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Research Article

Treatability of landfill leachate pre-treated with a two-stage biological treatment process to the level of receiving environment standards by using the fenton process

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ABSTRACT

Although two-stage (Anaerobic+Aerobic) biological treatment methods are applied to the landfill leachate formed in Istanbul-Kömürcüoda Sanitary Landfill, discharge standard to the sewer cannot be achieved. Treatability with the classical Fenton process, which can provide the discharge standard to the sewer and is applicable, is the subject of this study. In this context, batch treatability studies were carried out with the Fenton process for both raw landfill leachate and pilot biological treatment plant effluent samples. The independent variables selected in this study are pH, Fe^{2+} and H_2O_2 doses. The dependent variables are organic pollutant removal (as chemical oxygen demand) and change in biodegradable organic matter content (as biological oxygen demand). In the study, treatability studies were primarily carried out with the Fenton process for the two-stage biological treatment plant effluent. When 50% theoretical H_2O_2 dose was applied, chemical oxygen demand removal efficiency was calculated as 74.8%. The biodegradability rate of the remaining organic pollution (as 25.2 of COD) was determined as 23%. However, the pollutant removal efficiencies obtained in the treatability studies conducted with the batch Fenton process for both raw leachate and anaerobic treatment plot plant effluent samples remained at low levels. In addition, optimum chemical doses for the two-stage biological treatment plant effluent within the scope of the Fenton process were determined as 0.5 kg $Fe(SO)_4.7H_2O/m^3$ and 5 L H_2O_2/m^3 wastewater. According to the results obtained in these studies; the organic pollution load removal efficiency can reach 97.4% (in terms of chemical oxygen demand) with the Fenton process applied to the pretreated water coming out of the two-stage biological treatment. As a result, with the Fenton process option where the procedure specified in this study will be applied, it will be possible to treat the landfill leachate at a level that will meet the sewer discharge standard in terms of organic pollutant parameters. It is thought that the treatment process proposed here can be used in the treatment of all industrial wastewaters with similar properties.

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INTRODUCTION

In terms of waste hierarchy, sanitary landfill (SLF), also known as the final waste management option, is known as the traditional method that is needed regardless of which management option is applied. In addition to landfill gas, odor, and dust emissions, leachate is also formed in SLF facilities [1-3]. The leachate formed in SLF facilities contains a content that cannot be treated with traditional methods in terms of quantity and pollution intensity. Especially young landfill leachate contains quite complex, resistant, and dense pollutant types such as humus, ammonia nitrogen, heavy metals, inorganic salts, and organics [3-6]. Some factors affecting the amount and characteristics of leachate can be listed as the content of the stored solid waste (e.g. organic matter and moisture content), the depth of the landfill (assessment of groundwater and surface water exposure), the age of the stored waste (e.g. fresh domestic waste increases the leachate flow rate), top cover characteristics (permeability, slope, drainage status), operational conditions (e.g. compaction status of solid waste), climatic conditions (e.g. annual rainfall height, mm/year), hydrogeological conditions around the landfill and conditions in the landfill body [7]. The leachate flow rate likely to occur in SLF facilities is important in terms of planning treatment processes and varies from country to country and even from region to region in any country. In Denmark, leachate formation during operation is given as 350 mm/year, which is understood to be approximately 49% of the annual average rainfall height. In Sweden, the average leachate amounts formed by SLFs are 250-300 mm/year during the operation process, while this value is 10-40 mm/year for closed facilities with a clay top cover. The amount of leachate measured in SLFs in Istanbul, where leachate samples were taken within the scope of this article, varies between 730-1095 mm/year on average, and this value is approximately ten times the values estimated during the facility planning process [7]. Therefore, although literature information emphasizes that the amount of leachate likely to occur in SLFs will be 15-50% of the annual precipitation height, it is thought that it would be appropriate to take the average annual precipitation height in leachate estimates in Türkiye [7, 8]. SLF-related leachate must be treated at the level of the receiving environment standards. Otherwise, it is inevitable that irreversible negative situations will occur in the water, air, and soil environments around SLF [9-11]. In landfill leachate treatment, biological (anaerobic, aerobic, facultative), physical-chemical (chemical precipitation, chemical oxidation, flotation, air stripping, adsorption, coagulation-flocculation) and membrane (ultrafiltration, nanofiltration, reverse osmosis, membrane bioreactor) methods are generally used [12]. However, it is not possible to treat leachate at a level that will meet the receiving environment standards using traditional biological and chemical methods. Therefore, advanced oxidation methods must be used in SLF-related leachate treatment processes. Advanced oxidation methods can be evaluated within the scope of physicochemical treatment methods. One of the advanced oxidation methods is the Fenton reaction. The Fenton reaction plays an important role in converting organic pollutants into a range of products ranging from organic molecules to small inorganic molecules, thereby achieving complete degradation of pollutants and eliminating environmental pollution [13]. It is known that the Fenton reaction is successful in oxidizing many types of recalcitrant pollutants with its oxidation ability [14]. This success is due to the hydroxyl radicals (·OH) produced from H_2O_2 catalyzed by iron ions in acidic conditions and capable of removing recalcitrant organics in landfill leachate [15-19]. Transition metals (Fe, Cr, Mn) have catalytic effect ability resulting in faster oxidation reactions. In the presence of iron ions and relatively low pH values (pH: 3-4), H_2O_2 is converted to ·OH radicals. OH radical is the strongest oxidant except fluorine gas (F_2). In the treatability processes here, chemical oxygen demand (COD) and total organic carbon (TOC) were also taken into consideration as the two most important pollution indicators. Fenton reagents are effective on recalcitrant organic pollutants with the reaction mechanisms given in reactions from (1) to (9) [20-23].

$$\mathrm{Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \cdot OH + OH^-} \quad (1)$$

$$\mathrm{RH + \cdot OH \rightarrow H_2O + \cdot R} \quad (2)$$

$$\mathrm{Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2 \cdot + H^-} \quad (3)$$

$$\mathrm{Fe^{3+} + HO_2 \rightarrow Fe^{2+} + O_2 + H^-} \quad (4)$$

$$\mathrm{\cdot OH + H_2O_2 \rightarrow H_2O + HO_2 \cdot} \quad (5)$$

$$\mathrm{Fe^{2+} + HO_2 \cdot \rightarrow Fe^{3+} + HO_2 \cdot} \quad (6)$$

$$\mathrm{\cdot OH + Fe^{2+} \rightarrow Fe^{3+} + OH^-} \quad (7)$$

$$\mathrm{(COD/TOC)} + H_2O_2\mathrm{(Fe^{2+},Fe^{3+})} + H_2O_2 \rightarrow \text{Intermediate products} \quad (8)$$

$$H_2O_2\mathrm{(Fe^{2+},Fe^{3+})} + \text{Intermediate products} \rightarrow CO_2 + H_2O + \text{Inorganic salts} \quad (9)$$

All these reactions (1-9) can occur simultaneously in the water environment. While reactions (1) and (2) are desired to reduce organic pollution through hydroxyl radical activity, reactions (3)-(7) are undesirable because they produce parameters that are non-oxidant or have weak oxidation potential. pH, temperature, alkalinity, and the presence of electron-charged anions negatively affect the Fenton oxidation efficiency by reducing the hydroxyl radical. In order to increase the Fenton oxidation efficiency, pH optimization is required. The independent criteria for pH optimization, catalyst Fe(II) and oxidant H_2O_2 doses, should also be optimized. For some wastewaters and different leachate types, the optimum COD removal efficiencies obtained depending on the Fenton reactions and the independent criteria affecting the reactions are summarized in Table 1 [24-30].

The average values for the treatment performance of Fenton reagents on leachate types are given in Table 2 [32].

In this study, landfill leachate (LL) samples taken from Kömürcüoda Sanitary Landfill (SLF) facility, where approximately 10 tons per a day solid waste generated on the European side of Istanbul from 1994 to 2019 was regularly stored, were used. The treated water obtained as a result of the physical and biological treatment carried out in Kömürcüoda SLF can only meet the receiving environment standard by applying membrane processes. In membrane processes, there are problems of clogging and disposal of concentrate. Therefore, a lower-cost alternative treatment solution compared to membrane processes is the subject of this article by applying classical Fenton application before and after the physical and biological treatment in the SLF facility. In this study, it was planned to apply the classical Fenton process, one of the advanced oxidation methods, to the LL samples, which are well known to contain high-resistant organic matter. Here, controlled batch treatability studies were carried out to determine the levels of the main independent parameters (pH, H_2O_2, Fe^{2+}) that can reach the highest removal efficiencies for the main dependent criteria (COD and color removal efficiency). The main objective of this study is to analyze the treatability of both raw leachate samples and biological treatment plant effluent samples with the Fenton process. The treatability studies with the Fenton process were carried out separately for three different sample types: (1) raw landfill leachate from Kümürcüoda SLF, (2) pre-treated LL from anaerobic biological process effluent, and (3) pre-treated LL from anaerobic+aerobic biological process effluent available on Kümürcüoda SLF.

MATERIALS AND METHODS

In this study, the performance of Fenton Reactions, which are thought to provide an additional contribution to the insufficient biological treatment stages in landfill leachate treatment, was investigated. Raw leachate samples were obtained from Kömürcüoda SLF in Şile one of the districts on the Anatolian side of Istanbul. The wastewaters used in treatability studies, except for raw LL, were taken from the outlets of plot anaerobic and aerobic biological treatment reactors established for experimental purposes at Marmara University. The characterization of these samples is given in Table 3.

Table 1. Fenton's reaction reactant doses and COD removals for some wastewater types

Wastewater types	COD, mg/L	Fe(II), mg/L	H_2O_2, mg/L	COD removal, %	Fe(II)/COD, g/g	H_2O_2/COD, g/g	Ref.
[1]SS	1500	300	200	70	0.28	0.18	[18]
[2]SS	1100	900	900	63	1.30	1.30	[19]
[3]SS	1720	700	200	60	0.68	0.19	[20]
[4]AS	-	1500	1500	35	-	-	[21]
[5]SS	2000	200	1500	68	0.15	1.10	[22]
[6]AA	1190	100	1700	>95	0.09	1.50	[23]
[7]TE	250	200	200	78	1.03	1.03	[24]
[8]TE	590	-	310	86	-	0.53	[3]
[9]DWW	770	100	835	88	0.13	1.08	[31]

[1]Landfill leachate, upflow sequencing anaerobic reactor effluent; [2]Landfill leachate, Aerobic+ anaerobic reactor effluent; [3]Landfill leachate, aerobic reactor effluent; [4]Wastewater, upflow sequencing anaerobic reactor effluent; [5]Landfill leachate; [6]Aromatic amine production wastewater effluent; [7]Textile industry wastewater; [8]Textile industry wastewater, elektro-Fenton; [9]Domestic wastewater

Table 2. Fenton's reaction performance on landfill leachate types

Landfill leachate	H_2O_2/COD, g/g	H_2O_2/Fe(II), g/g	CODremoval, %	Reaction time, hour
Raw landfill leachate*	1.22 (n:10)	2.27 (n:12)	68.4 (n:19)	1.41 (n:14)
Pre-treated biologically	1.5 (n:11)	2.82 (n:12)	76.7 (n:16)	1.99 (n:16)

n: Runs numbers

Table 3. Characteristics of the landfill leachate used in the study

Sample types	**Characteristics of landfill leachate used in the study**				
	pH	**COD, mg/L**	**BOD_5, mg/L**	**Total Fe, mg/L**	**Color, Pt-Co**
Raw landfill leachate	6.5	17000	1400	85	460
Effluent from anaerobic process	6.7	6700	3800	59	410
Effluent from anaerobic +aerobic process	6.8	1750	620	21	142

In a study, it is emphasized that the independent criteria selected in experimental studies should be determined on a process basis [3]. In the study, the parameters that are widely used for the Fenton process and evaluated within the scope of the studies given in Table 1 were selected as independent process parameters. In this study, pH, H_2O_2 dose and Fe(II) dose were considered as independent criteria; and COD removal efficiency, color removal efficiency, turbidity removal efficiency and biodegradability rate were considered as dependent criteria.

Experimental studies were carried out in Ika-Werk brand jar test equipment. Five jars filled with 500 mL samples were used in each series of studies. Before the studies, the optimum pH was set as 3 for all sample types since it was reported in the literature that acidic pH conditions between 2 and 4 increase the oxidation performance of Fenton reactions [33, 34]. This data was also confirmed by the pH optimization study conducted within the scope of this study. Fe(II) was added to the leachate sample before H_2O_2 addition. H_2O_2 was added slowly to prevent possible foam formation. First, all samples were mixed at 300 rpm for one minute and then operated at 30 rpm for two hours. Considering the values given in Table 2 for both aged raw leachate and biologically treated wastewater, the reaction time was selected as two hours. The theoretical dose requirement of H_2O_2 for each sample was calculated from reactions (10) and (11) given below.

$$CH_4 + 2O_2 \cdot \rightarrow CO_2 + 2H_2O \tag{10}$$

$$CH_4 + 4H_2O_2 \rightarrow CO_2 + 6H_2O \tag{11}$$

According to reaction (10), 1000 mg O_2 can be taken as 1000 mg COD equivalent. In this case, H_2O_2/COD ratio can be calculated as 2.125 from reactions (10) and (11) (4*34/2*32= 2.125 g H_2O_2/g COD). According to this stoichiometric calculation, 2.125 mg H_2O_2 oxidant is required for each 1mg COD equivalent organic pollutant to be oxidized [31]. At the end of the reaction period, before COD, biological oxygen demand (BOD_5), color and turbidity measurements, H_2O_2 residue in the processed samples was removed. Removal of H_2O_2 residue was done by permanganometric method [35]. Residual H_2O_2 in the processed samples was removed by increasing the temperature of the processed sample to 70°C and pH to 10-11 before analysis. Then, the processed sample was left to settle for 40 minutes. COD, BOD_5, color and turbidity analyses were performed using samples taken from the upper clear phase of the sample after precipitation. The amount of Fe^{+2} remaining in the clear phase of the purified sample was determined to correct the COD value. The settled sludge volumes during and after the studies were also determined. $FeSO_4.7H_2O$ crystals were used as Fe^{+2} source and 30% H_2O_2 solution (d: 1.1 g/cm³) was used as H_2O_2 source. pH adjustments were made with 1N H_2SO_4 and 1N NaOH. Remaining H_2O_2 determinations were made with 0.01 N $KMnO_4$ solutions. All reagents used were selected as Merck quality. pH was measured with Jenway 3040 Ion Analyzer. Color and turbidity were determined with Merck SQ 118 spectrophotometer. Total iron analysis was performed with UNICAM 929 AAS device. All analyses in the study were performed in accordance with standard methods [36]. All studies were performed in three repetitions and the arithmetic means obtained were evaluated in this study. All processes implemented in this study are given in detail as a flow chart in Figure 1.

RESULTS AND DISCUSSION

In this section, the results of the treatability studies conducted with the Fenton process are presented and discussed considering the existing literature. The mentioned evaluations were made separately within the scope of the treatability of anaerobic+aerobic treatment plant effluent, anaerobic treatment plant effluent and raw young landfill leachate as three different wastewater types. The pH optimization studies conducted for the anaerobic treatment plant effluent are shown in Figure 1 and Figure 2.

According to Figure 2, the highest COD removal efficiency was obtained as 73% at the pH 3. Optimum treatment efficiency by Fenton process for the anaerobic treated effluent sample was achieved at pH 3. This optimized pH was used as an independent criterion level in the studies conducted here. Literature data also confirms these pH levels [33-34].

Figure 3 shows that the treated wastewater after the Fenton process is 30% biodegradable. The highest biodegradability rate (0.33) is obtained at pH 3 as 33% (or 0.33). This shows that in addition to the direct COD removal

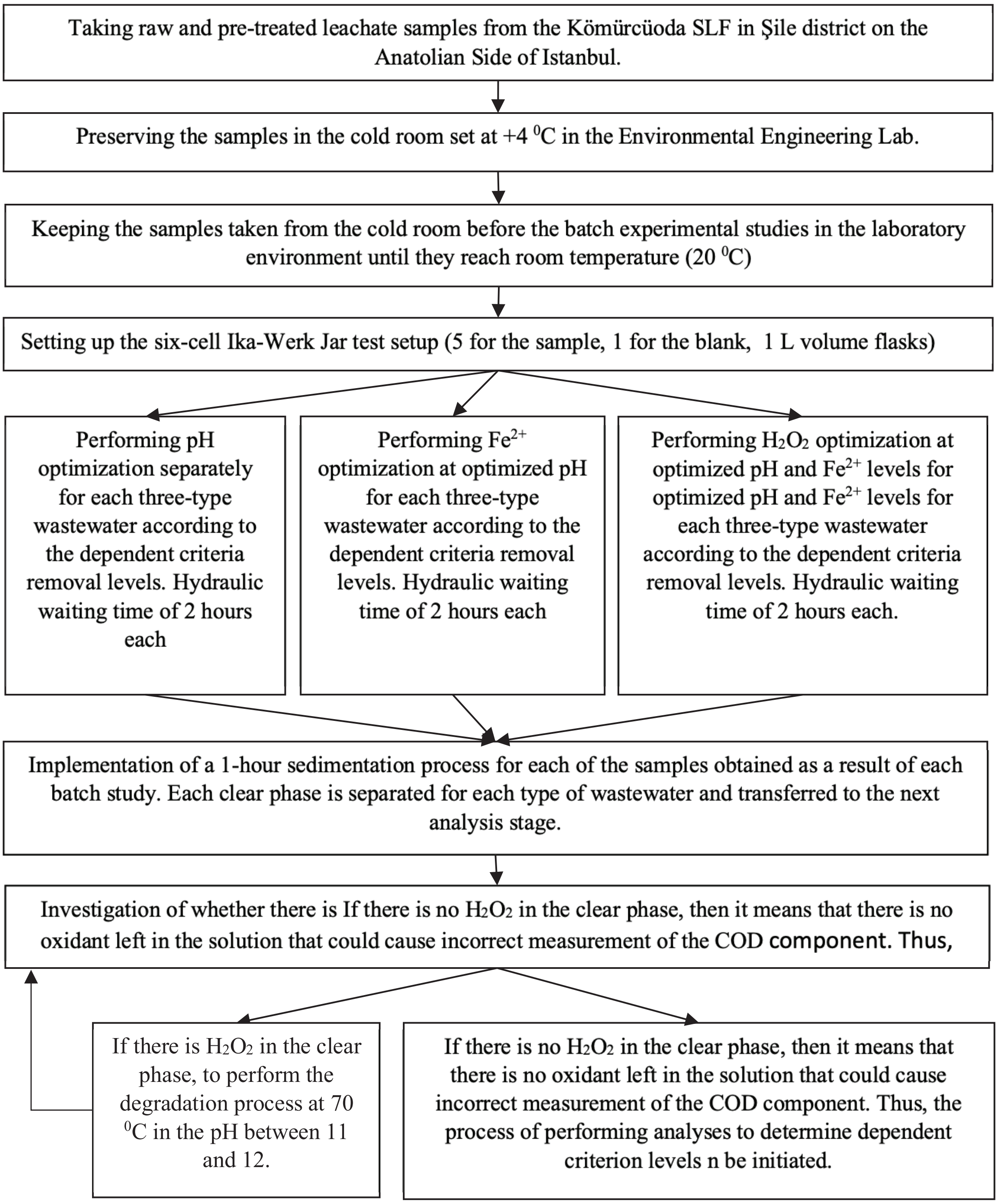

Figure 1. Flow diagram of the overall process for the current study.

effect of the Fenton process on the leachate, it also makes the remaining organic pollution partially biodegradable.

Results of Treatability Study Conducted with Fenton Process for Anaerobic+Aerobic Biologically Treated Landfill Leachate

The results of the treatability study conducted with anaerobic+aerobic biologically pretreated landfill leachate using Fenton process are given here. The results of the studies conducted to determine the optimum Fe^{2+} requirements are shown in Figure 4 (Working conditions; pH=3 and H_2O_2=2670 mg/L= 72 % of theoretical demand, COD= 1750 mg/L, reaction time:2 hour).

According to Figure 4, considering the consumed Fe^{2+} and the sludge volume to be produced, it would be appropriate to select the optimum value of 100 mg Fe^{2+}/L.

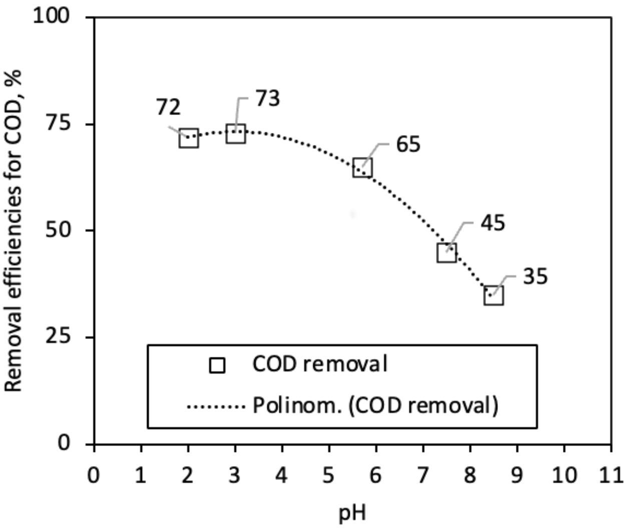

Figure 2. The results of pH optimization performed for COD removal in this study.

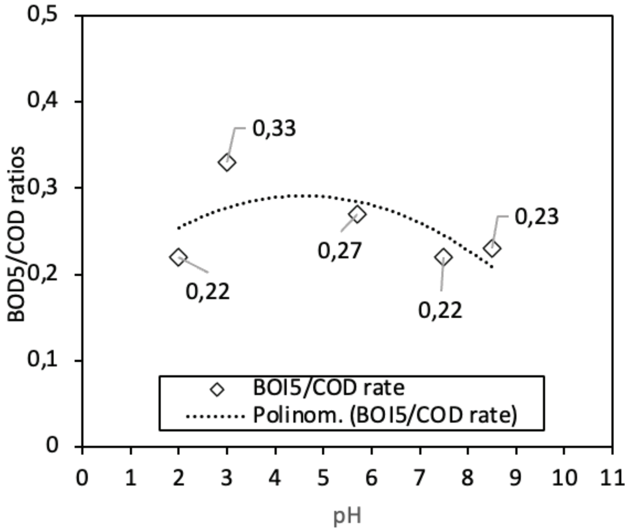

Figure 3. The results of pH optimization performed for BOD_5/COD ratios in this study.

Because, although there is a 3.9% increase in COD removal efficiency when a 200 mg Fe^{2+}/L dose is used, the amount of sludge that is likely to be formed may double. According to the results obtained from the studies, the removal efficiencies of COD, color and turbidity dependent variables were calculated as 76%, 99% and 99%, respectively. The final COD concentration decreased from 1750 mg/L to 440 mg/L. It was determined that 22.4% of the COD value (93 mg BOD_5/L) of the obtained treated landfill leachate (LL) was biodegradable. Optimum H_2O_2 doses, taking into account the previously determined optimum values of pH and Fe^{+2} the independent variables (pH: 3.0; Fe^{+2}: 100 mg/L) were given in Figure 5 for dependent parameters.

According to Figure 5, the optimum H_2O_2 dose is 1860 mg/L (50% of theoretical demand) and the optimum COD removal efficiency is 75%. The COD value of the treated sample with the Fenton process is 440 mg/L (Table 4). It is seen that this COD meets both the standards for sewer (<600 mg/L) and the standards for receiving environment standard [37]. In addition, it was understood in the analyzes that approximately 23% of the remaining COD is still biodegradable. Considering the 23% biodegradability level, the treated wastewater to be obtained after the Anaerobic + aerobic+ Fenton process can be biologically treated in large lagoons and the amount of organic matter can be further reduced. For the optimum Fe^{+2} dose (100 mg Fe^{+2}/L), the

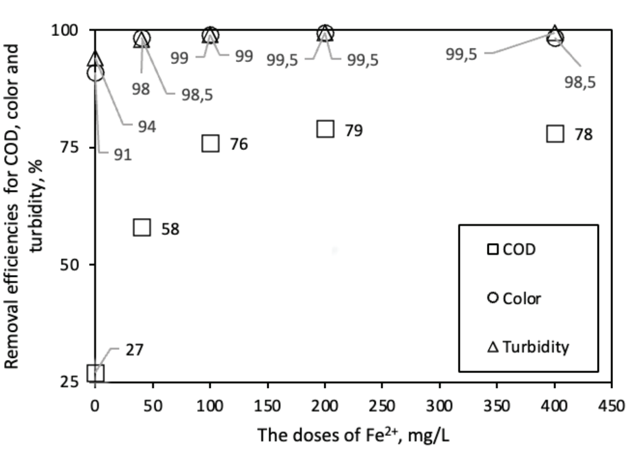

Figure 4. Studies to determine the optimum Fe^{2+} dose for the treated landfill leachate coming out of anaerobic+aerobic biological process.

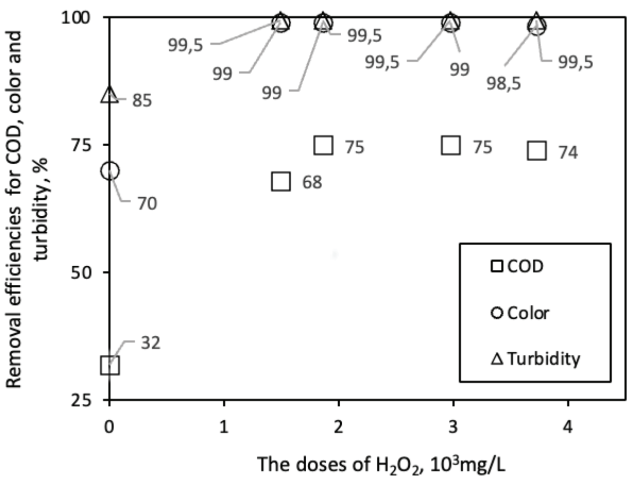

Figure 5. Studies to determine the optimum H_2O_2 dose for the treated landfill leachate coming out of anaerobic+aerobic biological process.

sludge volume obtained after 1 hour of sedimentation was obtained as 10% of the entire solution volume. In order to increase the COD removal efficiency, the following processes were specified for the clear phase obtained after the Anaerobic + aerobic + Fenton process + sedimentation:

-In order to get an idea about the additional removal possibilities of organic groups, 1000 mg/L of active carbon was added to the clear phase and mixed for 2 hours. The addition of active carbon provided an additional improvement in COD removal and the total treatment efficiency reached 83.5%. Therefore, as a result of the treatability studies, the final COD concentration could be reduced to 290 mg/L.

-The application of $KMnO_4$ at an amount of 1740 mg/L to the obtained clear phase for 2 hours was investigated. In this case, 85% COD removal and 260 mg/L final COD concentration were reached (reaction 12).

$$5CH_4 + 2\ 4H^- + 8KMnO_4 \rightarrow 5CO_2 + 8Mn^{2+} + 22H_2O \quad (12)$$

Using equations 10 and 12, the $KMnO_4$/COD ratio can be calculated as 4 ($KMnO_4$/COD = 8*158/10*32 = 4 mg $KMnO_4$/mg COD). As can be seen; $KMnO_4$ and activated carbon applications give good results in increasing COD removal efficiency, but still, the treatment of young landfill leachate to the level that will meet the required receiving environment standard (condition: COD<500mg /L [37]) can be provided.

The results obtained in this section by applying Fenton process to anaerobic + aerobic biological process effluent is compared with Table 1 in terms of COD removal level (75%), and given below:

-A higher COD removal efficiency was achieved than the COD removal efficiency (70%) obtained by applying Fenton process to pre-treated landfill leachate taken from upflow sequencing batch anaerobic reactor effluent [20]. While the amount of iron consumed is less (100 mg Fe^{2+}/L< 300 mg Fe^{2+} /L), the oxidant dose is higher (1860 mg H_2O_2/L>200 mg H_2O_2/L) than the specified study.

-A higher COD removal efficiency was achieved than the COD removal efficiency (63%) obtained by applying the Fenton process to pre-treated landfill leachate from the effluent of the aerobic + anaerobic reactor series [19]. While the amount of iron consumed is also less (100 mg Fe^{2+}/L<900 mg Fe^{2+}/L), the oxidant dose is higher (1860 mg H_2O_2/L>900 mg H_2O_2/L) than the specified study.

-A higher COD removal efficiency was achieved than the COD removal efficiency (60%) obtained by applying the Fenton process to the pretreated wastewater from the outlet of the aerobic biological reactor [20]. While the amount of iron consumed is also less (100 mg Fe^{2+} /L<900 mg Fe^{2+}/L), the oxidant dose is higher (1860 mg H_2O_2/L>900 mg H_2O_2/L) than the specified study. As can be seen, when compared to previous leachate treatability studies, the optimized Fe^{2+} dose in this study is lower. On the contrary, the oxidant dose obtained in this study is higher than other studies. However, the COD removal efficiency was obtained as 7.14% and 25% higher than the other studies mentioned. It is thought that the most important reason for the higher COD removal in this study compared to other studies is the high optimum oxidant dose used.

From experimental studies, it is seen that the optimum Fe^{2+} dose applied to anaerobic + aerobically pretreated wastewater is 100 mg/L. While the increase in the Fe^{2+} doses used does not increase COD removal significantly, it is understood that the amount of sludge increases in proportion to the iron used. While the Fenton reaction decreases the fraction of biodegradable organic matter that is likely to occur, it increases the inorganic fraction as $Fe(OH)_3$. The inorganic fraction does not cause any toxicity. On the contrary, it is known that Fenton has detoxification properties [13].

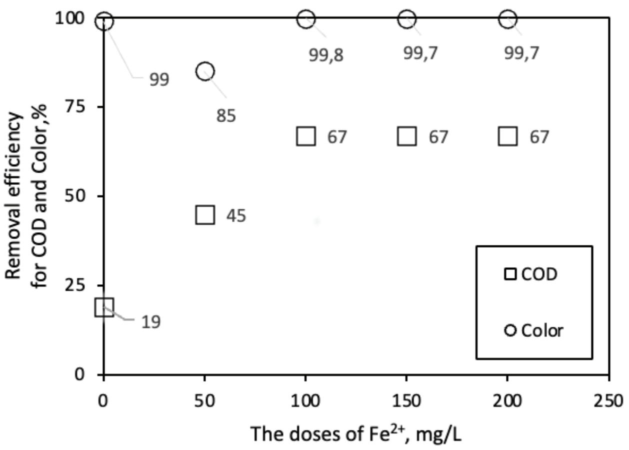

Figure 6. Studies to determine the optimum Fe^{+2} dose for the treated landfill leachate coming out of anaerobic biological process.

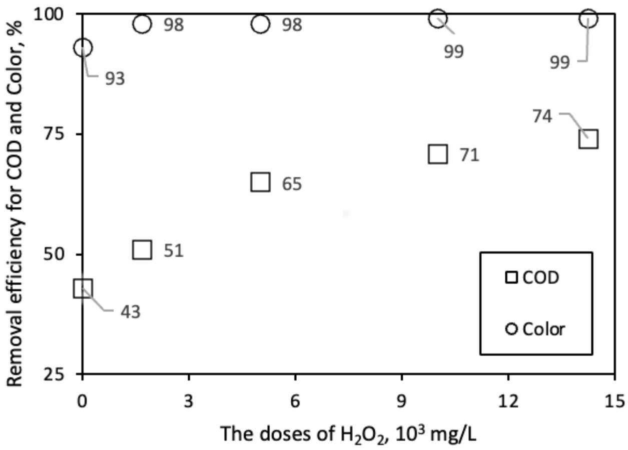

Figure 7. Studies to determine the optimum H_2O_2 dose for the treated landfill leachate coming out of anaerobic biological process.

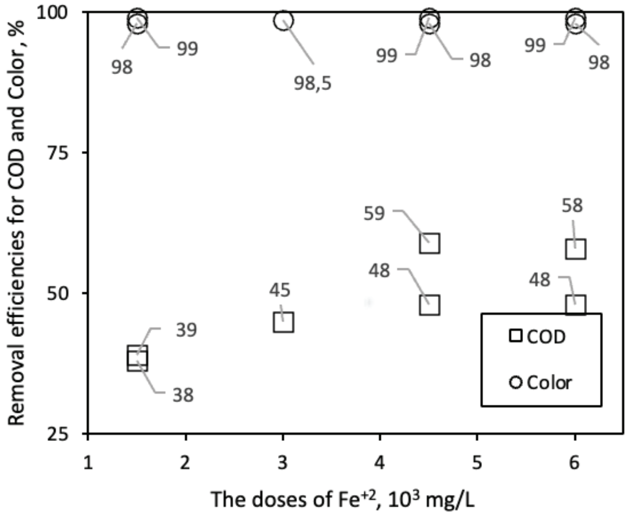

Figure 8. Studies to determine the optimum Fe^{+2} dose for the raw landfill leachate.

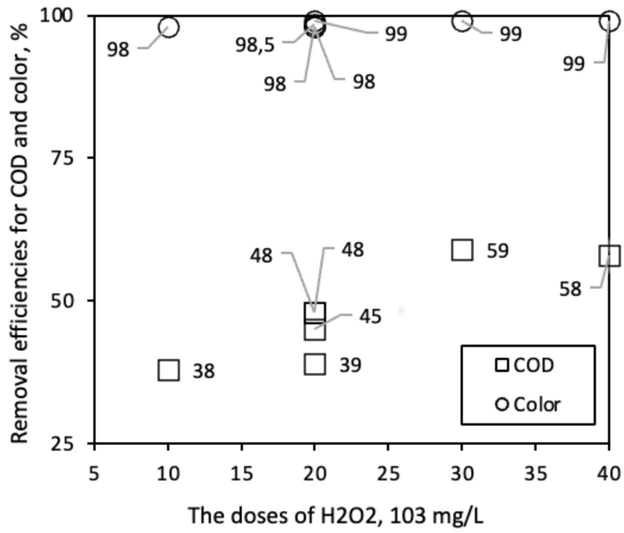

Figure 9. Studies to determine the optimum H_2O_2 dose for the raw landfill leachate.

Results of Treatability Study Conducted with Fenton Process for Anaerobic Biologically Pretreated Wastewater

In the studies conducted to determine the Fenton process efficiency for anaerobic treated leachate, optimum Fe^{2+} and H_2O_2 doses were determined and are given in Figure 6 and Figure 7, respectively.

For the special case of using optimum values of other independent variables (pH=3; 6660 mg H_2O_2/L= 47% of theoretical requirement; inlet COD= 6700 mg/L; reaction time: 2 hours), optimum Fe^{+2} dose was determined as 100 mg Fe^{+2} /L (Fig. 6). According to Figure 6, when 100 mg Fe^{+2}/L dose is applied to anaerobic treatment plant outlet samples, COD concentration in the clear phase after treatment decreases to 2190 mg/L (COD removal 67%). In addition, it is seen from Figure 6 that color removal efficiency is above 99% at the same Fe^{+2} dose.

For the special case where the optimum values of other independent variables are used (pH: 3, Fe^{+2}: 100 mg/L = 72% of the theoretical dose, inlet COD: 1750 mg/L, reaction time: 2 hours), the optimum H_2O_2 dose was determined as 5000 mg/L (Fig. 7).

As can be seen from Figure 7, when the H_2O_2 dose is 70% of the theoretical demand (10000 mg H_2O_2 /L), the COD removal efficiency is 71%. On the contrary, when the optimum dose, which is only 35% of the theoretical H_2O_2 dose, is applied, a COD removal efficiency of 65% is possible. As can be seen, in case of applying two times more H_2O_2, only a 9.23% increase in COD removal efficiency can be achieved. Therefore, the optimum H_2O_2 dose was accepted as 5000 mg/L in the section. In addition, in this study, biodegradability for the optimum H_2O_2 dose was approximately 50% (BOD_5: 1170 mg/L) and color removal efficiency was 99% (Fig. 7). In the determined optimum conditions, H_2O_2/COD (w/w), H_2O_2/Fe^{+2} (w/w) ratios were calculated as 0.75 and 50, respectively. At the end of the one hour settling period applied to all samples, the measured sludge volumes were obtained as 10% of all studies runs.

Results of Treatability Study for Raw Landfill Leachate Using Fenton Process

In the studies conducted to determine the Fenton process efficiency for raw landfill leachate, the optimum Fe^{+2} and H_2O_2 doses were determined and are given in Figure 8 and Figure 9, respectively.

As can be seen from Figure 8, when 20000 mg H_2O_2/L dose was applied to raw landfill leachate; COD removal efficiencies of 39%, 45%, 48% and 48% were obtained for 1500 mg Fe^{+2}/L, 3000 mg Fe^{+2}/L, 4500 mg Fe^{+2}/L and 6000 mg Fe^{+2}/L doses, respectively. Although the Fe^{+2}/L dose was increased by four times the initial dose (1500 mg/L H_2O_2/L, 6000 mg H_2O_2/L), only a 23.07% increase in COD removal could be obtained. On the other hand, when the same oxidant dose was applied (20 g H_2O_2/L), the COD removal efficiency was the same for both 4500 mg Fe^{+2}/L and 6000 mg Fe^{+2}/L doses (48%). Therefore, it is thought that 4500 mg Fe^{+2}/L can be accepted as the optimum Fe^{+2} dose. In the treatability studies conducted with the Fenton process for raw landfill leachate, no significant increase in COD removal was achieved by increasing the Fe^{+2} dose. It is understood that for doses higher than 6000 mg Fe^{+2}/L, no additional coagulation success will be provided in the removal of organics. From Figure 8, it is understood that the color removal efficiency is over 99% in all experimental studies conducted here. It was determined that the settled sludge volume in the experimental studies was at the level of 20% of the total sample volume.

As can be seen from Figure 9, when 15000 mg Fe^{+2}/L dose is applied to raw landfill leachate within the scope of Fenton process, 38% and 39% COD removal efficiency can

Table 4. Biodegradability results of raw landfill leachate and biologically pretreated landfill leachate before and after fenton process

Landfill leachate samples	Concentrations of pollutants before and after the application of the Fenton process			
	Before	After	Before	After
	COD, mg/L		BOD_5, mg/L	
Raw landfill leachate	17000	6850	14000	5950
Effluent from anaerobic process	6700	2380	3800	1170
Effluent from anaerobic+aerobic process	1750	440	620	101

be obtained for 10000 mg H_2O_2/L and 20000 mg H_2O_2/L oxidant doses, respectively. Despite doubling the oxidant dose, only a 2.63% increase in COD removal can be obtained. For the 4500 mg Fe^{+2}/L dose determined as optimum for Fe^{+2}, 48% and 59% COD removal efficiency can be obtained for 20000 mg H_2O_2/L and 30000 mg H_2O_2/L oxidant doses, respectively. Despite doubling the oxidant dose, only a 22.91% increase in COD removal efficiency can be obtained. According to Figure 9, when the oxidant dose is applied as 40000 mg H_2O_2/L, the COD removal efficiency decreases to 58% (for 6000 mg Fe^{+2}/L). Therefore, the optimum oxidant dose determined within the scope of the treatability of raw waste leachate, the properties of which are given in Table 3, by the Fenton process is 30000 mg H_2O_2/L. In other words, the optimum H_2O_2 dose was determined as 83% of the theoretical demand (For 17000 mg COD/L given in Table 3, theoretically 2.125*17000 mg H_2O_2 is required. The dose used here is 30000 mg H_2O_2/L, which is 83% of the theoretical dose). It is understood from Figure 9 that the color removal efficiency is above 98% in all experimental studies conducted here.

The effects of Fenton oxidation on biodegradability in raw and biologically pretreated landfill leachate samples are given in Table 4. Although the COD removal efficiency of the Fenton process seems to be higher in landfill leachate with low organic matter content (Anaerobic+aerobic pretreated effluent) compared to landfill leachate with high organic matter content (Raw young landfill leachate), the situation is exactly the opposite in terms of biodegradability. The BOD_5/COD ratios of the remaining COD load after Fenton application were calculated as 87% (5950/6850), 49% (1170/2380) and 23% (101/440) for raw landfill leachate, aerobic pretreatment effluent and anaerobic+aerobic pretreatment effluent samples, respectively. According to the obtained biodegradability results, it will be possible to meet the discharge criteria to the sewer with biological treatment following the Fenton process.

In this study, *firstly*; COD removal efficiencies obtained from the treatment of raw landfill leachate with Fenton process and anaerobic process were compared. COD removal efficiencies obtained from the treatment of raw waste leachate with Fenton process and anaerobic process were obtained as 59.7% and 60.6%, respectively. These results show that the same level of COD removal can be achieved with Fenton process and anaerobic biological processes. In BOD_5 removal, quite different removal levels were obtained (57.5% removal efficiency with Fenton process and 72.9% removal efficiency with anaerobic biological process). However, while the biodegradable level in the remaining organic matter load after Fenton process was 86.9%, this value could be only 56.7% as a result of anaerobic biological treatment. *Secondly*; COD removal efficiencies obtained from the treatment of anaerobically treated landfill leachate with Fenton process and aerobic biological process were compared. COD removal efficiencies obtained from the treatment of anaerobically treated waste leachate with the Fenton process and the treatment with the aerobic biological process were obtained as 64.5% and 73.9%, respectively. In BOD_5 removal, 69.2% and 83.7% levels were obtained for the Fenton process and the aerobic biological process. Although the BOD_5 removal efficiency was lower in the Fenton process than in the aerobic biological process, the situation is very different in terms of biodegradability levels. The remaining biodegradable organic matter levels after the treatment of anaerobically treated leachate with the Fenton process and the aerobic biological process were determined as 49.2% and 35.4%, respectively. In other words, it is understood that the amount of biodegradable organic matter remaining in the waste leachate treated with the Fenton process is 42.85% higher than in the aerobic biological process. *Thirdly*; After the application of the Fenton process to the two-stage biological treatment (Anaerobic + aerobic serial sequence) effluent sample, the basic pollutant removal and biodegradability levels were evaluated (Table 4 can be seen for the pollutant removal levels of each process). Two-stage biological treatment (anaerobic + aerobic) and anaerobic treatment and the application of a separate Fenton process after these processes can be evaluated in terms of non-biodegradable pollution. The remaining non-biodegradable pollution loads as a result of two-stage biological treatment and only anaerobic biological treatment were determined as 1130 mg/L (1750 mg/L -620 mg/L) and 1210 mg/L (2380 mg/L -1170 mg/L), respectively (Table 4). While the biodegradability levels of these non-biodegradable pollution loads were determined as 35.4% at the two-stage biological treatment effluent, this value could be increased to approximately 50% with the help of the Fenton process in this study. Although the COD removal efficiency is 90% with two-stage biological treatment, the discharge criteria to the sewer cannot be met. As a solution, with the help of the Fenton process that will follow the two-stage biological treatment, the wastewater can be treated at a level that will provide the receiving environment standard for the sewer, as in this study. Because, with the Fenton process, an additional COD removal of 75% (COD decreases from 1750 g/L to 440 mg/L with the Fenton process) can be achieved and the total COD removal reaches 97% (COD decreases from 17 000 mg/L to 440 mg/L).

CONCLUSION

In this study, the Fenton process was applied as an advanced oxidation technique to young wastewater leachates that had undergone two-stage biological (first anaerobic then aerobic) and single-stage (only anaerobic) biological treatment processes. The most important independent criteria affecting the process, pH, Fe^{2+} and H_2O_2 doses, were optimized, and the reaction time was limited to 2 hours. It was determined that with Fenton process, both additional COD removal could be provided in pretreated waste leachate during biological treatment process and a significant part of the remaining organic pollutant load could be made biodegradable. Receiving environment; it refers to the near or distant environment such as lake, stream, coastal and sea waters, and groundwater where wastewater is discharged or indirectly mixed. It is known that it is very difficult to treat landfill leachate to a level that will meet the standards of the receiving environment with conventional treatment methods. In this study, this fact was brought to light once again as a result of treatability studies applied to young landfill leachate (five-year-old). As follows; although the young landfill leachate was treated with both anaerobic and anaerobic+aerobic biological methods, COD could only be reduced to three times the standards of the receiving environment (1750 mg/L>500 mg/L). Therefore, in this study, the Fenton process was applied separately to both anaerobic pretreated landfill leachate and anaerobic+aerobic pretreated landfill leachate. The remaining COD in the pretreated landfill leachate after anaerobic+aerobic treatment was measured as 1750 mg/L. In order to provide the receiving environment standard, the Fenton process was applied to the pretreated landfill leachate after anaerobic+aerobic treatment. Fenton process applications were carried out within the scope of the values of the independent criteria selected for the process optimized in this study (pH: 3, Fe^{2+}: 4500 mg/L, H_2O_2: 30000 mg/L). After the Fenton process, the COD value of the treated leachate can be reduced to the level of 440 mg/L (<500 mg/L). As a result, when the Fenton process is applied to the two-stage biological pretreatment effluent, compliance with the receiving environment standard in terms of COD can be achieved. With this study, it will be possible to overcome receiving environment limitation by using the Fenton process in the landfill leachate treatment processes planned to be carried out in the future. In addition, the biodegradable content of the pollutant forms remaining after treatment by Fenton process will also enable a sustainable landfill leachate treatment process. In the studies performed here with three different wastewater samples, the optimum chemical and oxidant doses required for the treated LL were also determined. When the Fenton studies performed on anaerobic+aerobic outlet were evaluated for optimum efficiency; the chemical substance and oxidant doses were calculated as 0.5 kg $Fe(SO)_4.7H_2O/m^3$ wastewater and 8 L H_2O_2(30%)/m^3 wastewater. In anaerobic Fenton studies, the chemical substance and oxidant doses were calculated as 0.5 kg $Fe(SO)_4.7H_2O/m^3$ wastewater, 20 L H_2O_2(30%)/m3 wastewater. In the Fenton process studies performed with raw wastewater, the chemical substance and oxidant doses were obtained as 22.5 kg $Fe(SO)_4.7H_2O/m^3$ wastewater, 100 L H_2O_2(30%)/m^3 wastewater. From the obtained results, it is considered that the Fenton process is applicable only for anaerobic+aerobic effluent samples. Another chemical requirement in the Fenton process is to lower or raise the pH of the water to be treated. Because, the requirement to keep the pH between 3 and 4 in during process brings the cost of additional acid use to the agenda. Base addition may also be required after treatment. It is possible to use low pH industrial wastewater (e.g. metal industry) as an acid source in the treatment of wastewater with high pollution potential such as LL. In this case, it is important to control the heavy metal content of the wastewater planned to be treated. It is thought that detailed studies on the subject are needed for a sustainable wastewater treatment process. It is suggested that similar wastewaters, which cannot be sufficiently treated by conventional methods, should be treated with the classical Fenton process. According to the data obtained as a result of this study; it is clear that the classical Fenton process should be applied to the effluent, especially in facilities that cannot provide the receiving environment standard despite being biologically treated. Otherwise, membrane processes will need to be applied to provide the receiving environment standard, and this requires high costs due to the resulting concentrate and blockages in the system. It will be possible to increase the removal of organic pollutants by recycling the sluice output, whose biological treatability increases after the Fenton application, back to the biological processes. Detailed kinetic studies based on independent process parameters for similar wastewaters containing high organic pollution are considered as potential research areas based on the wastewater content.

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DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

STATEMENT ON THE USE OF ARTIFICIAL INTELLIGENCE

Artificial intelligence was not used in the preparation of the article.

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