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Feasibility of radiation technology for wastewater treatment

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ABSTRACT

A wide range of remediation technologies have been employed for degradation of domestic, agricultural, industrial and municipal wastewater. Besides being economically attractive, each remediation technology has to be flexible enough to span a wide range of applications. Ionizing radiation is recognized valuable procedure for this rationale, as the radical initiated degradation can switch various pollutants into comparatively lesser disparaging substances. However, the lack of comparative data in studies using radiation technique is a main concern in further using up of this method for wastewater treatment. The main purpose of this review is to conclude the optimized radiation dose and procedures, in combination with other processes, to treat wastewater contaminated with low and high concentration of organic compounds. This review will highlight on studies carried out by various workers for exploiting ionizing and non-ionizing energies for the comparative competence in wastewater treatment.

Keywords: Ionizing radiation; Non-ionizing radiation; Wastewater; Remediation technologies

1. Introduction

Environmental pollution has become a worldwide concern. One of the main sources of such pollution is sewage wastewater and sludge. Sewage sludge constitutes the most critical and voluminous source of the end product of the conventional sewage treatment plant. To be attractive then, remediation technologies should be flexible enough to span such a wide range of needs remaining, at the same time, economically attractive [1]. Application of radiation processing for drinking water, wastewater and groundwater treatment is a cost-effective process which may insure adequate availability of that resource worldwide. Further a good understanding of the underlying chemistry and disadvantages of chemical disinfectants render use of radiation processing for its potential implementation [2]. However, for large scale implementation of radiation technology, we need to check the feasibility of studies that has been carried so far and had led to maximizing the wastewater reuse and sludge added value. This might lease the monetary code of "polluter pays" into an accessible system. Further, ionizing radiation processes have emerged as successful alternative for the destruction of non-biodegradable matter

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released from textile industries [3]. In order to understand the efficiency of irradiation techniques, we will put before the review for comparison between non-ionizing and ionizing radiation techniques employed for wastewater treatment. These studies have been carried out in several kinds of contaminated waters including municipal and industrial wastewater. The response of ionizing and non-ionizing radiation can be studied under diverse disciples such as represented schematically below:

(1) Type of energy (ionizing and non-ionizing radiation e.g. gamma, UV and Electron beam), dose rate and irradiation dose.
(2) Physicochemical studies including removal of harmful impurities, element dynamics, pH, colour, biochemical oxygen demand (BOD) and chemical oxygen demand (COD).
(3) In activation of microbes (Coli forms and pathogenic organisms).
(4) Degradation of natural, synthetic chemicals such as nonyl phenols and its derivatives.
(5) Economic feasibility.

Though ample literature is available for the fundamental studies on utilization of irradiation technology, however, for an introduction of new comers and for avoiding ambiguity among sources, doses Table 1 has been inculcated. This table represents radiation units, conversion factors and output power of various radiation sources.

2. Advanced oxidation processes vs. irradiation technology

Advanced oxidation process (AOP) degrade vast range of water and wastewater effluents via chemical oxidization furnishing (·HO) hydroxyl radicals. These hydroxyl radicals are regenerated by the combined application of ozone/hydrogen peroxide (O_3/H_2O_2), ultraviolet radiation/ozone (UV/O_3), UV/titanium dioxide (UV/TiO_2) or UV/H_2O_2 [4]. Numerous research studies have proved the tremendous potential of UV/ H_2O_2 in the removal of micro-pollutants and other different pharmaceuticals from water and wastewater effluents [5,6]. In the midst of the O_3 and UV-based AOPs, the permutation of UV, O_3 and H_2O_2 ($UV/H_2O_2/O_3$) which utilize numerous direct and indirect degradation mechanisms presenting striking alternative for the degradation of an extensive variety of refractory pollutants. Numerous researches have previously confirmed the benefit of $UV/H_2O_2/O_3$ over other AOPs in the degradation of phenol [7] and 2-propanol [8] from water. The execution of AOPs and the determination of their efficacy are complicated on numerous grounds. The efficacy of AOPs will be principally determined by the detailed water quality medium of the polluted water. Though among AOPs, the effects of background water quality on impurity exclusion are discreetly well implicit than for other technologies. In water treatment applications, AOPs generally refer to a precise division of processes that entail O_3, H_2O_2 and/or UV light. However, in this review, AOPs will be used to refer a more broad faction of processes that also occupy TiO_2 catalysis, cavitations, E-beam irradiation and Fenton's reaction. All of these processes can generate hydroxyl radicals, which can react with and annihilate an extensive variety of organic contaminants [4].

AOPs can be divided into conventional and emerging technologies based on the existing literature and the water treatment industry's practice with the technology. All of AOP technologies are evaluated in Table 2 on the basis of their performance cited in the

Table 1
Radiation units, conversion factors and output power of various sources

Conversion and SI unit equivalence	Output power of various radiation sources ($kGykg^{-1}\,h^{-1}$)		
	Type of radiation source	Dose rate ($Mrad\,h^{-1}$)	
1 rad	6.24×10^{13}ev g^{-1}	X-rays	0.18
100 rad	$1\,Jkg^{-1} = 1$ Gray (Gy)	*^{60}Co gamma-source*: 0.5 × 10 6 curie	65
1krad	$10\,Jkg^{-1} = 10$ Gy	1.0 × 10 6 curie	130
1Mrad	$10^4\,Jkg^{-1} = 10^4$ Gy	*Electron accelerator*: Van de Graaff (1 mA, 5 MeV)	7.2×10^5
1 kW	$3.6 \times 10^6\,J\,kg^{-1}\,h^{-1} = 360\,M\,rad\,kg^{-1}\,h^{-1}$	Dynamitron (40 mA, 5 M eV)	7.2×10^7
		Linac (50 mA, 10 eV)	1.8×10^8

Source modified [9].

engineering literature, outcomes of manufacturer or vendor studies and the industry's experience with the technology. The table will provide detailed discussions of each technology's chemistry, advantages and disadvantages, key variables and design parameters and available performance data from bench, pilot and field-scale tests. The following AOP technologies are compared with respect to gamma irradiation in Table 2. Since, review focuses on comparative studies among non-ionizing and ionizing radiations but concise comparative summary of conventional technologies to that of upcoming radiation technologies will be presented in tabular form to provide greater efficacy of emerging technologies.

Table 2
Comparative summary of conventional vs. emerging technologies

Characteristics	Conventional technologies			Emerging technologies			
	H_2O_2/UV	H_2O_2/O_3	O_3/UV	E-beam radiations	Sonication hydrodynamic cavitation	TiO_2-catalyzed UV oxidation	Fenton's Reaction
Method description	Degradation of organic contaminants including MTBE via ·HO radicals. These radicals are generated via MP-UV are used compared to LP- UV lamps	Combination of and H_2O_2 and O_3 used for radiolysis of water to furnish ·HO that could be used for degradation of organic effluents	Hydroxyl radicals are generated via UV light that degrade organic compounds by hydroxyl radical reactions followed by combination of direct photolysis and ozonolysis	Hydroxyl radicals, hydrated electrons and hydrogen atoms are produced when electrons react with water. These intermediates degrade organic compounds	It involves formation of cavitation microbubbles which implode violently and generate free radicals and high temperature which bring thermal decomposition of organic pollutants	TiO_2 illuminated by UV shifts electrons from Valence bond to conduction bond resulting in holes which react with water to produce free radicals. These free radicals then oxidise organic pollutants	When iron reacts with H_2O_2 resulting in radicals ·HO which degrade organic matter
Advantages	95% degradation rate for MTBE compared to UV or H_2O_2 alone	More effective than O_3 or H_2O_2 alone	More effective than O_3 or UV alone. More efficient at generating ·HO than H_2O_2/UV process for equal oxidant concentration	Minimal by product formation Performance minimally reduced by turbidity No off gas treatment required	Energy usage comparable to AOPs using UV. Less heat transfer relative to UV system. No off gas treatment required	This method can be performed at higher 300–380 nm other than UV oxidation processes	It is less energy intensive than O_3 or UV
Disadvantages	UV light penetration interfered by turbidity and nitrates	pH and H_2O_2/O_3 ratio required. Ozone off gas supply required	UV light penetration interfered by turbidity and nitrates. Ozone off gas supply required	Controversial perception about radiation technology. No extensive use	Commercially inapplicable. High expenditure requisite for inflow of supplemental oxidants such as O_3 and H_2O_2	Required TiO_2 storage is too high. pH sensitivity and oxygen sparging required	No full scale application n Requires low pH and low iron extraction

Conventional technologies	Emerging technologies
Hydrogen peroxide/Ozone (H_2O_2/O_3)	High-energy electron beam irradiation (E-beam)
Ozone/Ultraviolet irradiation (O_3/UV)	Cavitation (Sonication & hydrodynamic
Hydrogen peroxide/ Ultraviolet irradiation (H_2O_2/UV)	TiO_2-catalyzed UV oxidation
	Fenton's reaction

3. Mechanism of irradiation technology

The wastewater treatment by means of non-ionizing radiations (e.g. UV-253.7 nm) and ionizing radiations (e.g. gamma rays, electron beams) is based on entirely different mechanisms. The ionizing radiation can be produced by the use of gamma irradiation source (such as ^{60}Co or ^{137}Cs) or the use of an accelerator that generates a high-energy electron beam. Based on the insight status of scientific attainment, proclivity is given to the electron- accelerator machines owing to their very high dose rate [9]. The interaction between the radiation and matter is best explained through radiolysis of water which is an instantaneous process and accomplished in three steps involving:

(i) The physical state which lasts for 10^{-15} sec leading to formation of ionized water molecules (H_2O^+), excited water molecule (H_2O^*) and sub-excitation electrons (e^-). However, the distribution of the energy absorbed is not uniform due to "build up" effects and electron scattering [10–12].

(ii) The second step was initiated by physicochemical stage which last for (10^{-15}–10^{-12}s) involving ion-molecule reaction, dissociative relaxation, auto ionization of excited states, salvation of electrons and hole diffusion consequently leading to production of very reactive primary species (·HO, e^-_{aq}, H) and molecular products (H_3O^+ and H_2O_2) as represented in Eq. (1). These hydrated electron species is a powerful reductant, it reacts with nucleophile in an electron transfer process and it can react by passageway over distances greater than the encounter distance. The electron can also react in its "dry" or presolvation state. The hydrogen is the minor reducing radical slightly less powerful reductant than e_{aq} in neutral solution and in strongly acid solutions can act as an oxidant. The hydroxyl radical is strong oxidant which readily oxidizes inorganic ions and reacts with organic molecules **.**OH adds readily to centres of instaurations and abstracts H from **.**C–H bonds. In the latter case, it is more reactive and less selective than the hydrogen atom because of the greater exothermicity [9].

$$H_2O^+ + H_2O \rightarrow H_3O^+ + {\cdot}HO$$

$$H_2O^* \rightarrow {\cdot}HO + H{\cdot}$$

$$e^- \rightarrow 3e^-_{aq}$$

$$e^-_{aq} + H_3O^+ \rightarrow H + H_2O$$

(iii) During this stage, called as chemical stage which lasts for 10^{-12}–10^{-6} s, the species evolved above react with each other as well as with surrounding molecules and these three steps are expressed in Table 3 generated as result of different energy sources. The radiolytic yield expressed in G- values, absorbed dose and rate constant (k) is presented in Table 3.

Table 3
The radiolytic yield (in μmol J^{-1}) expressed in G-values, absorbed dose and rate constant (k)

Reaction	Rate constant ($dm^3\,mol^{-1}s^{-1}$)	Radiolytic yields G value (μmol J^{-1})						
$H^+_{aq} + OH^-_{aq} \rightarrow H_2O$	1.4×10^{11}	e^-_{aq}	H^+	·OH	H·	H_2	H_2O_2	HO_2
$H + H \rightarrow H_2$	1.0×10^{10}	2.7	0.6	2.8	0.45	0.7	3.2	0.5
$H + OH \rightarrow H_2O$	2.5×10^{10}	G-value = number of changed molecules per 100 eV						
$H + e^-_{aq} \rightarrow H_2 + OH^-_{aq}$	2.0×10^{10}	(1.60×10^{-17}) absorbed energy						
$OH + OH \rightarrow H_2O_2$	6.0×10^9							
$OH + e^-_{aq} \rightarrow OH^-_{aq}$	2.5×10^{10}							
$e^-_{aq} + e^-_{aq}$ $H_2 + 2OH^-_{aq}$	3.0×10^9							
$e^-_{aq} + H^+_{aq}$ H·	2.3×10^{10}	For conversion into S·I. units: multiply the G-value by 0.10364 to obtain G(x) in (μmol J^{-1})						
H· + OH^-aq e^-_{aq}	2.5×10^7							
In the presence of oxygen	2.1×10^{10}							
$H + O_2 \rightarrow HO_2$								
$e^-_{aq} + O_2 \rightarrow$ $O_2{\cdot}^-$	1.9×10^{10}							

G-value = number of changed molecules per 100 eV (1.60×10^{-17} J) absorbed energy. For conversion into Sl-units: multiply the G-value by 0.10364 to obtain G(x) (in μmol J^{-1}).

These radicals are however short lived which bring out both oxidation and reduction reactions simultaneously [13].

Ionizing radiations are equally efficient in electron degradation both in aqueous solutions as well as in different concentrations. Furthermore, ionizing radiation is most efficient in generating high free radical yield per unit energy input. Radiation-induced degradation of wastes and sludge in infested water is carried through varied mechanism employed by different kinds of radiations.

4. Wastewater remediation by means of electron beam, gamma radiation and ultraviolet radiation

Electron beam irradiation was carried out for bacterial infestation as well as on some major wastewater attributes such as biochemical oxygen demand and chemical oxygen demand [14]. Electron beam has been found as the most effective treatment of wastewater in reducing pathogenic microbes as well as organic decontaminants [2]. Electron beam dose of 1.5 kilogray (kGy) was found as an effective dose for the elimination of all coliforms [15]. Electron beam brings out simultaneous degradation of numerous organic compounds as well as inactivation of microbes depending on source of energy, dose rate and absorbed radiation dose [16,17]. Electron beam irradiation furnishes main products as hydrated electrons (e^-_{aq}), ·HO, H atoms, H_3O^+ radicals, H_2O_2 and H_2 produced as result of radiolysis of water [18]. Among these oxidizing hydroxyl radicals ·HO, the reducing hydrated electrons e^-_{aq} and ionized hydrogen atoms are major products, all these are highly reactive transient species due to presence of unpaired electron [13]. The presence of unpaired electron makes them an effective oxidizing agent which brings the degradation of heavy metals, various organic and inorganic pollutants and simultaneously carries out disinfestations of pathogenic microbes [15,16,19]. The ionization and free radical release is different in different oxidation processes. In case of UV irradiation, radiation is absorbed by the solutes not by water, there is always one source for generation of ·HO that is ozone and hydrogen peroxide, respectively [20]. Electron beam irradiation is absorbed by the water not by the solutes, so there are two sources for OH release after water radiolysis and O_3 decomposition [21].

The current technology of wastewater treatment and recycle, radio sensitivity of micro-organisms, disinfection and microbiological control, physical and chemical modification of aqueous pollutants, technological and economic considerations and radiation treatment of gaseous and solid wastes was summarized in 48 papers presented in the meeting on the "Use of high level radiation in waste water treatment-Status and prospectus". This was attended by 160 participants from 26 member states and by representatives of two international organizations. In these meetings, different applications of electron beam irradiation were discussed under various sub-headings as follows:

4.1. Disinfestation and microbiological control

Application of electron beam accelerator was extensively employed for treating organic load rich in pathogenic microbes such as *E.coli, Salmonella* sp., viruses (e.g. Poliovirus) and protozoans major carrier of diseases [22,23]. Radiation effects on micro-organisms are associated with physical parameters such as dose rate, dose distribution, radiation quality and radiation type and exposure pattern [24]. The physiological factors like growth phase, sensitivity, number of microbes, etc. further determine effectiveness of EB irradiation. On the whole, the effects of radiations on living organisms can be direct or indirect. In case, radiation dose is absorbed by DNA molecule of living cell or some other critical cellular component which endangers survival of organism and halts its reproductive cycle, it is termed as direct effect. The effect of EB irradiation can be indirect if free radicals furnished as result of water radiolysis interact with major macro- and micro-molecules of cell [25,26]. Inactivation of microbes by EB irradiation was explained by its shear degradative effect on cell wall, alteration of cell permeability, variation in physical components of cell protoplasm and inactivation of some crucial metabolic enzymes [16,19]. Inactivation of bacteria and bacterial spores depends on dose of EB irradiation applied. The dependence of inactivation on EB irradiation dose follows the logarithmic rule. The logarithmic number of microbes in unit volume is linearly decreasing with the dose. The total appreciation of irradiation effects on microbial contamination is described in two main factors: (i) The lethal dose at which all microbes are eradicated or killed and (ii) The D_{10} value that corresponds to the radiation dose required to reduce the microbial concentration by a factor of 10- or by 1-log cycle [27]. Reduction in *E.coli* and total coliforms was sufficient enough even at 0.2 kGy. Almost 100% reduction in *E.coli* and other coliforms was reported at 0.8 kGy [28]. The estrogen activity which is of major concern about wastewater and is parallel to microbial contamination is also reduced by EB irradiation [29]. The total bacterial and total coliforms count reduced from 6.1×10^5 and 4.8×10^4 to 0 at 3.0 kGy,

respectively, whereas total *Salmonella* and *Shigella* count reduced from 4×10^2 to 0 at 0.75 kGy EB irradiation [15]. The faecal and total coliforms appeared to be very sensitive to radiation with low D_{10} value. Electron beam irradiation is also found to be very effective for decontamination of *Bacillus* spores. DNA damage, altered membrane permeability and subsequent spore leakage has been suggested as the mechanism by which electron beam irradiation *Bacillus* spores [30]. Further research reported electron beam doses of 2,900, 520, 80 and 550 Gy were adequate to achieve 4-log inactivation of PHI X 147, B40-8, MS-2 and *E. coli*, respectively [31].

Gamma sources were also identified as an alternative source for municipal wastewater sludge treatment. Different gamma irradiation facilities around the world were installed to investigate wastewater and sludge treatment. The efficiency of gamma irradiation for chemical degradation and microbial decontamination has been demonstrated in various countries like India, United States, Canada and Japan [32]. Almost 4-log and 1-log investigation of coliphages and total coliforms was reported at dose of 500 and 200 Gy, respectively [33]. Gamma irradiation inactivates strains of *Cryptosporidium parvum* at similar dose as required for bacteria and viruses. About >3 $\log_{10}$ units of inactivation by gamma irradiation were achieved for *C. parvum* at doses comparable with the doses required for bacterial and viral inactivation [34]. Gamma irradiation at dose rate of 463 kilo radon (krad) resulted in 3-log inactivation for Coliphages and 4- to 5-log inactivation for coliphages and heterotrophic plate count [35]. Irradiation of raw sewage with dose of 200 krad results in 4- to 5-log reduction of coliform bacteria [36]. Spore viability reduction of *Bacillus anthracis* was reduced to 6-log at high dose of 2.5 Mega radon (Mrad) of gamma irradiation [37]. The efficacy of gamma radiation for the disinfection of municipal sewage via Sludge Hygienization Research Irradiator (SHRI) determined dose of 2 kGy reduce the coliform load in raw sewage to acceptable safe levels of less than 100 colony forming unit (cfu/ml) [36]. At dose 1 kGy resulted in reduction of 99.8 and 99.3% in total and faecal coliforms, respectively [38]. In the similar study, complete inactivation of both total and fecal coliform with no regrowth was achieved at a dose of 1.3 kGy in the unchlorinated effluent samples. UV radiation treatment was found to be less efficient (96%) in the inactivation of total coliforms than the gamma radiation treatment (99.97%) at dose rate of 0.8 $kGyh^{-1}$ and at absorbed dose of 0.6 kGy [39]. Relatively low doses of gamma irradiation are required for microbial decontamination as compared with UV radiations [40]. Radiobiologists have determined 3–5 kGy dose of ionizing radiation is adequate to completely inactivate pathogenic microbes in sewage sludge [27]. Researchers found that 1 and 6 kGy dose of gamma radiation are sufficient for disinfection of sewage water and sewage sludge, respectively [41]. Virus has relatively high resistance to inactivation by ionizing radiation [42]. The D_{10} value is 2.5 kGy for virus inactivation in sewage sludge.

About 300 wastewater treatments plant using ultraviolet radiation for disinfection was first operated in 1988 [43]. The application of ultraviolet irradiation to disinfection has been an accepted practice since the mid-twentieth century and popularity of UV wastewater plants increased significantly thereafter [44]. The amount of UV energy required to inactivate micro-organisms is dependent on the UV transmittance of the liquid and suspended solids concentration [45]. The lower the transmittance, the lower the amount of UV light that reaches the micro-organism. Pathogenic microbes including bacteria and viruses are often bonded together as a floc or associated with particulate matter in wastewater. It has been estimated that about 1% of all microorganisms in wastewater are associated with particles [46]. Though UV radiation has been found effective for poor quality and primary wastewater effluents, but the presence of particle associated microbes has negative effects on the disinfestation process [47,48]. Suspended solid concentration can increase the microbial survival by shielding the microbes from UV irradiation [49]. Researchers reported particle-associated coliform exhibit a slower inactivation rate and tailing, whereas non-particle-associated coliform is more easily and rapidly inactivated [50]. A 4-log reduction was achieved for susceptible phage PH X 174 and for phage B40–8 at UV fluence of 100 and 290 Jm^{-2}, respectively [51]. A 4-log inactivation of MS2 at fluence of 750 Jm^{-2} [52] and 650 Jm^{-2} [53] has been reported for wastewater treatment. Polioviruses and ss DNA viruses PHI X 174 are highly UV sensitive and 4-log inactivation of polio viruses can be achieved at fluences between 220 and 350 Jm^{-2} [54,55]. UV lamp technology can achieve a 4-$\log_{10}$ inactivation of *Cryptosporidium* oocysts at cost effective dose [56,57]. Researchers demonstrated >3.59 $\log_{10}$ units of inactivation for *C. parvum*. A minimum UV transmission of 60% is required for effective disinfestation of water containing fungi like *Phytophthora* spp., *Pythium* spp., *Colletotricum* spp. and *Fusarium* spp. spread in irrigation water [58,59]. Table 4 has been inculcated to provide comparative efficacy of different radiation technologies on micro-organism inactivation.

Table 4
Comparative efficacy of different radiation technologies on microorganism inactivation

Treatment method	Microorganism	Dose	Log inactivation	Ref.
Electron Beam	*Total coliforms*	5,000	3	[33]
UV rays	*Alternaria zinnia*	850,000 μW.s.cm^{-2}	6	[59]
UV rays	*Fusarium oxysporum*	300,000	6	[59]
UV rays	*Pythium ultimum*	40,000	4	[59]
UV rays	*Phytophthora cinnamomi*	43,000	4	[59]
UV rays	*Colletotrichum capsici*	31,000	6	[59]
Gamma rays	*Coliforms*	200	1	[36]
Gamma rays	*Coliforms*	1,000	3	[36]
Gamma rays	*Coliforms*	2,000	4	[36]
Gamma rays	*PHI X147*	900	4	[102]
Gamma rays	*B40-8*	610	4	[51]
Gamma rays	*MS-2*	140	4	[51]
Gamma rays	*E. coli*	250	4	[51]
Gamma rays	*Total coliforms*	7 kGy	5	[103]
UV rays	*MS-2*	750 Jm^{-2}	4	[51]
Gamma irradiation	*Total Coliforms*	1,500 Gy	2	[104]
	Salmonella sp.	900 Gy	1	
	Enterococcus sp.	900 Gy	3	
	Fecal Streptococci	900 Gy	3	
UV rays	*PHIX147*	100 Jm^{-2}	4	[51]
UV rays	*B40–8*	290 Jm^{-2}	4	[51]
UV rays	*E. coli*	40 to 52 mWs/cm^2	3.4 to 3.8	[105]
UV rays	*Enterococcus*	40–52	3.1 to 3.3	[105]
UV rays	*Fecal coliforms, E. coli, Enterococci, C. perfringens spores*	13 mJ/cm^2	2.2, 2.1, 1.3, 0.2, and 2.3, respectively	[106]
UV rays	*Cryptosporidium, Giardia*	20 mJ/cm^2	3	[107]

4.2. Physical and chemical modification of aqueous pollutants

Various papers have been presented on different persistent and non-persistent aqueous pollutants of environmental importance such as organic solvents, phenols, linear alkylsulphonates surfactants, pesticides, anthraquinone dyes and polychlorinated biphenyls released either from laboratory mechanistic approach or under simulated practical conditions. Though some of reports favour radiation-induced decomposition of major pollutants, the yield is mostly too low to be competitive with other methods such as ozone treatment [7,32,60,61]. Electron irradiation at 23 kGy dose of polypropylene and after 2 MeV electron and 62 MeV proton irradiation of polytetrafluroethylene (PTFE), polyimide (PI), polyethyleneterephthalate (PET) and polypropylene (PP) has improved thermal stability and conductivity of polymer as characterized by different techniques, viz. Fourier transform IR spectroscopy, electron spin resonance spectroscopy, thermogravimetric analysis, differential scanning calorimetry and X-ray diffraction analysis [62,63].

Wastewater is often rich in mostly halogenated hydrocarbons, halonitromethanes like trichloronitromethane, chloropicric formed by chlorination and ozonation in presence of nitrite ion via disinfestation process [64]. Polychlorinated biphenyls (PCB) are associated with large range of hydrophobicity and volatility that can persistent even through anaerobic digestion [65]. These compounds are basically released from industries and plants uptake them through roots and atmospheric deposition [66]. Haloalkanes can be effectively decomposed by ionizing radiation using pulse radiolysis. In a remediation of polluted waters containing concentration of 100 μgdm^{-3}, a dose of 1 kGy was found to be sufficient to remove 98% of the trihalomethanes [67]. Trihalomethanes concentration in drinking water like $CHCl_3$ in 78.00 μgdm^{-3}, $CHBrCl_2$ in 12.25 μgdm^{-3} and $CHBr_2Cl$ in 168.32 μgdm^{-3} was subjected to pilot plant experiment. Results indicate removal of $CHCl_3$ to approximately 87.4% at dose of 2 kGy while the concentration of the other three THMs was decreased under the limit of detection [68]. Experiments with water having 145–780 μgdm^{-3} showed reduction efficiency near to

95% at doses below 6 kGy [17]. A free radical pathway has been established for radiolysis of air-free water containing halogenated aromatic compounds. This pathway often leads to the formation of suite of radical species. These free radicals react rapidly with halogenated organic compounds through dissociative electron capture [69,70]. In a reductive decomposition of family of chlorophenols, the chlorine removal rate of 2,4-DCP was found almost as five times fast as that of 2-CP or 4-CP [71]. Researchers reported diclofenac elimination using irradiation technology; approximately 1 kGy dose was required for degradation of 10^{-4} mol dm^{-3} diclofenac concentration [72]. Decomposition of nitrophenols at the initial concentration of 1 μmoldm^{-3} by gamma irradiation under O_2, air or He saturated resulted in degradation at the dose of 5 Gy [73]. Electron beam irradiation efficiently decomposes nonylphenol and its derivates with doses of about 1 kGy [74].

Pesticides are most common contaminants of surface and ground water. Pesticide compounds being highly stable, radiation treatments presents as an effective method to degrade them [75,76]. Different methods have been utilized for the removal of organic matter in water. Ionizing and non-ionizing radiations can decompose organic compounds by direct or indirect interaction with radiation via radiolysis [77]. The destruction of 96% of PCBs and water-dissolved herbicide was demonstrated in research conducted by MIT in 1980 [16]. Degradation of polycyclic aromatic hydrocarbon (PAH) in moist and dry sewage was carried out at 2, 4, 6, 8 and 10 kGy of gamma irradiation dose [78]. Carbofuran and prochloraz pesticides present an increase on degradation percentage from 5 to 10% at 5 kGy dose while methiocarb pesticide solution presents the highest degradation up to 99.9% [79]. The degradation percentage of the pesticides in the post-harvest samples due to the electron beam irradiation in the range of 6.7–88.3% with 5 kGy irradiation dose. Gamma radiolysis of carbofuran makes decomposition six times more efficient via oxygen participation than in anaerobic solutions [21]. The complete radiolytic degradation of dicamba at the concentration level of 110 ppm in aqueous solution requires an irradiation dose of 5 kGy [80]. This study suggests radiolytic degradation for pesticides like 2,4 D, MCPA and Carbendazim as an effective method for wastewater treatment. Four different pesticides (4-chloro phenoxyacetic acid (4-CPA), 2.4-dichloro phenoxyacetic acid (2,4-D), 2.4-dichloro phenoxyacetic propionic acid (2,4-DP) and 2.4-dichloro phenoxyacetic butanoic acid (2,4-DB) in aqueous solutions were irradiated with doses of 0.1–10.0 kGy at a 0.07 kGy/h dose rate and highest degradation was observed after a 1.0 kGy dose for all pesticides [81]. Complete degradation of organophosphates like dichlorovinyl dimethyl phosphate (DDVP) and chlorpyrifos-ethyl (CPF-E) was carried following 10 and 50 kGy gamma doses, respectively [82]. Complete degradation of 10^{-6} mol dm^{-3} trichloroethylene (TCE) solution was observed following dose of 150 Gy [9] and perchloroethylene (PCE). The radiolysis of chlorinated ethylenes in presence of air resulted in the formation of aldehydes and simple carboxylic acids [83]. Irradiation of aqueous solutions of various chlorophenols demonstrates efficient removal of all detected species by doses from 1 to 2 kGy [84]. For the pesticide 2,4-dichlorophenol (2,4-DCP), at doses not exceeding 1 kGy, the yield of decomposition essentially depends on initial concentration of 2,4-DCP. For 50 ppm 2,4-DCP, only 40% has been decomposed, and a dose 10 kGy is needed for complete decomposition. Institute for Energetic and Nuclear Research (IPEN) employed the existing gamma and electron beam to study the removal and degradation efficiency of toxic and refractory pollutants including organic compounds mainly from industrial origins. The experiments were performed with different trihalomethanes concentrations and its reduction with irradiation doses and the pH variation and results showed reduction efficiency near 95% at the doses below 6 kGy in chloroform concentrations varying from 145 to 780 μg/L while rest of $CHBrCl_2$, $CHBr_2Cl$ and $CHBr_3$ were found below detection limits [85].

Organic dyes and surfactants even at comparatively low concentration, determine the objectionable properties of the wastewater such as colour and foaming, so the concentration of these substances must be substantially reduced. Radiation treatment of wastewater report on decolouration focus mainly on the waste degradation by ·OH radicals only a few papers deal with the reaction of e^-_{aq} and H atom. A pilot plant for treating 1,000 m^3/day of dyeing wastewater by e-beam together with biological facility had been constructed since 1998 in Daegu, Korea. Pilot plant inlet flow was mixture of two flows: raw wastewater from dyeing process and wastewater from polyester fibre production enriched with Terephthalic acid (TPA) and Ethylene Glycol (EG). Electron beam irradiation resulted in radiolytic transformation of TPA that proceeded via radical or molecular products from TPA [86]. TPZ-enriched wastewater can also be efficiently purified by biological treatments. However, preliminary electron beam improves the process resulting in more significant decrease in TOC, COD_{Cr} and BOD_5 [87]. High-energy radiation-induced degradation of an H-acid containing azo dye Apollofix Red (AR-28), H-acid, (4-amino-5-hydroxynaphthalene-2,7-disulfonic acid, I) and its derivative, 4-hydroxynaphthalene

Table 5
Comparative efficacy of different radiation technologies on aqueous and non-aqueous pollutant degradation

Treatment method	Non-aqueous and aqueous pollutants	Dose	Percent decrease	Refs.
Electron beam	Phenol, chloroform, tetrachloroethylene (PCE), carbon tetrachloride, trichloroethylene (TCE), 1,1-dichloroethane, dichloromethane, benzene, toluene and xilene	2 kGy	80%	[108]
Electron beam	Non-chlorinated" aromatic VOC : Toluene, ethylbenzene, o-, m-, p-xylenes and chlorobenzene	55–65% 85%	10 kGy	[87]
Electron beam	Ca, Si, P, Al, Fe, Cr, Zn, Co, As, Se, Cd and Hg	Ca and P-80% Al and Si-96% Ca, Fe,Cr, Zn and Co- 99% Hg-71% Cd-44%	20 kGy 100 kGy 200 kGy 500 kGy 500 kGy	[109]
Electron beam irradiation	Metiocarb		67%	[79]
Electron beam irradiation	Procloraz, Imidacloprid, Carbofuran and Dimetoato	5 kGy	99%	[110]
Electron beam irradiation	SO_2, NO_2	3.93 kGy	90–99% 85–90%	[112]
Gamma irradiation	MCPA	4 kGy	97%	[80]
Gamma irradiation	Carbendazim	0.6 kGy	100%	[80]
Gamma irradiation	2–4 D	1 to2 kGy	95%	[80]
Gamma radiation	4-chloro phenoxyacetic acid 2.4-dichloro phenoxyacetic acid 2.4-dichloro phenoxyacetic propionic acid 2.4-dichloro phenoxyacetic butanoic acid	1.0 kGy	100%	[81]
Gamma irradiation	Chlorfenvinphos	0.2 kGy	100%	[79]
Gamma irradiation	Diclofenac	5.0 kGy	95%	[79]
Gamma irradiation	2.2 dichlorovinyl dimethyl phosphate DDVP	10 kGy	100%	[82]
Gamma irradiation	Chlorpyrifos-Ethyle (CPF-E)	50 kGy	100%	[85]
Gamma irradiation	Organo chlorines 2-CP, 4-CP, 2, 4-DCP	3 kGy	28.9%, 35.5% 86.9%	[71]
Gamma irradiation	Pharmaceuticals: Diclofenac (DCF).	1 kGy	10^{-4} mol dm^{-3}	[72]
Gamma irradiation	Total organic carbon	4 kGy	90%	[111]
Electron beam irradiation	Total organic carbon	122 kGy	97%	[111]
Gamma irradiation	Dyes; Acid fast yellow G, Maxilon C. I. Basic, Reactive red SH.B, Direct blue 3B	1–3 kGy	100%	[112]
Gamma irradiation	Alizarin Yellow	9 kGy	30%	[113]
E-beam irradiation	Iopromide	19.6 kGy	90%	[114]

-2,7-disulphonic acid (II) was studied in aqueous solution using pulse radiolysis and also by gamma radiolysis. Results obtained suggest that H-acid (I) and its derivative (II) can be efficiently destroyed by the ·HO and e^-_{aq} intermediates formed during water irradiation. In the case of compound I, the e^-_{aq} intermediate decayed faster, while in the case of II the decay of ·OH intermediate was more rapid [88]. Two different reactive dyes (Reactive Blue 15 and Reactive Black 5) in aqueous solutions were irradiated with doses 0.1–15 kGy at 2.87 and 0.14 kGy/h dose rates. The complete decolouration was observed after 1 and 15 kGy doses for RB5 and RB15, respectively. In a similar study, two other different Apollo fix dyes, Apollofix Red (AR) and Apollofix Yellow (AY), in aqueous solutions, were irradiated in air with doses of 1.0–8.0 kGy at 0.14 kGy/h dose rate. The complete decolouration was observed after 3.0 and 1.0 kGy doses for AR and AY, respectively [89]. Comparative efficiencies of irradiation technologies for degradation of different pollutants are summarized in Table 5.

4.3. Radiation treatment of non-liquid waste

Use of radiation in recycling solid scrap is already an industrialized process with proven economic merit, a Japanese delegate reported. Simultaneous removal of sulphur dioxide and oxides of nitrogen from flue gases and hygienization of sewage sludge has been carried out extensively utilizing radiation technology for environmental remediation [90]. Heavy metals constitute potentially toxic portion of organic contaminant in sludge especially from industrial wastewater. These toxic metals from industrial effluent include heavy metals like lead, mercury, cadmium, nickel, silver, zinc and chromium. The bioaccumulation of heavy metals disturbs the food chain thereby resulting in ecological imbalance [91]. Aqueous solution generates free radicals, radical ions and stable products via radiolysis of water at pH 7. The hydrated electron e^-_{aq} is the strongest reducing agent as represented below in Eqs. (2), (3) and (4). These equations were presented in different research studies carried for water remediation utilizing radiation [92,93].

$$e^-_{aq} + H_3O^+ \rightarrow H^{\cdot} + H_2O$$

$$Cr(VI) + H^{\cdot}Cr(V) \quad (2)$$

Cr(V) is unstable and is further reduced to stable Cr^{3+} ions.

$$Pb + e^-_{aq} \rightarrow Pb^+$$

$$2Pb^+ \rightarrow Pb + Pb^{2+} \quad (3)$$

Lead can also be reduced by H· atoms

$$H^{\cdot} + Pb^+ \rightarrow PbH^+$$

PbH^+ decays to produce Pb

$$2PbH^+ \rightarrow H_2 + Pb^{2+} + Pb$$

$$HgCl_2 + e^-_{aq} \rightarrow HgCl + Cl^-$$

$$HgCl_2 + H^{\cdot} \rightarrow HgCl + Cl^- + H^+ \quad (4)$$

HgCl is not stable and dimerizes to Hg_2Cl_2 as a final insoluble products schematically represented in Eq. (5):

$$2HgCl \rightarrow Hg_2Cl_2 \quad (5)$$

The hydroxyl radical (·OH) is one of the powerful oxidizing species, which lead to transformation of metal ions to higher valence states [94]. As the concentration of heavy metals in wastewater is very low (ppms), so the process seems technically not feasible, hence different mechanical approaches are employed for their separation. But for higher concentration of heavy metals in aqueous solution different chemical (precipitation, ion exchange) or physical methods (membranes, electrolysis) are comparatively more feasible from economical and technical point of view. Bulgaria has constructed new pilot plant to treat high humidity, high SO_x gases from combustion of low grade lignite [95]. Japanese scientists in 1970 demonstrated the conversion of SO_2 to an aerosol of sulphuric acid droplets which can be easily removed [96]. Electron accelerator in Russia required 300 kGy to decompose 10^{-3} mol/dm^3 non-biodegradable emulsifier Nickel to biodegradable form. Electron beam treatment combined with conventional purification methods has been constructed since 1998 for reduction of chemical reagent consumption and also reduction in retention times with increase in removal efficiencies of COD and BOD up to 30–40% [28,97]. Cadmium toxicity was extensively studied due to its high concentration in agricultural products and its release in sewage sludge [98,99]. Though the effect of ionizing radiation is well investigated, however, Massachusetts Institute of Technology (MIT 1980) found that the

electron beam significantly reduces the water soluble fraction of several toxic metals via binding of water-dissolved metals to sludge components. Ionizing radiation technology, exclusively electron beam irradiation, presents a valuable and cost-effectively feasible substitute to conventional wastewater treatment techniques for degradation of non-liquid wastes. Scientists further investigated a Co-60 facility design for sludge irradiation plant anticipated for anaerobically digested sludges with solids concentrations of 8–10% [100]. This irradiation treatment process included an irradiation tank with a recirculation system employed to irradiate batches of 6.0 m^3 in 30-min intervals. The removal of heavy metal ions from water using electron beam and gamma irradiation has been investigated for the cases of Pb^{2+} and Hg^{2+} ions. Mercury can be completely (>99.9%) removed from aqueous solution of 1×10^{-3} mol L^{-1} mercury (II) chloride using a 3 kGy dose. However, a 40 kGy dose is required to remove 96% of lead ions from 1×10^{-3} mol L^{-1} of $PbCl_2$ solution [101]. Following 20 kGy γ-ray treatments of raw wastewater and effluent from a rubber products factory, filtrations both at pH 3 and at the initial pH (pH 3.6) exhibited striking alteration (9–77% and 29–85%, respectively) in toxicity reduction, resulted in the formation of toxic filterable materials which are stable even at acidic conditions. Unlike raw wastewater, there was no significant change in toxicity identification evaluation results after γ-ray treatment at 20 kGy for rubber effluent [102].

Co-ordinated Research Project by IAEA has worked on the irradiation and toxicity evaluation for the progress of dyes standard solutions and real complex effluents (textile) to facilitate optional value for recycle of irradiated effluents during the process as well as for an appropriate ecological release. Three different types of effluents were exposed to irradiation together with standard solution of two reactive dyes (Remazol Black B-RPB and Remazol Orange 3R—R3AR). The discolouration of remazol black B and orange 3R solutions was effective at 1 and 2.5 kGy which is a comparatively low dose and possibly will add to an appropriate expenditure for irradiation technology. Irradiation of real textile BVT effluents resulted in high-quality colour elimination but the sharp toxicity reduction varied widely 97% (0.5 kGy), 54% (2.5 kGy) and 19% (1.0 kGy), BTV1—BTV3 (IAEA, 2013).

5. Conclusions

Studies so far conclude irradiation of wastewater as a well established technology. Though gamma rays and electron beam machines have potential application in solving environmental problems however, it appears that additional studies are warranted to provide additional data to potential users. The electron beams are relatively capital intensive and very few technological innovations exist presently. Ionizing radiation brings out destruction of organic molecules such as halogenated compounds, dyes and pesticides in a relatively less time than non-ionizing radiations. The ionizing radiation together with oxidants such as ozone or hydrogen peroxide, further improves the removal efficiency. Further, UV rays which were previously employed for microbial inactivation have proved to be detrimental by generating mutations in microbes. Gamma rays together with electron beam can be suggested as most efficient method for wastewater remediation.

Conversely, there subsists no ideal procedure whether conventional or else that can resolve all tribulations. However, ionizing radiations alone or in combination with other techniques has impending efficacy of contributing towards the elucidation of certain problems of waste treatment and reprocessing of exhausted resources. Radiation treatment of sewage sludge exhibited recovered sedimentation and dewatering furnishing enhanced sludges used as fertilizer/animal feed additives. Cost-benefit contemplations are still ambiguous. The alternative of radiation source to be utilized is still contentious in terms of type and nature of effluent, dose to be applied, technological clarifications subsist and expenditure approximation can be made for the explicit technology. For the assessment of performance, especially in comparison with other alternatives, mutual hard work from scientists, engineers and administrative authorities of all pertinent disciplines is indispensable.

6. Proposals /Recommendations

Concluding the review seems to be challenge, however we would like to forward some intricate proposals concerning the actions indispensable to advance this particular application of ionizing radiation. Selection of the proposals is as follows:

(a) Efforts focussed to comprehend surface properties of suspended sewage particles and their reaction with radiolytic species.
(b) Conventional technologies do not guarantee effluent without human pathogens; however, radiation technology might provide an effective method to release pathogen-free water. But very scarce information is available regarding radiation effects on pathogens

especially on parasites. Radio resistivity of some microbes indicates adaptation of specific microbes to continuous radiation exposure marking them insensitive to radiation treatment. So there is greater need to interrupt transmission cycle of such pathogenic microbes.

(c) Collaborative research teams including chemists and microbiologists functioning in radiation treatment of sludge and wastewater should be framed to put forth experimental procedure.

(d) Antagonistic consequences of radiation with chemicals like H_2O_2, chlorine, ozone, air, etc. and physical properties heat, vibration, etc. ought to be followed.

(e) Numerous research findings on various aqueous pollutants of environmental significance such as organic solvents, phenols, linear alkylsulphonates surfactants, pesticides, anthraquinonic dyes, and polychlorinated biphenyls signify the improvement of ionizing radiation in decaying these pollutants; the yield is predominantly too low to be competitive with other methods such as ozone treatment. However, there is urgent need to frame channelized network of techniques that could ensure replenishment of exhausted resources especially in developing countries like India and lead us to sustainable development.

Abbreviations

kGy	–	kilo gray
krad	–	kilo rad
Mrad	–	mega rad
cfu/ml	–	Colony forming unit per millilitre
IAEA	–	International Atomic Energy Agency
ITRC	–	Industrial Toxicology Research Centre

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