

Sequential Treatment of Landfill Leachate By Advanced Oxidation Processes and Struvite Precipitation

Sushma lavudya

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

maneesha vodnala

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

Bhagawan Dheeravath

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

kiran kumar Panga

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

Vijaya krishna saranga

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

Shankaraiah Golla

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

Himabindu V (✉ drvhimabindu@jntuh.ac.in)

JNTUH IST: Jawaharlal Nehru Technological University Hyderabad Institute of Science & Technology

Srinath Suranani

NIT Warangal: National Institute of Technology Warangal

Research

Keywords: Chemical Oxygen Demand, Electrooxidation, Fenton, Landfill leachate, and Struvite

Posted Date: July 7th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-627238/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

1 **Sequential treatment of Landfill Leachate by Advanced Oxidation Processes (AOP's)**
2 **and Struvite Precipitation**

3 Sushma Lavudya¹, Maneesha Vodnala¹, Bhagawan Dheeravath¹, Kirankumar Panga¹,
4 Saranga Vijaya Krishna¹, Shankaraiah Golla¹, Vurimindi Himabindu^{1*}, Srinath Suranani²

5 1. Centre for Environment, Institute of Science and Technology, JNTUH, Hyderabad,
6 India-500085

7 2. Department of Chemical Engineering, National Institute of Technology, Warangal,
8 India- 506004

9
10
11
12
13
14 **Corresponding Author:**

15 Dr. V. HIMABINDU, Ph.D.,

16 PROFESSOR

17 Centre for Environment

18 Coordinator for Centre for Alternative Energy Options

19 Institute of Science and Technology

20 JAWAHARLAL NEHRU TECHNOLOGICAL UNIVERSITY HYDERABAD,

21 Kukatpally, Hyderabad 500085, TELANGANA, INDIA

22 Email: drvhimabindu@jntuh.ac.in

23 **Abstract:**

24 Landfill leachate contains organic, inorganic compounds, heavy metals, ammonia, and
25 xenobiotic compounds which are considered unsafe for discharging into surface water which
26 requires to be treated before its discharge into the water. In this paper, preliminary studies are
27 reported on the application of Fenton, Struvite, and Electrooxidation processes for the removal
28 of Chemical Oxygen Demand (COD) and ammonia from landfill leachate. Various operational
29 parameters like pH, dosage, reaction time, and applied voltage were optimized in laboratory
30 batch experiments and evaluated for removal of COD and ammonia. Results demonstrated that
31 the Fenton process could effectively remove COD and ammonia by 75% and 23% respectively
32 at 210 min for $\text{Fe}^{+2}:\text{H}_2\text{O}_2$: 1:5 at a fixed pH 3. The Struvite process has been effective in the
33 removal of ammonia by 74% at pH 9 with the dosage of $\text{Mg}^{+2}:\text{PO}_4^{3-}:\text{NH}_4^+$ at 1:1:1 ratio. Results
34 from Electrooxidation for COD and ammonia were observed as 58.25% and 44% respectively
35 at the applied voltage 8 V for a reaction time of 60 min. The efficiency of treatment processes
36 was also evaluated in Sequential processes for COD and ammonia i.e., Sequence-I (Fenton-
37 Electrooxidation-Struvite) and Sequence-II (Fenton-Struvite) at pre-optimized conditions. The
38 sequential processes have been depicted, the removal efficiencies of COD and ammonia of
39 89% and 82% by Sequence-I; 76.77%, and 77% by Sequence-II respectively. The present study
40 demonstrates that Fenton followed by Electrooxidation and Struvite is an effective treatment
41 process that can enhance the treatment of landfill leachate.

42 **Keywords:** Chemical Oxygen Demand, Electrooxidation, Fenton, Landfill leachate, and
43 Struvite

44

45

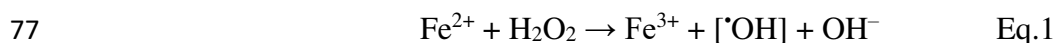
46

47 **1. Introduction**

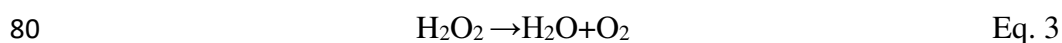
48 Management of Landfill Leachate is a serious problem that contains organic and inorganic
49 compounds, heavy metals, high ammonia content, and xenobiotic compounds. The physical
50 and chemical characteristics of landfill leachate vary significantly with time, age of landfill,
51 engineering aspects of the landfill and characteristics of dumped waste [1]. Depending on the
52 characteristics, leachate is classified into legacy (old) landfill leachate and new landfill
53 leachate. To prevent environmental impacts from landfill leachate, it is necessary to treat it
54 before its discharge into the aquatic environment. With the diversified characteristics of landfill
55 leachate, conventional treatment methods may not be feasible in the treatment of landfill
56 leachate. Several techniques implemented for the treatment of landfill leachate were adsorption
57 methods, chemical precipitation, anaerobic and aerobic biological methods, and advanced
58 oxidation processes (AOP). The above-mentioned treatment techniques have limitations like
59 less sensitivity, scaling and less biodegradability [2]. AOP's have increased attention in recent
60 times because of the oxidation potential of Hydroxyl radicals [$\cdot\text{OH}$]. AOP's were successfully
61 applied for the treatment of a wide variety of low biodegradable organic compounds in
62 industrial wastewaters as well as landfill leachate. In the treatment of landfill leachate, the
63 organic compounds are converted into less toxic intermediates or completely get mineralized.
64 AOP's entirely depends on the liberation of hydroxyl radicals in Fenton, PhotoFenton,
65 ElectroFenton, Electrooxidation, Ozonation, and Photocatalytic oxidation. Along with AOP's,
66 the Struvite precipitation process contributes to the enhancing of treatment efficiency of landfill
67 leachate

68 Fenton reaction is one of the most successful and widely used AOP's and is based on
69 the chemistry of hydroxyl radicals [$\cdot\text{OH}$], which has the strongest oxidation potential when
70 compared to ozone. The liberated [$\cdot\text{OH}$] oxidizes the organic compounds and mineralizes them
71 into either intermediate compounds or completely degraded products in presence of catalyst

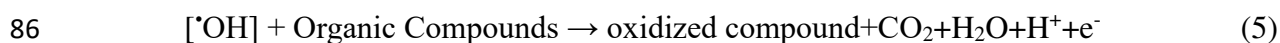
72 [3]. The reaction occurring in Fenton systems results in the formation of ferric hydroxo
 73 compounds depending on the ratio of H₂O₂ and Fe²⁺. Catalysts of Iron salts like FeSO₄·7H₂O
 74 and FeCl₃ can be used in the Fenton process have a synergistic effect on organic compounds
 75 when combined with Hydrogen peroxide (H₂O₂) under strong acidic conditions as shown in
 76 Eqs (1) and (2) [4].



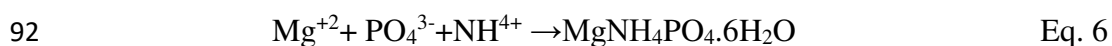
79 The net decomposition of H₂O₂ is as follows:



81 Electrooxidation is a powerful technique used in treating landfill leachate. In
 82 Electrooxidation, direct current is applied to generate free radicals as in Eq. (4). The generated
 83 radicals display a substantial role in the complete degradation of organic compounds by
 84 mineralization along with carbon dioxide and water as expressed in Eq. (5) [5].



87 Struvite precipitation is one of the most effective in the treatment of ammonia [6].
 88 Various Magnesium and Phosphorus salts like MgCl₂·6H₂O, Na₂HPO₄·12H₂O, MgO, H₃PO₄,
 89 Ca(H₂PO₄)₂·H₂O and MgSO₄·7H₂O, etc., are extensively used in precipitating ammonia Eq.
 90 (6) [7]. Struvite is insoluble which can be easily separated from water and can be used as
 91 fertilizer for plants that require magnesium for growth.



93 Fenton, Electrooxidation, and Struvite process are entirely dependent on operating
 94 conditions and initial characteristics of landfill leachates. In the present study, the experiments

95 were carried out for the removal of Chemical Oxygen Demand (COD) and ammonia from
96 landfill leachate wastewater using Fenton, Struvite, and Electrooxidation processes. Different
97 operational parameters like pH, reaction time, dosage, inner Electrode distance and applied
98 voltage were optimized. Although a lot of studies based on Fenton, Struvite, and
99 Electrooxidation techniques were reported in the literature, limited studies were carried out on
100 sequential treatment processes. This study is mainly focused on the enhanced treatment of
101 landfill leachate by sequential processes to overcome the limitation of individual techniques.

102 **2. Materials and Methodology**

103 All the laboratory experiments were carried out in a 500 ml batch reactor with a working
104 volume of 250 ml. All the experiments were conducted in triplicate and their mean values are
105 represented.

106 **2.1 Initial characterization of Landfill Leachate**

107 Landfill leachate was collected from a local landfill site located in Hyderabad. The initial
108 characterization of landfill leachate was done in triplicates and is presented in Table 1.

109 **2.2 Fenton process**

110 Fenton process was optimized for reaction time and H_2O_2 dosage and experiments were
111 performed. The calculated dosages of H_2O_2 and Fe^{+2} ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) were added based on the
112 initial concentration of COD of landfill leachate at 100 rpm [8]. The ratio of constant Fe^{+2}
113 varying H_2O_2 from 5 to 100 with the optimized reaction time (Fig. 1a). Samples were drawn at
114 an interval of 30 min for 240 min and analyzed for COD and ammonia.

115 **2.3 Struvite process**

116 The struvite process has been optimized for reaction time and pH with salts $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ and
117 $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$ at 100 rpm. The dosages of the salts were calculated based on stoichiometric
118 calculations and initial concentration of ammonia and were fixed for molar ratio $\text{Mg}^{+2}:\text{PO}_4^{3-}$

119 :NH⁴⁺ as 1:1:1. Samples were drawn at an interval of 10 min for 60 min and were analyzed for
120 ammonia. With the optimized conditions, ammonia was analyzed for Magnesium salt MgO
121 (Fig. 1b).

122 **2.4 Electrooxidation process**

123 Electrooxidation studies were performed for optimization of reaction time, applied voltage, and
124 inner Electrode distance. The Electrode material used was Stainless steel with of dimensions
125 100 mm×50 mm×2 mm with a reactive surface area of 40 cm². The setup was connected to a
126 DC power supply and pH was maintained at pH 7. The effect of the applied voltage was studied
127 at 5 V to 9 V, inner Electrode distance at 1 cm, 1.5 cm, and 2 cm with the optimized reaction
128 time (Fig. 1c) [9].

129 **2.5 Sequential treatment process**

130 To enhance the COD and ammonia removal, the sequential processes have been performed
131 with the above optimized operating conditions. The removal efficiency of COD and ammonia
132 were evaluated in two Sequential processes given below (Fig. 2):

- 133 1. Sequence-I Fenton followed by Electrooxidation and Struvite
- 134 2. Sequence-II Fenton followed by Struvite

135 **2.6 Analytical methods**

136 The pH of the leachate was adjusted with 5 N H₂SO₄/5 N NaOH using a pH meter (FE20,
137 Mettler Toledo, Switzerland). The COD (MN, Hach, Germany) was estimated by digestion
138 method and Ammonia was estimated using Kjeldahl (Pelicon) titrimetric as per standard
139 methods from APHA [10-11]. The samples drawn were filtered by Whatman filter paper No
140 45 prior to analysis for COD and ammonia. The percentage removal was calculated using the
141 following Eq. (7) [12].

$$142 \quad \text{Removal \%} = \frac{\text{Initial Concentration} - \text{Final Concentration}}{\text{Initial Concentration}} * 100 \quad \text{Eq. 7}$$

143 **2.7 Operational Cost (OC) analysis**

144 Economical aspect is a major parameter for assessing the performance of the treatment. The
 145 economic aspects for assessing the leachate treatment. The treatment process applied should
 146 be preferred based on both efficiency and operating cost. The treatment cost of Fenton,
 147 Electrooxidation and Struvite are given by following equations:

$$148 \quad \text{Fenton, Struvite } OC = \frac{M * P}{[V * (C_i - C_f)]} \quad \text{Eq. 8}$$

149 Where, OC is the operating cost, M is the amount of the chemical consumed (H₂O₂ in
 150 Liters and FeSO₄. H₂O in Kg), P is the commercial market price of each chemical in \$, V is the
 151 volume of waste water in Liters, C_i is the initial concentration of pollutant, C_f is the final
 152 concentration of pollutant under optimal conditions [13].

$$153 \quad \text{Electro oxidation } OC = aC \text{ energy} + bC \text{ electrode} + cC \text{ chemicals} \quad \text{Eq. 9}$$

$$154 \quad C \text{ energy} = \frac{(U * I * t)}{V} \quad \text{Eq. 10}$$

$$155 \quad C \text{ electrode} = \frac{I * t * M}{Z * F * V} \quad \text{Eq. 11}$$

156 Where, OC is the operating cost, U is the voltage (V), I is the current (A), t is the operating
 157 or reaction time (h), V is the volume of the sample taken (m³), M is the molecular weight of
 158 the electrode, Z is the electrons transferred, F is the Faraday's constant, a is the electricity
 159 consumed (KWh m⁻³), b is the electrode material consumed (Kg m⁻³), c is the quantities of the
 160 chemicals consumed [14]. The operating cost of all the individual and sequential processes are
 161 expressed in \$ m⁻³. The cost of the energy and chemicals used in the treatment process are
 162 calculated based on the Indian market.

163 **3. Results and Discussions**

164 **3.1 Fenton process**

165 Reaction time is an important operational parameter and plays a significant role in degrading
166 organic matter due to the reaction occurring between the [$\cdot\text{OH}$] and ferrous ions. Hence, in the
167 present study, the effect of the Fenton process was carried out from 30 min to 240 min with
168 constant pH. The influence of [$\cdot\text{OH}$] liberated from H_2O_2 may mineralize organic matter which
169 can be correlated with COD. Reduction of COD and ammonia were found linearly increasing
170 with reaction time. It has been evident from Fig. 3a that the maximum removal of COD and
171 ammonia were 75% and 23% at 210 min. As per Eqs. (1) and (2), with enough reaction time,
172 the [$\cdot\text{OH}$] degrade a wide range of organic matter. Further degradation of organic matter
173 decelerated down due to complete consumption of [$\cdot\text{OH}$]. Additionally, some inorganic ions
174 like chlorides, carbonates, bicarbonates, and bromides present or generated in degrading
175 organic matter may alter the rate of reaction [15]. Similar experiments were performed by
176 Maharaj et al [7] with synthetic wastewater and observed a maximum COD reduction of 70%
177 at the reaction time of 360 min. Studies were conducted by Perica et al [16] on heterogeneous
178 Fenton in the removal of COD and observed 75% COD removal at 400 min.

179 The effect of the dosage of H_2O_2 has been investigated with a constant $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$
180 dose. In the present study, H_2O_2 dosage was varied as it is directly proportional to the liberation
181 of [$\cdot\text{OH}$]. The calculated dosages of H_2O_2 and $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ were added in ratios 5, 10, 20, 25,
182 50, and 100. Among all the dosage ratios, ratio 5 has been effective in reducing COD and
183 ammonia by a maximum of 75% and 2.5% respectively as presented in Fig. 3b Further increase
184 in the dosage of H_2O_2 decreased the efficiency of degrading organic matter which may be due
185 to scavenging of OH radicals and therefore considered as a negative effect in treatment [17].
186 The unused H_2O_2 also may contribute to COD additionally and hence optimum dosage of H_2O_2
187 is recommended. Besides, the existence of chlorides and sulphates (Table 1) further inhibits
188 the Fenton process throughout as for their affinity to [$\cdot\text{OH}$]. This may also lead to the formation
189 of Ferric complexes preventing the reformation of ferrous ions [18]. Tony et al also investigated

190 $\text{H}_2\text{O}_2:\text{FeSO}_4\cdot 7\text{H}_2\text{O}$ and obtained a maximum reduction of COD 75% with a ratio of 15 using
191 industrial wastewater. In the present study, maximum COD removal efficiency of 75% was
192 observed with landfill leachate with an initial COD concentration of $24,000 \text{ mgL}^{-1}$ with
193 $\text{H}_2\text{O}_2:\text{FeSO}_4\cdot 7\text{H}_2\text{O}$ dosage a ratio of 5.

194 **3.2 Struvite process**

195 The Struvite process is effective in the removal of ammonia by chemical precipitation on the
196 addition of Magnesium and phosphorous salts with an equal molar ratio. Appropriate reaction
197 time enhances the reaction between Magnesium, Phosphorous salts, and ammonia for proper
198 formation thereby increasing the settleability of Struvite. The dosages of $\text{MgCl}_2\cdot 6\text{H}_2\text{O}$ and
199 $\text{Na}_2\text{HPO}_4\cdot 12\text{H}_2\text{O}$ were added as per the initial concentration of ammonia with the equal molar
200 ratio of $\text{Mg}^{+2}:\text{PO}_4^{3-}:\text{NH}_4^{+}$ (1:1:1) as given in Eq. 6. Experimental observations depict a direct
201 and linear influence of reaction time on Struvite precipitation as shown in Fig. 4a. The
202 maximum Struvite precipitation of 77% was observed at a reaction time 60 min. Although the
203 maximum removal was attained at 60 min, a significant change in removal was observed at 40
204 min, therefore, struvite can be operated for 40 min. Based on the observations further increase
205 in the reaction may result in redissolution of precipitated struvite and also affect settleability
206 of Struvite [19]. The presence of competitive ions like Ca^{2+} , Na^+ , K^+ , CO_3^{2-} , and HCO_3^- in
207 landfill leachate may interfere in the crystal formation and have a negative effect on the
208 treatment process. The formation of sodium and calcium salts along with Struvite also reduces
209 the ammonia in landfill leachate [20]. Studies by Musa et al., observed ammonia precipitation
210 of 85% with a dosage of 1.5:1.5:1 with a reaction time of 90 min with calcium pre-treatment
211 [22].

212 In the Struvite process, pH plays also a vital role in the ammonia precipitation as
213 Struvite at alkaline conditions. The pH also affects the balance of ions in crystallization,

214 therefore the pH at the induction time linearly influences the reaction and equilibrium in the
215 system. The effect of pH has been investigated from 3-11 at a variation of 2. At pH 9, 74% of
216 efficiency was observed in Struvite precipitation as represented in Fig. 4b. The solubility of
217 struvite is minimum at a higher pH range from 7 to 9, so pH in the mentioned range is used in
218 the Struvite process. Above pH 9, Struvite solubility increases which leads to a decrease in
219 ammonia concentration [21]. Similar findings were reported by Wang et al., who stated that
220 pH 9 is appropriate for the Struvite process and above pH 9 solubility of struvite increases [22].

221 The effect of ammonia reduction by replacing $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ with MgO has been
222 investigated at the above-optimized reaction time and pH of 40 min and 9.0 respectively.
223 Additionally, using $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ in Struvite precipitation may have some interferences with
224 chlorides, carbonates, and bicarbonates present in it, so the Struvite process with MgO was
225 investigated. From the experimental study, it was observed that both the Magnesium salts have
226 a significant effect in reducing ammonia in landfill leachate by 74% and 58.33% respectively
227 as shown in Fig. 4c. Despite the interferences caused by using $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, higher efficiency
228 has been observed when compared to MgO . The dissolution of MgO in aqueous solutions is
229 minimal and forms intermediates like $\text{Mg}(\text{OH})_2$ which further increases the pH of landfill
230 leachate. This increase in pH may have a negative effect on crystal formation [23].

231 **3.3 Electrooxidation process**

232 In Electrooxidation, reaction time was investigated at a time interval of 10 min for 60 min. Fig.
233 5(a) depicts the effect of COD and ammonia removal with reaction time. The maximum of
234 COD and ammonia reduction was found to be 58.25% and 44% respectively at a 60 min
235 reaction time. On further increase in the reaction time, no significant increase in the removal
236 of COD and ammonia was observed. After 60 min, the formation of a passive layer on the
237 electrodes may have occurred which reduces the effectiveness of electrooxidation [24-25].

238 Similar studies were also conducted by Mussa et al [26] who observed maximum COD removal
239 of 39% at 105 min with 5 V.

240 The applied voltage directly influences the performance of Electrooxidation for landfill
241 leachate. The experiment was carried out with different voltages range between 5 V and 9 V at
242 a reaction time of 60 min and current 2.5 A and its effect were investigated. It has been observed
243 that with an increase in applied voltage, reduction in COD and ammonia also increased. The
244 maximum reduction of COD and ammonia were found to be 58.25% and 44% respectively at
245 8 V as shown in Fig. 5b. The applied voltage is the potential difference that may lead to the
246 formation of certain reactive species like hydroxyl radicals. The gradual increase in applied
247 voltage leads to an increase in the generation of hydroxyl radicals, thereby enhancing the
248 degradation of organic matter [26]. Similar results were reported by Li et al [6] and stated that
249 further increase in applied voltage above 8 V slows down the removal of COD and ammonia.

250 The inner Electrode distance and its role in degrading the organic matter by
251 Electrooxidation were investigated. Inner Electrode distance of anode and cathode was varied
252 in the range 1, 1.5, and 2 cm. Fig. 5c illustrates that a decrease in COD and ammonia were
253 observed with an increase in inner electrode distance. The removal efficiency may have
254 increased due to faster discharge of anions at the anode and accelerated oxidation [28].

255 **3.4 Sequential treatment process**

256 Due to the diversified characteristics of landfill leachate, individual treatment processes may
257 not be effective in treating COD and ammonia. To overcome this problem two Sequential
258 treatments were examined:

- 259 1. **Sequence-I** includes Fenton followed by Electrooxidation and Struvite
- 260 2. **Sequence-II** includes Fenton followed by Struvite.

261 The Fenton, Electrooxidation and Struvite experiments in Sequential processes were carried
262 out from the above-optimized conditions. From Figure 6, in the Sequence-I (Fenton followed
263 by Electrooxidation and Struvite) the removal of COD and ammonia were 89% and 82%
264 respectively. In Sequence-II (Fenton followed by Struvite) the removal of COD and ammonia
265 were 76.67% and 77% respectively. The Electrooxidation followed by Struvite further
266 enhances the removal of COD and ammonia after the Fenton process.

267 **3.5 Cost Estimation**

268 The overall cost for estimation for the treatment processes have been given in the Table 2 which
269 are calculated by Eqs. (8) (9) (10) and (11). The cost evaluated for maximum COD and
270 ammonia removal at the optimized operating parameters. The operational cost was maximum
271 for the Electrooxidation as for its energy consumption while Fenton and Struvite has less
272 operational cost individually. Both the sequential process has shown maximum COD and
273 ammonia removal and their respective operational costs are 7.5 and 0.12 \$ m⁻³. Sequence-I has
274 high cost as it includes the Electrooxidation process. Very few studies are reported in the
275 literature regarding the determination of operating cost for Sequential treatment of landfill
276 leachate treatment. Guvence and Varank [13] have reported the individual Fenton cost as 0.238
277 € g⁻¹ COD removed. Cortez [29] has determined the operating cost of leachate treatment by the
278 Fenton process as 8.2 € m⁻³ g⁻¹ of COD removed. Bhagawan et al [27] has determined the step
279 by step cost evaluation for the Electro-coagulation process. Hence it can be concluded that
280 Sequential treatment of landfill leachate operational cost by optimizing the operational
281 parameters. The removal of COD and ammonia were removed better in Sequential treatment
282 than the individual processes.

283 **4. Conclusions**

284 The present study aims towards the enhancement of COD and ammonia reduction in the landfill
285 leachate by Fenton, Electrooxidation and Struvite techniques in individual and sequence
286 treatment process. The results showed that all the individual process were considerable in the
287 reduction of COD and ammonia at the respective optimized conditions. The sequential
288 treatment process were found to be more effective than the individual treatment process since
289 they overcome the limitations of individual process. The maximum reduction of COD and
290 ammonia were 89% and 82% in Sequence-I and 82%; 76.67% in Sequence-II. It can be
291 concluded that Sequence-I process (Fenton followed by Electrooxidation and Struvite) is
292 feasible for COD and ammonia reduction in the landfill leachate. Further studies would be
293 conducted for the effect of Sequence-I treatment process in pilot scale.

294 **Declarations:**

295 **Availability of data and material:** All the related to this manuscript was included in the
296 manuscript.

297 **Conflict of interests:** All authors have seen and approved the manuscript being submitted.
298 Hence, on behalf of all authors, the corresponding author states that there is no conflict of
299 interest.

300 **Funding:** The research leading to these results received funding from the Department of
301 Science and Technology (DST), Ministry of Science and Technology, Government of
302 India/Bharat Sarkar, under Grant Agreement DST/TDT/WM/2019//27(G).

303 One of the authors Dr. D Bhagawan received research support from the University Grants
304 Commission (UGC), Government of India/Bharat Sarkar with Grant Order no. F./31-
305 1/2017/PDFSS-2017-18-TEL- 14164.

306 **Code availability:** Not applicable.

307 **Authors' contributions:** Vurimindi Himabindu- conceptualization, methodology and review,
308 Srinath Suranani -conceptualization, project administration, Sushma Lavudya -

309 conceptualization, methodology, writing original draft, Maneesha Vodnala - methodology,
310 writing original draft, Kirankumar Panga - review and editing draft, Saranga Vijaya Krishna -
311 review and editing draft, Bhagawan Dheeravath - methodology, review final draft Shankaraiah
312 Golla - review and editing draft.

313 **Acknowledgements:** The research leading to these results received funding from the
314 Department of Science and Technology (DST), Ministry of Science and Technology,
315 Government of India/Bharat Sarkar, under Grant Agreement DST/TDT/WM/2019//27(G).

316 One of the authors Dr. D Bhagawan received research support from the University Grants
317 Commission (UGC), Government of India/Bharat Sarkar with Grant Order no. F./31-
318 1/2017/PDFSS-2017-18-TEL- 14164.

319 **References**

- 320 1. Jaafarzadeh Haghighifard NA, Jorfi S, Ahmadi M, Mirali S, Kujlu R. Treatment of
321 mature landfill leachate by chemical precipitation and Fenton advanced oxidation
322 process. Environmental Health Engineering and Management Journal. Winter. 2016
323 ;3(1):35-40.
- 324 2. Reshadi MAM, Bazargan A, McKay G. A review of the application of adsorbents for
325 landfill leachate treatment: Focus on magnetic adsorption. Science of The Total
326 Environment. 2020;138863.
- 327 3. Zhang MH, Dong H, Zhao L, Wang DX and Meng D. A review on Fenton process for
328 organic wastewater treatment based on optimization perspective. Science of the total
329 environment. 2019;11(9);670:110-121.
- 330 4. Sruthi T, Gandhimathi R, Ramesh ST, Nidheesh PV. Stabilized landfill leachate
331 treatment using heterogeneous Fenton and electro-Fenton processes. Chemosphere.
332 2018:38-43.

- 333 5. Bashir Mohammed, Lim Jun-Wei Aziz, Shuokr Qarani, Abu Amr, Salem.
334 Electrochemical Oxidation Process Contribution in Remediating Complicated
335 Wastewaters. 2014;7(5): 103352.
- 336 6. Li Z, Ren X, Zuo J, Liu Y, Duan E, Yang J, Chen P, Wang Y. Struvite precipitation for
337 ammonia nitrogen removal in 7-aminocephalosporanic acid wastewater. *Molecules*.
338 2012; 17(2):2126-2139.
- 339 7. Maharaja P, Gokul E, Prabhakaran N, Karthikeyan, S, Boopathy R, Swarnalatha S,
340 Sekaran, G. Simultaneous removal of NH₄⁺-N and refractory organics through
341 sequential heterogeneous Fenton oxidation process and struvite precipitation: kinetic
342 study. *RSC advances*. 2016;6(5): 4250-4261.
- 343 8. Tiwari AK. and Upadhyay VK. Fenton's reagent dose calculation with respect to COD
344 value and the process requirement optimization for effective oxidation of Aqueous
345 Mother Liquor Effluent of an API manufacturing industry at large scale. *International*
346 *journal of advanced research*. 2013: 158-164.
- 347 9. Krishna SV, Kumar PK, Verma K, Bhagawan D, Himabindu V, Narasu ML, Singh R.
348 Enhancement of biohydrogen production from distillery spent wash effluent using
349 electrocoagulation process. *Energy, Ecology and Environment*. 2019;4(4):160-165.
- 350 10. American public health association. Standard methods for the examination of water and
351 wastewater.2005.
- 352 11. Saranga VK, Kumar PK, Verma K, Bhagawan, D, Himabindu V, Narasu ML. Effect of
353 biohythane production from distillery spent wash with addition of landfill leachate and
354 sewage wastewater. *Applied biochemistry and biotechnology*. 2020;190(1):30-43.
- 355 12. Kumar PK, Krishna SV, Verma K, Pooja K, Bhagawan D, Himabindu V.
356 Phycoremediation of sewage wastewater and industrial flue gases for biomass generation
357 from microalgae. *South African journal of chemical engineering*. 2018; 25:133-146.

- 358 13. Guvenc SY, Varank G. Degradation of refractory organics in concentrated leachate by
359 the Fenton process: Central composite design for process optimization. *Frontiers of*
360 *Environmental Science & Engineering*. 2021;1-16.
- 361 14. Bhagawan D, Poodari S, Pothuraju T, Srinivasulu D, Shankaraiah G, Rani MY,
362 Himabindu V, Vidyavathi S. Effect of operational parameters on heavy metal removal by
363 electrocoagulation. *Environmental Science and Pollution Research*. 2014;21(24):14166-
364 14173.
- 365 15. Pani, N, Tejani V, Anantha-Singh TS Kandya A. Simultaneous removal of COD and
366 Ammoniacal Nitrogen from dye intermediate manufacturing Industrial Wastewater using
367 Fenton oxidation method. *Applied Water Science*. 2020; 10(2):1-7.
- 368 16. Pereira CAM, Brito NN. Integration of treatment technologies with Fenton reagent for
369 laboratory effluent remediation. *Revista Ambiente & Água*.2018;13(5).
- 370 17. Tony MA, Bedri Z. Experimental design of photo-Fenton reactions for the treatment of
371 car wash wastewater effluents by response surface methodological analysis. *Advances in*
372 *Environmental Chemistry*.2014.
- 373 18. Deng Y, Zhao R. Advanced oxidation processes (AOPs) in wastewater treatment. *Current*
374 *Pollution Reports*. 2015;1(3):167-176.
- 375 19. Siciliano A, Limonti C., Curcio GM, Molinari R. Advances in Struvite Precipitation
376 Technologies for Nutrients Removal and Recovery from Aqueous Waste and
377 Wastewater. *Sustainability*. 2020;12(18):7538.
- 378 20. Rahaman M S, Ellis N, Mavinic D S. Effects of various process parameters on Struvite
379 precipitation kinetics and subsequent determination of rate constants. *Water Science and*
380 *Technology*.2008;57(5):647-654.

- 381 21. Tonetti AL, de Camargo CC, Guimarães JR. Ammonia removal from landfill leachate by
382 struvite formation: an alarming concentration of phosphorus in the treated effluent. *Water*
383 *Science and Technology*.2016;74(12):2970-2977.
- 384 22. Wang H, Tian Z, Wang H, Yan Q. Optimization and reaction kinetics analysis for
385 phosphorus removal in struvite precipitation process. *Water Environment*
386 *Research*.2020;92(8):1162-1172.
- 387 23. Stolzenburg P, Capdevielle A, Teychené S, Biscans B. Struvite precipitation with MgO
388 as a precursor: Application to wastewater treatment. *Chemical Engineering*
389 *Science*.2015;133:9-15.
- 390 24. Bhagawan D, Poodari S, Chaitanya N, Ravi S, Rani YM, Himabindu V, Vidyavathi S.
391 Industrial solid waste landfill leachate treatment using electrocoagulation and biological
392 methods. *Desalin. Water Treat*. 2017;68:137-142.
- 393 25. Sirés I, Brillas E, Oturan MA, Rodrigo MA, Panizza M. Electrochemical advanced
394 oxidation processes: today and tomorrow. A review. *Environmental Science and*
395 *Pollution Research*. 2014:8336-8367.
- 396 26. Mussa ZH, Othman MR, Abdullah MP. Electrochemical oxidation of landfill leachate:
397 investigation of operational parameters and kinetics using graphite-PVC composite
398 electrode as anode. *Journal of the Brazilian Chemical Society*. 2015;26(5):939-948.
- 399 27. Bhagawan D, Poodari S, Pothuraju T, Srinivasulu D. Shankaraiah,G. Rani,MY,
400 Himabindu V, Vidyavathi S. Effect of operational parameters on heavy metal removal by
401 electrocoagulation. *Environmental Science and Pollution Research*. 2014;21(24):14166-
402 14173.
- 403 28. Bashir MJ, Aziz HA, Aziz SQ, Abu Amr SS. An overview of electro-oxidation processes
404 performance in stabilized landfill leachate treatment. *Desalination and water treatment*.
405 2013;51(10-12):2170-2184.

406 29. Cortez S, Teixeira P, Oliveira R, Mota M. Evaluation of Fenton and ozone-based
407 advanced oxidation processes as mature landfill leachate pre-treatments. Journal of
408 environmental management. 2011:749-755.

409

410

411

412

413

414

415

416

417

418

419

420

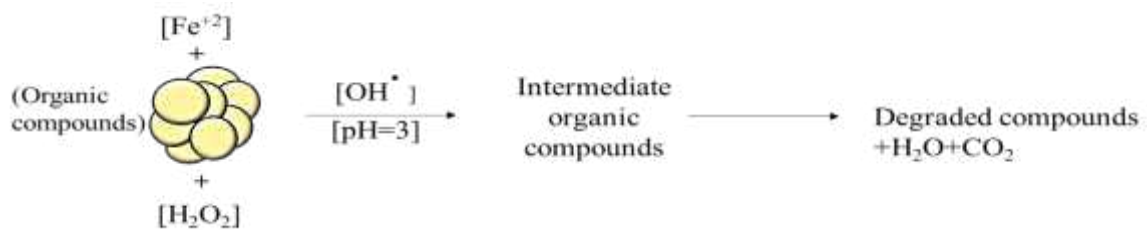
421

422

423

424

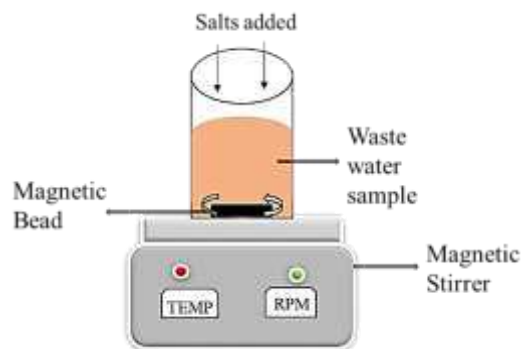
425



426
427

428

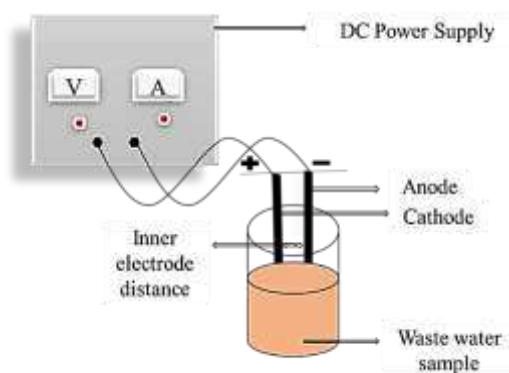
Fig. 1a Depiction of Fenton process



429

430

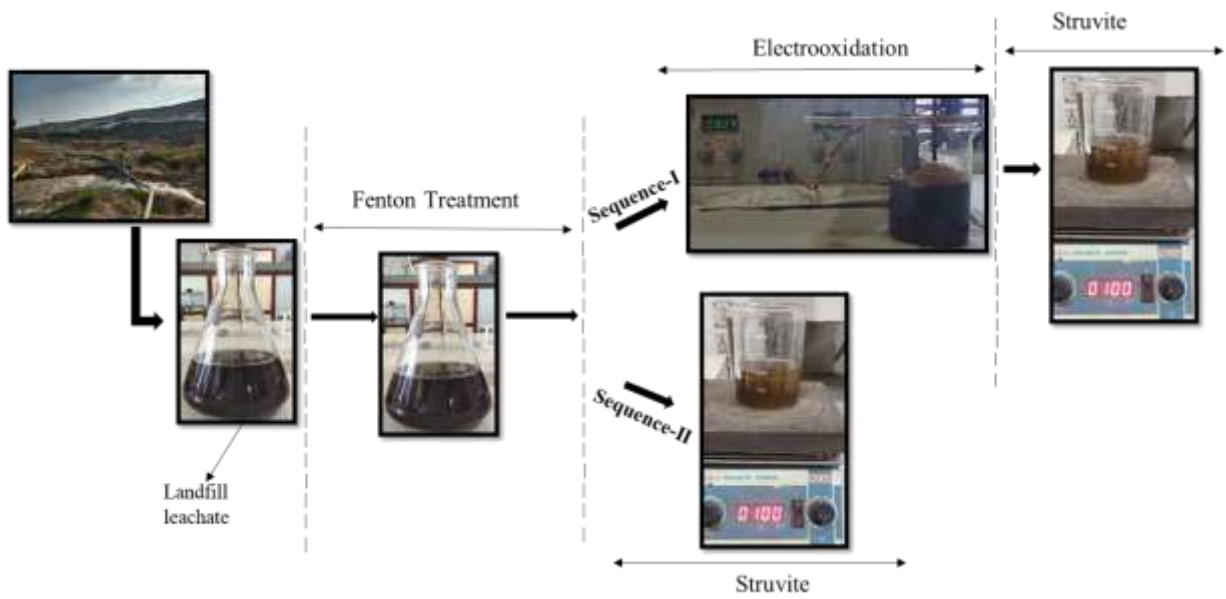
Fig. 1b Schematic representation of laboratory Struvite Precipitation setup



431

432

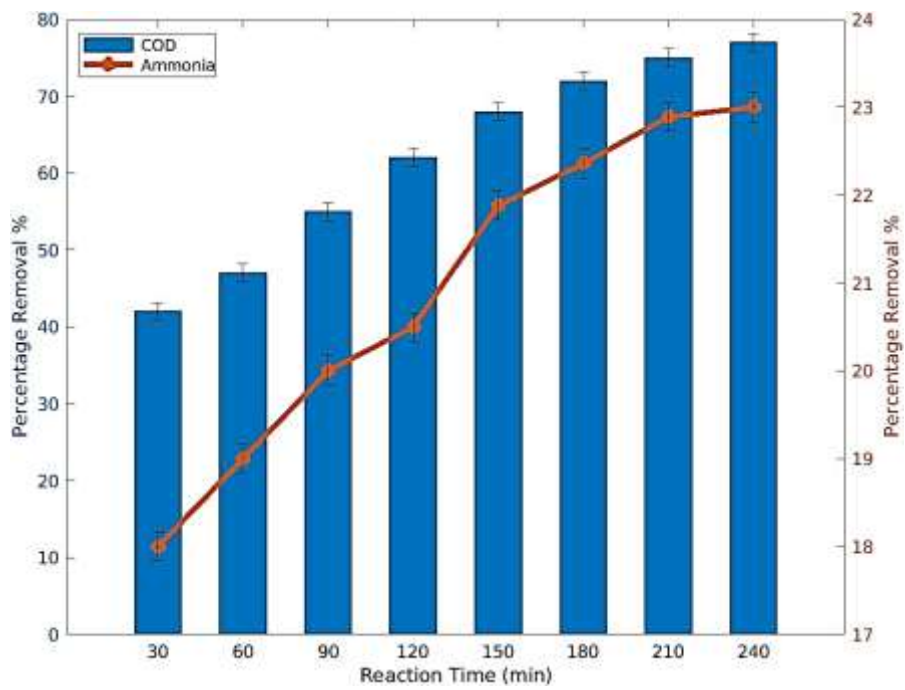
Fig. 1c Schematic representation of laboratory Electro-oxidation setup



433

434

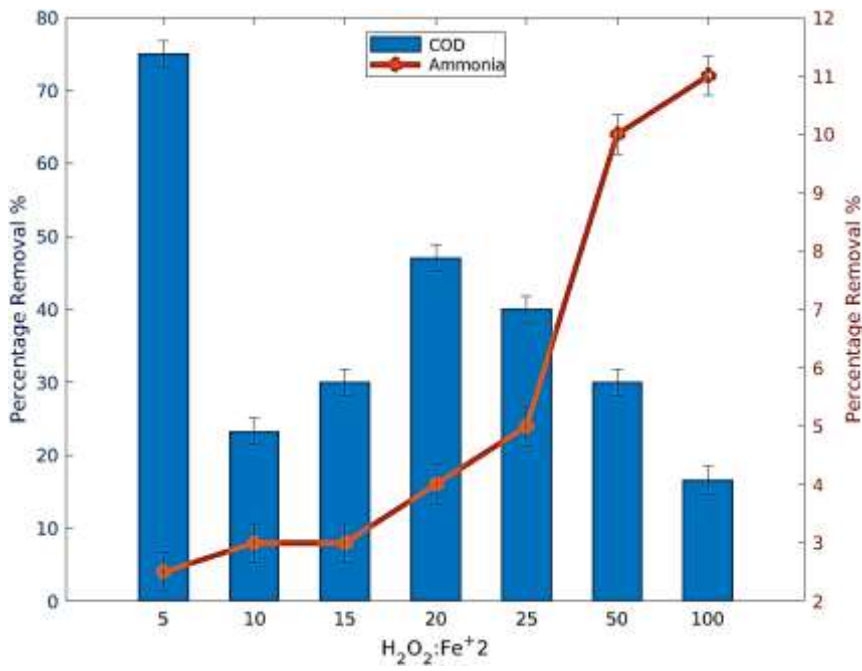
Fig. 2 Process flow diagram of Sequential treatment



435

436

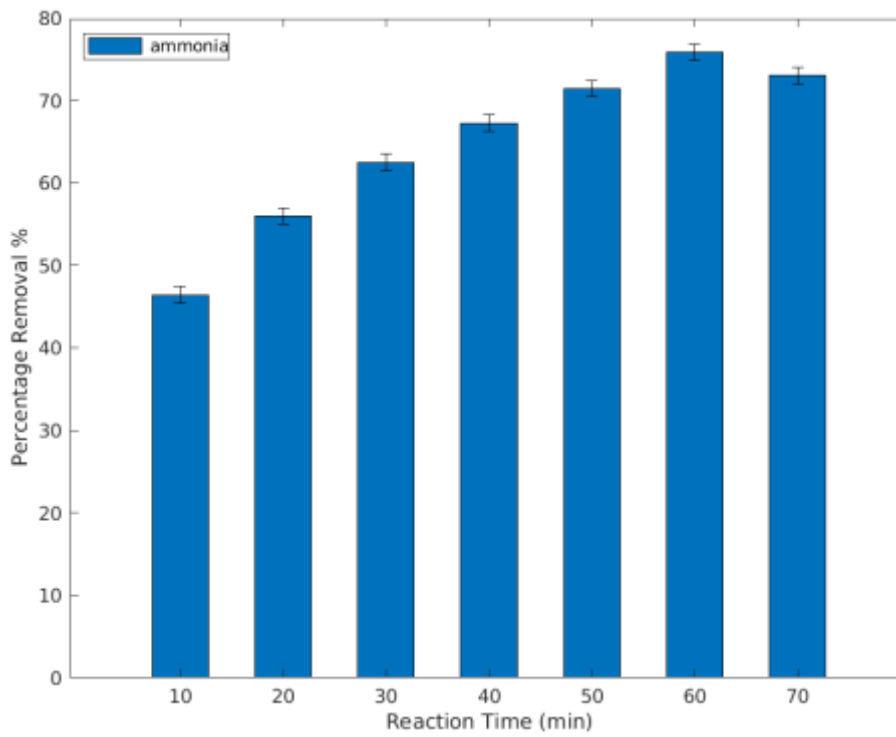
Fig. 3a Effect of reaction time on the removal of COD and ammonia in Fenton process



437

438

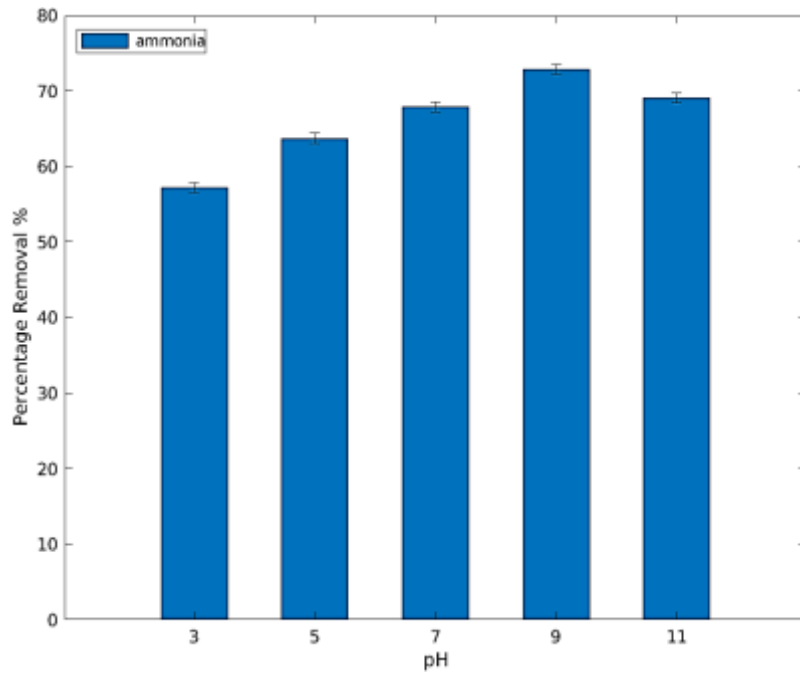
Fig. 3b Effect of H₂O₂ dosage on removal of COD and ammonia in Fenton process



439

440

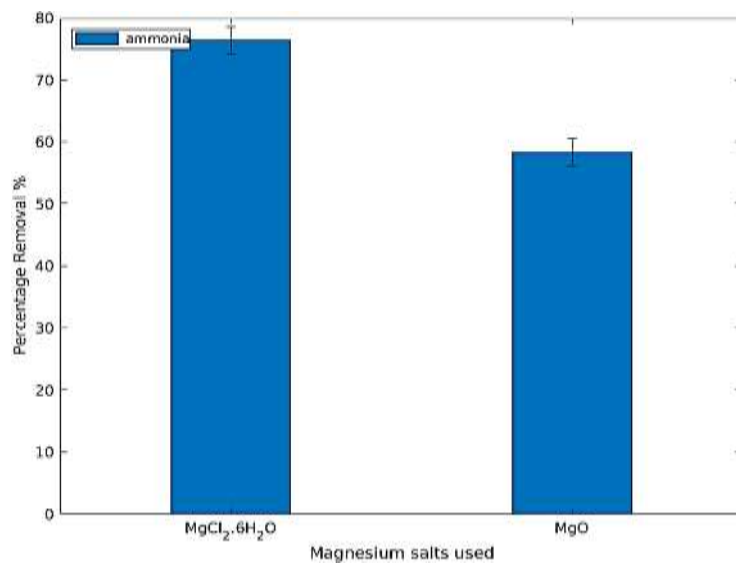
Fig. 4a Effect of reaction time on ammonia in Struvite precipitation



441

442

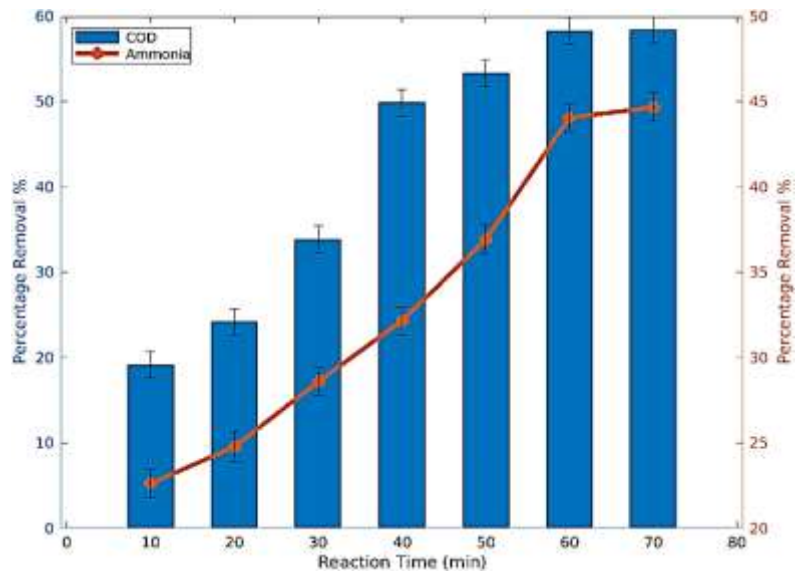
Fig. 4b Effect of pH on ammonia in Struvite precipitation



443

444

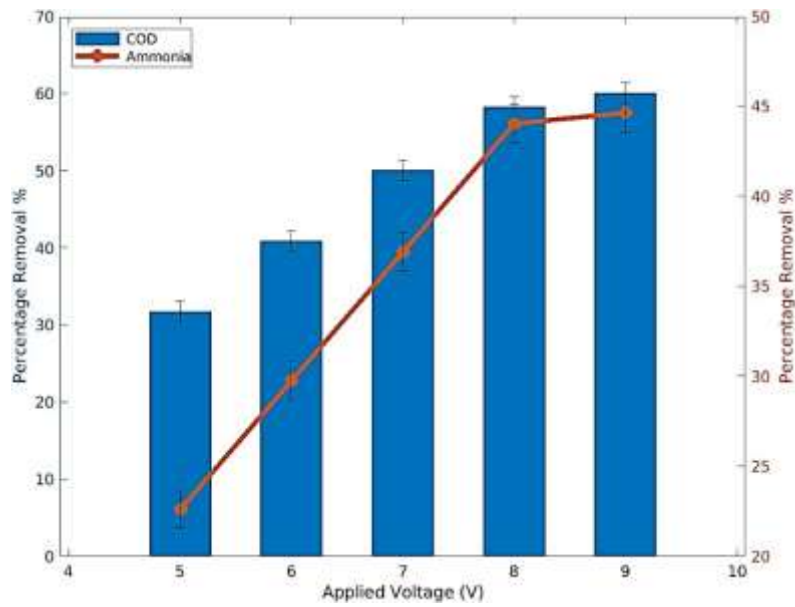
Fig. 4b Comparison of the effect of MgCl₂.6H₂O and MgO on ammonia in Struvite process



445

446

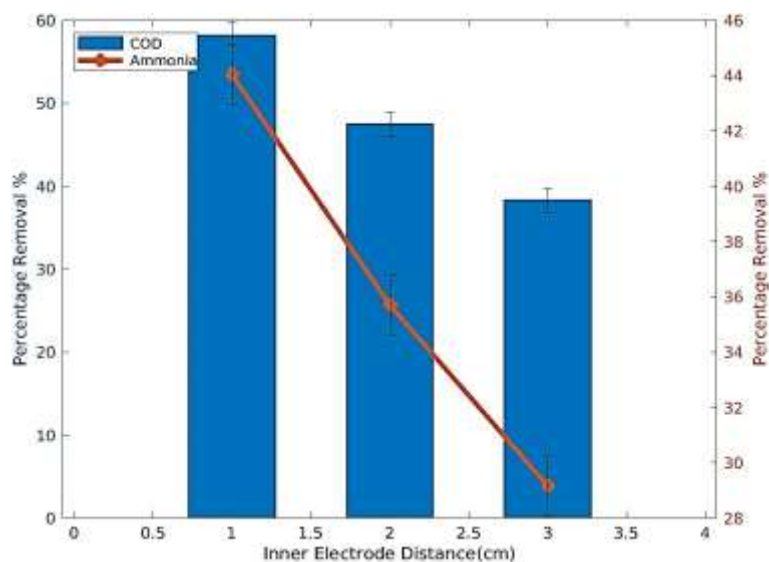
Fig. 5a Effect of reaction time COD and ammonia by Electrooxidation



447

448

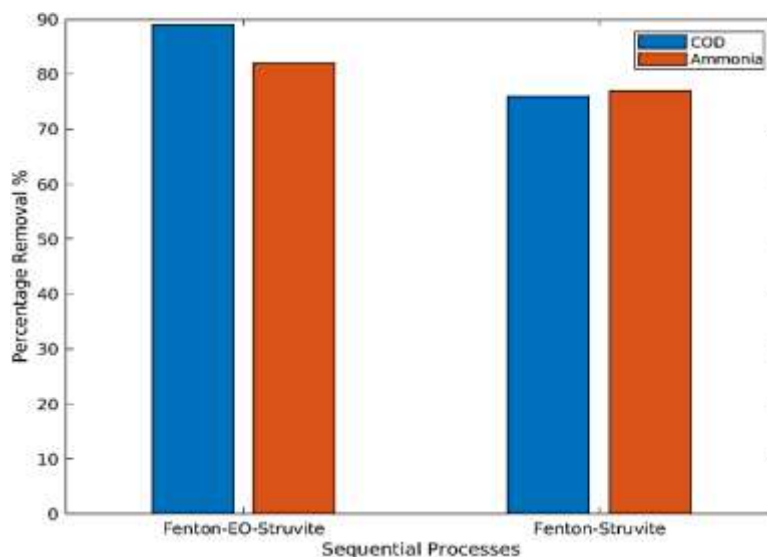
Fig. 5b Effect of Applied voltage



449

450

Fig. 5c Effect of Inner Electrode distance



451

452

Fig. 6 COD and ammonia removal % in Sequence-I and Sequence-II

453

Table 1 Initial Characterization of landfill leachate

S. No.	Parameters	Concentration \pm S. D
1	pH	8.58 \pm 0.5
2	Electrical conductivity (mS)	40.6 \pm 0.3
3	Chemical Oxygen Demand (COD) (mgL ⁻¹)	24,000 \pm 50

454	4	Chlorides (mgL ⁻¹)	8,330±30
455	5	Sulphates (mgL ⁻¹)	2,233±40
456	6	Ammonia (mgL ⁻¹)	1,680±20
457	7	Total Dissolved Solids (TDS) (mgL ⁻¹)	25,000±150

459

460

Table 2 Cost estimation of the treatment process.

S. No	Treatment Process	Total Cost at optimised parameters (\$ m⁻³)
1	Fenton	0.0214
2	Struvite	0.095
3	Electrooxidation	7.38
4	Sequence-I	7.5
5	Sequence-II	0.12

461

462