Management of metals resulting from an Italian nuclear facility: techniques for clearance and unconditional release.

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1 . INTRODUCTION

The start of decommissioning of nuclear facility in Italy leads to an appreciable increase in the volume of metal materials that will need to radiological characterization for the unconditional release.
The nuclear fuel reprocessing facility ITREC, located in Rotondella (MT) in the south of Italy, is currently in safety maintenance. As part of these activities was necessary, the replacement of the drainage pipe (approximately 5000 m) that discharges facility's liquid effluents.
The entire pipeline is in treatment within a small Waste Management Facility suitably equipped for the cutting, the separation of the non-metallic residue and decontamination. 100% of the pipe portions are characterized by measures of gross alpha - beta - gamma total range and high resolution spectrometry in order to verify the removal without radiological constraints. The comparison of the levels of surface activity and specific activity prescribed by the national Competent Authority is through measurement activities implemented according to specific MQO defined for specific processing.
The methodology described in this article provides a good example of management, treatment and decontamination of metallic materials for unconditional release.

2. REGULATORY FRAMEWORK

The version of the Basic Safety Standards [1] suggests the levels of exemption (Exemption Level), which are defined in a more restrictive sense in national law. [2] The unconditional clearance levels (Clearance Level) are also suggested in the publications Radiation Protection (RP) [3], [4], [5]: in particular, the RP 89 offers specific levels of removal (Specific Clearance Level) for metals.

The definition of clearance levels, whether they are general or specific, guarantees the radiological non-relevance of materials removed without needing additional controls: the same RP 122 [4] Part I to par. 5.1 states that the materials removed should not be subjected to further examination, otherwise it would contradict the principle of Unconditional Removal that prescribes instead the release of materials from the regulatory system (Clearance = release from regulatory requirements).
The Italian legislation on radioprotection [2] (Article 154 paragraph 3 bis of Legislative Decree 230/95 and subsequent modifications and additions) states that "... the removal of installations subject to authorization ... of materials containing radioactive substances intended to be disposed of, recycled or reused .... is subject to specific requirements to be included in authorization measures... The clearance levels ... take into account the guidelines, recommendations, and technical guidance provided by the European Union". The Operating License of ITREC Trisaia [5], issued by the Ministry of Economic Development pursuant to art. 50 of Legislative Decree 230/95, and subsequent modifications and additions, requires compliance with technical and management requirements - issued by the Control Authority (ISPRA - Institute for Environmental Protection and Research), in particular the management prescription 2.9 [6] indicates the surface and mass concentration levels established for the unconditional removal of metal solid materials by ITREC facility.
An indispensable operative indication about the methods and procedures of radiological control for

the purpose of removal is provided by the UNI 11458:2012 [7], and it outlines strategies for the measurement of low levels of radioactivity in solid materials from nuclear facility. ☒

The foregoing represents the regulatory framework for the verification of conditions for metals unconditional removal from nuclear installations.

3. PROCESSING AND CHARACTERIZATION

3.1 Operational history of drainage discharge

Over the years of operation of the ITREC (pre- nuclear tests : 1970 ÷ 1975; nuclear tests : 1975 ÷ 1978) , the liquid effluents were made by cooling water of process components, by the drainage of building process premises , by condensation from the monitoring tank, by intermediate layer evaporator condensation and the water drained from the aquifer to ensure water protection of underground rooms (Local HLW , LLW , Storage Pool , Underground Repository 7.1 and 7.2). After the stop of nuclear activities, from the beginning of the eighties to the present, the discharges are made predominantly from groundwater drainage for water protection of underground rooms. These drains are collected in 3 containment tanks , each with a working volume of about 600 cubic meters , which after analysis and verification of their compatibility with the formula of discharge authorized by the control authority , are drained in the Ionian Sea through a special metal pipe. The sea drainage discharge has been entirely replaced in 2008 and the old pipe with a length of about 5400 m, was stored in 12 containers in pieces from about 5 m, for a total gross volume of about 140 m^3 and a total mass of about 120 tonnes.

3.2 Preliminary operations to characterization

A treatment process as described below preceded the activity of radiological characterization [8, 9]:

• Identification of container and positioning in front of the tunnel overlooking to the equipped cells (WMF Wast Management Facility);
• extraction of some sections of the pipe from the container, control of the exposure rate and positioning in the tunnel entrance of the WMF;
• subdivision of the pipe contained in a specific container in batches made of 4÷5 sections each;
• transfer of each lot in the cells of treatment;
• detection and identification of each section of pipe;
• carrying out preliminary radiometric checks;
• removal of bitumen and mineral tissue protective coating;
• collection and packaging of removed bituminous material in 380 liter overpack for subsequent characterization;
• cutting into two parts, each of about 2 meters, of each section;
• univocal identification of the pipe's parts produced by cutting;
• inner cleaning of each of the two parts of pipe' section, using appropriate brushes, wire brushes and / or other tools, with simultaneous collection of the powders by dedicated aspirators;
• further cutting into about 50 cm parts, already identified in the section of treated pipe;
• crushing, with press of 60 Mg. of 50 cm parts and longitudinal cutting of each single piece with plasma torch
• controls the total surface contamination and surface contamination controls transferable through two smear tests for each of the two portions of single 50 cm piece, both on the outer and on the inner surface;

• insertion of treated drainage pipe lot in 380 liters Overpack for subsequent characterization by ISOCS system.

In the event that the activities of pre-characterization should highlight traits of pipe potentially not releasable, they would be inserted into standard 220 liter oil drums and treated as radioactive waste.

3.3 Characterization Plan

For the purposes of unconditional release of materials, it was verified that the levels of mass concentration and surface were respected for various radionuclides listed in Table 1 extracted from the document [6] and in particular by the prescription management 2.9.

Radionuclide (i)	(C_{li}) Metallic materials		(C_{li}) Concrete materials		(C_{li}) Other materials
	Surface (Bq/cm^2)	Mass (Bq/g)	Surface (Bq/cm^2)	Mass (Bq/g)	Mass (Bq/g)
^{3}H	10.000	1	10.000	1	0,1
^{55}Fe	1.000	1	10.000	1	0,1
^{60}Co	1	1	1	0,1	0,1
^{59}Ni	1.000	1	10.000	1	0,1
^{63}Ni	1.000	1	10.000	1	0,1
^{90}Sr	1	1	100	1	0,1
^{134}Cs	1	0,1	1	0,1	0,1
^{137}Cs	10	1	1	1	0,1
^{152}Eu	1	1	1	0,1	0,1
^{154}Eu	1	1	1	0,1	0,1
α emitters (*)	0,1	0,1	0,1	0,1	0,01
(*) As U and/or Th					

Table 1: *Levels of unconditional removal*

The condition to be met in the case of multiple radionuclides is the following:

$$\sum_i \frac{C_i}{C_{li}} < 1$$

being C_i the concentration of the i-radionuclide and Cl_i the appropriate limit value.
In order to have conditions even more conservative was assumed the value 0.8, so the condition to be met will be the following: $\sum_i \frac{C_i}{C_{li}} < 0{,}8$
The verification of condition "Surface" recalled in Table 1 was carried out on 100% of the areas to be investigated.
The determination of surface contamination of radionuclides emitting alpha and beta emitters was carried out in terms of concentration "gross alpha", "beta total" and "gamma total" using portable instrumentation described in paragraph 3. Not being able to discriminate the contribution of each radionuclide present in Table 1, was adopted the most restrictive level among those present in the

table. The instrumental data have been corrected conservatively for the efficiency of radionuclide worse detectable and for the extension of the sensitive area.
In order to estimate the concentration of activity was used a system of high-resolution gamma spectrometry with HPGe detector as described in paragraph 3. Measures of activity concentration were conducted on a sample representative of the total quantity of material to be removed (see par. 4.1.2). For the determination of the scaling factors between radionuclides easily detectable (Easy To Measure ETM) and hard to detect radionuclides (Hard To Measure HTM) the analyzed material has been considered, for quantity and radiological spectrum, as belonging to the facility section pertaining to "Liquids Waste Tanks" (LLW and HLW) or to "Process Cells Laboratories", as described in document [11].
To verify compliance with the limits given in Table 1 were measured values, expressed in Bq / g, of the radionuclides ^{137}Cs, ^{134}Cs, ^{60}Co, ^{152}Eu and ^{154}Eu determined by gamma spectrometry, considering conservatively the value of MDC (Minimum Detectable Concentration) for not measurable concentration. The activity concentrations of HTM radionuclides such as ^{90}Sr, ^{233}U, ^{235}U, ^{238}U and ^{232}Th, were estimated by correlation with ^{137}Cs, using the factors listed below (see documents [10] and [11])

$$\frac{^{235}U}{^{137}Cs} = 3.64\times10^{-6} \qquad \frac{^{233}U}{^{137}Cs} = 1.28\times10^{-4} \qquad \frac{^{238}U}{^{137}Cs} = 5.16\text{x}10^{-8}$$

$$\frac{^{90}Sr}{^{137}Cs} = 1 \qquad \frac{^{232}Th}{^{137}Cs} = 6.18\times10^{-6}$$

The activity of ^{3}H was evaluated by correlation with the ^{137}Cs using the factor obtained from a calculation code reported to element of the fuel (Elk River EER R22) with the highest Burn Up among those stored in the ITREC facility pool.

$$\frac{^{3}H}{^{137}Cs} = 1.08\times10^{-3}$$

The activities of ^{55}Fe, ^{59}Ni and ^{63}Ni were evaluated by correlation with the ^{60}Co using the scaling factors below

$$\frac{^{55}Fe}{^{60}Co} = 1.7\times10^{-3} \qquad \frac{^{59}Ni}{^{60}Co} = 1{,}67\times10^{-1} \qquad \frac{^{63}Ni}{^{60}Co} = 19$$

3.3.1 EQUIPMENT USED

For surface contamination's measurements it has been used the portable contamination monitor Berthold LB 124. It has a detection surface of 170 cm^2 equipped with a scintillation detector ZnS for measurement of "total alpha" and "beta / gamma total". For the determination of the concentration of activity is used a system of high-resolution gamma spectrometry. It is constituted by an HPGe detector and a hardware unit with a MCA (multichannel analyzer) that operates associated to a computer equipped by a dedicated software, which controls the entire flow of information from the MCA to the computer. The calculation of the activity is determined by factor

of proportionality between the activity and net counts obtained from the spectrum output multichannel analyzer MCA, using the calibration models Gamma Vision.
Combining the data response characteristic of the detector with the measuring geometry and the chemical, physical and geometrical characteristics of the object analyzed, the software is able to determine the efficiency curve.

The specifications of the measuring system are listed below:

• coaxial HPGe detector Detector System

• energy range: 50-3000 keV / 5-1500 keV

• relative efficiency: 50% - 55%

• resolution: 1.9 to 1332 keV; Peak / Compton: 66:1

4. RESULTS AND CONCLUSIONS

At the end of the decontamination and characterization process, has been verified that 90% of the processed material is releasable without radiological constraints. Below is a sample of data analysis collection form for determination of surface contamination of alpha and beta - gamma emitters radionuclides (Table 1) and a copy of the final report on high-resolution gamma spectrometry in ISOCS configuration (Table 2).

SCHEDA IDENTIFICATIVA TRATTO TUBAZIONE CONDOTTA A MARE (dopo pulizia interna) **TRATTO N° ____4.02.A_____**																					
Spezzone n°	CSR (Bq/cm²) (superficie **E**sterna parte ***a*** e ***b*** spezzone)								CSR (Bq/cm²) (superficie **I**nterna parte ***a*** e ***b*** spezzone)								CST (Bq/cm²) Parte **a**		CST (Bq/cm²) Parte ***b***		NOTE
	1***a*** Est		2***a*** Est		1***b*** Est		2***b*** Est		1***a*** Int		2***a*** Int		1***b*** Int		2***b*** Int						
	β/γ	α	β/γ	α	β/γ	α	β/γ	α	β/γ	α	β/γ	α	β/γ	α	β/γ	α	β/γ	α	β/γ	α	
1	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
2	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
3	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
4	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
5	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
6	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
7	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
8	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
9	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	
10	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	NR (α)= ≤ $3{,}42 \cdot 10^{-4}$
11	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	NR (β/γ)= ≤ $2{,}48 \cdot 10^{-3}$
12	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	-1,80E-01	-1,25E-02	-1,80E-01	-1,25E-02	

Table 1. *Data collection form for the determination of surface contamination of alpha, beta - gamma emitters radionuclides*

Radionuclide (i)	C_i Massa (Bq/g)	C_{li} Massa (Bq/g)
^{3}H	1,21E-06	1
^{55}Fe	1,09E-06	1
^{60}Co	6,39E-04	1
^{59}Ni	1,07E-04	1
^{63}Ni	1,21E-02	1
^{90}Sr	1,12E-03	1
^{134}Cs	1,02E-03	0,1
^{137}Cs	1,12E-03	1
^{152}Eu	3,12E-03	1
^{154}Eu	2,01E-03	1
^{233}U	1,43E-07	0,1
^{235}U	4,08E-09	0,1
^{238}U	5,78E-11	0,1
^{232}Th	6,92E-09	0,1
$\Sigma(C_i/C_{li})$ Massa = 3,05E-02		

Table 2. *Final report of high-resolution gamma spectrometry.*

The recorded values turned out much less than the instrument sensitivity and in any case not exceeding the limits of unconditional release.
In Table 1, it is possible to verify the surface condition for the section of analyzed pipe.
The results of the determinations collected in Table 2 show the values of activity concentration per unit mass for HTM and ETM radionuclides: the values of ^{60}Co , ^{137}Cs , ^{134}Cs , ^{152}Eu and ^{154}Eu were detected directly by gamma spectrometry and the values ^{3}H , ^{55}Fe , ^{59}Ni , ^{63}Ni , ^{90}Sr , ^{233}U , ^{235}U , ^{238}U and ^{232}Th were calculated using scaling factors, the last row shows the result of the sum defined in paragraph 3.3 .
At the end of the activities of radiological characterization, the releasable metal parts will be launched in fusion with the constraint on the mixing ratio of 1 to 10 with metallic material from different origin.
Activities , subject to the Control Authority, allow to remove unconditionally metallic materials

originated from the removal of the discharge pipe's liquid effluents from nuclear fuel reprocessing facility ITREC managed by Sogin S.p.A. The methodology described provides a good example of management, treatment and decontamination of metallic materials for unconditional removal.

BIBLIOGRAPHY

[1] Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation (OJ no. L 159, 29.6.96, p. 1-114).

[2] D.Lgs. 17 marzo 1995, n. 230 "Attuazione delle direttive Euratom 80/836, 84/467, 84/466, 89/618, 90/64, 92/3 in materia di radiazioni ionizzanti" e ss.mm.ii.

[3] Radiation protection 89 "Recommended radiological protection criteria for the recycling of metals from the dismantling of nuclear installations".

[4] Radiation protection 122 "Practical use of the concepts of clearance and exemption – part I" Guidance on General Clearance levels for Practices.

[5] Decreto Ministeriale del 26 luglio 2006. Ministero dello Sviluppo Economico, Direzione Generale per l'Energia e le Risorse Minerarie, Uff. B6. "Licenza di Esercizio dell'Impianto ITREC di Trisaia –Rotondella (MT) e della stazione SIRTE-MOWA".

[6] APAT RIS ITR 02/2006 "Prescrizioni per l'esercizio ai fini del mantenimento in sicurezza dell'impianto ITREC e per l'esecuzione delle attività propedeutiche alla disattivazione".

[7] UNI 11458:2012 "Materiali solidi provenienti da impianti nucleari. Metodi e procedure per il controllo radiologico ai fini dell'allontanamento"

[8] SOGIN IT BR 00015 Rev.01, "Caratterizzazione radiologica, ai fini dell'allontanamento incondizionato, di tratti dismessi della condotta di scarico a mare degli effluenti liquidi dell'impianto ITREC Trisaia".

[9] SOGIN IT BR 00036 Rev.01, "Trattamento e caratterizzazione radiologica della tubazione dismessa della condotta di scarico a mare effluenti liquidi, Impianto ITREC – Trisaia".

[10] SOGIN IT G 0003 Rev03 "Rapporto Finale di Sicurezza dell'impianto ITREC".

[11] Doc. IT ME 0004 "Presupposti Tecnici al Piano di Emergenza Esterna dell'Impianto ITREC".