


Review

# Application of Ionizing Radiation in Wastewater Treatment: An Overview

Rehab O. Abdel Rahman <sup>1,\*</sup>  and Yung-Tse Hung <sup>2</sup>

<sup>1</sup> Hot Lab. Center, Atomic Energy Authority of Egypt, Cairo P.O. No. 13759, Egypt

<sup>2</sup> Department of Civil and Environmental Engineering, Cleveland State University, Cleveland, OH 44115, USA; yungtsehung@gmail.com

\* Correspondence: alaarehab@yahoo.com; Tel.: +2-010-6140-4462

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**Abstract:** Technological applications of nuclear science and technology in different sectors have proved their reliabilities and sustainability over decades. These applications have supported various human civilization needs, ranging from power generation to industrial, medical, and environmental applications. Environmental applications of radiation sources are used to support decision making processes in many fields; including the detection and analysis of pollutant transport, water resources management, and treatment of municipal and industrial wastewaters. This work reviewed recent advances in the research and applications of ionizing radiation in treating different wastewater effluents. The main objective of the work is to highlight the role of ionizing radiation technology in the treatment of complex wastewater effluents generated from various human activities and to address its sustainability. Results of both laboratory and industrial scale applications of this treatment technology have been reviewed, and information on operational safety of industrial irradiators, which affect the sustainability of this technology, has been summarized.

**Keywords:** ionizing radiation; agricultural effluents; dye treatment; pharmaceutical effluents; disinfection

## 1. Introduction

Providing clean water and sanitation is one of the sustainable development goals that were proposed by the United Nation (UN). One of the problems that affect this goal is the reduction of freshwater quality. This reduction is attributed to the continuous increase in untreated wastewaters volumes and poor management practices, which led to the introduction of hazardous materials into freshwater sources [1]. Wastewater is defined, as indicated in UNEP/UN-Habitat, as a combination of one or more of the following effluents: domestic, commercial, industrial, horticultural, aquaculture, and storm water [2]. Recent advances in wastewater management helped in addressing some of the problems in water supply, pollution control, water recycling, and environmental protection. Now wastewaters are proposed as a resource, where many wastewater treatment plants are operated by the biogas generated from the anaerobic digestion of sludge, and the effluents from these plants could be used after appropriate treatment to meet the industrial, agricultural, and potable water requirements. Currently, wastewater management in developing countries is characterized by the discharge of large quantity of wastewater into surface water bodies without proper treatment. It is also challenged by the difficulties to sustain financing, operating and maintaining infrastructure for wastewater treatment.

Conventional wastewater treatment plants (WWTP) aims at reducing the contamination levels to acceptable limits required by the national regulatory agencies and at complying with international guidelines, which allow its safe discharge or reuse. Multi-stages of treatment processes are used, where pre-treatment stage is applied to remove coarse and large solids from the waste stream, using physical treatment technologies such as screens and grit chambers [3]. Primary treatment methods

are then applied to remove suspended solids that could be settled using gravity sedimentation with or without coagulation and flocculation [3,4]. The effluent from the primary treatment is directed to the secondary treatment stage to remove residual suspended solids and organic materials by using biological treatment processes [3]. The effluents from secondary stage contain some heavy metals, synthetic bio-refractory organic pollutants, and soluble microbial products derived during biological treatment [5,6]. The synthetic bio-refractory organic pollutants may include emerging micro-pollutants and disinfection by-products. In the tertiary stage of treatment, the effluents from the secondary stage are polished by removing persistence organic containments and heavy metals using advanced wastewater treatment technologies [3,4,7]. These technologies include filtration, sorption, gas stripping, ion-exchange, advanced oxidation processes (AOP), and distillation [7]. Finally, disinfection could be applied depending on the potential use of the treated effluents and the effluents characteristics [3].

Regulations on discharge/reuse indicators varied from country to another. In Denmark, Belgium, Spain, Germany, France, and Netherlands, the regulation covers heavy metals concentrations, total suspended solids (TSS), chemical oxygen demand (COD), 5 days biochemical oxygen demand (BOD<sub>5</sub>), total nitrogen (TN), total phosphorus (TP), and quantity of effluent discharge [8]. On the other hand, toxicity is only considered as reuse/discharge indicator in Germany, France, and United Kingdom [8]. Table 1 lists some international guidelines on the maximum limits for the reuse of effluents containing heavy metals, and U.S. regulation for their reuse and discharge [9–12]. Table 2 presents effluent discharge and reuse quality requirements in USA, Canada, and EU [11,13–15]. It should be noted that three classes of effluents are listed in the Canadian regulation based on the degree of treatment processes used in the treatment plants, namely, A, B and C, that refers to effluent from tertiary and disinfection treatment, tertiary treatment, and secondary treatment, respectively. For reuse regulations in EU, the limit of each class is determined based on the processes used in the treatment plants, irrigation methods, and the type of crops. Class A represents effluents from tertiary and disinfection treatment used any irrigation method and to produce all food crops. Classes B and C are effluent from secondary treatment and disinfection, which are used to produce food and processed food and non-food crops using all irrigation methods, and drip irrigation, respectively. Finally class D in EU regulation represents effluents from secondary treatment and disinfection, where all irrigation methods are allowed to produce industrial, energy, and seed crops [13]. Discharges from un-complied conventional WWTP can lead to the introduction of persistent chemicals and eco-toxic micro-pollutants into the aquatic systems [16]. The incomplete removal of these containments (even in  $10^{-9}$ – $10^{-6}$  g/L concentration range) was reported to induce potential long-term detrimental impact on the environment and the human health [17]. Recent research studies reported in the literature supports the application of advanced oxidation processes (AOP) for wastewater treatment to remove these contaminants [16,17].

**Table 1.** Guidelines of maximum limits for discharge and reuse of some inorganic pollutants in treated wastewater.

	WHO, Reuse, ppb [11]	FAO, Reuse, ppb [12]	US, ppb [9]			WHO, Reuse, ppb [11]	FAO, Reuse, ppb [12]	US, ppb [9]	
			Discharge	Reuse				Discharge	Reuse
As	50	100	3000	50	Hg	1	NA	2000	2
Cd	5	100	15,000	5	Ni	0	200	12,000	100
Cr	NA	100	10,000	50	Ag	NA	NA	5000	50
Cu	1	200	15,000	NA	Zn	5000	200	25,000	5000
CN	NA	NA	10,000	5.2	pH	6.5–8.5	6.5–8.4	6–9	
Pb	50	5000	40,000	50					

NA: Not Available.

**Table 2.** Effluent reuse and discharge limits in different regulations [11,13–15].

Indicator	Reuse				Discharge				
	EU [14,15]				EU [14,15]	US [11]	Canada [13]		
	A	B	C	D			A	B	C
BOD <sub>5</sub> , ppm	10			25		45	10	10	45
TSS, ppm	10			35		45	10	10	45
COD, ppm	NA	NA	NA	NA	125	NA	NA	NA	NA
E. coli, (CFU/100 mL)	10	10 <sup>2</sup>	10 <sup>3</sup>	10 <sup>4</sup>	NA	2.2	2.2	400	NA
Turbidity (NTU)	5	NA	NA	NA	NA	2	2	NA	NA
TN, ppm	NA	NA	NA	NA	15	10	20	NA	NA

NA: Not Available.

AOP use in-situ generated hydroxyl or sulfate radicals for organic pollutants degradation and heavy metals toxicity reduction [5,18,19]. AOP convert synthetic organic pollutants and soluble microbial products into simple biodegradable and harmless products, which lead to the reduction of COD and BOD in the treated effluents. AOP technologies employ various activation methods, where induced oxidation is achieved via exposure to photochemical or ionizing radiations, or chemicals [18,20,21]. Several studies were conducted to assess the feasibility of using ionizing radiation, in the form of gamma ( $\gamma$ ) rays or electrons ( $e^-$ ), to remove persistence contaminants and to disinfect treated water and sludge. The results of these studies indicated that the ionizing radiation treatment is technically and economically promising. The efficiency of this technology was also proved in the reduction of persistence heavy metals under different treatment conditions. Fifteen pilot plants and several full scale irradiation treatment facilities that employ electron accelerator or  $\gamma$  irradiators were established with varying capacities. International Atomic Energy Agency (IAEA) reported that there is a need to continue research in this area to increase the general awareness of these processes in the environmental engineering community [22]. This awareness could be achieved by presenting integrated reviews on the advances in ionizing radiation applications in industrial effluents treatment from technical, operational safety, and economical aspects. Some review papers were published that reviewed the role of ionizing radiation in the degradation of azo dyes [23], summarized the results of the IAEA coordination research project [24], and have reported the optimized doses and procedures [25]. There is a lack in review papers that summarize recent advances in this field and that provide integrated insights into the role of this technology in eliminating hazardous biodegradation and disinfection products. In this work, the effort is directed to summarize the current understanding of the decomposition and removal mechanisms for organic and inorganic pollutants, respectively. Principles and advances in investigating the scientific basis of the applicability of this technique in the treatment and disinfection of agriculture, dyes, pharmaceutical, and petrochemical effluents will be presented. Operational safety and economical factors that affect the sustainability of this technology will be summarized. Finally, knowledge gaps will be identified and research areas that need to be addressed will be highlighted.

## 2. Decomposition and Removal Mechanisms

Generally any emitted radiation is characterized by its ability to deposit some of its energy in the surrounding media. This energy excites the media atoms by stripping their electrons or break the chemical bonds between its molecules [26]. This particular characteristic leads to the application of ionizing radiation in treating wastewater, where the radiation imparts some of its energy in the radiolysis of water molecules. The effectiveness of the irradiation process is evaluated by calculating

the radiation chemical yield (G-value,  $\mu\text{mol}\cdot\text{J}^{-1}$ ) that quantifies the number of formed species due to the absorption of 100 eV and is given by:

$$G = \frac{6.023 \times 10^{23} C}{D \times 6.24 \times 10^{16}} \quad (1)$$

where C is the formed species concentration (mol/L) and D is the absorbed dose (Gy). Figure 1 illustrates the two stages radiolysis process for water molecules and its corresponding time scale, radiolysis products (primary intermediates), and their G-values. Primary intermediate (PI) then reacts with the pollutants (A) leading to the formation of secondary intermediates (SI) that are less persistence in a bi-molecular reaction to produce the degradation products (C + D), as follows [25,27,28]:

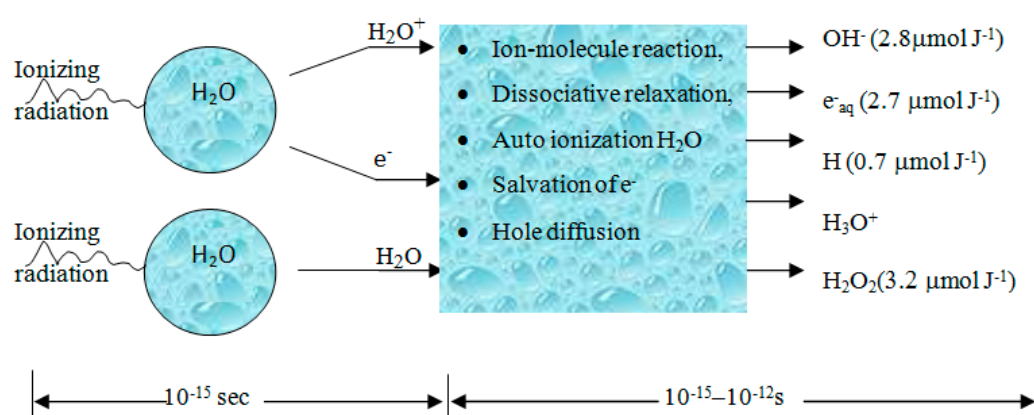


Figure 1. Two stages radiolysis process.

In actual wastewater, there is a competition between the pollutants and the anions in the solution, i.e.,  $\text{Cl}^-$ ,  $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ , for the reaction with the primary intermediates. This competition can affect the efficiency of the overall treatment process [27]. Gehringer studied the competition kinetics of two pollutants (A, B) in wastewater. The reaction rate was attributed to the bi-molecular rate constant ( $k_A$ ) between the solute concentration ( $[A]$  or  $[B]$ ) and the primary intermediates concentration  $[\text{OH}]$  as follow:

$$-\frac{dA}{dt} = k_A[A][\text{OH}] \quad (3)$$

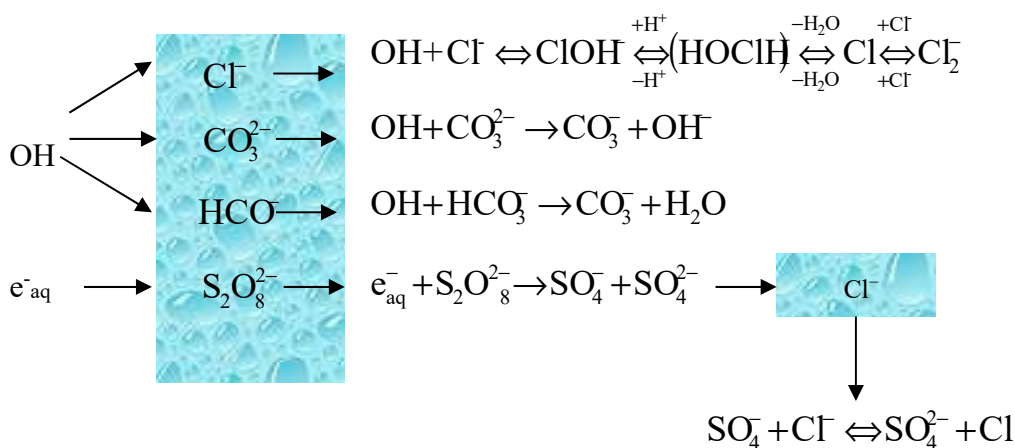
Table 3 lists the bi-molecular reaction rate constants for some pollutants [22,25,27,29]. The amount of the specific radicals available for interaction with certain solute could be calculated using the reaction probability ( $P_A$ ) according to the following equation:

$$P_A = \frac{k_A[A]}{k_A[A] + k_B[B]} \quad (4)$$

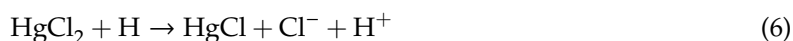
Figure 2 illustrates the reactions between the primary intermediates and the available anions in the wastewater effluents. Wojnarovits and Takacs [30] indicated that intermediate reaction with chloride ions is dominant at  $\text{pH} < 5$ , whereas the reaction with carbonate and bicarbonate ions are dominant at neutral or slightly alkaline pH. The produced free radicals will subsequently react with dissolved organic matters in the wastewater via direct electron transfer (outer sphere electron), addition to double bonds (inner sphere electron) or abstraction of H-atoms from C-H bonds. The last reaction takes place between oxidizing radicals and saturated molecules. It should be noted that in some cases, it is very hard to determine if the reaction is electron transfer or radical addition/elimination.

**Table 3.** Bimolecular reaction rate constants for some pollutants [22,25,27,29].

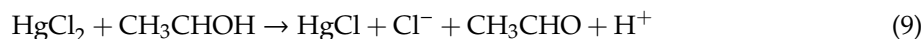
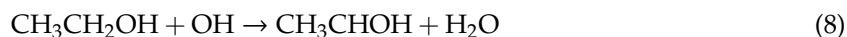
Compound	Rate Constant, $M^{-1} s^{-1}, \times 10^9$			Compound	Rate Constant, $M^{-1} s^{-1}, \times 10^9$		
	OH	$e^-$	H		OH	$e^-$	H
Perchloroethylene	1.7	13		Bicarbonate	0.0085		$4 \times 10^{-3}$
Trichloroethylene	2.6	19		Nitrogen		9.7	
Dichloroethylene	7	7.5		Oxygen		19	
Vinylchloride	12	0.25		Methyl t-butyl ether	2	0.018	
1,1,1-Trichloroethane	0.04			Ethyl t-butyl Ether	1.8	0.01	$7 \times 10^{-3}$
Chloroform	0.005	30		Diisopropyl Ether	2.5	$7 \times 10^{-3}$	0.067
T-Amyl Methyl Ether	2.4	$3 \times 10^{-3}$	$3 \times 10^{-3}$	Acetylenes	0.1–1		
Alcohols	0.1–1			Aldehydes	1		
Alkanes	0.001–1			Aromatics	0.1–100		
Carboxyl Acids	0.01–1			Ketones	1		
Organo-Nitrogen	0.1–100			Olefins	1–10		
Phenols	1–10			Organo-Sulfure	1–10		
Trichloronitromethane (TCNM), Chloropicrin	$0.0497 \pm 0.28$	$21.3 \pm 0.03$		Carbofuran	6.6		
Benzene	7.8			Carbendazim	2.2		

**Figure 2.** Schematic diagram of intermediate reaction with free radicals.

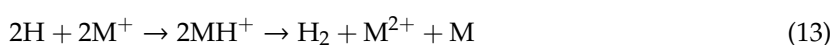
Ionizing radiation can reduce various forms of mercury via reaction with  $e^-_{aq}$  (Equation (5)) and H (Equation (6)) to form unstable compound that dimerized (Equation (7)) to produce insoluble form [31,32].



The presence of hydroxyl radicals inhibited the production of insoluble mercury ( $Hg_2Cl_2$ ) through re-oxidation of  $HgCl$  [31]. To enhance the removal performance of this technology the use of organic radical to act as hydroxyl scavenger was proposed. For example, ethanol may be used (Equations (8) and (9)) with the following reactions [9]:



Successive reduction of Cr species from (VI) state to (III) state is achieved via reaction with H [32]. On the other hand, cadmium (II) and lead (II) are reduced via reaction with H or/and  $e^-_{\text{aq}}$  (Equation (10)) to produce Cd(I) that undergoes disproportionation (Equation (11)) or oxidation through a reaction with  $\text{OH}^-$  or  $\text{H}_2\text{O}_2$  (Equation (12)), or react with hydrogen to produce unstable  $\text{MH}^+$  species that decay (Equation (13)) [18].



To reduce the oxidation effect and subsequently to enhance cadmium and lead precipitation, an organic  $\text{OH}^-$  scavenger could be used or the process could be operated in the absence of oxygen [31–33].

### 3. Advances in Treating Agricultural Wastewaters

Despite chemical pesticides, herbicides, and fungicides are applied according to national agricultural guidelines to enhance the agricultural production efficiency. Residues of these persistence pollutants and their toxic byproducts exist in agricultural wastewater, and could migrate to surface-and ground-waters. Ionizing radiation treatment technology proved its effectiveness in decomposing a varied number of these pollutants using tertiary treatment method. Most of the researches conducted in this area focused on optimizing the irradiation conditions, investigating the effect of other waste components on the degradation process, and the possibility of using combined treatment technologies to enhance the overall efficiency of the treatment process. Table 4 summarizes the performance of ionizing radiation in removing some pollutants in agricultural wastewaters.

**Table 4.** Performance of agricultural wastewater pollutants degradation using irradiation.

Pollutant	Origin	Degradation Performance	Initial Contaminant Concentration, ppm	Dose, KGy	References
Polychlorinated Biphenyls (PCB)	Pesticide	96%	NA	<0.1	[34]
Alkali Halides	Herbicides, fungicides, insecticide	98%	100	1	[25]
Trihalomethanes	Disinfection by products	87.4%	NA	2	[25]
		95%	145–780	6	[35]
		87.8%	264	8	[35]
Nitrophenols	Degradation products		0.139	5	[25]
Carbofuran, Dimethoate, Imidacloprid, Prochloraz	Pesticide	99%	50, (pH 5.5)	5	[36]
Methiocarb	Pesticide	67%			[36]
2,4-Dichlorophenol	Pesticide	Complete degradation	50	10	[22]

NA: Not Available.



The optimal irradiation conditions for six commercial pesticides, i.e., diazinon, dimethoate, prochloraz, metiocarb, imidacloprid, and carbofuran, using electron-beam facility, were determined for different wastewater compositions and at different operational conditions [36]. The initial pesticide concentrations ( $C_i$ ) varied in the range  $40 < C_i < 400$  ppm, the initial pH in the range ( $4.5 < \text{pH} < 8$ ) and the irradiation doses were varied from 2–10 KGy. The application of 5 KGy was found to be the most efficient for the treatment of prochloraz, dimethoate, imidacloprid, and carbofuran (99% degradation), but not efficient for the treatment of metiocarb. Low irradiation doses ( $<1$  KGy) led to the formation of hydroxylated intermediates that build-up in the solution with the progress of the irradiation process. By increasing the irradiation period, a complete removal of the pollutants is achieved as a result of hydroxylation of the accumulated intermediates. The effect of the combined process of aeration and irradiation treatment was evaluated. It was found that the use of aeration can improve the metiocarb removal by 18%. In general, the hydroxylation of the aromatic ring to produce hydroxyl-cyclohexadienyl radicals is reversible [37]. Using aeration to inhibit the formation of these reversible radicals by peroxidation will lead to more efficient degradation [37]. The optimum irradiation conditions for the degradation of three herbicide and one fungicide, i.e., 2,4-dichlorophenoxyacetic acid (2,4-D), 3,6-dichloro-2-methoxy-benzoic acid (dicamba), 4-chloro-2-methylphenoxyacetic acid (MCPA), and carbendazim were determined. It was found that complete radiolytic degradation of dicamba ( $C_i = 110$  ppm) is achieved using an irradiation dose of 5 KGy and the process is insensitive to the presence of other waste constituents, i.e.,  $\text{NO}_3^-$ . 2,4-D-degradation was found to be affected by the presence of other waste constituents [35]. Another study addressed the use of ionizing radiation in the treatment of 2,4-D, and MCPA at lower concentration levels ( $<50$  ppm), in the presence  $\text{Cl}^-$ ,  $\text{Br}^-$  and  $\text{NO}_3^-$ . The results confirmed the role of oxidative radicals ( $\text{OH}^\cdot$ ) in the degradation of these pollutants and the sensitivity of the irradiation process to the presence of the anions, where the secondary intermediate formed at low doses ( $<1$  KGy) are more toxic than the original pollutants [38,39].

The feasibility of using ionizing radiation in the treatment of agricultural effluents containing chlorinated organic pesticide, i.e., (4-chloro phenoxyacetic acid (4-CPA), 2,4-dichlorophenoxyacetic acid (2,4-D), 2,4-dichlorophenoxyacetic propionic acid (2,4-DP), and 2,4-dichlorophenoxyacetic butanoic acid (2,4-DB) was studied. The results indicated good efficiency of the irradiation process at 1 KGy in decomposing these pollutants. It was observed that chlorine was released as a result of their degradation. To reduce the disinfection byproducts in the treated wastewater prior to its use in chicken and fish livestock, an irradiation of the wastewater stream at 16.2 KGy dose was proposed, and 27 KGy was proposed for sludge treatment [40].

Swine wastewaters, which are alkaline agricultural wastewaters, that contain different pollutants, i.e., carbohydrates, proteins, lipids nitrates, nitrite, phosphate, and ammonia, were treated using a combined process of irradiation and ion-exchange biological treatment method. The use of electron-beam at 75 KGy achieved 85.1% removal efficiency of chemical oxygen demand (COD) at organic loading rate of  $1.41 \text{ kg/m}^3 \cdot \text{day}$  and achieved 75% removal efficiency of total nitrogen. The nitrogen removal was found to be sensitive to variation in current density [41].

#### 4. Advance in Dyes Treatments

The presence of organic dyes in industrial effluents can lead to serious health and environmental problems. Ionizing radiation treatment was proposed to remove these contaminants. Research efforts were directed to study model compounds, single polluted solution, simulated wastewater, and real effluents. Table 5 summarizes the kinetic reactions of model compounds with their primary intermediates, their corresponding rate coefficients and their secondary intermediates [23]. Most of these compounds, except Azobenzene, decompose as a result of hydroxylation reaction, where the rate coefficients are slightly varied depending on the molecular structure of the contaminant. On the other hand, reactions with  $e^-$  and  $\text{H}$  have small contribution to the decomposition process [23]. G value for phenol degradation was found to be inversely proportional to the irradiation dose. Degradation reaction is first order and favors neutral pH. Phenol degradation is enhanced with the addition of

oxidants, i.e.,  $O_3$ , or  $S_2O_8^{2-}$ .  $S_2O_8^{2-}$  was more efficient due to the selectivity of  $SO_4^{\cdot-}$  radical to the formed by-products (carboxylic acids) [42].

**Table 5.** Degradation kinetics of model compounds [23].

Model Compound	Primary Intermediate	Rate Coefficient, $\times 10^{10} \text{ mol}^{-1} \cdot \text{L} \cdot \text{s}^{-1}$	Secondary Intermediate
Aniline	$OH^{\cdot-}$	0.86–1	-
	$H^+$	0.2	Anilino radical (directly and after water elimination.)
Phenol	$OH^{\cdot-}$	0.66–1.4	Cyclohexadienyl-type radical
	$H^+$	0.17	
Atrazine	$OH^{\cdot-}$	0.24	
Azobenzene	$e^-$	1–3.3	Hydrazyl radicals
	$OH^{\cdot-}$	2	
4-amino-5-hydroxynaphthalene-2,7-disulphonic acid	$OH^{\cdot-}$	-	Anilino-type

65% de-coloration of aqueous solution containing Alizarin Yellow GG (AY-GG,  $C_i = 100$  ppm) was achieved using 9 KGy dose. Irradiation pre-biodegradation was proposed to improve the efficiency of treatment by 30% due to the formation of heterocyclic aromatic amines and cyanides. Increasing the irradiation dose can enhance the biodegradation due to the elimination of toxic secondary intermediates [43]. The aqueous solutions of Reactive Blue 15 (RB15) and Reactive Black 5 (RB5) dyes were irradiated with doses 0.1–15 KGy and at 2.87 and 0.14 KGy/h dose rates. Complete de-coloration was observed at 1 and 15 KGy doses for RB5 and RB15, respectively [25]. The de-coloration mechanisms for Apollofix Red (AR) and RB5 in aqueous solutions were investigated using  $\gamma$ -rays irradiation [44]. It was found that the de-coloration that was due to reactions with  $e^-$  and  $H$ , and increased linearly with increasing the irradiation dose, whereas de-coloration that was due to reaction with  $OH^{\cdot-}$  radical increase logarithmically. De-coloration was attributed to the destruction of the color bearing part of the dye as a consequence of  $OH^{\cdot-}$  addition to the aromatic ring [25]. Aqueous solutions containing AR and Apollofix Yellow (AY) were irradiated with doses of 1.0–8.0 KGy and at 0.14 KGy/h dose rate. The complete de-coloration was observed using 3.0 and 1.0 KGy doses for AR and AY, respectively [25].

The use of ionizing radiation to decompose wastewater effluents of varying pH ( $1.6 < \text{pH} < 11.5$ ) and COD concentration ( $650 < C_i < 2210$  ppm) from dye manufacturing factory was evaluated [45]. The use of 15 KGy decreased the optical density by 95% and removed COD by 72%. Use of aeration and  $H_2O_2$  enhanced the degradation. The treatment of simulated wastewater contains four dyes, i.e., direct and reactive azo and anthraquinone dyes, using electron beam and  $\gamma$  radiation at different dyes concentration and irradiation doses in the presence and absence of  $H_2O_2$  was studied [46]. The highest de-coloration performance ( $\approx 100\%$ ) was obtained via electron beam irradiation using 7 KGy for direct and reactive azo dye ( $C_i = 1000$  ppm). A combined process of electron-beam and biological treatment was used in a pilot plant study in treating two waste streams. The first stream was generated during the operation of dyeing process and the second stream was from polyester fiber production enriched with ethylene glycol and terephthalic acid in 1998. An industrial facility was later commissioned and operated based on the same technology in 2005 [24,25]. The use of electron-beam technology to de-color and detoxify three effluents that represent chemical, final, and standard textile effluents was conducted. The effluents were de-colored (96, 55, and 90%) using 40, 2.5, and 2.5 KGy, respectively. The use of irradiation pre-biological treatment was found to reduce the toxic effect to the subsequent biological treatment process [47]. The potential use of combined treatment for alkaline and



nearly neutral textile effluent of COD ( $632 < \text{COD} < 127$  ppm), BOD ( $311 < \text{BOD} < 490$  ppm), and turbidity (75–77%) at irradiation doses ( $< 3$  KGy) was investigated. It was found that the application of coagulation prior to irradiation enhanced the de-coloration performance [48]. Electron beam irradiation of simulated effluents that contains reactive dye (reactive yellow 15), size (starch), synthetic size (PVA), alkali, color, and pigment (pigment red 139) was studied [49]. Application of 1 KGy irradiation for post-biodegradation treatment enhanced the quality of the treated effluents.

## 5. Advances in Wastewater and Sludge Disinfection

Disinfection of treated wastewaters using ionizing radiation has been studied extensively. The application of 4 KGy reduced the BOD<sub>5</sub>, COD, and total organic carbon (TOC) to acceptable limits [50]. In another study, anaerobic digested sludge irradiation at 1 KGy eliminated 98% of the total and fecal coliform, whereas BOD<sub>5</sub> was reduced to an acceptable limit at 4 KGy and COD was not affected by irradiation up to 20 KGy [44]. The disinfection of acidic industrial effluents and sludge ( $2 < \text{pH} < 3$ ) of high concentration of BOD<sub>5</sub> and COD (7093, and 32,664 ppm, respectively) using  $\gamma$  irradiation was investigated [17]. Sludge and treated water disinfection could be achieved using 7 and 4 KGy, respectively, whereas the reduction of the BOD<sub>5</sub> and COD concentration to acceptable limits was obtained at 18 KGy. A comparative study was conducted to evaluate the use of different advanced oxidation methods in the disinfection of the municipal wastewater, re-growth control, and the associated operating costs [51]. The results revealed that UV efficiency was affected by the seasonal variation in the wastewater composition, whereas ionizing radiation efficiency was respectively unaffected by this factor. Ionizing radiation provided high stable disinfection efficiency (95%) for the total colony count and total coliform at radiation doses  $> 0.25$  KGy and inhibited the re-growth. From economical point of view, the electric power consumption for UV and ozone is three orders of magnitude higher than that required for ionizing radiation. Electron beam irradiation used to disinfect a municipal wastewater at irradiation doses ( $< 3$  KGy) and removed 90% of coliforms [51].

## 6. Advances in Pharmaceutical and Petrochemical Wastewater Treatments

The possibility of treating pharmaceutical effluents was addressed by studying the effect of ionizing radiation on the biodegradability and toxicity of individual drugs. Changes in biodegradability and toxicity induced in aqueous solutions containing sulfamethoxazole (SMX,  $C_i = 0.1$  mmol/L) using ionizing radiation treatment revealed that SMX biodegradability was improved by applying 0.4 KGy dose. At 2.5 KGy dose, SMX conversion to biologically treatable substances was noted [52]. The degradation of carbamazepine (CBZ) by ionizing radiation was enhanced by the application of oxidant, 10 mM H<sub>2</sub>O<sub>2</sub> [20]. The decomposition of mutagenic and carcinogenic secondary intermediates, i.e., acridine (ACIN), was enhanced in the presence of H<sub>2</sub>O<sub>2</sub>. The ionization of aqueous solutions containing ciprofloxacin (CIR) and norfloxacin (NOR) ( $C_i = 10^{-4}$  M) was investigated. The degradation reaction proceeds via OH<sup>−</sup> and e<sup>−</sup> reactions with comparable rate constant ( $\approx 10^9$  mol<sup>−1</sup>·L·s<sup>−1</sup>). At low irradiation doses, the antibacterial activities of the secondary intermediates vanished. Pollutants hydroxylation during  $\gamma$  irradiations proceeds on the hydroxylated molecules and desethylene derivatives and during pulse radiolysis is attributed to absorbance of hydroxyl-cyclohexadienyl radicals. In hydrated electron reactions, electron adduct is formed then it underwent protonation yielding cyclohexadienyl type radical [53]. The treatment of real pharmaceutical effluent using combined process of coagulation, biological treatment, and  $\gamma$  irradiation was investigated [54]. Two tested neutral effluents, namely, low organic strength (LSW; BOD  $< 6730$  ppm, COD  $< 12,715$  ppm) and high organic strength (HSW; BOD  $< 27,242$ , COD  $< 51,223$  ppm). The use of irradiation led to maximum reduction in COD of 45% in acidic media at 50 KGy (LSW) and 30% in acidic media at 100 KGy (HSW). The application of coagulation pre-treatment was found to affect the efficiency, where 55% and 50% could be achieved using 100 KGy, for LSW and HSW, respectively. The use of H<sub>2</sub>O<sub>2</sub> led to enhanced COD and TOC removal efficiency, when compared to S<sub>2</sub>O<sub>8</sub><sup>2−</sup>. The combined treatment led to overall  $92.7\% \pm 2.3\%$  and  $90.2\% \pm 2.9\%$  removal of COD from LSW and HSW, respectively. In a separate study, combined

process of coagulation, electron beam treatment, and biological treatment was performed. An overall reduction in COD of 94% and 89% was achieved LSW and HSW, respectively [55]. The radiation doses were varied from 25–100 KGy at different pH that represent acidic, neutral and alkaline media. The slightly improved performance of the electron irradiation process was related to the reaction of  $e^-_{aq}$  with H through parallel reaction with the organic contaminants to generate  $H^+$ , which subsequently inhabits the recombination of  $e^-_{aq}$  and  $OH^-$ . The consideration of the total cost of electron beam irradiation facility (20 MeV, 100 kW), its shielding and maintenance, and capital costs was estimated and the cost of the treatment was estimated to be 0.6 USD/m<sup>-3</sup>.

The decomposition of naphthalene ( $C_i = 5\text{--}32$  ppm) in aqueous solution was studied using  $\gamma$  irradiation combined with both  $H_2O_2$  and  $TiO_2$  nano-particles. The application of 3 KGy dose led to high naphthalene removal performance (>98%) and TOC reduction (28–31%) due to hydroxylation reaction. This performance is enhanced by 35% due to the presence of 40 ppm of  $H_2O_2$  and 48% due to the presence of 0.8 g/L  $TiO_2$  [56].

## 7. Sustainability of the Technology

In general, the sustainability of the nuclear industry is governed by its technical competitive performance, economical feasibility, and safe operational practice. As presented in the previous sections, the technical performance of the application of ionizing radiation technology in wastewater treatment is effective in disinfection and reduces the bio-refractory nature of several persistence organic pollutants. Several studies proved the economical feasibility of the e-beam and  $\gamma$  irradiation technologies [21,32,41,48,51,54–63]. For e-beam technology, the capital costs include the accelerator price, building shield, conveyer, cooling and ventilation systems, and monitoring system [57]. The capital cost of the e-beam accelerator is dependent on the power ( $P$ , kW) consumed to produce optimum dose ( $D$ , KGy) for specified plant flow rate ( $Q$ , m<sup>3</sup>/h) taking into account the utilization factor ( $\varphi$ ) [48,60]:

$$P = \frac{DQ}{\varphi} \quad (14)$$

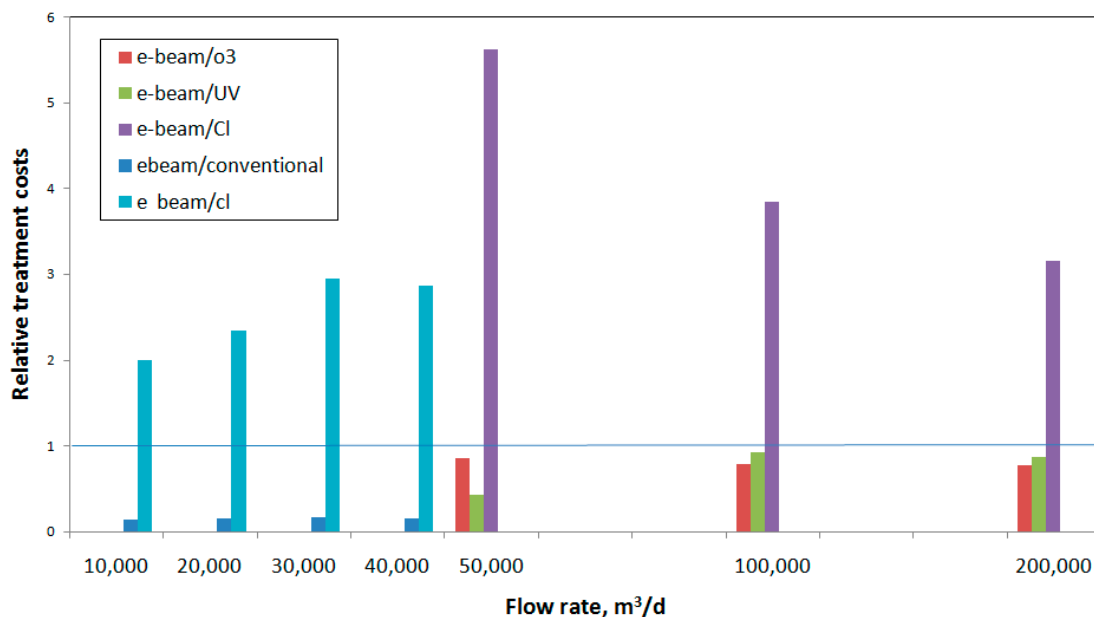
The cost of the accelerator installation ( $K_i$ , k\$) is determined using the applied electron energy ( $E$ , MeV), power, accelerator type and manufacturer ( $b$ ,  $d$ ), and installation coefficient ( $a$ ) as follows [48,57]:

$$K_i = a \cdot b(1 \pm d)E\sqrt{P} \quad (15)$$

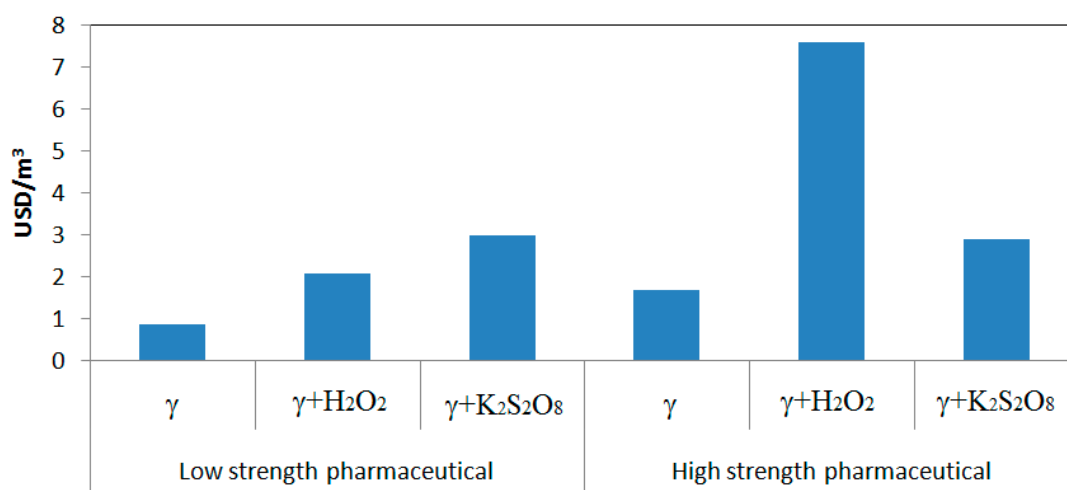
The values of the coefficients  $b$  and  $d$  varied with time due to the evolution and advances in the manufacturing process [48]. On the other hand, the costs of  $\gamma$  irradiator are not accompanied by power consumption [54], where  $\gamma$  source irradiate spontaneously. The average cost (ATC, k\$/m<sup>3</sup>) of this irradiation technique is determined based on the price of the used radioactive source ( $R$ , k\$/ $C_i$ ), required activity ( $I$ ,  $C_i$ ), irradiation time ( $t$ , h) and half life ( $t_{0.5}$ , h), and the irradiation chamber volume ( $v$ , m<sup>3</sup>)

$$A(C_i) = R \times I \times t / (t_{0.5} \times v) \quad (16)$$

To facilitate the cost comparison between the use of e-beam technology and other treatment technologies the relative treatment costs estimated at different time is presented in Figure 3 [48,64]. In terms of cost, only disinfection process, which uses chlorine, is better than the use of e-beam technology. Similar data are not available for  $\gamma$  irradiation. To compare the cost of this technique alone and cost of this technology combined with coagulation, the data reported by Changotra et al. [54] were plotted for two types of industrial effluents (Figure 4).



**Figure 3.** Relative treatment costs for wastewater using e-beam and other disinfection and conventional treatment methods [48,64].



**Figure 4.** Treatment costs for  $\gamma$  irradiation and combine coagulation- $\gamma$  irradiation [54].

There are some technical and non-technical issues that need to be resolved to ensure the sustainability of using this technology. These issues are listed below [22]

1. The usable output power of the electron accelerator limits its wide scale use as a safe substitute for chlorination for medium scale wastewater treatment plants.
2. Capital costs of electron accelerators needs to be reduced to ensure economical feasibility, where capital cost of installing an electron accelerator of 1MeV/400KW, could be attributed to half the accelerator price, whereas the design, construction, transportation, and installation of the facility contribute to 37.5% of capital cost.

From safety point of view, radiological accidents associated with different application of industrial irradiator, either  $\gamma$  irradiators or accelerators are limited, and could be classified as level 4 on the international nuclear and radiological event scale (INES). This level is used to describe accident with local consequence; Figure 5 summarizes the radiological exposures in these historical accidents [65]. Except the 1967 accelerator accident, one worker was exposed to radiation in each accident with a

probability 0.092 and  $0.12 \text{ a}^{-1}$  for accelerators and  $\gamma$  irradiators, respectively. It should be noted that due to the more stringent regulatory safety requirements that were issued later, similar accidents were not reported over three decades ago. These regulatory requirements are not only related to safety aspects but also to the security aspects to ensure the application of 3S concept (Safety, Security, and Safeguards) [66,67]. To reduce these accidents, and based on the national regulation, the following measures should be considered [68–70]:

1. Appropriate safety measures, i.e., shielding requirements, operational procedures, provision of safety assessment documents, should be applied to ensure radiological containments.
2. Appropriate security measures should be applied to ensure safety of workers and public.

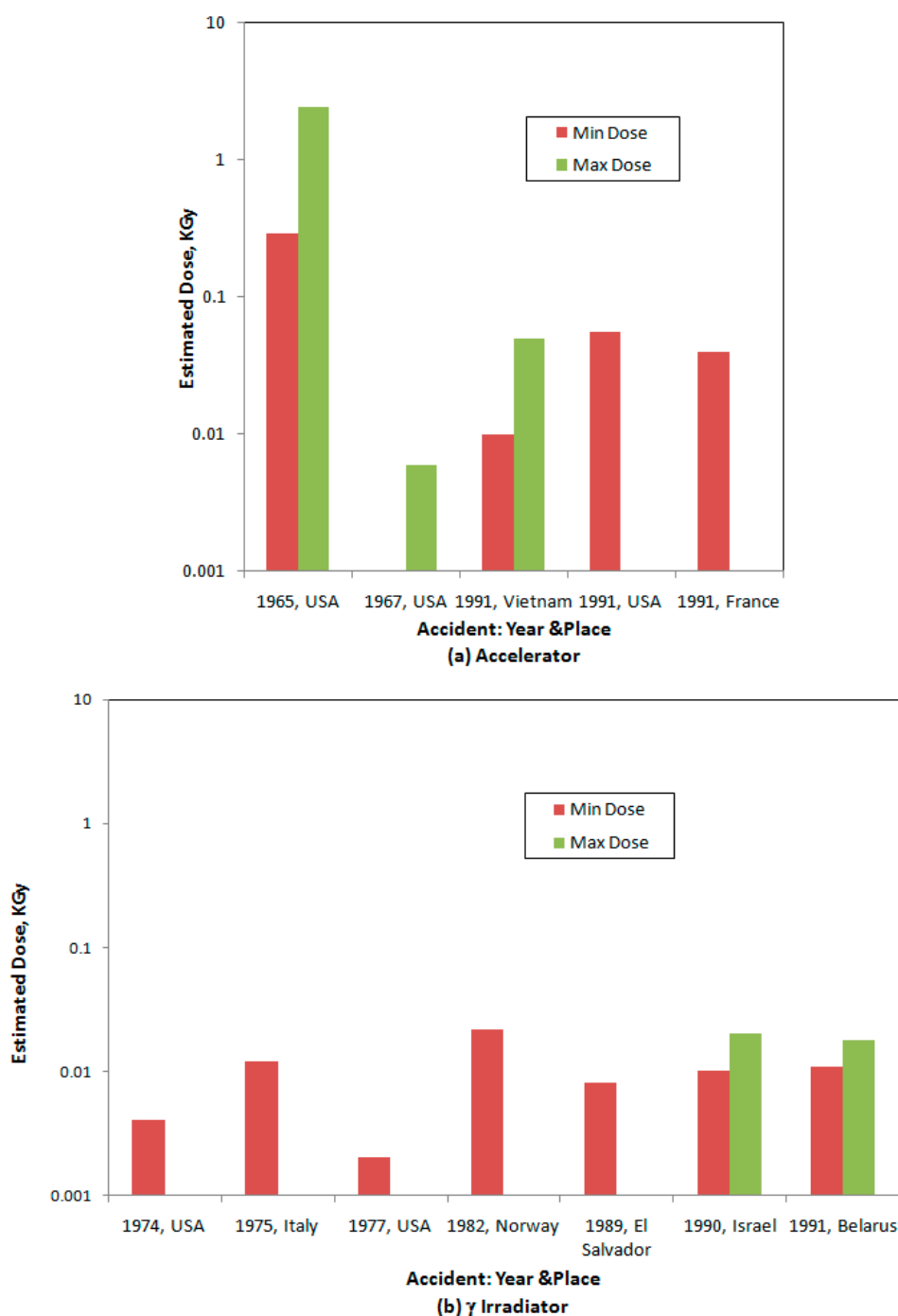


Figure 5. Summary of the historical industrial irradiator accidents; (a) accelerators, (b)  $\gamma$  irradiator.

Currently, the control strategy in these facilities is based on the combination of physical protection means, i.e., shields, barriers, and interlocks, and operational procedures [65,71]. This strategy ensured an accident probability for industrial irradiator in the order of  $2 \times 10^{-4} \text{ a}^{-1}$ .

## 8. Conclusions

The review of ionizing radiation technology in decomposing bio-refractory organic contaminants and disinfecting different wastewater effluents were presented in this study. Factors that affect the sustainability of this technology were summarized. From this review the following conclusions could be drawn.

1. Most of the published work focused on the quantification of ionizing radiation effects on primary pollutants and the secondary intermediates toxicity, and determination of optimum irradiation conditions in different effluents. Research efforts are needed to study the feasibility of ionizing radiation treatment for petrochemical wastewater effluents,
2. Despite real industrial wastewater effluents were studied, there is a need to study the kinetics reactions in these complex systems to enable a better understanding and a better design for combined treatment schemes.
3. Compared to other disinfection technologies, the ionizing radiation technology provides economical, reliable, and safer operations that are not affected by the seasonal variation in the effluent composition, and reduces the generation of secondary toxic intermediates.
4. Operational safety of industrial irradiators has been improved due to the increasing stringent regulatory requirements and the updating the operational procedures, which lead to the reduction of accidents probability from  $10^{-2}$  to  $10^{-4} \text{ a}^{-1}$ .

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