

Chapter 7

Safety Decontamination System for Combustion of Forestry Wastes

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Abstract The safety decontamination system of the contaminated forestry wastes by combustion was developed. Under the laboratory scale test with 10 g of cedar bark, about 35 % of radiocesium in the contaminated bark flowed out as a gaseous state by the combustion above 500 °C. The developed system consisted of three modules, the smoke extraction apparatus by water, the combustion ash coagulate apparatus and the radiocesium filtration unit from the sewage water. The demonstration combustion tests were carried out in March 2012. Forestry wastes (6.3 kg), pine needles, Japanese cedar bark and sapwood chips including radiocesium were combusted at 550–700 °C. The exhaust smoke was washed by the jet stream of water, the sewage water included small amount of soot and the radiocesium concentration of sewage water without soot was 50 Bq/kg. After the filtration of 550 L of sewage water by the radiocesium absorption filter consisting of wool dyed by Prussian blue, the radiocesium concentration decreased less than 0.2 Bq/kg. The filtrated water was recyclable in this system. No gaseous radiocesium was detected in the exhausted air from this system during the decontamination of forestry wastes. The combustion ash (140 g), consisting of cesium oxide alloy including various metal ions, was collected and packed under the reduced pressure automatically.

Keywords Forestry wastes • Combustion • Smoke extraction • Radiocesium • Decontamination • Sewage water • Cedar • Bark • Pine needles • Ash

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7.1 Introduction

Although 4 years have passed after the accident of Tokyo Electric Power Co. (TEPCO) Fukushima Daiichi Nuclear Power Station (FDPS), a huge amount of radionuclide remains in the forest at Fukushima [1]. The main components of radionuclide from FDPS disaster, cesium134 (^{134}Cs) and cesium137 (^{137}Cs), became the environmental contamination problem and disturbed both agricultural and forestry works in Fukushima [2, 3]. Most radiocesium fallout on the ground tightly bound with soil and formed the complex with clay minerals [4], the ionic ^{134}Cs and ^{137}Cs were a few in the environment [5, 6].

Fukushima Daiichi Nuclear Power Station accident induced the serious situation to revive the forestry works in Fukushima, especially in Abukuma forest area located at west from FDPS with heavy radioactive contamination. The regular maintenance of forest became difficult due to the high air dose environment, the low demand of woods and the forestry wastes including radionuclide. The fallout radiocesium (^{134}Cs and ^{137}Cs) existed on bark and leaves of trees [7–9], and the Japanese cedar woods absorbed radiocesium through bark and leaves in 2011 and 2012 [10, 11].

In Fukushima prefecture, about 970,000 ha of forest is the subject area for the periodic thinning from 2013 to 2030 in the current plan. For 1000 ha of the thinning area in forest, about 10 % of woods corresponding to 150,000 m³ of Japanese cedar will be cut down. The thinning of 1000 ha of forest will produce about 30,000 m³ of twigs including leaves and about 15,000 m³ of barks. The total forestry wastes produced by the periodic thinning is estimated at 3,445,000,000 m³ for each year. Before FDPS accident, the forestry wastes were used as composts for forestry and agriculture uses. The forestry wastes including radiocesium or other radionuclide have no way for utilization. These forestry wastes are disposable by the combustion or the fermentation as the energy resource.

One of the possible ways of forestry waste disposal is the biomass power generation. Ministry of the Environment planed the construction of two biomass power generations with 12 MW at Samekawa village and Hanawa town in 2013. The benefit of biomass power generation is the effective utilization of forestry waste and the power generation; on the other hand, the release of gaseous radionuclide and the mass transportation of biomass contaminated with radionuclide from the forest to the power plant are the risk of biomass power generation in Fukushima at present. The construction of biomass power generation (10 MW scale) in Hanawa town was canceled in October 2013 by the local residents' campaign based on the uncleanness of these risks [12]. However, Fukushima prefecture decided to construct again the biomass power generation in Hanawa town in May 2015 [13]. The insecurities of large-scale biomass power station are the leak of radionuclide from the discharge gas during the burning of forestry wastes, the fly out risk of the concentrated ash and the radiation exposure of workers in the biomass power plant.

In this study, the risk of radiocesium released in the environment during the combustion of forestry waste was estimated by the laboratory-scale combustion experiment. The demonstration combustion test of forestry wastes was carried to

evaluate the possibility of safety combustion method. The safety combustion system designed based on the results of laboratory-scale experiments was developed for the decontamination of forestry wastes.

7.2 Experimental

7.2.1 Samples

Samples used in this study were Japanese cedar bark (from 5000 to 21,000 Bq/kg), cedar leaves (from 5000 to 20,000 Bq/kg), cedar sapwood (less than 20 Bq/kg), cedar heartwood (less than 10 Bq/kg) and pine needles (from 10,000 to 25,000 Bq/kg) collected at Fukushima Forestry Research Center in autumn 2011. After drying at 60 °C for 3 days, samples were used for the experiments. The water content of samples evaluated from the mass loss after drying was 28.6 % for cedar bark, 56.8 % for cedar leaves, 61.8 % for sapwood, 54.3 % for heartwood and 10 % for pine needle based on the wet mass.

7.2.2 Combustion Test

Laboratory-scale combustion tests of radio cesium-contaminated Japanese cedar bark (20,000 Bq/kg) were carried out using the combustion instrument made of glass shown in Fig. 7.1. About 10 g of cedar bark in alumina crucible (A) was put in the quartz sample tube (B) and combusted at 300–900 °C under air flow condition. The combustion gas was cooled at the glass tube (C) by ice/water bath and was collected

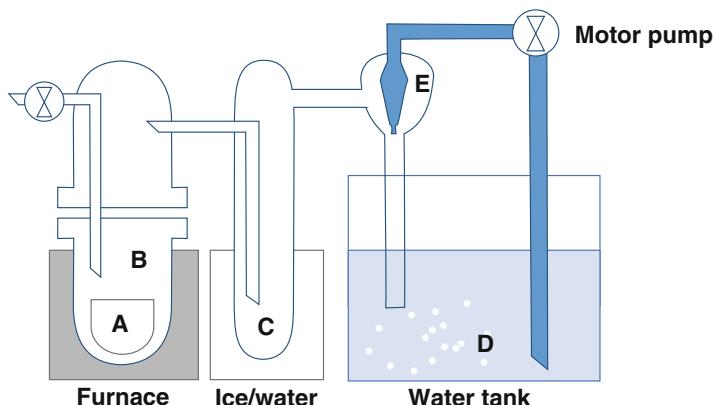


Fig. 7.1 Schematic drowning of combustion instrument for laboratory scale test

in 2 L of water sink (D) using an aspirator (E). Before and after the combustion, the radioactivity of sample and water in sink D were determined. The mass loss by the combustion was also determined.

Demonstration tests of forest waste were carried out at Fukushima Forestry Research Center in March 2012. Two times demonstration tests using 2.3 kg of pine needles, 3.6 kg of Japanese cedar bark and 0.4 kg of cedar sapwood were carried out by the combustion system described later.

7.2.3 Measurements

Radioactivity of samples was measured by the Germanium semiconductor detector (SEG-EMS: SEIKO EG&G Inc., Japan) with 100 mL of U8 container at 10,000–40,000 s of accumulation time to obtain 1–2 Bq/kg of the lower limit. The radiation decay was compensated at March 1, 2012.

Thermogravimetry measurements of forest wastes, Japanese cedar sapwood chip, cedar bark, cedar leaves and pine needles were carried out by TG/DTA 7200 (Hitachi High-Tech Science Co., Japan) under air and nitrogen gas flow atmospheres. About 3 mg of samples were heated at 20 K/min from room temperature to 900 °C. The evolved gas during combustion was analyzed by the online Fourier Transfer Infrared Spectroscopy FTIR 650 (JASCO, Japan) connected with TG/DTA [14].

Transmission electron microscopic observation of ash was performed by JEM 3200FS (JEOL, Japan) operated at 300 kV. The ash sample was dispersed on the copper glide and coated with carbon. The energy-dispersive X-ray spectroscopic analysis (EDX) was carried out for ash sample. Powder X-ray diffraction of ash was observed by Rint TTR III (Rigaku Co., Japan) operated at 50 kV and 300 mA with $2\theta/\theta$ scanning at 2°/min.

7.3 Results and Discussion

7.3.1 Laboratory-Scale Tests

Mass loss curves of Japanese cedar sapwood chip, bark, leaves and pine needles obtained by TG/DTA were shown in Fig. 7.2 in air flow (left) and nitrogen gas flow (right) atmospheres. The mass loss occurred in three steps below 200 °C, 200–400 °C and above 400 °C in both conditions; however, the third mass loss in air atmosphere occurred in the narrow temperature range; in contrast, the third step in nitrogen atmosphere occurred in the wide temperature range. The first mass loss below 200 °C was caused by the evaporation of water and volatile components without the combustion of main components of forestry waste, such as cellulose,

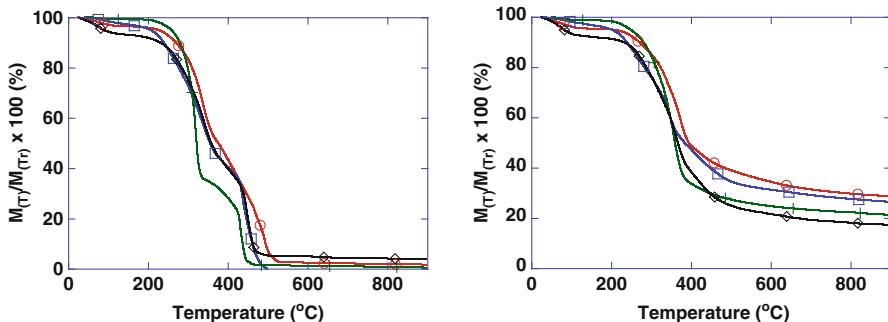


Fig. 7.2 Mass change of Japanese cedar sapwood chip (cross), cedar bark (circle), cedar leaves (square) and pine needles (rhombus) under air (left) and nitrogen gas (right) gas atmosphere

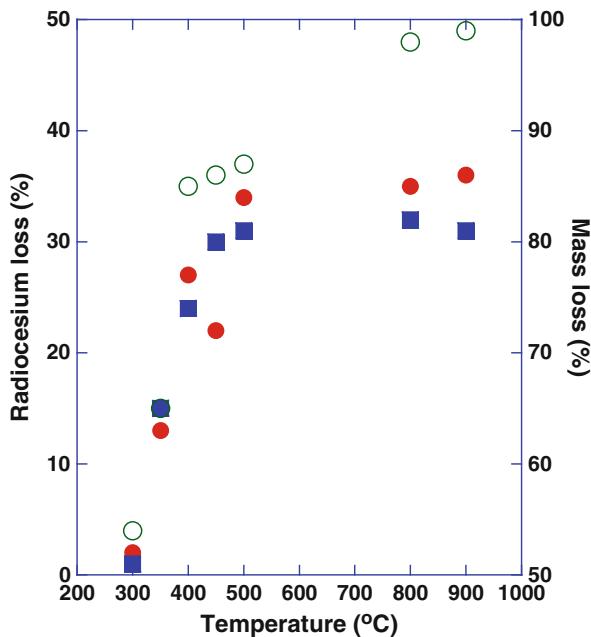
lignin and hemicellulose. The main component of mass loss below 150 °C was water from the evolved gas analysis. The second mass loss occurred above 250 °C and levelled off below 400 °C for both conditions; these mass losses were caused by thermal decomposition of cellulose and hemicellulose. The major components evolved in this stage were CO₂ and CH₄ in air atmosphere and CO and CH₃OH in nitrogen atmosphere. The third mass loss stage was caused by the dehydration of samples; the major components in this stage were CO₂, CH₄ and phenolic derivatives for air atmosphere and CO and H₂ for nitrogen atmosphere which were almost similar to the compounds obtained by the pyrolysis of wood [15].

The residues of each sample at 800 °C and 890 °C were 0.8 % and 0.6 % for Japanese cedar sapwood chip, 1.9 % and 1.7 % for bark, 0.5 % and 0.4 % for leaves, 4.2 % and 4.0 % for pine needles in air atmosphere and 22.4 % and 21.4 % for sapwood chip, 29.8 % and 28.9 % for bark, 27.7 % and 26.6 % for leaves and 18.2 % and 17.3 % for pine needles in nitrogen atmosphere, respectively. The residues obtained in air and nitrogen atmospheres were inorganic materials and char, respectively, by XRD analysis of residues.

The mass loss and the radio cesium loss, normalized by the initial mass and the initial concentration of ¹³⁴Cs and ¹³⁷Cs, during the combustion of cedar bark at various temperatures were shown in Fig. 7.3. Under the laboratory scale combustion test, the mass loss at the combustion temperature below 500 °C was less than 90 %, which was lower than the mass loss evaluated by TG measurement under air flow condition. However, the mass loss behavior occurred in three steps, which was consistent with TG behavior shown in Fig. 7.2.

For the combustion at 300 °C, most radio cesium (98 %) remained in the residues, which was 46 % of the initial mass; however, radio cesium in the residues decreased with the increase of combustion temperature. About 15–25 % of radio cesium evaporated by the combustion at temperature from 350 to 450 °C and about 30–35 % of radio cesium released by the combustion at temperature above 500 °C. After seven combustion tests at temperatures from 300 to 900 °C, the radio cesium concentration (¹³⁴Cs + ¹³⁷Cs) of trapped water was 153 Bq/L, which corresponded

Fig. 7.3 Mass loss (open circle) and radiocesium loss of ^{134}Cs (closed circle) and ^{137}Cs (closed square) at various combustion temperature



to 98 % of radiocesium evaporated during the combustion tests. These results indicated radiocesium in forestry wastes vaporized by the combustion, and the evaporated radiocesium was trapped efficiently by water.

7.3.2 Development of Decontamination System for Forestry Waste

In Fukushima, the non-industrial wastes including contaminated materials are burned at about 900 °C in the waste incineration plant equipped with the smoke extraction apparatus such as the desulfurization equipment and the bag filter for fly ash. The desulfurization of exhaust gas is processed generally by the absorption of sulfuric acid gas (SO_2) with aqueous dispersion of limestone (CaCO_3), which has the possibility to trap the evaporated radiocesium; however, the desulfurization equipment is not designed for the radiocesium absorption. Fly ashes are filtrated by the bag filter made of poly(ethylene tetrafluoride) with 0.4 μm of mesh size.

The safety decontamination system for forestry wastes was designed as shown in Fig. 7.4 [16]. The design criteria of this system were the compact size and the flexible attachment to connect with the existing small combustion furnace (100 kW scale) in sawmill and wood market yard. The developed system consisted of three modules, the smoke extraction apparatus by water (I), the combustion ash coagulate apparatus (II) and the radiocesium filtration unit from the sewage

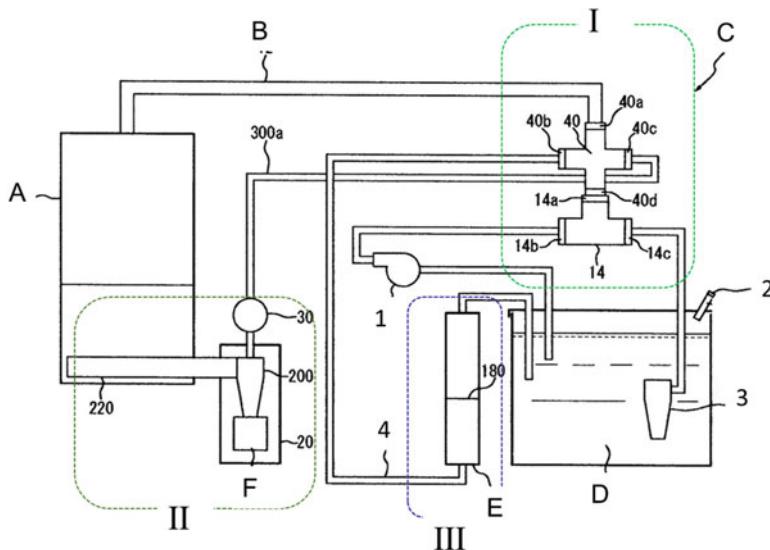


Fig. 7.4 Design of combustion system consisted of furnace (A), smoke tube (B), water scrubber absorbed smoke (C), water tank (D), sewage water filtrate column (E) and ash collecting apparatus (F)

water (III). The furnace (A) connected with the smoke extraction apparatus by jet stream of water (C) through the smoke tube (B), water supplied from the water tank (D), the combustion ash was collected by the ash enclosure apparatus (F) and the radiocesium in the sewage water was filtrated by the absorption column unit (E).

The woodstove model 1630CB (Morsø, Finland) connected with the water scrubber (14) through the smoke tube and three channel connector (40). About 600 L of water stored in water tank (D) pumped up and compressed by motor (1), and water jet (2 L/s) flowed into the scrubber from 14b and flowed out from 14c induced the reduced pressure at 14a. The smoke from the woodstove was absorbed in the scrubber from 14a and mixed with water jet in the scrubber. The mixed smoke and water flowed in the water tank through the wet cyclone (3) where the radiocesium in smoke dissolved in water and the excess pressured air flowed out through the air filter made of wool dyed by Prussian blue (2).

After the combustion, the water-extracted smoke was decontaminated by radio cesium absorption filter made of wool dyed by Prussian blue (18) through the column (E) absorbed by the scrubber through three mouth connector (40b). The combustion ash including radiocesium was collected and separated particles by air cyclone (200), and the separated ash particles were packed automatically under the reduced pressure by the ash enclosure (F: ATA Co, Japan) connected with the scrubber through three mouth connector (40c).

The outlook of combustion system was shown in Fig. 7.5. The smoke extraction apparatus (1) consisted of the water tank (A), the water motor (B), the scrubber (C)

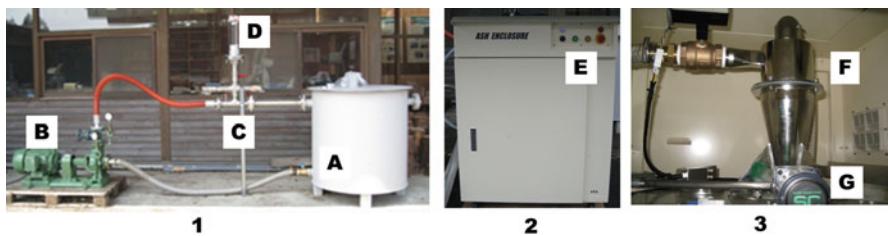


Fig. 7.5 Photographs of the smoke extraction system (1), the ash enclosure instrument (2) and its inside view (3)

and the three mouth connection to furnace (D) and the front view of ash enclosure (2) with control panel (E) and its inside consisted of air cyclone (F) and ash packing instrument (G) were shown in Fig. 7.5.

7.3.3 Demonstration Test of Combustion for Forestry Wastes

The decontamination experiments were carried out at Fukushima Forestry Research Center in March 2014. Two combustion tests using 2.3 kg of pine needles with 1.0 kg of cedar bark and 2.5 kg cedar bark with 0.4 kg of cedar sapwood were burned with slow temperature increment from 400 to 700 °C for 1 h. The mass and radiocesium concentration in forestry wastes used for the combustion tests were listed on Table 7.1. About 550 L of water was used for the smoke extraction. During the demonstration tests, the surrounding temperature was about 4–6 °C; the initial temperature of water was about 5 °C and became 45 °C after the combustion tests. After the combustion test, about 5 L of water was lost by evaporation. From these values, the heat transfer from smoke to water was 2.9 kWh, which corresponded to 200 kWh scale combustion furnace, such as wood chip boiler and incinerator. No radiocesium was detected (the lower limit of 0.2 Bq/kg) in the air filter where the exhausted steam passed from this system during the decontamination.

Immediately after the combustion tests, the clear water changed the black soot-suspended water with pH 7.2. The sewage water absorbed the acidic compound caused by the decomposition of wood components and the soot. The suspended soot in sewage absorbed on the surface of water tank overnight; the optical clear and pale yellow sewage water was obtained. The smoke tube with 5 m length took off from the system and separated to pieces; the soot absorbed in smoke tube was washed by 100 L of water. The sewage water (50 Bq/L, about 545 L) and the soot-washed water (soot suspended water, 60 Bq/L, about 100 L) were decontaminated by the filter system with 300 g of Prussian blue-dyed wool filter. After 1 h filtration, no radiocesium was detected by Ge semiconductor detector with 0.2 Bq/L of the lower limit. The decontaminated water was recyclable in this system.

Table 7.1 Mass and radiocesium concentration of materials used for the combustion test and obtained after the combustion

Raw materials of combustion			Collected materials after combustion		
Materials	$^{134}\text{Cs} + ^{137}\text{Cs}$	Mass	Materials	$^{134}\text{Cs} + ^{137}\text{Cs}$	Mass
	Bq/kg	kg		Bq/kg	kg
Pine needle ^a	22,000	2.3	Ash	440,000	0.14
Cedar bark ^a	10,800	1.0	Soot in water ^b	290,000	>0.0015
Cedar bark ^c	15,000	2.5	Soot in flue ^d	60	100
Sapwood ^c	50	0.4	Sewage water ^e	50	545

^aMaterials used for the first combustion test

^bSoot suspended in sewage water absorbed on water tank, collected from the water tank surface

^cMaterials used for the second combustion test

^dSoot absorbed in smoke tube washed by 100 L of water

^eSewage water after filtration of suspended soot

The ash was collected by the enclosure under the reduced pressure and was packed and sealed automatically in the antistatic polyethylene bag under vacuum. After sealing the ash, the package of ash was easily handled without further contamination. The mass and radiocesium concentration of ash, sewage water, separated soot and washed soot from the smoke tube were shown in Table 7.1. The ratio of mass of ash against the raw materials was 22.2 %, which indicated that the combustion proceeded in imperfect condition. The soot suspended in sewage water and absorbed in smoke tube also suggested the imperfect combustion. The collected ash included white powder and black charcoal particles caused by the imperfect combustion.

The mass balance of radiocesium before and after the combustion was evaluated. The initial amount of radicesium in the raw materials was 98,920 Bq; the amount of radiocesium in the collected materials was 95,285 Bq. About 95 % of radiocesium was recovered by this combustion system. The difference between total amount of radiocesium before and after the combustion was caused by the uncertainty of initial concentration due to the inhomogeneity of radiocesium distribution in pine needles and bark, and the radiocesium remained as soot in the woodstove, smoke tube and water tank.

The transmission electron microscopic observation and EDX spectrum of white ash were shown in Fig. 7.6. The main component of white ash was calcium oxide from X-ray diffraction analysis. EDX spectrum analysis indicated the existence of boron (0.187 keV, BK α), magnesium (1.25 keV, MgK α), silica (1.74 keV, SiK α), phosphor (2.01 keV, PK α), manganese (5.89 keV, MnK α), iron (6.38 keV, FeK α), copper (8.0 keV, CuK α), zinc (8.63 keV, ZnK α) and strontium (14.1 keV, SrK α) addition to calcium (3.69 keV, CaK α). Mg, P, Mn, Zn and Ca were the base element of plants, and B, Si, Fe, Cu and Sr came from the soil contamination of plants. The existence of radiocesium was not clear due to the low concentration in ash, because the estimated radiocesium concentration was about 12 ppb evaluated from 440,000 Bq/kg. The main component of calcium oxide formed the solid solution

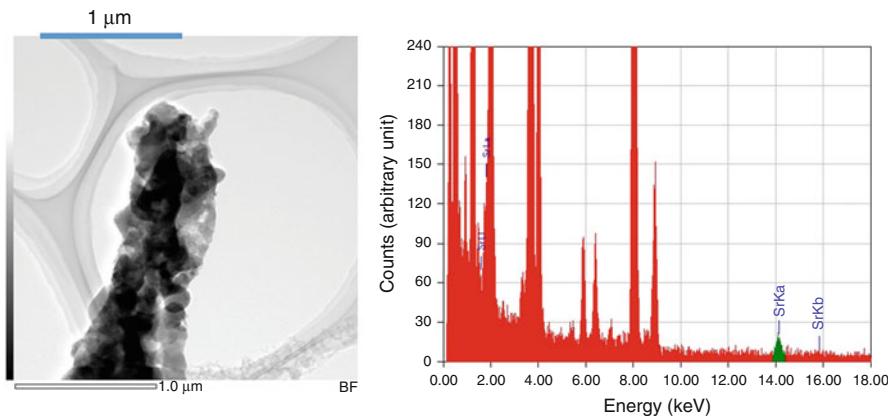


Fig. 7.6 Transmutation electron microscopic image (*left*) and EDX spectrum (*right*) of ash

with these elements. The water-soluble radiocesium from 5 g of ash was extracted by 4 L of water stirring for 5 h at room temperature. After the filtration of insoluble ash, the filtered water contained 260 Bq/L of radiocesium, which indicated 47 % of radiocesium in ash was water soluble. This result suggested that the storage of the ash from forestry wastes should be done with the extreme care not to leak the water-soluble radiocesium to the environment.

7.4 Conclusion

The mass balance of radiocesium in forestry wastes by combustion was evaluated with the laboratory-scale combustion tests and the demonstration combustion tests. About 30–35 % of radiocesium in cedar bark evaporated by the combustion under air flow condition at temperature above 500 °C. Most evaporated radiocesium was trapped efficiently by water.

Based on the laboratory combustion test, the decontamination system for the combustion of forestry wastes was developed. The decontamination system consisted of three modules, the smoke extraction apparatus by water, the combustion ash coagulate apparatus and the radiocesium filtration unit from the sewage water.

The demonstration combustion test for forestry wastes using the developed decontamination system was carried out. No gaseous radiocesium was detected in the exhausted air from this system during the decontamination of forestry wastes. About 80 % of mass of forestry waste decreased by the combustion, and 95 % of radiocesium was collected safely by the developed system. The ash caused by the combustion of forestry waste contained about 45 % of water-soluble radiocesium.

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