

# THE SAFETY OF ENVIRONMENT IN FINAL DISPOSAL OF ULTIMA GOLD SCINTILLATION LIQUID COCKTAIL USED FOR DETERMINATION OF THE RADIOACTIVE CONTENT IN VARIOUS SAMPLES AT CERNAVODA NUCLEAR POWER PLANT

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The Ultima Gold scintillation liquid waste samples was counted in a 2100 TRI-CARB® Packard model Liquid Scintillation Analyzer using the Ultima Gold XR scintillation liquid cocktail. To determine tritium content, three large sample/cocktail plastic vials were prepared and four sets of measurements were performed. Counting errors and statistics were applied on them to obtain specific activity of the waste. The immobilization and packaging of Ultima Gold liquid scintillation waste were carried out by cementation, using Portland cement and an adequate solidification additive in a standard dimension container. Radioactive waste packages produced comply with waste acceptance criteria developed by the National Commission for Nuclear Activities Control for disposal at the Băița National Repository, Bihor County, and thus the protection of human health and safety of the environment are ensured.

## INTRODUCTION

To assure the safety of waste disposal, radioactive waste undergoes some forms of immobilization and packaging, in a controlled and properly managed manner. Both the waste form and container are important for the safety of waste packages during handling and interim storage, as well as the post-closure phase of a repository. For low and intermediate level radioactive waste (LILRW) a number of immobilization processes are available including cementation, bituminization and polymer fixation. The following two waste conditioning processes<sup>1</sup> are most commonly used: placement of liquid waste in a container and immobilizing it within a suitable matrix, and the mixing of liquid waste with an immobilization matrix and pouring the mix into a container.

Cementation is the most widely applied technique for the immobilization of LILRW. The quality of the final cemented waste forms depends very much on the composition and the type of the waste. Various kinds of additives and chemicals are used to improve the cemented waste form.<sup>2,3</sup>

The rationale for the immobilization of radioactive waste using cementation materials is to provide a solid, stable and durable material that can be more easily stored and disposed than unconditioned wastes. The main advantages of cementations materials include:<sup>4,5,6</sup> favorable long-term chemical and mechanical stability, high strength facilitating handling, low permeability, high-pH conditions which reduce the solubility of many radionuclides, physical and chemical properties generally well understood, low cost.

Cernavodă Nuclear Power Plant generates a wide variety of radioactive wastes, which are in liquid, solid and gas form.<sup>7</sup>

Liquid radioactive waste class includes liquid organic and aqueous wastes resulting from process system operating and from decontamination and maintenance operations.

Liquid organic wastes consist of: Ultima Gold scintillation liquid cocktails with various radioactive liquids, miscellaneous organic solvents (acetone, white spirit, ethylene glycol, alcohol ethyl, toluene, chloroform), and lubricating oils.

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Ultima Gold scintillation liquid cocktails are used by the radio-chemical analysis laboratory from Cernavodã Nuclear Power Plant to determine the radioactive content in organic and aqueous samples.

The organic samples are lubricating oils, from pumps and motors used in Zones 1 and 2, and solvents from the decontamination area and from the laboratories and maintenance activities. Tritium atoms may diffuse into these organic materials and form organically-bounds. The terms "organically-bound tritium solvent", and "organically-bound tritium oils" are used to describe the compounds of tritium with the two types of organic samples. Organically-bound tritium oils (OBT oils) or tritiated pump oils are considered to be a mixture of three components: original molecules of oil labelled by tritium, small molecule created by radiolytic and degradation processes and labelled by tritium, and tritiated water (HTO). As a reasonable hypothesis, the tritium in OBT oils should be expected to partition into the above three components in approximately an 80:10:10 ratio, respectively.<sup>8</sup>

The aqueous samples (containing tritium as HTO) are obtained from Cernavodã Nuclear Power Plant Liquid Effluents System.

Tritium in the organic and aqueous samples can be detected through liquid scintillation counting<sup>9</sup> (LSC), which is an analytical technique defined by the incorporation of the radioactive sample into uniform distribution with the Ultima Gold liquid scintillation cocktail. To assure efficient transfer of the energy between the beta particle, which is emitted in a radioactive decay, and the scintillation cocktail, some solvents are included in the Ultima Gold chemical composition. The energy of the beta particle is absorbed by the medium in three forms: heat, ionization and excitation. Some of the beta energy is absorbed by solvent molecules making them excited (not ionized). Energy of the excited solvent is emitted as UltraViolet (UV) light and the solvent molecule returns to ground state. The energy from these molecules passes back and forth among the solvent system, allowing efficient capture by dissolved phosphors (primary and secondary scintillators). Primary scintillator is capable of being excited to a light emitting state by excited solvent molecules and provide the conversion of captured energy to the emission of light. Secondary scintillator captures the fluorescence energy of the excited primary scintillator, and re-emits it as a longer wave length signal for the

spectral response of the photo multiplier tubes (PMTs). Phosphor molecules emit blue light flashes (scintillation) upon return to the ground state. The intensity of the light is proportional to the beta particle's initial energy. The scintillation is captured by photo cathode of the PMTs and transformed by these to an electrical pulse proportional to the intensity of the scintillation. The amplitude of the electrical pulse is converted to a digital value that represents the beta particle energy. This digital value is memorized in a proper analyzer channel. The number of pulses in each channel is printed out or displayed as the spectrum of the sample, providing information about the energy of the radiation or the amount of radioactive material dissolved in the cocktail.

The gathering of scintillation liquid waste is made in 220 L drums in an appropriate collection point at Cernavodã Nuclear Power Plant. Then, these waste drums are transported to Radwaste Treatment and Conditioning Plant of Institute for Nuclear Research Pitesti to convert scintillation liquid waste into a stable monolithic form, which minimizes the probability to release radio nuclides in the environment during interim storage, transportation and final disposal.

Conditioning of scintillation liquid waste consists of three main process steps:

Tritium measurement of Ultima Gold scintillation liquid waste;

Immobilization of Ultima Gold scintillation liquid waste into a matrix to produce a solid form which retards radionuclide mobility;

Packaging the solid form into a standard dimensions container which retards the ingress of water.

The goal of these processes is to produce a waste form which is suitable for disposal and is compatible with the intended transportation and disposal operation. The waste in its final conditioned form can then be transfer to the Bãița National Repository for Low and Intermediate Radioactive Waste, in an old exhausted uranium mine sited in Apuseni Mountains, Bihor county.

## EXPERIMENTAL PART

### Establishing of tritium content in Ultima Gold liquid scintillation waste

To determine tritium content, the Ultima Gold XR liquid scintillation cocktail was used, whose chemical composition<sup>10</sup> is shown in Table 1.

Table 1  
Chemical composition of the Ultima Gold XR cocktail

Component	Name	Composition [weight %]
Solvents	di-isopropyl naphthalene (DIN)	40-60
	ethoxylated alkylphenol	20-40
	bis(2-ethylhexyl) hydrogen phosphate	2.5-10
	triethyl phosphate	2.5-10
	sodium di-octylsulphosuccinate	2.5-10
	3,6-dimethyl-4-octyne-3,6-diol	1.0-2.5
Scintillators	2,5 diphenyloxazole (PPO)	0-1.0
	1,4-bis (2-methylstyryl)-benzene (Bis-MSB)	0-1.0

The solvents portion of the Ultima Gold XR cocktail comprises from (68.5...99)% of the total solution, and is based on di-isopropyl naphthalene (DIN). This solvent is considered biodegradable and has a very high flash point (152°C) and it is therefore classified as non-dangerous in accordance with national and international traffic regulations.<sup>11</sup>

The primary scintillator (2,5-diphenyloxazole) is soluble in the solvents at a sufficient concentration to efficiently pass captured energy. The secondary scintillator (Bis-MSB) captures the energy of the excited PPO primary scintillator, and re-emits it as a longer wave length signal suitable for LSC technique.<sup>9</sup>

In these experiments, the preliminary research was conducted using glass vials to allow visual verification that a homogeneous solution is obtained at the desired cocktail/sample ratio. Then, the research was moved into plastic vials to reduce background and improve the counting statistics.<sup>11</sup>

Three sample/cocktail plastic vials, D<sub>1a</sub>, D<sub>1b</sub>, D<sub>1c</sub>, were prepared. Waste sample volume of 0.1 mL was accurately

pipetted directly into the vials, and the liquid scintillation cocktail was added so that the total volume was 20 mL. Each vial was shaken vigorously for several seconds after each addition to ensure homogeneity. The best counting efficiencies were achieved when samples were uniformly dispersed into the cocktail to produce a clear, colorless, pH neutral emulsion.<sup>9</sup>

Background counts were very low relative to the activity of the sample so these counts were ignored.<sup>11</sup>

The sample vials were counted in a 2100 TRI-CARB® Packard model liquid scintillation analyzer. The temperature in the counting room was (22±3)°C during a typical 24 h period of time.

The Spectralyzer™ spectrum analyzer is calibrated in keV, and the user can choose any region between 0 keV and 2000 keV to measure a sample. The region of interest<sup>9,11,12,13</sup> for <sup>3</sup>H is from 0 keV to 18.6 keV.

Typical Performance Data<sup>12</sup> (background, efficiency, Figure of Merit values for both <sup>3</sup>H and <sup>14</sup>C regions) published by PerkinElmer™, measured in factory at Downers Grove, Illinois, are shown in Table 2.

Table 2  
Typical performance data for TRI-CARB® 2100

Parameter	Radionuclide			
	<sup>3</sup> H		<sup>14</sup> C	
	Energy Range [keV]	Value	Energy Range [keV]	Value
Minimum Acceptable Efficiency, E (NCM)	(0...18.6)	60 %	(0...156)	95 %
Background, B (NCM)	(0...18.6)	17.3 cpm	(0...156)	24.3 cpm
Figure of Merit, E <sup>2</sup> /B (NCM)	(1...18.6)	180	(4...156)	380

Note: The efficiencies, backgrounds, and E<sup>2</sup>/B values for the Normal Count Mode (NCM) were determined using NIST traceable PerkinElmer™ sealed large glass vial standards sets

The 2100 TRI-CARB® Packard model liquid scintillation analyzer has the ability to automatically verify its performance. Normal Count Mode reduces background by 30% to 40% compared to conventional liquid scintillation

counters, and it is typically recommended for samples above 750 DPM.

Depending on the placement of the instrument in the laboratory, the background may differ from those values stated

in the Table 2. Altitude and environmental radiation sources must be taken into account when the local specifications are established. The term Figure of Merit (FOM), expressed as a function of efficiency and background, is currently the most widely used parameter to assess counter sensitivity and performance.

Four sets of measurements were performed on the three samples, and the values for Time, CPM (Counts per Minute),  $2\sigma\%$  (the statistical precision of measuring a sample), DPM (Disintegration per Minute), tSIE (transformed Spectral Index of the External standard) and SIS (Spectral Index of Sample) were displayed by analyzer and shown in Table 3.

Table 3

Samples measurements values

Measurement set	Sample code	Time [minutes]	CPM [cpm]	$2\sigma$ [%]	DPM [dpm]	tSIE	SIS
I	D <sub>1a</sub>	5.00	20467.95	0.63	42366.30	452.28	14.90
	D <sub>1b</sub>	5.00	21056.04	0.62	43982.78	445.49	14.71
	D <sub>1c</sub>	5.00	20027.14	0.63	41586.00	449.89	14.72
II	D <sub>1a</sub>	0.84	21362.61	1.50	45894.28	454.16	15.15
	D <sub>1b</sub>	0.85	21060.81	1.50	43869.73	447.56	14.78
	D <sub>1c</sub>	0.88	20327.73	1.50	42158.59	450.80	14.77
III	D <sub>1a</sub>	0.88	20337.96	1.50	41962.96	454.70	15.14
	D <sub>1b</sub>	0.84	21180.50	1.50	43834.37	452.39	15.10
	D <sub>1c</sub>	0.89	20137.23	1.50	41587.90	453.98	15.03
IV	D <sub>1a</sub>	0.86	20745.35	1.50	42936.14	452.35	16.14
	D <sub>1b</sub>	0.84	21395.24	1.50	44579.82	447.33	16.07
	D <sub>1c</sub>	0.87	20493.10	1.50	42511.93	450.62	16.24

The first measurement set was performed with protocol option "count time", that is sample counting was automatically interrupted at the completion of the selected time (5.00 minutes). The following three measurement sets were carried out with protocol option "2 Sigma Coincidence", so the counting was continued until the assigned sigma was reached ( $2\sigma\% = 1.50$ ).

The Direct DPM method was used in this experiment because the CPM values are greater than 500 cpm. This data mode does not require quench curves or standard sets for reporting accurate DPM results for most single label samples including tritium.<sup>13</sup>

Both tSIE and SIS are used as Quench Indicating Parameters (QIP). The minimum values for the tSIE and SIS parameters (445.49, respectively 14.71) are high enough to reflect a low degree of quenching in the sample and high measurement efficiency.

At the end of the measurement every detected event from the sample was stored into analyzer memory, spanning (0...20) keV, including those events due to background and other phenomena which could affect the counting of the sample.<sup>12</sup>

#### Immobilization of Ultima Gold liquid scintillation waste

The immobilization of Ultima Gold liquid scintillation waste was carried out by cementation, using Portland cement and some adequate solidification additives (aluminium stearate, sodium silicate and lime).

There were studied three compositions which differ from one another by the ingredients used and the order in which they were mixed:

A composition: waste: aluminium stearate: water: cement: sodium silicate;

B composition: waste: aluminium stearate: water: cement: lime: sodium silicate;

C composition: waste: aluminium stearate: water: cement.

The molecular formula of aluminium stearate (stearic acid aluminium salt) additive is  $C_{54}H_{105}AlO_6$ , and the chemical composition is the following:<sup>14</sup>

Assay (as Al): (3.0...4.0)%;

Chloride (Cl):  $\leq 0.05\%$ ;

Heavy metals (as Pb):  $\leq 0.01\%$ ;

Fe (Iron):  $\leq 0.01\%$ .

The physical solidity of the waste form was established by measurement of mechanical strength.<sup>15</sup> The values<sup>16</sup> of mechanical strength were greater than  $50 \cdot 10^3 \text{ N/m}^2$ , that is the minimal required disposal value, imposed by waste Acceptance Criteria of disposal site (Băița National Repository for Low and Intermediate Radioactive Waste, Bihor County).

The composition C had the highest values of mechanical resistance; therefore long-term leaching test was conducted on composite C.

Chemical stability was assessed by determining of leach ability.<sup>17</sup> Leach ability is a parameter that refers to the rate of release of radioactive species from the waste form as a result of interaction with water. The test method requires the waste form specimen to be cylindrical, and the value of the ratio of leaching agent volume to the exposed surface area of the specimen is of (0.1...0.2) m. The method uses demineralized water as the leaching agent. Testing is performed at a temperature of  $20^\circ\text{C}$ . The leaching agent is sampled and

completely replaced with fresh leaching agent on the following schedule:

Initially, after 1, 3 and 7 days;

Then once in second, third, fourth, fifth and sixth week;

Once per month during the following seven weeks, and

Twice per year for as long as is considered necessary.

The sampled leaching agent was analyzed to measure tritium activity content in a 2100 Tri-Carb® Packard model liquid scintillation analyzer.

The values<sup>16</sup> of the leach rate were less than  $10^{-3}$  cm/day, which is the maximal value<sup>18</sup> required by Băița National Repository, Bihor county.

#### Packaging the solid waste form in a container of standard dimensions

There were two types<sup>18</sup> of containers used to package the solidified organic waste:

218 L drum for the direct immobilization of waste with tritium activities values less than  $10^8$  Bq/L;

70 L drum packaged into a 218 L container (backfilling the interstices between the two drums with concrete) for the immobilization of waste with tritium activities values greater than  $10^8$  Bq/L.

The function of the drums was to provide a fixed volume into which the waste encapsulated in a matrix can be fed during the immobilization process.

To ensure long-term stabilization of waste, and guarantee the protection of human health and safety of environment during storage, the 218 L and 70 L drums were specially designed and tested by the certified producer.

The cementation method included into-container mixing for batch processing of organic waste volumes. The procedure was to adequately dose and mix the waste and water into a 218 L drum (for direct immobilization) and into a 70 L drum (for tritium activities values over  $10^8$  Bq/L). Then, the cement and aluminium stearate were dosed and added to the

waste/water emulsion and the whole composition stirred with the mixing blades until the obtained batch was homogeneous.

The mixing blades could then be removed and the same operations were performed on the 70 L drums.

The entire process was controlled from a separate command desk. Ventilation equipment was installed to avoid spreading of cement and aluminium stearate dust during filling of the drums.

The 70 L drum was packed into the 218 L drum by concrete, and the two containers were closed and stored for product cure to take place. Adequate cure required 28 days.

It is important that the recipe for 218 L and 70 L waste forms should contain enough water to hydrate the cement adequately, otherwise the solidified waste form may not have sufficient cohesion.

Performing tests on the two 218 L and 70 L packaged into 218 L drums were performed to confirm the quality of waste package content. These performing tests were carried out by certified laboratories of Institute for Nuclear Research Pitesti, and the following quality and technical characteristics were verified: analysis bulletins of the working materials, shape and size of the drums, external radioactive contamination,<sup>18-20</sup> and gamma radiation equivalent dose rate at the drum wall<sup>18</sup>. Also, the following tests were performed: penetration proof,<sup>18,19</sup> compression resistance,<sup>18,19</sup> free fall,<sup>18,19</sup> watering proof.<sup>18,19</sup>

## RESULTS AND DISCUSSION

### Establishing of tritium content in Ultima Gold liquid scintillation waste

The Instrument Performance Assessment Data carried out by 2100 TRI-CARB® Packard model liquid scintillation analyzer are shown in Table 4.

Table 4

The instrument performance assessment data

Parameters	Radionuclide	
	<sup>3</sup> H (0...18) keV	<sup>14</sup> C (1...156) keV
Efficiency, E	65.35 %	96.3 %
Background, B	18.18 cpm	26.62 cpm
Figure of Merit (E <sup>2</sup> /B)	235.28	431.14
Chi Square ( $\chi^2$ )	19.72	13.34

Comparative to the Instrument Performance Assessment Data in Table 2:

The values of measurement efficiency for <sup>3</sup>H and <sup>14</sup>C are greater than admissible minimum efficiencies values of 60% for <sup>3</sup>H and 95% for <sup>14</sup>C;

The values of Figure of Merit are greater than the values for both <sup>3</sup>H and <sup>14</sup>C;

The values of background for <sup>3</sup>H and <sup>14</sup>C are placed into the limits of normal background level;

The  $\chi^2$  test conducted with the 2100 TRI-CARB® Packard model liquid scintillation analyzer produced values of  $\chi^2$  within the (10.10...30.1) range indicated by the 20 measurements.<sup>12</sup>

The  $\chi^2$  test compares two distributions: the observed distribution and the theoretical distribution. The test involves repeating the measurement of a sample  $n$  times. For a 95% confidence level, the

values of  $\chi^2$  should lie within certain limits determined by the number of measurements.

If the system does not meet performance specifications, the verification procedures of the

four parameters (Efficiency, Background, Figure of Merit, Chi Square) are repeated.

Samples spectrums, in the Region of Interest for  $^3\text{H}$  (0...20) keV, are shown in Fig. 1.

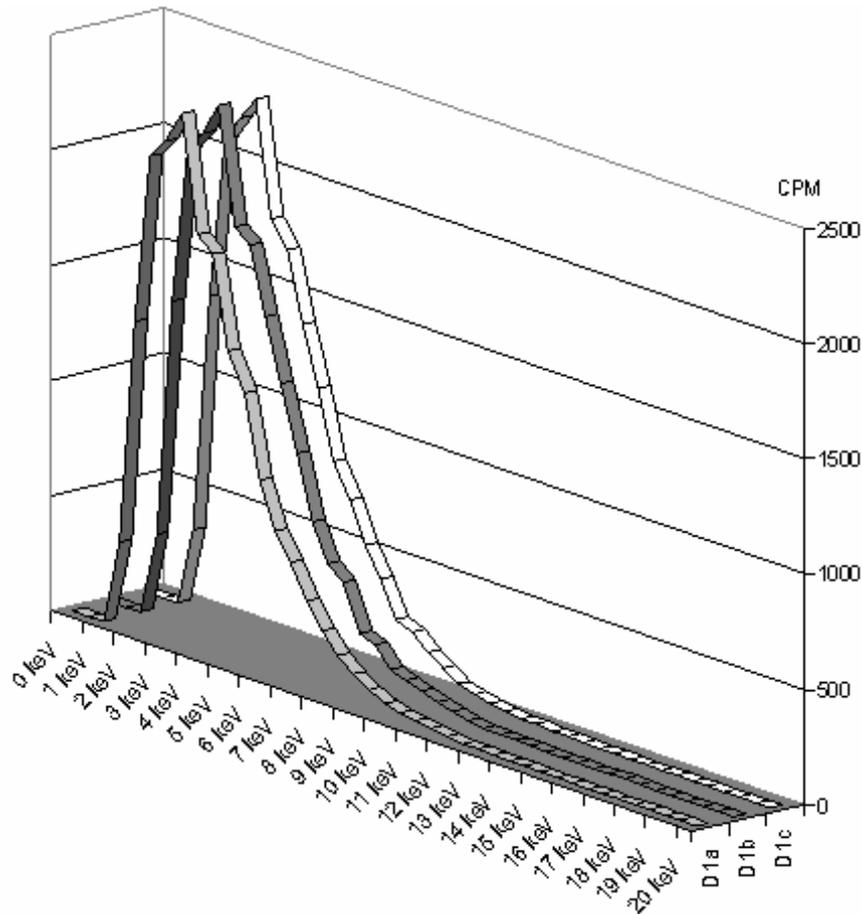


Fig. 1 – Samples spectrums.

$\beta$ -particles are high energy electrons, produced during the conversion of a neutron to a proton in the nucleus.  $\beta$ -particles are emitted in concert with a neutrino, which is almost impossible to detect and it is of no interest in liquid scintillation counting apart from the fact that it carries away some of the decay energy. The sum of the energies of the neutrino and  $\beta$ -particle is a constant for a given isotope, and defines the maximum energy of that radionuclide ( $E_{\max}$ ). The maximum energy emitted in a nucleus decay of  $^3\text{H}$  is 18.6 keV, and it is approached only for  $\beta$ -particles emitted with a low energy neutrino. In practice a distribution of

energies is observed which is characteristic of the emitting isotope. Thus, theoretically, the  $\beta$ -particle for tritium can possess any energy<sup>9,12,13</sup> between 0 and 18.6 keV.

The comparative samples spectrums illustrated in Figure 1 show that the  $\beta$ -particles emitted by a tritium nucleus produce a continuous energy spectrum from zero up to the maximum value. The energy used for calculations of absorbed energy is an average energy based on the maximum energy and the number of the nucleus decays. The average kinetic energy,  $E_{\text{av}}$ , also called the mean pulse height energy, is defined by Equation (1).

$$E_{\text{av}} = \frac{\int_0^{E_{\max}} E \cdot N(E) \cdot dE}{\int_0^{E_{\max}} N(E) \cdot dE} \quad (1)$$

where:

$E_{av}$  = average kinetic energy;

$N(E)$  = number of particles of energy  $E$ ;

$E$  = energy of particle.

The calculated value is the first moment of the  $\beta$ -spectrum, also called the center of gravity.

The calculated average energy for tritium is less than 6 keV.

The values of measurement efficiency are shown in Table 5.

Table 5

Tritium measurement efficiency in analyzed samples

Sample Code	$Efficiency = \frac{CPM}{DPM} \cdot 100$ [%]			
	I	II	III	IV
D <sub>1a</sub>	48.31	46.55	48.47	48.32
D <sub>1b</sub>	47.87	48.01	48.32	48.00
D <sub>1c</sub>	48.16	48.22	48.42	48.21

The counting efficiency, as a percentage, is defined as the ratio of the number of observed counts ( $CPM$ ) to the number of decays ( $DPM$ ) which occurred during a measurement time.

Counting efficiency is affected by the degree of quenching in the sample (mainly, chemical quench and color quench). The Ultima Gold XR scintillation liquid cocktail has exhibited good

efficiency, around 48%, which is higher than typical efficiency<sup>21</sup> of 40%.

The three radioactive samples were counted four times and counting errors and statistics were applied on the measurement values.

For the twelve measurements, the ascending arranged DPM values are shown in Table 6.

Table 6

Count data summary of counting samples

Count #	DPM [dpm]	DPM range [dpm]	Frequency
1	41586.00	(41586.000...42447.656)	5
2	41587.90		
3	41962.96		
4	42158.59		
5	42366.30		
6	42511.93	(42447.656...43309.312)	2
7	42936.14		
8	43834.37	(43309.312...44170.968)	3
9	43869.73		
10	43982.78		
11	44579.82	(44170.968...45032.624)	1
12	45894.28	(45032.624...45894.280)	1

The twelve sorted values were grouped into five equal ranges of 861.656 dpm, and the frequencies of the values occurrences into that range were noticed.

The graphical representation of the distribution of the frequency reported to the width interval is depicted in Fig. 2.

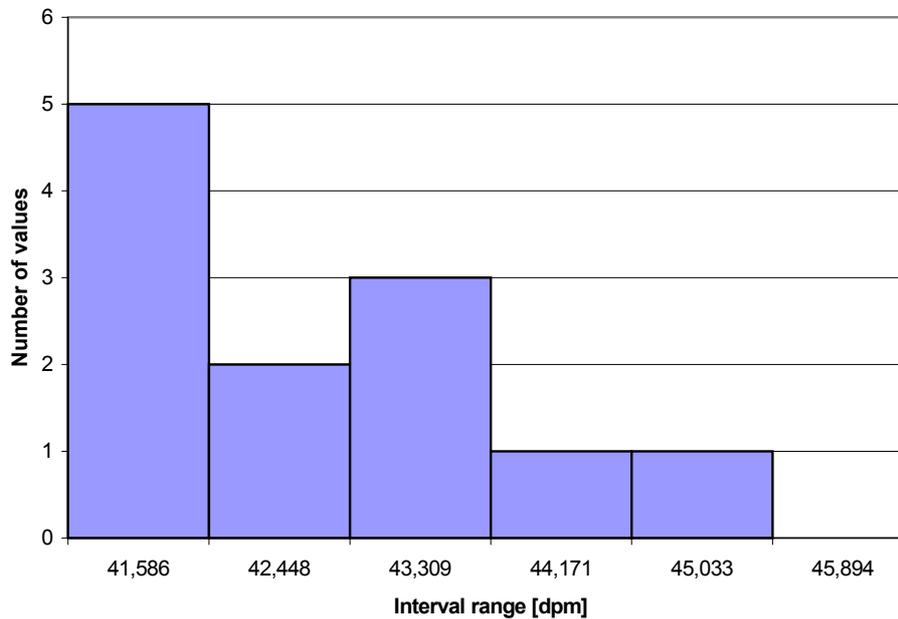


Fig. 2 – Distribution according to values.

The histogram shows a Poisson distribution of values, which is a good approximation of the Gaussian (normal) distribution for a sufficient number of samples.<sup>11</sup> The measured values illustrate the probabilistic behaviour of the disintegration phenomena, so the mathematical statistics methods can be applied to radioactivity calculations.

The two Gaussian parameters, the mean and the standard deviation values were calculated.

The compensation method of the direct values with unequal measurement precisions was used to calculate the error because the precisions of measurements are different<sup>22</sup>.

The values of the statistical precision of measuring the samples,  $2\sigma_i$ , and the calculated values of the ratio  $(2\sigma_{\max}/2\sigma_i)^2$  are shown in Table 7.

Table 7  
 $2\sigma\%$  and  $(2\sigma_{\max}/2\sigma_i)^2$  ratio values

Sample Code	$2\sigma_i$ [%]				$(2\sigma_{\max}/2\sigma_i)^2$			
	I	II	III	IV	I	II	III	IV
D <sub>1a</sub>	0.63	1.50	1.50	1.50	5.67	1.00	1.00	1.00
D <sub>1b</sub>	0.62	1.50	1.50	1.50	5.85	1.00	1.00	1.00
D <sub>1c</sub>	0.63	1.50	1.50	1.50	5.67	1.00	1.00	1.00

The compensation method consists in square ratio of the less precise value of the measurements, defined as  $2\sigma_{\max}$ , to all of the measurement precisions, so that the most precise measurement have the greatest rank of trust, and the less precise

measurement gets the minimum ratio of trust of 1. In this way the calculated mean of the values is compensated by shifting its value to the most precise measurements. In Table 7, the less precise measurement is  $2\sigma_{\max}$  [%] = 1.5, so ratios

$(2\sigma_{\max}/2\sigma_i)^2$  is calculated for each statistical precision value of the measurements.

The estimated mean value of the activity is defined, then, by Equation (2):

$$\overline{DPM} = \frac{\sum_{i=1}^{12} p_i \cdot DPM_i}{\sum_{i=1}^{12} p_i} \quad (2)$$

where:  $p_i = (2\sigma_{\max}/2\sigma_i)^2$

Result:  $\overline{DPM} = 42865.4$

The value of the standard deviation of the activity,  $2\sigma_{DPM}$ , is illustrated in Equation (3):

$$2\sigma_{DPM} = \frac{2\sigma_{\max}}{\sqrt{\sum_{i=1}^n p_i}} \quad (3)$$

where:  $n$  = number of data points.

Result:  $2\sigma_{DPM} = 0.29$  [%]

Because:  $DPM = \overline{DPM} \pm 2\sigma_{DPM}$

Result:  $DPM = 42865.4$  [dpm]  $\pm 0.29$  [%],

With the standard deviation expressed in [dpm], DPM becomes:

$$DPM = (42865.4 \pm 125.6) \text{ [dpm]}$$

The sample activity was calculated using Equation (4):

$$A = \frac{DPM_{sample}}{60 \cdot V_{sample}} \quad (4)$$

where:  $A$  = the activity of the sample [Bq/L];

$DPM_{sample}$  = the activity of the sample in units of nuclear decays per minute;

$V_{sample}$  = the volume of the sample [L].

Result:  $A = (7.14 \pm 0.02) \cdot 10^6$  [Bq/L]

The radioactivity of Ultima Gold liquid scintillation waste was generated by a tritium content of  $(7.14 \pm 0.02) \cdot 10^6$  Bq/L. Presence of another radionuclides in the Ultima Gold liquid scintillation waste was not detected by gamma spectrometry.

Generally, this type of waste may content<sup>7</sup> small quantities of Nb-95, Zr-95, and Sb-124, with the half-life very short of 35, 65, respectively 60 days. Consequently, the waste is stored a certain period of time for radionuclides decay to take place.

### Immobilization of the liquid waste and packaging of the solid waste

Immobilization and packaging of Ultima Gold liquid scintillation waste were carried out with all the operations conducted in a single container, without generating any secondary wastes.

The 218 L and 70 L drums were not filled completely, since an ullage space was necessary to allow mixing to take place.

Performing tests carried out on the two 218 L drums to confirm the quality of waste package content complied with the requirements of the waste package specifications. Analysis bulletins of the working materials were compared to operative normative: empty drum,<sup>23</sup> cement,<sup>24,25</sup> aluminium stearate.<sup>14</sup> The sizes of the two 218 L drums (height, diameter and weight) were within the normal values.<sup>18</sup> The values of the external radioactive contamination were less than 0.0036 Bq/cm<sup>2</sup>, and the values of the gamma radioactive equivalent dose at the drum wall was less than 0.2  $\mu$ Sv/L, relative to the admission limit<sup>18,19</sup> of 4 Bq/cm<sup>2</sup>, respectively<sup>18</sup> 2  $\mu$ Sv/L. After performing penetration test, compression resistance test, and free fall test, visual verifications were carried out on the two conditioned waste drums which did not present deformations and kept their initial sizes. The watering test highlighted the tightness of the two 218 L drums. Consequently, the two conditioned waste drums were labeled and transferred to the National Repository.

There were two quality assurance procedures<sup>18,26</sup> to ensure the management and control of Ultima Gold liquid scintillation waste from its acceptance for conditioning to disposal at

Băița National Repository for Low and Intermediate Radioactive Waste, Bihor county. The main role of quality assurance in conditioning of Ultima Gold liquid scintillation waste is to provide confidence that the quality of waste packages meets the requirements of the national authorities (National Commission for Nuclear Activities Control).

Information on waste packages characteristics and attributes were submitted to the National Repository operator for acceptance prior to the dispatch of any waste packages for disposal.

A list of information was provided for individual waste packages: package number, name of the conditioning plant, date of conditioning, typical waste content (weight) per container (kg), overall package weight (kg), type of immobilizing matrix, activity content, dose rate at the surface, surface contamination.

## CONCLUSIONS

Conditioning of Ultima Gold scintillation liquid waste consists of planned and systematic actions to provide adequate confidence that the processes and products involved satisfy the given requirements for quality.

The Ultima Gold XR scintillation liquid cocktail has exhibited good counting efficiencies, with the waste sample well dispersed throughout the cocktail to allow a close contact between the radionuclide and solvent molecules. This is particularly important for the detection of tritium, which emits low energy.

Immobilization of Ultima Gold scintillation liquid waste into a cement and aluminium stearate matrix is a simple method, with low cost of working materials. The ratios of waste, additives and cement can be varied according to the laboratory studies results. The operator exposure is negligible because there is not any vapor problem. The great values of mechanical resistance and the low rate of tritium released from the waste form offer security for transportation and final disposal.

Radioactive waste packages produced comply with waste acceptance criteria developed by the National Commission for Nuclear Activities Control for disposal at the Băița National Repository, Bihor county, and thus the protection of human health and safety of the environment are ensured.

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