Treatment of Off-Gas from Lagoon Sludge Thermal decomposition

D.S. Hwang [a], J.H. Oh [a], Y.D. Choi [a], S.T. Hwang [a], J.H. Park [a], M.J. Ga [a]
a KAERI, 150 Deockjin-dong, Yuseong-gu, Daejeon
b KOCAT INC., Hanshin IT Tower II, 60-18 Gasan-dong, Geumchon-gu, Seoul

1. Introduction

Korea Atomic Energy Research Institute (KAERI) has launched a decommissioning program of the uranium conversion plant in 2001. The treatment of the sludge waste, which was generated during the operation of the plant and stored in the lagoon, is one of the most important tasks in the decommissioning program of the plant. The major compounds of the lagoon sludge are ammonium nitrate, sodium nitrate, calcium nitrate, calcium carbonate, and uranium compounds. The minor compounds are iron, magnesium, aluminum, silicon and phosphorus [1]. A treatment process of the sludge was developed as figure 1 based on the results of the sludge characteristics and the developed treatment technologies [2]. A treatment of off-gas evolved from the nitrate salts thermal decomposition is one of the important process. Off-gas treatment by using a selective catalytic reduction (SCR) method was investigated in this study.

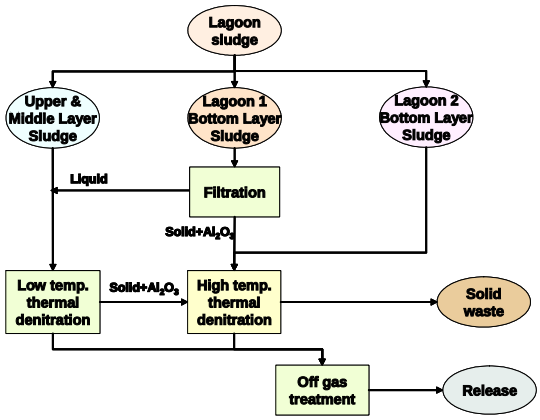

Figure 1. Lagoon sludge treatment process

2. Experimental

Samples were directly taken from each layer in the lagoon 1 and 2 as the process shown in figure 1. 50g of each sample was heated from normal temperature to 300℃ by a regular heating rate and kept at 300℃ for decomposing ammonium nitrate. And this was heated to 900℃ by a regular heating rate for decomposition of other nitrates and stabilizing of waste. Off-gas was treated by oxidation catalyst and SCR catalyst at 300℃. Oxidation catalyst is Pt/TiO_2 type and is 15(H) X 15(W) X 10(L) cm and 200 cells/in^2. SCR catalyst is V & W/TiO_2 and is 15(H) X 15(W) X 20(L) cm and 46 cells/in^2. Figure 2 shows a treatment process. NO, NO_2, N_2O, and NH_3, which are off-gas from decomposing of sludge, were analyzed by FT-IR Spectrometer Gas Analysis System (MIDAC I-4001). Flue Gas Analyzer (Eurotron Green Line MK II) was used for calibration of values analyzed from FT-IR.

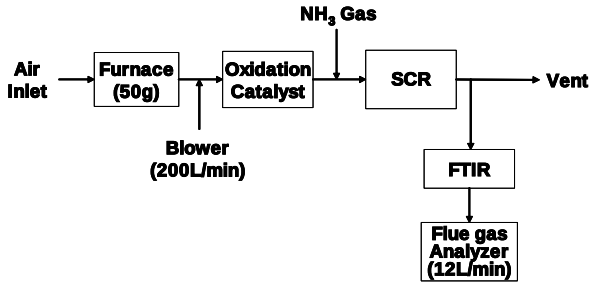

Figure 2. Off-gas treatment process

3. Results and discussion

Figure 3 shows a concentration of each gas before and after treatment of off-gas from thermal decomposition of lagoon 2 middle layer sludge. Lagoon sludge contains nitrate salts like ammonium, sodium, and calcium nitrate. It is known that these are decomposed by several mechanisms.

Ammonium nitrate is decomposed by several reactions as shown in table 1 [3]. It is confirmed that this is decomposed around 300℃ by several reactions as shown in figure 3 (a).

Calcium nitrate is decomposed according to the $Ca(NO_3)_2 = CaO + 2NO_2 + 1/2O_2$ equation at about 500℃ [4]. Sodium nitrate is decomposed as the following equation at around 600 ℃ [5].

$NaNO_3 = NaNO_2 + 1/2O_2$

$2NaNO_2 = Na_2O + NO + NO_2$

Therefore, it is thought that NO_2 and NO evolved from decomposition of calcium and sodium nitrate.

Off-gas was treated by second step. First, NH_4 evolved from decomposition of ammonium nitrate was oxidized by oxidation catalyst as shown in figure 3 (b). NOx was treated by SCR catalyst with reducing agent,

NH_3, around 300℃. NH_4 peak of figure 3 (c) was due to supply of excess NH_4. However, N_2O could not be treated SCR method. It is thought that N_2O should be treated by a additional method.

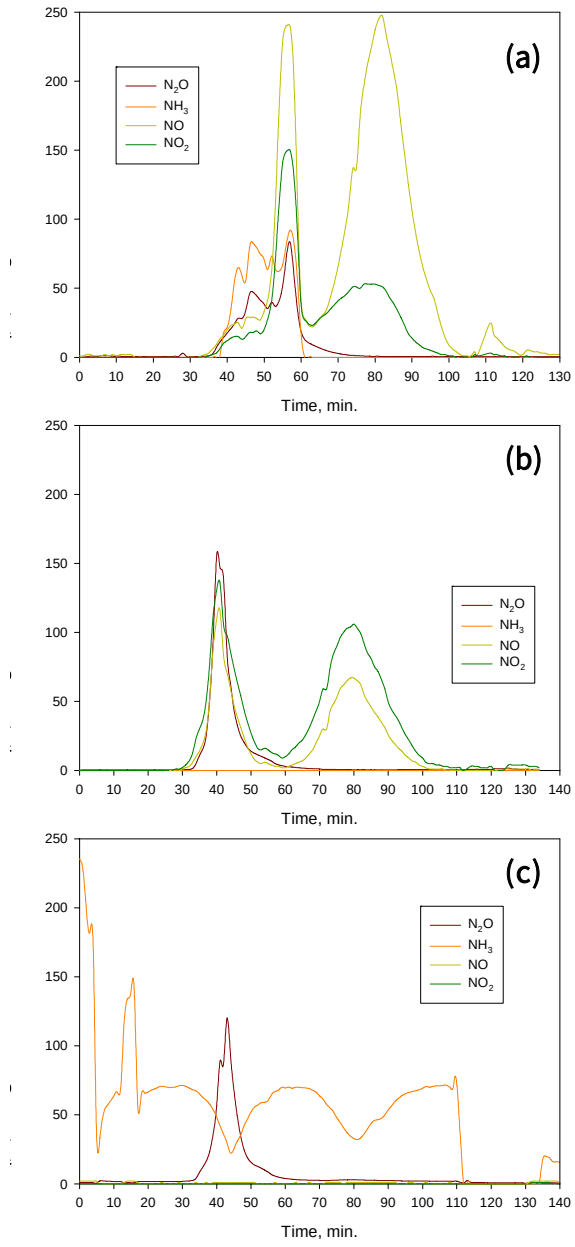

Figure 3. Concentration of each gas with time
(a) before treatment, (b) after oxidation cat. Treatment, (c) after oxidation and SCR cat. treatment

Table 1. Modes of thermal decomposition of NH_4NO_3

Reaction	Heat evolved (cal/g)	Gas volume (ml/g)	Temp (°C)
i) $NH_4NO_3 \rightarrow$ $NH_3(g)+HNO_3(g)$	- 521		
ii) $NH_4NO_3 \rightarrow$ N_2O+2H_2O	108	840	320
iii) $NH_4NO_3 \rightarrow$ $3/4N_2+1/2NO_2+2H_2O$	316	910	860
iv) $NH_4NO_3 \rightarrow$ $N_2+2H_2O+1/2O_2$	354	980	950
v) $8NH_4NO_3 \rightarrow$ $5N_2+4NO+2NO_2+16H_2O$	201	945	560
vi) $NH_4NO_3 \rightarrow$ $1/2N_2+NO_2+2H_2O$	86	980	260

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