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RADIATION PROCESSING WITH HIGH-ENERGY X-RAYS

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ABSTRACT

The physical, chemical or biological characteristics of selected commercial products and materials can be improved by radiation processing. The ionizing energy can be provided by accelerated electrons with energies between 75 keV and 10 MeV, gamma rays from cobalt-60 with average energies of 1.25 MeV or X-rays with maximum energies up to 7.5 MeV. Electron beams are preferred for thin products, which are processed at high speeds. Gamma rays are used for products that are too thick for treatment with electron beams. High-energy X-rays can also be used for these purposes because their penetration in solid materials is similar to or even slightly greater than that of gamma rays. Previously, the use of X-rays had been inhibited by their slower processing rates and higher costs when compared with gamma rays. Since then, the price of cobalt-60 sources has been increased and the radiation intensity from high-energy, high-power X-ray generators has also increased. For facilities requiring at least 2 MCi of cobalt-60, the capital and operating costs of X-ray facilities with equivalent processing rates can be less than that of gamma-ray irradiators. Several high-energy electron beam facilities have been equipped with removable X-ray targets so that irradiation processes can be done with either type of ionizing energy. A new facility is now being built which will be used exclusively in the X-ray mode to sterilize medical products. Operation of this facility will show that high-energy, high-power X-ray generators are practical alternatives to large gamma-ray sources.

1. INTRODUCTION

Radiation processing is a means to change the physical, chemical or biological characteristics of commercial products and materials by treatment with ionizing energy. In many cases, this technique can improve their market value or reduce their environmental impact. Examples of ongoing applications for polymeric products are the curing of inks, coatings and adhesives on metal, plastic and paper substrates, the crosslinking of plastic film, foam, tubing and pipe, molded plastic parts, wire insulation and rubber components for automobile tires. Examples of biological applications are the sterilization of medical devices and the disinfection of packaging materials and fresh foods. Some emerging applications are the curing of carbon-fiber reinforced plastic materials and the treatment of toxic substances, including solid, liquid and gaseous waste materials.

Practical sources of ionizing energy for these applications are high-energy electrons, X-rays emitted by accelerated electrons and gamma rays emitted by radioactive nuclides. These can all remove atomic electrons to create ions or free radicals, which will initiate similar chemical reactions. The preferred type of radiation source for a particular application is usually determined by practical process requirements, such as minimum absorbed dose, dose

uniformity and dose rate, material thickness, density and shape, production rate, capital and operating costs. Ease of use, source maintenance and safety are also important considerations.

High-power electron beams can process materials at high line speeds, but their penetration is limited to a few centimeters in unit density materials. Gamma rays from cobalt-60 sources are mainly used to irradiate larger packages of medical devices and foods at slower processing rates. High-energy X-rays emitted by electron beams with energies greater than 3.0 MeV are somewhat more penetrating than such gamma rays. Recent increases in the price of cobalt-60 sources and increases in the electron beam powers of industrial electron accelerators have made X-ray processing a practical alternative to gamma-ray processing for high-capacity irradiation facilities.

2. X-RAY CHARACTERISTICS

X-rays can be produced by two different effects. Relatively low-energy X-rays are emitted by electrons when they make transitions between different energy levels in an atom. Such X-rays have well-defined energies, which are determined by the atomic number and the particular electronic transition. The highest energy obtainable in this manner is the transition of an unbound electron to the k level of uranium, which is 115.6 keV. Higher-energy X-rays with a broad energy spectrum are emitted by accelerated electrons when they pass near to atomic nuclei and are deflected by their electric fields. This effect is called by the German term bremsstrahlung, which means braking radiation. X-rays produced in this manner have a continuous spectrum of photon energies extending from about 30 keV up to a maximum energy equal to the kinetic energy of the incident electron. For example, with a 5 MeV electron, the most probable photon energy is about 300 keV, the average photon energy is near 1.0 MeV, while the maximum photon energy is 5.0 MeV. The X-ray intensity increases with the electron energy and beam current and with the atomic number of the target material.

Heavy metals, such as tantalum and tungsten, are practical target materials. They have high atomic numbers and high melting temperatures. Tantalum is usually preferred versus tungsten for industrial, large-area, high-power targets because it is more workable than tungsten and has a higher threshold energy for induced nuclear reactions. Lead and bismuth have lower melting temperatures, gold and platinum are expensive, and uranium has other problems. The highest X-ray yield can be obtained with an optimum target thickness, which is about 40% of the electron range in that material. The maximum CSDA electron range (continuous slowing down approximation) for 7.5 MeV electrons in tantalum is about 5.0 g/cm^2. With a volume density of 16.6 g/cm^3, this is equivalent to a thickness of 3.0 mm. So the optimum thickness of tantalum at this energy is about 1.2 mm. This is not a critical dimension because the X-ray yield decreases slowly with increasing target thickness. With an optimum tantalum target thickness, the efficiency for converting electron beam power to X-ray power emitted through the target in the direction of the incident electron beam increases in proportion to the electron energy. It is about 8% with an electron energy of 5.0 MeV, 12% with 7.5 MeV and 16% with 10 MeV. These relatively low efficiencies can be compensated by using high-energy, high-power electron accelerators to produce X-ray dose rates sufficiently high for industrial applications of radiation processing.

In order to avoid inducing nuclear reactions in a tantalum target, the electron energy should not be more than 7.5 MeV. The US Food and Drug Administration has recently increased the

X-ray energy limit for food irradiation from 5.0 MeV to 7.5 MeV when a tantalum target is used. This regulation was issued in response to a petition and data submitted by IBA [1].

The X-ray intensity decreases exponentially with increasing thickness of irradiated material. The tenth-value layers in water are 39 cm, 44 cm and 49 cm for maximum X-ray energies of 5.0 MeV, 7.5 MeV and 10 MeV, respectively. In contrast, the tenth-value layer in water for a large area source of gamma rays from cobalt-60 is 31 cm. For treatment from opposite sides, the optimum thicknesses of water for the most efficient use of X-ray power are 34 cm, 38 cm and 43 cm for 5.0 MeV, 7.5 MeV and 10 MeV, respectively. The max/min dose uniformity ratio (DUR) is about 1.5 with these conditions. In contrast, the optimum thickness for gamma rays from a large area cobalt-60 source is 28 cm of water. These thicknesses are not critical because the X-ray power utilization efficiency is not very sensitive to changes in thickness. Common plastic materials, such as polyethylene and polypropylene with densities slightly less than water, would have nearly the same values of optimum thickness. The properties of high-energy X-rays listed in Table 1 have been calculated with the ITS Monte Carlo code [2].

Table 1. Physical properties of high-energy X-rays

Electron Energy	Mean Energy	Emission Efficiency	Tenth Value Layer (cm) Present Work			Optimum Thickness Double Sided	
(MeV)	(MeV)	(%)	Calculated	Measured	Previous	(g/cm2)	Max/Min
10	1.56	16.2	49.0	47.9	49.0	43	1.54
7.5	1.38	13.3	44.3	N/A	N/A	38	1.54
5.0	1.19	8.2	39.0	39.5	38.0	34	1.54
Co-60	1.25				31.0	28	1.75

With electron energies greater than 2.0 MeV, the X-ray emission is greatest in the direction of the incident electron beam, and the angular dispersion of the X-ray beam decreases as the energy increases. The divergence at half of the maximum intensity is about 20 degrees with an electron energy of 5.0 MeV, about 15 degrees with 7.5 MeV and about 10 degrees with 10 MeV. This important characteristic is different from the isotropic emission of gamma rays from radioactive nuclides. The narrow angular distribution of high-energy X-rays enables this kind of radiation to penetrate deeper in irradiated materials and to be used more efficiently than gamma rays. It also makes it easier to accommodate changes in product density and dose requirements in an industrial irradiation facility, because only a few product carriers are in the X-ray beam at any time. The X-rays are mainly concentrated on the product carrier that is in front of the target. The absorbed dose is relatively small in adjacent carriers when they are not in the most intense part of the X-ray beam [3-6].

3. HIGH-ENERGY, HIGH-POWER X-RAY GENERATORS

The beam power ratings of IBA's high-energy electron accelerators have been increased to provide higher X-ray intensities for radiation processing. The Dynamitron® direct-current accelerator has been upgraded from 200 kW to 300 kW of electron beam power at 5.0 MeV. The emitted X-ray power with 8% power conversion efficiency is 24 kW, which can provide

nearly the same processing capacity as 2.0 MCi of cobalt-60 sources. With recent prices of about 2.25 USD per curie, the capital cost of this source loading would be more than the capital cost of a 300 kW Dynamitron. The cost for replenishing the radioactive decay of cobalt-60 at 12.3% per year is slightly more than the annual electricity cost for a 300 kW Dynamitron, which draws about 600 kW from the electric power service. This comparison of power costs is for a four-shift, 8000 hour per year operation of the accelerator. If the process does not require continuous operation, then the accelerator can be turned off and the cost of electric power would be reduced. In contrast, the emitted power from a radioactive source is always on. Electricity rates in the USA vary with the location of the facility, but the national average is about 10 cents per kilowatt hour. Other costs, such as land, building, radiation shield, product conveyor, management, labor, etc., for industrial irradiation facilities with the same processing capacity are comparable for either X-ray or gamma-ray processing [7, 8].

The Rhodotron® radio-frequency accelerator has been upgraded from 135 kW to 500 kW at 5.0 MeV and to 700 kW at 7.0 MeV. The emitted X-ray power with 8% power conversion efficiency at 5.0 MeV is 40 kW, which is equivalent to the gamma-ray power from 3.0 MCi of cobalt-60. The emitted X-ray power with 11% power conversion efficiency at 7.0 MeV is 77 kW, which is nearly equivalent to the gamma-ray power from 6.0 MCi of cobalt-60. The capital cost for this source loading would be much more than the capital cost of a 700 kW Rhodotron. The cost for replenishing the radioactive decay of cobalt-60 at 12.3% per year is substantially more than the annual electricity cost for a 700 kW Rhodotron, which draws about 1.4 MW from the electric power service. This comparison of power costs is for a four-shift, 8000 hour per year operation of the accelerator. If the process does not require continuous operation, then the accelerator can be turned off and the cost of electric power would be reduced [8, 9].

4. X-RAY AND ELECTRON BEAM PROCESSING FACILITIES

Dual-purpose electron accelerators equipped with removable X-ray targets can accommodate both electron-beam and X-ray irradiation processes. There are now three facilities like this in Japan, three in the United States, one in Germany and one in France. One of the Japanese facilities has a 5.0 MeV, 150 kW Cockcroft Walton accelerator. Another has a 5.0 MeV, 200 kW Dynamitron, and a third has a Rhodotron with two separate beam lines. The X-ray line is rated for 135 kW at 5.0 MeV and the electron beam line is rated for 200 kW at 10 MeV. One of the USA facilities is equipped with an L-band microwave linac rated for 100 kW at 5.0 MeV. Another has a Dynamitron rated for 90 kW at 3.0 MeV, and the third has a Rhodotron with three beam lines. The X-ray lines are rated for 135 kW at 5.0 MeV and 190 kW at 7.0 MeV. The electron beam line is rated for 200 kW at 10 MeV. The German installation has a similar Rhodotron. The French facility is equipped with an S-band microwave linac rated for 20 kW at 10 MeV [8].

The Rhodotron facility in the USA had been designed to irradiate bulk plastic materials and commercial products with 10 MeV electrons and packages of medical devices and food with 5.0 MeV and 7.0 MeV X-rays. After the anthrax attack occurred in 2002, the Postal Service acquired exclusive use of this facility for sterilizing mail addressed to government offices in Washington, D.C. This protective service is still ongoing. Flat mail is being irradiated with 10 MeV electrons and packages of mail are being irradiated with 5.0 MeV X-rays [8].

5. A NEW X-RAY PROCESSING FACILITY FOR MEDICAL DEVICES

A new irradiation system intended for the continuous use of high-energy X-rays has been designed using 3D Monte Carlo simulation tools. This system, called eXelis®, will offer optimal performance for the treatment of low-density products such as medical devices [10].

The eXelis system makes use of two levels of pallets moving in front of a long X-ray target, which is oriented vertically to irradiate pallets from the side. The total height of the pallet stack is larger than the electron scanning width in order to optimize the capture of X-rays emitted by the target. The top and bottom pallets of a stack are exchanged between passes in front of the X-ray beam in order to obtain a uniform dose distribution along the vertical axis of the pallets. Also, the pallets are rotated by 180° between two passes in order to perform a two-sided irradiation, which gives a rather uniform dose distribution along the beam axis. Thus, each pallet must pass 4 times in front of the beam to be treated. The deep penetration of high-energy X-rays allows obtaining excellent dose uniformities for low-density products.

The eXelis system has been designed with the Monte Carlo simulation toolkit GEANT 3.21 from CERN [11]. The main elements of the system have been modelled:

- The electron scanning horn;
- The X-ray target and its support;
- The product (as a homogeneous box);
- The wood pallet supporting the product;
- The slave pallet to carry the pallet.

Simulations have been performed for industrial pallets with dimensions 100 x 120 x 180 cm^3. The longer pallet width (120 cm) is aligned with the electron beam axis in order to optimize the X-ray capture inside the product. As the wood pallet can absorb part of the X-ray energy before reaching the product, a great care has been taken to model it as accurately as possible. The same is true for the slave pallet and a preliminary design allowing pallets with density up to 0.5 g/cm^3 has been used. The slave pallet is made of aluminium tubes and has a height of 2.5 cm. In addition to the wood pallet and the slave pallet, an extra gap of 22.5 cm has been taken into account between the two levels, to allow for the conveyance mechanism.

Figs. 1 and 2 show a side view and a top view of the model implemented in GEANT 3, respectively. Fig. 2 shows that 3 stacks of two pallets are modelled along the conveyance axis in order to correctly reproduce the shadowing and scattering effects from the middle stack to the adjacent stacks. A conservative value of 20 cm has been used as separation gap between two pallet stacks. A gap of 20 cm is also used along the beam axis between the X-ray target and the product surface.

All pallets have the same density but the dose deposition is only recorded in the central stack. The pallet stacks move along the conveyor axis with a uniform speed. This is simulated by displacing the pallets by small steps of 3 cm along the conveyor axis. In order to integrate over the full X-ray field, the centre of the middle pallet stack is displaced between -2L and +2L, where L is the distance separating the centre of two pallet stacks along the conveyor axis (L = 100 + 20 cm in this case).

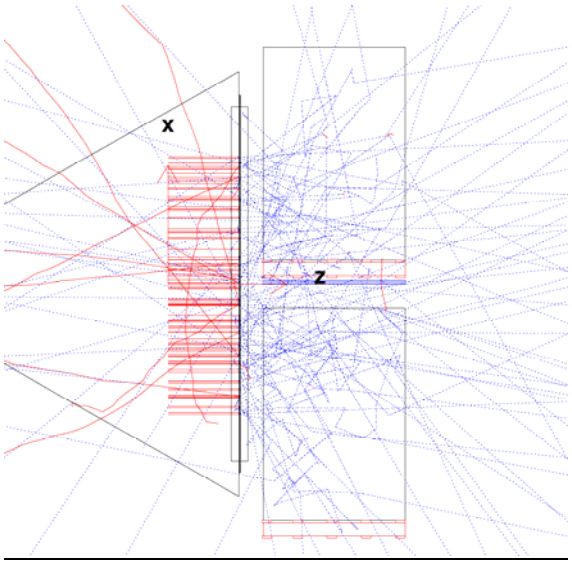

Figure 1. Side view of the 2-level pallet system modelled in GEANT3.

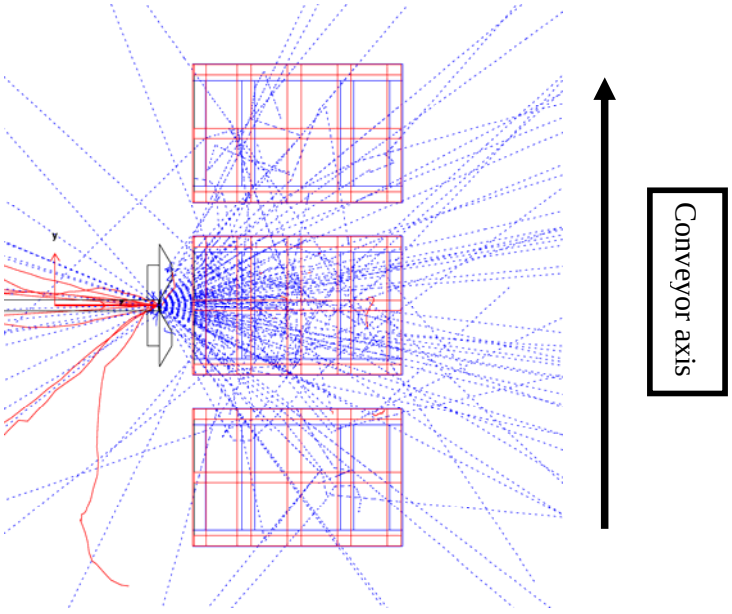

Figure 2. Top view of the 2-level pallet system modelled in GEANT3.

The electron beam is scanned along the X-ray target in order to obtain a uniform dose distribution along the vertical axis of the pallet. Thanks to the use of a 2-level system, it is possible to use a homogeneous electron distribution along the vertical axis. This distribution does not give a homogeneous dose distribution along the vertical axis by itself but, as each pallet will stay the same amount of time in the upper and lower levels, the dose distribution becomes rather uniform along the vertical axis of the pallet. This is demonstrated in Fig. 3 showing the dose distribution obtained along the Z (vertical) axis during each pass and the total dose obtained by summing the two passes.

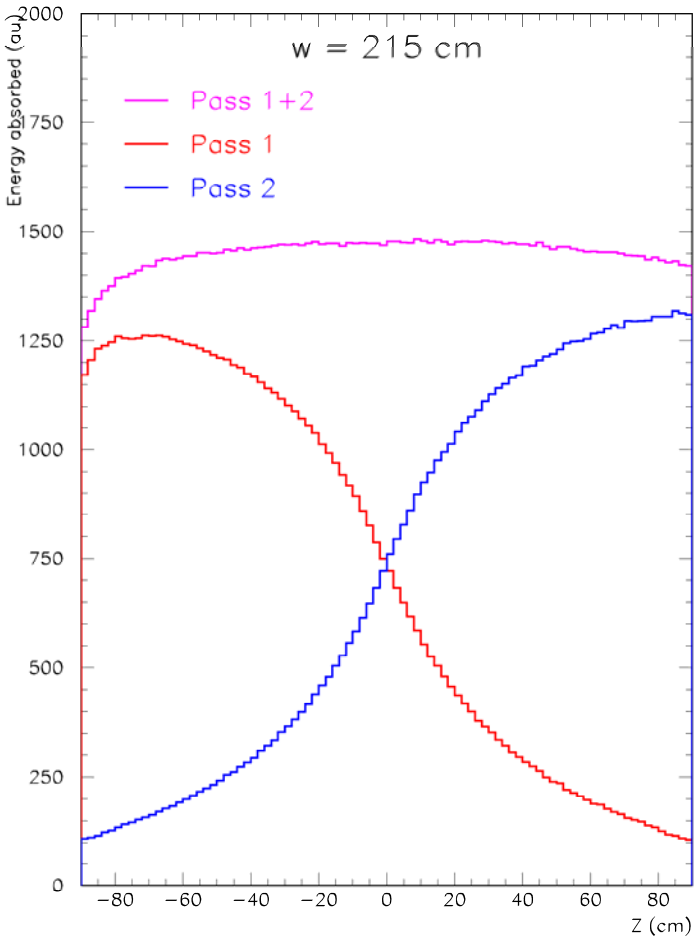

Figure 3. Variation of the dose along the vertical axis inside one pallet. The left curve shows the dose in the top location while the right curve shows the dose in the bottom location. The total dose collected after two passes is shown by the upper curve.

All simulations were done using a semi-parallel electron beam on the X-ray target. It is also possible to use a divergent beam on the target but, in that case, the performance figures become very sensitive to the scanning width value. This sensitivity is strongly reduced for a semi-parallel beam. The divergence of a scanning electron beam can be controlled or even cancelled by means of a special magnet system mounted just before the exit window at the end of the beam scanning chamber. This parallel beam scanning system can be provided with IBA's electron accelerators.

To determine the performance figures of the eXelis system, the irradiated product was divided into 5 cm thick slices along the vertical Z axis. Each slice was subdivided into a 2D map of 20x20 bins and the energy deposition was recorded into each of the 400 bins. Then, the values of the minimal and maximal energy depositions were searched for and recorded. Fig. 4 shows the evolution of these minimal and maximal energy depositions as a function of the Z position. In this figure, all the doses are normalized to the smallest dose obtained inside the product. Finally, the smallest value of the minimal doses gives the global minimal dose, D_{min}, while the largest of the maximal values gives the global maximal dose, D_{max}. The D_{min}

value is obtained at the bottom of the pallet, because part of the X-ray energy is absorbed inside the wood pallet supporting the product. Beyond the first 30 cm of the product, the maximal and minimal doses become rather constant and their ratio is about 1.15.

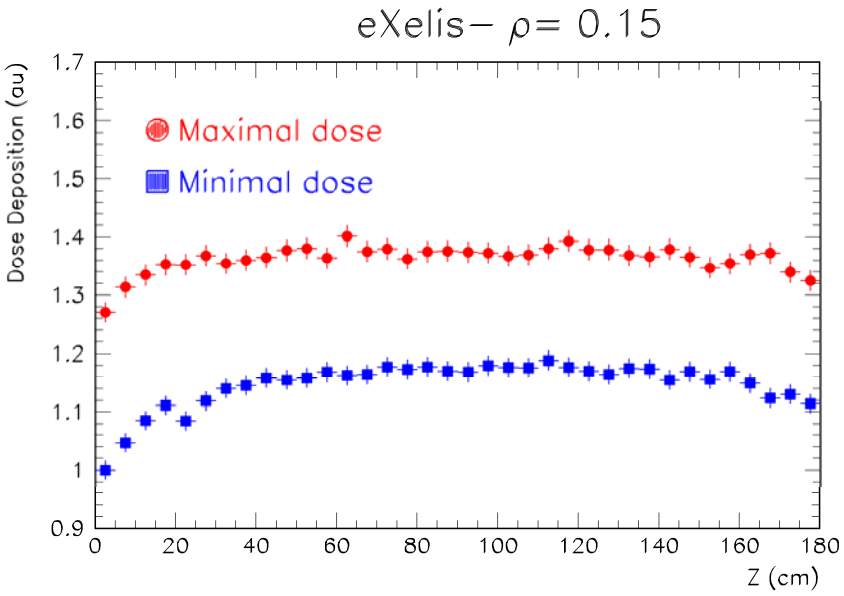

Figure 4. Evolution of the minimal and maximal doses along the vertical axis of a pallet.

Fig. 5 shows the evolution of the global Dose Uniformity Ratio, DUR = D_{max} / D_{min}, as a function of the product density ρ. This DUR remains below 1.5 for product densities below 0.25 g/cm^3. This is usually the case for packaged medical devices. The system throughput is computed based upon the D_{min} quantity, the required sterilization dose and the accelerator power. The treatment capacity obtained for an electron beam power of 700 kW and for a minimal sterilization dose of 25 kGy is shown in Fig. 6 as a function of the product density ρ. For a product density of 0.15 g/cm^3 (typical density for medical device packages), the eXelis system allows the sterilization of about 21 m^3/h. These performance figures may vary with different parameters such as:

- The type of conveyance mechanism used to convey the pallets in front of the X-ray beam;
- The sizes of the gaps separating the pallets along the vertical and the conveyance axes.
- The size of the gap separating the pallet and the X-ray target.
- The structure of the slave pallet.

Fig. 7 shows a photograph of the Model TT1000 Rhodotron accelerator, which can produce an electron beam power of 700 kW with an energy of 7.0 MeV. Fig. 8 is a three-dimensional drawing of an X-ray facility with a dual-pallet irradiation system as described above. Fig. 9 is a three-dimensional drawing of this facility showing the location of the accelerator inside the radiation shield and the path followed by the product carriers as they pass through the facility.

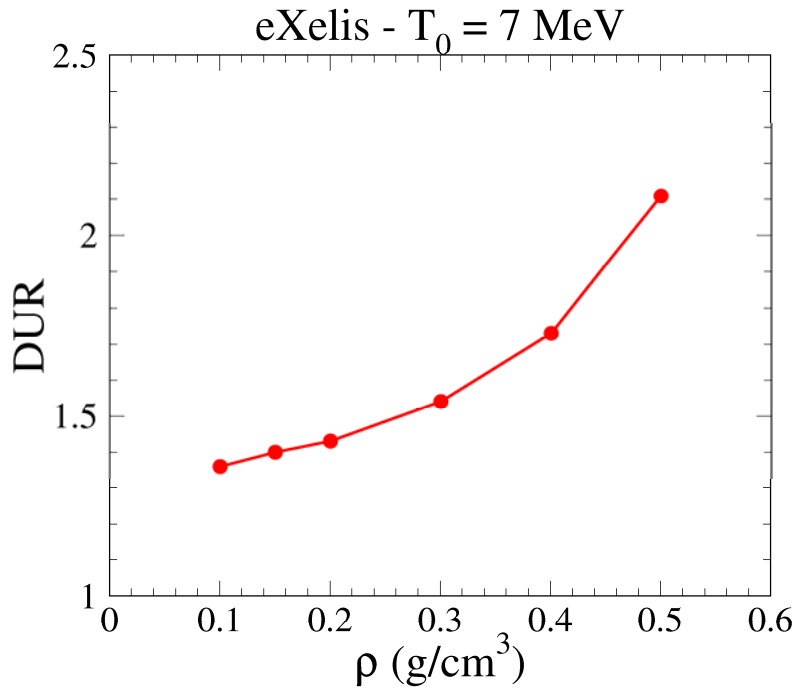

Figure 5. Evolution of the DUR as a function of product density.

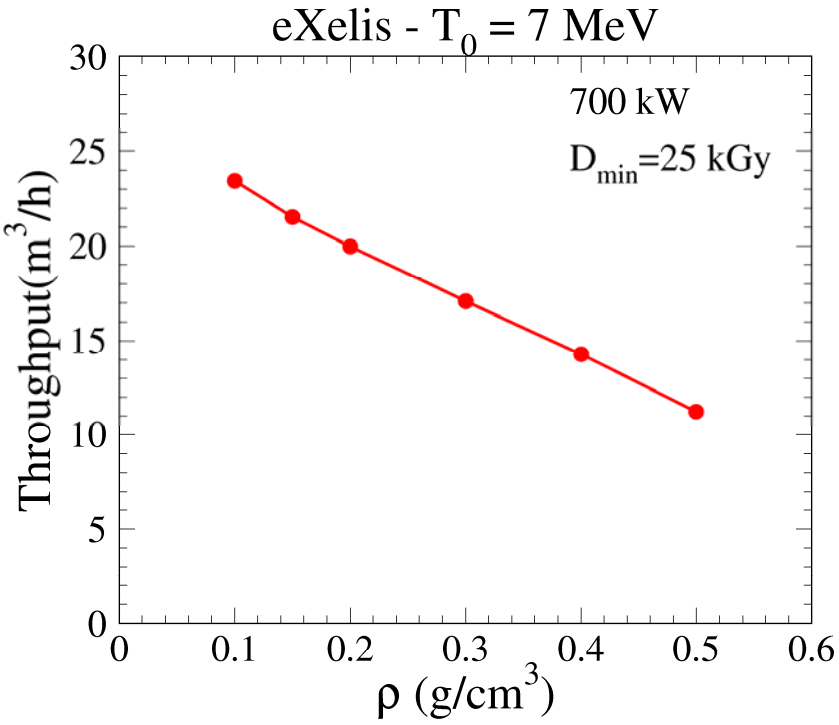

Figure 6: Evolution of the treatment capacity as a function of product density.

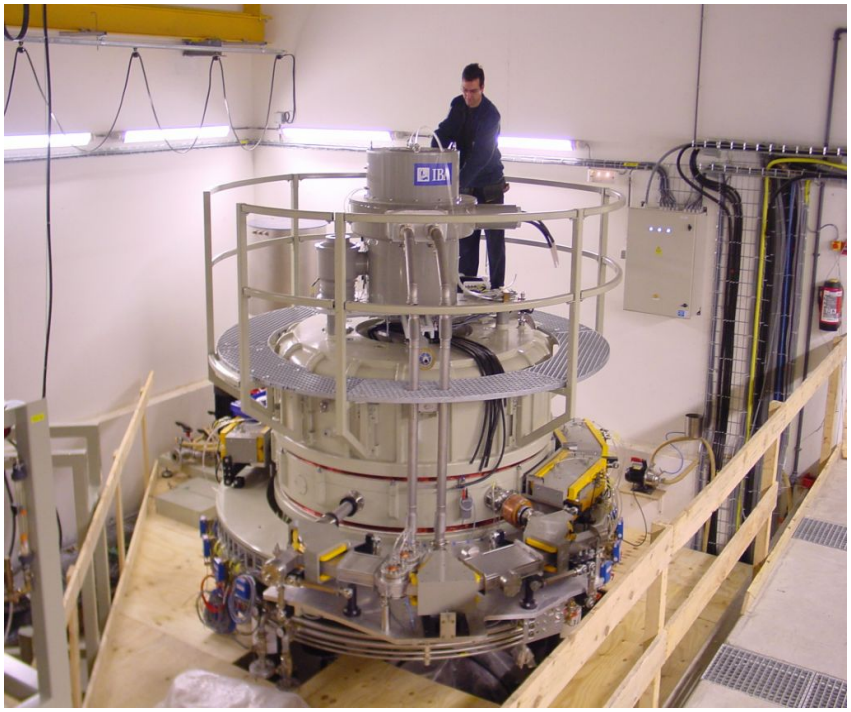

Figure 7. A Photograph of the Model TT1000 Rhodotron ─ 7.0 MeV, 700 kW.

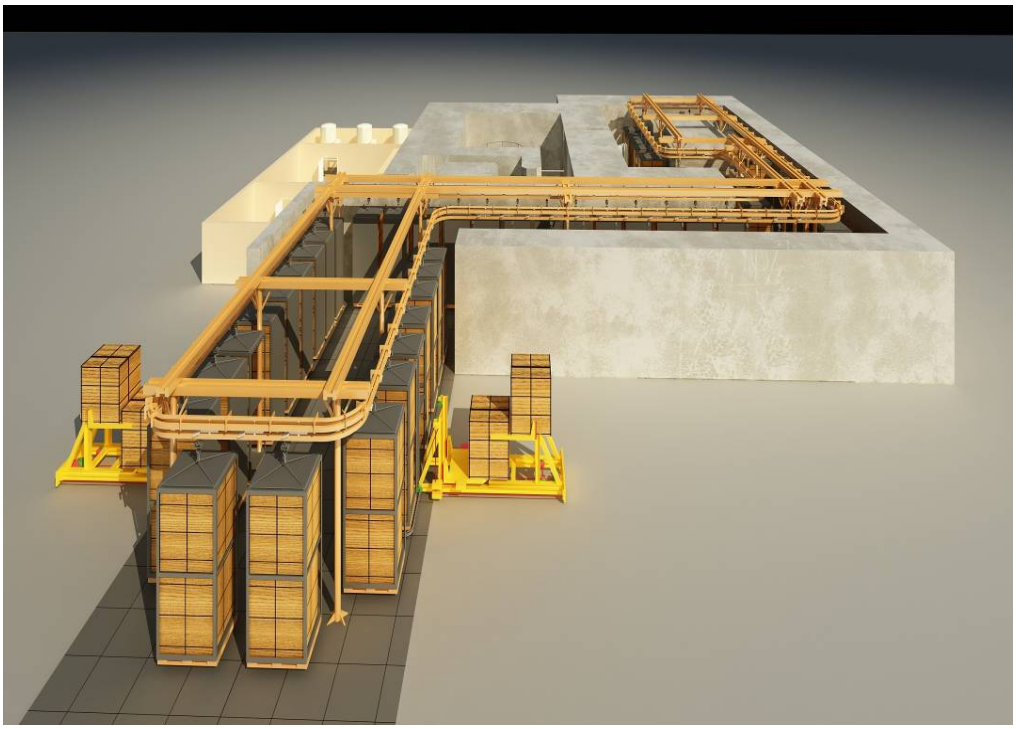

Figure 8. A three-dimensional drawing of an X-ray facility with dual pallet carriers showing how the top and bottom pallets are exchanged outside of the facility.

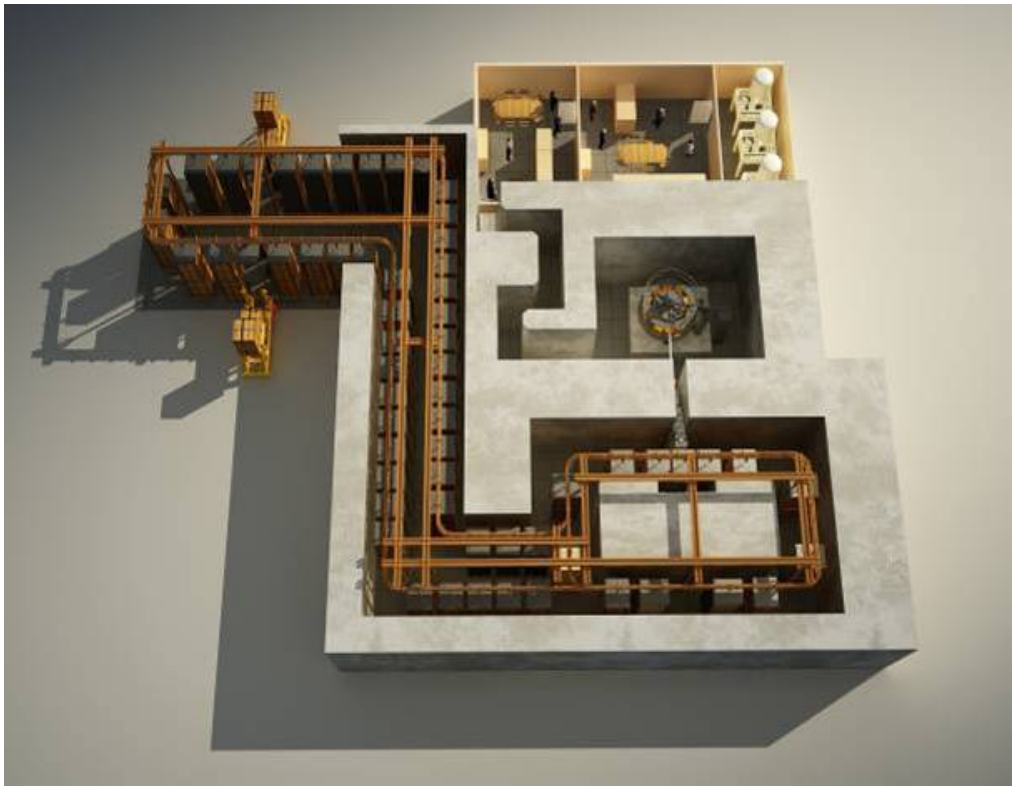

Figure 9. A three-dimensional drawing of an X-ray facility with dual pallet carriers showing how the pallets are turned around for treatment on both sides.

6. CONCLUSIONS

The development of high-energy electron accelerators with very high-power electron beams has made X-ray processing a practical alternative to gamma-ray processing for applications, such as the sterilization of packaged medical devices and the preservation of foods, which require greater penetration than can be provided by energetic electron beams. The feasibility of radiation processing with high-energy X-rays has been demonstrated in various industrial facilities in several countries. Monte Carlo simulations can provide useful information about the physical properties of high-energy X-rays, as well as the absorbed dose distributions and process throughput rates that can be achieved in irradiated products and materials. Recent comparisons have shown that the capital costs and electric power costs for accelerators with electron energies of 5.0 MeV to 7.0 MeV can be lower than the capital costs and source replenishment costs for cobalt-60 source loadings greater than 2.0 MCi. The capability to turn the radiation source on and off and to control the X-ray intensity are attractive features of an accelerator facility.

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