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Electron Beam Driven Industrial Chemistries

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The purpose of this white paper is to provide an overview of radiation chemistry and existing industries that use radiation processing, highlight the advantages and disadvantages of the current technology, introduce new emerging electron accelerator beam applications and provide a summary cost comparison with conventional technologies.

Brief History of Radiation Chemistry

The discovery of X-rays by Wilhelm Roentgen in 1895, radioactivity by Antoine Henri Becquerel in 1896 and polonium and radium by Marie and Pierre Curie in 1898 set the early stages of radiation chemistry. Radioactive sources enabled studies on the effects of ionizing radiation on various materials; however, the experiments were very limited due to the absence of high-energy ionizing radiation. With the invention and development of charged particle accelerators in the 1930s, radiation chemists began investigating radiation effects on a range of materials (food and pharmaceuticals) [1]. In 1940, the Manhattan Project motivated scientists to discover radiation resistant materials for the purpose of building nuclear reactors. The 1950s were dedicated to utilizing radiation processing in various industries. Arthur M. Bueche and Elliott J. Lawton from General Electric did some of the pioneering work on using radiation to crosslink plastics [1]. Since then, the following scientists have studied the effects of radiation on materials and greatly contributed to the field of radiation processing: Arthur Charlesby (UK) [2] and Adolphe Chapiro (France) [3] studied radiation-induced polymeric systems in the 1960s, John Spinks and Robert Woods (UK) [4], Sueo Machi and Yoneho Tabata (Japan) [5], Vivian Stannett and Joseph Silverman (USA) [6], and Alexei Pikaev (Russia) [7] developed radiation chemistry handbooks in the 1990s.

Current US Chemical Industry

There are roughly 13,500 chemical plants in the United States supplying both the domestic and international market, exceeding $800 billion in profits, employing over 3 million people, dominating research and development and making this country one of the top leaders in chemical industry globally [8], [9].

Typically, industrial chemical reactions are driven by heat, pressure and catalysts. All these factors play an important part in many chemical processes. Temperature tends to excite particles' movement and high-pressure draws particles together, both increasing the probability of collisions. Catalysts are required to increase the reaction rate. The attractiveness of catalysts is that they are not consumed by the reaction and are only needed in very small amounts; however, they require removal of chemical contamination from the product. In 1992 efforts spent on developing industrial catalysts was estimated at 500M to 1B dollars [10]. From 1993 to 2016 the number of US original patents relating to catalysts has increased from 479 to 721 [11]. The U.S. chemical industry consumes more than 8.5% of the power in the end use sector [12] and significantly impacts the environment by emitting over 1.5 million tons of air pollutants and toxic waste [9]. The amount of money spent, energy consumed to run chemical plants, and number of patents awarded in the field of catalysis demonstrates a desire to make chemicals in new and more energy efficient ways.

Chemical Manufacturing via Electron Beams

Electron beams are an exceptional source of energy that are capable of initiating chemical reactions without the need for catalysts, high temperature or high pressure. The high kinetic energy and penetrating nature of the electrons provides significant benefits over typical chemical methods. When an electron with sufficient energy interacts with matter, it can remove a tightly bound electron from an atom, which results in an ionized or charged atom [13]. Such charged atoms are unstable and react quickly allowing for the formation of new molecules.

Industrial electron beam processing commonly employs carbon-based materials that are typically monomers, oligomers or polymers. The chemical reaction of interest, for instance, when treating polymers is the formation of free radicals from the homolytic cleavage of carbon-hydrogen bonds, which leaves unpaired electrons [5]. Electron beam processing involves the absorption of large doses of energy from accelerated electrons in materials in order to modify them in some beneficial manner. The main processes initiated by electron beam are polymer modification by crosslinking or scission, curing of coatings, decomposition of industrial effluents, or synthesis of a new substance. Some materials that have been successfully processed via electron beam include plastics and rubber, wire and cable insulation, crosslinking of ultra-high molecular weight polyethylene for hip and joint replacement in the medical industry and many more. Beneficial changes produced in treated materials are improved thermal and chemical resistance, stability at elevated temperatures, improved tensile strength and other mechanical properties. Electron beam technology provides an efficient, safe and environmentally friendly way to drive chemical reactions. This technology is used in a vast array of industries and common consumer products, in which sales eclipse $2B annually, providing an estimated added value to products of more than $500B every year worldwide [14], [15].

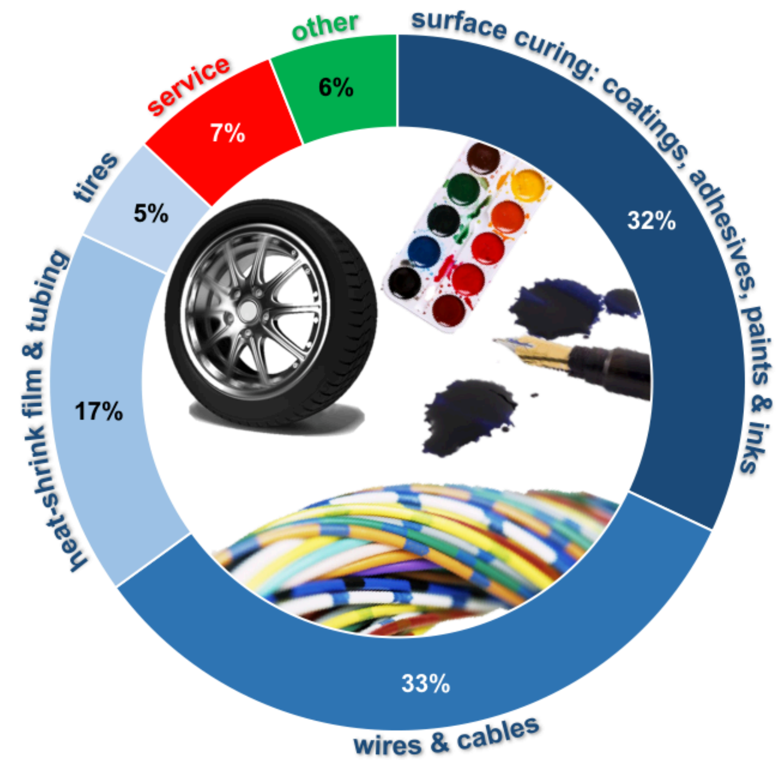

Figure 1: Current End-use Market Distribution of Electron Beam Industrial Applications – 87% of e-beam-induced processes involve crosslinking, occurring in applications represented by the blue segments in the pie-chart [14], [16];

The energy, the current and the line speed of the process are key characteristics in electron beam accelerators. The energy determines the penetration depth of the beam; higher energy equals deeper penetration. The current is a count of electron flow past a given point, which is closely related to the dose that the product receives. The dose required to perform the physical and/or chemical changes of the product brought by the electron beam treatment is associated with the processing cost. Finally, the line speed represents the speed at which the product passes through the stream of electrons; for a specified dose, a rapid line speed will require a higher current, while a slow line speed will require a comparatively lower current to deliver the same dose. In addition, the power capability of an accelerator plays a vital role in the product throughput. Typical industrial electron beams can operate at up to 10 million electron volts (10 MeV) without any material activation and have a power from 15 kW to 150 kW contingent on the process application [17]. Current accelerator capabilities are not enough for some applications. Therefore, there is a growing need for high-power, higher efficiency and mobile accelerators to drive different applications.

Advantages of Current Electron Beam Driven Chemistry

- Create Products with Different Properties
  - It can reduce product damage due to oxidation and prevent polymer embrittlement.
  - It can make purer products because the chemistry can be driven without initiators, catalysts or other processing aids.
- Environmentally friendly – Reduction of chemicals; hence, reduction of toxic byproducts
  - Electron beam processing does not leave chemical residues in the products being treated. Because the radiation sources in electron beam accelerators are electric-powered, they do not entail the handling, licensing, shipping, disposal or use of radioactive isotopes. Electron beam driven curing, for example, can be designed with no Volatile Organic Compounds (VOCs) .
- Safe – No residual radiation
  - The electron beam's penetration into a material is determined by the mass and density of the target and the kinetic energy of the electrons striking the target. Most of the energy is used to crosslink or damage the target. Activation of materials can be avoided by careful matching of the electron beam energy with the materials' elemental composition.
  - A heavy-duty concrete shield effectively prevents radiation from leaving the irradiation chamber. A whole system of safety features such as controlled access, failsafe systems, radiation and ozone level monitoring, auto power down sensors etc. ensures that electron beam processing is thoroughly safe.
- Efficient
  - Electron beam accelerators can operate nonstop with minimal interruption. There is no loss in efficiency as the radioactive source is not dependent on any radioisotopes that decay over time. The radiation is switched on or off with a flick of the electric switch.
  - The reduction and in some cases even elimination of high temperatures reduces the energy cost.

- Current Savings

Industry	E-beam processing	Benefits	Compared to conventional technology
Tire	Crosslinking of rubber	Improved thermal and mechanical properties while low consumption of rubber;	16% savings of total manufacturing cost per tire [18]
Wire & Cable	Crosslinking of polymeric material such as PE, PVC, EVA or EPDM	Improved thermal properties, chemical resistance, tensile strength; fire retardant;	18% savings of average cost per km of product [19]
Coatings, Adhesive, Paints & Printing Inks	Chain scissioning of PTFE	Improved mechanical properties and functionalization;	25-65% savings of manufacturing cost [20]

Table 1: Examples of Industrial E-beam Processing, Benefits and Estimated Savings

Current Drawbacks of Electron Beam Driven Chemistry

- The process is dose/dose rate dependent and low chemical yields of the product are possible if the process is done without a catalyst or unless a chain reaction is present.
- Operation of current electron beam accelerators is more expensive than use of traditional chemical processes for most chemistries.

Due to the relatively high cost of currently available electron beam accelerators, electron beam processing is economically feasible only if the value or amount of the product treated is larger in comparison to the energy used. For instance, electron beam chemistry is commonly used in the modifications of various industrial products including crosslinking of polymer-based materials to improve materials' properties, sterilization of medical devices, cosmetics, pharmaceuticals and degradation of materials in the recycling industry where the cost of the energy used to treat the product is not relevant in comparison to the dramatic value added to the product. However, new advances in the accelerator field have the potential to mitigate this disadvantage.

Conventional surveys show that over 1400 high-current electron beam accelerators, not counting about 1000 low-current machines utilized for research, are commercially employed and have added over $85 billion in various products' value [18], [21]–[23]. In summary, e-beam accelerators are dependable and robust instruments that can drive industrial chemistry when needed for a specific commercial usage [24], [25].

Current Industrialized Applications

Hoses, Tubing and Cables

One of the first and most well-known uses of electron beam processing is crosslinking of the insulation jacketing on cables. The benefits of this process lie in creating a tougher and fire-resistant material. Depending on the industry necessity, cable jacketing is most frequently made from polyethylene. If the application requires elasticity, then mixtures of polyethylene and ethylene-propylene rubber are used [26]. If temperature resistance is essential, polyvinylidene fluoride or other fluoropolymers are used [27]. Fluoropolymers benefit from being flame retardant but are also more expensive base materials.

Heat-shrinkable Films and Tubing

Heat-shrinkable tubing is also made from electron beam treated polyethylene. The process involves extrusion, electron beam to crosslink, heating to above the melt transition temperature of the plastic which makes the plastic behave like a rubber and can be strained. Once the plastic is cooled, the crystal structure of the crosslinked polymer helps preserve the shape in the stretched state [28]. In a similar fashion, heat shrinkable films have been constructed for the purpose of food packaging.

Polyethylene Foams

The automotive industry uses crosslinked polyethylene foams in cushioning for safety and protection. These are conventionally made by using thermo-chemical reaction to crosslink the polyethylene and then another to blow it into foam. With electron beam processing both polyethylene and a blowing agent are crosslinked at ambient temperatures, eliminating the need for thermal input [29].

Rubber Manufacturing

In 1933, electron beam processing to “vulcanize” natural rubber was patented by the Goodrich Company [30]. Vulcanization is the crosslinking process of elastomers [7]. The method was based on extrusion and then treatment with electron beam to bring the elastomer to a gel state which in fact is tougher than uncured materials. The finished tire is then knitted and fused together via thermal molding process [31]. Electron beam induced crosslinking is responsible for creating strong bonds in elastomers used in the tire industry [32], [33].

Printing and Other Thin Coatings

Electron beam cured inks, coatings and adhesives can be manufactured 750 times faster than conventional paint and drying practices [34]–[36]. One advantage of electron beam curing in this application is that the pigmentation does not affect the crosslinking which is a problem when using ultraviolet radiation. During the radiation treatment, electrons create free radicals in the vinyl terminated monomers leading to double bond opening, polymerization and crosslinking. This process is much faster and more efficient than the generally known convection heating in combination with thermo-chemical reactions in the presence of a catalyst [37].

Unindustrialized and Emerging Applications

Utilization of Bio-Fuels

Studies have shown that when exposing cellulose to ionizing radiation it causes degradation of the cellulose by scissioning of the polysaccharide [38]. This area is of very high environmental interest due to the ability to produce bio-fuels from cellulosic plant materials using electron beam treatment. Utilizing electron beam treatment as a degradation source can protect the environment by eliminating toxic chemicals in conventional methods and decreasing the energy consumption.

Water Treatment

Much interest has been generated in water treatment by electron beam irradiation due to the effectiveness of ionizing radiation in the decomposition of microorganisms and undesirable organic contaminants [39]–[41]. Applying radiation to treat water is a viable solution because when water is subjected to high-energy electrons it produces highly reactive species such as the hydrated electron, hydroxyl radicals and hydrogen atoms that destroy organic materials. Electron beam treatment is the only form of energy that produces highly oxidizing and reducing species at the same time.

Fluoro-Chemistry

Another area of interest is the development of proton exchange membranes for fuel cells that are used to power virtually everything, from drones to cars. The production process involves modification of a fluoropolymer such as polytetrafluoroethylene (PTFE), polyethylene-co-tetrafluoroethylene (ETFE), polyvinylidine fluoride (PVDF), polyethylene (PE), polypropylene (PP) and polytetrafluoroethylene-co-hexafluoropropylene (FEP) through radiation grafting copolymerization, followed by sulfonation. In comparison with ultraviolet or plasma irradiation, electron beam treatment has been demonstrated to be one of the most useful techniques in inducing grafting copolymerization reactions for membrane production, due to its penetration depth, which leads to a higher uniformity of grafting throughout the depth of the product. Electron Beam-induced reactions are chemically simpler than those involved in the use of conventional synthesis techniques, because they do not involve the use of initiators or catalysts. To obtain the desired membrane composition, control of the degree of grafting can be achieved through the adjustment of the grafting process parameters such as dose, dose rate and temperature. [42]–[44]

Petroleum Industry

Current refining capabilities for heavy oils are limited, inefficient and often environmentally unfavorable. Pyrolysis, also known as thermal cracking, is a decrease of the molecular weight of petroleum fragments by heating the material under pressure to about 600℃ to give lower molecular weight alkenes. Another way to carry out this job conventionally is catalytic cracking that involves the presence of zeolite (aluminum silica), again at high temperatures (about 500℃), which produces mixtures of branched-chain alkenes, cycloalkanes, and aromatic molecules [7].

New developments in electron-beam technologies for processing of heavy oils indicate a sound foundation for commercializing an operational process that has environmentally friendly features for exploiting largely untapped heavy oil resources to meet increasing world demand. Electron beam treatment of petroleum induces significant changes in molecular weight distribution, unsaturation and branching. Historically, there has been little interest in electron beam processing of petroleum due to the availability of low-cost, high-quality crude for which conventional catalytic cracking meets all requirements.

Electron beam-initiated cracking of heptane, for example, occurs at lower temperatures (450℃) than thermal cracking and provides greater yield of alkenes and methane [7]. The yield for alkene is indicative of a chain reaction wherein both the chain length and the yield are higher at lower dose rates. Electron beam-induced cracking of natural petroleum occurs in a similar fashion with heptane. Even though the process can be achieved at low temperatures, the yield and composition of the products are contingent on temperature, pressure, dose rate and total dose. The relatively high yields of unsaturated products under suitable conditions make the electron beam-induced reaction a possible source of feedstock for the petrochemical industry. [7]

A patent has been granted describing methods to decrease the molecular weight of heavy oil by exposing it to electron beam in the presence of oxygen or air, which is claimed to result in the main chain scission of the hydrocarbon molecules [45]. Under irradiation, hydrocarbons form transient species that when reacted with air or other radical species result in a decrease in the molecular weight of petroleum [45]. In order to take full advantage of the radiation-induced reaction, the surface area-to-volume ratio of the treated petroleum play a vital role; e.g. larger surface area and more exposure to air (oxygen) result in more reactive species [45]. Rapidly increasing dependence on high viscosity crude to meet expanding global demand, together with recent advances in applied electron beam chemistry of petroleum and electron beam accelerator technology, provide new cost-effective and environmentally-friendly options for refining bitumen, oil (tar) sands, and heavy crude.

Economics of Electron Beam Driven Chemistry

The wide-ranging commercial application and high-throughput potential of modern electron beam facilities demonstrates the potential for electron beam accelerators to add substantial economic value to industrial products [46]. It is well known that irradiation treatment is a capital-intensive technology that requires a considerable initial investment. This includes cost and regulation of radiation sources, shielding design and costs, conveyors, construction costs, land prices, wages, and other variables specific to each situation. Electron beam accelerators do not require a radiation source (such as gamma irradiators); therefore, the main cost in operation comes from electricity.

The accelerator's beam power and efficiency dictate the electricity consumption to complete a treatment. Actual costs associated with irradiation treatment are contingent on the required total dose to the product. The total dose needed, the throughput rate and the actual absorbed dose determine the beam power requirements. The percentage of absorbed dose by the product versus absorbed by the product, conveyer and "empty space" in between is the net utilization efficiency. Electron beam accelerators have much higher efficiency (almost double) in comparison to gamma irradiators since their beam can be directly focused versus streaming in all directions, as is the case with Cobalt-60 sources. System efficiency for DC accelerators is anywhere from 60 to 80% while RF linear accelerator system efficiency is 20 to 30% [47]. A detailed economic analysis of electron accelerators is described by Rosanna Morrison [48].

While nuclear reactors fall under federal jurisdiction, gamma irradiators and electron beam accelerators are only regulated at a state level and these regulations can differ from state to state. Some states may only require accelerator registration, which is comparable to x-ray registration utilized by medical doctors, while other states are more rigorous and demand a license registration that is typically required for gamma irradiators. The license should include the design and drawings of the electron beam accelerator facility and its safety systems description, thickness, type and density of the surrounding shielding materials, entrances and exits identification, interlocks description and finally, accelerator operation and training of authorized personnel. In most cases, states have agreements with the U.S. Nuclear Regulatory Commission (NRC) and the involvement of the Department of Labor's (DOL) Occupational Safety and Health Administration (OSHA) and the Environmental Protection Agency (EPA) to run particle accelerators, regulate radioactive material if produced from particle accelerators, ensure the safety of the workers, and regulate the impact on the environment, respectively [49]. Overall, the process and cost of administrative regulations for electron beam accelerators is fairly hassle-free and inexpensive.

<u>Economics Examples</u>

Commercial electron beam systems for water treatment applications are yet to be competitive with most existing technologies due to the cost of electron beam treatment. However, select applications such as sludge and specific industrial liquid or gaseous waste possibly will benefit more from e-beam treatment in comparison to traditional technologies due to the capability of faster and more effective treatment. Electron beam technology has the ability to effect treatment via oxidation or reduction reactions, consequently, competing primarily with chemical oxidants such as chlorine, ozone, or peroxide. The price of the oxidants can vary from a few cents per cubic meter treated for chlorine to more than $1 per cubic meter treated for ozone [50]. Water treatment businesses use the practical comparison of cost to deionize water; if this treatment is higher than $1 per cubic meter to use electron beam, it is not cost competitive. Economic analysis reported in the Miami wastewater treatment plan reveals that a factor of ten reduction in the cost of electron beam is required in order to be cost-competitive for environmental applications [50]. New high-power, high-efficiency electron beam accelerators would be able to lower the cost of power consumption ($/kW) in an electron beam system, resolving this major challenge and adding electron beam treatment as a viable and more attractive technology on the market.

The medical waste industry currently spends roughly $0.18 per pound of hospital waste by autoclaving, which is a cheaper treatment method than incineration. Electron beam treatment of medical waste would cost approximately $0.0026 per pound of hospital waste, which includes the electricity and capital cost [50]. Undoubtedly, this is a much cheaper option than the currently employed technology.

Novel Accelerator Work at Fermi

Since the field of High Energy Physics demands accelerators with ever increasing capabilities, much effort has been put into developing high energy, high power and energy efficient accelerators. Because of breakthroughs achieved in multiple accelerator technologies, the Illinois Accelerator Research Center (IARC) at Fermilab is developing a novel compact electron beam accelerator for industrial use that is high power, high energy, energy efficient and highly reliable. Such a system is sufficiently different than accelerators currently available from industry, which are largely unchanged for over 30 years. Some advantages of this novel accelerator compared to conventional accelerators include:

- High Efficiency

- Use of niobium cavities versus copper means almost all of the input power is transferred directly to the beam, rather than wasted as heat. We are currently targeting 80% wall plug power efficiency, which is much more efficient than traditional accelerators.

➢ High Throughput

- High average power is possible due to continuous wave operation. The current compact accelerator design is for 250 kW. Multiple 250 kW units can be used simultaneously, and a 1 MW version is being designed. This will allow for the treatment of on the order of hundreds of thousands or millions of gallons of chemicals per day.

➢ Mobility

- Smaller footprint allows for portability.

One system could process roughly 500,000 gallons per day of product (assuming a 10 kGy dose), be small enough to fit on a flatbed truck (mobile), operate at greater than 80% wall plug power efficiency and on average operate continuously for more than 8 years before having to replace any components. This makes the electron beam accelerator an excellent tool that can be used in application areas such as materials modification, cargo scanning, advanced manufacturing, sterilization of medical devices, and food and water treatment. One area of particular interest is the ability to drive industrial chemistries. The novel design coupled with the portable nature of the accelerator will allow for increased application in industrial chemistry.

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