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RADIATION PROCESSING USING ELECTRON LINACS

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Summary

The performance capabilities of a modern linac make it a commercially viable tool for enterprising applied radiation chemists and biologists. Good economics requires efficient transformation of mains power to beam power and although pulsed travelling wave linacs have been used in industrial applications for several years, their high cost per watt is a disadvantage. The cw linac with its variants is a developed technology which could be used profitably in selected applications. More integrated designs like the self-excited linac and the induction linac also offer promise for the near future. Linac design optimization in relation to the physical and chemical processes desired in the irradiated product is discussed.

Introduction

The ability of energetic electrons or photons to break chemical bonds directly, and release chemically active free radicals, has led to important applications. Accelerators supply 93% of existing industrial radiation power[1] while radioactive sources such as Co^{60} and Cs^{137} supply the remainder. Choice of a source is determined mainly by the intensity and penetration of the radiation hence low voltage electron accelerators dominate surface, high-dose applications while gamma-ray sources are used for high value products requiring penetration, such as in the sterilization of medical disposables. The linear accelerator has the potential to compete over the whole field with its ability to combine the positive features of these traditional technologies for application to bulk materials. The radiation from the linac can be penetrating, cover a wide power range and its flexibility can be tailored to many established manufacturing processes. Moreover the enormous unit power can be expected to uncover new applications within the chemical and resource industries.

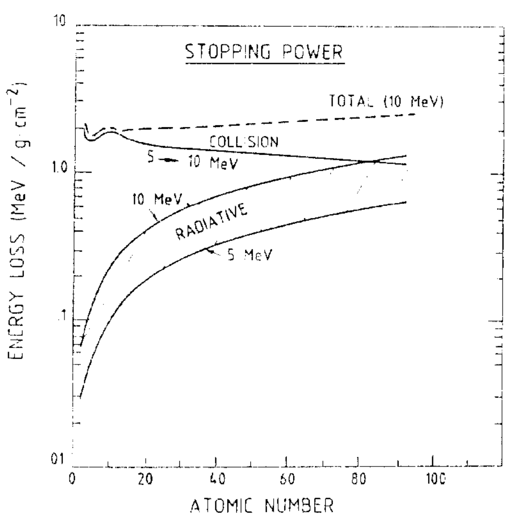

Fig. 1(a) Variation of electron energy loss with atomic number.

Radiation Physics

The linac can provide either penetrating electrons or photons as a consequence of the basic interaction processes[2] shown in Fig. 1(a). For low atomic number targets, ionizing collisions dominate, giving rise to emission of electrons. Radiation of photons (bremsstrahlung) is more probable in targets having nuclei with high atomic number. The radiative probability increases rapidly with energy as shown in Fig. 1(b) and for a thin tungsten target exceeds the ionizing collision probability at energies greater than 10 MeV. For thick targets, source absorption of

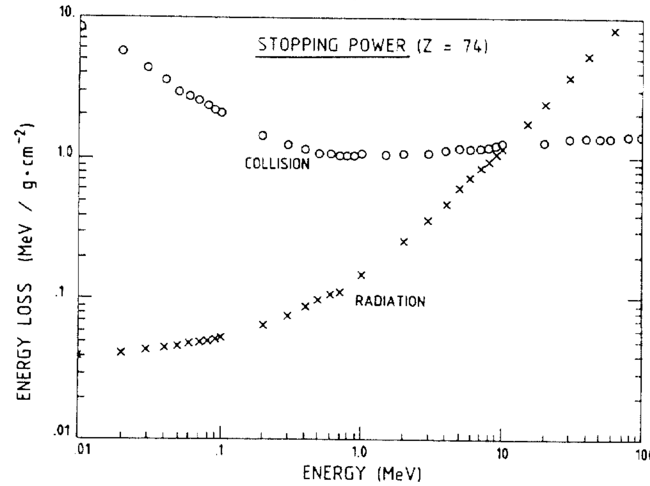

Fig. 1(b) Variation of collision and radiative cross-sections with energy.

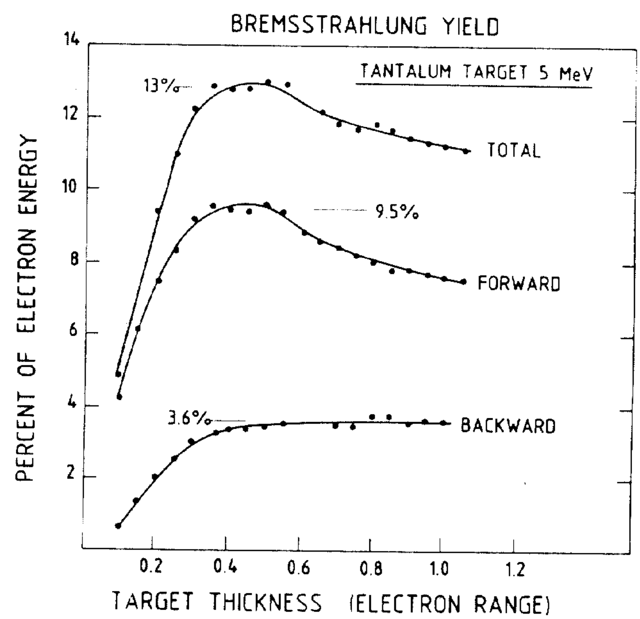

Fig. 2(a) Bremsstrahlung yield vs thickness of radiator.

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low energy photons must be included and combined with the lower radiative cross section of electrons deenergized by collisions. The net photon yield[3] for 5 MeV electrons is shown in Fig. 2(a). The bremsstrahlung yield[2] in the forward direction is further enhanced by kinematic factors as shown in Fig. 2(b).

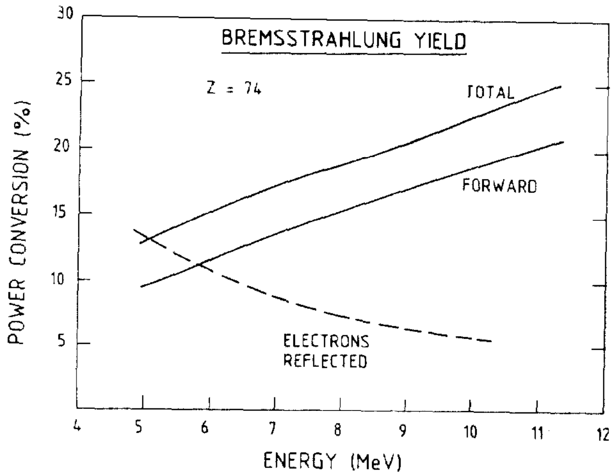

Fig. 2(b) Variation of bremsstrahlung yield with energy.

Chemical or pathologenic changes in an irradiated object are related to the power of the accelerator by the simple relation

$$P = (D/f)(M/T)/3600 \qquad (1)$$

where P is the exposure power in kilowatts, D is the absorbed dose in kGray, M is the mass of treated material in kilograms, T is the exposure time in hours and f is the power utilization factor. All of these quantities have simple definitions but, f, is critically dependent on absorption processes and geometric factors. In some bulk applications, such as food irradiation maximum and minimum doses are specified and Fig. 3(a) shows that for a single sided irradiation with electrons only the shaded area in a homogeneous target like water results in a useful dose for a specified max/min ratio of 1.5, the associated value of f is 0.7. However for two sided irradiation not only is the treatable thickness more than doubled but f is increased to 0.85. For bremsstrahlung, two sided irradiation is even more desirable, the thickness being increased by a factor of 5 and f increased from 0.2 to 0.3 (see Fig. 3(b)).

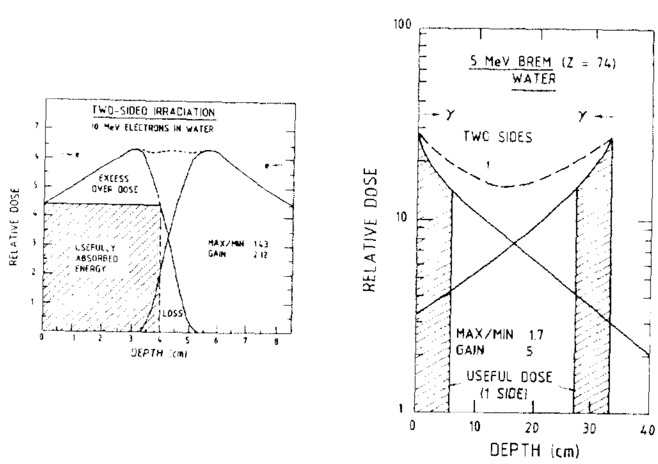

Fig. 3(a) Energy utilization efficiency for electrons.
(b) Energy utilization efficiency for photons.

High energies (≈ 10 MeV) bring attendant activation hazards and this is of particular concern in irradiating food. The Codex Alimentarius[4] has taken a conservative approach to such perceived problems and has set specific limits of 10 MeV and 5 MeV respectively for electron and photon irradiation of food. The problem arises mainly from neutrons created in the radiator and the target by photon interaction. Figure 4(a) shows the number of neutrons generated from (γ,n) reactions in a thin tungsten radiator and a 'thick' water target[2]. In the latter case the neutrons arise from the 150 ppm concentration of D_2O in water. The neutrons are emitted isotropically but as the photon yield is mainly forward peaked the geometry is not critical in assessing the rate of neutron production.

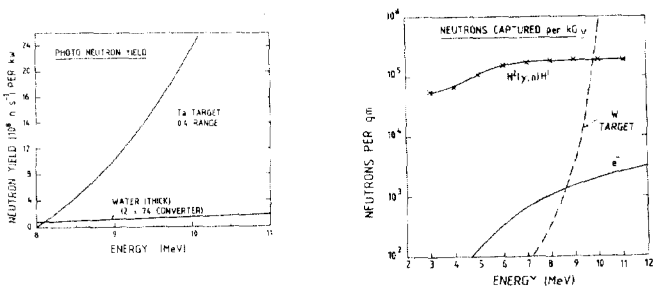

Fig. 4(a) Number of neutrons produced from a thin tungsten radiator and a thick water target.
(b) Variation with energy of neutron absorption in food for electron and photon irradiation assuming elemental composition of human body at a source/produce distance of 30 cm.

The situation is quite different when capture probability is considered and dose absorption efficiencies must be combined with neutron diffusion parameters. Slowing down is due mainly to hydrogen atoms, hence Leboutet in a recent study[5] assumed a pure water medium and diffusion lengths the same as for fission neutrons in a reactor. An 8.5 cm thick planar water volume formed the irradiated medium for electrons and a 20 cm thickness for γ irradiation. The relative effects of photon and electron produced neutrons in wet food are compared with those resulting from the target in a typical irradiator geometry in Fig. 4(b). The activity created by these absorbed neutrons (Na^{24} is the major contributor) and other competing nuclear processes is typically similar to C^{14} contamination one hour following irradiation and a few percent of that from K^{40} contamination after one day.

Irradiator Requirements

A typical gamma-ray facility with one MCi of ^{60}Co sterilizing disposable medical devices in shipping cartons will process annually 28 300 m^3 with density 0.2 and dose 25 kGy. As dose distributions are rather more favourable for electrons, an electron accelerator of ≈ 10 kW and having sufficient energy could duplicate such a facility[6]. However, the linac's ability to provide much greater power can enable one shift operation and compete with the larger cobalt units (up to 4 MCi) already in use, hence a competitive linac might have a beam power of 120 kW.

The conventional linear accelerator consists of several subsystems and these are depicted in Fig. 5. The poor overall energy efficiency of the linac is a disadvantage and the figure shows the maximum subsystem efficiencies[7]. Even though simultaneous operation at peak efficiencies of all components may be possible, mains to electron beam conversion at 10 MeV with an efficiency greater than 50% will be difficult to achieve. The linac therefore lags behind the widely used insulated core transformer (efficiency

85%), or Dynamitron (50%) dc accelerators in terms of efficiency, hence a premium is paid for penetration.

The subsystems of Fig. 5 have evolved over several decades of linac development and a review of the current status of dc power supplies, modulators, rf sources, electron injectors and detailed structure design lies outside the scope of this review. A decision to use a linac would be dominated by economics and compatibility with industrial manufacturing practices. The accelerator physics problems at high current such as higher order mode excitation, beam break-up, field gradient limits and beam emittance parameters are secondary considerations.

Microwave Accelerators

Several decades of design evolution of the microwave linear accelerator for high energy applications makes it a strong candidate for the core of an industrial irradiator.

Pulsed Linacs

Travelling wave accelerators have reached a mature state of development and in the past have been favoured for pulsed electron machines. However, standing wave linac structures have higher shunt impedance than travelling wave linacs especially for particles not fully relativistic. For high gradient applications with a source of relatively low power (e.g., a magnetron) and a pulse length of a few rf filling times (such as for medical accelerators) they are especially suitable. These structures have also found wide application in $e^{\pm}$ storage rings where particles of opposite charges are accelerated simultaneously in opposite directions. However they are generally built with a fixed coupling and at best 80% of the rf energy can be delivered to the structure during the pulse. Also as the fields take several filling times ($2Q_L/\omega$) to build up to their final value the beam will vary in energy throughout the pulse. More fundamentally, as the system works in stored energy mode, to achieve 40% beam loading, the average energy will drop by about 20% unless fast control of the rf power system is achieved. So far such control has not been demonstrated.

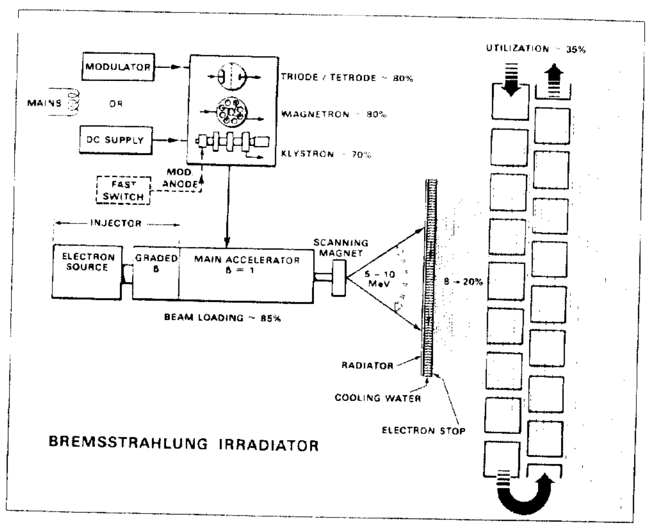

Fig. 5 Component efficiencies for linac irradiator.

To illustrate the problem, Fig. 6 gives measured data from an S-band standing wave structure recently developed at Orsay for the 30 MeV LEP injector[8]. It is believed that this machine's performance will be duplicated for a 10 MeV end point energy as the particles are relativistic after the first two cells and hence their phase is fixed. For the LEP injector the gradient of 13.6 MeV/m is provided by an 11 MW magnetron and the expected beam loading parameter is 0.0563 MeV/nC. The measured energy spreads containing 80% of the electrons are 5, 11.5 and 19.6% for 3, 50 and 100 nC per pulse. The measured field droop of 11% for an accelerated charge of 50 nC is consistent with 20% loading for the 2.5 A pulse. The consequences of these beam loading effects were investigated with the SANDYL Monte Carlo code at CRNL[2] assuming a 10 MeV machine and Fig. 7(a) shows the change in depth dose distributions for the three loading conditions above. The effect is most evident in the min/max ratios for two-sided electron treatment (Fig. 7(b)). However the most dramatic consequence is likely to result from contamination of the beam by high energy particles above photo-neutron threshold where neutron yields increase rapidly with energy.

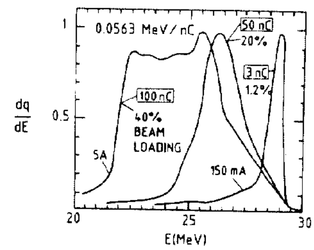

Fig. 6 Typical energy spectra for the LEP injector.

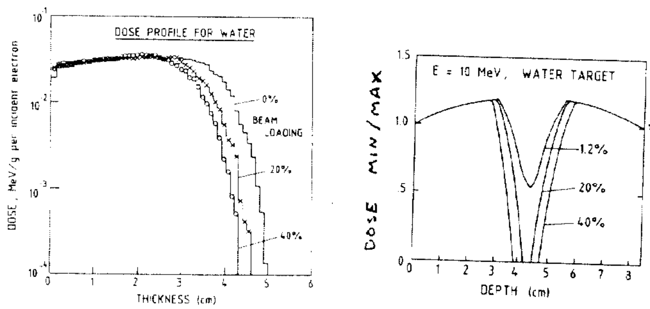

Fig. 7(a) Change in depth dose curves with beam loading.
(b) Variation of min/max ratios with beam loading.

Such a dramatic beam loading effect does not occur in an equivalent travelling wave machine. Haimson[9] has designed and constructed a 10 kW machine which has demonstrated beam loading of 87.5% with two-thirds of the 1.1 A beam passing through energy analyzing slits with $\Delta E/E$ of 3%. This machine has been running at Riso[10], Denmark since 1975 and is used for product irradiation (40%) and pulse radiolysis experiments (60%). Heavily loaded travelling wave linacs are also used in radiation chemistry research and Tabata[11] has developed a system with two parallel S-band linacs which can accelerate electron beams with a width of shorter than 10 ps.

CW Linacs

Because rf power dominates linac capital and running costs the cw linac provides an attractive radiation source. At CRNL we have been experimenting with cw linac structures for almost 20 years and have succeeded in developing side-coupled, on-axis coupled and coaxial coupled structures which are stable up to gradients of 2 MeV/m. Numerical methods to optimize cavity geometry, analyze thermal stresses and design cooling systems for minimum thermal detuning of the structures[12] has been complemented with similar approaches to studies of the beam cavity interaction, higher order mode excitation and beam blow-up phenomena. These computer studies have been supported by a program of beam experiments using the Electron Test

Accelerator (ETA) as a test bed for beam loading work and as a prototype irradiator[13]. Figure 8 shows the effects of beam loading at high average power and demonstrates the high degree of control that is possible with the cw linac. Over a beam loading range of 0 to 50% the accelerating field remains constant, hence the output electron energy also remains constant.

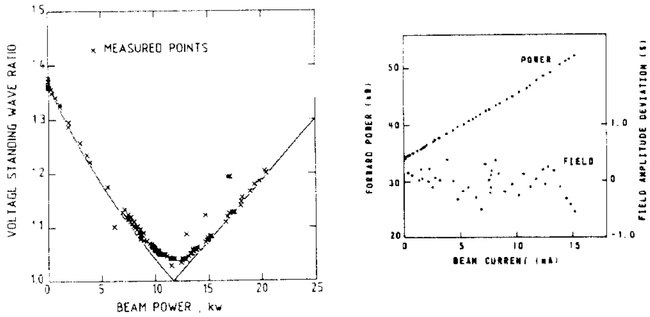

Fig. 8 Effect of beam loading in a cw linac (a) shows the change in impedance match and (b) demonstrates that the fields can be held constant (≈ 1%) at up to 50% loading.

Recently a design group at CRNL[14] has studied the technical viability of building a cw linac that can produce 500 kW of beam power at 10 MeV. The design, which incorporates a novel start-up procedure, has a frequency of 2.45 GHz, gradient of 2 MeV/m, beam loading of 63% and a consequent overall efficiency of 30% for 50 mA operation.

The cw linac can also be used for irradiations at a lower power level by operation at less than 100% duty factor as a long pulse linac. 'Ohm's Law' for linacs is $P = E^2 \cdot D/[(ZT^2) \cdot L]$ where P is the structure dissipated power, E is the energy gain, D the duty factor, (ZT^2) the shunt impedance and L the length. Considering arbitrarily two conditions of the CRNL study, E = 10 MeV and loading of 62%, Fig. 9 shows how duty factor effects the average power levels. The arrows are drawn at 15 kW/m which is the level where radial cooling is necessary. The curve for a structure length of 2.5 m is a special case as this can be driven from a single source of 1.6 MW peak eliminating multi-tank control systems that require resonance control at a fixed frequency and phase control loops. Frequency shifts can be easily compensated using a voltage controlled oscillator thus eliminating the need for tuners. These 'pseudo-cw' conditions have two main advantages, namely elimination of the modulator of a pulsed linac by using the modulated anode of the klystron (see Fig. 5) and the

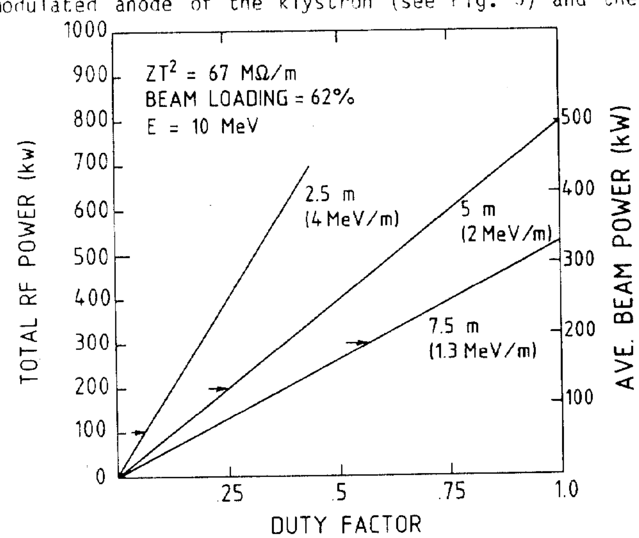

Fig. 9 Design curves for long pulse machine.

use of precise 'cw linac' control of the accelerating fields. As all configurations of Fig. 9 are heavily beam loaded there is no advantage to be gained with more complicated recirculation schemes which carry with them the hazards of regenerative beam blow-up.

New Linac Concepts

The previous chapter described accelerator structures conceived in an era where the achievement of high energy was the main goal. Industrial linacs are increasingly more reliable, with improved vacuum and cooling systems, klystrons lifetimes greater than 10 000 hours, high brightness dispenser cathodes and installation lifetime expectations in excess of 20 years, nevertheless the fundamental pedigree of the microwave linac remains unchanged.

Russian scientists have recently patented[15] an industrial version of a self-excited linac. The coupling loop of the accelerating resonator is capacitively coupled to the anode of the oscillator tube to form a two-circuit oscillation system. This permits a considerable improvement of electron efficiency, reduction in size and simplification of construction. A drawing of this machine, ILU 6, is shown in Fig. 10(a). The frequency is 110 MHz, gap voltage 1.5 MV and with a 2 MW triode (≈ 2% duty factor) the power output is 20 kW with 4 kW dissipated in the structure. In one run it operated continuously for 500 hours.

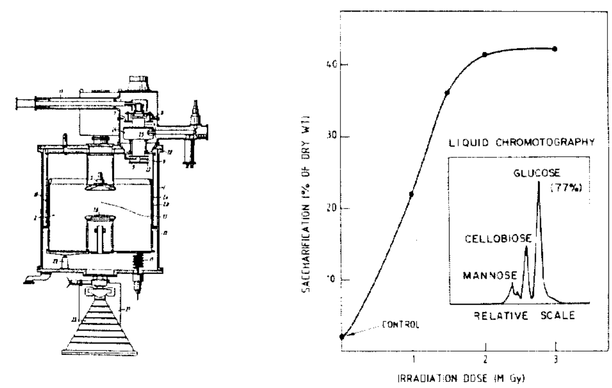

Fig. 10(a) Drawing of industrial self excited linac taken from U.S. Patent No. 4 140 942.
(b) Saccharification of spruce wood as a function of dose.

A French structure which combines the rugged simplicity of the TH 116 triode (used in most proton injectors) with a 3-cell π-mode structure at 166 MHz forms the nucleus of the CASSITRON[16]. Both Russian and French design operate in a long pulsed mode. The Russian machine is used extensively for vulcanization of rubber. The French machine is in an advanced stage of construction and one is scheduled to begin irradiating mechanically separated poultry meat in July 1985 in Paris.

These self excited linacs eliminate many beam control devices but the induction linac[17] makes an even more direct contrast with the traditional high energy gradient, low current research accelerator. Recent work with the experimental test accelerator at Livermore has demonstrated 5 MeV, 10 kA at 1 kHz operation. Power levels up to 1 MW have been accomplished and with the development of new magnetic switches allowing burst rates of 15 kHz, enormous radiation fields are possible.

In the 50 MeV Advanced Test Accelerator also at Livermore, the self-focusing electron beams and the low-Q ferrite structures are observed to overcome beam break-up, which further emphasizes the potential of the induction linac. Although some demonstration food irradiation experiments have been done at Livermore with bremsstrahlung radiation at 2 MeV, assimulation of induction linac technology by the radiation community is likely to be protracted.

Radiation Applications

There are many excellent reviews of radiation processing in the literature[18] and only those applications that might benefit from recent linac developments will be mentioned here. The penetration of the radiation from linacs may expose new opportunities. These opportunities are enhanced by impending government bans on ethylene oxide as a disinfectant and ethylene dibromide as a fumigant. Even though the Codex Alimentarius recommendations for food have been accepted by 30 countries and commercial scale gamma activities are already under way in several countries, the application of linacs has, so far, been limited as shown in Table 1.

Table 1

Accelerators for Food Irradiation, February 1985

COUNTRY	LOCATION	TYPE	APPLICATION DESCRIPTION	STATUS
CHINA	Beijing Radiation Centre	LINAC (5 MeV)	Experimental, Demonstration	Operating being dismantled
ECUADOR	Escuela Politecnica Nacional, Quito	LINAC (6 MeV, 2 kW) USSR	Pilot Plant	Expected 1986
FRANCE	SODETEG, Paris	LINAC (6 MeV, 7 kW)	Demonstration, Multipurpose	Operating since 1968
	Société GUYOMARCH, BRITANNY	'CASSITRON' (CGR) (10 MeV, 10 kW)	Commercial, frozen deboned poultry	Expected July, 1985
FRG.	Karlsruhe	d.c. (2), LINAC (Varian) (10 MeV, 6 kW)	Experimental, Demonstration Multipurpose	Program Terminating
	Hamburg 'Anton Dohrn'	X-ray, 200 kV, 30 kW	Demonstration Multipurpose	Probably out of use
GDR.	Leipzig	LINAC	Food, multipurpose	Planning
ISRAEL	Sorcy NRC, YAVNE	ICT, 1.5 MeV 75 kW	Pilot Plant, Poultry feed	Operating
MEXICO	Institute of Physics UNAM, Mexico City	Dynamitron 3 MeV, 25 kW	Experimental Maize Disinfestation	Program Terminating
MALAYSIA	PUSPATI, Selangor	Japanese ICT.	Multipurpose Pilot scale	Planning
NETHERLANDS	ITAL, Wageningen	Van de Graaff	Experimental	Program Terminating
POLAND	Technical University Lodz	LINAC (Soviet)	Multipurpose	Completed 1983
USA	LLNL, Livermore, CAA	Induction Linac	Experimental, Demonstration	Operating 1985
USSR	Odessa Port Elevator RDU, Odessa	Two ELV-2 1.4 MeV, 20 kW each	Grain disinfestation Commercial 200 t/h	Operating since 1980
	VNIIKOP, Birjulovo(Moscow)	LINAC	Multipurpose	Being dismantled

* Based on compilation by J. Farkas, IFFIT, Wageningen (IAEA).

The increased penetration of linac based radiation is unlikely to change the rate of growth of applications relating to the environment[19] mainly because of the economics of competing processes. However, waste water treatment (with ozone enhancement), sludge disinfection for reuse, international airport waste disinfection and simultaneous removal of SO_2 and NO_x from fossil fuel power plants, could all benefit from the high unit power of the linac. Economic considerations are important in many of these applications, applying direct pressure to keep capital and operating costs at a minimum.

Adoption of linacs by manufacturing industries for in-line processes is likely to be inhibited by the 10^{10} to 10^{11} shielding attenuation factor needed for even a modest sized linac. Shielding costs are almost independent of power (a factor of 10 in power will change shielding cost by ≈ 10%). Self shielding is not possible at energies above a few MeV. Where penetrating radiation is required the standard manufacturing processes must be reorganized to suit the source. Where this can be done, the linac is likely to be preferred over radioactive sources because of its flexibility.

The applications mentioned above continue to be studied and although many are not yet commercial, it is likely that linear accelerators will be increasingly adopted as the preferred source. However the greatest opportunities for the machines discussed in previous sections undoubtedly lie in activities still in the laboratory stage. These include applications in the forest product industry, particularly the degradation of waste cellulosic materials for the production of liquid fuels and valuable sugars[20]. Enzymatic saccharification of lignocellulosics, pretreated with electron beam irradiation, has been studied by Kumakura and Kaetsu[21] and increased saccharification of 10 to 15% in newsprint was obtained. Figure 10(b) shows results of a recent experiment with spruce wood chips irradiated in the 4 MeV electron beam at Chalk River and subsequent enzyme hydrolysis carried out by A.W. Khan at National Research Council, Ottawa. The sugars released were estimated both by calorimetry and, as indicated in the insert of the figure, by liquid chromatography. In spruce wood 46% is cellulose and the figure shows complete conversion to sugars is achieved at 2 MGy with glucose, the most valuable sugar, comprising 77% of the total.

This last example serves to illustrate the beneficial use that could be made of waste products. Production of new chemicals and detoxification of by-products from the chemical industry have not been studied in detail although radiolytic destruction of PCB's has been demonstrated[22]. Already radiation can compete favourably with steam processes in the food industry and it could provide energy savings in many traditional thermal processes. Accelerator technology has reached a level of development which is not easily adopted by a fledgling radiation processing industry accustomed to low power radioactive sources for bulk processing, however, the modern linac provides radiation chemists with many new challenges and exciting opportunities.

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