA and PGAA and compared with previously measured [1] and [2]. In the Fig. 3 the actual measurements were compared with the minimal and maximal limits of impurities concentrations used by now in the modeling [7]. Generally, for all measured impurities, concentrations may vary by order of magnitude comparing stack/sleeve and bushing materials and results show that the stack is "cleaner" comparing with graphite sleeve and bushing material. The obtained information about different impurity concentrations of some clue nuclides as Cl, Mn, Fe, Co, Ni, Cu, Zn, Sr, Cs, Ba, Eu, U which can be detected by gamma spectrometry or by destructive beta and alpha spectrometry analysis after chemical preparation in the spent graphite are summarized in Table 1, where optimized concentration limits are determined. The obtained new impurities data will be used for MCNP6 and SCALE6.1 model adapted for RBMK-1500 graphite activation case .

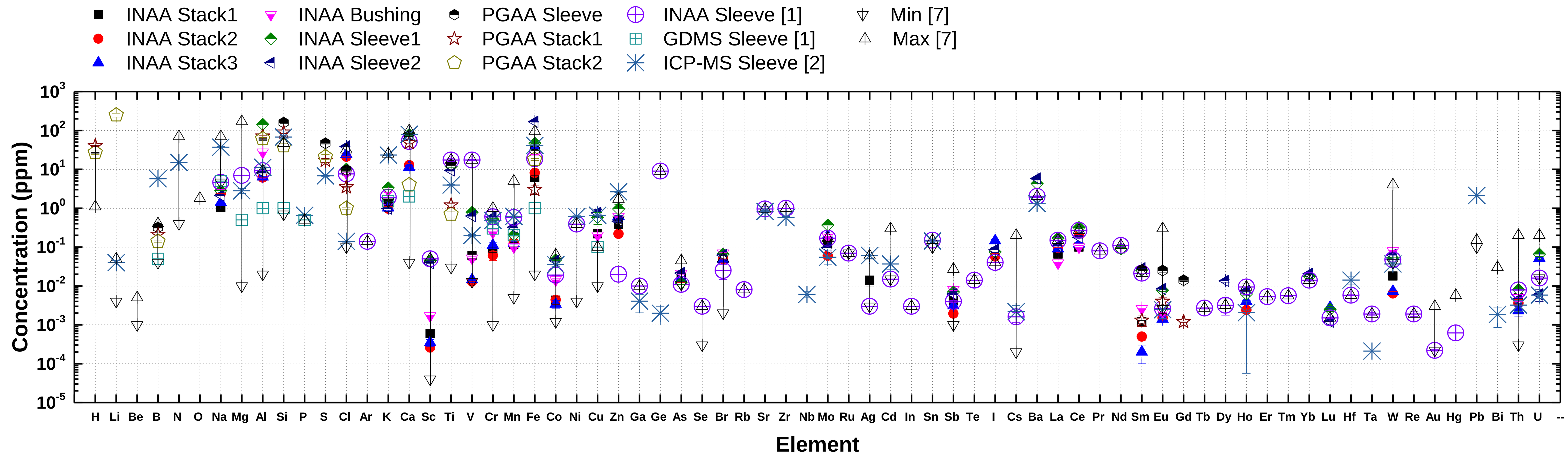


Fig. 3. Measured concentration of impurities in the virgin RBMK-1500 graphite constructions and it comparison to existing limits.

Table 1. Optimized limits of RBMK-1500 reactor graphite basic impurities**.**

Element	Conc. Min, ppm	Conc. Max, ppm
N	0.4	15
Cl	0.1	39
Mn	0.01	1.1
Fe	0.02	168
Co	0.001	0.1
Ni	0.004	0.61
Cu	0.01	0.76
Zn	0.52	5.3
Sr	0.83	0.96
Nb		0.006
Cs	0.016	0.0022
Ba	1.3	5.8
Eu	0.001	0.025
Th	0.003	0.008
U	0.0012	0.066

Conclusions

- Activity of radionuclides in irradiated graphite is mostly determined by concentration of impurities (by few orders of magnitude) in virgin graphite as neutron flux is sufficiently reliable for models used.
- The lower concentrations for Na, K, Sc, V, Cr, Mn, Sb, Cs, Sm, Eu and Ho but higher concentrations for Al, Si, S, Fe, Br, Mo, Ba, Eu, Gd, and U were obtained from INAA and PGAA analysis comparing with other measurements [1-2].
- Concerning the actual measurements comparison with the current maximal limits [7], the concentrations values were exceeded for Al, Cl, Fe, Ni, Mo and Ba. Gd impurity have been measured for the first time, the others were below maximal limit.
- Optimized limits for RBMK-1500 reactor graphite basic impurities have been determined: the maximal value have been raised up for Cl and Fe, Co, Ni, Zn, Ba and reduced for Mn, Cu, Cs and Th , as these impurities could have high contribution to the graphite activation modeling.

References

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