The Off Gas Treatment In The Process Of Vitrification And Incineration of Nuclear Waste

Herlan Martono, Aisyah

Radioactive Waste Technology Centre, BATAN
Kawasan Puspiptek, Serpong, Tangerang Selatan INDONESIA
***E-mail** : herlanmartono@yahoo.com*

Abstract :

The off gas treatment in the process of vitrification and incineration of nuclear waste has been studied for the purpose of the effectiveness treatment system evaluation and using its equipment. The off gas from vitrification process leaves the melter at temperature of 600 ^{0}C composed of NO_x, Ru and particle. The treatment consist of cooling, scrubbing and filtering of the off gas. The cooling is performed by air film cooler, the scrubbing is performed by a series of submerged bed scrubber, venturi scrubber and water scrubber. Then the un-scrubbed particles are captured by HEME and HEPA filters. Before entering the Ru srubber, the off gas is warmed up to a temperature of 65 ^{0}C, scrubbing the Ru by silica gel and filtering the particles by HEPA filter. The off gas from the vitrification process has a fixed composition corresponding to the composition of the vitrified waste which has relatively fix composition. The off gas from incineration process leaves the incinerator at temperature of 1100 ^{0}C composed of CO_2, NO_x, CO, HCl, HF and SO_2 with the composition depend on the incinerated waste type. The treatment consist of cooling and filtering. The cooling is performed with the air injection by double fan. The treated off gas, either from the vitrification or incineration to be released into the environment via a stack. The off gas treatment system for vitrification process can be used for treatment of off gas from incineration process by passing the Ru scrubber, vice versa by connecting Ru scrubber after HEPA filter.

Keywords: incineration, off gas, vitrification

1. Introduction

At the present time, nuclear technology application in the many fields have been conducted such as in the medicine, industry, agriculture, research and development and so on. Based on the ctivities rose any kinds of waste such as high level active waste and low intermediate active waste. High level liquid waste (HLLW) is generated as by product of the first cycle extraction of spent nuclear fuel reprocessing. The HLLW contains many fission product radionuclide and a small of actinide. Transuranic liquid waste is by product of the second cycle extraction of the spent fuel reprocessing which contain many actinide and a small fission product. The Management of HLLW is very important because contain of long live radionuclide. Based on this reason, vitrification process of HLLW is conducted by borosilicate glass, which is corosif, heat, and radiation resistance and stabilized for a long time **[1,2]**. Vitrification process in the melter need to operate on high temperatur, so that treatment of off gas for safety aspect criteria must be conducted in order off gas is out through the stack into the environment. Low and intermediate solid waste generated coming from reactor operation, and research laboratory. Choosing of radioactive waste treatment technology is based on several consideration such as, capacities volume reduction, weight reduction, safety and simplification process, storage safety and economic. Treatment of low and intermediate solid waste is developed in Radioactive Waste Technology Center (RWTC) - BATAN, through the inceneration process. Inceneration process is conducted for treatment of burning waste such as laboratory wears, spent ion exchange resin, liquid organic waste, used filter, alpha emitter waste, plastic waste, biology waste. Before inceneration of solid radioactive waste, there are many kinds for considerations **[3]**;

a. There are significan different in the treatment cost as conditioning, transportation, interm storage and disposal when waste treatment are conducted by inceneration process and non inceneration.
b. There are more advantage in the interm storage and disposal for inceneration ash which have it's volume lower than without operation.
c. Ash as burning product are stable form in the interm storage and disposal.

Volume reduction in the inceneration process can be obtained about 10 - 100 %, it depend on density, composition and homogenity of waste, burning method, treatment of off gas method, and method of condisioning ash as burning product. Volume reduction will increase, if separation of non combustible materials and compatible materials are conducted **[3,4]**. One of the problems will be met in the inceneration process is off gas as by product contain radioactive materials in solid aerosol to form the dust contain 60Co , $14CO_2$ and liquid aerosol in relative to use H_2O in the cooling system contain 3H, or contain corosive gas NO_x, CO, HCl, HF and SO_2, it depend on waste chemical composition. The various gas must be treated by off gas treatment system to protect environment toward chemical and radiology dangerous **[3,5,6]**. In this paper it will be presented off gas as by product of treatment of high level liquid waste with vitrification process. and low and intermediate radioactive waste with inceneration process.

2. Material and Methods

2.1. Treatment Of High Level Liquid Waste By Vitrification Process.

High level liquid waste is generated from the first cycle of the reprocessing plant of spent reactor fuel. The waste contain many fission products and a little actinide so that need to manage for million years. There are many kinds materials have been studied for immobilization HLLW i.e glass, synroc, and vitromet. Glass is chosen because production of glass easier than vitromet and synroc, and glass is stabel for along time. There were many kind of glass have been developed such as phosphate glass, aluminosilicate glass, borosilicate glass, and ceramic glass. Base on technical consideration, stability in the long term, and waste loading, borosilicate glass is choosen for HLLW conditioning. This method has been used in the industrial scale by many countries. Borosilicate glass has formed at temperature of 1150 ^{0}C, good chemical resistance, while devitrification is occur at higher temperature than phosphate glass. Immobilisation HLLW with glass materials is vitrification process **[1,2]**. For example vitrificasion process is conducted by Japan Nuclear Fuel Cycle Development Institute in Japan. Vitrification process use borosilicate glass in the melter, until the position of waste-glass higher than electrode although melter is not operated. Start up of melter by heater and microwave until temperature 600 °C. At the 600 °C, electrode is on and waste glass can be electric current as heat producing. This system is called Joule heating system. Joule heating system is conducted until waste glass temperature is 1150 ºC. At the 600 ºC, electrode is on and waste-glass can be electric current as heat producing. At this time, feeding of glass frit and HLLW are begun. The ratio of borosilicate glass frit and HLLW is 3/1 which is fed into the melter with feeding rate is 9 kg/hour. The feed composition is 6,75 kg/hour glass frit and 2,25 kg/hour (16,10 L/hour) is HLLW. The capacity of feeding according to melter capacity is 300 kg. Melting of glass formed materials and HLLW is done at temperature 1150 ºC for 2 hours. Generally cooling of HLLW is 4 years before immobilisation with vitrification process using borosilicate glass. Vitrification product is waste glass has activity 4 x 105 Ci and decay heat 1,4 kW put into canister of stainless steel has volume 118 litre. One canister contains 300 kg (110 litre) waste glass (93% volume canister) [2]. Canister contains waste-glass store in the interm storage with cooling air for 30 - 50 years. The purpose of cooling to avoid waste-glass crystallization. Gas is out from melter, and treatment by off gas treatment system.

2.2. Treatment Of Off Gas From Vitrification Process

Off gas treatment system from the melter is presented at Figure 1. At this system off gas from the melter is cleaned up by taking the particle and gas. High efficiency apparatus was used to take very small particle and ruthenium (Ru) gas **[7]**. Off gas treatment system from melter is arranged according to a apparatus function are as cooler, for gas absorption, to take particle, and Ru gas. Apparatus for treatment of off gas from melter with capacity 240 – 300 Nm^3/hour at temperature 300 – 500 ^{0}C are as the following to **[6,7,8]**:

1. Air Film Cooler.

   Off gas temperature from the melter is cooled by air which enter into air film cooler until air mixed with off gas. Air flow rate enter into air film cooler about 70 Nm^3/hour. Temperatur of off gas decrease until 200 ^{0}C.

2. Submerged Bed Scrubber (SBS).

   The capacity of water in SBS is 1,5 m^3. Decontamination factor (DF) for NO_x particle is 3-5 and volatile gas Ru is more than 50. Pressure drop at SBS about 800 – 900 mmH_2O. Apparatus of SBS contains ceramics ball (Al_2O_3) which diameter 10 – 20 mm. The function of ceramic ball for increasing surface area of contact beetween gas and water, also has function as stirer. In the SBS off gas is cooled until temperature about 40 ^{0}C.

3. Venturi Scrubber

   Venturi Scrubber is used for treatment off gas coming from SBS. Recirculation rate of scrubbing water is 275 l/hour and gas flow rate of air input and off gas is 315 m^3/jam. The ratio of water/gas about 0,85. Gas velocity about 85 m/s. Pressure drop at venturi scrubber about 1200 mmH_2O. Decontamination factor of venturi scrubber for NO_x, particles and Ru gas are 1, 10, and 3 respectively. In the venturi scrubber off gas is cooled until temperature is 32 ^{0}C.

4. Water Scrubber

Type of water scrubber is perforated plate. In the water scrubber, recirculation of scrubbing water is conducted. Water circulation rate is 70 l/hour. At the every plate cooling water is needed. Pressure drop of water scrubber about 400 mmH_2O. Every perforated plate has 300 holes which diameter of every hole is 3 mm. The decontamination of water scrubber for particles is 3 - 5 and for Ru gas is 50. Off gas in the water scrubber is cooled until temperature is 28 ^{0}C.

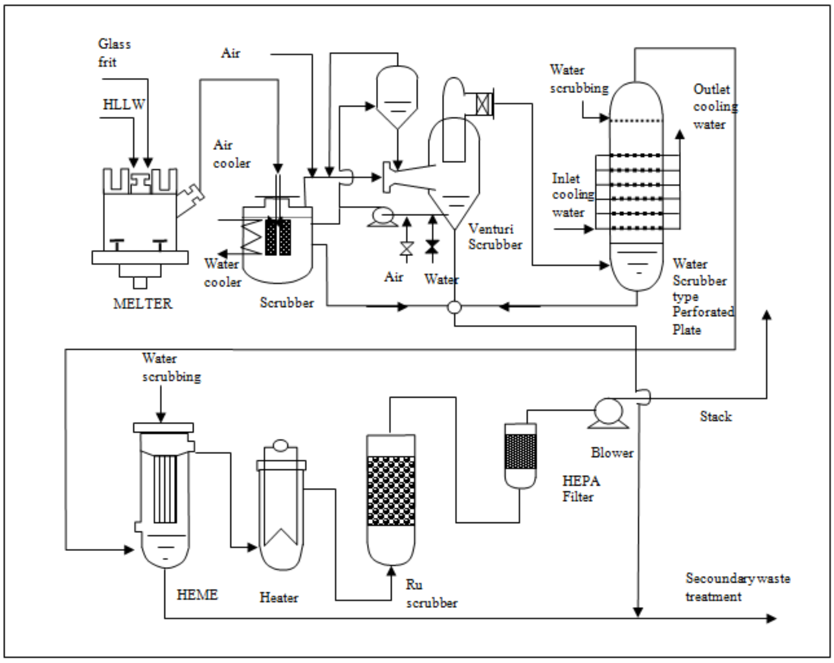

Figure 1. Off Gas Treatment from Vitrification [6,8].

5. High Efficiency Mist Eliminator (HEME)

HEME apparatus contain fiber glass and has high effisiency for particle decontaminasion. Decontaminasion factor of HEME to the particle more than 100. Pressure drop of HEME about 200 – 300 mm H_2O. After operation has finished, the fiber glass has washed or flow by water through water spray nozzle. Water flow down to the HEME which contain particles. Off gas from HEME is heated with heater, and then is flow to the Ru absorber apparatus.

6. Ruthenium Absorber.

Ruthenium absorber rate in the melter depends on temperature and composition of HLLW. Evaporation of Ru is pressed with many various reductan, such as formaldehide, formic acid and sugar. Inert gas will decrease Ru evaporation. At the high temperature process in the vitrification, Ru in the off gas as RuO_4 and at more than 800 ^{0}C as RuO_3. Ruthenium fraction in the gas form out from the melter is 1 – 15 %. For sorbing Ru in the off gas is used silica gel. Absorbing process is done at temperature 65 ^{0}C. Ruthenium gas is fed to silica gel absorber, with off gas heating before enter to ruthenium absorber. Off gas is heated before enter to the Ru absorber apparatus. Decontamination factor of Ru absorber apparatus more than 1000. Pressure drop Ru absorber is 600 mmH_2O. Because of solid gel silica, it is difficult for analysis Ru. In the experiment, sampling vaporated Ru is done by sorben solution mixed is 6 N HCl + C_2H_5OH at temperature 0 ^{0}C. The comparation of volume of HCl 6 N/C_2H_5OH is 99. Analysis of Ru is done by Induction Coupling Plasma Fluoresence Photometer.

7. HEPA Filter

HEPA filter can be used for end step treatment of off gas in the vitrification. From the all filtration componen, HEPA filter is the most effective for scrubbing small particle which minimum size about 0,1 – 0,3 μm.

Decontamination factor of HEPA filter for the same diameter particles are effected by managing off gas flow rate, temperature, pressure drop and relative humidity. Long life of HEPA filter for treatment gas has not studied. Decontamination factor of HEPA filter to the particles is about 1000. Using of HEPA filter was needed for assuring off gas decontamination through stack and then to the environment. Pressure drop of HEPA filter is about 50 mm H_2O.

2.3. Treatment Of Low And Intermediate Level Waste Using Inceneration.

In the nuclear technology application, will be arisen many kinds of radioactive waste in various amount and activities with 70 % as burnable solid radioactive waste. This waste including low and medium solid radioactive waste catagories. The purpose of inceneration process of radioactive waste is for reduction waste volume, change of physic form, chemist, and radiology waste for disposal. In the radioactive waste disposal want to be characteristic change of waste into stable anorganic material with high reduction volume **[9]**. To take high volume reduction, then are needed initial treatment of burning waste before inceneration process, are grouping of kinds waste, waste packing in the according form, measurement of weight or volume and dimention of waste, solid waste cuting and so on. In the inceneration process, there are many burning technique of radioactive waste, are **[3]**:

a. Incenerasion with excess air with burning are conducted at 800 – 1100 ^{0}C
b. Incenerasion with controlled air amount when burning is conducted at 800 – 1000 ^{0}C.
c. Pirolysis, when burning are conducted at 500 – 600 ^{0}C
d. Incenerasion at high temperature, when burning process conducted at 1400 – 1600 ^{0}C
e. Fluidized bed inceneration when incenerasion is conducted at 800 ^{0}C

Inceneration with excess air, using excess oxygen for burning process, so that gas and solid fraction can be burned in the one of burning room. Incenerasion with excess air is simple inceneration technique. Inceneration with controller air amount, need certainly air supply in the first burning room, with amount below the stoikhiometri. In the second burning chamber it is need air with rich oxigen for completing combustion of gas out from the first combustion chamber. Pyrolisis process or thermal decomposition need combustion in the reduction condition and generally have gotten with limited air feed until more than below in stoikhiometri. Pyrolisis from organic material give thermal degradation and evaporation of volatile material fraction, so that will be form burn liquid and vapor. The biggest composition of vapor is methane, hidrogen, carbon monoxide, carbon dioxide, air, and complex of hydrocarbon composition such as etane, propane, oil and asphalt. Spent materials after pyrolisis process is carbon with the first content is fixed carbon. Pyrolisis is endothermic process and is used heat resources continued for keeping. Incenerator for producing slag using relative high temperature. Heat producing from combustion of oil for conversion all burning waste loading, so that will become molten slag. Fluidized bed incenerator using different combustion technique with generally insenerator, the combustion area contain inert particle in suspended condition by air has flown throught particle with high flow rate. Waste will burn when enter to the fluidized bed incenerator. The burning solid fraction and gas of waste is occured in the one combustion area and fly ash combustion product will out from combustion area with off gas. Incenerator unit in RWTC - BATAN include in incenerator catagory with excess air for treatment burning solid radioactive waste, organic liquid waste and animal waste with capacity is 50 kg/hour. Volume reduction factor is 90 - 100 times and weight reduction factor is 70 %.

The characteristics of waste can be treated with incenerator in RWTC BATAN, are **[10]**:

1. Solid radioactive waste with the composition rubber 15 %, clothes 25 %, PVC 33 %, paper 20 %, wood 5 %, non burn able material 2 %, combustion heat 5.500 kcal/hour, Maximum activities for alpha and beta 10^{-4} Ci/m^3 and gamma 10^{-2} Ci/m^3. Before burning solid waste put in the carton box with size 500 x 350 x 300 mm and weight of every box is 5 – 7 kg.
2. Liquid radioactive waste of tributyl phosphate (TBP) in the dodecane solution with maximum concentrasion, combustion heat 10.000 kcal/hour, maximum activities for alpha and beta are 10^{-4} Ci/m^3, gamma 10^{-2} Ci/m^3. Combustion is conducted with waste injection throw nozzle to the combustion burner. The combustion of liquid organic waste containing TBP, must be added oxalate calcium to avoid pluging in the filtration system at baghouse filter and HEPA filter.
3. Experimental animal waste, for examples mouse with combustion heat 4000 kcal/hour, maximum activity of alpha and beta are 10^{-7} Ci/kg, gamma 10^{-5} Ci/kg. Before combustion, animal waste put into plastic bag and input in the carton paper, waste store in the cooler storage and then burn like burning solid waste.

4. Inceneration process will be begun with air intake in the furnace chamber and pressure constant at -15 daPa. Initial combustion is used electric system, kerosine, and air. Heating of refractory furnace is done until temperature of 850 – 1000 ^{0}C by feeding of kerosine and air with fixed ratio. Combustion reaction of kerosine and air in the furnace, so that exothermal heat will increase refractory heat in the furnace and then occured of waste combustion reaction. Gas of combustion is cooled by cooling air, so that temperature gas out from cooler is 180 – 220 ^{0}C. Gas of combustion product out of washing column, temperature decreasing to 60 ^{0}C. Combustion product i.e ash collected in the bottom part of incenerator, and immobilization in the 100 litre drum with the composition are 15 kg ash, 8 kg coral with size 0 – 80 mm, 65 kg cement, and 30 litre water. For the mixture homogenisation, the 100 litre drum then is played in the mixture unit for 1 hour with the velocity is 28 rpm. Drum contain immobilized ash are stored in the interm storage of radioactive waste **[10,11,12]**.

2.4. Treatment Of Off Gas From Incenerator.

The purpose of off gas treatment is for polutan out to the environmment has been minimal radiation or chemical and according to regulation standar. Analyse of waste characteristic and combustion condition must be conform to understand content materials in the off gas, so that can be determined off gas treatment method has been chosen. Inceneration of radioactive waste will produce gas from combustion area at 1100 ^{0}C. Radionuclide in the initial waste will be in combustion ash which collected in the first combustion area and in the off gas will be collected in the bottom part of baghouse filter. Radioactive materials in the off gas are $14CO_2$, $3H_2O$, $35SO_2$ and also corosive gas i.e NO_x, CO, HCl, HF and SO_2 depend on chemical composition of waste and combustion temperature **[3,5,6]**. Treatment of off gas in the inceneration process is conducted in RWTC - BATAN, show in the Figure 2. In the cooling step, filtrasion using baghouse filter with efficiency 99%, HEPA filter with efficiency 99%, and sorpsion using NaOH solution for eliminate acid gas in the off gas, so that gas out from stack is clearly **[10, 13]**.

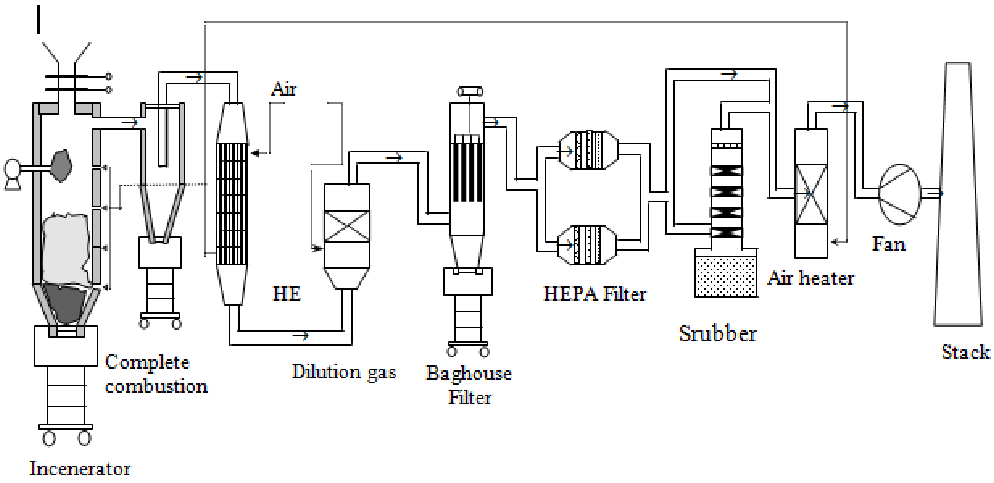

Figure 2. Off Gas Treatment from Inceneration **[10,13]**.

1. Incenerator chamber.
   Incenerator has 2 combustion chamber, and respectively has 1 burner with the capacity is 5 kg/hour, and 20 kg/hour. Temperature of combustion chamber I is 800 ^{0}C with 1,3 m^3 in volume, 1 m in diameter, and 1,5 m in height. Temperature of combustion II is 1100 ^{0}C with 1,9 m^3 in volume, 1 m in diameter, and 2,5 m in height. Operation pressure in the combustion chamber kept about - 15 daPa. Combustion chamber is coated with refractory with classificasion ASTM C401A.
2. Off gas dilution chamber.
   Hot gas from combustion chamber I and II have temperature is 1100 ^{0}C before enter to gas treatment unit, temperature is down by mixed with air until is get temperature is 180 ^{0}C. Addition of fresh air is supplied by 2 fans.
3. Particles sieving unit in the off gas.
   Out gas from dilute chamber through 2 filtrasion units are baghouse filter and HEPA filter. Off gas flow rate is filtered 7700 Nm^3/hour. Baghouse filter consist of 120 piece of bag made in teflon with filtrasion efficiency is 99 %. Out gas from baghouse filter through HEPA filter wih efficiency is 99,991 %.
4. Neutralization unit.

After filtration, gas is cooled until 50 ^{o}C in the venturi scrubber and acid vapor in the gas is netralized in the closed column with NaOH solution is flown by pump from the tank. If pH of NaOH solution reach to 5 – 6, then injection of NaOH solution is performed for operasion. Injection system consist of tank and pump. Injection of NaOH solution is conducted by pump operasion if low pH detection and injection is finished if pH detection is enough. The function of NaOH solution in the netralization unit is also as absorber.

5. Extraction and release off gas to the stack.
After off gas treatment in the neutralisation unit, then gas is heated by coil. Extraction of off gas is conducted by fan and release off gas to the stack after radiation activity monitoring using radiation protection system.

3. Result and Discussion.

The composition of low and intermediate solid radioactive waste, if compared with high level liquid waste, then radionuclide content in the HLLW more complex. The HLLW contains actinide and Ru, it there is not in the low and intermediate solid radioactive waste, then for treatment off gas from vitrification process is completed Ru gas absorber i.e silica gel or mixed of 6 N HCl and C_2H_5OH. Off gas from vitrification process is not contain actinide, but contain NO_x which is coming from saturated HNO_3 in the HLLW. Generally the first content low and intermediate activity solid waste is Cs **[14]**. The composition of HLLW more complex, so that off gas more in amount and more complex. Off gas out from the melter more difficult, because through molten waste-glass has high viscosity. Vitrificasion process is operated at 1150 ^{0}C, then temperature distribution in the melter show that waste-glass temperature at the surface of the melter about 700 ^{0}C and waste-glass temperature near electrode is about 1130 ^{0}C. The effect of temperature distribution is waste glass flown because there is density difference. In the incenerator, there is not barrier for resisting out of off gas, so that temperature of off gas from incenerator is 1100 ^{0}C more than temperature of off gas from the melter is 600 ^{0}C. Capacity of air for cooling off gas from incenerator more than vitrification for the same capacity of off gas. Off gas at the vitrification process has fixed composition because HLLW is processed has fixed relative composition. In the inceneration process, off gas composition will be different depend on kind waste is burn i.e solid waste, organic liquid waste and experiment animal waste. Off gas treatment system in the inceneration process has been design for burning different waste composition. The component off gas in the inceneration and vitrification process are the same in which the gas and particles at high temperature. Off gas treatment system consist of temperature decreasing, gas sorption, and particles filtration. At the incenerator, cooling use 2 air fans. Air is mixed with off gas from incenerator so that decreasing of off gas temperature. Cooling at the melter use air film cooler, air mixed with off gas from the melter, so that decreasing off gas temperature. Treatment of off gas from the incenerator, filtration is conducted after cooling. Filtration is conducted by baghouse filter. At the baghouse filter, off gas temperature must low in order to filter unburn material and then off gas can not high humidity. The baghouse filter must be saturated, then is cleaned and particles is collected in the drum. Gas is not containing particles, then is absorbed by NaOH solution and discharge to environment through stack. After baghouse filter, off gas still contain small particles pass through HEPA filter.Treatment of off gas from the vitrification process, after air film cooler gas flow through submerged bed scrubber (SBS), venturi scrubber, and water scrubber for gas absorption, and can help for cooling and particle resistance. In the next step off gas with low temperature and remain particles flow to the HEME. Particles will filtered by HEME. Gas out of HEME is heated until temperature 65 ^{0}C and input to Ru absorber. Absorption Ru by silica gel at the temperature of 65 ^{0}C. Output gas from Ru absorber through HEPA for filtering small particles has size 0,1 – 0,3 μ. Output off gas from the HEPA filter to the environment through stack hopes not dangerous to the environment. Treatment of off gas from inceneration process through cooling step, particle sieving, and gas absorption. Treatment of off gas from the melter consist of step are cooling, gas absorbtion, and particle sieving. Off gas treatment system at the vitrification process, the life time of filter is longer because at the gas sorbtion much particles is filtered. On the off gas treatment system from inceneration process, although saturated baghouse filter is fast but the next can be able to cleaned. Sorber can not put before bag house filter because output gas has high humidity. Gas input baghouse filter may not high humidity.Off gas treatment system at the inceneration process, can be used for treatment off gas of vitrification with addition of Ru absorber after HEPA filter. Absorption of Ru by silica gel at temperature 65 ^{0}C, and off gas out of sorber at temperature 60 0C, so

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that heater is not needed. Absorption Ru is used sorber of mixed HCl 6 N + C_2H_5OH, can be put after sorber NaOH. Off gas treatment system of vitrificasion process can be used treatment off gas from incenerasion with delete of Ru absorber. The part of the other system of off gas can not be changed.

4. Conclusion

Treatment of off gas from vitrification process more complex because there are Ru absorber. Off gas release from melter more difficult because there are molten waste glass as barrier which has lower temperature so that high viscosity. This condition is not occur at the incenerator that off gas temperatur is 1100 ^{0}C higher than off gas temperatur of melter is 600 ^{0}C that air is needed for cooling of off gas with the same capacity more than incenerator.

Off gas treatment system from inceneration process consist of cooling, filtration, and sorption systems. This system using baghouse filter for filtration. Although filter saturated rapidly, but every time filter can be cleaned again. Sorber is put behind the baghouse filter because off gas entered baghouse filter can not in high humidity condition.

Off gas treatment system from the vitrification process consist of cooling, sorption, and filtration. Besides function of absorber for gas absorbtion, also for cooling and resist of particle so that the function of filter is decrease, then life time of melter increase. If filter put before sorber, then filter saturated rapidly.

The off gas treatment system from inceneration process can be used for treatment off gas from vitrification process with addition Ru absorber after HEPA filter, if using gel silica absorber. If used mixed of 6 N HCl and C_2H_5OH then Ru absorber put after NaOH absorber. Off gas treatment system from vitrification process can be used for off gas treatment from inceneration process without Ru adsorber apparatus. Filter and sorber component in the system can not be changed.

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