SIMULATION TEST OF AEROSOL GENERATION FROM VESSELS IN THE PRE-TREATMENT SYSTEM OF FUEL REPROCESSING*

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Abstract

Aerosol concentration and droplet size are measured in off-gas of vessel under various conditions by changing off-gas flow rate, stirring air flow rate, salts concentration and temperature of nitrate solution. Aerosols are also measured under evaporation and air-lift operation.

I. Introduction

Non-volatile radioactive elements such as Pu, Np, Am, Tc, Ru, Sr, Cs and other FPs are confined in the process under normal operation of reprocessing. Almost all radioactive non-volatile fission products are actually confined in highly radioactive liquid waste. However, there are pathways for release of these non-volatile elements to the environment through off-gas and/or aqueous effluents. Radioactivity migration of non-volatile elements takes place in the facility primarily by aerosols. Aerosol generation under normal operation are caused by liquid aeration, evaporation, transportation and other handling of radioactive solutions[1-3]. Important area of facility for the radioactivity migration is particularly pre-treatment process which contains highly radioactive solutions in vessels. Not only decontamination factor of filters but also radioactivity behaviors in the process play the important role in assessment of source terms of environmental releases from large scale reprocessing plant[4]. Role of aerosol behavior is significant especially for the migration of non-volatile nuclides. Influences of operation condition of vessels are studied on the concentrations and droplet size of particles of aerosols in off-gas.

II. Experimental

Experimental facility(**Fig.1**) is composed of a slab-type vessel, a condenser, an air-lift and an exhaustive blower, those are connected with pipes and valves. The slab-type vessel is aerated with air

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nozzle submerged in the liquid of vessel. Liquid in vessel can be heated with two 5kW electric heaters. Liquid can be circulated outside by air-lift attached to the vessel. Aerosols are sampled on off-gas pipes(Inner diameter 28mm) with Andersen sampler(AS-500). Standard sampling time is 5 hours. Nitrate aqueous solutions, listed in **Table 1**, are used as the simulated radioactive solution of reprocessing. Salts concentration of sodium, lithium, and nitrate ions are total 100 and 350 g/L. Lithium is added as tracer of the solution. Li in aerosols is analyzed by ICP.

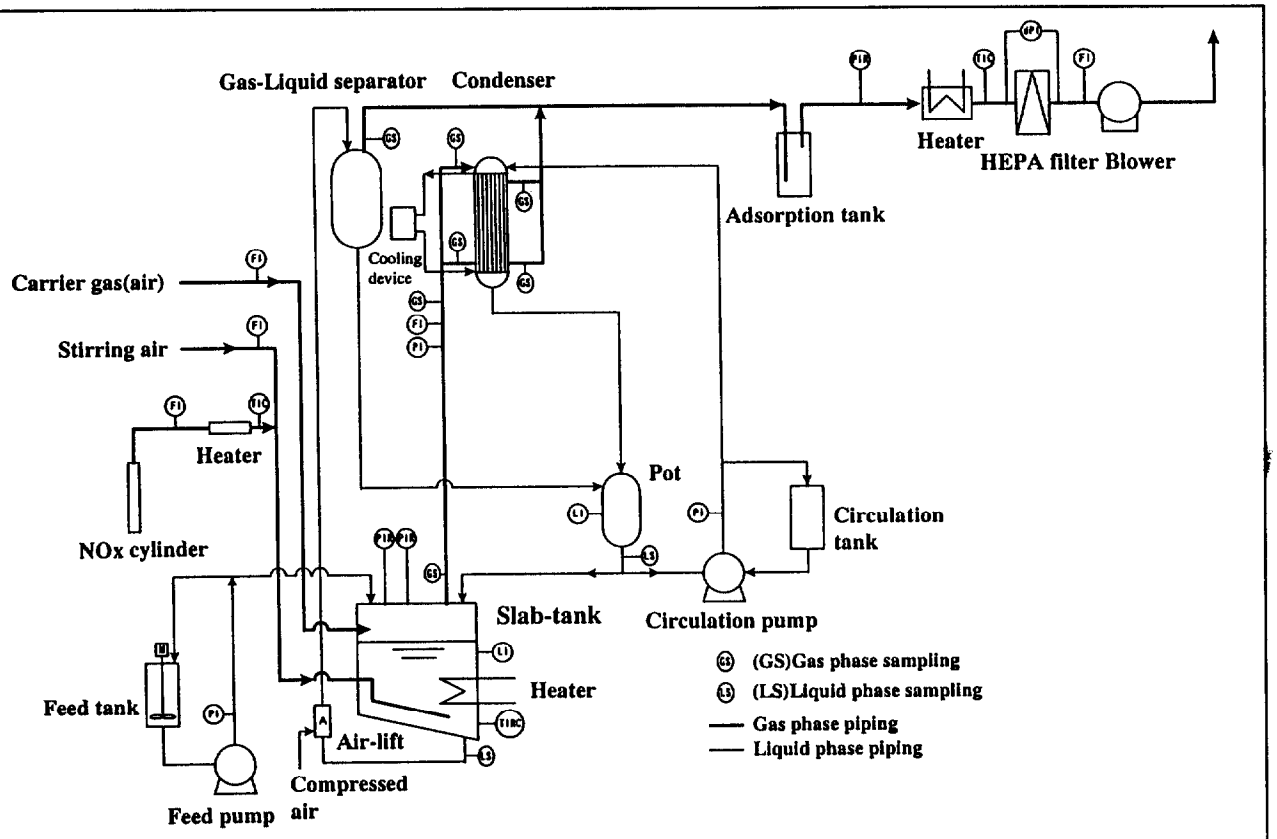

Fig. 1 Flow sheet of experimental equipment.

Liquid volume in vessel is 30L under standard condition of experiment. Distance from air nozzle submerged in solution to liquid free surface is 44cm. Distance from free surface to vessel ceiling is 55cm. Standard air stirring flow rate is 10 m^3/hm^2-free surface area of vessel which corresponds to a weak stirring condition of chemical process. Linear flow velocity of off-gas is in the range from 2 to 10 m/s. Required off-gas flow rate is adjusted by controlling sweeping air flow rate of the volume above the liquid free surface in vessel.

Aerosols in off-gas are measured at the outlet of vessels under aeration by changing the sweeping air flow rate or stirring air flow rate as operation parameters. Properties of solutions in vessel are changed in some cases. In the case of evaporation operation, aerosols in off-gas are measured at the outlet of condenser of vessels. In the case of air-lift operation, aerosols in off-gas are

measured at the outlet of air-liquid separator.

Table 1 Properties of salt solutions used in the experiment.

solution	concentration g/L	density g/cm^3	viscosity cP	surface tension dyn/cm
$NaNO_3$+$LiNO_3$ in water	100	1.065	1.17	60.0
$NaNO_3$+$LiNO_3$ in 3N-nitric acid	350	1.290	2.33	50.3
$NaNO_3$+$LiNO_3$ TBP 100ppm in water	100	1.065	1.12	36.9

III. Results

Off-gas Flow Rate

Aerosol concentrations in off-gas decreases inverse proportionally to off-gas flow rate, as shown in **Fig.2**. Required off-gas flow velocity is adjusted by changing the sweeping air flow rate on the liquid free surface. Stirring air flow rate is 10 m^3/hm^2 per free surface area of liquid in slab-tank. Aerosol concentrations are observed in the range from a few to a dozen mg/m^3 under the experimental conditions. Numerical values near plots are 50% mean diameters. It is observed that mean particle size decreases from 5.3 to 2.3 μ m by increasing off-gas flow rate. There must be the bigger possibility of trapping for the larger aerosol particles near outlet of slab-tank to off-gas pipe under the higher off-gas flow rate. The outlet is located at ceiling of the tank. Saturation by humid of sweeping air influences to reduce both aerosol concentration and particle size, that is shown in Fig.2 as well.

Stirring Air Flow Rate

Aerosol concentrations in off-gas increases roughly proportionally to stirring air flow rate, as shown in **Fig.3**. This figure shows the relations between aerosol concentration at off-gas outlet of tank and stirring air flow rate for several kind of solutions. As for $NaNO_3$ 3M-nitric acid solution,

aerosol concentrations are less than 10 mg/m^3 in the range of less than 30 m^3/hm^2 stirring air flow rate. A separate peak of the smaller particle size in addition to the main peak of the bigger particle size is observed in the aerosol size distribution for the higher range of stirring air flow rate in the experiment.

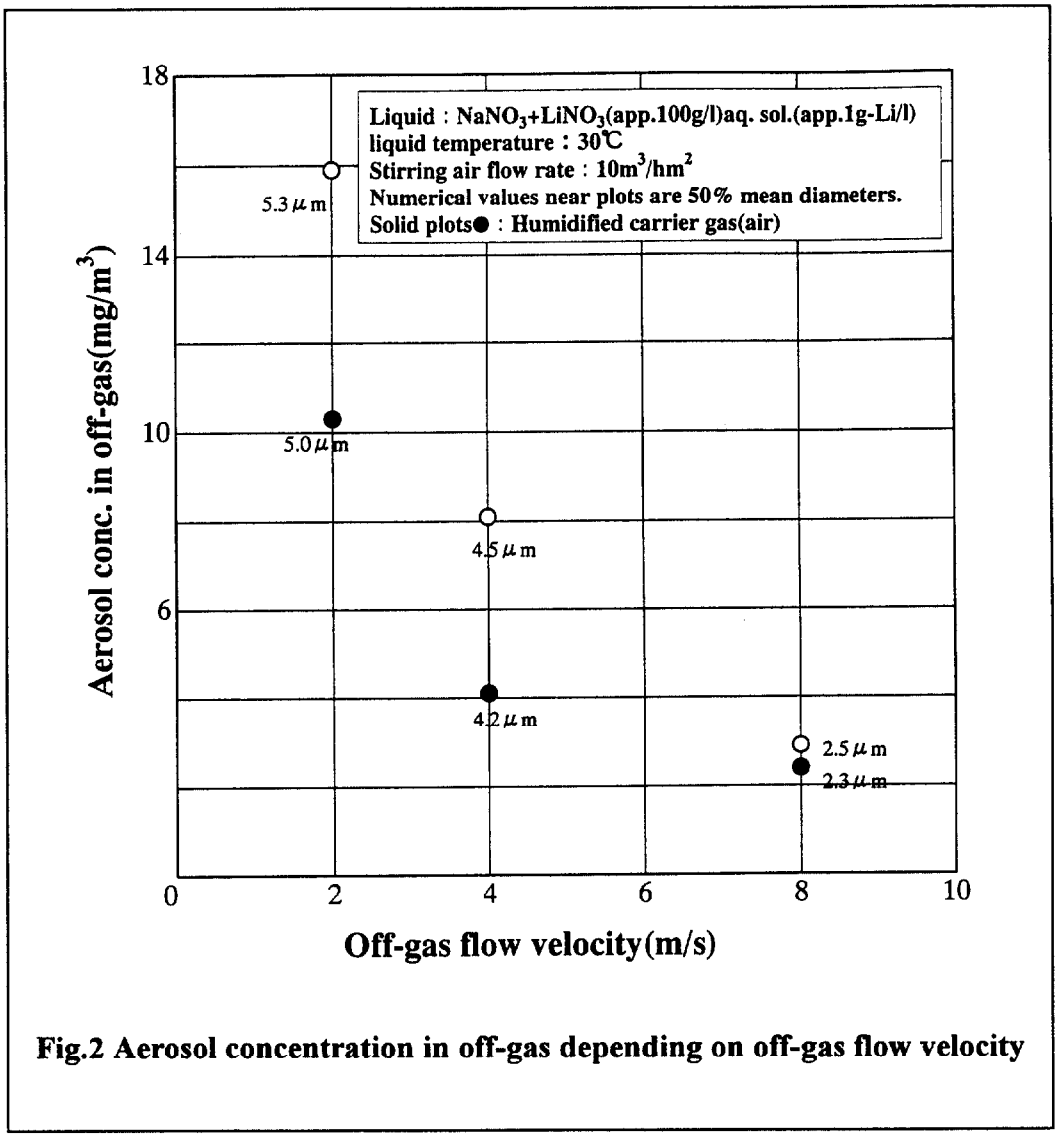

Fig.2 Aerosol concentration in off-gas depending on off-gas flow velocity

Salts Concentration

Figure 3 shows that a solution of 350 g/L $NaNO_3$+$LiNO_3$-3M nitric acid gives the smaller aerosol concentrations in off-gas approximately by half than a 100 g/L $NaNO_3$+$LiNO_3$ aqueous solution does. Both viscosity and density are bigger for 350 g/L $NaNO_3$+$LiNO_3$-3M nitric acid solution. Viscosity is 2.33 cP and density is 1.290 g/cm^3 for the solution. On the other hand, viscosity is 1.17 cP and density is 1.065 g/cm^3 for 100 g/L $NaNO_3$+$LiNO_3$ aqueous solution. Two peaks size distribution are more often observed for the higher salts concentration.

TBP Addition

Extractant TBP is added by 100 ppm to a 100 g/L $NaNO_3$+$LiNO_3$ aqueous solution. Surface tension of the solution is decreased to 36.9 dyn/cm from 60.0 dyn/cm of 100 g/L $NaNO_3$+$LiNO_3$ aqueous solution. **Figure 3** shows that aerosol concentration in off-gas increases significantly by 2 to 5 times by TBP addition.

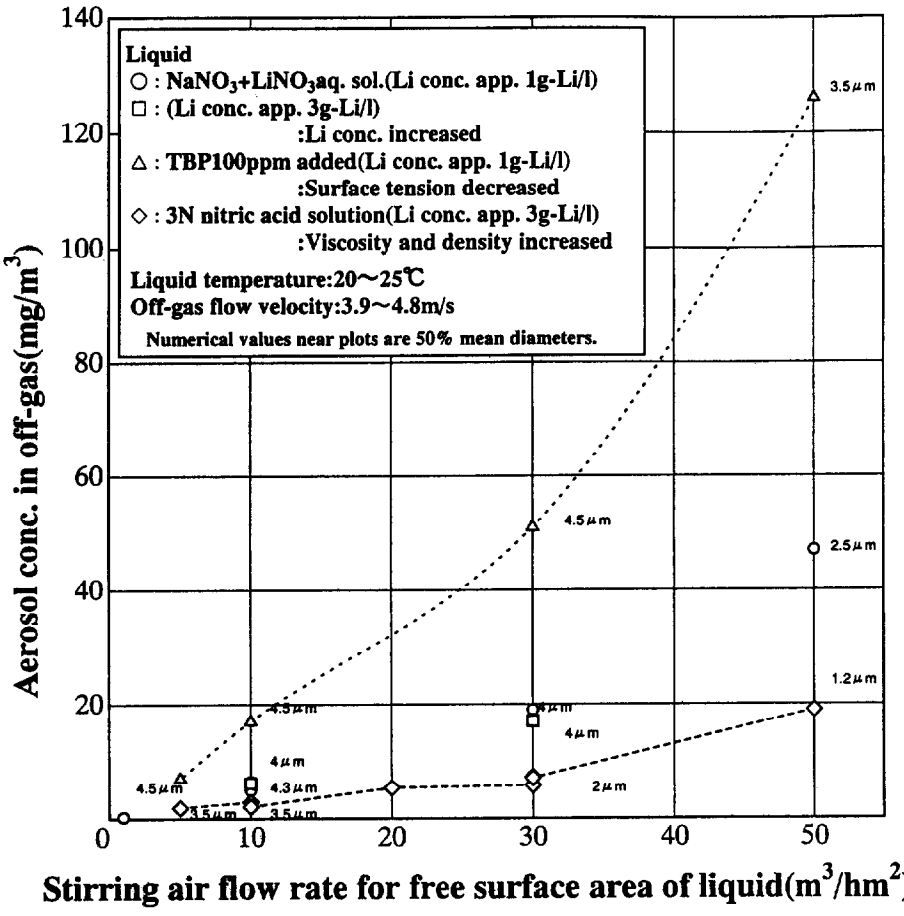

Fig.3 Aerosol concentration in off-gas depending on stirring air flow rate for different kind of solutions.

Temperature Increase

Liquid temperature in slab-tank was changed in the range from 20 to 60℃ by using two 5kW electric heaters submerged in the solution.

Aerosol concentrations in off-gas are increased by increasing temperature, as shown in **Fig.4**. Surface tension decrease by increasing temperature must be primary cause for the aerosol increase.

Significant aerosol increase is observed at 60℃ in particular. More particles of smaller size are observed for the higher temperature.

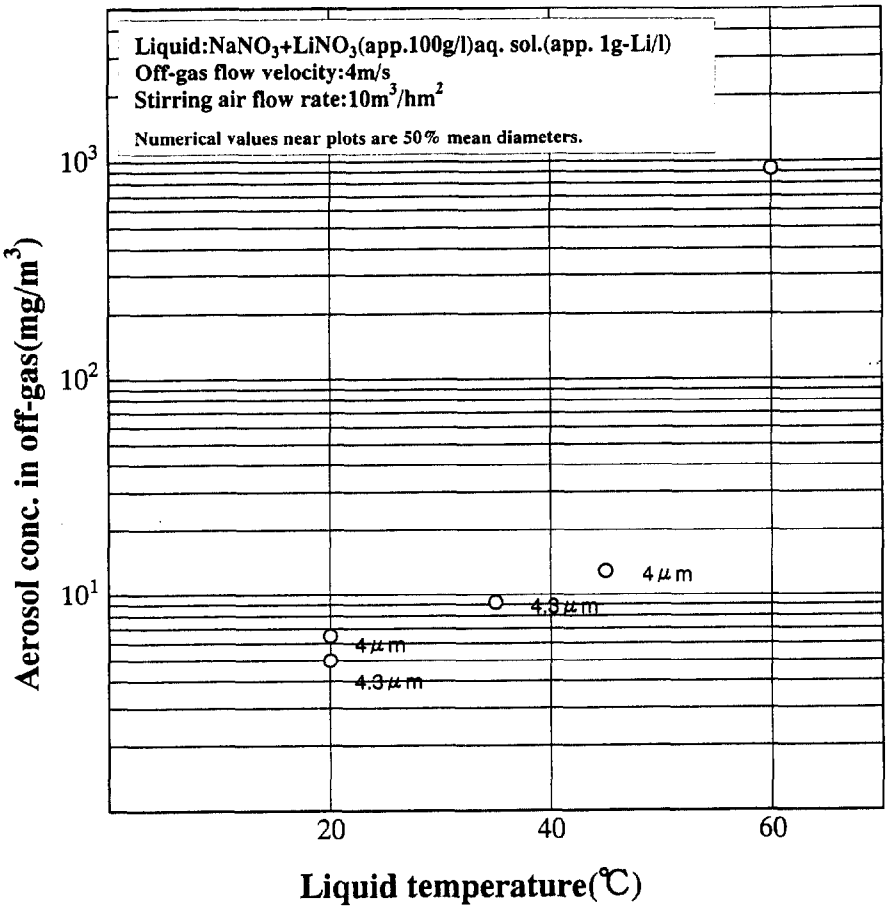

Fig.4 Aerosol concentration in off-gas depending on liquid temperature.

Air-lift Operation

Characteristics of air-lift depends on submergence ratio. Submergence ratio is defined as the ratio of depth of air-lift submergence beneath free surface to total head of lifting liquid. Air-lift was operated by adjusting submergence ratio at 0.3 by keeping app. 30L solution volume in slab-tank. Solution was recycled through air-liquid separator and pot to slab-tank. Diameter of air-lifting pipe is 12.7 mm. Aerosol sampling was carried out for 5 hours on vent pipe from air-liquid separator.

Aerosol concentration in off-gas from air-liquid separator are related to linear velocity of air in air-lifting pipe, as shown in **Fig.5**. Lifted liquid velocity is also shown in the figure. Aerosol concentrations are roughly proportional to air velocity. And it is observed that aerosol concentrations are in the range from a few to several hundreds mg/m^3 in this experimental range. Air-liquid separator

actually has no function to remove aerosol from off-gas. Demister should be applied to reduce aerosol migration to the downstream of off-gas from air-liquid separator.

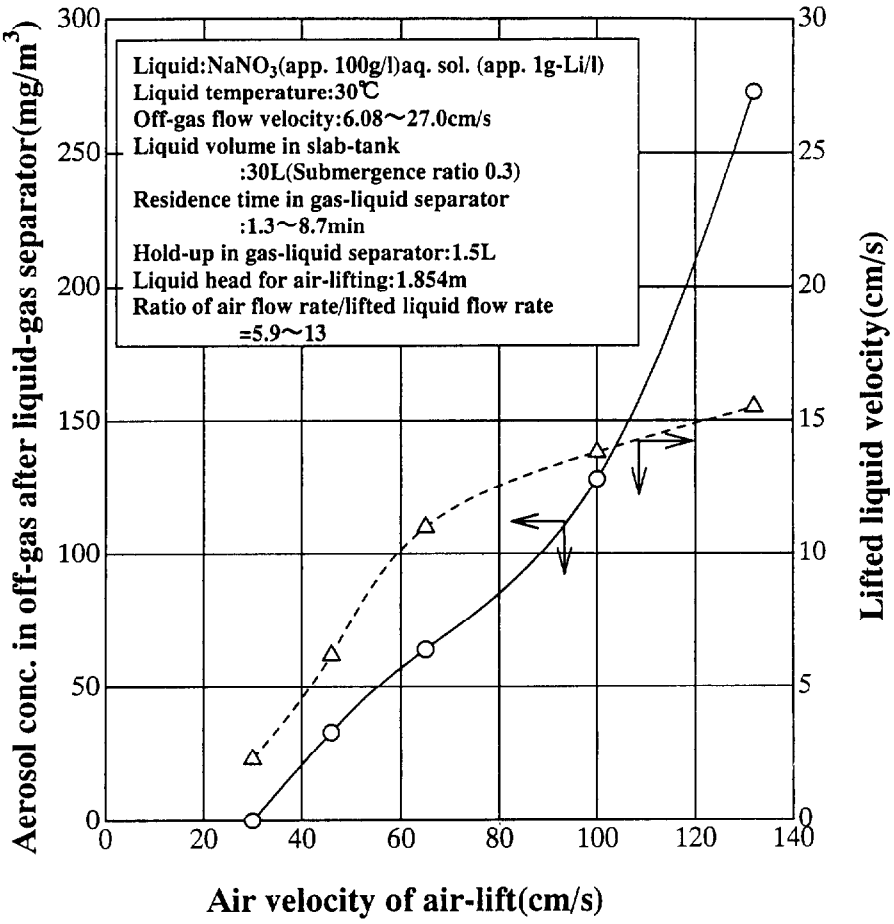

Fig.5 Aerosol concentration in off-gas at outlet of air-liquid separator and lifted liquid velocity depending on air velocity of air-lift.

Evaporation

Evaporation of solution in slab-tank was carried out at the evaporation rate in the range from 1.4 to 7 L/h. Standard evaporation rate is assumed to be 10% liquid volume in one hour, this rate corresponds to 3 L/h in this experiment. Evaporation rate was controlled by heater output and measured by condensate level increase in pot. Inner pressure of slab-tank was adjusted in the range of app. -80 to -200 mmH_2O by exhaustive blower operation. Off-gas flow velocity was adjusted at 4 m/s at off-gas outlet of condenser by controlling carrier air which sweeps the volume above free surface in

slab-tank. Aerosol sample was collected at the off-gas outlet of condenser for one hour.

Aerosol concentration and migration rate to outlet of condenser are related to evaporation rate, as shown in **Fig.6**. Dependence of aerosol migration rate on evaporation rate is not apparently clear in this experiment. Aerosol concentrations are observed in the range from 2 to 7 mg/m^3 in off-gas. Hourly migration ratios are calculated in the range from 6.2×10^{-4} to 1.7×10^{-3}. Hourly migration ratios are defined by the ratio of migration rate to outlet of condenser divided by total amount of solution in slab-tank.

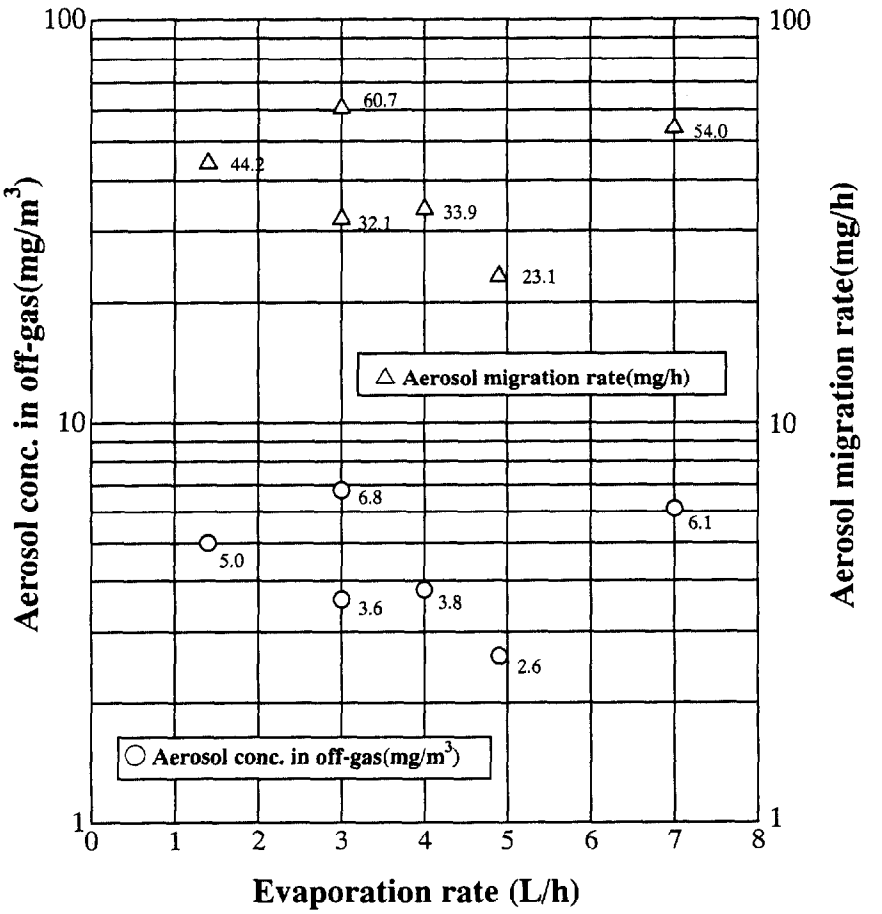

Fig.6 Aerosol concentration in off-gas and aerosol migration rate to the outlet of condenser, depending on evaporation rate.

IV. Conclusion

Aerosols in off-gas are measured at outlet of vessel under various operation condition using

simulated nitrate solutions to understand the behavior of non-volatile radioactive elements under normal operation of reprocessing. Experimental results show the following conclusions.

(1) Aerosol concentrations in off-gas at outlet of vessel are observed in the range from a few to 20 mg/m^3 in the range of off-gas flow rate 2 to 10 m/s, stirring air flow rate up to 50 m^3/hm^2, liquid temperature 20 to 45 ℃. Aerosol concentration of less than 10 mg/m^3 is observed at stirring air rate less than 30 m^3/hm^2 for 350 g/L $NaNO_3$+$LiNO_3$-3M nitric acid solution.

(2) Decrease of liquid surface tension by adding trace amount of TBP increases aerosol concentration in off-gas by 2 to 5 times that of nitrate solution without organic additives.

(3) Aerosol concentrations in off-gas from air-liquid separator are observed in the range from a few to several hundreds mg/m^3. Aerosol removal should be applied to reduce aerosol migration to the downstream of off-gas from air-liquid separator.

(4) Aerosol concentrations in off-gas from condenser of vessel under evaporation operation are observed in the range less than 10 mg/m^3.

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DISCUSSION

DORMAN: One would expect fairly rapid evaporation of some of the aerosols so size would depend markedly on position of sampling. Did you carry out size measurements at more than one point?

FUJINE: Yes, we measured the aerosol sizes at different points, namely, different distances less than 2 m from the vessel in which aerosols are generated. Average sizes of aerosols observed were not different markedly. Regarding evaporation, we made measurements on the size distribution of aerosols in humidified and non-humidified carrier air. Aerosols in humidified air consist of smaller sizes than those in non-humidified air. However, the difference is not big, approximately 10%. Sampling point was just at the outlet of the vessel. I think the non-humidified air was partially humidified to some extent in our experiment.