

動燃プルトニウム燃料部におけるプルトニウム 廃棄物の管理

Management of Plutonium-contaminated wastes at the
Plutonium fuel facilities in P N C

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Management of plutonium-contaminated wastes
at the plutonium fuel facilities in PNC

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ABSTRACT

A summary is presented concerning the experience in the management of Pu contaminated wastes at the plutonium fuel facilities of the Power Reactor and Nuclear Fuel Development Corporation (PNC). Description are given of waste generation, current handling techniques, Pu measurement techniques and process techniques now under research and development such as incineration system and a closed cycle liquid waste treatment by using decomposition of ammonium nitrate contained in the liquid.

Introduction

During the last few years, about 9.6 metric tons of mixed oxide fuels and 1.6 metric tons were fabricated for deuterium critical assembly (DCA) and JOYO reactor, respectively. The amount of plutonium-contaminated wastes is markedly increased along with these fabrication campaigns.

The purpose of this paper is to describe the current situation concerning the management of plutonium-contaminated wastes and further development expected in future at the plutonium fuel facilities of the Power Reactor and Nuclear Fuel Development Corporation.

1. Generation of plutonium-contaminated wastes and current handling techniques

Excluding the inside of glove boxes and hoods, in which plutonium and uranium are handled, operation areas have been maintained essentially at the zero contaminated level, since the beginning of the facilities.

Therefore most of the solid wastes generated in the controlled areas have been disposed as the non-radioactive waste and only those generated in contaminated areas have been stored in the storage as the radioactive wastes. In principle, these wastes are classified by the origin into the special containers which are classified by color separately as shown in Table I and treated as shown in Figure 1.

Liquid wastes originated in the controlled areas have been treated as low level wastes, separated from non-process wastes and laundry wastes, which are sent to separate vessels and monitored. These wastes, after being decontaminated, have been

discharged below the rate of 200 m³/month as shown in Table II.

Exhaust gas from glove boxes, hoods, incinerator and controlled areas etc. is discharged to the atmosphere, after treatment with High Efficiency Particulate Air (HEPA) filters and monitoring.

1.1 Solid wastes

During the last 10 years the radioactive solid wastes at the facilities have accumulated to be about 720 m³ in net volume of the containers. The amounts of the wastes generated each year is shown in Figure 2. These wastes are stored in 8 storages in which the net volume is totalized to be 2.000 m³.

The total amount wastes contained in drums and spent HEPA filters occupy 65 % and 26 %, respectively.

Glove boxes are scrapped in the following steps:

Bag out of the installed equipment.

Decontamination and painting of the glove box.

Loading of the box sealed in PVC-sheet, into a steel container.

The waste containers for the scrapped glove boxes have been accumulated to be about 37 m³ until March 1975.

Table III shows the amounts of radioactive solid wastes which were generated in the controlled rooms directly concerned with fabrication and inspection of the fuels for JOYO and DCA. Typical drum wastes distribution in 1974 is as follows; the ratio of paper to the total drum wastes was 29.8 %, that of rubber and plastics was 58.8 % and that of metal and glass was 9.1 %.

Solid wastes generated in the corridors and process control rooms etc., where fissile materials are not handled, are disposed after the check for contamination. On the other hand solid wastes

generated in process rooms where fissile materials are handled, are classified into two groups; the combustibles mainly composed of paper and the others. The former, after being contained into 20 litres yellow cartons and stored temporary in the facilities, is hand-sorted and incinerated. During the last 10 years the amounts of incinerated wastes have accumulated to be 108 m^3 . The other wastes, which are mainly rubber, plastics and glass etc., are put into 200 litres drums to be laid at the special out-door storage place. About 800 drums have been generated until March 1975 for the beginning.

1.2 Liquid wastes

Liquid process wastes originated in chemical analyses and wet recovery processes at Plutonium Fuel Development Facility (PFDF) and Plutonium Fuel Fabrication Facility (PFFF) have been decontaminated by following equipments. The PFDF has the liquid waste decontamination equipment consisting of an evaporator and an active carbon bed with the capacity of 8 l/h. The decontamination factor (D.F.) of nearly 10^4 is obtained. On the other hand, the PFFF has the equipment consisting of three flocculators and an active carbon bed etc. with the capacity of 30 l/h. The D.F. of $10^4 \sim 10^5$ is obtained. Until March 1975, 27.8 m^3 of process wastes were decontaminated at the PFDF and PFFF. The waste treated in the above together with the liquid waste originated from chemical hoods have been treated as the low level liquid waste.

The amount of liquid wastes generated at the PFDF and PFFF is summarized in Figure 3.

1.3 Gaseous waste

The amount of exhaust gas was about $1.1 \times 10^9 \text{ m}^3/\text{year}$ before 1971 and then increased to $3.5 \times 10^9 \text{ m}^3/\text{year}$ with the start-up of the PFFF and other facilities. The amounts of alpha emitters contained in these gaseous waste is always below the detection limit ($1 \times 10^{-16} \mu\text{Ci}/\text{cm}^3$).

2. Improvements in alpha waste treatment

2.1 Solid wastes

Major object in near future is stabilization of the plutonium-contaminated solid wastes after volume reduction in a retrievable condition. This may be attained, for example, by incineration of combustible waste, melt and re-solidification of plastics waste, pressing of the bulk waste, and finally, melt and re-solidification of glass and metal wastes.

2.1.1 Incineration of the plutonium-contaminated wastes

An incinerator with a capacity of $\sim 20 \text{ kg/h}$ was constructed in 1974 in the PFDF in order to reduce the volume of stored wastes by burning the plutonium-contaminated wastes.

The combustion chamber and post-combustion chamber consist of refractory brick lined on casing of stainless steel shell body. Ignition was accomplished with a heavy oil fueled jet. The combustion chamber and post-combustion chamber are maintained by heavy oil at $200 \sim 800^\circ\text{C}$ and $800 \sim 1.000^\circ\text{C}$, respectively. An off-gas treatment system consists of the post-combustion chamber with a silicon carbide brick bed, a high temperature filter (HTF) with 222 candle filters pre-coated with asbestos, a dilutor for cooling the gas with air

below 150°C, four high temperature HEPA filters, a heat exchanger for cooling the gas below the temperature of 60°C, and a wet scrubber for removing toxic elements such as hydrogen chloride and sulphur dioxide.

As shown in Figure 4, the combustible wastes are hand-sorted to be free from non-combustibles, put into 20 litres cartons, and hand charged to the combustion chamber. For the purpose of preventing the contamination spreading in the operating room, this incinerator has four glove boxes for feed preparation, ash-removal, pulling out the HTF elements and exchanging the high temperature HEPA filters. Further caution is paid to minimize the possibility of leakage of combustion products to the operating room caused by pressure fluctuation.

After the completion of this incinerator, pre-operational tests using artificially made uranium wastes shown in Table IV have been performed for testing and improving the equipment. Results of these tests are summarised in Table V.

2.1.2 Measurement of plutonium in the solid waste containers

(A) drum scanning

The equipment itself is the conventional type. It consists of a platform on which a 200 litres drum is placed and a single channel gamma scanning system. The platform is rotated 14 times uniformly during the counting period of 45 minutes. An NaI(Tl) detector is moved vertically along one of the generatrix of the barrel during this period. A 2 mm thick lead shield covers the detector, eliminating some of the soft gamma rays of americium. The minimum detectable

quantity of plutonium by the waste-drum scanner is 20 mg.

About 1,700 drums have been measured since 1973.

Percentage of the drums containing plutonium less than 1 g was 83 % and that more than 10 g was 0.5 %.

(B) carton scanning

It is better to classify the cartons at the origin by the plutonium content before loading them in a drum. An equipment using the active neutron technique has been installed and now under testing.

2.1.3 Treatment of spent HEPA filters

As the first step for developing the method to treat spent HEPA filters, an examination to melt and re-solidify the glass fibre filter media is started using a small electric crucible furnace.

2.2 Liquid waste

Development of a new liquid waste treatment system aiming a closed cycle system was started in 1975. In this system, solid waste would be left minimized and water be treated for re-use.

A feasibility study for the closed cycle system was performed before September 1975. As shown in Figure 5, model flowsheet and the processes to be developed were proposed.

The liquid process waste at the plutonium fuel facilities contains a large quantity of salt (~20 wt% of ammonium nitrate) which should be removed for full decontamination of the liquid waste.

From the results of the feasibility study, a thermal decomposition process is considered to be the most promising process to be developed. In this process, the liquid waste containing ammonium nitrate is decomposed to innocuous gas and the minimum quantity of solid residue.

3. Summary

- 1) During the last 10 years total of 720 m³ of solid waste was generated, sealed, and stored within facilities.
- 2) About 2,000 m³ of liquid waste, most of which in non-process waste, is yearly discharged after decontamination.
- 3) An incinerator for the combustible wastes including plastics and rubbers is now under pre-operational tests.
- 4) Solid plutonium wastes in drum is measured by gamma-scanning and another method using an active neutron technique is now under testing.
- 5) A new liquid waste treatment program for the closed cycle system was started in 1975.

Table I Containers for solid wastes

Standard containers		Wastes
20 litres carton	(red)	radioactive combustibles (paper, rags etc.)
	(green)	radioactive combustibles (plastics, rubber)
	(white)	radioactive non-combustibles (glass, metal etc.)
	(yellow)*	non-radioactive combustibles (paper, rags etc.)
200 litres drum	(yellow)	radioactive
	(blue)*	non-radioactive (rubber, glass, metal etc.)
paper bag	(light brown)**	non-radioactive combustibles (paper, rags)

* solid wastes generated in process rooms

** solid wastes generated in the corridors

Steel containers and PVC-sheet are also used, but not yet standardized.

Table II Present liquid waste treatment process

Waste sources	Categories	Total alpha activity ($\mu\text{Ci}/\text{cm}^3$)
Chemical analyses		
Wet recovery —————> decontamination		0.1 ~ 1.0 ↓ < 1.5×10^{-5}
Hoods Decontamination rooms —————> LOW LEVEL		< 1.5×10^{-5} — treatment —> discharge
Floor drain	NON-PROCESS	under detection limit ($< 3 \times 10^{-8}$)
Laundry room	LAUNDRY	under detection limit ($< 3 \times 10^{-8}$)

Spent oils and solvents are stored.

Table III Amount of plutonium-contaminated solid wastes generated along with the fabrication of "JOYO" and "DCA" fuels

JOYO		
period	amounts of mixed oxide fuels treated (1,000kg)	amounts of solid wastes generated (in 200l drums) (m ³)
Nov. 1972 ~ Mar. 1973	0.04	1.2
Apr. 1973 ~ Mar. 1974	0.86	21.4
Apr. 1974 ~ Mar. 1975	0.70	10.0
Total	1.60	32.6

DCA		
period	amounts of mixed oxide fuels treated (1,000kg)	amounts of solid wastes generated (in 200l drums) (m ³)
Apr. 1972 ~ Mar. 1973	5.1	21.5
Apr. 1973 ~ Dec. 1973	4.5	15.2
Total	9.6	36.7

Table IV Solid wastes for pre-operational test of the incinerator

material	wt %
polyethylene	48
rags	23
paper	23
rubber	4
PVC	2
natural uranium	(0.1)

Table V Results of pre-operational test of the incinerator

items	results
capacity	10 ~ 15 kg/h
U distribution	ash ≈ 100 %, HTF elements = 0.1 %, gas after HTF < 0.001 %
volume reduction	1/85
weight reduction	1/20 ~ 1/40

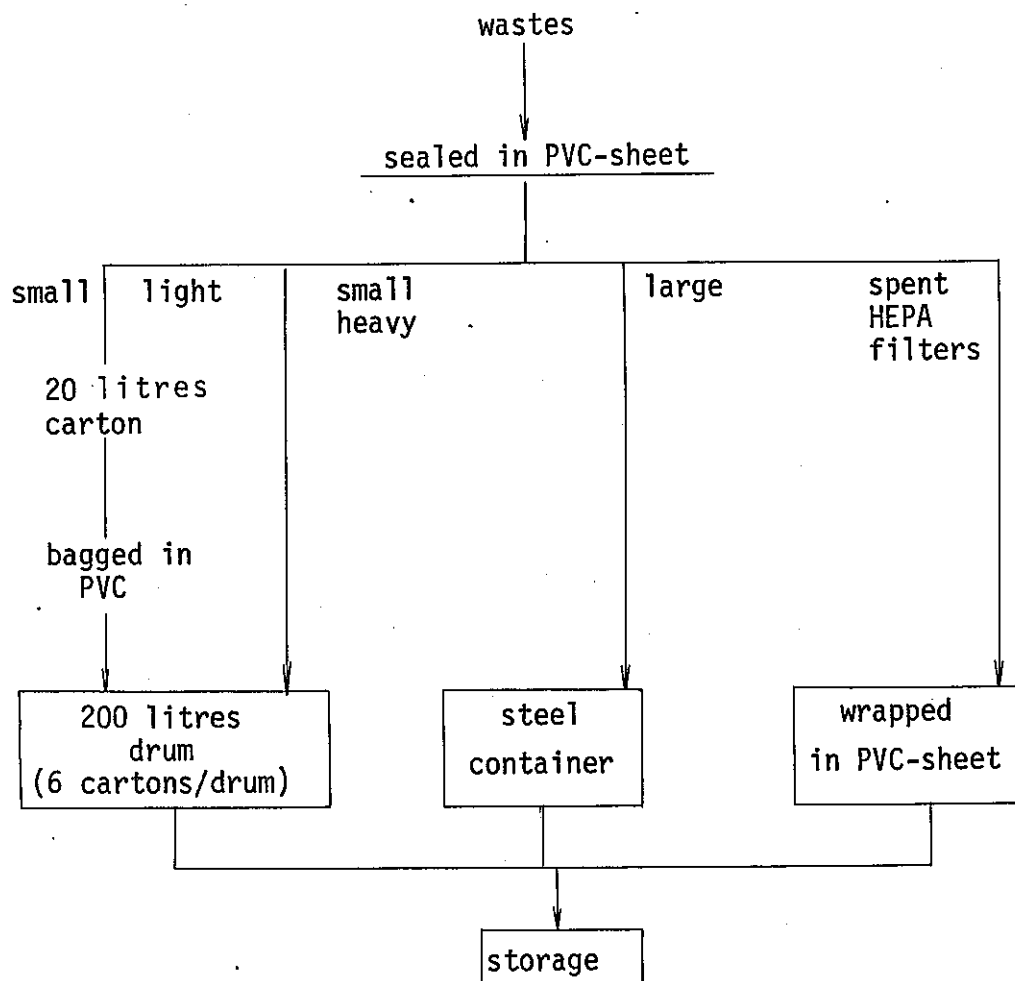


Fig. 1 Treatment of plutonium-contaminated solid wastes at the plutonium fuel facilities of PNC

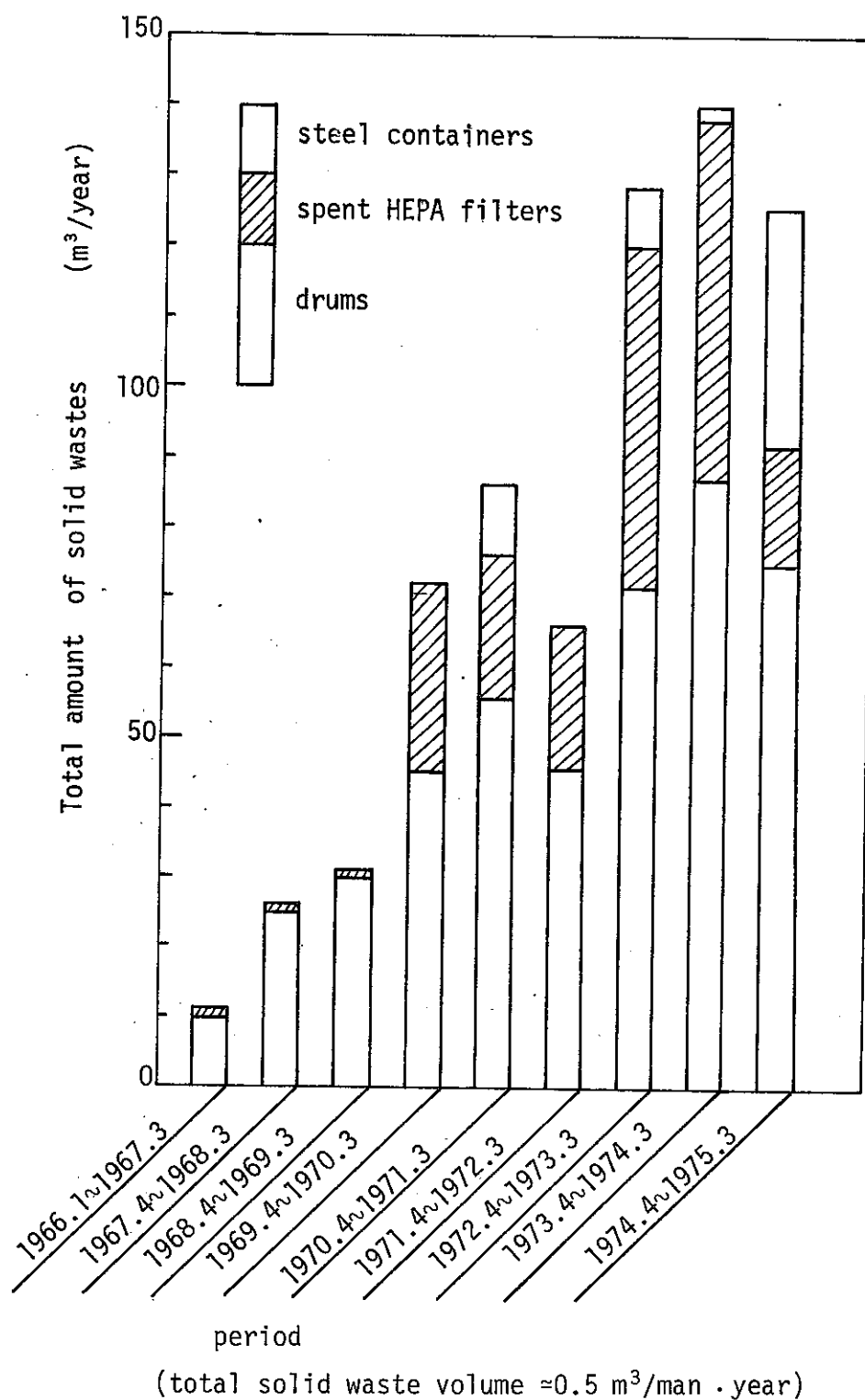


Fig. 2 Total amount of plutonium-contaminated solid wastes generated annually at the PFDF and PFFF

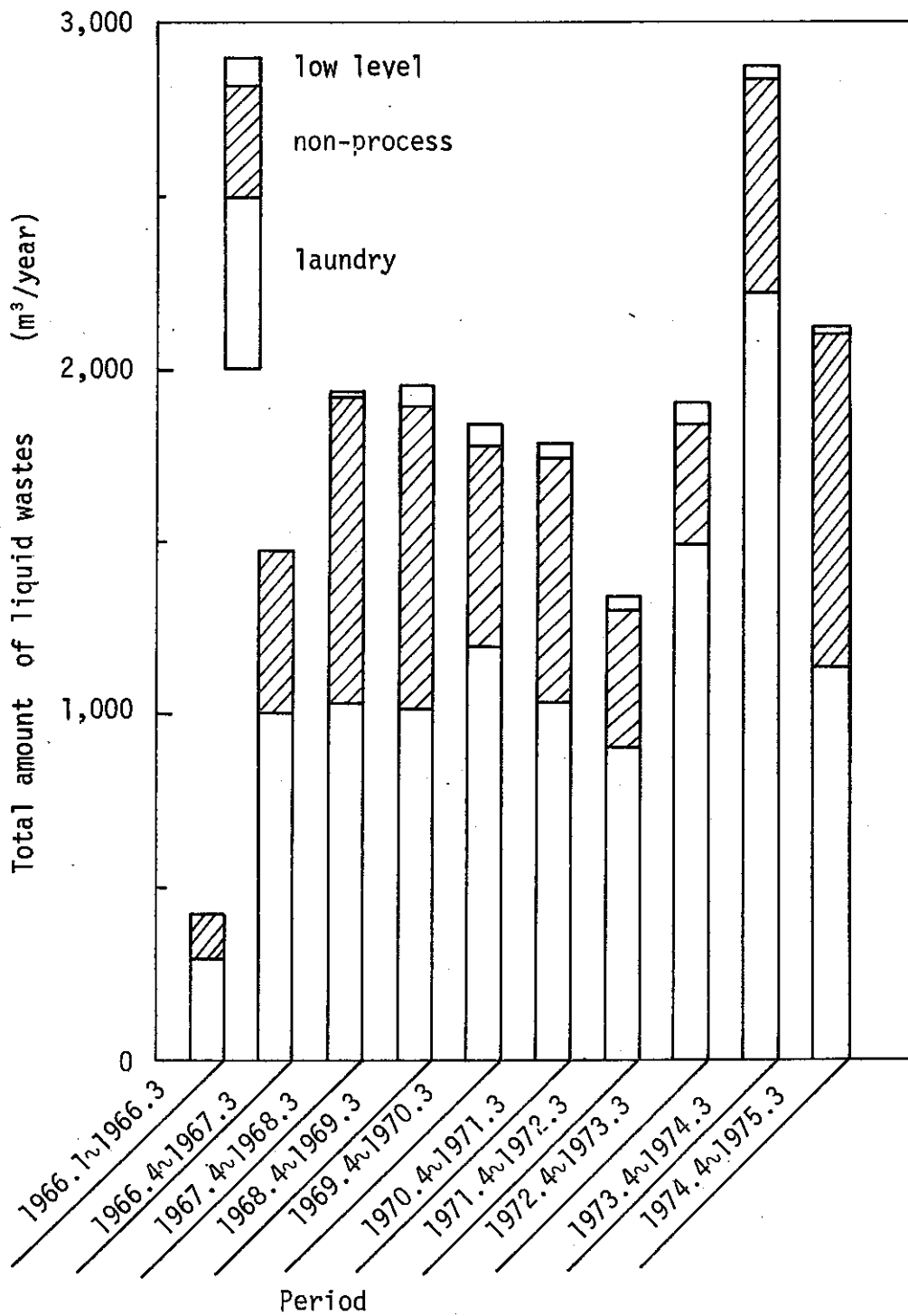


Fig. 3 Total amount of liquid wastes generated annually at the PFD and PFF

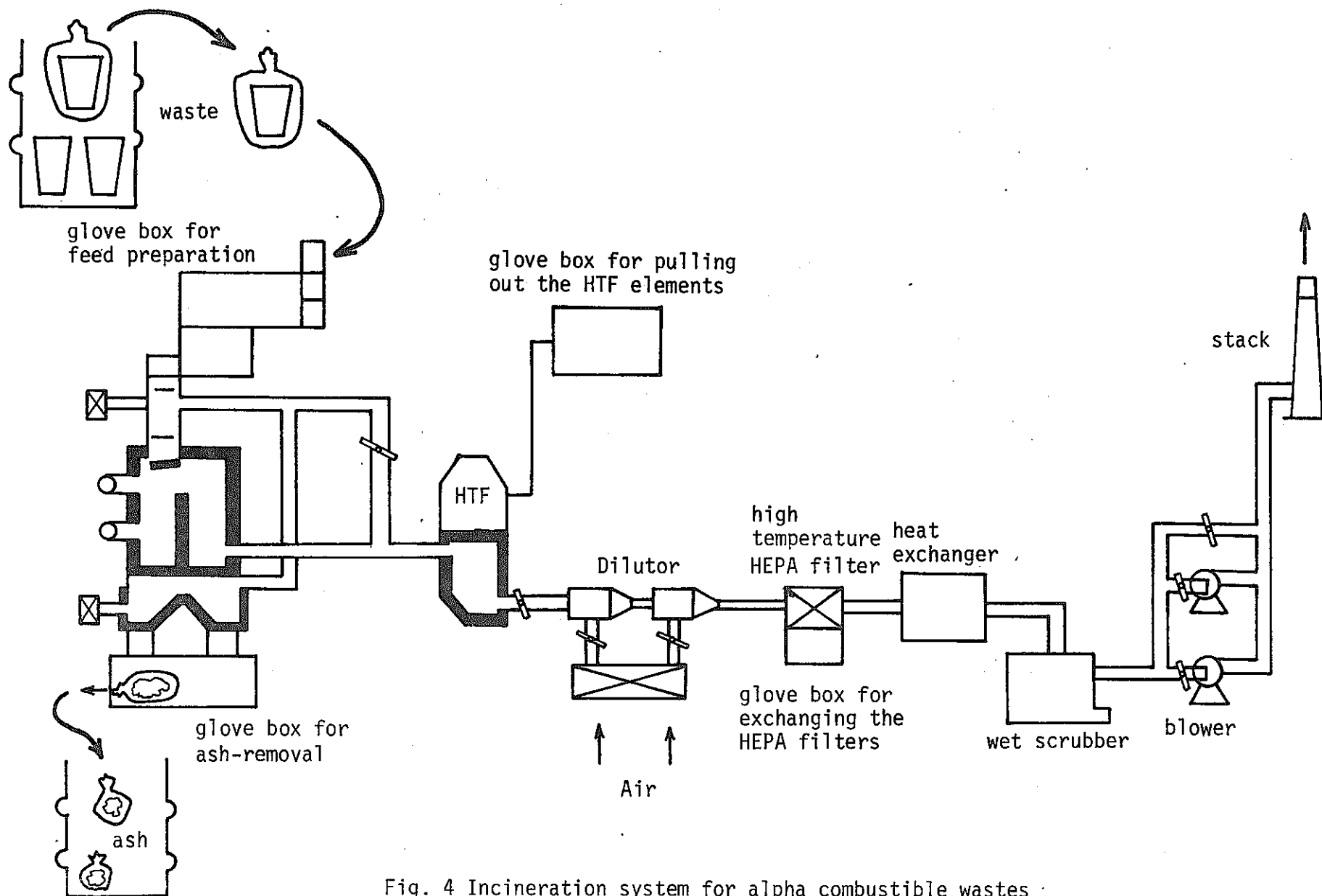


Fig. 4 Incineration system for alpha combustible wastes

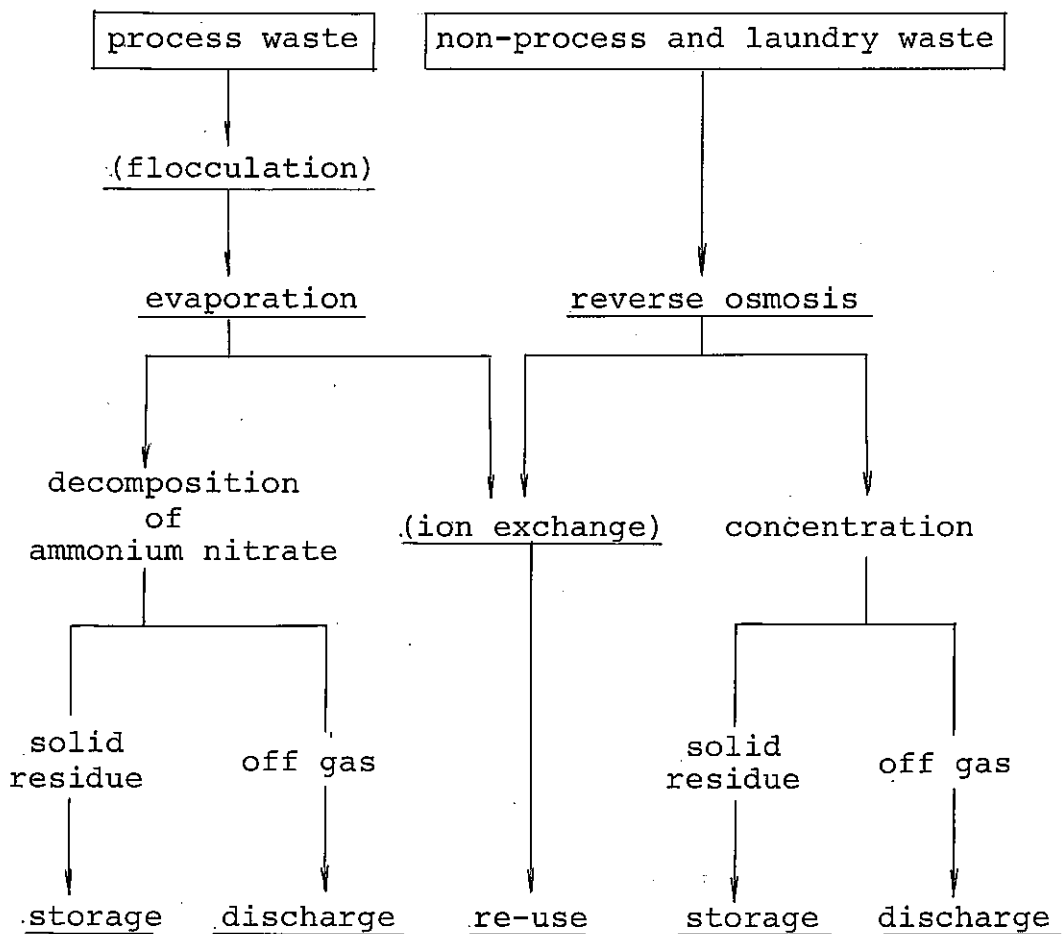


Fig. 5 A flowsheet for a closed cycle system of liquid wastes.