RADIOLOGICAL AND STRUCTURAL CHARACTERIZATION OF RBMK-1500 REACTOR GRAPHITE AND APPLICATION OF THE ION IMPLANTATION METHOD TO INVESTIGATE IRRADIATION DAMAGE IN GRAPHITE MATRIX

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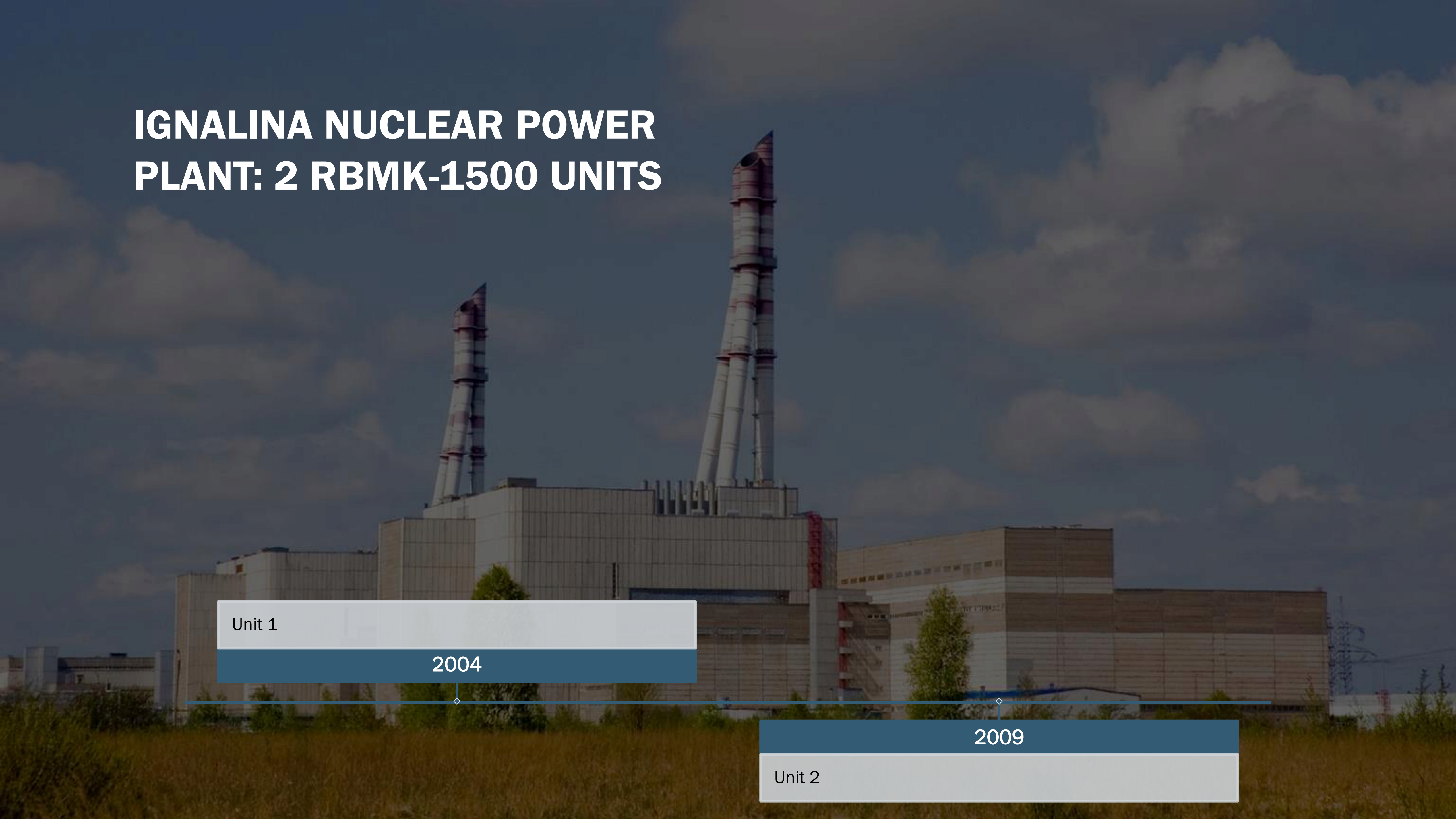
PRESENTATION CONTENT

- Nuclear graphite: function and statistics
- RBMK-1500 case: radiological and structural characteristics
- Rapid analysis method for the ^{14}C specific activity determination
- Ion implantation as a tool to study radiation induced structural changes in materials
- Plans and ideas for the future

NUCLEAR GRADE GRAPHITE

- a purity level < 5 ppm (boron equivalent)
- a density greater than 1.50 g/cm^3
- produced from selected pitch or petroleum coke
- Reflector and moderator

IGNALINA NUCLEAR POWER PLANT: 2 RBMK-1500 UNITS



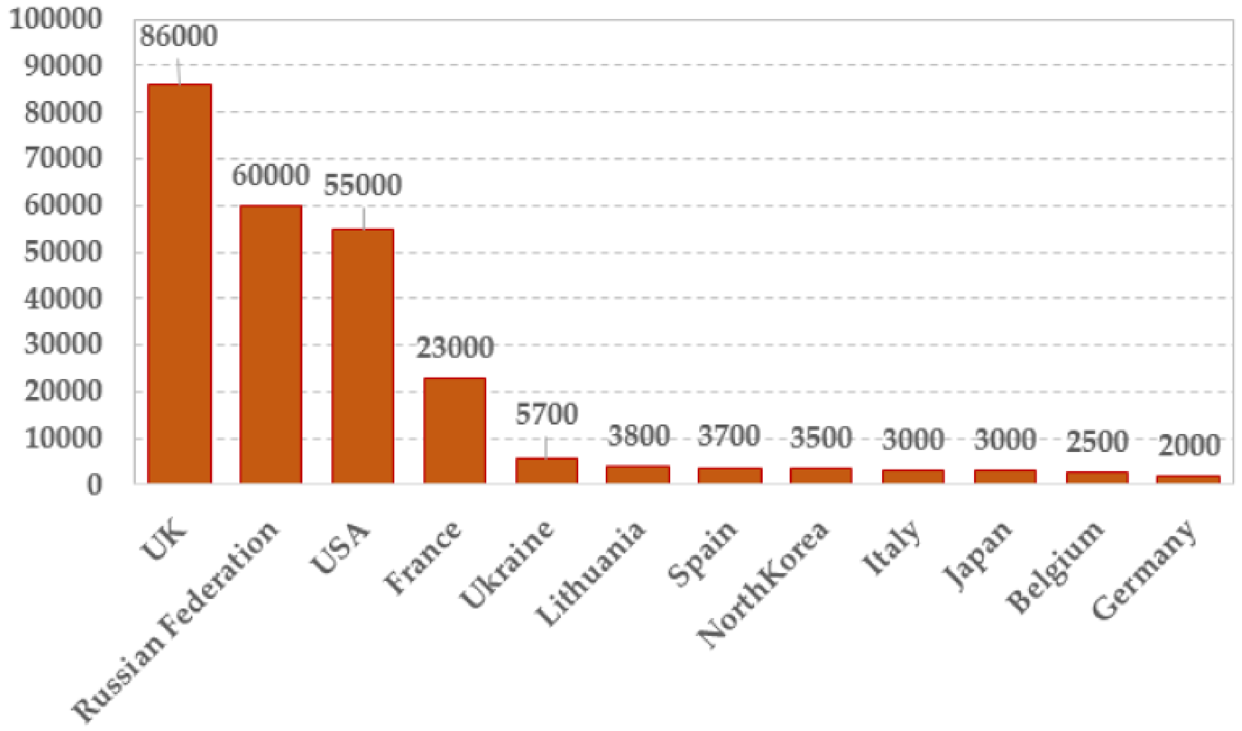
Unit 1

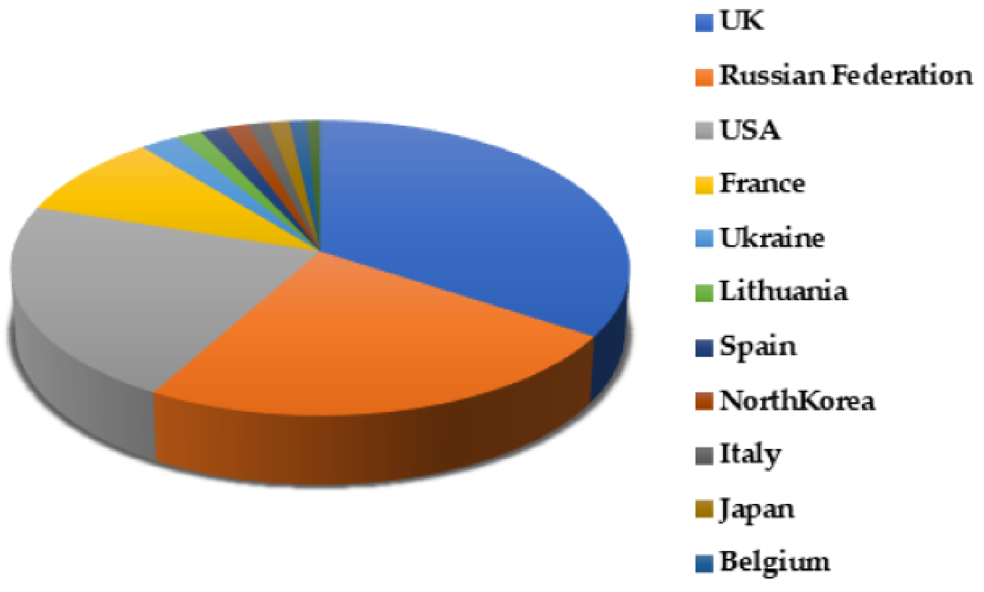
2004

2009

Unit 2

INTRODUCTION





- World resources of irradiated graphite waste by country (tons) (doi:10.3390/en13184638)

RESEARCH AREAS - QUESTIONS

- Radionuclide distribution
- Structural changes
- Radionuclide stability

RESEARCH AREAS - TOOLS

- Modelling (MCNP, SRIM-2013, GEANT4, SCALE, etc.)
- α-,β-,γ-spectrometry; NAA; ICP-MS
- Ion implantation; Raman spectroscopy; SIMS, SEM

RADIONUCLIDES

- Corrosion and activation products (^{57}Co, ^{60}Co, ^{54}Mn, ^{59}Ni, ^{63}Ni, ^{22}Na),
- fission products (^{134}Cs, ^{137}Cs, ^{90}Sr, ^{152}Eu, ^{154}Eu, ^{155}Eu; ^{144}Ce)
- And small amounts of uranium and transuranium radionuclides (^{258}Pu, ^{259}Pu; ^{241}Am, ^{243}Am)

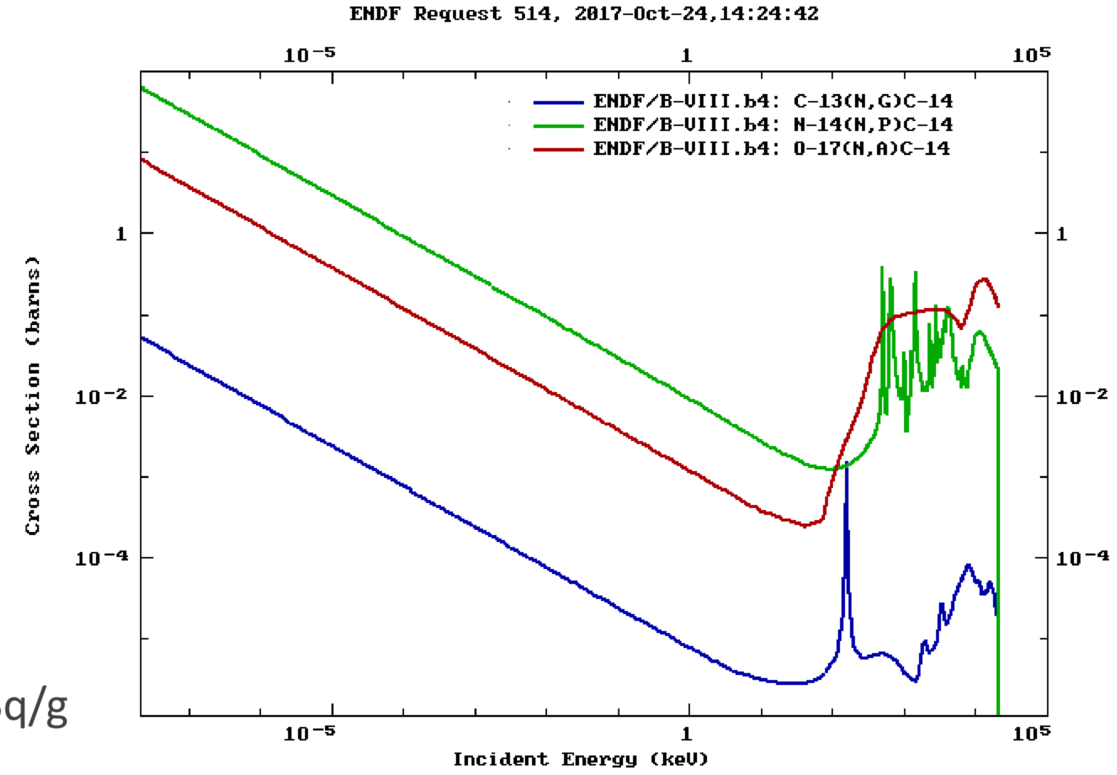
Even if present in small amounts (usually less than 0.01%), after 20–30 years of irradiation in the neutron flux these impurities may still pose a radiological risk to the environment.

(doi:10.3390/en13184638)

^{14}C

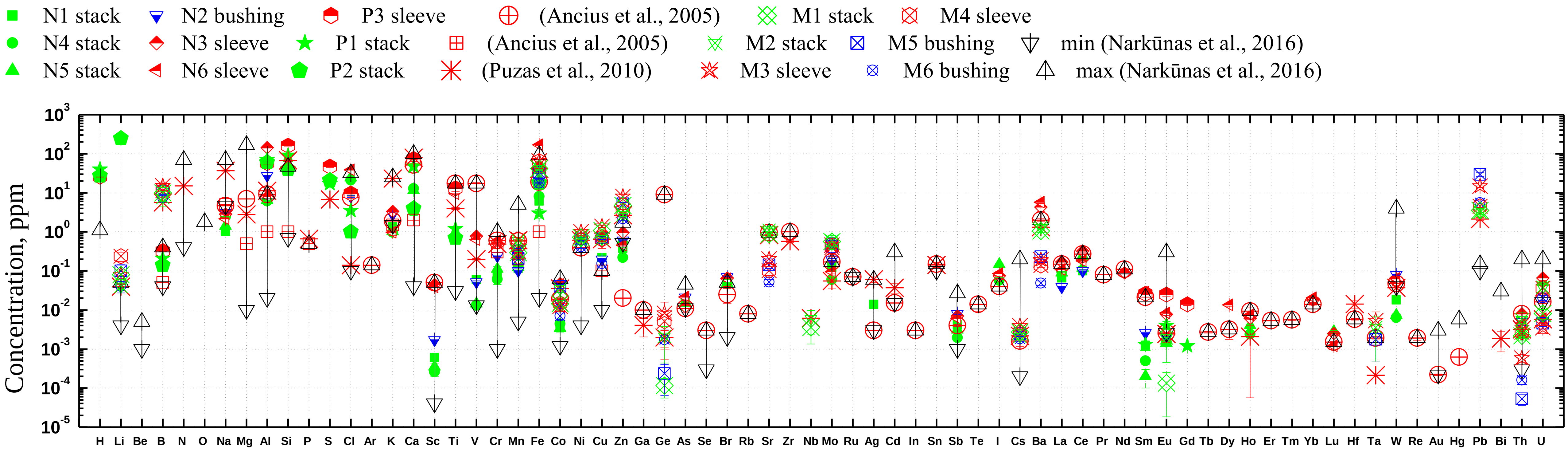
- Limting radionuclide due to long half-life (~5730 years) and possibility to release into the atmosphere

Reaction	Natural abundance	Neutron capture cross-section (barns)		
		$E_n < 1$ eV	Resonance	$E_n > 10$ keV
$^{17}O(n,\alpha)^{14}C$	0.038%	0.235	0.106	0.095
$^{14}N(n,p)^{14}C$	99.6%	1.82	0.818	0.0355
$^{13}C(n,\gamma)^{14}C$	1.1%	1.37×10^{-3}	5.93×10^{-4}	5.16×10^{-5}



- ^{14}C activity in the RBMK-1500 reactor central zone ~10^5 Bq/g

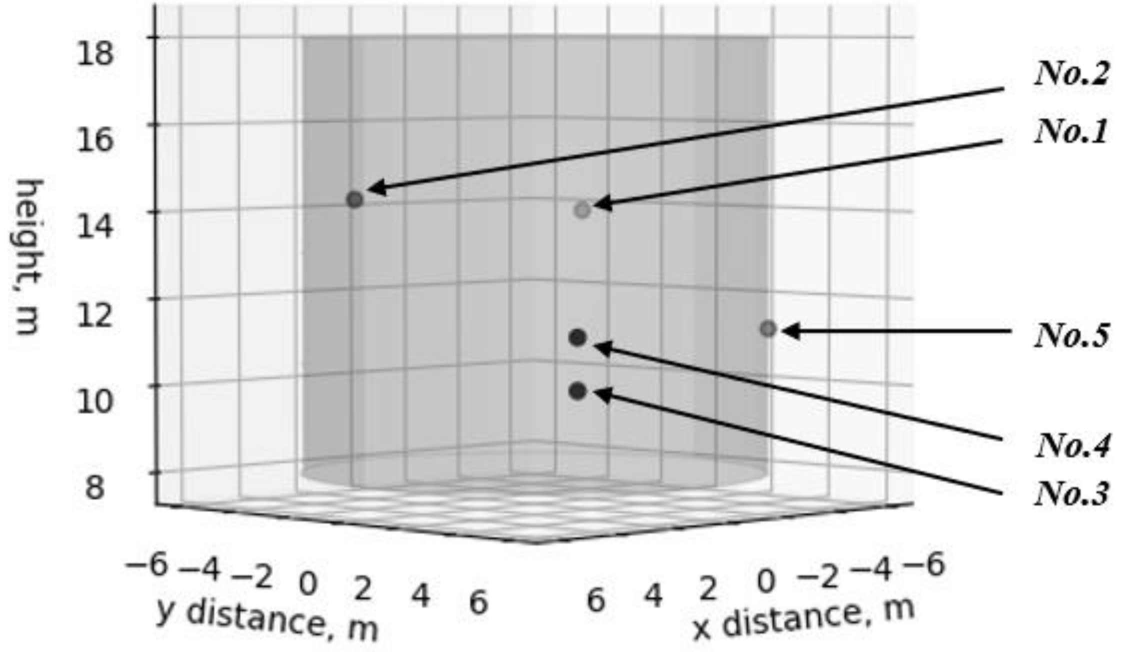
IMPURITIES

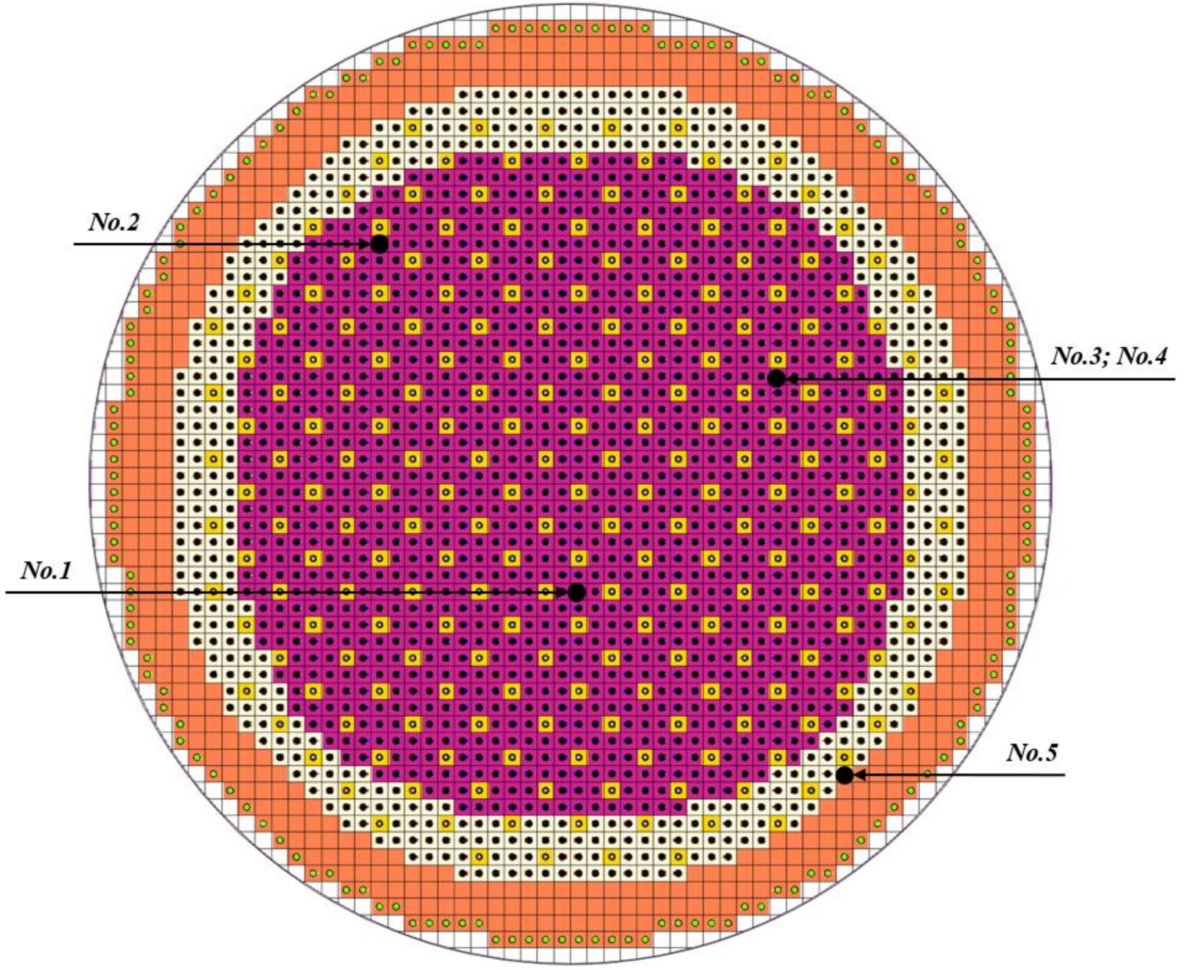


- Glow Discharge Mass Spectrometry (GDMS) in collaboration with CEA (France) (Ancius et.al 2005)
  (ICP-MS) technique (Puzas et al., 2010)
- Prompt gamma activation analysis (PGAA) - Heinz Maier-Leibnitz Zentrum 20 MW water cooled heavy-water moderated thermal neutron flux
- Instrumental neutron activation analysis (INAA) - LVR-15 experimental reactor (Research Centre Řež, Ltd.)

https://doi.org/10.1017/RDC.2018.93

EXPERIMENTAL ANALYSIS





The positions of the samples in the RBMK-1500 reactor core.

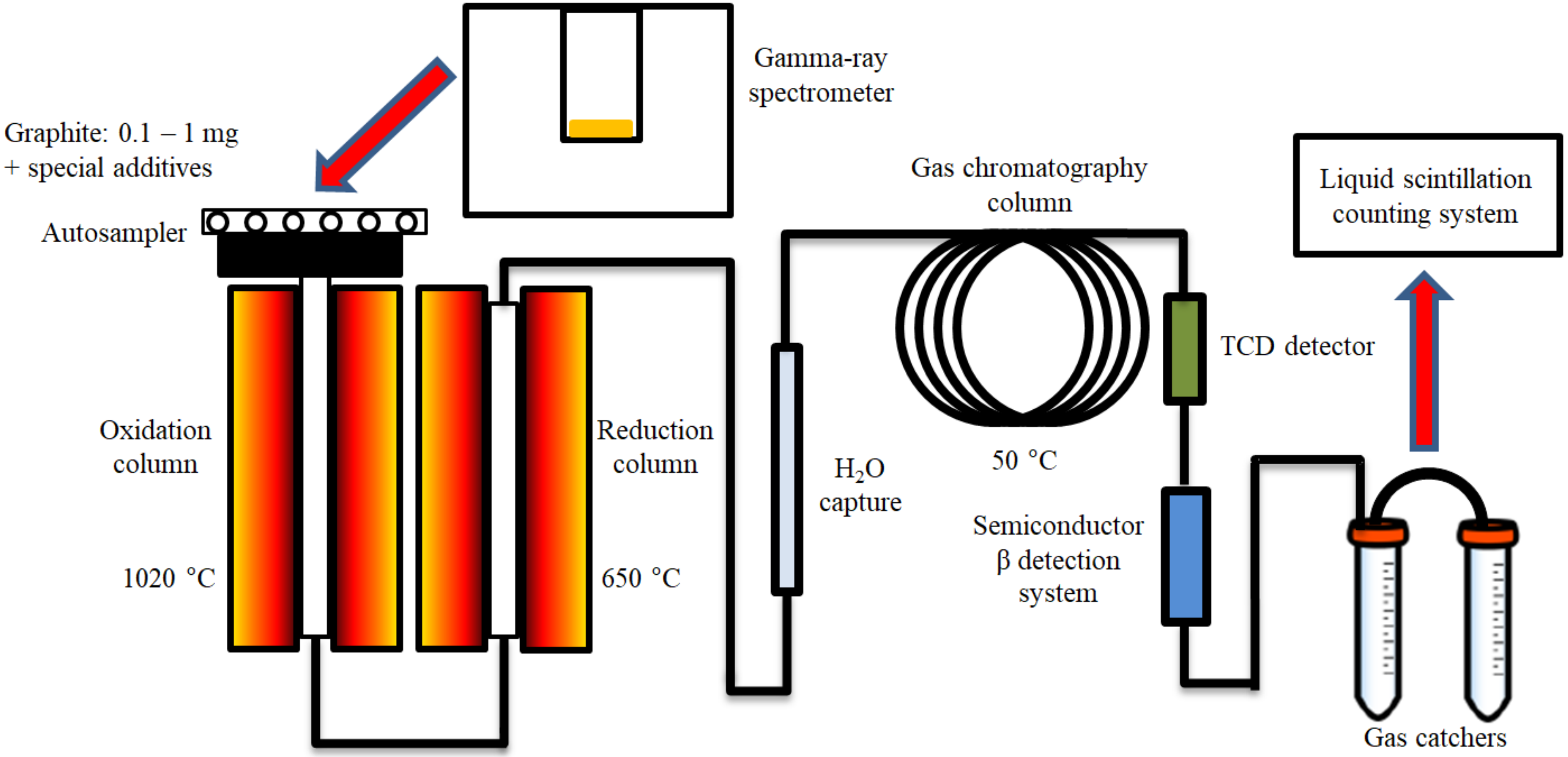
https://doi.org/10.1016/j.net.2021.07.039

ACTIVITY OF GAMMA-RAY EMITTERS

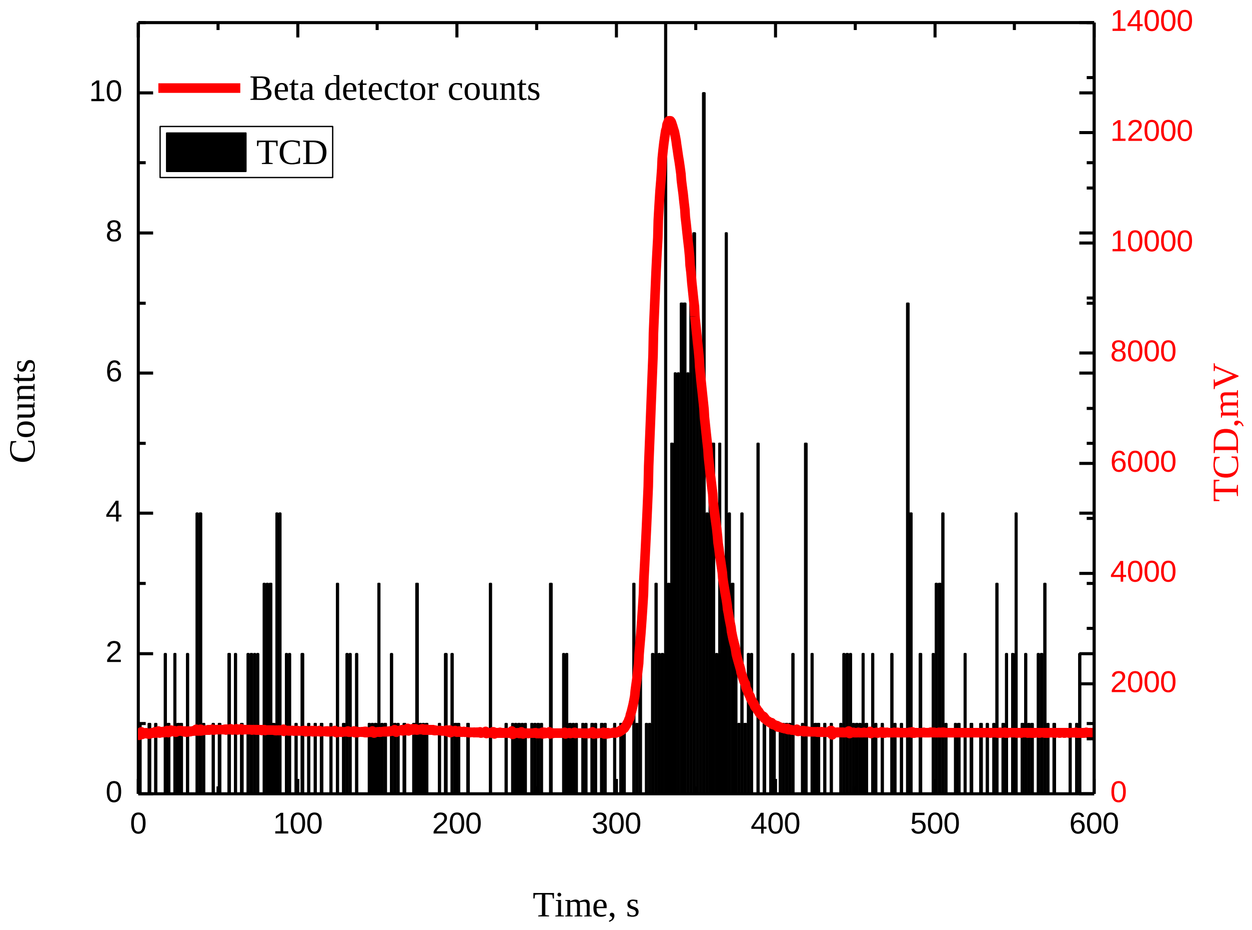
Sample No.	Sample mass, g	Co-60 A, Bq/g	Ba-133 A, Bq/g	Cs-134 A, Bq/g	Cs-137 A, Bq/g	Eu-152 A, Bq/g	Eu-154 A, Bq/g	Eu-155 A, Bq/g
No.1	0.02245	12970±778	153±14	40±4	321±20	< 7.3	83±8	100±7
No.1b	0.00019	39500±3160	363±44		257±24		-	70±10
No.1a	0.000074	2100±168	165±21		145±18		-	-
No.2	0.02333	10830±650	< 3.0	10±2	96±7	< 6.7	< 2.4	< 4.4
No.2b	0.000422	4900±392	-		66±6		-	-
No.2a	0.0001	4560±365	-		73±10		-	-
No.3	0.0251	12540±752	75±7	35±4	1060±64	< 6.1	167±15	173±11
No.3a	0.000278	21700±1740	112±15		987±60		354±58	111±11
No.3b	0.00038	33500±2680	341±42		1370±83		517±58	393±25
No.4	0.01869	1351±82	< 1.5	< 2.5	66±4	< 3,1	< 1.4	< 2.5
No.4a	0.000152	4310±345	-		139±16		-	-
No.4b	0.000204	2100±168	-		70±7		-	-
No.5	0.02041	4010±241	13±2	17±2	91±6	< 4.5	78±7	80±5
No.5b	0.000414	4240±340	-		61±5		-	-
No.5a	0.000272	331±27	-		102±8		-	-

https://doi.org/10.1016/j.net.2021.07.039

RAPID ANALYSIS METHOD FOR THE ^{14}C SPECIFIC ACTIVITY DETERMINATION



https://doi.org/10.1371/journal.pone.0191677

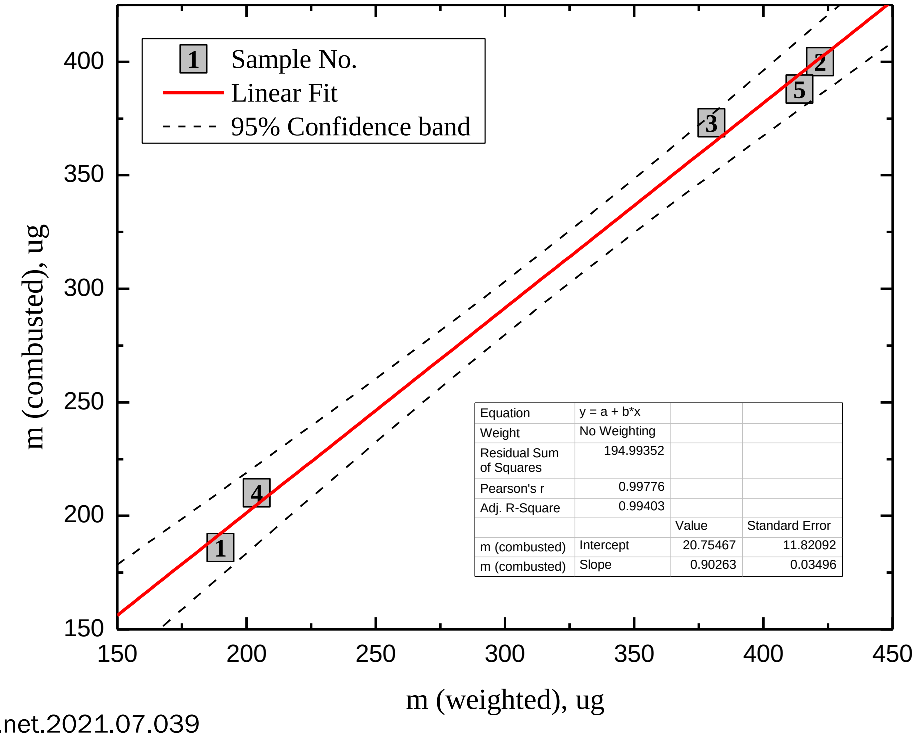


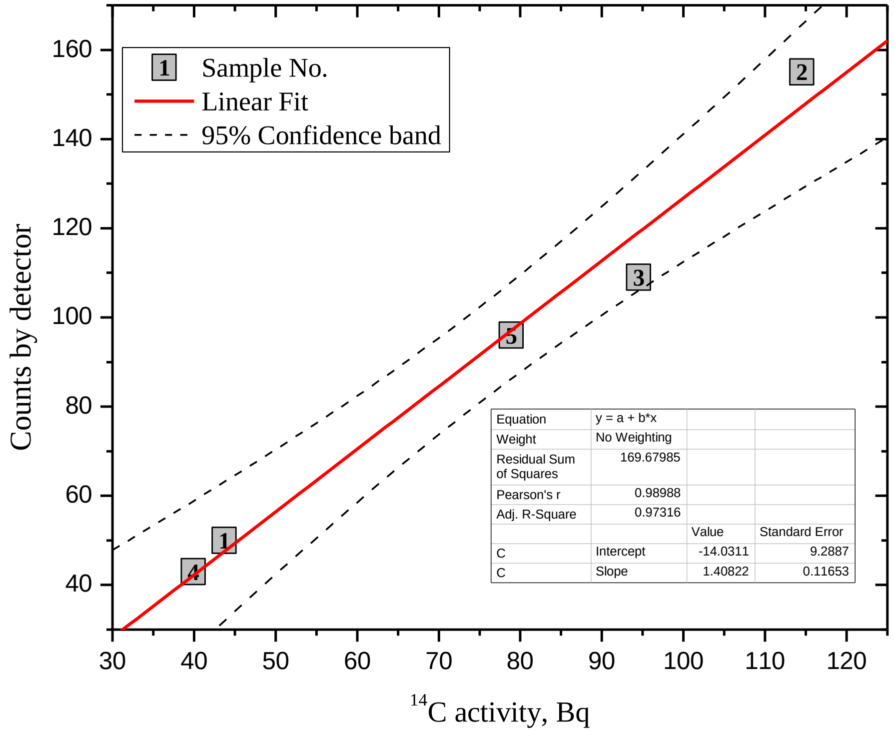
The TCD signal indicating intensity of CO_2 flow and β counts registered by semiconductor detector during the combustion of the graphite sample.

https://doi.org/10.1016/j.net.2021.07.039

^{14}C ACTIVITY IN IRRADIATED RBMK-1500 GRAPHITE SAMPLES

Sample No.	Sample mass, µg		^{14}C activity, Bq	^{14}C activity, Bq/g
	By weighing	By CO_2 amount		
No.1b	190	186	43.7 ± 2.5	$(2.30 \pm 0.13) \times 10^5$
No.2b	422	400	114.5 ± 6.5	$(2.71 \pm 0.16) \times 10^5$
No.3b	380	373	94.5 ± 5.5	$(2.49 \pm 0.14) \times 10^5$
No.4b	204	210	39.9 ± 2.3	$(1.96 \pm 0.11) \times 10^5$
No.5b	414	388	78.9 ± 4.6	$(1.91 \pm 0.11) \times 10^5$





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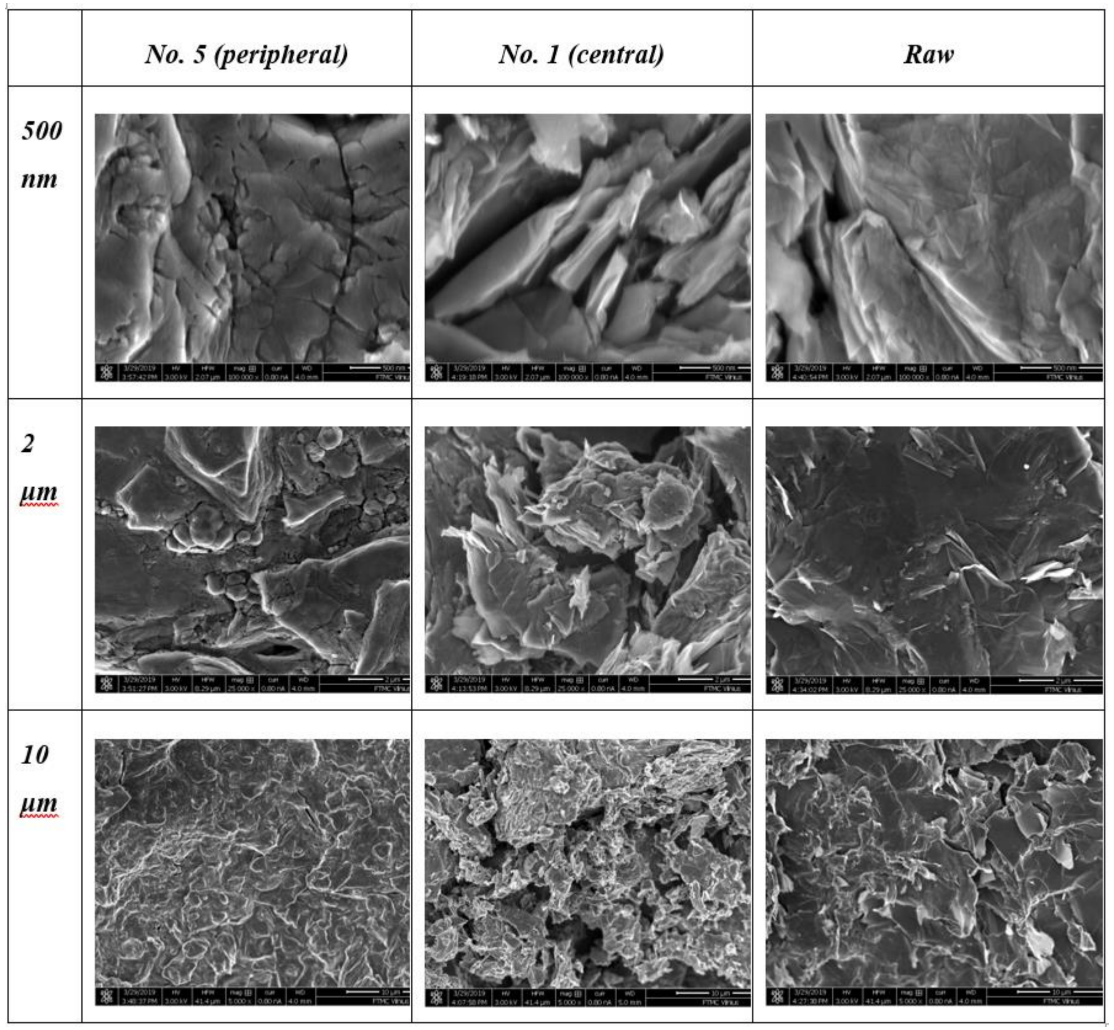
THE FULL SCALE MCNP6 3D MODEL OF RBMK-1500 CORE

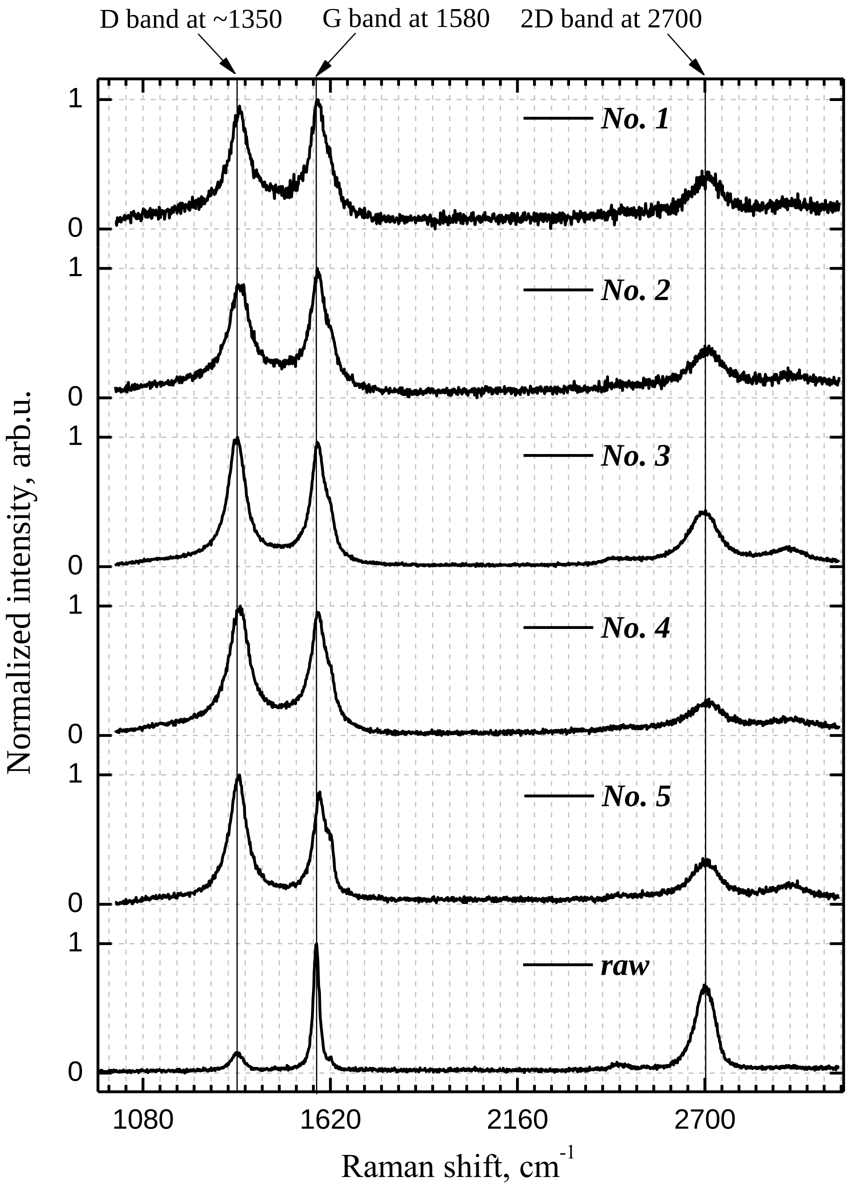
Graphite construction	Flux, n/cm^2*s	^{14}C A, Bq/g
Graphite stack (plateau region)	1.0×10^{14}	1.44×10^{5}
Graphite stack top&bottom	1.4×10^{13}	3.24×10^{4}
Graphite sleeve	9.9×10^{13}	1.16×10^{5}
Periphery graphite stack	3.0×10^{13}	4.26×10^{4}
Periphery graphite stack top&bottom	4.1×10^{12}	9.67×10^{3}
Graphite reflector	9.7×10^{12}	2.38×10^{4}
Graphite reflector top& bottom	1.4×10^{12}	3.71×10^{3}
Graphite reflector with cooling channel	1.2×10^{12}	3.26×10^{3}
Graphite reflector with cooling top&bottom	1.8×10^{11}	5.07×10^{2}
Graphite CPS channel	9.0×10^{13}	1.60×10^{5}
Graphite CPS channel top& bottom	1.2×10^{13}	2.81×10^{4}

The results of modelling of neutron flux and 14C activity in the RBMK-1500 reactor graphite constructions for average power of 2152MW (for 2018-01-01 date)

https://doi.org/10.1016/j.net.2021.07.039

SEM IMAGES AND RAMAN SPECTRA





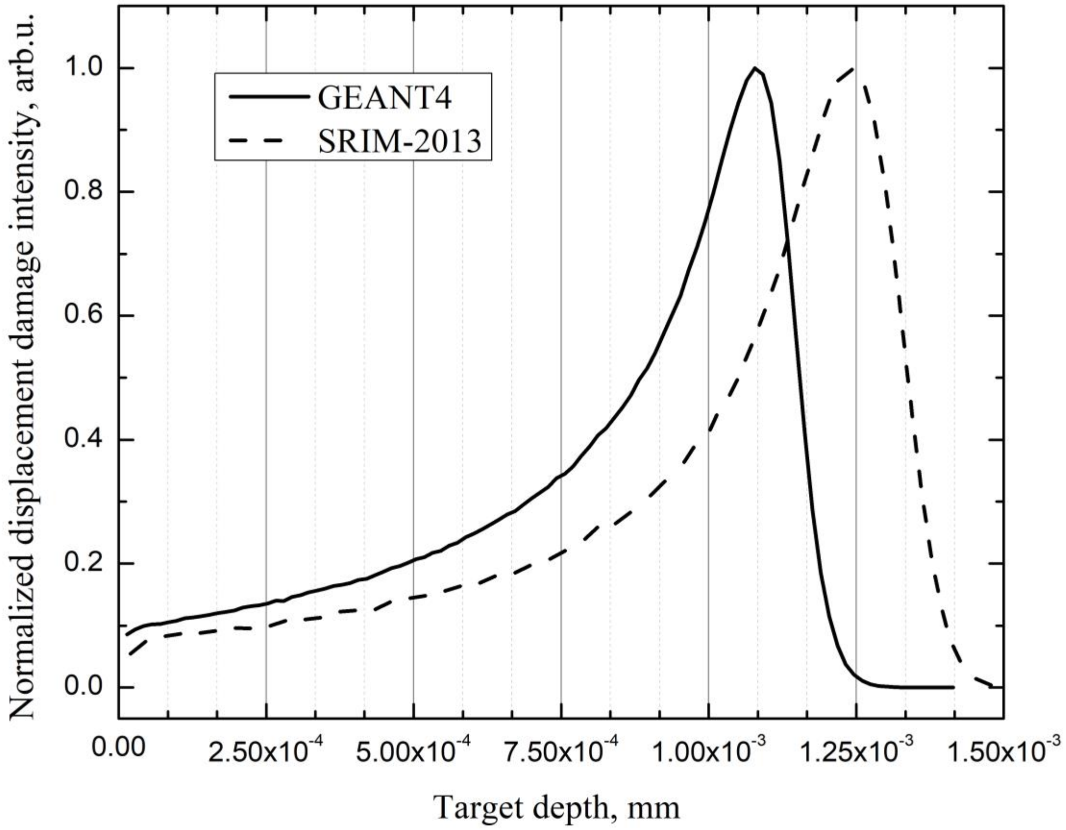
https://doi.org/10.1016/j.net.2021.07.039

ION IMPLANTATION AS A TOOL TO STUDY RADIATION INDUCED STRUCTURAL CHANGES IN MATERIALS

- no sample activation
- time saving
- a variety of choice (ion type, energy, fluence)

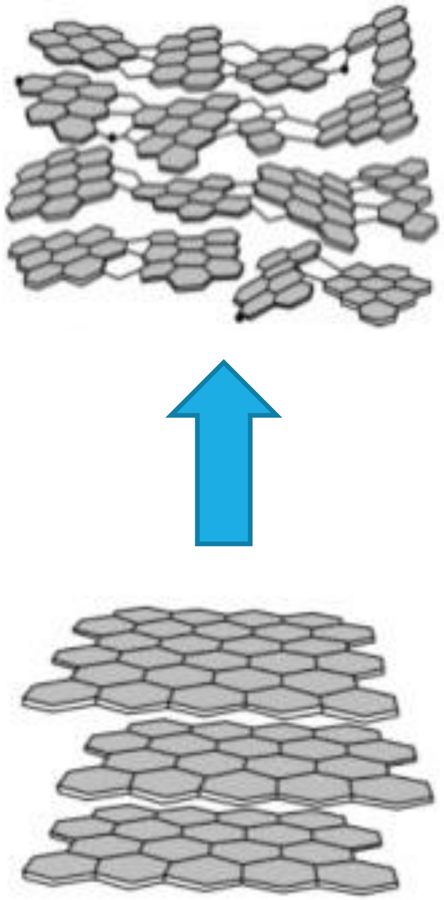
Tandetron 4110A

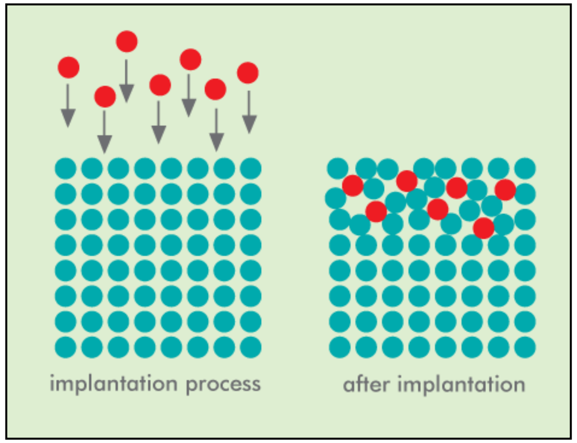
IMPLANTATION PARAMETERS: GEANT4 AND SRIM-2013 MODELLING



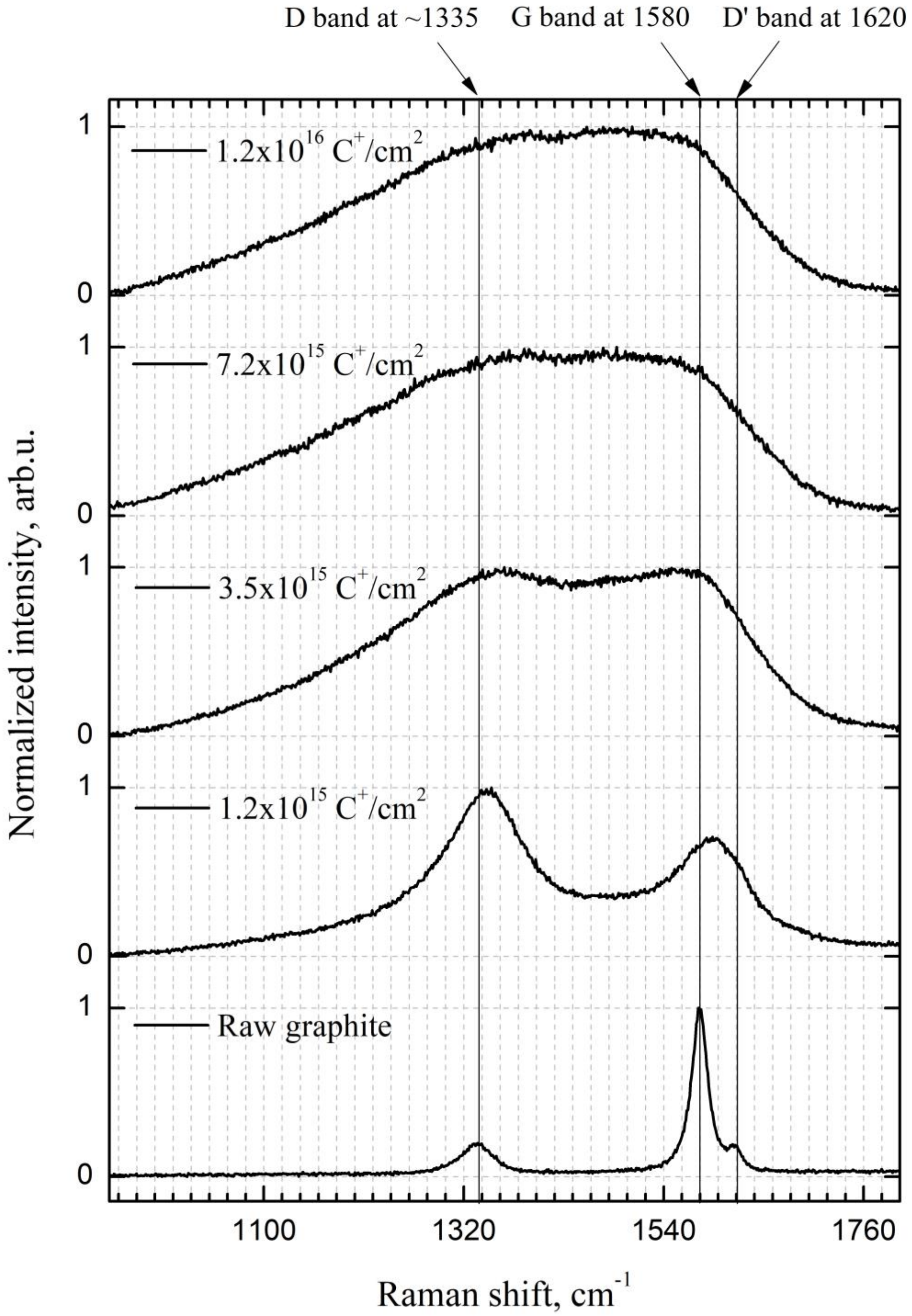
https://doi.org/10.1016/j.nimb.2019.01.049

700 keV $^{12}C^{+}$ ION IMPLANTATION



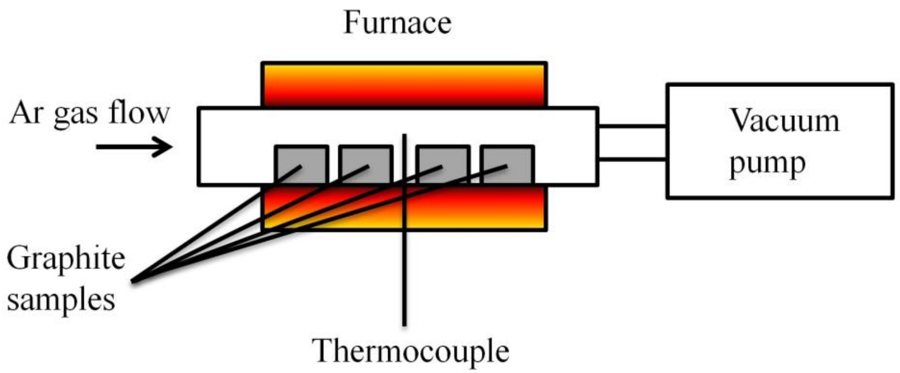


G band : C-C bond
D band : sp3 hybridization, point defects
D1 band : amorphous carbon
D´ band: specific/complex defects



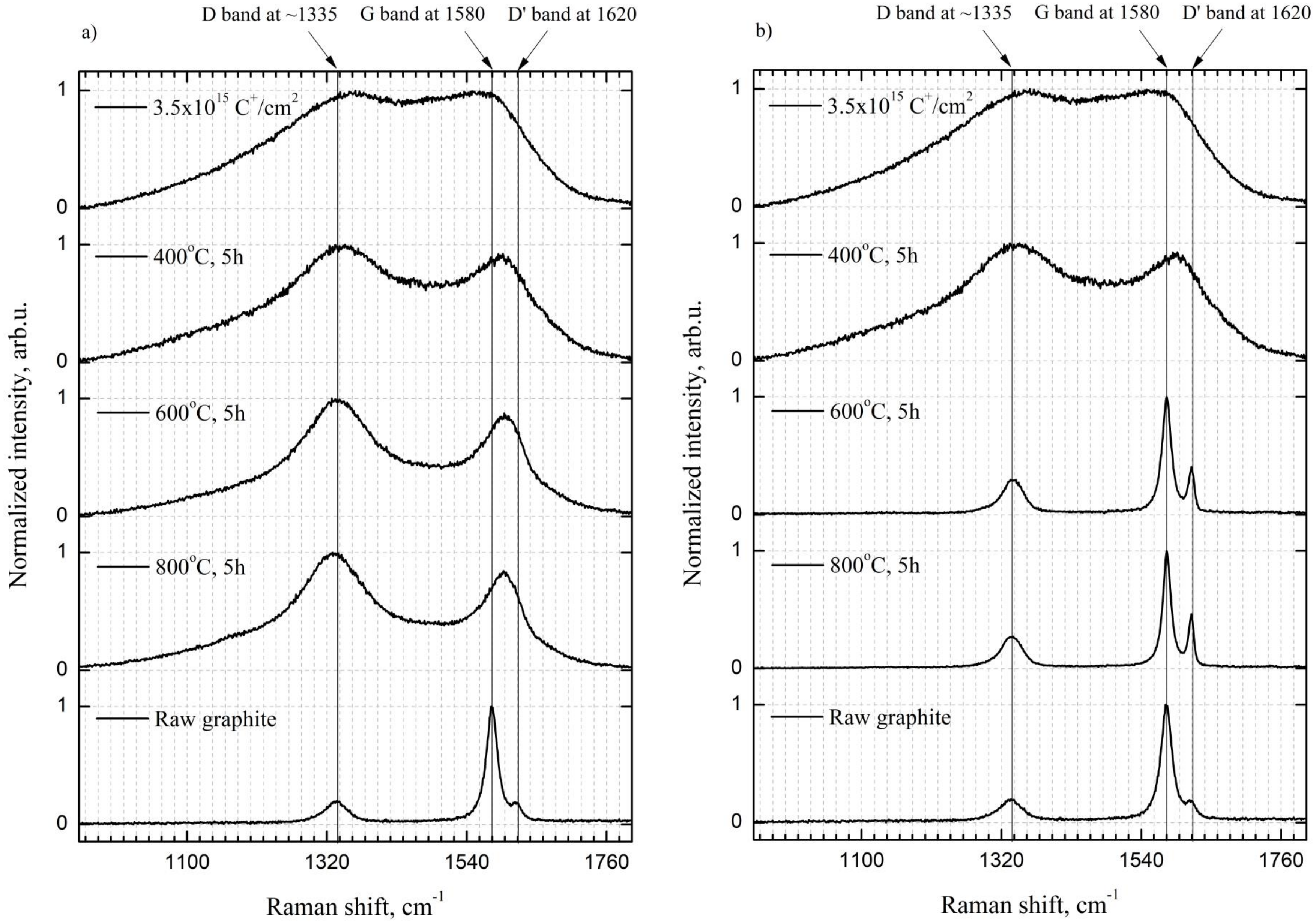
https://doi.org/10.1016/j.nimb.2019.01.049

$^{12}C^+$ ION IMPLANTATION: ANNEALING



https://doi.org/10.1016/j.nimb.2019.01.049

$^{12}C^{+}$ ION IMPLANTATION: ANNEALING

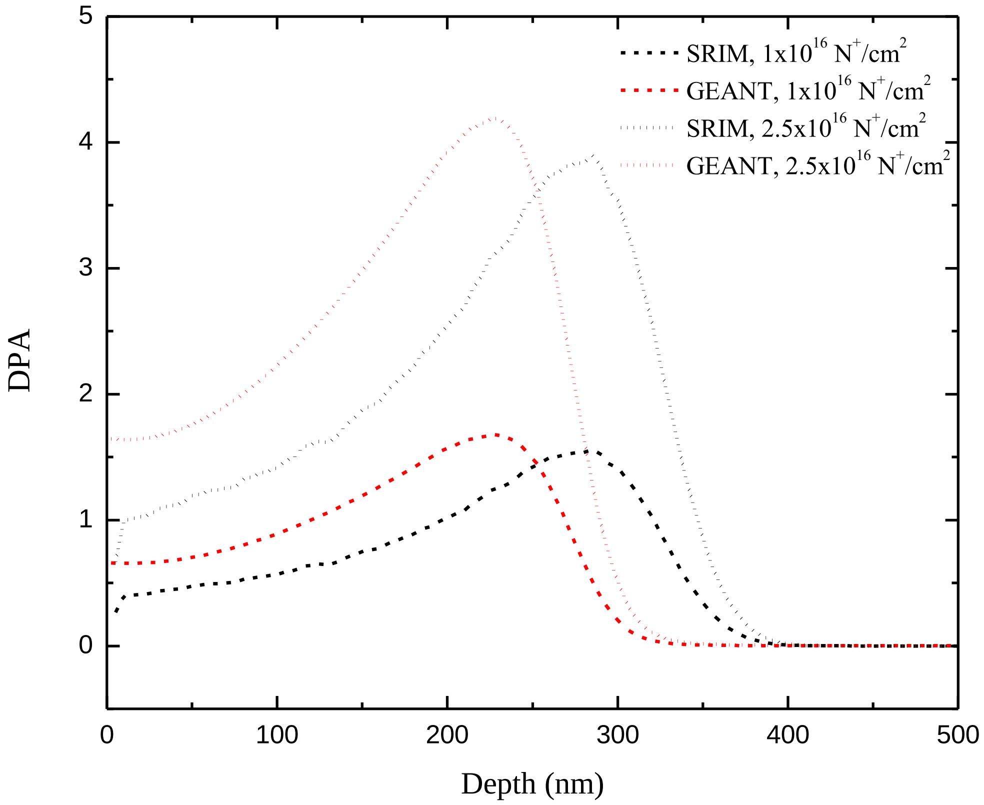


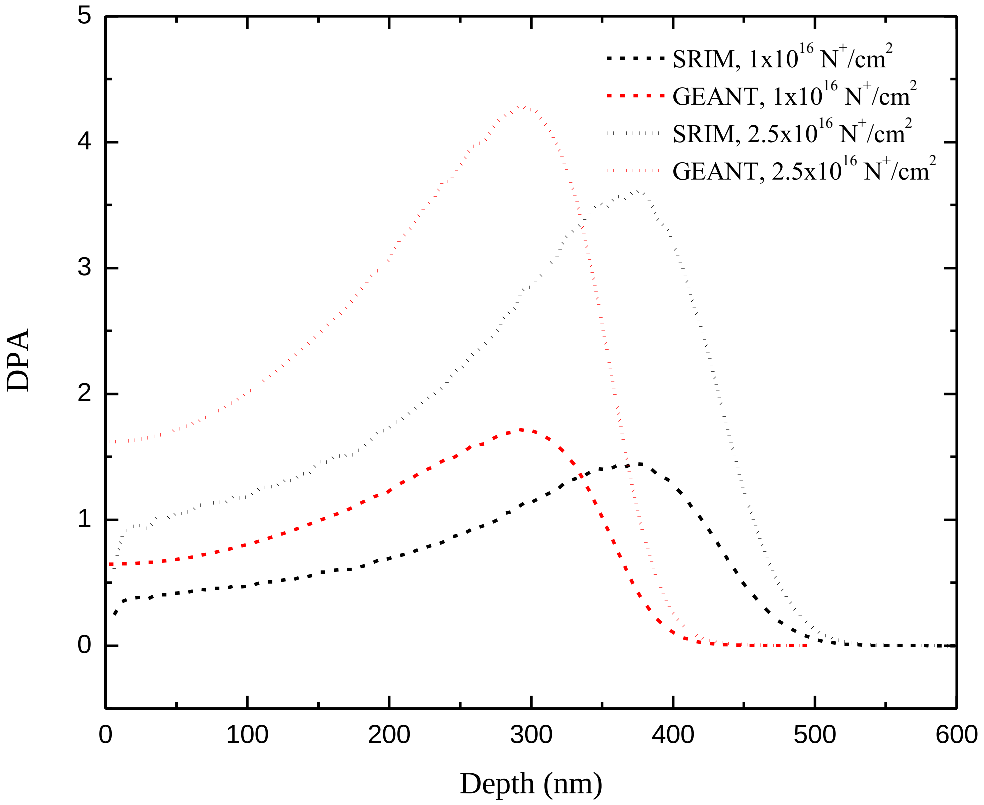
https://doi.org/10.1016/j.nimb.2019.01.049

$^{14}N^{+}$ ION IMPLANTATION

- $^{14}N^{+}$ ions at the energy of 180 keV
- fluence of 1.0×10^{16} ions/cm^2 and 2.5×10^{16} ions/cm^2
- Temperature: RT and 500°C
- Graphite: HOPG and RBMK

$^{14}N^{+}$ ION IMPLANTATION: SRIM-2013 AND GEANT4 MODELLING

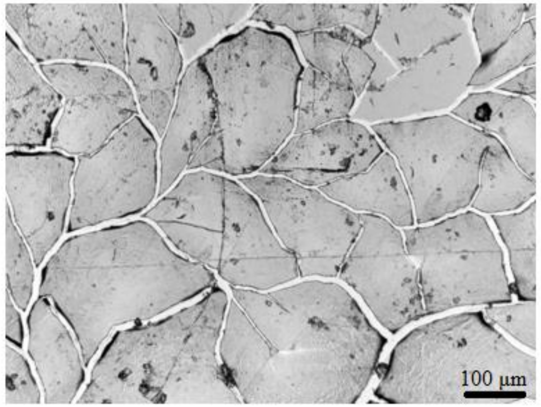
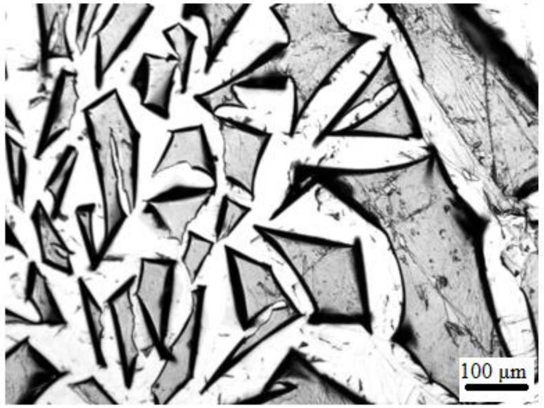
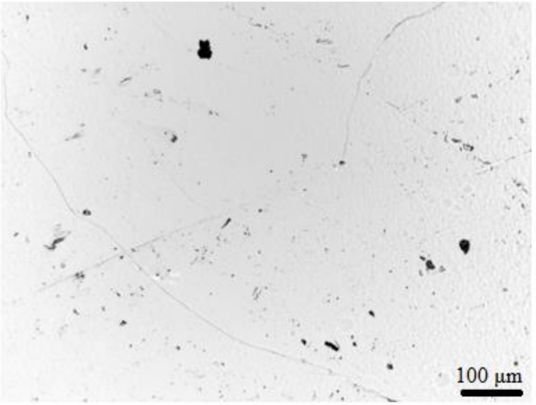
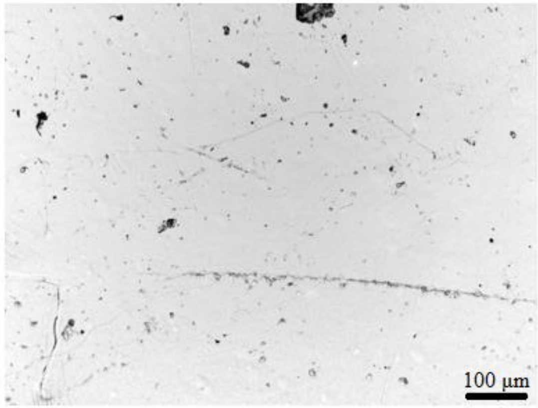




SRIM-2013 AND GEANT4 MODELLING

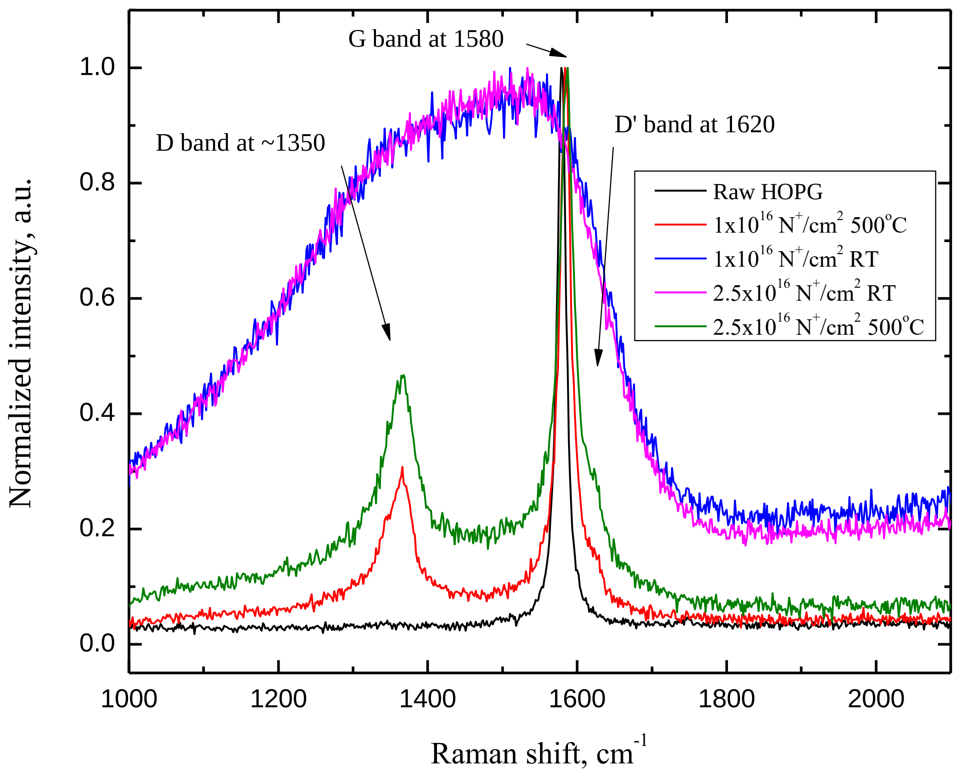
Graphite type	HOPG	RBMK
Density	2.25 g/cm^3	1.7 g/cm^3
Projected range of ions: SRIM-2013 GEANT4	 299 ±45 nm 292 ±58 nm	 397 ± 60 nm 368 ± 60 nm
Maximum number of displacements per atom (DPA) SRIM-2013 GEANT4	 1.75 (1×10^{16} ions/cm^2) 4.37 (2.5×10^{16} ions/cm^2) 1.88 (1×10^{16} ions/cm^2) 4.70 (2.5×10^{16} ions/cm^2)	 1.61 (1×10^{16} ions/cm^2) 4.04 (2.5×10^{16} ions/cm^2) 1.93 (1×10^{16} ions/cm^2) 4.82 (2.5×10^{16} ions/cm^2)
Average number of defects on the surface (~50 nm) (DPA) SRIM-2013 GEANT4	 0.46 (1×10^{16} ions/cm^2) 1.13 (2.5×10^{16} ions/cm^2) 0.75 (1×10^{16} ions/cm^2) 1.88 (2.5×10^{16} ions/cm^2)	 0.41 (1×10^{16} ions/cm^2) 1.03 (2.5×10^{16} ions/cm^2) 0.74 (1×10^{16} ions/cm^2) 1.86 (2.5×10^{16} ions/cm^2)
Average amount of defects in the nuclear reactor due to the neutron damage (DPA/full power year)	-	0.51

$^{14}N^{+}$ ION IMPLANTATION: MICROSCOPY IMAGES

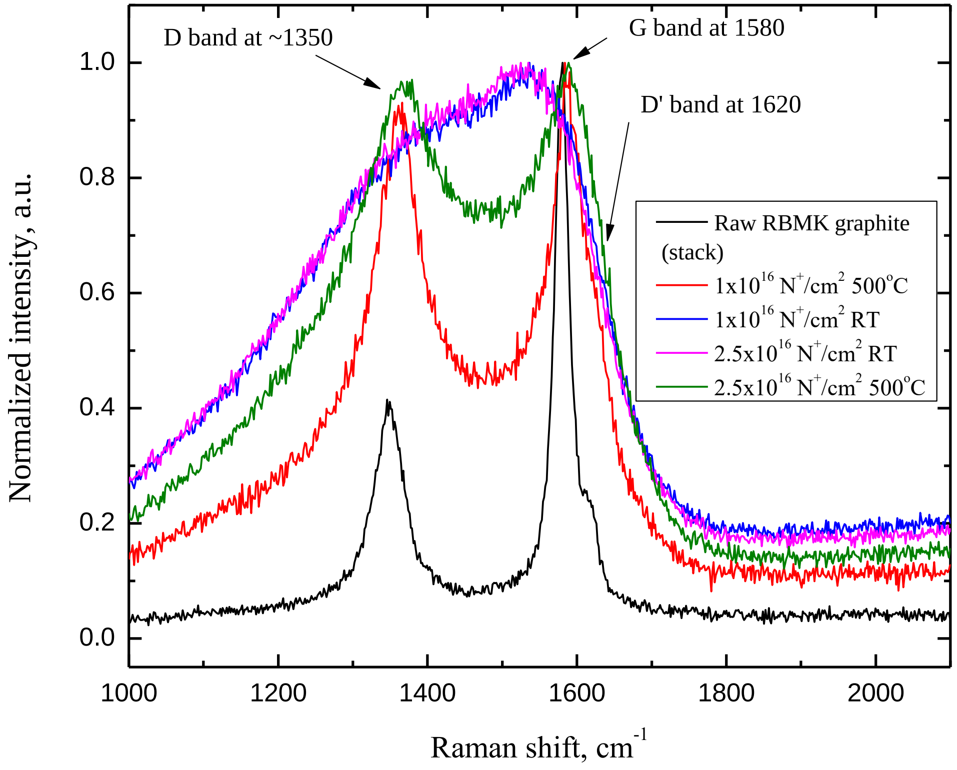
	1.0×10^{16} ions/cm^2	2.5×10^{16} ions/cm^2
RT		
500°C		

$^{14}N^{+}$ ION IMPLANTATION: TEMPERATURE EFFECTS

HOPG

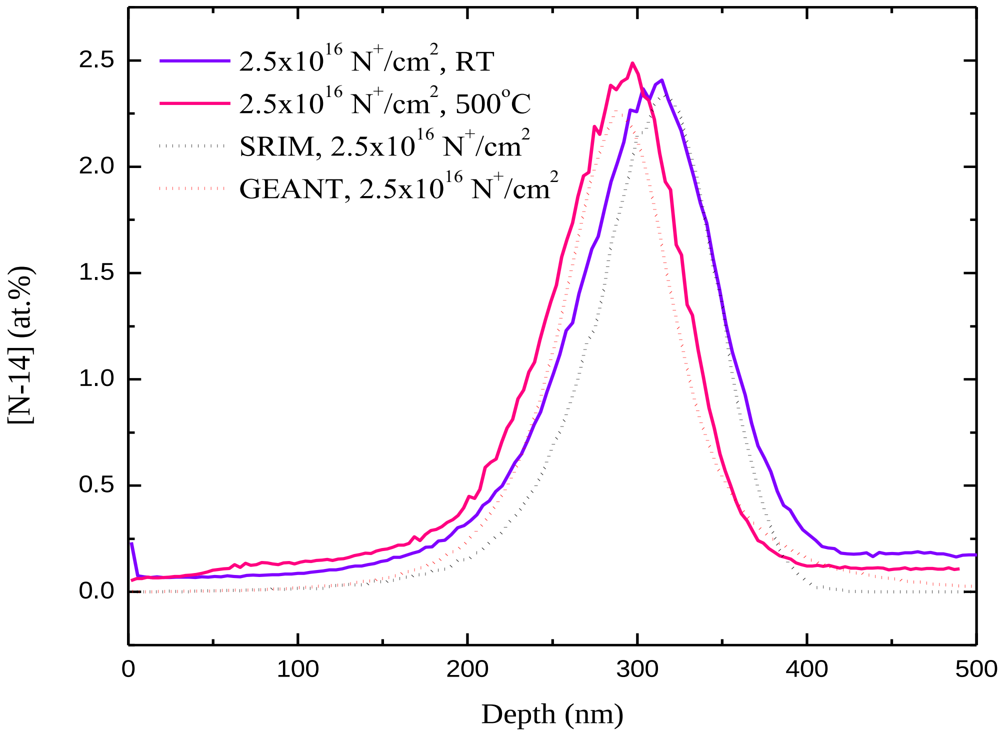


RBMK

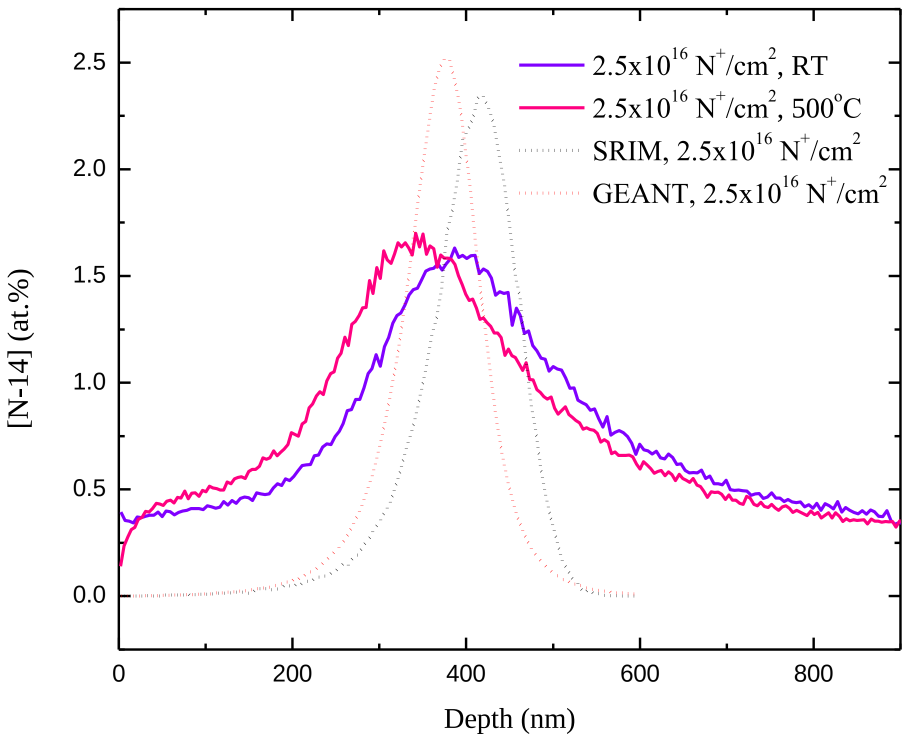


$^{14}N^{+}$ ION IMPLANTATION: NITROGEN PROFILE (SIMS)

HOPG



RBMK



CONCLUSIONS

- Radionuclide production in the reactor core is highly dependent on impurity concentrations. Distribution of γ-ray emitters is non-homogeneous in the irradiated RBMK graphite matrix; the main γ-ray emitters are ^{60}Co and ^{137}Cs.
- Measured ^{14}C specific activity values in the irradiated RBMK-1500 graphite samples vary from 190 kBq/g to 270 kBq/g. This corresponds to 25−35 ppm ^{14}N impurity concentration.
- Operational temperature of nuclear reactor (~350-550 ºC) is high enough to ensure that both defect creation and structural reordering process occur at the same time. Due to this, the full amorphization of the crystal matrix is avoided and the functional properties of graphite are not lost, which ensures proper functionality of the material.
- During the dynamical annealing of the graphite structure in the operating RBMK-1500 reactor, most of the $^{14}N(n,p)^{14}C$ reaction dependent ^{14}C is immobilized in the graphite lattice. This analysis of ^{14}C behaviour in RBMK graphite suggests that the graphite matrix is a highly effective ^{14}C dispersion barrier, which is one of the determining factors in the assessment of the surface/geological storage choice strategy.

PLANS AND IDEAS FOR THE FUTURE

- Investigation of the spatial distribution of ^{14}C activity in the irradiated RBMK-1500 reactor graphite
- Improvemets of the proposed ^{14}C specific activity determination method
- Characterisation of the metal waste: determination of the surface contamination vs. activation induced activity

THANK YOU FOR YOUR ATTENTION!

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