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THE USE OF IONIZING RADIATION IN SEWAGE SLUDGE TREATMENT FOR HYGIENIZATION AND DISINTEGRATION

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The use of ionizing radiation in sewage sludge treatment for hygienization and disintegration

Amounts of sewage sludge (SS) produced every year have a tendency to increase due to developing urbanization and rising number of people all over the world. SS is a waste product obtained in wastewater purification process conducted in wastewater treatment plants (WWTPs) as the effect of sedimentation in primary settlers and aerobic purification process in activated sludge tank. As a source of nutrients, it can be utilized as a feedstock for biogas fermentation process or as a fertilizer, and forms of SS disposal, such as incineration or landfilling, not only can be considered as tremendous waste but also are disadvantageous for the environment. Nevertheless, as the SS is the product originated from municipal wastewater, it contains variety of pathogents such as parasitic worms ova, protozoa, pathogenic bacteria, viruses, pathogenic fungi, *etc*. Therefore certain steps must be taken to eliminate mentioned threats before any agricultural use. Also before feeding SS into anaerobic digester to produce biomethane, hygienization process has to be performed while methane fermentation does not guarantee the removal of pathogens and waste from methane fermentation – the digestate is later distributed on the fields as an organic fertilizer. Introducing pathogens into the soil intended for agriculture may cause entering these threats to crops and subsequently infecting humans and animals. The amounts of chosen pathogens, which are *Salmonella* spp. and viable ova of three species of helminths: human roundworm (*Ascaris* sp.), carnivorous animal roundworm (*Toxocara* sp.), and human whipworm (*Trichuris* sp.), are regulated by the EU law as well as internal regulations of member states, including Poland. One of the possible ways to remove pathogenic organisms from SS is irradiation, as it can interact with DNA of living cells, damaging it, or interact with water molecules, leading to water radiolysis which produces highly reactive radical species. These species can also damage DNA or cell membranes, leading to cell death. An irradiation dose required to remove pathogens from SS can range from 3-4 to 15 kGy. Irradiation of sewage sludge brings one more profit which is disintegration – the effect causing the release of easily soluble organic nutrients from solid to liquid phase. Biomass processed in this way can be digested more easily, so methane fermentation process can be conducted more efficiently.

Wykorzystanie promieniowania jonizującego w obróbce osadów ściekowych w celu ich higienizacji oraz dezintegracji

Ilości wytwarzanych corocznie osadów ściekowych wzrastają ze względu na postępującą urbanizację i rosnącą liczbę ludności na całym świecie. Osady ściekowe są produktem odpadowym powstającym w procesie oczyszczania ścieków w oczyszczalniach ścieków jako efekt sedymentacji w osadnikach wstępnych oraz tlenowego procesu oczyszczania w zbiornikach osadu czynnego. Będąc źródłem składników odżywczych mogą być wykorzystywane jako surowiec w procesie fermentacji biogazu lub jako nawóz, a formy unieszkodliwiania osadów ściekowych, takie jak spalanie czy składowanie, nie tylko mogą być uznane za ogromne marnotrawstwo, ale są również niekorzystne dla środowiska. Osady ściekowe zawierają różnorodne patogeny, takie jak: jaja robaków pasożytniczych, pierwotniaki, bakterie chorobotwórcze, wirusy, grzyby chorobotwórcze itp., które należy usunąć, jeżeli omawiane osady mają być wykorzystane w rolnictwie w jakiejkolwiek formie. Również przed wprowadzeniem osadów ściekowych do komory fermentora w celu wytworzenia biometanu należy przeprowadzić proces higienizacji, gdyż fermentacja metanowa nie gwarantuje usunięcia patogenów, a odpad z fermentacji metanowej – poferment – jest później kierowany na pola uprawne jako nawóz organiczny. Wprowadzenie patogenów do gleb przeznaczonych na cele rolnicze może spowodować przedostanie się ich do upraw, a następnie zarażenie ludzi i zwierząt. Ilości wybranych patogenów, którymi są *Salmonella* sp. i żywe jaja trzech gatunków robaków: glisty ludzkiej (*Ascaris* sp.), glisty zwierząt mięsożernych (*Toxocara* sp.) i włosogłówki ludzkiej (*Trichuris* sp.), regulowane są zarówno przez prawo UE, jak i wewnętrzne rozporządzenia krajów członkowskich, w tym Polski. Jednym z możliwych sposobów usuwania organizmów chorobotwórczych z osadów ściekowych jest napromieniowanie, ponieważ może ono oddziaływać na DNA żywych komórek, uszkadzając je, lub wchodzić w interakcje z cząsteczkami wody, prowadząc do radiolizy wody, w wyniku której powstają wysoce reaktywne rodniki. Te indywidua chemiczne również mogą uszkadzać DNA lub błony komórkowe, co prowadzi do śmierci komórek. Dawka promieniowania wymagana do usunięcia patogenów waha się od kilku (3-4) do 15 kGy. Napromieniowanie osadów ściekowych daje jeszcze jedną korzyść, jaką jest dezintegracja – efekt powodujący uwolnienie łatwo rozpuszczalnych organicznych składników pokarmowych z fazy stałej do ciekłej. Tak przetworzona biomasa łatwiej ulega fermentacji, dzięki czemu proces fermentacji metanowej jest wydajniejszy.

CONTENTS

1. INTRODUCTION

Sewage sludge (SS) is a waste product obtained in the process of wastewater treatment, both municipal and industrial, usually in the form of a thick paste containing about 2-5% w/w of solid particles. It is rich in valuable organic nutrients and elements such as phosphorus, nitrogen, and potassium [1]. This entails that SS can be considered as a good soil fertilizer or a renewable source of energy as a substrate for the methane fermentation process, with the later utilization of digestate, a waste product of the methane fermentation process, as a fertilizer. This is also important due to the fact that large amounts of SS are produced every year. In Poland, it is about 1 million tons of dry mass per year, also connected with the prohibition of landfilling sewage sludge since 2016 [2, 3]. Innovative ways to exploit constantly recovering amounts of SS are then needed. Nevertheless, due to the origin of sewage sludge, the presence of health-threatening pathogens is nearly guaranteed. Chemical pollutants, *e.g.* heavy metals, drugs, or pesticides, are also often found in sewage sludge. For this reason, some sort of treatment is required to remove the mentioned negative factors and make sewage sludge safe for agricultural use.

2. SEWAGE SLUDGE

2.1. Wastewater treatment plant method of operation

A wastewater treatment plant (WWTP) is a facility designed for mechanical and biological purification of wastewater entering the system. WWTPs use natural biological aerobic processes but in intensified form by providing appropriate conditions such as temperature, pH, aeration, or organic matter concentration.

The first step is the mechanical separation of large objects (larger litter, branches, *etc*.) from the stream of wastewater entering the WWTP. After that step, the stream is directed to grit removal, where smaller particles (smaller litter, pieces of plastic, wood fragments, *etc.*) are removed on another sieve. Then, the sand removal process takes place. It is usually done by passing a stream of raw wastewater through a wide and relatively shallow channel at low speed so that sand grains can settle freely without being carried by the stream. Sometimes sand removal is performed before grit removal. The grit and sand are then landfilled. The next step is degreasing, which is the removal of fats and oils. This process is usually conducted by flotation. Air bubbles stick to hydrophobic droplets, and such agglomerates form a layer of foam at the top of the chamber, where they can be collected. Also, limiting wastewater stream flow can ensure laminar flow so that oil droplets with a density lower than water can float at the top of the chamber. Then, roughly cleared wastewater undergoes primary treatment to remove easily settling particles of organic matter, where primary clarifiers are used. Primary clarifiers are settlers, which can have a round, square (stream is directed to the center of the settler and spreads towards the exterior), or rectangular shape (stream enters the settler from one side and moves to the opposite one). This process is similar to sand removal by reducing flow speed. Because the density of organic matter is just slightly higher than the density of water (about 1.1 g/cm^3), surface loading rate of the primary clarifier cannot be too high to allow sedimentation of particles without interruptions. Settled particles form sludge that is constantly removed from the bottom of the settler as preliminary sludge. The water phase, still containing solid particles and soluble organic compounds, is discharged through the upper edge of the settler and directed to the next step of purification process, which is secondary treatment. Secondary treatment is based on the same processes which occur in natural microbiological aerobic water purification, where microorganisms transform organic matter into mineral compounds such as carbon dioxide, nitrates, and nitrogen (reaction 1).

$$C_{organic} + O_2 \xrightarrow{bacteria} CO_2 + H_2O + bacteria \tag{1}$$

However, it is intesified by ensuring appropriate conditions. Secondary treatment can be carried out in tanks with an aeration system that allows for optimal oxygen levels in the whole volume of the tank where bacteria form flocks (activated sludge process). Another option is the “trickling filters” process, where bacteria form biofilm on fixed beds made of different materials (rocks, ceramic, plastic media, polyurethane foam, *etc.*) and purified wastewater is sprayed onto these beds. In both cases, bacteria utilize organic compounds to feed themselves and produce new bacterial cells. When the process is conducted in an aeration tank, after the time necessary to digest organic compounds by the aerobic microorganisms, content of the tank is directed to secondary settlers, where flocks composed of bacteria cells undergo sedimentation, forming secondary sludge (or waste activated sludge – WAS). Part of this sludge is recirculated to the aeration tank to inoculate new portions of primarly purified wastewater, while the rest forms a waste called secondary excess sludge. In trickling filters, flocks taken away by streams of wastewater are sedimented or removed from the water phase by flotation. Water from secondary storage tanks with low content of dissolved organics can be considered purified and dumped into the natural water reservoir or subjected to other purification processes such as denitrification. Obtained sludges: preliminary sludge and secondary excess sludge are waste, however, containing large amounts of nutrients as well as biological threats [4].

2.2. Structure and composition of sewage sludge

As previously mentioned, there are two main types of sewage sludge: preliminary sludge and secondary excess sludge. The main parameters to characterize sewage sludge are:

- Total solid (TS) – the total amount of organic and inorganic substances suspended in the water phase of sewage sludge. It is measured gravimetrically after drying the sample at 105°C and given in g/L, kg/m^3, or weight percentage of solids in sludge.
- Volatile solid (VS) – the organic fraction of TS measured gravimetrically by loss in ignition of dried sludge samples and usually expressed as a weight percentage of organic substances included in the solid fraction of sewage sludge.
- Chemical oxygen demand (COD) – an indicator of the total amount of organic and some inorganic substances that are possible to oxidize by strong oxidizing agents such as potassium dichromate in sulfuric acid. COD is given as an amount of oxygen needed to react with measured substances in mg per 1 liter of sample and can be related to the amount of substances dissolved in the water phase (SCOD – soluble COD) or to the amount of substances in both the solid and liquid phases (TCOD – total COD).
- Biological oxygen demand (BOD) – an indicator of the amount of biodegradable substances in the sample given as the amount of oxygen in mg used by the bacteria to oxidize these substances at 20°C within 5 (BOD_5) or 20 days (BOD_{20}) per 1 liter of sample.
- Total organic carbon (TOC) – the amount of carbon originating from organic compounds in mg per 1 liter of sample [1, 4].

Due to their origin, both have different content and structure, but they do share some similarities. Preliminary sludge obtained during organic matter sedimentation in raw wastewater consists of human and animal feces (including dead cells), toilet paper, industrial wastewater (including food processing wastewater, leftovers of fruits and vegetables), hair, *etc*. Organic parts of these ingredients consist of 40-60% proteins and 25-50% carbohydrates. Fat content is about 19%. Usually, TS ranges between 2 and 6%, and VS is about 60-75% in reference to TS. This type of sludge is black or gray, has a slimy consistency, is maldodrous, and due to the high content of easily biodegradable organics, it often ferments in storage tanks [4]. Secondary excess sludge is a suspension of flocks formed during the sedimentation of suspension from the aerobic purification process. Bacteria taking part in the process of aerobic treatment (mostly heterotrophic bacteria species: *Pseudomonas*, *Bacillus*, *Micrococcus*, *Alcaligenes*, *Moraxella*, and *Flavobacterium*) produce extracellular matrix consisting of polymeric substances. It is the composition, also called extracellular polymeric substances (EPS), of metabolic products of bacteria cells that form a protective layer for these cells and can also be used as a source of energy in cases of a lack 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 also allow for bacterial cells aggregation into flocks [5]. These flocks contain not only bacteria cells and EPS but also other organic compounds: soluble high molecular weight substances and colloidal substances adsorbed on the surface of flocks, insoluble particles occluded inside flocks. In the case of both types of sludge, there are also soluble organic compounds dissolved in the water phase.

2.3. Pathogens present in sewage sludge

The presence of different pathogens in sewage sludge is a matter of concern. Human and animal feces excreted into sewage, especially onto hospital wastewater systems, may contain helminth ova, protozoa, bacteria, fungi, and viruses even in highly developed countries; however, their types, diversity, and amount in sewage sludge depend on climate, life standards, area, *etc*. Some of the listed pathogens are persistent in the environment, so they can survive in the sewage system and enter WWTP, becoming more concentrated in primary sludge during the primary settling process. Some of them are also able to survive treatment processes. Another worrying fact is that commonly performed tests for the presence of pathogens are ineffective or done carelessly, giving false-negative results [1, 6].

2.3.1. Bacteria

In sewage sludge, a large number of bacteria cells can be found [7]. In addition to sparophytes and fecal bacteria (*Escherichia coli*), the previously mentioned pathogenic bacteria species are also present. Bacteriological analyses of municipal wastewater and sewage sludge allowed to identify some of the pathogenic bacteria species present there: *Salmonella* sp., *Shigella* sp., *Clostridium perfringens, Clostridium botulinum, Baccillus anthracis, Vibrio cholerae, Mycobacterium tuberculosis, etc*. Czeszejko *et al.* detected *Listeria* sp., mostly *Listeria monocytogenes,* bacteria responsible for listeriosis, in samples from 60 different WWTPs located in Poland [8]. *Salmonella* sp. was found in wastewater from slaughterhouses by Paszkiewicz [9]. Machnicka and Grübel [7], testing disintegration and anaerobic digestion as factors removing pathogens from waste activated sludge, tested reference samples for *Salmonella* sp., *Escherichia coli*, and *Clostridium perfringens* and received positive results.

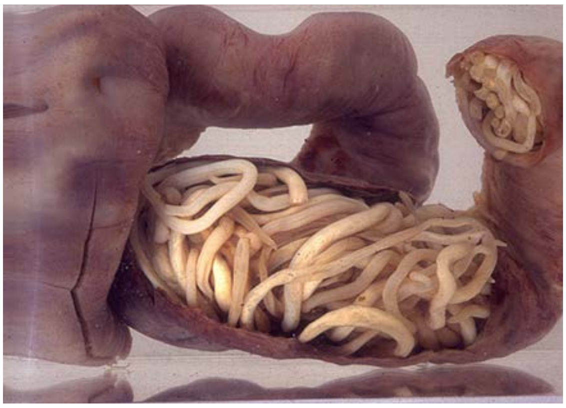
2.3.2. Helminths

In sewage systems, intestinal parasitic helminths [10] (Fig. 1) and their viable ova are present, due to the fact that they are excreted with the feces of infected humans and animals. Similarly to the previous example with bacteria, this problem concerns not only parts of the world with poor hygienic standards but also highly developed countries. There are a number of species of parasites that can occur in sewage sludge. It can be Nematodes (for example *Ascaris* sp., *Toxocara* sp., *Trichuris* sp., *Enterobius vermicularis, Strongyloides stercoralis, etc*.), Cestoda (for example *Taenia solium, Taeniarhynchus saginata, Echinococcus granulosus*), or Digenea (*Schisostoma haematobium, Opisthorchis felineus, Paragonimus westermani, Fasciola hepatica*). Many researchers indicate the presence of helminth ova in sewage sludge. Amahmid *et al.* tested municipal wastewater and municipal wastewater sediment for the presence of *Giardia* cysts and roundworm (*Ascaris* sp.) ova. *Ascaris* sp. ova were found in 39.5% of raw wastewater samples, in 83.3 and 70.8% of wastewater sediment samples (depending on the sampling point). *Giardia* cysts were found in 50% of raw wastewater samples, in 25 and 6% of wastewater sediment samples (depending on the sampling points) [11].

Chaoua *et al*. tested the presence of parasite ova in raw sewage and sewage sludge from two different WWTPs in Morocco. *Ascaris lumbricoides* ova were found in 88.32% of the wastewater samples tested from the WWTP in Marrakech. *Ancylostoma duodenale, Trichuris trichuria, Capillaria* sp., *Taenia* sp., and *Hymenolepis* sp. were also detected in 4.96, 0.97, 0.89, 3.04, and 1.82% of the samples tested, respectively. In sewage sludge from Marrakech WWTP,

A

B



C

Fig. 1. Human parasitic helminths: A – human whipworm (*Trichuris* sp.), B – human hookworm (*Ancylostoma duodenale*) and C – human roundworm (*Ascaris* sp.) inside a surgically removed part of the intestine.

Ascaris lumbricoides ova were found in 95.11%, *Ancylostoma duodenale* in 3.83%, *Trichuris trichuria* in 0.52%, and *Taenia sp.* in 0.4%. Another WWTP in Chichaoua also showed the presence of parasites. 88.08% of the wastewater samples were positive for *Ascaris lumbricoides* ova. *Ancylostoma duodenale*, *Trichuris trichuria*, and *Capillaria* sp. were found in 5.07, 3.53, and 3.3% of the samples tested, respectively. *Taenia* sp. and *Hymenolepis* sp. ova were absent. Chichaoua WWTP sewage sludge contained *Ascaris lumbricoides* ova in 29% of the samples tested, *Ancylostoma duodenale* in 2.33%, *Trichuris trichuria* in 0.66%, and *Capillaria* sp. in 1% [12]. Schilling *et al*. tested *Salmonella typhimurium*, *Listeria monocytogenes*, and *Escherichia coli* persistence in digestate from mesophilic anaerobic digestion processes of agricultural origin feedstock: cattle, horse, chicken, and swine manure, and maize silage, in different proportions (five variants). Inoculated with the previously mentioned bacteria load (10^8 CFU of *Escherichia coli* and *Salmonella typhimurium* and 10^7 CFU of *Listeria monocytogenes*), the samples were stored under different conditions to simulate storage during different seasons: January-June, April-September, July-December, and October-March. The time necessary to reduce the number of tested bacteria by 4 logs was measured. The storage period resulted in reduction in bacteria number; however, the time to achieve the assumed effect for all tested bacteria was never shorter than 16 weeks (for the July-December variant or the October-March variant). *Salmonella typhimurium* number was reduced to a given level after 12 weeks, but other species exceeded the limit even after 24 weeks, when the experiment was stopped. It can be concluded that digestate storage has to be long enough to achieve safe fertilizer and additional hygienization processes are required [13]. As it was already mentioned, the occurrence of helminth ova in sewage sludge is not only a problem in African countries. Human whipworm, human roundworm, and animal roundworm ova (*Ascaris* sp., *Trichuris* sp., and *Toxocara* sp. – ATT) can occur in sewage sludge across Poland. Zdybel *et al.* worked with samples from 17 WWTPs located in seven separate districts in Poland. The samples were collected at different points in the WWTPs visited. The following types of samples were tested: raw sewage, sludge from grit removal, preliminary sludge, secondary sludge, digestate, and thickened sludge. Experiments showed the presence of ATT ova in all types of samples; tests were positive in 46, 11, 76, 44, 100, and 82% of samples, respectively [14]. Another paper presented by Zdybel contains the results of testing 92 samples from WWTPs of different methods of operation and sizes from 16 regions of Poland. ATT ova occurred in 91 of the 92 examined samples; *Toxocara* sp. were the most common [15]. Hudzik and Wodzisawska-Czapla worked with 546 samples of sewage sludge taken from WWTPs in southern Poland. Experiments were carried out within 6 years (between 2003 and 2009), and the presence of *Ascaris* sp. *and Trichuris* sp. was tested. Tests showed the presence of ova of these parasites in 35 samples (6.56%), and *Ascaris* sp. seemed to occur more frequently [16].

2.3.3. Other pathogens

Sewage sludges are also abundant in viruses and fungi. Viruses can enter sewage system while being excreted by infected humans or animals and these are: Enteroviruses (Poliovirus, Coxsackieviruses), Adenoviruses, Rotaviruses, Reoviruses, and Coronaviruses (including SARS-CoV 2) [17]. Fungi are very widely found in the whole environment, including humans and animals bodies, but in opposite to viruses, they can grow outside organisms. Thus sewage sludges can be infected with different types of fungi, which then are growing in sludges. Fungi types occurring in sewage sludges are: yeast (*Candida* sp., *Cryptococcus neoformans, Trichosporon* sp.) and moulds (*Aspergillus* sp., *Phialophora richardsiae, Geotrichum candidum, Trichophyton* sp., *Epidermophyton* sp.) [10].

3. CURRENT REGULATIONS ON AGRICULTURAL UTILIZATION OF SEWAGE SLUDGE

The use of untreated sludge or wastewater in agriculture poses a serious risk of bacterial and parasitic infection among humans [18]. Epidemiological studies carried out by a number of authors showed a significant association between roundworm or hookworm infections and agricultural use of wastewater, human excreta, and sewage sludge, especially in children [19]. Pathogens are commonly found in sewage sludge, but can also be found in commercially available organic fertilizers and soil conditioners [20]. They are also persistent in the soil and even long-term storage does not guarantee successful pathogen removal. For this reason, regulations on sewage sludge and organic waste for agricultural use had to be introduced.

3.1. European Union regulations on agricultural use of sewage sludge

The European Union (EU) encourages the use of sewage sludge in agriculture. EU Council Directive concerning urban waste water treatment (91/271/EEC) [21], 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 met before dumping sludge on the field, and these are regulated as well. The EU Council Directive of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC) [22] contains many regulations related to agricultural sewage sludge usage. 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 that has undergone biological, chemical, or heat treatment, long-term storage, or any other appropriate process so as to significantly reduce its fermentability and the health hazards resulting from its use.
In article 4, permissible concentrations of heavy metal are given:
Values for concentrations of heavy metals in soil to which sludge is applied, concentrations of heavy metals in sludge and the maximum annual quantities of such heavy metals which may be introduced into soil intended for agriculture are given in Annexes I A, I B, and I C.
The limit values for heavy metal concentrations in sewage sludge for use in agriculture are given in Annex I B (Table 1).

Table 1. Limit values for heavy metal concentrations in sewage sludge for use in agriculture taken from Annex I B of EU Council Directive on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture (86/278/EEC) [22].

Parameters	Limit values [mg/kg of dry matter]
Cadmium	20 to 40
Copper	1000 to 1750
Nickel	300 to 400
Lead	750 to 1200
Zinc	2500 to 4000
Mercury	16 to 25
Chromium*	-

* It is not possible at this stage to fix limit values for chromium. The Council will fix these limit values later on the basis of proposals to be submitted by the Commission within one year following notification of this directive.

EU directives do not indicate any limits for pathogen content in sewage sludge intended for agricultural use; EU member states establish such limits for themselves.

3.2. Permissible amounts of heavy metals in sewage sludge for agricultural use in Poland

Limits for heavy metal content in organic fertilizers and sewage sludge intended for agricultural use are established by Polish law in:

- Regulation of the Minister of Agriculture and Rural Development of June 18, 2008 on the implementation of certain provisions of the Act on fertilizers and fertilization [23],
- Regulation of the Minister of the Environment of February 6, 2015 on the use of municipal sewage sludge [24].

The Regulation of the Minister for Agriculture and Rural Development of June 18, 2008 on the implementation of certain provisions of the Act on fertilizers and fertilization says that *the permissible value of impurities in organic and organic-mineral fertilizers as well as organic and organic-mineral agents supporting plant cultivation may not exceed 100 mg of chromium, 5 mg of cadmium, 60 mg of nickiel, 140 mg of lead, 2 mg of mercury.*

The Regulation of the Minister of the Environment of February 6, 2015 on the use of municipal sewage sludge gives slightly different values, which are connected with different possible ways of utilizing this waste. These details are given in Annex 1 (Table 2).

Table 2. Permissible content of heavy metals required for sewage sludges for agricultural and land reclamation purposes by the Polish law under the Regulation of the Minister of the Environment of February 6, 2015 on the use of municipal sewage sludge [24].

Metal	Content of heavy metals in mg/kg of dry matter when used municipal sewage sludge not higher than		
	in agriculture and for land reclamation for agricultural purposes	for land reclamation for non-agricultural purposes	when adapting land to specific needs resulting from waste management plans, zoning plans, or decision on the conditions of development and land development, for growing crops intended for compost production, for growing crops not intended for consumption and feed production
Cadmium	20	25	50
Copper	1000	1200	2000
Nickel	300	400	500
Lead	750	1000	1500
Zinc	2500	3500	5000
Mercury	16	20	25
Chromium	500	1000	2500

3.3. Permissible amounts of parasites and bacteria in sewage sludge for agricultural use in Poland

In Poland, sewage sludge is allowed for agricultural use under certain circumstances. The Act of December 14, 2012 on waste [25] says that:
Recovery based on the use of municipal sewage sludge:
1) in agriculture, 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 plans 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.
And also says:
The use of municipal sewage sludge is possible if they are 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 July 16, 2015 on the admission of waste for storage in landfills [3] imposed restrictions on landfilling some types of waste. Annex 4 defines conditions for storage of waste other than hazardous and inert at a landfill, where it is given that amount of total organic carbon cannot exceed 5% of total solid, which disqualifies sewage sludge. Thus, the only method of disposal, other than incineration, is agricultural use or land reclamation. In Poland, any form of organic fertilizers (sewage sludges, biogas plant digestate, manure, *etc.*) must meet the restrictions referring to pathogens occurrence. The current Polish regulations with these restrictions are the same as previously mentioned.

In the first regulation [23] it can be found that:

Fertilizers and agents supporting the cultivation of plants referred to in paragraph 1 must not contain: viable ova of intestinal parasites Ascaris sp., Trichuris sp., Toxocara sp., Salmonella bacteria.

In the case of fertilizers referred to in paragraph 5 point 7, in addition to meeting the requirements set out in paragraphs 1 and 2, the number of bacteria of the Enterobacteriaceae family, determined on the basis of the number of aerobic bacteria, should be less than 1000 colony-forming units per gram of fertilizer.

The second regulation [24] according to what was given in the first one says:

§ 2. 1. Municipal sewage sludge may be used on land if the following conditions are met: (…) in the case of using these sludge in agriculture and for land reclamation for agricultural purposes – Salmonella bacteria have not been isolated in a representative sample of sediments weighing 100 g obtained in accordance with § 5 section 3;

3) the total number of viable ova of the intestinal parasites Ascaris sp., Trichuris sp., Toxocara sp. in 1 kg of dry matter, called hereinafter "d.m.", of sludge intended for applied research:

a) in agriculture and for land reclamation for agricultural purposes – is 0,

b) for land reclamation – no more than 300,

c) to adapt land to specific needs resulting from waste management plans, zoning, or zoning decision – not more than 300,

d) for the cultivation of plants intended for the production of compost – not more than 300,

e) for the cultivation of plants not intended for consumption and for the production of fodder – is not more than 300.

4. IONIZING RADIATION

4.1. Linear energy transfer, radiation chemical yield and dose absorbed

Ionizing radiation is the emission of different types of particles: alpha (particles consisting of two protons and two neutrons), beta (electron), and gamma (photon) with energy enough to ionize the medium through which these particles are passing. For a single act of ionization, UV light energy is enough (several eV), but particles with energy exceeding 0.1 keV can cause multiple ionizations [26]. These particles, which differ in radius and mass, while penetrating medium, are losing their energy as a result of inelastic collisions with electrons and nuclei. Energy loss is different for each of these particles; alpha radiation loses its energy after a very short travel distance, causing a high concentration of ionizing products along the particle travel track, while gamma photons can penetrate deep, producing a lower concentration of reaction products along their track. To quantify the energy loss of particles, a parameter called linear energy transfer (LET) has been introduced, which is expressed as energy loss per length unit. LET units can be keV/m or eV/nm. The effects of irradiation in a water environment can be quantified using a parameter called radiation chemical yield (G), also known as G-value, which says about a quantity of produced or decomposed substance per energy unit. G-value is typically expressed in mol/J (the previous G-value unit was number of molecules per 100 eV). Another physical quantity used to express the interaction of ionizing radiation with matter is absorbed dose (D), which is the mean energy deposited in the irradiated medium per mass unit, as in the below equation:

$$D = \frac{dE}{dm}$$

SI unit for absorbed dose is 1 gray (Gy), which is 1 J/1 kg [27].

4.2. Water radiolysis: reactions and products

Ionizing radiation causes the ionization of water molecules, and the effect is the formation of cation radicals $H_2O^{\bullet+}$ and a free electron (reaction 2).

$$H_2O \rightarrow H_2O^{\bullet+} + e^-_{prehyd} \quad (2)$$

The cation radical as a very strong acid immediately (measured lifetime of $H_2O^{\bullet+}$ is 200 fs, 1 fs is 10^{-15} s) deprotonates in water with the formation of hydroxyl radical (reaction 3).

$$H_2O^{\bullet+} + H_2O \rightarrow OH^{\bullet} + H_3O^+ \quad (3)$$

Free electrons are hydrated by surrounding water molecules, forming solvated electrons (e^-_{aq}) – reaction 4.

$$e^-_{prehyd} + nH_2O \rightarrow e^-_{aq} \quad (4)$$

Along particle penetration tracks, there are areas where water radiolysis products occur. These areas are unevenly distributed throughout the whole track length and are called spurs. Spurs can form agglomerates called blobs (lower LET radiation, gamma, beta) or short tracks (higher LET radiation, alpha). Inside spurs, before diffusing into the bulk volume, water radiolysis products can react with water or with themselves, resulting in the production of secondary radicals or molecules (reactions 5-12) [27].

$$2\,e^-_{aq} + 2H_2O \rightarrow H_2 + 2OH^- \quad (5)$$
$$e^-_{aq} + OH^{\bullet} \rightarrow OH^- \quad (6)$$
$$e^-_{aq} + H_3O^+ \leftrightarrow H^{\bullet} + H_2O \quad (7)$$
$$e^-_{aq} + H^{\bullet} + H_2O \rightarrow H_2 + OH^- \quad (8)$$
$$H^{\bullet} + H^{\bullet} \rightarrow H_2 \quad (9)$$
$$OH^{\bullet} + OH^{\bullet} \rightarrow H_2O_2 \quad (10)$$
$$OH^{\bullet} + H^{\bullet} \rightarrow H_2O \quad (11)$$
$$H_3O^+ + OH^- \rightarrow 2H_2O \quad (12)$$

G-values of selected water radiolysis products are given in Table 3.

Table 3. Radiation chemical yields of selected water radiolysis products for low LET radiation [27].

Water radiolysis product	G [μmol/J]
$OH^{\bullet}$	0.28
e^-_{aq}	0.28
$H^{\bullet}$	0.062
H_2	0.047
H_2O_2	0.073
H_3O^+	0.28

4.3. Properties of water radiolysis products

The obtained hydroxyl radicals ($OH^{\bullet}$) are very strong oxidants, while hydrogen radicals ($H^{\bullet}$) and solvated electrons are very strong reductants. Thus, water environments can show oxidative and reducing properties after irradiation. The reduction potentials of the discussed species are given in Table 4.

Table 4. Reduction potentials for main water radiolysis products [27].

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

To obtain a reaction environment with only reducing or only oxidative properties, some specific substances can be added to the irradiated system that can selectively react with only reducing or only oxidative species. To scavenge solvated electrons in the system, nitrous oxide (N_2O) saturated solution can be used (reaction 13).

$$e^{-}_{aq} + N_2O \rightarrow N_2 + O^{\bullet-} \xrightarrow{H_2O} N_2 + OH^{\bullet} + OH^{-} \quad (13)$$

Solvated electrons are transformed into more hydroxyl radicals, increasing the $OH^{\bullet}$ yield to 0.56 mol/J. Hydrogen radicals do not react like that, but their contribution to the total amount of radicals in neutral solutions after irradiation is only about 10%. Hydroxyl radicals can be removed by the addition of tert-butyl alcohol ($(CH_3)_3C\text{-}OH$) – reaction 14.

$$OH^{\bullet} + (CH_3)_3C\text{-}OH \rightarrow H_2O + {}^{\bullet}CH_2(CH_3)_2C\text{-}OH \quad (14)$$

The obtained environment has reducing properties with the content of hydrogen radicals and solvated electrons with the yield of 0.062 and 0.28 μmol/J, respectively. Reducing primary transients can be scavenged with hydrogen peroxide (H_2O_2), producing additional hydroxyl radicals (reactions 15 and 16).

$$e^{-}_{aq} + H_2O_2 \rightarrow OH^{\bullet} + OH^{-} \quad (15)$$

$$H^{\bullet} + H_2O_2 \rightarrow OH^{\bullet} + H_2O \quad (16)$$

Adjusting pH can also determine the type of primary transients present in the system. Hydrogen radicals are in acid-base equilibrium with solvated electrons (reactions 17 and 18).

$$H^{\bullet} + OH^{-} \leftrightarrow e^{-}_{aq} + H_2O \quad (17)$$

or

$$e^{-}_{aq} + H^{+} \leftrightarrow H^{\bullet} \quad (18)$$

So under low pH conditions, $H^{\bullet}$ yield of even 0.34 μmol/J can be achieved. It is also possible to obtain systems with dominance of hydrogen radicals by irradiation of highly acidic H_2-saturated solutions (a pressure cell is needed due to low hydrogen solubility under normal pressure conditions), in which hydroxyl radicals react with hydrogen, resulting in the production of additive hydrogen radicals (reaction 19).

$$OH^{\bullet} + H_2 \rightarrow H^{\bullet} + H_2O \quad (19)$$

Hydroxyl radicals are in acid-base equilibrium with the oxide radical anion ($O^{\bullet-}$),

$$OH^{\bullet} + OH^{-} \leftrightarrow O^{\bullet-} + H_2O \quad (20)$$

so higher concetrations of $O^{\bullet-}$ can be achieved in basic solutions (reaction 20) [27-29].

4.4. Sources of radiation used in industry

Industrial and laboratory irradiation facilities designed for biomass hygienization, depending on preferences, can use different types of ionizing radiation to obtain the intended result. It can be achieved by introducing different sources of ionizing radiation, such as electron accelerators able to produce high-energy electron beams (EB) or isotopes producing gamma radiation.

4.4.1. Isotope gamma sources and gamma radiation properties

^{60}Co is a radioactive isotope of cobalt obtained by neutron activation of ^{59}Co. This isotope emits mostly gamma radiation and low-energy β-radiation. During decay, ^{60}Co transforms into ^{60}Ni by emitting electrons with an energy of 0.31 MeV (reaction 21).

$$^{60}Co \rightarrow {}^{60}Ni + e^{-} \quad (21)$$

During transition into the ground state, the ^{60}Ni nucleus emits two gamma photons: 1.1732 and 1.3325 MeV. In rare cases (0.12%), the energy of the emitted electron is 1.48 MeV, and then one gamma photon with an energy of 1.3325 MeV is emitted [30]. The ^{60}Co decay scheme is presented in Fig. 2.

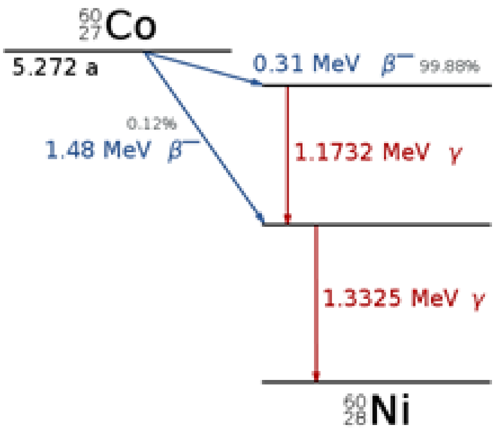


Fig. 2. ^{60}Co decay scheme [30].

The half-life of ^{60}Co is 5.27 years. This isotope can be used in laboratory gamma irradiators, medicine, food, or disposable medical equipment sterilization.
The radioactive cesium isotope ^{137}Cs is also used in medicine and industry. As a result of its decay, in 94.6% of cases, electrons with an energy of 0.512 MeV are emitted and a ^{137m}Ba isotope is obtained (reaction 22).

$$^{137}Cs \rightarrow {}^{137m}Ba + e^{-} \quad (22)$$

^{137m}Ba emits gamma photons with an energy of 0.6617 MeV, transferring into the stable ^{137}Ba isotope (reaction 23)

$$^{137m}Ba \rightarrow {}^{137}Ba + h\nu \quad (23)$$

In 5% of cases, ^{137}Cs decays directly into ^{137}Ba by the emission of a gamma photon with an energy of 1.174 MeV. The half-life of this nuclide is 30.17 years. The ^{137}Cs decay scheme is presented in Fig. 3.

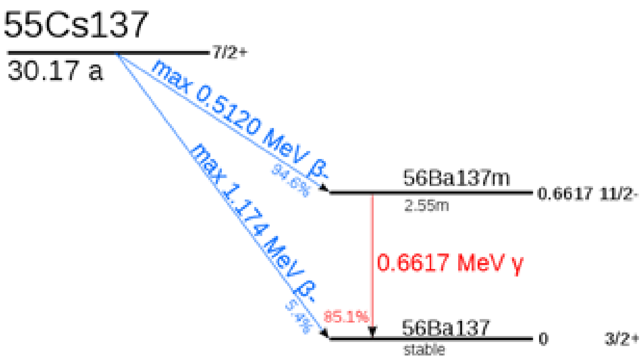


Fig. 3. ^{137}Cs decay scheme [31].

Because photons are neutral, they are not affected by atomic Coulomb forces, unlike charged particles, so gamma photons can achieve good penetration depth, which increases with photon energy [31]. On the other hand, for gamma sources, a dose rate is low, and a long period of irradiation is necessary to achieve the assumed dose.

4.4.2. Electron accelerators and electron beam properties

Another source of ionizing radiation are electron accelerators, which are electric devices that provide energy to electrons and operate in a similar way to old-type cathodic-tube TVs.

Electrons, as charged particles, undergo propelling when placed in an electric field, and this phenomenon is used in the operation of electron accelerators. Electrons are first emitted from the cathode in large amounts and then accelerated in a high vacuum to form a beam characterized by beam current, usually given in miliamperes (mA), and mean beam energy, usually given in multiples of electronvolts (eV, keV and MeV). There are different types of electron accelerators, varying in the method of generating the electric field, and thus in the energy, power, and efficiency of the beam. Accelerators, due to the method of operation, can be divided into: linear accelerators using microwaves and small concentric resonators (LINACs), resonant accelerators using high-frequency electromagnetic waves (about 100 MHz) or resonant cavity (radio frequency – RF accelerators), direct accelerators using a high potential difference and two electrodes (high voltage direct current – DC accelerators). LINACs allow to obtain high electron beam energy (5-10 MeV) required for high penetration depths, but their cost is high and their efficiency is low (about 10%). For industrial applications, DC and RF accelerators are usually preferred for their low cost and high efficiency (up to 50% for RF accelerators), but obtained EB energies do not exceed 5 MeV (usually 100 keV to 1 MeV), thus penetration depths are limited, while the higher the EB energy, the higher the penetration depth is. EB energies have to be high enough (more than 0.1 MeV) since too low energy will end up with inadequatly low penetration depth (Kurucz *et al.* claimed that 0.74 cm is the maximum penetration depth for 1.5 MeV EB in water) [32], but not too high, while EB energy exceeding 10 MeV may cause induced radioactivity of the irradiated material [33-35]. It is worth mentioning that the distribution of electron energy in the irradiated medium is not linear with the layer thickness. Electrons deposit their energy in the medium due to elastic and inelastic interactions with atomic electrons in this medium, so energy loss is dependent on the EB energy and the time the electrons spend nearby nuclei. The speed of electrons decreases with energy loss, so the time that

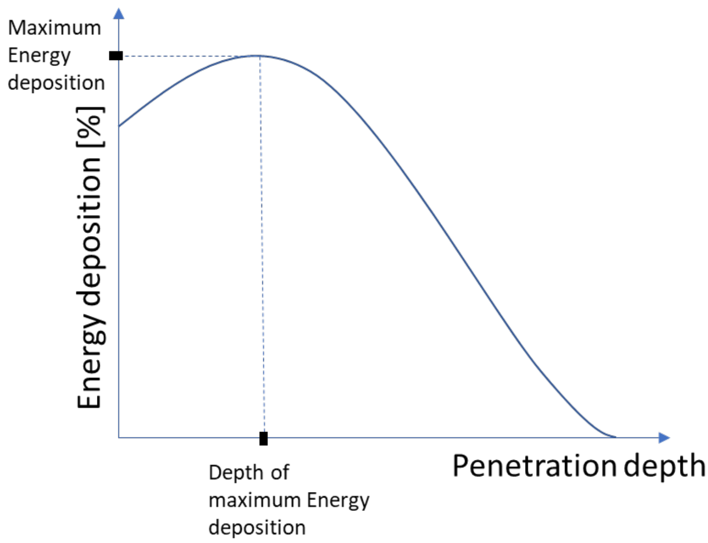


Fig. 4. Energy deposition of electrons in irradiated medium as a function of penetration depth (based on [35]).

electrons spend close to nuclei increases, which results in larger energy deposition. As the electrons energy loss is progressing, the total energy deposit in the irradiated medium is decreasing. As a result, EB energy deposition does not reach its maximum value at the surface of the medium, but at some depth, called the “depth of maximum dose”, after which energy deposition drops to 0 at the maximum penetration depth. A graph showing energy deposition of EB as a function of penetration depth is presented in Fig. 4 [35].

5. EFFECTS OF IONIZING RADIATION ON LIVING CELLS

5.1. DNA damages induced by ionizing radiation, direct and indirect effects

Ionizing radiation is lethal for living cells, and one of the reasons for this fact is that it damages 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. Mutation, which is the modification of genetic information, is another result of DNA damage. DNA strand damage can be caused by two separate mechanisms. The first is an indirect effect based on the chemical reaction of DNA with water radiolysis products, mainly hydroxyl radicals. Hydroxyl radicals interact with base (thymine, guanine, citosine, or adenine) where double bonds are formed, resulting in the formation of radicals that undergo further reactions with ring rearrangement as well as opening or reactions with molecular oxygen. $OH^{\bullet}$ radicals can also attack hydrogen atoms in the phosphate backbone of 2-deoxyribose, resulting in strand breaks. The second type of action is a direct effect when radiation ionizes DNA strands, transforming the bases into radical species that undergo further reactions. Such modified bases are no longer able to perform their previous function. Ionizing radiation can also ionize the sugar-phosphate backbone, leading to strand breaks [36, 37]. The scheme of direct and indirect effects is shown in Fig. 5.

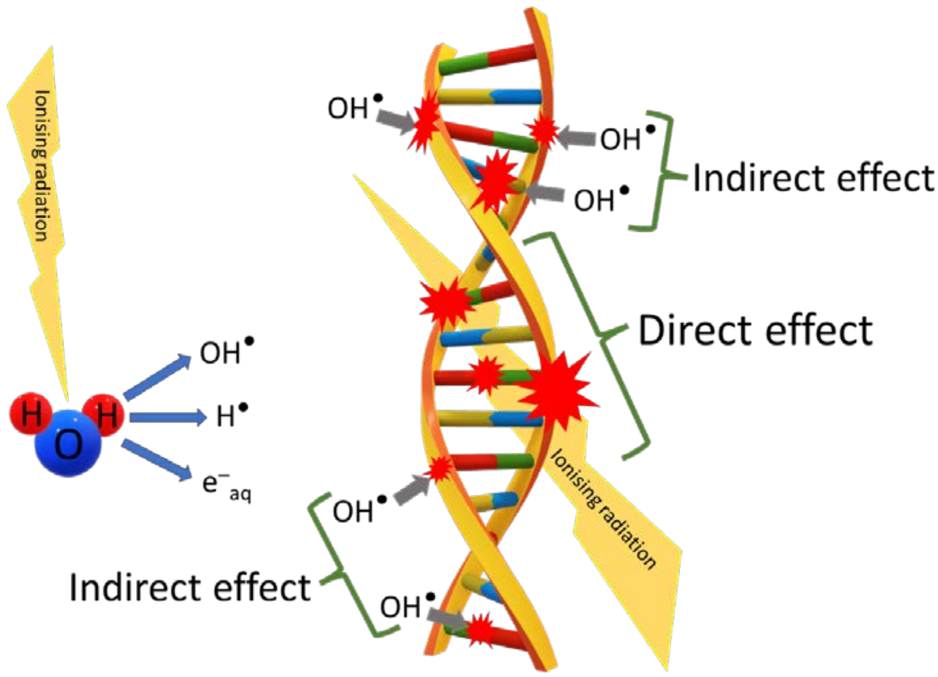


Fig. 5. Schematic drawing presenting the direct and indirect effects of ionizing radiation damaging a DNA molecule (based on [36]).

5.2. Lipids peroxidation

Living cells can be damaged by ionizing radiation not only by damaging DNA but also by causing chemical changes in lipids that create cell membranes. Similarly to DNA damage, cell membranes can be ionized directly by radiation or can react with water radiolysis products as water molecules surround cell membranes on both sides. $OH^{\bullet}$ radicals formed by water radiolysis can abstract hydrogen atoms from unsaturated lipid molecules, leading to the formation of carbon-centered radicals. Such a radical reacts rapidly with oxygen molecules (O_2), forming peroxyl radicals. Peroxyl radicals can then act as hydroxyl radicals and abstract hydrogen atoms

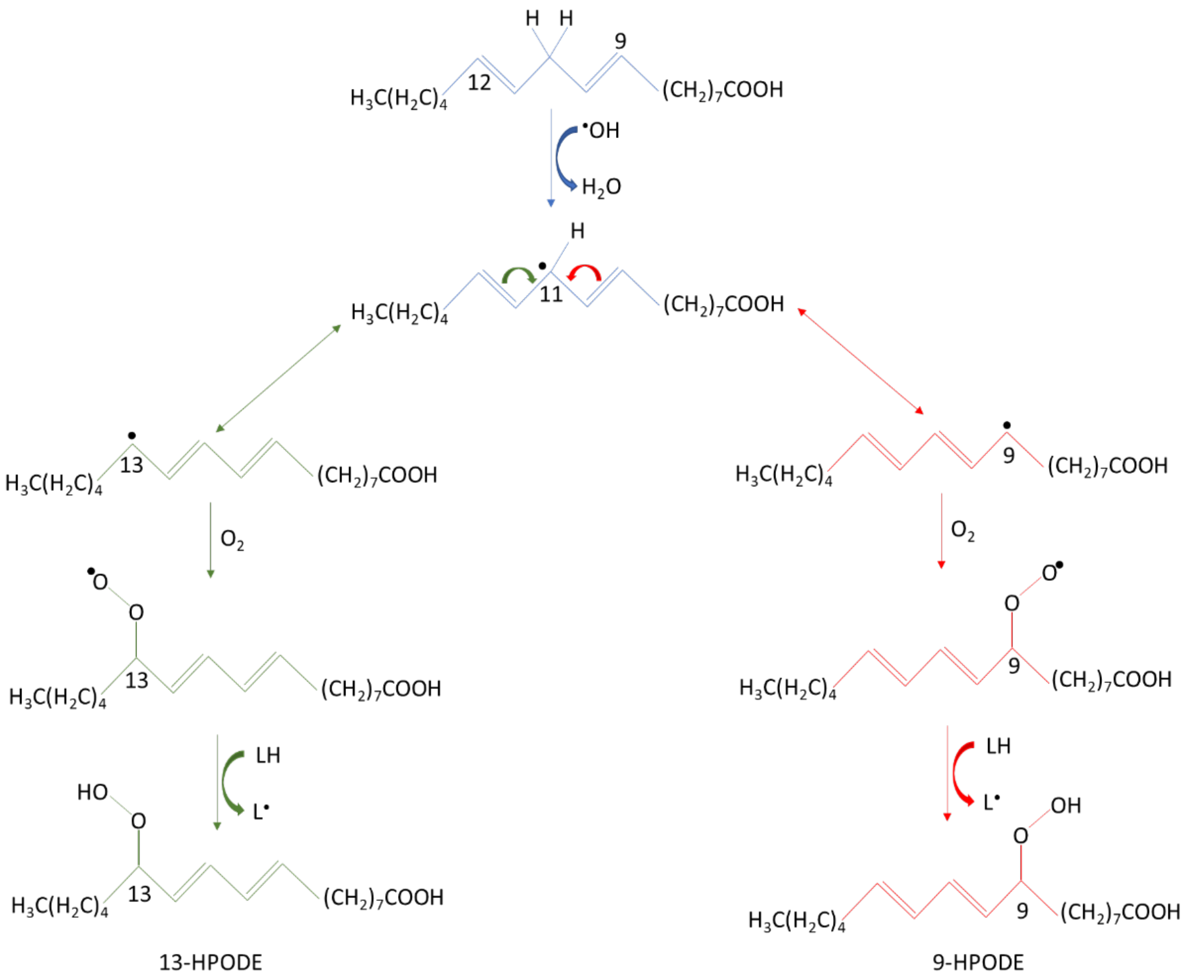


Fig. 6. Scheme of linoleic acid peroxidation initiated by hydroxyl radical and resulting in forming two optional compounds: 9 and 13-hydroperoxyoctadecadienoic acids. LH is another linoleic acid molecule (based on [38]).

from another lipid molecule, transforming themselves into lipid hydroxyperoxide. Because of such chain reactions, the presence of one hydroxyl radical may result in the formation of many radicals and peroxidized product molecules. These chain reactions can also be terminated by the recombination of radical products. A scheme of linoleic acid peroxidation as an example of the radical-chain mechanism of lipid peroxidation is shown in Fig. 6 [38].

6. THE EFFECTS OF IRRADIATION OF SEWAGE SLUDGE

6.1. Hygienization

Ionizing radiation, including electron beams and gamma rays, showed its effectiveness in pathogen removal from sewage sludge and municipal wastewater over the years of laboratory and industrial-scale tests. Asgari Lajayer *et al.* [39] experimented with gamma irradiation of effluent and sewage sludge from WWTPs in southern Teheran, Iran. A ^{60}Co source was used for irradiation and the dose range was 5-20 kGy. The presence of total coliform (TC) units and fecal coliform (FC) units was examined. Tests showed that for effluent, doses of 5-10 kGy reduced TC load by 99.66-100% and EC load by 99.62-100%, while for sewage sludge (TS = 28%), doses of 5-20 kGy reduced total and fecal coliforms by 99.1-99.96 and 96.32-99.72%, respectively. The authors claimed that higher doses needed to hygienize sludge result from higher TS content because solid particles may react with water radiolysis products, affecting indirect effects, and can also work as a shield against radiation for bacteria cells, affecting direct effects of radiation. Chmielewski *et al.* [40] irradiated thickened sewage sludge with 35% TS from municipal WWTP and tested samples for total bacteria content, spore-forming bacteria content, coliform counts, *Clostridium perfringens* counts, and ATT ova presence. The used dose range was 5-7 kGy. Total bacteria content was reduced by 2 logs after 5 kGy irradiation, 3 logs for 6 kGy, and 4 logs for 7 kGy. Spore-forming bacteria number was reduced by 1 log for 5 kGy and 2 logs for 6 kGy. Interestingly, coliform counts did not change after irradiation at 5 kGy, reduced by 2 logs at 6 kGy and by 3 logs at 7 kGy. *Clostridium perfringens* also stayed intact in the sample after 5 kGy treatment and was 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 an amount of 30 from the initial 90, and the total number of ATT ova was reduced from the initial 240 to 30. 6 kGy was enough to destroy all ATT ova. The doses required to remove pathogens appear to be high, but this could be due to the high TS content in the examined sewage sludge, as previously mentioned by the authors. Praveen *et al.* tested the effectiveness of 10 MeV EB treatment of sludges from aerobic and anaerobic treatment at two separate WWTPs in Texas, USA. Samples with indigenous *Escherichia coli* and aerobic and anaerobic spores were spiked with *Salmonella typhimurium*, somatic coliphages, male-specific coliphages, Polioviruses, and Rotaviruses. D_{10} was defined as an absorbed dose giving a 1 log reduction. For *Salmonella typhimurium*, D_{10} was 0.28 ± 0.01 kGy for aerobically digested sludge and 0.23 ± 1.01 kGy for anaerobically digested sludge; for *Escherichia coli* these values were 0.31 ± 0.01 and 0.25 ± 0.01 kGy; for aerobic spores 3.75 ± 0.24 and 4.04 ± 0.33 kGy; for anaerobic spores 4.96 ± 0.34 and 3.12 ± 0.38 kGy; for somatic coliphages 4.02 ± 0.38 and 4.07 ± 0.31 kGy; for male-specific coliphages 2.25 ± 0.19 and 2.45 ± 0.16 kGy, respectively. Poliovirus and Rotavirus were determined only in anaerobically digested sludge, and D_{10} was 2.07 ± 0.69 and 1.53 ± 0.03 kGy, respectively. Another experiment showed that *Salmonella typhimurium* and *Escherichia coli* are inactivated by EB; irradiation to about 1 kGy reduced *Escherichia coli* by 3.2 log for aerobic sludge and 3.8 log for anaerobically digested sludge, while for *Salmonella typhimurium*, this dose caused a reduction of 3.7 log for aerobic sludge and 4.5 log for anaerobically digested sludge [41]. Engohang-Ndong *et al.* used a 3 MeV electron accelerator to irradiate 15% TS primary sludge in a cascade system. Doses obtained during the experiments were: 2.7, 6.7, 13.2, 25.7, and 30.7 kGy. The number of viable *Ascaris* sp. ova and CFU (colony forming unit) counts per gram of TS of total heterotrophic bacteria (THB), total coliforms, and fecal coliforms were determined in the study. After irradiation sludge with a dose of 2.7 kGy, 93.3 ± 8.5% of THB, 21.1 ± 11.4 % of TC, and 67.2 ± 1.8% of FC of the initial number survived; for a dose of 6.7 kGy, these values were 31.0 ± 15%, 0.85 ± 0.23%, and 1.85 ± 0.65%, respectively. 8.9 ± 1.3% of the initial number of THB remained alive after 13.2 kGy treatment, and no TC or FC were found while irradiation with a dose of up to 25.7 kGy removed all tested bacteria. D_{10} values of 8.94, 3.16, and 3.17 kGy were determined for THB, TC, and FC, respectively. It was also mentioned that a dose of 6.7 kGy was sufficient to meet the requirements for A class sludge for agricultural use, which are 180 CFU of TC per 1 g TS. In the untreated sludge, *Ascaris* sp. viable ova number was 312 ± 24 per 4 g TS; 23 ± 8% of this amount were still alive after 2.7 kGy treatment, 11 ± 1.6% survived 6.7 kGy irradiation, 2 ± 0.03% were still viable after receiving a dose of 13.2 kGy. No viable *Ascaris* sp. ova were found after irradiation with a dose of 25.7 kGy. The estimated D_{10} value was 7.93 kGy [42]. Naing and Lay used a ^{60}Co gamma source to irradiate wastewater from Mandalay, Myanmar, sewage sludge samples (the authors did not mention TS content in the sludge). Initial bacterial counts were 24×10^7 CFU for wastewater and 38×10^8 CFU for sludge. Irradiation of wastewater with a dose of 1 and 2 kGy reduced bacteria counts by 2 and 4 logs, respectively. After 3 kGy irradiation, only 21 CFU were left, while after irradiation with a dose of 4 kGy, no bacteria were found. Bacteria counts in sewage sludge irradiated with a dose of 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, respectively. Treatment with a dose of 6 kGy left 23 CFU, and no bacteria were detected after 7 kGy irradiation. Lower irradiation doses required to remove bacteria from wastewater seem to confirm previous statements that higher TS content causes the deposition of part of the energy of radiation on solid particles instead of microorganisms cells, making the hygienization process more difficult [43].

6.2. Disintegration

Park *et al.* tested an EB treatment of sewage sludge (25 000 ± 2 000 mg/L TS) prior to the methane fermentation process using a 1 MeV electron accelerator. Irradiations were performed under various conditions: sludge layer thicknesses from 2.5 to 10 mm, exposure times from 0.3 to 1.2 s, and doses of 1, 3, 5, 7, 10, and 20 kGy. Measurements of soluble chemical oxygen demend (SCOD) showed a tremendous increase in this parameter after the irradiation up to 20 kGy; an increase of 49, 54, 97, and 147% were recorded after reducing sludge layer thickness from 10 mm through 7.5 and 5 to 2.5 mm. Such changes were caused by the limited penetration depth of the electron beam with an energy of only 1 MeV – for thicker layers bottom portions of sludge remained unirradiated or irradiated with lower dose than sludge portion located closer to the electron accelerator exit window. The total COD remained almost unchanged. The authors claimed that irradiation solubilized some of the organic compounds present in the solid phase of sludge. Experiments with exposure time changes with dose and layer thickness fixed showed no significant changes in SCOD. The authors tested the protein concentration in the liquid phase after irradiation and noticed an increase with dose increasing and layer thickness decreasing, which was explained by cell rupture caused by EB treatment. A decrease in volatile fatty acid (VFA) concentration after irradiation (by 66.7% for propionic acid after 20 kGy irradiation of a 5 mm thick layer) was also noticed. Tests with irradiated sewage sludge as anaerobic digestion reactor feedstock were carried out in mesophilic conditions, and enhancement in the hydrolysis process occurred, which was manifested by an increase in SCOD in comparison to reference reactor content measurements. Higher biogas yields for reactors fed with irradiated sludge were also observed [44]. Chu *et al.* irradiated sludge from the last step of the A^2/O process (the Bardenpho process) using a ^{60}Co gamma source. The sludge contained 1.1-1.4% TS, and doses used in the study ranged from 5 to 25 kGy. The authors noticed distinct changes after irradiation. Microscopic observations showed flock breakage and changes in filamentous bacteria cells, including rupture. The release of proteins and polysaccharides into the supernatant was also observed after gamma treatment. The measured values of total organic carbon, total nitrogen (TN), and total phosphorus (TP) also increased after treatment. TOC and protein concentrations were higher by 2 orders of magnitude at a dose of 25 kGy, and polysaccharide concentrations increased by 1 order of magnitude at the same dose [45]. Kim *et al.* studied the influence of gamma radiation on solubilization of waste activated sludge (WAS). The authors irradiated WAS from Jeongeup WWTP in South Korea with an average TS of 16 200 mg/L using a ^{60}Co gamma source. The irradiation doses used ranged from 0 to 50 kGy. SCOD, TCOD, biological oxygen demand (BOD_5), and extracellular substances (proteins, carbohydrates, and humic acid) were measured. While TCOD remained stable after 50 kGy irradiation, 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 increased from 160 to 787 mg O_2/L, which was explained by the hypothesis that gamma irradiation transforms non-biodegradable organics into biodegradable compounds. The BOD_5/SCOD ratio was calculated and the results showed an increase in that ratio for a dose of 10 kGy from 22.9% for the nontreated sample to 40.0% for the irradiated one. Nevertheless, further absorbed dose increase caused a decrease in this ratio value, and for 50 kGy, it was only 26.6%, so the authors concluded that the amount of non-biodegradable compounds converted into biodegradable ones is lower than the amount of compounds released into the water phase by irradiation. Carbohydrate and humic acid concentrations in the liquid phase increased with the increase of the absorbed dose and for 50 kGy, the results were: for carbohydrates – 85.7 mg/L from an initial 4.9 mg/L and for humic acids – 260.2 mg/L from an initial 22.4 mg/L. The protein concentration increased from 5.8 to 53.9 mg/L for 10 kGy, but irradiation with higher doses showed no further changes [46]. Shin and Kang worked with EB-irradiated WAS with 1.5% TS and 73% VS and thickened sludge (a mixture of WAS and preliminary sludge) containing from 2.4 to 3.2% TS as a biogas fermentation reactor feedstock. The authors used a 1 MeV electron accelerator to determine the effects of sludge irradiation on SCOD, pH, specific resistance to filtration (SRF), sludge volume index (SVI), alkalinity, protein, and carbohydrate concentrations in the liquid phase. The anaerobic digestion (AD) tests were also carried out in 18 L bioreactors in mesophilic conditions (35 ± 1°C) for 50 days to evaluate the influence of irradiation on organic compound conversion in the AD process. The sludge samples were irradiated with doses of 0.5, 1, 3, 6, and 10 kGy. An influence of EB treatment on pH was negligible for both types of sludge; samples were slightly more acidic for the 10 kGy dose. Alkalinity increased slightly for WAS and decreased slightly for thickened sludge with dose increase. TS and VS kept stable after irradiation, but SCOD, carbohydrates, and soluble protein concentrations changed dramatically after treatment. For WAS, the initial SCOD level was 52 mg O_2/L, but after irradiation with doses of 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 doses of 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-dose treatment, while after irradiation with higher doses, the SCOD increase is not so intense. A similar situation occurred when examining the influence of EB irradiation on carbohydrates and protein concentrations in the liquid phase. Doses of 0.5, 1, 3, 6, and 10 kGy increased protein content from initial 14.4 to 121.8, 240.1, 306, 379.1, and 397.3 mg/L for WAS and from initial 62.4 to 230.7, 235.4, 383.8, 469.2, and 559.9 mg/L for thickened sludge, respectively. EB treatment using the same dose set increased the concentration of carbohydrates in the supernatant from 5.9 to 92.2, 108.4, 110.3, 119.7, and 116.8 mg/L for WAS and from 17.1 to 152.0, 158.3, 197.0, 243.2, and 262.7 mg/L for thickened sludge, respectively. Again, the most significant results were obtained at lower doses. SVI, a parameter used to describe the settleability of treated sludge, decreased from 110-1160 to 60-70 mg/L. However, dewaterability worsened after EB treatment; for 10 kGy, the SRF parameter increased from 0.45×10^{16} to 2.24×10^{16} m/kg in the case of WAS and from 0.37×10^{16} to 2.27×10^{16} m/kg for thickened sludge. Anaerobic digestion studies showed a substantial increase in biogas yield when fermenting irradiated samples; for this experiment, only WAS and irradiation doses of 1, 3, and 6 kGy were used. The maximum biogas yield for a reactor with untreated sludge was 95 L/(m^3d) on day 15; for a 1 kGy reactor, it was 180 L/(m^3d) on day 15; for a 3 kGy reactor, it was 260 L/(m^3d) on day 15; and for a 6 kGy reactor, it was 290 L/(m^3d) on day 15, which resulted in a 189, 274, and 305% increase in biogas yield, respectively. Also, methane concentrations in biogas were slightly higher in reactors with irradiated samples. The VFA concentration on day 15 was higher by 107% for 1 kGy, 132% for 3 kGy, and 153% for 6 kGy in comparison to the reference reactor [47].

7. CONCLUSIONS

Large amounts of sewage sludge produced every year all over the world have a great potential as an energy source and soil fertilizer with its protein, carbohydrates, fats, nitrogen, phosphorus, and potassium content, and no use of it would be a tremendous waste contradictory to the circular economy philosophy. Nevertheless, the pathogen content of sewage sludge constitutes a problem that must be overcome. The use of ionizing radiation seems to be a remedy for this inconvenience, as it allows for the removal of pathogens with relatively low energy input, short treatment time, and high effectiveness. In particular, the electron beam is an appropriate tool as it is safe to maintain and has stable parameters for all exploitation periods. The dose range necessary to remove pathogens from sewage sludge is wide and depends on pathogen type (bacteria are more resistant to irradiation than helminth ova) and TS content in the sludge (for higher TS content, there are more molecules to react with water radiolysis products, more solid particles that are working as shields from irradiation, and more bacteria cells and helminth ova).

For sludges with lower TS content, several kGy doses are required to make them safe as organic fertilizer, while higher concentrations of solid particles in sludge make it necessary to use doses exceeding 15 kGy. The EB penetration depth should be also taken into consideration, as it is limited and depends on electron beam energy and density. Ionizing radiation treatment can also be used to increase the efficiency of the anaerobic digestion process. Disintegration of feedstock induced by irradiation causes the increase in SCOD and sewage sludge flocks to rupture, making the hydrolysis step of the methane fermentation process easier. An increase in the dose is followed by a SCOD increase, but the greatest changes are noted for lower doses not exceeding 4-5 kGy.

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8. LITERATURE

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