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Trial Burns of Low-Level Radioactive Wastes the Demonstration-Scale Incineration Plant at KAERI

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한국원자력연구소 실증소각시설에서의 저준위방사성폐기물 시험소각

양희철 · 김인태 · 김정국 · 김준형
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Abstract

Behavior of radionuclides such as ^{60}Co, ^{54}Mn and ^{137}Cs in the incineration process was studied by trial burns of simulated wastes with radio-isotope tracers. Behavior of nonvolatiles, ^{60}Co and ^{54}Mn, was similar to that of particulate matters in the process. Decontamination factors(DFs) for ^{60}Co and ^{54}Mn were 4.7×10^5 and 6.2×10^5, respectively. Behavior of semivolatile radio-isotope, ^{137}Cs, was temperature dependent. DFs for ^{137}Cs at two different incineration temperature of 850°C and 700°C were 2.8×10^3 and 2.6×10^4, respectively. Trial burns of dry active waste(DAW) transported from nuclear power station(NPS) Kori 3, 4 were also performed. DF for gross β/γ radioactivity in DAW was 1.1×10^5. This was a little higher than the estimated value, which was calculated from the tracer test results and nuclides distribution in the DAW. Average emission concentration was 0.019 Bq/Nm^3, which could meet the maximal permissible concentration(MPC) in stack emission.

요 약

방사성 동위원소 추적자를 포함한 모의폐기물의 시험소각을 통하여 ^{60}Co, ^{54}Mn 및 ^{137}Cs의 소각공정에서의 거동을 고찰하였다. 공정내에서 비휘발성 방사성 핵종들인 ^{60}Co 및 ^{54}Mn의 거동은 입자상

물질의 거동과 유사하였다. ^{60}Co 및 ^{54}Mn의 제염계수(DF)는 각각 4.7×10^5 및 6.2×10^5이었다. 반휘발성인 핵종의 거동은 소각온도의 의존성을 보여주었다. 반휘발성 ^{137}Cs의 제염계수는 850°C 및 700°C의 다른 소각온도에서 각각 2.8×10^3, 2.6×10^4이었다. 원자력발전소(NPS) 고리 3, 4호기에서 운반된 건조 방사성폐기물(DAW)에 대한 시험소각도 실시하였다. 폐기물에 포함된 총 베타/감마 방사능에 대한 제염계수가 1.1×10^5이었다. 앞의 추적자 시험의 결과 및 건조 고체폐기물 내 핵종분포에 기준을 둔 예상제염계수보다 다소 높은 값을 보였다. 굴뚝에서의 배출농도는 0.019 Bq/Nm^3으로 기체상 배출물에 대한 최대허용농도(MPC)를 만족시킬 수 있었다.

1. Introduction

Incineration is an effective means for the volume reduction and stabilization of burnable radioactive wastes before final disposal. However, airborne effluent release of radioactive materials is one of the concerning issues. Nevertheless, currently operating radwaste incineration systems adequately meet safe regulatory limit for radionuclides emission[1]. This is because radionuclides exited with a condensed phase in the off-gas are readily removed through an appropriate off-gas treatment system and those that are volatile or semivolatile in waste are presented in sufficiently small concentrations.

Also, a high volume reduction can be obtained by the retention of a major part of nonvolatile radionuclides in the furnace bottom ashes. A minor part of them are entrained in the off-gas with particulate matters(PMs) such as fly ash and unburned carbon. The entrained nonvolatile nuclides can be easily trapped by conventional filtration technology. Volatile radionuclides such as radioiodine, radiocarbon and tritium are referred to as "permanent gas"[2]. They may be present in gas phase through the whole dry off-gas treatment system which should be kept above dew point. The retention in an incinerator furnace as well as the decontamination through the off-gas system for those volatiles can not be expected. Semivolatiles such as Cs and Ru vaporize to a certain extent under normal incineration conditions. Vapor phase semivolatile nuclides can be condense out of the gas stream during off-gas cooling. Condensed phase semivolatile nuclides can be removed through ensuing filtration.

The reliability on radwaste incineration is mainly dependent upon the removal efficiency of those entrained nonvolatile and semivolatile radionuclides. Trial burns of simulated wastes with radioactive tracers were carried out to evaluate the behavior of typical nonvolatile and semivolatile nuclides in demonstration-scale incineration plant(DSIP). The entrainment of nonvolatile and semivolatile radionuclides in the off-gas during incineration and the removal of entrained radioactive matters(RMs) through the off-gas treatment system were discussed. The removal efficiencies for RMs through the off-gas system were compared to those for PMs. In addition, the results of the demonstration test using DAW from NPS Kori 3, 4 were reviewed by observing β/γ activity distribution in the process and the final emission in stack.

2. Process and Experiment

The schematic diagram of demonstration-scale incineration process is shown in Fig. 1. The process consists of two combustion chambers and an off-gas cooling and treatment system. The dry off-gas system without utilizing wet scrubber consisted of a heat exchanger, an air mixing cooler, a baghouse filter, and a HEPA filter in series.

Table 1 lists the summary of a series of test-burns using tracer nuclides. Operation conditions for the trial-burns were based on the results of test runs conducted in 1992~1993[3, 4]. After preheating of the incinerator up to 400°C, inactive simulated wastes were continuously fed to get a desired temperature. The active simulated wastes containing RI tracers were fed to maintain desired incineration temperatures.

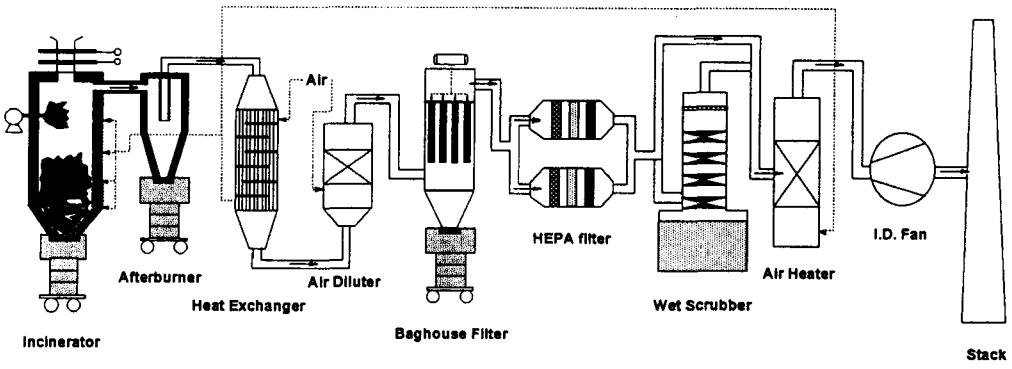


Fig. 1. The Schematic Diagram of Demonstration-scale Incineration Process

Table 1. Summary on the Program of Tracer Test Series

test No.	tracer			simulated waste	target temperature of incinerator (°C)
	nuclide	activity (kBq)	chemical form		
1	Co-60	23,950	$CoCl_2$	cloth	850
2	Mn-54	22,940	$MnCl_2$	paper	850
3	Cs-137	18,870	CsCl	paper	700
4	Cs-137	18,870	CsCl	paper	850
5	dry active waste (11,800 kBq)[1)]				850

[1)] distribution: Co-58(3.7%), Co-60(55.0%), Mn-54(15.4%), Cs-134(5.1%) Cs-137(15.3%), others(5.5%)

During all the trial-burns, immediately after feeding the first active simulated waste package, off-gas samples of the incinerator, the baghouse filter and HEPA filter were simultaneously taken by three identical particulate sampling trains. The schematic diagram of sampling train installed at the outlet of the incinerator is shown in Fig. 2. Sampling trains are arranged in a series of a sampling probe, a sample transport tube, a filtering unit, rotameter, a moisture removal system and a flow control system. A sample transport tube and a filtering unit are optionally cooled or heated according to the off-gas temperature. Flat HEPA sampling filters made of fine glass fiber matrix with a diameter of 110mm and pore size of 0.45μm was used.

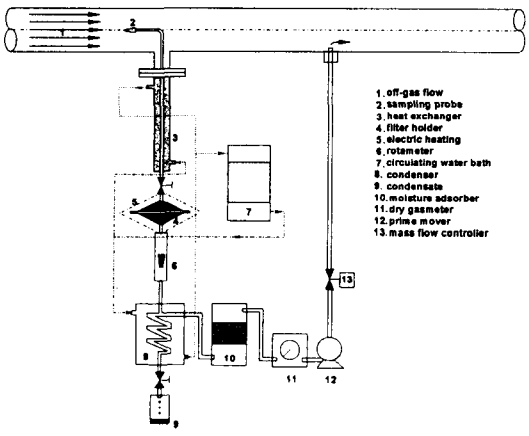


Fig. 2. The Schematic Diagram of the Particulate Sampling Train Installed at the Outlet of the Incinerator

Sample flowrates were set before the sampling operation according to the isokinetic rates determined by off-gas linear velocities measured by a hot-wire anemometer. There had been no significant pressure drop across sampling filter during all the sampling operations. Main off-gas flowrates in the sampling positions also have not changed significantly. Therefore sample flowrates were maintained within the 5% deviation of isokinetic conditions.

Specific activities of the off-gas streams and resultant emission concentrations were calculated from the measured radioactivities of beta particles on the sample filters, sample gas volumes and off-gas volumetric flowrates in the sampling positions. The specific beta-radioactivities of sample filters were measured by a low-level manual sample changer (LB 110, manufactured by Berthold). The weight changes of sample filter also were measured to evaluate particle collection efficiency.

3. Results and Discussion

3.1. Decontamination Factor and Removal Efficiency

The system decontamination factor(DF) is the ratio of the radioactivity in the feed waste to the radioactivity released subsequent to incineration and off-gas treatment. Removal efficiency(RE) or effectiveness of a system is defined as follows;

$$DF(-)=\frac{\text{Input radioactivity in wastes}}{\text{Output radioactivity at stack}}$$

$$RE(\%)=\frac{\text{Input activity-Output activity}}{\text{Input activity}}\times 100\%=(1-1/DF)\times 100\%$$

The values of related parameters to calculate DF, the distributions of radioactivity at sampling positions and determined DFs are listed in Table 2.

Table 2. Related Values of the Parameters in Measurement Position for the Evaluation of DFs and Calculated DF

measurement position	parameter	test No. 1	test No. 2	test No. 3	test No. 4	test No. 5
tracer	A_{tf} (activity in feed, Bq)	23,950,000	22,940,000	18,870,000	18,870,000	11,800,000
incinerator off-gas	A_{gi} (activity of sample filter, Bq)	2,936.2	1,004.8	1,219.0	9,178.0	9,644.5
	G_i (sample gas volume, Nm^3)	1.950	1.814	1.156	1.769	3.251
	V_i (off-gas flowrate, Nm^3/h)	310.4	310.0	310.1	309.7	310.4
Bagfilter off-gas	A_{gb} (activity of sample filter, Bq)	1.516	0.521	34.230	164.080	32.950
	G_b (sample gas volume, Nm^3)	2.670	2.205	2.305	2.135	2.355
	V_b (off-gas flowrate, Nm^3/h)	840.3	840.1	840.2	840.0	840.4
HEPA filter off-gas	A_{gh} (activity of sample filter, Bq)	0.066	0.054	1.682	12.003	1.154
	G_H (sample gas volume, Nm^3)	6.600	7.500	11.750	9.250	60.010
	V_H (off-gas flowrate, Nm^3/h)	1,140.2	1,140.0	1,140.1	1,139.8	1,140
DF for process unit	parameter relation	calculated value				
DF_1 incinerator	$A_{tf}/(A_{gi}\times E_d\times V_i/G_i)$	1.1×10^1	3.0×10^1	1.3×10^1	2.6×100	$2.6\times$
$DF_2\times DF_3$ afterburner baghouse filter	$(A_{gi}\times V_i/G_i)/(A_{gb}\times V_b/G_b)$	9.8×10^2	8.7×10^2	2.6×10^1	2.5×10^1	7.8×10^1
DF_4 HEPA filter	$(A_{gb}\times V_b/G_b)/(A_{gh}\times V_H/G_H)$	4.2×10^1	2.4×10^1	7.6×10^4	4.4×10^1	5.4×10^2
DF overall DF	$DF_1\times DF_2\times DF_3\times DF_4$	4.7×10^6	6.2×10^5	2.6×10^4	2.8×10^3	1.1×10^5

1) effective operation time

3.2. Entrainment and Vaporization in the Furnace

After being fed into the incinerator, $CoCl_2$ and $MnCl_2$ may be converted into nonvolatile CoO and MnO through the following reaction[5].

$$MeCl_2(s) + H_2O \rightarrow MeO(s) + 2HCl(g) \quad (1)$$

The converted oxides are very stable and do not vaporize at the normal incineration temperatures. The possible process causing these nonvolatile monoxides to vaporize is the reduction reaction.

$$MeO(s) \rightarrow Me(g) + 1/2\ O_2(g) \quad (2)$$

The above reaction can locally occur only in an extremely oxygen-deficient atmosphere due to the rapid combustion of high oxygen-demanding hydrocarbons. However, the vapor pressure of reduced oxide, which acts as a driving force for vaporization, is extremely low in the incineration temperature ranges. For these reasons, nonvolatile nuclides in the waste behave like ash particle in the furnace. The bulk of cobalt and manganese remained in the bottom ash as shown in Table 2. Less than 10% of RM was entrained in the incinerator off-gas. It can be, therefore, positively said that entrained RM of nonvolatile nuclides in the combustion process are mostly combining with carry-over PM.

Semivolatile CsCl may not be converted into oxide with temperature increase in the combustion condition and chemically stable CsCl in the combustion condition has enough vapor pressure to generate gas-phase itself through following vaporization process[5].

$$CsCl(s) \rightarrow CsCl(l) \rightarrow CsCl(g) \quad (3)$$

The equilibrium vapor pressure of cesium chloride is 7.83×10^{-4} atm at 700°C and 2.13×10^{-2} atm at 850°C[6]. Because of low vapor pressure, DF_1 in test 3 was nearly the same as that in test 1. Due to its relatively high vapor pressure, DF_1 in test 4 was about 1/50 of that in test 3. The difference between entrainment of CsCl in test 3 and in test 4 is basically attributable to the difference in mass transfer rate of gas phase CsCl in the incinerator.

The gas-phase mass transfer rate in an incinerator can be estimated using the model proposed by Biswas and Lin[7]. Original Lin's model is as follows:

$$M_{v,i} = (0.037\ Re_i^{0.8}\ Sc_i^{0.6}\ D_i\ W\ C_{sat,i}) f_i = M_{max}\ f_i \quad (4)$$

where, $M_{v,i}$(g/s) is the total mass of metal compound i transferred to the gas phase per time, Re_i is the Reynolds number($=LV_\omega/\nu$), Sc_i is the Schmidt number($=\nu/D_i$), L(cm) is the length of the incinerator, ν(cm^2/s) is the kinematic viscosity of gas, D_i (cm^2/s) is diffusion coefficient in air, W(cm) is the diameter of the incinerator, $C_{sat,i}$(g/cm^3) is the saturation concentration of metal compound and f_i is a factor that accounts for the binding of metal i to the waste matrix. The factor f_i has to be determined empirically, but "f_i" for simulated waste in this experiment was not meaningful. In Fig. 3 the maximum vaporization rates of three kinds of tested radionuclides are compared using following Eq.(5).

$$A_{v,i,max} = 0.037\ Re_i^{0.8}\ Sci_i^{0.6}\ D_i\ W\ C_{sat,i}\ a_i \quad (5)$$

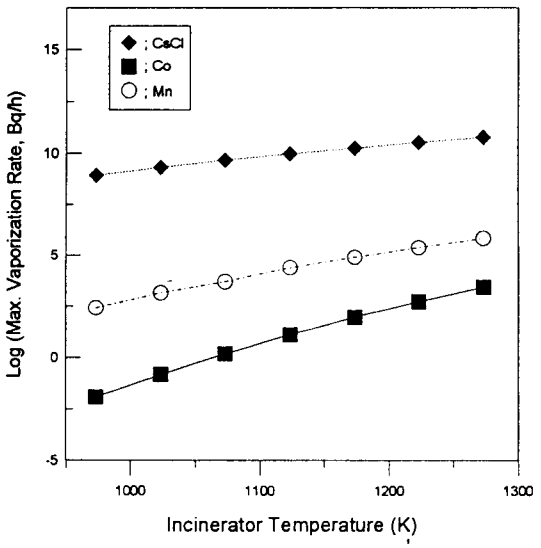


Fig. 3. Maximun Vaporization Rates in the Furnace According to the Combustion Temperatures

Table 3. Activity Balance and Entrainment of PM in the Furnace

	tracer	Incineration Temp. (°C)	radioactivity balance(Bq) input	retention(%)		entrainment(%)	
	^{60}Co	820−880	23,950	21,849	(91.2%)	2,101	(8.8%)
	^{54}Mn	815−875	22,940	22,167	(96.6%)	773	(3.4%)
	^{137}Cs	685−730	18,870	17,399	(92.2%)	1,471	(7.8%)
		835−885	18,870	11,632	(61.6%)	7,238	(38.4%)
DAW	actual	820−880	11,800	7,202	(61.0%)	4,598	(39.0%)
	predicted[1)]	820−880	11,800	9,912	(84%)	1,988	(16%)

[1)] Retention(%) = 60% × 0.2(fraction of Cs) + 90% × 0.8(other nonvolatiles)

where a_i (Bq/g) is the specific activity of tracer radionuclide. D_i was calculated by empirical correlations developed by Fuller, Schetter, and Giddings[8, 9]. Maximum gas phase transfer rates($A_{v,i,max}$) for tested tracers in the furnace were calculated based on the vaporization equation (2) and (3). The vapor phase entrainment of Co and Mn is extremely low in the combustion temperature ranges even in the possible maximum vaporization assuming f_i to be unity. However, chemically stable cesium chloride exists in liquid phase at normal incineration temperatures. It has relatively high gas-phase entrainment rate. As the combustion temperature increases, the resultant entrainment rate increases exponentially as shown in Fig. 3.

Radioactivity balances for incinerator in test series are shown in Table 3. Over 90% of the input nonvolatile radionuclides remained in the bottom ash. The small difference of retention between Co and Mn was not because of volatility but because of the entrainment rate of fly ash. Difference in the amount of fly ash entrainment is due to the characteristics of simulated material and its combustion condition. In the case of semivolatile Cs, more than 90% of input remained in bottom ash in test 3. But only about 60% did in test 4. These results show that the strong effect of the combustion temperatures on the vaporization of semivolatile nuclide. The balance of radionuclides in the DAW was estimated on the basis of the results of radioisotope tracer tests. It was compared to the actual test results. The fraction of entrainment was much higher than that of estimated value. It was nearly the same as that of Cs in test 3.

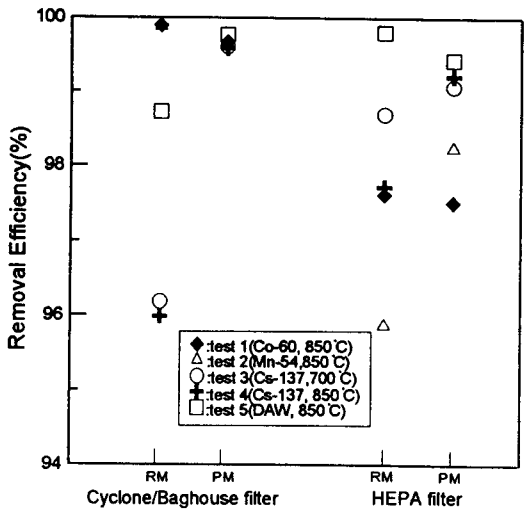
3.3. RM and PM Removal

3.3.1. Afterburner and Baghouse filter

RM and PM removal efficiencies through the off-gas treatment system are shown in Fig. 4. PM removal efficiencies through the combination of cyclone afterburner and baghouse filter were in the range between 99.6 and 99.8%. They showed stable high values in all the test series. Removal efficiencies of nonvolatile Co and Mn were not less than 99.8%. They were somewhat higher than that of PM. In case of Cs, RM removal efficiencies through this combination were 96.2% in test 3 and 96.0% in test 4. Removal efficiency for entrained mixed radionuclides in test 5 was 98.7%. All these comparisons indicate that removal efficiency for entrained nonvolatile nuclides is somewhat higher than those for fly ash particles and removal efficiency for entrained semivolatiles is just the opposite.

3.3.2. HEPA Filter

RM removal efficiency through the HEPA filter could change from 99.8 to 95.9% under tested conditions. And PM removal efficiency changed from 4 to 99.5%. RM removal efficiencies for nonvolatile



4. Removal Efficiency for RM and PM Entrained Through the off-gas Treatment System

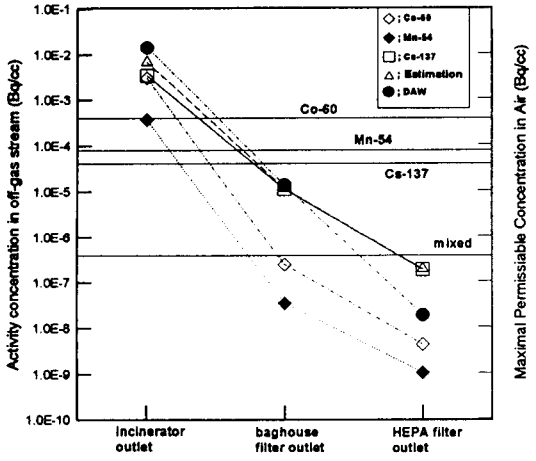


Fig. 5. Change of Activity Concentration in off-gas Stream and Emission Concentration vs. MPC (Estimation: Summation of fractional efficeincy based on tracer test results with assumption of the fraction as Co 60%, Cs 20% and Mn 20%)

Co and Mn were much lower than that for semivolatile Cs. In case of DAW, RM removal efficiency as well as PM removal efficiency was higher than any other tracer test. There is no relationship between RM and PM removal efficiency through the HEPA filter.

3.3.3. MPC versus Emission

Changes of radioactivity concentration in off-gas stream through off-gas system units are shown in Fig. 5. By tracer tests, it was shown that about 5 to 40% of RM input was entrained in incinerator off-gas according to volatility. Fig. 5 shows how these entrained RM was captured by off-gas system units. For illustration of reliability on DSIP's off-gas system, maximal permissible concentrations (MPCs)[10] for ^{137}Cs, ^{54}Mn, ^{60}Co and mixed nuclides in DAW were marked as grid lines. The resultant emission concentration for mixed nuclide was much lower than the estimated value based on the DFs for tested radionuclides and about one twentieth of MPC.

4. Conclusions

The trial burns of simulated wastes including three kinds of major radionuclides in DAW were performed. The results were compared to those of DAW and the following conclusions were derived.

1. Over 90% of nonvolatile radionuclides was remained in the incinerator ash. The entrained nonvolatile radionuclides in the incinerator off-gas were effectively removed through off-gas treatment system. Overall DF for nonvolatile radionuclide was an order of 10^5.
2. Retention of semivolatile cesium in the incinerator was largely dependent upon the furnace temperature. Over 90% of Cs remained in the incinerator bottom ash in 700°C run and only about 60% in 850°C run. However, entrained cesium could be effectively removed by low-temperature filtration process via cooling process. Overall DFs for cesium were 2.6×10^4 and 2.8×10^3 for 700°C and 850°C run, respectively.
3. Overall decontamination factor for the treatment of real DAW generated from NPS was an order of 10^5. It was somewhat higher than the estimated value based on the previous tracer test results and

nuclides distribution in the DAW. The emission concentration was one twentieth of the maximum permissible concentration in air.

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