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Ozone Generation in Air during Electron Beam Processing

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Abstract

Ozone, the triatomic form of oxygen, can be generated by exposing normal diatomic oxygen gas to energetic electrons, X-rays, nuclear gamma rays, short-wavelength ultraviolet radiation (UV) and electrical discharges. Ozone is toxic to all forms of life, and governmental regulations have been established to protect people from excessive exposures to this gas. The human threshold limit values (TLV) vary from 60 to 100 parts per billion (ppb) in air, depending on the agency or country involved. Much higher concentrations can be produced inside industrial electron beam (EB) facilities, so methods for ozone removal must be provided. Equations for calculating the ozone yield vs absorbed energy, the production rate vs absorbed power, and the concentration in the air of an EB facility are presented in this paper. Since the production rate and concentration are proportional to the EB power dissipated in air, they are dependent on the design and application of the irradiation facility. Examples of these calculations are given for a typical EB process to cross-link insulated electrical wire or plastic tubing. The electron energy and beam power are assumed to be 1.5 MeV and 75 kW.

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Keywords: ozone; ozone yield; ozone production rate; ozone concentration

1. Ozone yield

The production of a product obtained by radiation processing is proportional to the G value of the irradiated material. The G value is defined as the number of molecules produced per 100 electron volts (eV) of ionizing energy absorbed. The yield in SI units, moles/joule, can be obtained from the G value with the following equation:

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$$Y = G / (100\ e\ N) \quad (1)$$

where Y is the yield in gram moles per joule, e is the number of joules per electron volt (the same number as the electron charge in coulombs), 1.602×10^{-19}, and N is the number of molecules per gram mole (Avogadro's number), 6.022×10^{23}. Therefore:

$$Y = 1.037 \times 10^{-7}\ G\ mol/J \quad (2)$$

The yield in gram moles /kilowatt hour can be obtained by modifying Equation (2) as follows:

$$Y = 1.037 \times 10^{-7} \times 1000 \times 3600\ G \quad (3)$$

$$Y = 3.73 \times 10^{-1}\ G\ mol/kW\ h \quad (4)$$

The yield in kilograms/kilowatt hour, can be obtained by modifying Equation (4) as follows:

$$Y = 3.73 \times 10^{-1}\ G \times (M / 1000) \quad (5)$$

$$Y = 3.73 \times 10^{-4}\ G\ M\ kg/kW\ h \quad (6)$$

where M is the gram molecular weight of the substance produced by the irradiation process [1-5]. The value of M for ozone is three times the molecular weight of oxygen, that is, $3 \times 15.9994 = 47.998$ g/mol.

Reported G values for the production of ozone by irradiating pure oxygen gas range from 9 to 13, but the G values for the production of ozone by irradiating air at normal temperature and pressure are about half the value with pure oxygen [6-11]. A median value of 5.0 for air is used in the examples presented below in this paper. With this lower value, the ozone yield, Y, is given by:

$$Y = 3.73 \times 10^{-4} \times 5.0 \times 48.00 \quad (7)$$

$$Y = 8.95 \times 10^{-2}\ kg/kW\ h \quad (8)$$

2. Ozone Production Rate

In an electron beam irradiation process, the production rate of ozone is proportional to the incident beam power and the fraction of this power absorbed in the irradiated air. An estimate of the power fraction can be obtained by dividing the thickness of the air space by the maximum range of the incident electrons in air. Another method is to multiply the average stopping power of the electrons by the thickness and the density of air. Neither of these methods are accurate for electron energies below 1.5 MeV because the range decreases and the stopping power increases non-linearly as the energy decreases.

A more accurate way is to use a Monte Carlo computer program [12] to calculate the energy deposition per electron in units of MeV cm^2/g or MeV / (g/cm^2) in many thin air spaces between the beam window and the irradiated material. When these energy deposition values are multiplied by the associated area or areal density values Z in g/cm^2, the result is the energy deposition per electron in MeV units in each of the thin air spaces.

The ratio of the accumulated energy depositions in the air spaces, AED, in MeV units to the incident electron energy E in MeV gives the fraction (AED / E) of the electron energy and the beam power absorbed in air. This concept is illustrated in Figures 1 - 4. The beam power absorbed in air P(air) can be obtained as follows:

$$P(air) = P(beam)\ (AED / E)\ kW \quad (9)$$

Equation (9) can be combined with Equation (8) to obtain the ozone production rate in kilograms per hour as follows:

$$\text{Ozone Production Rate} = Y \times P(\text{air}) \text{ kg/h} \quad (10)$$

$$\text{Ozone Production Rate} = 8.95 \times 10^{-2} P(\text{air}) \text{ kg/h} \quad (11)$$

3. Ozone concentration

In this section, the ozone concentration in the effluent air is defined as the dimensionless ratio of the production rate divided by the air exhaust rate. The density of dry air at normal temperature and pressure is 1.205 mg/cm^3 or 1.205 kg/m^3. So, this ratio can be obtained by modifying Equation (11) as follows:

$$\text{Ozone Concentration (weight)} = 8.95 \times 10^{-2} P(\text{air}) / (1.205\ F) \quad (12)$$

$$\text{Ozone Concentration (weight)} = 7.43 \times 10^{-2} P(\text{air}) / F \quad (13)$$

where F is the air exhaust rate in m^3/h. If the exhaust rate F is given in m^3/min, then the ozone concentration by weight is given by:

$$\text{Ozone Concentration (weight)} = 7.43 \times 10^{-2} P(\text{air}) / (60\ F) \quad (14)$$

$$\text{Ozone Concentration (weight)} = 1.24 \times 10^{-3} P(\text{air}) / F \quad (15)$$

According to Avogadro's Law, one mole of any ideal gas occupies the same volume at the same temperature and pressure, regardless of its molecular weight. Therefore, the ozone concentration by volume is less than its concentration by weight by the ratio of the average molecular weight of air, 28.965, to the molecular weight of ozone, 47.998, as follows:

$$\text{Ozone Conc. (volume)} = \text{Ozone Conc. (weight)} (28.965 / 47.998) \quad (16)$$

$$\text{Ozone Conc. (volume)} = \text{Ozone Conc. (weight)} \times 0.603 \quad (17)$$

The average molecular weight of air at normal temperature and pressure is derived from the volume composition of air and the molecular weights of the component gases, as shown in Table 1 below.

TABLE 1. MAIN COMPONENTS OF NORMAL AIR

Air Components	Concentration By Volume (%)	Molecular Weight (g)	Component Weight (g)
Nitrogen	78.084	28.013	21.874
Oxygen	20.947	31.999	6.703
Argon	0.934	39.948	0.373
Carbon Dioxide	0.033	44.009	0.015
Totals	99.998		28.965

4. Examples

In the following examples, the TIGER code of the ITS3 Monte Carlo program is used to calculate the fractions of electron beam power that are absorbed in the beam window, the air space and the irradiated material [12]. The amount of beam power dissipated by electron backscattering can be obtained by subtracting the absorbed power in

the beam window, the air and the irradiated material from the incident electron beam power. The results of these calculations are shown in Table 2 below. It is assumed that the electron energy is 1.5 MeV, the beam current is 50 mA and the beam power is 1.5 x 50 = 75 kW.

TABLE 2. ELECTRON BEAM POWER FRACTIONS

Irradiated Material	Back Scattering	Titanium Window	Air Space	Irradiated Material	Total Fraction
PE	0.0093	0.0157	0.0215	0.9535	1.0000
SS	0.0935	0.0220	0.0651	0.8194	1.0000

4.1 Example 1

In Example 1, the irradiated material is a flat sheet of low-density polyethylene (PE) 0.80 cm thick with a density of 0.92 g/cm^3. This material, in combination with the 40 micron titanium beam window and the 15 cm air space, is thick enough to absorb all of the transmitted electron beam energy. The values of electron energy deposition versus depth in these materials are shown in Figures 1 and 2. The areas under these curves give the absorbed energy in units of MeV per electron. The electron beam power fractions in Table 2 were obtained by dividing the energies in separate areas by the incident electron energy. Therefore, the beam power fraction in the air space is 0.0215 and the beam power deposited in the air space is 75 x 0.0215 = 1.613 kW.

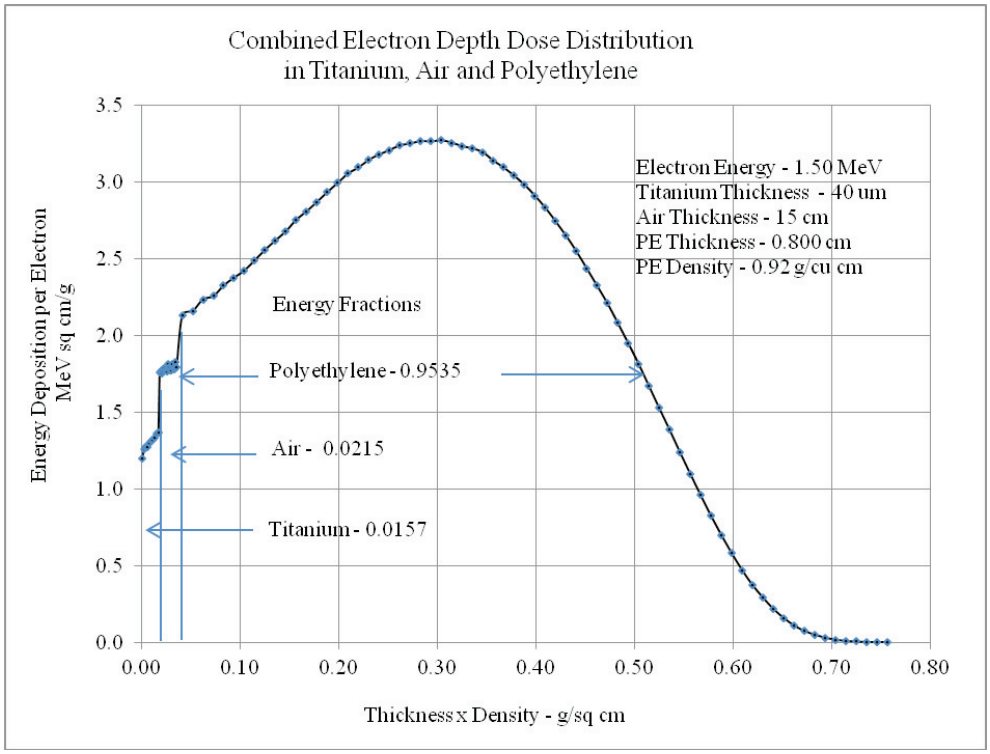

Figure 1. Combined electron depth dose distribution in titanium, air and polyethylene.

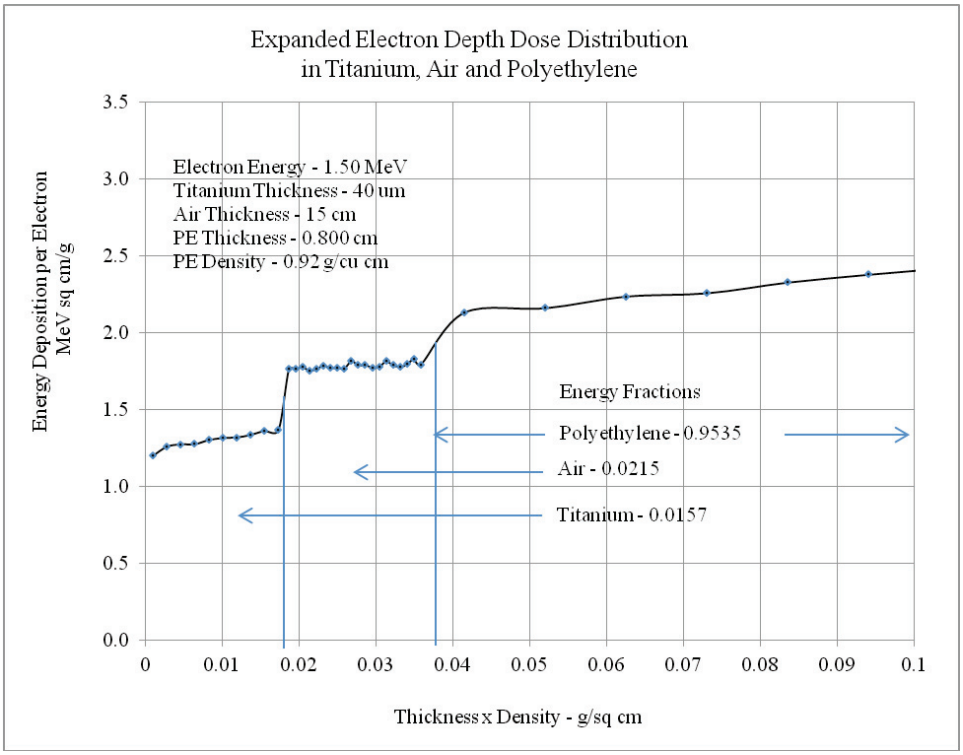

Figure 2. Expansion of Figure 1 to show the energy deposition in the air space in more detail.

OZONE PRODUCTION RATE

The ozone production rate can be obtained by using Equation (11) above and the beam power deposited in the air space, 1.613 kW, as follows:

$$\text{Ozone Production Rate} = 8.95 \times 10^{-2}\ P(\text{air})\ \text{kg/h} \quad \text{(repeat)} \tag{11}$$

$$\text{Ozone Production Rate} = 8.95 \times 10^{-2} \times 1.613 \tag{18}$$

$$\text{Ozone Production Rate} = 1.44 \times 10^{-1}\ \text{kg/h} \tag{19}$$

OZONE CONCENTRATION

The ozone concentration by weight (the ratio of the production rate divided by the air exhaust rate) can be obtained by using Equation (15) above and the beam power deposited in the air space, 1.613 kW, as follows:

$$\text{Ozone Concentration (weight)} = 1.24 \times 10^{-3}\ P(\text{air}) / F \quad \text{(repeat)} \tag{15}$$

$$\text{Ozone Concentration (weight)} = 1.24 \times 10^{-3} \times 1.613 / 850 \tag{20}$$

$$\text{Ozone Concentration (weight)} = 2.35 \times 10^{-6}\ \text{or } 2.35\ \text{ppm} \tag{21}$$

In this example, the air exhaust rate is assumed to be 850 m^3/min or 30,000 ft^3/min by using a typical external dilution fan on the roof of the irradiation facility. The value from Equation (21) is higher than the allowable

threshold limit values (TLV) of 60 to 100 ppb, which depend on the agency or country involved. However, the ozone concentration at ground level outside of the facility will be much less than the allowable TLV because of further dilution in the external air.

4.2 Example 2

In Example 2, the irradiated material is a flat sheet of stainless steel (SS) 0.10 cm thick with a density of 8.007 g / cm^3. This material, in combination with the 40 micron titanium beam window and the 30 cm air space, is thick enough to absorb all of the transmitted electron beam energy. The values of electron energy deposition versus depth in these materials are shown in Figures 3 and 4. The areas under these curves give the absorbed energy in units of MeV per electron. As in Example 1, the electron beam power fractions in Table 2 were obtained by dividing the separate areas by the incident electron energy.

The stainless steel sheet is the beam stop which limits the amount of air that can be irradiated if there is no PE in the beam. In comparison to Example 1, the higher energy fractions from backscattering and power absorption in the titanium beam window are caused by increased electron scattering from the high-density stainless steel beam stop. The higher energy fraction in the air space is mainly caused by its increased thickness, 30 cm versus 15 cm, but the increased scattering from the beam stop also contributes to this effect. From Table 2, the beam power fraction in the air space is 0.0651 and the beam power deposited in the air space is 75 x 0.0651 = 4.883 kW.

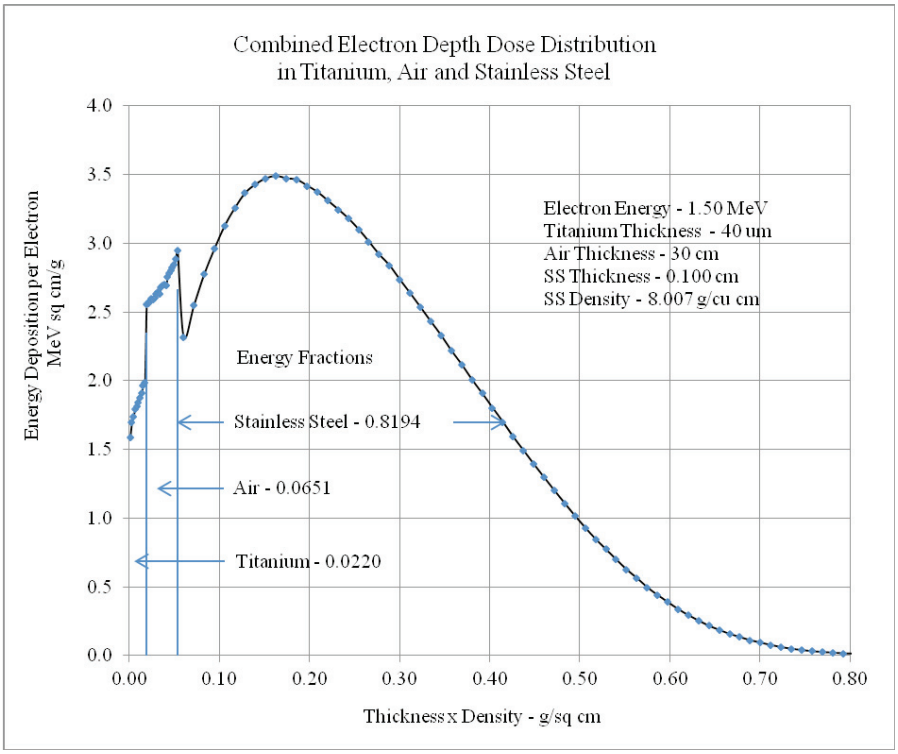

Figure 3. Combined electron depth dose distribution in titanium, air and stainless steel.

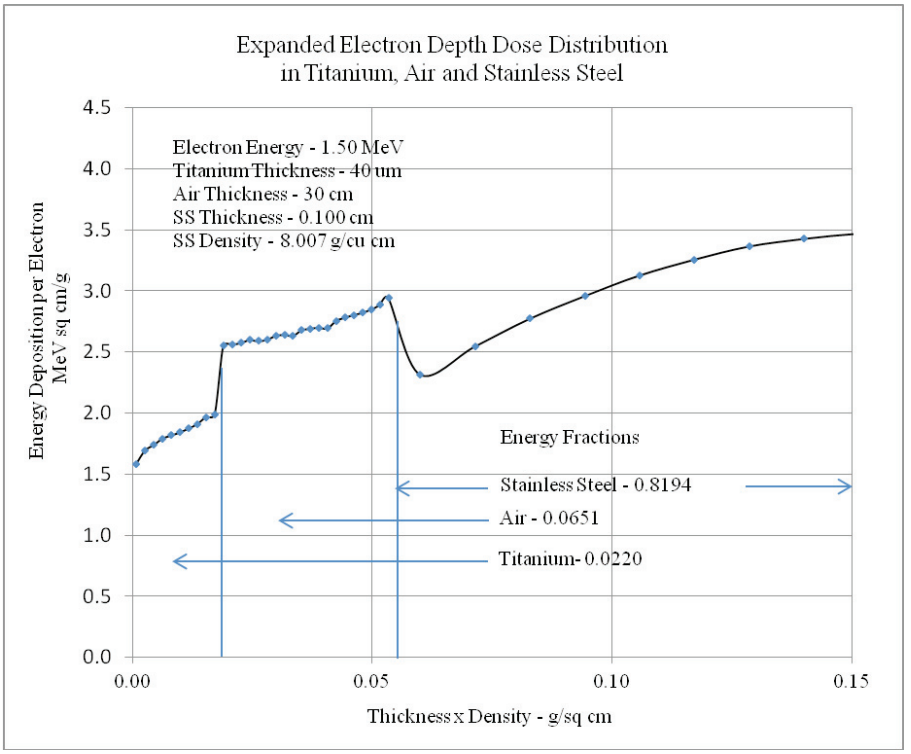

Figure 4. Expansion of Figure 3 to show the energy deposition in the air space in more detail.

OZONE PRODUCTION RATE

The ozone production rate can be obtained by using Equation (11) above and the beam power deposited in the air space, 4.883 kW, as follows:

$$\text{Ozone Production Rate} = 8.95 \times 10^{-2}\ P(\text{air})\ \text{kg/h} \qquad \text{(repeat)} \qquad (11)$$

$$\text{Ozone Production Rate} = 8.95 \times 10^{-2} \times 4.883 \qquad (22)$$

$$\text{Ozone Production Rate} = 4.37 \times 10^{-1}\ \text{kg/h} \qquad (23)$$

OZONE CONCENTRATION

The ozone concentration by weight (the ratio of the production rate divided by the air exhaust rate) can be obtained by using Equation (15) above and the beam power deposited in the air space, 4.883 kW, as follows:

$$\text{Ozone Concentration (weight)} = 1.24 \times 10^{-3}\ P(\text{air}) / F \qquad \text{(repeat)} \qquad (15)$$

$$\text{Ozone Concentration (weight)} = 1.24 \times 10^{-3} \times 4.883 / 850 \qquad (24)$$

$$\text{Ozone Concentration (weight)} = 7.12 \times 10^{-6}\ \text{or } 7.12\ \text{ppm} \qquad (25)$$

In this example, the air exhaust rate is the same as in Example 1, 850 m^3 / min or 30,000 ft^3 / min, by using a typical external dilution fan on the roof of the irradiation facility. The value from Equation (25) is higher than the maximum allowable threshold limit value, 0.10 ppm, but the ozone concentration at ground level outside of the facility will be much less than the allowable TLV because of further dilution in the external air.

4.3 Example 3

In Example 3, the PE is in the form of narrow ribbons to simulate a multiple-pass process to irradiate insulated electrical wire or small plastic tubing. It is assumed that the PE ribbons intercept 75 % of the beam current and that 25 % of the beam current passes through the air spaces between the ribbons and strikes the SS beam stop.

COMBINED OZONE PRODUCTION RATE

With the assumed combination of Examples 1 and 2, the net ozone production rate will be as follows:

$$\text{Net Ozone Production Rate} = 0.75 \times 1.44 \times 10^{-1} + 0.25 \times 4.37 \times 10^{-1} \quad (26)$$

$$\text{Net Ozone Production Rate} = (1.08 + 1.09) \times 10^{-1} \quad (27)$$

$$\text{Net Ozone Production Rate} = 2.17 \times 10^{-1} \text{ kg/h} \quad (28)$$

COMBINED OZONE CONCENTRATION

With the assumed combination of Examples 1 and 2, the net ozone concentration by weight will be as follows:

$$\text{Net Ozone Concentration} = 0.75 \times 2.35 \times 10^{-6} + 0.25 \times 7.12 \times 10^{-6} \quad (29)$$

$$\text{Net Ozone Concentration} = (1.76 + 1.78) \times 10^{-6} \quad (30)$$

$$\text{Net Ozone Concentration} = 3.54 \times 10^{-6} \quad \text{or } 3.54 \text{ ppm} \quad (31)$$

In this example, the air exhaust rate is the same as in Examples 1 and 2, 850 m^3 / min or 30,000 ft^3 / min, by using a typical external dilution fan on the roof of the irradiation facility. The value from Equation (31) is still higher than the maximum allowable threshold limit value, 0.10 ppm, but the ozone concentration at ground level outside of the facility will be much less than the allowable TLV because of further dilution in the external air.

5. Conclusion

This paper presents basic information about the generation of ozone in an irradiation facility, including the ozone yield in kilograms per kilowatt hour of ionizing energy absorbed in the air, the ozone production rate in kilograms per hour and the ozone concentration in the exhaust air (in parts per million) from a typical electron beam facility with an electron energy of 1.5 MeV, a beam current of 50 mA and a beam power of 75 kW.

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