Letter to the Editors

Computer Simulation to Predict Ozone Concentration in an Electron Accelerator Room[†]

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1. Introduction

Ozone (O₃) and nitrogen oxide compounds are generated through the radiolysis of air by the operation of an electron accelerator. Of these gases, O₃ is the most toxic and may be produced in such quantities as to constitute a health hazard and damage of the machine components by corrosion within the accelerator room¹⁾.

We have developed a computer simulation method to predict O₃ concentration in an electron accelerator room. This method consists of two computer codes, an electron-photon coupled radiation transport code EGS4²⁾ and a numerical integration code FACSIMILE³⁾. The EGS4 code calculates the absorbed dose in the air by the Monte Carlo method for the geometry and electron beam energy under consideration. On the basis of the absorbed dose, FACSIMILE solves a

set of chemical kinetic equations expected for the radiolysis of air. This paper describes a gas phase radiation chemistry model, together with a benchmark calculation to show its validity, to describe the generation of O₃. The chemistry model along with EGS4 simulation is applied to predict O₃ concentration in an electron accelerator room. The prediction is compared with both experiment and empirical equation, and the validity of the present method is discussed.

2. Methods

In order to simulate the chemical reactions resulting from the radiolysis of air, the absorbed dose in the air, the number of the primary radiolytic products, and a set of kinetic equations with their rate constants are required. The absorbed dose in the air was simulated using the EGS4 code. In the present modeling, chemical reactions of nitrogen, oxygen and water molecules and their radiolytic products were considered. The number of the primary radiolytic products was calculated based on *G*-values, the number of species formed or destroyed per 100 eV of radiation energy absorbed. The *G*-values employed in this model

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were those given by Willis et al.⁴⁾. The relative amount of energy imparted to each molecule was defined by the ratios of their stopping powers. In order to simplify the chemistry model, we ignored electronic states of ions and excited molecules, as Willis et al. did⁵⁾. The reaction scheme involves 94 reactions⁶⁾ which are adequate for the range of conditions of interest. The rate constants for radical-radical and radical-molecule reactions were obtained from the NIST chemical kinetics database⁷⁾. These rate constants contain a temperature dependence factor, so that the reaction scheme is applicable over a wide temperature range. The chemical kinetic equations were simultaneously solved using FACSIMILE and O_3 concentration was calculated as a function of time.

3. Results and Discussion

It is necessary to benchmark the radiation chemistry model before it can be applied with confidence for prediction purposes. We calculated using the present radiation chemistry model the G -value for the formation of O_3 to compare it with that of pulse radiolysis of air⁵⁾. Moist-free air was irradiated by a single pulsed electron beam at 298 K and 1.0×10^5 Pa. The total dose, imparted for 60 ns, was 1.7×10^3 Gy. The pulse shape of the electron beam was approximated by a step function with 5 ns steps.

Figure 1 shows calculated concentrations of ionic and neutral species during and after the pulsed electron beam irradiation. The peak of the pulse, shown as DR in Fig. 1(a), corresponds to 6.6×10^{10} Gy/s. The concentrations of O_2^+ and

Fig. 1 Calculated concentrations of the ionic and neutral species.
(a) During irradiation. DR is the absorbed dose rate.
(b) After irradiation.

electron increase and decrease concomitantly by the pulse, but N_2^+ does not increase in spite of the primary radiolytic product, because of charge transfer to O_2 ,

and/or neutralizations by an electron and O_2^- ,

The concentration of O atoms existing immediately after the pulse decreases very rapidly whereas the concentration of N atoms falls much more slowly (Fig. 1 (b)). The O_2^+ is neutralized by an electron or O_2^- and produces O atoms. The resultant O atoms react with O_2 molecules and produce O_3 ,

where M is a third body. The N produced from the neutralizations of N_2^+ reacts with O_2 and NO to produce O atoms that build up the O_3 concentration in the later stages.

The reaction of N with NO also depresses the consumption of O_3 caused by the following reac-

tion.

The modeled concentration of O_3 reached almost a steady state at 3 ms after the pulse, and $G(O_3)$ was 11.2. The $G(O_3)$ is in good agreement with the experimental result of $10.3 \pm 0.5^{(5)}$.

The chemistry model along with EGS4 simulation was applied to predict O_3 concentration in an accelerator facility. The experiment used for comparison was the measurement of O_3 concentration in a target room of an electron linear accelerator (linac) at the Rensselaer Polytechnic Institute (RPI)⁽⁸⁾. An electron beam of 50 MeV with 80 μ A average current was incident on a 4 cm thick beam stopper of lead, and O_3 concentration in the target room was measured to be 2.7×10^{-8} mol/l. EGS4 simulation was performed for the above conditions and the absorbed dose rate in the air of the room was calculated and found to be 68 mGy/s. By using the absorbed dose rate, the concentration of O_3 in the air, of 30% relative humidity at 298 K, was calculated as shown in Fig. 2.

The irradiation time in the figure corresponds to the average resident time of air in the target room. The time is mainly determined by the room

Fig. 2 O_3 concentration as a function of the irradiation time.

size and ventilation rate, and the air flow in the room, which is influenced by the room geometry and arrangement of accelerator equipment, would also affect it. From the room size and ventilation rate, the irradiation time is roughly estimated to be 4–7 min. The corresponding O_3 concentration in Fig. 2 ranges from 8.0×10^{-9} to 1.2×10^{-8} mol/l. The concentration agrees with the observed value (2.7×10^{-8} mol/l) within a factor of 4. The estimation of the irradiation time does not take the effect of the air flow in the room into account, because the information is not mentioned in the literature⁸⁾. However, it is considered from the above result that the present method gives reasonable estimation for the O_3 concentration in the RPI electron linac.

The generation rate of O_3 was also calculated using an empirical equation presented in the IAEA Technical Report¹⁾ under the same conditions. The calculated generation rate was 5.8×10^{-8} mol/l min; that is about 40 times greater than the predicted one from the present modeling method (1.5×10^{-9} mol/l min). There are two possible reasons for the discrepancy between the empirical equation and the present method. The first reason is the difference in G -value employed. In the empirical equation, a value for $G(O_3)$ of 10.3, derived from the high dose rate pulse radiolysis of air⁵⁾, is used. The $G(O_3)$ is dose rate dependent and this value is applicable for dose rates ranging from 10^8 to 10^{11} Gy/s. In the RPI linac the dose rate is fairly low (68 mGy/s), so that $G(O_3)$ should be 6 at most. On the other hand, the present method considers the dose rate dependence of $G(O_3)$ in the chemistry model

itself. The second reason is the estimation of the absorbed dose in the air. The empirical equation is generalized for all target materials and other parameters, so that the deposited energy in the air is overestimated. In the present method, the deposited energy in the air was calculated from the Monte Carlo simulation that takes into account the incident beam energy and the dimensions and materials of the target and target room; therefore the absorbed dose rate in the air is more accurate. It is considered from the above comparison that although the empirical equation of the IAEA Technical Report is convenient to obtain a rough estimate of O_3 concentration, the present method is useful for calculating a realistic value when the geometry and materials of the accelerator components and the operating conditions of the machine are given.

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