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Evaluation of potential induced radioactivity in medical products as a function of electron energy in electron beam sterilization

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ABSTRACT

Commercial sterilization of medical devices may be performed using electron beam irradiators at various electron energies. The potential for activating components of the devices has been discussed, with current standards stating that electron energy greater than 10 MeV requires assessment of potential induced radioactivity. This paper evaluates the potential for induced activity in medical products sterilized in electron beam as a function of the electron maximum energy. Monte Carlo simulation of a surrogate medical device was used to calculate photon and neutron fields resulting from electron irradiation, which were used to calculate concentrations for several radionuclides.

The experiments confirmed that 10 MeV is a conservative assumption for limiting induced radioactivity. However, under the conditions as evaluated, which is a limited total quantity of metal in the material being irradiated and absent a limited number of elements; the amount of induced activity at 12 MeV could also be considered insignificant. The comparison of the sum-of-fractions to the US Nuclear Regulatory Commission exempt concentration limits is less than unity for all energies below 12.1 MeV, which suggests that there is minimal probability of significant induced activity at energies above the 10 MeV upper energy limit.

1. Introduction

The issue of whether radioactivity may be induced in medical devices irradiated in electron beams has been considered for some time. In International Standards Organization publication ISO 11137-1, the relevant standard on radiation sterilization, the problem is stated as (ISO, 2006)

> "If energy for electrons exceeds 10 MeV or energy level for electrons used to generate x-rays exceeds 5 MeV, the potential for induced radioactivity in product shall be assessed."

However, the standard and other relevant regulations, such as the US Food and Drug Administration regulations on food irradiation at 21 CFR 179.26, which also gives the 10 MeV electron energy limit, do not give a technical justification for imposing this energy limit. As discussed in subsequent sections, most relevant publications tacitly assume that 10 MeV is a practical limit for considering activation, which may be in part based on the most prevalent availability of commercial accelerators used for electron beam sterilization.

Publications from the International Atomic Energy Agency (IAEA, 1995,2002) give extensive analysis of the potential activation mechanisms. From this analysis, supported by other publications, three mechanisms are considered significant for inducing radioactivity in products irradiated by electrons: photoneutron production, neutron capture, and isomeric excitation.

Other potential activation mechanisms have been studied in other published papers, specifically photoproton production (Gregoire et al. 2003;Findlay et al., 1992). Findlay's analysis of the photoproton production mechanism, in particular, shows that this pathway is an insignificant contribution to the total induced radioactivity in electron beam processing as compared to photoneutron production and neutron absorption.

Direct activation from electron interactions, rather than from bremsstrahlung, is addressed in detail in one publication (IAEA, 2002). In this analysis, the amount of radioactivity that could be expected from the electron interactions is small in comparison to that resulting from bremsstrahlung interactions. As a result, the focus of this paper is on the photon interaction and excludes consideration of direct activation by electrons.

2. Calculating induced radioactivity concentrations

As discussed in a previous publication (Smith, 2008), the induced activity in an electron beam system is calculated for three mechanisms: photoneutron reactions, neutron capture, and isomeric excitation.

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2.1. Photoneutron production

The photon interaction with a nucleus can result in the ejection of a neutron from the atom, leaving a resultant radioactive species. Estimating the activity produced in target atoms following ejection of the photoneutron can be estimated to within a factor 10 by a simple relationship. (IAEA 2002)

$$A_{xrays} = 10^{-13} D \frac{N_A \cdot f_{Is} \cdot \ln(2)}{A_{Is} \cdot T_{1/2}} (E - E_{th})^3 \quad (1)$$

where A_{xrays} is the induced activity in Bqg^{-1}; D is the delivered dose in kGy; N_A is Avogadro's number; f_{Is} is the fraction of photoneutron reactions that result in radionuclide, assumed to be 1.0 for the purpose of this analysis; A_{Is} is the atomic weight of the nuclide under consideration; $T_{1/2}$ is the half-life in seconds; E is the electron energy in MeV; and E_{th} the threshold energy for photoneutron production, also in MeV.

In the derivation of Eq. (1), (IAEA, 2002) the x-ray energy spectrum was evaluated from bremsstrahlung in a single element, high atomic number target, as would be used in an industrial x-ray irradiator. This energy spectrum removes the low-energy component that corresponds to the K-shell cut-off in the target material, which was estimated to give an overestimate of activity per unit dose of approximately 20%. However, in the situation considered here, the x-rays are produced within the irradiated product itself, which consists of materials with lower atomic number. The equation was for food irradiation applications, with the composition given as oxygen, nitrogen, and carbon, similar to the assumed medical product composition. The lower energy cutoff then is related to the K-shell of the product material and not the target. The actual radioactivity produced per unit x-ray dose in product irradiated with 10 MeV electrons is approximately 6% lower than would be produced in the irradiated product with the same intensity x-rays produced from a tungsten target. (IAEA 2002).

2.2. Neutron capture

The photoneutron production process is the source of neutrons, which are generated within the irradiated product and subsequently absorbed to create additional induced radioactivity. While many neutron-generating reactions may occur, depending on photon energy (IAEA, 2002), the most significant source of such neutrons in the material being considered here is the $^{2}H(\gamma,n)^{1}H$ reaction, which has threshold energy of 2.23 MeV. (Wakeford and Blackburn, 1991) This reaction dominates neutron production for absorption reactions, such that the contribution from other photoneutron reactions, e.g., $^{13}C(\gamma,n)^{12}C$ and $^{17}O(\gamma,n)^{16}O$, do not contribute significantly to the neutron yield in considering photoneutron production from bremsstrahlung in these conditions. (Stichelbaut et al., 2006)

The estimated induced activity from neutron capture in a single element can be calculated from the equation (Lieser, 2001)

$$A = N\phi\sigma(1 - e^{-\lambda t}) \quad (2)$$

where A is the induced activity in Bq g^{-1}; N is the number of target atoms per gram; ϕ is the neutron fluence rate; σ is the thermal neutron absorption cross-section; λ is the decay constant for the radioactive species and t is the irradiation time.

2.3. Isomeric excitation

The equation for calculation of induced radioactivity from isomeric excitation is the same as Eq. (2) above, changing the fluence rate to be photons instead of neutrons and the cross-section to be that for the isomeric excitation reaction. As noted previously, (Smith, 2008) these cross-sections are difficult to locate in published literature. For this experiment, the same isomers used in the previous paper were considered as the most probable to occur as induced radioactivity.

3. Description of methodology

The experimental methodology consisted of two phases: (1) mathematical modeling of the electron beam irradiation process as it is expected to be encountered with medical device sterilization and (2) confirmatory measurements at two electron energies for a given radionuclide production reaction. Because the scope was conceived as being broad, specifically evaluating the potential for activation for the entire range of radionuclides with a production mechanism that fell within the defined electron energy parameters, confirmatory measurements could only be conducted on a limited scale.

3.1. Calculation of photon and neutron fluence

The modeling work described below was performed with MCNPX 2.6.0 (Los Alamos National Laboratory (LANL), 2008). In MCNPX, the simulated medical device product, which was essentially a phantom to be irradiated, was represented by a right prism 20 cm by 10 cm in the horizontal dimension and 10 cm in height, comprising twenty laminar sheets of unit density polyethylene with 0.5 cm thickness. Natural isotopic abundances for carbon, hydrogen, and oxygen were assumed as the composition of the irradiated material. A titanium layer was added at the bottom of the stack, simulating a conveyor in a commercial system.

A directional electron beam was placed 10 cm above the top surface of the polyethylene stack and directed perpendicular to the surface. The typical scanning function of a commercial electron beam irradiator was not simulated in the model. Instead, the beam was assumed to cover the entire surface of the polyethylene phantom, which will also account for the horizontal transit of the product along the conveyance system under the beam.

MCNPX tallies were used to calculate the neutron and photon fluence across the surfaces of the product laminar sheets and to calculate energy deposition at the same locations. The total electron plus photon energy deposition was calculated at the interface of each laminar sheet.

The same geometry was used to evaluate different initial electron energies. All electrons generated from the source term were assumed to be monoenergetic, with the model being repeated for initial electron energies of 8, 9, 10, 11, 12, and 13 MeV.

In the model, it was assumed that the photon field would only be present as the beam spot was passing over the surface of the phantom directly above the location of the metal foil. Induced radioactivity would only occur during direct irradiation, but a penumbral photon field would be present immediately before and after the beam spot passes over the foil due to scattering within the phantom, thereby creating fringe photon fields outside of the principal field. This effect has not been quantified, but could contribute to an underestimate of the induced radioactivity concentration.

Potential bremsstrahlung production in the window of the electron beam accelerator also needed to be taken into account. These windows are typically thin titanium foils through which the electrons pass from the vacuum into the air (IAEA and iiA, 2010). Production of x-rays within the window would occur, but the photons would not be attenuated in the window itself and would contribute to the surface dose to the polyethylene stack and to the number photons interacting with the metal foil to create induced radioactivity, taking into account attenuation in the polyethylene to the depth of maximum photon fluence.