DECONTAMINATION AND MELTING TEST FOR METAL WASTES IN JPDR DECOMMISSIONING PROJECT - TOWARD RECYCLING OF LLW -

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ABSTRACT

Decontamination technology is important in the decommissioning of nuclear reactor facility, not only for the reduction of radiation level in the working environment but also the management of dismantled metal wastes. As part of the technology development in the Japan Power Demonstration Reactor(JPDR) decommissioning program, several decontamination methods have been developed in JAERI. The methods developed for dismantled metal wastes includes electropolishing method, Sulfuric acid-Cerium method, chemical immersion method and flowing abrasive method. It is demonstrated that these methods provide sufficiently high performance in decontaminating dismantled metal wastes into reusable level.

While such decontamination methods are very effective to make low-level metal waste reusable, metal melting is beginning to be a concern as simpler and straight forward way for the reuse of metal wastes from nuclear facilities. However, the safe and rational techniques for recycling the dismantled metal wastes have not been established yet, especially in accordance with the strict practice on the radioactive materials management in Japan.

In conjunction with such circumstance JAERI has been conducting basic melting and casting test. This is the part of first stage of decommissioning material reuse research in JAERI. The objective of the test is to reveal the behavior of radionuclide during melting and casting processes of metal wastes and the influence of radioactive materials on working environment quantitatively. The basic parameters are melted material type such as stainless steel or carbon steel, kind of the additives or flux, temperature of melted steel, and so on. One of the important parameter to be measured is a radionuclide concentration in slug because it determines the decontamination ratio by melting. The core of the test facility is 500kg high frequency induction furnace with a capacity of 350kVA which is newly constructed and installed for this test program. The furnace and the chamber surrounding the melt facility are equipped with separate ventilation system from the building. The preliminary melting/casting tests using non-radioactive metals has been carried out, and the results shows the test facility has sufficient capability to carry out coming RI tracer tests.

INTRODUCTION

A large amount of low level solid wastes will be generated from decommissioning of nuclear facility, and much of these low level wastes(LLW) are also expected to be generated during specific period especially in its dismantling phase. In Japan, large scale power reactors are assumed to be eventually dismantled after shutdown and 5 to 10 years of safe storage in order to use their site for new power reactors, due to lack of sufficient siting locations. Establishing the rational methods to reduce the amount of LLW is, therefore, very important activity in preparing for the power reactors decommissioning especially in such country which has limited siting area for the final repository, as Japan.

In many nuclear facilities, decontaminations are carried out to reduce radiation level in the working environment and many kinds of decontamination methods have been developed according to various conditions of contaminations. In decommissioning of nuclear facilities, however, decontamination is important not only to reduce the radiation level in the facility, but to make a part of metal wastes be able to be released in unrestricted manner. Decontamination method for such purpose should achieve high decontamination factor(DF). Also a decommissioning of nuclear power reactor will generate a huge amount of LLW during relatively short interval. Therefore, decontamination of each dismantled component must be carried out in a short time without sacrificing high DF. In addition to such high performance, following characteristics are the subjects on the development of decontamination methods for the decommissioning:

- Only small amount of secondary wastes is generated.
- Secondary wastes generated is to be easily processed.
- Efficient and quick measurement for residual very low radioactivity is possible.

Japan Atomic Energy Research Institute(JAERI) has been engaging in development of efficient decontamination methods (1, 3) as part of the technology development in the Japan Power Demonstration Reactor(JPDR) decommissioning program (4, 5). Several methods have been applied to decontamination of dismantled component of JPDR. They are electropolishing method and chemical

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immersion method. Mechanical method using grid blasting is also being developed and will be applied in this year.

While such decontamination methods have been proved as very effective to make low-level metal wastes reusable, metal melting is beginning to be a concern as a simpler and straight forward way for the reuse of metal wastes from nuclear facilities. JAERI has been conducting basic melting and casting test in conjunction with such circumstance.

In the following sections, these activities of JAERI related to possible future reuse of dismantled metals are described.

DECONTAMINATION METHODS DEVELOPED IN JAERI

Decontamination works for the metal structure in reactor system are usually categorized into two groups: decontamination before and after dismantling. The former is referred as "system decontamination" and the latter as "decontamination of dismantled equipments/components". Typical decontamination methods for each category are shown in Fig.1. Both chemical and mechanical methods are available for each category, but decontamination of dismantled equipment usually requires higher DF than the system decontamination. This is because main purpose of the system decontamination is usually to reduce occupational exposure and, therefore, does not need high DF. On the other hand, decontamination of dismantled equipment is usually intended to reduce contamination down to the level of free-release as non-radioactive wastes. Hence, even some methods are commonly available for both categories of decontamination, modifications to improve the efficiency are usually required when such methods are applied to the decontamination of dismantled equipment.

As such example, the redox method using sulfuric acid and Cerium solution (SC-process) 3), which has been developed in JPDR decommissioning project and has been proved its high performance for system decontamination, is more improved and utilized in the chemical immersion decontamination system for dismantled components.

Even the system decontamination does not require high DF, flowing abrasive method, another original method developed in JPDR project, has been shown as a very effective method with DF of more than 1000 through system decontamination test. This method, a kind of mechanical decontamination, uses the water flow and suspended abrasive particles and, therefore, can be utilizes under room temperature. Moreover only an abrasive collector and a cartridge filter are required in the liquid wastes treatment system. This makes a decontamination system very simple and easy to handle.

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1. DECONTAMINATION BEFORE DISMANTLING FOR
   REDUCTION OF OCCUPATIONAL EXPOSURE

                                  ┌ CHEMICAL METHOD
          ┌ PIPE LINE SYSTEM ─┤
          │                       └ MECHANICAL METHOD
   ───┤
          │                       ┌ HYDRO-JET METHOD
          └ POOL, TANK ───────┼ BLAST METHOD
                                  ├ STRIPPABLE COATING METHOD
                                  └ -----

2. DECONTAMINATION AFTER DISMANTLING FOR
   RECYCLE OF METAL WASTE AND
   REDUCTION OF RADIOACTIVE WASTES

                                  ┌ ELECTROPOLISHING METHOD
                                  ├ CHEMICAL IMMERSION METHOD
   ─── PIPES, COMPONENTS ─┼ BLAST METHOD
                                  ├ ULTRASONIC WAVE METHOD
                                  └ GEL METHOD
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Fig. 1. Decontamination for decommissioning.

In addition to these methods, electropolishing method have been developed in JPDR and utilized in decontamination system for dismantled components. This methods will be described in next section.

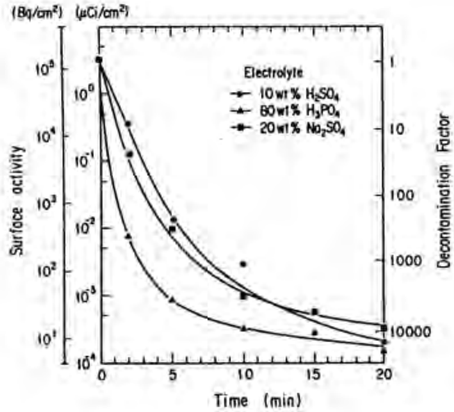
DECONTAMINATION OF DISMANTLED COMPONENTS

Since there is no decontamination methods which can apply to all kinds of dismantled component, several methods are used depending on the shapes and materials of the objects. The electropolishing method has been used in decontamination of straight pipes or components with relatively simple shape. Equipments/components with more complicated shape will be decontaminated by the chemical immersion method.

Electropolishing Decontamination

From the results of the basic examination of electropolishing, optimum decontamination conditions for each electrolyte were determined to be as follows:

Electrolyte	Concentration	Current Density	Temperature
phosphoric acid	80 wt%	0.2 A/cm^2	60°C
sulfuric acid	10 wt%	0.2 A/cm^2	60°C
neutral salt (Sodium sulfate)	20 wt%	0.4 A/cm^2	60°C



Sample : RTD of Commercial power plant (PWR)
Decon. Method : Anodic Electrolysis with Immersion System
Material : SUS-316
Temp. of Electrolyte : 60℃
Current Density : 0.2 A/cm²

Fig. 2. Result of basic examination of electropolishing using stainless steel samples taken from JPDR.

The electropolishing decontamination under these condition was performed using the samples from the JPDR and the commercial power plants. Figure 2 shows the decontamination results for JPDR's primary coolant system pipes (SUS 304) for each decontamination method. The results indicated that the anodic decontamination method could rapidly decontaminate the samples in every electrolyte and also indicated that the DF of more than 5000 was obtained. In addition, the results also showed that the decontamination time required for the alternating current(a.c.) decontamination method was twice that for anodic decontamination method when sodium sulfate solution was used as an electrolyte. However, sufficient results were attained by every method after 20 minutes of decontamination.

The result of decontamination test using removed equipment sample of stainless steel from commercial power plant also showed very high DFs. A case of RTD (resistance temperature detector) of PWR, initial contamination of more than 10E5 Bq/cm^2 were completely removed within 20 minutes of decontamination (Fig.3).

Even such high performance, some limitation are posed in application to decontamination of dismantled components from JPDR.

- **Material**

  Comparing with the case of stainless steel, in which DF of over 5000 was attained, surface contamination on carbon steel is rather difficult to remove. In order to get efficient result, care must be taken in selecting electrolyte liquid and in applying electric current.

- **Shape of the object**

  A component with asymmetric or complicated shape takes longer time to be decontaminated and needs some complement measure such as mechanical rotation of the cathode and others.

- **Effect of cutting method and others**

  Figure 4 summarized affected factors on the performance of electropolishing decontamination for dismantled pipes.

Chemical Immersion Decontamination

Chemical decontamination is suitable method to such objects as component with complex shape and/or with pinholes or gaps, to which the electropolishing method is not effective. The redox method using SC- process, which has been proved its high performance for system decontamination, is also applied to the decontamination of dismantled components. This is called as chemical immersion decontamination system. Major improvements in the method were:

- **recycled use of decontamination reagents (H_2SO_4, Ce^{3+}),**
- **using higher concentration of reagents, (H_2SO_4: 0.25 -> 1.0 mol, Ce^{4+}: 0.002 -> 0.1 mol)**

Test apparatus has been installed in 1990 and the decontamination of the JPDR's dismantled components is in progress.

Mechanical Decontamination

The efficiency of mechanical decontamination method using abrasive is not so much affected by material of the object and contamination level. There is, in principle, no upper limit of DF value obtained by such kind of method because the operation continues the longer time, the higher DF is achievable. The flowing abrasive method for the system decontamination of JPDR cooling loop used the water as carrier and B_4C as abrasive particles. For decontamination of the dismantled components, however, steel grid will be blasted by flowing medium, for easiness of handling and high efficiency. Details of the method are now under discussed in preparing for the decontamination work planned in his year.

METAL MELTING TEST IN JAERI

Advantages of the Metal Melting

While many decontamination methods have been developed and some methods are very effective to reduce contamination of dismantled component down to the level

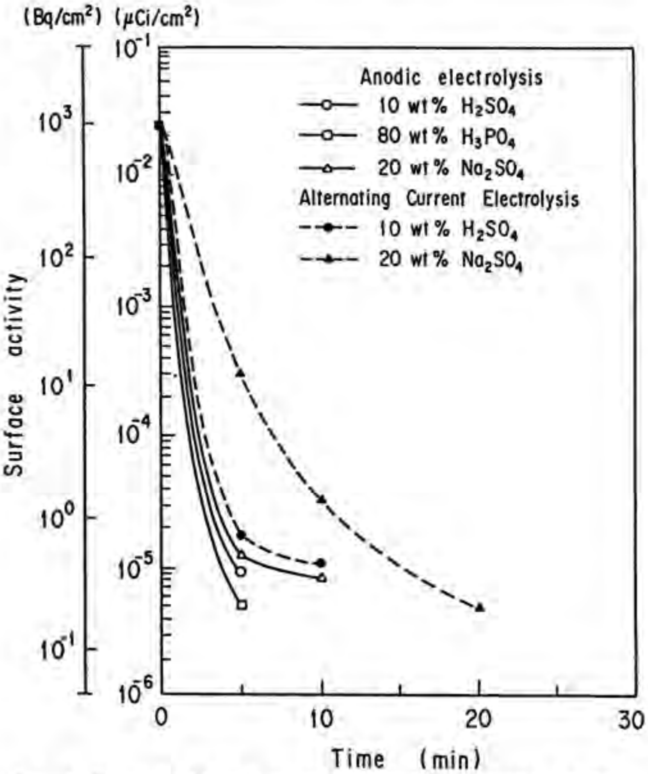


Fig. 3. Result of basic examination of electropolishing using components of PWR.

of free-release as non-radioactive wastes, metal melting is beginning to be a concern as a simpler and straight forward way for the reuse of metal wastes from nuclear facilities.

As mentioned previously section, contamination taken into melted metals and activated radioactivity inside the metal cannot be removed by usual decontamination methods. In concerning such contamination, melting is a suitable method to treat low level metal wastes.

Of course the most significant merit of melting from waste management view is volume reduction. Low level metal wastes from reactor dismantlement are usually packed into steel drums or containers. Average density of these containers are lower compared to the density of material itself. In our experience, average density of $1m^3$ containers used in JPDR decommissioning is about 1.0 while dismantled component are mostly made of steel with density about 8. Therefore the melting is almost ideal solution in volume reduction of metal wastes, even recycle of such materials are not the scope of the waste management.

Beside such volume reduction, melting has other advantages. By melting, many volatile radioactive materials are vaporized and transferred into slug or exhausting gas treatment system and removed from casted metal ingot. This means melting process reduce specific radioactivity in the metal matrix and is regarded as a partial decontamination method. Among the radioactive materials in LWR

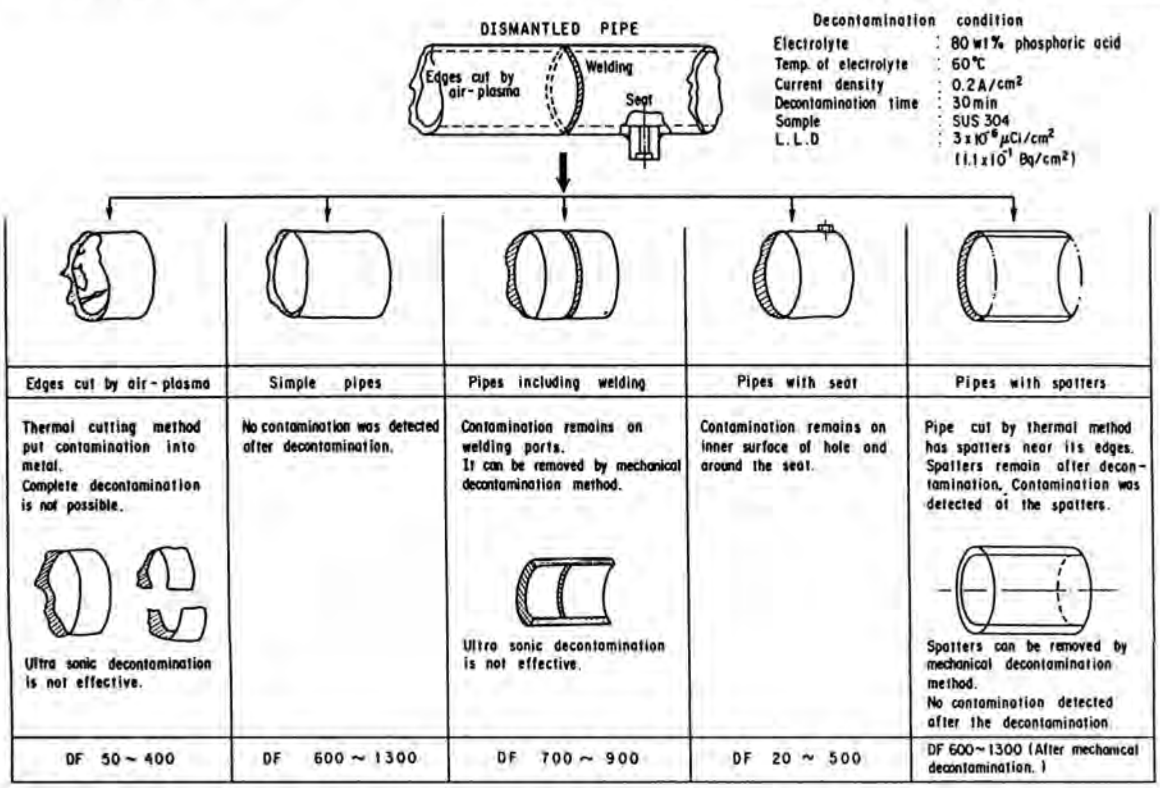


Fig. 4. Effect of surface condition of piping on electropolishing decontamination.

power plant contamination, for example, such as ^{137}Cs and ^{54}Mn will vaporized and move to slug and dust. In addition to such decontamination effect, melted metal has fairly uniform composition and this makes radioactivity measurement of solidified metal ingot very simple by using only small amount of samples taken from ingot.

Another merit of melting is that it fits into the industrial reuse of metal wastes because recycled metal will be at least melted once anyway in order to be used as materials in the industrial processes.

Research on the Recycle of Metal Waste in JAERI

With regard to the industrial aspect, the recycle system for ordinary metal wastes is already established, because metal wastes has inherent economical value. Therefore it is expected that the existing recycle system will function well for the wastes under an exemption level. Although the exemption level is not yet determined in Japan, it will be a subject to be discussed in near future, in order to prepare for the huge amount of low level wastes generation from decommissioning of power reactors. Moreover, reuse of radioactive metal wastes with slightly higher activities than exemption level have been discussed, considering the application within the nuclear installations, with the ease in promoting recycle taken into account.

In establishing the recycle system for wastes metal and necessary criteria, the behavior of radionuclides during various industrial processes in recycling system must be revealed for the safety of workers and the public. Especially transport of radionuclides during melting and solidification of the radioactive metals should be quantified because it is an important factor to the radiation dose evaluation for the downstream of the recycle process. It is also a major factor to decide upper limit of radioactive concentration in the metal wastes to be reused, even for the limited applications within the nuclear installations. At present in Japan, however, there is no relevant data on melting process, as well as the safe and rational techniques for recycling the dismantled metal wastes, especially in accordance with the strict practice on the radioactive material management in Japan.

In conjunction with such circumstance JAERI has been conducting basic melting and casting experiments (7, 8). This is the part of first stage of decommissioning material reuse research in JAERI, performed under contract with the Science and Technology Agency since 1987. In this research, melting tests are planned using radioactive metal wastes from JPDR decommissioning and other materials. The objective of these tests are to investigate and assess the behavior of radionuclides during melting and casting processes and its influence on working environment quantitatively. A series of cold test using fresh steel bars have

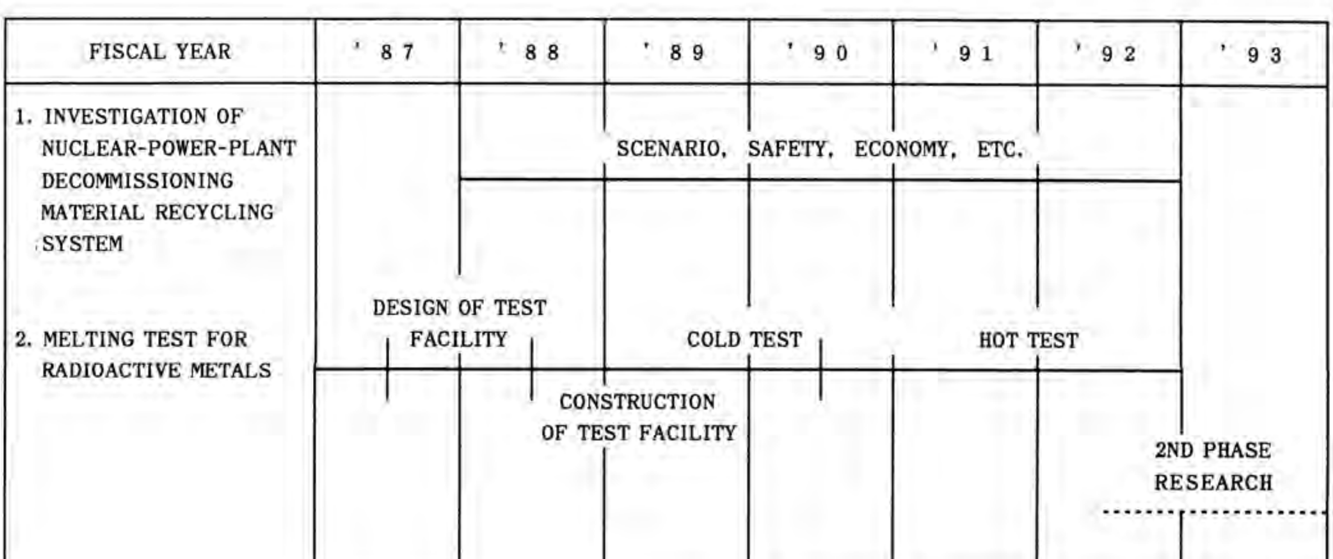


Fig. 5. Schedule of current research program in JAERI on the recycle of metal waste.

completed in 1990, as shown in the program schedule in Fig.5.

Experimental Facility

Melting equipment and related facilities have been build and installed in a building in JPDR which was formerly used as dump condenser building, by March 1990. The core of the test facility is 500kg high frequency induction furnace which is specially designed for this program. Induction furnace was selected because:

- Feasible melting of both stainless steel and carbon steel,
- Less secondary wastes(aerosol, dust, etc.),
- Easiness to make air lock by hood.

Its maximum electric capacity is 350kVA and a ring hood is equipped for air lock during melting. The melting furnace and casting equipments are placed in a containment steel chamber. Separate ventilation system is also equipped to the chamber to keep its inside slightly negative pressure and to minimize the spread of contamination by radionuclides during operation. Another local ventilation using air curtain system is installed to collect aerosol generated during casting. The filtering system in exhaust line is composed of a cyclone, bag filters and HEPA (high efficiency particulate air) filters. Outlines of the experimental facility are shown in Fig.6.

The chamber is installed in order to satisfy practical regulation in Japan. Because the radiation exposure of the workers by inhalation of radioactive materials are regulated in very strict manner based upon "The Law concerning Prevention of Radioactive Hazard" and it is usually recommended to keep a space surrounding the equipment negative pressure.

Contents of the Tests and Present Status

Basic parameters controlled in the tests are:

- type of material melted such as stainless steel or carbon steel,
- condition of radioactivity(activated, contaminated or artificially added tracer)
- kind of the additives or flux,
- temperature of melted steel, and so on.

In order to avoid extra exposure for the workers, three types of tests have been planned: cold test, radioactive isotope(RI) tracer test, and actual decontamination test using dismantled JPDR components. Data on the material balance, radioactivity balance, and working environment condition (including dose rate and dust concentration) will

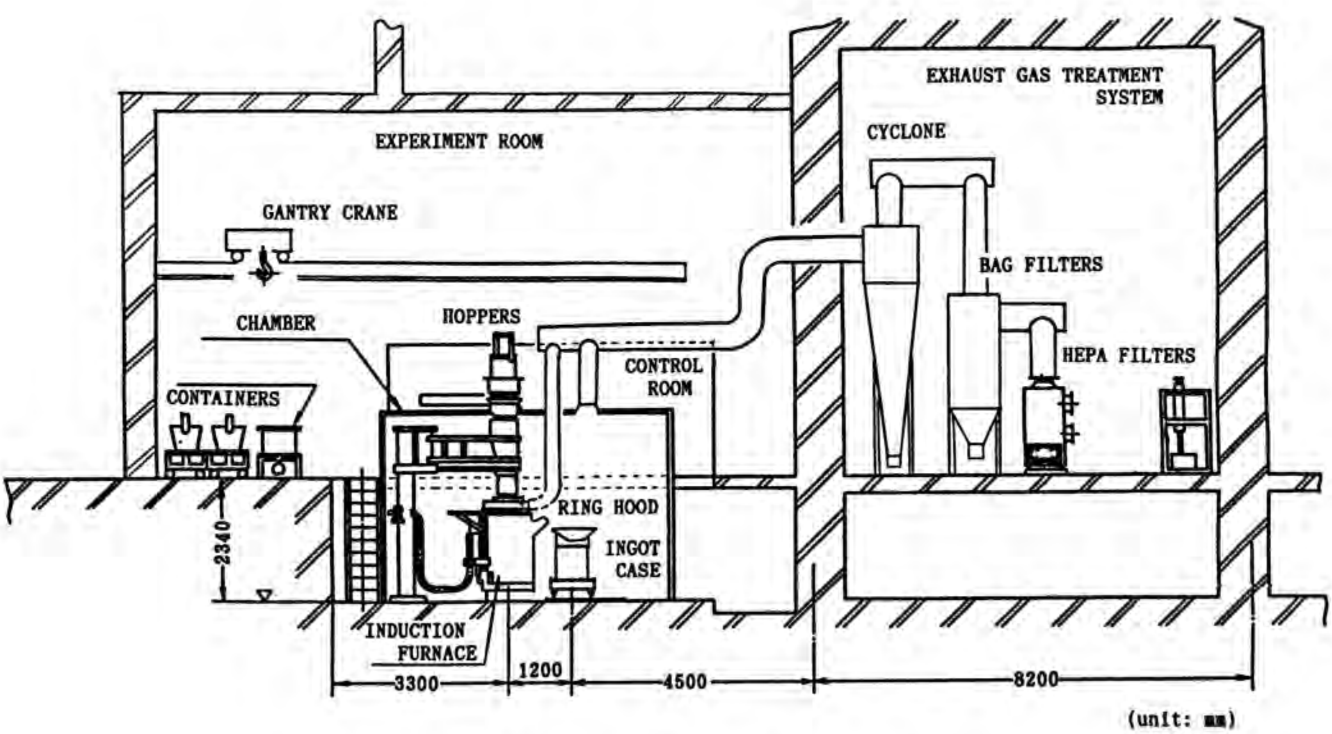


Fig. 6. Outline of the metal melting experimental facility.

be collected. One of the important parameter to be measured is a radionuclides concentration in slug because it determines the decontamination ratio by melting. Testing parameters and items assessed are summarized in Table I.

TABLE I

Test Parameters and Assessment Items in Melting Test with RI Tracers

Test Parameter

a. Material (carbon steel, stainless steel)
b. Radioactivity
c. Condition of radioactivity activated, contaminated)
d. Nature of flux
e. Meltin temperature

Assessment Item

a. Material movement from charges material to ingot, slag, off-gas, etc.
b. Movement of radionuclides to ingot, slag, off-gas, etc.
c. Radioactivity distribution within an ingot
d. Radiation dose rate in working place
e. Radioactive dust and aerosol in working place

Cold Test

Cold tests using non-radioactive metals were served as trial runs of the melting facility. Nine runs of cold tests have been completed by September 1990. Operational data on melting equipments (such as melting time, off-gas temperature, air flow balance, etc.) and the characteristics data of melted and casted steel were also collected. An understanding of the material balance and useful information on the characteristics of melting equipments were provided, such as;

- Oxidation of melted components easily progressed due to sufficient air supply by ring hood air extraction and air-curtain. This means careful control of additives and flux is necessary to get ingots of qualified level of contents.
- The spread of aerosol and splashed metals during casting process can be limited inside the chamber. This means the melting test with radioactive material will be able be performed in accordance with the practical regulation on RI treatment in Japan.

Generally speaking, it has been shown that test facility has sufficient capability to carry out coming tests using RI trace or JPDR dismantled components.

Tracer Test With RI

The evidence suggests that some metal wastes are contaminated with radionuclides which are produced both by the activation of the base metal and impurities and by fission products arising from the breakage of fuel rods. The aim of this test is to investigate the behavior of these radionuclides in the melting and solidifying processes.

This is difficult to examine using JPDR decommissioning materials because of its decreased radioactivity. For this reason, the amount of radionuclide movement from melting metal to solidified metal, furnace wall, slag and off-gas will be measured in commercially available metals coated with RI tracers that imitate the nuclides found in contaminated material of the light water reactor. The radionuclides to be used are ^{54}Mn, ^{60}Co, ^{65}Zn, ^{85}Sr, ^{137}Cs, etc. In addition, the radiation dose rates on the equipment surfaces and in the working environment will be measured to obtain radiation control data.

Actual Decontamination Test using Dismantled JPDR Components

Contaminated and activated metals such as piping, valves, and a piece of the pressure vessel from decommissioning the JPDR will be melted. By these melting tests, the safety of melting real radioactive metal will be demonstrated.

CONCLUSION

According to our preliminary study on the relevant system for the recycled radioactive materials, it is estimated that approximately 0.6 PWRs and 0.8 BWRs in average will be dismantled in Japan during the second decade of 21st century. The key to success in decommissioning a large commercial power reactor is to treat and dispose of large volumes of dismantled wastes. In the future, establishing better ways of treating and disposing of wastes will become increasingly important. In particular, the economics of treatment and disposal of wastes, and environmental protection from wastes, will be regarded as major problems. Therefore, we must find a way to solve these problems, from a long-term standpoint, irrespective of the types of wastes. One of the effective measures for reduction of amount and management cost is to establish the recycle system of the LLW.

JAERI has started research on the recycle of decommissioning materials. As described in this paper, metal melting and surface decontamination are complements of other each in establishing feasible recycling system of wastes metals from decommissioned reactors. The most effective combination of various decontamination methods and the melting in respect to the cost will be an item to be discussed hereafter.

REFERENCES

1. E. Tachikawa, et al.,"Research and Development on LWR system Decontamination, Mechanochemical and Redox Decontamination Method," Proc. 1988 JAIF Int.

Conf. on Water Chem. in Nucl. Power Plants, Vol.2, p.443, Tokyo(1988)

2. T.Suwa, N. Kuribayashi, E.Tachikawa, "Development of Chemical Decontamination Process with Sulfuric Acid-Cerium(IV) for Decommissioning. Single Stop Process to Dissolve chromium-rich Oxides," J.Nucl.Sci.Technol., 23, p.622(1986)
3. T. Suwa, N. Kuribayashi, E. Tachikawa,"Development of Chemical Decontamination Process with Sulfuric Acid-Cerium(IV) for Decommissioning. System Decontamination Process with Electrolytic Regeneration of Ce(IV) from Ce(III)," J.Nucl.Sci.Technol., 25,p.574 (1988)
4. M. Ishikawa, et al.,"Present Status of JPDR Decommissioning Program," Proc. 1987 Int. Decommissioning Symposium, Westinghouse Hanford Co., Vol.1, p.III-18(1987)
5. M. Tanaka, et al.,"The Japan Power Demonstration Reactor Decommissioning Program," Proc. Int. Conf. on Decommissioning of Major Radioactive Facilities, IMechE, p.25, London(1988)
6. H. Yasunaka, T.Kozaki,and T.Gorai,"Decontamination Technology for Decommissioning of Nuclear Facility," Proc. Residual Radioactivity and Recycling Criteria Workshop, USEPA/JAERI, p.64(1989)
7. M. Tanaka and H. Nakamura,"A Research Program on the Recycling of Decommissioning Material at JAERI," ibid., p.257(1989)
8. M. Tanaka and H. Nakamura,"Research Program of Decommissioning Material Reuse in JAERI," Proc. 1989 Joint Int. Waste Management Conf., ASME/JSME/AESJ, Vol.2, p.213(1989)