INCINERATION OF RADIOACTIVE WASTES CONTAINING ONLY C-14 AND H-3 CALCULATIONS TO KEEP INDIVIDUAL AND COLLECTIVE DOSES AS LOW AS REASONABLY ACHIEVABLE

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ABSTRACT

Carbon 14 (^{14}C) and tritium (^{3}H) are preferentially used in chemical and biological research institutions in the Philippines. Most of the solid radioactive wastes generated by these institutions consist of combustible materials such as paper and accumulated environmental samples. Liquid wastes usually contain organic substances and biological components. The method proposed by this paper for the management of ^{14}C and ^{3}H wastes is incineration will be able to provide a means of disposal for ^{14}C and ^{3}H and their hazardous organic and biological constituent while keeping the level of radioactive releases in the environment within accepted limits.

In the incineration process, the radioactively contaminated wastes which may consist of pure combustible solid wastes or a mixture of combustible solid and absorbed liquid wastes will be mixed with non-radioactive combustible wastes to lower the activity concentration and to improve the efficiency of combustion. The incineration may be carried out in an ordinary locally fabricated incinerator.

The calculations presented determines the concentration limit for the incinerable wastes and the restriction on specific activity of the particles of the incinerable wastes containing ^{14}C or ^{3}H on the basis of the accepted air concentration and on the annual dose limit for an average radiation worker in the country.

The study is intended to determine the concentration limits for the incineration of ^{14}C and ^{3}H wastes. The results of the calculations will be essential in the establishment of future regulatory requirements in the management of wastes in the country.

INTRODUCTION

Carbon 14 (^{14}C) and tritium (^{3}H) are preferentially used in chemical and biological research institutions in the Philippines. Most of the solid radioactive wastes generated by these institutions consist of combustible materials such as paper and accumulated environmental samples. Liquid wastes usually contain organic substances and biological components. The method proposed by this paper for the management of ^{14}C and ^{3}H wastes is incineration which if properly conducted can provide an effective means of disposal for ^{14}C and ^{3}H and their hazardous organic and biological constituent while keeping the concentration of gaseous releases in the environment as low as reasonably achievable.

In the incineration process, the radioactively contaminated wastes which may consist of pure combustible solid wastes or a mixture of combustible solid and absorbed liquid wastes will be mixed with non-radioactive combustible wastes to lower the activity concentration and to improve the efficiency of combustion. The incineration may be carried out in an ordinary locally fabricated incinerator.

The calculations presented determine the concentration limit for the incinerable wastes and the restriction on specific activity of the particles of the incinerable wastes containing ^{14}C or ^{3}H on the basis of the accepted air concentration and onthe annual dose limit for an average radiation worker in the country.

OBJECTIVES

The purpose of this study is to be able to determine the concentration limit for Carbon 14 or Tritium wastes which can be incinerated using allowed regulatory and internationally accepted values. The results of the calculations presented are essential in the establishment of future regulatory requirements in the management of radioactive wastes.

GENERAL CONSIDERATIONS

Incineration is a method that can be applied for the disposal of wastes containing ^{14}C or ^{3}H. The proposed conditions for the method considered is found conservative as far as minimizing the hazards of releasing ^{14}C and ^{3}H to the environment is concerned. It can also be considered practical and economical with respect to the needs of the users being simple and effective in destroying hazardous organic and biological components of the radioactive wastes.

Carbon 14 is one of the most commonly encountered elements in living matter, being a component of all food we eat and also present in the air we breath. Both ^{14}C and ^{3}H undergo rapid dispersion in the atmosphere together with their stable element counterparts in nature. In its transformation, ^{14}C is reduced to CO_2 which becomes fixed in vegetation through photosynthesis. Carbon dioxide is a chemically stable gas and it does not bind itself with the molecules of the tissue. In other chemical forms, the only way by which ^{14}C reaches man is through the ingestion pathway.

The uptake of ^{3}H results in the incorporation of the radionuclide in the organic molecules of the body tissue and its mixing with the body water.

Since carbon and hydrogen are components of plants and animals, ^{14}C and ^{3}H are preferentially used and becoming very useful in most biological and chemical researches in the Philippines.

BASES FOR RECOMMENDATION

The majority of the ^{14}C and ^{3}H solid wastes generated in the Philippines consist of combustible materials such as paper and accumulated environmental and biological samples. Liquid wastes generally contain organic substances and biological components. Users of the radionuclides segregated waste materials generated according to radionuclide following licensing requirements on radiation safety program.

Calculations concerning incineration of ^{14}C and ^{3}H wastes are presented in the suceeding sections. The calculations are based generally on accepted maximum permissible concentration in air and annual dose limit for radiation workers in the Philippines.

In the calculation for ^{14}C, no considerations were taken on the fact that exposure can only be occasional and that additional dilution occur as stack gases are released to the atmosphere. The dilution factor which is expected to occur within a 10 meter radius around the incinerator is about 400. The allowed limit for the public is 1/10 to 1/15 of that in restricted area.

In the absence of available data on the heating value of mixture of combustible waste materials, the heating value for garbage provided in Ref. 5 is used.

Since combustion results in the reduction of biological and organic ^{14}C to CO_2, restriction in the specific activity of the waste materials is established because incineration may result in the release of dust and smokes which may contain unoxidized particles of carbon. These particles may be inhaled and fixed in the lungs causing long-term radiation exposure.

In the calculation for ^{3}H, it is assumed that the concentration of ^{3}H in the body tissue is equal to that in the environment. Values and assumptions used concerning average radiation worker are taken from data for a reference man provided in Ref. 2.

CALCULATIONS FOR ^{3}H

The calculations for ^{3}H are based on the following assumptions:

a. The Average Relative Humidity of Air is 70%.
b. Air Temperature is 25°C.
c. The permissible concentration of ^{3}H in air within a restricted area is the Derived Air Concentration (DAC) which is 8×10^5 Bq-m^{-3} as provided in Ref. 1.
d. Vapor Pressure of H_2O @ 25°C is 101353 Pascals.
e. Average Energy for ^{3}H is 0.005 MeV
g. Weight of an average radiation worker 70 Kg with body containing 7 kg of hydrogen, approximately 4.8 kg of which is in body water and 2.2 kg in organic molecules (4).
h. Annual exposure limit is 5 Rems.
i. The heating value of combustible material is 4440 calories per Kg. (5). The corresponding theoretical volume of dry air required in m^3 per Kg combustible materials is 4.55 (Table I).

Restriction on Specific Activity

The body burden corresponding to the limiting annual dose rate will be calculated as follows:

$$\frac{5\,Rems}{year} \times \frac{100\,ergs-q^{-1}}{1\,Rem} \times \frac{1 MeV}{1.6 \times 10^{-6}\,ergs} \times \frac{dis}{0.006\,MeV} \times \frac{1\,year}{3.2 \times 10^{7}\,sec} \times \frac{1\,Bq-sec}{1\,dis.} \times \frac{7 \times 10^{4}\,g}{1\,ave.\,worker} = \frac{1.14 \times 10^{8}\,Bq}{ave.\,worker}$$

The resulting limiting activity will be obtained from the following:

$$\frac{1.14 \times 10^{8}\,Bq}{1\,ave.\,worker} \times \frac{1\,ave.\,worker}{7 Kg\,H} \times \frac{2\,Kg\,H}{118\,Kg\,H_2O} \times \frac{1\,Kg\,H_2O}{1000\,ml} \times \frac{7\,Kg\,H}{[4.8\,Kg + 0.85(2.2\,Kg)]} = 1898\ Bq-ml^{-1}$$

Concentration Limit for Incineration

The molar density of dry air can be calculated as follows:

$$1\,m^3 \times \frac{273}{298} \times \frac{454q-mole}{10.052\,m^3} = 41.4\,mole-m^{-3}$$

The resulting ratio between the amount of water vapor per unit volume of air will be as follows:

$$\frac{moles\ H_2O}{m^3\ dry\ air} = 0.7287$$

The ratio between the volume of moist air to the volume of dry air will be:

$$\frac{m^3\ wet\ air}{m^3\ dry\ air} = 1.006$$

The maximum concentration of ^{3}H in the incinerable wastes will be:

$$\frac{8 \times 10^{5}\,Bq}{m^3\ wet\ air} \times \frac{1.006\,m^3\ wet\ air}{m^3\ dry\ air} \times \frac{4.55\,m^3\ dry\ air}{Kg.\ combustibles} = 3.66 \times 10^{6}\ \frac{Bq}{Kg\ combustibles}$$

CALCULATIONS FOR ^{14}C

The calculations for ^{14}C are based on the following assumptions:

a. The largest particle that can penetrate the lungs and become fixed has a diameter of 10 u.
b. Beta ray from the 10 u particle will be distributed in the tissue within a sphere of 40 u.
c. Specific gravity of tissue is 1.0 (6).
d. The specific gravity of the carbon particle is assumed to be identical to graphite which is 2.25.
e. Annual dose limit for the lungs of an adult workers is 500 mSv (50 Rems) (1).
f. Average Energy of ^{14}C is 0.049 MeV.
g. The permissible concentration in air within a restricted area is the Derived Air Concentration (DAC) which is 3×10^6 Bq-m^{-3} (1).
h. The Theoretical Volume of dry air in m^3 needed in 1 Kg of combustible materials is 4.55 following instruction in Table I using Heating Value of Garbage provided in Ref. 5.
i. Air temperature is 25°C.

The theoretical volume of dry air required per Kg of combustible materials for each substance considered was calculated on the assumption that Air Temperature is 25°C.

TABLE I
Theoretical Volume of Dry Air Required for Some Selected Combustible Substances

Combustible Substance	Heating Value Calories/g	Theoretical Air Volume Cu. m./Kg
Amorphous C	8075.25	9.63
Carbon Monoxide (CO)	2413.14	2.06
Methane (CH_4)	13296.07	14.45
Benzene (C_6H_6)	10082.13	11.11

The following relationship was developed using Linear Regression:

$$V = 0.0011HV - 0.3368 \qquad \text{(Eq. 1)}$$

where:

V = Volume of dry air in cubic meter per Kg. of, combustible materials

HV = Heating Value in calories per gram of substance

Restriction on Specific Activity

The volume occupied by a 10 u particle is:

$$\frac{4}{3} \times 3.1416 \times [5\,u \times 10^{-4}\,cm{-}u^{-1}]^3 = 5.24 \times 10^{-10}\,cm^3$$

The volume of a 40 u spherical tissue that will be exposed to beta ray will be determined as follows:

$$\frac{4}{3} \times 3.1416 \times [20\,u \times 10^{-4}\,cm{-}u^{-1}]^3 = 3.35 \times 10^{-}8\,cm^3$$

The average energy released by ^{14}C will be:

$$\frac{0.049\,MeV}{dis} \times \frac{2.22 \times 10^{12}\,dpm}{1\,Ci} x \frac{60\text{ min}}{1\,h} = 6.5268 \times 10^{12}\,Mev{-}Ci^{-1}{-}h^{-1}$$

The energy emitted by the 10 u particle will be:

$$6.5266 \times 10^{12}\,Mev{-}Ci^{-1-h^{-1}} \times 2.25\,g{-}cm^{-3} \times 5.24 \times 10^{-10}\,cm^3 = 7695\,Mev{-}Ci^{-1}{-}h^{-1}$$

The approximate mass of tissue which will be exposed will be:

$$3.35 \times 10^{-8}\,cm^3 \times 1\,g{-}cm^{-3} = 3.35 \times 10^{-8}\,g$$

The dose that will be received by the tissue from Beta radiation will be:

$$\frac{76694.86\,MeV{-}g{-}Ci^{-1}{-}hr^{-1}}{3.35 \times 10^{-8}\,g\,tissue} = 2.3 \times 10^{11}\ Mev{-}Ci^{-1}{-}h^{-1}$$

The energy of material corresponding to the annual dose limit will be:

$$\frac{50\,Rads}{year} \times \frac{1\,year}{365\,days} \times \frac{1\,day}{24\,hours} \times 6.242 \times 10^7\,MeV{-}g^{-1} = 3.56 \times 10^5\,MeV{-}g^{-1}{-}h^{-1}$$

The limiting specific activity of the incinerable radioactive waste must be:

$$3.56 \times 10^5\,\frac{MeV}{g{-}hr} \times \frac{1\,Ci{-}hr}{2.3 \times 10^{11}\,MeV} \times \frac{3.7 \times 10^{10}\,Bq}{1\,Ci} = 5.73 \times 10^4\,Bq{-}g^{-1}$$

Concentration Limit for Incineration

The concentration limit for the incinerable materials will be:

$$3 \times 10^6\,Bq{-}m^{-3} \times \frac{4.55\,m^3\,dry\,air}{kg.\,combustibles} \times \frac{1.006\,m^3\,wet\,air}{m^3\,dry\,air} = \frac{1.37 \times 10^7\,Bq}{Kg.\,combustibles}$$

RECOMMENDATIONS

Combustible wastes containing ^{14}C or ^3H may be incinerated in concentration that do not exceed 1.37 x 10^7 Bq per Kg of combustible materials for ^{14}C or 3.66 x 10^6 Bq per Kg of combustible materials for ^3H. Since non-radioactive combustible wastes are usually incinerated together with the radioactive components, the calculations presented made use of data concerning combustible garbages. The combustion, however, should be supported by auxiliary fuel.

Restrictions on specific activity of ^3H or ^{14}C are determined because of the possibility of formation of radioactive particles. Incineration of ^{14}C wastes may produce dusts or smokes containing unoxidized Carbon which when inhaled may be deposited in the lungs. Incineration of ^3H may cause contamination of the water vapor in air which when taken in by inhalation will be distributed in the body fluid.

In view of the above, it is recommended that waste materials containing ^{14}C burned in an ordinary incinerator must contain ^{14}C of activity 5.73 x 10^4 Bq per gram of Carbon in the region of highest ^{14}C concentration. On the other hand, materials containing ^3H must have a specific activity of 1898 Bq per ml of water of liquid wastes absorbed in the region of highest concentration.

In the absence of specific requirement or regulation to be followed in the design of the incinerator, it is recommended that the height of the stack be such that no buildings within the fifty meter radius will be directly exposed to the flume. It has always been recommended in any combustion process to supply air in at least 100% excess. This will minimize the hazards resulting from the possible formation of unoxidized particles and will reduce the concentration of the radioactive materials in the exhaust gases. The exhaust system should be provided with an appropriate gas filter to prevent releases of unburned particulates in the atmosphere.

Periodic sampling of the stack gases taken at a selected exit point of the incineration should be conducted. Counting result should be such that the recommended DAC value will not be exceeded. Activity measurement of ash samples should be conducted to ensure complete combustion of radioactive components. Provisions should be established for safe handling of contaminated ashes and to re-charge them to the incinerator for re-burning.

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