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ScienceDirect
Journal of Hazardous Materials 143 (2007) 2–7
Journal of Hazardous Materials
www.elsevier.com/locate/jhazmat

Review

Application of radiation technology to sewage sludge processing: A review

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Received 15 July 2006; received in revised form 5 January 2007; accepted 5 January 2007
Available online 12 January 2007

Abstract

Sewage sludge is unwanted residual solid wastes generated in wastewater treatment and its management is one of the most critical environmental issues of today. The treatment and disposal of sludge contribute a considerable proportion of the cost for running a wastewater treatment plant. The increasing amount of swage sludge and more and more legislative regulation of its disposal have stimulated the need for developing new technologies to process sewage sludge efficiently and economically. One ideal consideration is to recycle it after proper treatment. Radiation technology is regarded to be a promising alternative for its high efficiency in pathogen inactivation, organic pollutants oxidation, odor nuisance elimination and some other characteristics enhancement, which will facilitate the down-stream process of sludge treatment and disposal. Here we present a brief review of application of radiation technology on sewage sludge processing. Some basic information of two currently available irradiation systems and fundamental radiation chemistry are introduced firstly; then the world-wide application of this promising technology is reviewed; various effects of radiation on sludge is discussed in detail; and some concluding remarks are given and some future directions are also proposed.

Keywords: Radiation technology; Sewage sludge; Disinfection

Contents

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0304-3894/$ – see front matter
doi:10.1016/j.jhazmat.2007.01.027

1. Introduction

The treatment and disposal of sludge is one of the most critical environmental issues of today. The need for water supply is increasing along with the improvement of life quality. Wastewater is the product resulting from fresh water after being used for life-supporting for human or the manufacturing processes [1]. Sewage sludge is an inevitable by-product of wastewater treatment process. The most common steps of wastewater treatment directed towards removal of pollutants with least effort involve screening, primary sedimentation, biological treatment, secondary sedimentation and disinfection [2]. Sedimentation both before and after biological treatment generate sewage sludge [3]. Production of sewage sludge has reached an alarming dimension with the growth of urban population, the improvement of human life quality and the development of industry and agriculture [2]. It is estimated that approximately 26 kg of sludge is generated per person per year, on a dry weight basis [4]. About 4000 wet tonnes sewage sludge is produced each day in Korea [5]. And in the United States, more than 2 billion US dollars is spent annually treating and managing approximately 5–7 million dry metric tonnes of biosolids from over 13,000 publicly owned treatment works [6]. In China, sewage sludge produced can be estimated to be 3.5 million tonnes (dry weight) in 2005.

Proper treatment and disposal of sewage sludge avoiding harm to environment is an integral part of wastewater treatment and environmental protection. Municipal sludge has high water content, high load of organic pollutants and high content of pathogens as well as considerable concentration of heavy metals. Generalized processes for sewage sludge handling are gravity thickening, chemical conditioning, anaerobic digestion, dewatering and disinfection, followed by final disposal, including incineration, composting, land application, landfill, ocean dumping, etc.

Sewage sludge handling contributes a considerable portion of the total cost for running a wastewater treatment plant. The treatment and disposal of sewage sludge have seriously challenged the development of wastewater treatment industry and environmental protection. The increasing amount of sewage sludge generated and relatively high cost for disposal as well as the more and more stringent legislative restrictions governing the disposal practice have warranted the needs for developing some cost-effective solutions that can significantly address the current sludge handling goals, the best of which is to consider its beneficially recycling after nuisance reduction and pathogen elimination.

One of the most promising alterative is the radiation processing. Radiation treatment of sewage sludge has several advantages, for example:

(1) It can reduce pathogens efficiently to a safe level [2].
(2) It can oxidize toxic and hazardous organic pollutants such as pesticides, herbicides, PCBs, etc., and convert non-biodegradable substances into more readily degradable ones [4].
(3) Odor nuisance elimination is also desirable during irradiation.
(4) It can also enhance the dewatering property and biodegradability.
(5) It is a relatively simple, efficient and reliable way of sludge processing.

Early studies on applying radiation technology to waste treatment were started in 1950s [4]. And numerous publications have appeared regarding the basic principle research as well as some cases study in the past few decades. However, this promising technology has attracted little interest until recent years and has not yet been widely accepted mainly due to the conservative attitude of government for safety concern resulting from knowing little about this technology. In order to achieve a better understanding of the prospective application of ionizing radiation technology in the processing of municipal sewage sludge, here a brief review of this promising technology is presented and the possibility of its application is demonstrated. The development and the basic principle of radiation technology processing sewage sludge are briefly introduced. World-wide application of this technology and some results reported from both laboratory and full-scale studies are reviewed. The effects of radiation processing are discussed in detail. Finally, some concluding remarks are given and future directions are also proposed.

2. Radiation technology employed in sewage sludge processing

2.1. Two radiation systems

Two options are currently available that can be used for the radiation processing of sewage sludge. One is gamma sources, and the other is electron accelerators. These two are considered equivalent for a given radiation dose.

Either Cs-137 or Co-60 can be used efficiently to generate gamma rays for the application on sludge processing. The most common used industrial gamma source is Co-60, which is produced by exposing non-radioactive Co-59 to neutron flux in a nuclear reactor [7]. Co-60 is generally more efficient which emits two gamma rays with the energy of 1.17 and 1.33 MeV, respectively [6,8]. Co-60 has a half-life of 5.26 years. So after prolonged use of these sources for 20 years, only 6% activity of its original is left. And then the residual is generally returned to the irradiator suppliers for reuse, recycling or disposal [8]. The steady decay of Co-60 is one of its main disadvantages, making the eventual replacement of radioactive source is necessary

Another option of radiation source is Cs-137 in the form of salt CsCl. Its radiation energy is 0.66 MeV with a half-life of 30 years. CsCl is water-soluble, which restrains its application because it may pose serious problem in case of leak or other accident [7]. So Co-60 is almost solely used as the gamma radiation source for industrial use currently [8].

Gamma rays penetrate well in water and sludge, which ensures the radiation effect in thick layer of sludge. The half-value thickness of Co-60 gamma rays (1.3 MeV) is about 28 cm in water and no less than 25 cm in normal liquid sludge, and the half-value thickness of Cs-137 is 24 cm in water [7].

As far as high-energy electron beam is concerned, it is generated by electron beam accelerator which accelerates charged particles in a single direction through electric and magnetic field. Typically, the electrons emitted from cathode are accelerated in a vacuum system. Then they pass through a thin metal window before hitting the targets [4]. The type of machine is generally classified according to the energy range of electron beams. And the energy of accelerated electrons can achieve 3 MeV with conventional and relatively low-cost machines [7]. High-energy electrons are often produced by linear accelerators which are much more expensive and complicated.

The penetrating power of electron is approximately 3 mm/MeV in water. Thus, thin cross-section of sludge passed through electron beam is required to ensure the contact of accelerated electron with its targets. And obtaining a homogenous condition of sewage sludge as well as the dose distribution plays an important role when applying accelerated electron beam to process sewage sludge.

One of the main advantages of accelerated electron beam over gamma radiation is its absence of radioactive source, which significantly simplify some safety-concerning issues [6]. Another obvious advantage of employing electron beam accelerator is that it can be turned on and off instantaneously thus facilitating control compared with gamma radiation, especially in case of emergency or maintenance. However, it has been limited to some application in the treatment of some exhaust gas due to its rapid energy loss with the penetration depth in aqueous media as just mentioned above.

2.2. Radiation chemistry

Two basic concepts are fundamentally important in aqueous radiation chemistry. One is absorbed dose, which describes the amount of energy deposited in the material exposed to an ionizing radiation field. Formally, the absorbed dose can be defined as the amount of energy absorbed divided by the mass of material irradiated. SI unit used to describe the absorbed dose is Gy. And 1 Gy is equivalent to 1 J/kg.

The other important concept in radiation chemistry is *G*-value, which measures the radiochemical yield by the number of specified chemicals species in an irradiated substance produced per 100 eV of energy absorbed from ionizing radiation.

In water, wastewater as well as sludge matrix, the principal component is water. Therefore, it would be expected that the effect of ionizing radiation may be dominated by the interaction of radiation and water [6]. As far as pure water is concerned, when exposed to ionizing radiation, the radiolysis of water can be presented as following equation [9]:

$$H_2O \rightarrow [2.7]OH^* + [2.6]e_{aq}{}^- + [0.6]H^* + [2.6]H_3O^+ + [0.7]H_2O_2 + [0.45]H_2$$

So if we consider pure water, each 100 eV absorbed by water will result in the generation of 2.7 radical OH^*, $2.6e_{aq}{}^-$, 0.6 radical H^*, $2.6H_3O^+$, 0.7 molecule of H_2O_2 and 0.45 molecule of H_2 [4].

In aqueous media, the oxidizing hydroxyl radical OH^*, the reducing hydrated electron $e_{aq}{}^-$ and the hydrogen radical H^* are the predominant products, all of which are highly reactive transient species and are responsible for the various effects including the reduction of pathogens, the oxidation of hazardous organic pollutants, the destruction of molecular structures of targeted pollutants, the elimination of odor nuisance as well as various characteristics changing when the ionizing radiation is employed in the processing of sewage sludge.

3. History, development and world-wide application of ionizing radiation in sewage sludge processing

Ionizing radiation is any of several types of particles and rays given off by radioactive material, nuclear reactions, and radiation producing machines. Its energy is high enough to break molecular bonds and ionize atoms, but not high enough to affect the structure of atomic nucleus, which avoids the induction of radioactivity to the irradiated products. Radiation with such high energy is referred to ionizing radiation, which includes gamma radiation, high energy electrons and X-rays generated from high energy electrons [8].

Ionizing radiation has been present ever since the earth was created. But before 1890s when magical presence of X-rays was discovered by Wilhelm Roentgen, there were only natural sources of radiation from cosmic sources and radioactive material.

3.1. Gamma radiation

As far as environmental application of ionizing radiation is concerned, Lowe et al. [20] carried out one of the earliest studies applying radiation technology to wastewater purification using Co-60 gamma irradiator to disinfect actual effluent. After the irradiation sources became available in 1960s, the environmental application of ionizing radiation in the treatment of water, wastewater and sewage sludge was intensively studied [4].

The first irradiator used for the disinfection of sewage sludge was installed in Geiselbullach Treatment Plant [7], 10 km east of Munich, Germany. The irradiation shaft with a built-in central tube was built underground. The radiation sources had a cylindrical form (diameter 30 mm; length 300 mm). Each contained 30 plaquettes of Cobalt-60, which were doubly encapsulated with an initial activity of about 18,000 Ci. Additional sources were installed every 2 or 3 years [7].

The irradiator was operated in a batch mode, treating approximately 5.6 m^3 of sludge with an absorbed dose of 3 kGy. For irradiation, the sludge containing 4% solids flowed from the silo into the irradiation shaft, where it also entered the connection pipes. To provide a homogenous dose distribution, the sludge was mixed with a recirculation pump. After a preset time, which depended on the installed radioactivity and the required dose, the sludge was pumped out by evacuation. The batch-wise operation ran automatically for 24 h a day. All pumps, valves, indicators and control equipment were located either in the permanently accessible pump shaft or in the building above ground, so that services and inspections were possible at any time, without need for special safety precautions [7]. Unfortunately, the operation of the plant was stopped in the springtime of 1993 for major repairs, and at that time new regulations in Germany disallowed further use of sewage sludge on grassland and areas for fodder production. As only those areas required the disinfection of sewage sludge, the treatment was no longer necessary and the decision was made to stop operation of the irradiation plant [7]. But the 20 years operation experiences are very important and precious for the researchers interested in this field all over the world.

A pilot and demonstration plant for the treatment of dewatered and compost sludge, also mixed with other solid wastes, was built and commissioned in 1979, at Sandia National Laboratories in Albuquerque, NM, USA [7]. The radiation source employed was Cs-137 with maximum activity of 1,000,000 Ci. The designed capacity was 8 tonnes of dried sludge (about 50% dry solid) per day with a required dose of 10 kGy. But the operation of the plant was stopped due to some unknown reasons.

A facility using gamma source for demonstrating appropriate radiation technology in treating the entire generated sludge of a conventional municipal wastewater treatment plant was built up by Isotope Division of the Bhabha Atomic Research Centre in collaboration with M.S. University of Baroda, Gujarat Water Supply & Sewerage Board and Municipal Corporation of Baroda, India [2]. The employed gamma source was Co-60 with the maximum activity of 500,000 Ci and the maximum treatment capacity designed was 110 m^3/day [2]. The municipal wastewater processing plant contains a grit removal chamber followed by primary sedimentation tank, trickling filter, secondary sedimentation tank and an anaerobic digester. The irradiation plant is located at the end of the process channel. The irradiated sludge is released in a drying bed for sun drying [10]. The sludge from primary sedimentation tank was collected in a cemented tank and then the irradiation process was carried. Since it's commissioning in 1992, the facility is in daily operation and working smoothly [10,2]. The results showed that a dose of 2 kGy is adequate for disinfection of raw sewage sludge.

3.2. Electron beam

Electron beam has not been so widely used in sewage sludge treatment. But there were still some laboratory studies as well as some demonstration projects. However, most of them were only in operation for several years before they were decommissioned.

Two electron accelerators were once employed for the processing of municipal sewage sludge in USA, one was in Boston and the other in Miami [7]. The former was built for the treatment of sewage sludge as a pilot and demonstration project in 1976 at Deer Land Wastewater Treatment Plant in Boston. The design and supervision were completed by Massachusetts Institute of Technology. The capacity was reported to be 400 m^3/day. But the investigation was conducted in laboratory and the project was decommissioned without scale-up research in 1984. The latter was built during 1981–1983 at wastewater treatment plant in Virginia Key, Miami, FL. The employed accelerator had a power of 75 kW (1.5 MeV, 50 mA), allowing daily capacity of 645 m^3 at a dose of 4 kGy [7].

There were a few reports regarding the application of accelerated electron beam on the processing of sewage sludge from Germany, Australia and Japan [7]. But as just mentioned, most of them were operated only for 2–4 years. Although considerable reports concerning the treatment of wastewater were appeared and some industrial scale wastewater treatment plants were constructed all over the world using accelerated electron beam, no full-scale plant employing accelerators for sewage sludge processing has been arisen.

4. Various effects of irradiation on sewage sludge

4.1. The effect of radiation on pathogens

Sewage sludge generally contains high concentration of pathogens, even after digestion or other conventional stabilization treatment. Some of the most important pathogens in sewage sludge are bacteria (e.g. salmonellae), viruses (e.g. poliovirus) and parasites.

Sewage sludge is hazardous waste requiring expensive disposal procedures. On the other hand, the municipal sewage sludge is a rich source of plant nutrient and has potential use in agriculture as a soil conditioning agent or fertilizer. Agricultural use of sewage sludge after careful elimination of pathogen and the control of organic contaminants and heavy metals to a safe level is the favored end outlet in some developing countries such as Latin American countries, China and India, for it has the advantage of simultaneously reducing the cost for final disposal when compared with some conventional end outlet such as incineration and thermal solidification and economically benefiting both the municipal administration and the farmers.

The high concentration of pathogens presents a major obstacle of the practical application of sewage sludge in agriculture. So the effect of radiation on pathogens is of primary concern when applying the radiation technology on the processing of sewage sludge.

Radiation effects on microorganisms are mainly associated with the chemical changes but also depended on physical and physiological factors [4]. Dose rate, dose distribution, radiation quality, radiation type and exposure pattern are important physical parameters. Species, temperature, moisture content and oxygen concentration are some of the most important physiological factors. Both physical and physiological factors may largely modify the response of a living organism to a given dose of radiation.

The action of radiation on a living organism can be divided into direct and indirect effect. If the radiation interacts with the atoms of the DNA molecule, or some other cellular component critical to the survival of the cell, it is a direct effect, which will eventually affect the ability of the cell to reproduce and survive. The formation of major radiolysis products from water and their subsequent interactions with activated sludge particles are described as indirect effect, which is generally caused by energy deposition in the medium resulting in the formation of secondary reactants generated through free radical production, sensitizer reaction and secondary ionizations. Indirect ionization effects are rapid and typically occur within about 10^{-7} s of exposure [6]. The radiolysis products OH^*, e_{aq}^- and H^* are responsible for indirect effects caused by ionizing radiation. And indirect effect is generally considered more significant than direct effect in the application of ionizing radiation to process sewage sludge.

Both direct and indirect effect on microorganisms caused by radiation may result in the damage of genetic material, i.e. DNA and RNA, thus the cell will be killed and the aim of disinfection is achieved. But the sensitivity of microorganisms to radiation varies significantly from one species to another. Decimal reduction dose (D_{10}) is usually employed to describe the radiation sensitivity, which means the dose required for a one-log inactivation or the dose required for killing 90% of the presented population. The D_{10} for *Escherichia coli* and *Salmonella typhimurium* was reported to be 0.34 and 0.30 kGy, respectively [4].

Most literatures released suggested that an absorbed dose of 2–4 kGy was enough to inactivate the pathogens to a safe level. Researchers in India reported that a dose about 2 and 2.7 kGy was found to be adequate for disinfecting raw sewage sludge and digested sludge, respectively. Total coliforms and salmonella-shigella counts were reduced below 100 cfu/ml. After four hours radiation at an absorbed dose of 2 kGy along with aeration using Co-60 gamma radiation, average log unit reduction in total coliforms, total bacteria and total salmonella-shigella in raw sludge was 3.6, 1.9 and 3.5, respectively [2].

Capizzi et al. [11] found that *Ascaris* ova were no longer viable after exposure to radiation dose of 0.75 kGy and D_{10} value was 0.39 kGy using accelerated electrons. Although the outer coat protects against low-dose radiation (0.20 kGy), no difference in ovum viability was observed after exposure to 0.30 kGy. A similar study using accelerated electron beam to destruct *Ascaris* ova was conducted by Capizzi-Banas and Schwartzbrod [12]. The D_{10} value obtained for radiation of residual sludge contaminated with ova depended on the source of ova, 788 ± 172 Gy for suspensions of ova extracted from slaughter house sludge and 1125 ± 145 Gy for suspension freshly prepared ova by worm dissection. Results obtained also suggested that the storage and the medium in which the ova were radiated (deionized water or sludge) would affect D_{10} value.

Researchers in Poland carried out an experiment applying accelerated electrons to sewage sludge to inactivate the pathogens and eliminate the parasite eggs [13]. Results suggested that an absorbed dose of 6 kGy was capable of killing all parasite eggs and decreasing bacteria content by three logs, spore-forming bacteria and *Coliforms* count by two logs 99% and *Clostridium perfringens* count by one-log. All adult parasites were also killed as they are much more radiosensitive than their eggs.

Compared with other options that are conventionally used for sludge disinfection, such as pasteurization, aerobic–thermophilic treatment, composting, thermal conditioning, incineration, lime treatment and long-term storage, ionizing radiation is carried out at ambient temperature and it's not an energy intensive process. Besides, it has small foot-print and is capable of efficiently eliminating pathogens and parasites to a safe level within a rather short time. In addition, no chemicals are needed in the process which will consequently reduce the operational cost. And at last, this technology is compatible with the exiting wastewater treatment unit, for the sludge for irradiation can be taken directly from primary sedimentation tank, secondary sedimentation tank or digestion tank. Thus, ionizing radiation is a promising alternative for the disinfection of sewage sludge and more application may be expected in the coming decade.

4.2. The effect of radiation on organic pollutants

Sewage sludge is residual solids of biological wastewater treatment, and thus is usually characterized by high organic content and especially concentrated refractory or non-biodegradable compounds such as pesticides, herbicides, PCBs, etc.

The ionizing radiation-induced oxidation of organic pollutants has been extensively studied in radiation processing of water and wastewater in the past few years [14–18,21,22]. Ionizing radiation has been demonstrated as an effective way to oxidize organic pollutants. Although the effect of radiation on a specific pollutant in sewage sludge has not been investigated in the past, the radiation-induced degradation of organic pollutants is well acknowledged in aqueous medium. So from this perspective, the oxidation of these organic pollutants and conversion of them to more biodegradable ones may be expected.

A few researchers examined the effect of ionizing radiation on bulk parameters such as chemical oxygen demand (COD) [6]. Generally, COD exhibited a decreasing trend with radiation, which indicated the occurrence of the oxidation and degradation of some organic pollutants. Research carried out by Meeroff showed that ionizing radiation stimulated the release of COD at low dose (1–4 kGy) and exhibited mineralization at higher dose, which demonstrated a competing mechanism effect.

4.3. Other effects caused by ionizing radiation

Besides biological inactivation of pathogens and degradation of organic pollutants, other effects caused by ionizing radiation, such as the effect on suspended solids (SS), thickening capacity effect and anaerobic stabilization were also investigated [6,19]. Zheng et al. [19] studied the effect of radiation on total suspended solids (TSS) and volatile suspended solids (VSS) using electron beam. Decrease of TSS and VSS was confirmed but no distinct change of COD. However, the mechanism by which caused the changes of TSS and VSS was still unknown.

Meeroff [6] studied the effect of ionizing radiation on anaerobic digestion, conditioning and thickening property systematically. The results from the research of radiation-assisted anaerobic digestion indicated that no enhancement of biogas production occurred. But the conditioning effect caused was noticeable, which was induced by neutralizing charge. Although the radiation alone could not achieve the point of zero zeta potential without polymer addition, the amount of polymer might be reduced by 60%. But at a higher dose, the competition mechanism effect showed undesirable results. As far as the effect on thickening property is concerned, results showed that filtration was improved by 65% with radiation up to 4 kGy during polymer addition. Surface charge modification, more compaction characteristics and the enhanced thickening property may significantly benefit the down-stream processes and result in the reduction of the overall cost for running a wastewater treatment plant since the handling and disposal of sewage sludge represents such a considerable proportion that either enhanced filtration or enhanced dewatering property of sewage sludge will eventually reduce this considerable cost.

5. Concluding remarks

Ionizing radiation is very efficient in elimination of pathogens, destruction of organic pollutants, modification of conditioning and thickening property of sewage sludge. And the sludge irradiated to a safe level has potential use in agriculture within the legislative permission as a soil conditioner or fertilizer, which can economically benefit the municipal administrations and the farmers. And this may have great significance for some developing countries in the enhancement of crop yield and eventually the reduction of poverty.

Although the radiation processing of municipal sewage sludge was of many researchers' interest in the past, little progress in practical use especially full-scale application has been achieved, mainly due to the conservative attitude held by government. However, this conservative attitude largely results from knowing little about this technology. In fact, radiation processing is a regular industry and has been under operating safely for more than 40 years. The workers in the facilities live a normal life comply standard health and safety regulations. The past 40 years' successful operation experiences, the advanced in the control and monitoring technology, together with careful design, operation and maintenance under the guidelines of IAEA and other international organizations can provide enough safety guarantee.

In the past few decades, most projects employing ionizing radiation to process sewage sludge were pilot- and demonstration- or even laboratory-scale, and full-scale application were only reported in Germany and India using gamma radiation. No literature regarding the full-scale application of accelerated electron beam in sludge handling has been released.

Numerous basic researches of employing radiation technology to process municipal sewage sludge have been reported in the past few decades. However, more work still need to be done to promote this promising technology towards its wide application. One of the most urgent issues needed to be addressed is economical feasibility analysis and demonstration. More pilot and demonstration plants also play an important role in the development of its practical application. The combination of radiation technology with conventional treatment is also recommended for the future research.

Acknowledgment

The work was financially supported by the National Natural Science Foundation of China (Grant no. 50678089).

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