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Continuous electrocoagulation process for the treatment of high colloidal clay in coal mine water using lab scale baffle channel reactor

ABSTRAK

Proses elektrokoagulasi (EC) telah dipelajari secara luas dalam beberapa tahun terakhir untuk menghilangkan berbagai kontaminan yang ada dalam air limbah seperti logam, bahan organik, pewarna, minyak, senyawa farmasi, hingga emerging pollutant lainnya. Di industri pertambangan, pengembangan EC terus dilakukan untuk menyisihkan kontaminan didalam air tambang yang memiliki karakteristik asam maupun *suspended solid* yang tinggi. Studi ini bertujuan untuk mengembangkan EC dalam menyisihkan *suspended solid* yang disebabkan karena kehadiran *colloidal clay* dari batuan penutup (*overburden*) selama kegiatan penambangan. EC dioperasikan menggunakan lab *scale baffle channel reactor* bervolume 15 Liter secara kontinyu dengan menggunakan pasangan elektroda besi dan alumunium monopolar. Untuk mendapatkan kinerja yang optimal, studi difokuskan pada pengaruh tiga parameter yang mempengaruhi EC, yaitu kepadatan arus (2 A; 4 A; dan 6 A), jenis elektroda (besi dan alumunium), dan waktu tinggal di dalam reaktor (15 menit; 30 menit; dan 45 menit equal to debit 0,3; 0,5; 1 Lpm). Hasil penelitian menunjukkan penyisihan suspended solids tertinggi sebesar 99,66% terjadi pada reaktor elektroda besi dengan kuat arus 6 ampere dan HRT sebesar 45 menit atau debit 0,3 liter per menit atau. Reaktor dengan elektroda aluminium memiliki presentase penyisihan 99,11% dengan kuar arus 6 ampere dan HRT sebesar 15 menit atau debit 1 liter per menit. Penelitian ini merupakan tahap lanjutan dari operasi batch dalam upaya mengembangkan EC skala lapangan untuk pengolahan air tambang dengan kandungan *colloidal clay* yang tinggi.

Kata kunci: air tambang, colloidal clay, elektrokoagulasi

ABSTRACT

The electrocoagulation (EC) process has been extensively studied in recent years for the removal of various contaminants present in wastewater, such as metals, organic materials, dyes, oils, pharmaceutical compounds, and other emerging pollutants. In the mining industry, EC development continues to be conducted for the removal of contaminants in mine water, which is characterized by high acidity and suspended solids. This study aims to develop EC for the removal of suspended solids caused by the presence of colloidal clay from overburden during mining activities. EC was operated using a lab-scale baffle channel reactor with a volume of 15 liters in continuous mode, utilizing monopolar iron and aluminum electrode pairs. To obtain optimal performance, the study focused on the influence of three parameters affecting EC: current density (2 A, 4 A, and 6 A), electrode type (iron and aluminum), and residence time in the reactor (15 minutes, 30 minutes, and 45 minutes, equivalent to flow rates of 0.3, 0.5, and 1 LPM). The results demonstrated that the highest suspended solids removal of 99.66% occurred in the iron electrode reactor with a current strength of 6 amperes and an HRT of 45 minutes or a flow rate of 0.3 liters per minute. The reactor with aluminum electrodes achieved a removal percentage of 99.11% with a current strength of 6 amperes and an HRT of 15 minutes or a flow rate of 1 liter per minute. This research represents an advanced stage of batch operation in efforts to develop field-scale EC for the treatment of mine water with high colloidal clay content.

Keywords: mine water, colloidal clay, electrocoagulation

1. Introduction

Clay minerals are significant geological components that cover a substantial portion of the Earth's surface and are prevalent in sediments, soils, and rocks that have undergone weathering and hydrothermal alteration (Kumari and Mohan, 2021; Huggett, 2005). These minerals, known as hydrous phyllosilicates, possess particle sizes less than 2 micrometers with a layered structure, resulting in high specific surface area (SSA) and varying quantities of inorganic ions such as iron, magnesium, alkali metals, alkaline earth metals, and other cations (Abfertiawan et al., 2023; Hamza et al., 2023). The unique characteristics of clay minerals have prompted numerous researchers to investigate their significance in various industries and their behavior in the environment. In environmental contexts, clay minerals are known to exhibit high water-holding capacity (WHC), adsorption, and cation exchange capacity (CEC) per unit mass (Lambooy, 1984; Huggett, 2015; Arthur et al., 2019; Zhang, 2021). These properties render clay minerals highly suitable for use as adsorbents in the removal of metal contaminants from water (Uddin, 2017; Orucoglu et al., 2022; Barakan and Aghazadeh, 2021; ElBastamy et al., 2021; Otunola and Ololade, 2020). However, these characteristics may potentially impact water quality. The adsorption of metals by clay minerals may result in their transport over long distances along river systems (Chen et al., 2016; Chen et al., 2018; Cao et al., 2022; Mirandi et al., 2022). This phenomenon may lead to the presence of metal content detected in association with clay fractions in sediments along river streams (Ali et al., 2018; Taha, 2023), potentially causing bioaccumulation in benthic organisms (Gnanasekaran and Amal Raj, 2023; Zhang et al., 2014). Furthermore, due to their interlayer space characteristics, nearly all clay minerals in the kaolinite, smectite, vermiculite, illite, and chlorite groups possess the ability to absorb and retain water to varying degrees and for relatively extended periods. The swelling properties of these minerals, also known as hydrous alumino silicates or hydrous aluminum phyllosilicates, are recognized as a contributing factor to the presence of colloidal clay in wastewater or water bodies that cannot settle through gravitational means (Abfertiawan, et al., 2023; Lagaly, 2005; Lagaly, 2006; Sposito, 2016). Consequently, engineered coagulation-flocculation processes remain the sole method for removing colloidal clay from wastewater.

In open-pit coal mining operations, mine water potentially exhibits acidic characteristics, commonly referred to as acid mine drainage, and is characterized by high levels of suspended solids. The elevated concentration of suspended solids is attributed to the presence of clay minerals abundantly found in the overburden. During overburden excavation and deposition activities, precipitation falling and flowing over active mining areas induces crystalline swelling, also known as surface hydration (Karpiński and Szkodo, 2015). This phenomenon can occur during the adsorption process of monomolecular water layers on the surface of layered clay minerals (Roderick and Demirel, 1963; Parker et al., 1982; SATO, 1996; Chen et al.m 2022). Notably, the type and quantity of exchangeable cations within clay minerals may further augment the swelling volume. Colloidal particles in mine water present a significant challenge for removal in mine water treatment facilities. Water management challenges are particularly pronounced for mining industries in tropical rainforest countries with high precipitation rates (Wantzen and Mol, 2013; Gilsbach, et al., 2019). With an average annual temperature of approximately 28°C, these tropical rainforest countries are characterized by relatively high rainfall, with 75 percent of precipitation generated through evaporation and transpiration. In Indonesia, the average annual rainfall can reach 3,000 mm, with burst rainfall intensities potentially exceeding 150 mm/hour. Due to the settling-resistant nature of colloidal clay, mine water treatment is typically conducted using active-conventional methods involving chemical coagulant addition, which tends to require relatively high energy inputs (Wang et al., 2021; Mollah et al., 2004; Dang and Dang, 2018). Ongoing research and development efforts are focused on treatment technologies for high suspended solid concentrations to achieve more effective and efficient treatment methodologies.

Electrocoagulation is a technology that has been extensively developed for the removal of contaminants in wastewater due to its relatively high effectiveness and efficiency (Mostefa and Tir, 2004; Oncel et al., 2013; Mamelkina et al., 2019; Merma et al., 2020; Syafila et al., 2023; Shahedi et al., 2023). This technology is also recognized for its potential application due to its advantages such as simplicity, environmental friendliness, and low cost (Moussa et al., 2017; Nariyan et al., 2018; Shahedi et al., 2023). Previous research has demonstrated that electrocoagulation using a batch reactor has proven capable of removing suspended solids due to the presence of colloidal clay in coal mining by 99.58% with a current of two amperes and a contact time of fifteen minutes (Abfertiawan et al., 2023). Electrocoagulation is a technology that involves the in-situ formation of coagulants by dissolving metal anodes followed by the simultaneous formation of hydroxyl ions and hydrogen gas at the cathode (Hakizimana et al., 2017; Shaker et al., 2023). Factors influencing the electrocoagulation process include temperature, contact time, electric current strength, voltage, and acidity or pH (Kobya et al., 2007; Jane et al., 2023; Mousazadeh et al., 2023). In recent years, research on the development of electrocoagulation technology has focused on identifying optimal parameters, particularly using batch reactors. However, for implementation, especially in mine water treatment, the development of electrocoagulation must continue using continuous reactors, considering factors such as reactor configuration, process performance, and operating parameters. This study aims to determine the effectiveness of the electrocoagulation process using a continuous reactor with iron and aluminum electrodes in removing colloidal clay from coal mine water.

2. Materials and Methodology

2.1. Mine Water Sample

The study was conducted utilizing artificial mine water formulated based on the characteristics of mine water from coal mines in East Kalimantan, Indonesia. The original characteristics of this mine water have been published in Abfertiawan et al. (2024), with a total suspended solid (TSS) concentration of 5400 mg/L. To ensure that the artificial mine water characteristics closely resembled those of the original mine water, 81 g of clay was added, resulting in artificial mine water with a TSS concentration of 3865 mg/L. A comparison of the characteristics between the original and artificial mine water can be observed in Table 1 below.

2.2. Electrocoagulation Reactor Design

The electrocoagulation reactor is constructed from 5 mm acrylic, with dimensions of 41.4 cm x 26 cm x 20 cm, a total capacity of 15 liters, and a baffle flow channel type. The reactor model with a horizontal flow baffle channel ensures the wastewater remains homogeneous due to hydraulic mixing that utilizes water flow as a mixing force (Holt, 2002). Twelve electrode plates (6 cathodes and 6 anodes) are placed along the reactor with a 3 cm inter-plate distance and arranged in a parallel monopolar configuration. The electrode plates have a thickness of 2 mm with a length of 23 cm and a width of 20 cm. The submerged portion of the electrode is 13 cm with a total wet surface area of 0.358 m2. All electrodes are connected to an electric current sourced from a DC power supply (MDB 0-60 V and 0-20 A). Current strength and voltage measurements are conducted using a MASDA DT830B multimeter. The electrocoagulation reactor is continuously filled with synthetic wastewater using a peristaltic pump (Pulsafeeder DC21C1PT) with a maximum flow rate of 75 L/hour.

2.3. Research Procedure

The electrocoagulation process was conducted with 9 experiments performed in duplicate, resulting in a total of 18 trials. Three variations of current (2A, 4A, 6A) and three variations of hydraulic retention time (15 minutes, 30 minutes, and 45 minutes) were employed, yielding influent flow rates into the reactor of 0.3 L/minute, 0.5 L/minute, and 1 L/minute, respectively. Post-treatment water sample quality parameters were assessed through testing methods in accordance with the Indonesian National Standard (SNI). The methodologies for sample testing and analysis are presented in Table 2 below.

Table 1. Characteristics of Coal Mine Water

No	Parameters	Unit	Value	
			Original	Artificial
1	pH	-	8	6,65
2	Konduktivitas	μs/cm	538	310
3	Temperatur	^{o}C	26,7	24,6
4	TDS	mg/L	320	150
5	TSS	mg/L	5400	3865

Tabel 2. Hasil Pengujian Air Limbah RPH Ayam

No	Parameter	Metode Pengujian	Metode Analisa
1	pH	SNI 6989.11 : 2019	pH meter
2	Temperatur	SNI 6989.23-2005	Termometer
3	Konduktivitas	SNI 6989.1 : 2019	*Conductivity* meter
4	TDS	-	TDS meter
5	TSS	SNI 6989.3 : 2019	Gravimetri

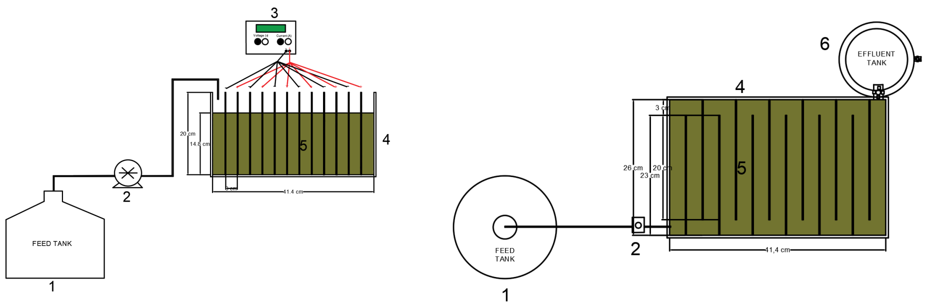


Figure 1. Electrocoagulation reactor design and schematic

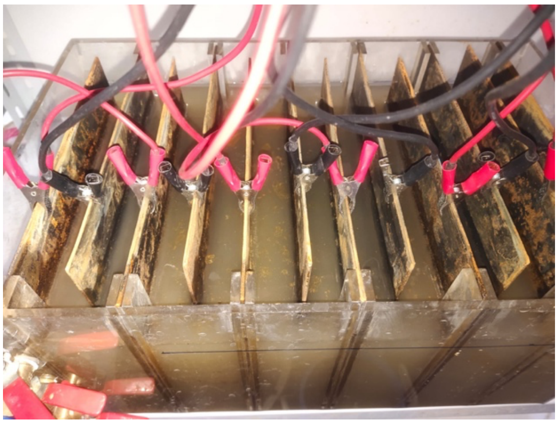


Figure 2. Electrocoagulation reactor with continuous flow in baffle channel

3. Result and Discussion

3.1. Change in pH

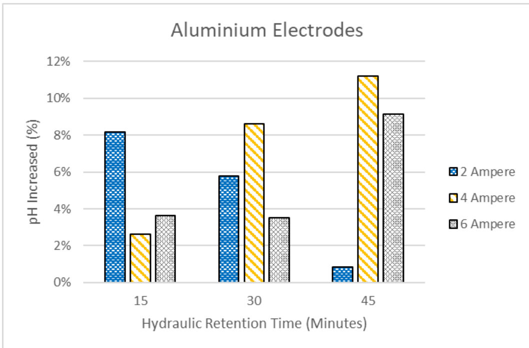
The pH of a solution is considered a critical parameter prior to treatment as it influences solution conductivity, electrode dissolution, and the zeta potential of colloidal species. As illustrated in Figure 3(a), the pH exhibits an increasing trend across all flow rate variations from the initial sample pH. This increase in pH occurs due to the release of OH^- ions from the cathode, as described by the following equation:

$$2H_2O + 2e - \rightarrow H_2 + 2OH^- \qquad (1)$$

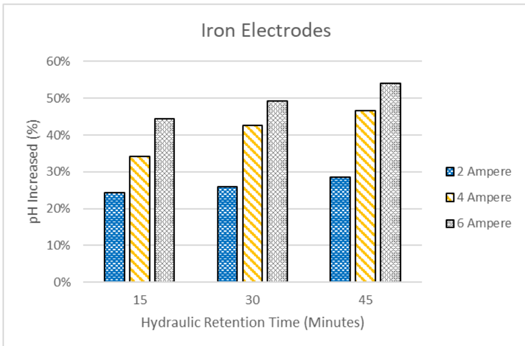
A similar trend was observed in the iron electrode. As shown in Figure 3(b), the highest pH increase was achieved with a current strength of 6 Amperes and a hydraulic retention time of 45 minutes. Under these conditions, the solution pH reached 10.37 from an initial pH value of 6.65. The lowest pH increase occurred at a current strength of 2 Amperes and a hydraulic retention time of 15 minutes. In this variation, the average solution pH reached 8.46 from an initial pH value of 6.65. Therefore, it can be concluded that higher current strengths and longer hydraulic retention times applied to the system result in greater increases in the solution pH. This phenomenon can be attributed to the increased release of OH- ions from the cathode into the solution. OH^- ions are basic in nature and thus can elevate the solution pH. The pH value of wastewater will lead to the formation of various iron compound species in the form of monomer and polymer ions (Mollah et al., 2004).

3.2. Change in Total Dissolved Solid

Total Dissolved Solids (TDS) is a parameter that quantifies the amount of dissolved solids or the concentration of ions (both cations and anions) in water, expressed in mg/L. As illustrated in Figure 4(a), the fluctuations in TDS observed in this study do not exhibit a significant pattern of change. The highest increase in TDS was observed at a current strength of 2 Amperes with a flow rate of 1 L/minute, demonstrating a percentage increase of 6.15%, from 179 mg/L to 190 mg/L. The most substantial decrease was recorded at 23.83%, from 195 mg/L to 147 mg/L, at a current strength of 6 Amperes and a flow rate of 0.3 L/minute. The changes in TDS concentration for iron electrodes are presented in Figure 4(b), with the highest TDS reduction achieved at a current strength of 6 Amperes and a hydraulic retention time of 45 minutes. The TDS value of the solution under these conditions was observed to decrease from 155 mg/L to 47.5 mg/L.

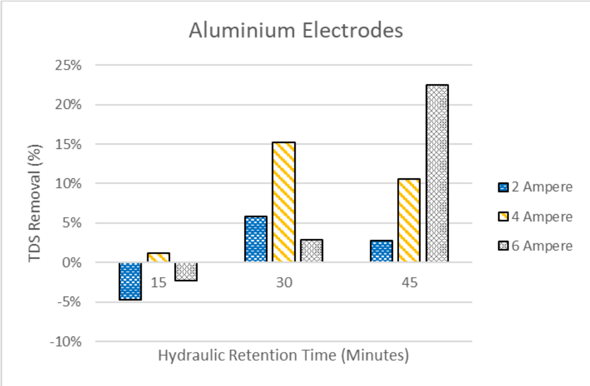


(a)

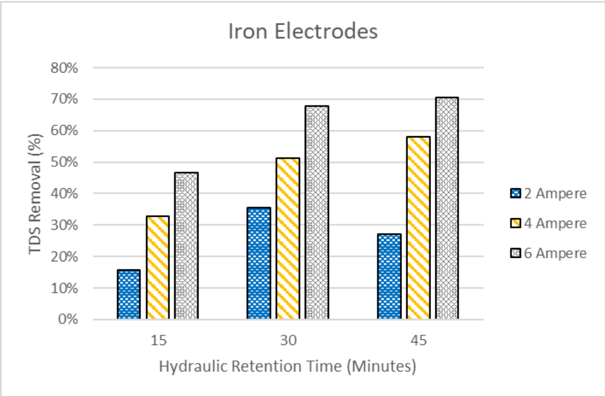


(b)

Figure 3. Percentage increase in pH (a) aluminum electrode, (b) iron electrode



(a)



(b)

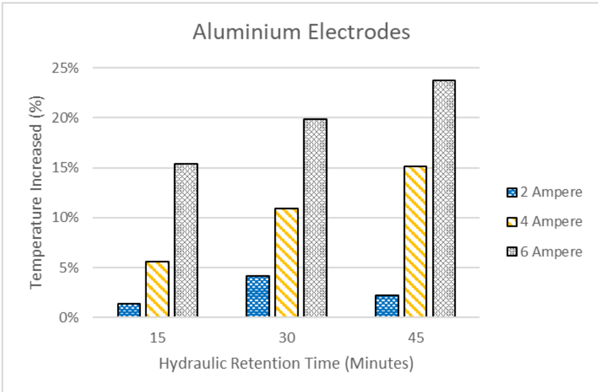
Figure 4. Percentage removal of TDS (a) aluminum electrode, (b) iron electrode

Changes in TDS concentration is known to be significantly influenced by flow velocity, wherein a decrease in flow velocity results in prolonged contact time between the flow and the reactor. The established current strength leads to an increase in ions with different charges in the dissolved substances within the waste, resulting in the formation of flocs that subsequently coalesce into larger flocs. According to Nur and Effendi (2014), the presence of Al^{3+} ions can neutralize contaminant ions dissolved in water by opposing ions generated from the cathode. These ions reduce the repulsive forces between particles in the wastewater, thereby facilitating the coagulation process.

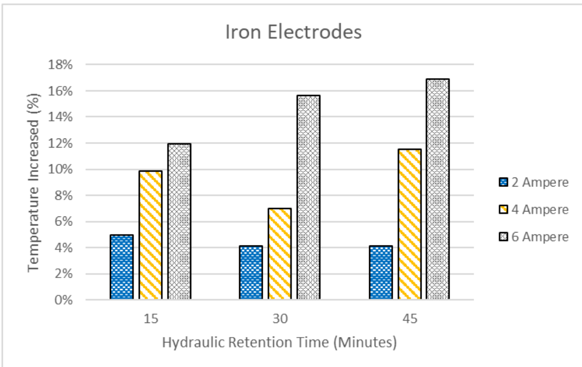
3.3. Temperature Change

The temperature behavior can be observed in Figure 5(a) for aluminum electrodes and 5(b) for iron electrodes. Figure 5(a) demonstrates that the highest temperature increase occurs from 25.5°C to 31.1°C at a current strength of 6 Amperes with a flow rate of 0.3 L/minute. Conversely, at a current strength of 2 Amperes with a flow rate of 1 L/minute, the temperature increases from 25.2°C to 25.5°C. This indicates that temperature elevation occurs in correlation with increased current strength and contact time.

Based on Figure 5(b), for iron electrodes, the highest temperature increase was achieved at a current of 6 Amperes and a hydraulic retention time of 45 minutes. At this variation, the solution temperature reached 28.85°C from an initial temperature of 24.3°C. The lowest temperature increase was achieved at a current strength of 2 Amperes and a hydraulic retention time of 15 minutes. The solution temperature at this variation was 25.35 °C from an initial temperature of 24.3°C. According to Holt (2002), the temperature increase occurs due to the release of Fe^{2+} metal hydroxide during the electrocoagulation process, which releases energy in the form of heat, affecting the increase in reaction rate. The temperature increase promotes the formation of metal hydroxides and causes greater particle mobility (El-Ashtoukhy et al., 2009). The temperature increase observed in this study led to enhanced mass transfer and increased particle collision kinetics. Additionally, there was an increase in anode solubility and the amount of hydroxide formed, which is necessary for binding pollutants, with more hydroxide formed at higher temperatures compared to lower temperatures (Khaled et al., 2019). Higher temperatures enable the production of larger hydrogen bubbles, thereby increasing the flotation rate (Koren and Syversen, 1995).



(a)

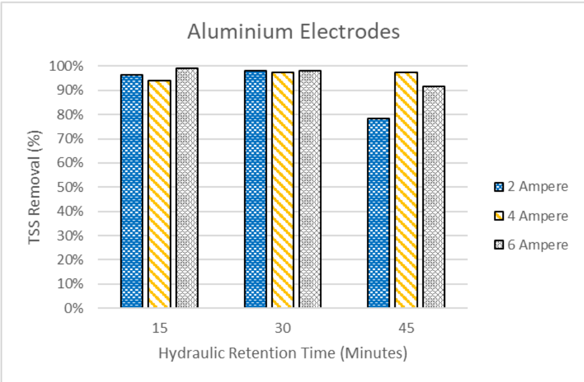


(b)

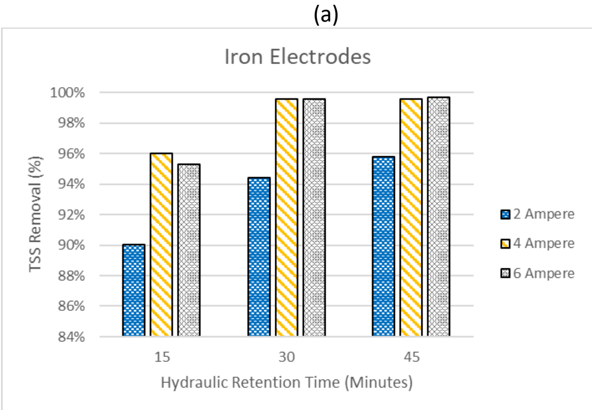
Figure 5. Percentage temperature increase (a) aluminum electrode, (b) iron electrode

3.4. Total Suspended Solid Removal

Total Suspended Solid (TSS) represents the fraction of solids in water that is retained on a filter with pore sizes between 0.45 µm and 2 µm (Tchobanoglous et al., 2003). Figure 6(a) demonstrates that the highest TSS removal efficiency was achieved at a current strength of 6 Amperes with a flow rate of 1 L/minute, resulting in a removal percentage of 99.1%, reducing TSS from 1,528.8 mg/L to 13.6 mg/L. The lowest removal efficiency was recorded at 78.49% with a current strength of 2 Amperes and a flow rate of 0.3 L/minute, reducing TSS from 2492 mg/L to 536.1 mg/L. For iron electrodes (see Figure 6(b)), it was observed that the highest TSS removal efficiency was achieved at a current strength of 6 Amperes and a hydraulic retention time of 45 minutes, with an average TSS reduction efficiency of 99.18%.



(a)



(b)

Figure 6. TSS removal percentage (a) aluminum electrode, (b) iron electrode

These results indicate that as the current strength and hydraulic retention time increase during electrocoagulation, the removal of TSS concentration tends to increase proportionally. This phenomenon can be attributed to the fact that particles contained in mine water are generally negatively charged, where like charges cause repulsion between particles, leading to their stability. During the electrocoagulation process, positive and negative ions produced by iron and aluminum electrodes can destabilize the particles present in the wastewater (Yulianto et al., 2009). The anode electrode undergoes an oxidation reaction with anions (negative ions) to form Fe^{2+}, which then binds with OH- to form $Fe(OH)_2$ compounds capable of binding pollutants. The flocs formed gradually increase in size and eventually settle to the bottom of the electrocoagulation tank through gravitational force (Mollah et al., 2004). Additionally, the cathode during the electrocoagulation process generates hydrogen gas (H_2), causing the formed flocs to rise to the surface.

3. Conclusion

A study on the development of continuous electrocoagulation technology has been conducted to remove mine water containing colloidal clay with relatively high total suspended solids (TSS). The results of the study indicate that electrocoagulation with a continuous process can remove total suspended solids with high efficiency. Using iron electrodes, with a current of 6 Amperes and a hydraulic retention time of 45 minutes, the highest TSS efficiency of 99.66% was achieved. For aluminum electrodes, the optimum TSS removal occurred at a current of 6 Amperes and a hydraulic retention time of 15 minutes, with a removal efficiency reaching 99.11%. Further studies are necessary to examine other variables that potentially influence the effectiveness of the electrocoagulation process, including variatio ns in influent load and continuous reactor configuration.

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