Integration of techniques for the radiological characterization of waste containing Special Nuclear Materials

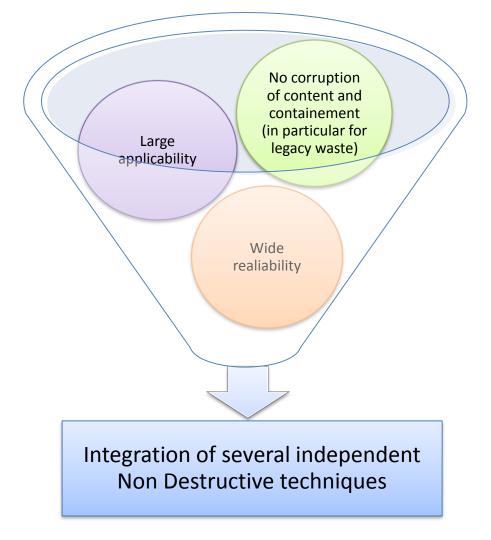
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- Introduction
- Characterization procedure
- Characterization systems
- Case study: high Uranium mass
- Conclusions and outlook



# Introduction

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# The characterization systems

In order to use several independent techniques for the NDA radiological characterization, SOGIN has implemented several gamma assay devices (SGS, ISOCS, TGS) with a active/passive neutron assay system (PANWAS), creating an integrated gamma/neutron waste characterisation facility.







ISOCS Open Geometry Gamma Assay System

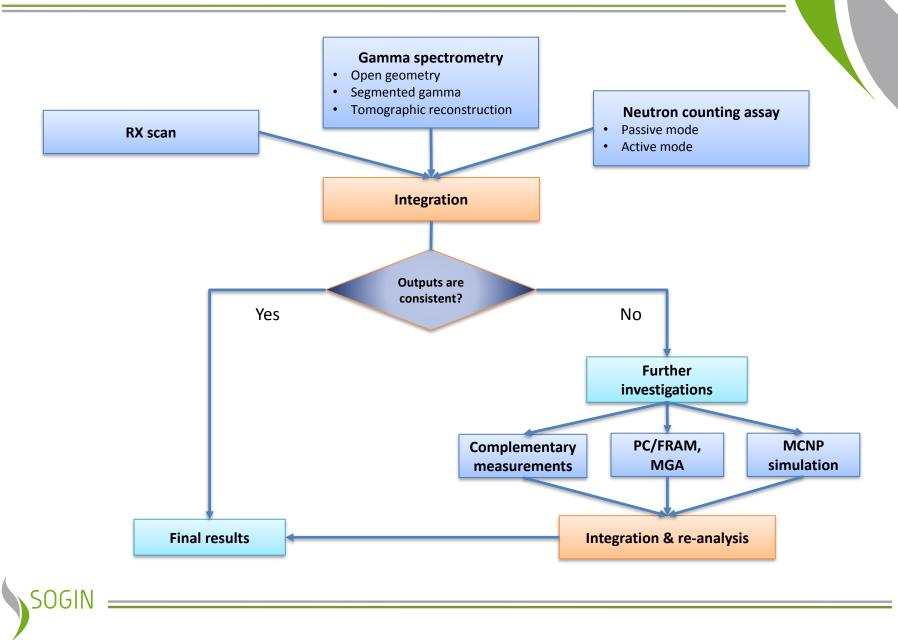
**PANWAS** Passive/Active Neutron Waste Assay

SGS Segmented Gamma Assay System

The instruments feed a common database so that when items are measured on more than one station a consolidated record is available for analysis.



# The complete procedure



### Gamma spectrometry systems







MGAS (Mobile Gamma Assay System), based on ISOCS

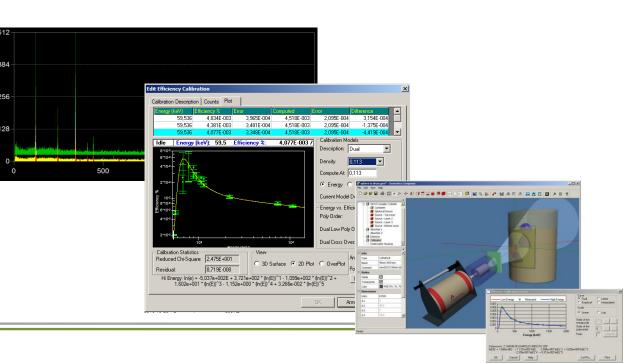
SGS Segmented Gamma Scanner

**TGS** Tomographic Gamma Scanner

The efficiency is evaluated by means of

- Transmission sources
- Multi-curve calibration
- Monte Carlo simulations

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# Neutrons counting system





- Technique: counting of time-correlated neutrons coming from
  - Spontaneuos fission reactions (<u>Passive mode</u>)
  - Induced fission reactions (<u>Active mode</u>)
- The system is able then to identify both fertile and fissile radioisotopes with mimumum detectable masses (for a typical 50 kg drum) of
  - Pu-239eq: 1 mg
  - Pu-240eff: 2 mg
- If the neutron-emitter material is a small source, the system is also able to localize its position in the drum

# The RX system

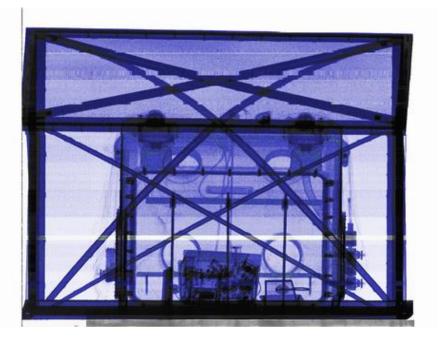
- Useful information on the matrix distribution are obtained by means of a RX scanner
- The RX is also very useful if the waste has to be re-conditionated and/or reduced in volume



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Nucleco has a mobile, daily-used radiographic system for drums and in general for items with maximum transversal dimensions 1m x 1m.

<u>A new mobile radiographic</u> <u>system without dimensional</u> <u>constraints is now in</u> <u>commissioning phase.</u>



#### Investigation of SNM: self-absorption studies

The **self-absorption** is the shielding effect that a radioactive material has on the photons that it emits. It depends on the element (i.e. the atomic number) and on the density, and has a huge relevance for Special Nuclear Materials.

- Methods or algorithms for the automatic correction of the gamma self-absorption are not generally implemented in the analysis software because without expert review they can be misleading;
- The commonly used methods to correct for the matrix effects are not fully sufficient for the self-absorption:
  - Transmission source

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• Multi-curves (efficiency vs energy profiles as a function of the matrix density)

In order to evaluate this phenomenon, we introduce a correction factor calculated via Monte Carlo simulations. If the geometrical model used to simulate the sample is close to the reality, then this correction factor is reliable.

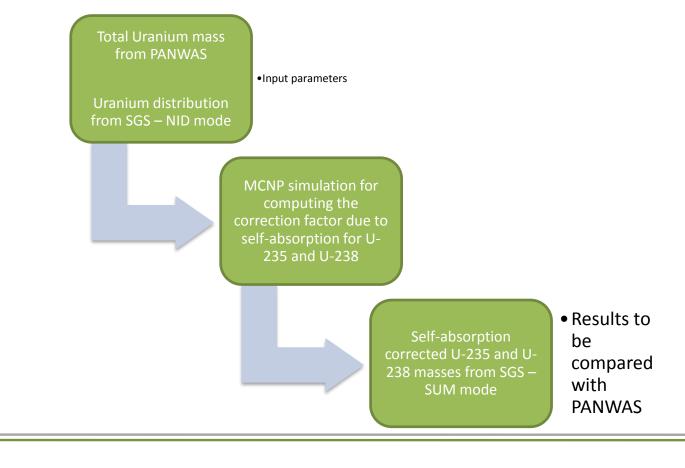
A realistic sample model can be obtained only in two ways:

- > From a detailed knowledge of material and its elemental composition (historical information);
- Using other techniques (i.e. NDE techniques)

# Case study: high Uranium mass

- The first activity involving the integration process was the characterization of drums containing Uranium mass ranging from few mg up to 50 kg
- Historical information were unavailable or not reliable
- SGS + PANWAS + MCNP

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### Results

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N°	Drum information			Neutron assay		Non corrected gamma assay		Corrected gamma assay		Discrepancy [%]	
	Gross weight [kg]	Source height [cm]	Historical information	<sup>238</sup> U mass [kg]	<sup>235</sup> U mass [g]	<sup>238</sup> U mass [kg]	<sup>235</sup> U mass [g]	<sup>238</sup> U mass [kg]	<sup>235</sup> U mass [g]	<sup>238</sup> U	<sup>235</sup> U
1	72	20	Sources U <sub>nat</sub>	0,8	2,4	0,5	1,8	0,6	2,8	-25%	21%
2	128	30	Sources U <sub>nat</sub>	3,4	16,8	2,5	7,7	2,9	19,4	-15%	16%
3	78	41	U <sub>dep</sub> spezzano	53,9	196,6	30,3	14,5	47,8	179,8	-11%	-9%
4	78	41	U <sub>dep</sub> spezzano	53,4	180,9	32,6	15,6	51,4	193,3	-4%	7%
5	78	41	U <sub>dep</sub> spezzano	51,8	180,5	33,8	17,0	53,3	210,4	3%	17%
6	78	41	U <sub>dep</sub> spezzano	53,6	179,7	32,6	16,2	51,4	200,4	-4%	12%
7	78	46	U <sub>dep</sub> spezzano	53,6	178,8	34,6	17,1	51,5	192,8	-4%	8%
8	196	46	U <sub>dep</sub> caesar	30,2	70,2	23,0	38,8	29,9	280,1	-1%	299%
9	106	30	U <sub>dep</sub> caesar	10,7	20,9	5,7	3,2	7,0	20,3	-35%	-3%
10	78	41	U <sub>dep</sub> spezzano	53,9	175,6	33,6	16,5	53,0	204,2	-2%	16%
11	78	45	U <sub>dep</sub> spezzano	51,4	151,3	33,5	16,4	49,7	188,1	-3%	24%
12	78	41	U <sub>dep</sub> spezzano	54,0	167,5	30,8	13,7	49,8	176,9	-8%	6%
13	78	48	U <sub>dep</sub> spezzano	51,8	171,1	33,4	16,1	45,7	161,3	-12%	-6%
14	74	35	U <sub>dep</sub> spezzano	48,6	173,2	28,2	13,0	47,5	177,3	-2%	2%
15	78	41	U <sub>dep</sub> spezzano	53,0	166,4	31,5	14,3	49,4	175,8	-7%	6%
16	164	41	U <sub>dep</sub> caesar	25,7	59,6	16,1	10,5	21,1	92,4	-18%	55%
17	80	44	U <sub>dep</sub> spezzano	52,4	174,0	33,8	17,1	51,0	199,1	-3%	14%
18	174	52	U <sub>dep</sub> spezzano	29,4	63,3	18,7	10,3	23,9	83,5	-19%	32%
19	180	44	U <sub>dep</sub> caesar	28,8	71,4	18,1	13,3	23,8	118,5	-18%	66%
20	190	52	U <sub>dep</sub> spezzano	32,9	68,5	21,0	16,1	26,5	135,7	-20%	98%
21	78	41	U <sub>dep</sub> spezzano	53,5	168,3	31,1	14,1	48,9	173,7	-9%	3%
22	172	50	U <sub>dep</sub> caesar	30,4	66,7	20,8	33,5	26,1	275,8	-14%	313%

For drums with a uniformly distributed matrix and source, the average discrepancy between neutron and correctedgamma results are

- <15% for U-238
- <25% for U-235 (excuding the 2 outliers)

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N°	Drum information			Neutron assay		Non corrected gamma assay		Corrected gamma assay		Discrepancy [%]	
	Gross weight [kg]	Source height [cm]	Historical information	<sup>238</sup> U mass [kg]	<sup>235</sup> U mass [g]	<sup>238</sup> U mass [kg]	<sup>235</sup> U mass [g]	<sup>238</sup> U mass [kg]	<sup>235</sup> U mass [g]	<sup>238</sup> U	<sup>235</sup> U
23	84	30	U <sub>dep</sub> daytona	6,6	2,0	2,4	1,3	2,7	5,6	-59%	179%
24	74	30	Sources U <sub>nat</sub>	7,1	27,1	3,5	7,2	4,0	32,4	-44%	20%
25	68	62	Sources U <sub>nat</sub>	1,8	5,5	1,3	1,9	1,3	2,6	-24%	-53%
26	66	41	Sources U <sub>dep</sub>	38,9	34,9	12,3	2,0	17,8	22,5	-54%	-36%
27	106	30	Sources U <sub>dep</sub> , U <sub>nat</sub>	51,5	51,2	5,5	0,6	10,5	9,8	-80%	-81%
28	40	35	Sources U <sub>nat</sub>	1,1	5,9	1,1	1,3	1,2	1,8	8%	-69%
29	62	30	Sources U <sub>dep</sub> , U <sub>nat</sub>	24,3	28,7	8,8	1,7	15,1	24,7	-38%	-14%
30	100	25	Sources U <sub>dep</sub>	57,9	79,5	17,4	5,4	39,1	105,1	-32%	32%
31	166	30	Sources U <sub>nat</sub>	61,9	85,2	17,2	34,2	31,9	208,3	-49%	144%
32	84	30	Sources U <sub>nat</sub>	3,6	10,4	2,7	4,0	3,0	11,1	-17%	6%

As the matrix and the contamination are far from being homogeneous, the discrepancies increase

• ≈40% for U-238

 ≈40% for U-235 (excuding the 2 outliers)

In general, the corrected gamma results are lower than the neutron results.

This means that the source should be modeled by a bulk and the MCNP simulations underestimate the self-absorption effect. This procedure was approved by EURATOM/IAEA during a dedicated audit!

# Conclusions

- > By means of the presented procedure, we integrate
  - gamma spectrometry (segmented and far-field modes)
  - passive/active neutron counting
  - Monte Carlo simulation

in order to compensate the limits of the single technique and elaborate consistent results;

- The reliability of the MCNP simulations is strongly related to the goodness of the input data;
- Even when the source information is missing, the results are still useful since they supply a warning for a possibile measurement fault;
- When detailed information on the sample to characterize are available, the presented procedure can be reduced by using only the gamma assay corrected using MCNP;
- > The presented procedure is applicable also to other radionuclides, e.g. Plutonium.

# Future improvements

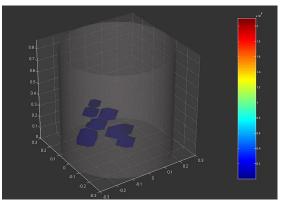
In order to obtain further information on drums content and improve the accuracy and reliability of the results, we are now using other ND techniques and methods. In particular:

- Improve the digital X-ray radiographic system making it:
  - More powerful;
  - More applicable;
  - ≻ 3D
- Improve the NIT (Neutron Imaging Technique) analysis algorithm for the PANWAS system, which provides a gross neutron tomography;

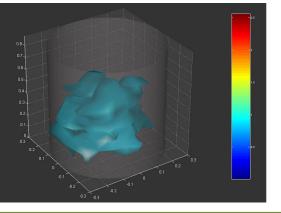


 Transportable Tomographic Gamma Scanner (TTGS), actually in commissioning phase





Matrix distribution





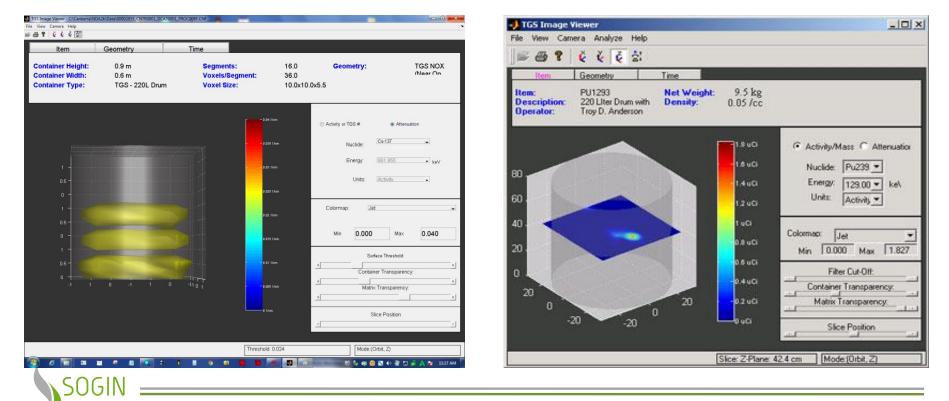






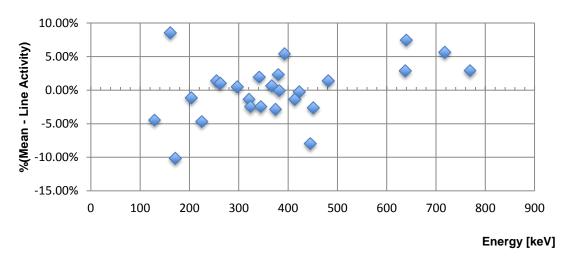
# The Tomographic Gamma Scanner

- We are now introducing a new gamma spectrometry system able to work also in Tomographic mode
- In that mode, a 220L drum is divided in small cubes (with typical dimensions 5cm x 5cm x 5cm) called "voxel"
- For each voxel are determined the photon attenuation and the activity, allowing for the 3D reconstruction of matrix and contamination distribution



# Extention of the procedure: ISOCS-only

- When detailed information on the sample to characterize are available, the current procedure is applied by using only the gamma assay in open geometry
- For each measurement, the efficiency is evaluated via Monte Carlo simulation and the analyst can invoke the self-absorption correction including the distribution of the SNM in the sample model



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#### **Application**

- Simple cylinder containing <u>hundreds</u> grams of Pu and U oxide
- The activities of the energy keylines agree with the mean value with only a 10% discrepancy
- The discrepancy between the ISOCS results and the other data available for the sample (historical data, destructive analysis) is below 3% for both Plutonium and Uranium