Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2018.22684

123 (2018) 74–81
August

Extraction of DMAC from the pharmaceutical wastewater by a multistage countercurrent model

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Received 2 November 2017; Accepted 26 June 2018

ABSTRACT

A multistage countercurrent solvent extraction model was used for recovering *N*,*N*-dimethylacetamide (DMAC) from the pharmaceutical wastewater. The theoretical stages were obtained to be five by graphical method in the light of the fitted phase equilibrium data. And then the five-stage countercurrent extraction experiments were performed in the actual and simulant effluent systems, respectively. It was found that the optimum conditions of the experiments were as follow: pH value of 8, phase ratio (O/A) of 1:1, and addition of 0.05 mass fraction of NaCl solid into wastewater at room temperature. The reliability of the theoretical stages was verified on the basis of the relative errors (<0.0641) calculated by the theoretical and experimental data, as well as the high extraction efficiency obtained by the actual and simulant wastewater systems. The results showed that the content of DMAC in the simulant wastewaters decreased to 0.0074, the actual one dropped to 0.0078, and thus the extraction efficiency of DMAC was 97.53% and 97.40%, respectively.

Keywords: Multistage countercurrent model; Graphical method; Pharmaceutical wastewater; *N*,*N*-dimethylacetamide; Extraction efficiency

1. Introduction

Amide compounds as pollutants have aroused widespread attention due to their toxicity and difficult degradation in the environment, which are kind of typical organic components in industrial wastewaters from plastics plants, paint factories, pharmaceutical factories, and textile mills [1]. Wastewaters containing amide compounds are threats to the ecological environment and public health when released without proper treatment [2]. For instance, *N*,*N*-dimethylacetamide (DMAC) is an excellent solvent in pharmaceutical industry, and generally exists in industrial wastewaters, but its toxicity can intensely irritate human eyes, skin, and respiratory tract mucosa. The abnormal symptoms of liver, kidney, heart, blood vessels, and nerves will appear if people often get contact with DMAC [3]. Therefore, investigations on the treatment of effluent containing DMAC are urgently needed.

The current treatment methods of pharmaceutical wastewater are batch biofilter, microbial fuel cells (MFCs), catalytic ozonation, membrane separation, and solvent extraction. Buitróu and Melgoza [4] removed nitroaromatic compounds in pharmaceutical wastewater using the method of the sequencing batch biofilter and the removal efficiency was about 80%. Liu and Liu [5] studied the method of MFCs on the treatment of DMAC-containing wastewaters and found that this way is possible to realize the power extraction from DMAC wastewaters in the form of electricity by the bioconversion process of MFC. However, some substances in the effluents have a negative impact on the survival of microorganisms, which limits the efficiencies and applications of the batch biofilter and MFC methods [6]. In the method of catalytic ozonation, copper ferrite ($CuFe_2O_4$) magnetic nanoparticles (MNPs) were used for the

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degradation of DMAC by Zhang and Ji [7]. The results suggest that the synergetic effect between $CuFe_2O_4$ MNPs and O_3 in the $CuFe_2O_4/O_3$ process was significant and the degradation efficiency of DMAC reached 95.4%. This method is an effective way to deal with wastewater containing DMAC, and the removal efficiency is satisfactory, while a deficiency is that DMAC cannot be recovered and reused. Recently, membrane separation technology has been studied extensively to the industrial outlet water treatment, which can separate high-purity compounds, but cannot be large-scale applied in industry now because of its immature technology in domestic and the high operating costs [8]. Liu et al. investigated the method of solvent extraction using cumene as the extractant to treat the phenol-containing wastewaters. The results report that cumene shows excellent extraction performance on phenol and the efficiency of DMAC is above 99%, which indicate that solvent extraction is an effective way to remove and recover phenol from the wastewaters [9]. In addition, the comparison of the methods of SBB, catalytic ozonation, MFCs, and solvent extraction mentioned earlier was shown in Table 1.

Therefore, the extraction method is a promising technique for wastewaters treatment due to its mature technology, simple operation, low energy consumption and high extraction efficiency [9,10]. Most importantly, this method can recover DMAC from wastewaters and the extractant can be separated by low-temperature distillation and cyclic utilization in the process of extraction in this paper. The key to the method is to obtain a high extraction efficiency, so optimal extractant, operational conditions, and extraction stages need to be determined by experiments to achieve this goal.

The aim of this work is to calculate the extraction efficiency of DMAC by simulation experiments. The optimum conditions of the experiment were determined by the method of control variable, and the theoretical stages of extraction were calculated by using liquid–liquid equilibrium (LLE) data, then dichloromethane was selected as an extraction solvent and the extraction efficiencies of DMAC were calculated by the multistage countercurrent extraction experiments.

In order to achieve the desired result of the extraction efficiency and touch the anticipative target of the DMAC content (mass fraction reached 0.01), the number of theoretical stages was calculated by graphical method according to the LLE data published before [11] and the extraction experiments were simulated in the light of the optimum conditions.

2. Experiments of single factor

2.1. Chemicals and apparatus

All the chemicals used in the experiment are analytical reagent grade and without further purification and the purity of them was examined by gas chromatography. Deionized water was prepared in our laboratory and used in all experiments (electrical conductivity k = 3.427 μs cm^{-1}; pH = 6.7). The source, mass fraction and relative molecular mass of the chemicals are listed in Table 2.

The water content of the two phases in the samples was determined by the Automatic Trace Moisture Analyzer (ATMA Model SF101). The other components of each sample were analyzed by gas chromatograph (Agilent GC 6890n) with an HP-5 (30 m × 0.32 mm × 0.25 μm) capillary column and a hydrogen flame ionization detector. The uncertainty of the calculated mass fraction was estimated less than ±0.0005.

The simulation of countercurrent cascade experiments was completed in separating funnels.

Table 1
Comparison of the methods of SBB, catalytic ozonation, MFCs, and solvent extraction

Method	SBB[a] [4]	Catalytic ozonation [7]	MFCs[c] [5]	Solvent extraction [9]
Wastewater composition	Phenols, *o*-nitroaniline	150 mL DMAC aqueous solution C_{DMAC} = 200 mg/L	NH_4Cl: 0.1 g/L, NaH_2PO_4: 0.5 g/L Na_2PO_4: 1.0 g/L, DMAC: 0.5 mL/L	Phenol ($C_{p,ini}$[d] = 1,004 ppm) HCl, H_2SO_4
Applicable condition	Pharmaceutical wastewater containing nitroaromatic compounds	Industrial wastewater containing DMAC	Industrial wastewater containing DMAC	Pharmaceutical wastewater containing phenol
Optimal operating condition	The anaerobic stage: 8–12 h The aerobic stage: 4–12 h	Catalyst: $CuFe_2O_4$ MNPs[b]/O_3 Catalyst dosage: 30 g/L O_3 flow rate: 0.6 L/min T = 120min	Culture exoelectrogens: 350 h Influent/effluent flux:0.86 mL/min HRT = 12 min	T = 25°C, extractant: cumene PH: neutral or weak acid O/A[e] = 1:1 (Volume ratio) extraction time = 30 min
Removal/extraction efficiency	Above 80%	95.4%	15%–50%	Above 99%

[a] Sequecing bath biofilter.
[b] Magnetic nanoparticles.
[c] Microbial fuel cells.
[d] The initial concentration of phenol.
[e] Phase ratio(organic/aqueous).

Table 2
Source, initial purity in mass fraction (*w*) and molar weights (*M*) of chemical reagents

Chemical name	Source	*w*	*M*	Analysis method	Purification method
n-Butanol	Beijing Chemical Works	0.995	74.12	GC[a]	None
Ethanol	Beijing Chemical Works	0.997	46.07	GC[a]	None
DCM[b]	Beijing Chemical Works	0.995	84.93	GC[a]	None
DMAC	Tianjin Guangfu Chemical Research Institute	0.998	87.12	GC[a]	None
NaOH	Beijing Chemical Works	0.960	39.99	–	None
Water	Made by Our Group	–	18.02	–	Redistilled twice
NaCl	Tianjin Chemical Reagent Factory	0.995	58.44	–	None

[a] Gas chromatography.
[b] DCM = dichloromethane.

2.2. Experiment on single influencing factor

In order to determine the optimum operating conditions, the effects of temperature, pH value, salt content, concentration of DMAC, and phase ratio on the extraction efficiency were investigated by adopting control variable. The detailed experimental data are shown in Tables 3(a)–(e), and the results are shown in Fig. 1.

The experiments were carried out in a glass cell (4 cm × 1.5 cm × 13 cm) placed in a constant temperature water bath with a magnetic stirrer (Model DF-101S) to ensure temperature stability and homogeneity [12]. A thermometer (with an accuracy of ±0.1 K) was used to measure the temperature in the equilibrium cell and ensure its constant. First, a certain amount of extractant and wastewater weighted by electronic balance (with an accuracy of ±0.0001 g) was added into the equilibrium cell in a certain conditions. Then, the stirring magnet was put into the cell and stirred for 4 h to ensure that all components were fully mixed. After that the mixture was placed for 6 h in a hermetic condition to reach the thermodynamic equilibrium. At last the system was divided into two liquid phases by a well-defined interface. The sample of organic-rich phase was taken by a syringe (5 mL) from the upper layer and that of water-rich phase was obtained from a sampling tap at the bottom of the cell. The concentrations of water in the both phases were analyzed by ATMA and the other organic components were determined by GC. The mean value is obtained by three parallel sampling tests. All experiments were carried out at atmospheric pressure.

3. Experiments of multistage extraction and simulation

3.1. Diagramming theoretical stages in triangular coordinate

The fitting solubility and auxiliary curves obtained according to the LLE data were plotted in the triangular coordinate, and the points of F (0, 0.3), S (1, 0) (pure extractant) and R_n (X_n = 0.01) were determined in the diagram on the basis of the known composition of the raw material liquid, the extractant, and the final raffinate phase, respectively. The composition point of the mixture M was acquired in the light of the amount of F and S as well as the lever rule. After that the point of E_1 was confirmed in accordance with Eqs. (1) and (2). Then, the operating line of multistage countercurrent extraction (Eq. (3)) and the operating point O were obtained by the material balance of overall and each stage. At last, the composition of each stage was determined by the auxiliary curve and the operating line equation. The requirement of the separation was completed when the composition of the raffinate phase at a certain stage $x_i \le x_n$, and the number of the tie-lines is the required theoretical stages [13,14]. The result is shown in Fig. 2 from which it can be seen that the theoretical stages are five.

$$E_1 = M\frac{MR_n}{E_1R_n} \tag{1}$$

$$R_n = M - E_1 \tag{2}$$

$$F - E_1 = R_1 - E_2 = R_2 - E_3 = \cdots = R_{n-1} - E_n = R_n - S = O \tag{3}$$

3.2. Diagramming theoretical stages in rectangular coordinate

The distribution curve was plotted according to the relation of phase equilibrium [12,13]. In the triangular coordinate, the operating lines and the solubility curve intersected at R_{i-1} and E_i, so the operation line in the rectangular coordinate was obtained by the points of (x_{i-1}, y_i). As shown in Fig. 3, started with the point of (x_F, y_1), the stages were drawn between the distribution curve and the operation line, and it cannot be finished until $x_i \le x_n$. In the figure, the number of the stages drawn is the required theoretical stages. The result shows that the theoretical stages are also five, which in accordance with the result of above.

3.3. Simulation of multistage countercurrent extraction

The wastewaters used in the extraction experiment were actual and simulant effluent systems, respectively. The actual effluent mass fraction mainly consists of 17%–22% of DMAC, 3%–4% of ammonium chloride, 1%–2% of dichloromethane, 8%–9% of hydrochloric acid, and 7%–8% of phosphoric acid. The simulant effluent was composed of 70% of water and 30% of DMAC. The effluent was pretreated with salt (sodium chloride) and alkali (sodium hydroxide) according to the optimum operating conditions. The mass fraction of 0.05 of the sodium chloride was added into wastewaters. The pH value was adjusted to 8 by adding sodium hydroxide and

Table 3
The effects of temperature (*T*), pH value, salt content (m_{NaCl}), concentration of DMAC (m_{DMAC}), phase ratio (O/A) on the distribution (*D*), and selectivity coefficient (*S*)

(a)

Constant	pH = 8, O/A = 1, m_{NaCl} = 5%, m_{DMAC} = 30%			
T (°C)	15	25	35	40
AVG (*D*)[a]	4.9873	4.4508	3.8275	3.1827
SD[b]	0.0821	0.0845	0.0652	0.0763
AVG (*S*)[a]	50.560	47.761	25.134	19.516
SD	0.0925	0.1022	0.1150	0.0874

[a] Average distribution coefficient (*D*) and selectivity coefficient (*S*).
[b] Standard deviation.

(b)

Constant	*T* = 25°C, O/A = 1, m_{NaCl} = 5%, m_{DMAC} = 30%						
pH	2	4	6	7	8	9	10
AVG (*D*)	4.8973	5.1675	5.2033	5.6243	5.7707	5.7723	5.7736
SD	0.0852	0.0927	0.0534	0.0512	0.0623	0.0750	0.0657
AVG (*S*)	64.034	72.049	73.977	74.6404	79.002	79.070	79.347
SD	0.0878	0.1022	0.1034	0.0987	0.1120	0.0975	0.1056

(c)

Constant	*T* = 25°C, O/A = 1, pH = 8, m_{DMAC} = 30%				
m_{NaCl} (%)	0	2	4	6	8
AVG (*D*)	3.9627	5.2063	5.8672	5.9038	5.8265
SD	0.0856	0.0752	0.0924	0.0723	0.0758
AVG (*S*)	51.889	54.576	55.375	50.3617	50.672
SD	0.1024	0.1124	0.0954	0.0862	0.0795

(d)

Constant	*T* = 25°C, O/A = 1, pH = 8, m_{NaCl} = 5%					
m_{DMAC} (%)	5	10	15	20	25	30
AVG (*D*)	6.4567	5.8675	4.7526	4.2034	4.3065	4.2126
SD	0.0756	0.0685	0.0821	0.0749	0.0685	0.0876
AVG (*S*)	50.313	45.179	40.484	38.048	36.834	31.1162
SD	0.1028	0.0856	0.1123	0.0958	0.1156	0.1134

(e)

Constant	*T* = 25°C, m_{DMAC} = 30%, pH = 8, m_{NaCl} = 5%				
O/A	0.5	1	1.5	2	2.5
AVG (*D*)	4.2357	5.1267	5.2106	5.0326	4.9869
SD	0.0725	0.0689	0.0612	0.0832	0.0654
AVG (*S*)	31.010	35.580	35.189	33.161	31.877
SD	0.0958	0.1032	0.0865	0.1052	0.0965

monitored with a pH meter. The follow-up experiments were performed using the simulant and the actual waste systems, respectively.

The simulation experiments of five-stage countercurrent extraction are shown in Fig. 4. The experiments were performed at room temperature and the phase ratio of organic and aqueous is 1. The aqueous phase was contacted with organic phase in a separatory funnel that was placed in a thermostatic oscillator for oscillating about 1 h, and then the separatory funnel was settled for completing

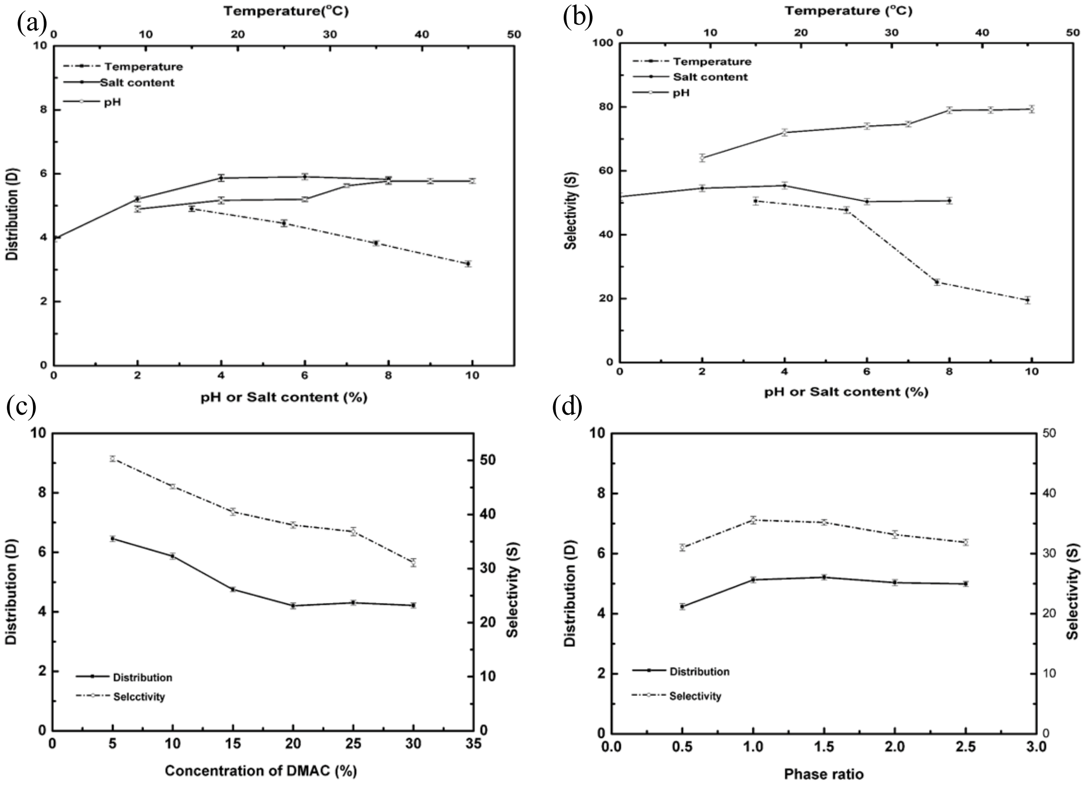

Fig. 1. The effects of temperature, pH value, salt content on distribution (a) and selectivity coefficient (b), the concentration of DMAC (c), and the phase ratio (d) on the distribution.

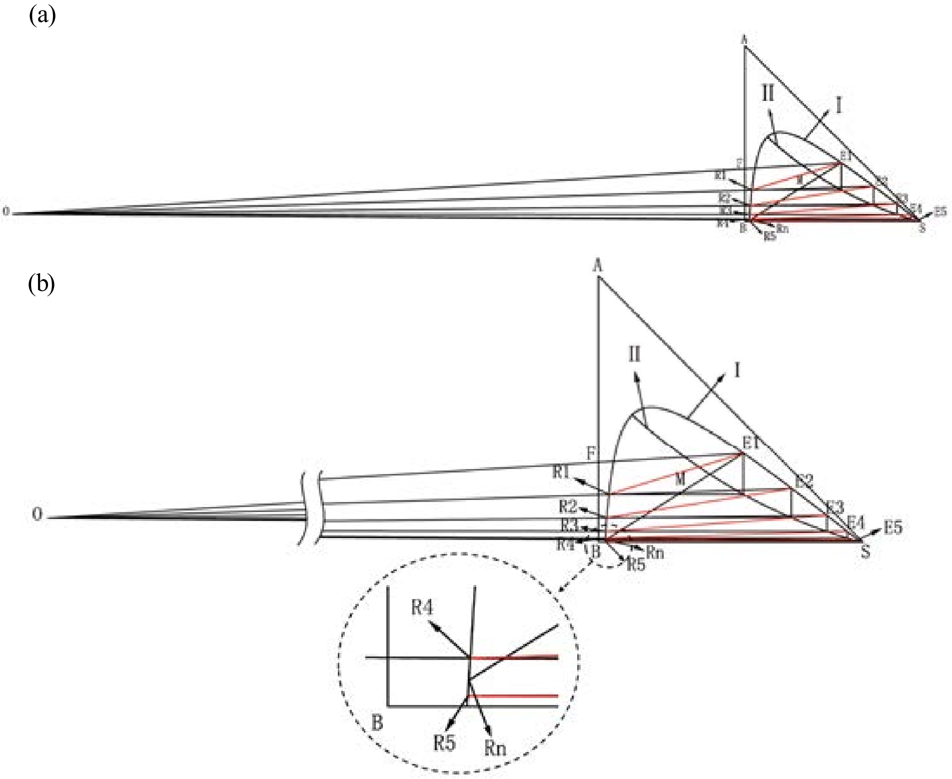

Fig. 2. The theoretical stages are diagrammed in triangular coordinate: (a) panoramagram; (b) partial enlarged detail (I = the solubility curve, II = the auxiliary curve).

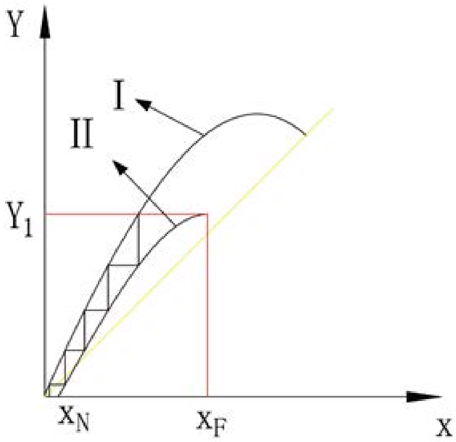

Fig. 3. The theoretical stages are diagrammed in rectangular coordinate (I = the solubility curve, II = the auxiliary curve).

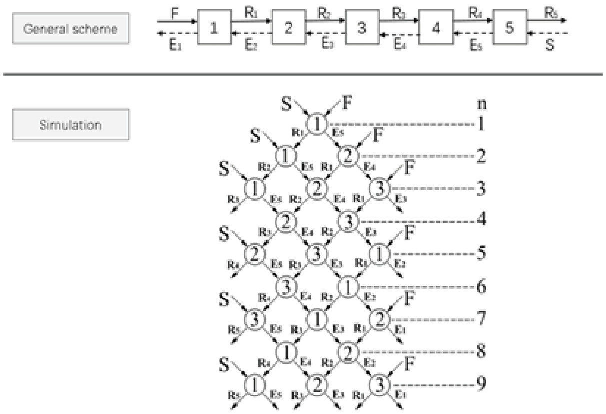

Fig. 4. The simulation of five-stage countercurrent extraction (*F* = wastewater, *S* = fresh solvent, *E* = extract phase, *R* = raffinate phase, *n* = row number, ①②③ = separatory funnel).

phase disengagement, settling time was about 1 h. The serial multistage countercurrent extraction experiments were carried out using three separatory funnels and the simulation was described as follow: three separatory funnels were labelled $1^{\#}$,$2^{\#}$, and $3^{\#}$, the wastewater (*F*) and the fresh extractant (*S*) were added into $1^{\#}$, and oscillated together for 1 h, this was the first row [15,16]. After settling, the aqueous phase of $1^{\#}$ was still stayed in $1^{\#}$ and the fresh extractant was added into it, the organic phase of $1^{\#}$ was transferred to $2^{\#}$ and added the wastewater, $1^{\#}$ and $2^{\#}$ were oscillated at the same time, this was the second row … and so on and so forth [17]. Continue to follow the directions shown in the arrows, it could not be ceased until the concentration of raffinate remained unchanged. The experimental data of each multistage countercurrent extraction are given in Table 4.

4. Results and discussion

4.1. Distribution and selectivity coefficient

The distribution coefficient (*D*) [18] and selectivity coefficient (*S*) [19] were calculated by the experimental data in order to evaluate the performance of the extractant to extract DMAC from the wastewater and the feasibility of the experimental scheme. The distribution and selectivity coefficients are defined as Eqs. (4) and (5), respectively, and the results are shown in Table 4.

$$D = \frac{x_1^1}{x_1^2} \tag{4}$$

$$S = \frac{x_1^1 x_2^2}{x_1^2 x_2^1} \tag{5}$$

where $x_1{}^1$ and $x_1{}^2$ are the mole fractions of DMAC in organic and in aqueous phases, respectively, x_2^1 and x_2^2 are the mole fractions of water in organic and in aqueous phases, respectively.

From the data of Table 4 it can be seen that the values of distribution and selectivity coefficient can increase with the deepening of extraction in most cases. However, the selectivity coefficient of the fifth stage is slightly reduced compared with the fourth. In large part it is because the increase of the concentration of DMAC in organic phase leads to the diminished efficiency of the mass transfer between the two phases. Nevertheless, all the data indicate that dichloromethane as extractant is satisfactory with the request of the separation.

Table 4
Distribution (*D*) and selectivity (*S*) coefficient of the five-stage countercurrent extraction for the {DMAC (1) + water (2) + dichloromethane (3)} system at room temperature[a]

Stage number	Organic phase(mass fraction)			Aqueous phase (mass fraction)			*D*	*S*
	w_1^1	w_2^1	w_3^1	w_1^2	w_2^2	w_3^2		
1	0.0720	0.0255	0.9025	0.0548	0.9418	0.0034	5.7752	51.8525
2	0.0422	0.0219	0.9359	0.0370	0.9601	0.0029	5.0093	51.9790
3	0.0272	0.0222	0.9506	0.0219	0.9767	0.0014	5.4447	56.0145
4	0.0182	0.0247	0.9571	0.0123	0.9864	0.0013	6.4261	60.0668
5	0.0111	0.0255	0.9634	0.0074	0.9893	0.0033	6.4690	58.7747
5[b]	0.0114	0.0265	0.9630	0.0078	0.9894	0.0028	6.3032	57.0649

[a] Standard uncertainty $u(w) = 0.0005$.
[b] The actual effluent.

4.2. Effects of single factor

It can be concluded from Figs. 1(a) and (b) that the distribution and selectivity coefficient exhibit basically the same trend by increasing the salt content (mass fraction %), pH value, and temperature. Distribution and selectivity coefficient all together increased slightly with the increase of pH value and decreased with the raise of temperature indicating that alkaline wastewater is favorable to the extraction of DMAC at room temperature. Distribution shows a modest growth with the increase of salt content, but selectivity rises slowly at the first and then begins to decline, which indicates that a moderate amount of salt is beneficial to the extraction, so 0.05 mass fraction of sodium chloride of the wastewater was chosen in this work. And the conclusion is in line with Wang and Liu [11].

Fig. 1(c) shows that the distribution and the selectivity coefficient are both minished with the DMAC concentration (mass fraction %) enhanced. However, the extraction can be completely carried out on account of the concentration of DMAC in wastewater no more than 30% and the selectivity greater than 1 in this range. The extraction develops best in the phase ratio of organic and aqueous is 1, which is revealed in Fig. 1(d).

Therefore, the effluent in this experiments is pretreated with salt (add 5% NaCl) and alkali (pH = 8) at room temperature and the phase ratio is 1.

4.3. Analysis and verification of extraction experiments

The extraction efficiencies (E) of the simulant and actual effluent were calculated to evaluate the feasibility of the five-stage countercurrent extraction scheme [20]. And the relative errors (δ) between the data of experiment (underlined in Table 2) and simulation (obtained by graphic method) were analyzed for the sake of confirming the credibility of the theory result. Their equations are as follows and the results are displayed in Table 5.

$$E\left(\%\right)=\frac{\left\{[\mathrm{DMAC}]_{\mathrm{aq0}}-[\mathrm{DMAC}]_{\mathrm{aq}}\right\}}{[\mathrm{DMAC}]_{\mathrm{aq0}}}\times 100\% \quad (6)$$

$$\delta=\frac{\left|x^{E}-x^{S}\right|}{x^{E}} \quad (7)$$

Table 5
The extraction efficiencies (E) and the relative errors (δ) data for the five-stage countercurrent extraction[a]

Stage number	Experimental values	E (%)	Theoretical values	δ
1	0.0548	–	0.0543	0.0091
2	0.0370	–	0.0364	0.0162
3	0.0219	–	0.0212	0.0320
4	0.0123	–	0.0116	0.0569
5	0.0074	97.53	0.0073	0.0135
5[b]	0.0078	97.40	0.0073	0.0641

[a] Standard uncertainty $u(w)$ = 0.0005.

where $[\mathrm{DMAC}]_{\mathrm{aq0}}$ and $[\mathrm{DMAC}]_{\mathrm{aq}}$ are the DMAC concentrations in the initial wastewater and in the aqueous phase, respectively. x^E and x^S are the experimental and theoretical mass fractions, respectively.

Table 5 reveals that the content of DMAC in the simulant effluent fell from 30% to 0.74% and dropped to 0.78% in the actual wastewater, reaching the separation target completely (1%). Furthermore, the extraction efficiencies of DMAC were 97.53% and 97.40%, respectively. In addition, the maximum relative error between the experimental and theoretical values is 0.0641, indicating that the theoretical stages calculated is very reliable.

In summary, DMAC can be effectively removed and recovered from the wastewaters using the multistage countercurrent extraction model. Moreover, the method has been practically applied to recover DMAC from the cefuroxime acid pharmaceutical wastewaters produced by Siping Fine Chemicals Co. Ltd. of Jilin Province (China) and the extraction efficiency is above 90%. The yield of recycled DMAC can reach about 800 tons per year. Therefore, the solvent extraction is suitable for recycling and processing refractory organic compounds of low concentration from pharmaceutical industry wastewaters and it will play an increasingly important role in the treatment of refractory organic effluent.

5. Conclusions

The optimum conditions of the simulation experiments were as follow: pH value of 8, the phase volume ratio (O/A) of 1:1, and addition of 0.05 mass fraction of sodium chloride into wastewaters at room temperature. The theoretical stages were calculated to be five by the graphical method in the triangular and the rectangular coordinate. The content of DMAC decreased from 30% to 0.74% for the simulant effluent and to 0.78% for the actual one, and thus the total extraction efficiency was 97.53% and 97.40%, respectively. In addition, the maximum relative error between the experimental and theoretical values is 0.0641. In this work, the secondary pollution, the main problem of solvent extraction, has been avoided by choosing dichloromethane which originally existed in the actual wastewaters as the extractant, but extraction efficiency will be reduced unless the DMAC content is not greater than 30%. Above all, the theoretical extraction stages have a high degree of credibility, and the extraction target has completed satisfactorily and efficiently by the five-stage countercurrent extraction.

Conflict of interest

The authors declare no competing financial interest.

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