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TOXIC GAS PRODUCTION AT ELECTRON LINEAR ACCELERATORS*

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ABSTRACT

Data relevant to toxic gas production by ionizing radiation in air are reviewed. Recent studies of radiolytic yields have found higher values of G for ozone production in air than have usually been assumed in radiation-protection planning. Calculations are made for ozone production in four types of radiation environments common at electron linear accelerators.

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Toxic gases produced in air by ionizing radiation are listed in Table 1. Of these, ozone is the most toxic, and may be produced in such quantities as constitute a health hazard within the radiation room of an electron linear accelerator installation. Ozone and nitric acid formed from the interaction of nitrogen oxides and water vapor may also gradually damage equipment by corrosion. Of the gases listed, ozone production will almost always be the limiting factor, owing to its much lower TLV (0.1 ppm) (ACGIH, 1971a; ACGIH, 1971b; Occupational Safety and Health Administration, 1974) and relatively high radiolytic yield.

The production rate p of a chemical species is related to the integral dose to the air volume irradiated via the yield G, the number of molecules formed per unit of energy deposited. Recent work has found the radiolytic yield of O_3 in pure oxygen to be around G = 13 molecules per 100 eV (Riley, 1962; Johnson and Warman, 1964; Shah and Maxie, 1966; Meaburn et al., 1968; Sears and Sutherland, 1968; Ghormley et al., 1969; Boyd et al., 1970; Willis et al., 1970). In air, an efficient charge-transfer mechanism (positive charge transfer from N_2^+ ions to O_2) enhances the O_3 yield, and G-values are believed to be in the range 7.4-10.3, depending on the instantaneous dose rate, even though air is only one-fifth oxygen (Willis et al., 1970). Such values imply that more than one ozone molecule is formed for every ion formed (34 eV per ion pair). This yield represents an efficient conversion mechanism for O_3 and the yields for the other toxic gases are smaller.

Ozone decomposes spontaneously, reacts chemically with air impurities and other materials, and is decomposed by the radiation itself. The effective decomposition time will therefore depend on room size, wall material, temperature, impurities in the air, and ozone concentration. The decomposition time

has been found to be about 50 min in a typical research installation (George et al., 1965).

Provisions for mitigating the effects of toxic gases are the same as those for reducing concentrations of radioactive gases and include reduction of the integral dose imparted to the air by limiting the beam path length and increasing the ventilation. The pattern of accelerator usage and radiation-room occupancy (no persons present during irradiation times and no ozone produced during off-times) provides considerable protection in itself and should be considered in assessing these risks. The smell of ozone can be detected at concentrations in the range 0.02-0.05 ppm, so that any room free of the characteristic odor may be regarded as safe from these radiolytic gases.

I. Concentration Buildup and Removal

The first step in estimating the concentration of ozone is the determination of its production rate p for the situation in question (see examples below). In this discussion we express p in units of liters per minute, which is convenient when the room volume is also expressed in liters (a large room is on the order of 10^6 liters), and buildup and decay times are expressed in minutes. Once the production rate p has been determined, factors to account for buildup and removal are applied, to determine the concentration at any time, just as for radioactivation. The pattern of accelerator use should be considered here.

The concentration buildup is described by the formula

$$C(t_b) = \frac{p\bar{T}}{V}\,[1 - \exp(-t_b/\bar{T})] \qquad (1)$$

where C is the concentration (dimensionless), V is the room volume in liters, t_b is the "on-time" or buildup time in minutes, and $\bar{T}$ is determined by the removal processes:

$$\bar{T} = \frac{T(\text{vent}) \cdot T(\text{decomp})}{T(\text{vent}) + T(\text{decomp})} , \tag{2}$$

where T(vent) is the room volume divided by the air volume exhausted per unit time. For ozone, we take the decomposition time T(decomp) to be 50 min. In the usual situation, the ventilation time T(vent) is sufficiently less than the decomposition time that it essentially controls ozone removal, and $\bar{T} \approx T(\text{vent})$ would be a conservative value.

Eq. (1) shows that the concentration buildup for running times short compared to the effective removal time $\bar{T}$ is proportional to the buildup time:

$$C(t_b) = \frac{pt_b}{V} \quad (\text{short running times; } t_b \ll \bar{T}) \tag{3}$$

The saturation concentration C_s derived from Eq. (1) for long buildup times is evidently

$$C_s = \frac{p\bar{T}}{V} \quad (\text{saturation; } t_b \gg \bar{T}) . \tag{4}$$

In the case of no ventilation, the saturation concentration of ozone is proportional to the effective decomposition time:

$$C_s = \frac{p \cdot (50 \text{ min})}{V} \quad (\text{ozone, no ventilation}) \tag{5}$$

Following turnoff, the concentration will diminish as an exponential in the decay time t_d:

$$C(t_d) = C_{\text{turnoff}} \exp(-t_d/\bar{T}) . \tag{6}$$

II. Production Rates

Production rates for toxic gases and their radiological safety implications have been discussed in Brynjolfsson and Martin, 1971; Brobeck, 1968; NBS and NCRP, 1964; NCRP, 1977; Ladu et al., 1965 - trans. 1968; Less and Swallow, 1964. We consider here the production rate of ozone for four cases frequently encountered at electron linear accelerators.

A. External Electron Beam (No Showering)

The rate of ozone production by an external electron beam is based on an assumed average collision mass stopping power of 2 MeV g^{-1} cm^2, the G value of 10.3 molecules per 100 eV, corresponding to high instantaneous dose rates to air within a narrow beam path. These assumptions lead directly to

$$p(\text{liters min}^{-1}) = 350\ I(\text{amps})\ L(\text{m}) , \tag{7}$$

where I is the average electron current in amperes and L is the path length in air (meters). This situation applies whenever the electron beam does not cause showering in any thick materials before entering the air. This is the case if the beam window is thin ($X < X_0$) or if the energy E_0 is less than about twice the critical energy E_c for the material(s) through which the beam passes. The critical energy is approximately given by

$$E_c(\text{MeV}) = \frac{800}{Z + 1.2} , \tag{8}$$

where Z is the atomic number of the material in question. Currents accelerated at almost all electron linacs are capable of producing ozone concentrations above the TLV in an average room, if the air path is sufficiently long.

B. External Electron Beam with Showering

Whenever a high-energy electron beam (E_0 greater than about twice E_c) passes through a solid material (such as a beam window, pipe, or flange) before entering the air path, buildup of particle fluence can enhance the ozone production considerably, easily by an order of magnitude or more in some cases. The extreme situation is where the thickness of solid material corresponds to the shower maximum. In this case the enhancement factor will be on the order of E_0/E_c (Rossi, 1952):

$$\text{Maximum enhancement of electron fluence} = \frac{0.31(E_0/E_c)}{[\ln(E_0/E_c)-0.37]^{\frac{1}{2}}} \quad \text{(Approx. B)} \qquad (9)$$

The shower maximum occurs at a thickness measured in radiation lengths, which can be estimated by

$$X(\text{shr max})/X_0 = 1.01[\ln(E_0/E_c)-1] \quad \text{(Approx. B)} \qquad (10)$$

It should be assumed that the maximum enhancement will occur, unless there is positive assurance that these conditions cannot be achieved.

The production rate given by Eq. (7) should be multiplied by the enhancement factor.

C. Bremsstrahlung or Diffuse Electron Beams Specified in Units of Exposure Rate or Dose Rate

At radiotherapeutic and radiographic installations, a bremsstrahlung or diffuse electron beam is usually specified in terms of exposure rate or absorbed dose rate (R min^{-1} m^2 or rads min^{-1} m^2), in a field area $S(\text{m}^2)$ measured at 1 m from the target. Here we make the simplifying assumption that the absorbed dose (rads) in the irradiated air at the same distance is equal to the exposure (R) or dose to tissue in rads (in the case of a therapeutic situation). This leads directly to the production rate for ozone:

$$p(\text{liters min}^{-1}) = 2.1 \cdot 10^{-7} \dot{D}(\text{rads min}^{-1}\text{m}^2)S(\text{m}^2)L(\text{m}) , \qquad (11)$$

assuming G = 7.4 molecules per 100 eV, appropriate for "low" dose rates typically found in situations where the accelerator output is specified in this manner. The parameters of representative therapeutic or radiographic accelerators indicate that the ozone production rate is generally not critical for standard installations, under normal operating conditions.

D. Uncollimated Bremsstrahlung Beam from an Optimum Target, where the Incident Electron Beam Power is Specified in kW

For this case, we make use of the approximation for the absorbed-dose rate from bremsstrahlung from optimum targets: $\dot{D}(\text{rads min}^{-1}\text{m}^2\text{kW}^{-1}) = 80\ E_0^2$, where $\dot{D}$ is the bremsstrahlung dose rate and E_0 is in MeV. We further assume that all of the bremsstrahlung energy is contained within a solid angle given by $\pi(100^\circ/E_0)^2 \approx 10 \cdot E_0^{-2}$ steradians. These approximations, together with a G-value of 7.4 molecules per 100 eV directly yield

$$p(\text{liters min}^{-1}) = 1.7 \cdot 10^{-4} L(\text{m}) P(\text{kW}) , \qquad (12)$$

where the electron energy E_0 cancels and we have a production rate proportional to the incident electron beam power P in kW, independent of energy.

The approximations used are valid for $E_0 \lesssim 50$ MeV. In this energy range, the lower G value of 7.4 molecules per 100 eV would normally hold. At higher energies, the effect of the showering in the target material complicates the situation and the discussion given above (IIC) would apply. The parameters of typical research accelerators are such that ozone production by bremsstrahlung may be a problem without adequate ventilation.

References

American Conference of Governmental Industrial Hygienists (ACGIH), 1971a, Threshold Limit Values of Airborne Contaminants and Physical Agents with Intended Changes Adopted by ACGIH for 1971 (ACGIH: Cincinnati).

American Conference of Governmental Industrial Hygienists (ACGIH), 1971b, Documentation of the Threshold Limit Values for Substances in Workroom Air, 3rd ed. (ACGIH: Cincinnati).

Boyd, A. W., Willis, C., and Cyr, R., 1970, "New Determination of Stoichiometry of the Iodometric Method for Ozone Analysis at pH 7.0," Anal. Chem. 42 6, 670. (G = 12.8 ± 0.6.)

Brobeck, W. M., 1968, Particle Accelerator Safety Manual (U.S. Dept. of Health, Education, and Welfare: Rockville, Md.).

Brynjolfsson, A., and Martin, T. G., III, 1971, "Bremsstrahlung Production and Shielding of Static and Linear Electron Accelerators Below 50 MeV. Toxic Gas Production, Required Exhaust Rates, and Radiation Protection Instrumentation," Int. J. Appl. Radiat. Isotopes 22, 29.

George, A. C., Breslin, A. J., Haskins, J. W., Jr., and Ryan, R. M., 1965, "Evaluation of the Hazard from Radioactive Gas and Ozone at Linear Electron Accelerators," Proc. USAEC 1st Symposium on Accelerator Radiation Dosimetry and Experience, Brookhaven, 3-5 Nov 1965, USAEC Report No. CONF-651109 (Washington, D.C.),539.

Ghormley, J. A., Hochanadel, C. J., and Boyle, J. W., 1969, "Yield of Ozone in the Pulse Radiolysis of Gaseous Oxygen at Very High Dose Rate. Use of this System as a Dosimeter," J. Chem. Phys. 50 1, 423. (G = 13.8 ± 0.7.)

Johnson, G. R. A., and Warman, J. M., 1964, "Formation of Ozone from Oxygen by the Action of Ionizing Radiations," Discussions Faraday Soc. 37, 87. (The value 12.8 given by these authors has been corrected to 10.2 ± 0.6 by Ghormley et al. - see preceding reference.)

Ladu, M., Pellicioni, M., and Roccella, M., 1965, trans. 1968, Produzione e scarico di gas tossici e radioattivi nel tunnel del Linac de Frascati, Laboratori Nazionali di Frascati del CNEN, Internal Report No. 282; translated as Production and Discharge of Toxic and Radioactive Gases in the Linac Tunnel in Frascati, National Research Council of Canada, Ottawa,

Technical Translation No. NRC-TT-1332.

Less, L. N., and Swallow, A. J., 1964, "Estimating the Hazard Due to Radiolytic Products from Air," Nucleonics 22 9, 58.

Meaburn, G. M., Perner, D., LeCalve, J., and Bourene, M., 1968, "A Pulsed-radiolysis Study of Atomic Oxygen Reactions in the Gas Phase," J. Phys. Chem. 72 11, 3920. (G = 10.5.)

National Bureau of Standards and National Council on Radiation Protection and Measurements, 1964, Shielding for High-Energy Electron Accelerator Installations, NBS Handbook No. 97 and NCRP Report No. 31, p. 8.

National Council on Radiation Protection and Measurements, Scientific Committee No. 22, 1977, Radiation Protection Design Guidelines for 0.1 - 100 MeV Particle Accelerator Facilities, Report No. NCRP-51, pp. 133-134.

Occupational Safety and Health Administration, 1974, Occupational Safety and Health Standards, U.S. Department of Labor, Washington, D. C., Federal Register 39 125, 23541.

Riley, J. F., 1962, Chemistry Division Annual Report, Oak Ridge National Laboratory, Report No. ORNL-3320 (Oak Ridge, Tennessee). (This author reports G = 12.5.)

Rossi, B., 1952, High-Energy Particles (Prentice-Hall, Inc.: Englewood Cliffs, N.J.), p. 257.

Sears, J. T., and Sutherland, J. W., 1968, "Radiolytic Formation and Decomposition of Ozone," J. Phys. Chem. 72 4, 1166. (G = 10.5 ± 0.5.)

Shah, J., and Maxie, E. C., 1966, "Gamma-Ray Synthesis of Ozone from Air," Int. J. Appl. Radiat. Isotopes 17, 155. (These authors find G = 11 for flowing air.)

Willis, C., Boyd, A. W., and Young, M. J., 1970, "Radiolysis of Air and Nitrogen-Oxygen Mixtures with Intense Pulses: Determination of a Mechanism by Comparison of Measured and Computed Yields," Can. J. Chem. 48, 1515.

Table 1

Toxic Gases Produced by Radiation at Electron Linac Installations

Gas		TLV[a] (ppm)	G (Air)[b] (molecules per 100 eV)		Decomposition[c] Time Assumed (min)
			Low $\dot{D}$	High $\dot{D}$	
Ozone	O_3	0.1	7.4	10.3	50
Nitric oxide	NO	25			
Nitrogen dioxide	NO_2	5[d]	(4.8)	(< 0.15)	
Nitrogen trioxide	NO_3				
Nitrogen tetroxide	N_2O_4	5[d]			
Nitric anhydride	N_2O_5				
Nitrous oxide	N_2O				

(a) Threshold Limit Value. Maximum concentration averaged over any 8-hour shift, assuming 40-hour work week.

(b) Values from Willis et al., 1970. "High dose rate" means instantaneous dose rate (during beam pulse) greater than about $5\ 10^{10}$ rad s^{-1} ($\approx 3\ 10^{24}$ eV $g^{-1}\ s^{-1}$) to air. Theoretical values are in parentheses.

(c) Decomposition time for ozone may depend strongly on size of room and nature of materials present.

(d) Value also represents Ceiling Value: Maximum concentration allowed at any time.