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Treatment of hospital wastewater by electron beam technology: Removal of COD, pathogenic bacteria and viruses

Jianlong Wang \textsuperscript{a,b,*}, Shizong Wang \textsuperscript{a,b}, Chuanhong Chen \textsuperscript{c}, Jun Hu \textsuperscript{a}, Shijun He \textsuperscript{a,c}, Yuedong Zhou \textsuperscript{c}, Huanzheng Zhu \textsuperscript{c}, Xipo Wang \textsuperscript{c}, Dongming Hu \textsuperscript{c}, Jian Lin \textsuperscript{c}

\textsuperscript{a} Laboratory of Environmental Technology, INET, Tsinghua University, Beijing, 100084, PR China
\textsuperscript{b} Beijing Key Laboratory of Radioactive Wastes Treatment, Tsinghua University, Beijing, 100084, PR China
\textsuperscript{c} Dasheng Electron Accelerator Technology Co., Ltd., China Guangdong Nuclear Group, Suzhou, Jiangsu, 215214, PR China

HIGHLIGHTS

- Full application of electron beam radiation to treat hospital sewage was reported.
- COD, facies \textit{Escherichia coli} and virus could be effectively removed by EB technology.
- Electron beam technology can provide a solution for treating hospital sewage.

GRAPHICAL ABSTRACT

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ABSTRACT

The effective treatment of hospital sewage is crucial to human health and eco-environment, especially during the pandemic of COVID-19. In this study, a demonstration project of actual hospital sewage using electron beam technology was established as advanced treatment process during the outbreak of COVID-19 pandemic in Hubei, China in July 2020. The results indicated that electron beam radiation could effectively remove COD, pathogenic bacteria and viruses in hospital sewage. The continuous monitoring date showed that the effluent COD concentration after electron beam treatment was stably below 30 mg/L, and the concentration of fecal \textit{Escherichia coli} was below 50 MPN/L, when the absorbed dose was 4 kGy. Electron beam radiation was also an effective method for inactivating viruses. Compared to the inactivation of fecal \textit{Escherichia coli}, higher absorbed dose was required for the inactivation of virus. Absorbed dose had different effect on the removal of virus. When the absorbed dose ranged from 30 to 50 kGy, Hepatitis A virus (HAV) and Astrovirus (ASV) could be completely removed by electron beam treatment. For Rotavirus (RV) and Enterovirus (EV) virus, the removal efficiency firstly increased and then decreased. The maximum removal efficiency of RV and EV was 98.90% and 88.49%, respectively. For the Norovirus (NVLII) virus, the maximum removal efficiency was 81.58%. This study firstly reported the performance of electron beam in the removal of COD, fecal \textit{Escherichia coli} and virus in the actual hospital sewage, which would provide useful information for the application of electron beam technology in the treatment of hospital sewage.
1. Introduction

Effective treatment of hospital sewage is critical to human health and environmental sustainability. Since the Pandemic of COVID-19 outbreak, the treatment of hospital sewage, especially from the hospital that accepts infected patients, has received extensive attention due to the potential transmission of COVID-19 by sewage discharge (Bhowmick et al., 2020; Kataki et al., 2021; Wang et al., 2020a, b). Disinfection is thus considered as necessary measure to control the spread of virus and to mitigate the potential health risk (Ilyas et al., 2020). To date, the common disinfection processes include chlorination, UV, ozone and advanced oxidation processes (AOPs) (Herraiz-Carboné et al., 2021a; Herrera-Carboné et al., 2021b; Herrera-Carboné et al., 2022a; Herrera-Carboné et al., 2022b). Chlorination could completely inactivate bacteria, such as Escherichia coli and Enterococcus faecalis, but partially inactivate virus (Tree et al., 2003). To guarantee the chlorination disinfection effect for bacteria and virus, especially for COVID-19, higher dose of chlorine and longer contact time were required (Chu et al., 2021b; Wang et al., 2020a, b), which could cause potential risk, such as generating viable but non-cultivable bacteria (Wang et al., 2021). UV could also inactivate Escherichia coli and Enterococcus faecalis, but the bacteria could regrow after UV disinfection (Li et al., 2017; Wen et al., 2019). Ozone is globally recognized as a green, broad-spectrum and efficient disinfectant (Morrison et al., 2022; Zhang et al., 2020), but it is relative high cost and lack of lasting disinfection effect (Morrison et al., 2022).

In addition to pathogens, the hospital sewage also contains emerging organic pollutants, such as antibiotics, which are difficult to remove by conventional wastewater treatment processes (Wang et al., 2020a, b; Wang and Chen, 2022). During the process of disinfection by chemicals, the possible formation of the disinfection by-products (DBPs) is also a matter of widespread concern. For example, chlorination and ozonation could result in the formation of disinfection by-products (Kozari et al., 2020; Mazhar et al., 2020). How to control the formation of disinfection by-products is an urgent problem to be solved at present, especially for the hospital sewage.

Electron beam technology is a novel wastewater treatment process, which is partially similar to the conventional advanced oxidation processes (AOPs) (Wang and Xu, 2012; Wang and Zhuan, 2020). Electron beam radiation can degrade organic pollutants in wastewater by direct action of high energy beam, and indirect action of reactive species formed by the radiolysis of water molecules (Wang and Chu, 2016; Ponomarev and Ershov, 2020; Trojanowicz, 2020; Wang and Wang, 2020).

Many studies have demonstrated the capacity of electron beam in removing organic pollutants, bacteria and antibiotic resistance genes (Chen et al., 2021; Chu et al., 2021a; Shen et al., 2019; Wang et al., 2019; Lee et al., 2015; Liu et al., 2014; Liu and Wang, 2013). But there are relatively a few practical applications of electron beam irradiation due to the limitation of electron accelerator and special wastewater radiation reactor.

Due to its unique advantages, such as no demand of chemical addition, fast reaction rate, high reaction activity, and short treatment time, electron beam technology has broad application prospects in the field of wastewater treatment, especially for the advanced treatment of recalcitrant industrial wastewater.

Our research group at Tsinghua University, cooperated with China Guangdong Nuclear Group, successfully researched and developed the electron accelerator and radiation reactor for wastewater treatment, and realized the first application of electron beam technology for the treatment of dyeing wastewater in Jinghua City, Zhejiang Province, in 2016, and established the largest wastewater treatment facility using electron beam technology for the advanced treatment of dyeing wastewater in the world in Jiangmen City, Guangdong Province, in 2019 (Wang and Wang, 2022a,b). Electron beam technology showed good performance in practical application (Wang et al., 2022).

We established the first demonstration project for the hospital sewage treatment using electron beam technology during the outbreak of COVID-19 pandemic in Shiyan City, Hubei Province, China, in order to prevent the potential transmission of virus via hospital sewage, which was finished in July 2020.

This paper reported the performance of electron beam technology in the treatment of hospital sewage, including the removal of COD, the inactivation of pathogenic bacteria, such as fecal Escherichia coli, and virus. Compared to the synthetic wastewater, the practical wastewater can accurately reflect the performance of electron beam in the treatment of hospital sewage (Mwatondo and Silverman, 2021). This study could provide technical support for the treatment of actual hospital wastewater by electron beam technology.

2. Material and methods

2.1. Chemicals

Aluminum chloride (99% purity) and urea (99% purity) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. Sulphuric acid was obtained from Beijing Tong Guang Fine Chemicals Company. Filter membrane (0.45 μm) was provided by Millipore, Ireland.

2.2. In-situ removal of fecal Escherichia coli and COD by electron beam

The self-shielding electron accelerator was in-situ used in the Xiyuan Hospital, Shiyan City, Hubei Province, to treat the hospital sewage. The outside view of the accelerator is depicted in Fig. 1. The energy was 1.5 MeV, and the beam current was 15 mA.

2.3. Virus enrichment

The actual hospital sewage before and after electron beam treatment was collected to enrich the virus. The total volume of hospital sewage taken was 10.0 L. The enriched procedure was as described before (Fout et al., 2015; Li et al., 2011). Specifically, 2.0 L of sewage was taken, and 10.0 mL of 0.1 M aluminum chloride was added into the sewage. The sewage pH was adjusted to 3.5 using sulphuric acid. Thereafter, the sewage was stirred and centrifuged at 3500 rpm/min for 5 min. The supernatant and precipitates were filtered by filter membrane, respectively. It is noted that the sample was filtered by several times. After each time, the filter was washed by 40.0 ml of 0.5 mM sulphuric acid to remove the cations on the membrane. Finally, the filter membrane was placed in petri dish and sealed, stored at −20 °C refrigerator for further analysis. The samples for supernatant and precipitates were prepared in triplicate.

![Fig. 1. The outside view of the electron accelerator used in this study.](image-url)
2.4. Analytical methods

The fecal *Escherichia coli* was determined using the filter membrane method based on the Environmental Protection Industry Standard of the People’s Republic of China (HJ/T 347–2007). The virus concentration was determined according to the quantitative reverse transcription PCR (RT-qPCR) method (Ahmed et al., 2020). In this study, five common viruses, including enterovirus, hepatitis A virus, astrovirus, norovirus and rotavirus, were selected. The used primer and corresponding sequence were listed in Table S1.

3. Results and discussion

3.1. The treatment processes of hospital sewage

The treatment process of the hospital sewage mainly included biological treatment and electron beam radiation, and the flow chart of the treatment process is depicted in Fig. 2.

The water quality of the effluent of biological treatment, i.e., moving bed biofilm reactor, was analyzed and listed as follows: solution Ph, 6–9; COD concentration, 40–90 mg/L; suspended solidS, 20–30 mg/L; NH$_3$–N, 10–15 mg/L; fecal *Escherichia coli*, $1.4 \times 10^6$–$1.6 \times 10^7$ MPN/L.

3.2. Removal of COD by electron beam

The performance of electron beam in the removal of COD and fecal *Escherichia coli* was firstly investigated using batch experiment. The hospital sewage (1.0 L) was taken and treated by electron beam in the laboratory. The beam current for batch experiment was set to 1.0 mA, and the residence time was 10.0 s.

The removal performance of COD concentration was in-situ monitored within the continuous operation of 41 d (Fig. 3). The influent COD concentration of electron beam treatment unit was in the range of 40–87 mg/L. After the treatment of electron beam, the concentration of COD in the effluent steadily ranged from 13 to 27 mg/L, which meets the discharge standard of water pollutants for medical institutions in China (GB18466-2005).

3.3. Inactivation of fecal *Escherichia coli* by electron beam

The concentration of fecal *Escherichia coli* decreased from $\geq 2.4 \times 10^6$ MPN/L before the treatment unit of electron beam, to $< 20$ MPN/L after the treatment of electron beam, and the COD concentration decreased from 79.0 mg/L before the treatment of electron beam to 24.0 mg/L after the treatment of electron beam.

In-situ continuous experiment in the hospital was also conducted to further determine the performance of electron beam in the removal of fecal *Escherichia coli*. Trial results showed that the treatment capacity of electron beam was closely related to the beam current and sewage treatment capacity (Table S2). The operational conditions for in-situ continuous experiment was as follows: 15.0 mA of beam current and 10 m$^3$/h of sewage treatment capacity. Under the experimental conditions, the concentration of fecal *Escherichia coli* in the effluent of electron beam was steadily below 50 MPN/L within 41 d, that meets the discharge standard of water pollutants for medical institutions in China (GB18466-2005).

According to Eq. (1), the absorbed dose was about 4 kGy.

Absorbed dose $= \frac{E \cdot BA \cdot 0.5}{Q}$  

(1a)

Where, $E$ represents the energy of the used accelerator (1.5 MeV in this study); $BA$ was the beam current (A); and $Q$ was the sewage treatment capacity (m$^3$/h).

3.4. Inactivation of targeted viruses by electron beam technology

The targeted virus genes were found in both supernatant and precipitates (Fig. 4). Among them, Rotavirus (RV) had the highest concentration, followed by Norovirus (NVLII), Enterovirus (EV), Astrovirus (ASV) and Hepatitis A virus (HAV). For all the viruses, their concentration in the precipitates was higher than that in the supernatant, which was due to the fact that virus would adsorb and precipitate to the particles (Kim et al., 2020).

As shown in Fig. 5, the concentration of virus decreased after the treatment of electron beam in both supernatant and precipitate, indicating that electron beam was effective to inactivate the targeted viruses.

![Fig. 2. The flow chart of the hospital wastewater treatment process.](image-url)
viruses. The removal efficiency of viruses in both supernatant and precipitate was related to the absorbed dose, which was listed in Table S3.

HAV and ASV in both supernatant and precipitates were completely removed after the treatment of electron beam, no matter what the absorbed dose was. RV removal efficiency was higher than 99% in the supernatant. Its removal efficiency in the supernatant had no obvious change when the absorbed dose increased from 30 to 50 kGy. Interestingly, its removal efficiency in the precipitate presented the trend of first decrease and then increase, when the dose increased from 30 to 50 kGy. For EV in the supernatant, it was completely removed when the absorbed dose was 50 kGy. In addition, the removal efficiency of EV in the supernatant was higher than that in the precipitates. For NVLII, its removal efficiency in the supernatant slightly decreased with the increase of absorbed dose. In comparison, the removal efficiency of NVLII increased with the increase of adsorbed dose in the precipitates. The opposite trend in the supernatant and precipitates indicated that electron beam might change the structure of virus or the dissolved particles, which induced the difference of its affinity to virus. In theory, the higher the absorbed dose, the higher the removal efficiency of virus. However, the removal efficiency of RV in the precipitates at the absorbed dose of 40 kGy was lower than that at the absorbed dose of 30 kGy.

To further analyze the removal efficiency of virus, the total removal efficiency of virus was calculated, in which the total concentration of virus in the supernatant and precipitates was considered as a whole. The final removal efficiency was shown in Table S4.

For RV and EV, their removal efficiency firstly decreased, and then increased when the absorbed dose increased from 30 to 50 kGy. The possible reason may be that electron beam radiation could not only destroy the RNA structure of virus, but also degrade the particles and organic pollutants in wastewater. The affinity of virus to the particles could vary with the change of structure of the dissolved particles and organic pollutants induced by electron beam. In addition, the dissolved particles and organic pollutants could compete the reactive species with virus, decreasing the concentration of reactive species reacting with virus. Thus, the final removal efficiency of virus was not only related to the absorbed dose, but also to other components in the hospital sewage. Compared to that at the absorbed dose of 30 kGy, the removal efficiency decreased at the absorbed dose of 40 kGy, the possible reason needs to be further investigated.

4. Mechanism of the removal of COD, fecal escherichia coli and virus

Previous studies have demonstrated that, the mechanism of wastewater treatment by electron beam include direct reaction of high energy electron beam and indirect reaction of reactive species induced by high energy electron beam (Fig. 6).

In general, indirect reaction is mainly responsible for the wastewater treatment. Indirect reaction involves that the water molecules are decomposed into different reactive species with the help of electron beam, as described in Eq. (1).

$$\text{H}_2\text{O} \rightarrow \text{HO}^- (2.7) + e^- (2.6) + \text{H}^- (0.55) + \text{H}_2(0.45) + \text{H}_2\text{O}_2(0.71) + \text{H}_3\text{O}^+(2.6)\quad (1b)$$

The number in the brackets represents the radiation chemical yield G-value, which is defined as the number of product molecules formed (or initial molecules changed) for every 100 eV of energy absorbed. It can be seen that the main reactive species include hydroxyl radicals (HO•), hydrated electron (e^-), and hydrogen radicals (H•). In the presence of oxygen, e^- and H• can be converted into oxidative species, DNA/RNA.
Based on the above analysis, the oxidation species, especially hydroxyl radicals, should dominate the removal of pollutants. The conclusion has been verified by our previous studies, in which hydroxyl radicals make major contribution to the removal of organic pollutants and antibiotic genes (Chu et al., 2018, 2021a; Wang and Wang, 2018, 2022; Wang et al., 2019; He et al., 2021). Therefore, in the practical application of electron beam for the removal of pollutants in the hospital sewage, hydroxyl radicals played major role in the removal of COD, the inactivation of fecal Escherichia coli and virus. In addition, hydroxyl radicals can react with other components in the wastewater, such as dissolved organic matters. Thus, the components of wastewater and the variation of organic matters induced by hydroxyl radicals could affect the removal of fecal Escherichia coli and virus. The influential mechanism needs to be further investigated, based on the absorbed dose, variation of organic matters and the structural change of Escherichia coli and virus.

5. Conclusion

Electron beam can be used as an advanced treatment process, which could effectively remove COD, fecal Escherichia coli and virus in hospital sewage. In practical application, the operational conditions were as follows: the beam current was 15 mA, and the sewage treatment capacity was 10 m³/h. Under such conditions, the concentration of COD was steadily below 30 mg/L, and the concentration of fecal Escherichia coli was steadily below 50 MPN/L. In the hospital sewage, RV had the highest concentration, followed by NVLII, EV, AVS and HAV. All the viruses were removed effectively by electron beam when the absorbed dose ranged from 30 to 50 kGy. In general, electron beam technology could be an effective advanced treatment process for the simultaneous removal of COD and disinfection.

CRediT author contribution statement

Jianlong Wang: Conceptualization, Project administration, Supervision, Writing - review & editing. Shizong Wang: Investigation, Formal analysis, Writing - Original Draft. Chuanhong Chen: Investigation, Formal analysis. Jun Hu: Investigation. Shijun He: Investigation, Project administration, Jianlin Wang: Conceptualization, Project administration. Jian Lin: Conceptualization, Project administration. Huanzheng Zhu: Conceptualization, Project administration, Xipo Wang: Conceptualization, Project administration, Chuanhong Chen: Formal analysis. Jun Hu: Investigation. Shijun He: Investigation. Jian Lin: Conceptualization, Project administration, Huanzheng Zhu: Conceptualization, Project administration, Jian Lin: Conceptualization, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.136265.

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