

## Electrocoagulation for spent coolant from machinery industry

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### ABSTRACT

Spent coolant is considered as one of the most polluting industrial wastes and causes environmental problems. It mostly contains high non-biodegradable organic carbon and oil contents; the biodegradability index was very low at 0.04, which is difficult to be effectively treated by common treatment systems. Electrocoagulation (EC) was proposed for a pre-treatment of coolant. The lab-scale of EC reactor was developed with Al electrodes and 10 mm of interelectrodes. The efficiency of the EC reactor on COD removal was investigated at various current densities and electrolysis times. The highest current density of 50 mA/cm<sup>2</sup> induced a short electrolysis time of 10 min to reach the steady state of approximately 65% COD removal. When lower current densities of 20–40 mA/cm<sup>2</sup> were supplied to the EC reactor, maximal COD removal efficiency of 65% can be achieved at longer electrolysis times. According to the specific energy consumption and sludge production, the optimal condition for spent coolant treatment was the current density of 20 mA/cm<sup>2</sup> and electrolysis time of 30 min in which a COD removal of efficiency of 65% was obtained, 0.88 kWh/kg-COD of the specific energy consumption and 0.04 kg/kg-COD of the sludge production.

**Key words** | current density, electrocoagulation, electrolysis time, spent coolant treatment

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### INTRODUCTION

Coolant is one of the principle chemicals in machinery manufacturers. The coolant is used to cool cutting tools and parts to minimize overheating during machining and grinding applications. In the manufacturing process, the coolant is continuously recirculated through the cutting tools until

the coolant becomes less effective and dirty. Subsequently, the spent coolant is collected as a waste for further treatment and disposal. Due to the water-soluble oil property of the coolant, it cannot be effectively treated by traditional treatment processes, such as biological processes and chemical precipitation which are commonly used in industry for wastewater treatment. To avoid the expensive cost for developing alternative efficient technology to treat the spent coolant, the industry mostly agrees to incinerate it in the

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doi: 10.2166/wrd.2017.057

cement industry as an alternative fuel in furnaces. In the meantime, the illegal disposal of spent coolant to lakes and rivers is also found in several areas, which causes serious environmental issues affecting human and aquatic lives (Marfe & Stefano 2016).

A large volume of spent coolant is generated in the machinery manufacturing industry which contains high non-biodegradable organic carbon, oil and metals contaminations. The concentrations are as follows: biological oxygen demand (BOD; refer to biodegradable organic carbon) of 5,200 mg/L, chemical oxygen demand (COD; refer to both biodegradable and non-biodegradable organic carbon) of 122,000 mg/L, oil and grease were 6,800 mg/L, and Fe was 32 mg/L (Siam Kubota Corporation 2015). The above pollutants concentrations exceed the effluent standard limit of the Industrial Estate Authority of Thailand to be further treated in central wastewater treatment plants (PCD 2015). According to a pre-treatment of chemical precipitation, four coagulants of  $\text{Al}_2(\text{SO}_4)_3$ ,  $\text{AlCl}_3$ ,  $\text{Fe}_2(\text{SO}_4)_3$  and  $\text{FeCl}_3$  were suggested for the spent coolant in a previous study (Hilal *et al.* 2004). The pre-treatment can decrease the total organic carbon (TOC) content from 44,200 mg/L in the initial concentration to 5,300 mg/L. When the spent coolant was treated by the pre-treatment and together with nanofiltration, the lowest TOC concentration of 3,800 mg/L was found (~91% removal efficiency). Further, a pilot plant included peat-bed filtration, and demulsification centrifugation and ultrafiltration were proposed for the spent coolant treatment (Benito *et al.* 2002). According to the plant design, an efficient COD removal of 90% was observed and the oil and grease content was effectively removed from 22,400 mg/L in the spent coolant to 30 mg/L in the final effluent.

Membrane technology is known to be a reliable process for oily wastewater treatment via pressure driven and pore size selection. The process is a direct realization of oil and water separation with no phase change. Abadi *et al.* (2011) employed a tubular ceramic microfiltration system for a typical oily wastewater. The TOC removal efficiency was greater than 95% and the treated water contained a low oil and grease content of 4 mg/L. Salahi *et al.* (2013) employed a sheet nano-porous membrane in order to treat oily wastewater from a petroleum refinery. The nano-porous membrane was an efficient system to treat the wastewater

and the treated water was able to be reused for agriculture; the efficiencies of oil and grease content, COD and BOD reached 99.9, 80.3 and 76.9% respectively. However, Yu *et al.* (2017) suggested that multi-membranes such as ultrafiltration and reverse osmosis were required for oily wastewater treatment, or a single membrane combined with other traditional methods. The membrane system is compact, easy to operate and cost-effective for a high membrane area per unit volume, however the cleaning system is required to periodically clean the membrane and restore the membrane flux.

Another treatment technology of electrochemicals is a potential newly developed technology for water and wastewater treatment. Among the electrochemical technologies of electrodeposition, electrocoagulation, electroflotation and electrooxidation, the electrocoagulation in which aluminium (Al), iron (Fe) or hybrid of Al/Fe was used as electrodes has been widely used in various applications. For example, the applications were for turbidity and colour removal, nutrient removal, oil and grease removal, and metal removal (Hong *et al.* 2013; Matias *et al.* 2015; Mores *et al.* 2016; Perez *et al.* 2016; Tian *et al.* 2016). Various operating factors of current density, electrolysis time, NaCl concentration, pH, electrode material and system design were suggested to significantly effect the performance of electrocoagulation (Chen 2004). In addition, the optimal conditions for electrocoagulation operation were different in typical wastewater; (i) current density of 30 mA/cm<sup>2</sup>, electrolysis time of 30 min, NaCl of 1 g/L and pH of 8 were appropriate conditions for livestock wastewater (Tak *et al.* 2015), (ii) current density of 35 mA/cm<sup>2</sup>, electrolysis time of 100 min and pH of 5 were suitable for fibre manufacturing wastewater (Gong *et al.* 2014), (iii) current density of 21 mA/cm<sup>2</sup>, electrolysis time of 20 min and hybrid electrodes of Al/Fe were the optimal conditions for swine manure effluent (Rahman & Borhan 2014).

The objective of this study is to evaluate the performance of a developed electrocoagulation (EC) reactor to treat the spent coolant from a machinery manufacturing industry. The four monopolars of Al electrodes were parallel in the EC reactor with an interelectrode distance of 10 mm. The effects of current density and electrolysis time, which were the most important operating factors, were studied to optimize the COD removal efficiency and the specific

energy consumption. The floating scum (sludge) which was a by-product from the treatment process was measured and characterized. According to optimal conditions, the experiments were further conducted at various pH values and NaCl additions.

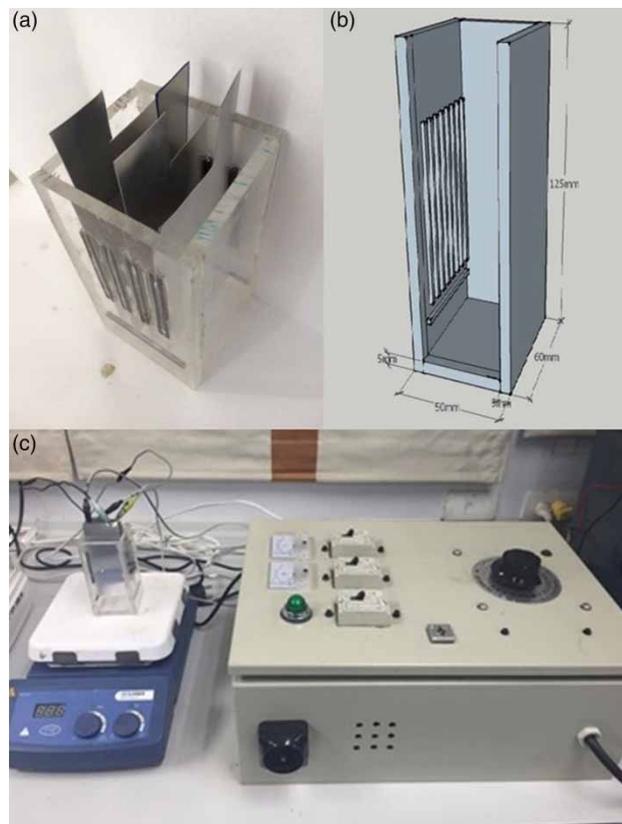
## METHODOLOGY

### Spent coolant

The spent coolant was collected from a machinery manufacturing industry in Thailand and kept in polypropylene bottles at 4 °C before being used in the experiments. The colour was milky brown. The spent coolant initially contained a high non-biodegradable organic carbon and oil, and the other pollutants parameters were as follows; COD of 112,000 mg/L, BOD of 5,200 mg/L, oil and grease of 6,800 mg/L, SS of 5,200 mg/L, total dissolved solids (TDS) of 11,500 mg/L and Fe 32 mg/L. The pH was around 10.5. During the experiments, the spent coolant was used with no pre-treatment, no dilution and no pH adjustment.

### Electrocoagulation (EC) reactor

The experimental setup is shown in Figure 1. The lab-scale EC reactor was made from a 5-mm acrylic sheet with a dimension of 5 cm (width) × 6 cm (length) × 12.5 cm (height) and a working volume of 0.2 L. Four parallel mono-polar electrodes were made from an Al sheet with the dimensions of 0.1 × 4 × 12.5 cm. The total effective electrode was 30 cm<sup>2</sup> and the interelectrode distance was 1 cm. The EC reactor was placed on a magnetic stirrer. A DC power supply (2 kW, 0–400 V, 0–5 A) was used to provide the electricity current to the EC reactor. The experiments were operated at a room temperature of ~25 °C, mixing speed of 150 rpm and NaCl concentration of 5 g/L. At the end of the experiments, the sludge was removed and the dry weight was measured. Then, the liquid was measured providing the remaining organic carbon as the COD value, which commonly represents the organic carbon content in wastewater.



**Figure 1** | (a) Actual electrocoagulation (EC) reactor, (b) drawing EC reactor and (c) experimental setup.

### Experimental design

In this study, the optimization of experimental conditions for treating the spent coolant from the machinery manufacturing industry was conducted under the developed EC reactor. The COD removal efficiency and the specific energy consumption were two parameters to evaluate the effectiveness of the system. The experiments were operated under various current densities (10–50 mA/cm<sup>2</sup>) and electrolysis times (2–60 min). Both variables extensively affected the overall performance, reaction rate and energy consumption.

### Analytical techniques

The efficiency of the EC reactor for the spent coolant treatment was evaluated by the reduction of the COD value, which is commonly referred to as the organic carbon content of the wastewater. The COD value was measured by using a COD analyzer (AL200 COD Vario, Aqualytic).

The other parameters, such as biochemical oxygen demand (BOD), suspended solids (SS), TDS and oil and grease content, were measured by an environmental consultant company; the methods of measurement are presented in Table 1. The pH value was detected by using a pH meter (Eutech Instruments). The COD removal efficiency and the specific energy consumption were calculated using Equations (1) and (2) (Gong *et al.* 2014).

For sludge characterization, the floating scum was collected. The remaining treated water was allowed to settle overnight. Then, the settle sludge was filtered and oven dried at  $103 \pm 2$  °C until the weight of the sludge samples stabilized. A composition of sludge content was characterized by an acid digestion method (EPA 2016).

$$\text{Efficiency (\%)} = \left(1 - \frac{\text{COD}_{\text{final}}}{\text{COD}_{\text{initial}}}\right) \times 100 \quad (1)$$

$$\text{Energy consumption} \left(\frac{\text{kWh}}{\text{m}^3}\right) = \frac{U \times I \times t}{V} \quad (2)$$

where  $U$  is the voltage (in V),  $I$  is the applied current (in A),  $t$  is the reaction time (in hours) and  $V$  is the treated volume (in L).

## RESULTS AND DISCUSSION

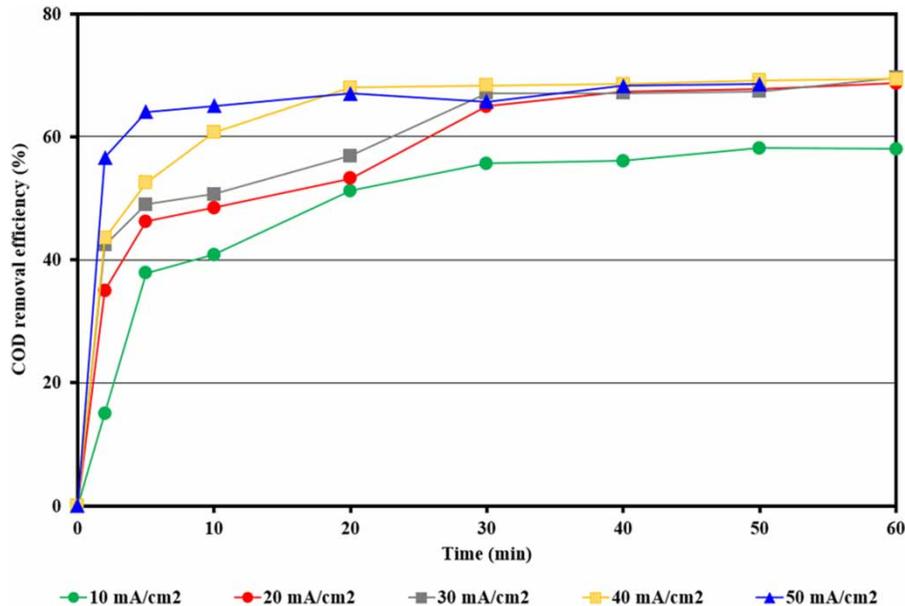
### Optimization of experimental condition

The EC reactor experiments were performed at various current densities of 10, 20, 30, 40 and 50 mA/cm<sup>2</sup> with the

actual pH of the spent coolant being 10.5. At the electrolysis time <30 min, the increasing current densities gradually improved the COD removal efficiency, as shown in Figure 2. At the electrolysis time of 10 min, the highest COD removal efficiency of 65% was observed at the current density of 50 mA/cm<sup>2</sup>, whereas the COD removal efficiency was around only 60, 50, 48 and 40% at the lower current densities of 40, 30, 20 and 10 mA/cm<sup>2</sup> respectively. These results revealed that the increasing current density effected the enhancement of the COD removal rate. This is because the readily degradable organic carbon content in the spent coolant was immediately removed when starting the electrocoagulation process. The readily degradable organic carbon content was completely removed by 10 min when the high current density of 50 mA/cm<sup>2</sup> was supplied to the EC reactor (presented in the steady state of COD removal). However, the period to complete the readily degradable organic carbon removal was increased to 20 and 30 min for the lower current densities of 40 and 30 mA/cm<sup>2</sup>. It can be summarised that the increasing current density can enhance the COD removal efficiency, however the electrolysis time also positively correlates to the performance of the EC reactor; the reactor operating under the lower current densities required longer electrolysis times to achieve stable COD removal efficiency of approximately 65–70%. In the meantime, a reduction of pH was observed during the electrocoagulation process. The pH sharply decreased from 10.5 to 8.6 when the current density of 50 mA/cm<sup>2</sup> was supplied to the EC reactor for 10 min, whereas the pH was slightly decreased to 9.5–9.8 at the lower current densities. When the steady state of COD removal was

**Table 1** | Characteristics of the spent coolant and treated water by electrocoagulation

Parameters	Concentration		Removal efficiency (%)	Analysis method
	Spent coolant	Treated water		
pH	10.5	7.7	–	Electrometric method
BOD (mg/L)	5,200	410	92.1	5-Day BOD test
COD (mg/L)	122,000	35,500	70.9	Close reflux
SS (mg/L)	3,500	1,200	65.7	Dried at 103–105 °C
TDS (mg/L)	11,500	25,300	–120.0	Dried at 103–105 °C
Oil and grease (mg/L)	6,800	350	94.8	Liquid-liquid, partition gravimetric
Fe (mg/L)	32	1.6	95.0	Acid digestion



**Figure 2** | COD removal efficiency from the spent coolant at various current densities and electrolysis times.

reached, the pH was insignificantly different for all current densities; the pH range was 7.7–8.0. The pH reduction was mainly due to the oxidation of hydroxyl ions at the anode surface during electrolysis (Aswathy *et al.* 2016).

At >30 min of the electrolysis time, the current density had no effects on improving the COD removal efficiency. The COD removal efficiency was in the range of 65–70% at current densities of 20–50 mA/cm<sup>2</sup>. However, the lowest current density of 10 mA/cm<sup>2</sup> was insufficient to effectively treat the spent coolant; the COD removal efficiency was found to be only 58% at the steady state. This is because at the low current density, small quantities and particles of aluminium hydroxide were generated, which do not allow efficient adsorption of the destabilized pollutants. The biodegradability index (BOD<sub>5</sub>/COD) in the treated water was evaluated at the electrolysis time of 30 min in which the steady state of the spent coolant treatment was observed. The results show that the biodegradability index rose with increasing current densities (see Figure 3). The biodegradability index was 0.09 at the current density of 20 mA/cm<sup>2</sup>, and reached 0.20 at the highest current density of 50 mA/cm<sup>2</sup>. It can be seen that the concentration of COD was stable whereas the BOD concentration rose with the current densities. This phenomenon shows that more non-biodegradable organic carbon was decomposed to readily

biodegradable organic carbon when the higher current density was supplied. The performance of the EC reactor to treat the spent coolant was not enhanced at long electrolysis times (>30 min), however the biodegradable organic carbon content increased in the treated water, and could be possibly further treated by traditional treatment plants.

The operating cost of the electrocoagulation process is dominated by energy consumption. To optimize the operating condition for the industry site, the COD removal efficiency and the specific energy consumption was plotted (see Figure 4). The specific energy consumption was increased by current densities and electrolysis times. The optimal condition was the current density of 20 mA/cm<sup>2</sup> and the electrolysis time of 30 min, which achieved COD removal efficiency of 68% and consumed the energy of 0.88 kWh/kg-COD. According to the electricity cost of Industrial Estate of Thailand, the cost for the spent coolant treatment by electrocoagulation was approximately 3.5 Baht/kg-COD, which was a more economical treatment than incineration. Although the lower treatment cost by electrocoagulation was reported in previous studies (Bensadok *et al.* 2011; Al-Shannag *et al.* 2015), typical wastewater characteristics (i.e. dairy wastewater and metal plating wastewater) containing a low COD and oil content is the significant reason.

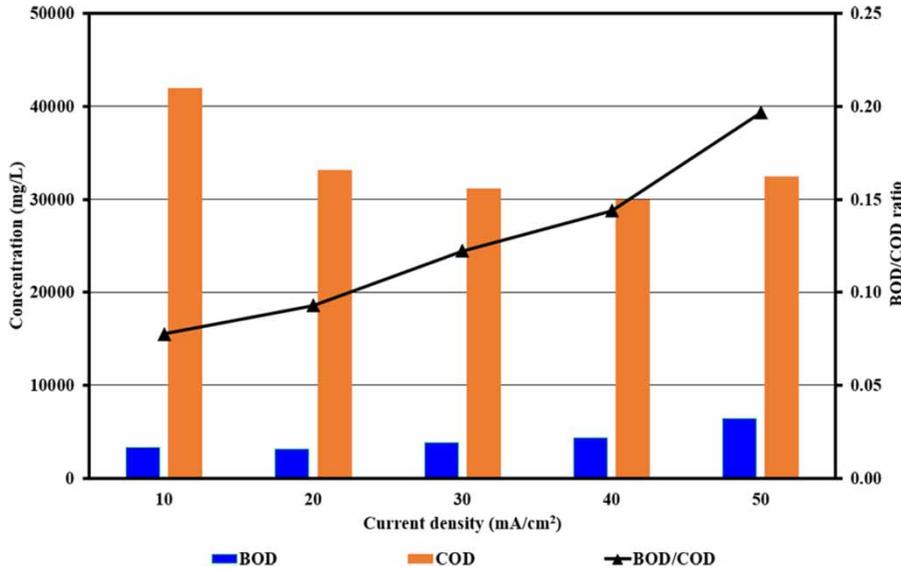


Figure 3 | BOD and COD concentrations and biodegradability index (BOD<sub>5</sub>/COD ratio) at various current densities (electrolysis time of 30 min).

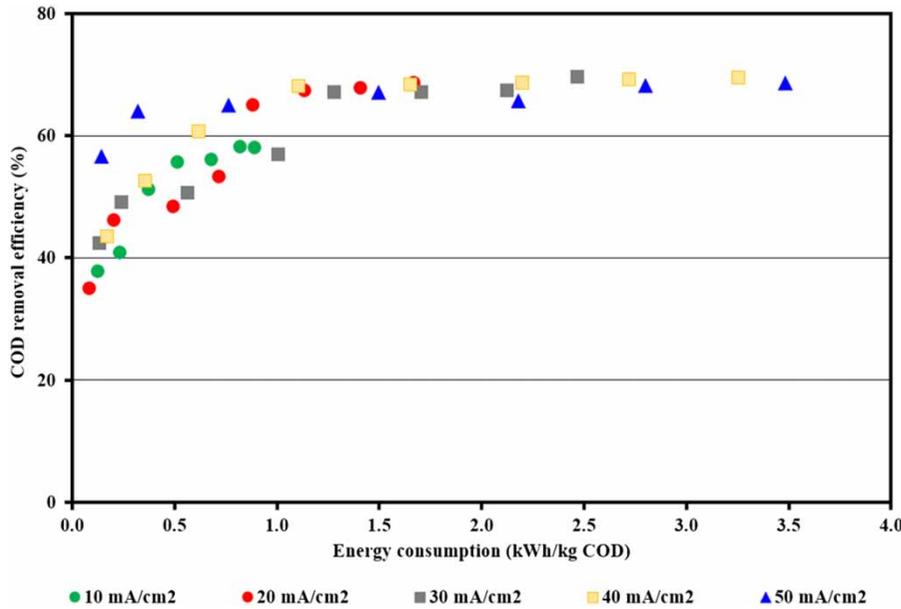
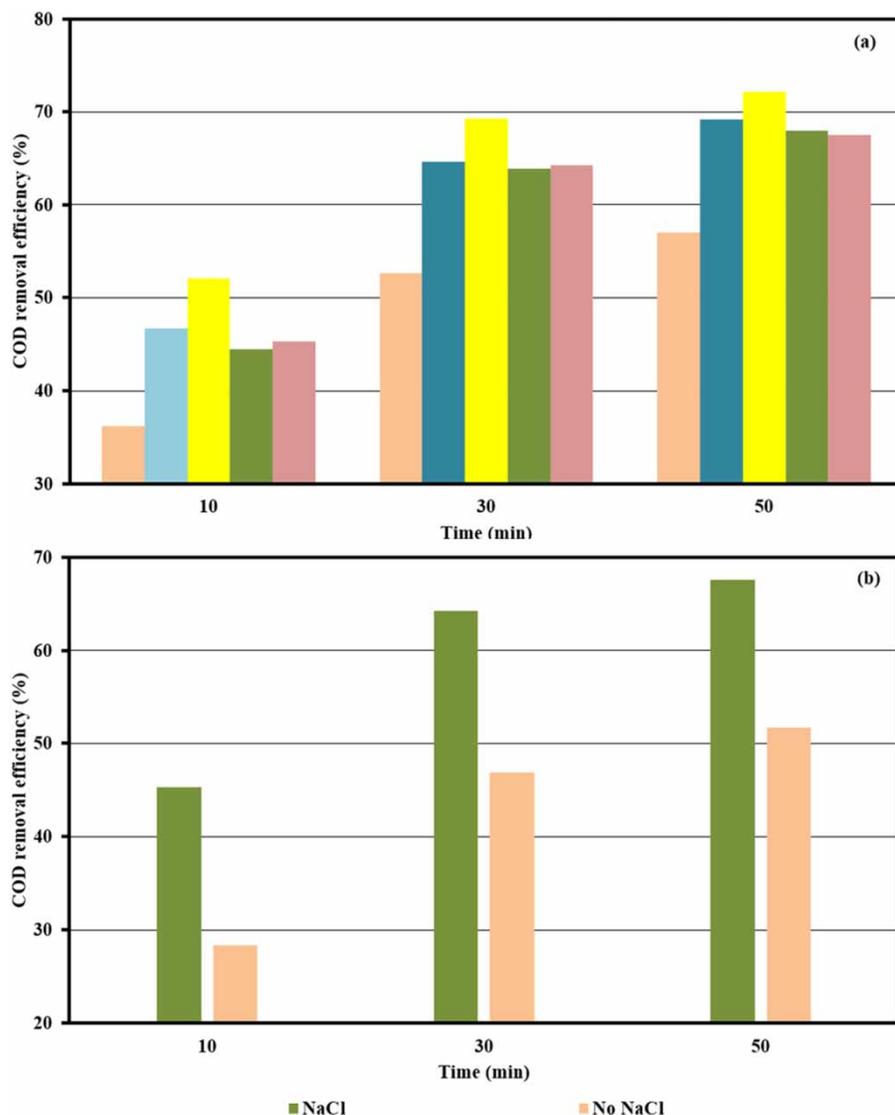


Figure 4 | Relationship between COD removal efficiency and specific energy consumption at various current densities and electrolysis times.

Under optimal conditions, the experiments were further studied at various pH values and no NaCl addition. As the pH increased from 3 to 7, the COD removal efficiency was increased from 52 to 70% (Figure 5(a)). The COD removal efficiency decreased to 64–68% at the higher pH of 9 and no pH adjustment. The different removal efficiency

at various pH values was due to the dissolution of Al electrodes. The Al<sup>3+</sup> and Al(OH)<sub>2</sub><sup>+</sup> were predominant at the lower pH, and the hydroxide ions (OH<sup>-</sup>) were neutralized by the photon, resulting in less amounts of aluminium hydroxide formation (Daneshvar *et al.* 2007). In the meantime, the Al<sup>3+</sup> and OH<sup>-</sup> ions were generated by Al electrodes to



**Figure 5** | COD removal efficiency at (a) various initial pH values and (b) NaCl additions (current density of 20 mA/cm<sup>2</sup>).

transform into the insoluble form of  $\text{Al}(\text{OH})_3$  at the pH range of 5–7 (Akyol 2012). The flocs precipitated with organic carbon in the spent coolant, resulting in the higher removal efficiency obtained. When pH was 9 and higher, the negatively charged form of  $\text{Al}(\text{OH})_4^-$  occurred and it caused the redissolution of flocs (Linares-Hernandez *et al.* 2009). A similar phenomena was observed at the electrolysis time of 10 and 50 min. It is noted that to achieve the high COD removal efficiency at pH 7, the cost of chemical consumption should be included due to the alkaline condition of the actual spent coolant.

The experiments were further studied with no external NaCl addition. The results are shown in Figure 5(b), the COD removal efficiency was decreased from 64 to 47% at the electrolysis time of 30 min. The electrical conductivity of the spent coolant was around 10.7  $\mu\text{S}/\text{cm}$  and it was raised to 17.5  $\mu\text{S}/\text{cm}$  after maintaining NaCl of 5 g/L. The results revealed that the increasing conductivity showed the fast reduction of organic carbon during the electrocoagulation process. This is because the high electrical conductivity resulted in a large amount of electrode cations ( $\text{Al}^{3+}$ ) released, which were responsible for the organic

carbon removal (Franco *et al.* 2017). As mentioned above, the neutral pH and high electrical conductivity of the spent coolant enhanced the efficiency of COD removal, however the cost-effectiveness should be a concerning aspect to judge the appropriate operating condition to treat the spent coolant at the industrial site.

### Sludge characterization

At the end of the experiments the sludge was collected and measured as a dry weight. The results Figure 6 show that the amount of sludge generated was continuously increasing by current densities and electrolysis times. This is because numerous Al ions were produced at the high current density and/or long electrolysis time, and the ions were formed to aluminium oxide such as  $\text{Al(OH)}_3$  and  $\text{Al(OH)}_4^-$  (Hong *et al.* 2013). Subsequently, the aluminium oxide particles were floated to the EC reactor surface by  $\text{H}_2$  gas generated during the process and adsorbed the destabilized pollutants (Chen 2004). Further, the aluminium oxide particles were also deposited as a sediment in the EC reactor base. It can be noted that the amount of sludge was not in correlation with the COD removal efficiency. At the optimal condition (current density of  $20 \text{ mA/cm}^2$  and electrolysis time of 30 min), sludge of 3.5 g was observed which was approximately 0.04 kg in 1 kg-COD removal. The sludge

production increased to the maximal value of 5.4 g at the conditions of  $50 \text{ mA/cm}^2$  and 60 min. Therefore, the increasing cost for further sludge treatment and disposal must be spent when the EC reactor is operated under higher current densities and electrolysis times rather than optimal conditions. To support the above explanation of pollutants removal mechanisms, the main compositions of sludge content were characterized. The results showed that the sludge contained high carbon which represented the organic carbon removal from the spent coolant. In addition, the sludge contained high Al of 4.6 g/kg from the electrodes decomposition and Fe of 0.4 g/kg from the spent coolant composition.

The water quality of treated water was measured and is summarized in Table 1. The treated water was neutralized to pH of 7.7 with no acid and chemical addition after the treatment. The electrocoagulation process also improved other parameters of water quality; the BOD and Fe was effectively reduced by 92–95% and the SS was reduced by 66%. On the other hand, the TDS concentration was increased from 11,500 mg/L in the spent coolant to 25,300 mg/L. The increasing TDS caused the addition of NaCl for maintaining the good performance of the EC reactor. It can be seen that not only the organic carbon was removed, but the electrocoagulation also removed other pollutants such as BOD, SS and Fe. This study indicated that the EC reactor using

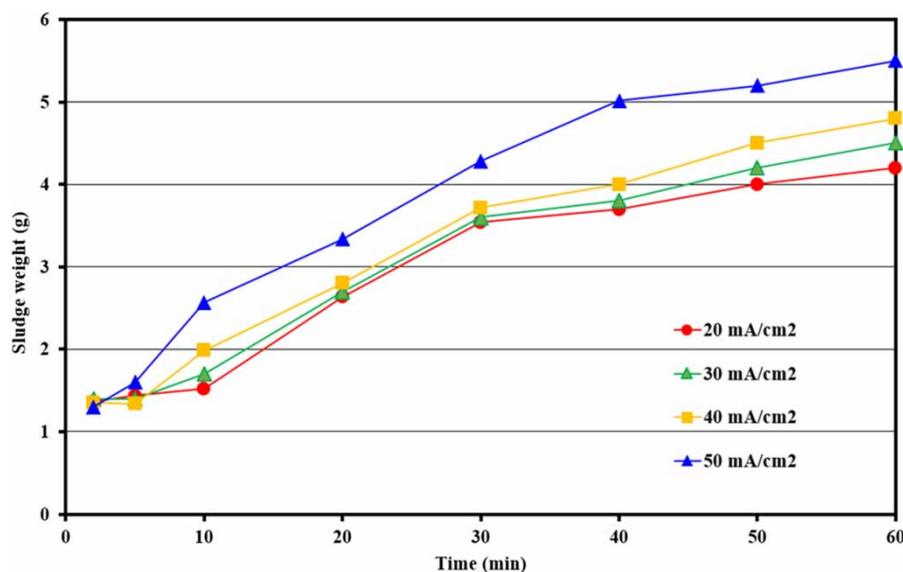
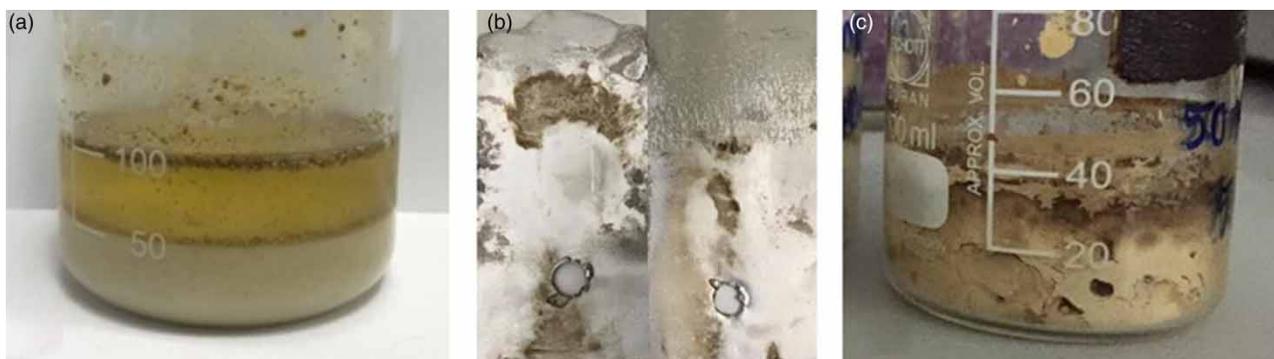


Figure 6 | Amount of sludge from electrocoagulation at various current densities and electrolysis times.



**Figure 7** | (a) Spent coolant after treatment, (b) Al electrodes after treatment, and (c) sludge after treatment.

Al-Al electrodes was a potential pre-treatment system with the purpose of reducing the organic carbon prior to sending to centralized wastewater treatment plants. The actual spent coolant, electrodes and sludge after the treatment are shown in [Figure 7](#). Further, the enhancement of the EC reactor and/or combined with other post-treatment processes should be studied.

## CONCLUSIONS

In this study, the EC reactor was demonstrated for a pre-treatment of spent coolant from a machinery industry. According to the results, the non-biodegradable organic carbon was effectively removed at the high current density of  $50 \text{ mA/cm}^2$  with around 65% COD removal efficiency after 10 min of electrolysis time. Although the maximal COD removal efficiency in the steady state was of a similar value under varying current densities, the increasing current density significantly enhanced the biodegradability index ( $\text{BOD}_5/\text{COD}$ ) of the spent coolant. Under the optimal conditions (current density of  $20 \text{ mA/cm}^2$  and electrolysis time of 30 min), the EC reactor can remove 70.9% of COD, 92.1% of BOD and 94.8% of oil and grease. In the meantime, 0.04 kg of sludge was produced to remove 1 kg of COD in the spent coolant, and the specific energy consumed was about 0.88 kWh/kg-COD. This preliminary study shows that the electrocoagulation is a potential and feasible process for pre-treating the spent coolant prior to post-treatment, such as a bioreactor.

## ACKNOWLEDGEMENTS

The authors would like to thank Naresuan University Research Funding for financial support during the experiment.

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First received 26 September 2017; accepted in revised form 1 December 2017. Available online 30 December 2017