## A 'Method Based Survey' Succeeds in a Spanish Scrap Clearance Project – 15664

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## ABSTRACT

During the last years, a Waste Minimization Project has been implemented in Cofrentes NPP in a collaborative effort performed by IBERDROLA and ENRESA. The final result has been more than 760 t of metallic scrap including containerized materials but also massive surfacial contaminated objects have been managed and finally cleared. The initial project scope included 10 spent fuel pool racks weighting 130 tones but limited testing demonstrated they presented high contamination levels and were managed as radwaste including downsizing and conditioning in ENRESA standard radwaste containers for metallic scrap (CMT`s).

This project started in 2008 and finished just in the summer of 2014. During these years planning and licensing, procurement, execution, including, a huge effort in decontamination and clearance measurement, was developed. The project closure is developing actually.

The residual materials were stored and initially characterized previously its downsizing and decontamination using chemical baths (warm diluted sulfuric acid and ultrasounds), blasting (water jetting with iron grit) and in special cases mopping using industrial cleaners). Some big items were decontaminated previously its downsizing.

The materials were cleared using an approach similar to MARSAME approach based in MARSSIM guidelines. The 160 containers needed six in situ gamma spectrometry measurements each and 115 massive (from 0.45 t to 13.7 t) items need at least so many measurements as total surface in square meters. In total 763 t were cleared with only 0.6 t rejected and treated as radwaste.

#### INTRODUCTION

In the Spanish Cofrentes NPP, a BWR NPP type, a huge amount of contaminated metallic materials were collected from different plant modifications and refurbishments and stored in the Low Activity Parts Warehouse (LAPW). Around 860 t were designated to be managed in a specific Waste Minimization Project supported by ENRESA the Spanish Waste Management Company. A specific project to manage these materials was developed since 2007 to this year. Initially, more than 600 tons of metallic scraps were classified as potentially recyclable scrap. The goal of this work is Recycle for use in a controlled environment (i.e., authorized disposition). Then these scraps will be shipped to a scrap dealer and finally to a steel foundry. The last July 2014 the clearance project was finished and more than 700 t have been cleared.

The NUREG 1757 Supplement 1, well known as MARSAME (ref 1), discusses three principal survey designs: Scan-only, In Situ and "MARSSIM-Type". A fourth survey design, "Method Based", is also mentioned. A similar approach as a "MARSAME method based survey" was developed in Spain since 2004 (ref.2) and currently applied by IBERDROLA in this project. The approach uses a special device holding a HPGE equipment to locate the detector in a 2D geometry. The measurements (gamma spectra)

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performed in such locations are interpreted in the containers measurement as soil MARSSIM approach, and in surface contaminated objects measurements as a surface structure MARSSIM approach. The commissioning phase of clearance process was presented in previous meeting (ref.3). However some information of this phase is included to show consistency and the importance of this phase in all the

project implementation.

# METHODS

The main characteristics of IBERDROLA approach are:

- 1) On site processes in the Candidate Material is considered:
  - a. Historical Material Information and inventory
  - b. Feasibility study and planning
  - c. Initial Characterization using judgmental sampling, and Physical and Radiological Sorting,
  - d. Decontamination (availability and feasibility), including secondary Radwaste management.
  - e. Post decontamination characterization using judgmental sampling and Physical and Radiological Sorting,
  - f. Clearance Measurement and Decision taking.
  - g. Disposition of material: Recycling in Foundry or Radwaste disposal

Typical process was planned as is depicted in Figure 1.



Figure 1. - Typical Material Management Process Flow diagram

2) In each material process step, **Residual Activity Index (RAI)**, defined as a spatial random variable in the material lot or item, is the Quantity of Decision defined by:

$$RAI = \sum_{k=1}^{m} \frac{\widehat{C_k}}{CL_k} = \sum_{j=1}^{p} \widehat{C_j} \left[ \frac{1}{CL_j} + \sum_{l=1}^{q} \frac{\widehat{F_{jl}}}{CL_l} \right]$$

Where  $\widehat{C_k}$  is the estimated mean of the k- nuclide concentration.

 $\tilde{CL}_{k,j,l}$ , are the clearance level of the nuclide of concern in RP-89 (Ref.4)

 $\widehat{C}_{j}$ , is the estimated mean of the j-key nuclide concentration for j=1,...,p

 $\hat{F}_{Jl}$ , is the clearance scaling factor as estimated mean of the concentration ratio between j-th key nuclide and the l-difficult –to-measure nuclide.

According the above formula the analytical uncertainty of the RAI is approximately. 1/2

$$u(RAI)_{anal.} = \left[\sum_{j=1}^{p} u(C_j)^2 \left[\frac{1}{CL_j} + \sum_{l=1}^{q} \frac{F_{jl}}{CL_l}\right]\right]^2$$

Operationally is more convenient to use the relative uncertainty  $\varphi_j = \frac{U(C_j)}{C_j}$  and this becomes:

$$u(RAI)_{anal.} = \left[\sum_{j=1}^{p} \varphi_j^2 C_j^2 \left[\frac{1}{CL_j} + \sum_{l=1}^{q} \frac{\widehat{F_{jl}}}{CL_l}\right]\right]^1$$

And  $\varphi_i$  can be estimated conservatively using a classic least squares curve fitting.

- 3) The *F<sub>jl</sub>* estimator is the non-parametrical upper 95% confidence limit for the true median, the sample order statistic f<sub>(u)</sub>. It is defined in pages 173 and 174 in ref. 5. It is used because the distribution of this scaling factor is unknown, so only non-parametrical robust estimators are statistically sound. In accordance with Tukey (ref. 6), the minimum number of paired data is 5 and usually it will be below 20 data. For instance, if the ordered sampled: f(1)<f(2)<...<f(l)<...<f(u)<...<f(n), this statistic is determined depending "n", i.e. when n=6 to 8, u=n; If n=9,10, and 11, u=n-1; If n= 12,13 or14, u=n-2, etc. If n>20: u = 0.5(n + 1 + 1.96√n) Of course, a graphical and statistical testing (e.g. Spearman correlation coefficient or Kendall tau test) of the relationship between the paired concentrations of the two nuclides is necessary to use this estimator.
- 4) The Decontamination factor is determined comparing pre-decontamination sample RAI mean (or median),  $\widehat{RAI}_{in}$ , with post-decontamination sample RAI mean (or median),  $\widehat{RAI}_{out}$ :

Decontamination factor, 
$$DF(RAI) = \frac{\hat{RAI}_{in}}{\hat{RAI}_{out}}$$

- 5) Clearance Rules of Decision are:
  - a. If the RAI if the average is greater than 1, the lot is rejected,
  - b. If all RAI values are less than 1, the lot is cleared with around 100% of confidence. It is the Sign test. (see ref. 5)
  - c. If the RAI if the average is less than 1 but any value is greater than 1, two tests must be performed:
    - i. Sign test (Median test based in binomial distribution) and
    - ii. Elevated Value Comparison test (Using Grubb's test or 10 CL test)

The use of Sign test is recommended by MARSSIM and our experience indicates that more or less asymmetric measurement distributions are more realistic than Gaussian distributions non parametrical tests are more efficient than parametrical test such as Z-test or T-test when normal or lognormal distributions are applicable.

# **Measurement Quality Objectives**

In total agreement with the approved Methodology, identified Measurement Quality Objectives were:

MQO1: Limit of detection will be less than 0.5 CL, of key nuclides

MQO2: Mathematical calibration will be used assuming uniform activity concentration in the source but it will be validated in a test program, as required by Regulatory body.

MQO3: Each single measurement will not average in volume higher than 1  $m^3$ , or surface higher than  $1m^2$  or mass higher than 1 ton, in accordance with Ref. 8

MQO4: Limit of Detection will be determined on Field Blank Lots or Item.

MQO5: Experimental Measurement results will be validated using Field Reference Lots or Items prepared with traceable sources.

MQO6: Detector Uncertainty will be less than 10% using point check sources without background uncertainty.

MQO7: Detector Relative bias will be less than 10 %.

MQO8: Valid measurements will be greater than 90%.

## **Measurement Equipment Selection**

Typical Portable beta/gamma contamination counters were used in material sorting during the material process.

Specific Clearance Equipment was a commercial portable HPGE gamma spectrometer placed in an automatic –programmable positioning system to monitor 2m<sup>3</sup> containers and massive big items (e. g. fully decontaminated BWR turbine rotor parts).



## Figure 2. - HPGE detector with collimator and placed in automatic positioning device.

Additionally to the supplied commercial software, a specific software was developed to:

- 1) Incorporate the complete clearance methodology including the automatic issuance of clearance reports for each Clearance Unit
- 2) To automatize the detector positioning.

This software will allows to produce automatically the clearance records and to move the detector to specific grid locations in big items clearance and/or to measure a batch of containers placed in lane.

## **Standard Operational Procedures**

To perform these activities eight new SOPs were developed.

- 1. Material Interim Storage Operations. Ref. PA PR-14
- 2. ISOCS Clearance Equipment Calibration. Ref. P-PR/1.4.34
- 3. Material Management and Control in Clearance. Ref. P-PR/2.6.19
- 4. Metallic Scrap radiological Clearance Process Operations", Ref. P-PR/2.6.20.
- 5. Management of Rejected Materials on Clearance Process. Ref. P-PR/2.6.21
- 6. Clearance Process Quality Control. Ref P-PR/2.6.22.
- 7. Radiological Clearance of Metallic Scrap Management. Ref. PG-061
- 8. Management and Control of Cleared Metallic Scrap Shipping. Ref. PG-062

All operational procedures were tested and modified, as it was applicable, according the experience obtained during the Commissioning phase.

#### Low Background Clearance Building

A new warehouse was built in a low background area to place the clearance equipment and candidate materials (see figure 3)



Figure 3.- New Low Background Clearance Building

#### **Clearance Commissioning Phase**

A commissioning program was submitted to the Regulatory Body and developed in two phases.

First phase with planning, field and reporting activities during three last months of 2010 and the three first months of 2011 were performed. Once it was finished the Regulatory Body required some minor change and performing additional assays.

The second phase start with planning since April 2012, field activities during October and November of 2012 and the final report was issued in January of this year. Finally it was approved in August 2013. Main tests and assessments performed in both phases were:

- Energy and efficiency calibrations, using point sources
  - o Energy vs. channel equqtion was tested wih Co-60 and Cs-137 point sources
  - FWHM were tested lower than 9%
  - o Relative bias was clearly lower than source- detector uncertainty
- Background was determined and reported using control charts.,
- Geometry selected was Segmented Box model not single box model it perform the divison of the box in 3 segment: one central and 2 lateral with any volume lower thaan 1m<sup>3</sup>. So each segment was two time measured (one for each side). (see figure 4). It is equivalent to the ISOCS "Single Box Template" but used 3 times.



**Figure 4 Measurement Geometry** 

• Disks, diaphragms and plates were measured using the ISOCS geometry template: "*Circular plane template*" to use in grid design (see enclosed figure 5)



Figure 5. - Hemi-disk: Item picture and grid for scanning

- Cylindrical-like items were measured using the "*Round Tube Int/Ext Contamination*" template.
- Software produces the Efficiency calibration curves for each above template
- Equipment performance checking, using blank material tests: 2 m<sup>3</sup> container with real scrap decontaminated tubes (CMD-CO-15-01 BLANC), steel solid cylindrical coupling, steel solid hemi-disk with 4.24 m<sup>2</sup> lateral area,, and unreal dummy container from ENRESA.
- Measurement data ANOVA was performed to demonstrate optimal measurement times: It demonstrates no significant differences between means obtained with different measurement time:
  - o 300s and 600 s for containers for volumetric concentrations.
  - o 600s and 900 s for singular items with surface contamination
- Field reference sources tests: using Blank material with Point Single High Activity Sources: 5.29 MBq Co-60 and 1.68 E+05 Bq Cs-137 (placed in 21 positions within the container and 6 gamma

spectrometry data for each position: 126 spectra). and Uniformly distributed sources using a set of 100 Cs-137 & Co-60 Exempted sources (760 Bq average source) totalizing 76000 Bq

- Complete Clearance process and procedures tests to demonstrate the whole operation with containers and big items were performed including the issuance of completely automatized clearance records
- Container sample data were compared with clearance equipment in situ measurement results. The ISGS measurement statistics were higher than the laboratory data statistics.
- Rejected container simulation was performed giving to the clearance software a set of simulated measurement data with high values.

# **Execution phase**

The main issues during this phase were:

- 1) Contracting heavy transportation, cutting, handling and decontamination services.
- 2) Coordination of different agents including the clearance measurement device and operator free supplied by ENRESA.
- 3) Developing and incorporating the software to the ENRESA equipment.
- 4) Residual materials handled are in tables I and II.
- 5) In total, 162 containers were produced but two were rejected and newly decontaminated and finally cleared. The total containerized scrap cleared was around 155 tons.
- 6) A total of 115 massive items surface contaminated were decontaminated and measured. Only one item was rejected by the clearance process. The contaminated part was decontaminated but decontamination failed and the contaminated part (around half ton) was cut and managed as radwaste.

	Container		
Containerized materials	amount	Mass (kg)	%
MSR tubes	61	58910	37.98%
MSR beam plates	26	26465	17.06%
Low Pressure turbine A rotor blades	22	22284	14.37%
Low Pressure turbine B rotor blades	15	15734	10.15%
Miscellaneous	36	31,698	20.44%
Total	160	155,091	100.00%

7) The total cleared material was around 763 t.

 Table I. Cleared Containerized materials (100%)

	Items	
Surface (potentially) contaminated massive items	amount	Masa (kg)
Low Pressure Turbine A rotor parts	22	150,045
Low Pressure Turbine B rotor parts	21	137,550
Total	43	281,235
Low Pressure turbines diaphragm parts	72	320,845
Rejected Low Pressure Turbines diaphragms parts	1	555
Low pressure turbine B diaphragm parts cleared	72	320,290
Massive items cleared	115	607,885

Table II. Cleared Massive items (99.91%)

#### DISCUSSION

## **Commissioning phase**

The main results were during the calibration validation tests :

- The high variability of single measurements with Co-60 single sources in blank containers. From 10 to 0.01 times the real container Co-60 source. Only in two cases the source was not correctly (or not conservatively) measured.
- The high variability of single measurements with Cs-137 single standard sources placed in blank containers: 50 to 0.01 times the source. In all cases the Cs-137 source was correctly (or conservatively) measured.
- The summarized comparison between the container estimated concentration mean vs. the real concentration mean is in Table III.

Nuclide	Real mean	Minimum	Maximum	Averaged	Average
	concentration	estimated	estimated	Estimated	bias
		mean	mean	Mean	
Co-60	5.34 Bq/g	2.66 Bq/g	73.2 Bq/g	24.67	19.33
				Bq/g	Bq/g
Cs-137	0.17 Bq/g	0.21 Bq/g	5.56 Bq/g	1.46 Bq/g	1.29 Bq/g

**Table III. Measurement Mean Bias Estimations** 

In only 2 cases to the 21 (less than 10%) the Co-60 mean was underestimated. Both underestimations could be originated from the known spatial location of the source in respect the detector and the real variability in matrix attenuation.

- The results using dummy container with uniform source distribution was consistent with this results but, as it was foreseeable, presenting lower variability.
- The results demonstrate it is a very conservative clearance system for containers:

	Container with single high activity sources		
Nuclide	Measurement type	Relative bias average	
Co-60	Single (maximum)	362,32%	
	Average	55,26%	
Cs-137	Single (maximum)	826,45%	
	Average	186,24%	

#### **Table IV. Single Measurements Bias Estimations**

It suggests the convenience of correction factors for Cs-137 measurement at least.

• The total uncertainty of the mean in containers depends of sampling, analytical and matrix effect uncertainties (ref. 8). It can be estimated using the following equation:

$$u_{\bar{c}} = \frac{\bar{c}}{\sqrt{n}} \sqrt{\varphi_s^2 + \varphi_a^2 + \varphi_T^2}$$

Nuclide	Relative uncertainty Sampling	Relative uncertainty Analytical	Relative uncertaint y Matrix	Mean Total Relative uncertainty (N=6)	Mean Total Relative uncertainty (N=12)
Co-60	111%	7.5%	26.3%	46.9%	33.1%
Cs-137	110%	10.0%	25.6%	46.6%	32.9%

Table V. Measurements Uncertainty Estimation	Table V.	Measurements	Uncertainty	<b>Estimations</b>
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According this the analytical uncertainty could be not quoted in respect the matrix variability uncertainty and the radioactivity distribution uncertainty. In terms of container concentration average uncertainty the most important variable is the spatial distribution of the radioactivity (around 45% with six measurements), in second position is the matrix heterogeneity (around 11%) and in last position is the measurement uncertainty (around 4%)

#### **Execution Phase**

The long duration of the project (from 2008 to 2014) is explained by different circumstances including:

- 1) The unusual and complex licensing process, 2008-2013
- 2) The necessary compatibility with the operation of the NPP: Outages
- 3) The availability of human and technical resources: Heavy load transport, measurement equipment, software testing, diamond wire contractor, etc.

The logistic was another complex issue due the heavy load to be managed. The turbine rotors weight was around 20 tons each so requiring contracting of heavy load transport, and many other items weighted more than 5 tons.

Cutting of massive items was performed using diamond wire by a contracted specialist.

Decontamination was performed with the plant equipment when it was available in Hot Workshop using chemical baths (warm diluted sulfuric acid and ultrasounds), blasting (water jetting with iron grit) and in special cases mopping using industrial cleaners. Some big items were decontaminated in a Turbine Dock SAS previously its downsizing. Decontamination factor between 10 but 30 was achieved with strongest decontamination techniques. Higher values had been non economical.

Estimations	Initial	Post-decontamination	Estimated
	Characterization	Characterization	Decontamination
	RAI	RAI	Factor
Mean	1,048	0,05	19
Median	0,680	0,05	12
95% UCL mean	2,369	0,07	35
95% UCL median	3,982	0,07	57

**Table VI. Decontamination Factor Estimations** 

The clearance measurement and decision process was performed in less than eight months.

The eight new operational `procedures were developed, reviewed, as necessary, and tested

The main conclusions from statistical analyses performed according past projects (see ref 7) of the container measurements were as follows:

- 1) Around the 40.34% of Co-60 values have null value .They are censored values due the optimized measurement time (300 s) . The lower measured value was 0.0183 Bq/g (1.83% of 1 Bq/g clearance level in RP-89 y al 18,3% of 0.1 Bq/g IAEA exemption/clearance level).
- The Co-60 data distribution can be fitted to a joint distribution from adding a degenerated distribution with null value and a tri parametrical lognormal distribution with mean =0.143 Bq/g, standard deviation= 0.166 Bq/g and threshold =0.010. The degenerated distribution weight is 40.34% and the lognormal distribution weight is 59.7%. According this total average is 0.085 Bq/g and total standard deviation is 0.114.
- 2) Similar results are found for Cs-137 data. The data fitted a joint distribution from a degenerated distribution with value zero (due optimized measurement time) and a lognormal distribution  $LN(\mu=0.093, \sigma=0.071, t=0.006)$ .
- 3) The RAI data are fitted to a joint distribution from a degenerated distribution but in the 1.41E-5 value (It is the result to use conservatively the fixed value 1.41E-5 Bq/g for Tc-99 concentration in the RAI calculations) and other distribution that could be log logistic or lognormal. We decide use lognormal only because it is the type of distribution well fitted in Co-60 and cs-137 data. This lognormal distribution is with mean = 0.199045, standard deviation= 0.176099 and t = -0.0092. From the data we estimate the degenerated distribution weight is 14.5% and the lognormal distribution weight is 85.5% so the joint average will be 0.17 similar to the sample average is 0.175. The joint standard deviation is 0.177 is similar to the sample standard deviation is 0.226.

# CONCLUSIONS

- 1) The project planning used Data Quality Objectives For instance, optimal measurement times were selected: 300 seconds for each container measurement (less than 1 hour per container) and 600 seconds for each surface activity measurement.
- 2) The statistical distributions of the results demonstrates the necessity to use non parametrical hypothesis testing for clearance processes because data do not fit Lognormal or Normal distributions
- 3) All the measurement system performs automatically not only the measurement but the detector location (minimum operator intervention).
- 4) Clear SOP's and QC protocols are implemented to ensure the correct decision making.
- 5) The next Cofrentes NPP Clearance operations can be performed using the new SOP's.
- 6) The approaches of this project can be used in other projects such as current and future decommissioning projects or operational residual materials management project
- 7) Finally, our estimated total cost of this project was around 3.7 M€but the alternative to disposal all the materials as radwaste was 6.9 M€ Taking into account other costs this project has saved around 3 M€ to the Spanish radwaste system.

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