



Separation Science and Engineering

Using bipolar membrane electrodialysis to synthesize di-quaternary ammonium hydroxide and optimization design by response surface methodology

Jiangnan Shen^{*}, Zhendong Hou, Congjie Gao

Center for Membrane Separation and Water Science & Technology, Zhejiang University of Technology, Hangzhou 310014, China

ARTICLE INFO

Article history:

Received 19 December 2016

Received in revised form 21 February 2017

Accepted 4 March 2017

Available online 27 March 2017

Keywords:

Di-quaternary ammonium hydroxide

Bipolar membrane electrodialysis

Response surface methodology

N,N-hexamethylenebis(trimethyl ammonium hydroxide)

ABSTRACT

Bipolar membrane electrodialysis (BMED) has already been described for the preparation of quaternary ammonium hydroxide. However, compared to quaternary ammonium hydroxide, di-quaternary ammonium hydroxide has raised great interest due to its high thermal stability and good oriented performance. In order to synthesize *N,N*-hexamethylenebis(trimethyl ammonium hydroxide) (HM(OH)₂) by EDBM, experiments designed by response surface methodology were carried out on the basis of single-factor experiments. The factors include current density, feed concentration and flow ratio of each compartment (feed compartment: base compartment: acid compartment: buffer compartment). The relationship between current efficiency and the above-mentioned three factors was quantitatively described by a multivariate regression model. According to the results, the feed concentration was the most significant factor and the optimum conditions were as follows: the current efficiency was up to 76.2% (the hydroxide conversion was over 98.6%), with a current density of 13.15 mA·cm⁻², a feed concentration of 0.27 mol·L⁻¹ and a flow ratio of 20 L·h⁻¹:26 L·h⁻¹:20 L·h⁻¹:20 L·h⁻¹ for feed compartment, base compartment, acid compartment, and intermediate compartment, respectively. This study demonstrates the optimized parameters of manufacturing HM(OH)₂ by direct splitting its halide for industrial application.

© 2017 The Chemical Industry and Engineering Society of China, and Chemical Industry Press. All rights reserved.

1. Introduction

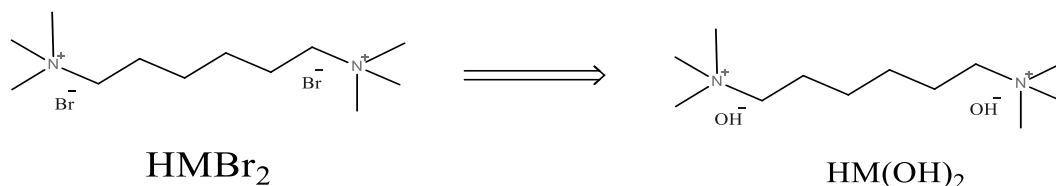
Quaternary ammonium hydroxide [1] is an organic alkali compound, which is more alkaline than sodium hydroxide or potassium hydroxide. Quaternary ammonium hydroxide has industrial applications as catalyst for synthesis of organic silicon, as phase transfer catalyst and as a template agent for the synthesis of zeolites or molecular sieves [2,3]. The synthesis of quaternary ammonium hydroxide was mainly restricted to the traditional quaternary ammonium hydroxide, such as tetra-methyl ammonium hydroxide [4,5], and tetrapropyl ammonium hydroxide. Research on di-quaternary ammonium hydroxide is scarce, in spite of its potential benefits. Compared with the commonly used quaternary ammonium hydroxide, di-quaternary ammonium hydroxide has many advantages: rich raw material resources, low cost, easy preparation process and excellent thermal stability [6]. Di-quaternary ammonium hydroxide can be also used as an alkali catalyst with advantages of good stability, high target product selectivity, and as template agent [7]. Traditionally, the preparation of quaternary ammonium hydroxide is mainly based on the ion exchange method

and the electrolytic method [8]. These methods have some disadvantages, like environmental pollution, equipment corrosion, and low product purity [9].

Bipolar membranes can be used for water splitting, causing a different pH on each side of a membrane [10]. Without introducing new components, it allows to convert salts in aqueous solution into acid and base [11,12]. According to a theoretical calculation [13], the required electrical potential is only 0.83 V, which is less than the potential of 2.1 V required for the electrolysis of water for the preparation of 1 mol·L⁻¹ acid and alkali. Therefore, the use of bipolar membranes to prepare acids and bases is more economic. With the development of BMED in research and real applications, it has been widely applied in desalination, preparation of acid and alkali in chemical industry [14], purification of proteins, amino acids [15, 16], and industrial wastewater [17]. Bipolar membrane electrodialysis has been proposed as an effective alkali production technique without the generation of oxygen and hydrogen gas as byproducts. Therefore, the synthesis of di-quaternary ammonium hydroxide by BMED is of great interest.

This study explores the feasibility of the synthesis of HM(OH)₂ by BMED from hexamethonium bromide (HMBBr₂) solution. An optimization was carried out by the response surface methodology [18], based on single factor experiments. The preparation route is shown in Fig. 1.

^{*} Corresponding author.E-mail address: shenj@zjut.edu.cn (J.N. Shen).

Fig. 1. Schematic diagram of HM(OH)_2 synthesis route.

2. Experimental

2.1. Materials

Sulfate acid (analytical grade) was provided by Nanjing Chemical Reagent Co., LTD, Nanjing, China and hexamethonium bromide (analytical grade) was provided by Hangzhou Greenda Chemical Co., LTD, Hangzhou, China. Deionized water was used throughout all experiments.

2.2. Experimental set-up

As shown in Fig. 2, a four-compartment EDBM stack of BP- C_1 -A- C_2 configuration (BP, bipolar membrane, FBM, FuMA-Tech GmbH; C_1 , cation-exchange membrane, CMX, Tokuyama Co.; A, anion-exchange membrane, AMX, Tokuyama Co.; C_2 , cation-exchange membrane, JCM-II-05, TMb-Tech) with five repeating units was used as the experimental set-up. The effective membrane area was 189 cm^2 . The BMED stack was composed of two electrode compartments between which two uniform cells were inserted. Seleminon® CMTE membranes (Asahi Glass Co., LTD., Japan) were used as the electrode membrane. The stack included four compartments, i.e., a buffer compartment (between BP and C_1), an acid compartment (between C_1 and A), the feed compartment (between A and C_2) and an alkaline compartment (between C_2 and BP). Compartments were isolated by a 0.70 mm-thick polypropylene gaskets and spacers. Each compartment was connected to a separate external 1000 cm^3 beaker, allowing for continuous circulation by five magnetic driving pumps. The acid and base compartments were filled with pure water, respectively. Electrodes were made of titanium coated with ruthenium. The two electrode compartments were rinsed with a $0.3 \text{ mol} \cdot \text{L}^{-1} \text{ H}_2\text{SO}_4$ solution to prevent the introduction of alkali metal ions. A $0.1 \text{ mol} \cdot \text{L}^{-1} \text{ H}_2\text{SO}_4$ solution was added to the intermediate compartment, and the solution was replaced before each experiment. When

Table 1

Main properties of the membranes used in BMED

Membrane type	Thickness/ μm	Ion exchange capacity/ $\text{mmol} \cdot \text{g}^{-1}$	Area resistance/ $\Omega \cdot \text{cm}^2$	Efficiency/%
FBM	200		<3	98
CMX	220–260	1.5–1.8	2.0–3.5	
AMX	120–180	1.4–1.7	2.0–3.5	
JCM -II-05	160–230	2.0–2.9	1–3	

a direct current passes through the stack, di-quaternary ammonium ions (HM^{2+}) are transported across the cation-exchange membrane to the corresponding compartment, where they combine with OH^- produced by the bipolar membranes to form HM(OH)_2 (Table 1).

2.3. Analytical methods

The concentrations of HM(OH)_2 and HBr produced by BMED were determined by titration with a standard sulfuric acid and sodium hydroxide solution using methyl orange and phenolphthalein as indicators, respectively. The content of Br^- in the HM(OH)_2 solution was determined by the Mohr method. The voltage and current across the stack were recorded directly by a CV/CC regulated power supply (WYL 1703 $\times$ 2, Hangzhou Siling Electrical Instrument Ltd., China). The conductivity and pH of the HMBR_2 solution were monitored during the entire process by a conductivity meter and a pH-meter, respectively.

2.4. Data calculation

The performance of BMED for di-quaternary ammonium hydroxide production was evaluated in terms of current efficiency (η), energy consumption (E) and conversion rate (X) of HM(OH)_2 . The current

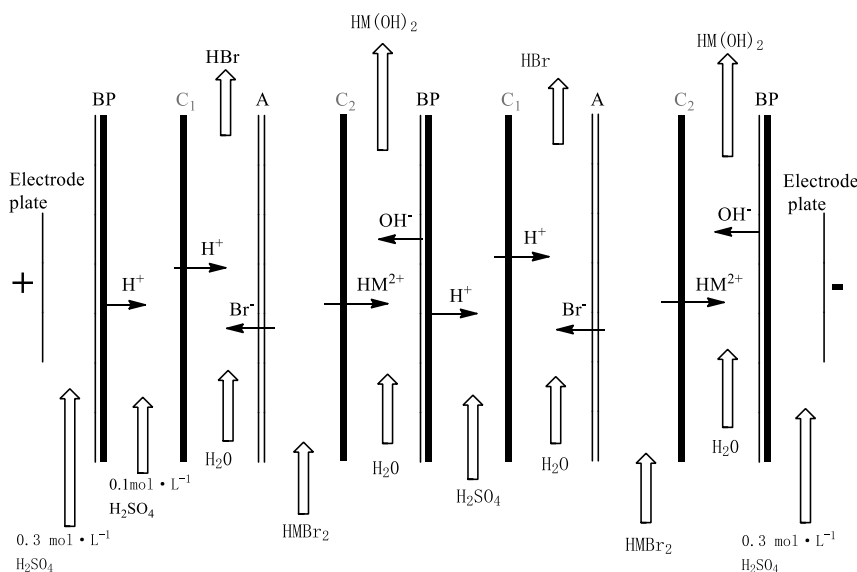


Fig. 2. Bipolar membrane electro dialysis scheme to obtain acid and base. (BP, bipolar membrane, FBM; C_1 , cationic membrane, CMX; A, anionic membrane, AMX; C_2 , cationic membrane, JCM-II-05).

efficiency was calculated by Eq. (1).

$$\eta = \frac{z(C_t V_t - C_0 V_0) F}{N I t} \times 100\% \quad (1)$$

where z is the ion valence ($z = 2$). C_t and C_0 ($\text{mol} \cdot \text{L}^{-1}$) are the $\text{HM}(\text{OH})_2$ concentrations at time t and initially, respectively, V_t and V_0 (L) are the volume of $\text{HM}(\text{OH})_2$ solution at time t and initially, respectively, F is the Faraday's constant ($F = 96,500 \text{ C} \cdot \text{mol}^{-1}$), N is the number of repeating units of the stack, I is the current used in the stack (A) and t is the operation time (s).

The energy consumption ($\text{kW} \cdot \text{h} \cdot \text{kg}^{-1}$) was calculated by the following Eq. (2).

$$E = \int_0^t \frac{U I}{(C_t V_t - C_0 V_0) M} dt \quad (2)$$

where U is the voltage drop (V) across the membrane stack at time t , M is the molar mass ($\text{g} \cdot \text{mol}^{-1}$) of the acid (or base).

The conversion rate (X) of $\text{HM}(\text{OH})_2$ was calculated with Eq. (3).

$$X = \frac{(C_t V_t - C_0 V_0) M}{m} \quad (3)$$

where m (g) is the initial weight of HM^{2+} ion in the feed compartment.

2.5. Response surface methodology (RSM) design of experiments

On the basis of single-factor experiments, RSM design experiments were carried out [19,20]. A three-level-three factor box-betoken design (BBD) with 15 runs (3 repetitive runs and 12 factorial runs) was carried out using Design-Expert 8.0.5 Trial software [21]. Current density (X_1), feed concentration (X_2) and flow ratio (X_3 , feed compartment: base compartment: acid compartment: buffer compartment) were selected as independent variables with three levels of -1 , 0 , $+1$. Table 2 shows the variable factors with the coded and actual values.

Table 2
Variables and experimental design levels for response surface

Factor	Code	None code	Code level		
			-1	0	1
Current density/ $\text{mA} \cdot \text{cm}^{-2}$	X_1	A	10	13	16
Feed concentration/ $\text{mol} \cdot \text{L}^{-1}$	X_2	B	0.2	0.3	0.4
Flow ratio	X_3	C	1:0.5:1:1	1:1:1:1	1:2:1:1

Note: the flow rate of 1:1:1:1 was equal to the actual values of $20 \text{ L} \cdot \text{h}^{-1}$: $20 \text{ L} \cdot \text{h}^{-1}$: $20 \text{ L} \cdot \text{h}^{-1}$: $20 \text{ L} \cdot \text{h}^{-1}$.

Experiments data from BBD were represented using a second order polynomial model as follows [17]:

$$Y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \beta_{ij} X_i X_j$$

where Y is the estimated response variable; β_0 is the constant coefficient; β_i , β_{ii} and β_{ij} are the linear, quadratic and interaction constant coefficients, respectively; X_i ($i = 1, 2, 3$) and X_j ($j = 1, 2, 3$) represent the coded independent variables. To test the significance of the obtained model, analysis of variance (ANOVA) was used. To analyze the goodness-of-fit of the model, the regression coefficients (R^2), lack-of-fit, and adjusted and predicted R^2 were also evaluated.

3. Results and Discussion

3.1. Single-factor experiments

3.1.1. Influence of current density on BMED

Fig. 3 indicates the results of the BMED experiments at different current densities. The voltage drop and the conductivity curve of HMBR_2

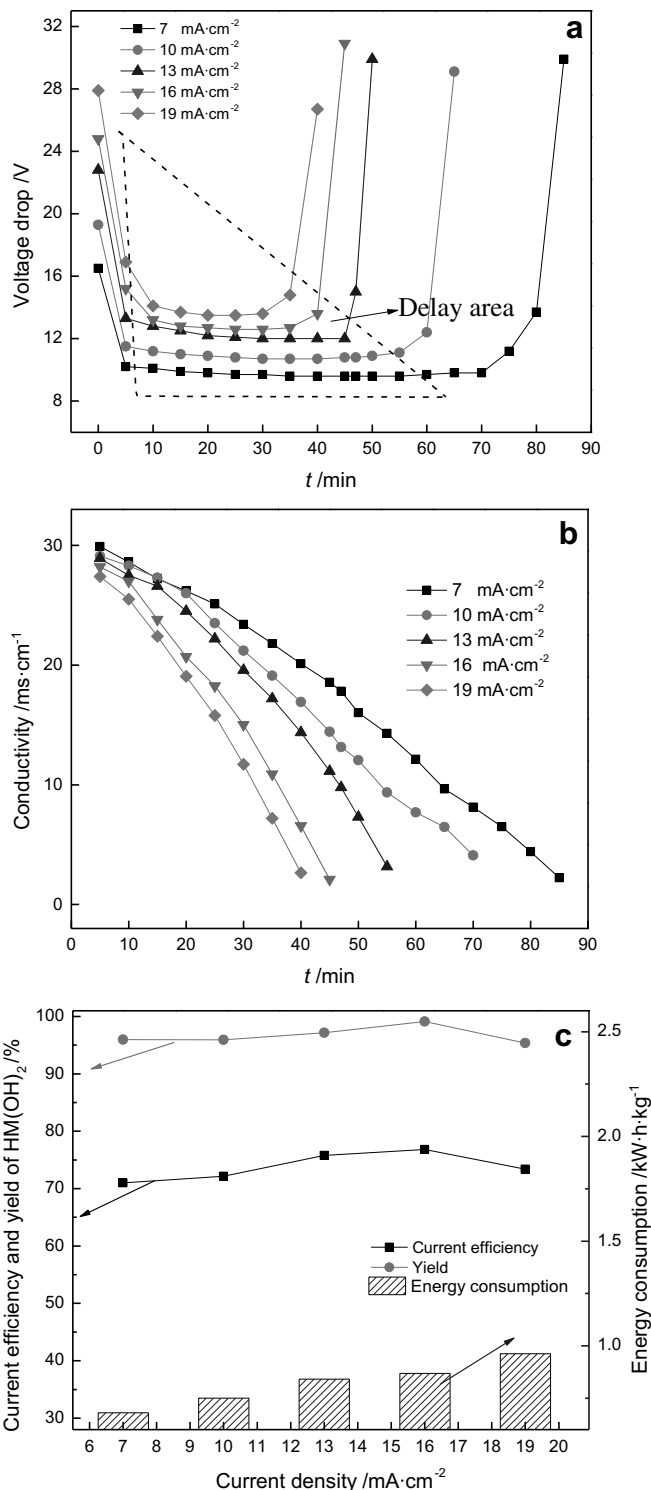


Fig. 3. The influence of the current density on the BMED process: a, the voltage drop across the stack as a function of time; b, the conductivity of the HMBR_2 solution as a function of time; c, the current efficiency and conversion of $\text{HM}(\text{OH})_2$, the column chart of energy consumption at different current densities.

solution are shown in Fig. 3a and b, respectively. Fig. 3a shows the time dependency of the applied voltage across the BMED stack at various current densities, i.e., 7, 10, 13, 16 and 19 $\text{mA} \cdot \text{cm}^{-2}$. During the electrodiolysis process, it was found that the voltage drop across the BMED stack has a U-shape, as shown in Fig. 3a. The applied voltage declined sharply at the start of the experiments, which was mainly related to the increase of the conductivity of the $\text{HBr}/\text{HM}(\text{OH})_2$ solution in the acid/base compartment,

reducing the electrical resistance of the BMED stack. Subsequently, the BMED stack reached a steady state in the intermediate phase, presenting a slight increase in the voltage. This suggests that the electrical resistance of the stack remains constant. The increase of the resistance of the salt compartment, resulting from the exhaustion of HMBR_2 in the solution, can be offset by the decrease of the electrical resistance of the acid and base compartments due to the increase of the HBr and HM(OH)_2 concentrations induced by the transfer of Br^- and HM^{2+} ions from the feed solution. However, the concentration of HMBR_2 in the feed decreases with time, which increases the electrical resistance of the stack, and then the voltage drop across the stack increases accordingly at the end of the experiment. In the BMED stack, at high current densities, water dissociation and transmission of HM^{2+} and Br^- ions was accelerated, so that a large amount of HM(OH)_2 and HBr was produced, which reduced the electrical resistance. Therefore, the operation time is shortened due to the fast exhaustion of HMBR_2 in the feed solution at higher current densities. It can be observed from Fig. 3b that the conductivity of the HMBR_2 solution continuously decreased. A possible explanation could be that under the electric field, the conductive ions HM^{2+} and Br^- continuously migrated across the corresponding ion exchange membrane into the acid/base compartment.

As shown in Fig. 3c, the current efficiency increased with the current density at the initial stage and then decreases again. The increase of the current efficiency is due to the fact that the transport rate of HM^{2+} was enhanced, while at the same time, the transport of the counter-ions was inhibited. At a higher current density, the membrane surface was prone to concentration polarization. As a result, the HM^{2+} migration was inhibited, and the migration rate of H^+ generated by concentration polarization in higher current density increased. Thus, the current efficiency decreases, and the energy consumption increases with the current density. This can be ascribed to the fact that a larger part of the total electrical energy was used to overcome the electrical resistance at higher current densities. The HM(OH)_2 conversion increases linearly with the increase of the current density in the range of $10\text{--}16\text{ mA}\cdot\text{cm}^{-2}$, but then decreases in the range of $16\text{--}19\text{ mA}\cdot\text{cm}^{-2}$. This is caused by a competitive migration in the feed compartment (competition between HM^{2+} and H^+).

3.1.2. Influence of HMBR_2 concentration

Aqueous solutions with various HMBR_2 concentrations were used as feed solutions (Fig. 4). It can be seen that the current efficiency increases in the concentration range of $0.2\text{--}0.3\text{ mol}\cdot\text{L}^{-1}$, and decreases in the range of $0.3\text{--}0.6\text{ mol}\cdot\text{L}^{-1}$. The increase of the current efficiency can be explained by the following reasons: when the concentration of HMBR_2 increases, water electrolysis is impeded on the membrane surface. Thus, when the

quantity of HM^{2+} in the solutions increases, the migration ability of HM^{2+} increases and restrains the migration of H^+ , so that the energy consumption declines. When the feed concentration exceeded $0.3\text{ mol}\cdot\text{L}^{-1}$, the thermal effect is enhanced because a large part of the electric energy is used to produce heat rather than effective ion migration, so that the current efficiency declines and the energy consumption increases. Therefore, as the HMBR_2 concentration increases, a longer time is needed to transport HM^{2+} and Br^- from the salt compartment. The HM(OH)_2 conversion was over 90.0%, and can reach 97.8%, as shown in Fig. 4.

3.2. Response surface Box–Behnken analysis

3.2.1. Model fitting

Three-level factors (current density, feed concentration and flow ratio) and the effect of their interactions on the current efficiency were estimated through the Box–Behnken center united design of RSM. Table 3 shows the experimental and predicted current efficiency. By applying analysis of variance (ANOVA) to the regression model, the predicted response Y can be expressed by a second order polynomial model in terms of coded factors, as:

$$Y = 74.92 - 0.18X_1 - 6.43X_2 + 0.80X_3 - 0.082X_1X_2 - 0.79X_1X_3 + 0.11X_2X_3 - 5.11X_1^2 - 8.04X_2^2 - 2.93X_3^2$$

Table 3
Experimental design and results of RSM

Run order	Factors			Current efficiency/%	
	A	B	C	Actual	Predicted
1	0	−1	1	69.98	71.08
2	1	0	−1	66.42	66.69
3	1	1	0	54.26	55.08
4	0	1	−1	57.71	56.62
5	0	−1	−1	69.68	69.69
6	0	1	1	58.44	58.43
7	1	−1	0	68.39	68.11
8	−1	0	−1	64.65	65.46
9	0	0	0	74.24	74.92
10	−1	1	0	55.32	55.60
11	0	0	0	75.49	74.92
12	1	0	1	67.52	66.71
13	−1	−1	0	69.12	68.30
14	−1	0	1	68.92	68.65
15	0	0	0	75.04	74.92

The significance of the equation for the current efficiency was verified by the F -test, as shown in Table 4. The F -value was 59.96 and the probability values (0.0001) were sufficient to demonstrate that

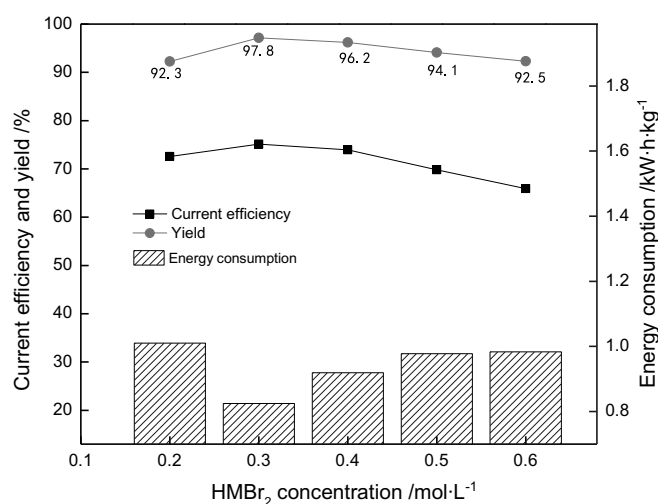


Fig. 4. Synthesis of HM(OH)_2 by EDBM: energy consumption, current efficiency and conversion of HM(OH)_2 at different concentrations of HMBR_2 .

Table 4
Analysis of variance (ANOVA) for the regressive model

Source	Sum of squares	df	Mean square	F value	p-Value Prob > F	Significant
X_1	0.25	1	0.25	0.20	0.6705	
X_2	330.76	1	330.76	267.55	<0.0001	**
X_3	5.12	1	5.12	4.14	0.0975	
X_1^2	96.52	1	96.52	78.08	0.0003	*
X_1X_2	0.027	1	0.027	0.022	0.8878	
X_1X_3	2.51	1	2.51	2.03	0.2133	
X_2^2	238.55	1	238.55	192.96	<0.0001	**
X_2X_3	0.046	1	0.046	0.037	0.8543	
X_3^2	31.76	1	31.76	25.69	0.0039	*
Model	667.14	9	74.13	59.96	0.0001	**
Residual	6.18	5	1.24			
Lack of fit	5.38	3	1.79	4.47	0.1881	
Pure	0.80	2	0.40			
Corr total	673.32	14				

R-squared (R^2) = 0.9908 Adj R-square = 0.9743

the model was well adapted to the response. The ANOVA results show that the current efficiency is significantly affected by all linear variables. The feed concentration (X_2) was the most significant factor to influence the model, with a p -value for X_2 below 0.0001.

Fitting the experimental results to a quadratic polynomial model (see Table 4), ANOVA indicates a regression coefficient (R^2) of 0.9908 with no significant lack of fit ($p = 0.1881$). These values suggest that the calculated model was able to explain 99.08% of the results. Regarding the models of the current efficiency, the adjusted R^2 value was 0.9743 and the predicted R^2 was 0.8695, which is in good agreement with the predicted R^2 values. Fig. 5 and Table 3 confirm a satisfactory correlation between the experimental and predicted values of the current efficiency. The points cluster along the diagonal line, which indicates that the model developed in this study was reasonably accurate.

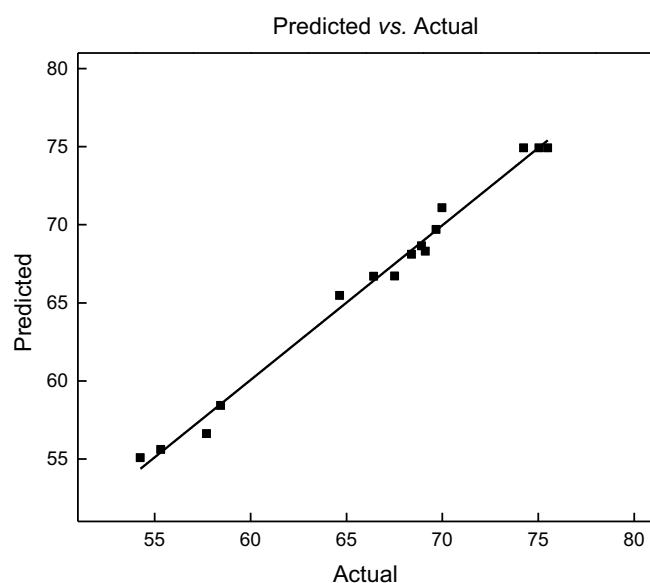


Fig. 5. Comparison between actual values and predicted values.

3.2.2. Effect of variables and optimization

The 3D response surface plots were obtained by the software, and are shown in Fig. 6. The effect of the independent variables and their mutual influence on the current efficiency can be seen. According to the p -value and the 3D response curves, the effect of the feed concentration on the current efficiency was more significant than that of the current density and the flow ratio. When two variables were fixed, the trends of the current efficiency with a single variable showed similar trends to those obtained in the single-factors optimization.

The response surface plots show the current efficiency for each pair of interactive variables from current density, feed concentration, and flow ratio (see Fig. 6). The optimum values of the variables, which maximize the responses (i.e., the current efficiency of BMED process to produce $\text{HM}(\text{OH})_2$), were found as: current density $13.15 \text{ mA} \cdot \text{cm}^{-2}$, feed concentration $0.27 \text{ mol} \cdot \text{L}^{-1}$ and flow ratio $20 \text{ L} \cdot \text{h}^{-1} : 26 \text{ L} \cdot \text{h}^{-1} : 20 \text{ L} \cdot \text{h}^{-1} : 20 \text{ L} \cdot \text{h}^{-1}$ (1:1.3:1:1). With the three parallel experiments under optimized conditions, the current efficiencies were 76.2%, 75.3% and 75.9%; the $\text{HM}(\text{OH})_2$ conversion were over 98.2%, 98.6% and 97.9%, respectively.

3.2.3. Process economics

The process cost was estimated on the basis of the above experimental equipment under the conditions shown in Table 5. The total cost of BMED comprises the energy consumption in electrodialytic water

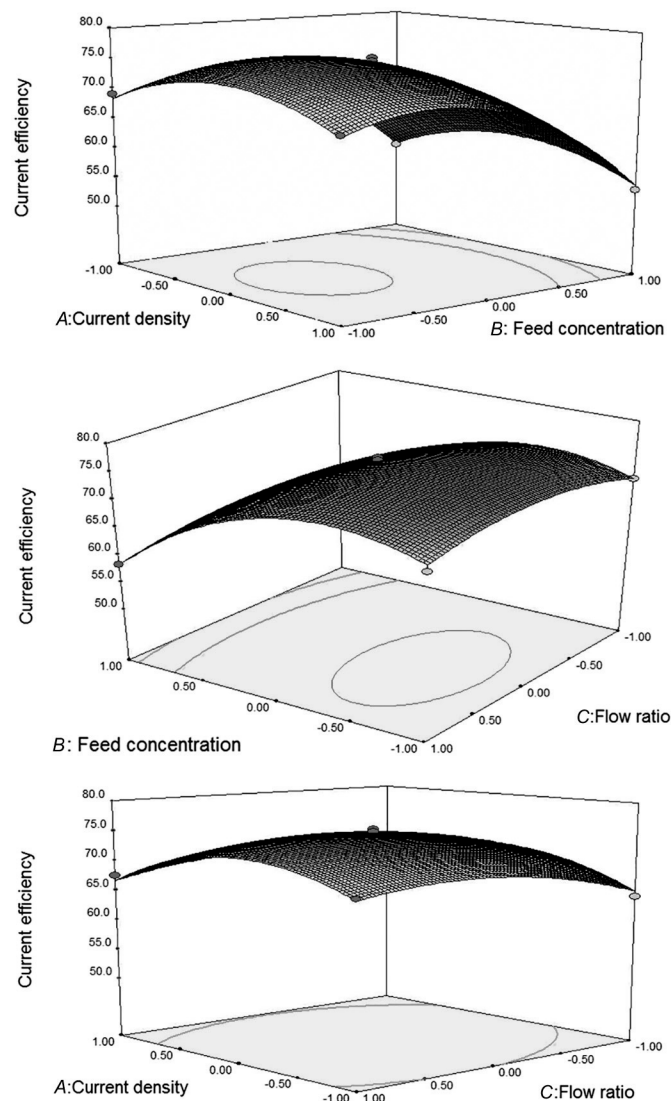


Fig. 6. Response surface plots showing the effect of the current density (A), feed concentration (B), and flow ratio (C) on the current efficiency.

dissociation and the investment cost. The total investment cost includes the stack cost and peripheral equipment cost. The stack cost is proportional to the membrane cost and is estimated to be 1.5 times the total membrane cost. The peripheral equipment cost (pumps, monitoring, control panels, etc.) is estimated at 1.5 times the stack cost. As shown in Table 5, it can be found that the process cost is $364 \text{ USD} \cdot \text{t}^{-1}$. The process cost of BMED is higher than that of the conventional synthesis method; the main reason is the high membrane price, especially for the bipolar membranes. However, the BMED technology can provide products of higher value and higher quality; in addition, the process is environmentally friendly, and relatively pure byproducts can be obtained, such as HBr , which can account for the cost disadvantage.

4. Conclusions

In this study, the synthesis of $\text{HM}(\text{OH})_2$ with BMED was studied. On the basis of single-factor experiments for the response surface design, RSM was used to optimize the BMED process to produce $\text{HM}(\text{OH})_2$ from HMBR_2 solution. The maximum current efficiency was 76.2% at a current density of $13.15 \text{ mA} \cdot \text{cm}^{-2}$, a feed concentration of $0.27 \text{ mol} \cdot \text{L}^{-1}$ and a flow ratio of $20 \text{ L} \cdot \text{h}^{-1} : 26 \text{ L} \cdot \text{h}^{-1} : 20 \text{ L} \cdot \text{h}^{-1} : 20 \text{ L} \cdot \text{h}^{-1}$ for the feed compartment, base compartment, acid

Table 5

Operation condition and its corresponding estimation of the process cost in experimental equipment

Operation conditions	Quantity	Remarks
HMBR ₂ concentration/mol·L ⁻¹	0.27	
Current density/mA·cm ⁻²	13.15	
Flow ratio	20 L·h ⁻¹ :26 L·h ⁻¹ :20 L·h ⁻¹ :20 L·h ⁻¹	
Operation time/min	50	
Energy consumption/kW·h·kg ⁻¹	0.86	
Electricity charge/USD·(kW·h) ⁻¹ ·kg ⁻¹	0.09	
Total investment cost/USD	622	
Membrane area/cm ⁻²	189	
Membrane cost/USD	166	Bipolar membrane (1350USD·m ⁻²) and ion-exchange membrane(135USD·m ⁻²)
Stack cost/USD	249	1.5 times membrane cost
Peripheral equipment cost/USD	373	1.5 times stack cost
Amortization/USD·a ⁻¹	207	3 years
Interest/USD·a ⁻¹	48	Interest rate, 8%
Maintenance/USD·a ⁻¹	62	10% of total investment cost
Total investment cost /USD·a ⁻¹	317	
Process cost/USD·t ⁻¹	364	

compartment, and intermediate compartment, respectively. The regression model predicts the current efficiency by BMED; the feed concentration was the most significant factor to synthesize di-quaternary ammonium hydroxide. The experiments indicate that the proposed process is promising for manufacturing HM(OH)₂ by direct splitting its halide, and that this method can be applied in the industry.

References

- [1] M. Kumar, B.P. Tripathi, A. Saxena, *et al.*, Electrochemical membrane reactor: Synthesis of quaternary ammonium hydroxide from its halide by in situ ion substitution, *Electrochim. Acta* 54 (5) (2009) 1630–1637.
- [2] T.H. Goswami, B. Nandan, S. Alam, *et al.*, A selective reaction of polyhydroxy fullerene with cycloaliphatic epoxy resin in designing ether connected epoxy star utilizing fullerene as a molecular core, *Polymer* 44 (11) (2003) 3209–3214.
- [3] I. Zubel, M. Kramkowska, K. Rola, Silicon anisotropic etching in TMAH solutions containing alcohol and surfactant additives, *Sensors Actuators A Phys.* 178 (2012) 126–135.
- [4] K.-H. Jun, B.-J. Kim, J.-S. Kim, Effect of additives on the anisotropic etching of silicon by using a TMAH based solution, *Electron. Mater. Lett.* 11 (5) (2015) 871–880.
- [5] J. Sun, D.R. Macfarlane, M. Forsyth, Novel alkaline polymer electrolytes based on tetramethyl ammonium hydroxide, *Electrochim. Acta* 48 (14) (2003) 1971–1976.
- [6] D.J. Kim, C.H. Park, S.Y. Nam, Characterization of a soluble poly(ether ether ketone) anion exchange membrane for fuel cell application, *Int. J. Hydrog. Energy* 41 (18) (2016) 7649–7658.
- [7] B. Marler, P. Daniels, J. Sañé i Muné, Synthesis and structure of RUB-35, a disordered material of the EUO-NES-NON family, *Microporous Mesoporous Mater.* 64 (1–3) (2003) 185–201.
- [8] T. Geng, X.-G. Teng, Q.-X. Li, *et al.*, Synthesis and properties of long-chain quaternary ammonium hydroxides, *Chin. Chem. Lett.* 24 (6) (2013) 494–496.
- [9] J.N. Shen, J. Yu, J. Huang, *et al.*, Preparation of highly pure tetrapropyl ammonium hydroxide using continuous bipolar membrane electrodialysis, *Chem. Eng. J.* 220 (2013) 311–319.
- [10] Y. Li, S. Shi, H. Cao, *et al.*, Bipolar membrane electrodialysis for generation of hydrochloric acid and ammonia from simulated ammonium chloride wastewater, *Water Res.* 89 (2015) 201–209.
- [11] C. Jiang, Q. Wang, Y. Zhang, *et al.*, Separation of methionine from the mixture with sodium carbonate using bipolar membrane electrodialysis, *J. Membr. Sci.* 498 (2016) 48–56.
- [12] M. Reig, C. Valderrama, O. Gibert, *et al.*, Selectrodialysis and bipolar membrane electrodialysis combination for industrial process brines treatment: Monovalent-divalent ions separation and acid and base production, *Desalination* 399 (2016) 88–95.
- [13] T. Xu, Development of bipolar membrane-based processes, *Desalination* 140 (3) (2001) 247–258.
- [14] J. Jaimeferrer, E. Couallier, P. Viers, *et al.*, Three-compartment bipolar membrane electrodialysis for splitting of sodium formate into formic acid and sodium hydroxide: Role of diffusion of molecular acid, *J. Membr. Sci.* 325 (2) (2008) 528–536.
- [15] J. Balster, I. Pünt, D.F. Stamatialis, *et al.*, Electrochemical acidification of milk by whey desalination, *J. Membr. Sci.* 303 (1–2) (2007) 213–220.
- [16] F. Lin Teng Shee, L. Bazinet, Cationic balance and current efficiency of a three-compartment bipolar membrane electrodialysis system during the preparation of chitosan oligomers, *J. Membr. Sci.* 341 (1–2) (2009) 46–50.
- [17] M.-L. Lameloise, R. Lewandowski, Recovering l-malic acid from a beverage industry waste water: Experimental study of the conversion stage using bipolar membrane electrodialysis, *J. Membr. Sci.* 403–404 (2012) 196–202.
- [18] D. Baş, İ.H. Boyacı, Modeling and optimization I: Usability of response surface methodology, *J. Food Eng.* 78 (3) (2007) 836–845.
- [19] Z. Kong, M. Li, J. Chen, *et al.*, Processing factors of triadimefon and triadimenol in barley brewing based on response surface methodology, *Food Control* 64 (2016) 81–86.
- [20] X. Qin, Y. Wang, C. Lu, *et al.*, Structural acoustics analysis and optimization of an enclosed box-damped structure based on response surface methodology, *Mater. Des.* 103 (2016) 236–243.
- [21] G. Zhao, H. Wang, G. Liu, *et al.*, Box–Behnken response surface design for the optimization of electrochemical detection of cadmium by square wave anodic stripping voltammetry on bismuth film/glassy carbon electrode, *Sensors Actuators B Chem.* 235 (2016) 67–73.