VOLUME REDUCTION OF NON-COMBUSTIBLE RADIOACTIVE SOLID WASTES BY PLASMA MELTING

Noboru Moriyama, Shigeru Dojiri
Japan Atomic Energy Research Institute

Hiroo Ono, Tako Aizu
The Chubu Electric Power Co., Inc.

Moritaka Tamura, Toshihiko Nishikawa
Daido Steel Co., Ltd.
Nagoya City, Japan

ABSTRACT

Non-combustible radioactive solid wastes generated from the nuclear power plants are types of metals such as pipes, structural materials, mechanical parts, etc. Types of non-metals such as heat insulator, concrete, filters, etc. A large space is needed to store them because the major part of the waste is stored without treatment. Rational storage and safe disposal of the waste is occupied in a large space. It is desirable to reduce the volume and to stabilize radionuclides effectively.

Close attention was given to the treatment of melting, from the point of view that we can reduce the volume of the waste to the condition of real density theoretically, and expect to stabilize radionuclides. We adopted and developed the method of melting by plasma arc because of the following items:

1. Not only metals but non-metals can be melted easily by high temperature of plasma arc.
2. The melter furnace is an airtight chamber and a water cooled structure, so we can design it compactly.
3. The electrode is non-comsumable and heat insulator, etc. are consumed scarcely, so we can maintain easily.

As a result of various tests, we recognized the effectiveness and safety of this treatment technique.

BACKGROUND AND HISTORICAL OVERVIEW

We studied about the ability of melting and other various characteristics of melting wastes. As a result, we made it clear that we could reduce the volume of the wastes regardless of metals and non-metals. And we obtained the volume reduction ratio from 8 to 20.

In order to investigate effectiveness and stability of this treatment, we installed a pilot plant in Japan Atomic Energy Research Institute and investigated the ability of melting, behavior of radionuclides at melting, efficiency of off-gas treatment and leaching of radionuclides in solids obtained finally. On the basis of the results, we tried a conceptual design of a practical plant.

This work was developed by the Hokkaido Electric Power Co., Inc. (Yoshifumi Watanabe), The Tohoku Electric Power Co., Inc. (Yasufumi Fujii), The Tokyo Electric Power Co., Inc. (Takashi Mikoshiba), The Hokuriku Electric Power Co., Inc. (Toshio Kamizawa), The Kansai Electric Power Co., Inc. (Toshio Taniguchi), The Chugoku Electric Power Co., Inc. (Junji Yamashita), The Shikoku Electric Power Co., Inc. (Seiichi Fujito), The Kyushu Electric Power Co., Inc. (Tatsuo Mazaki) and The Japan Atomic Power Co. (Megumu Kanba) in addition to three that are indicated at the top column of this paper during two years from April 1983 to March 1985.

ABOUT PLASMA MELTING

The conceptual figures of plasma arc furnace structure are shown in Fig. 1 (the skull melting furnace) and in Fig. 2 (the direct melting furnace).

The skull melting: Method of melting that forms solidified layer (skull) and molten layer during melting.

The direct melting: Method of melting the wastes directly in a crucible.

The skull melting furnace (Fig. 1) has plasma torches and a guide pipe at the upper shell for charging the wastes. Inside the lower shell, there are bricks for insulating heat and a basemetal on the bricks. After melting the basemetal, the wastes are inserted on the molten surface and are melted. The molten waste is discharged by overflow from the discharge hole and is molded in a mold. The entire furnace shell is a water cooled structure.

The direct melting furnace (Fig. 2) has a plasma torch at upper shell and a water cooled crucible. After the wastes are inserted in the crucible, plasma arc is sparked between the torch anode and the wastes, the wastes are then melted by high temperatures of plasma arc. The molten material generated by melting the wastes is cooled in the crucible. Only heated parts of the furnace are water

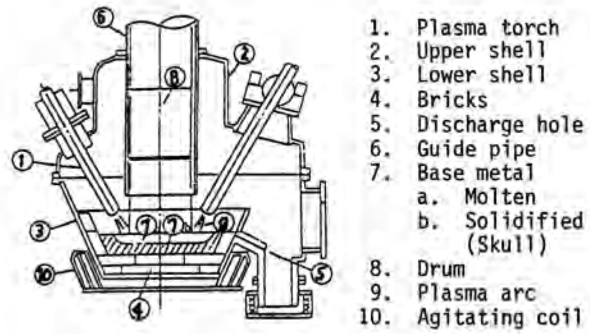


Fig. 1. The Structure of the Plasma Furnace (Skull Melting Method).

water cooled structure. This study was proceeded by the former method mainly. But we discussed the conceptual design of the later method also. Therefore, we also introduce about it.

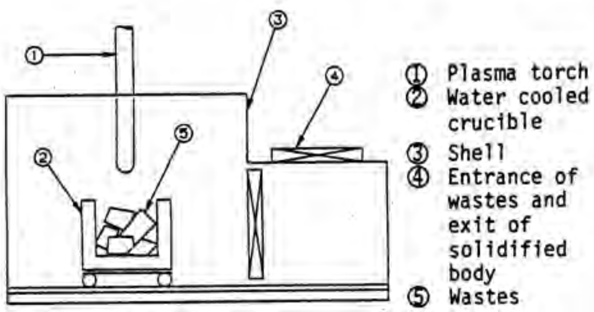


Fig. 2. The Structure of the Plasma Furnace (Direct Melting Method).

A CONCEPT OF THE MELTING TREATMENT PROCESS

The type of non-combustible radiactive solid waste is shown in Fig. 3 (except filters).

They include pipes, structural materials, heat insulators, parts of machine, concrete, glass, filters, electric-wires, etc. The size, quality, and shape of them are various. We make the most suitable concept of melting treatment process from following three points, (1) application to the various type of wastes, (2) safe operation, (3) condition of solidified body (ingot). The process is shown in Fig. 5.

In this hot test, we investigated about melting, solidifying and off-gas treatment mainly in the process.

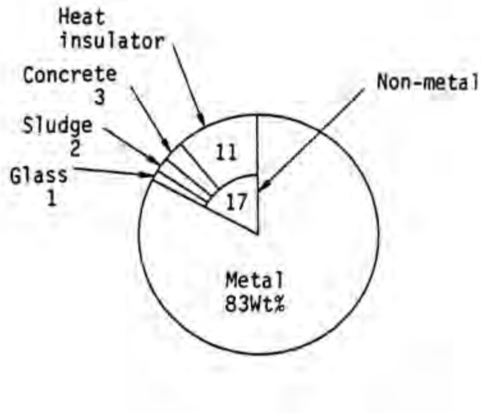


Fig. 3. Type of Non-Combustible Radioactive Solid Waste.

OUTLINE OF THE HOT TEST EQUIPMENT

The Flow of the Equipment

The flow of the equipment is shown in Fig. 6. The main equipment consists of the furnace which melts the simulated wastes, and the equipment of off-gas treatment which has a gas mixing chamber, a dustcollector and a high efficiency filter unit. The furnace structure is basically the same as the conceptual figure (Fig. 1).

The Arrangement of the Equipment

The above equipment was installed in Japan Atomic Energy Research Institute. The equipment was installed in the radioactive controlled room except for the electrical source, water cooling line and gas supplying line, and the main equipment such as the furnace and the equipment of off-gas treatment were installed in the temporary house (8m x 7m by 5m high). The general view of them is shown in Fig. 4.

Specification of the Main Equipment

1. The furnace:

a. Melting rate: 16kg (as steel)/20min

b. Melting method: skull melting method

c. Plasma power: 160kW (80kWx2 torches)

d. Temperature of molten surface: 1500∿1700°C

e. Molding method: Slow cooling in a mold

2. Off-gas treatment

a. Flow rate: $2.2m^3/h$

b. Construction: A gas mixing chamber: Dilution and reduction of temperature by air.
A dustcollector: Bag filter
A high efficiency filter unit: Prefilter+HEPA filter 2 sets

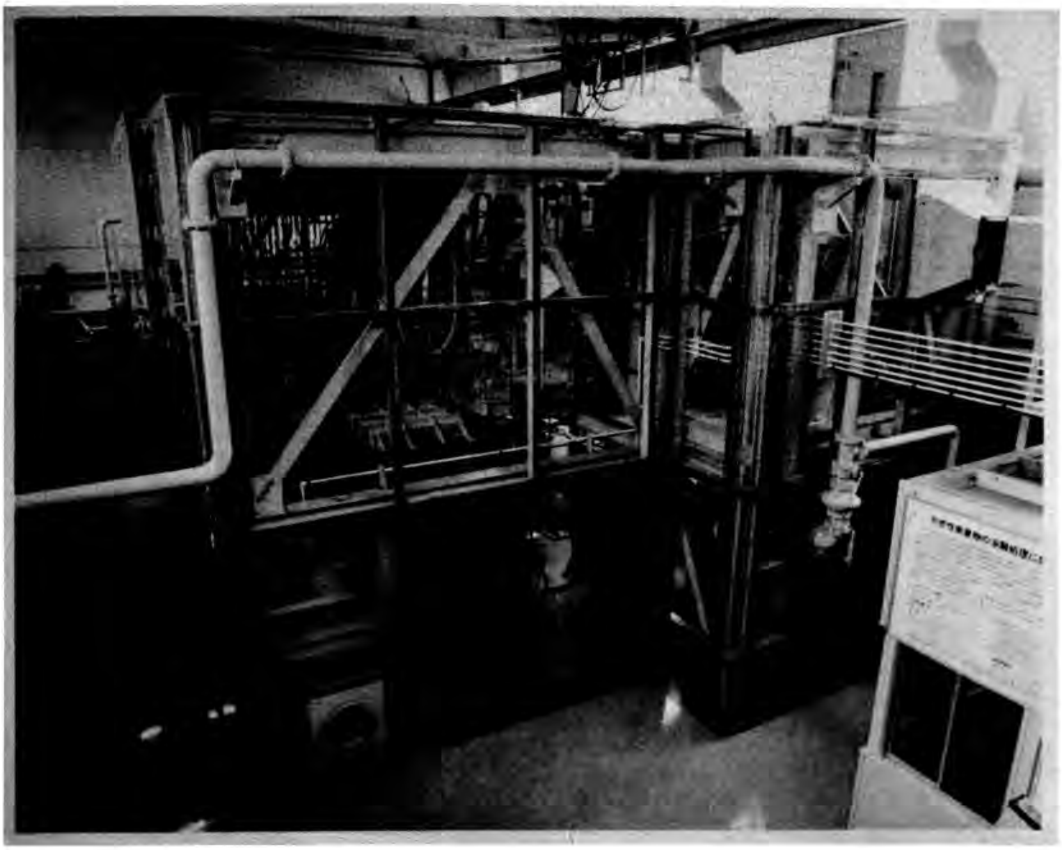


Fig. 4. Appearance of the Hot Test Equipment.

TEST METHOD

Test Procedure

1. The radionuclides solution is deposited on the simulated waste and it is dried.

2. A simple drum (7 ℓ drum about 100mmøx1100mmH) with the simulated waste is set in the guide pipe of the furnace.

3. The furnace and the accessary equipment are operated. After the basemetal is melted, the drum is inserted on the surface of the basemetal and the waste is melted. The molten waste is molded after melting. During operation, dust in the off-gas are collected properly.

4. After operating, various samples are collected to obtain the necessary data.

An example of the simulated waste and the simple drum is shown in Fig. 7. The appearance of the solidified body is shown in Fig. 8. Its size is approximately 260mmx180mmx80mm-h.

Radionuclides Used

Types and quantity of radionuclides used at one test were Co-60, Cs-137 (each about 1mCi) Zn-65, Mn-54, Sr-85 (each about 0.2m Ci). Activity concentration in the simulated waste was 0.16μCi/g (waste).

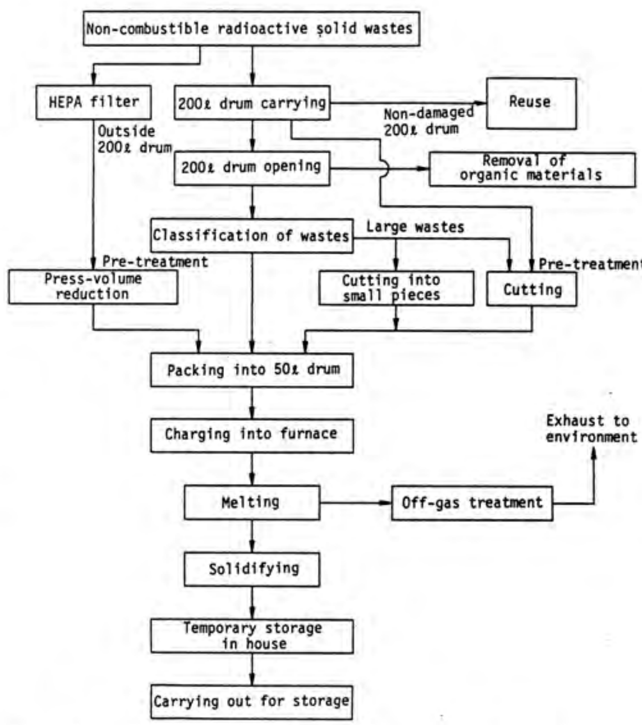


Fig. 5. Flow Sheet of Volume Reduction Process.

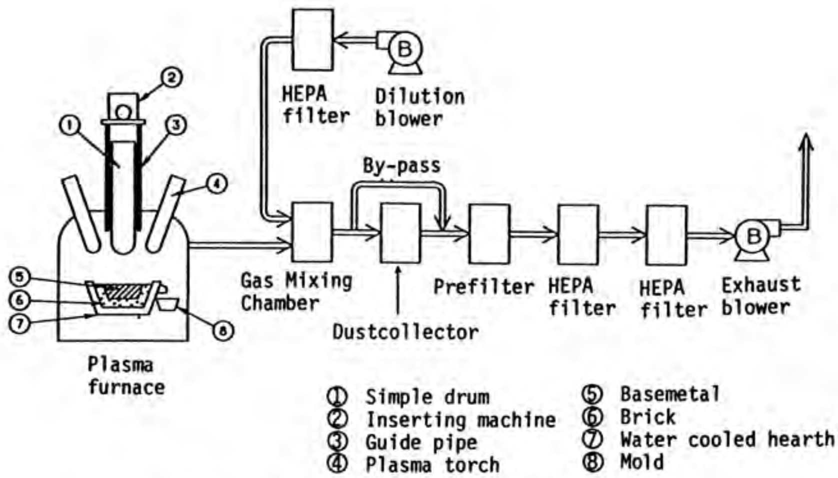


Fig. 6. Flow Sheet of the Hot Test Equipment.

Fig. 7. The Simulated Wastes and Simple Drum (Steel+ Silicaboard+Polyethylene).

Fig. 8. Appearance of the Solidified Body.

The Simulated Waste

The simulated waste is shown in Table I.

TABLE I

Composition of the Simulated Wastes

Wastes	Composition
C.S. (Carbon Steel)	C.S. 100%
C.S. + Silicaboard	C.S. 80%, Silicaboard 20%
C.S. + Silicaboard + Polyethylene	C.S. 82.5%, Silica-board 16.5%, Poly-ethylene 1%
C.S. + Silicaboard + Concrete	C.S. 80%, Silica-board 10.4%, Concrete 9.6%
SUS	SUS 50%, C.S. 50%

TEST RESULT

Behavior of Radionuclides

A melting test was performed and 11 charges and behavior of radionucledes were investigated.

1. The major part of Co-60 transfers to metal regardless of slag being or not (Fig. 9). Mn-54 transfers to metal under melting only metal, and transfers to slag as slag generates (Fig. 10). The almost part of Sr-85 transfers to slag and it transfers scarcely to metal. Cs-137 doesn't transfer to metal, and the transfer ratio to slag is different accouding to the composition of slag. Its transfer ratio to slag under melting concrete is greater than that under melting silicaboard. Because viscosity of slag made of concrete is higher than that of silicaboard. The major part of Zn-65 transfers to dust.

2. The transfer ratio to dust is in order of Sr-85<Co-60<Mn-54<Cs-137<Zn-65, and they volatilize according to each element's vapor pressure.

3. The non-clear parts are due to situation, method, etc. of sampling. Judging from each element's vapor pressure, basic test, etc., we think that

Co-60 transfers to metal, Mn-54 transfers to metal or slag, Sr-85 transfers to slag, Cs-137 and Zn-65 transfer to dust.

Distribution of Radionuclides

In the case of the simulated wastes made of SS, silicaboard, and polyethylene, the distribution of radionuclides after melting is shown in Table II. Through all charges, radionuclides are less than lower limit of detection at the outlet of the high efficiency filter unit.

High Loaking Test of Radionuclides to the High Efficiency Filter Unit by Using By-pass of the Dustcollector

On the assumption of an accident by bag filter's damage in a dustcollector, we tried the test that off-gas passed from the gas mixing chamber directly to the high efficiency filter unit by using by-pass of the dustcollector. The radionuclides were used as standard concentration. The simulated waste was only metal. Activity concentration was 0.16 μCi/g (waste).

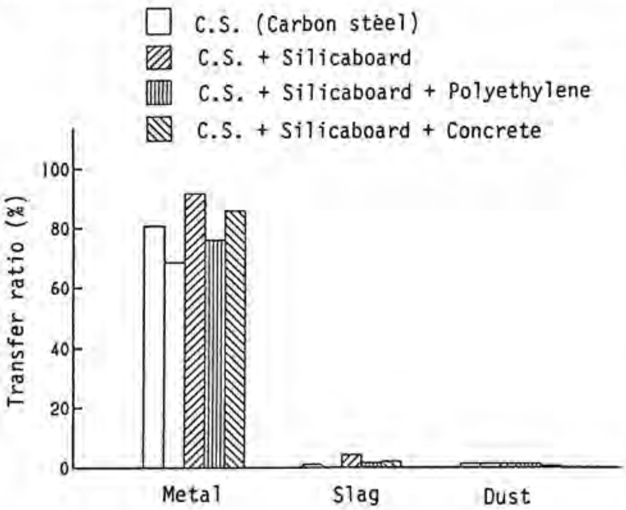


Fig. 9. The Transfer Ratio of Co-60.

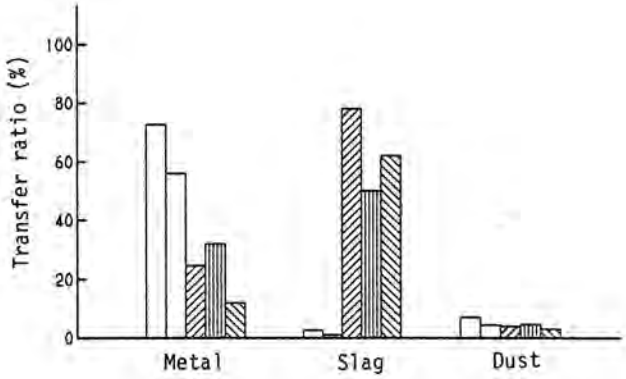
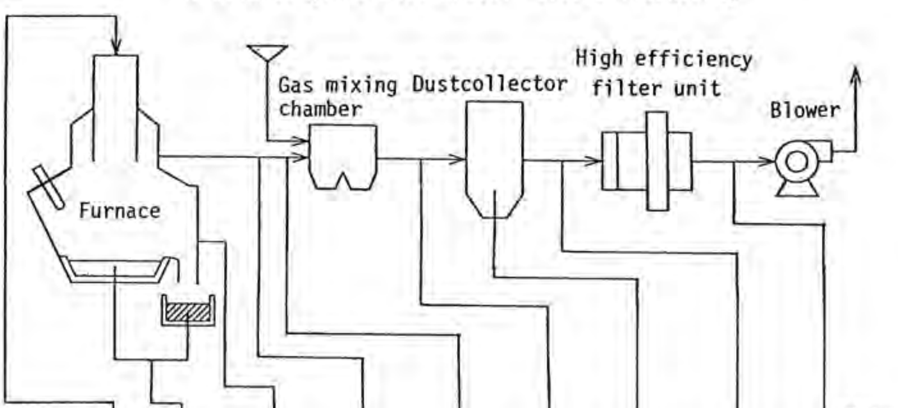


Fig. 10. The Transfer Ratio of Mn-54.

TABLE II

RI Balance (Simulated Waste: C.S.+Silicaboard)



Unit:μci

	Waste	Solid *	Furnace	Pipe	Off-gas	Off-gas	Dust	Off-gas	Off-gas
Co-60	2001	1959	12.1	0.22	0.54	2.45	6.22×10^{-3}	5.4×10^{-3}	5.1×10^{-3}
Mn-54	419	400.4	8.74	0.82	1.52	8.52	1.84×10^{-2}	5.3×10^{-3}	5.1×10^{-3}
Sr-85	430.4	365.1	3.12×10^{-2}	3.94×10^{-2}	4.40×10^{-2}	ND	4.84×10^{-4}	6.7×10^{-3}	6.4×10^{-3}
Cs-137	2192	269.6	530.7	43.3	114.6	340.1	6.74	1.35×10^{-2}	7.0×10^{-3}
Zn-65	267.4	28.0	120.7	10.4	26.7	124.6	0.29	1.3×10^{-2}	1.2×10^{-2}
Total	5310	3022	672.3	54.8	143.4	475.7	7.06	4.4×10^{-2}	3.6×10^{-2}

*: (Basemetal + Solidified body)

As a result, we could not detect radionuclides at the outlet of the high efficiency filter unit and we could recognize the safety of the off-gas.

Decontamination Factor (DF) of Each Equipment

By melting, radionuclides are distributed to solid (metal, slag) and dust.

Radionuclides exhausted from the furnace to the off-gas treatment line are eliminated at the gas mixing chamber, the dustcollector and the high efficiency filter unit. DF of each equipment are as follows:

DF of Co-60, Mn-54, and Sr-85 for the furnace are from 3×10^2 to 2×10^3. DF of Cs-137 and Zn-65 are about 20. DF of volatile Cs-137 is 1.9×10^4 for the dustcollector and 96 for the prefilter.

We recognized that Cs-137 exhausted from the furnace was caught and not detected at the outlet of the off-gas treatment line. The result is shown in Table III.

TABLE III

DF of Each Equipment

	Co-60	Mn-54	Sr-85	Cs-137	Zn-65
Furnace	2250	310	960	15	14
Gas mixing chamber	1	1	1	1	1
Dust-collector	57 (1.9×10^4)	29x10 (1.9×10^4)	9.2 (1.9×10^4)	1.9×10^4	2.3×10^3 (1.9×10^4)
Prefilter	51.2	86.4	53.1	196	392
HEPA filter (first)	(1000)	(1000)	(1000)	(1000)	(1000)
HEPA filter (second)	(100)	(100)	(100)	(100)	(100)
Over all DF	6.5×10^{11} (2.2×10^{14})	7.7×10^{11} (5.1×10^{13})	4.6×10^{10} (9.7×10^{13})	5.6×10^{12}	1.2×10^{12} (1.0×10^{13})

( ):Presumption

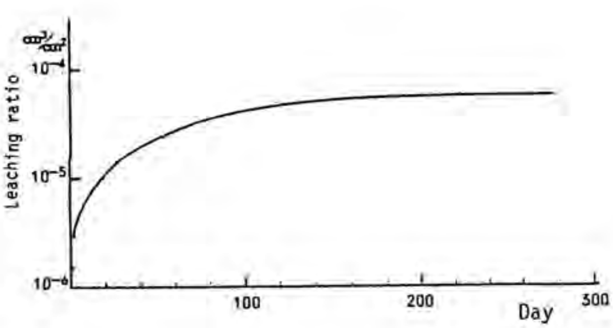


Fig. 11. Cumulative Leaching Ratio of Co-60.

Condition of Radionuclides Accumulation in the Equipment

The radionuclides are caught and grow up on the wall of the furnace, inside the pipe of the off-gas line and the dustcollector etc. We examined its condition by measuring gamma-dose rate on the surface of the equipment. Gamma-dose rate is saturated by several charges of melting. It shows that the dust repeats growing and falling when it grows up to a fixed weight. Therefore radionuclides are caught at the wall, and are saturated by repeating melt. We presume that all parts of the radionuclides volatilized on the molten surface are exhausted to the off-gas line. The other equipment has a tendency to the same as above mentioned.

Leaching from Solidified Bodies

Leaching rate from the solidified bodies (10 17kg) generated finally were measured. Diffusion coefficient of each nuclides are about

Co-60 $4 \times 10^{-11} cm^2/d$, Mn-54 $6 \times 10^{-10} cm^2/d$,
Sr-85 $9 \times 10^{-6} cm^2/d$, Cs-137 $6 \times 10^{-6} cm^2/d$,
Zn-65 $7 \times 10^{-2} cm^2/d$

We could recognize stability of the solidified bodies. An example of results is shown in Fig. 11.

CONCEPTUAL DESIGN OF PRACTICAL PLANT

On the basis of the results, we tried the conceptual design of practical plants about specification and arrangement of the equipment, balance of radionuclides, safety of off-gas treatment, distribution of gamma-ray intensity from the solidified bodies, accumulation of radionuclides in each equipment etc. An example of contents discussed is shown as follows:

Condition for the Conceptual Design

We presume the following conditions:

1. Type of the waste and capacity of treatment, as shown in Table IV.
2. Activity Concentration 1.0×10^{-2} μCi/g (waste)
3. Composition of radionuclides
   Co-60 56.6%, Mn-54 33.1%, Sr-90 1.0%
   Cs-137 1.0%, Zn-65 8.3%

Specification an Arrangement of the Equipment

The specification of the main equipment is shown in Table V. As an example of skull melting method, the bird's eye view for the arrangement of the equipment is shown in Fig. 12.

Discussion for the Off-Gas Treatment

We could not determine accurate DF because concentration of radionuclides was less than lower detection limit at the outlet of the dustcollector except Cs-137. Therefore, we presumed that DF of each radionuclides were the same as that of Cs-137. We could not determine accurate DF for the HEPA filter, so we presume the value which is safer than designed value. By using the DF shown in Table III, we calculated the activity concentration at the outlet of the off-gas line and compared it with the permissible value. As a result, we recognized safety of the off-gas treatment because it was low enough. For an accident of bag filter's damage, we calculated as the same and recognized its safety. We obtained the same result about the direct melting method.

CONCLUSION

By the mock up "cold" test equal to actual scale, we recongized volume reduction of the waste and adequacy of the equipment ability by plasma melting.

By the "hot" test, we recognized behavior of radionuclides at melting, ability of the equipment, leaching rate from solidified bodies, etc. On the basis of the results, we tried the conceptual design of practical plant and recognized safety in this process.

REFERENCES

1. The submitance report of 1980 Annual Meeting of the Atomic Energy Society of Japan.

2. Eguchi, Isamu, et al., "Processing of Nuclear Wastes by Plasma Arc Melting; Seminar on Pilot Plant Experiences-Melting and Processing Technology" Sponsored by Vacuum Metallurgy Division.

3. Yokoi, Shinji, et al., "Behavior of Radionuclides at Smelting of Non-Combustible Solid Wastes," 1981 Annual Meeting of Materials Research Society.

4. Tamura, et al., The submitance report of Meeting of The Chemical Engineering Society of Japan F11 (1982).

5. Miyagawa, et al., The Journal of the Thermal and Nuclear Power, Vol. 135, No. 6, p-27-46 (1984).

6. Moriyama, Noboru, et al., The submitance report of 1985 Fall Meeting of the Atomic Energy Society of Japan.

TABLE IV

Type of Waste and Capability of Treatment

		Skull melting method	Direct melting method
Kind	Metal	82.5%	90%
	Non-metal	16.5	10
	Organic material	1	-
Treatment rate		28 drums (50ℓ drum)/day 8h/day 700kg (as steel)/day	200kg/day 8h/day 2 batches/day

TABLE V

Specification of the Equipment

		Skull melting method	Direct melting method
Plasma furnace	Method of inserting wastes	Continuous inserting by 50ℓ drum	Batch inserting by bucket
	Plasma power	420kW (105kWx 4 torches)	180kW (1 torch)
	Solidifying method	Slow cooling in a mold	Direct solidifying in a water cooled crucible
	Size of solidified body	ϕ457mmx170mmh 80kg	ϕ400mmx170mmh 100kg
Off-gas treatment equipments	Component	A gas mixing chamber A dustcollector A high efficiency filter unit (pre+HEPA+HEPA)	The same as left
	Flow rate	About 220Nm^3/h	About 70Nm^3/h
Space occupied by the main equipments		15mx15m by 10m-h	8mx8m by 8m-h

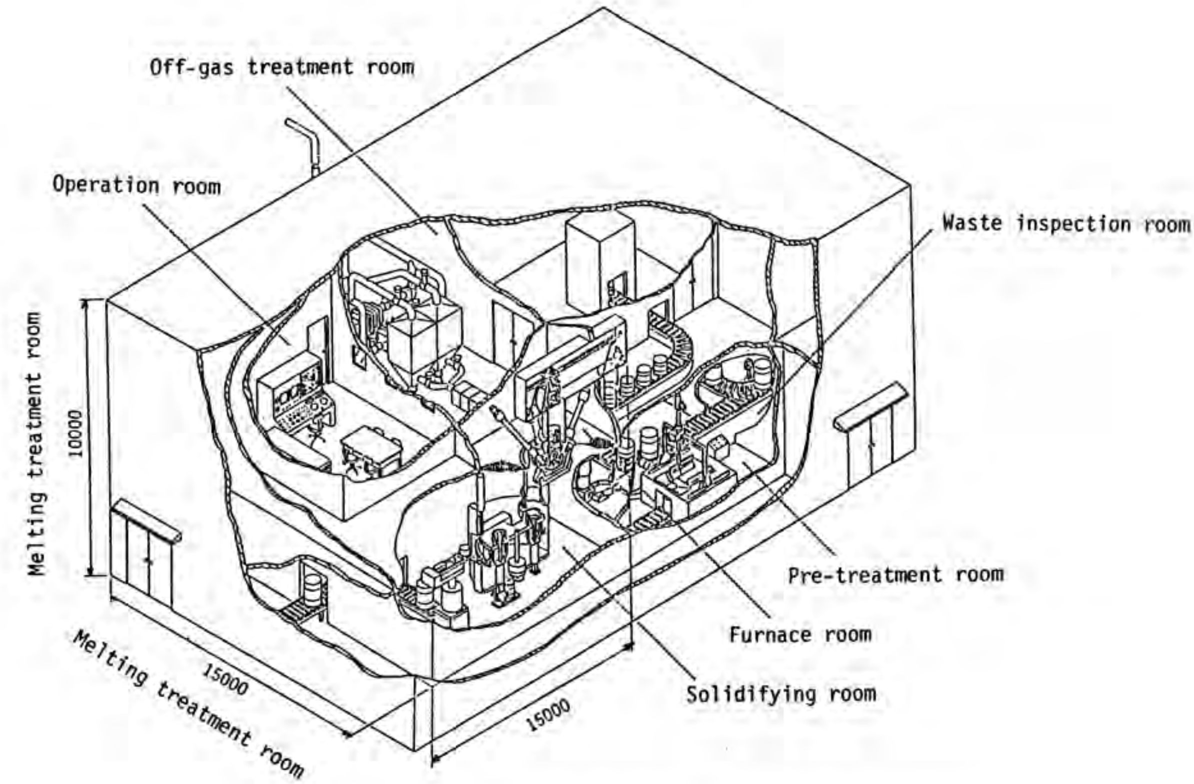


Fig. 12. Bird's Eye View for the Arrangement of the Equipment.