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Research paper

Degradation of Matured Landfill Leachate by Electro Fenton Treatment and Optimization by Response Surface Methodology

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Abstract

The landfill is the most widely utilized approach for solid waste management over the world. The fluid side-effect of the procedure, leachate, is a genuine danger to the earth. The ordinary biological treatment of matured landfill leachate is restricted by the presence of toxic contaminants and recalcitrant organics. A novel approach for management of landfill leachate by Electro Fenton process is researched here for the technical feasibility. The experimental outcome demonstrated that there was extensive reduction in COD-78.2%, Ammonia nitrogen-42.0%, Chloride-63.5%, Sulfate-60.0%, Sulfide-57.3%, and Phosphate-62.4% at the optimum working state of pH-2.85, Current density-140.5 A/m^2, H_2O_2 dosage- 53% of theoretical value, Reaction time-44min. The biodegradability index was enhanced from 0.21 to 0.43 because of the treatment. The examination not just uncovers the likelihood of degradation of contaminants by giving a mild electrochemical treatment additionally extends the possibility of utilizing the approach as a compelling pretreatment for its biological treatment.

Keywords: Advanced Oxidation Process; Electro Fenton process, Matured landfill leachate; Municipal Solid Waste;, Response Surface Methodology

1. Introduction

The improved population development and urban advancement throughout the most recent decades have brought about a demand in more successful and regular management of residential solid waste. Managing municipal solid waste without causing an antagonistic effect on nature and human health has turned out to be tedious. The Municipal Solid Waste (MSW) has been a major ecological issue in numerous nations. Landfills are the most well-known practice for MSW transfer everywhere throughout the world [1]. Landfills are intended to dispose high amounts of waste at a lesser cost with less ecological impacts; nonetheless, improper landfill management may posture genuine natural dangers through the release of high quality contaminated wastewater otherwise called leachate. Landfill leachate if not appropriately gathered and treated will cause significant contamination of groundwater and surface water. The amount of leachate produced relies upon rainwater permeated through wastes, biochemical procedures inside the waste cell, the inherent water content of waste and its level of compaction [2]. By and large, leachates contain a lot of organic contents, ammonia nitrogen and inorganic salts which are a greater threat to groundwater, surface water and encompassing soil [3, 4]. The residential waste contains just the small amount of heavy metals; subsequently, its commitment in the leachate is less [5]. Leachate quality is affected by many variables like the synthesis of waste, local climate, method for land filling and age of the landfill. As the landfill matures the organic part in the leachate winds up plainly overwhelmed by refractory compounds [6, 7, 8]. In the later stable methanogenic stage, the pH increases and the BOD/COD ratio diminishes, thus reducing in the biodegradability. In different nations, leachate discharge quality standards have turned out to be more stringent and henceforth increased efforts are being placed in the improvement of leachate treatment system.

For the most part, an ideal method for controlling the contamination because of landfill leachate is by evacuating the hazardous compound before it penetrates the water system [9]. The significant portion of matured or biologically treated leachate is substantial refractory organic molecules that are not easily removed amid biological treatment. Non biodegradable or recalcitrant substances like organic halogens, phenolic compounds, and poly chlorinated biphenyls could be removed from leachate utilizing physical and chemical treatment techniques. Among the different treatment advancements, AOP has been perceived as an extremely powerful device in detoxifying hazardous compounds and min-

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eralization of organic compounds that are non biodegradable [10]. The landfill leachate properties, technical applicability and constraints, effluent discharge alternatives, cost adequacy, regulatory prerequisites and ecological effects are essential elements to the choice of the most appropriate treatment procedure for landfill leachate treatment [11].

1.1. Advanced Oxidation Processes (AOPs)

For treating the recalcitrant substances present in wastewater, Advanced Oxidation Processes (AOPs) are promising techniques [12]. The fundamental component of AOPs is the era of exceedingly receptive free radicals. Hydroxyl radicals (•OH) have an oxidation capability of 2.8V and display quicker rates of oxidation responses when contrasted with conventional oxidants [10,13].Thus AOPs are successful in disintegrating numerous toxic and bio-resistant organic pollutants without delivering additional hazardous byproducts or sludge which necessitate additional managing.

A tremendous number of strategies are ordered under the wide meaning of AOPs. The vast majority of them utilize a blend of strong oxidizing agents e.g. H_2O_2, O_3 with catalysts (e.g. transition metal particles). Fenton process is measured as the most encouraging treatment among AOPs for remediation of profoundly contaminated wastewater [10, 14]. Fenton's reagent is a blend of ferrous iron catalyst and hydrogen peroxide oxidizing agent. It was founded by Henry J.H. Fenton. Within the sight of transition metal, for example, iron or copper, hydrogen peroxide responds more vigorously and aggressively than without the catalyst. Fenton process can oxidize and mineralize all the organic matter to CO_2 and water.

The mechanism of the Fenton procedure is said in conditions (1) - (5) [14].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH- + \bullet OH \tag{1}$$

$$Fe^{2+} + \bullet OH \rightarrow Fe^{3+} + OH- \tag{2}$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + + HO_2\bullet + H^+ \tag{3}$$

$$\bullet OH + H_2O_2 \rightarrow H_2O + HO_2\bullet \tag{4}$$

$$2\bullet OH \rightarrow H_2O_2 \tag{5}$$

To maintain a strategic distance from the impediments of conventional Fenton oxidation, for example, loss of reactivity of chemicals and sludge generation, an altered procedure called Electro Fenton process was developed [15].

1.2. Electro Fenton Process

The Electro Fenton (EF) process is the utilization of electrically aided Fenton response. Ordinarily, there are two diverse EF applications. In the first, the Fe $^{2+}$ and H_2O_2 are added to the reactor from outside and inert electrodes having high catalytic movement are utilized as anode material shown in figure 1. In the second one, $H_2O_2$is included from outside and Fe $^{2+}$is given from conciliatory cast iron anodes. In this strategy, dissolvable Fe3+ can be cathodically reduced to Fe $^{2+}$. Thus, the quick generation of Fe $^{2+}$quickens the creation of Hydroxyl radicals (•OH). Fig 1 demonstrates the schematic chart of the experimental set up of EF process [16, 17]. The relevant reactions are specified in equation (6)-(9)

At anode:

$$Fe(s) \rightarrow Fe^{2+} (aq) + 2e^- \tag{6}$$

$$Fe^{2+} (aq) + 2OH^- (aq) \rightarrow Fe (OH)_2 (s) \tag{7}$$

$$\text{At cathode:} 2H_2O (l) + 2e^- \rightarrow H_2 (g) + 2OH^- (aq) \tag{8}$$

$$\text{Overall: } Fe(s) + 2H_2O (l) \rightarrow Fe (OH)_2 (s) + H_2 (g) \tag{9}$$

EF process can self-control the supply of a steady measure of iron particles up and down the response time and furthermore the simple reusing of the iron catalyst after treatment. It stays away from the utilization of a greater number of chemicals instead of customary Fenton treatment [18]. EF process has quick reaction rates as contrasted and other chemical treatments. In addition, electricity as a perfect energy source and it is utilized as a part of the procedure, so the general procedure does not make any secondary pollutants [19].

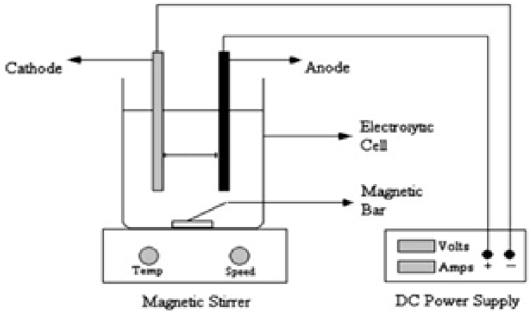

Fig 1: Schematic diagram of Electro-Fenton process

The non selective pathway permits the treatment of numerous organics at once [20]. The other basic points of interest of the electrolytic procedure are less maintenance (no moving parts) and exact control of the reaction. The need of substitution of the sacrificial anode, the conductivity of the pollutant stream and the utilization of electricity can be recorded as the procedure necessities. Yet, the monetary attainability of electrolytic procedures over their conventional counterparts has been established by many researchers.[21].

2. Materials and Methods

The landfill leachate samples were gathered from Municipal Solid Waste Disposal Facility at Brahmapuram, Kochi. The qualities of the leachate were analyzed according to standard techniques [22]. Every one of the chemicals utilized as a part of the examination was diagnostically immaculate. The qualities of original leachate acquired are given in table 1.

Table 1: Characteristics of landfill leachate sample collected from Brahmapuram Municipal Solid Waste Disposal Facility

Parameters	Content
pH	8.1
TDS	32760 mg/l
TSS	498mg/l
COD	23200 mg/l
BOD	4872 mg/l
Sulphate	587 mg/l
Phosphate	185mg/l
Sulphide	20.1 mg/l
Chloride	2670 mg/l
Ammonia nitrogen	2196 mg/l
BOD/COD	0.21

2.1. Experiment Design and Optimization

Response Surface Methodology (RSM) was utilized to enhance the procedure for the removal of Chemical Oxygen Demand (COD) as the objective response. pH, current density, H_2O_2 dosage and reaction time were considered as the significant parameters. Variable levels were resolved in view of both literature reports and preparatory runs to guarantee that the design points fell within the design space. The computation of H_2O_2 dosage depended on the stoichiometric ratio with respect to complete oxidation. H_2O_2 dosage fluctuated from 20% to 80% of the theoretical value. Additionally, the current density was varied from 15 A/m^2 to 150 A/m^2, the reaction time from 15min to 60min and pH from 2 to 4.

The experiment design, optimization, and the contour plots were proficient utilizing the Minitab 17 programming. Box

Benhken experiment design of Response surface methodology (RSM) was adopted to discover the connection between the response functions and variables. Sequentially, to examine the factors (pH, current density, H_2O_2 dosage and reaction time) required in the Electro Fenton reaction, 27 sets of experiments with differing working conditions as indicated by the test configuration were performed.

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2.2. Electro-Fenton Process

The EF experiment was conducted in a 1000ml beaker as a reactor with cast iron plates as electrodes. The measurements of electrodes were 12 cm x 6.5 cm x 0.1 cm. 800ml of the wastewater was filled in the reactor which plnges 55.25 cm^2 of the surface of electrode bringing about a particular electrode surface of $0.069 cm^2/cm^3$. The pH of the solution was balanced utilizing H_2SO_4. Hydrogen peroxide (H_2O_2) was added externally. The reaction mixture was blended completely utilizing magnetic stirrer. The separation between two electrodes was kept 2.5 cm. Examinations were conveyed by experimental design utilizing RSM 27 sets of investigations with changing working conditions were performed. Samples were gathered and examined for COD, Ammonia nitrogen, Chloride, Sulfate, Sulfide, and Phosphate. The best-working conditions giving most extreme pollutant removal was discovered utilizing Response optimizer. The trial was repeated at optimum working conditions four times so as to validate the performance of the process.

3. Result and Discussion

Table 2 demonstrates the design of experiments according to Box Behnken design and responses, for example, the removal of COD, Ammonia nitrogen, Chloride, Sulfate, Sulfide, and Phosphate. A second order polynomial model was fitted to the exploratory data for COD removal as given underneath.

$$Y = b_o + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_4 + b_{11}X_1^2 + b_{22}X_2^2 + b_{33}X_3^2 + b_{44}X_4^2 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{14}X_1X_4 + b_{23}X_2X_3 + b_{24}X_2X_4 + b_{34}X_3X_4 \quad (10)$$

Where Y is the predicted value, X_1, X_2, X_3, X_4 are the variables, b_o is a constant, b_1, b_2, b_3, b_4 are regression coefficients for linear effects. b_{11}, b_{22}, b_{33}, b_{44} are quadratic coefficients and b_{12}, b_{13}, b_{14}, b_{23}, b_{24}, b_{34} are the interaction coefficients.

The optimum level of every variable and the impact of their associations on the COD removal were considered by plotting two-dimensional contour lines (Fig 2-5). The figures depend on equation (10) with two variables kept steady at its centre point and changing the other two factors within the experimental range. COD removal increased with increment in treatment period. After that, COD removal was steady which might be because of the development of items which are not further degradable and likewise the reaction amongst H_2O_2 and Fe^{2+} was finished inside this time interim [23, 24]. Fig.2, 3 and 5 demonstrate that maximum COD removal is acquired at pH around 3. At lower pH iron species forms stable mixes with H_2O_2 which prompts deactivation of the iron catalyst. H_2O_2 is insecure in fundamental arrangement and it deteriorates quickly to O_2 and H_2O [25]. At lower pH, H_2O_2 would stay stable because of the formation of oxonium ion (H_3O^{2+}) [24]. COD removal was higher at a higher current density (Fig.2 and Fig. 4) which might be because of the higher hydroxyl radical concentration. As the concentration of H_2O_2 builds the hydroxyl radical concentration likewise increases. In any case, when the dosage of H_2O_2 was increased further, COD removal diminishes because of hydroxyl radical scavenging effect of H_2O_2 and development of hydroperoxyl radical ($HO_2\bullet$) which has lesser oxidation capacity than that of hydroxyl radical. Fig. 6 demonstrates the optimum estimations of every parameter. The optimum conditions acquired from the Minitab are pH = 2.85, current density = 140.5 A/m^2, H_2O_2 dosage = 53% and reaction time = 44 min.

Maximum COD removal is acquired at pH range 3 to 3.5 and reaction time more than 50minutes. COD removal was higher at H_2O_2 dosage around 60%. For COD removal, the acquired coefficient of determination (R^2) is 0.9506. This infers 95.06% of the varieties for percent removal of COD are clarified by the independent variables. Table 3 demonstrates the regression coefficients and a p value of various independent variables for removal of COD. Low p value shows that the corresponding coefficient value is more significant. For this situation, the linear effect and quadratic effect of the factors were huge with the exception of the linear effect of pH. Higher p value for interaction coefficients demonstrates their lesser influence. pH - reaction time and H_2O_2 dosage - reaction time were related and had a huge interactive impact.

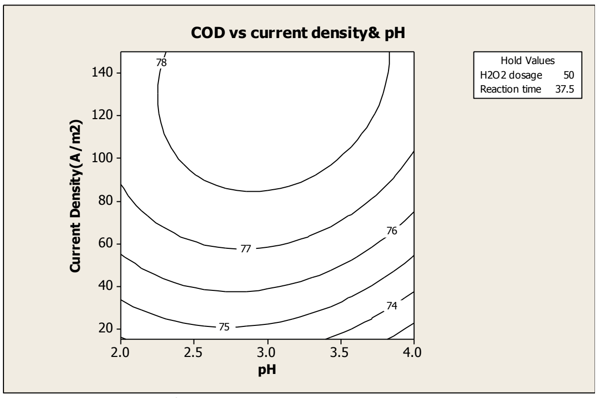

Fig. 2: Contour plot of COD removal at Reaction time -37.5 minutes and H_2O_2 dosage 50%

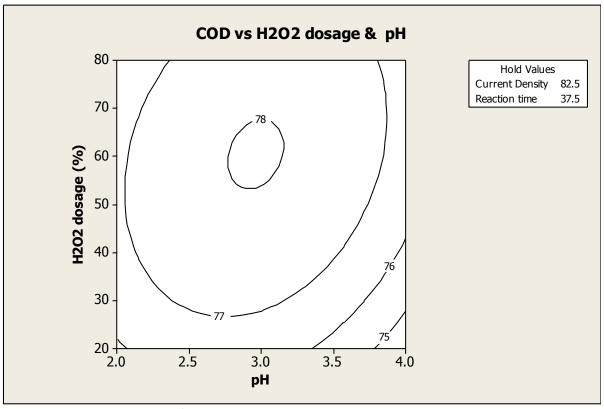

Fig. 3: Contour plot of COD removal at Reaction time -37.5 minutes and current density -82.5A/m2

Table 2: The design of experiment and experimental response for Electro Fenton treatment

SL. No:	pH	Current density (A/m^2)	Dosage (%)	Time (min)	Pollutant removal (%)					
					COD	Ammonia Nitrogen	Chloride	Sulphate	Sulphide	Phosphate
1	2	15	50	37.5	74.2	39.1	59	56.1	52	59.1
2	4	15	50	37.5	72	37.3	57.2	52.9	50	57.4
3	2	150	50	37.5	78.2	41.8	63.1	58.8	56	62.4

4	4	150	50	37.5	77.9	42	60.5	59	55	62.1
5	3	82.5	20	15	74.2	37.2	59.7	57.6	52.5	57.1
6	3	82.5	80	15	78.1	39.3	62.9	57.2	54.6	59.3
7	3	82.5	20	60	78	39	60.9	56.3	54	59.4
8	3	82.5	80	60	76.3	40.9	61	58.1	54	61.2
9	2	82.5	50	15	76.4	40	60.7	56.9	53	60.1
10	4	82.5	50	15	73.9	38.3	59.2	57.2	52	58.3
11	2	82.5	50	60	74.8	41.7	60	57.3	53	62.3
12	4	82.5	50	60	78	40.1	60.2	56.8	54	60
13	3	15	20	37.5	73.1	37.2	58.3	52	51	57.4
14	3	150	20	37.5	76.5	40.8	61	58.8	56.4	61.2
15	3	15	80	37.5	75	38.2	60	57	52.9	58.1
16	3	150	80	37.5	77.9	41	63.8	59.9	56.1	60
17	2	82.5	20	37.5	76	40	60	56.9	53.2	59.8
18	4	82.5	20	37.5	74.2	37.1	59.4	55.4	51.9	57.4
19	2	82.5	80	37.5	76	40	61.1	59.2	54	60.2
20	4	82.5	80	37.5	76.2	40.2	59.4	57.1	53.2	60.1
21	3	15	50	15	73	37.2	58.2	53.6	51.3	57.3
22	3	150	50	15	78.4	39.9	62.1	59.2	54	60
23	3	15	50	60	74	40.1	59	54	52	60.4
24	3	150	50	60	77.9	42	62.5	58.9	57.3	62.3
25	3	82.5	50	37.5	77.8	40.9	62.9	60	55.9	61.6
26	3	82.5	50	37.5	78	41	62.8	60	56.2	61.7
27	3	82.5	50	37.5	78	41.1	63	59.6	56.1	61.8

Table 3: Regression coefficients and corresponding p values for COD removal

Term	b_o	b_1	b_2	b_3	b_4	b_{11}	b_{22}	b_{33}	b_{44}	b_{12}	b_{13}	b_{14}	b_{23}	b_{24}	b_{34}
Regression Coefficient	62.7	3.93	0.07	0.151	0.054	-1.33	-0.0003	-0.001	-0.001	0.01	0.017	0.063	-0.0001	-0.0002	-0.002
P	0	0.15	0	0.005	0.044	0	0.001	0.008	0.03	0.16	0.144	0.001	0.703	0.264	0.001

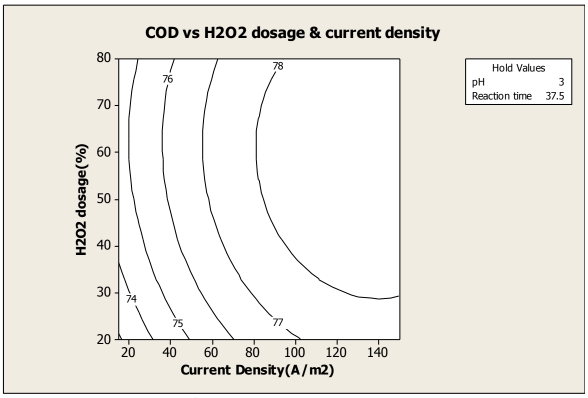

Fig. 4: Contour plot of COD removal at Reaction time -37.5minutes and pH-3

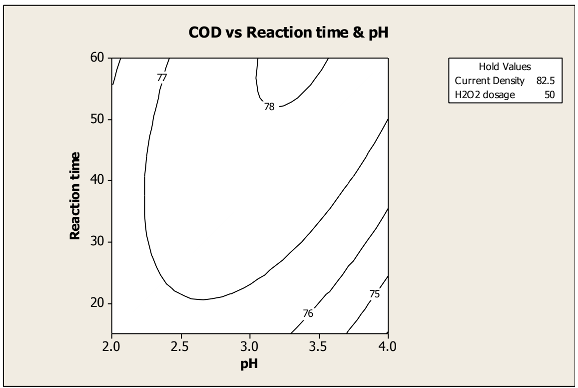

Fig. 5: Contour plot of COD removal at current density -82.5A/m^2 and H_2O_2 dosage 50%

The procedure was approved by running the experiment at ideal working conditions (Table 4).The average COD removal gotten after EF treatment is 78.2% i.e., the COD value was diminished to 5058mg/l and BOD to 2175mg/l. The test estimation of COD decrease practically coordinates with the anticipated COD removal percentage. Percentage decrease of different parameters is additionally close to the predicted values.

As indicated by the general standards for release of environmental pollutants into surface water Part-A: Effluents [Schedule VI] according to The Environment (Protection) Rules, 1986, Govt. of India, the COD ought to be under 250 mg/l. Thus, the leachate effluent after EF treatment can't be securely released into surface water. BOD to COD ratio increased from 0.21 to 0.43.

Table 4: Average result of experimental runs at optimal conditions

Parameter	Average Experimental value (%)
COD removal	78.2
Ammonia nitrogen removal	41.0
Chloride removal	62.2
Sulphate removal	59.0
Sulphide removal	55.8
Phosphate removal	60.9

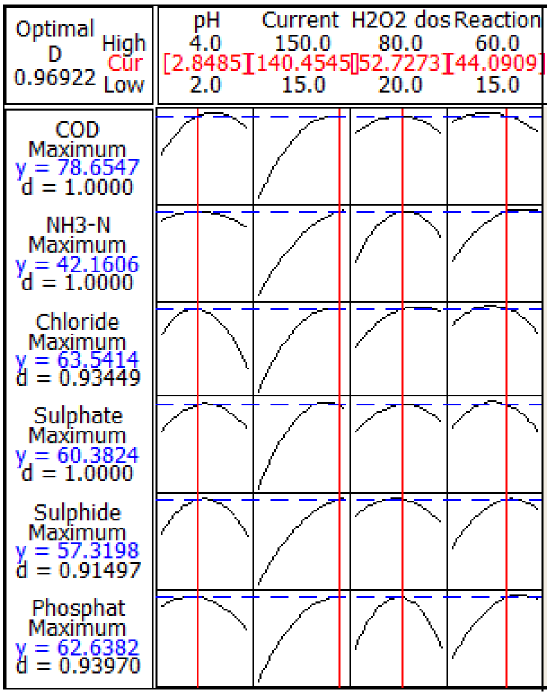

Fig. 6: The graph showing optimum values of the independent variables

For complete biodegradation, the effluent must have a BOD to COD ratio (biodegradability index) of no less than 0.4 [21]. This demonstrates EF process oxidizes the refractory natural or inorganic compounds of the landfill leachate in a more proficient way. Matured landfill leachate after EF treatment can be subjected to subsequent biological treatment for further degradation.

4. Conclusion

A laboratory examination was done to assess the appropriateness of Electro Fenton (EF) treatment of a matured municipal landfill leachate gathered from Municipal Solid Waste Disposal Facility at Brahmapuram, Kochi. The impacts of significant parameters on EF procedures and optimization were assessed utilizing Response surface methodology. The factors affecting EF process, pH, reaction time, current density, hydrogen peroxide dosage were completely explored and optimized. Exact prediction of COD removal of the test response and higher coefficient of determination (R^2=0.9506) demonstrate that the quadratic model satisfactorily depicts the response inside the exploratory region. The ideal conditions obtained from the Minitab programming are pH =2.85, current density = 140.5 A/m^2, H_2O_2 dosage = 53% and reaction time = 44 min for EF treatment. After EF treatment, the percentage removal obtained for COD, ammonia nitrogen, chloride, sulfate, sulfide, and phosphate are 78.2, 41.0, 62.2, 59.0, 55.8 and 60.9% respectively. BOD to COD ratio increased from 0.21 to 0.43 which shows that the biodegradability of the waste water has improved. In light of the important Indian Standards, the treated leachate effluent can't be released into surface water. It can be reasoned that the EF procedure can be utilized as a successful pretreatment technique for matured landfill leachate that is hard to treat with conventional biological treatment technologies.

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