

INDUSTRIAL APPLICATIONS OF IRRADIATION AS A SERVICE

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Industrial use of electron beams began in the 1950's with the crosslinking of polyethylene film and wire insulation. Today the number of electron beam Processing Systems installed for industrial applications throughout the world has grown to more than six hundred stations in over 35 countries.

Total installed power is now approaching 40 megawatts (over 8 million tons of products per year).

Electron Beam is now utilized by many major industries including plastics, automotive, rubber goods, wire and cable, electrical insulation, semiconductor, medical, packaging, or pollution control.

The principal effect of high-energy electrons is to produce ions in the materials treated, resulting in the liberation of orbital electrons. As a result, the original molecule is modified and the free radicals combine to form new molecules with new chemical reactions or disorganization of the DNA chains of living organisms (insects, fungus, microorganisms, etc.).

The use of ionizing radiation in medicine, industry and research continues to grow in the developed countries. There are currently more than 170 product irradiation facilities using gamma rays and more than 800 that use beta rays produced by electron acceleration. Japan alone has more than 180 particle accelerator machines distributed among research and special application centers.

Of the many applications of these radiations, the most common of all is medical, pharmaceutical, cosmetic and food product sterilization.

The application of this know-how to industry has been and continues to be impressive, including everything from the development of nuclear power and medical applications to the use of industrial X-rays and of carbon 14 in archaeology, as well as biological product sterilization, plastic or component polymerization and synthesis of radiopharmaceuticals by means of PET techniques, etc. The therapeutic, and harmful, effect of radiation has been a well-known phenomenon since the early days of this science. The simplest procedure, from an operational point of view, for applying this technology in therapy has been to provide individualized, suitably encapsulated natural radiation sources, as in the case of facilities that use cobalt sources for irradiating. The importance of this method lies in the fact that it does not require complicated equipment or installations. The major drawback, however, is that operation cannot be interrupted, as the radioactive source is continuous-

ly emitting radiation and also its activity declines over time; this means that the source must be replaced, which involves radioactive material handling and transport operations. An additional risk is the possibility that the material could be misplaced or stolen.

The search for another source that can be controlled at will and put into operation as needed has led to the development of Accelerators. These machines are capable of transmitting energy to particles and drawing them out of electromagnetic fields as projectiles aimed at a selected material.

There are many kinds of accelerators with very different forms of energy and power. They can be divided into three major groups: Low, Medium and High energy, according to whether they have less than 5 or greater than 5 MeV, respectively.

Energy, Power and Dose are interrelated terms:

Electron Energy is measured in electron volts (e.V.). Power is the measure of energy given off by an electron in one second. The greater the power, the higher productivity (treatment speed) is.

Absorbed Dose is the energy deposited by the radiation in the unit of mass. It is measured in Gy. $1 \text{ Gy} = \frac{\text{Joule}}{\text{kg}}$.

In 1956, Johnson & Johnson developed the first industrial application for medical product sterilization using an electron accelerator. The driving force behind accelerator development was scientific curiosity in the use of accelerators, which has transcended the field of research and is now fully tar-

geted at industrial applications, especially Medical Applications.

The process of sterilization with electrons is the shortest of all those known. Product exposure takes just a few seconds and the total process, including transport, lasts a few minutes.

Another advantage and opportunity for shortening process time even more is the ability to instantaneously change the parameters from one batch to another depending on the needs of each product. This also cuts down on associated inventory stocks and costs.

BASIC PRINCIPLES OF ACCELERATORS

Atoms are basically composed of three components: protons, neutrons and electrons. The protons and neutrons form the nucleus, i.e. the core and most solid part of the atom. The electrons, which are lighter, circle the nucleus just as the planets orbit the sun. Because they are so light, orbital electrons are relatively easy to separate from the atom. In fact, an atom's electrons that enter into contact with one another can be diverted from their orbits like billiard balls.

This facility of displacing electrons from their orbits makes it easy to accelerate electrons, i.e. increase the speed at which they travel in their orbits. The skill of scientists in accelerating electrons and focusing accelerated electron beams has resulted in important, practical innovations such as television, X-rays, computer screens, etc. All these are based on electron acceleration.

A television "Tube" is a vacuum container. The vacuum is required to prevent the larger atoms from interfering with the smaller electrons, as in the above mentioned example of the billiard balls. An overheated filament produces a flow of electrons in a cavity at one end of the "Tube", the so-called "electron gun". The beam is aimed towards the front of the "Tube" (screen) by a system of magnets. There the electrons are "scanned" at the bottom of the screen, which has a special coating, and this generates the images.

The principles that operate a television set are similar to those that govern an Electron Accelerator. However, a TV requires only that the electrons be distributed on the screen. In industry, electrons must penetrate and go through the surface of the materials being treated. To do so, a flow of electrons, i.e. a powerful, concentrated "electron beam", is needed, which means that more electrons must be accelerated at a much higher speed.

Accelerators can be either linear or circular, and the latter are the more modern ones. Both kinds use an electron gun, the same as in TV sets, to continuously supply a flow of electrons.

Linear accelerators accelerate their electrons through several chambers and, as they go from one chamber to another, increasingly stronger forces are applied to them so that they move at ever faster speeds. A system of magnets is used to make sure that this electron flow is going in the right direction. When these electrons reach the end of this series of chambers, they are moving at extraordinarily high speeds, which vary according to the type of accelerator. They then pass through another magnetic system that makes this unilinear focus do a sweep, which turns it into a beam of preconceived width that can be varied as needed, i.e. convert a point of light into another of the required width.

Circular accelerators are based on the same principles but we are going to take a closer look at these, as this is the type of accelerator used in Spain and the latest one to be marketed.

Electron Accelerator

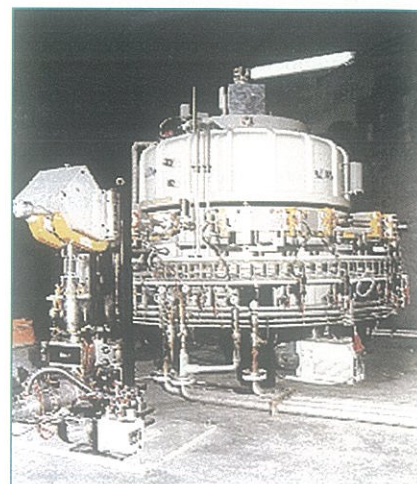
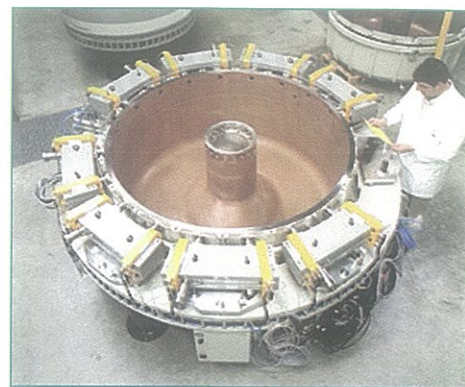
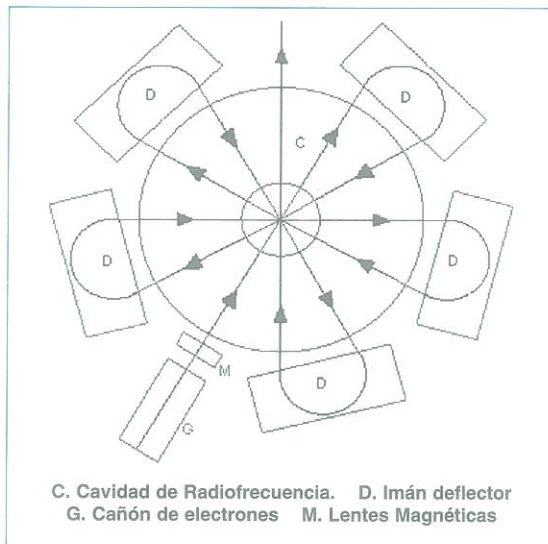
The RHODOTRON is a revolutionary design in the world of electron accelerators. It is based on the technology applied to construction of Cyclotrons, which means it is more compact and can reach higher powers than classic, similarly sized designs such as Linacs or Van der Graff.

The rated power of these accelerators spans a wide range of up to 200 KW depending on the commercial model, although the accelerator in question has a power of 50 kW.

Just as all electron accelerators, it operates on the basic principle that electrons experience an energy gain when they cross through a region where there is an electric field parallel to their direction of movement. The Rhodotron is original in that the electric field is radial and not axial, and it accelerates the electrons in a vacuum chamber as they follow a trajectory composed of several loops in the shape of a daisy (Rhodos means flower in Greek) and pass through the chamber's center several times. This means the machine can be operated in continuous mode (107 Mhz frequencies), offering obvious industrial advantages over other competing designs.

The electrons are generated in the electron source and are fired inside the chamber, where they connect to a variable electromagnetic field with the above mentioned frequency of 107 MHz; its direction changes at the same rate as the electrons passing through the center of the cavity, producing an energy gain after each pass of 1 MeV originated by the 1 MV voltage existing between the cavity's center and the outside walls.

After traveling around the diameter, the 9 deflecting magnets send them back into the cavity, where they again pass through the center to the point diametrically opposite. Finally, the electrons exit the acceleration chamber with an energy of 10 MeV. They are directed along the beam conveyor line, diverted 90° and dispersed in the raster cone by a system of magnets that makes them sweep a width of approxi-



mately 1 m. (typical blade size) with a 100 Hz frequency.

APPLICATIONS

The main applications, both in terms of volume and of revenue and demand, are medical product sterilization and food disinfection or sanitization.

Sterilization of materials and products used in certain professional, industrial or consumer activities is a process that is increasingly needed to address growing social demands for quality and safety.

A sterile product is one that is free of viable microorganisms.

Although products are manufactured under controlled conditions, they are usually contaminated with microorganisms. These products are, by definition, considered as unsterile.

The purpose of the sterilization process is to destroy microbial contamination of unsterile products. Microbial destruction by physical or chemical agents follows an exponential law that allows us to calculate a microorganism's probability of survival based on the dose administered in treatment. The probability of survival depends on the number and types of microorganisms (biological



load), on the lethality of the sterilization process and, in some cases, on the environment in which the microorganisms reside during sterilization.

In actual practice, the *Sterility Assurance Level (SAL)* is used. This defines the probability of finding one or more viable microorganisms in a product or, in other words, the probability that a product or unit will not be sterile after the sterilization process. The recognized SAL value worldwide is 10^{-6} for sterilization of any product.

FOOD SANITIZATION

Irradiating food with β rays causes very few chemical changes in the food. In addition, it has been confirmed that these changes are neither harmful nor hazardous for purposes of consumption.

Some of the chemical changes form the so-called "radiolytic" products, which are well-known products such as glucose, formic acid, acetaldehyde and carbon dioxide that are naturally found in food or formed by heat treatments. In the last 30 years, numerous scientific tests have been performed using extremely sensitive analytical techniques to isolate and detect radiolytic products formed by irradiation. No substance has actually been detected that is produced exclusively in irradiated food.

Ionizing radiation or irradiation of food for preservation and asepsis has gradually won over more and more supporters around the world in spite of the controversy that these issues provoke, often for reasons that are more visceral than purely scientific.

There is a very long list of foods that are authorized for irradiation around the world. This list varies from one country to another, but Europe overall has a wide range of irradiated products fit for consumption with full guarantees and the assurance that this treatment will prevent innumerable cases of salmonella and intestinal disorders that on occasion are life-threatening.

The European Union has recently enacted a Directive that regulates the use of food ionization throughout the Union.

The USA, with the FDA's endorsement, permits irradiation of numerous fruits, species, perishable products and finally meat.

MEDICAL PRODUCT STERILIZATION

Nowadays, the process of sterilization by ionizing rays is technically and industrially developed, both in terms of the process and its application to medical-surgical products. This treatment is now fully accepted after more than thirty years of experience and, therefore, everything related to its implementation, processes and controls is sufficiently verified and regulated on an international scale.

These sterilization plants have been developed at the level of manufacturing firms where production capabilities justify full operation, and also by firms whose aim is to provide a SERVICE to different manufacturers whose production volumes alone do not make an investment of this magnitude advisable.

More than 160 facilities are currently in operation around the world for treating medical-surgical products and components, thus validating the expansion of the treatment.

In 1956, JOHNSON & JOHNSON developed the first commercial electron-beam sterilization application for medical products. Because the system was not very reliable at the time, this technology was replaced by Cobalt.

With major laboratories working on high-energy physics research, greater efforts were made to improve the reliability of this method.

In the 1970s, industry's involvement in the development of X-ray and oncology machines helped to increase accelerator durability and reliability. All this, together with the development of computerized control systems, encouraged the industry to reassess the application of accelerators to sterilization.

The process involves the use of high-energy electrons, typically energies ranging from 3 to 10 million electron volts (MeV).

These high energies are required to penetrate products without the need to remove them from their boxes or wrapping.

When electrons pass through the product, they interact with the materials and create secondary energy species, such as electrons, ions and

free radicals. These secondary energy species are responsible for inactivation of the microorganisms, as they break the chains of DNAs and make them sterile.

Standard ISO 11137 defines a product as sterile if it is free of viable microorganisms. If, in spite of being manufactured under very controlled conditions, the products are contaminated, they are considered as unsterile. The purpose of sterilization is to eliminate microbial contamination of unsterile products. This elimination follows an exponential law that allows us to calculate the probability of survival based on the dose administered in treatment. This probability is also a function of the product's biological load, i.e. the number and types of microorganisms and the environment in which they reside during sterilization. The probability of survival of viable microorganisms in a product is expressed in terms of SAL (Sterility Assurance Level), which is defined in section 3.6.4 of ISO 11137.

The sterilization company is responsible for administering the sterilization dose in accordance with the specifications set down during the validation process of each product.

PLASTIC MATERIAL RETICULATION

Daily experience could make the use of ionizing radiation for the purpose of obtaining enhanced material properties seem contradictory to us. It is known that, in general, materials deteriorate when subjected to radiation fields, regardless of whether these fields are as energized as those generated by β radiation or X-rays or apparently as inoffensive as solar radiation. However, acquiring a thorough understanding of the effects of radiation interactions with matter can provide us with very important information for ascertaining under what conditions, for what compositions, for what types of radiation and at what dose they can positively affect the behavior of different materials of technological interest. Specifically, in the case of polymeric materials, there is no end of very interesting economic-industrial possibilities.

The effect caused by ionizing radiation when it strikes a polymeric material can, in terms of its molecular structure, be explained as follows. We are going to limit the explanation to the case of β radiation (accelerated electrons), although it would be similar for other types of ionizing radiation.

When highly energized electrons strike the molecular chains forming

polymers, a series of ionizations occur and free radicals are formed in the materials. We all know that free radicals are highly reactive and tend to rapidly trigger certain chemical reactions. The most important effects of these reactions are chain scission and reticulation. These two opposing effects are explained below.

Chain scission, which results in a decrease in the polymer's mean molecular weight, is thought to be favored by the material's crystallinity, by the presence of oxygen and by the time that elapses during irradiation. One of the most well-known degradation mechanisms is oxidative degradation caused by diffusion of the air's oxygen through the polymer's structure during irradiation. The presence of this oxygen leads to formation of peroxide ($-O-O-$), carboxylic ($-COOH$), aldehyde ($-CHO$), ketone ($-C=O$) and hydroxylic ($-OH$) groups. The presence of these functional groups usually negatively affects the material's properties. Obviously, the appearance of these functional groups will depend a lot on the complete material composition (additives such as antioxidants, plasticizers and coloring agents can improve the polymer's radiostability). But the polymer's response to irradiation also depends in large part on the amount of radiation received (dose) and the speed at which it has received it (dose rate). Thus, in the case of polymers that tend to degrade with irradiation, a partial solution is to make it receive the desired dose in the shortest possible time so that oxygen diffusion will not take place on a generalized basis. Other options include performing the process in the absence of oxygen (in a vacuum or inert atmosphere). However, one of the drawbacks of these options is their high cost, and thus their use is very limited.

The other effect caused by the appearance of free radicals created during irradiation is reticulation, i.e. some chains joining to others through covalent links. This results in a three-dimensional structure that provides the material with some very advantageous properties for a multitude of applications. Reticulation changes a thermoplastic polymer into a thermostable polymer with completely different properties. Herein lies the major benefit of plastic material irradiation, as simple, cheap materials can be used to obtain other materials with a considerable added value thanks to their remarkable properties.

One of the major challenges that the plastics industry has successfully confronted during the last few years, thanks to polymer irradiation with

accelerated electrons, has been to find materials capable of replacing PVC in applications such as electric installation protection. As we all know, PVC has been widely used in this field. One of the reasons that it is suited to this field is its good performance against fire (it is flame-retardant), but one of the drawbacks is its very poor response to high temperatures, as it decompose and gives off toxic hydrogen chloride fumes. Thus its use in installations with a significant fire hazard is a serious safety problem. The solution to this drawback is to use halogen-free, reticulated polyolefins, and the best way to achieve reticulation, from a technical and economic perspective, is to irradiate with accelerated electrons.

The most important of all industrial applications, because of the volumes involved, is precisely reticulation of plastic wire, cable and pipe coatings.

Reticulation notably improves the heat and fire resistance properties of these materials, which are in high demand for automobiles, electronic equipment, nuclear power plants, etc.

SPECIFIC INDUSTRIAL APPLICATIONS

The main industrial fields where electron beams have been most actively and effectively applied are industrial hardwiring, the polyolefin foam industry, plumbing and thermo-retractile piping or films. In the last decade, the rubber tire industry has also grown in importance.

All these applications are related to polymers: modification of plastic materials or elastomers by subjecting them, for example, to reticulation of their molecules and transformation of monomers into polymeric chains.

The following table shows further details on reticulated products and the effects on their chemical or physical properties, the applicable polymers and the major uses.

The reticulated polymers most suited to industrial applications include the following:

Polyolefins: polyethylene (PE), polypropylene (PP), ethylene-acetate vinyl copolymer (EVA), ethylene-ethylacrylate copolymer (EEA), chlorated polyethylene (CIPE).

Vinyls: polyvinyl chloride (PVC)

Fluoropolymers: polyvinylidene fluo-



Productos tratados por irradiación β

ride (PVdF), ethylene-tetrafluoroethylene copolymer (ETFE), ethylene-chlorotrifluoroethylene copolymer.

Elastomers: natural rubber (NR), chloroprene rubber (CR), styrene-butadiene rubber (SBR), butadiene rubber (BR), isoprene rubber (IR), ethylene-propylene-diene multi-polymer (EPDM), isobutylene rubber (IIR), chlorosulfuric polyethylene rubber (CSPE).

OTHER INDUSTRIAL APPLICATIONS

• SURFACE TREATMENT

For example, on computer disks, electrical conductors, packing cases, motorcycle pieces, etc.

Electron beam treatment of compounds used in the aeronautical industry has proved to be a much more competitive process than thermal treatment, primarily because it is faster and the ability to penetrate is greater, thus providing greater homogeneity.

• GEM TREATMENT

Semiprecious stones (topazes, sapphires) are treated at high doses to make their colors purer and more intense. In this way, pieces with a high added value can be obtained from otherwise nearly worthless materials.

Gem treatment combines thermal treatments with electron ionization to obtain optimum results.

• ENVIRONMENTAL APPLICATIONS

Application of an accelerator to the effluent gases from a rubbish incinerator is being developed, especially in Japan, with a view to reducing the contaminant load.

Effects of Radiation on Polymers					
Classification	Product	Physical-Chemical Effects	Purpose of Effects	Applicable Polymers	Applications
Cables and Wires	Small-section cable Cables for electronics	Increased resistance to heat of polymeric insulating materials. Enhanced mechanical properties	Increase the range of temperatures for use. Improve resistance to abrasion. Increase resistance to chemical agents.	PE, EVA, EEA, CIPE, PVC, PVdF, ETFE, ECTFE	Hardwiring in electric & electronic equipment such as computers, audio equipment, lighting, etc. Hardwiring in cars, planes and spacecraft. Hardwiring in distribution boards, switches, etc.
	Medium-to large-section cables. Power and control cables	Increased resistance to heat of insulating materials.	Increase maximum current to be supported. Increase short-circuit current.	PE, EVA, EEA, EPDM	Hardwiring in plants such as thermal or nuclear power plants, chemical plants and blast furnaces.
		Increased resistance to heat of sleeve materials. Enhanced mechanical properties and chemical resistance of sleeves.	Increase range of operating temperatures. Improve response to especially hostile working conditions.	PE, EVA, EEA, CIPE, CSPE	Hardwiring in large buildings, theaters, shopping centers, tunnels, etc. Hardwiring in ships, transportation vehicles such as locomotives, etc.
	Heating cables. Self-regulated heaters.	Increased resistance to heat of insulating and lining materials. Enhanced mechanical properties and chemical resistance of sleeves.	Increase range of operating temperatures. Improve response to especially hostile working conditions.	Polyolefins Fluoropolymers	Pipe and tank heating.
Plastic Foams	Plastic foam sheets	Modification of viscoelastic properties in molten state. Increased resistance to heat.	Enhance performance in foaming process. Improve processability for a second processing. Increase working temperature.	PE, EVA, PP	Thermal insulation materials.
Pipes and Film	Thermo-retractile pipes. Thermo-retractile film.	Give memory effect. Increased resistance to heat.	Give thermo-retractile properties. Increase range of operating temperatures.	Pipe: PE, EVA, EEA, CIPE, PVdF, ETFE, CR, PVC Film: PE	Pipe: Cable joint insulation. Piping protection against corrosion in joints. Film: Packaging in general. Piping protection against corrosion in joints.
	Non-thermoretractile pipes. Non-thermoretractile film.	Increased resistance to heat. Enhanced mechanical properties.	Increase range of operating temperatures. Improve resistance to traction (film)	Pipe: PE, EVA, EEA, PVC Film: PE, EVA	Pipe: Cable joint insulation. Film: Extra-strong containers.
Piping	Piping	Increased resistance to heat. Enhanced mechanical properties at high temperatures.	Increase pressure and temperature allowed in hot water or gas distribution systems. Minimize wall thickness.	PE, PVC	Ventilation systems. Central and radiant floor heating.
Medical Products	Plastic medical products	Increased resistance to heat.	Increase resistance to heat to allow pasteurization with pressurized steam.	EVA	Medical liquids capsules.
Tires	Tires	Decreased deformation of untreated rubber (providing a semi-treatment)	Prevent rubber deformation in the shaping process. Enhance product dimensional stability.	NR, SBR, BR, EPDM, IR	Automotive in general.

Several facilities have been built to disinfect sewage. It has been demonstrated that 99.9% of coliforms can be eliminated with a dose of around 500 Gy.

Reticulating rubber with electron beams is also more beneficial than classic vulcanization with sulfur from the environmental standpoint because, by avoiding the addition of sulfur, eventual product incineration after the product's effective lifetime is over will not generate acid smoke.

• INDUSTRY RESIDUAL SLUDGE DISINFECTION

Excellent results are being obtained from the treatment of sludge for producing compost for fertilizers.

FACILITIES

The facilities for industrial application of accelerators should be equipped with the following elements:

- *Ionization area.*- This is the place where the electron generation source is located and where the material sterilization process is carried out. In this area, which is lined with concrete walls, the products are moved through a labyrinth on an automated conveyor, the speed of which will depend on the density, volume and dosimetry of the products to be treated.

The ionization area is connected to the two warehouses via the labyrinth and conveyor.

The electron generation source (Accelerator) is located inside the so-called *irradiation cell*. Once the electrons are accelerated, they are conducted through a window or *scanner* that, by way of a screen, projects these electrons onto the products to be treated.

- *A special conveyor system*, which is fully automated and controlled, that will carry the product to the window (scanner) for exposure, at the speed required according to the different parameters and adequately identified and controlled.

- *A management and control system* for the main equipment, accelerator and conveyor system, as well as for auxiliary safety, ventilation, cooling, etc. equipment.

- *A general warehouse* for receiving, classifying, storing and controlling both untreated products and treated products. To prevent them from getting mixed up, a physical barrier will be

provided to partially divide this warehouse.

- *A dosimetry laboratory*, provided with all elements required for monitoring, reading and certifying the dose applied in each case, as well as the Radiological Protection system used in this type of facility.

The purpose of the Plant is to certify predetermined doses applied to the products to be treated, and therefore parameter control and traceability or quality control systems in particular are essential for this type of activity.

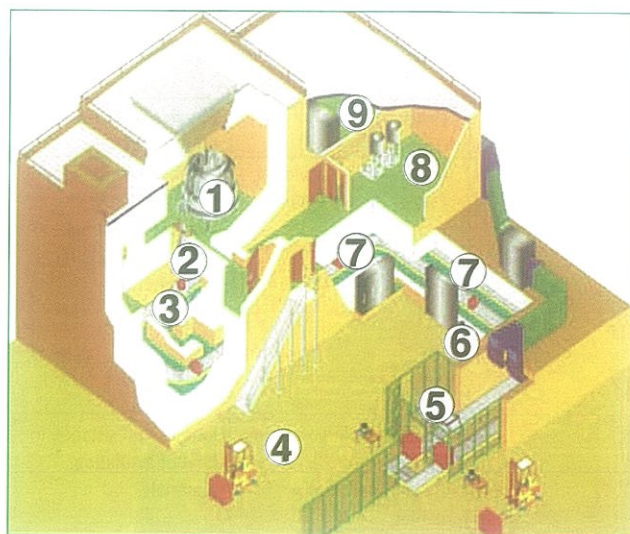
REGULATION AND LICENSING REQUIREMENTS

Electron accelerator facilities in Spain are considered as second-category nuclear facilities, and as such they must be licensed by the *Consejo de Seguridad Nuclear*. They must comply with applicable Safety and Radiological Protection standards.

As dedicated medical or food product sterilization plants, they must also be approved by the Ministry of Health and Consumption. International Standards have been established to regulate the qualification and use of products, processes and facilities. The two main standards are ANSI / AAMI / ISO 11137 and EN 552, among others.

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| 3- ZONA DE TRATAMIENTO | 8- SALA DE REFRIGERACIÓN |
| 4- ALMACÉN | 9- SALA DE CONTROL |
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