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EXPERIMENTAL STUDY TO OPTIMIZE THE TREATMENT EFFICACY OF PHARMACEUTICAL EFFLUENTS BY COMBINING ELECTRON BEAM IRRADIATION

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Abstract

Here, we report our first step towards tackling this issue at the roots by irradiating the pharmaceutical effluents from a stages of their existing treatment plant with an Electron Beam (EB) with doses varying from 25 kGy to 200 kGy. We have used a normal conducting pulsed wave linear accelerator developed by SAMEER. It produced a pencil beam of electrons of energy 6 MeV with an average current of 16 µA. To ensure optimum dose delivery, Fluka-Flair Simulations have been used. We have successfully demonstrated that electron beam irradiation along with the use of conventional techniques like coagulation after the irradiation can further increase the efficacy of the process with a final reduction in Chemical Oxygen Demand (COD) to be as large as 65% in some of the cases.

INTRODUCTION

Every day, the global demand for fresh water rises, putting additional strain on available water resources such as rivers and lakes. Furthermore, human activities have resulted in the release of new pollutants known as Contaminants of Emerging Concern (CECs) into these resources, which include micropollutants, endocrine disruptors (EDs), pesticides, pharmaceuticals, hormones, toxins, and synthetic dyes [1]. Pharmaceutical effluents are the most difficult to treat conventionally due to the presence of ammonium nitrogen, toxic and complex compounds produced as byproducts in drug manufacturing. The conventional treatments primarily aim to convert pollutants from one phase to another, pollutants can still contaminate groundwater after purification because the residue is disposed of in a landfill. As a result, we need a treatment process that can breakdown and degrade these complex compounds into smaller, less harmful byproducts such as salts or water.

According to the research, electron beam irradiation is an efficient method for degrading complex compounds [2], which also improves the efficiency of conventional wastewater treatment plants. This inspired us to develop a compact, high intensity superconducting electron accelerator that can be easily integrated into existing industrial effluent treatment systems as shown in the Fig. 1.

In this paper, We reported our first step toward this technology by using an Electron Beam (EB) to irradiate pharmaceutical effluent collected from existing industrial treatment plant. Furthermore, the EB process has been investigated in combination with a number of enhancement processes, including chemical coagulation and scavengers such as titanium dioxide (TiO_2), air, and ozone flow.

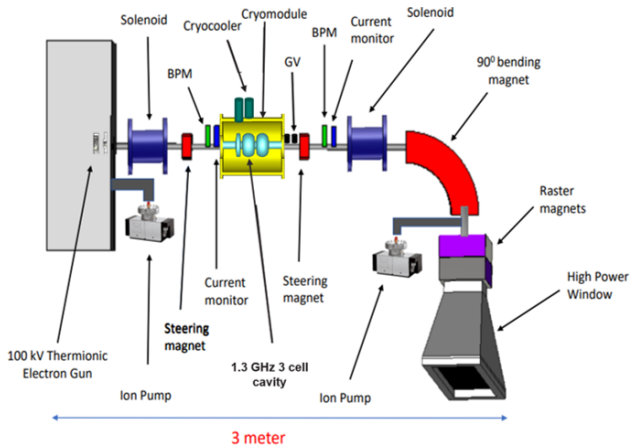

Figure 1: A Schematic figure of proposed accelerator structure.

MATERIAL AND METHODS

The industry from which samples are collected has wastewater purification scheme from which sample S is collected after the stage soil biotreatment (SBT) as shown in the Fig. 2.The initial chemical oxygen demand (COD), pH, and total dissolved solids (TDS) values for this sample are 704 ppm, 7.74, and 1421 ppm, respectively. The Titrimetric

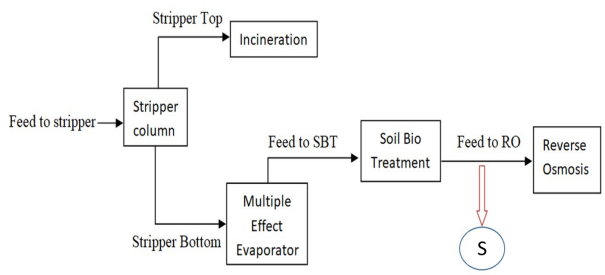

Figure 2: Stage of sample S collection from the industrial wastewater treatment plant.

Method is used for quantitative COD analysis and is used to determine the quality of irradiated water sample S. For

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this all required COD solutions are created using the APHA manual [3]. For Coagulation, ferric sulphate ($Fe_2[SO_4]_3$) is used as a coagulant. TiO_2 powder is also used in combined with air flow to achieve better results.

EXPERIMENTAL SETUP

A 6 MeV, S band, side coupled, normal conducting pulsed linac from the Society of Applied Microwave Electronics Engineering and Research (SAMEER) at IIT Bombay was used to irradiate the sample. We kept the average current constant at 16μA all across our experiments. For uniform electron beam dose distribution, samples were irradiated in a 100 mL standard cylindrical borosilicate glass beaker with a diameter of 46.2 mm, as shown in Fig. 3.

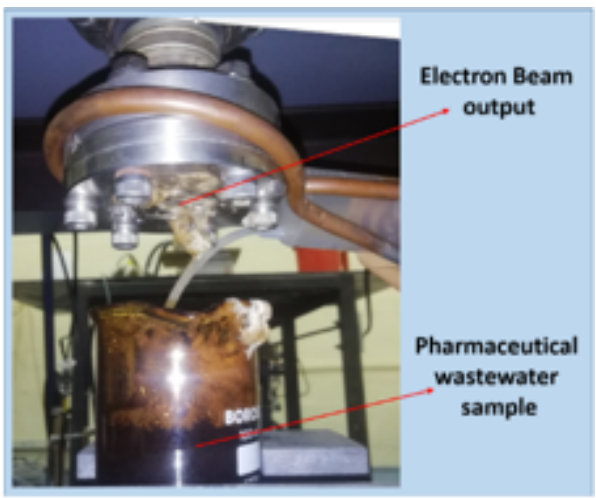

Figure 3: Experimental setup of electron beam accelerator with wastewater.

Fluka-Flair simulation was performed to optimise the dose deposition in the sample S. The absorbed dose in the sample was determined using simulations with the same geometry. Table 1 shows the dose associated with the irradiation time.

Table 1: Dose as a function of irradiation time

Irradiation time (s)	Associated Dose(kGy)
15	25
30	50
60	100
120	200
480	800

RESULTS & DISCUSSION

The effects of irradiation are discussed in terms of COD as a function of irradiation dose. To evaluate COD improvement, samples were subjected to a variety of processes, including coagulation after irradiation (EBC), irradiation + airflow (EBA), irradiation + ozone-flow (EBO) and irradiation + TiO2 + airflow (EBTA).

Electron Beam Irradiation

As discussed earlier, Sample S is collected after Soil Biotreatment (SBT), which is an environmentally friendly wastewater treatment process that uses microorganisms (Indigenous soil microflora) to convert organic impurities into H_2O and CO_2. In this case, SBT reduces additional 10% of COD, indicating that non-biodegradable compounds are the primary source of the remaining COD. Phenol, which has a concentration of 18.75 ppm, is one of the nonbiodegradable compounds still present in SBT effluent.
When water molecules are irradiated with an EB, they produce a variety of reactive species such as OH*, H_3O^+, e^-, H_2O_2, H_2, and H* in water. As shown in the Fig. 4, the most reactive species i.e. OH radical, attacks the benzene ring of nonbiodegradable aromatic compounds in water. When

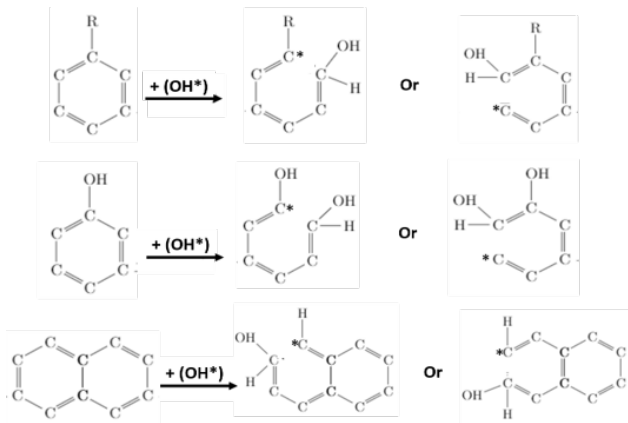

Figure 4: The OH radical produced by water radiolysis attacks aromatic compounds in various positions [4].

the benzene ring is broken, long chain aliphatic compounds are formed. Figure 5 depicts additional possible reactions of these aliphatic compounds with reactive species, and in the presence of oxygen molecules, they can be converted into carboxylic acid and esters [2]. This could be one of the

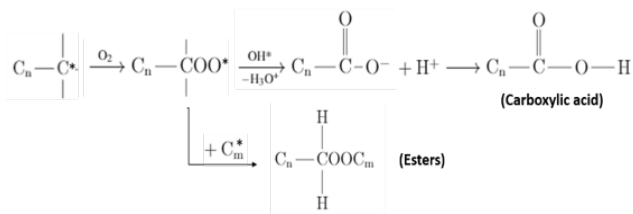

Figure 5: Possible combination of water radicals with intermediate compounds [2].

main reasons for the increase in COD value as the dose is increased (in Fig. 6). Because long carboxylic acid chains have low water solubility, the larger ester group may cause polymerisation in the wastewater during irradiation. Coagulation aids in the extraction of these insoluble compounds, resulting in a further reduction in the COD value of the sample of up to 65% at the absorbed dose of 200 kGy (Fig. 6).

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However, only EB treatment increases the COD value by up to 41% for this water sample.

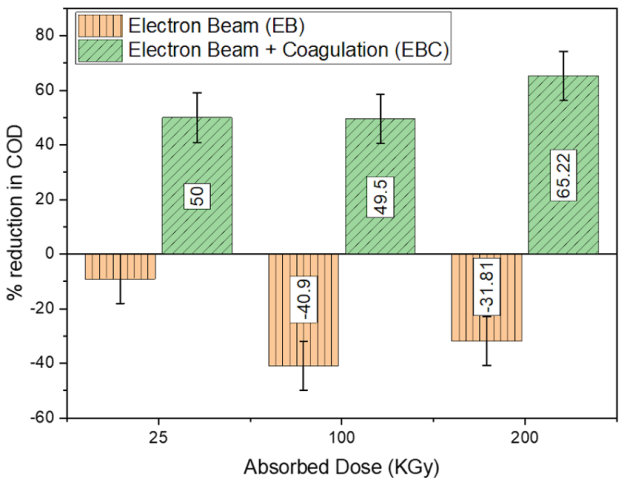

Figure 6: Percentage COD reduction of sample S for EB and EBC process with different absorbed dose.

The Scavengers Effect

In the water radiolysis process, compounds such as nitrate (NO_3^-), nitrite (NO_2^-), carbonate (CO^-), and bicarbonate (HCO_3), oxygen molecules (O_2), ozone molecules (O_3), and titanium dioxide powder (TiO_2) act as radical scavengers. These scavengers interact with free radicals before they trying to act on impurities. The three radical scavengers

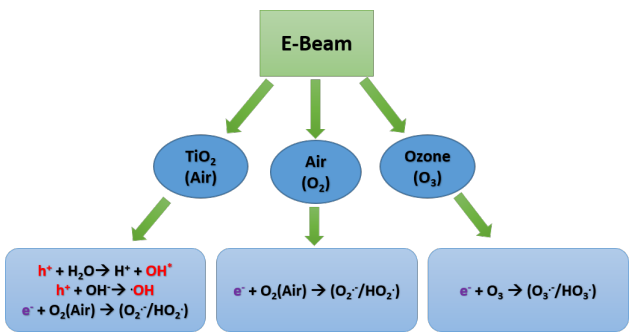

Figure 7: EB with scavengers and their byproducts.

(TiO_2,O_2, and O_3) absorb reducing species and convert them to oxidising species, as shown in the Fig. 7. These newly formed oxidising species aid in the mineralization of impurities during EB treatment. h^+ in Fig. 7 represents the holes produced after the interaction of electrons (energy >3.2 eV) with TiO_2, a semiconductor of band gap 3.2 eV. the COD reduction is observed in the S sample as shown in Fig. 8. The percentage reduction in COD value for all combinations varies between 27 to 30% with error bar of ±4.5%. The results show that EB with scavengers reduces COD more effectively than EB alone.

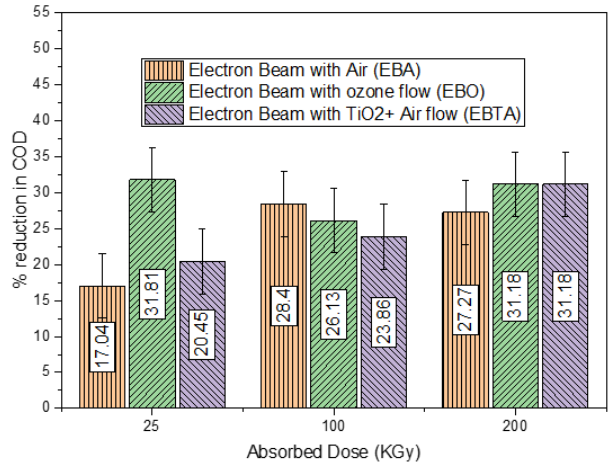

Figure 8: Percentage COD reduction of sample S for EBA, EBTA and EBO process with different absorbed dose.

CONCLUSION

In this study, we used a 6 MeV electron accelerator to irradiate a pharmaceutical wastewater sample. The aim of this study was to improve the EB treatment process by combining coagulation and scavenging agents. Electron Beam with coagulation (EBC) achieved a reduction of 66% in COD just after 200 kGy of radiation dose. This results clearly show that combining EB treatment with coagulation is more efficient than EB treatment alone. Moreover, the scavenging agents also help EB treatment to reduce COD efficiently (% COD reduction was between 27 to 30 %).

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