

Organic Matter Pollutant Removal Using Continuous Baffle Flow Electro-Oxidation Reactor (CBFER) Equipped with Al⁰-Al⁰ Electrodes Configuration

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Abstract

Electrooxidation experiment using continuous baffle flow electro-oxidation reactor (CBFER) equipped with Al⁰-Al⁰ electrodes configuration provides the high removal efficiency for TSS, COD and TDS with certain detention time and applied voltage. This technique is suitable for pretreatment of liquid waste with high concentration of TSS, COD and TDS. Handheld Niton XL3t XRF Analyzer confirmed that beside Al³⁺, there are other elements released during electrooxidation. All metals contained in plate sheet electrodes were attacked by OH⁻ during electrooxidation to form metal hydroxide together with increased detention time and applied voltage. Electroconductivity (EC) values of liquid waste were found to decrease together with increase of detention time due to decrease of metal ions that form hydroxide during electrooxidation and not only Al(OH)₃.

Keywords: Electro oxidation; Reactor; TSS; COD; TDS

Introduction

The global tofu market size was estimated at USD 2.31 billion in 2018 and is expected to expand at a CAGR of 5.2% from 2019 to 2025. Rising preference for vegan diet in developed countries on account of an increasing number of campaigns aimed at reducing animal slaughter is expected to remain a key driving factor. Tofu is considered as healthy and gluten-free food and has been observed that one out of every 133 Americans suffers from celiac disease. The prevalence of celiac disease was 4% in South America, 0.5% in Africa and North America, 0.6% in Asia, and 0.8% in Europe and Oceania [1]. Tofu contains moisture, fat and protein ranged from 4.91–6.13/100 g; 13.77–19.82/100 g and 31.78–36.56/100 g fresh weight, respectively [2]. Consumption of Tofu in Indonesia is 676 gr/month/capita in 2018 according to Statistics Indonesia and will be increase together with the growth of population. Water footprint of soybean curd product (tofu) is 2.154 m³/kg. Water footprint of soybean curd (Tofu) and soybean fermented (Tempe) consumption in Bandung city (West Java) is 93 m³/capita/year or 223 million m³/year [3]. Huge of clean water uses for making tofu must be considered as depletion of clean water resources. COD_{Cr} and BOD₅ contents of tofu liquid waste vary from tofu processing facilities in Sumatra and Java provinces, was 5000-8500 and 3500-7000 mg/L respectively [4,5]. Increasing of tofu production remains a large volume of liquid waste which discharge directly into waterbodies. This wastewater, once discharged, not only causes irreversible damage to the environmental balance but also contributing to the depletion of fresh water reserves, in this way creating a threat to the coming generations. The sustainability of water usage is the crucial concern of new world era for modern life style in terms of recovery and reused as a part of MDGs. Various advanced technologies have been developed to meet the requirements for wastewater treatment such physical (screening, filtration, adsorption and membrane separation), chemical (fenton's reagent, alum, ozonation etc), biological (bacteria growth, biofilter) and electro-oxidation. Electro oxidation, an electrochemical technology, has received growing attention due to involves no chemicals during pollutant removal [6,7]. Electro oxidation process is known has ability to reduce high concentration of organic matter pollutant (OMP) as mentioned in literatures [8-13]. Electro oxidation involved an electric current and sacrificial metallic plates (electrodes) instead of expensive chemical reagents uses as described elsewhere in electro oxidation process [14-16]. The principle of this method that is by immersing metallic Fe⁰ and/or Al⁰ in wastewater with apply of certain electric current will release metallic ion Fe³⁺ and Al³⁺ for destabilizing the electrical charge of colloidal particles. This colloidal particle forms a bigger aggregates which settle down for separation process [17-20]. Electro oxidation process has been utilized effectively for removal many kind of pollutant sources as mentioned in many literatures [21] except for tofu industry. The main uses of electro-oxidation are as a pretreatment to enhance the biodegradable properties of raw wastewater and/or as a treatment to decompose residual (refractory) matter [22]. Electro-oxidation processes have a greater effect on persistent and non-degradable pollutants from secondary biological treatment than traditional physicochemical methods [23]. Some researchers have systematically assessed the operation or capital costs of electro-oxidation in comparison with another process [24]. These analogous explorations have established a sound foundation for the application of electro-oxidation technologies. The efficiency and operation cost are the two key driving factors for determining its industrial-scale application on wastewater treatment plant.

Electro-oxidation technique was introduced to get the better removal performance in the continuous baffle flow electro-oxidation reactor (CBFER) and this technique could be said as simply and promising technique. In this study the effects of detention time, various of electric current and applied voltage against the organic matter pollutant removal was observed relate to the performance of continuous baffle flow electro-oxidation Reactor. Rectangular baffle flow reactor was constructed in laboratory scale and equipped with Al⁰ electrode which enable the pollutant's contact time a bit longer with the large square area of electrode. Experiment results are providing the basis for the subsequent industrial-scale application of wastewater treatment subjected to high concentration of degradable and undegradable of organic matter pollutant. Detailed information on its experimentally treatment is presented, which also provides valuable information condition for this process.

Materials and Methods

Electro-oxidation reaction of tofu liquid waste was estimated for determining the reactor sizes based on the baffle flow model. The sectional area of the wastewater flowing through the reactor was calculated based on the mass transfer [25]. The design capacity of this laboratory-scale electro-oxidation was 30 x 10 x 12 cm (L x W x H) and the operational upper level of reactor was 80 mm with total volume of operational reactor was 2400 cm³ (Figure 1). It was consisted of cell bodies, electrodes, and constant current DC power supplements. There were 8 of Al⁰-plate electrodes immersed in this reactor and the effective volume of each electrode was 12.800 mm³ (Length x Width x Thickness). There were four of Al⁰-plate electrodes connected to positive charge and the rest one connected to negative charge with monopolar configuration [(+), (-), (+), (-), (+), (-), (+), (-)]. These electrodes were positioned alternately at distances of 28 mm, perpendicularly to the waste-water flow direction through each electrode. The density of Al⁰-plate electrodes is 2,7 g/cm³. A constant DC power supplement was supplied with two voltages of 5 and 12 V. Detention time were 40, 50, 60, 70 and 80 minutes and flow rate were 60, 48, 40, 34.3, 30 ml.min⁻¹ respectively.

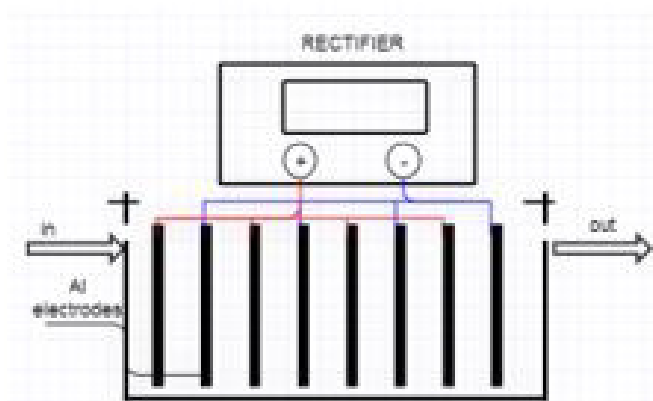


Figure 1: Rectifier power supply and continuous flow reactor with Al⁰-Al⁰ electrodes configuration

Tofu liquid waste was obtained from CV. NJ FOOD Babakan Ciparay, Bandung City, West Java Bandung-Indonesia, exactly before discharging to the sewer. It was taken to the laboratory in closed containers and kept at 5°C in freezer before performing analysis. The characteristic of tofu liquid waste and the standard methods were presented in table 1.

Parameter	Standard methods	Technique	Result	Unit
Temperature	SNI 06-6989.23-2005	Thermometer	23,20	°C
pH	SNI 06-6989.11: 2004	pH meter (electrode)	4,67	-
EC	SNI 06-6989.1: 2004	Conductometry	6,32	ms/cm
COD	SNI 6989.73: 2009	Closed reflux	10472	mg/L
TSS	SNI 06-6989.3: 2004	Gravimetry	16574	mg/L
TDS	SNI 06-6969.27: 2005	Gravimetry	10513	mg/L

Table 1: Characteristic of tofu liquid waste and standard methods

Each removal efficiency (RE%) of TSS, COD, and TDS was calculated using equation (1)

$$(RE\%) = \frac{C_0 - C_t}{C_0} \times 100 \dots\dots\dots (1)$$

Where C₀ is the initial concentration of TSS, COD and TDS (mgL⁻¹) and C_t is the concentration of TSS, COD and TDS at detention time (t). Faraday’s law of electrolysis could be written as an expression for the amount of substance, undergoing electrochemical change, in terms of the amount of electrical energy involved. Rate of Al weight lost in experiment was calculated before and after

electro-oxidation by weighting and theoretical rate of Al weight lost per unit area per unit detention time was calculated according to Faraday’s law [26] (equation 2).

$$-\frac{1}{A} \frac{dW}{dt} = \frac{(\sum M)j}{nF} xt \dots\dots\dots (2)$$

where $-\frac{1}{A} \frac{dW}{dt}$ is the weight lost of Al (g cm⁻²) in certain detention time, M is molecular weight of Al hydroxides (gmol⁻¹), j is applied current density (A cm⁻²) F is Faraday constant (96485 A.s.mol⁻¹), t is detention time (second) and n the number of electrons involved in reaction (n=3).

The sludges and floatation foams obtained from the reactor were collected and dried furthermore for determining the elements was released using handheld Niton XL3t XRF Analyzer.

Results and Discussion

Experiment was performed on a wide range of testing. The results are presented in form of figures, graphs and tables.

pH, Temperature and EC effect in hydrolysis

Figure 1 shows the values profile of pH, temperature and electroconductivity during Al⁰ oxidation on 40, 50, 60, 70 and 80 minute of detention time. Results shows that pH was increased together with the enhanced of detention time. On the voltage of 12 Volt, the pH values were bigger than voltage of 5 Volt. On the voltage of 12 Volt, electrocoagulation was occurred in the pH range 5-6 and on the voltage of 5 Volt, electrocoagulation happened in the pH range of 4.9-5.6. Enhanced of pH values is caused in cathodic the reduction of liquid waste forming OH ions and hydrogen gas occurred (equation 4). were formed in cathode.

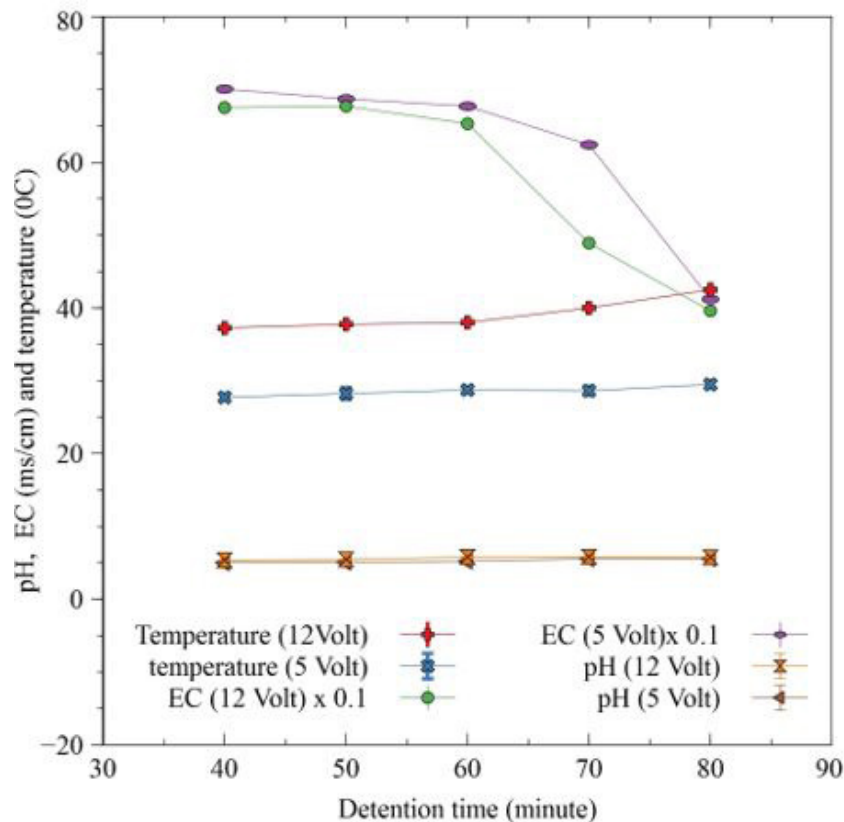
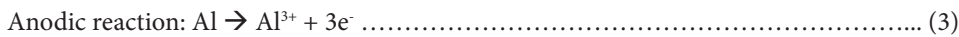


Figure 1: pH, EC and temperature electro-oxidation of tofu liquid waste at two applied voltages

While temperature tends to be increase with enhance of detention time and the applied voltage. This could be seen that voltage of 12 volt the temperature of tofu liquid waste was 37-43 °C and voltage of 5 volt was 27-30 °C. Increasing of temperature was caused of Al released electrons and its mobility in tofu liquid waste. This mobility of electrons tends decrease when the ions of metals is bind with inorganic and organic ions formed flocks as shown in the EC values. Increase of the detention time the EC was drop down from 6-7 ms.cm⁻¹ at 40-60 minutes to 4 ms.cm⁻¹ at 80 minutes for two applied voltages.

TSS, COD and TDS effects in electro-oxidation

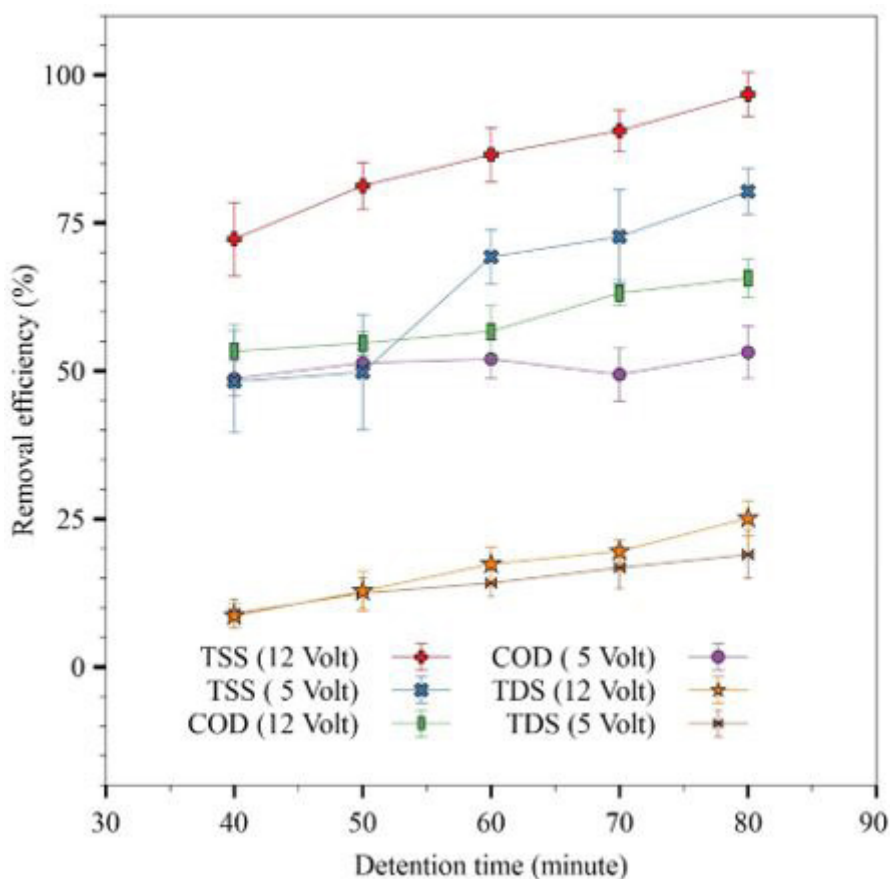
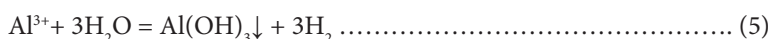


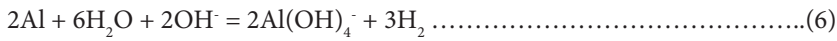
Figure 2: Removal efficiency of TSS, COD and TDS with different of detention time

Interesting results can be seen of removal efficiency profile for total suspended solid (TSS), chemical oxygen demand (COD) and total dissolve solids (TDS). These parameters were increase together with enhancing of detention time and voltage (Figure 1). Total suspended solid occupied the higher position of removal efficiency followed by COD and TDS respectively. Removal efficiency of total suspended solid was achieved 98.9% (12 volt) and 83.23% (5 volt), chemical oxygen demand was 70% (12 volt) and 57.5% (5 volt) as well as total dissolve solid was 29.78% (12 volt) and 23.55% (5 volt) at the 80 minute of detention time. The lower of removal efficiency at a pH less than 5 and higher than 7 was observed by many investigators and was attributed to an amphoteric behavior of Al(OH)₃ which leads to soluble Al³⁺ cations, when the initial pH is low and monomeric anions Al(OH)₄⁻, when the initial pH is high [27]. Al(OH)₃ could be possible present when reacted occurred between Al³⁺ with water in pH less than 7 (equation 5)



However, when the initial pH was kept in the range between 4.9 and 6, all aluminum cations produced at the anode with possibility formed various polymeric species such as Al₆O₁₅(OH)₂₄³⁺, Al₇O₁₇(OH)₂₄⁴⁺, Al₁₃O₄(OH)₂₄⁷⁺ and precipitated as Al(OH)₄ due to attack of OH⁻ ions [28] to Al⁰ in anodic (equation 6) leading to a more effective treatment in electrocoagulation. Adsorption of pollutants on these species needs time to create of agglomeration until settle down in the bottom of reactor and the same time hydrogen gas was produced in cathodic elevated species at the surface of reactor as flotation mechanism. As seen, the highest removal efficiencies have

been obtained at a level pH 6 (Fig. 2) and detention time was 80 for TSS, COD and TDS.



High removal efficiency of organic pollutants with electro-oxidation which described with the range of applied pH, detention time, electrolytic concentration, the configuration of electrode, type of electrodes and reactors (batch and continuously) for dealing with degradable and undegradable pollutants as demonstrated by many investigators. [29-35]

No	Analyse	Cd	Pd	Ag	Mo	Nb	Zr	Sr	Rb	Bi	Pb	Zn	Cu	Fe	Mn	Total (%)
	Units	%	%	%	%	%	%	%	%	%	%	%	%	%	%	
1	Al foam	0,237	0,163	0,275	0,020	0,010	0,005	0,003	0,002	< LOD	0,006	0,110	0,072	1,580	0,138	
2	Al Sludge	0,222	0,148	0,253	0,016	0,008	0,005	0,002	0,004	0,004	0,003	0,100	0,032	0,494	0,089	
3	Sum	0,459	0,311	0,528	0,036	0,018	0,010	0,005	0,006	0,004	0,009	0,210	0,104	2,074	0,227	4

Table 2: Elements of electrode was released during electro-oxidation

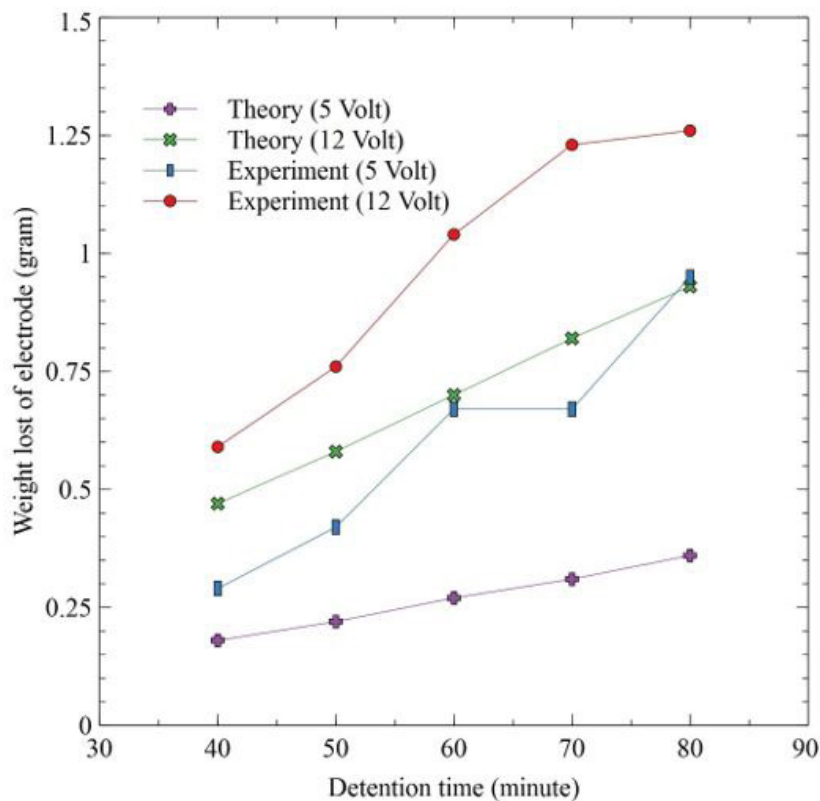


Figure 3: Weight loss of electrode was calculated theoretical and experimental

Plate No	Detention time (40 minute)			Detention time (50 minute)			Detention time (60 minute)			Detention time (70 minute)			Detention time (80 minute)					
	5 volt	0,08 A/cm2	Fara day (gr)	5 volt	0,08 A/cm2	Fara day (gr)	5 volt	0,08 A/cm2	Fara day (gr)	5 volt	0,08 A/cm2	Fara day (gr)	5 volt	0,08 A/cm2	Fara day (gr)			
1	0,29	0,17	0,11	0,49	0,22	0,66	0,61	0,27	1,55	0,70	1,17	0,31	0,72	0,82	1,17	0,36	0,87	0,93
2	0,19	0,18	0,11	0,17	0,22	1,13	0,42	0,27	0,76	0,70	0,37	0,31	1,25	0,82	0,37	0,36	0,57	0,93
3	0,49	0,18	1,35	0,47	0,40	0,64	1,09	0,27	1,70	0,70	0,50	0,31	1,46	0,82	1,50	0,36	1,46	0,93
4	0,09	0,18	0,24	0,47	0,24	0,57	0,39	0,27	0,30	0,70	0,47	0,31	1,29	0,82	0,47	0,36	1,29	0,93
5	0,52	0,18	1,28	0,47	0,65	0,85	1,06	0,27	1,65	0,70	0,79	0,31	1,68	0,82	1,79	0,36	1,88	0,93
6	0,13	0,18	0,43	0,47	0,18	1,20	0,49	0,27	1,19	0,70	0,63	0,31	1,69	0,82	0,63	0,36	1,69	0,93
7	0,55	0,18	1,09	0,47	0,69	0,72	1,11	0,27	0,98	0,70	1,19	0,31	1,39	0,82	1,39	0,36	1,39	0,93
8	0,08	0,18	0,09	0,47	0,52	0,30	0,18	0,27	0,17	0,70	0,27	0,31	0,40	0,82	0,27	0,36	0,96	0,93
Aver age	0,29	0,18	0,59	0,47	0,42	0,76	0,67	0,27	1,04	0,70	0,67	0,31	1,23	0,82	0,95	0,36	1,26	0,93
Total	2,35	1,43	4,71	3,76	3,34	6,07	5,34	2,16	8,28	5,60	5,38	2,48	9,87	6,56	7,58	2,88	10,11	7,44

Table 3: Weight lost of electrodes was calculated based on theory and experiment

Electrode weight lost

Many investigators have been paid attention for calculation the weight loss of electrodes in term of total energy consumption during electro-oxidation [34,35]. We compare the amount of Al weight lost by gravimetry with the theoretical amount was calculated using equation (2) and refers to stoichiometric reaction of equations 5 and 6. Table 2 shows the elements concentration of electrode was released during electro-oxidation in the dried foam and sludge using handheld Niton XL3t XRF Analyzer. Table 3 shows amount of each electrode weight lost in experiment with various applied electric current compare with the theory one.

Figure3 showed that the weight loss of electrode theoretically smaller than the experiment one, based on the applied voltage and also detention time, as was demonstrated by others. We believe that the theoretical calculation must involves the others elements of electrodes was released during electro-oxidation and not only Al^0 since the availability of plate sheet aluminum in the market with various grade and it is not pure enough for proving that only Al^0 was released during electro-oxidation process in reactor. It also possible if Al^0 and another element are dissolved in the liquid and influence the electro-oxidation process. Indeed, for proving this hypothesis the comprehensive experiment must be done.

Conclusion

This experiment has described that there is increase of removal efficiency of TSS, COD and TDS by increasing of detention time and voltage as well as giving the effect also for increasing of temperature from 27-30 °C on voltage 5 volt to 37 – 43 °C on 12 volts during electro-oxidation. pH range was increase from 4.94 – 5.6 become 5-6 in voltage 12 volt, shows that formation of Al hydroxide occurred together with increase of detention time and applied voltage. Reactor continuous baffle flow electro-oxidation reactor (CBFER) shows high performance removal efficiency for TSS, COD and TDS and was depended on the detention time and applied voltage as described in this experiment.

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