Characterization of ^{14}C in neutron irradiated RBMK-1500 graphite

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Nuclear graphite waste management strategy during decommissioning of Ignalina NPP is the pending decision in Lithuania. In the RBMK type reactor graphite is a neutron moderator and reflector. The total mass of radioactive graphite from the both Ignalina NPP units is up to 3,800 t. ^{14}C is the limiting radionuclide for long-term disposal of irradiated graphite due to half-life of 5730 years and relatively high activity as well as mobility in geological media. Characterization of irradiated graphite in terms of both ^{14}C activity and chemical bonds in the lattice is crucial for the optimization of treatment technology (e.g. geological disposal, landfill storage, recycling, etc.). For this purpose numerical simulations and experimental analysis are performed.

RBMK-1500 graphite

Experimentally validated numerical 3D model of RBMK-1500 (MCNP6 and SCALE 6.1.) is effective for description of the change of radiological characteristics of different parts of nuclear reactor during operation and decommissioning periods [1]. Both experimental measurements and modeling data are used for scaling factor determination [2], which subsequently could be used for sorting of spent graphite radioactive waste.

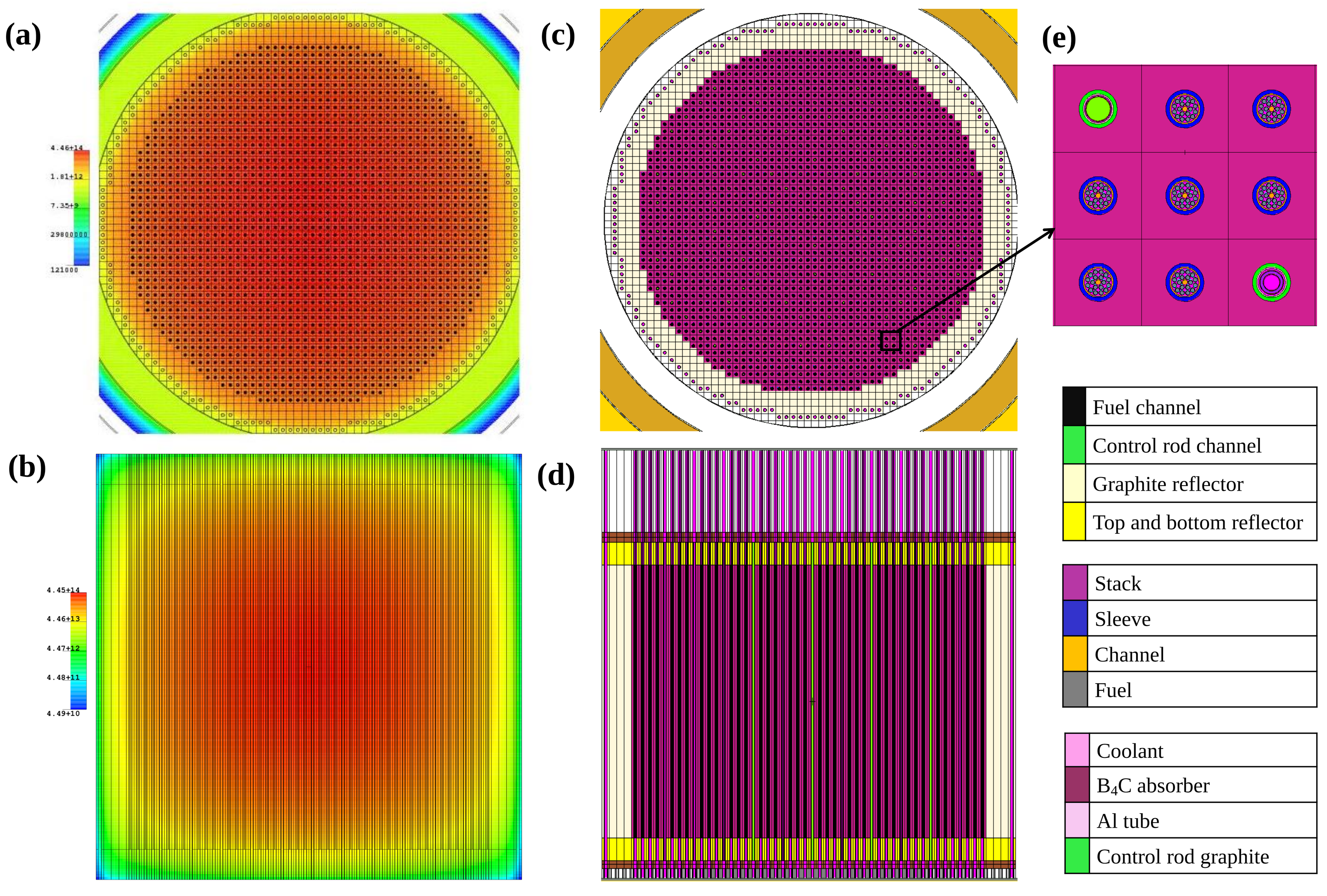

Fig. 1. (a) Neutron flux distribution in the RBMK reactor graphite horizontal and **(b)** vertical cross sections. **(c)** Horizontal cross section full scale reactor core; **(d)** vertical cross section of reactor core with bottom, top reflectors, metal plates and cooling tubes system on the top. **(e)** Magnified view of RBMK-1500 core fragment (3x3) with fuel assemblies, inserted and extracted control rods.

Determination of ^{14}C specific activity in irradiated graphite

^{14}C measurements are usually carried out by using liquid scintillation counting (LSC) technique after time consuming sample preparation procedure. Recently we proposed an express analysis method for the specific ^{14}C activity determination in small graphite samples in the range of 1-100 µg [3]. This method is based on the graphite sample combustion in the commercial elemental analyzer and determination of ^{14}C specific activity by using the semiconductor detectors. This method is planned to apply for determination of the graphite homogeneity profile in terms of ^{14}C activity.

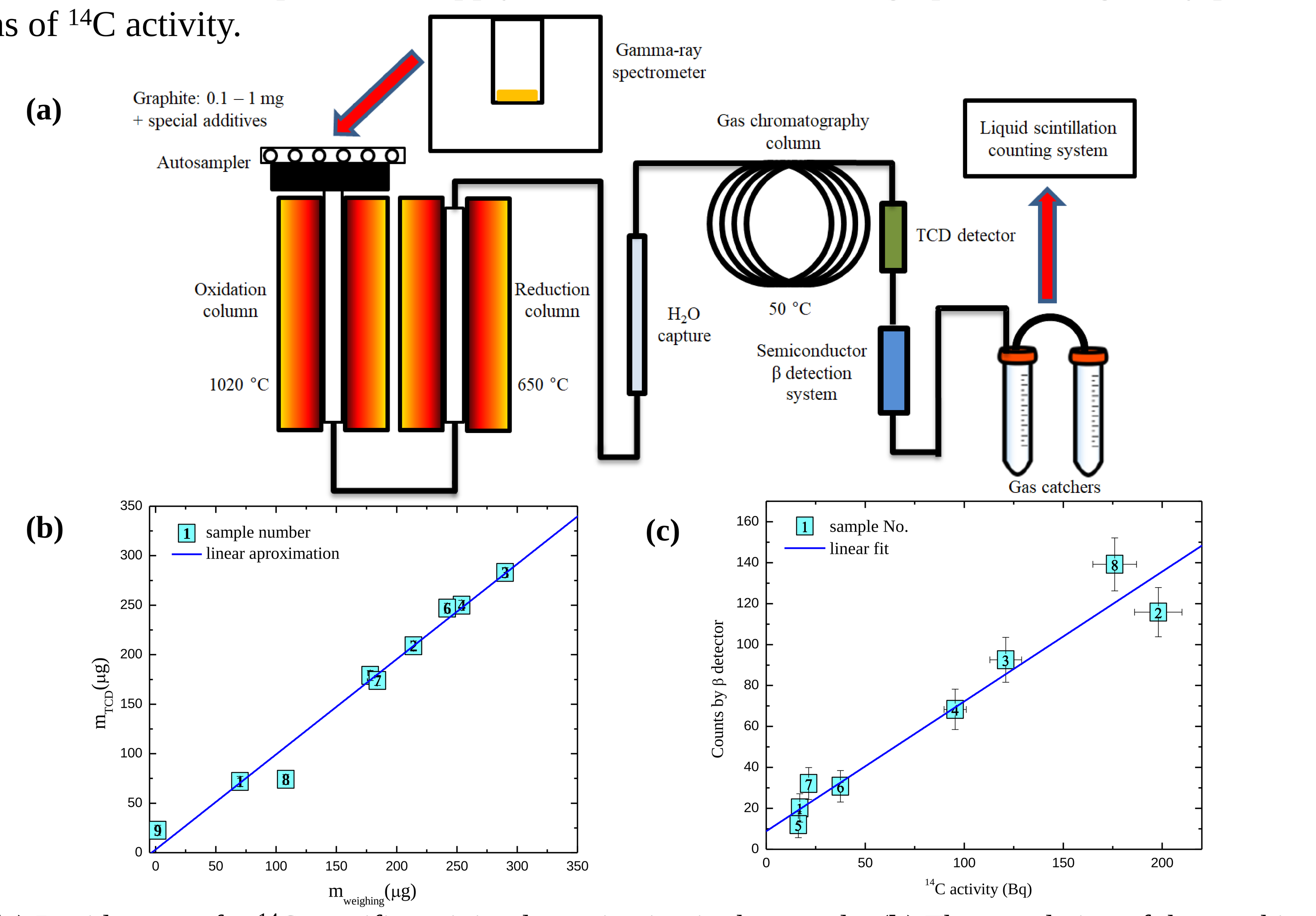

Fig. 2. (a) Rapid system for ^{14}C specific activity determination in the sample. **(b)** The correlation of the graphite sample mass as determined by two independent methods: weighing and combustion in the elemental analyzer. **(c)** Correlation between LSC and semiconductor detector data.

$^{12}C^+$ ion implantation and thermal treatment

For further graphite treatment technology optimization the structural investigations of graphite should be performed. ^{14}C mobility and position in graphite matrix is determined by neutron irradiation in the reactor at certain operation conditions. In order to understand the processes in the irradiated graphite we observe the propagation of defects induced by $^{12}C^+$ ion implantation at energy of 700 keV at varying fluences. The structural changes after implantation and thermal treatment later on are investigated by Raman spectroscopy. The SRIM-2013 code is also used to estimate the damage profile in the surface of the graphite samples.

Theoretical calculations (SRIM) → **Sample preparation** → **Ion implantation (ion accelerator Tandetron 4110A)** → **Raman characterization** → **Thermal treatment** → **Raman characterization**

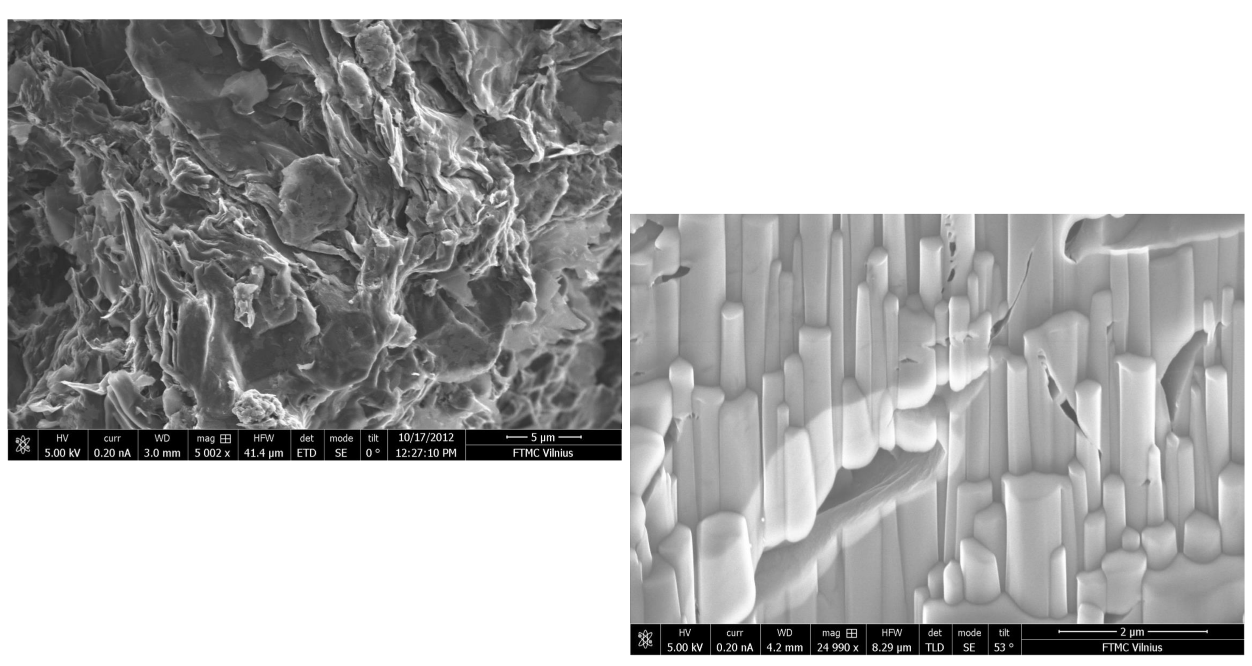

Fig. 3. SEM images of raw RBMK-1500 stack graphite samples.

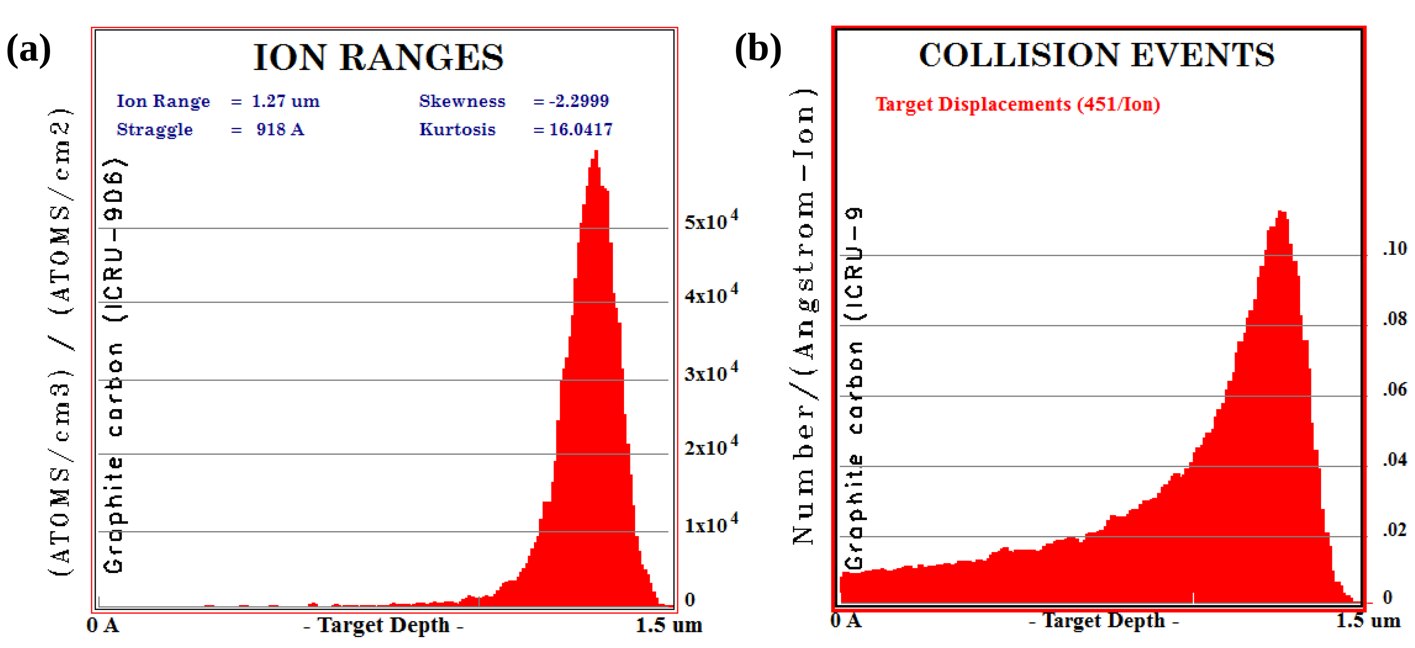

Fig. 4. 700 keV $^{12}C^+$ ion implantation parameters according to SRIM: **(a)** the projected range of implanted ions; **(b)** the damage profile.

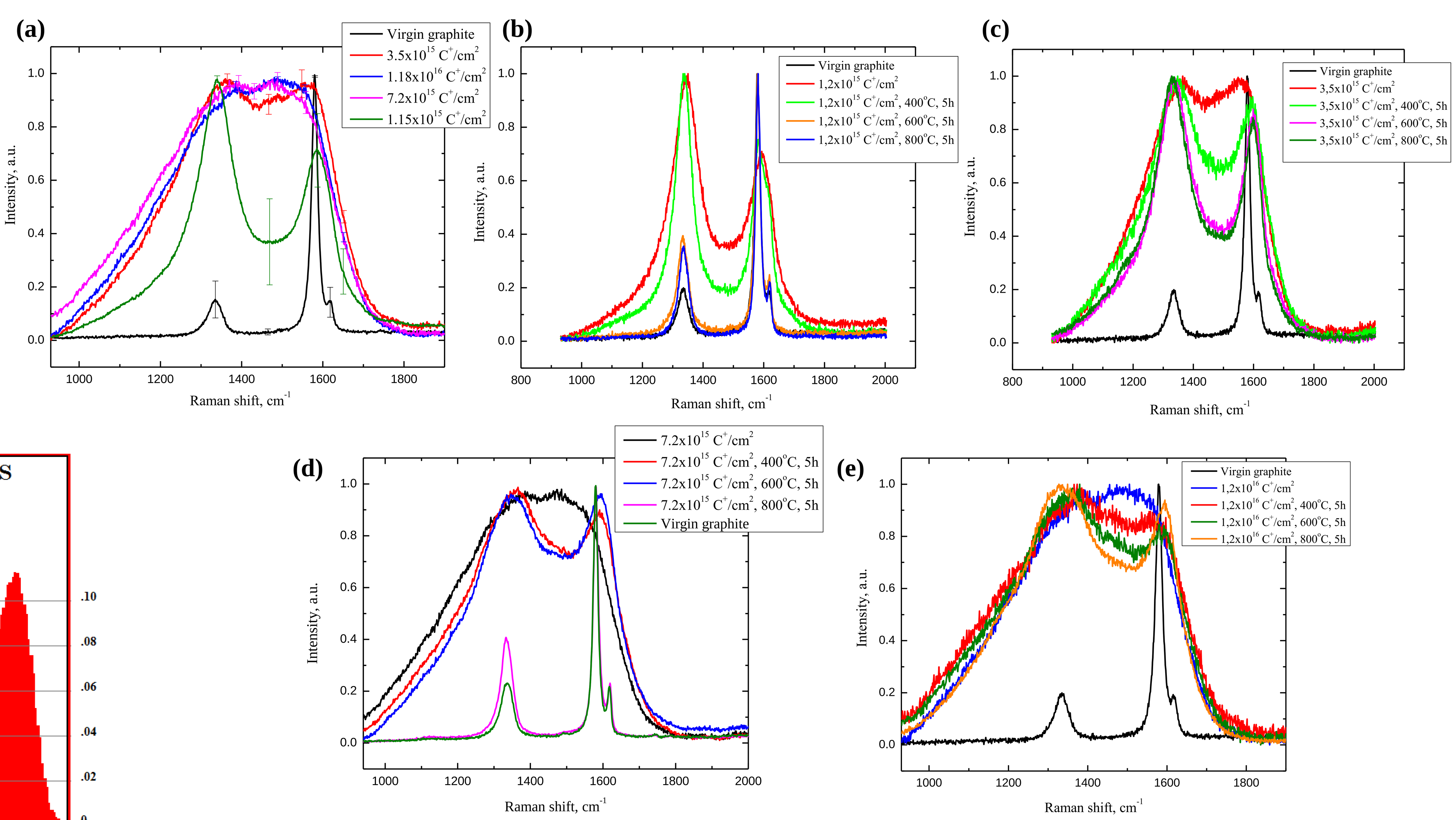

Fig. 5. Raman spectra of raw RBMK-1500 stack graphite samples **(a)** implanted at a fluence of **(b)** 1.2×10^{15} ions/cm^2 **(c)** 3.5×10^{15} ions/cm^2 **(d)** 7.2×10^{15} ions/cm^2 **(e)** 1.2×10^{16} ions/cm^2 and subsequently annealed at various temperatures ranging from 400ºC to 800ºC for 5h.

Conclusions

- Experimentally validated numerical 3D model of RBMK-1500 is used for ^{14}C profile determination in different graphite constructions (stack, sleeve, top, bottom, side reflectors). ^{14}C activity measurements in graphite samples is carried out by using express method or liquid scintillation counting (LSC) technique.
- The evolution of graphitic sp^2-related content as well as formation of an amorphous structure serves for understanding of location and stability of ^{14}C in graphite matrix, while the thermal treatment carries information about recrystallization process. Further structural investigations are currently in progress.

References

1. Plukiene et al., 2014, Nuc. Eng. Des. 277, 95-105
2. Remeikis et al., 2009, Nucl. Eng. Des. 239, 813-818
3. Remeikis et al., 2018, PLOS ONE, In Press