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Industrial packaging and characterization problems of the graphite blocks, sealing rings and graphite chips resulting from the dismantling of the RBMK-1500 reactor facility

Workshops

“Experience of Reactors dismantling”

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General information

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The production of graphite is based on the use of cokes of various microstructures as fillers and as binders of pitches, obtained, as a rule, from coal tar. Fillers for graphite nuclear reactors can serve as coke obtained from petroleum or from condensed coking coal products, which are close to isotropic in their microstructure. The technology for producing these graphite for laying reactors and solid contact rings consists in preparing the filler (calcining, grinding and fractioning), mixing the prepared mixture with pitch and pressing the resulting mass into the required dimensions. Then the “green” blanks are fired (1500ºC), impregnated with coal tar pitch until the required density is reached, and after the last firing operation (2800ºC), they are grated. At the same time, graphite tends to get “as clean as possible”, without admixtures of various elements (especially boron), which is essential for the efficient operation of the reactor as a whole.

General information

In general, the radioactivity of irradiated UGR graphite reactor can be caused by the following processes:

1) activation of impurities in graphite (the dominant nuclides ^{3}H, ^{14}C, ^{36}Cl, ^{60}Co);

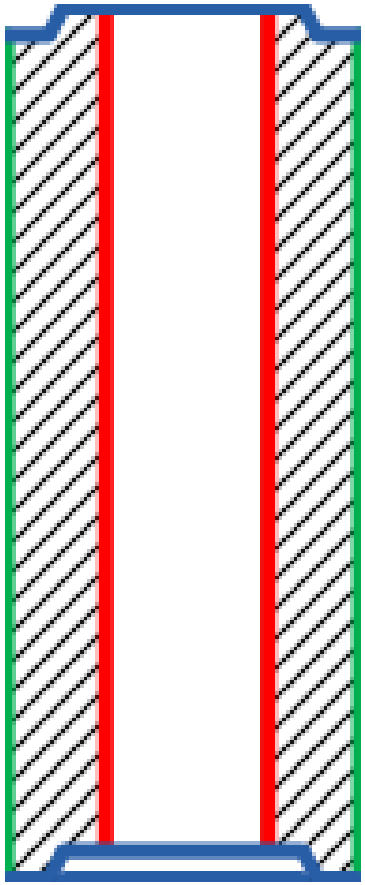
2) contamination of the surfaces of graphite products with products of activation of purge gas (for example, ^{14}C from nitrogen) and other products that come into contact with masonry, for example, channels (mainly, ^{60}Co, ^{55}Fe, etc.);

3) contamination of the surfaces of products with nuclear materials and fission products of nuclear fuel in the event of channel leakage incidents (mainly, ^{137}Cs, ^{134}Cs, etc.);

As summary ^{14}C provide 95% activity, but external gamma-emissions provide by ^{60}Co and others ^{154}Eu, ^{133}Ba, ^{155}Eu, ^{137}Cs, ^{134}Cs, ^{94}Nb, ^{54}Mn.

General information

Depending on the nature of the accumulation of radionuclides, we have a complex structure of the distribution of activity by volume graphite stack:



The shaded volume of the graphite block - conditionally evenly contains ^{14}C, ^{3}H, ^{36}Cl

Red – corrosion products ^{60}Co, ^{55}Fe by direct contact (60 - 70%)*

Blue – corrosion products and other by leakage incidents and diffusion (15-30%).

Green – diffusion and leakage incidents.

* Of course, much of the corrosion pollution is deposited on the thrust rings.

(Info based on investigation Beloyarskaya NPP)

Proximally activity levels

Nuclide	Mass activity, [Bq/g]	Activity per graphite block (54kg), [Bq]
^{3}H; 12,3year; β	$3,3\times10^{5}$	$1,6\times10^{10}$
^{14}C; 5730year; β	$1,1\times10^{6}$	$5,9\times10^{10}$
^{36}Cl; $0,3\times10^{6}$year; β	$1,0\times10^{3}$	$5,4\times10^{7}$
^{60}Co; 5,3year; β, γ	$3,0\times10^{3}$ - $5,0\times10^{4}$	$1,6\times10^{8}$ - $2,7\times10^{9}$
^{55}Fe; 2,737year; E	$5,3\times10^{4}$	$2,7\times10^{9}$
^{137}Cs; 30,2year; β, γ **	$2,8\times10^{3}$	$1,5\times10^{8}$
^{134}Cs; 2,06year; β, γ **	$9,4\times10^{2}$	$5,2\times10^{7}$

(Info based on investigation Beloyarskaya NPP)

Other causes of heterogeneity

The reactor plant has channels for various applications - heat, control, measurement, emergency protection, reflector. Accordingly, all the channels operate in different conditions (temperature, neutron fluence, contact with the pipe of the channel) and acquire a different nature and levels of radioactive contamination.

During repairs associated with the replacement of a graphite column, bore for a channel, a situation arises in which the nature of radioactive contamination changes.

There is an assumption that the blocks of the upper and lower reflector are enriched with iron, which significantly effects the level of activation of corrosion products. On the other hand, outside the core (zirconium channel), stainless pipes are used that pass through the layers of the upper and lower reflector. At the points of the SS-Zr adapter, the activity of ^{60}Co will be significantly higher than the length of the channel, and it is depended by neutron fluence.

General information sub-conclusion

The main radionuclide is ^{14}C, but it is a beta emitter.

The key Nuclide that can be measured by spectrometr is ^{60}Co, but its activity does not directly correlate with ^{14}C.

Accordingly, there are few ways:

1) Apply the nuclide vector to the entire volume of graphite, conservatively overestimating the activity on other nuclides with great uncertainty.

2) Develop a special strategy for the classification of graphite blocks according to the purpose of the channel and the position in the reactor (sub-zone), and for each type determine an individual nuclide vector.

3) To characterize each graphite block individually - mass spectrometry and radiochemical analysis (expensive). In this case, we measure only the dose rate for safety requirements during transportation and handling.

General principles and purposes of methodology support

Document purposes

The Russian Regulator (and also the countries of the former USSR) makes demands for metrological and methodical support of the measurement process.
There are two ways:

1) Metrological tests and certification procedures for the approval of the Type of measuring instrument, like a “Standard Instruments”.

2) Certification of a special document – “Measurement Methodology” for Standard Instruments used in other conditions for obtaining other types of results.

Document purposes

Radiation monitoring methodology are developed and applied to ensure that consistent results of the “Radiation Control” (RC) are obtained according to the nomenclature of parameters established for a given monitoring object with a certain uncertainty.

There are three types of documents used for the RC:

- methods (regulations) for radiation monitoring of objects (MRMS);

- methodology of performing measurement of quantities by certain methods and means of measurement (MM);

- sampling techniques and methods for preparing countable samples (MPCS).

Document structure

Measurement Methodology consists of:

- name of measurement procedure;
- purpose of measurement procedure;
- field of use;
- measurement conditions;
- measurement method (methods);
- permitted and (or) assigned measurement uncertainty or error rate and (or) assigned characteristics of measurement uncertainty;
- used measuring instruments, standard samples and their metrological characteristics, information regarding confirmation of their types (measuring instrumentation);
- appendix shall include procedure of RAW package formation, requirements to auxiliary devices, materials and chemical agents, technical characteristics and references to documents in accordance to which this operations shall be implemented;

Document structure

- operations during preparation for measurements, including description of RAW stream formation on conveyer system, their sorting and stocking in RAW containers;
- operations during measurements;
- operations on measurement processing, algorithm and sequence of calculations depending on set conditions, quality criteria and acceptance criteria of RAW to measurement (according to inhomogeneity), to further storage (by levels of activity or dose rates) or resorting (by other criteria);
- requirements to presentation of measurement results of the basis of current regulations of the Customer and National Regulator;
- procedures and frequency of accuracy control of received measurement results; (It may be necessary to expand functionality of the software and supply additional tools, for example, reference sources, to control integrity of metrological characteristics);

Document structure

- requirements to qualification of operators with corresponding references to regulatory documents active on the National Regulator Standards at the time of development of procedures and operation manuals for technical and software tools;
- requirements to safety assurance of implemented activities with corresponding references to regulatory documents active on the National Regulator Standards at the time of development of procedures;
- requirements to environmental safety assurance with corresponding references to regulatory documents active on the National Regulator Standards at the time of development of procedures;
- other requirements and operations required according to requirements of regulatory authorities or Customer specialists.

Document structure

- procedures and frequency of accuracy control of received measurement results; (It may be necessary to expand functionality of the software and supply additional tools, for example, reference sources, to control integrity of metrological characteristics);
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- requirements to safety assurance of implemented activities with corresponding references to regulatory documents active on the National Regulator Standards at the time of development of procedures;

Document purposes

In other words, MM is an independent document of direct application, which step by step describes all the questions concerning the measurement procedure. Starting with the preparation of the measured object and equipment, ending with obtaining the measurement results.

The main goal of MM is to ensure the traceability of the measurement result to the primary metrological standards and to justify the uncertainty of the measurement results.

The MM should contain a description of the entire path of conversion of physical quantities from the initial recorded effect to the final result.

Typical customers issues

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Typical customers issues

- Customers want to fill measuring container by unsorted non-homogeny (by activity distribution, by density distribution, by size of elements) waste – as faster and cheaper as possible.
- Customers want to use large volume container, with high absorbing walls and want to receive measurement result for large amount nuclides (also alpha- and beta- emitting nuclides) without additional investigation, for example, for receiving Nuclide Vector.
- Customers want easy software solution – like a “one button = one result”, without complicated setup and tuning detector parameters.

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Typical customers issues

- Customers want cheap systems.
- Customers want fast systems (short measure time).
- Customer want high resolution, high sensitivity, high accuracy measuring systems.
- Customers want high reliable systems, with short time for repairy and cheaper repairing parts.
- Customers want a system with very-very wide diapason measuring activity.
- Customer don’t want to think about physical principles and possibility applying of measuring system.

Typical producers limitation

Typical producers limitation

- Typical producers are wide world known Company.
- Producers gives common characteristic of measurement equipment in different conditions (meas.time, MDA, etc.) as comfortable for their – it can confuse Customer for compare equipment and makes complicated tender procedures.
- Producers does not determinate real diapason of measured activity for used packages of RAW – as result some times it makes problems with project of facilities RAW, because real activity can be higher then throughput of detectors.

Typical producers limitation

- Producers gives uncertainty only for calibrated parameters (energy and efficiency calibration) for ideal measurements conditions (homogeneity), achievable values around 5 – 10 % – it can confuse Customer for compare equipment and as result can makes mistake for checking storage limitation.
- Producers does not determinate “dead volume” for measuring packages – it makes to much trouble to proof for Regulator of carrying out of instruction for checking storage safety limitation.

Typical producers limitation

- Producers does not determinate uncertainty for non-homogeneity packages and confidence level for distribution real RAW (rules for separating and filling) – it makes to much trouble to proof for Regulator of carrying out of instruction for checking storage safety limitation.
- Some producers does not determinate uncertainty real chemical material of RAW and use “universal approximated material” – it makes to much trouble to proof for Regulator...
- Producers does not give full information of measuring process and modeling transfer emissions for packages RAW – it makes to much trouble for adequate calculate all components of uncertainty.

The need for methodological support

1) Based on the above, in order to eliminate problems with obtaining a license from the National Regulator, an MM should be developed for any type of measuring installation.
2) MM will close all issues related to the presentation of information:

- according to the measurement methodology, approving the order of magnitude conversion,
- by calculating all the components of uncertainty,
- to justify the limits of measurement and the adequacy of the results with reference to real conditions,
- ensure the traceability of the result to the primary standards of activity,
- postulate additional measures, usually not sufficiently described in the technological process.

Measurement uncertainty

Measurement uncertainty

On the basis of averaged uncertainty relative expanded measurement uncertainty is calculated (lower and upper evaluation) by formulas:

$$U^{-}_{Nuclide} = k \cdot \sqrt{u^2_{NuclStat} + \sum_{i=1}^{N} u_i^2}$$

$$U^{+}_{Nuclide} = k \cdot \sqrt{u^2_{NuclStat} + \sum_{i=1}^{M} u_i^2}$$

where: k – coverage coefficient for set type of distribution and selected confidence interval (Note: coefficient k = 2.00 with confidence probability P=0,95 taken for normal distribution of measured value is more conservative than k = 1.96 (for gamma-measuring, which is used during determination of limits of confidence interval for statistical constituents with Poisson distribution)).

N – number of constituents of standard combined uncertainty related with calibrated, passport and reference values. For typical package type N = 3.

M – number of all constituents of standard combined uncertainty, including components related to inhomogeneity by density and by activity, as well as with averaging during rotation and conservative approach during selection of approximation for densities higher than the measured one, etc. For typical package type M = 5 ÷ 6.

Measurement systematical uncertainty

u_1 – is determined by error of reference sources, error of measurement of distance to them (taking into consideration displacement of energy effective center of detector), statistical uncertainty of measured complete absorption peaks and mean square error of construction of approximation function of detector response from energy:

$$u_1 = \sqrt{\frac{1}{3}\cdot\delta^2 + \frac{1}{3}\sigma_D^2 + u_{Stat}^2 + u_{Eff}^2}$$

Therefore, value of u_1 falls within range from 5% to 10% for reference nuclides ^{137}Cs and ^{60}Co.

u_2 – is determined by error of construction of functional dependency – one curve for all energies of measured reference sources. This constituent u_2 is determined during commissioning of the facility for each collimator and at any energy of gamma-quantum does not exceed 10%

Measurement systematical uncertainty

u_3 – is determined by error of linear coefficients of gamma-radiation attenuation, incompliance of filling material of real package with material of the model, uncertainty on position packages on the measuring table during transportation by roller conveyor, etc:

$$u_3 = \sqrt{u_\mu^2 + u_\upsilon^2 + u_\varsigma^2 + u_\theta^2}$$

u_μ – error of reference data of linear attenuation/absorption coefficient for set in the model material, including uncertainty of approximation for arbitrary energy. Reference data are defined up to 3-4 significant character, with error not more than 0,1% for certain values of energy, but approximation of dependency by energy lead to value of uncertainty of approximately 1%.

u_υ – (**υ**λικό – material) – as material for filling package in set radiation transfer model for facility “carbon” is taken. If we measure 1 Graphite Block with correction at “channel hole” or measuring “graphite cracks” that uncertainty will not exceed 1%, but if we measured few GrBlocks in container uncertainty u_μ will be depended by filling efficiency and increased up to 10-20%.

Measurement systematical uncertainty

$u_ς$ – (μέγεθο**ς** – size) – uncertainty is related to incompliance of package of type “container” size with ideal parallelepiped described in measurement model. Of course the container will have rigid walls ($u_ς$ is evaluated at 1%), but if we have significant space from placed Gr.Blocks in filled container, then this uncertainty $u_ς$ is evaluated at 5-10%.

$u_θ$ – (**θ**έση – position) – uncertainty is related to the fact that we have 1 measuring per “container” will be depended by way of transport/placed container for measure - this uncertainty $u_θ$ is evaluated at 5%. But if we will measure 4 side rotated container we can decrees uncertainty down to 1%. (Far field measuring at 2 meters)

Therefore, constituent of uncertainty u_3 is evaluated at a level of not more than 6% for reference nuclides ^{137}Cs and ^{60}Co during package rotation and measured Homogeneity “graphite cracks” or 1 Gr.Block with “channel hole” correction. Otherwise, it can be higer than 30%

$$u_3 = \sqrt{u_\mu^2 + u_\upsilon^2 + u_\varsigma^2 + u_\theta^2}$$

Visible volume for package

Half-absorbing path in graphite for ^{60}Co is around 7,5 cm.

We can measure whole Gr.Block just only Far Field method around 2 meters for 1 Gr.Block by HPGe10% efficiency.

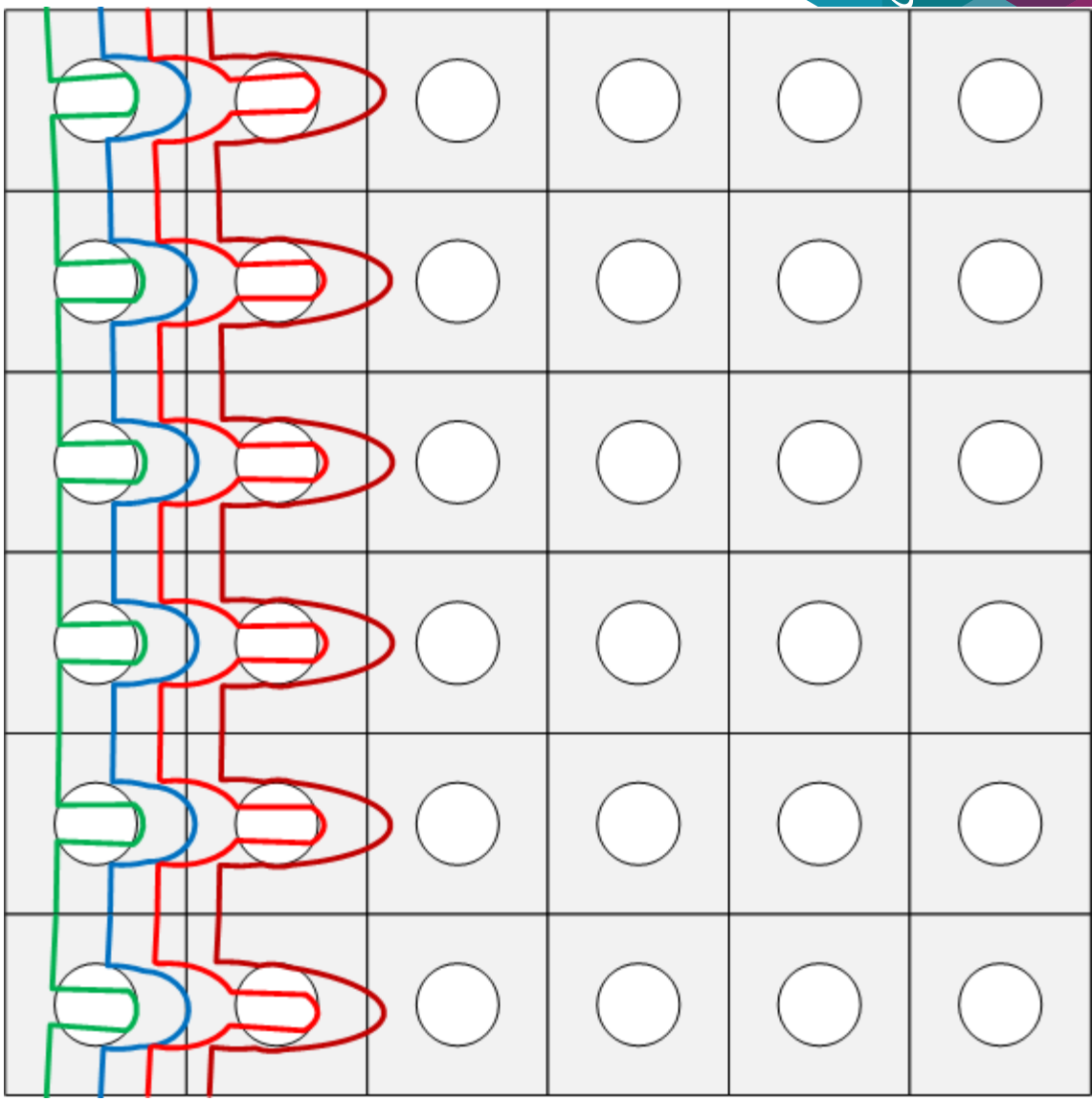
In another word in this case we will have good angle anisotropy and distance will be higher than viewing depth.

Green – 50% emission;

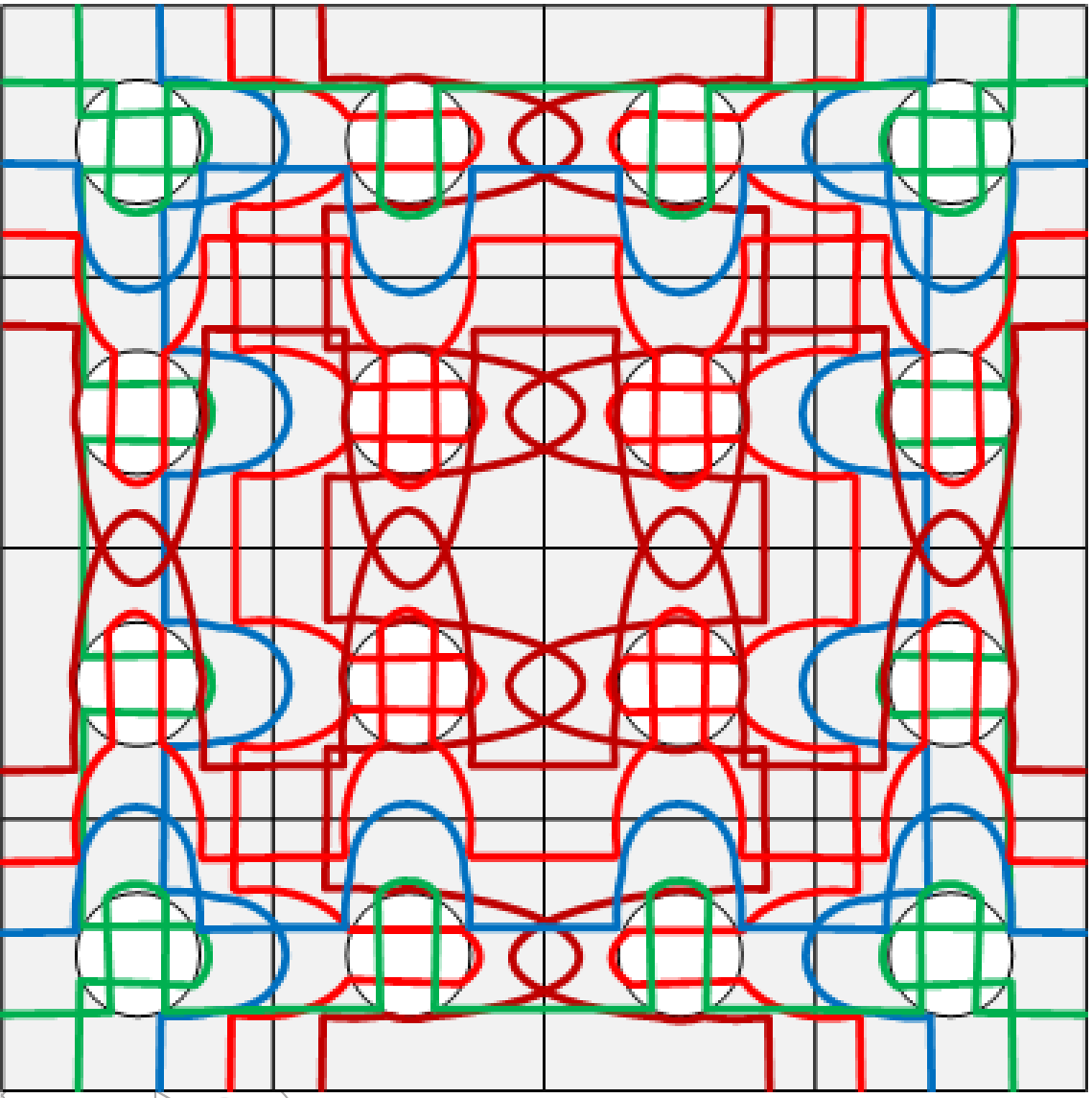
Blue – 75%;

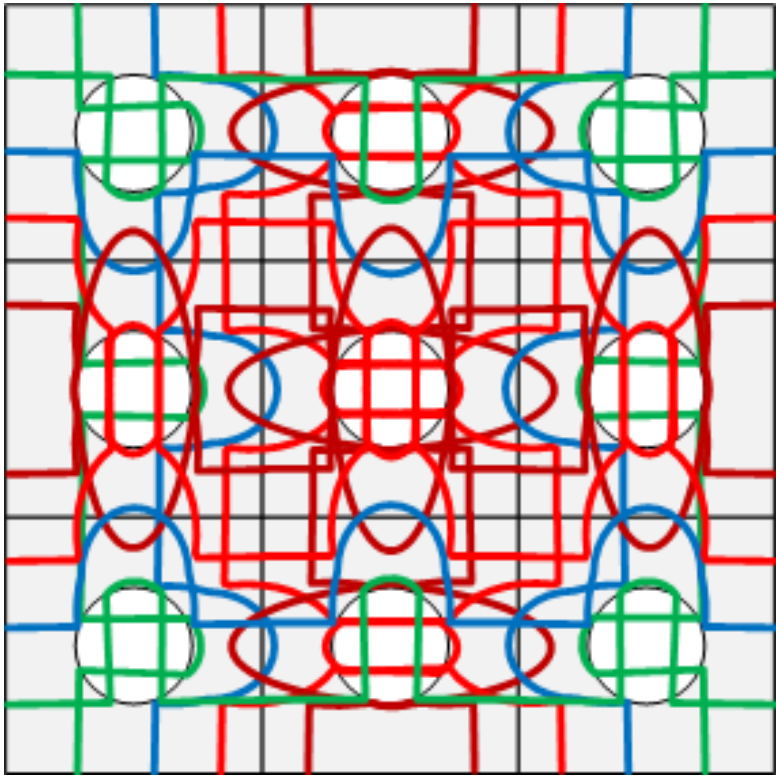
Red – 82%;

Dark Red – 95%



Visible volume for package





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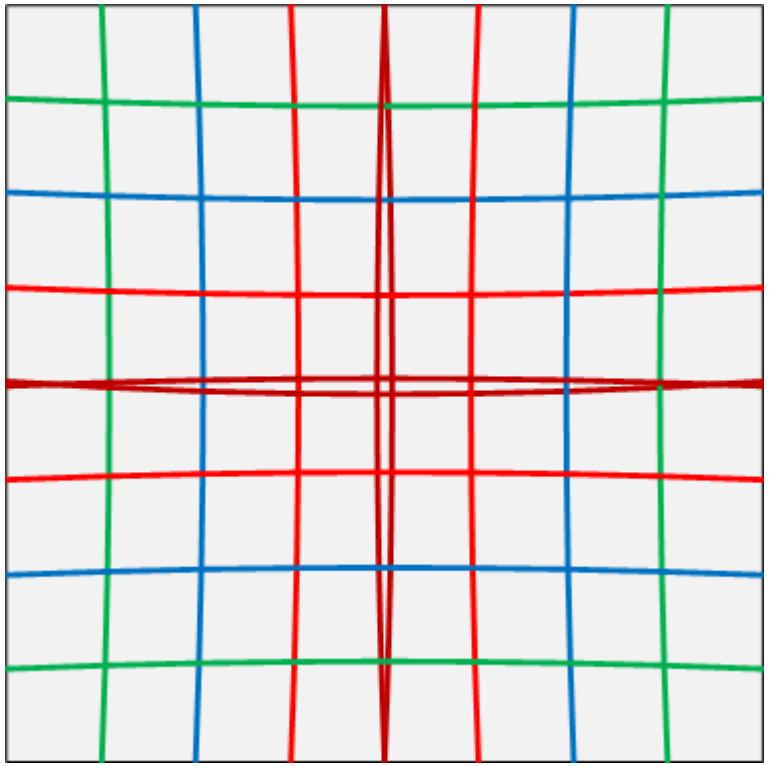
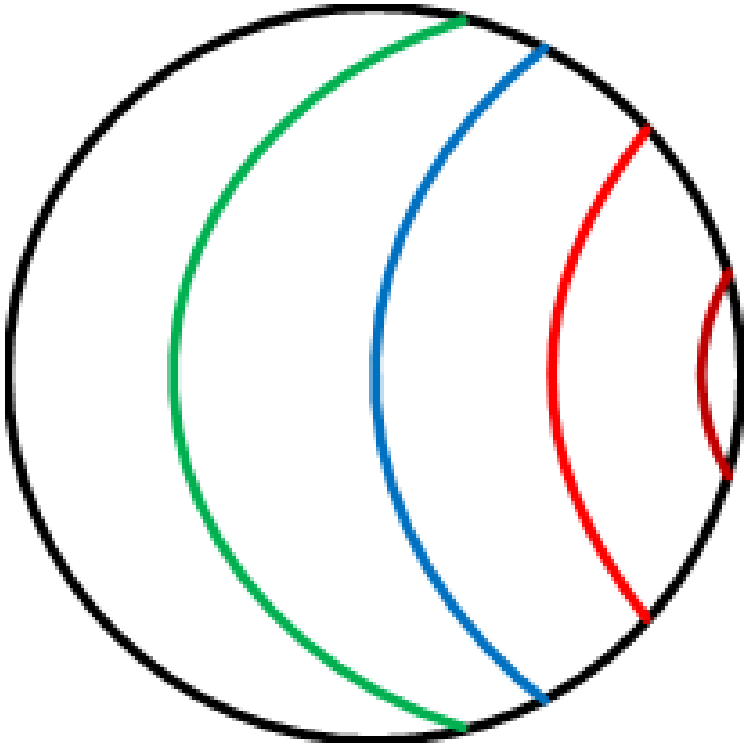
Visible volume for graphite cracks 60%

Half-absorbing path for Gr.cracks 60% filling eff. for ^{60}Co is around 12,5 cm.

Cube Container 1×1 ×1 m



Drum 200 l = Ø540×860



Measurement systematical uncertainty

Inhomogeneity by density is determined by presence/forming of fragments measuring material during sorting and mixing, as a result cavity areas and high density areas may appear areas. If taking into consideration “graphite cracks” (with 60% filling eff. – with low density 1g/cm^3) of RAW package – uncertainty u_4 will be around 1%.

If consider collection of Gr.Blocks without correction on “channel hole” as uncertainty u_4 will be 15%.

Inhomogeneity by activity is determined by by presence/forming of fragments measuring material during sorting and mixing, as a result cavity areas and high activity parts may appear areas. . If taking into consideration “graphite cracks” – uncertainty u_5 will be around 5% (depended quality of mixing).
If consider collection of Gr.Blocks without correction on “channel hole” as uncertainty u_5 will extremely high – it is needed apply additional special rules for sorting Gr.Blocks (for example by Dose rate) and in this case it can be decreased down to 15%.

Measurement systematical uncertainty

u_6 – systematical constituent of standard measurement uncertainty related to un-viewable volume (“dead zone”, “black hole”) of radiation flux cannot cross out by absorbing in material.
Rotation around its’ vertical axis, can decrease this constituent, but not in all case.

If taking into consideration “graphite cracks” of “graphite rings” in drum – uncertainty u_6 will be around 0%.

If taking into consideration “graphite cracks” in cube container – uncertainty u_6 will be not exceed 10%.

In other case with Gr.Blocks uncertainty u_5 is depended of package size, but can be decreased by filling rules (pre sorting). A proximally u_5 will be not exceed:

0% – for matrix 1×1 Gr.Blocks;

10% – for matrix 2×2 Gr.Blocks;

15% – for matrix 3×3 Gr.Blocks;

20% – for matrix 4×4 Gr.Blocks;

30% – for matrix 5×5 Gr.Blocks;

50% – for matrix 6×6 Gr.Blocks;

Result uncertainty for “drum”

On the basis of averaged uncertainty relative expanded measurement uncertainty is calculated (lower and upper evaluation) for confidence interval P=0,95

Material	$U^{-}_{Nuclide} \approx$	$U^{+}_{Nuclide} \approx$
Graphite cracks in cube stainless steel container	30%	45%
Graphite cracks in drum	28%	34%
Graphite rings in drum	35%	58%
For matrix 1×1 Gr.Blocks (hole correction);	30%	40%
For matrix 2×2 Gr.Blocks ;	40%	60%
For matrix 4×4 Gr.Blocks ;	50%	90%
For matrix 6×6 Gr.Blocks ;	60%	150%

Sub-conclusions

- For good uncertainty you need fragmentation graphite to the “cracks” as low size as possible.
- You need good mixing fragments in filled packages.
- You shall to use a low volume packages as possible (drum – is ideal, but some times it is not useful).
- You need sorting Gr.Blocks by activity before loading in package.
- You need check distribution limits before final measuring.
- You need develop filling rules and investigate possibilities before planning (designing) facilities factory for your RAW.

Sub-conclusions

- You need good investigation for your graphite with destructive methods – in ideal for each Gr.Block
- You need increase your personal's competences in questions of measuring possibilities different type of equipment before buying.
- You need methodological support for each step of characterization of Gr.Blocks
- You need full-done Information system for manage process of handling Gr.Blocks and storing characterization results

Do not afraid meeting and exchange experience.

Thank you for attention!

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