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ORIGINAL ARTICLE

Removal efficiency of nickel and lead from industrial wastewater using microbial desalination cell

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Abstract Microbial desalination cell (MDC) is a new method of desalination. Its energy is supplied through microbial metabolism of organic materials. In this study, synthetic samples were provided with concentration of 25, 50, 75, 100 mg/L Ni and Pb. Removal efficiency of each metal was analyzed after 60, 90, 120 min, psychrophilic, mesophilic, thermophilic and 3–4, 4–5, 5–6 mg/L dissolved oxygen. Optimum conditions for removing Ni and Pb were achieved in 100, 4.5 and 4.6 mg/L dissolved oxygen, respectively, 26 °C and 120 min. Nickel and led were removed from wastewaters of Isfahan electroplating industry and steel company. The maximum removal efficiencies of Ni and Pb in real samples were 68.81 and 70.04%. MDC can be considered as a good choice for removing Ni and Pb from industrial wastewater. Due to microorganisms for decomposing organic material in municipal wastewater, metals from industrial wastewater can be removed simultaneously.

Keywords Microbial desalination cell · Isfahan electroplating wastewater · Isfahan steel company wastewater

Introduction

Large volumes of wastewater with high pollution intensity are produced by different processes annually and they must comply with discharge regulations before being discharged into the environment (Min et al. 2005). Many ways, including conventional treatment systems, have been proposed for wastewater treatment, but this process requires high setup costs and energy consumption (Oh and Logan 2005, Ghangrekar and Shinde 2007). This shows that humans are dramatically energy-dependent (Liu et al. 2004; Logan 2004; Rabaey and Verstraete 2005; Lovley 2006; Mohan et al. 2007).

Chemical and physical processes are used for the removal of heavy metals from wastewater, such as ion exchange (IX), redox, precipitation, and ultrafiltration (Bitton 2004). Such methods include nickel (II) removal from industrial plating effluent by Fenton process (Malakootian et al. 2015), use of hybrid nanoparticles of Tio2/Sio2 in removal of lead from paint industry effluents (Malakootian et al. 2012), and photocatalytic processes using silica and zirconia nanoparticles in the bivalent nickel removal of aqueous solutions and determining the optimum removal conditions (Malakootian and Cholicheh 2012).There is investigation of Fe_3O_4 nanoparticles modified with orange peel efficiency in removal of lead and copper ions from aqueous environments (Malakootian et al. 2014), Pb and Co removal from paint industries effluent using wood ash (Malakootian et al. 2008)were used for removing metals from industrial wastewater.

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Sometimes, microorganisms obtain methods for separating and removing metals that can replace physical and chemical methods (Bitton 2004). For this reason, some studies have been carried out to collect renewable energy from organic waste sources and it is well known that microorganisms can produce fuels, such as ethanol, methane, hydrogen and electricity from organic materials (Shukla et al. 2004; Oh and Logan 2005; Kim et al. 2005; Liu et al. 2005; Logan and Regan 2006).

Microbial desalination cell (MDC) is a new method of desalination and its energy is supplied through microbial metabolism of organic materials. In other words, organic matter decomposition and energy production are conducted by microorganisms in water solution and chemical oxidation regardless of whether it is organic or inorganic substrate (Maier et al. 2000; Kim and Logan 2013; Brastad and He 2013). The first MDC that is proposed for desalination consists of three parts of anode, cathode and desalination chamber between the anode and the cathode (Cao et al. 2009).

In the anode, exoelectrogenic microorganisms oxidize organic materials. During the oxidation process, electrons are removed from the substrate and transferred to the terminal electron acceptor (TEA, i.e., cathode) from the electron transport chain. In the cathode, oxygen usually acts as the TEA. The reaction of these electrodes creates an electrical potential gradient (Maier et al. 2000, Kim and Logan 2013).

In the electric field created, anions are absorbed by the anode and cations are led to the cathode. Water desalination is done in the middle chamber by IX membranes. AEM and CEM are located after the anode and cathode, respectively (Kim and Logan 2013).

Exoelectrogenics is a feature that is granted to the bacteria. "Exo" means outside the cell and "electrogens" means the ability to transfer electrons to insoluble electron acceptors (Zuo et al. 2008).

The concept of MDC was first introduced by Cao et al. (2009) on a small scale (capacity of 3 mL of salt water) and later by Jacobson et al. with larger scale (approximately, 1 L salt water capacity) (Cao et al. 2009; Jacobson et al. 2011a, b). A Chinese study showed reduction of hexavalent chromium by MDC method by 75.1% (An et al. 2014).

In this study, the removal of nickel (Ni) and lead (Pb) was investigated by MDC method. Polyester fabrics such as AEM and CEM were used. In most other studies, Gel polystyrene has been used. Ion exchange capacity of AEM and CEM was 1.6 and 1.9 meq/g. For swelling membranes, demineralized water instead of NaCl is used.

Materials and methods

Materials and reagents

Anion exchange membranes (fumasep FTAM-E) and cation exchange membranes (fumasep FTCM-E) were purchased from FuMA-Tech GmbH Company of Germany. Carbon graphite electrodes were purchased from Pariz Fan Company in Iran. All chemical materials, including phosphate buffer, nickel (II) chloride hexahydrate, lead (II) nitrate, sulfuric acid, sodium hydroxide were purchased from Merck.

Analytical methods

The quantity of removed metals in each sample was measured by atomic absorption spectrometric device (Younglin AAS 8020 manufactured by YL Instrument South Korea). The pH, EC and turbidity were measured by pH meter, EC meter and turbid meter, respectively. Experiments, such as TSS, BOD and COD were performed in accordance with procedures noted in standard methods for examinations of water and wastewater 2540 D, 5210 and 5220 Cr. Sulfate, ammonia and volatile fatty acids (VFCs) were performed in accordance with 4500 D, 4500 E and 5560 C. Cu, Zn, Cr, Ni, Cd, Pb and Hg were performed in accordance with 3111 B and CN were performed in accordance with 4500 D. (A.P.H.A. et al. 1999). SPSS, version 16 software was used for data analysis.

Experimental procedure

This experimental study was conducted on 108 synthetic samples containing Ni and 108 synthetic samples containing Pb, and real samples containing Ni and Pb from wastewater of plating industries and Isfahan steel company.

First, a batch system reactor was built of Plexiglas with a thickness of 10 mm and outside dimensions of length, width and height were 48, 18 and 18 cm, respectively. Useful volume of MDC was about 8 L. Included in the reactor was a bioelectrochemical chamber or anode and an electrochemical chamber or cathode with aerobic conditions and a middle chamber. Inside dimensions of the anode, cathode and middle chambers were each $14 \times 14 \times 14$ cm^3 and the volume of each of them was about 2.6 L. Inlet and outlet of all three chambers were located at the top and the bottom of each chamber, respectively.

These three chambers were separated by an anion exchange membrane (AEM) and a cation exchange membrane (CEM) with dimensions of 18×18 cm^2. Then AEM was located between the anode chamber and the middle chamber, and the CEM between the middle chamber and the cathode chamber, and the membranes were soaked for 48 h in deionized water. To prevent membrane damage caused by the fluid pressure in chambers, the membrane was placed between two perforated backing Plexiglass plates.

The anode and cathode electrode made of carbon graphite were in the form of a rectangular cube with the dimensions of 4 cm length, 1 cm width and 14 cm height. The electrodes were connected to each other and then to a digital ohm meter device by copper wires. It should be noted that the electrodes were soaked in deionized water for 24 h before applying.

The anolyte in anode chamber included a return activated sludge from the Kerman wastewater treatment plants as a source of microorganisms, and municipal wastewater in aeration tanks as a source of organic matter. The catholyte in the cathode chamber included 0.1 M phosphate buffer. The cathode chamber was aerated by aquarium aeration pump and the amount of dissolved oxygen (DO) was measured by a digital DO meter. The middle chamber included a synthetic sample containing Ni and Pb that were used from nickel (II) chloride hexahydrate and lead (II) nitrate, and its pH was set on 7 sing 0.1 M sulfuric acid and 0.1 M sodium hydroxide. The MDC pilot that was used during operation is shown in Fig. 1.

In Fig. 2a, b its performance and supporting plates are shown, respectively.

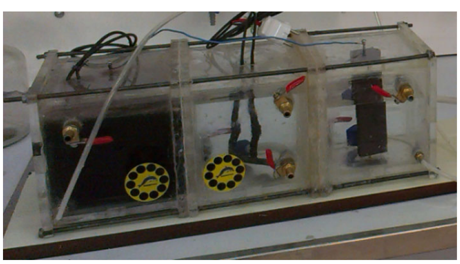

Fig. 1 MDC pilot

To study the removal efficiency, a 25 mL sample was taken from the outlet of the reactor in each case. Experiments for any of the tested concentrations of metals were conducted in the retention times of 60, 90 and 120 min and at different psychrophile temperatures (0–20 °C), mesophile (20–35 °C) and thermophile (40–55 °C) and the amount of DO in cathode chamber of 3–4, 4–5 and 5–6 mg/L.

The results were read by a spectrophotometer atomic absorption device. Experiments were carried out under the same conditions on wastewater of Isfahan electroplating industry and wastewater of Isfahan steel company as real examples with determined quality. At the end, each metal removal rate was calculated from Eq. (1).

$$\eta = \frac{C_0 - C_1}{C_0} \times 100 \qquad (1)$$

Where η is the removal efficiency, C_0 and C_1 are the primary and secondary concentrations of metal. Data analysis was performed using descriptive statistics.

Results

Dissolved oxygen and Ni and Pb removal

The effect of DO on the removal of Ni and Pb in synthetic samples by MDC method is shown in Fig. 3.

Increased DO in the cathode chamber, Ni and Pb removal rate was initially increased and then decreased. Maximum efficiency of removal of Ni and Pb, occurred in 4.4 mg/L DO. Removal rate of Ni and Pb in 3.46, 5.44 mg/L and 3.49, 5.48 mg/L DO was decreased because the number of TEA in cathode chamber and nutrient in anode chamber was decreased, respectively.

Retention time and Ni and Pb removal

The effect of retention time on the removal of Ni and Pb in synthetic samples by MDC method is shown in Fig. 4.

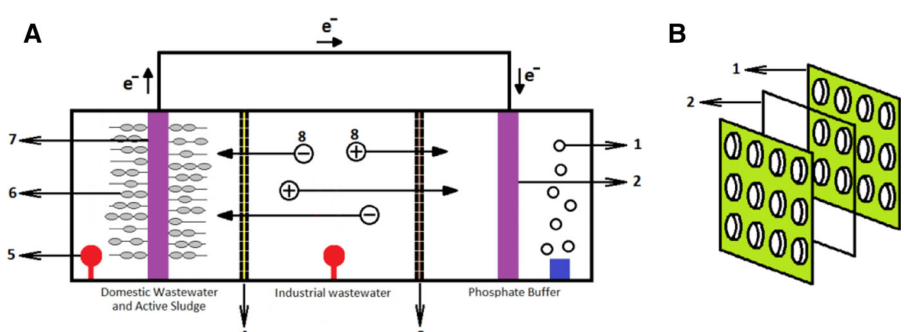

Fig. 2 **a** The process of MDC: (1) oxygen bubbles; (2) cathode electrode (in cathode chamber); (3) CEM; (4) AEM; (5) heater; (6) exoelectrogenes bacteria; (7) anode electrode (in the anode chamber); (8) cations and anions. **b** AEM and CEM supporting plates (1) supporting (2) membrane

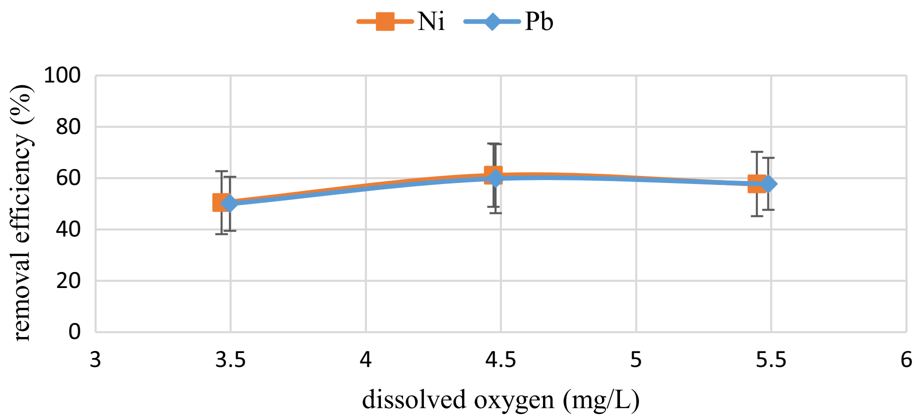

Fig. 3 The relationship between DO and removal of Ni and Pb by MDC method in synthetic sample

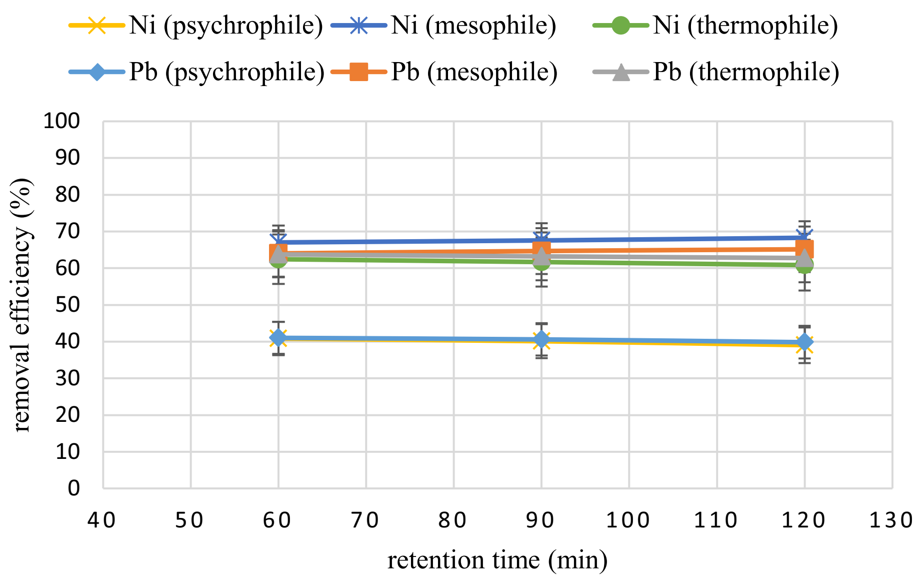

Fig. 4 The relationship between retention time and removal of Ni and Pb by MDC method in synthetic sample

Increasing retention time in the psychrophile and thermophile phases decreased the removal of Ni and Pb, and in the mesophile phase, the removal rate was increased because there were suitable conditions for microorganisms' metabolism.

Temperature and Ni and Pb removal

The effect of temperature on the removal of Ni and Pb in synthetic samples by MDC method is shown in Fig. 5.

As the temperature increases from psychrophile phase to mesophile phase, the removal of Ni and Pb was increased, and then it was decreased from mesophile phase to thermophile phase.

The temperature is one of the parameters affecting microbial growth and survival. Since the temperature inside a bacterial cell is similar to the temperature of the surrounding environment, increased environment temperature increases microbial activity to the point that rising temperatures will cause the loss of cells (Zazouli and Bazrafshan 2009). Therefore, the voltage produced by exoelectrogenic bacteria during the removal of Ni and Pb by MDC method in psychrophile and thermophile phases is declined due to poor growing conditions and the removal of Ni and Pb are also declined.

Concentration and Ni and Pb removal

The effect of the concentration on the removal of Ni and Pb in synthetic samples by MDC method is shown in Fig. 6.

A direct relationship was observed between concentration and removal of Ni and Pb, and increasing concentration increased removal due to the presence of more ions in synthetic solution. Maximum efficiency of removal of Ni and Pb occurred in 120 mg/L concentration. Removal efficiency of Pb is more than Ni, because electron capacity of Pb (4+) is more than Ni (3+).

According to the statistical analysis of the synthetic sample of Ni and Pb, with 0.94 and 0.99 line slope, the maximum efficiency of removal of Ni and Pb in the

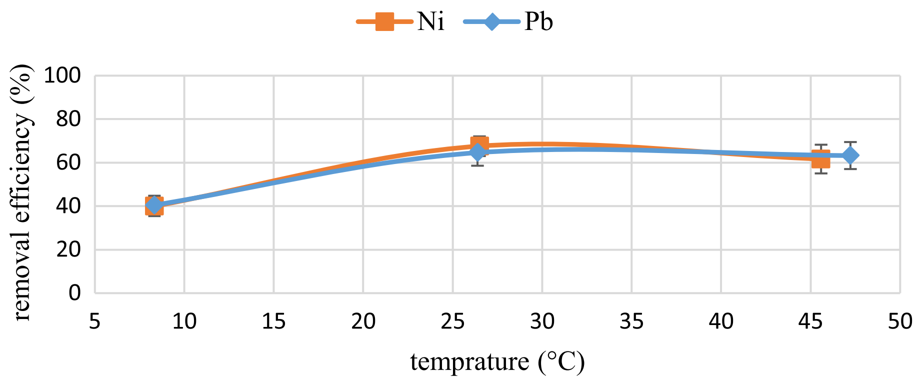

Fig. 5 The relationship between temperature and removal of Ni and Pb by MDC method in synthetic sample

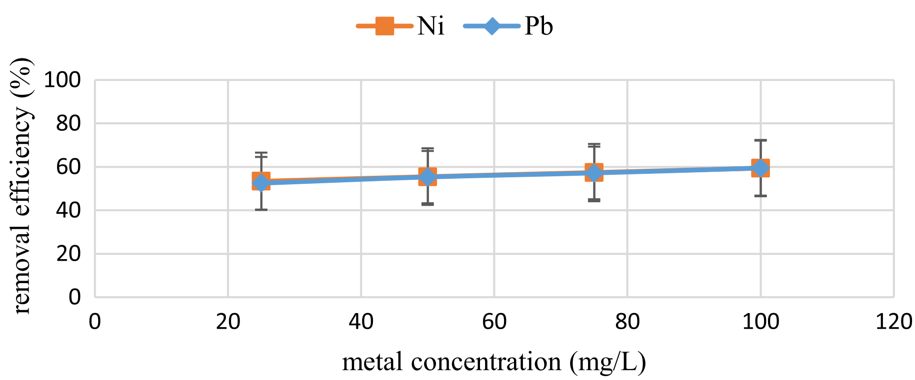

Fig. 6 The relationship between concentration and removal of Ni and Pb by MDC method in synthetic sample

Table 1 Quality of wastewater of Isfahan electroplating industries

pH	EC (ds/m)	Turbidity (mg/L)	TSS (mg/L)	BOD (mg/L)	COD (mg/L)	SO_4^{2-}(mg/L)
6.1	14.8	90	128	140	310	11.26
NH_4^+ (mg/L)	VFC (μg/L)	Cu (mg/L)	Zn (mg/L)	Cr (mg/L)	Ni (mg/L)	CN (mg/L)
2.91	422	14.22	3.85	14.51	24.43	0.012

concentration of 100 mg/L, and the DO of 4–5 mg/L, at 20–35 °C and retention time of 120 min was 74.5 and 75.1%, respectively. The highest voltage produced by the conditions listed above was 0.88 and 0.89 V, respectively.

Electroplating is a common industry in Isfahan. Metals enter wastewater through washing and electroplating tanks. So if the process of electroplating is modified, metals cannot discharge in the wastewater. The quality of wastewater of Isfahan electroplating industries that was used as a real solution containing Ni was obtained as in Table 1.

Steel is a major industry in Isfahan. Wastewater types produced from this industry are domestic, industrial, salty and phenolic wastewater. We choose industrial wastewater because it contains heavy metals. The quality of wastewater of Isfahan steel company that was used as a real solution containing Pb was obtained as in Table 2.

The results of applying optimal conditions on real samples are provided in Table 3.

Discussion

Malakootian et al. (2011) in Iran, in a study aimed at producing electricity through simulated food industry wastewater treatment using two-chamber MFCs and nafion membrane, concluded that the maximum voltage is produced in the oxygen concentration of 4–5 mg/L (Malakootian et al. 2011). Clauwert et al. (2007) in Australia, in a study entitled open air biocathode enables effective electricity generation with MFCs concluded that reducing DO in the cathode is one of the constraints of MFC (Clauwert et al. 2007). The listed studies are consistent with the results of this study because oxygen in the cathode chamber is as the terminal electron acceptor, and the depletion of DO, leads to reduced terminal acceptors and low electricity generation.

Luo et al. (2011) in the United States of America, in a study aimed at concurrent desalination and hydrogen

Table 2 Quality of wastewater of Isfahan steel company

pH	EC (ds/m)	Turbidity (mg/L)	TSS (mg/L)	BOD (mg/L)	COD (mg/L)	SO_4^{2-}(mg/L)	NH_4^+ (mg/L)
7.4	20.7	55	62	97	50	15.68	3.7
VFC (µg/L)	Cu (mg/L)	Zn (mg/L)	Cd (mg/L)	Pb (mg/L)	Ni (mg/L)	Hg (mg/L)	Cr (mg/L)
481	0.06	0.12	0.33	0.02	0.038	0.41	0.1

Table 3 Removal of Ni and Pb and the voltage produced by the MDC in real samples

Metal	DO (4–5 mg/L)	Temperature (20–35 °C)	Retention time (min)	Removal amount (%)	Voltage (*V*)
Ni	4.6	26	120	68.81	0.86
Pb	4.5	26	120	70.04	0.84

generation using microbial electrolysis and desalination cells found that increased retention time increases desalination (Luo et al. 2011). Jacobson et al. (2011a) in America, in a study with the aim of efficient salt removal in a continuously operated up flow MDC with an air cathode found that long retention time involves more ions in the flow production and increases ions removal (Jacobson et al. 2011a). The mentioned studies are consistent with this study due to the increased retention time and the removal of ions.

Werner et al. (2013) in Saudi Arabia, in a study with the aim of wastewater treatment, energy recovery and desalination using a forward osmosis membrane in an air–cathode microbial osmotic fuel cell found that removal efficiency increases at 30 °C (Werner et al. 2013). Lio et al. (2005) in Pennsylvania, in a study entitled power generation in Fed-Batch MFC as a function of ionic strength, temperature and reactor configuration concluded that reduced temperature results in decreased electricity production (Lio et al. 2005). The results of these studies are consistent with our study because lower temperature can slow metabolism and increased temperature speeds up metabolism, so bacteria grow faster and die quickly (MCkinney 2008).

Lio et al. (2005) in Pennsylvania, in a study entitled power generation in Fed-Batch MFC as a function of ionic strength, temperature and reactor configuration, and Brastad et al. (2013) in the United States of America, in a study aimed at water softening using MDC technology found that higher number of ions results in greater electrical conductivity (Lio et al. 2005, Brastad and He 2013). The results of these studies are consistent with this study due to increased number of ions and electricity generation caused to increasing removal.

Results of Brastad et al. (2013) in the United States of America, aimed at water softening using MDC technology are consistent with this study because lower electron capacity of the metal leads to reduced removal (Brastad and He 2013).

Ammonia, sulfate and volatile fatty acids are common pollutant in industrial wastewater (Siles et al. 2010; Singhania et al. 2013; Zacharof and Lovitt 2013). There are some methods to recover them or resolve their inhibitor, such as recovery of ammonia and sulfate from waste streams and bioenergy production via bipolar bioelectrodialysis (Zhang and Angelidaki 2015c), submersible microbial desalination cell for simultaneous ammonia recovery and electricity production from anaerobic reactors containing high levels of ammonia (Zhang and Angelidaki 2015d), counteracting ammonia inhibition during anaerobic digestion by recovery using submersible microbial desalination cell (Zhang and Angelidaki 2015b), microbial electrochemical monitoring of volatile fatty acids during anaerobic digestion (Jin et al. 2016), and bioelectrochemical recovery of waste-derived volatile fatty acids and production of hydrogen and alkali (Zhang and Angelidaki 2015a).

In this MDC, at the top of the reactor, an anode chamber, there was a small hole for exiting gases such as ammonia that were produced during decomposition of organic materials.

Conclusion

Removal efficiencies of Ni from wastewater of Isfahan electroplating industry and Pb from wastewater of Isfahan steel company in optimum condition were 68.81 and 70.04%. Optimum conditions for removing Ni and Pb from industrial wastewater were 4.6 and 4.52 mg/L DO, respectively, 26 °C and 120 min retention time for both. Advantages of MDC were the saving of energy, available exoelectrogenic bacteria and nutrient for them, simultaneous treatment for municipal and industrial wastewater. Thus, MDC is an effective method for removal of heavy metals, such as Ni and Pb present in industrial wastewater.

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