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Electrocoagulation by rotated anode: a novel reactor design for textile wastewater treatment

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Malaysian Batik

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RESEARCH BACKGROUND

Textile industry is one of the major water-intensive chemical processes. It generates a huge amount of wastewater and creates significante conomic and environmental problems (Erick Butler et al. 2011).

Electrochemical technologies, such as electrodeposition, **electrocoagulation (EC)**, electroflotation (EF), electrooxidation and electrokinetic remediation, have received significant attention during the past two decades because they offer the possibility to be easily distributed, and require minimum amount and number of chemicals (Nazih et al., 2010).

We focus on **EC technology** as a **treatment prior** to **sedimentation** and **filtration** units in TWWTP.

ELECTROCOAGULATION

The coagulated ions are produced *in situ* and it involves 3 successive stages

1. Formation of coagulants by electrolytic oxidation of the sacrificial electrodes.
2. Destabilization of the contaminants, particulates suspension and breaking of emulsions.
3. Aggregation of the destabilized phase to form flocs.

Electrodes which produce coagulants into water are made from either iron or Al. Iron and Al cations dissolve from the **anodes** according to Eqs. 1 and 2.

At anode,

$Fe(s) \rightarrow Fe^{n+}(aq) + ne^-$ (1)

$Al(s) \rightarrow Al^{3+}(aq) + 3e^-$ (2)

At cathode according to Eq. 3

$2H_2O + ne \rightarrow H_2 + 2OH^-$ (3)

In solution the +ve ions are attracted to the -ve hydroxide ions to produce ionic hydroxides that have a strong attraction towards dispersed particles as well as counter ions to cause coagulation layer. (**Yadav, 2010**).

PROBLEM STATEMENT

1. Conventional coagulation treatment approach in textile mill has a serious setback (Ajjam, and Ghanim, 2012) and can be summarized as :
   - Creating a discrete addition of low dense and weak coagulant
   - Low efficiency in improving water quality (COD, BOD, TSS, Turbidity, and color) from 25% to 60%,
   - Long retention time 60-90 min,
   - Large quantity of sludge after treatment 6-10 kg/m^3,
   - Large quantity of chemical coagulants 10kg/m^3 which are high cost, a
   - Addition of chemicals CaO, NaOH, NaCl, and Na_2SO_4 for pH adjustment where it creates high cost (10.66USD/m^3)

PROBLEM STATEMENT Cont..

2.According to previous research of EC process, no set configuration is applicable to all needs, the problem of conventional EC reactor, can be summarized (Gunukula, 2011-Wang *et al.*, 2007-Molla,2004-Moreno,2009 –Ali and Yakob, 2012) :

The generated hydrogen (H_2) bubble accumulates onto the electrode surface forms an insulating layer (oxide film). Due to this layer:

- The electrodes will be passive and reduce activity of electrodes ,
- large electrical energy required to achieve optimum efficiency,
- IR drop between electrodes , and long reaction time gives high operational cost.

RESEARCH OBJECTIVES

1. To design new electrocoagulation system to treat TWW in order to
   i. Reduce energy from fossil fuel
   ii. Reduce accumulation of hydrogen at the cathode
   iii. Improve removal efficiency of TWW pollutants
   iv. Improve time for water treatment
   v. Applicable for batch and contnous flow system
2. To investigate the mechanism of EC treatment and hydrogen production
3. To verify the unique design by studying the reactivity of the anode (passivation and adsorption phenomenon)

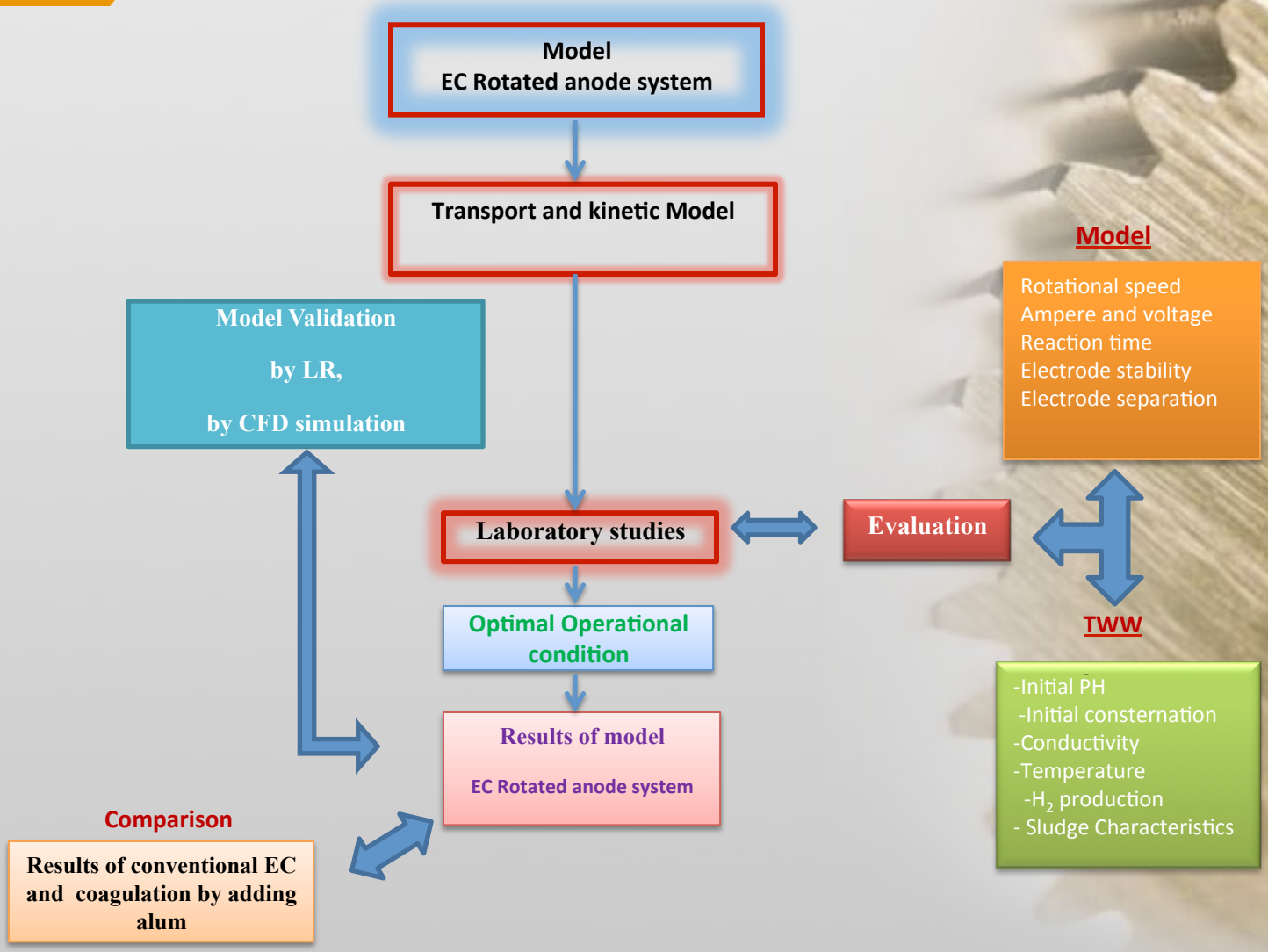
RESEARCH SCOPE

1. Application of solar energy to the system
2. Design unique model of EC with rotated electrode
3. Optimise on the operational conditions such as current, rotation speed, etc.
4. Test on effluent quality from the real and synthetic textile wastewater
5. Measure water quality parameters on effluent to check efficiency.
6. Characterise the sludge produced using zeta potential test and XRD, etc.

LITERATURE REVIEW

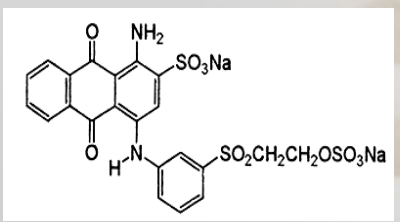
References	Waste Type	Model Description (static electrode)	Optimal condition	Results	Advantage	Disadvantage
Sarala (2012)	DWW	EC Fe electrode	CD=3.5mA/cm2 RT=90min,PH7	COD 76%, TSS 91%,Color 98%.	Low current, High TSS%, Color %	Low COD%, adjust pH, Long RT , High Cost
Zongo et al (2012)	Tannery WW	EC Pilot, Al or Fe Electrode	CD=67.5A.m-2, 62.5 V, RT=60min	COD 85%, turbidity 96%, color 91%, Chromium 99%	High Cr %, Turbidity%	High current, High EEC , Low COD% , High Cost
Ali and Yaakob (2012)	POME WW	EC Al Electrode	C= 5 A - 4V RT=8 hr, pH=4.5	COD57% , turbidity 62%, H_2=22.6 L/h	H_2 production	Low removal %, Long RT , High EEC , and High Cost
Quinones et al (2009)	Leather WW	EC Al Electrode	C=5A-30V RT=60min,PH7	COD 90%, Turbidity 85%, (TSS) 90%	High COD%	Low turbidity%, High EEC , Adjust PH, and High Cost
El-Ashtoukhy et al. (2013)	ORWW	EC fixed bed Al anode raschig rings (NM)	CD=8.59mA/ cm2, RT=2 hr, pH = 7	phenol 99% COD 91%	High removal %	High current, Adjust pH, Long RT High EEC, and High Cost

RESEARCH METHODS

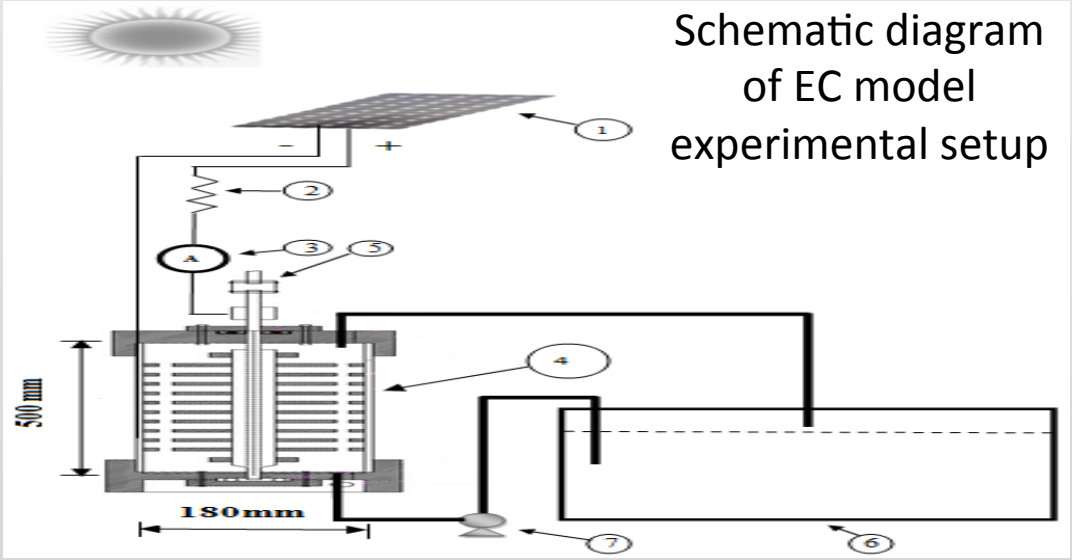


TEXTILE WASTE WATER

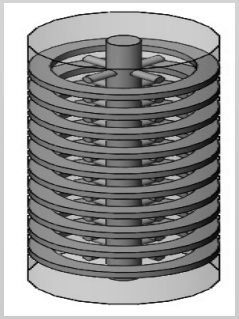
Characteristics of textile wastewater and properties of Reactive Blue 19 dye

Parameters	Values and Colour properties
Electrical conductivity (μS/cm)	19000
COD (mg/L)	4010
pH	7.00
Colour generic name	Reactive Blue 19
Synonym	Remazol Brilliant Blue R
Chemical Structure	
Chemical Formula	$C_{22}H_{16}O_{11}N_2S_3Na_2$
Molecular Weight (g/Mol)	626.50
λ max (nm)*	455

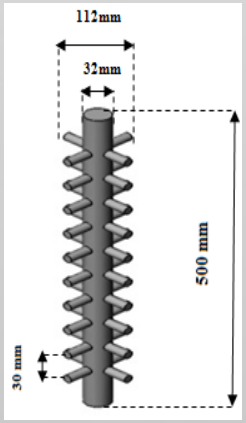
EC model with rotated electrode



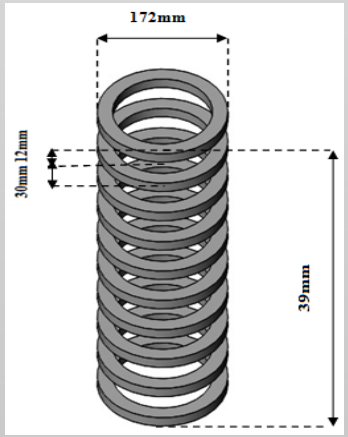
Pictures of EC model experimental setup



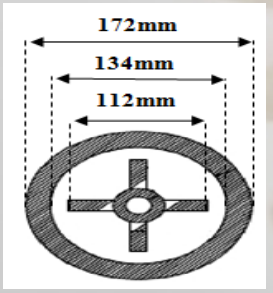
Anode and Cathode



Impellers Anode



Rings Cathode



Top view / Anode and cathode

Solar Feed

Experimental Procedure

1. Cathode and anode were attached with a solar energy cell (TPSM6U-300W, maximum current 8.02 A, maximum voltage 37.42 V, dimensions 1956*992 mm, 72 cells). Solar panel was fed by the sun simulator inside the lab to ensure continuity of energy.
2. The EC batch 10L working volume in 72 times, for real textile wastewater 36 times and for synthetic reactive blue 19 dye were 36 times. The colour was 300 mg/L of reactive blue 19 dye. Operational parameters of EC batch reactor : RT (ranging from 10 to 30 Min), Current densities of 2, 4 ,6, 8, 10, and 12 mA/cm^2, Rotational speed of anode (75, 100, 150, 200, 250) RPM,
3. Recirculation flows rate (20 L recirculated at 6, 12 and 21 L/min) and continuous flow system (Q= 0.333 L/min) at the optimal conditions of EC batch.

Experimental Procedure

4. Mechanism of treatment using zeta potential test and XRD techniques to elucidate the reaction behavour of aluminium hydroxyl with pollutants during the treatment process.
5. The enhancement of Hydrogen gas production using H2 detector under optimal current density with respect to rotational speed of anode.
6. The effect of rotational speed of anode on passivation and adsorption phenomenon by Impedance spectroscopy experiments.
   - The potential amplitude of the AC signal kept at 10 mV, and the measured frequency range was 0.01 –105 Hz. An EC-Lab SP-300 potentiostat with EC-Lab software V10.12 was used.
   - Impedance runs by three-electrode system, consisted of an aluminium electrode (1:25 of the original size) as the working electrode, Ag/AgCl (3 M KCl) electrode as a reference electrode, and a platinum wire as counter electrode.

Results and Discussion EC Rotating Model

Results: EC Rotated Anode (batch reactor)

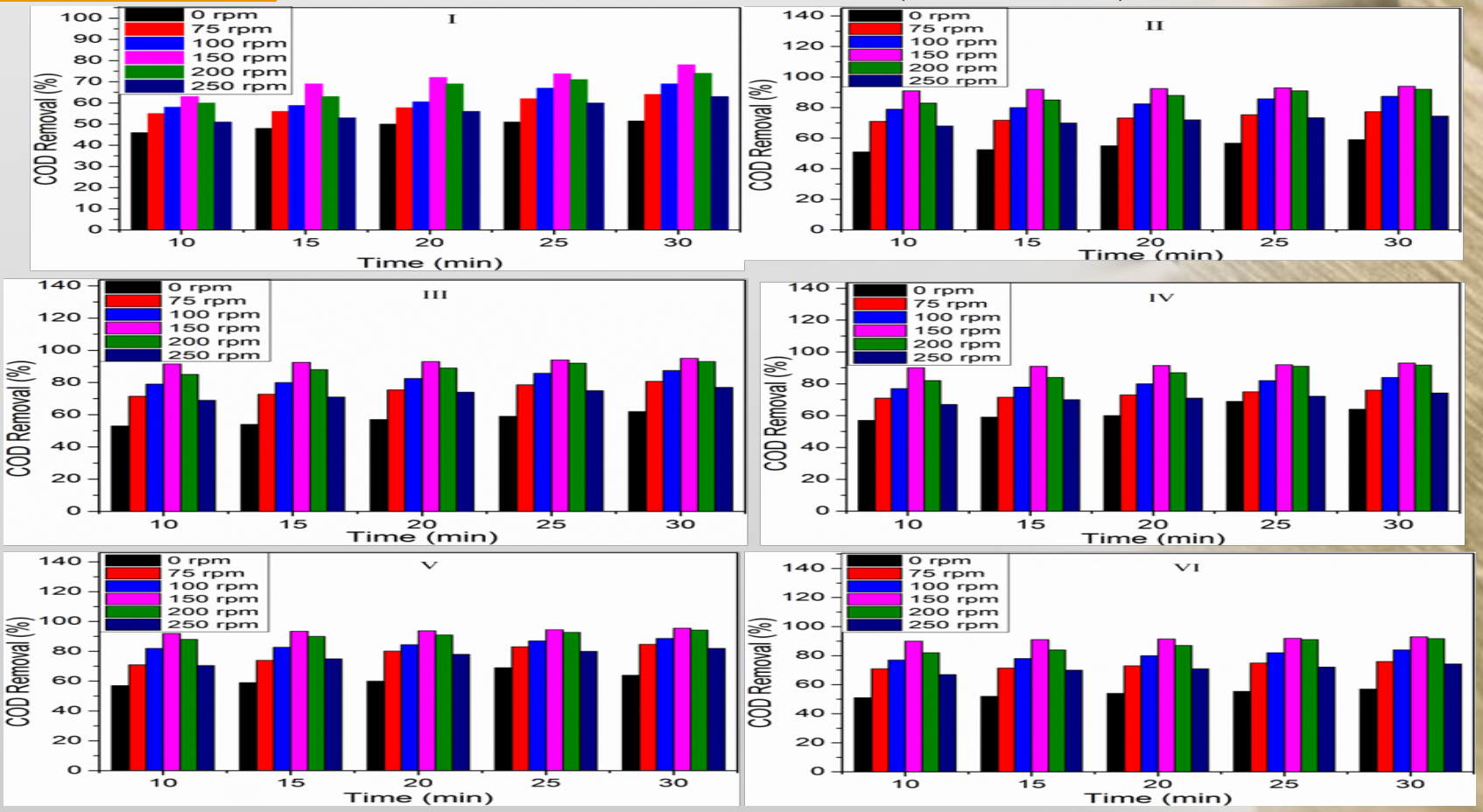


Fig. 2 (a). Effect of rotational speed on COD removal of real textile wastewater : I. CD = 2 mA/cm^2; II. CD = 4 mA/cm^2; III. CD = 6 mA/cm^2; IV. CD = 8 mA/cm^2; V. CD = 10 mA/cm^2; VI. CD = 12mA/cm^2

(optimal conditions for 92% COD removal was CD=4mA/cm^2, RT=10min, 150RPM)

Results and Discussion

EC Rotated Anode (batch reactor)

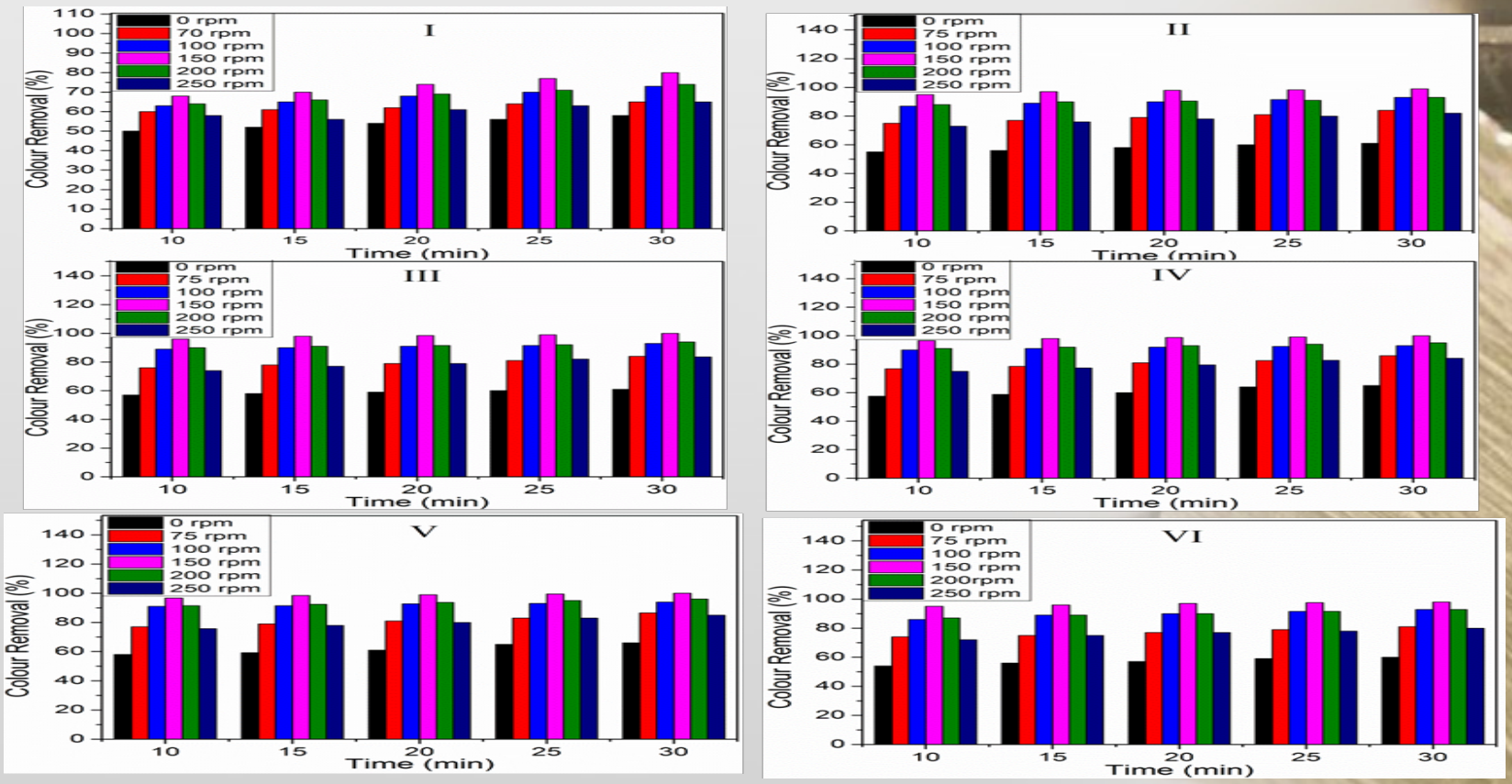


Fig.2 (b). Effect of rotational speed on colour removal (reactive blue 19) : I. CD = 2 mA/cm^2; II. CD = 4 mA/cm^2; III. CD = 6 mA/cm^2; IV. CD = 8 mA/cm^2; V. CD = 10 mA/cm^2; V. CD = 12mA/cm^2;

(optimal conditions for 95% colour removal was CD=4mA/cm^2, RT=10min, 150RPM)

Results and Discussion

EC Rotated Anode (batch reactor)

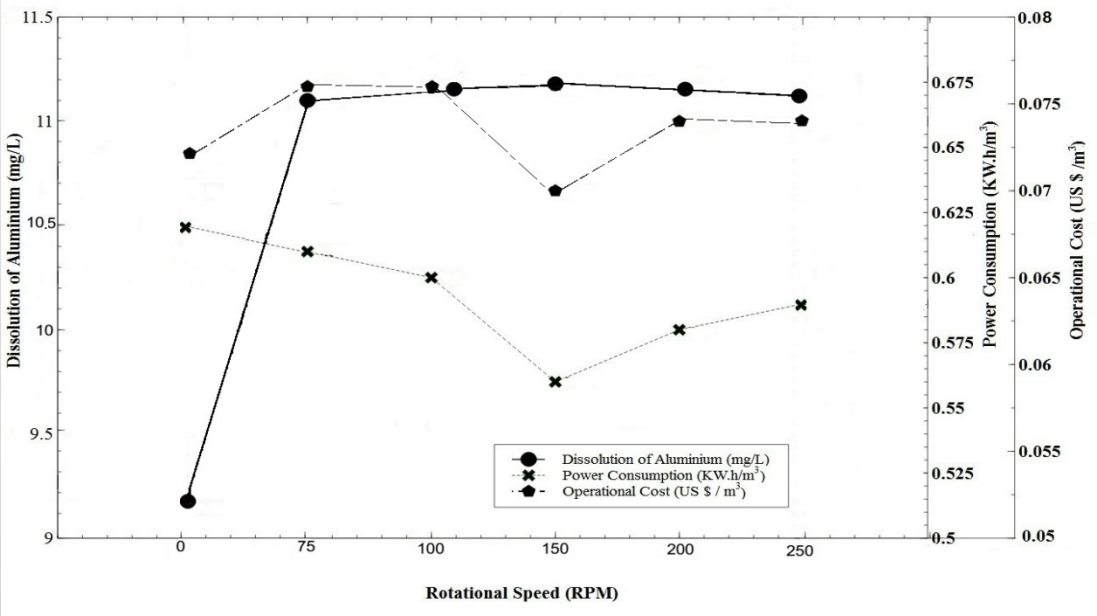
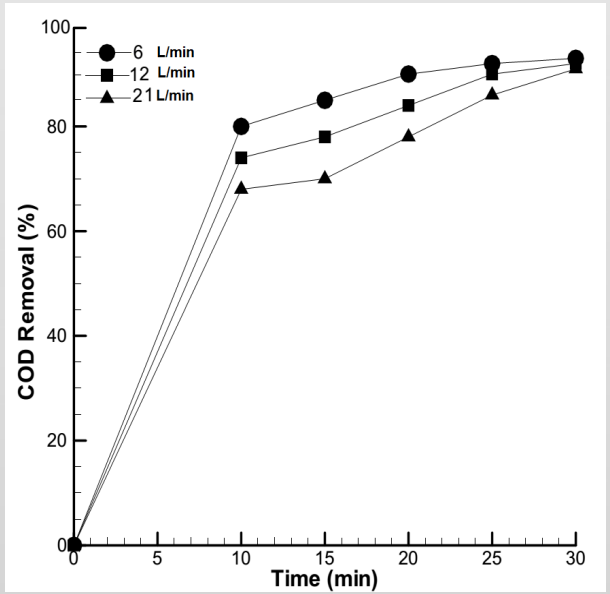


Fig. 2 (c). Effect of anode rotational speed on dissolution of aluminium, power consumption and operational cost at condtions (CD=4mA/cm^2, RT=10min).

- Maximum value of **aluminum dissolution** was **11.18 mg/L** at 150 rpm. This increase due to passivation of the anode reduced with increasing rotational speed.
- Main **voltage** decrease with increasing rotational speed (18.5, 17, 16.30, 15.10, 15.00 and 15.00 V for 0, 75, 100, 150, 200, and 250 rpm)
- Minimum overall **electrical energy consumption** was **0.56 kW.h/m^3** at 150 rpm due to the decreasing in main voltage. Minimum **operational cost** was **0.072 US$/m^3** at 150 rpm.

Results and Discussion

EC Rotated Anode (semi-continous flowrate, continous flowrate)



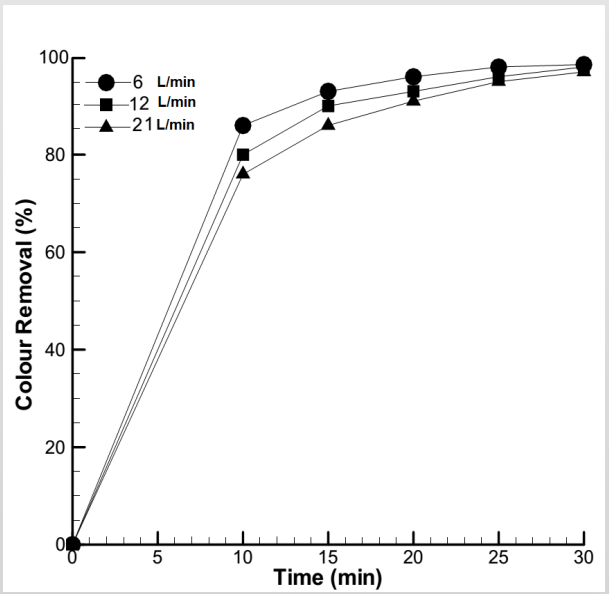


Fig. 3. Effect of recirculation flow rate at conditions (CD=4mA/cm^2, 150rpm) (a). COD removal (%) ; (b). Colour removal (%).

- At higher flow rate, lower COD and colour removal were detected at first 10 min
- the effectiveness of the COD and colour removal achieved were approximately comparable to all the recirculation flow rates reported at 30 min of the EC treatment.

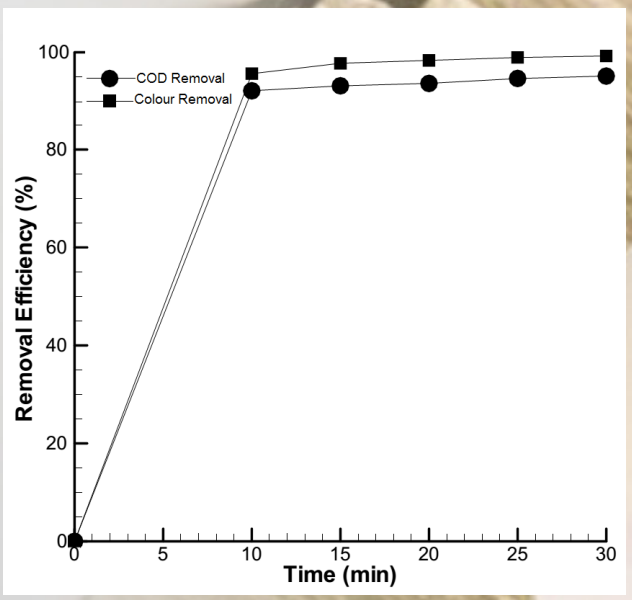


Fig.4. EC process at a continuous flow system with conditions (CD=4mA/cm^2, 150rpm, Q= 0.333 L/min and pH of wastewater =7).

- COD and colour removal were 91.5%, 95.5% at first 10 min of the continuous flow system in one pass.

Results and Discussion (Mechanism of treatment)

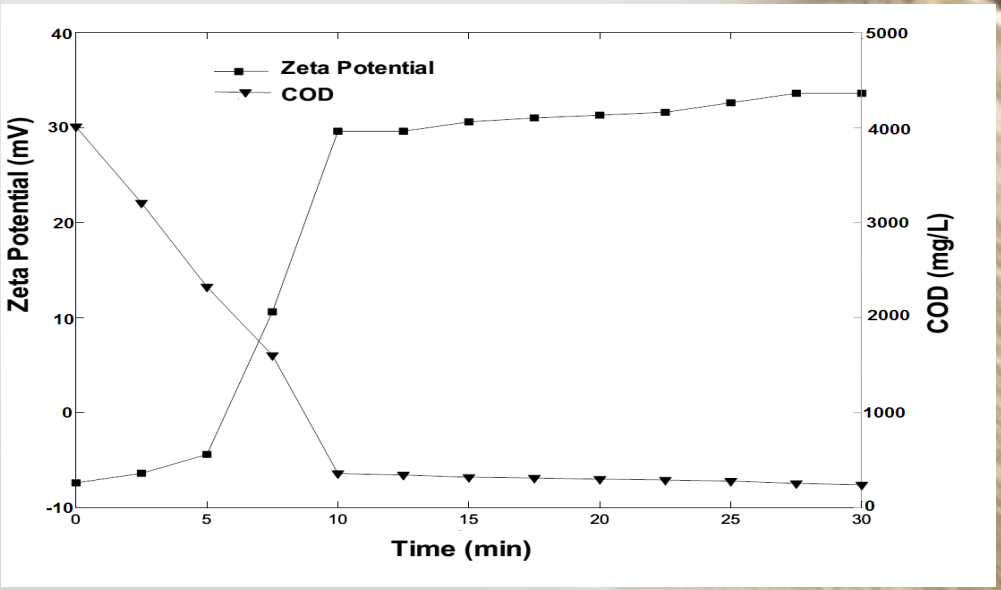
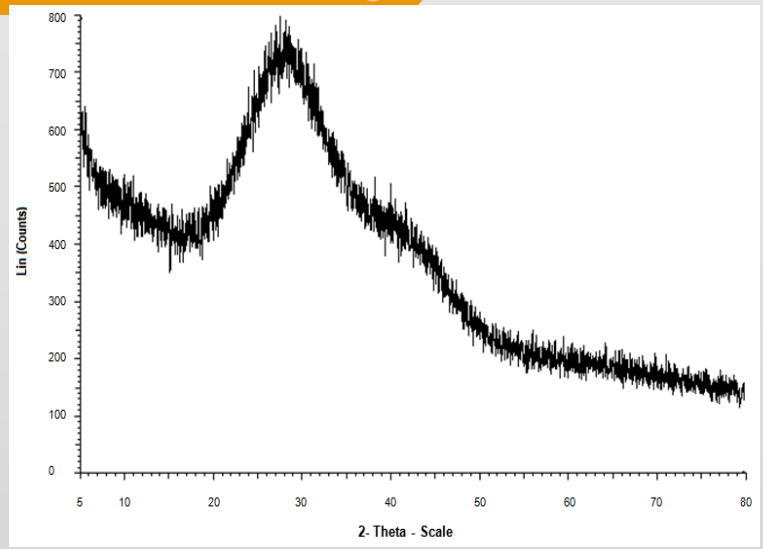


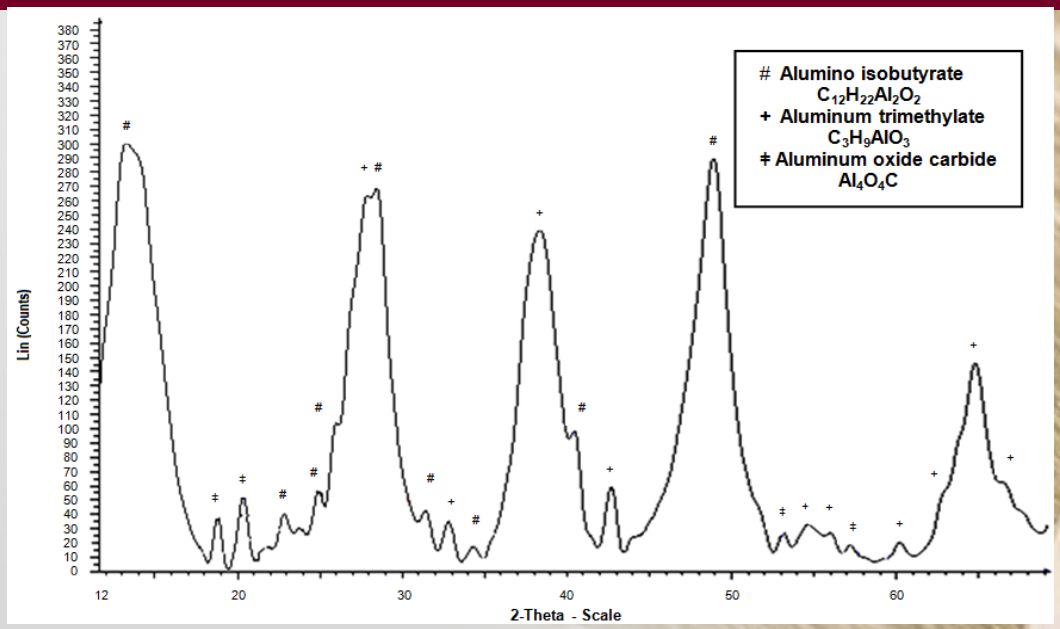
Fig. 5. Variation of zeta potential and residual COD concentration in solution with respect to electrolysis time at conditions (4mA/cm^2, 150 rpm and pH of wastewater =7).

- Change of zeta potential values with time show that chemical reaction occurred.
- Al dissolved at the anode increases, the residual COD concentration decreases and the zeta potential increases.
- As the system moved into more electrolysis time (>10 min), the rate of COD removal continued to decrease from 360 to 240 mg/L, while the zeta potential increased from 29 to 33 mV. It seems that after 10 min reaction time, the low increase in zeta potential indicates stabilization of the solution when COD removal is slow in the stable stage.
- The slow change of zeta potential reveals there was a drop in the reaction rate between aluminium hydroxyl and the pollutants, where the decreasing of COD concentration leads to reduce the reaction rate.

Results and Discussion (Mechanism of treatment)



(a)



(b)

Fig.6. XRD analysis of sludge product after EC process at conditions (4mA/cm^2, RT=10min): (a). 0 rpm; (b). 150 rpm;

- At **0 rpm**, aluminium coagulant showed a very broad spectrum and no peaks. The coagulant is amorphous.
- At the optimum rotational speed (**150 rpm**), the crystallization took place, which were identified as **new products,** namely **aluminum isobutyrate, aluminum trimethylate** and **aluminum oxide carbide**.
- According to these element products, the adsorption between the colour and $Al(OH)_3$ was **chemo-adsorption.**
- An oxidation reaction first took place between the colour and $Al(OH)_3$, leading to breakage of the **aromatics rings** of the colour to produce an **aliphatic substrate as shown in Fig. 7 .**
- **Aliphatic chemicals** are more **environmentally friendly** than aromatic sludge.

Results and Discussion (Mechanism of treatment)

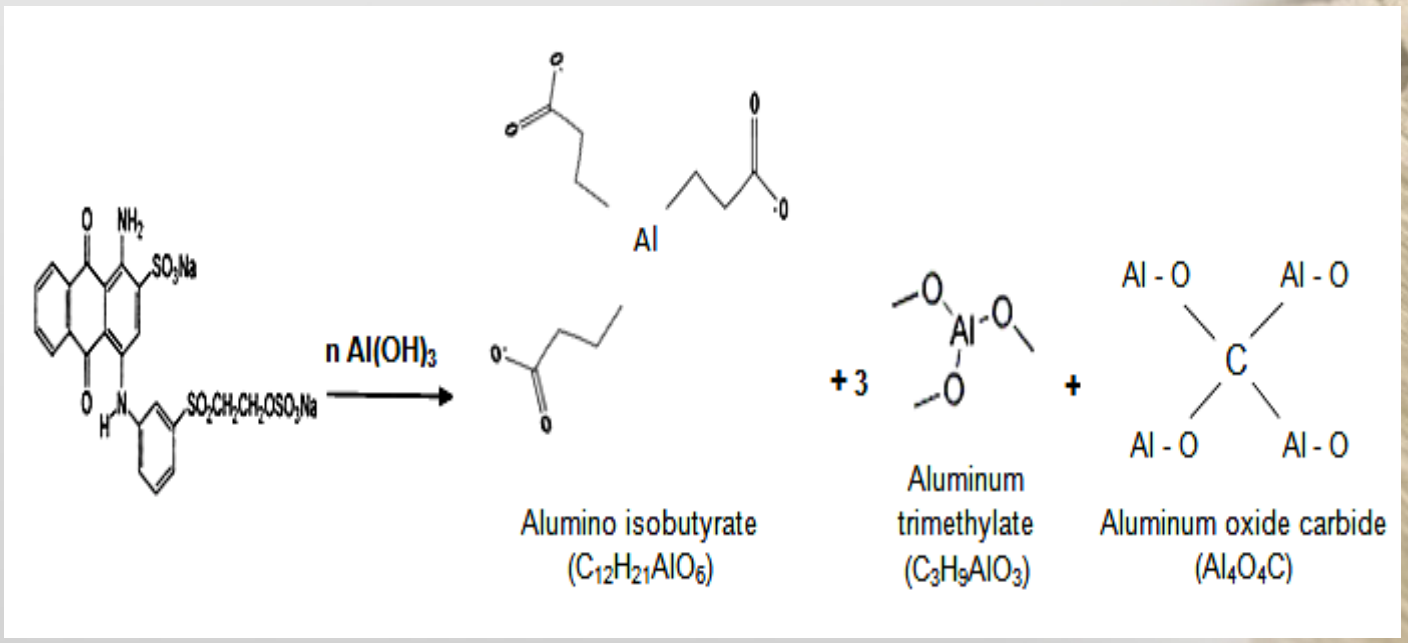


Fig.7 . The suggested equation of EC reaction at optimal conditions according to the XRD products

Results and Discussion (H2 production)

Table 2. Values of experimental, theoretical hydrogen yields and energy content with different rotational speed (rpm) at conditions (CD=4mA/cm^2, RT=10min).

Theoretical amount of H_2		Experimental amount of H_2				H_2 yield	Energy content
n_{H2} (mole)	V_{H2} (L)	Rotational speed (rpm)	n_{H2} (mole)	m_{H2} (g)	V_{H2} (L)	(experimental / theoretical)	$E_{H2}\times10^{-2}$ (KW.h/m^3)
0.0062	0.15	0	0.0045	0.009	0.110	0.73	346
		75	0.0053	0.0106	0.130	0.87	407
		100	0.0055	0.0110	0.135	0.90	422
		150	0.0061	0.0122	0.148	0.99	470
		200	0.0058	0.0116	0.140	0.93	446
		250	0.0052	0.0104	0.125	0.83	399

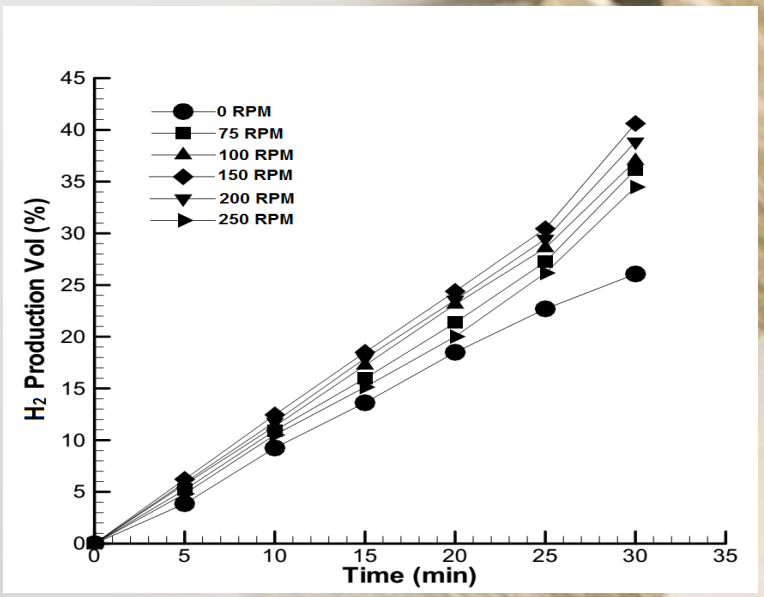


Fig. 8. Effect of anode rotational speed on H_2 production as a volume percent of gas with respect to reaction time at conditions (CD=4mA/cm^2, pH of wastewater =7).

- From Fig. 8, the maximum values of H_2 at 150 rpm (12.45% at 10 min and 35% at 30min).
- From table 2, the hydrogen yield was 0.73 at 0 rpm. That is, exactly 23% of H_2 theoretical amount was lost in solution. The hydrogen yield was enhanced for the rotational case to nearly 1 at 150 rpm. There was no significant difference in the rotation case.
- The maximum energy content (E_{H2}) was 0.047 kW.h/m^3 at 150 rpm, which can reduce the electrical energy consumption of the EC process by 9.4%

Results and Discussion

Table 3. Electrochemical impedance data extracted from the Nyquist plots at different rotational speed (rpm).

Rotational speed (rpm)	R_s(Ω)	R_{ct}(Ω)	C_{dl}(μF)	R_{ads} (Ω)	C_{ads}(μF)
0	63.30	96.98	0.128	1774	7.18
75	56.90	88.89	0.129	1531	7.36
100	59. 08	90.00	0.145	1369	7.13
150	40.54	41.65	0.412	1151	8.31

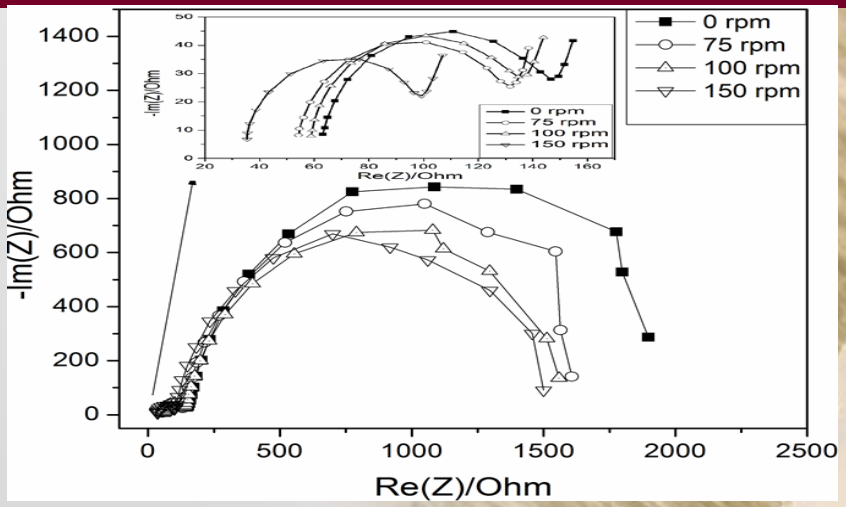


Fig. 9 Nyquist plots of the aluminium anode in an aqueous solution of textile wastewater at different electrode rotation speed and 25℃ temperature.

- From Fig. 9 and Table 3 that the values of solution resistance (R_s), passivation resistance (R_{ct}) and adsorption resistance (R_{ads}) decreased significantly with the increase in the rotation speed of the aluminium anode from 0 reaching a minimum at 150 rpm.
- Adsorption capacitance (C_{ads}) of electrode increases with increasing rotational speed.

CONCLUSION 1

1. A different design using EC rotated anode reactor was achieved,
2. Create turbulence by rotation of anode to solve problems of CC and EC by:
   - Sweep out the bubbles formed near the electrode surface,
   - The motion of the anode has a self-cleaning effect, reducing fouling ,
   - Increasing mass transfer efficiency due to high shear stress at the electrode surface ,
   - Fast interaction ionic hydroxide as coagulants with contaminants (Fast reaction),

CONCLUSION 2

- Solar Energy can be used to run the electrocoagulation system
- COD and color improved were obtained with a relatively low current density at initial 10 minutes with rotational speed of 150 rpm.
- The overall electrical energy consumption was 0.56 kW.h/m^3, which resulted in lower operational costs of 0.072 US$/$m^3$, which can be considered low compared with previous studies.
- EC process is efficient at continuous flow system.

CONCLUSION 3

- The zeta potential values with time indicate that the chemical interaction happened and showed that the optimum reaction time was 10 min.
- Producing sludge without any environmental hazards
- Enhancing the hydrogen production under the same current density a minimum operating area to working volume ratio.
- The hydrogen produced is trapped and can be used to run the system.
- The impedance results show a significant reduction in passivation of the anode and an increase in the adsorption when the anode is rotated. This explains the high removal rate at 150 rpm.

Activities

- **Intellectual Property**

Copyright for invention

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Attn: Dr C. Shreeshivadasan

By Email & Post

Dear Sir,

Patent Novelty Search for "Rotary Electro-Coagulator System"

1. We have conducted a patent search on the above matter to determine its patentability.

2. We enclose the related patent documents with their summaries.

2.1 US 2012186992 (A1)
Title: Electrocoagulation For Treating Liquids
Publication Date: 26 July 2012
Applicant(s): Berrak Abderrazak [CA]

A method, a system and a kit for removing colloid contaminants from a fluid by destabilization thereof with addition of kinetic energy thereto is provided, the method to overcome the energetic barrier preventing an efficient fluid-solid separation comprises injecting the colloidal fluid containing contaminants in an electrolytic system including an electrocoagulation module comprising an anode and a cathode, the anode and the cathode being adapted to be electrically connected to perform electrolysis of the fluid, providing an electric current, between the anode and the cathode, to form electro-coagulated contaminants flocs in the agitated fluid, separating the electro-coagulated flocs from the fluid, and extracting the fluid from the electrolytic system.

2.2 Separation and Purification Technology, Vol. 38, Issue 1, P. 11–41
Title: Electrochemical Technologies In Wastewater Treatment
Publication Date: 15 July 2004
Author(s): Guohua Chen

This paper reviews the development, design and applications of electrochemical technologies in water and wastewater treatment. Particular focus was given to electrodeposition, electrocoagulation (EC), electroflotation (EF) and electrooxidation. Over 300 related publications were reviewed with 221 cited or analyzed. Electrodeposition is effective in recover heavy metals from wastewater streams. It is considered as an established technology with possible further development in the improvement of space-time yield. EC has been in use for water production or wastewater treatment. It is finding more applications using either aluminum, iron or the hybrid Al/Fe electrodes. The separation of the flocculated sludge from the treated water can be accomplished by using EF. The EF technology is effective in removing colloidal particles, oil & grease, as well as

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Publication

- Naje, A. S., Chelliapan, S., Zakaria, Z., & Abbas, S. A. 2015. Enhancement of an Electrocoagulation Process for the Treatment of Textile Wastewater under Combined Electrical Connections Using Titanium Plates. *Int. J. Electrochem. Sci*, *10*, 4495-4512.
- Ahmed Samir Naje, Shreeshivadasan Chelliapan, Zuriati Zakaria, Saad A. Abbas. 2015. Treatment Performance of Textile Wastewater Using Electrocoagulation (EC) Process under Combined Electrical Connection of Electrodes. *International Journal of Electrochemical Science.* ISSN: 1452-3981.

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THANK YOU

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CERTIFICATE OF PARTICIPATION

Awarded to AHMED SAMIR NAJE

Has participated in
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Organized by
INNOVATION & COMMERCIALISATION CENTRE (ICC), UTM

Date : **23 – 24 MARCH 2014**
Venue : **S01, UTMCSI**

PROF. IR. DR. AZHAR BIN DATO' ABDUL AZIZ
Director
Innovation and Commercialisation Centre
Universiti Teknologi Malaysia

THANK YOU