Italian Foundry Contamination Due To Cs-137

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ITALIAN FOUNDRY CONTAMINATION DUE TO CS-137

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ABSTRACT

In late October 2005 at the Beltrame plant in Susa Valley (Italy) a radioactive source was accidentally burned in the blast furnace. The source was not discovered by the detectors at the entrance, evidently because of some shielding effect. This caused the contamination of foundry dust, air intake system, and of the filters. There was no dispersion in the environment and risks to workers. All contaminated dust was collected in big bags and placed within containers waiting to know for their destination. The global activity is now estimated at 4 GBq. Five different contamination scenarios, have been analyzed, i.e., possible ways by which such an amount of radioactive material could have been introduced into the foundry. The considered contamination (4 GBq) is compatible with the total activity of some calibration sources. In both cases, Cs-137 is in the form of a quite small radioactive source capsule, sealed and surrounded by a shielding material assembly, such as Pb. The source capsule would have a quite high radioactive concentration, and then a total mass in the order of grams. It has therefore to be classified – for sure – as a High Radioactivity Material, or, once it is inadvertently thrown away, as High Level Waste (HLW), according to the Italian regulation (III Categoria). The lead assembly shielding explains why the assembly passed through the check of the gamma sensor without creating alarm.

INTRODUCTION

The use of radioactive materials for medical treatment is established and controlled throughout the entire life of the sources. Nevertheless, the Goiânia (Brazil) accident revealed the possibility that unattended sources may become extremely dangerous items. In September 1987, in Goiânia, a teletherapy unit was abandoned after the moving of a private radiotherapy institute and the materials abandoned in the premises where left to the investigation activities of the local population. The capsule containing the caesium chloride salt was stolen and opened with the subsequently release of the radioactive material in the environment. The contamination was both direct and indirect and it was distributed among the population as the capsule itself was sold as scrap and small particles of the caesium salt were also sold or distributed as they were glowing in a blue light in the dark, thus fascinating the unknowing people.

Four people died for the consequences of the internal and external irradiation, while other 250 were contaminated and treated at local hospitals [1]. Thorough environmental clean up took place in order to remove the potential residual sources of contamination and to bring the area back to the original situation which allowed for normal living conditions.

The remembering of this accident, for a long time indicated as one of the most relevant accidents in the nuclear history, was revamped when in an Italian foundry, in October 2005, an abnormal radioactivity level was measured in the dust and the suspect that an unexpected, and undetected at entrance in the plant, radioactive source was burnt among the other scraps became evident.

BACKGROUND

CAESIUM PROPERTIES AND USE

Caesium is a chemical element in the periodic table that has the symbol Cs and atomic number 55. It is a soft silvery-gold alkali metal and it is liquid at or near room temperature.

Major caesium deposits can be found at Bernic Lake in Manitoba. The deposits in that area are estimated to contain 300,000 metric tons of pollucite at an average of 20% caesium.

Caesium has a great number of isotopes (39 known) and their atomic masses range from 112 to 151. Of these, only one isotope, ^{133}Cs, is naturally stable, while most of the others have half-lives from a few days to fractions of a second.

^{137}Cs is a radioactive isotope which is formed mainly by nuclear fission in power plants and from the detonation of nuclear weapons. Its half-life is 30 years, and it decays by pure beta decay to a metastable state of ^{137}Ba. Barium-137m has a half-life of minutes and is responsible for all the of gamma ray emission. The ground state of barium-137 is stable (Table1).

Ba-137m can be used in food irradiation or in cancer treatment, thanks to the energy of its photon (about 800 keV).

TABLE 1
Data on Caesium-137 (adapted from [1])

Emissions		
	Gamma	0,66 MeV (84%)
	Beta	from 0,51 MeV (95%) to 1,17 MeV (5%); mean 0.187 MeV
Half-life		30 years

^{137}Cs is used for brachytherapy in the form of small spherical sources. Compared to ^{60}Co, caesium has the advantage of a lower price, due to its direct derivation from the normal operation of nuclear power plants; besides, ^{137}Cs has a longer half-time (30 years) and thus it is not necessary to change the source to maintain a high activity as often as with ^{60}Co; however, the energy of its γ photons is lower (0,662 MeV) and its specific activity is 10 times lower: thus, keeping constant the activity, the volume of the source is quite bigger than that of ^{60}Co.

^{137}Cs can be used in teletherapy machines, it is used in the form of highly soluble caesium chloride salt and it is contained in a sealed radioactive source capsule. This is set in a source wheel, made of lead and stainless steel, to form the rotating shutter mechanism. To produce a radiation beam, the shutter is rotated electrically to align the source capsule with the radiation aperture. As a measure to reduce the risk of uncontrolled contamination, the sealed radiation sources are usually produced in selected laboratories and are identified with a serial number.

CLASSIFICATION OF RADIOACTIVE WASTES

US regulations. In the United States, radioactive waste is classified according to five categories which range from high-level to low-level radioactive wastes. Materials are also classified according to the amount (quantity) of elements heavier than uranium, to their belonging to the uranium milling residues, or to the family of naturally occurring radioactive materials (a class to which also radioactive materials produced in an accelerator are associated).

High-level radioactive waste is spent nuclear fuel or highly radioactive waste produced if spent fuel is reprocessed. Spent nuclear fuel is used fuel from nuclear power plants. If the spent fuel is reprocessed, the materials that are left over after the collection of the reusable material is classified as high-level radioactive waste. The United States is not presently reprocessing spent nuclear fuel.

Uranium mill residues, or tailings, are the materials which remain after uranium has been extracted from the mined ore: these tailings cannot be classified as low-level radioactive waste.

Certain materials, such as radioactive waste that contains more than specified concentrations of elements heavier than uranium (transuranics), naturally occurring radioactive material or radioactive material produced in an accelerator are not under the jurisdiction of the Federal Nuclear Regulatory Commission (NRC) and cannot be classified as Low-Level Radioactive Wastes.

As a consequence, low-level radioactive wastes include a variety of different materials. Some examples are wastes generated at facilities such as nuclear power plants, hospitals, and research institutions. Many materials and equipments can be associated to the low-level wastes category: these might me radioactive materials used in various processes or contaminated supplies and equipment. Hereafter some examples [2] are provided:

- from nuclear power plants:
  - tools, piping, components, general equipment used e.g. for ordinary maintenance;
  - unrecoverable equipment and spare parts, such as ion exchange resins and filter materials (these are used for processing water);
- from research sites:
  - any research equipment from laboratories which deal with and handle radioactive materials;
  - disposable equipment such as: protective coat, shoe covers, towels and other supplies that might have been used in a place where radioactive material is present;
- from hospitals and medical centres:
  - towels, containers, paper, liquids, etc. and equipment which might have been contaminated during their use for diagnose or treating in hospitals;

- air filters used in areas where airborne radioactive contamination has been tested.

No liquid waste can be received as low-level radioactive waste disposal facility.

Due to the variability of the nature of the low-level radioactive waste, its concentration of radioactive material can belong to a wide range of values. Because of this, low-level waste is divided into four classes with specific regulations for each class.

The four classes of low-level radioactive waste are Class A, Class B, Class C, and Greater Than Class C and are defined in the Code of Federal Regulations, Title 10, Part 61 (10CFR61) [3]. The materials belonging to all classes, except for the fourth, are generally acceptable for near-surface disposal. Section 10CFR61.55 lists the radioactivity concentration limits of specific radioactive materials (Table 2 reports values for the Cs isotope) allowed in each low-level waste class.

TABLE 2
Classification determined by short-lived radionuclides, (excerpt of ^{137}Cs) [curies per cubic meter] [3]

	Class A	Class B	Class C
Cs-137	1	44	4600

As it can be derived from Table 2, the description of low-level radioactive waste is usually given according its radioactivity i.e. the rate at which radiation is released by the material. Another common way is to provide the volume of the waste itself.

Italian Regulations - Low-Level Waste according to Italian Regulation. In most countries with a nuclear program, the waste management strategies are based on deep geological disposal of High-Level Waste (HLW), while a less sophisticated disposal method, mostly a near-surface type repository, is used for Low-Level or Intermediate-Level Waste (LLW/ILW)[4].

Italian waste management regulations deal with National Laws on radioactive materials [5], and with Technical Guides from the Italian nuclear regulatory committee [6]. Wastes are classified into three categories ("*I Categoria*" = First category = Very Low-Level Waste, "*II Categoria*" = Second Category = Low-Level Waste, "*III Categoria*" = Third Category = High Level Waste) according to concentration limits for radionuclides.

Second category waste may be defined "Low-Level Waste" since it may be disposed of in surface or near-surface disposal sites, in a similar way to Low-Level Waste eligible for Shallow Land Burial according to the United States Waste Regulation 10CFR61. First category waste may be defined "Very Low-Level Waste" since it decays down to radioactive concentration levels comparable to natural substances in a maximum decay period of some years. Tables 3 and 4 list a series of maximum concentrations for radionuclides either in the case of conditioned wastes or in the case of unconditioned ones.

TABLE 3
Concentration limits for second category radioactive wastes, conditioned (translated from [6])

Radionuclides	Concentration
α emitters $t_{1/2}$>5 years	370 Bq/g (10 nCi/g)
β/ γ emitters $t_{1/2}$>100 years	370 Bq/g (10 nCi/g)
β/ γ emitters $t_{1/2}$>100 years in activated metals	3,7 K Bq/g (100 nCi/g)
β/ γ emitters 5< $t_{1/2}$≤100	37 K Bq/g (1 μCi/g)
^{137}Cs and ^{90}Sr	3,7 M Bq/g (100 μCi/g)
^{60}Co	37 M Bq/g (1 mCi/g)
^{3}H	1,85 M Bq/g (50 μCi/g)
^{241}Pu	13 K Bq/g (350 nCi/g)
^{242}Cm	74 K Bq/g (2 μCi/g)
Radionuclides with $t_{1/2}$≤5 years	37 M Bq/g (1 mCi/g)

TABLE 4
Concentration limits for second category radioactive wastes, unconditioned (translated from [6])

Radionuclides	Concentration
Radionuclides with $t_{1/2}$>5 years	370 Bq/g (10 nCi/g)
^{137}Cs and ^{90}Sr	740 Bq/g (20 nCi/g)
Radionuclides with $t_{1/2}\leq$5 years	18,5 K Bq/g (500 nCi/g)
^{60}Co	18,5 K Bq/g (500 nCi/g)

CASE HISTORY

In late October 2005, at the Beltrame plant in Susa Valley (Italy) a radioactive source was accidentally burned in the blast furnace. The source was not discovered by the detectors at the entrance, evidently because of some shielding effect. This caused the contamination of foundry dust, air intake system, and of the filters. There was no dispersion in the environment and risks to workers. All contaminated dust was collected in big bags and placed within containers waiting to know for their destination. The global activity is now estimated at 4 GBq. In 2004, there was already a similar incident occurred in Vicenza, caused by an apparatus for gammagraphy: it is likely that the same has happened in this new incident. An investigation was carried out to identify possible originating causes for the contamination.

CONTAMINATION SCENARIOS

Considering the fact that the estimated ^{137}Cs contamination was around 4 GBq, it was possible to consider some (five) different contamination scenarios, i.e., possible ways by which such an amount of radioactive material could have been introduced into the foundry.

A – Lost Radiotherapy Source. Cs-137 is used for teletherapy machines for medical use. In this case, however, the required dose rate intensity (around 5 Gy/h) implies a radioactive source of around 1000 Ci of ^{137}Cs, that is, around 50 TBq. Several orders of magnitude higher than our case. A contamination of around 4 GBq is not compatible with the presence of such a source.

B – Lost Industrial Gammagraphy Source. ^{137}Cs is seldom used for this purpose also, even if its maximum gamma energy (660 keV) is not so high. In this case also, considering the specific gamma ray constant of ^{137}Cs (8.9 mGy/h per GBq at 1 m) it is unlikely that such a source could cause the contamination in exam. 4 GBq would cause a gamma dose rate at 1 m of about 35 mGy/h, insufficient for any industrial gammagraphy purpose. One could think to a portion of that source, however it would be unlikely that such a source would have been sealed and shielded, in order not to cause alarm when passing though the gamma sensor.

C – Lost gamma source for emoderivatives irradiation. Cs-137 sources are industrially used for irradiation of emoderivatives products for medical use. In this case, the source would have an activity compatible with the considered contamination (some GBq). The source would come inside a sealed shielding assembly, with a source capsule surrounded by a shielding materials such as lead.

D – Lost instrumentation calibration source. ^{137}Cs source are also used as calibration source for several instrumentation devices. Those sources are sealed and included into shielding assemblies too. Once the device is not operative anymore, the source may be thrown away with it and become an inadvertent contaminant inside a metallic waste. The considered contamination (4 GBq) is compatible with the total activity of some calibration sources.

E – Contaminated material – Italian II Categoria. One must consider the possibility of having contaminated waste in which ^{137}Cs is present as a contaminant in a bulky amount of material. However, we know that the material has passed a check through a very sensitive radiation sensor before entering the foundry.

This scenario is unlikely, and to demonstrate it, let us consider the upper limit for second category waste (II Categoria) in the Italian regulation, that is, for ^{137}Cs in unconditioned materials, 740 Bq/g (Table 5). If a contamination level compatible with, however inferior to, this limit (say around 400 Bq/g) is assumed, then we should have had at least 10000 kg (10 Metric tons) of contaminated material, in order to have a total radioactivity of 4 GBq. If the specific radioactivity is 40 Bq/g only, then we would have 100 metric tons of contaminated materials, and so on.

This situation is not possible in practice. The material has passed, without causing alarms, through a sensor (a gamma portal) with a sensibility that

excludes the presence of such a radioactivity concentration in the material.

In order to be diluted in such a way in order to avoid any alarm by the gamma sensor, the quantity would have been of several hundreds of tons of contaminated material: this is not possible. By definition, then, it is not possible to have ^{137}Cs in so low concentrations in order to classify the material in the Italian first category waste (I Categoria).

Further case – Contaminated material – US 10CFR61 Class C Waste. Considering finally the maximum concentration limits for LLW according to the US regulation, we may consider the highest possible concentration limit for ^{137}Cs in a material that can be classified as LLW in the US. According to the previously mentioned 10CFR61 regulation, we may consider the limit for Class C waste: it is - for ^{137}Cs - 5 10^4 Ci/m^3 (material still eligible for Shallow Land Burial). This translates into 18.5 10^{14} Bq/m^3, that is, 18.5 10^8 Bq/cm^3. If we have a material with a density similar to that of steel (7.8 g/cm^3) we then have a maximum radioactivity concentration of around 2.5 10^8 Bq/g. If the density is similar to concrete (2 g/cm^3) then we have a maximum radioactivity concentration of around 9 10^8 Bq/g.

The amount of material necessary for having 4 GBq is therefore varying from 3 to 16 grams of material. However, if ^{137}Cs is not exactly equal to the upper Class C waste limit, the amount of material becomes higher as the concentration of ^{137}Cs decreases.

This situation may be a likely scenario, and it may be included in cases C and D (see above) as an upper limit.

However, it is quite unlikely that a sealed and concentrated source of ^{137}Cs could be classified as Class C waste in the US and disposed of in Shallow Land Burial (see definitions in the previous paragraph).

It is much more likely that, given its small volume and its nature, the ^{137}Cs source would be classified and disposed of as High Level Waste.

CONCLUSION

In conclusion, we consider as likely scenarios for the contamination source the ones as C and D in our list:

C – Lost gamma source for emoderivatives irradiation

D – Some lost instrumentation calibration sources

In both cases, ^{137}Cs is in the form of a quite small radioactive source capsule, sealed and surrounded by a shielding material assembly, such as Pb.

The source capsule would have a quite high radioactive concentration, and then a total mass in the order of grams. It has therefore to be classified – for sure – as a High Radioactivity Material, or, once it is inadvertently thrown away, as High Level Waste (HLW), according to the Italian regulation (III Categoria).

The lead assembly shielding explains why the assembly passed through the check of the gamma sensor without creating alarm.

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