

Review

Advanced Technology for Energy, Plant Nutrients and Water Recovery at Wastewater Treatment Plants

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Abstract: In present times, with increasing emphasis on circular economies, municipal wastewater treatment plants (WWTPs) are considered resource recovery facilities. The targeted resources are water, biogas, and sludge, organic residuals containing nutrients and elements needed by plants (nitrogen and phosphorus). Sludge is a byproduct that constitutes the largest volume of all other byproducts obtained in wastewater treatment plants. Its processing and disposal are challenging for environmental engineers because of its complexity. Thus, quick development and implementation in industrial practice of sludge valorization and utilization technologies is required, where high nutrient content must be taken into account. Also, the occurrence of a variety of pathogens in sewage sludge is a matter of concern, even in the case of developed countries. The use of untreated sludge or wastewater in agricultural activities poses a serious risk of bacterial and parasitic infection in human beings. To overcome such issues, the application of ionizing radiation processing, especially electron beam (EB), can be considered a promising method. Its effectiveness in pathogen removal has been proven by researchers. Water radiolysis products created during irradiation of water are highly reactive and cause some effects such as DNA damage, $\bullet OH$ radical production, etc. Additionally, ionizing radiation technologies in sewage sludge treatment enhance the efficiency of the methane fermentation process. Depending on specific needs, different types of ionizing radiation sources can be discussed. Based on the review information and our research results, the basic engineering parameters of hybrid installation have been presented as the conclusion of the report. In this technical solution, a notably effective additional step would be the use of EB irradiation, combined with conventional wastewater treatment, to achieve efficient removal of pollutants.

Keywords: wastewater treatment plant; sewage sludge valorization; electron beam; ionizing radiation; water reuse; organic fertilizer; circular economy; biogas production; fertilizer



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1. Introduction

North America and Europe generate around 67 billion m³ of wastewater each year. That gives 231 m³ per capita in the case of the U.S. However, Africa's wastewater discharge gains around 95 m³ per capita annually and is significantly low compared to North America and Europe. A total of 56% of the household wastewater globally was properly treated in 2020 [1]. Significant differences in the amount of household wastewater safely treated were discovered in different regions (from 25% to 80% by SDG region). That indicates the inconsistency in global progress [2]. The most widely used biological wastewater treatment process worldwide is the activated sludge process, which is implemented in both domestic and industrial plants. The technology is based on converting dissolved and suspended organic compounds in aerobic conditions to bacteria cells and gases (CO₂, CH₄, N₂, and SO₂). Worldwide production of sewage sludge has increased dramatically over the last few years. For this reason, it is imperative to swiftly develop and apply

in industrial practice various technologies for sewage sludge valorization. Considering the advantageous high protein content, these technologies can enhance the sustainable management and utilization of sewage sludge [3]. In 1991/1992, the European Union (EU) member states had 40,300 working WWTPs that produced 6.5 million tons of dry sewage sludge annually. This amounts to approximately 30 kg of dry sludge per capita each year [4].

Treatment and disposal of excess sludge obtained during the activated sludge wastewater purification process is now more challenging for WWTPs. That is due to economic, environmental, and regulation factors. Municipal excess sludge (ES) can be co-combusted with coal, providing an alternative method for ES disposal. Nevertheless, it is disadvantageous due to CO₂ emission. Land disposal is also possible, but the environmental unfriendliness, limited number of available sites, and sewage sludge landfilling prohibitions in some countries make this method unviable. Additionally, the high cost further contributes to its impracticality. The presence of pathogens and toxic heavy metals contributes to a decrease in sewage sludge application in agriculture. Such issues stimulate researchers to look for alternative disposal methods.

Sewage sludge contains significant amounts of exploitable materials, making it a sustainable resource for energy, nutrients, or secondary raw materials. This aligns with the philosophy of a circular economy. In the EU countries, many sludge treatment technologies are implemented, but differences are observed in each member state. The most popular stabilization methods in EU countries seem to be anaerobic and aerobic digestion. The former is used in 24 nations and the latter in 20 countries. Sludge is obtained in large volumes in WWTPs, making it the largest byproduct of wastewater treatment processes. The processing and disposal of sludge present complex technological and environmental problems, motivating engineers to search for new solutions. Sludge from WWTPs is in the form of a suspension, which typically contains 0.25 to 12% w/w solid phase, depending on the operation and process used. The preferred method for sludge dewatering is mechanical instead of using drying beds. In EU-15 countries (EU member states before 2004), especially in Germany, Italy, France, and the U.K., thermal drying is mostly applied. In the EU-15 states, the predominant choice for sludge final disposal is reuse, accounting for 53% of produced sludge. This includes composting and agricultural utilization, as waste streams are important sources of water, energy, and nutrients. As for other disposal forms, incineration is less frequently used (21% of produced sludge) [5].

For economical and sustainability reasons, resources embedded in sludge (energy, nutrients, raw materials, and process byproducts) should be recovered and used. This can create a virtuous wastewater-based circular economy cycle. The water from municipal wastewater can be recycled using adequate treatment technologies. The organic fraction achieved from the same processes can be harnessed to yield energy. Phosphorus, a relatively scarce element in soil, can be extracted for use in inorganic fertilizers, as it typically constitutes an average of 1.83% of sewage sludge content. Nonetheless, to take advantage of all the mentioned benefits, the adoption of advanced sludge treatment technologies is important. Pathogen and toxic compound removal as well as odor control is necessary to ensure human health and environmental protection [6].

Therefore, different advanced technologies are pursued to close the consumer circle for water and organic biomass. Researchers are currently focused on developing new technologies to enable energy and resource recovery from wastewater. As a result, advancements include technologies for obtaining H₂ from various types of wastewater [7–9]. One universally applicable technology utilizes ionizing radiation for wastewater treatment, sludge hygienization, and sludge disintegration for biogas plant optimization. The combination of ionizing radiation with bio-fermentation enhances the microbiological disinfection of sludge. Furthermore, electricity generated at the biogas power station serves as a power supply for the plant installations. Also, the heat produced is utilized to dry the byproduct fertilizer pellets. The intention behind writing the present paper is to illustrate the potential applications of radiation techniques in sewage sludge processing. Additionally, it aims to

elucidate the reasons for employing such techniques. Familiarization of the reader with the basics of ionizing radiation sources and the effects induced by this type of processing is another intention.

2. Wastewater Treatment Plants in the Circular Economy Era

A wastewater treatment plant (WWTP) is a facility that is purposed for mechanical and biological purifying of wastewater entering the system. WWTPs use natural biological aerobic processes. These are intensified by providing appropriate conditions like temperature, pH, aeration, or organic matter concentration. The principles of technology have not changed too much in the last 50 to 100 years [10]. Two main streams leave WWTP, purified wastewater and sludge. Regarding the obtained sludge, preliminary sludge and waste activated sludge (WAS) are waste but contain large amounts of organic matter. Thus, they are desirable in every branch where nutrients are needed, e.g., biogas plants, agriculture, etc. WWTPs are now beginning to be considered not just wastewater and sewage sludge treatment facilities but also as an opportunity to exploit new resources of mineable materials and energy. WWTPs will play a key role in future “ecologically sustainable” technological systems in SMART cities [11].

3. Ionizing Radiation and Electron Beam Sources

In the spectrum of electromagnetic radiation (Table 1) with a wavelength of less than 1 nm, we are dealing with X-rays. This type of radiation is emitted during electron deceleration/braking in the positive nuclei field (Roentgen radiation or bremsstrahlung). Electromagnetic radiation with a wavelength of less than 10 nm is called ionizing radiation. These are the X-rays mentioned before, and some fraction of ultraviolet (UV). The majority of UV radiation falls within the category of non-ionizing radiation. Only the higher-energy portion of the UV spectrum, with wavelengths ranging from approximately 10 to 120 nm (referred to as “extreme” UV), is considered ionizing. Nevertheless, the whole spectrum of UV radiation has a negative influence on biological structures. Effects are similar to those caused by ionizing radiation.

Table 1. The wavelength of electromagnetic radiation shorter than visible light.

Type of Radiation	Wavelength [m]
Visible light	10^{-6}
UV	10^{-8} to 10^{-6}
X-rays	10^{-13} to 10^{-9}
Gamma-Rays	$<10^{-11}$

Another type of ionizing radiation is the emission of particles. Two main types of such radiation are the emission of α particles (nuclei of helium consisting of two protons and two neutrons) and β^- particles, which are electrons. α radiation is impractical for industrial use due to its extremely low penetration depth. Therefore, only the emission of electrons is suitable for applications in industry. However, isotopic sources of β^- radiation usually emit other types of radiation simultaneously, presenting other disadvantages. The downsides of isotopic sources include the need for continual shielding and the progressive loss of activity over time. Electron emission radiation has higher dose rates compared to γ /X-ray radiation and still has considerable penetration depth. Hence, to harness this, an electron accelerator is a more suitable option. An electron accelerator is an electricity-powered device capable of imparting energy to electrons. It results in the formation of a stable and relatively evenly energetic beam known as an electron beam (EB). The primary advantages of this unit include the ability to regulate parameters. Particle energy and dose rate can be adjusted. Advantages include long-term stability and the ability to turn off radiation emissions.

All types of radiation, including γ and β^- from isotopes or electron beams and X-rays, produce secondary electrons when absorbed. These secondary electrons form reactive ions or free radicals during the energy transfer phase in the material [12].

Providing energy to electrons is possible due to the negative charge of these particles. That means they can be accelerated in an electric field. Firstly, electrons are emitted from the cathode. Then, they are accelerated in a high vacuum, forming EB characterized by parameters such as beam current and average beam energy. Considering the method of electric field generation, there are three existing types of electron accelerators:

- (a) DC type, where the electric field is obtained by generating a high potential difference between two electrodes;
- (b) Linear accelerators (LINACs), where microwaves and a large number of small resonators are used;
- (c) Radiofrequency accelerators (RF accelerators), which use radiofrequency (100 to 200 MHz) electromagnetic waves and a large, single resonant cavity.

Electron accelerators can also work in continuous or pulse mode. All of the mentioned types of accelerators have become the workhorse of radiation processing. They all have their advantages and disadvantages, making them suitable for different applications. For example, DC accelerators deliver high average beam power. LINACs give low average power and have low efficiency. In their case, only 10 to 20% of energy input is transferred to an electron beam. It is caused by power losses in the microwave generator and accelerating tube. However, LINACs can accelerate electrons to high energies, reaching 10 MeV, and have high energy gain per unit length. Such capabilities make them more compact in comparison to DC accelerators. LINAC accelerating structure can produce an electric field with over 10 MV/m compared to 2 MV/m for DC accelerators. DC and RF accelerators are more efficient, with RF accelerators reaching up to 50%. However, these accelerators can only produce beam energy of up to 5 MeV. Continuous wave RF-type accelerators can produce a DC-like beam current at higher energies. DC voltage power supplies in DC accelerators are usually simple and dependable. They frequently comprise high-power oil- or gas-filled high-voltage (HV) transformers with a suitable rectifier circuit. For an accelerator with a beam energy below 0.8 MeV, an HV cable is usually used to connect a power supply to the accelerating head. For the higher energies, a few MV voltages are necessary. In a conventional transformer, such values are impractical due to problems related to the insulation thickness and dimensions. For such cases, inductance or capacitance coupling of different types makes it possible to raise the low primary voltage to 5 MV by multistage cascade systems [13].

These days, electron accelerators are multifunctional devices that produce beams with varying powers and energies. The required electron energy is directly related to the density and structure of the objects to be irradiated. Beam power, which specifies the installation's total processing capacity, determines which type of electron accelerator is most appropriate for a given application [13].

Over the past few decades, the fundamental designs of electron accelerators used in radiation technology have been progressively improved. These advancements have been driven by new technological opportunities, including advancements in accelerator technology (Table 2). Simultaneously, there has been a recent trend toward the practical application of accelerator technology advancements. These developments, once limited to nuclear physics research apparatus, are now being used to create tools beneficial for industrial radiation processing settings. One of these novel approaches is the electron fixed-field alternating gradient (eFFAG), a cyclic electron accelerator. It operates on a continuous wave with electron energy, beam power, and dimensions tailored to meet radiation technology standards. They use separated magnet segments and a constant magnetic field akin to cyclotrons. Also, electron dynamics similar to the synchrotron is a distinguishing characteristic of eFFAG accelerators [14].

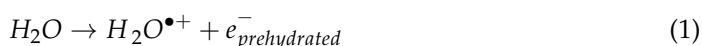
Table 2. Performance limits for electron accelerators used in radiation processing [13].

Parameter	Direct DC	UHF 100 to 200 MHz	LINAC 1.3 to 9.3 GHz
Beam mean current	<400 mA	<100 mA	<30 mA
Electrons energy	0.05 to 5 MeV	0.3 to 10 MeV	2 to 10 MeV
Beam power	~500 kW	700 kW	150 kW
Electrical efficiency	60 to 80%	20 to 50%	10 to 20%

An accelerator for radiation technology that uses a superconducting structure to accelerate electrons is another equally inventive design. This kind of acceleration section has 10^6 times less surface resistance. It results in very small HF power losses and boosts the device's efficiency. Simultaneously, the increased structural quality results in reduced energy consumption for system cooling. Thus, nearly all of the high-frequency power is transferred to the electron beam. Under these circumstances, a continuous wave (CW) operation with an acceleration gradient of 10 MeV/m is feasible [15].

4. Effect of Ionizing Radiation on Water Environment, Biological Effects

Irradiation of the water environment using ionizing radiation causes ionization and excitation of water molecules (Figure 1). The excited water molecules quickly return to the ground state while ionization forms cation radicals and free electrons (Equation (1)).



Ionized molecules react with water molecules to form hydroxyl radicals, $\bullet OH$ (Equation (2)):



The free electrons become hydrated, as presented in Equation (3):



The radicals react between themselves or with hydrogen ions (H_3O^+), forming molecules of $H^\bullet$, $\bullet OH$, H_2O_2 , and H_2 . The yield of radicals and molecular products is also dependent on pH. That is because $H^\bullet$ radicals and e_{aq}^- are in the acid–base equilibrium (Equation (4)). While some reactions involving e_{aq}^- are diffusion-limited, many exhibit activation energies varying between 4.2 and 33.6 kJ/mol. That indicates that control of kinetics is determined by the availability of a vacant orbital in the reacting partner species. But even for the simple reactions, the molecular-level mechanism is not always clear.



Radical water radiolysis products are highly reactive and most of the chemical reactions occurring in irradiated aqueous solutions are caused by them. The reduction potentials of selected water radiolysis products against Normal Hydrogen Electrode (NHE) are given in Table 3. An important parameter describing the effects of irradiation on the water environment is the radiation chemical yield (G or G-value). It gives information on the amount of water radiolysis products. It is defined as the number of moles of a given water radiolysis product per 1 J of absorbed energy (mol/J) [16].

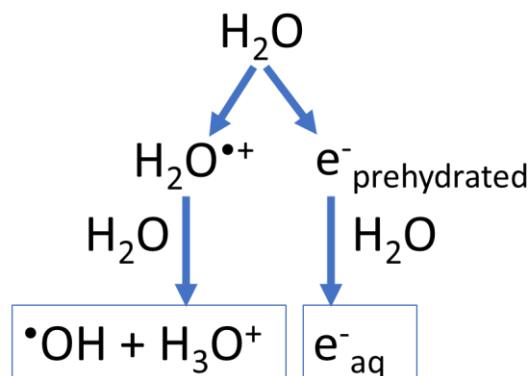


Figure 1. Water radiolysis: formation of free radical species and hydrated electrons in water utilizing ionizing radiation.

Table 3. Reduction potentials for main water radiolysis products [16].

Chemical Species	Reduction Potential vs. NHE
$\bullet OH$	+2.72 for redox couple $\bullet OH, H^\bullet / H_2O$ +1.90 for redox couple $\bullet OH / OH^-$
$H^\bullet$	−2.31
e^-_{aq}	−2.87

When this method is used, the products of water radiolysis and other reactive species generated in water are enhanced by ozone (Table 4), resulting in the elimination of organic and microbiological contaminants. This results in significant external effects, including the elimination of contaminants. However, the internal effects occur when ionizing radiation is penetrating cells. Ionizing radiation is lethal for living cells. One reason for this is the damage it causes to deoxyribonucleic acid (DNA), a biopolymer responsible for genetic information storage. If the DNA strand is damaged so severely that it cannot be fixed by repair mechanisms, the cell dies. Another consequence of DNA damage is mutation, entailing the modification of genetic information on the DNA strand; these are damages caused by two separate mechanisms. First is the indirect effect based on the chemical reaction of DNA with water radiolysis products, mostly $\bullet OH$ radicals. $\bullet OH$ radicals are added to the positions in heterocycles of bases (thymine, guanine, cytosine, or adenine) where double bonds result in the formation of radicals. Such molecules undergo further reactions with ring rearrangement, opening, or reactions with molecular oxygen. $\bullet OH$ can also attack H atoms of 2-deoxyribose in the phosphate backbone, resulting in strand breaks. The second type of action is the direct effect, where radiation ionizes the DNA strand. This ionization transforms the bases into radical species that undergo further reactions and are no longer able to perform their original functions. Additionally, the ionization of the sugar–phosphate backbone leads to strand breaks [17,18]. The scheme of direct and indirect effects is shown in Figure 2.

Living cells can be damaged by ionizing radiation, not only by damaging DNA but also by causing chemical changes in lipids creating cell membranes. Similarly, as in the case of DNA damage, cell membranes can be ionized directly by radiation or react with water radiolysis products. $\bullet OH$ radicals formed by water radiolysis can abstract hydrogen atoms from unsaturated lipid molecules, leading to the formation of carbon-centered radicals. Such radicals react rapidly with the oxygen molecule (O_2), forming a peroxy radical. The peroxy radical can then act as $\bullet OH$. It can abstract hydrogen atoms from another lipid molecule and transform itself into lipid hydroperoxide. Because of such chain reactions, the presence of one $\bullet OH$ radical may result in the formation of many radical and peroxidized product molecules. These chain reactions can also be terminated by the recombination of radical products [20].

Table 4. Comparison of different advanced oxidative processes (AOPs) applying ozone, UV radiation, and electron beam, combination of UV and EB with ozone [16,19].

Ozone	UV Radiation and Ozone of Aqueous Solutions	Electron Beam Irradiation of Aqueous Solutions + Ozone
$3O_3 + H_2O \rightarrow 2\bullet OH + 4O_2$	$O_3 + H_2O + h\nu \rightarrow H_2O_2 + O_2$ $O_3 + h\nu \rightarrow O_2 + O$ $O + H_2O \rightarrow H_2O_2$ $H_2O_2 \leftrightarrow HO_2^- + H^+$ $O_3 + HO_2^- \rightarrow \bullet OH + 2O_2$ $H_2O_2 + h\nu \rightarrow 2\bullet OH$	$H_2O \rightarrow 2.7\bullet OH + 2.7 H_3O^+ + 2.6 e_{aq}^- + 0.7 H_2O_2 + 0.6 H^\bullet + 0.45 H_2$ $O_3 + H^\bullet \rightarrow \bullet OH + O_2$ $O_3 + H_2O_2 \rightarrow 2\bullet OH + O_2$ $e_{aq}^- + H_3O^+ + O_3 \rightarrow H_2O + \bullet OH + O_2$
Direct oxidation by ozone molecules is a selective reaction, in which reactions of ionized and dissociated forms of organic compounds with O_3 are preferred over reactions with neutral form. When certain conditions occur, $\bullet OH$ is obtained from O_3 so indiscriminate oxidation may take place (indirect mechanisms).	Radiation is absorbed by solution, not water! Just one source for $\bullet OH$ (O_3 and H_2O_2 , respectively)	The coefficients in the above equation are the G values that are defined as the number of moles of a given radiolysis product per 1 J of absorbed energy. Radiation is absorbed by water, not by solution! Two sources of $\bullet OH$ (water radiolysis and O_3 decomposition).

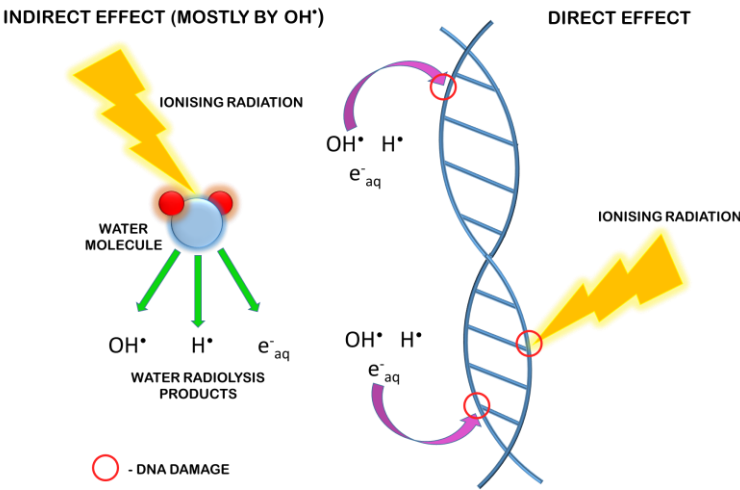
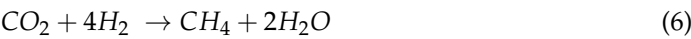


Figure 2. Schematic drawing presenting the direct and indirect effect of ionizing radiation damaging DNA molecules.

5. Methane Fermentation Process

One of the in-plant processing methods for the management of WAS is the methane fermentation process. The approach stabilizes sludge and partially converts its volatile compounds into biogas, which is mostly a mixture of methane and carbon dioxide [21]. Methane fermentation is a complex biochemical process in which high-molecular-weight substances such as carbohydrates, lipids, and proteins are degraded into lower-weight compounds, mainly methane and carbon dioxide. The whole process is usually divided into four steps. First is hydrolysis, where carbohydrates, lipids, and proteins are transformed into sugars, long-chain fatty acids, and amino acids. Second is acidogenesis, where volatile fatty acids (VFAs) and some alcohols are obtained from products of the previous step. During the third step—acetogenesis—an acetic acid is produced as well as some hydrogen. The last step is methanogenesis, a stage where methane is obtained by acetic acid digestion (Equation (5)) or by carbon dioxide (from the second, third, and fourth stages of methane fermentation) reduction by hydrogen (Equation (6)) [22].



Process operating conditions have a significant impact on biogas yield and quality. For instance, temperature changes exceeding some value can disturb methanogenesis, and pH values for each stage, hydrogen concentration in the gaseous phase, and mixing conditions are important [23]. For methane fermentation, microbial activity generally declines with temperature changes (temperature changes in the digester should not exceed 2 °C/d). pH must be maintained at the optimal level of 6.8–7.4 to ensure methane fermentation stability [24].

6. Sewage Sludge—A Resource, Not Waste

The right way to effectively manage the steadily rising waste sludge generation is to use waste sludge as a renewable resource for energy recovery. That will help meet strict environmental quality standards. It can also deliver dependable and economically priced energy for us and future generations. Sewage sludge has valuable qualities, such as its high energy and nutrient content. This, combined with the strict disposal guidelines, makes environmental engineers and scientists change their minds. Now, sewage sludge is considered as a viable energy resource rather than waste. It might be a significant step in the direction of creating a sustainable energy source. In particular, meeting current and future energy needs and lessening reliance on non-renewable resources is important nowadays [25].

Sewage sludge can be described with many parameters. The most important are:

Total solids (TS) representing the total amount of organic and inorganic compounds in sludge. Volatile solids (VS), the number of organic substances in the solid fraction of sludge. Chemical Oxygen Demand (COD), an indicator showing how much organic (and some inorganic) compounds can be oxidated using strong oxidative conditions present in sludge. COD can be divided into soluble COD (SCOD), COD of liquid phase, and total COD (TCOD), COD of both liquid and solid phases [26].

There are two main types of sewage sludges: preliminary sludge and waste activated sludge. Both have different content and structure due to their origins; however, both have some similarities. The preliminary sludge acquired during the initial stages of organic matter sedimentation in raw wastewater primarily comprises human and animal fecal matter, which includes deceased cells, along with toilet paper remnants. Additionally, it contains industrial wastewater residues, encompassing byproducts from food processing, particularly from fruits and vegetables, hair, and other similar substances. The organic part of these ingredients contains 40–60% proteins and 25–50% carbohydrates. Fat content is about 19%. Usually, TS content ranges between 2 and 6%, and VS content is about 60–75% in reference to TS. This type of sludge has a black or grey color, has a slimy consistency, and is malodorous. Due to the high content of easily biodegradable organic material, it is often fermented in storage tanks [27].

Secondary excess sludge is a suspension of flocs formed during the settling of suspension from the aerobic purification process. Bacteria taking part in the process of aerobic treatment (mostly heterotrophic bacteria strains: *Pseudomonas*, *Bacillus*, *Micrococcus*, *Alcaligenes*, *Moraxella*, *Flavobacterium*) produce an extracellular matrix consisting of polymeric substances. It is a mixture of bacterial metabolic products that forms a protective coating for these cells (extracellular polymeric substances (EPSs)). It can also be used as a source of energy in case of a deficiency of available nutrients. The main ingredients of EPS are carbohydrates and proteins. However, some other substances, including DNA and humic acids, can be present. EPS allows bacteria cells to aggregate into flocs [28]. Whatever the type of sludge is, all types represent sources of organic matter worth reusing.

7. Pathogens Present in Sewage Sludge

Even if a vision of sewage sludge utilization in various branches may be attractive, the occurrence of different pathogens in sewage sludge is still a matter of concern. Human and animal feces excreted into sewage, especially from hospital wastewater systems, may contain several types of pathogens. These include helminth eggs, protozoa, bacteria, fungi,

and viruses. And this takes place even in highly developed countries. The diversity and amount of pathogens depend on factors such as climate, living standards, and the specific area. Some of the mentioned pathogens are persistent in the environment. Therefore, they can survive in the sewage system and enter WWTP, growing more concentrated in primary sludge during the primary settling process. Furthermore, some of these organisms are able to survive during the treatment processes. Another worrying fact is that commonly performed tests for the presence of selected pathogens are ineffective or performed carelessly, leading to unreliable results [29].

In sewage sludge, a large number of bacteria cells can be found [30]. A number of bacteriological analyses carried out in municipal wastewater and sewage sludge indicated the presence of pathogenic bacteria species. These include *Salmonella* sp., *Shigella* sp., *Clostridium Perfringens*, *Clostridium Botulinum*, *Bacillus Anthracis*, *Vibrio holerae*, *Mycobacterium Tuberculosis*, etc. [31,32].

Czeszejko et al. [33] detected *Listeria* spp., typically *Listeria monocytogenes* (bacteria responsible for listeriosis), in samples from 60 different WWTPs. *Salmonella* sp. was found in wastewater from a slaughterhouse by Paszkiewicz [34]. Machnicka et al. [30] made a discovery while testing disintegration and anaerobic digestion as factors for removing pathogens from waste activated sludge. Tests were carried out on the reference samples for *Salmonella* spp., *E. coli*, and *Clostridium perfringens* and positive results were reported.

In sewage systems, intestinal parasitic helminths are present as well as their living ova due to the fact that they are excreted with feces of infected humans and animals [22]. Species commonly found include human roundworm (*Ascaris* sp.), animal roundworm (*Toxocara* sp.), and human whipworm (*Trichuris* sp.)—ATT, Figure 3. Others include, for example, *Enterobius vermicularis*, *Strongyloides stercoralis*, *Taenia solium*, *Taeniarhynchus saginata*, *Echinococcus granulosus*, *Schistosoma haematobium*, *Opisthorchis felineus*, *Paragonimus westermani*, *Fasciola hepatica*. This problem also affects not only portions of the world with low hygiene standards but even highly developed countries. There are a number of species of parasites present in sewage sludge. Many researchers indicate the availability of helminth eggs in sewage sludge.

Amahmid et al. [35] tested municipal wastewater and municipal wastewater sediment for *Giardia* cysts and roundworm (*Ascaris*) ova. *Ascaris* ova were found in 39.5% of samples of raw wastewater and in 83.3% and 70.8 % of samples of wastewater (depending on sampling point). *Giardia* cysts were found in 50% of raw wastewater samples and in 25% and 5.6 % of samples of wastewater sediments (depending on sampling points).

S. Chaoua et al. [36] tested the presence of parasite eggs in raw sewage and sewage sludge from two different WWTPs in Morocco. *Ascaris lumbricoides* ova were found in 88.32% of analyzed samples of wastewater from WWTP in Marrakech. The following were also detected: *Acylostoma duodenale*, *Trichuris trichuria*, *Capillaria* spp., *Taenia* spp., and *Hymenolepis* spp. in 4.96%, 0.97%, 0.89%, 3.04%, and 1.82% of examined samples, respectively. In sewage sludge from Marrakech WWTP, *Ascaris lumbricoides* ova were found in 95.11% of analyzed samples. Also, *Ancylostoma duodenale*, *Trichuris Trichuria* and *Taenia* spp. were found in 3.83%, 0.52%, and 0.4% of the tested samples, respectively. Another WWTP in Chichaoua also showed the presence of parasites. A total of 88.08% of the wastewater samples were positive for *Ascaris lumbricoides* ova. *Ancylostoma duodenale*, *Trichuris trichuria*, and *Capillaria* spp. were found in 5.07%, 3.53%, and 3.3% of tested samples, respectively. *Taenia* spp. and *Hymenolepis* spp. ova were absent. Chichaoua WWTP sewage sludge contained *Ascaris lumbricoides* ova in 29% of tested samples, *Ancylostoma duodenale* in 2.33%, *Trichuris Trichuria* in 0.66%, and *Capillaria* spp. in 1% of the analyzed specimens.

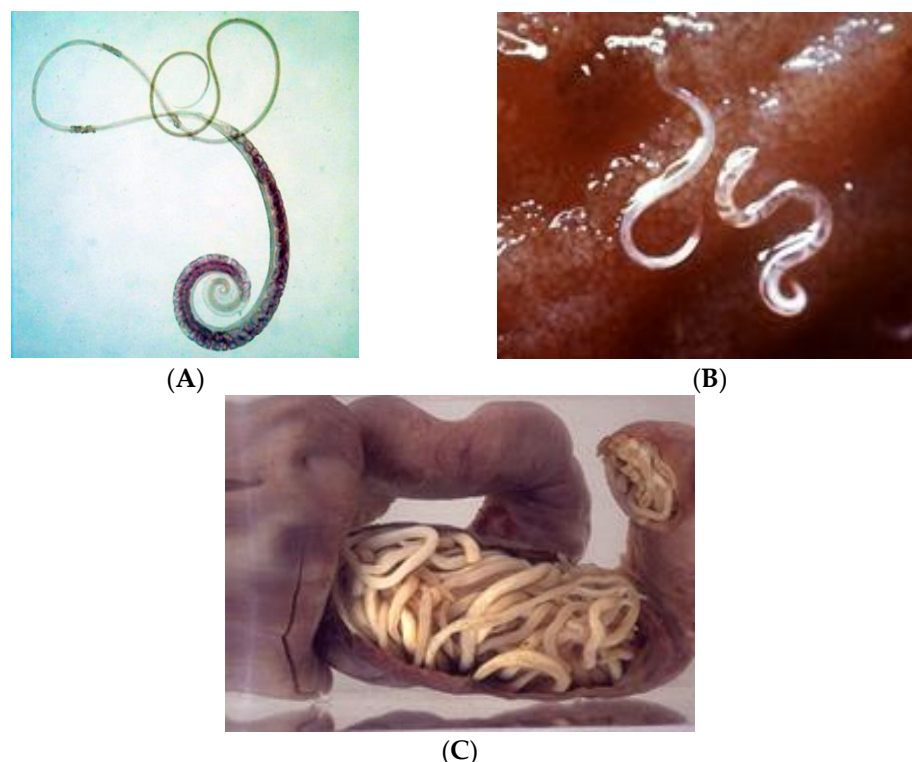


Figure 3. Human parasitic helminths: (A)—human whipworm (*Trichuris* sp.), (B)—human hookworm (*Ancylostoma duodenale*) and (C)—human roundworm (*Ascaris* sp.) inside surgically removed intestine.

T. Schilling et al. [37] studied *Salmonella typhimurium*, *Listeria monocytogenes*, and *Escherichia Coli* persistence in digestate from mesophilic anaerobic digestion process. Several types of agricultural origin feedstock were used for anaerobic digestion. These included cattle, horse, chicken, and swine manure and maize silage in different proportions, totaling 5 variants. Inoculated with mentioned bacteria load (10^8 CFU/mL of *E. Coli* and *S. Typhimurium* and 10^7 CFU/mL of *L. Monocytogenes*) samples were stored in different conditions to simulate storage during different seasons. Four types of conditions were introduced: 1—January-June, 2—April-September, 3—July-December, 4—October-March. The time necessary to reduce the number of tested bacteria by 4 logs was measured. The storage period caused a reduction in bacteria numbers. However, the time needed to achieve the assumed effect for all tested bacteria was never shorter than 16 weeks for the July-December variant. For the October-March variant, the number of *S. Typhimurium* was reduced to the given level after 12 weeks. Other strains remained over the limit even after 24 weeks when the experiment was stopped. It can be concluded that digestate storage has to be adequately long to achieve safe fertilizer and additional hygienization processes are required.

Human whipworm, human roundworm, and animal roundworm ova (*Ascaris* sp., *Trichuris* sp., and *Toxocara* sp.—ATT) can be present in sewage sludge across Poland. Zdybel et al. [38] worked with samples from 17 WWTPs located in seven separate districts in Poland. Samples were collected at different points in the visited WWTPs. The following types of samples were tested: raw sewage, sludge from grit removal, preliminary sludge, secondary sludge, digestate, and thickened sludge. Experiments revealed the occurrence of ATT ova in all types of samples. Tests were positive in 46, 11, 76, 44, 100, and 82% of samples, respectively. Zdybel et al., in another paper [39], presented the results for 92 samples from different WWTPs in 16 regions of Poland, obtained by varying methods of operation and size. ATT ova were found in 91 of the 92 examined samples, with ATT and *Toxocara* sp. being the most common. Hudzik et al. worked with 546 samples of sewage sludge collected from WWTPs in southern Poland. Their experiments, conducted

over six years (between 2003 and 2009), tested for the presence of *Ascaris* sp. and *Trichuris* sp. The research showed ova of these parasites in 35 samples (6.56%), with *Ascaris* sp. occurring more frequently [40].

Sewage sludge is also rich in viruses and fungi. Viruses can enter the sewage system while being excreted by infected humans or animals. Some of these are Enteroviruses (Poliovirus, Coxsackie viruses), Adenoviruses, Rotaviruses, Reoviruses, or Coronaviruses [22] (including SARS-CoV-2) [41]. Fungi are widely found throughout the environment and within the bodies of humans and animals. Unlike viruses, fungi can grow outside of living organisms. As a result, sewage sludge can become infected with various types of fungi, which can then proliferate within the sludge. Fungus types occurring in sewage sludge include yeast (*Candida* spp., *Cryptococcus neoformans*, *Trichosporon* sp.) and molds (*Aspergillus* spp., *Phialophora richardsii*, *Geotrichum candidum*, *Trichophyton* sp., *Epidermophyton* sp.) [22].

8. Sewage Sludge as Fertilizer—Legal Regulations

The use of untreated sludge or wastewater in agriculture poses a serious risk of bacterial and parasitic infection amongst humans [42]. Epidemiological studies carried out showed a significant association between roundworm or hookworm infections and the agricultural use of wastewater, human excreta, and sewage sludge [43]. This association is particularly evident in the case of children. Pathogens commonly occur in sewage sludge but can also be found in commercially available organic fertilizers and soil conditioners [44]. They are also persistent in soil. Long-term storage cannot be considered as a method for the removal of this threat. For this reason, legal regulations on sewage sludge and organic waste agricultural use must be introduced.

The EU generally encourages the use of sewage sludge in agriculture. EU Directive 91/271/EEC on urban wastewater treatment [45] in article 14 says, “Sludge arising from waste water treatment shall be reused whenever appropriate. Disposal routes shall minimize the adverse effects on the environment”. Nevertheless, strict standards must be fulfilled before dumping sludge on the field, and these are regulated as well. The Council directive of 12 June 1986 on the protection of the environment [46], particularly regarding soil protection, addresses the use of sewage sludge in agriculture (86/278/EEC). It contains numerous regulations related to the agricultural use of sewage sludge. In article 2 of this directive, definitions of sludge and treated sludge can be found:

- (a) “‘sludge’ means:
 - (i) Residual sludge from sewage plants treating domestic or urban waste waters and from other sewage plants treating waste waters of a composition similar to domestic and urban waste waters;
 - (ii) Residual sludge from septic tanks and other similar installations for the treatment of sewage;
 - (iii) Residual sludge from sewage plants other than those referred to in (i) and (ii);
- (b) Treated sludge’ means: sludge which has undergone biological, chemical or heat treatment, long-term storage or any other appropriate process to significantly to reduce its fermentability and the health hazards resulting from its use”.

EU directives do not indicate any limits for pathogen content in sewage sludge purposed for agricultural use. EU member states establish such limits on their own.

In Poland, sewage sludges are allowed for agricultural use under certain circumstances. Act of 14 December 2012, on waste, [47] says, “Recovery based on the use of municipal sewage sludge:

- (1) In agriculture, is understood as the cultivation of all agricultural products placed on the market, including crops intended for feed production,
- (2) For the cultivation of plants intended for the production of compost,
- (3) For the cultivation of plants not intended for consumption and for the production of fodder,
- (4) For land reclamation, including land for agricultural purposes,

- (5) When adapting the land to the specific needs resulting from the plan waste management, zoning plans or decisions on the conditions of development and land development—takes place in accordance with the conditions set out in par. 2–13”. It says: “The use of municipal sewage sludge is possible if it is stabilized and prepared appropriately for the purpose and method of their use, in particular by subjecting them to biological, chemical, thermal or another process that reduces the susceptibility of municipal sewage sludge to putrefaction and eliminates the threat to the environment or human life and health.”

It is also worth mentioning that the Regulation of the Minister of Economy of 16 July 2015 on the acceptance of waste for storage in landfills [48] imposed restrictions on landfilling some types of waste. Annex 4 defines conditions allowing for landfilling wastes other than hazardous and inert. The amount of total organic carbon cannot exceed 5% of total solids, which disqualifies sewage sludge. Thus, the only solution to dispose of it, other than incineration, is agricultural use or land reclamation. In Poland, any form of organic fertilizer (sewage sludges, biogas plant digestate, manure, etc.) has to fulfill restrictions referring to pathogen occurrence. Current Polish regulations with these restrictions are the same as previously mentioned:

- Regulation of the Minister of Agriculture and Rural Development of 18 June 2008, on the implementation of certain provisions of the Act on fertilizers and fertilization [49];
- Regulation of the Minister of the Environment of 6 February 2015, on the use of municipal sewage sludge [50].

In the first act, it can be found that “Fertilizers and agents supporting the cultivation of plants referred to in paragraph 1 must not contain: live eggs of intestinal parasites such as *Ascaris* sp., *Trichuris* sp., *Toxocara* sp. and *Salmonella* bacteria. In the case of fertilizers referred to in paragraph 5 point 7, additional requirements must be met beyond those set out in paragraphs 1 and 2. The number of bacteria of the Enterobacteriaceae family, determined based on the number of aerobic bacteria, should be less than 1000 colony-forming units per gram of fertilizer”.

The second act says, “§ 2. 1. Municipal sewage sludge may be used on land if the following conditions are met: (...) in the case of using this sludge in agriculture and for reclamation of land for agricultural purposes—*Salmonella* bacteria have not been isolated in a representative sample of sediments weighing 100 g obtained under § 5 section 3;

1. the total number of live eggs of the intestinal parasites *Ascaris* sp., *Trichuris* sp., and *Toxocara* sp. in 1 kg of dry matter, called hereinafter “d.m.”, of sludge intended for applied research:
 - (a) In agriculture and land reclamation for agricultural purposes—is 0,
 - (b) For land reclamation—is not more than 300,
 - (c) To adapt land to specific needs resulting from waste management plans, zoning, or zoning decisions—is not more than 300,
 - (d) For the cultivation of plants intended for the production of compost—is not more than 300,
 - (e) For the cultivation of plants not intended for consumption and the production of fodder—is not more than 300;”.

9. Sewage Sludge Disintegration for Anaerobic Digestion

Disintegration is a process for delivering energy (in various forms: mechanical, chemical, thermal) to biomass to cause a release of organic substances into the aqueous phase. It can be based on a variety of mechanisms and results in the disruption, lysis of microbial cells, and the destruction of floc structure. Such operation enhances the availability of nutrients for microorganisms responsible for tracking biological processes, e.g., methane fermentation [51–53]. There are several types of disintegration processes, which are presented in the scheme (Figure 4).

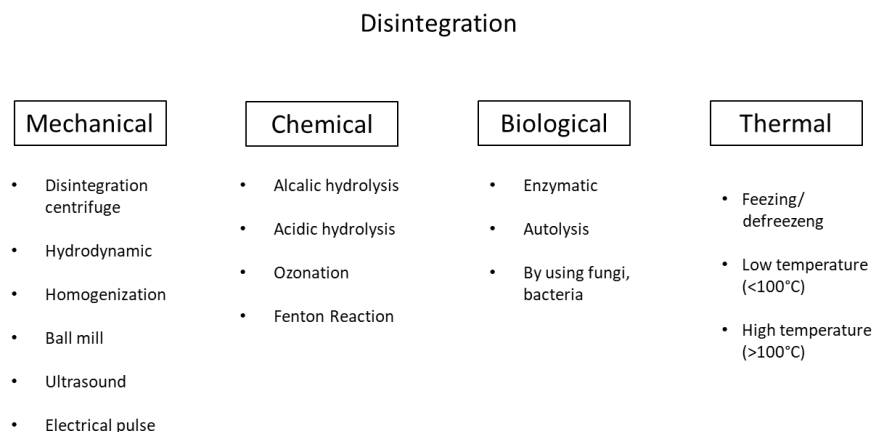


Figure 4. A schematic diagram illustrating the methods of biomass disintegration.

Many previous studies showed significant effectiveness in increasing SCOD for methane fermentation process enhancement. Sludge can be pre-prepared using a disintegration technique to increase the amount of biogas or to accelerate the methane fermentation process [53]. The INCT's biogas experimental plant has been studying novel low-temperature disintegration (process temperature: 55 °C, oxygen concentration: 0.2 mg/dm³). Low-temperature thermal disintegration of waste activated sludge mixed with distillation residue increased biogas production by 30% and methane production by 65% (over 26 days) [54]. Garlicka et al. used a mechanical device (causing cavitation in the disintegrated medium) for the disintegration of waste activated sludge and distillation residue. The study demonstrated that the energy input value in the hydrodynamic cavitation (HC) process is a critical factor. It can be defined as either ES (specific energy) or EL (energy density). Energy input is helpful in determining the likelihood of increasing methane production during the anaerobic digestion process. This parameter measures the sludge's disintegration-related changes (such as sludge floc deagglomeration, cell lysis, re-flocculation, and the associated release of compounds that can biodegrade from sludge flocs). EL = 140 kJ/L produced the greatest increase in methane yield (MY) of 152%, which is equivalent to ES = 3636 kJ/kg TS. Sludge floc deagglomeration in this instance was primarily caused by HC [55].

Żubrowska-Sudoł et al. tested a mechanical disc disintegrator in conjunction with a full-scale biogas plant. This featured a 5000 m³ mesophilic digester operating on mixed sludge (preliminary and waste activated sludge). Energy densities used in this study varied from 70 to 280 kJ/L for laboratory tests. A value of 20 kJ/L was used for the whole WAS digester feedstock. SCOD and volatile fatty acid (VFA) analysis showed a significant increase in both parameters; however, VFA stopped rising at 210 kJ/L. SCOD was higher for every next energy density used. Experiments with methane fermentation resulted in a biogas yield increase from 2400 m³/d to 3213 m³/d [56]. The sludge disintegration significantly improved the methane fermentation process.

10. EB Irradiation as an Alternative to Other Disintegration Methods

EB irradiation can be considered as another disintegration method. The impact of using EB disintegration of WAS on the efficiency of biogas production was investigated using the same techniques. The ILU-6 electron accelerator, which is a single-cavity accelerator with electrons of energy 2 MeV, and the Elektronika model 10/10 electron accelerator, which is a linear accelerator with electrons of energy 10 MeV, were used for the EB tests [57]. Absorbed dose underpins electron beam processing, which is then followed by chemical processes in which water radiolysis products are crucial [58]. The amount of biogas produced in 11 to 14 days was similar to the amount generated over 21 days for untreated samples (Figure 5).

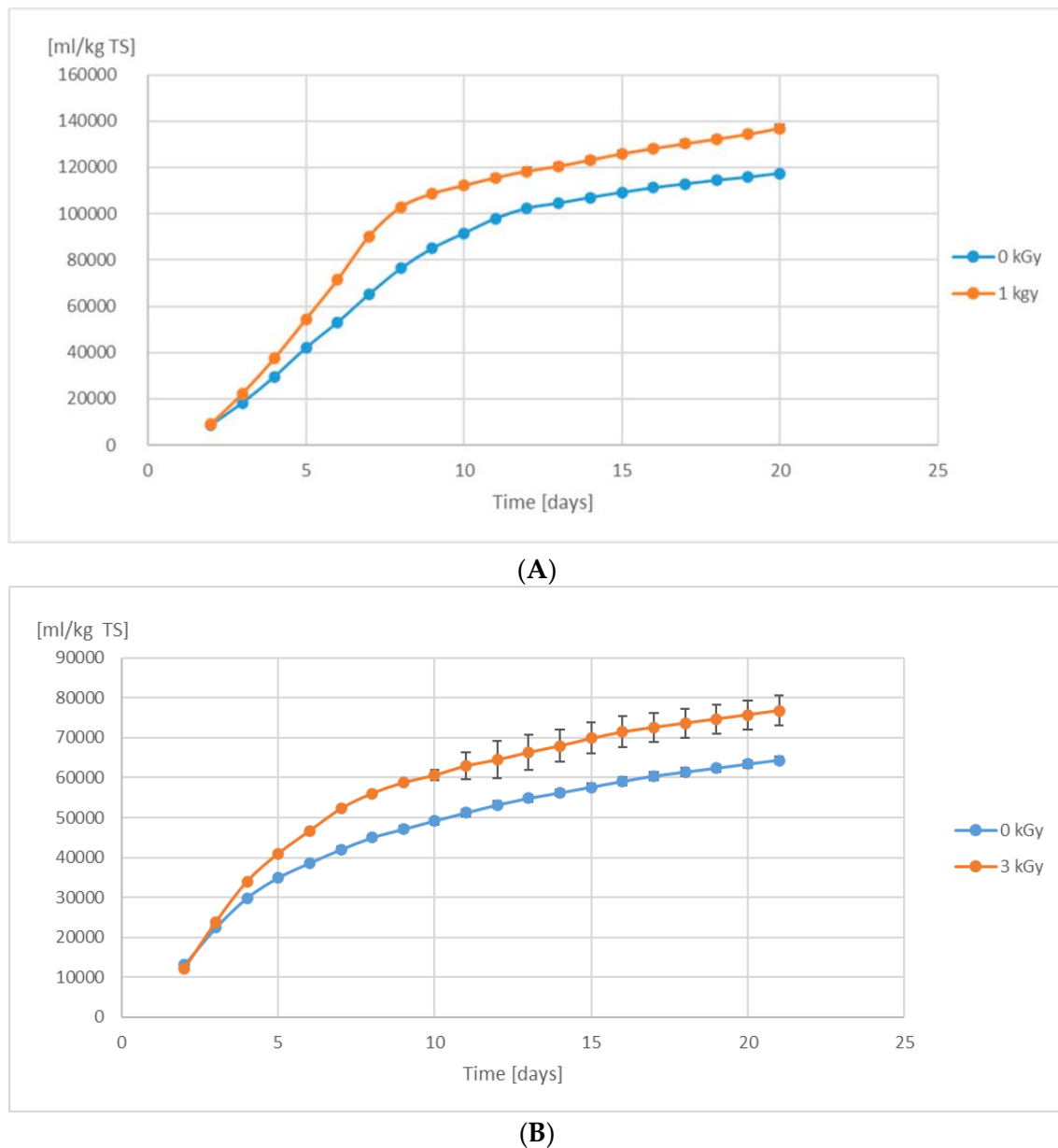


Figure 5. Methane generation over 21 days of mesophilic (38 °C) digestion of wastewater treatment plant sludge pretreated using EB (orange) at a dose of 1 kGy (A) and 3 kGy (B) and data for references samples not irradiated (blue: 0 kGy) [58].

There are significant process and financial ramifications when digester residence times can be shortened without sacrificing biogas production. Optimization research can shorten digester residence times and increase the efficiency of biogas production.

Park et al. [59] assessed an EB sewage sludge ($25,000 \pm 2000$ mg/L TS) treatment prior to the methane fermentation process. They used a 1 MeV electron accelerator and different sludge layer thicknesses varying from 2.5 to 10 mm. Exposure times varied from 0.3 to 1.2 s, and dose ranges of 1, 3, 5, 7, 10, and 20 kGy were used. Measurements of SCOD showed a tremendous increase in this parameter. After the irradiation to 20 kGy, increases of 49, 54, 97, and 147% were recorded after reducing sludge layer thickness from 10 mm to 7.5, 5 mm, and 2.5 mm. Such changes were caused by the limited penetration depth of the 1 MeV energy electron beam. Therefore, the thinner the layer, the more the treated sludge received the irradiation dose, and the total TCOD remained almost unchanged. Authors claimed that irradiation solubilized some of the organic compounds present in the solid

phase of a sludge. Experiments with exposure time changes with dose and layer thickness fixed showed no significant changes in SCOD. The authors evaluated protein concentration in the liquid phase after irradiation and noticed an increase with dose and layer thickness decrease. This was explained by cell rupture caused by EB treatment. VFA concentration decrease after the irradiation (by 66.7% for propionic acid after 20 kGy irradiation of 5 mm thick layer) was also noticed. Tests with irradiated sewage sludge as anaerobic digestion reactor feedstock were carried out in mesophilic conditions. Enhancement in the hydrolysis process occurred, manifested by SCOD increase in comparison to reference reactor content measurements. Also, higher biogas yield for reactors fed with irradiated sludge has been observed.

Chu et al. [60] irradiated sludge from the last step of the A2/O process (Bardenpho process) using ^{60}Co gamma source. Sludge contained 1.1 to 1.4 % TS and doses used in the study ranged from 5 to 25 kGy. The authors noticed distinct changes after irradiation. Microscopic observations showed floc breakage and changes in filamentous bacteria cells, including rupture. Also, the release of proteins and polysaccharides into the supernatant was observed after gamma treatment. Measured values of total organic carbon (TOC), total nitrogen (TN), and total phosphorus (TP) also rose after treatment. TOC and protein concentration were higher by two orders of magnitude at a dose of 25 kGy, while polysaccharide concentration increased by one order of magnitude at the same dose. Kim et al. [61] studied the influence of gamma radiation on WAS solubilization. WAS from Jeongeup WWTP in South Korea, with the average TS equal to 16 200 mg/L, has been studied. Irradiation was performed using ^{60}Co gamma source. The used irradiation doses ranged from 0 to 50 kGy. SCOD, TCOD, biological oxygen demand (BOD_5), and extracellular substances, including proteins, carbohydrates, and humic acid, were measured. While TCOD remained stable after irradiation for 50 kGy, SCOD increased from 700 to 2850 mg O_2 /L, and the SCOD/TCOD ratio increased from 4.8% to 19.0%. For the same absorbed dose, BOD_5 was increased from 160 to 787 mg O_2 /L. That is because gamma irradiation transforms non-biodegradable organics into biodegradable compounds. Also, the BOD_5 /SCOD ratio was calculated. The results showed an increase in that ratio for 10 kGy from 22.9% for nontreated samples to 40.0% for irradiated ones. However, the absorbed dose increase caused a decrease in this ratio. For 50 kGy, it was only 26.6%. It was concluded that the amount of non-biodegradable compounds converted into biodegradable ones was lower than the amount of compounds released into the water phase by irradiation. Carbohydrate and humic acid concentration in the liquid phase rises with the increase in absorbed dose. For 50 kGy, carbohydrates were recorded at 85.7 mg/L from an initial 4.9 mg/L. Humic acid concentration increased to 260.2 mg/L from an initial 22.4 mg/L for the same dose. Protein concentration increased from 5.8 to 53.9 mg/L for 10 kGy, but irradiation with larger doses showed no further changes.

Shin and Kang [62] worked with two types of EB irradiated sludge as a biogas fermentation reactor feedstock. This involved WAS with 1.5% TS and 73% VS and thickened sludge (mixture of WAS and preliminary sludge) containing 2.4 to 3.2% TS. The authors used a 1 MeV electron accelerator to determine the effect of sludge irradiation. They measured SCOD, pH, specific resistance to filtration (SRF), sludge volume index (SVI), alkalinity, and protein and carbohydrate concentrations in the liquid phase. Also, anaerobic digestion (AD) tests were carried out in 18 L bioreactors in mesophilic conditions ($35 \pm 1^\circ\text{C}$) for 50 days. It was carried out to evaluate irradiation's influence on organic compound conversion in the AD process. Sludge samples were irradiated with 0.5, 1, 3, 6, and 10 kGy. The influence of EB treatment on pH was negligible for both types of sludge; samples were slightly more acidic for 10 kGy. Alkalinity increased slightly for WAS and decreased slightly for thickened sludge with an increase in dose. TS and VS remained stable after irradiation, but SCOD, carbohydrates, and soluble protein concentration changed dramatically after treatment. For WAS, the initial SCOD level was 52 mg O_2 /L. After irradiation with 0.5, 1, 3, 6, and 10 kGy, it was 390, 735, 828, 1072, and 1254 mg O_2 /L, respectively. For thickened sludge, the initial value was 442 mg O_2 /L and 1259, 1377, 1560, 1913, and 1970 mg O_2 /L for

0.5, 1, 3, 6, and 10 kGy, respectively. From both cases, it can be concluded that the largest differences in relation to the reference sample occur after low doses of treatment. However, after irradiation with higher doses, the SCOD increase is not so intense. The influence of EB irradiation on carbohydrates and protein concentration in the liquid phase was studied. Doses of 0.5, 1, 3, 6, and 10 kGy increased protein content from an initial 14.4 to 121.8, 240.1, 306, 379.1, and 397.3 mg/L for WAS. For thickened sludge, the protein content increased from an initial 62.4 to 230.7, 235.4, 383.8, 469.2, and 559.9 mg/L, respectively. EB treatment using the same dose set increased the concentration of carbohydrates in the supernatant. This parameter was increased from 5.9 to 92.2, 108.4, 110.3, 119.7, and 116.8 mg/L for WAS. In the case of mixed sludge, it changed from 17.1 to 152.0, 158.3, 197.0, 243.2, and 262.7 mg/L, respectively. Again, the most significant results were obtained for lower doses. SVI, a parameter used to describe the settleability of treated sludge, decreased from 110–160 to 60–70 mg/L. However, dewaterability evaluated using SRF worsened after EB treatment; for 10 kGy, the SRF parameter increased from 0.45×10^{16} m/kg to 2.24×10^{16} m/kg in the case of WAS and from 0.37×10^{16} m/kg to 2.27×10^{16} m/kg for thickened sludge. An anaerobic digestion study showed a substantial increase in biogas yield when fermenting irradiated samples. Only WAS was used for this experiment, and the irradiation doses were 1, 3, and 6 kGy. The maximum biogas yield for the reactor with untreated sludge was 95 L/[m³d] on day 15. For the 1 kGy reactor, it was 180 L/[m³d] on day 15, for the 3 kGy reactor, it reached 260 L/[m³d] on day 15, and for the 6 kGy reactor, it peaked at 290 L/[m³d] on day 15. That represents increases in biogas yield of 189%, 274%, and 305%, respectively. Also, methane concentration in biogas was slightly higher for reactors with irradiated samples. VFA concentration on the 15th day was higher by 107% for 1 kGy, 132% for 3 kGy, and 153% for 6 kGy in comparison to the reference reactor.

Changqing et al. determined the effect of EB irradiation of WAS using a 1.8 MeV electron accelerator. The authors used sludge with 1, 3, and 10% TS content and experimented with various irradiated sludge layer thicknesses. Irradiation doses used in this study were 2, 5, 10, 15, and 20 kGy. SCOD and soluble total nitrogen (STN) changes after irradiation were researched. Also, UV absorption measurements of supernatant were carried out. Tests of SCOD showed an increase with irradiation dose but also with TS content, e.g., the more solid fraction, the higher the SCOD after the irradiation. The proposed explanation of this effect was the microbial cell damage, resulting in the release of intramolecular substances into the liquid phase. Additionally, it was noted that the decomposition of insoluble intramolecular substances into soluble products also contributed to this phenomenon. Samples containing 1, 3, and 10% TS after receiving 20 kGy dose had SCOD reaching 685, 1605, and 2270 mg O₂/L (0.5 cm layer thickness). Nonetheless, all samples had the same initial SCOD, as they were obtained from the same initial sludge sample just by thickening, settling, or centrifugation. A sample with 3% TS was used to test the dependence of irradiated layer thickness on SCOD increase. The obtained results indicated that the highest SCOD increase with the dose was achieved for 0.5 cm for each dose repeatedly. Similar results were obtained for STN representing $N - NN_3$, $N - NO_2^-$, $N - NO_3^-$, soluble organic nitrogen, but mostly proteins and nucleic acid. However, for STN test differences between results for 1, 3, and 10%, TS values were not as high as in the previous case. The dependence of STN changes on layer thickness, with 3% TS sludge being examined. Again, the highest STN increment for each dose was found for 0.5 cm. Also, it was noted that for doses higher than 5 kGy, the efficiency of radiation processes was lower. UV spectra analysis showed an increase in absorption from 240 to 300 nm with the dose. A peak near 260 nm comes from nucleic acid, while a peak near 280 nm is often the absorption peak of amino acids. It was concluded that EB treatment increases the concentration of amino acids and nucleic acids produced by microorganisms present in the sludge. As in previous experiments, the increase in peak intensity with the dose was dependent on the total solid (TS) content. Higher TS content resulted in greater absorption for a given dose [63].

11. Sludge Hygienization by Irradiation

Ionizing radiation: electron beam and gamma showed their effectiveness in pathogen removal from sewage sludge and municipal wastewater over the years of laboratory- and industrial-scale tests.

Asgari Lajayer et al. [64] conducted gamma irradiation of effluent and sewage sludge from WWTP in southern Teheran, Iran. ^{60}Co source was used for irradiation at a dose range of 5 to 20 kGy. The presence of total coliform units and fecal coliform units was examined. The test showed that for effluent, a dose of 5 to 10 kGy reduced total coliform load by 99.66–100% and fecal coliform load by 99.62–100%. For sewage sludge (TS = 28%), doses ranging from 5 to 20 kGy reduced total and fecal coliforms by 99.1–99.96% and 96.32–99.72%, respectively. The authors claimed that the higher doses needed to hygienize sludge are the result of higher TS content. That is because solid particles may react with water radiolysis products, creating an indirect effect, and can also work as a shield against radiation for bacteria cells, affecting the direct effect of irradiation.

Chmielewski et al. [65] irradiated thickened sewage sludge with 35% TS from municipal WWTP. Total bacteria content, spore-forming bacteria content, coliform counts, clostridium perfringens counts, and ATT ova presence were assessed. Doses ranging from 5 to 7 kGy were applied. Total bacteria content was reduced by 2 logs after 5 kGy irradiation, 3 logs with a dose of 6 kGy, and 4 logs with a dose of 7 kGy. Spore-forming bacteria number was reduced by 1 log for 5 kGy and 2 logs for 6 kGy. Coliform counts, interestingly, were unaffected after irradiation to 5 kGy, reduced by 2 log at 6 kGy, and 3 logs at 7 kGy. Clostridium perfringens also stayed intact in the sample with 5 kGy treatment and were reduced by 1 log for 6 kGy and by 2 log for 7 kGy. ATT ova turned out to be less resistant; after 5 kGy irradiation, only Ascaris sp. ova were found alive in the amount of 30 from the initial 90. The total ATT ova number was reduced from the initial 240 to 30.6 kGy, which was enough to remove all ATT ova. The doses necessary to remove pathogens seem to be high. This could be caused by the high TS content in the examined sewage sludge.

Praveen et al. [66] studied the effectiveness of 10 MeV EB treatment of sludges from aerobic and anaerobic treatment from two separate WWTPs in Texas. Samples with indigenous *E. coli* bacteria and aerobic and anaerobic spores were spiked with pathogens. The following species were involved: Salmonella Typhimurium, somatic coliphages, male-specific coliphages, poliovirus, and rotaviruses. D_{10} as an absorbed dose giving log1 reduction was determined. For *S. Typhimurium*, D_{10} was 0.28 ± 0.01 kGy for aerobically digested sludge and 0.23 ± 1.01 kGy for anaerobically digested sludge. D_{10} values achieved for other pathogens in aerobically treated sludge were as follows: *E. coli*: 0.31 ± 0.01 kGy, aerobic spores: 3.75 ± 0.24 kGy, anaerobic spores: 4.96 ± 0.34 kGy, somatic coliphages: 4.02 ± 0.38 kGy, and 2.25 ± 0.19 kGy for male-specific coliphages. In the case of anaerobically treated sludge, the found D_{10} values were as follows: *E. coli*: 0.25 ± 0.01 kGy, aerobic spores: 4.04 ± 0.33 kGy anaerobic spores: 3.12 ± 0.38 kGy, somatic coliphages: 4.07 ± 0.31 kGy, and 2.45 ± 0.16 kGy for male-specific coliphages. Poliovirus and rotavirus were determined only in anaerobically digested sludge, where D_{10} was 2.07 ± 0.69 and 1.53 ± 0.03 kGy, respectively. Another study showed that *S. Typhimurium* and *E. coli* were inactivated by EB. Irradiation by 1 kGy reduced *E. coli* number by 3.2 log for aerobic sludge and 3.8 log for anaerobically digested sludge. Such a dose caused a reduction in *S. Typhimurium* by 3.7 log for aerobic sludge and 4.5 log for anaerobically digested sludge.

Engohang-Ndong et al. [67] used a 3 MeV electron accelerator to irradiate 15 ± 3 % TS primary sludge in a cascade system. Doses obtained during the experiments were as follows: 2.7; 6.7; 13.2; 25.7; and 30.7 kGy. The number of living *Ascaris* sp. ova and CFU counts per 1 g of TS of total heterotrophic bacteria (THB), total coliforms (TC), and fecal coliforms (FC) were determined. After irradiation of the sludge using 2.7 kGy, $93 \pm 8.5\%$ of THB, 21.1 ± 11.4 % of TC, and $67.2 \pm 1.8\%$ of FC of the initial number survived. Applying 6.7 kGy, these values were $31 \pm 15\%$, $0.85 \pm 0.23\%$, and $1.85 \pm 0.65\%$ for THB, TC, and FC, respectively. A total of $8.9 \pm 1.3\%$ of THB initial number were kept alive after 13.2 kGy treatment and no TC and FC were found. The irradiation at 25.7 kGy removed all tested bac-

teria. D_{10} values were determined for THB, TC, and FC, reaching 8.94, 3.16, and 3.17 kGy, respectively. It was also mentioned that a dose of 6.7 kGy allowed the achievement of the requirements for A-class sludge for agricultural use. This is 180 CFU of TC per 1 g of TS. In the untreated sludge, *Ascaris* sp. living ova number was 312 ± 24 per 4 g TS. A total of $23 \pm 8\%$ of this amount were still alive after 2.7 kGy treatment. A total of $11 \pm 1.6\%$ survived 6.7 kGy irradiation, while $2 \pm 0.03\%$ were still viable after receiving a dose of 13.2 kGy. No living *Ascaris* sp. ova were found after irradiation using 25.7 kGy. The estimated D_{10} value was 7.93 kGy.

Naing and Lay [68] used ^{60}Co gamma source to irradiate wastewater from Mandalay, Myanmar, and sewage sludge samples. Initial bacteria counts were 24×10^7 CFU/mL for wastewater and 38×10^8 CFU/mL for sludge. Irradiation of wastewater at a dose of 1 and 2 kGy reduced bacteria counts by 2 and 4 log, respectively. The 3 kGy dose left only 21 CFU/mL, while after irradiation at 4 kGy, no bacteria were found. Bacteria counts in sewage sludge irradiated to 1, 2, 3, 4 and 5 kGy were 13×10^6 , 36×10^5 , 12×10^5 , 16×10^4 , and 32×10^3 CFU/mL, respectively. The 6 kGy treatment left 23 CFU/mL, and after 7 kGy irradiation, no bacteria were detected. Lower irradiation doses required to remove bacteria from wastewater seem to confirm previous statements. Higher TS content causes part of the radiation energy to be deposited on solid particles instead of microorganism cells, making the hygienization process more difficult.

12. Discussion—Results of Laboratory, Pilot, and Industrial Works Leading to Hybrid WWTP Sludge Treatment Layout

EB irradiation in conjunction with traditional physicochemical and biological treatment could be a particularly effective supplementary step in the development of hybrid installations. This will lead to an overall more effective removal of pollutants. As demonstrated in multiple applications [69], the radiolytic processes can produce the desired effects at much lower doses than irradiation alone. This will lead to better energy efficiency in the wastewater field. Also, it can improve water reuse and reutilization opportunities, and optimize process train design for wastewater treatment. EB technologies have been developed in the last few decades to guarantee the environmental safety of liquid effluents discharged into the environment. Industrial-scale research has shown that EB technologies for sludge hygienization and wastewater purification can be used to reduce the degradation of the environment [70]. The water treatment facility was constructed at the Guanhua Knitting Factory in southern China. This installation treats water contaminated with industrial dye residues using electron beam technology. That is because the molecules of these dyes are difficult to break down with chemicals or bacteria. These lengthy and intricate molecules in the wastewater can be degraded by using ionizing radiation. The treated water can be used again by employing electron beam technology. Between CNY 2.0 and 2.5/m³ was the operating cost of the entire wastewater treatment process [71], which included the EB unit of service.

Subsurface pollutants such as pesticides, gasoline additives, per- and polyfluoralkyl substances (PFAS), aromatic hydrocarbons, chlorinated organic compounds, and others can be broken down by the irradiation technique. The state-of-the-art knowledge and research developments in ionizing radiation-based remediation of contaminated soils and groundwaters were covered by Bao et al. [72].

WWTPs are considered resource recovery facilities in the age of circular economies. The resources that are targeted are, at the very least, phosphorus, biogas, and water. Sludge and effluent from municipal wastewater streams, however, must be sufficiently treated to remove the possibility of microbial pathogens. Electron beam technology has been shown to be effective in enhancing methane (biogas) production and sludge hygienization [58]. It was also shown that biogas-powered cogeneration can produce energy in different forms. Heat could be generated for fertilizer granulation and drying, as well as electricity to run the electron beam system [73]. Recovery and recycling of waste are important actions taken to stop environmental deterioration. Using ionizing radiation to clean up sewage sludge

can make it suitable for use in agriculture. The method effectively removes biological threats without harming the environment. It does not require the addition of chemicals to the sludge. Using electron accelerators is safer and does not require restocking the irradiation chamber with new sources. Thus, EB treatment appears to be a better option than using isotope sources. Putting together all these features allows the creation of a zero-energy biogas plant concept. It is the idea of a complex comprising a biogas plant, an electron accelerator, and a biofertilizer manufacturing line. Digestate produced in the methane fermentation of sewage sludge will be exposed to an EB. The electron accelerator will be powered by electricity generated from the combustion of biogas in a co-generator. The process aims to eradicate all pathogens. The end product will be a biologically safe premium fertilizer [73]. The document has been approved by the licensed design office. The study concerns a radiation module equipped with an electron accelerator working with a biogas plant. The biomass (excess sludge from WWTP) throughput would be 22,300 t per year, with an average TS content of 7.4%. The study includes the following:

- Determining the geometry of the radiation treatment process.
- Determining the optimal parameters of the accelerator.
- Selecting accelerators that meet the requirements.
- Compiling a list of accelerator manufacturers to collect offers.
- Determining service requirements (frequency of replacements, cost of spare parts, service time).
- Determining the requirements for the room where the accelerator will be installed.
- Determining the requirements for the employment of personnel operating the accelerator.
- Preliminary determination of operating unit and investment costs of the radiation installation.
- To meet the requirements of the radiation treatment process, the basic parameters of the electron accelerator were determined (Table 5).

Table 5. Basic accelerator parameters for the irradiation installation [74].

Parameter	Value
Electron energy	2.5 MeV
Beam power	50 kW
Width of scanning	160 cm
Ti window thickness	50 μ m
Beam current	20 mA
Dose	5 to 7 kGy
Distance window-product	20 cm

Assuming the useful area of radiation treatment, defining the depth at which the depth dose is equal to the surface dose is crucial. For the mentioned installation, the thickness of the useful layer was set to 8 mm. Beam power losses related to the specified range of the electron beam (greater than the useful range) were assessed to be not greater than 20%. The design of the building intended for the installation of a radiation module in a biogas plant is closely related to the intended use of the plant. Therefore, it is a direct result of the parameters of the electron accelerator to be installed in the plant. This includes the need for shielding walls and the design of rooms associated with the operation of the accelerator. A typical solution is a building with two storeys. The lower level houses the room with the electron beam exit and process equipment, while the upper level is used to install the basic components of the accelerator (Figure 6) [74].

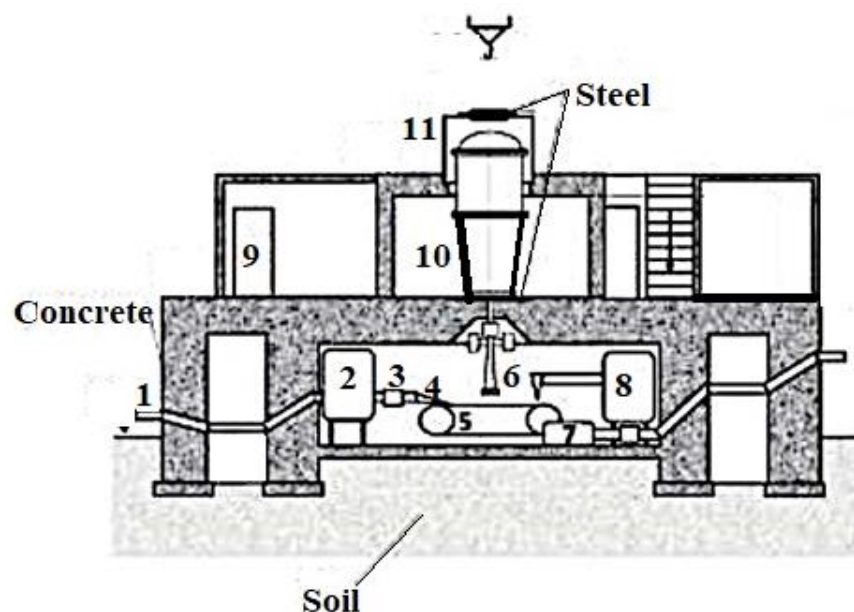


Figure 6. Cross-section of shielding for an accelerator with an electron energy of 2.5 MeV and a beam power of 50 kW along an axis perpendicular to the beam sweep direction with the location of process line elements, marked as follows: 1—sewage sludge transport system from the external tank; 2—intermediate sludge tank; 3—sludge pump providing the required pressure to form the sludge layer of the desired thickness with a nozzle forming the sludge layer; 4—nozzle; 5—conveyor; 6—output of accelerated electron beam; 7—installation for collecting sludge after radiation treatment; 8—lime tank and lime distribution nozzle for stabilization of sludge properties; 9—accelerator power cabinet; 10—electron accelerator; 11—steel enclosure [74].

The best selection among existing accelerators would be the transformer one, designed as industrial equipment being generally capable of 24-h operation. The advantages are relatively simple design, low cost in relation to the electron energy, and beam power offered. Also, unified systems easily adaptable to user requirements, ease of operation and reliability in operation, and electrical efficiency of 70% are important benefits. An important element in the operation of the accelerator is now computer-based control. This allows the accelerator operation to be easily adjusted to the user's needs, up to automatic operation as part of a process line. Application of this technology will reduce the amount of substrate needed to achieve the same heat/electrical power. It will allow for at least 10% higher methane content in the obtained biogas. These effects can be achieved by separating the processes of hydrolysis of the mixture of sewage sludge and biomass methane fermentation under commonly used conditions: mesophilic, thermophilic, and psychrophilic. This is performed by returning leachate obtained from these technological processes, containing appropriate bacterial cultures, for mixing and watering the biomass feed [75]. The process diagram is presented in Figure 7.

Environmental impact assessments (EIAs) are required for major building or development projects in the EU, as per the EU's Environmental Impact Assessment (EIA) Directive (2011/92/EU as amended by 2014/52/EU) [76]. Before the onset of the project, an EIA must be completed. For a number of projects, including waste treatment and disposal, an EIA is necessary. The investment project is recognized as a Project That Can Significantly Affect the Environment classified in § 2, sect. 1, item 47, processing systems as defined by Art. 3 section 1 item 21 of the act on projects that may significantly affect the environment, per the Regulation of the Council of Ministers of 10 September 2019, on projects that may significantly affect the environment (Journal of Laws 2019 item 1839) [77]. Excluding the system for agricultural biogas production as defined by Article 2, item 2 of the Act of 20 February 2015 on Renewable Energy Sources (Journal of Laws of 2018, item 2389, as amended) [78],

waste for waste other than those listed in items 41 and 46 includes storage yards for waste other than those listed in item 41. These facilities may process waste in quantities of not less than 10 tons per day or have a total capacity of at least 25,000 t. An extensive range of environmental factors are taken into consideration by the EIA when evaluating the direct and indirect significant impact of a project. These are population and human health, biodiversity, land, soil, water, air, climate, landscape, material assets, and cultural heritage. The report that the project developer submits to the approval authority must include some details, such as a description of the project (including its location, design, and size) and possible significant effects and reasonable alternatives. Others are project features and/or steps taken to prevent, lessen, or offset likely significant environmental impacts.

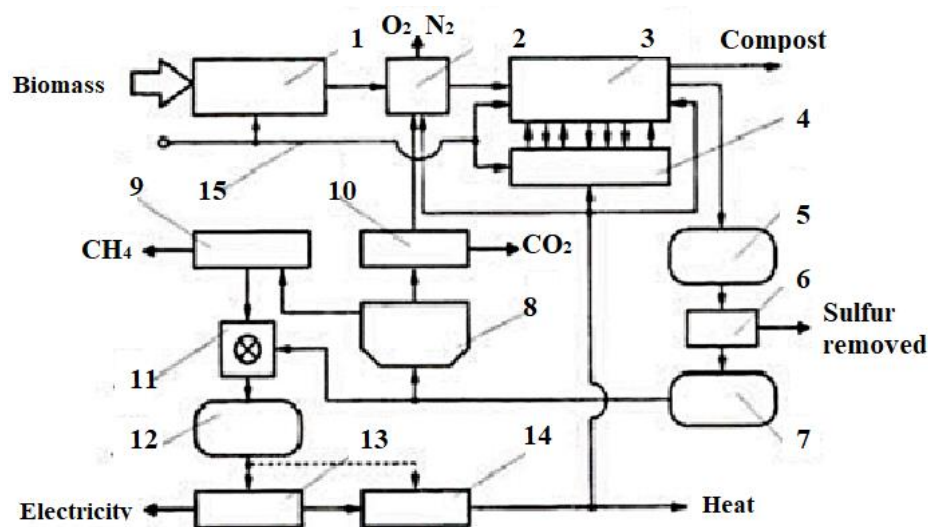


Figure 7. Process diagram. Biomass preparation system 1, hydrolyzer 2, series system of fermenters and composter 3, leachate return and enrichment system 4, crude biogas tank 5, biogas purification system 6, purified biogas tank 7, biogas separation system 8, methane processing system 9, carbon dioxide conversion system 10, gas mixer 11, standard fuel gas tank 12, electricity and heat generation system 13, heat conversion system 14, and external water intake 15.

13. Conclusions—Challenges for the Future Ionizing Radiation Applications in Future Waste-Free Technology Development: WWTP Case

Economically speaking, effective environmental control is far less expensive than irreversible environmental damage to the planet's life support systems. However, to meet these criteria, a variety of novel techniques and strategies are needed. Ionizing radiation may be one of the answers, much like frequently used UV radiation. Gamma sources are used for sludge processing in some countries. However, the most appropriate for most radiation processing applications are electron accelerators or EB/X systems. One special contribution to the application of this technological advance was the development of new, powerful electron accelerators. Such devices can be used for the online processing of massive flow streams of liquid or gaseous effluents. The accelerators were used to treat off-gas and wastewater in an incredibly efficient installation. However, more developments in accelerator technology are needed for the application of these processes in order to make this processing method competitive. This is due to the fact that the machines must function in demanding industrial sectors. In this field, a high degree of dependability, high power, and high energy consumption performance are required. A workshop examining the environmental applications of accelerators was conducted by Argonne National Laboratory [79]. The primary barriers preventing accelerator-related technology from being widely used are as follows:

- (i) The lack of accelerator devices that can function at full-scale industrial levels, which are typically ten times more advanced than the state of the art.

- (ii) The need to create accelerator systems that can be notably dependable and efficient while still being competitive with current technological advancements.
- (iii) The absence of pilot-level applications to demonstrate the effectiveness and efficiency of these new technologies. It is possible to use the provided data to compare these needs with the state of the market at this time. More complicated scenarios are associated with ecological applications that require high power, excellent electrical efficiency, and low-cost accelerators. Superconducting system-based solutions provided some hope, but their advantages and disadvantages were not thoroughly investigated on an industrial scale. They need magnet cooling systems, so they might not reach their predicted efficiency ratings. Utilizing clean energy sources to produce the electricity required to run accelerators could help solve the issue. The other challenge is the incorporation of an electron accelerator in the layout of the full sludge processing line. This is necessary for effective biogas and electricity production and the production of microbiologically safe fertilizer. The biogas generation in the fermenter should consider optimization of the process and proper organic carbon utilization. It is necessary to preserve outlet digestate value and the possibility of solid fertilizer production with its granulation with generator waste heat use. The composition of the final product can be upgraded by potassium compounds and other additives.

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