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ORIGINAL ARTICLE

Removal of Al, Ba and Mg from industrial wastewater using EAFDS and lime

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Abstract

Wastewater treatment and removal of hazardous metals is imperative for the well-being of living things and the environment. Different methods for the removal of hazardous metals have been proposed and practiced with varying success. In this study, dust slag generated by electric arc furnace is utilized for the removal of aluminium, barium and magnesium from industrial effluent. Slag which is a waste generated during the process of steel production is alkaline by nature. It was able to raise the pH of the acidic effluent to 8.7 removing most of the metals and reducing the amount of commercial alkali required to raise the pH to the target pH of 12.5. EAFDS in conjunction with lime removed 98% of Al, 95.8% of Ba and 99.7% of Mg. The method was simple, efficient and cost-effective since it utilizes a material considered hazardous waste for the treatment.

Keywords Aluminium · Barium · Magnesium · Wastewater · Metal removal · Dust slag · Treatment

Introduction

Rapid industrialization accompanied by urbanization specially in developing countries has increased pollution by hazardous metals, which is a concern for human well-being and the environment at large (van Neuss 2015; Aljerf 2018; Al-Makhadmeh et al. 2018; Rodriguez and Mandalunis 2018a, b). Technological development and implementation of machinery and factories led to increased production, which in turn led to the generation of increased environmental hazards, some of whose effects on the environment could only be seen years later (Inyinbor et al. 2016; Al-Makhadmeh et al. 2018). Pollution and its effects continue to exist in the environment for a prolonged time, even after the disappearance of the cause; our present actions determining the well-being of future generations as well (Bobylev 2016; Lu et al. 2019; Titah et al. 2019).

Many industries discharge organic and inorganic wastes containing hazardous metals into the environment including the water bodies (Islam et al. 2018). These may be acids, highly toxic minerals and metals, such as aluminium, barium, magnesium, mercury, zinc, chromium or lead (APHA 1989). Such pollution can have a variety of effects such as the water becoming uninhabitable for aquatic organisms, unsuitable for domestic use or irrigation; or the wastes may re-enter the food chain with deleterious effects on humans (Oribhabor 2016; Aniyikaiye et al. 2019; Omale et al. 2019). This pollution is caused mainly by atmospheric fallout from various sources, the most important being industrial and traffic emissions. Urbanized landscapes and industrial sites shown significantly higher metal contamination compared to agricultural areas (Sodango et al. 2018).

Aluminium (Al), barium (Ba) and magnesium (Mg) are the most common elements in industrial wastewater. Their effect on the environment and the biota cannot be overemphasized. Environmental institutions and industries are always in search of cost-effective and efficient wastewater treatment methods. Conventional methods used for the removal of metals from acidic effluents is raising the pH to alkaline level by using chemicals such as lime and calcium carbonate so that the metals form insoluble precipitate and removed from the water. To reduce cost associated

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with the procurement of these chemicals, the quest to find replacement materials that display similar characteristics as these alkaline materials is an ongoing process. The material selected for this study is the solid waste generated by a steel industry. Steel industry generates various types of wastes one of which is electric arc furnace dust slag (EAFDS).

The amount of EAFDS generated in EAF route is estimated to be 15–20 kg per ton of steel produced (Arnold et al. 2017; Ferreira et al. 2018; Silva et al. 2019). The composition of EAFDS differs based on the type of steel produced with a considerable variation of compositions. The major components of EAFDS are Fe_2O_3, FeO, metallic iron (Fe), $2FeO–SiO_2$, ZnO, SiO_2 (quartz), $3CaO–2SiO_2$ and 4PbO–PbSO (Wu et al. 2017; Al-Makhadmeh et al. 2018). Several research works have been carried out to characterize EAFDS and evaluate possible applications and utilizations for it (Omran, et al. 2017; Lozano-Lunar et al. 2019).

Experimental

Reagents and chemicals

All the reagents used in this study were analytical grade hence used without purification. The HCl was procured from SMM chemicals. The NaOH was obtained from Ace Chemical suppliers, and the $Ca(OH)_2$ was from Sigma-Aldrich. The acidic effluent and EAF dust slag were provided by a stainless steel industry that recycles scrap metal to produce steel.

Methods

The pH, acidity and the metal content of the effluent were analysed using the APHA (1989) method. The pH value was determined to be 0.64, whereas the acidity 36 g/l as $CaCO_3$.

Prior to the analysis, EAFDS was dried at 105 °C for two hrs. Then, the dry dust was cooled to room temperature and pulverized to powder. The powder was sieved through 300 μm pore size sieve. Total alkali in the slags were determined as $CaCO_3$ by using the following method before determining the amount of EAFDS required for the neutralization process.

2.5 g of the dried and pulverized EAFDS was put in a 250 ml beaker; 100 ml of one N HCl was added into the beaker and boiled for 2 min. The resulting solution was allowed to cool to room temperature before made up back to the 100 ml 100 ml mark of with DI water. The solution was filtered through Whatman No. 1 filter paper. 40 ml of the filtrate was measured into a beaker and titrated with 1 N NaOH to pH 7.

The total alkali content in the slag as $CaCO_3$ was determined from the mass and volumes of HCl and NaOH using the following formula given in Eq. 1.

$$\%\mathrm{CaCO}_3 = \frac{50 * C1 * V1 - C2 * V2}{1000} * \frac{M * V3}{V4} * 100 \tag{1}$$

where $C1$ is the concentration of HCl, V1 the volume of HCl, $C2$ the concentration of NaOH, $V2$ the volume of NaOH, $V3$ the volume of the solution titrated, v4 the total volume of the sample solution and M the mass of the slag weighed.

The total alkali content is given in Table 1.

Based on the acidity of the effluent and the total alkali content in the slag, the amount of slag required to achieve 3:2 alkalinity to acidity ratio was calculated. The precalculated amount of slag which was 54 g was weighed and added to 1 L effluent in a beaker and stirred at 380 rotations per minute (RPM). Samples were taken from the solution at 10 and 30 min, then every hour for the next 61 h., except during the night, and every two hours until it is 96 h. The reaction solution was made up to its initial 1 L volume after each sampling. The collected samples were filtered using No.1 Whatman filter paper and analysed immediately for pH and acidity. The remaining sample was analysed for the metal content with ICP-OES.

Results and discussion

Removal of metals

Aluminium

Aluminium (Al) is ranked as the third most abundant element with about 7–8% in the earth's crust next to oxygen and silicon (Stahl et al. 2017). Most of the Al in the environment originates from natural processes. Acidic precipitation initiates the release of Al from natural sources, whereas anthropogenic sources of Al compounds are industrial processes mainly releasing into the atmosphere. Some uses of Al products may also lead to the presence of Al in drinking water and foodstuffs.

Despite being one of the most abundant elements, Al is not an essential element nor has any biological significance in living organisms. So far, there is no evidence that shows

Table 1 Total alkali in EAFDS

Parameter	Sample mass(g)	EAFDS
Total alkali	2.50	92.4[a]

[a] $CaCO_3$

biological importance it plays in living things (Aljerf 2018). But toxicity by Al on plants is reported by various studies. It is a trivalent metal (Al^{3+}), and its behaviour in aqueous solutions varies with pH forming species including $Al(OH)_2^+$, $Al(OH)^{2+}$ and $Al(OH)^{4-}$ (Regina et al. 2015).

Al is used usually in household appliances and as a coagulant in wastewater treatment (Aljerf 2018). It is widely used in the manufacture of cans for beverage, for the production of antacid medicines, as a base for making pigments and cosmetics (Rodriguez and Mandalunis 2018a, b).

The largest consumers of Al metal and its alloys include the fields of transportation and construction as well as production of electrical equipment. Al powder is used in the production of paints, additive in fuel, propellants and explosives. Its oxides also have application as food additives and in the production of abrasive and heat-resistant materials, electrical insulators, catalysts, artificial gems, alloys and glass, while the hydroxide has extensive application in pharmaceutical and personal care products.

The large utilization of Al creates a considerable amount of waste containing Al and its compounds (Aniyikaiye et al. 2019). Al contamination can affect the environment, and alterations such as the colour change of groundwater, reduced soil fertility and the fish death have been reported (Kurniawan et al. 2018; Titah et al. 2019). Investigation of humans affected by Al indicated the formation of complexes with DNA and cross-linking with protein and DNA (WHO 2019). Exposure to Al is also confirmed to cause significant decrease in acquiring and retaining of learned behaviours and also has link to Alzheimer's and Down's syndrome (Klotz et al. 2017).

Other health risks to human beings associated with the intake of Al include deleterious effects to the bones, the nervous and hemopoietic systems (Stahl et al. 2017). Inhaling dust containing Al for an extended period of time can lead to a restrictive lung disease called pulmonary aluminosis. Exposure to Al has for a prolonged duration has also been implicated in chronic neurological disorders such as dialysis dementia and Alzheimer's disease (Masindi and Muedi 2018). Due to its tendency to accumulate in the body, The European Food Safety Authority (EFSA 2017) recommends a Tolerable Weekly Intake (TWI) for Al of 1.00 mg/kg body weight (BW) per week.

Al has a negative effect on different biological processes in human body. In addition to be considered a potentially cell and neurotoxic, it may also disrupt the enzyme activities as well as mitochondrial functions. It also may produce oxidative stress and may play a role in diseases such as breast cancer and Alzheimer's dementia. Clinical studies of Al indicated that poisoning of Al can cause bone diseases such as renal osteodystrophy, osteomalacia and osteoporosis (Rodriguez and Mandalunis 2018a, b).

Toxicity by Al is one of the main factors limiting plant growth in especially in acidic soils (Arnold et al. 2017). Industrial and mining wastes containing metals and there derivatives and compounds are hazardous to the environment and living things due to their toxicity and non-biodegradability, even when they are present at trace concentrations (Goher et al. 2015). Removing or reducing the amount of hazardous metals from the environment is imperative. Several methods for removal of metal ions from wastewaters have been developed. Any method used for removal must be simple and easy to operate, effective and inexpensive (Goher et al. 2015).

Some wastes containing Al, such as dross, scrap and foil are being recycled by aluminium recycling industries (Gomes et al. 2018). Bioaugmentation, the addition of pre-grown microbial cultures to enhance the treatment process, is a method widely applied to treat metal contamination (Nzila et al. 2016; Arnold et al. 2017). This method is usually used to treat metal contamination in water. The application of this method is also considered to be economically profitable due to the metal recovery potential after the treatment process (Rodriguez and Mandalunis 2018a, b).

The study to remove Al from the acidic steel industry effluent indicates that Al forms insoluble precipitate at higher pH. Hence, using the alkaline nature of the EAFDS as a replacement for commercial alkaline materials such as $Ca(OH)_2$ as a source of alkali can raise the pH to the required level to remove Al.

The Al content in the raw effluent was 89.8 mg/l. After the mixture of the effluent and EAFDS was agitated at 380 rpm for 30 min, the pH rose to 3.1 and the concentration of Al in the solution dropped to 22.3 mg/l achieving more than 75% removal. After 24 h at pH 8.1 Al concentration dropped further to 3.2, the removal was 95.4%. At pH 8.4, the concentration of Al dropped to 3.0 mg/l, but starting from 8.5 to 8.7 the concentration showed a slight increase reaching 3.9 mg/l. The slag alone was able to remove 95.7% of the aluminium reducing the concentration to 3.9 mg/l. To reach the target pH of 12.5, 0.65 g of pure $Ca(OH)_2$ was added to the reaction mixture. Two more samples were collected at pH 9.5 and 12.5. The concentration decreased again and reached 1.8 mg/l at pH 12.5 achieving 98% removal. The value even with EAFDS alone was far below the maximum permissible effluent discharge limit set by Food and Agriculture Organisation (FAO), which is 5.0 mg/l. The trend of aluminium concentration with respect to pH is given in Fig. 1.

Barium

Barium (Ba) a yellowish white metal, being in the same group of alkaline earth metals, it has similar chemical and physical properties to calcium and magnesium. Even though

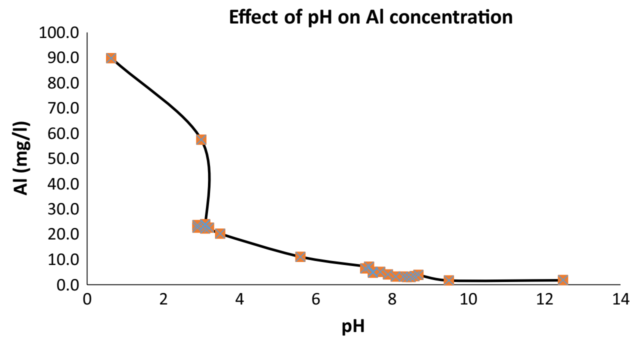

Fig. 1 Effect of pH on Al concentration

it is non-essential element, it is toxic to animals and plants (Lu et al. 2019).

In the natural state, barium occurs as a divalent cation (Ba^{2+}) combined with sulphate or carbonate to form barium sulphate or barium carbonate (witherite). Ba is the 14th most abundant element in the earth's crust. It reacts with almost all nonmetals forming water-soluble toxic compounds.

Ba is used in the manufacturing of barium-nickel alloys used for making spark-plug electrodes as well as a drying and oxygen-removing agent in the production of vacuum tubes. Since impure barium sulphide phosphoresces after exposure to light, it is also used in fluorescent lamps. Oil and gas industries use Ba compounds to make drilling mud, which lubricates the drill making drilling through rocks easier. Ba compounds are utilized to make paint, bricks, glass and rubber, and the nitrate and chlorate of Ba give fireworks a green colour.

Ba compounds have extensive applications in industrial fields, such as in the petroleum and steel industry as well as in the fields of medicine as an agent for gastrointestinal tract radiography (Kravchenko and Guber 2017). Due to its extensive application, significant amount of Ba has entered into the environment, resulting in an increased public awareness of its risk (ASTDR 2017). Because of its widespread use and rapid urbanization, Ba pollution is becoming a serious environmental issue (Lu et al. 2019).

Because of its vast use in the industries, human activities contribute immensely to the release of Ba to the environment. As a result around locations of manufacturing involving Ba, its concentration is considerably higher than the concentration occurring naturally. Sources of Ba pollution include mining and refining processes as well as the production of barium compounds. Coal and oil combustion also can contribute Ba to the atmosphere. Ba contamination could impact human health, various areas of agriculture, wildlife and the quality of air and water (FAO 2018).

Ba ion is toxic if enters to human body; oral exposure even at small amount may result in severe ototoxicity with degeneration of inner ears in mammals. It can also lead to hypertension in humans (Jabłońska et al. 2016). Short-time exposure to Ba may enhance malignant cellular characteristics in nontumorigenic cells.

Exposure to a large amount of Ba may cause different negative effects on animals, such as cardiovascular, renal, respiratory, hematological, nervous and endocrine systems damage and even death (Kravchenko and Guber 2017). Cancer-promoting effects of Ba on humans have also been observed. The toxicity of a Ba compounds is enhanced by its solubility (Lu et al. 2019).

Extensive usage of Ba compounds in various production and manufacturing processes such as drilling fluids, oil industry, explosives, fire extinguishers, soaps and insecticides resulted in elevated levels of Ba in the environment and exposure in daily life (Lu et al. 2019). Being readily water-soluble Ba compounds released during industrial processes can enter into water bodies in the environment enabling them to spread to large areas. When fish or other aquatic organisms consume Ba or its compounds, it will accumulate in their tissues. However, its insoluble salts such as carbonate and sulphate that are persistent usually remain on the soil surface or in the sediment of water.

The amount of Ba in food and water from natural sources and processes usually does not pose a health concern. People who work in Ba industry work in Ba industry face a greater risk to Ba exposure and related health effects. The health risks mostly originate from inhaling in air that contains barium compounds.

Small amount of Ba in water may cause physical discomforts such as difficulty in breathing, increase in blood pressure, stomach irritation, swelling of the brain, muscle weakness, damage to vital organs and changes heart rhythm. It might also be carcinogenic due to its nature of being bone seeking (ASTDR 2017).

Many industrial and mining waste sites may contain Ba resulting in the people who inhabit in the proximity to harmful levels through inhaling dust, consuming plants from the vicinity or water that is polluted with Ba (Lozano-Lunar et al. 2019).

The toxicity of Ba depends on the water solubility of its compounds. The readily soluble the compound is the more deleterious to human health it is. Large amounts of water-soluble Ba may result in paralyses and in some cases even death.

Wastewater treatment methods such as filtration, chemical oxidation, disinfection with UV, chemical precipitation, ion exchange, reverse osmosis, steam stripping and acidification are currently used for Ba removal (Kim et al. 2017a, b). Since Ba possesses similar properties with calcium and magnesium, treatment methods effective for the removal of Ca and Mg can also be applied for the removal of Ba.

Treatment techniques employed for the removal of Ba include ion exchange, lime application, coagulation, adsorption, reverse osmosis, electrodialysis and distillation. According to USEPA, the maximum allowed concentration of Ba in drinking water is 2 mg/l; however, WHO recommends the limit to be less than 0.7 mg/l (USEPA 2017).

In this study, the conventional chemical precipitation method was used. However, the source of alkali used the precipitation was EAFDS instead of commercial chemicals. The barium concentration in the raw effluent was 29.7 mg/l. The concentration showed sharp decrease in the first 10 min until it reached pH 3.0 removing 45.5% of Ba from the solution. The removal slowed down considerably after that. After treatment for 7 h with EAFDS, the pH reached 5.6 and the concentration dropped to 14.9 mg/l achieving 50% of Ba removal. After 24 h, the pH rose to 7.9 and the concentration of Ba dropped to 11.5 mg/l achieving 61.3% of the Ba removal from the solution. EAFDS alone raised the pH to 8.7 achieving removal of 87.5% of Ba from the solution reducing the concentration to 3.7 mg/l. After continuing the process with pure $Ca(OH)_2$, the concentration further decreased to 1.6 mg/l achieving 94.7% removal t pH 9.5. At the final target pH 12.5 the concentration was 1.3 mg/l achieving 95.8% removal of Ba from the effluent. The pattern of the effect of pH on the concentration of Ba is given in Fig. 2.

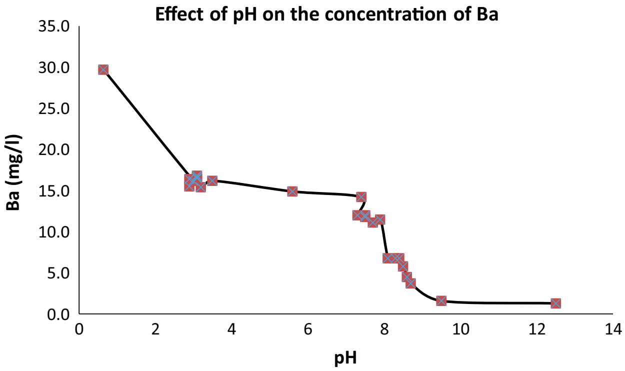

Fig. 2 Effect of pH on the concentration of Ba

Magnesium

Magnesium (Mg), one of the essential metals to humans, one of the most prevalent minerals in the human body and is the second most common intracellular cation. It is also part of more than 300 enzymatic systems and needed for fundamental life processes including the production of energy and synthesis of nucleic acid; it also plays a significant role in the synthesis of ATP (adenosine triphosphate) from ADP (adenosine diphosphate) and inorganic phosphate.

Mg along with calcium is the principal cause of water hardness. Water hardness creates problems in everyday life such as by causing human stone disease, atopic eczema and unfavourable taste of drinking water. Several studies confirmed a strong relation between the concentration of water hardening minerals (particularly calcium and magnesium) and cardiovascular diseases, Alzheimer's diseases and atopic eczema, undesirable spots on sinks and clothes as well as toughening of skin and hair, reducing the cleansing ability of soap and in industry by forming scales and clogging pipes, causing corrosion of machines. The scale buildup may also reduce the efficiency and performance of water pumps. Excessive Mg can also lead to diarrhoea and has a laxative effect.

Considering the problems of water hardness, removal of hardness is essential. Various physicochemical techniques employed for removing Mg ions from water supplies include chemical precipitation, adsorption, ion exchange, extraction electrocoagulation, reverse osmosis, electro dialysis, nanofiltration, crystallization, distillation and evaporation and membrane processes. WHO Recommendation for drinking water hardness is based on maximum 500 mg/l calcium carbonate (Helmy et al. 2017).

There are many different methods of wastewater treatment to remove or reduce the metal content. The most common techniques include the use of different adsorbents such as polymeric resins and activated carbon, or alternative matrices to adsorb metals in solution, reverse osmosis, nanofiltration, polymer-material membrane and electrodialysis and reverse electrodialysis. In addition, there are forward osmosis and membrane distillation technologies as well as biological process using microorganisms (Zhao et al. 2019). The technique used widely involves increasing of the pH of wastewater to an alkaline level in order to remove Mg through precipitation.

To get the pH to alkaline level, a source of alkali used in this study was EAFDS. After adding EAFDS to the effluent, the mixture was stirred continuously. The concentration of Mg in the raw effluent was 377.3 mg/l. But when the slag came in contact with the acidic effluent, Mg was leached from the slag. The slag contained a large amount of Mg. It was determined to be 106.5 mg/g. After 10 min of agitation, the pH rose to 3.0 and the concentration of Mg also increased to 399.9 mg/l. The concentration of Mg reached the highest value of 464.3 mg/l at pH 3.1 and started to decrease after that. However, the decrease was very slow and reached 214.9 mg/l at pH 8.7 before adding $Ca(OH)_2$ achieving a removal of 39.5%. After the addition of lime, one more sample was collected at pH 9.5 and there was no decrease in the concentration of Mg. But after pH 10, the decrease was sharp and the concentration of Mg dropped to 1 mg/l achieving removal percentage of 99.7%. The pattern of removal and the effect of pH on Mg concentration is given in Fig. 3.

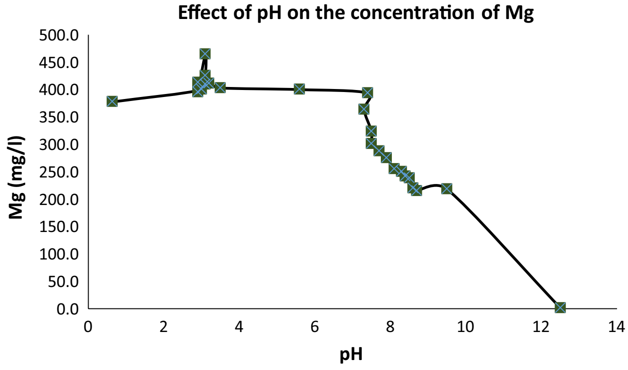

Fig. 3 Effect of pH on the concentration of Mg

Conclusion

The treatment method investigated in this study was chemical precipitation technique, which is one of the most common treatment methods to neutralize and remove hazardous metals from wastewater. Since the chemicals used for neutralization and metal removal incur cost, the aim was to find an alternative replacement which possesses similar property to the commercial costly chemicals. The material selected for the study was electric arc furnace dust slag (EAFDS). This material is a by-product generated by a steel manufacturing industry and usually considered a waste and disposed of into dump sites resulting in the pollution to the environment, disposal levy for the company and loss of potentially useful chemicals embedded in it.

The investigation of the composition of EAFDS indicated that it contains ample amount of alkali which can be exploited as a source for water treatment. Since it is mainly composed of metal oxides, the leachate is alkaline. This property was used for the treatment of industrial acid effluent from a steel industry containing various hazardous metals in considerably high concentration.

The study demonstrated that EAFDS can be used for the removal of metals AL, Ba and Mg from a highly acidic steel industry effluent.

The Al content in the raw effluent was 89.8 mg/l. The EAFDS alone was able to remove 95.7% of the aluminium reducing the concentration to 3.9 mg/l. The concentration decreased again and reached 1.8 mg/l at pH 12.5 achieving 98% removal.

The barium concentration in the raw effluent was 29.7 mg/l. EAFDS alone raised the pH to 8.7 achieving removal of 87.5% of Ba from the solution reducing the concentration to 3.7 mg/l. At the final target pH 12.5, the concentration was 1.3 mg/l achieving 95.8% removal of Ba from the effluent.

The concentration of Mg in the raw effluent was 377.3 mg/l. The decrease in the concentration of Mg was very slow and reached 214.9 mg/l at pH 8.7 before adding $Ca(OH)_2$achieving a removal of 39.5%. After pH 10, the decrease was sharp and the concentration of Mg dropped to 1 mg/l achieving removal percentage of 99.7%.

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Compliance with Ethical Standards

Conflict of interest The authors declare there is no conflict of interest whatsoever.

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