SAFETY EVALUATION FOR HULL WASTE TREATMENT PROCESS IN JNC

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ABSTRACT

Hull wastes and some scrapped equipment are typical radioactive wastes generated from reprocessing process in Tokai Reprocessing Plant (TRP). Because hulls are the wastes remained in the fuel shearing and dissolution, they contain high radioactivity. Japan Nuclear Cycle Development Institute (JNC) has started the project of Hull Waste Treatment Facility (HWTF) to treat these solid wastes using compaction and incineration methods since 1993. It is said that Zircaloy fines generated from compaction process might burn and explode intensely. Therefore explosive conditions of the fines generated in compaction process were measured. As these results, it was concluded that the fines generated from the compaction process were not hazardous material. This paper describes the outline of the treatment process of hulls and results of safety evaluation.

INTRODUCTION

Shared both ends of spent fuel are called 'end pieces'. The wastes remained in the fuel shearing and dissolution called 'hulls' (4-5 cm). Hull is the typical high radioactive waste from TRP. JNC has started to design the treatment facility for hull since 1993. Sharing cutter, incinerator and compactor will be installed in HWTF. JNC needed to evaluate topics of the safety for hulls treatment process. For example, Zircaloy fines generated from hull are easily oxidized material, it might cause dust explosion. Therefore JNC made examinations using Zircaloy fines to investigate ignition behavior and so on.
This paper describes results of safety evaluation and treatment process in HWTF.

PROCESS DESCRIPTION

At present, hulls and end pieces are stored in standard drums (350 L) with burnable, resistance-burnable waste, scrapped large equipment and water. Fig. 1 indicates a classification of the wastes in all standard drums. These wastes were called hull wastes. Filling the wastes to the standard drum is not efficient because the wastes are not compacted. It is anticipated that the storage facility will be filled with the wastes in future. Therefore, JNC has started to design the treatment facility of hull waste. Following two topics were mentioned to treat hull wastes.

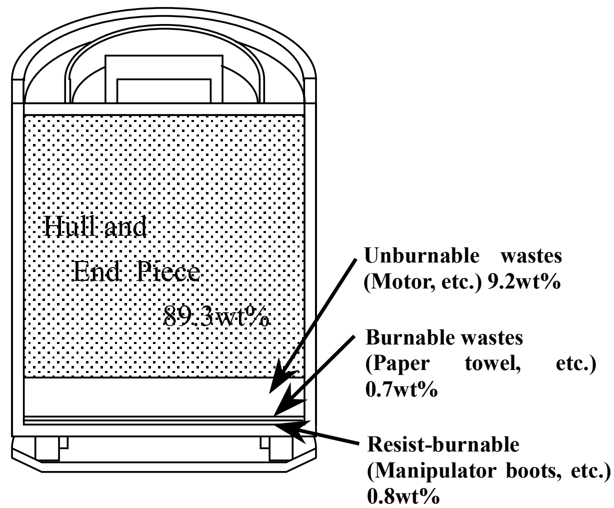

Fig. 1. Classification of the Waste in All Standard Drums in Tokai Works

The first topic is generation of hydrogen from hull wastes. Internal pressure of disposal canister would increase because hydrogen gas would be generated by radiolysis of burnable and resistance-burnable wastes. Therefore, burnable and resistance-burnable wastes, which generate hydrogen gas, must be removed from hull waste for the safety in the storage.

The second might fire and dust explosion of Zircaloy fines. Because the hull is made from zirconium alloy, fire and dust explosion might occur in the treatment process. Therefore burning behavior of Zircaloy fines need to be evaluated to prevent fire and dust explosion.

TREATMENT PROCESS IN HWTF (Hull Waste Treatment Facility)

In 1993 JNC started to design the treatment facility of high radioactive solid wastes. The facility is called Hull Waste Treatment Facility (HWTF). The process is shown in Fig. 2.

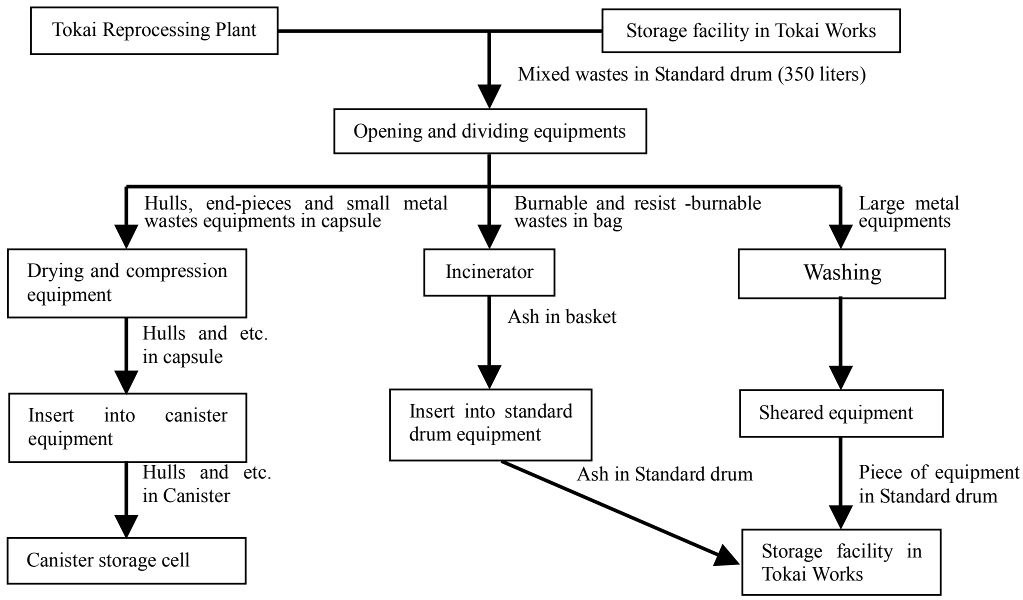

Fig. 2 Process Flow of HWTF (Hull Waste Treatment Facility)

Solid wastes in standard drums will be transported from TRP and Storage facility to HWTF. These wastes will be classified to burnable, resistance-burnable, large equipments and the other's wastes including hulls and end pieces at pretreatment room in HWTF. Classified large equipments will be sheared after water washing, filled into standard drums and transported to the storage facility. Burnable and resistance-burnable wastes will be put in the bags and incinerated after drying. Ash generated in incinerator will be filled into standard drums and transported to storage facility in the same way as large equipments. The other's wastes including hulls, end pieces and small metals, will be packed into the capsule (Diameter 400 mm, Height 500 mm) by about 50kg after drying and sealed with nitrogen (130torr). The capsules will be compacted by vertical direction press machine at 375 MPa and reduced to 1/5 volume. The compacted wastes will be packed into canister, which has same diameter as that used in Tokai Vitrification Facility. These canisters will be stored at the storage cell in HWTF.
Details and topics of each process are described as follows

Classification of wastes

Wastes transported to HWTF will be classified by two methods. Each method is described as follows.

-Removal by power manipulators

At first, large equipments and end piece in standard drums will be removed on separation table by power manipulators. Removed large equipments will be sent to the shearing process. End pieces will be filled in the capsule with hulls and sent to the compaction process.

-Division by specific gravity

A classification machine of waste is shown in Fig. 3. This machine is consisted of a water pool and a vessel (trommel). Burnable and resistance-burnable wastes adhering to the hulls will be removed by high-pressure water splaying in the trommel. Hulls, metals, burnable and resistance-burnable wastes discharged was designed to have the vessel will be dropped in the pool by a water stream as Fig. 3. The pool forms the double bottom structure. Wastes will be sent to the downstream by an oscillation conveyer on the upper bottom. Burnable and resistance-burnable wastes adhering to the hulls will be removed by ultrasonic oscillation units mounted in center of oscillation conveyer.

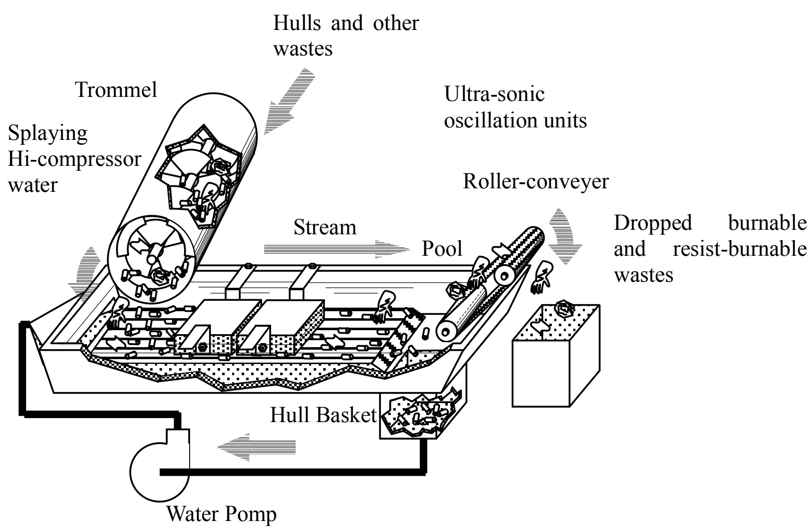

Fig. 3. Separator Unit

Burnable and resistance-burnable wastes will be floating in the pool because the specific gravity is low. These wastes will be recovered in a basket by a roller-conveyer. Then, they will be put in bags and incinerated. On the other hand, hulls and metal wastes whose specific gravities are high will flow in the lower part of the pool and recovered in other basket. They will be filled in the compaction capsule with the end piece removed and transported to the cell at first for the compaction treatment.

Hydrogen gas might be generated by radiolysis of organic materials with the hull. The generated gas might raise the pressure of the canister during storage. In addition, it might be possible that generated gas explode. Therefore, amount of burnable and resistance-burnable wastes adhering to the hulls in this process must be controlled.

Hull treatment process

-Drying and sealing of hull

Hulls, end pieces and metal wastes in the capsule will be dried in the chamber filled with nitrogen (130 torr) at 140 degree centigrade. After drying, capsule will be capped, welded by Tungsten Inert Gas (TIG welding) and transported to the compaction process.

In this process, temperature and oxygen concentration must be investigated so that hulls including Zircaloy fines cannot ignite.

-Compaction of capsule

A compaction machine of hulls, end pieces and metal wastes in the capsule is shown in Fig. 4. The machine will be composed of a main frame and a sub frame. A ram will be mounted with the sub frame. The capsule will be compacted by moving the sub frame down. The nitrogen will be enclosed in a mold. Hulls and end pieces in the capsule will be inserted to the mold enclosed with nitrogen. Afterward, the capsule will be compacted and ejected from the mold. When operation-oil leaks, the oil will not flow to the compaction cell because the oil pressure piston will be located in the lower floor.

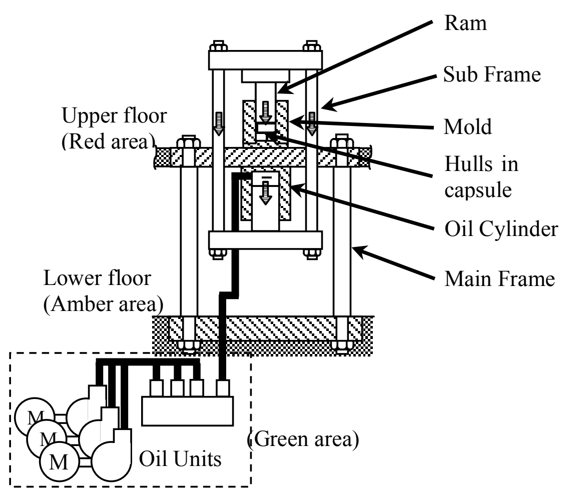

Fig. 4. Hull Compaction Machine

In this process, it is necessary to investigate the oxygen concentration when the generated fines ignite during compaction.

-Inserting in Canister and Storage

Ten compacted capsules will be inserted in one canister. Afterward, the canister will be capped and welded. The canister will be stored at the cell in HWTF where the air will be vented for cooling of capsules. In this process, it is necessary to investigate the oxygen concentration and temperature when the Zircaloy fines ignite during storage.

Incineration for burnable and resistance-burnable wastes

Burnable and resistance-burnable wastes transported from the division process in the bag will be incinerated. Wastes will be incinerated by feeding the high-temperature air. Generated ash will be taken from the bottom of the incinerator. The ash will be packed in standard drums and stored in the storage facility.

The fines generated in the fuel shearing process, might adhere on burnable and resistance-burnable wastes. In this process, the effect to the incinerator of rapid temperature increasing by oxidation heat of Zircaloy fines is necessary to investigate.

Shearing of large equipments

Transported large equipments the in removal process will be sheared by three devices. The thin equipments will be sheared by shearing-cutter. The thick equipments will be sheared by hacksaw. And unnecessary standard drums will be sheared by YAG laser (Yttrium Aluminum Garnet laser). These sheared and scrapped equipments will be packed in standard drums and transported to the storage facility.

RESEARCH AND DEVELOPMENT FOR HWTF

Compaction

JNC carried out compacting examination to observe inner compacted capsule and evaluate the relation between compaction force and volume reduction. The capsule (diameter 400 mm, height 500 mm), hulls and end pieces in the capsule were simulated in size and material of the real waste. Simulated hull was made from annealed zirconium alloy and cut by 40mm. Simulated end piece was made from stainless steel. These capsules were compacted at various pressures by the vertical direction press machine. The compacted capsule was cut in axial direction to be observed. As a result, large space in the capsule was not found. Compacted capsule was dropped from 3m high. As a result, the capsule did not change its shape. It is thought that, the capsule was difficult to change its shape in case of adding external force by 375 MPa.

Compaction force and change of height of the compacted capsule were measured. As a result, volume reduction rate was calculated. These results are shown in Fig. 5. The capsule was reduced to about 20 % volume at 375 MPa compaction. Judging from tendency in this Fig. 5, it is thought that the change of volume reduction will be quite small when the compacting force is over 375 MPa. For the above reason, hulls and end pieces in the capsule will be compacted at 375 MPa in HWTF.

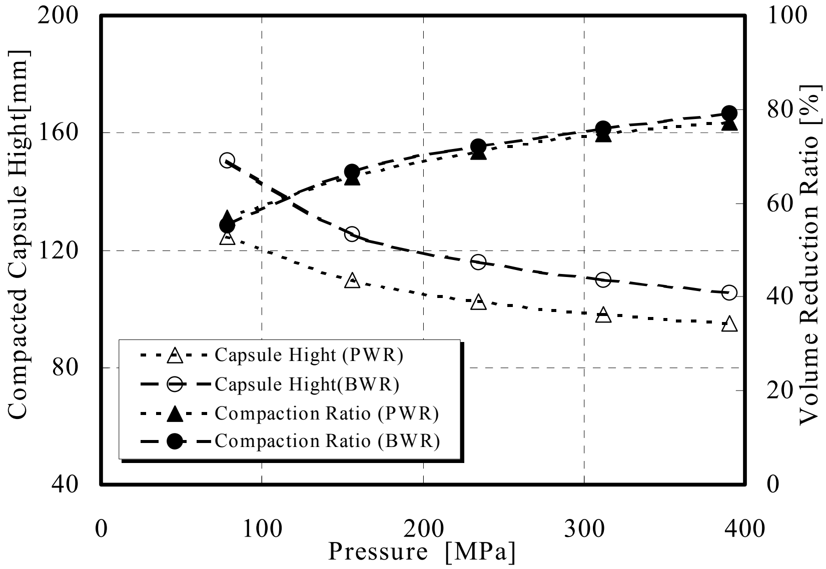

Fig. 5. Result of Compacting examination

The fines were observed during compaction. Examination about amount of generated the fines was carried out using real hull. Spent fuels of 30-40 GWD/t were cut to 40 mm, and meats (UO_2) of these pieces were removed from spent fuels by mechanical method. Four pieces of hulls were compacted at 375 MPa. As a result, amount of generated fines was 10 g per 1 kg hull (1 wt%). It was analyzed that these fines were almost metal oxides. The average particle size of these fines was 50-70 μm.

Zircaloy fines in the mold might be possible to explode because the Zircaloy fines were exposed in the air. As the result of above examination, simulated hull, which is easy to generate Zircaloy fines was produced, and remained amount of Zircaloy fines in the mold was evaluated. The capsule was included with hulls and fines by a weight ratio of about 100:1 and compacted at 375 MPa. It was found that about 50 ppb Zircaloy fines adhered to the compaction mold.

SAFETY EVALUATION FOR ZIRCALOY FINES

Minimum Ignition Temperature, Minimum Ignition Energy, Limiting Oxygen Concentration and Maximum Explosion Pressure of the Zircaloy fines were measured to prevent the fire and dust explosion of the Zircaloy fines under treatment of hull wastes because it is considered that the Zircaloy fines works as chemically active dust in dust explosion. These tests were assumed of the behavior in the storage and incineration. All tests except for the analysis of dust property were performed without radioactive materials.

Measurements for minimum ignition temperature

Minimum ignition temperature of dispersed dust was measured using Godbert-Greenwald equipment based on IEC (international electrotechnical commission) 1241-2-1 method B. In this method, the dust is dispersed in a glass tube (diameter 44 mm, height 216 mm) using high compressed air. After the dust is kept in constant temperature and oxygen concentration, visual observation will be made to judge if the ignition occurs. Fig. 6 indicates the results of minimum ignition temperature. Oxygen concentration and minimum ignition temperature are plotted in Fig. 6. As a result, ignition temperature was 110 degree centigrade in oxygen concentration of 15 vol%, and very high ignition temperature was measured at oxygen concentration of 12 vol%. When the oxygen concentration was kept under 10 %, no ignition was observed. Also the ignition was observed at oxygen concentration of 24 vol% and air temperature of 20 degree centigrade. These results indicated that the control of the oxygen concentration is very important for prevention of dust explosion.

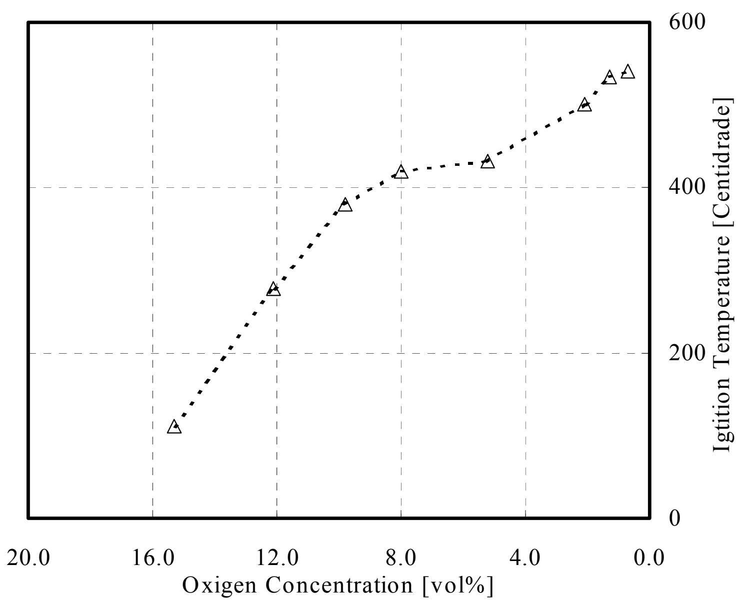

Fig. 6. Measurements for Minimum Ignition Temperature

Measurements for minimum ignition energy

Minimum ignition energy of dispersed dust was measured using Hartmann method based on IEC 1241-2-3 method A. Dust is dispersed in a glass tube (diameter 312 mm, height 70 mm) by compressed air, it is ignited by the electronic igniter which can vary the ignition energy. Ignition is observed visually 5 times at the same condition. Test results are indicated in Table I. From these results, the Zircaloy fines were ignited by the ignition energy under 1 mJ in the air. This result indicates that it is difficult to prevent the ignition, because the Zircaloy fines easily ignited by very small energy.

Table I. Hartman Test Results

Ignition Energy [mJ]	Dust Concentration [mg]	Ignited Number
30	300	4
10	300	5
10	600	5
3	600	1
1	600	5
1	900	1
1	1200	1

At atmosphere

Measurements for maximum explosion pressure and limiting oxygen concentration

Maximum explosion pressure and limiting oxygen concentration were measured by 30 liters of spherical chamber for testing dust explosion. In this chamber, after oxygen concentration was controlled and dust was dispersed using compressed air, dusts were ignited by the explosives which generates explosion of 7.5 kJ, and explosive pressure was measured. Measured maximum pressure is indicated in Fig. 7-a and Fig. 7-b. It is thought that explosive pressure was reached to 0.8 MPa in the air, because the maximum pressure became 0.6 MPa in oxygen concentration over 4.0 vol%. These figures indicate that the flame propagation is not kept stably due to the pressure increasing under oxygen concentration of 2.0 vol%. Based on this pressure tendency, the maximum pressure increase rate (dP/dt) was obtained. This rate reached almost 25 MPa.m/s under the oxygen concentration of 6.0 vol%, whose rate was agreed with that for magnesium in the air. This result and previous results indicate that the oxygen concentration control is very effective method to prevent the fire and explosion. It is important that the oxygen concentration is controlled under 2.0 vol%.

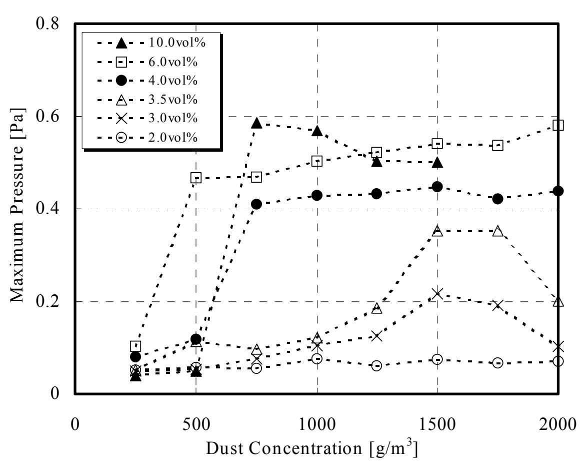

Fig. 7-a

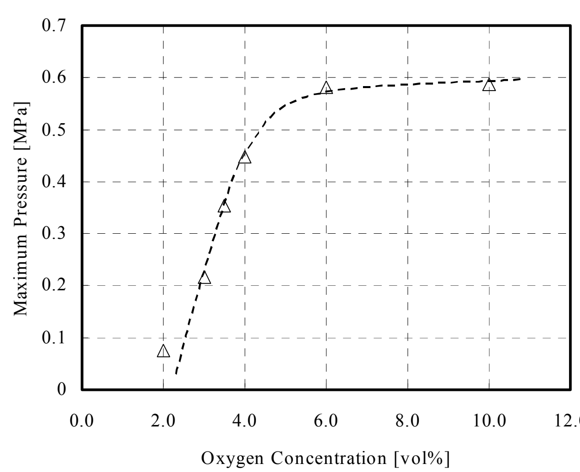

Fig. 7-b

Fig. 7. Maximum Explosion Pressure And Minimum Explosion Oxygen concentration

SAFETY EVALUATION ON TREATMENT PROCESS IN HWTF

Combustion test of Zircaloy fines

Minimum ignition temperature of the accumulated dust was measured using the equipment based on IEC 1241-2-1 method A. After the dust whose thickness of layer is 5,10,15 and 20 mm is set circularly on the hot plate, the ignition of Zircaloy fines is observed. This result is shown in Fig. 8. It was observed that the minimum ignition temperature decrease as the depth of dust layer became thick. It is impossible that the fire occurs in the process without the heat source, because the ignition was observed at the layer of 20 mm thick and the temperature of 233 degree centigrade.

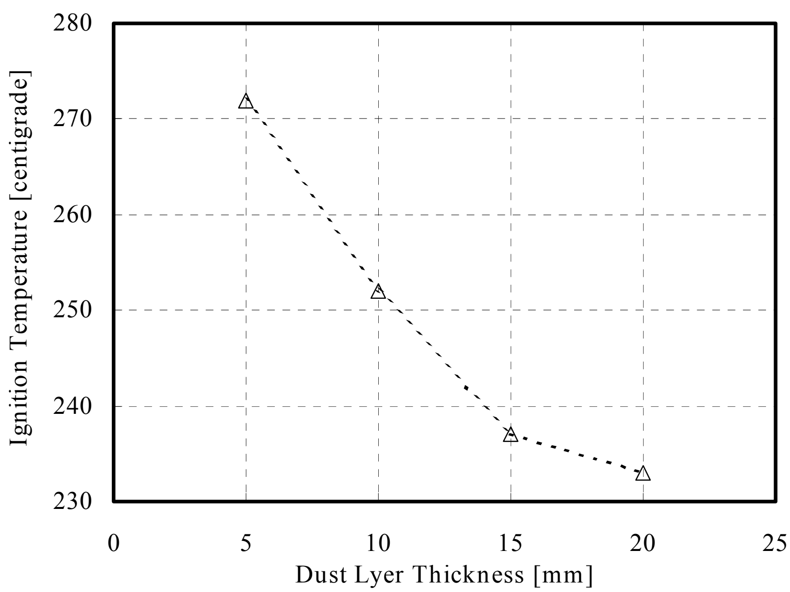

Fig. 8. Results of Combustion test

Incineration tests for burnable and resistance-burnable waste mixed with Zircaloy fines

It is thought that the incinerator was damaged by the heat generation of the Zircaloy fines that was mixed in the burnable and resistance-burnable waste. In the examination, small amount of the Zircaloy fines (1.5-15.0 g) were mixed with clothe, polyethylene and polyvinyl chloride (almost 15.0 kg), which are simulated burnable and resistance-burnable waste. The mixed waste was burnt in the incinerator. As a result of temperature measurement of the exhaust gas from this incinerator, the exhaust gas temperature from non-Zircaloy fines waste (containing no fine) was reached 355-705 degree centigrade, and that from mixed waste (containing small amount of fines) was 365-694 degree centigrade. There were no differences between each simulated waste. Abnormal temperature increasing and structural deformation of the reactor walls were not observed either.

Temperature control for capsule

There might be a risk of fire and explosion, if we could not control the cooling equipment for the compacted capsules. The behavior of the Zircaloy fines in the compacted capsules was observed, assuming some troubles in cooling equipment. Compaction capsules were filled with mixed 1 kg hulls and 115 g Zircaloy fines. This capsule was compacted until the diameter was 102 mm and height was 36 mm.

These capsules were set on the high temperature circumstances, which was kept 100-500 degree centigrade, and temperature of the inside and outside of the capsule were measured. Pressure around the capsule was also measured. These results are indicated in Fig. 9. In this figure, temperature and pressure around the capsule are plotted. Rapid change of the temperature and pressure were not observed. From this result, it is considered that continuous large heat source did not exist. Observed temperature slowly changed at the inside of the compacted capsule. In addition, it was analyzed that the fines after this examination were almost oxidized. Therefore, it may be confirmed Zircaloy fines filled in the compacted hulls were being oxidized slowly.

Even if the exothermic reaction of the Zircaloy fines occurs, this reaction must not be kept continuously, because it is considered that heat is transported from the Zircaloy fines to the hull. These results indicate that the compacted hulls should not cause fire and explosion, even if these include the Zircaloy fines.

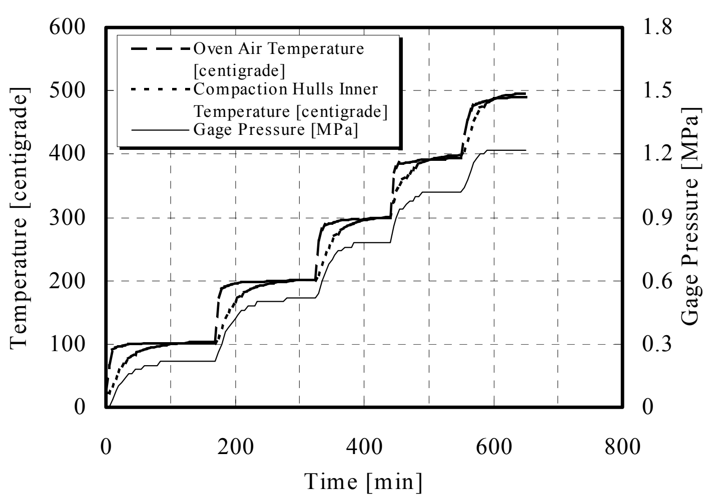

Fig. 9. Results of Temperature Control for Capsule

Thermal conductivity of compacted capsule

Hull generates decay heat. The concrete is needed to be controlled of the temperature at less than 65 degrees centigrade, because the concrete becomes brittleness at more than this temperature.

Therefore, storage cell made from concrete in HWTF needs to be cooled. Quantity of cooling air for temperature control in the cell needed to be calculated. Thermal conductivity of compacted capsule was measured and calculated.

High and low temperature plate were attached on top and bottom of compacted capsule. Temperature of top and bottom of the capsule was measured when temperature became constant. The thermal conductivity was leaded from those temperatures difference.

Thermal conductivity was calculated in same condition by FEM (Finite Elements Method). Those results are shown Table II. Table II indicates that the calculation results were agreed with the measurement results. The calculation method using FEM can be applied for evaluation of thermal conductivity in case that the treated condition and the type of fuel assemblies are changed. The thermal calculation of the cell in HWTF was well conducted with this thermal conductivity.

Table II. Thermal Conduction of Compaction Hulls

Thermal Conduction W/mK	Calculation		Measurement	
	Axle direction	Radial direction	Axle direction	Radial direction
PWR Type	0.92	3.80	0.74	Cannot Measured
BWR Type	0.85	3.70	0.87	
FBR Type	0.90	1.70	0.89	

CONCLUSION

JNC designed the hull treatment facility. Then, examinations were carried out for many points. It was found from these results that fines were generated from hulls by compaction. The fines were mainly metal oxide. Generally, it is said that brittleness of fuel assembly is varied with burn up, reactor type, location of reactor and etc. We carried out many examinations and research for safety of HWTF process. It was found that explosion did not occur under 2.0 vol% of oxygen in these results. We confirmed to safety of HWTF process from these results. These results had been used for the safety evaluation of HWTF. We are convinced that the process of HWTF can provide prevention methods against fire and dust explosion of the fine generated from sharing or compaction. We need to have some effort to confirm the integrity of mitigation methods to fire and dust explosion.

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