PLUTONIUM RETENTION IN THE ALPHA WASTE INCINERATOR AT CEA VALDUC

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ABSTRACT

The Valduc Research Center, which reports to the CEA's Military applications Division, generates solid waste contaminated with alpha emitters in the operation of its installations. An incineration plant has been built to treat this contaminated waste.

The selected process is the IRIS process, developed by the CEA's Nuclear Energy Division in the Marcoule Research Center. It features two-step incineration and produces ash with good reduction factors, and in a quality allowing further treatments. The active waste incineration has been operated for four years.

Criticality risk prevention is based on limiting the mass of active material undergoing treatment in the facility. A balance is compiled continuously by calculating the difference between the mass of active material entering the facility and the mass leaving it. These masses are evaluated by the activity measurements performed on qualified measurement units.

Due to measurement uncertainty, the balance must be zeroed periodically by cleaning and drainage of all the equipment and the absence of holdup in the components must be checked. The holdup measurement of the installation is made chiefly by gamma spectrometry, allowing overall or targeted measurements. Measurements are completed by passive neutron counting. Tests have been conducted, previous to beginning of the active incineration, with calibrated radioactive sources to qualify the systems for measuring holdup from outside the equipment.

This paper presents the salient facts concerning the plutonium retention, the feedback from the operations of cleaning and measurement in active conditions during these four years.

INTRODUCTION

Incineration is a promising weight and volume reduction technique for alpha-contaminated organic waste. An innovative process, IRIS, has been developed at CEA's Marcoule Center. In March 1999, the first highly chlorinated alpha-contaminated waste has been incinerated in the industrial facility at CEA's Valduc Center. So far, it can be said that IRIS process provides good performances for incineration of waste [1, 2].

THE INCINERATION PROCESS

The IRIS Process

The process selected was developed by the CEA at the Marcoule Research Center. It represents a cumulative 5000 h of tests on an inactive pilot facility. It was designed to meet the safety requirements for alpha waste. Incineration is carried out in two stages followed by off-gas postcombustion in small furnaces, enabling their installation in radioactive material containments.

The process also produces ash in a grade suitable for in-line vitrification, or plutonium recovery by argentic dissolution.
The different steps of the IRIS process are shown in the diagram below (fig. 1, see also fig. 4).

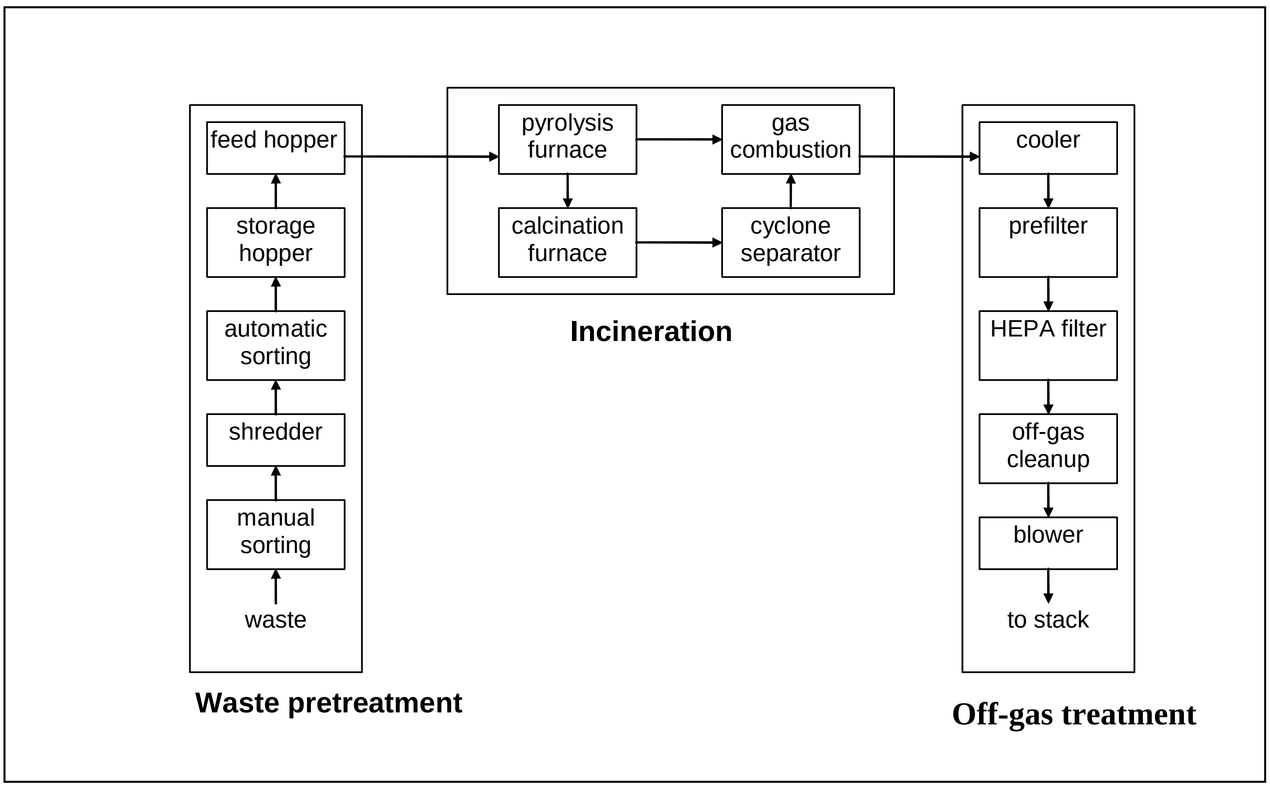


Fig. 1 diagram of the IRIS process

Waste preparation

This part includes the following steps :

- X-ray inspection to detect large metal parts,
- manual extraction of large metal parts if necessary,
- waste shredding,
- detection and automatic extraction of small metal parts,
- buffer storage of shredded waste.

Incineration

This part can be divided in the following steps :

- pyrolysis furnace feed at a maximum regulated rate of 7 kg/h,
- waste pyrolysis at 550 °C in reducing atmosphere, yielding a combustible gas and a solid residue called pitch essentially consisting of carbon,
- pitch calcination at 900 °C in oxygen-enriched atmosphere to obtain ash suitable for possible subsequent treatment,
- combustion of pyrolysis gases at 1100 °C in a post-combustion furnace to which the calcination gases are also sent.

Off-gas treatment

It consists in:

- gas cooling by passage through a pipe heat exchanger, followed by mixing with cold air,
- gas prefiltration in an electrofilter featuring 99.8% filtration efficiency,
- HEPA filtration to meet the site environmental release specifications,
- chemical purification of inactive gases by dry neutralization, so as to release no chlorine residue.

ACTIVE INCINERATION

After inactive waste incineration campaigns run in 1996, 1997 and 1998, the active incineration has begun since March 1999.

The waste to be incinerated are essentially PVC, latex, neoprene and cellulose.

The average activity of the waste is 7.5 10^8 Bq/kg (0.02 Ci/kg). The waste is only contaminated by plutonium without any fission products.

Waste

The table I gives the results of incineration of active waste for each year.

Table I : results of active incineration at CEA Valduc

Year	Weight of waste (kg)	Ashes (kg)	Dust (kg)	Reduction factor
1999	982	42.8	5.4	20.4
2000	1345	73.3	15.3	15.2
2001	1423	54.8	9.9	22
2002	685	33.1	17.8	13.5
2003 (first run)	369	15.4	9	15.1
total	4804	220	57.4	17.3

CRITICALITY SAFETY

Criticality risk prevention is based on limiting the mass of active material undergoing treatment in the facility. A balance is compiled continuously by calculating the difference between the mass of active material entering the facility and the mass leaving it. These masses are evaluated by the activity measurements performed on qualified measurement units.

We must always verify :

$$B = \Sigma_{\text{mass Pu in}} + 20\%^* \Sigma_{\text{mass Pu in}} - \Sigma_{\text{mass Pu out}} + 12\%^* \Sigma_{\text{mass Pu out}} \leq 350 \text{ g}$$

The balance must be zeroed periodically by cleaning and drainage of all the equipment. At the present time, there has been 2 'criticality runs', as shown in the table II :

Table II : Plutonium balance

Year	Weight of plutonium in waste (g)	Weight of plutonium in ashes (g)	Weight of plutonium in dust (g)	Plutonium losses (g)	B
1999	271.3	228.6	17.1	25.6	109.36
2000	525.3	514.3	8.4	2.6	276.67
Total 1st criticality run	796.6	742.9	25.5	28.2	276.67
2001	450.7	433.2	4	13.5	156.1
2002	370.3	328.8	9	32.5 *	303.2
2003 (first run)	91.1	107	23.1	-39 *	284.0
Total 2nd criticality run	912.1	869	36.1	7	284.0
Total	1708.7	1611.9	61.6	35.2	0

* : the explanation for these values is that the incinerator was not empty at the end of 2002.

The values of plutonium in dust depend on the cleaning. The losses are rather weak, and decrease with time.

PLUTONIUM RETENTION

The balance is zeroed periodically by cleaning and drainage of all the equipment. But, due to measurement uncertainty, the absence of holdup in the components must be checked.

The operator has conducted tests with calibrated radioactive sources to qualify the systems for measuring holdup from outside the equipment. Measurement are taken chiefly by gamma spectrometry, allowing overall or targeted measurement with the collimator, and, for the post-combustion furnace, by passive neutron counting. The instruments used are mobile, since no continuous holdup measurement is carried out. If necessary, gamma detectors will be introduced into the components to locate any holdup.


Fig. 2 Mobile gamma detector (HPGe)


Fig. 3 Mobile passive neutron counter

After the first criticality run, 26 points were measured using gamma spectrometry and neutron passive counter. The mass of plutonium measured by gamma spectrometry was 25 g ± 32%, the mass of plutonium measured by neutron passive counter was 12 g ± 70 %. These values, given in table III and IV, are coherent with the results of the first criticality run (as shown in table II).

Because of the initial calibration, only the measured points with a mass of plutonium greater than 1 g are looked forward.

Table III retention values measured by gamma spectrometry

	December 1999	November 2000	November 2001	April 2003
Pyrolysis furnace	2.2 g	3.0 g	2.8 g	3,4 g
Calcination furnace	1.2 g	1.4 g	2.7 g	3 g
Cyclone entry	1.4 g	2.0 g	3.3 g	2,2 g
Cyclone	2.5 g	8.2 g	3.8g	1,5 g
Cyclone exit	1.1 g	2.5 g	2.2 g	1,9 g

Table IV retention value measured by passive neutron counting

	March 1999	November 2000	November 2001	April 2003
post combustion furnace	3 g ± 60%	12.6 g ± 70%	17.2 ± 60 %	14.7 ± 60 %

Compared with the complexity of the process, the retention is rather low. It increases with treated waste, but after the initial sooty layer, it increases quite slowly.

Retention places are presented in figure 4. The furnaces are retention places, especially post-combustion furnaces. The layer of plutonium is very hard, and cannot be cleaned easily. The decrease of plutonium between 2001 and 2003 is connected with the change of some heating pieces.

The difficulties of cleaning pipes and bends of the cyclone separator explain the retention at these places.

In term of criticality, it must be taken into account that the different retention places are distant enough from one another.

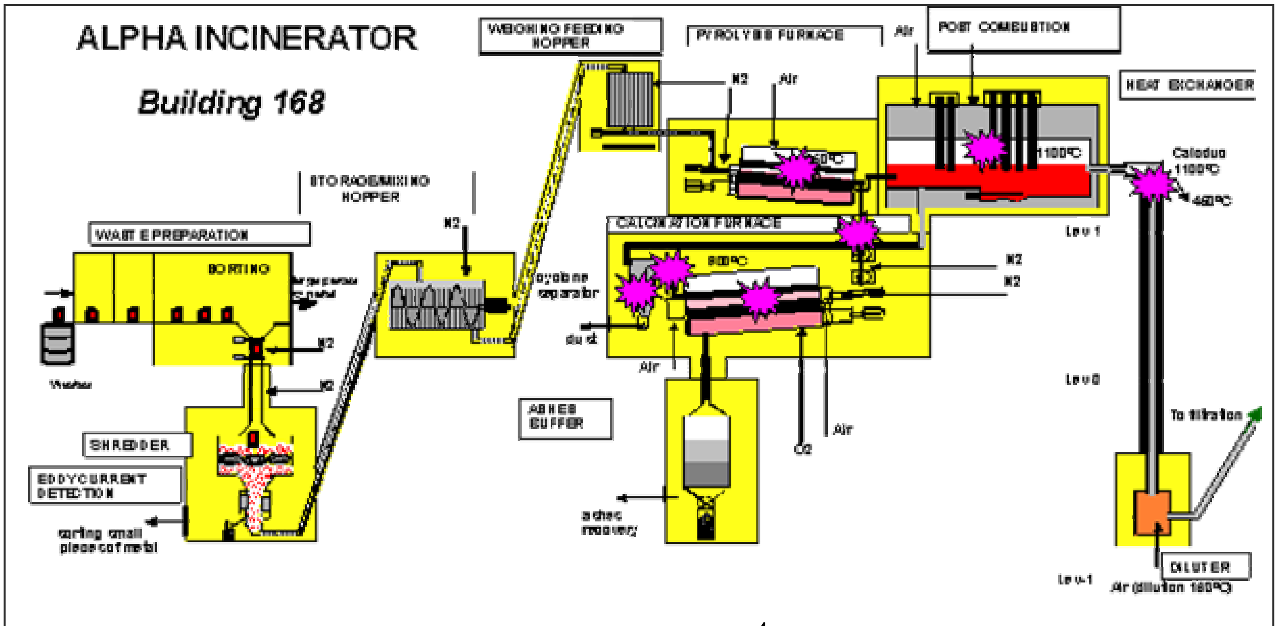
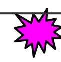


Fig. 4 Incinerator components and retention points : 

CONCLUSION

The alpha incinerator of CEA Valduc has been operated in active condition since 1999. The IRIS process provides performances in volume and weight reduction which are very interesting.

The balance between the mass of active material entering the facility and the mass leaving it is in very good accordance with the retention measurements, periodically checked. Furthermore, the plutonium retention is low, and well located, essentially in the furnaces.

Thus the alpha incinerator of CEA Valduc is a well adapted tool for the incineration of contaminated waste.

REFERENCES

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