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Irradiation of Sewage Sludge

Roberto M. Uribe and Jean Engohang-Ndong

Abstract

A review of the current status of sewage sludge decontamination using electron beam irradiation at industrial scale is presented. The chapter includes a historical development of the technology using both gamma and electron beam sources, a description of a facility using an electron accelerator, a discussion of the quality control techniques used to certify that satisfactory decontamination levels for safe use of treated sludge have been achieved, the effect of electron beam irradiation on the bacteria and virus present in a typical sample of municipal sewage sludge, and an analysis of the costs of decontaminating sewage sludge using electron beam irradiation compared to traditional and more routine technologies. Finally, the chapter concludes by emphasizing on the fact that electron accelerators described in this chapter are capable to decontaminate a typical municipal sewage sludge at competitive costs which are shown to be comparable and/or lower than routinely used technologies to achieve class A biosolids by the Environmental Protection Agency standards.

Keywords: sludge decontamination, e-beam applications, microbial pathogens

1. Introduction

The use of radiation to reduce the microbial contamination in foodstuffs and medical supplies is a well-established technology. Both gamma rays and high energy electrons are used with this purpose and their main effect is to damage the DNA of harmful microorganisms. Although the technology has also been used in environmental applications like the removal of contaminants from fume stacks and the decontamination of wastewater and sewage sludge, their application at a commercial stage has not been as successful as in the case of medical supplies or food irradiation.

This chapter presents first an historical overview of the area of sewage sludge irradiation and the technologies developed over time. Then a description of the technologies used for the electron beam treatment of sewage sludge will be presented. A typical facility that has been developed to irradiate sewage sludge with electrons will be described as well as a current state of the use of the technology in several places around the world.

The techniques used to assess the quality control of the process will be described including dosimetric techniques and the analysis of the effect of the electron beam on the reduction of microorganisms that contaminate sewage sludge.

2. Historical development of the technology

Sludges from municipal sewage systems are good soil fertilizer because of their high content of organic matter, nitrogen, phosphorus, and many trace metals

essential to plants pointing to its potential in agricultural applications, however they also contain a large density of pathogenic microorganisms, parasites and parasitic eggs that cause diseases to human beings, pets, and farm animals. To make the use of sewage sludge as a safe valuable soil fertilizer in agricultural applications it needs to be disinfected. To that end, several approaches have been used over the years including old techniques such as incineration and more modern techniques such as irradiation.

The use of irradiation to disinfect sludge started in 1973 when an industrial gamma ray facility from Geiselbullach near Munich (Germany) used Co-60 and Cs-137 sources [1]. The facility used 90,000 Ci of Co-60 and 570,000 Ci of Cs-137 and treated up to 180 m³/day of sludge. Similar activities were undertaken in the United States by the US Department of Energy Sandia National Laboratories using Cs-137 and capable of operating up to a maximum capacity of 7250 kg/day [2].

The use of electron accelerators to disinfect sludge was also started in the 1970's with the work by Trump and collaborators [3] in Cambridge Massachussets, USA. The system which was installed at the Deer Island Wastewater Treatment Plant in Boston Massachussets originally in 1976 consisted of a 50 kW High Voltage Engineering (HVE) electron accelerator and originally delivered up to 375,000 l/day (100,000 gpd) of sludge irradiated at a dose of 4 kGy. In 1980 the system was completely restructured to increase the capacity of the plant to 637,500 l/day (170,000 gpd). In 1982 a similar system was installed in Miami, Florida with a 1.5 MeV, 50 mA accelerator used to treat sludge at a throughput of 645 m³/day [4]. Aqueous streams were presented to the beam in a falling stream about 114 cm wide and 0.4 cm thick. The system was capable to irradiate water streams to doses up to 8 kGy by changing the beam current from 0 to 50 mA [5]. A similar system was installed in Brazil where a 1.5 MeV, 37.5 kW accelerator, with a maximum throughput of 45 l/min were described [4]. Chmielewski and collaborators [6] have described the activities developed in Poland in this area including feasibility studies on the technique and then the design of a 70 tons/day treatment plant for doses of 5–6 kGy using a 300 kW, 10 MeV electron accelerator.

Similar studies have been conducted in Japan since the 1970's by the Japan Atomic Energy Research Institute (JAERI). In this respect Washino [7] has compared electron and gamma ray treatment of wastewater in order to determine their bactericidal effect and found a larger reduction in the concentration of microorganisms for the latter one, and mention that this is due to an oxygen destruction effect produced by the higher dose rate generally used in the case of the electron irradiation. To overcome this problem, he proposed a reaction chamber for the irradiation with electrons consisting of a concentric dual-tube bubbling column with a 50 micron thick stainless steel window at the top to allow for the entrance of electrons. Oxygen is bubbling from the bottom of the reaction chamber to compensate for this reduction effect. Similarly, Hashimoto and co-workers [8] have described the use of process-control techniques to make the electron irradiation of waste waters more effective.

In 2014 through a collaboration between Arlington County in Virginia, USA and Kent State University, in Kent Ohio, USA a sample of sludge was irradiated to demonstrate the feasibility and the economic value of the process. The sample consisting of 33,750 l (9,000 gal) of sludge was irradiated at 6.7 and 25.7 kGy under typical production conditions using 3 MeV electrons provided by a Dynamitron accelerator and a flow rate of 184 l/min (50 gallons/min) and showed that the process is effective in reducing the concentration of some microorganisms and more economical than conventional disinfection techniques [9].



Figure 1.
 Example of a delivery system to irradiate sludge with high energy electrons. Photo (a) shows the entire system including the scanner of the accelerator (1), the air blower (2) to refrigerate the titanium window (3), the sludge delivery and collection tank (4), and the piping system to introduce the sludge in the tank from the bottom (5). The photo in (b) shows the scanner of the electron accelerator (1), the accelerator shutter (2), the weir with the irradiated sludge, simulated with water (3), and the incoming sludge (4). Photo in (c) shows an actual “curtain” of sludge flowing through the system.

3. Description of a typical electron beam treatment facility

Usually, sludge consists of wastewater with a low concentration of solid particles (around 15%). This is a fluid that can readily move from the storage area to the irradiation zone and back to the storage area and in a typical sludge irradiation facility this is done using a hydraulic system consisting of pipes and pneumatic pumps. Inside the electron beam room, the sludge is irradiated using a bulk irradiation system that will present a laminar like fluid to the electron beam with a thickness of a few millimeters depending on the energy of the electrons. An example of this could be a rectangular stainless-steel tank divided in the middle by a wall. At the top of this wall, a weir is built to produce a cascade of sludge. This weir is aligned with the length of the scanner of the accelerator in such manner that the sludge is uniformly irradiated by the system. The influent (pre-treated) sludge flows from the bottom of the first section of the tank, rising up to the height of the middle wall over the weir and then overflowing into the second section (**Figure 1** shows an example of this system, as installed for the Arlington County experiment described above). The effluent (post-treated) sludge is taken out from the bottom of the second section flowing through a second set of rubber pipes to a second storage tank.

4. Quality control of the process

The effect of radiation on the reduction of bacterial load and decontamination of sludge depends on the amount of energy from the radiation that is absorbed by the sludge. This energy is used to produce chemically active species that eventually disrupt structural integrity of DNA in bacteria, parasites, and viruses causing their partial or permanent inactivation and eventually death of microorganisms. The amount of energy from the radiation that is absorbed by the sludge is measured by the physical term dose which is defined as the amount of energy absorbed in a volume of sludge divided by the mass of that volume and is measured in kilogray (kGy). A dose of several kGy could be enough to cause a disruption of DNA molecules and inactivate a virus or kill a bacterium. In order to apply the technique in a municipal installation one needs to be sure that all the sludge that gets irradiated will receive the minimum amount of dose needed to inactivate the microorganisms. Several techniques have been proposed to determine the real dose received by the sample during the irradiation of

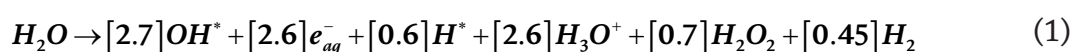
sludge. One of them makes use of the fact that the interaction of the radiation with the sludge will cause a certain amount of temperature increase in the sludge, therefore, the dose can be determined by measuring the difference in temperature in it before and after irradiation using thermocouples located in the influent and effluent parts of the water piping system near the falling stream; this set of thermocouples can also be used to monitor the irradiation treatment. An advantage of this technique is that once the thermocouples are installed the temperature difference between the input and output portions of the system is easy to be determined; the only two parameters needed to know are the specific capacity of the sample which needs to be determined experimentally for each type of sludge treated and the temperature difference. With these two values the energy absorbed in a sample of sludge can readily be obtained. One problem with this technique however is that it only provides an average value of the dose and does not measure the dose that individual samples of sludge will get when going through the system neither does it measure how uniform the dose delivered to the sludge is really. To overcome the first problem, a small dosimeter can typically be placed inside a small water tight plastic capsule and be allowed to run through the system from the supply tank to the collection tank. Several candidates of dosimeters can be used in this respect; one of them consists of alanine pellets which have dimensions of several mm. These dosimeters rely on the fact that under irradiation a concentration of relatively stable free radicals are produced in them; the free concentration of free radicals can be determined by the technique of electron paramagnetic resonance [9, 10]. Another dosimeter that can be used is Lithium fluoride in crystals or in powder; the crystals have dimensions of a few millimeters per side and two or three could be accommodated in a single capsule. When using it in powder a few mg can be placed in a capsule for irradiation. Upon irradiation electrons from the valence band of the crystal jump to the conduction band and when trying to be de-excited, some of these electrons get trapped in the energy gap of the material. The electrons can be liberated from these traps and allowed to return to the valence band by applying heat to the material. When the electrons return to the valence band, they emit light to lose the excess of energy they have in them. The technique to heat the crystals in a controlled way and measure the output light in the de-excitation process is called thermoluminescence and when applied to dosimetry it is called thermoluminescence dosimetry (TLD) [11]. Uribe and co-workers described the use of this technology in a similar application in the irradiation of corn kernels in a pilot plant using 1.0 MeV electrons [12].

To address the second issue and as part of the operation qualification (OQ) activities of the system [13] a verification of the dose uniformity along the weird where the electrons irradiate the sludge needs to be performed. This is useful to remove “dark” spots where the sludge does not get irradiated or to correct for instabilities in the electron beam scanning system that makes the electrons to stay longer times or to correct for misalignment of the sludge delivery system with the scanned electrons coming out of the electron accelerator. Film systems are the best candidates to perform dose uniformity measurements along the scanner of the electron accelerator. Several examples of these are available in the market; good examples of them are the cellulose triacetate film (CTA) [14] and the radio chromic films [15]. Both of these work on the principle that radiation induces a change in the optical absorption of the film that can be quantified using a spectrophotometer. Through a suitable calibration with a primary dosimeter these systems can be used to measure the dose along the length of the weird of the sludge delivery system. For instance, when performing the OQ activities using CTA film it was found that the length of the weird where the sludge was irradiated was longer than the extent of the scanned electrons. So, there were “dark” spots at both ends of the weird where the sludge was not irradiated. The situation was corrected by placing a couple of plates at both ends of the weird that reduced its length so all the sludge going through the weird was irradiated [9].

5. Bactericidal effect

Until a recent past, the radiation effects on the bacterial load and removal of noxious chemical compounds had only been performed in small samples of sludge irradiated under laboratory conditions and mainly address either only the microbiological or the chemical effect of radiation in a sample of sludge [16–20]. Processing and disposal of wastewater sludge is a critical problem worldwide [18] and especially in large metropolitan such as Washington DC in the USA and Tokyo in Japan. Therefore, new technologies to solve the problem of safely disposing of sewage sludge are constantly being sought. An attractive solution for the disposal of wastewater sludge is its utilization in crop fields and landscaping as a fertilizer due to its high content in natural nitrogen compounds. However, in order to be used as such, it must be converted into a class A biosolid [21] which is a form of treated sludge that is deemed safe for humans and animals. In recent years, electron beam technology have shown to be an economically alternative that could be used to meet these regulations by considerably reducing the number of potentially harmful bacteria such as the fecal coliform bacteria and helminth ova such as *Ascaris ova* which are both ubiquitous contaminants present in the sewage sludge. The fecal coliform bacteria are a group of bacteria that are released in the environment through the fecal excrement of humans, wildlife, and livestock. They typically occur in the digestive tract of humans and warm-blooded animals. When present in sewage sludge, they are indicative of the presence of human and animal pathogens including some strains of *Escherichia coli*, *Shigella flexneri*, and *Salmonella typhimurium*. Among the fecal coliforms, the *E. coli* O157:H7 strain which was responsible for many outbreaks has retain attention of media in the past several years. Indeed, *E. coli* O157:H7 was found contaminating drinking water and vegetables, causing cases of stomach cramping, vomiting, and bloody diarrhea in patients. However, in most cases infections with the *E. coli* O157:H7 strain were mild or with no symptoms. Most people infected with *S. typhimurium* develop diarrhea, stomach cramping, and fever which could cause the patient to experience fatigues and dehydration. An infection with *S. flexneri* is much more serious with severe stomach pain, dehydration, severe diarrhea, and fever. Patients suffering for shigellosis feel very sick and stay in bed at the pick of the infection. In farming areas where livestock is grown, beside occurrence of fecal coliforms, ascaris ova that when ingested by a person or an animal would hatch and develop into adult worms, *Ascaris lumbricoides* cause stomach pain, nausea, vomiting, and fatigue. In patients who are heavily infested with ascaris worms, the parasites could be expelled through feces and/or vomit.

Additionally, the electron beam technology could be used to reduce the concentration of volatile organic sulfides and other volatile organic compounds (VOCs) responsible for the unpleasant odor in sewage sludge. Moreover, electron beam treatment of sludge is not an energy intensive process which means that it has a small footprint and the processing times are usually short. The main effect of radiation on sewage sludge is in the radiolysis of water producing OH^* and H^* radicals and hydrated electrons, highly reactive chemical species which rapidly react with organic compounds in the sludge. The main intermediates in this process are:



The effect of electron beam irradiation on the microbial reduction in sludge is accomplished by a two-fold effect of the irradiation on the sludge [22]. The first one is the direct effect of radiation on the microorganisms disrupting the structural integrity of DNA molecule and eventually killing them. The other one is the indirect

effect caused by the radiolysis of water described above. The chemical active species which are produced in water because of the irradiation will cause oxidative damage on nucleic acids, proteins, and lipids in microorganisms leading to their death. Typically, at a given electron beam radiation dose, the killing of microorganisms occurs at a constant rate over time. Similarly, the effectiveness of the killing of the microbial populations including that of bacteria increases with the radiation dose. The effectiveness of electron beam radiation on the killing of microorganisms is better represented by the so called D_{10} value which is the dose of radiation required to kill 90% of the population of microorganisms present in the sludge sample. Because of the difference in structural complexity of microorganisms, D_{10} values may vary more or less significantly from one microorganism to another. Thus, for *Ascaris ova*, that D_{10} value was determined to be in the order of 0.39 kGy [23]. For other microbial contaminants of sludge including *S. typhimurium* and *E. coli*, these D_{10} values were determined to be 0.3 and 0.34 kGy respectively [4]. Recent studies performed by Engohang-Ndong and his research collaborators have shown that the electron beam technology could be used at industrial scale to eliminate potential microbial pathogens from sewage sludge [9]. The US-based research team showed that a dose of 25.7 kGy was enough to eliminate *Ascaris ova* to a level that is not detectable to available technique used to count the helminth eggs in sewage sludge including the use of Sedgwick Rafter cells to count the detectable ascaris ova. Thus, at that electron beam radiation dose, it was possible to achieve a class A sludge. According to the US Environmental Protection Agency (EPA) standards, to be considered class A biosolids, the sewage sludge must contain less than one *Ascaris ovum* per four gram of sludge dry weight [21]. The dose needed to eliminate fecal coliforms to the norm set by the US EPA to convert sewage sludge to class A biosolids is much lower. The experimental dose determined by Engohang-Ndong and collaborators is 6.7 kGy. In other words, when risks of contamination of sewage sludge by helminth eggs is reduced such as in heavily urbanized areas, the doses needed to convert sewage sludge to class A biosolids that in turn could be used to enrich the soil for landscaping and agricultural purposes is very beneficial and requires low consumption of energy.

6. Economic aspects

An important aspect in the implementation of a new technology such as an electron accelerator in a wastewater treatment plant is to anticipate its impact on the operation costs of the facility and on the environment. With respect to the irradiation of sewage sludge several authors have addressed this issue. A team in Florida reported a cost of \$2.50 per 1000 gallons of sludge for a 1.5 MeV electron irradiation facility running at 160 gallons per minute [24], while another group compared gamma and electron beam irradiations for a sample of activated sludge and obtained treatment costs of \$4.20/m³ for gamma irradiation and \$2.10/m³ for electron beam irradiation, which are lower compared to \$4.85–\$5.19 when using conventional technology at the Central District Wastewater Treatment Facility in Miami Dade County [25]. Furthermore, a comparison was made between irradiation at a dose 6 kGy and incineration of sludge samples and showed a cost of \$60.87/m³ for this latter compared to \$3.12/m³ when using gamma radiation. Similar results have been obtained by Engohang-Ndong et al. using a 3 MeV electron accelerator. In this case these authors reported that the cost to irradiate one cubic meter of sludge to a dose of 25.7 kGy will be \$1.26 [9]. That comparative cost analysis tends to show that electron beam irradiation of sludge consumes less energy than other technologies. Furthermore, electron beam irradiation requires processing times,

and very importantly more environmentally friendly technology compared to other technologies such as gamma radiation and incineration.

7. Conclusions

While the use of irradiation to control microbial populations in foodstuffs, on medical devices, and on the environment is relatively old, the use of electron beam irradiation to eliminate potential human and animal pathogen from sewage sludge is relatively new. At the present times electron beam irradiation is a mature technology and is capable to provide the required power to irradiate sludge at the flow rates used in a sewage sludge decontamination municipal plant. The quality control techniques are readily available to provide measurements of the dose needed to decontaminate the sludge and the technology has proved its capability to eliminate bacterial populations and ascaris ova to levels that are considered safe for humans and animals in landscape, agricultural, and landfill applications. Analysis of the costs involved in using electron beam technology to decontaminate sewage sludge showed that they are competitive with usual costs involving mechanical and chemical means to treat the sewage sludge. Application of this technology opens new possibilities for major agglomerations worldwide to safely repurpose municipal sewage sludges.

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