Sulfuric Acid and Ammonia Generation by Bipolar Membranes Electrodialysis: Transport Rate Model for Ion and Water Through Anion Exchange Membrane

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Regeneration of sulfuric acid and ammonia from ammonium sulfate by bipolar membrane electrodialysis (BMED) coupling with stripping ammonia by air-blowing was studied. The result showed that it was feasible to regenerate sulfuric acid and ammonia from ammonium sulfate solution using this method. Empirical models to describe the ion and water transport behaviors through anion exchange membrane for BMED system were successfully developed. The models were valid to evaluate water transport rate and ion transport behavior for anion exchange membrane under similar operation conditions. Comparison of calculated values with experimental data indicated that the models were reliable to describe the water and ion transport behavior through anion exchange membrane for BMED system and could also be used to predict the water transport and ion transport behaviors for other current density under similar operation conditions.

Key words:
Ammonium sulfate, anion exchange membrane, bipolar membrane, electrodialysis, transport

Introduction

Large amount of wastewaters containing ammonium sulfate are generated from organic acid, especially from amino acid such as lysine and glutamic acid production processes.[1,2] To remove ammonium sulfate from wastewaters, the processes of concentrated crystallization and desalted by electrodialysis have been studied.[3,4] However, the energy consumption is too high in the concentrated process and ammonium sulfate is not suitable to use as fertilizer in agriculture. Thus, a lot of wastewaters containing ammonium sulfate are discharged into the environment, resulting in resource waste and environmental pollution. For economic and environmental considerations, the ideal way is to find a path conjugating with neutralization. EDBM is such an advantageous approach: salts dissociate in the same way as in conventional electrodialysis and the corresponding ions form acids and bases with the H^+ and OH^- ions supplied by the water splitting in bipolar membranes. In such a way, EDBM can form closing loops with neutralization reactions and carry out acid and base regeneration and salt containing effluent treatment at the same time.

EDBM is a kind of electrodialysis (ED) technology, which is characteristic of water splitting in bipolar membranes. It has attracted more and more attention from the academia and industry application due to its technical advance, economic competence, and environmental benignity.[5-7] When applied to treat wastewater containing salt, EDBM can split salt into acid and base and overcome salt pollution if discharged without treatment. The applications of BMED in the treatment of wastewaters containing inorganic salt such as: NaCl;[8] Na_2CO_3;[9] $NaNO_3$;[10] Na_2SO_4;[11] Na_3PO_4;[12,13] NH_4NO_3[14] had been reported. Zhang *et al.*[15] investigated the direct treatment of ammonium sulfate using BMED coupling with ammonia stripping, which showed that BMED was available to treat the solution containing ammonium.

Lay-Pee *et al.*[16] have presented a mathematical model of acid concentration to describe the ion and water transport behavior of an electrodialysis process for concentrating citric acid under the influence of different current density. In this model, the ion transport behavior was only expressed by concentration variation. However, the concentration variation caused by volume variation of acid solution was neglected. In the present work, the three-compartment BMED coupling with stripping ammonia was investigated to regenerate sulfuric acid and ammonia from ammonium sulfate solution. A mathematical model was developed to describe the ion and water transport behavior through anion exchange membrane for regeneration of sulfuric acid and ammonia.

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Materials and methods

Ion-exchange membranes

The membranes used are listed below and their main characteristics are shown in Table 1:

– cation-exchange membrane: JCM-15 (Huanyu Lida);

– anion-exchange membrane: JAM-15 (Huanyu Lida);

– bipolar membrane: BP-1 (Tokuyama Soda).

Table 1 – *Main characteristic of the membranes used in experiments*

Membrane	Characteristic	
Cation-exchange membrane (JCM-15)	Type, group	Interpolymer type, sulfonic group
	area resistance (Ω cm^2)	≤7
	water fraction (%)	≥22
	transport number	≥95 %
	thickness (mm)	0.10 – 0.13
	ion exchange capacity (mg dg^{-1})	≥1.6
Anion-exchange membrane (JAM-15)	Type, group	Interpolymer type, quarternary ammonium group
	area resistance (Ω cm^2)	≤8
	water fraction (%)	≥18
	transport number	≥88 %
	thickness (mm)	0.10 – 0.12
	ion exchange capacity (mg dg^{-1})	≥1.3
Bipolar membrane (BP-1)	Type, group	Composition of the cationic and anionic membranes
	water splitting voltage (1N NaOH · 1N HCl, 100 mA cm^{-2}, 30 °C)	0.9 – 1.7 V
	water splitting efficiency	> 0.98
	burst strength (MPa)	0.4 ~ 0.7
	thickness (mm)	0.17 – 0.26

Electrodialysis experiment

Electrodialyzer supplied by Sanyuan Bada Co. (Beijing, China) with five cells was used in this work. The effective area of each membrane was A = 147 cm^2 and compartment width was about 2 mm. The current density during operation was kept constant by an AC-DC rectifier. The temperature of solutions was controlled by thermostat (DC-1015, China).

Batch-mode BMED with three-compartment cell was carried out at constant current density. The cell configuration to regenerate sulfuric acid and ammonia is shown in Fig. 1. It involved four independent streams: acid stream (AS), base stream (BS), salt stream (SS) and electrode rinse stream (ES). In the experiments, a predetermined volume of various solutions (c = 0.3 mol L^{-1} sodium sulfate, 1 L; c = 0.1 mol L^{-1} sulfuric acid, 1 L; c = 0.1 mol L^{-1} sodium hydroxide, 1 L) was circulated by diaphragm pumps (HIGHHfl0-8007, Taiwan) at the flow rate of Q = 20 L h^{-1} in electrode compartment, acid compartment and base compartment, respectively. The model solution containing c = 0.3 mol L^{-1} ammonium sulfate (2 L) was circulated at the flow rate of Q = 20 L h^{-1} in the salt compartment. The solution temperature was maintained at 30 °C and the operation time was 2 h. During BMED, the ammonia generated was extracted continuously from the base reservoir tank by air-blowing. The flow rate of air was Q = 0.5 m^3 h^{-1}.

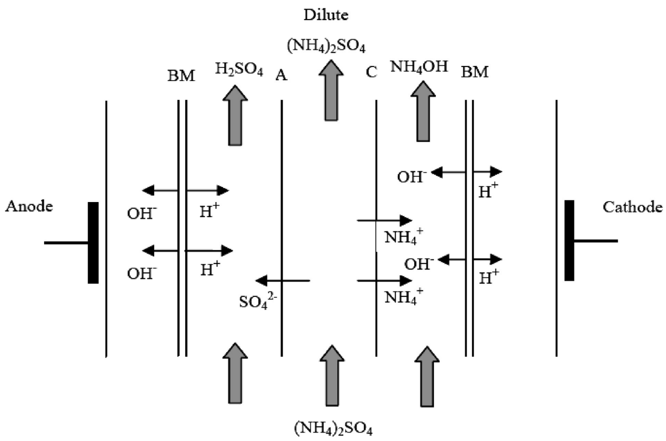


Fig. 1 – *Cell configuration of three-compartment cell (A: anion exchange membrane; C: cation exchange membrane; BM: bipolar membrane)*

3 mL of sample in acid circulation was taken at the interval of 20 min. The sulfuric acid concentration was measured by titration and sulfate ions concentration was determined by barium chromate spectrophotometry.[17] Volume variation for each solution was determined by reading the scale on the reservoir tank.

Calculations

The average current efficiency (ACE) of sulfuric acid regeneration by BMED at the acid stream could be defined by the equation:

$$\mathrm{ACE} = \frac{100\, z\, F\, n}{N\, I\, t} \qquad (1)$$

where ACE is an apparent quantity; n is the amount of sulfuric acid; z is the valence of sulfate; F is Faraday's constant, which is 96 500 C mol^{-1}; N is the number of membrane pairs; I is the current; t is the operation time.

Energy consumption (E_c) is defined as eq. (2) :

$$E_c = \frac{I \int U\, \mathrm{d}t}{m} \text{ (kWh kg}^{-1}\text{ salt treated)} \qquad (2)$$

where, U is voltage (V); I is the current; t is the operation time; m is the mass of salt treated.

Results and discussion

Regeneration of sulfuric acid and ammonia using BMED coupling with ammonia in situ stripping

Batch-mode experiments were carried out at four different current densities of j = 10, 20, 25 and 30 mA cm^{-2}, respectively. The ammonia generated in the base reservoir tank was continuously extracted by air-blowing. Average current efficiency (ACE) and energy consumption of sulfuric acid at different current densities are shown in Table 2. The final concentration of sulfuric acid and its average current efficiency reached c = 0.561 mol L^{-1} and 68.7 % at the current density of j = 30 mA cm^{-2} after t = 2 h of operation, respectively. The energy consumption in the experiment was E_c = 0.207 kWh mol^{-1} sulfuric acid. The results indicated that it was feasible to regenerate sulfuric acid and ammonia from ammonium sulfate solution using BMED coupling with stripping ammonia.

T a b l e 2 – *Average current efficiency (ACE) and energy consumption of sulfuric acid at different current densities in BMED*

Current density j/mA cm^{-2}	ACE (%)	Energy consumption E_c/kWh mol^{-1} sulfuric acid
10	60.3	0.235
20	63.0	0.223
25	65.4	0.211
30	68.7	0.207

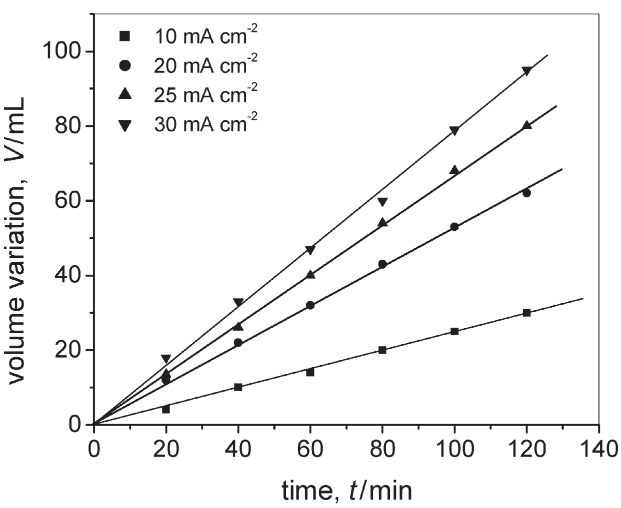
Transport rate model for water through anion exchange membrane

Volume variations of acid solution

Volume variations occurred during the mass transfer between the different solutions in BMED, which resulted in the increase of volume in acid solution followed by the decrease of volume in the salt solution. The volume variations of sulfuric acid solution at the current densities of j = 10, 20, 25 and 30 mA cm^{-2} are shown in Fig. 2. A linear relationship between the volume change in the acid stream and operation time was observed, which means the water transferred is directly proportional to the current density applied. The volume variation rate (dV/dt) could be evaluated from the slope of the straight line that was obtained by the linear regression based on the experimental data. Therefore, a relationship between the volume variation rate and current density (j) could be formed by plotting the volume variation rate (dV/dt) as a function of the current density. A straight line was obtained as shown in Fig. 3. The mathematical model relating the volume variation rate in terms of current density is expressed by the eq. (3):

$$\frac{\mathrm{d}V}{\mathrm{d}t} = f + k\, j \qquad (3)$$

where both f and k are coefficients.



F i g . 2 – *Volume variation of acid solution at different current densities during BMED*

Setting the initial solution volume in AS to $V_{\mathrm{sul}}^{\mathrm{AS}}(0)$ at the initial time $t = 0$ and solution volume in AS to $V_{\mathrm{sul}}^{\mathrm{AS}}(t)$ at the time t, eq. (4) is obtained by integration of eq. (3):

$$V_{\mathrm{sul}}^{\mathrm{AS}}(t) - V_{\mathrm{sul}}^{\mathrm{AS}}(0) = (f + k\, j)\, t \qquad (4)$$

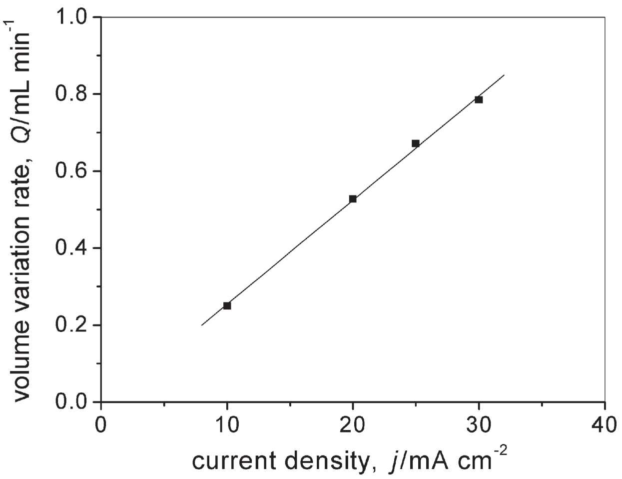


Fig. 3 – *Relationship between volume variation rate and current density*

The coefficients of $f = -0.01668$ and $k = 0.02706$ in eq. (4) are obtained by linear regression on experimental data. Eq. (4) integrates acid solution volume in AS as a function of time and current density.

Water-transport rate model

Water as concomitant is transported through the ion exchange membranes when ions transfer through the membranes in electrodialysis, which is the main result of electro-osmosis.[8] Thus, the volume variation of the acid solution is observed as the result of water transport. The amount of water transported (n_w) can be defined by eq. (5).

$$n_{\mathrm{w}} = \frac{V_{\mathrm{sul}}^{\mathrm{AS}}(t) - V_{\mathrm{sul}}^{\mathrm{AS}}(0)}{V_{\mathrm{m,w}}} \tag{5}$$

where, the term $V_{\mathrm{m,w}}$ is molar volume of water in mL mol^{-1}.

Substituting eq. (4) into eq. (5), to obtain eq. (6):

$$n_{\mathrm{w}} = \frac{(f + kj)\,t}{V_{\mathrm{m,w}}} \tag{6}$$

The water-transport rate (J_{w}) through the anion exchange membrane into AS of BMED system is defined as the amount of ions through unit area of membrane at the unit time, which is expressed by:

$$J_{\mathrm{w}} = \frac{n_{\mathrm{w}}}{N S_{\mathrm{m}} t} \tag{7}$$

where S_{m} is the effective membrane area; N is the number of anion exchange membrane.

Substituting eq. (6) into eq. (7), water-transport rate in terms of the current density is expressed by:

$$J_{\mathrm{w}} = \frac{f + kj}{N S_{\mathrm{m}} V_{\mathrm{m,w}}}. \tag{8}$$

Transport rate model for ion through anion exchange membrane

Acid concentration

The time course of acid concentration for AS at different current densities was investigated in BMED. All the experiments were carried out at the initial sulfuric acid concentration of $c = 0.1$ mol L^{-1}. The results are shown in Fig. 4. A straight line between acid concentration and operation time was observed. The variation rate of the acid concentration ($\mathrm{d}c/\mathrm{d}t$) could be evaluated from the slopes that were obtained by linear regression on the experimental data. Fig. 5 shows the variation rate data of the acid concentration at different current densities, which represents the anion-transport rate through the anion exchange membrane in BMED. Thus, a

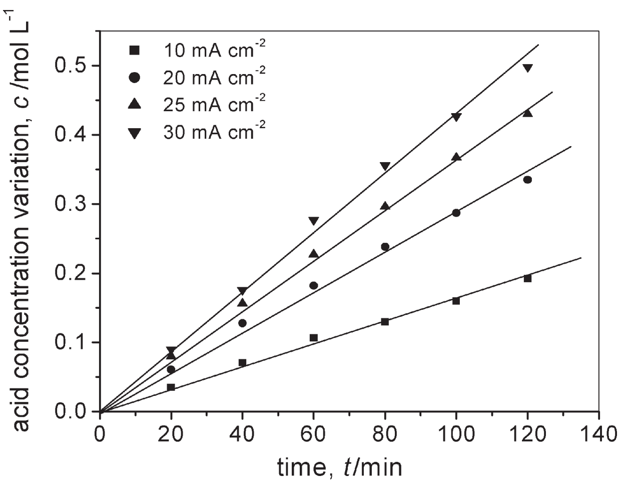


Fig. 4 – *Variation of sulfuric acid concentrations in AS as a function of time in BMED*

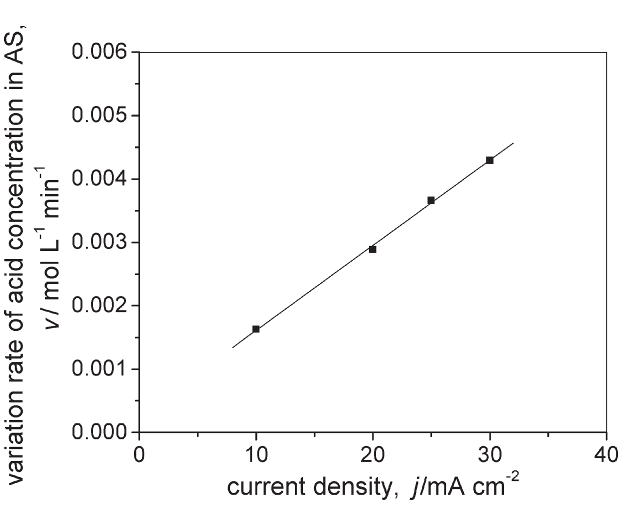


Fig. 5 – *Relationship between variation rate of sulfuric acid concentration in AS and current density*

relationship between the variation rate of sulfuric acid concentration and current density can be formed by plotting the values (dc/dt) as a function of current density (Fig. 5).

As shown in Fig. 5, a linear relationship was obtained between the variation rate of sulfuric acid concentration and current density, which can be expressed by the eq. (9).

$$\frac{dc}{dt} = m + nj \tag{9}$$

In the eq. (9), the intercept (m) indicates that the sulfate transport is not equal to zero when no current was applied. The term n represents the slope of the linear line as shown in Fig. 5. The values of $m = 2.678 \cdot 10^{-4}$ and n = $1.3428 \cdot 10^{-4}$ were calculated based on the linear regression.

Setting sulfuric acid concentration in AS equal to $c_{sul}^{AS}(0)$ at the initial time $t = 0$, eq. (9) is integrated to form eq. (10).

$$c_{sul}^{AS}(t) - c_{sul}^{AS}(0) = (m + nj)t \tag{10}$$

Eq. (10) integrates the acid concentration in AS as a function of time and current density.

Average current efficiency

ACE of sulfuric acid production at the AS is defined by eq. (1). Under constant current conditions, q is defined as shown in eq. (11).

$$q = \frac{0.06\, j\, S_m\, t}{F} \tag{11}$$

where F is Faraday's constant that is equal to 96 500 C mol^{-1}.

At the time t, the term n is defined as in eq. (12).

$$n = n_{sul}^{AS}(t) - n_{sul}^{AS}(0) \tag{12}$$

where $n_{sul}^{AS}(t)$ and $n_{sul}^{AS}(0)$ are the amount of sulfuric acid in the AS at the time t and the initial time, respectively.

Eq. (1) is expanded by substituting the eq. (11) and eq. (12) to obtain the equation of ACE for sulfuric acid as expressed by eq. (13).

$$\text{ACE} = \frac{100\, z\, F[n_{sul}^{AS}(t) - n_{sul}^{AS}(0)]}{0.06\, N j\, S_m\, t} \tag{13}$$

Since volume variation was observed in the AS, the effect of volume variation should be considered. Thus, a final expression of ACE is developed as expressed as in eq. (14). This equation is used in the ACE calculation.

$$\text{ACE} = \frac{z\, F[c_{sul}^{AS}(t)\, V_{sul}^{AS}(t) - c_{sul}^{AS}(0)\, V_{sul}^{AS}(0)]}{0.06\, N j\, S_m\, t} \tag{14}$$

Fig. 6 shows the results of ACE as a function of sulfuric acid concentration variation at the current density of $j = 20$ mA cm^{-2}. It was observed that the experimental data could be plotted on a straight line. This means that the ACE for AS obeys a simple mathematical equation:

$$\text{ACE} = \alpha - \beta[c_{sul}^{AS}(t) - c_{sul}^{AS}(0)] \tag{15}$$

where α is the intercept on the y-axis; term β represents the slope value of linear line.

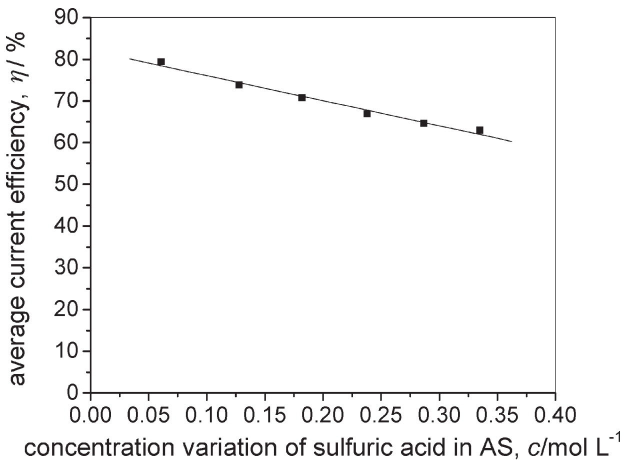


Fig. 6 – *Average current efficiency for regeneration of sulfuric acid in BMED against the variation of sulfuric acid concentration in AS at the current density of 20 mA cm^{-2}*

Substituting eq. (10) into eq. (15), we obtained the eq. (16).

$$\text{ACE} = \alpha - \beta(m + nj)t \tag{16}$$

The values of α and β at the different current densities were evaluated by linear regression on the experimental data as shown in Table 3, indicating that α and β were affected by current density and could be seen as the functions of current density. Based on the data, the mathematical models were developed to describe the relationship between $\alpha(\beta)$ and current density as expressed in eq. (17) and eq. (18), respectively.

$$\alpha = a_1 + b_1 j \tag{17}$$

$$\beta = a_2 + b_2 j + c_2 j^2 \tag{18}$$

The values of coefficients in eq. (17) and eq. (18) were evaluated by linear and non-linear regression: a_1 = 118.43, b_1 = –1.721, a_2 = 753.9, b_2 = –54.02, c_2 = 0.9842.

Table 3 – *Values of α and β at different current densities*

Current density j/mA cm^{-2}	α	β
10	102.21	313.3
20	82.08	60.1
25	75.34	27.9
30	67.83	15.51

Substituting eq. (17) and eq. (18) into eq. (16), we obtained:

$$\mathrm{ACE} = (a_1 + b_1 j) - [a_2 m + (a_2 n + b_2 m) j + (b_2 n + c_2 m) j^2 + c_2 n j^3]\, t \qquad (19)$$

ACE in AS could be evaluated by the eq. (19) at the time t for the system of BMED applied by the current density j.

Ion-transport rate model

The average transport rate (J_a) of sulfate through anion exchange in BMED is defined as the amount of ions through the membrane of unit area at the unit time. At the time t, the average transport rate is expressed by eq. (20).

$$J_a = \frac{n}{N S_m t} \qquad (20)$$

Then, eq. (20) can be expanded to obtain eq. (21):

$$J_a = \frac{c_{sul}^{AS}(t) V_{sul}^{AS}(t) - c_{sul}^{AS}(0) V_{sul}^{AS}(0)}{1000 N S_m t} \qquad (21)$$

As both eq. (14) and eq. (19) expressed the ACE in BMED simultaneously, these equations were rearranged to eliminate term ACE. Therefore, combining eq. (21) we obtained:

$$J_a = 6 \cdot 10^{-4} \cdot \left\{ \frac{(a_1 j + b_1 j^2)}{zF} - \frac{t[a_2 m j + a_2 n + b_2 m) j^2 + (b_2 n + c_2 m) j^3 + c_2 n j^4]}{zF} \right\} \qquad (22)$$

Based on the coefficients as described above, the average transport rate (J_a) of sulfate through anion exchange membrane into AS was determined by the time t for the system of BMED at any process of constant current density. Eq. (22) describes the behavior of the transport rate of sulfate through anion exchange membrane by using BMED. This equation is a general equation that represents the ion transport behavior through anion exchange membrane in the BMED system. The values of the coefficients are affected by the characters of membranes and solutions. Therefore, the coefficients should be determined based on experiments in different BMED systems.

An acid concentration model was also developed to describe the concentration change of sulfuric acid in AS for the BMED system by rearranging the eq. (21) and combining eq. (4). So, we obtained the eq. (23):

$$c_{sul}^{AS}(t) = \frac{c_{sul}^{AS}(0) V_{sul}^{AS}(0)}{V_{sul}^{AS}(0) + (f + kj)t} + 0.6 N S_m t \cdot \frac{(a_1 j + b_1 j^2) - t[a_2 m j + a_2 n + b_2 m) j^2 + (b_2 n + c_2 m) j^3 + c_2 n j^4]}{zF[V_{sul}^{AS}(0) + (f + kj)t]} \qquad (23)$$

Based on all coefficients described above and the initial experimental conditions, eq. (23) can be used to calculate the concentration of sulfuric acid in BMED process. Different ion exchange membranes as well as different solutes would affect the value of coefficients. Therefore, these coefficients should be determined experimentally for different ion exchange membranes and solutes used.

Reliability test of water transport and ion transport model

In order to verify the developed water-transport rate model and ion-transport model, the calculated values were compared with the experimental data. The results of experimental and the calculated values of water transport rate for different current densities are shown in Fig. 7, Fig. 8 and Fig. 9.

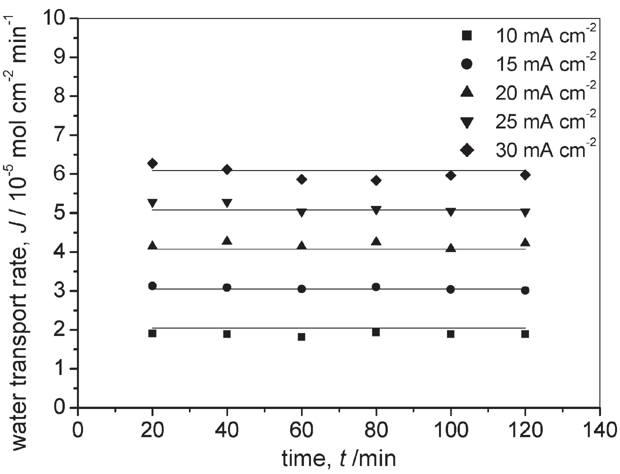


Fig. 7 – *Comparison of experimental and calculated data of water transport rate for different constant current densities (point: experimental data; line: calculated data)*

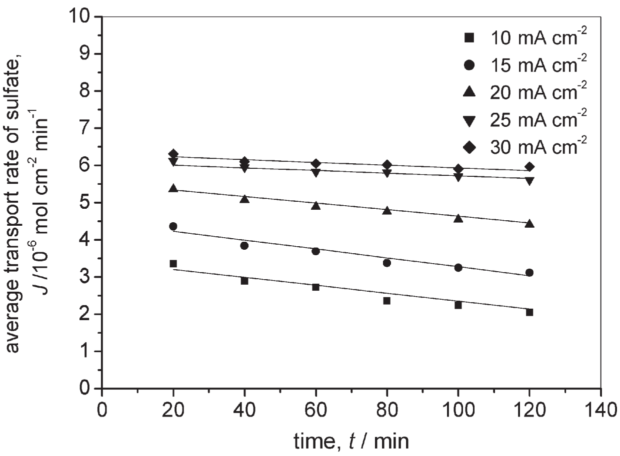


Fig. 8 – *Comparison of experimental and calculated results of sulfate average transport rate for different constant current densities (point: experimental data; line: calculated data)*

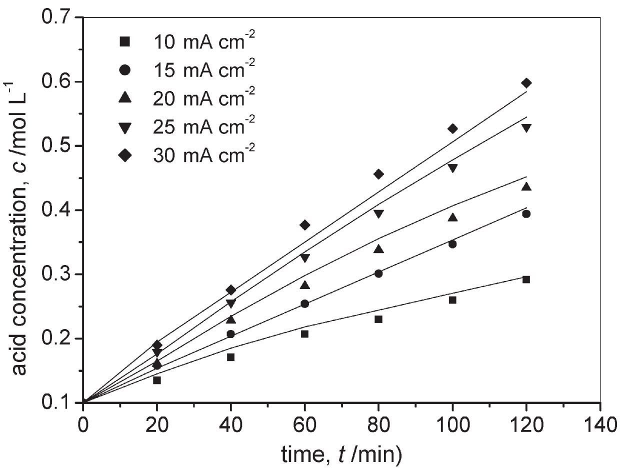


Fig. 9 – *Comparison of experimental and calculated results of sulfate concentration for different constant current densities (point: experimental data; line: calculated data)*

Fig. 7 compares the results of experimental and calculated data of water transport rate for different current densities, which shows a good agreement between the experimental and calculated results. This indicates that the water-transport rate model (eq. (8)) is reliable to describe the actual water transport behavior through anion exchange membrane for BMED system. This model could also be used to predict the water transport for other current densities under similar operational conditions. For example, a good prediction of water transport model was obtained by the model at the current density of $j = 15$ mA cm^{-2} (Fig. 7).

Fig. 8 and Fig. 9 compare the results of experimental and calculated data of ion-transport rate and the change of sulfuric acid concentration for different current densities, respectively. From Fig. 8 and Fig. 9, the calculated results of sulfate average transport rate and sulfate concentration in AS were almost in accordance with the experimental data. Therefore, the ion average transport rate model (eq. (22)) and acid concentration model (eq. (23)) are reliable to describe the sulfate transport behavior through anion exchange membrane for BMED system. These models could also be used to predict the sulfate transport for other current density under similar operational conditions. For example, sulfate average transport rate and sulfuric acid concentration at the current density of j = 15 mA cm^{-2} were predicted by the model very well.

Conclusions

It was feasible to regenerate sulfuric acid and ammonia from ammonium sulfate solution using BMED coupling with ammonia *in situ* stripping by air-blowing. The general models to describe water transport and ion transport behaviors for BMED system were successfully developed. The values of all coefficients in the models were different for different membranes and solutions and had to be determined experimentally. Comparison of the calculated values with the experimental data indicated that the developed models were reliable to describe the water transport rate and ion transport behavior through anion exchange membrane for BMED system. They could also be used to predict the water transport rate and ion transport behavior for other current densities under similar operational conditions.

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List of symbols

- $\mathrm{d}V/\mathrm{d}t$ – volume variation slope of acid solution with time, mL min^{-1}
- c – concentration, mol L^{-1}
- f, k – volume variation coefficient
- j – current density, mA cm^{-2}
- $V_{\mathrm{sul}}^{\mathrm{AS}}(t)$ – sulfuric acid solution volume in acid stream at time t, mL
- $V_{\mathrm{sul}}^{\mathrm{AS}}(0)$ – sulfuric acid solution volume in acid stream at time t = 0, mL
- t – time, min
- n_{w} – amount of water transport, mol
- $V_{\mathrm{m,w}}$ – molar volume of water, mL mol^{-1}

J_w – water transport rate through anion exchange membrane into acid stream, mol cm^{-2} min^{-1}

S_m – effective membrane area, cm^2

N – number of anion exchange membrane

dc/dt – variation slope of acid concentration with time, mol L^{-1} min^{-1}

m, n – acid concentration variation coefficient

$c_{sul}^{AS}(t)$ – sulfuric acid solution concentration in acid stream at time t, mol L^{-1}

$c_{sul}^{AS}(0)$ – sulfuric acid solution concentration in acid stream at time t = 0, mol L^{-1}

z – valence of ion

n – amount of sulfate transferred through anion exchange membrane, mol

q – amount of electrical charges carried by the current, mol

F – Faraday's constant, 96 500 C mol^{-1}

$n_{sul}^{AS}(t)$ – amount of sulfuric acid in acid stream at time t, mol

$n_{sul}^{AS}(0)$ – amount of sulfuric acid in acid stream at time t = 0, mol

Q – volume flow rate, L h^{-1}

α, β – phenomenological coefficients

a_1, b_1, a_2, b_2, c_2 – average current efficiency coefficients

J_a – ion transport rate through anion exchange membrane into acid stream, mol cm^{-2} min^{-1}

η – efficiency, %

References

1. *Atkinson, B., Mavtuna, F.*, Biochemical engineering and biotechnology Handbook, 2nd ed. New York: Stockton Press, 1991.
2. Monosodium glutamate production factory of Shanghai, The production of monosodium glutamate, Beijing, Light Industry Press, 1978.
3. *Shen, G.*, Commun. Fermentation Sci. & Tech. (in Chinese) **34** (2005) 39.
4. *Hong-Joo, L., Suk-Jung, O., Seung-Hyeon, M.*, Water Res. **37** (2003) 1091.
5. *Xu, T.*, Resources, Conservation and Recycling **37** (2002) 1.
6. *Huang, C., Xu, T.*, Environ. Sci. Technol. **40** (2006) 5233.
7. *Huang, C., Xu, T., Zhang, Y., Xue, Y., Chen, G.*, J. Membrane Science **288** (2007) 1.
8. *Mazrou, S., Kerdjoudj, H., Cherif, A., Elmidaoui, A., Molénat, J.*, New J. Chem. **22** (4) (1998) 355.
9. *Audinos, R.*, Chem. Eng. Technol. **20** (1997) 247.
10. *Chérif, A., Molénat, J., Elmidaoui, A.*, J. Appl. Electrochem. **27** (1997) 1069.
11. *Paleogou, M., Thibault, A., Wong, P., Thompson, R., Berry, R.*, Sep. Purif. Technol. **11** (1997) 159.
12. *Trivedi, G., Shah, B., Adhikary, S., Rangarajan, R.*, React. Funct. Polym. **39** (1999) 91.
13. *Trivedi, G., Shah, B., Adhikary, S., Indusekhar, V., Rangarajan, R.*, Reactive & Functional Polymers. **28** (1996) 243.
14. *Ben, A., Rakib, M., Laborie, S., Viers, P., Durand, G.*, J. Membrane Science **244** (2004) 89.
15. *Zhang, X., Lu, W., Cong, W.*, Chinese Journal of Process and Engineering, 2007, 4 (Accepted).
16. *Lay-Pee, L., Heng-Fatt, L., Sarmidi, M.*, J. Membr. Sci. **199** (2002) 59.
17. National bureau of Environmental Protection, Analyses and measurement methods of water and wastewater, Third ed., Beijing, 1989.