Optimization of Copper Removal by Photovoltaic Electrocoagulation from Aqueous Solution Using Response Surface Methodology Towards Sustainable Development

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ABSTRACT

This research aims at illustrating the optimal functions of removing copper ions in aqueous solution by means of the electrocoagulation process in which portable solar power generators are used as renewable energy. A solar photovoltaic cell (PV), producing approximately 48A current intensity for 4–7 h per day, was sufficient to charge the lithium batteries completely during the day. This system was connected directly to the electrocoagulation tank. The Box-Behnken design (BBD) was applied to evaluate three effects of process factors: current density, the dose of electrolyte (NaCl), and application time. The results showed that an optimal efficiency of 99.01% Cu removal plus an energy savings of 1.039 kWh/m³ were obtained at a current density of 4 A/m², the dosage of NaCl (electrolyte) of 1.87 g/L, and electrolysis time of 10 min. The chemical components of the sludge produced under these optimized conditions were determined by means of EDX. It was illustrated that the copper ions were the main elements of sludge, and nonhazardous compounds were contained. The PV-lithium battery system is considered to be an efficient alternative energy source toward sustainable development.

Keywords: photovoltaics; electrocoagulation; copper; Box-Behnken design; energy consumption; lithium battery; sustainable development

INTRODUCTION

Copper is considered one of the most toxic heavy metals for living cells, and it is one of the more widespread heavy metal pollutants in the environment [Kamaraj et al., 2013]. The elevated levels of copper ions occur from industrial sources, such as electroplating, paints and dyes, petroleum refining, and metal production The application of agricultural chemical, fertilisers, wood burning or waste incinerators, as well as sewage treatment processes commonly contributes to the generation of copper ions and their release to the environment [Shrivastava, 2009]. Besides, the lower concentration of copper is discharged naturally from the Earth's crust. The surface water can become polluted by acids, bases, and heavy metals, such as lead and copper that originate from various industrial processes, and they can have a severe effect on the people's health. Copper is a fundamental element demanded health and well-being in humans as it is included in a difference of proteins and metalloenzymes, which accomplish crucial metabolic functions for tissue and bone development. However, the excessive Cu^{2+} utilisation has an impact on the liver, and subsequent vomiting causes headache, nausea, respiratory problems, abdominal pain and, lastly, gastrointestinal bleeding [Simsek et al., 2013; El-Ashtoukhy and Abdel-Aziz, 2013; Parlak and Arar, 2018]. Degradation of copper ions from water can be achieved by using the chemical or physicochemical methods [Gunatilake, 2015] such as adsorption [Hossain, 2012; Balintova et al., 2012; Demcak et al., 2017] ion-exchange [Edebali and Pehlivan, 2016], photocatalysis [Kanakaraju et al., 2017], or electrochemical processes [Kamaraj et al., 2013; Dermentzis et al., 2011]. Compared to this research, electrocoagulation compares well as an alternative to the conventional wastewater

removal techniques [Al-Saydeh et al., 2017]. Electro-coagulation is the electrochemical generation of destabilisation operators that induce the charge of neutrality level for removing contaminants and has already been utilized for treating water and wastewater [Vasudevan, 2012]. The Al and Fe materials are most regularly applied as an electrode due to the following advantages: availability, low price, and non-toxicity. The iron and aluminium ions generated from these electrodes can withstand hydrolysis at the surface of an anode to perform a sequence of activated intermediates that can diminish the delicately dispersed molecules appearing in the wastewater. Then, the flocs can be formed from aggregating of destabilised particles [Vasudevan et al., 2009]. When aluminium is applied as an anode, the chemical reactions which are given below occur:
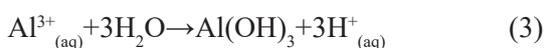
At the cathode:

$$2H_2O+2e^-\rightarrow H_{2(g)}+2OH^- \tag{1}$$

At the anode:

$$Al\rightarrow Al^{3+}+3e^- \tag{2}$$

In solution,

$$Al^{3+}_{(aq)}+3H_2O\rightarrow Al(OH)_3+3H^+_{(aq)} \tag{3}$$

However, the major disadvantage of EC is its high electricity consumption, which also constitutes a critical environmental issue and has economic impact. Thus, the application of traditional electrical energy to power an electrocoagulation reactor is the primary barrier to wider utilization in developing countries where the electrical grids are less reliable [Cucchiella et al., 2016; Ganiyu et al., 2019].

Photovoltaic panels (PV), have been used as a reliable, available and unlimited renewable energy source to replace the traditional electricity [Montiel et al., 2018]. The application of battery energy storage (capacitance) has been used to provide the energy produced on a bright sunny day, which is stored for use on a raining day or at night. Furthermore, using photovoltaic battery systems solves the issue of tremendous output power frequently encountered in PV arrays [Ganiyu et al., 2019]. As such, an electrocoagulation system powered by a PV-battery cell can operate full time, regardless of the solar intensity [Ganiyu et al., 2019]. Several studies have illustrated the feasibility of PV energy in the treatment of contaminants found in the water and wastewater [Ganiyu et al., 2019; Khemila et al., 2018;

Hussin et al., 2017; Millán et al., 2018; Nawarkar and Salkar, 2019]. Nevertheless, these studies paid attention to the availability of applying a PV energy source for degrading the organic pollutants from the water and wastewater. Thus, the PV applications must further be investigated in terms of heavy metals removal [Hussin et al., 2017].

The efficiency of the EC process influences many elements such as pH, applied electric current, the dosage of electrolyte, and the time reaction [Ölmez, 2009]. In traditional multi-factor experiments, optimization is usually realized by changing a factor while keeping other components fixed at a particular arrangement of conditions. Recently, Box – Behnken Design (BBD) in a response surface method (RSM) has become a valuable design tool for the optimisation of processes. BBD provides far-reaching consequence and detailed info indeed in a smaller number of experiments and can display the interactive effects of experimental components on all responses [Kabuk et al., 2014; Shokri et al.,2017].

Accordingly, this research was undertaken for removing copper ion (Cu^{2+}) -contaminated water by a photovoltaic-electrocoagulation (PV-EC) process using aluminium electrodes to optimise the degradation capabilities of copper and the system energy consumption. Additionally, the optimal process applying BBD tool of Design-Expert Software (11 trial, Stat-Ease Inc. Minnepolis) (DX11) was realized by three parameters, namely current density, experimental duration, and concentration of the of support electrolysis. The trial and model-predicted information obtained the best settings for the PV-EC process design with the least energy consumption and highest removal efficiency of Cu^{2+}. The sludge was produced at the optimum operating parameter of the EC process, examined by means of EDX.

MATERIALS AND METHODS

Chemicals

Copper sulfate $CuSO_4$•$5H_2O$ (Merck, Germany), sodium hydroxide (NaOH, Merck), sulfuric acid (HCl, Merck), sodium chloride (NaCl, Merck) of analytical reagent were employed to prepare the aqueous solutions for the experiments. The pH of the solutions was changed by adding 0.1M HCl or 0.1M NaOH. All solutions were made with double distilled water at room temperature (25°C ± 1) every day.

PV-lithium battery system and electrocoagulation process setup

The PV-EC experiments were conducted using two PV panels made of polycrystalline silicon (ArmSolar, Singapore) with a size of 659×387×25 mm, connected in arrays with a total power supply of 70W. The portable solar generator, including MPPT (Solar V, Vietnam) converted the electricity directly from the solar panels to the lithium battery (Solar V, 12 V/240 Ah each) (Fig. 1). The portable solar generator was regulated by the current supply to the electrocoagulation tank.

A lab-scale electrocoagulation reactor constituted a 1 L glass cell that was 10 mm (width) × 12 mm (length) × 15 mm (height) and was set up with two parallel monopolar electrodes with a 2-cm interelectrode distance. The anode and cathode of the electrocoagulation reactor were made of Al with dimensions of 2×7×17 mm. Before each test, the aluminium electrodes were prepared as follows: washing with 0.1 M HCl to remove surface matter and then rewashing with the distilled water, drying in the oven and weighing. Copper sulfate, $CuSO_4 \cdot 5H_2O$, (Merck, Germany), was used with distilled water to make a concentration of 50 mg/L copper. The pH of the aqueous solution, analyzed by means of a digital pH-meter (Eutech, Singapore), was adjusted up to 5 by 0.1 M HCl (Xichlong, China) and 0.1 M NaOH (Xichlong, China).

The PV-EC process was carried out in aqueous solutions containing 50 mg/L Cu^{2+} and concentrations of NaCl from 1g/L to 3 g/L, in batch mode. For retention times ranging from 10 to 30 minutes, different current densities varying between 1–4 A/m² were changed with a rheostat (PHCAN-B009PINJNS, China). Finally, the sample was filtered; in order to determine this filtrate, the electrodes were dried and reweighed. A quantity of sludge formed from the PV-EC system was analyzed by filtering, using a 0.45µm filter paper (Whatman, UK) then drying at 103–105°C in an oven (Blinder, UK).

Experiment design

The optimization of the experimental setting for the degradation of copper by PV-EC process was performed by applying the Box – Behnken design (BBD). Three main factors were chosen to estimate the impact on experimental parameters for the copper degradation and energy consumption: current density, A (A/m²), (X_1), initial the dosage of NaCl (g/L) (X_2), and electrolysis time (min) (X_3), as shown in Table 1.

Analytical methods

The copper was determined with the atomic absorption spectroscopy method (AAS) (Savant-AA Σ, GBC, Australia) according to the EPA 7000B standard. The sludge after the electrocoagulation process was analyzed with an X-ray diffractometer (JSX-1000S, JEOL, Japan).

The copper removal efficiency (% Cu) was calculated as:

$$\%Cu = (1 - \frac{H}{H_0})x100\%, \tag{4}$$

where: H_0 and H are the initial and final copper concentrations.

Energy consumption was calculated with the following equation:

$$W\ (Wh/m^3) = \frac{UxIxt}{V} \tag{5}$$

where: W is the energy consumption (Wh/m³),
U is the applied voltage (V),
I is the current (A), t is time (h), and
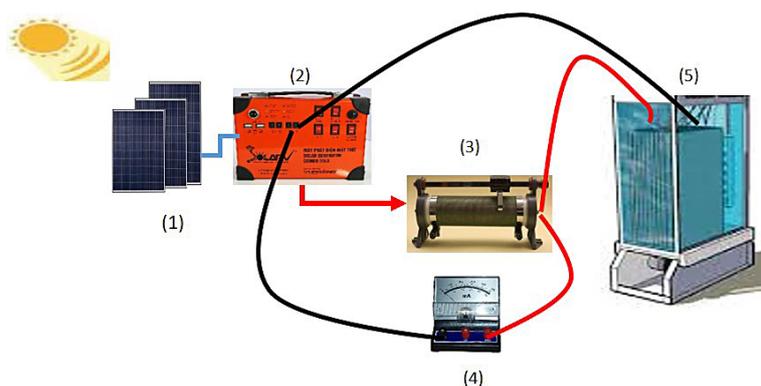V is the volume of the sample (m³).

Data analysis

For the calculations of the PV-EC operation setting, the response variable was fitted by a second-order model in the form of the quadratic polynomial equation, which is as follows:

$$Y = b_0 + \sum_{i=0}^{k} b_i X_i + \sum_{i=0}^{k} b_{ii} X_i^2 + \sum_{i=0}^{k} \sum_{j=0}^{k} b_i b_j X_i X_j \tag{6}$$

where: Y is the predicted response (Copper removal efficiency, % and energy consumption, kWh/m³) applied as the dependent variable;
X_i (i = 1, 2 and 3) are the independent variablefactors, and b_0, b_i (i = 1, 2 and 3), b_{ii}, and b_{ij} (i = 1, 2 and 3; j = 1, 2 and 3) are the model coefficients, respectively.

Figure 1. Diagram of the experimental setup: 1 – solar panel, 2 – solar generator, 3 – rheostat, 4 – ammeter, 5 – electrocoagulation reactor

Table 1. Coded and actual levels of the variables of the design of experiments for the overall electro-coagulation optimization

Factor	Variables	Coded levels of variables		
		-1	0	1
Current density	A/m²	1	2.5	4
NaCl concentration	g/L	1	2	3
Electrolysis time	Min	10	20	30

Experimental output was analysed by the DX11 (trial) program with ANOVA to show the interplay between the process values and the responding variable. 2-D contour plots and 3-D contours of the response surfaces were performed in the same program.

RESULTS AND DISCUSSION

Response analysis and explanation by BBD method

The 3-factorial experiments were carried out under the BBD with the DX11 software using the predicted and experimental data of the responses (copper removal (Y_1) and energy consumption (Y_2)), which are presented in Table 2. The response functions with the fix coefficients for copper and energy consumption are performed with equations (7) and (8)

Y_1 (copper removal, %) = 58.94333 + 9.13583X_1 + 5.20042X_2 + 0.851750X_3 − 1.25167 X_1X_2 − 0.13X_1X_3 − 0.009763 X_3^2 (7)

Y_2 (energy consumption, kWh/m³) = 1.87181 − 2.81528 X_1 − 5.58833 X_2 − 1.25167 X_1X_2 − 0.121667 X_1^2 + 0.151125 X_2^2 − 0.009763 X_3^2 (8)

The response function forecasts suited the experimental data completely (Table 3) (R^2=99.77% for Y_1 and R^2=99.66% for Y_2). Both of the correlation coefficient values (R^2) achieved in the current research for these dependent variables were higher than 0.80, pointing out that the regression modellings justified the reaction correctly. Thus, the response surface model promoted in this research for the prediction of removing copper feasibility and energy consumption was determined acceptably [Ölmez, 2009].

The quadratic regression technique is appropriate, being apparent from the F-test with a shallow probability value (Prob > F). Table 3 resulted in ANOVA analysis considering the significance of factors in PV-EC experiment through equation (7) and (8). Because the p-values of each coefficient are less than 0.05, it shows that indicate model terms are significant. From Table 3, the Y_1 model terms X_1, X_2, X_3, X_1X_2, X_2X_3, and X_3^2, as well as Y_2 model calls X_1, X_1^2, X_2^2, X_3^2 had a significance of impact on the energy consumption because the p-value is less than 0.05.

The surface response of the second-degree polynomial model with a variable was maintained at an average value and the two remained ones changing in an experimental range are described in Figures 2–4. In Figure 2, the response surface was shown as a function of current density and the initial NaCl concentration as electrolyte concentration within the operation time kept at 20 min. The average value, 98.6% Cu²⁺ removal, was obtained at 20 min application time while the current density was 4 A/m² and the electrolyte (NaCl) concentration was 3 g/L. Similarly, Figure 3 investigates the response under the dosage of variables such as current density and electrolyte

Table 2. Independent variables and response from Box-behken Design

Run	Independent variables			Response (Y)			
	X_1 (A/m^2)	X_2(g/L)	X_3(min)	Experimental		Predicted	
	Current density	Dose of NaCl	Time	Copper removal (%)	Energy consumption (kWh/m^3)	Copper removal (%)	Energy consumption (kWh/m^3)
1	4	2	10	96.8	1.07	96.81	1.11
2	2.5	3	30	95.3	1.96	95.40	2.13
3	1	2	10	82.7	0.64	82.66	0.6862
4	2.5	2	20	92.8	1.07	92.30	1.12
5	4	3	20	98.2	5.59	98.06	5.46
6	2.5	2	20	91.7	1.25	92.30	1.12
7	4	2	30	95.5	1.07	95.54	1.02
8	2.5	3	10	92.7	2.3	92.83	2.39
9	1	1	20	82.4	2.74	82.54	2.87
10	1	2	30	89.2	0.64	89.19	0.5987
11	2.5	1	10	87.6	0.23	87.50	0.0550
12	2.5	1	30	90.3	0.22	90.18	0.1325
13	2.5	2	20	92.4	1.04	92.30	1.12
14	1	3	20	91.7	5.75	91.61	5.62
15	4	1	20	96.5	3.74	96.59	3.87

Table 3. ANOVA outputs for copper reduction and energy consumption by PV-EC process

Source	Copper removal (%)					Energy consumption (Wh/m^3)				
	DF	SS	MS	F-value	Prob>F	DF	SS	MS	F-value	Prob>F
Model	9	313.20	34.80	243.78	<0.0001 significant	9	43.55	4.84	162.99	<0.0001 significant
X_1-Current density	1	211.05	211.05	1478.40	< 0.0001	1	0.4186	0.4186	14.10	0.0132
X_2- Dose of NaCL	1	55.18	55.18	386.52	< 0.0001	1	9.12	9.12	307.11	< 0.0001
X_3-Time	1	13.78	13.78	96.54	0.0002	1	0.0153	0.0153	0.5158	0.5048
X_1X_2	1	14.10	14.10	98.77	0.0002	1	0.2652	0.2652	8.93	0.0305
X_1X_3	1	15.21	15.21	106.55	0.0001	1	0.0000	0.0000	0.0000	1.0000
X_2X_3	1	0.0025	0.0025	0.0172	0.9008	1	0.0272	0.0272	0.7606	0.4230
$X_1{}^2$	1	0.2767	0.2767	1.94	0.2226	1	8.20	8.20	276.14	< 0.0001
$X_2{}^2$	1	0.0845	0.0845	0.5917	0.4765	1	12.13	12.13	408.62	< 0.0001
$X_3{}^2$	1	3.52	3.52	24.65	0.0042	1	11.37	11.37	383.10	< 0.0001
Residual	5	0.7138	0.1428			5	0.1484	0.0297		
Lack of fit	3	0.0938	0.0313	0.1008	0.9524	3	0.1226	0.0409	3.17	0.2491 not significant
Pure error	2	0.6200	0.3100			2	0.0258	0.0129		
R^2		0.9977					0.9966			
R^2 adjusted		0.9936					0.9905			
R^2 predicted		0.9908					0.9538			

concentration. It demonstrated a significant increase in the energy consumption (Wh/m^3) when the current density was increased. The electrolyte concentration was shown not to have much impact on energy saving.

In order to research the influence of electrolyte dosage and the operation time on copper reduction, the experiments were performed with the dose of electrolyte (NaCl) range were from 1–3 g/L in other contact times at a current density of 2.5 A/m^2. The results are discussed in Figure 4 showing that the copper degradation efficiency increased along with the application time at all investigated doses of electrolyte. The maximum copper concentration was removed at 30 min and with an electrolyte concentration of 3 g/L. Thus,
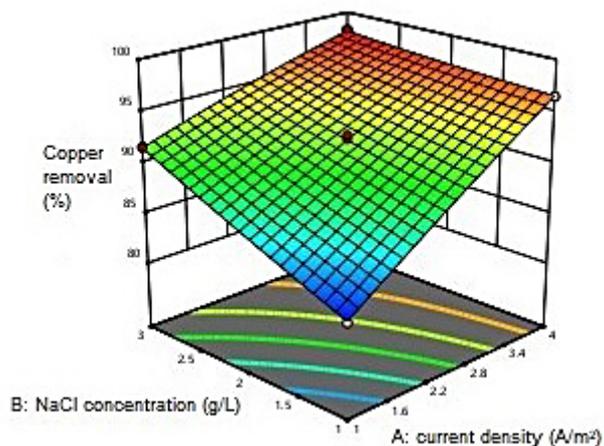
Figure 2. The effect of current density and electrolyte (NaCl) concentration on copper removal
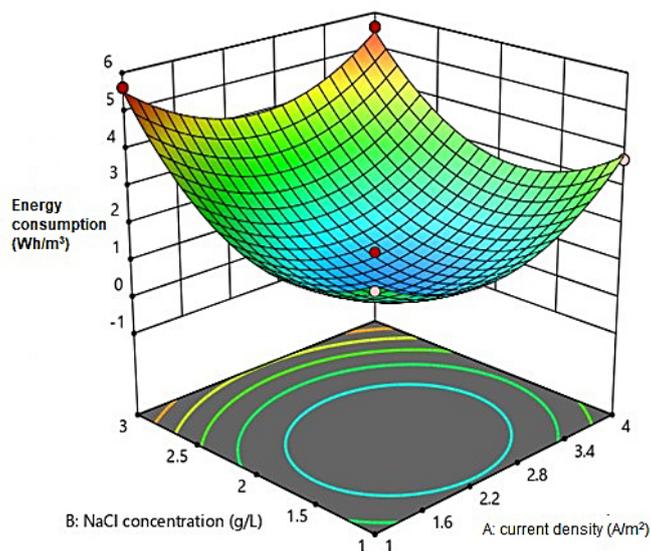(application time: 20 min)

Figure 3. The impact on current density and electrolyte concentration on energy consumption
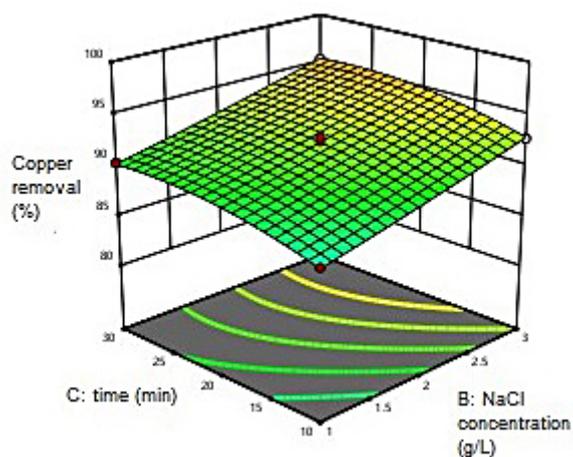(application time: 20 min)

Figure 4. The effect of the dosage of NaCl and application time on copper removal (the current density 2.5 A/m²)

it can be showed that the operation time has a more considerable influence on the Cu^{2+} removal efficiency than the dose of electrolyte.

Optimization of the PV-EC process for copper removal

The primary goal of optimization is to show the optimal values of the variables for copper reduction and energy saving with PV-EC from the variety achieved by applying experimental input. The aim desired for the response (the effect of removing copper) was selected to be a maximum target value with the lowest amount of energy consumption, which was affected by the applied voltage, the dose of electrolyte, and operation time. The optimization of the PV-EC process for the copper reduction is shown in Table 4. The optimal condition of the experimental variables was current density at 4 A/m^2, NaCl concentration = 1.87 mg/L, and time = 10 min, while the predicted responses were 96.72% and 1.03 kWh/m^3 for copper reduction and energy consumption, respectively. The desirability function value was determined to be 0.886 under the optimum conditions. As shown in Table 4, PV-EC is considered a suitable technology for optimum degradation of Cu^{2+} ions under a reasonable operating setting.

An additional experiment was illustrated under the optimal experimental setting based on the results showed in Table 5 to confirm the model accuracy and potency of the optimization procedure. The experimental results have approximately the same value as the predicted results under the optimum conditions. These results verify that BBD is a powerful tool for optimizing the operational requirements of EC for copper removal.

Sludge from the EC process

The EDX spectra show the elemental compositions of the sludge generated by the electrocoagulation process with Al electrodes, given in Figure 5. Copper ions are shown to be the primary component of the sludge at 15.85%. Other elements, including carbon, chlorine, oxygen, and sulfur, which could appear with the presence of organic and inorganic matter (Cl^-), and sodium, were shown with the weight percentage of each element in Table 6. Thus, the sludge formed by the PV-EC process in this research is also non-hazardous, which makes its management and disposal more convenient and less expensive.

Prospective application of solar energy-based electrocoagulation system

Alternatively, the coupling of these two technologies, electrocoagulation and PV with battery storage can provide a useful system and show many advantages, such as no requirement to be connected to an electric grid [Montiel et al., 2018] and low carbon emissions [Üçtuğ and Azapagic, 2018]. The research of Pirkarami et al. illustrated the removal of organic compounds: acid orange 2, reactive blue 19 [Pirkarami et al., 2013], phenol, and aldehydes in the wastewater of a resin factory [Olya and Pirkarami, 2013] with the use of a solar PV – EC with a battery system to save 17% of the total costs.

Lithium-ion batteries are an accessible technology with benefits over metal-acid ones, such as improved lifespan, faster charge and environmental friendliness [Üçtuğ and Azapagic, 2018]. Liang et al. compared the global warming potential (GWP) of lithium-ion and GWP of nickel metal hydride batteries and illustrated that the lithium batteries generated ten times less CO_2

Table 4. Optimum experimental conditions of PV-EC process variables for maximum copper removal and the lowest energy consumption.

Parameter	Optimum value
Current density (Am^{-2})	4
Electrolyte concentration (g/L)	1.862
Application time (min)	10

Table 5. Predicted and experimental levels for the responses at optimum operating conditions.

Type of value	Copper reduction, %	Energy consumption, kWh/m^3
Predicted	98.72	1.039
Experimental	99.01	1.3

Table 6. Elemental composition of sludge from the EC process based on weight (%).

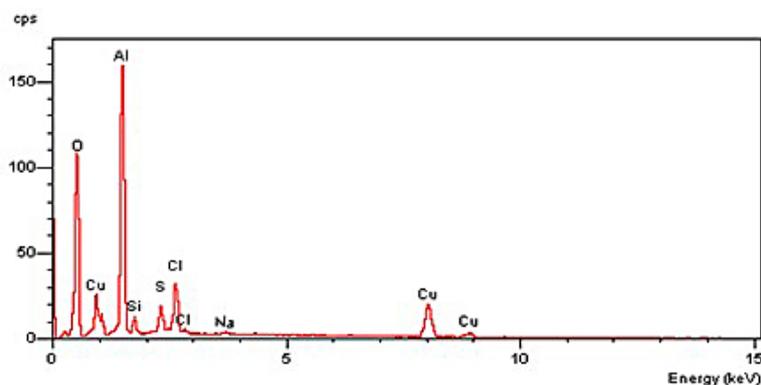| Elements	Weight (%)
C	0.7
O	21.21
Al	27.44
Na	13.69
S	2.86
Cl	18.25
Cu	15.85
Total	100

Figure 5. EDX images of sludge generated during the PV-EC process.

than the nickel metal battery (12.7 vs 124 kg CO_2-eq., respectively) [Liang et al., 2017]. The report concluded that lithium batteries were eco-friendly with lower CO_2 emissions, no pollution with metals and greater sustainability than other batteries such as lead acid, nickel cadmium batteries.

CONCLUSIONS

Solar PV-lithium battery powered electrocoagulation system has been successfully applied for the copper reduction in aqueous solutions:

1. BBD was a reasonable method to optimize the operating status of PV-EC for a decrease of ion Cu^{2+}. Evaluation of the variance described a high coefficient of dedication value ($R^2 = 0.9977$). The optimal conditions for the highest copper removal with the least amount of electric consumption were determined to be as follows: 4 A/m^2 current density, 1.8 g/L electrolyte (NaCl) concentration during 10 minutes.

2. The sludge formed after the EC process is non-hazardous and displayed excellent dewatering characteristics. Hence, the management and stabilizing of the sludge are easy and simple.

3. The developed solar PV-lithium battery device arrangement is beneficial for the reduction of copper ion and it is also cost-effective and eco-friendly.

Acknowledgements

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