Removal of organic contaminants from the water used to wash the tanks of trucks transporting diesel: electrocoagulation in batch mode

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abstract

The removal of chemical oxygen demand (COD) from the wash water of trucks transporting diesel by electrocoagulation using Al-Al and Fe-Fe electrode arrays with a spacing of 0.03 \overline{m} is a promising alternative. The experiments in batch mode were carried out with three current densities 80.2, 121.97, and 138.68 A/m². The initial pH of the washing water was 7.3. During the process it experienced changes of 0.6 and –0.8 with the Al-and-Fe electrodes, respectively. With a current density of 80.2 A/m2 , the COD removal efficiency stabilized in 20 min, reaching removals of 99.21% and 97.9% with Fe-and-Al electrodes, respectively. The energy consumption per unit volume of water treated and per unit mass of COD removed in both cases with Fe-and-Al electrodes were 3.97 and 4.03 kWh/m³ and 0.63 and 0.65 kWh/kg·COD, respectively. Electrocoagulation is a suitable process for the removal of organic contaminants from wastewater from washing the tanks of trucks that transport diesel fuel.

Keywords: Electrocoagulation; Al-and-Fe anodes; Chemical oxygen demand; Oil-and-grease removal

1. Introduction

Water is vital for all living organisms to exist [1–3]. Water is a limited resource and very sensitive to pollution caused by human activities and natural conditions. The quality and quantity of water are very important for human survival [4]. Many industrial activities generate water contamination with organic compounds (e.g., oil refineries etc.). Organic contaminants are persistent toxic substances that do not break down naturally in aquatic systems [5]. Animals, including humans, can bioaccumulate toxic organic compounds [5], triggering mutagenic diseases as well as cancer

[4,6,7]. Diesel fuels are complex hydrocarbon mixtures [8,9], containing all kinds of hydrocarbons: paraffins, naphthenes, aromatics, and in small concentrations, olefins [8,10]. Alkanes or paraffins, which are saturated hydrocarbons with straight or branched chains, but without any ring structure [11], are not soluble in water, which is highly polar [12]. Alkanes have densities between 0.6 and 0.8 g/cm^3 , so they are less dense than water [13]. Naphthenes are paraffins that have been "bent" into a ring or a cyclic shape [14]. They are saturated hydrocarbons containing one or more rings. Each of which may have one or more paraffinic side chains [11]. Aromatics are unsaturated cyclic compounds

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composed of one or more benzene rings. Crude oils from various origins contain different types of aromatic compounds in different concentrations [15]. Since diesel fuel is a mixture of numerous individual substances, absorption, metabolism, and excretion are very complicated and have not been fully characterized. Absorption of these substances can occur via all routes of exposure [16]. Petroleum refineries' wastewater contains high concentrations of oils, benzene, etc. [17,18], and is rich in biochemical oxygen demand (BOD) and chemical oxygen demand (COD) [19,20]. In this scenario, electrocoagulation (EC) has a wide range of applications and is capable of removing contaminants present in water in a wide pH range, from organic contaminants to heavy metals. EC is an effective process to destabilize finely dispersed particles [21] and remove hydrocarbons, greases, suspended solids, and heavy metals of wastewater [22]. Electrocoagulation is an attractive method due to its operational simplicity [23–25], and it does not require the addition of chemical coagulants [26], which are produced by the oxidation reaction of the anodic material. EC is a method of water and wastewater treatment that generates metal coagulant *in-situ* by electro-chemically dissolving sacrificial electrodes (anodes), mainly iron (Fe) and aluminum (Al) [27–29]. At the cathode, (Al or Fe) electrolyzes water producing H₂ gas and OH⁻ hydroxyl anions. The applied electric field generates the migration of metal cations and OH– anions, and they will combine to hydrolyze in solution to form hydroxide flocs with high specific surface area and rich surface hydroxyl groups. These hydroxide flocs have the ability to adsorb toxic substances in the water through the complexation formation, netting or bridging [30]. EC can remove organic [5,24,31,32] and inorganic [31,33–35] contaminants from water. The removal mechanisms of organic or inorganic contaminants by EC is influenced by electrodes used in the process, and in great detail the chemical equations are explained for aluminum [17,36–38] and iron [24,31,39]. The electrocoagulation with Al-and-Fe electrodes has an efficiency of 92.5%–99% to remove oil and grease

(O&G) [40,41]; it has an efficiency of 60%–98% to treat COD [42–44]; it has an efficiency of 62.5%–97% to treat turbidity $[45,46]$; and, it has an efficiency of 85%–99.7% to remove total suspended solids (TSS) [47,48]. In addition, maintenance and operation of the system is simple. The objective of this research work was to remove the COD by electrocoagulation from the washing water of tank trucks that transport diesel, considering the Peruvian Standard DS 010-2019 Ministry of Housing.

2. Materials and methods

2.1. Sample collection

Fig. 1 shows the wash water coming out of the tank truck. For this research work, a 300 L sample of wash water (wastewater) was collected. The experimental development was carried out at the company, and the analyses were carried out by an external laboratory accredited and recognized by Peruvian Environmental Institutions. The different parameters of the washing water (wastewater) were analyzed according to the following methods: Standard for O&G (SM 5520-B: Liquid–Liquid, Gravimetric Partition Method); Standard for BOD (SM 5210-B: biochemical oxygen demand, 5 days); Standard for COD (SM 5220-D: Closed Reflux, Colorimetric Method); Standard for pH (SM 4500 H+B: pH Value Electrometric Method); Standard for TSS (SM 2540-D: total suspended solids dried at 103°C–105°C); and, Standard for NTU (SM 2130-B: Nephelometric Method). SM: Standard Methods for Examining Water and Wastewater APHA, AWWA, WEF 22nd ed., 2012. Table 1 shows the characteristics of the wastewater to be treated. The COD, oil and grease and BOD exceed the Peruvian Standard by a factor of 6.4, 60.8 and 6.0, respectively.

2.2. Experimental set-up

Fig. 2 shows the batch system for the electrocoagulation process. The power source is (0–20 V/0–100 A). The reactor

Fig. 1. Washing water from tankers transporting diesel.

Table 1

Physico-chemical characteristics of wastewater from washing the tank of a tanker truck that transports diesel fuel and maximum admissible concentration according to the Peruvian Standard

Parameter	Symbology	Wastewater	Peruvian Standard DS 010-2019 Ministry of Housing
Oil and grease, mg/L	O&G	6,076	100
Chemical oxygen demand, mg/L	COD	6,369	1,000
Biochemical oxygen demand (mg/L)	BOD ₅	2,997	500
Total suspended solids (mg/L)	TSS	263	500
pH		7.3	
Turbidity (NTU)	NTU	950	
Conductivity $(\mu S/cm)$		2,880	

Fig. 2. The applied scheme for experimental development. (1) DC power supply; (2) reactor; (3) electrodes; (4) electrolyte; (5) ball valve; (6) treated wastewater.

is built with 0.01 m thick plexiglass and its dimensions are $0.19 \text{ m} \times 0.13 \text{ m} \times 0.25 \text{ m}$, with a working volume of 5 L. It also has a 1/2-inch diameter ball valve. The dimensions of the electrodes are $0.133 \text{ m} \times 0.15 \text{ m} \times 1.6 \text{ mm}$. The reactor has three anodes (0.1197 m^2) and two cathodes, and the separation between the electrodes is 0.03 m. The anode– cathode electrodes in each case were organized as follows: Al-Al and Fe-Fe.

2.3. Wastewater conductivity

Table 2 shows the effect that adding NaCl has on the conductivity of the wastewater to be treated. The conductivity of the solution is enhanced by adding anions in the form of salts, in our case NaCl. When adding 6.67 g/L of

Table 2 Effect of adding NaCl on the conductivity

NaCl, the conductivity was 26,522 µS/cm and the EC process developed steadily over time. The reactions in the electrodes – with an oxygen (anode) and a hydrogen (cathode) formation and in the effluent with a sludge formation that settles and floats towards the surface of the reactor – were constant. This parameter was measured with a HACH Model HQ14d conductivity meter (Colorado, USA). The conductivity provided by the supporting electrolyte has a direct influence on the current density. It is directly related to the efficiency of contaminant removal and the energy consumption of the EC process [49]. There are several electrolytes that can be used to improve conductivity including NaCl, KI, $CaCl₂$, etc. [50]. The role of the electrolyte is to increase the conductivity of the solution by reducing the cell voltage due to reducing the ohmic resistance of wastewater [24,51]. NaCl is commonly used for this purpose [17,52–55]. Water is a polar solvent, thus ionic compounds, as, for example, NaCl, dissolve and dissociate to form Na⁺ and Cl⁻ ions, which are electrolytes [56,57].

2.4. Energy consumption

Table 3 shows the energy consumption during the EC process. Eq.(1) was applied to calculate the energy consumption per unit volume of wastewater treated during the experiments (kWh/m³) with Al-and-Fe electrodes.

$$
\frac{E}{V_R} = \frac{V \times I \times t}{V_R} \tag{1}
$$

where *V* is voltage used in the process; *I* is intensity of the applied current (A); *t* is reaction time (h); V_p is reactor volume (m^3) .

Time	Voltage (V)		Ampere (A) Energy consumption (kWh/m^3) Voltage (V)		Ampere (A)	Energy consumption $(kWh/m3)$			
(min)	Aluminum			Iron					
$j_1 = 80.2$ A/m ²									
$\mathbf{0}$	θ	θ	0.0000	$\mathbf{0}$	$\boldsymbol{0}$	0.0000			
5	6.5	9.6	1.0400	6.7	9.6	1.0720			
10	6.4	9.6	2.0480	6.4	9.6	2.0480			
20	6.3	9.6	4.0320	6.2	9.6	3.9680			
40	6.3	9.6	8.0640	6.3	9.6	8.0640			
j_2 = 121.97 A/m ²									
$\mathbf{0}$	θ	$\mathbf{0}$	0.0000	$\mathbf{0}$	$\mathbf{0}$	0.0000			
5	9.8	14.6	2.3847	9.9	14.6	2.4090			
10	9.8	14.6	4.7693	9.8	14.6	4.7693			
20	10	14.6	9.7333	10.2	14.6	9.9280			
40	10.1	14.6	19.6613	10.2	14.6	19.8560			
j_3 = 138.68 A/m ²									
$\mathbf{0}$	θ	θ	0.0000	$\mathbf{0}$	$\mathbf{0}$	0.0000			
5	15.4	16.6	4.2607	15.5	16.6	4.2883			
10	15.5	16.6	8.5767	15.5	16.6	8.5767			
20	15.1	16.6	16.7107	15.3	16.6	16.9320			
40	15.3	16.6	33.8640	15.4	16.6	34.0853			

Table 3 Energy consumption with Al-and-Fe electrodes

Tables 4 and 5 show the energy consumption (kWh) per mass unit (kg) of pollutant removed oil and grease, COD, BOD and TSS, with Fe-and-Al electrodes.

3. Results and discussion

3.1. Effect of current density on pH with Al-and-Fe electrodes

Fig. 3 shows pH-performance at electrocoagulation times. The pH of the solution plays an important role in the electrocoagulation process as well as in chemical coagulation [58,59]. The EC process with Fe-electrodes causes this parameter to change rapidly to an acid condition. At the end of the process, its value remains in a range of 6 to 6.4. The process with aluminum electrodes has a slight increase in pH, maintaining an alkaline condition. At the end of the process (40 min), this parameter remains in a range of 7.4 to 7.8. A similar effect with Al-electrodes is observed in the research work carried out by Alkurdi and Abbar [55].

The processes involved with Fe-electrodes [60,61] are: Reaction at alkaline pH:

$$
2Fe_{(s)} + 6H_2O_{(l)} \to 2Fe(OH)_{3(s)} + 3H_2(g)
$$
 (2)

Reaction at neutral pH:

$$
3Fe_{(s)} + 8H_2O_{(l)} \to Fe(OH)_{2(s)} + 3Fe(OH)_{3(s)} + 4H_2(g)
$$
 (3)

Reaction at acid pH:

$$
2Fe_{(s)} + 6H_2O_{(l)} \rightarrow 2Fe(OH)_{2(s)} + O_{2(g)} + 3H_{2(g)}
$$
(4)

In an alkaline medium, ferric hydroxide Fe(OH), $[(Eq. (2)]$ is formed. If the medium is neutral, ferrous hydroxide $(Fe(OH)_2)$ and ferric hydroxide $[(Eq. (3)]$ are formed, both compounds contribute to floc formation. When the process develops in an acid condition ($pH > 6$) $[(Eq. (4)]$ ferrous hydroxide is formed. When the process is carried out with Fe-electrodes, by electrochemical reactions at the anode [(Eqs. (2) and (3)], Fe^{2+} and Fe^{3+} ions are formed, which hydrolyze in the water and form monomeric and polymeric compounds, which depend on the pH [62]: $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)_2^+$, $Fe(H_2O)_4(OH)_2^+$, $Fe_2(H_2O)_8(OH)_2^{4+}$, $\text{Fe}_{2}(\text{H}_{2}\text{O})_{6}(\text{OH})_{2}^{4+}$, $\text{Fe}(\text{OH})_{4}^{-}$ [63]. The pH and the concentration of dissolved oxygen have a strong influence on the oxidation of Fe²⁺ ions to Fe³⁺ ions [64]. In neutral media or alkaline media, $Fe²⁺$ is immediately transformed into ferrous hydroxide, which is quickly oxidized by dissolved oxygen to iron(III) hydroxide [Eq. (1)]. In acidic media, Fe²⁺ cations oxidize very slowly in contact with dissolved oxygen [62]. Aluminum at pH 4–9 forms complex compounds and polymers through the hydrolysis process. Some of these compounds are: Al(OH)²⁺, Al(OH)₂⁺, Al(OH)₂⁺, Al(OH)₃⁺, Al₆(OH)³⁺₁₅⁺, Al₇(OH)⁴⁺₁₇⁺ $Al_8(OH)_{20'}^{4+}$ and $Al_{13}(OH)_{32}^{7+}$ [17,24,38,65,66]. These cationic compounds are transformed into $AI(OH)_{3}$ [Eq. (5)] [58] and have a large surface area [65] which helps in the rapid adsorption of organic compounds and capture of colloidal particles [38].

$$
Al_{(aq)}^{3+} \to Al(OH)_{n}^{3-n} \to Al_{2}(OH)_{2}^{4+}
$$

\n
$$
\to Al_{13}complex \to Al(OH)_{3}^{4+}
$$
 (5)

Time (min)	Energy consumption (kWh/kg·O&G)	Energy consumption (kWh/kg·COD)	Energy consumption (kWh/kg·BOD)	Energy consumption (kWh/kg·TSS)				
$j_1 = 80.2$ A/m ²								
$\boldsymbol{0}$	0.0000	0.0000	0.0000	0.0000				
5	0.1780	0.1707	0.3675	4.9174				
10	0.3385	0.3248	0.6985	8.9825				
20	0.6553	0.6279	1.3428	16.6025				
40	1.3314	1.2772	2.7308	33.4606				
j_2 = 121.97 A/m ²								
$\boldsymbol{0}$	0.0000	0.0000	0.0000	0.0000				
5	0.3980	0.3828	0.8191	10.6123				
10	0.7874	0.7550	1.6134	20.3818				
20	1.6383	1.5706	3.3405	41.1950				
40	3.2776	3.1453	6.6855	83.0795				
j_3 = 138.68 A/m ²								
θ	0.0000	0.0000	0.0000	0.0000				
5	0.7076	0.6809	1.4483	18.4842				
10	1.4148	1.3575	2.8829	36.1885				
20	2.7941	2.6787	5.6953	69.9669				
40	5.6265	5.3984	11.4804	142.0222				

Table 5

Energy consumption per mass unit of pollutant removed with Al-electrode

These flocs $(Al(OH)_{3})$ can polymerize as shown in [Eq. (6)] [17,66].

$$
n\text{Al}\left(\text{OH}\right)_3 \to \text{Al}_n\left(\text{OH}\right)_{3n} \tag{6}
$$

The Fe(OH)_n [24,60] and Al(OH)₃ [67] form gelatinous suspensions flocs [17,68] interactive with the pollutant present in the wastewater to form sludge as shown in [Eq. (7)]:

$$
Pollutant + M(OH)n \to Sludge
$$
 (7)

3.2. Effect of current density on oil-and-grease removal with

Al-and-Fe electrodes

Fig. 4 shows the behavior of the oil-and-grease removal percentage. When the current density is 80.2 A/m^2 in 20 min, the oil-and-grease removals with Fe-and-Al electrodes were 99.65% and 99.57%, respectively. When the EC process time is 40 min, the removals were 97.96% and 99.14%, respectively. In both cases there is a slight decrease in the removal yield. When the current density is 121.97 A/m², the removal percentage with Fe-and-Al electrodes in 20 min were 99.25% and 99.01%, respectively, in 40 min of process the yields are 99%. With a current density of 138.68 A/m^2 , the removals in 20 min were 99.25% and 99.11% and in 40 min 99% with Fe-and-Al electrodes, respectively. Removals efficiencies in 20 min with both electrodes are high > 99%, so it is not necessary to increase the EC process time. The energy consumption per unit volume of treated wastewater (Table 3) as well as unit weight of pollutant removed (Tables 4 and 5) in 20 min at 80.2 A/m^2 were 3.97 and 4.03 kWh/m³ and 0.66 and 0.68 kWh/kg·O&G with Fe-and-Al electrodes, respectively. The initial pH value of 7.3, in 20 min of process at 80.2 A/m2 with Fe-and-Al electrodes changes to 6.5 and 7.9, respectively. Other studies report similar removal values for this parameter, for Al (>96%) [69,70] for Fe (99%) [41]. The removal efficiencies of oil and grease were calculated according to Eq. (8).

$$
\text{O&G Removal}(\%) = \frac{\left[\text{O&G}\right]_{o} - \left[\text{O&G}\right]_{t}}{\left[\text{O&G}\right]_{o}} \times 100\tag{8}
$$

Fig. 3. Effect of current density on pH. Fig. 4. Effect of current density on oil and grease removal.

Fig. 5. Effect of current density on chemical oxygen demand removal.

where $[O\&G]$ _{*o*} and $[O\&G]$ _{*t*} are the oil and grease at time initial $(t = 0)$ and at *t* (reaction time), respectively. A relationship similar to 7 was applied to calculate the COD, BOD, turbidity, and TSS removal percentages.

3.3. Effect of current density on COD removal with Al-and-Fe electrodes

Fig. 5 shows the COD removal behavior. This parameter has a high percentage of removal in 20 min of EC. The COD removal efficiency stabilizes after 20 min of the EC process. With a current density of 80.2 A/m², the removal percentages were 99.21% and 97.9% with Fe-and-Al electrodes, respectively. Al has a behavior similar to Fe only at 121.97 and 138.68 A/m². When the EC time is 40 min, with the Fe-electrode, the removal is 99%. For the Al-electrode, the removals were 97.96% with 80.2 A/m² and 99% with 121.97 and 138.68 A/m2 .

The formation of ferric hydroxide with the capacity to remove contaminants from water occurs in a range of $6 < pH < 9$. Fe(OH)₃(s) is stable and insoluble at neutral pH and can adsorb and remove hydrocarbon molecule complexes from wastewater using van der Waals forces [68]. Using Fe-and-Al anodes, COD removals > 95% of waters containing hydrocarbons are reported [17], which is consistent with this research work. Oil refinery COD removal from wastewater by electrocoagulation in a pH range (5–9)

in 30- and 60-min using Al-electrodes were (80%–88%– 82%) and (88.4%–96.8%–92.8%), respectively [55]. In this research work with Al-electrodes, 80.2 A/m², pH 7.9-7.8 with 20–40 min of process, 97.90%–97.96% COD removal was achieved. The energy consumption per unit weight of pollutant removed (Tables 4 and 5) in 20 min at 80.2 A/m2 were 0.63 and 0.65 kWh/kg·COD with Fe-and-Al electrodes, respectively.

3.4. Effect of current density on BOD removal with Al-and-Fe electrodes

Fig. 6 shows the BOD removal profiles with Al-and-Fe electrodes. The removal percentages are similar >98% at 20 and 40 min of the EC process. At 80.2 A/m2 using Al-and-Fe electrodes in 20 min, removals of 98 and 98.60% are achieved, respectively. With 40 min of process the yields were 97.93% and 98.53%. When the experiment was carried out with Al-electrodes at 121.97 A/m² in 20 and 40 min, removals of 99.17% and 99.10% were achieved, and with Fe-electrodes the yields were 99.17% and 99.10%, respectively. With 138.68 A/ m2 using Al-electrodes the yields were 99.43% and 99.27%, and with Fe these values were 99.20% and 99.07%, respectively. The BOD removal efficiency stabilizes after 20 min of EC process for each current density. The energy consumption per unit weight of pollutant removed (Tables 4 and 5) in 20 min at 80.2 A/m2 were 1.34 and 1.37 kWh/kg·BOD with Fe-and-Al electrodes, respectively. The removal percentage of this parameter is reported in the literature for wastewater from different industries: dairy wastewater (Al-electrodes, 60 min, 98%) [71]; palm oil mill effluent (Fe-electrodes, 60 min, 73%–91%) [72]; slaughterhouse wastewater (Al-electrodes, 120 min, 56.4%) [73].

3.5. Effect of current density on TSS removal with Al-and-Fe electrodes

The behavior of the TSS removal efficiency is shown in Fig. 7. This parameter also reaches a stable level of contaminant removal after 20 min of the process, and efficiencies >90% are obtained. The removal efficiencies achieved during this time with current densities j_1 , j_2 and j_3 were 90.11%, 91.63% and 92.02% with Al-electrodes and 90.87%, 91.63%, 92.02% with Fe-electrodes, respectively.

Fig. 6. Effect of current density on biochemical oxygen demand removal.

When the treatment is 40 min, a slight decrease of 1% in performance is observed 89.73%, 90.87% and 91.25% with Al-electrodes and 91.63%, 90.87% and 91.25% with Fe-electrodes, respectively. Tables 4 and 5 show the energy consumption kWh/kg·TSS removed with Fe-and-Al electrodes. The high levels of turbidity removal >90% are in agreement with the efficiencies achieved >90% for TSS. Turbidity is often assumed to be a surrogate for TSS [74–76]. It is generally true that the higher the TSS then more suspended particles are expected, and the turbidity should be higher [74]. The research work developed by Jasim and AlJaberi [77] reported 94% TSS removal treating oily wastewater at 35 mA/cm², 120 min of treatment, pH 6.

3.6. Effect of current density on turbidity removal with Al-and-Fe electrodes

Fig. 8 shows the turbidity removal profiles. High removal yields (>91%) of this parameter are obtained in 20 min. The turbidity removal efficiency stabilizes after 20 min of EC process for each current density. For Al with 80.2 A/m2 in 20 and 40 min the removal efficiencies were 93.68% and 93.89%, and for Fe these values were 92.11% and 91.58%, respectively. For Al in 20 min of EC process with 121.97 and 138.68 A/m2 the removal efficiencies were 94.42% and 94.74% and for Fe these values are 93.16% and 94.54%, respectively. In 40 min of process with 121.97 A/m² the removal efficiencies were 94.32% and 92.63% for Al and Fe and with

Fig. 7. Effect of current density on total suspended solids removal.

Fig. 8. Effect of current density on turbidity removal.

Table 6

Concentrations achieved with the EC process and Peruvian Standard

138.68 A/m2 the values are 94.42% and 94.21%, respectively. For the treatment of oily wastewater, efficiencies greater than >97% are reported with Al-electrodes [36,78–80]. For slaughterhouse wastewater the efficiencies are 99% and 88.5% in 25 min with Al-and-Fe electrodes, respectively [81].

3.7. Effect of the lowest current density on the Peruvian Standard parameters

Table 6 shows the concentrations achieved with the EC process, with the lowest current density $j_1 = 80.2$ A/m² in 20 min of treatment. The Fe-electrode proved to be more efficient than the Al-electrode for the removal of contaminants from diesel. However, both electrodes successfully satisfy the parameters established by the Peruvian Standard (Table 1).

4. Conclusions

Electrocoagulation is a suitable process for the removal of organic contaminants from wastewater from washing truck tanks that transport diesel fuel. Both Al-and-Fe electrodes have high levels of contaminant removal. The removal efficiency achieved at 80.2 A/m² and 20 min of treatment was >99%, for oil and grease, COD, BOD, and turbidity, and slightly higher than 90% for TSS. The control parameters of the washing water of tank trucks that transport diesel, after the electrocoagulation process, are below the standard that is required by the Peruvian Standard. Thus, the objective of the work was achieved.

Conflicts of interest

The authors declare no conflict of interest, financial, or otherwise.

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