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INCINERATION PROCESS FOR PLUTONIUM-CONTAMINATED WASTE

J.J. Vincent, T. Longuet, R. Cartier and L. Chaudon

Commissariat à L'Energie Atomique (CEA)
Rhône Valley Nuclear Research Center
BP 171, 30205 Bagnols-Sur-Cèze, France

ABSTRACT

A reprocessing plant with an annual throughput of 1600 metric tons of fuel generates 50 m^3 of incinerable α-contaminated waste. The reference treatment currently adopted for these wastes is to embed them in cement grout, with a resulting conditioned waste volume of 260 m^3. The expense of mandatory geological disposal of such volumes justifies examination of less costly alternative solutions.

After several years of laboratory and inactive pilot-scale research and development, the *Commissariat à l'Energie Atomique* has developed a two-step incineration process that is particularly suitable for α-contaminated chlorinated plastic waste. A 4 $kg \cdot h^{-1}$ pilot unit installed at the Marcoule Nuclear Center has now logged over 3500 hours in operation, during which the operating parameters have been optimized and process performance characteristics have been determined. Laboratory research during the same period has also determined the volatility of transuranic nuclides (U, Am and Pu) under simulated incineration conditions. A 100 $g \cdot h^{-1}$ laboratory prototype has been set up to obtain data for designing the industrial pilot facility.

INTRODUCTION

The reference treatment for incinerable α-contaminated wastes has been to embed them in cement grout. The high cost of mandatory geological disposal for these wastes justifies examination of less costly alternative solutions.

After several years of laboratory and inactive pilot-scale research and development, the *Commissariat à l'Energie Atomique* has developed an incineration process that is particularly suitable for α-contaminated chlorinated plastic waste. A two-step process was considered preferable to conventional direct incineration in view of the specific constraints of α-contaminated chlorinated waste. The CEA process minimizes fly ash entrainment while producing ashes of a quality compatible with plutonium recovery techniques. A dry off-gas treatment process was developed that complies with chemical and nuclear waste discharge standards (no α-contaminated liquid waste) and with existing safety regulations regarding containment, criticality, explosion and fire hazards. The process is compatible with wide variations in the waste composition and feed rate, and specifically limits corrosion problems.

ECONOMIC ADVANTAGES OF INCINERATING α-BEARING TECHNOLOGICAL WASTES

Technological waste contaminated with α-emitting radionuclides account for an annual volume of some 120 m^3 produced by a reprocessing plant with a capacity of 1600 metric tons per year. The incinerable fraction of this waste is estimated at 50 m^3, representing about 12.5 tons (assuming an apparent density of 0.25 $t \cdot m^{-3}$). These wastes are currently conditioned either in 20 or 50-liter canisters or in 400 or 800-liter drums, which are placed in concrete containers and immobilized with cement grout. The containers are then transferred to interim storage facilities pending geological disposal.

The overall management cost (conditioning, transportation and storage) of the reference solution was evaluated and compared with an alternative solution implementing the incineration process developed by the CEA. Two ash conditioning methods were considered: cement embedding and melting (the residual α activity does not warrant a recovery treatment). The economic assessment was based on available economic data.

Comparative Volume

Process	Conditioned Waste Volume (m^3)	Waste Disposal Volume (m^3)
Direct cement embedding	260	340
Incineration +		
- ash cementation	5	7
- ash melting	3.8	5

Comparative Waste Management Cost

The estimates (in arbitrary units) are based on a 20-year depreciation period.

Direct cementation

• Annual depreciation and operating costs:	3.8
• Road or rail transportation to the storage site:	1.0
• Deep geological disposal cost in drifts assigned to Type B waste, based on a cost estimate prepared by the Commission of the European Communities [1]:	95.2
Total:	**100.0**

Incineration

α-contaminated technological wastes incinerated in a unit operated 16 hours a day in two shifts, with a throughput rate of 8 $kg \cdot h^{-1}$, and a weight reduction factor of 30. Secondary waste generated by the process is estimated at 1 wt% of the incinerated material, and is cemented in concrete containers. The ashes are either embedded in cement or melted [2,3].

• Annual depreciation and operating costs:	
- Ash cementation	28.5
- Ash melting	29.6

- Rail or road transportation (ashes in Type B casks, cemented secondary waste on standard rail or road platform):
  - Cemented ashes + secondary waste 1.9
  - Melted ashes + secondary waste 0.5
- Geological disposal (same conditions as above):
  - Cemented ashes + secondary waste 2.0
  - Melted ashes + secondary waste 1.4

Total:
- Incineration + ash cementation 32.4
- Incineration + ash melting 31.5

The following summary table highlights the significant economic advantage of incineration in the overall waste management cost. By significantly reducing the waste volume, incineration allows appreciable savings in transportation and disposal costs, which rapidly offset the higher investment cost for the incineration facility. The choice of ash conditioning options depends more on considerations of containment quality than on economic criteria.

Cost Itemization (arbitrary units)	Direct Cement Embedding	Incineration	
		+ Ash Cementation	+ Ash Melting
Depreciation and operating costs	3.8	28.5	29.6
Transportation costs	1.0	1.9	0.5
Disposal costs	95.2	2.0	1.4
Total	100.0	32.4	31.5

DESCRIPTION

Waste Material

The average waste composition used to define and test the process is based on the estimated composition of a MOx fuel fabrication plant:

PVC	50 wt%
Neoprene	17.5
Latex	17.5
Cellulose	10
Polyethylene	5.

The mean activity is estimated at 7.5×10^9 Bq·kg^{-1} (0.2 Ci·kg^{-1}).

The Incineration Process

The two-step incineration process was tested and developed in an inactive 4 kg·h^{-1} pilot facility at the Marcoule Nuclear Center. The following flowsheet provides an overview of the process, and shows the breakdown into three separate operational sequences: waste pretreatment, incineration, and off-gas treatment.

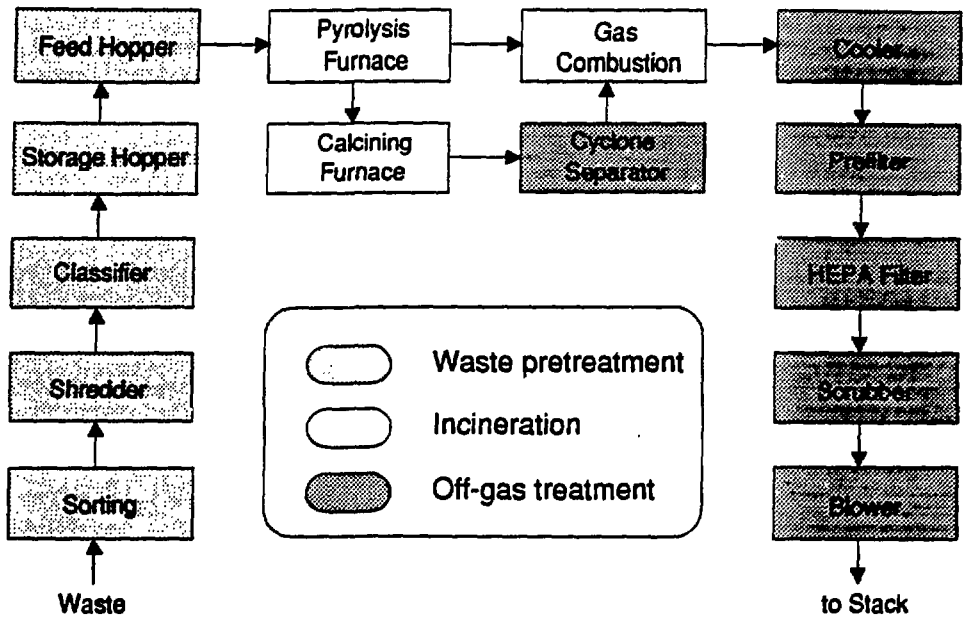


Figure 1. Process Flowsheet

The *waste pretreatment* operations include X-ray detection and manual removal of any large metal objects liable to damage the shredder, then waste shredding followed by separation of the incinerable waste from denser metallic items (nuts, bolts, Pu fragments, etc.). The shredded waste is placed in a buffer storage hopper prior to metered feeding.

The actual *incineration* process comprises two steps:

- Pyrolysis of the shredded waste in an electrically heated rotating kiln under reducing atmosphere. This operation produces a combustible gas and a solid "pitch" residue containing 85% carbon.
- Calcining of the pitch in an electrically heated rotating kiln under oxygen-enriched atmosphere to produce ashes of sufficient quality to allow Pu recovery from highly contaminated waste.

Total combustion of the pyrolysis and calcining off-gases is ensured in an electrically heated furnace with largely excess air concentration.

The *off-gas stream* is subjected to physical and chemical purification:

The calcining off-gases are partially decontaminated in a cyclone separator at the calciner outlet. The afterburner gases are cooled by dilution in air in a double-jacket heat exchanger, then filtered successively across bag filters and HEPA filters to trap any active or inactive particles in suspension.

- The decontaminated gases are scrubbed and the residual hydrochloric acid is neutralized.

PERFORMANCE

The 4 $kg \cdot h^{-1}$ pilot unit at Marcoule has now logged 3500 hours in operation, during which 14 metric tons of waste have been incinerated. Three additional test runs, each lasting at least 500 continuous hours without intervention, have also been completed.

Incineration

The overall waste-to-ash weight reduction for typical wastes is 38. This factor depends to a great extent on the inorganic content of the waste. The overall ash entrainment in the off-gas stream is less than 1%. The carbon content of the final ashes is 1 to 2%. The combustion efficiency at the afterburner outlet is 99.98%, with a CO concentration of 7 ppm, a CO_2 concentration of 4.5% and an O_2 concentration of 18%.

Off-Gas Treatment

The preliminary bag filter retention is 99.8%, with a purification factor (i.e. the ratio of particles in suspension in the incoming and filtered off-gas stream) of 500. The purification factor for the HEPA filter stage is 4×10^5, and the overall filtration purification factor is 2×10^8.

Following process improvements, for example the use of a cyclone separator to further limit fly ash entrainment, the Pu entrainment factors were reassessed. Plutonium was simulated by cerium oxide, since this element is not present in the incinerable waste, and most closely simulates plutonium oxide under the specified thermodynamic conditions. Plutonium distribution in the facility is shown in the Figure 2.

Some inorganic oxides in the waste materials become chlorinated during the pyrolysis step, notably ZnO, which forms $ZnCl_2$. Most of the resulting metallic chlorides are volatile, and are

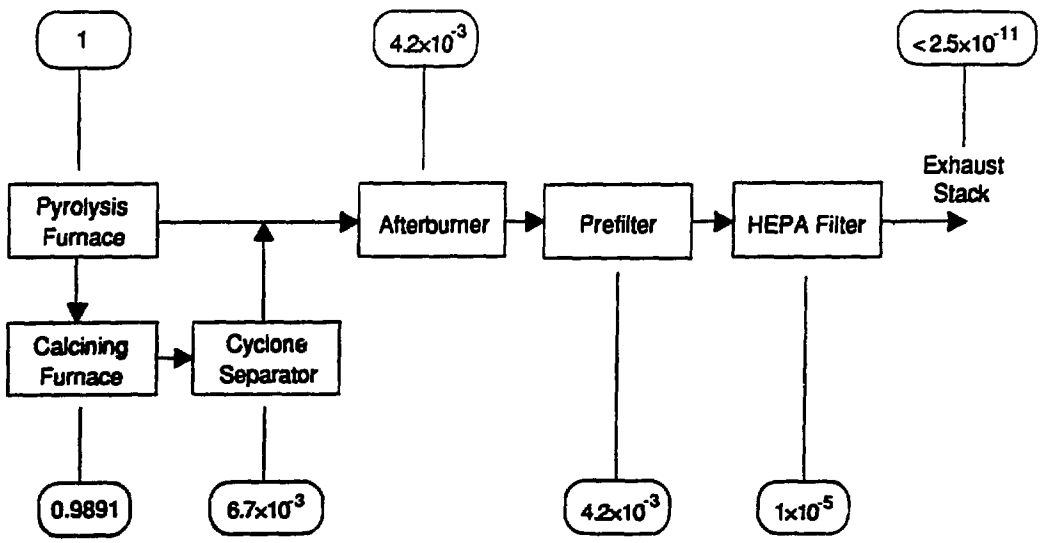


Figure 2. Plutonium Trapping in the Incineration Facility

entrained in the off-gas stream in appreciable quantities. The particle matter is extremely fine (80% of the particles are less than 0.35 μm in diameter) with significant humidity and a tendency to clog the filters. These two constraints were taken into account in designing the off-gas system: the bag filter efficiency is 99.8%, and the clogging problems typical of this type of particles have been solved.

Flexibility

Sensitivity tests were conducted on the inactive 4 $kg \cdot h^{-1}$ prototype to test the process flexibility regarding the waste feed composition and throughput rate.

Various waste compositions were tested, in which the lower heat capacity, the chlorine content and the inorganic matter content were individually varied in turn by ± 40%, and in which ethylene vinyl acetate (EVA) or polyurethane was substituted for PVC. The results show that any change in the waste composition affects the afterburning temperature, but that these variations may easily be corrected by regulating the process air flow rate. In addition, without any prior modification, the pilot unit was successfully operated at 7 $kg \cdot h^{-1}$.

The test results demonstrated considerable process flexibility regarding the nature of the waste stream and the throughput rate. On the basis of the data acquired from these tests, it is now possible to design and build a unit capable of handling 20 kg of waste per hour. The same process could be extrapolated to 100 $kg \cdot h^{-1}$ if necessary.

LABORATORY RESEARCH

Transuranic Nuclide Volatilization during Simulated Incineration

Actinide volatility was determined for the pyrolysis, calcining and afterburning steps. Wastes containing plutonium/americium oxide and uranium oxide were incinerated in a glove box under conditions similar to an indusrial pilot facility.

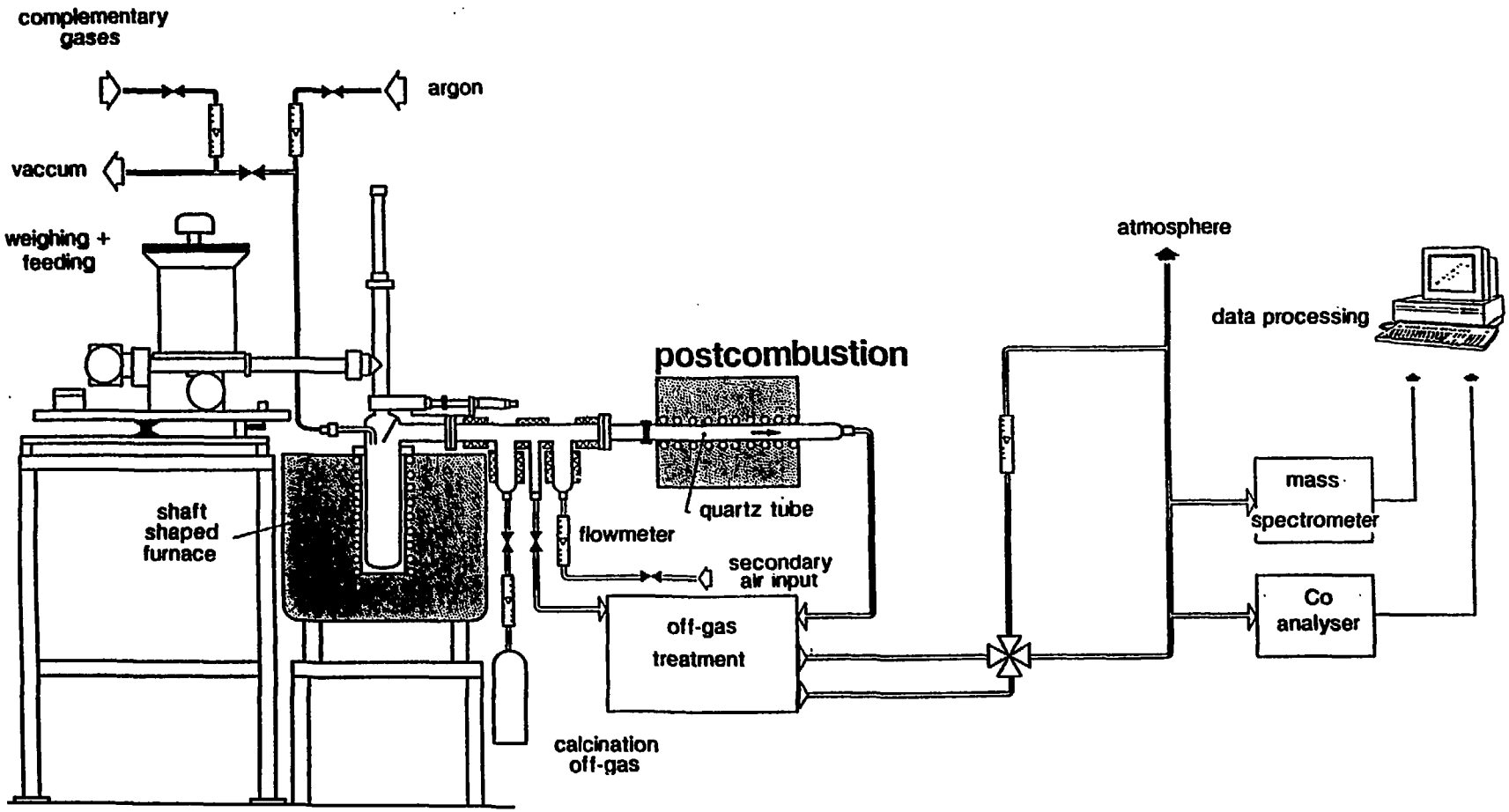
The quantities of volatilized transuranic nuclides were extremely low compared with the quantities in the feed stream: < 0.1% for uranium, < 0.01% for americium and < 0.001% for plutonium. Volatilization of transuranic nuclides thus accounts for only a minute fraction of the activity entrained in the off-gas stream: the entrainment is attributable primarily to mechanical phenomena (turbulent gas flow).

Laboratory-Scale Incineration Facility

A laboratory device with a nominal throughput rate of 100 $g \cdot h^{-1}$ was developed to acquire process data and prepare for the design of an industrial pilot. The unit (Figure 3) comprises a continuous waste feed hopper, a pyrolysis furnace and an afterburner. Pitch calcining off-gases may be simulated and injected at the afterburner inlet. The unit is coupled with an off-gas analysis system comprising a 4-pole mass spectrometer and an infrared detector for quantitative online analysis of H_2, N_2, O_2, CO, CO_2, CH_4, C_2H_6, C_3H_6 and Ar.

The facility has been used to date to analyze the pyrolysis off-gases and optimize the pyrolysis air flow rate to prevent soot from forming. Preliminary sensitivity studies have also been conducted to assess the effects of the waste composition.

FIG. 3 : LABORATORY APPARATUS



CONCLUSION

A reliable, flexible and safe process has been developed from a combination of laboratory studies and technological testing. All process equipment items are compact for easier nuclearization.

The resulting volume reduction allows substantial reductions in waste transportation and disposal costs that largely offset the investment cost necessary for the waste treatment facility.

The *Commissariat à l'Energie Atomique* is currently investigating a melting process for ash conditioning. This process ensures higher quality containment than direct cementation for practically the same cost.

The process has been adopted by a CEA defense center, and has contracted with USSI INGENIERIE to build the first industrial facility based on a throughput capacity of 7 $kg \cdot h^{-1}$.

REFERENCES

1. *Evaluation of Storage and Disposal Costs for Conditioned Radioactive Waste in Several European Countries*, Report EUR 12871 EN, Commission of the European Communities: Nuclear Science and Technology.

2. A. Jouan, N. Jacquet-Francillon, M. Cler and L. Chaudon, *Economic and Technical Advantages of High-Temperature Processes in High-Level Radioactive Waste Management*, 2nd International Conference on High-Level Radioactive Waste Management, Las Vegas, April 28-May 3, 1991.

3. A. Jouan, H. Frotscher, B. Ganser, N. Jacquet-Francillon, C. Kertesz and M. Klein, *New Volume-Reduction Conditioning Options for Solid Alpha-Bearing Waste*, 3rd European Community Conference on Radioactive Waste Management and Disposal: Luxembourg, Sept 17-21, 1990.