Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 10-11, 2007

Estimation of the Production of Ozone and Nitric Acid in a Proton Accelerator Facility of the Proton Engineering Frontier Project

Cheol Woo LEE, Young-Ouk LEE, Young-Sik CHO
Nuclear Data Evaluation Lab., Korea Atomic Energy Research Institute,
105 Deogjin-dong, Yuseong-gu, Daejeon 305-353, cwl@kaeri.re.kr

1. Introduction

The operation of the high-energy particle accelerator leads to the production of not only radioactive gases, but also radiolytic noxious gases, such as ozone and nitrogen compounds in the air of the facility. Of the radiolytic products, ozone production is usually taken into account for the evaluation of the heath hazard in the operation of the particle accelerator facilities, owing to its high radiolytic yield and much lower maximum acceptable concentration. Nitrogen compounds do not commonly constitute a heath hazard but are of important concern because of its chemical properties of being a strong acid and a strong oxidizing agent. Among the nitrogen compounds, nitric acid is a principal radiolytic compound produced in large quantities and has a potential for the corrosion of the components in the facility[1]. It has founded that at a high-energy proton accelerator facility, nitric acid was produced in the radiolysis of air in beam-loss region. The nitric acid was desorbed on the surface of the magnet, walls, floors[2]and local lead shields[3]. It also tends to be more uniform through the tunnel due to air circulation.

In these backgrounds, the production of ozone and nitric acid in proton accelerator facility from Proton Engineering Frontier Project (PEFP) was estimated in this study, because very strong radiation environment leads to an abundant production of nitric acid in such a high-current proton accelerator[4].

2. Methods

The formation of radiolytic products, ozone and nitrogen compounds can be commonly characterized by the so-called G-value which is the number of molecules formed per 100 eV of the radiation energy absorbed. And moisture in the air is important to the formation of the nitric acid because of the $OH\cdot$ from the moisture.

The energy deposition in the irradiated air from gamma-ray is simulated by using MCNPX code. To estimating ozone and nitric acids, the theoretical formulation and various empirical constants from the experiments of LEP[5], L.N. LESS and A. J. Swallow[1] and Y. Kanda[6] were used

3. Calculations & Results

3.1 Energy Deposition calculation

The most considerable beam-loss region is beam dump rooms for the accelerator facility of the PEFP. From the MCNPX code simulations, the energy deposition in the air from gamma-ray was calculated with the previous study[7].

The information on the energy deposition in the beam dump rooms was shown in Table 1.

Table 1. Characteristics of the beam dump rooms and gamma-ray energy deposition in the PEFP

Room	Dump 1	Dump 2
Proton Energy [MeV]	20	100
Beam Current [mA]	4.8	1.6
Target Material	C, Cu	Cu
Room Volume [m^3]	40	96
γ-ray Flux [#/sec]	1.09×10^{14}	4.23×10^{14}
Energy Deposition [eV/cm^3]	1.71×10^{7}	5.40×10^{7}

3.2 Estimating the Concentration of Ozone

The equation (1) was used in estimating the concentration of ozone.[5]

$$N_{(t)} = \frac{IG}{\alpha + kI + Q}\left(1 - e^{-(\alpha+kI+Q)t}\right) \quad \cdots\cdots(1)$$

where,

$N_{(t)}$: Concentration of ozone, [$/cm^3$]
I : Energy deposition in the unit irradiated volume, [$eV/cm^3 sec$]
G : Number of molecules of ozone per deposited energy of gamma-ray, [#/eV]
α : Dissociation constant of ozone, [/sec]
k : Decomposition constant per radiation, [cm^3/eV]
Q : Ventilation rate for the irradiated volume, [/sec]

From equation (1), the saturated concentration of ozone is derived and shown in equation (2)

$$N_{(t)} = \frac{IG}{\alpha + kI + Q} \quad \cdots\cdots(2)$$

The G -value applied in this calculation was in the range from of 6 to 8 (G-value: 6[1], 6.4[4] and 7.4[5]). And 2.3×10^{-4} for α and 1.4×10^{-16} for k were applied in this calculation.[5]. For the ventilation factor, Q, was

neglected from assuming no ventilation in this calculation. The estimated concentration of ozone is shown in Figure 1.

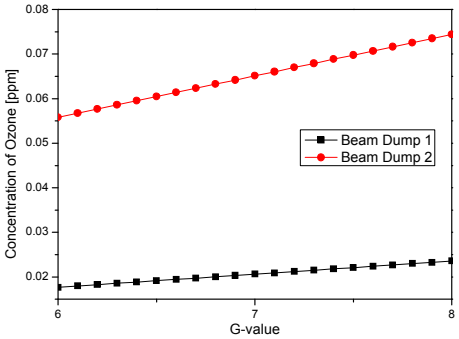

Figure 1. Saturated concentration of ozone produced in beam dump room 1 and 2 with the G-value of the range from 6 to 8

3.3 Estimating the Concentration of Nitric Acids

The radiolysis of air produces active species such as ions, excited molecules and electrons. The predominant reactive products of the secondary reactions of these species are O and N atoms. In dry air, the production mechanisms of the radiolysis products follow as.

$$O + O_2 + O_2 \rightarrow O_3 + O_2$$
$$N + O_2 \rightarrow NO + O$$
$$NO_2 + O + (M) \rightarrow NO_3 + (M)$$
$$NO_2 + NO_3 \leftrightarrow N_2O_5 + (M)$$

In the case of moist air, HNO_2 and HNO_3 are produced additionally because of $OH\cdot$ from moisture. The production mechanisms for these species follow as.

$$NO + HO_2 \rightarrow NO_2 + OH$$
$$NO + OH + (M) \rightarrow HNO_2 + (M)$$
$$NO_2 + OH + (M) \rightarrow HNO_3 + (M)$$

It is known that NO_3 reacts rapidly NO_2 to form N_2O_5 , and that NO_3 and N_2O_5 are in rapid equilibrium with each other[8] and the NO_3 concentration was estimated to be less than 0.01 of that of N_2O_5 [6]. In conclusion, the nitrogen-compound in air are produced mainly NO_2 and N_2O_5 for dry air, NO_2 , HNO_2 and HNO_3 for moist air. In this study, the concentration of nitric acid, HNO_3 , was estimated with the G-values of the range from 1 to 2, in the view of shielding concerning worker's maintenance. The estimated results are shown in Figure 2.

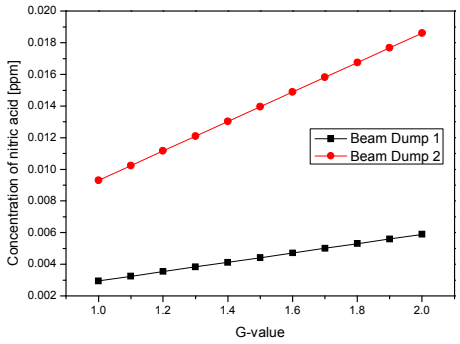

Figure 2. Saturated concentration of nitric acid in beam dump room 1 and 2 with the G-value of the range from 1 to 2

4. Conclusion

The concentrations of ozone and nitric acid produced in the beam dump rooms of the proton accelerator facility of the PEFP were estimated with the variance of the G-value by using MCNPX simulation, theoretical formulation and empirical constants. Now, measurement has not yet performed for PEFP because the facility is not constructed. Therefore, the variance of the G-value was applied to estimate the level of radiolysis products. As a result, it is expected that the concentrations of the radiolysis product in the interested area is under 0.1 and 0.02 ppm for ozone and nitric acid.

REFERENCES

[1] L. N. Less and A. J. Swallow, Estimating the Hazard due to Radiolysis Products from Air, Nucleonics, Vol. 22(9), p. 58, 1964.
[2] Y. Kanda, T. Miura and H. Nakajima, Observation of gaseous nitric acid production at a high-energy proton accelerator facility, Radiation Physics and Chemistry, Vol. 73, p. 213, 2005.
[3] T. Momose and H. Ishimaru, Radiation damage in TRISTAN vacuum system, Journals of Vacuum Science Technology, Vol. A 9 (4), p. 2149, 1991
[4] Y. Kanda, et al., Measurement of radiolysis yield of nitric acid in air, Radiation Physics and Chemistry, Vol. 74, p. 338, 2005.
[5] A. Fasso et al., Formation D'ozone dans L'anneau Principal du LEP et sa Dispersion dans L'atmosphere par le System de Ventilation, LEP Note 379, CERN, 1982.
[6] Y. Kanda, T. Momose and M. Taira, Charaterization of radiolysis products from air at a high-energy electron-positron storage ring, Radiation Physics and Chemistry, Vol. 48, p. 49, 1996.
[7] C. W. Lee, Y. Lee and Y. Cho, Analysis of the Radiation Field for 100 MeV Beam Line of the PEFP, Transactions of the Korean Nuclear Society Autumn Meeting Busan, Oct. 27-28, 2005, KNS
[8] R. A. Graham and H. S. Jonston, The photon chemistry of NO3 and the kinetics of the N2O5-O3 system, Journals of Physics Chemistry, Vol. 82, p. 254, 1978.