Membrane Water Treatment, *Vol. 7, No. 4 (2016) 341-353*
DOI: http://dx.doi.org/10.12989/mwt.2016.7.4.341

Desalting of tobacco extract using electrodialysis

Shaolin Ge [1,2], Wei Li [3], Zhao Zhang [2], Chuanrun Li [1,3] and Yaoming Wang *[1]

[1] CAS Key Laboratory of Soft Matter Chemistry, Laboratory of Functional Membranes, School of Chemistry and Materials Science, University of Science and Technology of China, Hefei, Anhui 230026, People's Republic of China
[2] China Tobacco Anhui Industrial Co., LTD, Hefei, Anhui,230088, People's Republic of China
[3] Hefei ChemJoy Polymer Materials, Co., LTD, Hefei, Anhui,230601, People's Republic of China

(*Received August 13, 2015, Revised March 23, 2016, Accepted April 11, 2016*)

Abstract. Papermaking reconstituted tobacco is an important strategy for recycling the waste tobacco residues. To indentify the influences of the inorganic components on harmful components delivery in cigarette smoke, a self-made electrodialysis stack was assembled to desalt the tobacco extract. The influences of the applied current and extract content on the removal rate of the inorganic ions were investigated. Results indicated that the applied current was a dominant impact on the desalination performance. High currents lower than the limiting current density could accelerate the desalting efficiency but cause higher energy consumption. A current of 2 A, or current density of ~11 $mA \cdot cm^{-2}$, was an optimal choice by considering both the energy consumption and desalting efficiency. A 20% tobacco extract was an appropriate content for the electrodialysis process. More than 90% of inorganic ions could be removed under the optimum condition. The preliminary result indicated that removal of inorganic components was beneficial to decrease the harmful component delivery in cigarette smoke. Naturally, ED is an environmentally friendly and high-effective technology for desalting the tobacco extract.

Keywords: reconstituted tobacco; tobacco extract; electrodialysis; ion exchange membranes

1. Introduction

Nowadays, reconstituted tobacco, or known as tobacco sheet, has played an increasingly significant role in tobacco industry. The sheet tobacco manufacturing process includes the extraction, pretreatment, fabrication, cutting, and cigarettes wrapping etc. Papermaking reconstituted tobacco is the most prevalent method for tobacco sheet, which is made by taking tobacco by-product materials and putting them through a process of repetitive hot water extraction to obtain the tobacco extracts in an aqueous medium. Papermaking tobacco sheet has inherent superiority in structural strength, combustion performance, tar delivery, and waste recycling (Wang *et al*. 2005). The tobacco stems, tobacco dust, mid-ribs, leaf scraps and other waste tobacco residues can be recycled as raw material for cigarettes and the chemical, physiological and biological properties of cigarette can be manually manipulated with the addition of external chemicals and additives. For these reasons, papermaking tobacco sheet is considered as the one of the most paramount strategies to improve the quality of cigarette without the loss of flavor

*Corresponding author, Ph.D., E-mail: ymwong@ustc.edu.cn

http://www.techno-press.org/?journal=mwt&subpage=7 ISSN: 2005-8624 (Print), 2092-7037 (Online)

components.

There are thousands matters in the tobacco extract including sugars, amino acids, nicotine, proteins, inorganic ions such as potassium, sodium, chloride, nitrate, sulfate, and etc. Numerous studies have indicated that the inorganic ions have significant influence on the sensory taste of the cigarette and the harmful component delivery in cigarette smoke. It is often believed that chloride ions can affect the moisture absorptivity and combustibility of the flue-cured tobacco (Müller *et al*. 2011, Zarrelli *et al*. 2012). Previous research has indicated that cigarette has a better combustion performance when the chloride ions concentration in tobacco sheet is lower than 1.0wt%; while the burning cigarette will self-extinguish when the chloride ions concentration in tobacco sheet is higher than 2.0wt% (Johnson *et al*. 2009). Potassium ions are conducive to the combustion behavior of cigarette. The ammonium salt is a potential burn additive to reduce carbonyl compounds delivery in the cigarette mainstream smoke (Chen *et al*. 2014). The nitrate and nitrite ions can react with alkaloids during the combustion processes to generate the carcinogen tobacco-specific nitrosamines (TNSAs), so these ions are also considered as the precursors of TNSAs (Seyler *et al*. 2013, Narkowicz *et al*. 2013, Nunes-Alves *et al*. 2013). Nunes-Alves and co-authors have found that nitrate ions are close related with NO_X release in the mainstream tobacco smoke (Nunes-Alves *et al*. 2013). The removal of nitrate ions will increase the puff number, leading to an increase in the total particulate matter (TPM), nicotine and carbon monoxide delivery in the smoke (Flischer *et al*. 1989). Chen and co-authors have found that the ammonium salt was a potential burn additive to reduce carbonyl compounds delivery in the cigarette mainstream smoke of reconstituted tobacco (Chen *et al*. 2014).Therefore, there is no doubt to conclude that the inorganic salts in the tobacco sheet have significant impact on the thermal degradation and combustion properties of restituted tobacco definitely. In most case, the majorities of inorganic salts have negative impact on the quality of cigarette. However, more detailed information about the correlation between inorganic salts and harmful carbonyl compounds release in the mainstream smoke is still lacking in the literature. Moreover, the influences of particular salts on harmful carbonyl compound delivery in the most previous studies were conducted by additionally adding extra corresponding salt in the tobacco extract. In this case, it is not scientific enough since these salts levels may have exceeded the threshold limit for a performance metrics. To investigate the influence of a particular ion on the cigarette, it is more desirable to vary the concentration of the corresponding ion while keep the other ions at their original levels. Therefore, the demineration of the tobacco extract is beneficial not only to improve the sensory quality of smoke but also to decrease the harmful carbonyl compounds delivery in tobacco smoke.

A number of methods have been developed to desalt the inorganic salts contents in the tobacco extract. The multistage continuous countercurrent extraction was developed for the reduction of nitrate in reconstituted tobacco by Uhl. R in B&W Company in 1977 (Uhl 1977). The process was capable of a 90% reduction of nitrate content but all the soluble extracts were discarded. The Philip-morris company developed the combined extraction, condensation and frozen centrifugation method for the removal of nitrate in the reconstituted tobacco (Jeanneret and Morris 1981). More than 50% of nitrate could be removed and the reduction rate could be further enhanced with further microbial fermentation process. Gaisch and co-authors have employed the microorganisms for reducing the nitrate, nitrite, and ammonium compounds content of an aqueous tobacco extract (Gaisch *et al*. 1986). The nitrate, nitrite, and ammonium compounds were eliminated via an aerobic assimilatory metabolic pathway. But a large amount of nutrients such as phosphate and carbons sources were consumed for microbial growth. Mattina and Selke have investigated ion exchange resin for the removal of potassium nitrate from the tobacco extract (Mattina and Selke

1973). The ionic material and specifically potassium nitrate, is selectively retarded in these ion retardation resins. But a large amount of acid and base were needed for the resin regeneration and large amount of wastewater was discharged. The absorption method was also applied by Baker and co-authors to absorb the TNSAs and Hoffmann analytes contents (Baker *et al*. 2004). In 2009, Verma *et al*. patented a process for the manufacture of reconstituted tobacco sheets with much reduced percentage of nicotine, sugar and chloride using un-conventional raw materials derived from jute and/or bamboo for papermaking (Verma *et al*. 2009). But these desalting technologies usually present some limitations mainly related to low-efficiency, high consumption of chemicals, loss of desirable constituents of the tobacco extract, as well as secondary pollution to the environment. Therefore, it is necessary to explore some environmentally friendly and high-effective technologies for desalting the tobacco extract.

Electrodialysis (ED) is a mature technology with multitude of new applications in chemical cleaner production, environmental protection and energy conversion (Huang and Xu 2008, Strathmann 2011, Tanaka *et al*. 2012, Aghajanyan *et al*. 2013, Ali and Hamrouni 2016, Majewska-Nowak 2013). As opposed to other separation techniques, electrodialysis (ED) does not suffer from major drawbacks, such as generation of large amounts of waste, use of hazardous solvents, short lifetimes of adsorbents, etc. ED is thus considered an environmentally friendly and sustainable technology and may be a very competitive one for tobacco industry. In our previous study, a preliminary experiment was conducted to test the feasibility of ED for the removal of chloride and nitrate ions (Zhang *et al*. 2014). In this study, more detailed results of the ED process for the removal of most inorganic salts will be presented and the influences of demineration of tobacco extract on the delivery of the mainstream smoke will also be proposed.

2. Experimental

2.1 Materials

Concentrated tobacco extract (total solid content ~40%) used in this experiment was supplied by China Tobacco Anhui Industrial CO., Ltd, Hefei, Anhui Province, China. This tobacco extract was pretreated by a centrifugation at 3000rpm for 5min to remove the suspended solids before the ED process. The membranes used for the electrodialysis (ED) experiments were CJ-MA-2 (Hefei ChemJoy Polymers Co., Ltd, China) and CJ-MC-2 (Hefei ChemJoy Polymers Co., Ltd, China), and their properties are listed in Table 1. All the chemicals were of analytical grade.

2.2 ED set-up

A schematic diagram of ED set-up is illustrated in Fig. 1. The ED set-up was composed of (1) a cathode and an anode were respective fixed on two organic glasses plate each with a rectangular notch; the electrodes were made of Iridium-Tantalum with a thickness of 1.5 mm. The electrodes were fixed into the notch with epoxy glue to make an even surface on the organic glasses. A direct current power supply (WYL1703, Hangzhou Siling Electrical Instrument Ltd.) was connected on the electrodes. The voltage drop across the stack was directly read from indicators on the power supply. (2) Eleven pieces of cation exchange membranes and ten pieces anion exchange membranes, which are alternatively arranged. The effective area of each membrane was 189 cm^2. (3) Sealing spacers made from PE with a thickness of 0.75 mm was used to separate the membranes; (4) Beakers to store the feed. Each beaker was connected with a submersible pump

Table 1 The main characteristics of the membranes used in the experiments

Properties	CJ-MC-2	CJ-MA-2
Thickness /mm	0.200	0.145
Ion exchange capacity /mmol/g	1.50	1.25
Water Uptake /%	35	32
Resistance /$\Omega\cdot cm^2$	1.5	1.2
Transfer number /%	98	99
Break stress /MPa	>3.5	>3.5

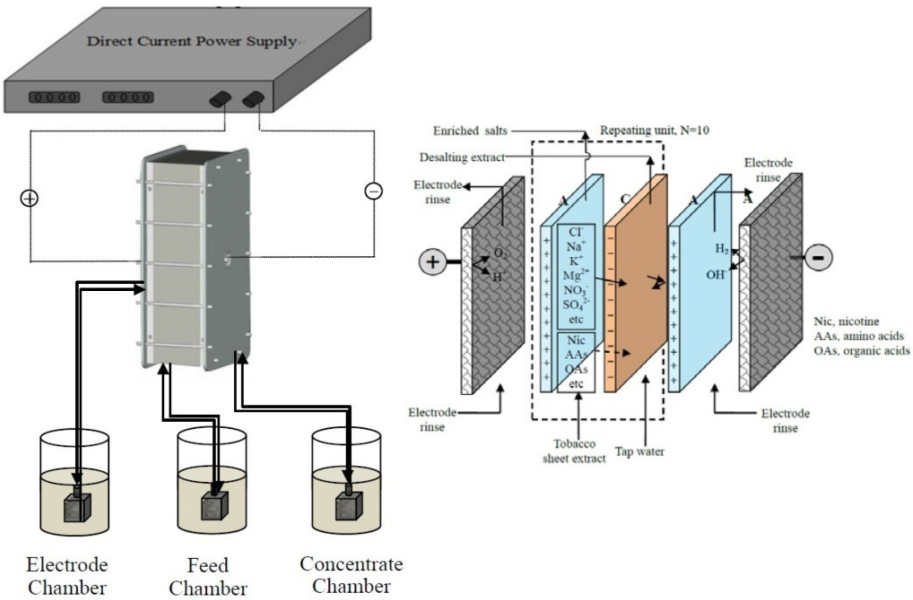


Fig. 1 Schematic diagram for the experiment stack and the main principle of the electrodialysis process

(AP1000, Zhongshan Zhenghua Electronics Co. Ltd., China, flow rate of 22 L/h) to form a circulating loop. In our experiments, three circulating loops, namely, electrode chamber, concentrate chamber, and diluted chamber were established in this ED stack. A 400 mL Na_2SO_4 solution (0.3 mol/L) was fed into the electrode chambers as the rinse electrolyte. A 400 mLtobacco extract was fed into the dilute chambers. A 400 mL tap water was fed in the concentrate chamber. Before the experiment, each chamber was circulated for 30 min to eliminate the visible bubbles. All the experiment runs were kept at room temperature.

2.3 Current-Voltage (I-V) curves

A self-made experimental set-up was used to measure the current-voltage curves (Xu *et al*. 2006). The effective area of membrane in the stack is 7.07cm^2. In this stack, a 0.3 mol/L, 500 mL Na_2SO_4 solution was used as electrolyte rinse solution. Different solid contents of tobacco extract were fed in the concentrate and dilute chamber. The current was supplied by a programmable DC power system (PPE-3323, Good Will Instrument Co., Ltd., Taiwan) and applied to the cell by two ruthenium-coated titanium electrodes. The voltage drop was measured using a voltmeter (ZW1418, Qingzhi Instruments Co., Ltd., China) by two platinum filament located approximately 1.0 mm away from each membrane surface. The membrane sample was inserted between two half-cells with two platinum filament spaced near both sides of the membrane.

2.4 Energy consumption

The energy consumption (E, $kWh \cdot L^{-1}$) of the ED process was calculated as Eq. (1)

$$E = \int_0^t \frac{U_t I_t dt}{C_t VM} \tag{1}$$

U_t (V) is the voltage drop across ED stack at t time; I (A) is the current applied at t time; V is the volume of feed compartment; Removal rate of a particular i ions (R_i) was calculated as Eq. (2)

$$R_i = \frac{C_{i,0}}{C_{i,t}} \tag{2}$$

$C_{i,0}$ and $C_{i,t}$ are the concentration of i ions at time 0 and t, respectively.

2.5 Analytical methods

The conductivity of the feed solution (dilute chamber) was monitored by a conductivity meter (DDS 307, Shanghai INESA Scientific Instrument Co. Ltd., China). The concentrations of inorganic ions were determined by Ion Chromatography (ICS3000 multifunctional ion chromatography and ED electrochemical detector, DIONEX Company, USA). To determine the yields of CO, HCN, NH_3, phenol, crotonaldehyde and total particulate matter (TPM) in cigarette mainstream smoke, cigarettes made from PRT were smoked by a smoking machine (Borgwaldt RM200A), according to the selected smoking conditions (35 mL puff volume, 2s duration, 1 puff per min) of International Standard (ISO) recommendations (ISO 4387, 1991) The harmful components in cigarette mainstream smoke were determined according to previous study (Zhou *et al*. 2014).

3. Results and discussion

3.1 Limiting current density

The applied current is the most prominent factor affecting the performance of electrodialysis process. A high current can save the capital cost of the ED project, but high current density will reduce the lifespan of membranes. Based on the industrial experience of concentration polarization, no currents larger than the limiting current density can be expected in an electrodialysis process (Krol *et al*. 1999). It is recommended that the current density should not be higher than 90% of the limiting current density. Fig. 2 shows the current-voltage curves of the ED stack for different contents of tobacco extracts. Table 2 shows the limiting current densities of ED stacks for different contents which are indicated from the plateau values of the I-V curves in Fig. 2. It is clearly indicated that the limiting current density is decreased with a decrease in the contents of tobacco

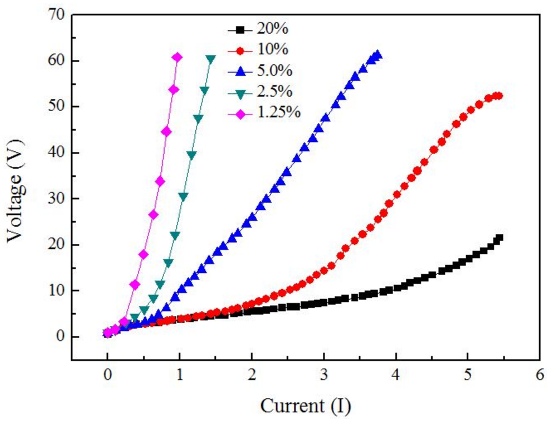


Fig. 2 The I (current)-V (voltage) curves of the ED stacks for different contents of tobacco extract

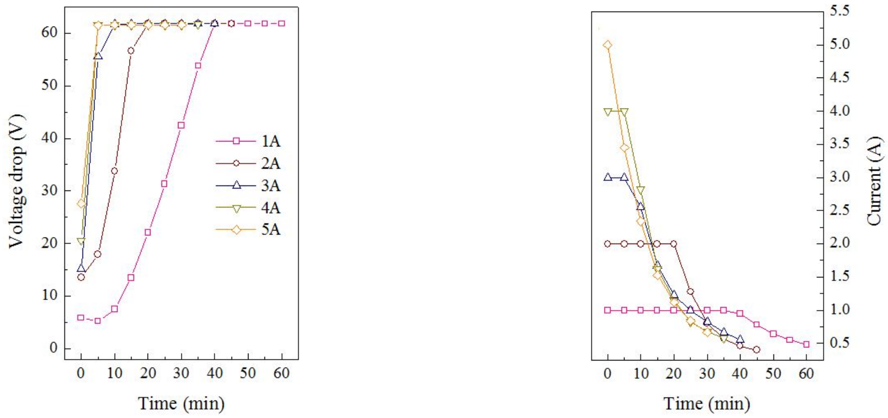
Table 2 The limiting current of ED stacks for different contents of tobacco extract

Contents of tobacco extract /(w/w)	20%	10%	5%	2.5%	1.25%
Limiting current /(A)	4.10	3.08	1.00	0.80	0.36
Limiting current density /(mA/cm^2)	21.69	16.30	5.29	4.23	1.90

extract. This is consistence with the literature since the limiting current density depends on the solution concentration (Lee *et al*. 2006). On the diluate side the salt concentration decreases with respect to the bulk concentration whereas it increases on the concentrate side of the membrane. It should point out that electrodialysis in our experiment is operated at a constant-current mode to give a steady desalination. In this case, the limiting current density of the electrodialysis process will gradually decrease with a continuous desalting of electrolytes in the dilute compartment. This suggests the operating current must be carefully monitored to below the values of limiting currents.

3.2 The effect of current density

As discussed before, the applied current density in ED process affects the current utilization and the concentration polarization as well as the cost of the process. Fig. 3 shows the effect of the initial current on the voltage and current drops of the electrodialysis process when the tobacco extract content is 20%. It is clearly indicated that voltage drops increase with the elapse of time. This suggests the inorganic ions in the dilute chamber are depleted under the electric field in the experiments. Meantime, the higher the current is, the higher the initial voltage will be. This is consistence with the Ohm's law. It can be seen from Fig. 3(b) that the operation mode shifts from a current-constant mode to a voltage-constant mode considering the maximum value is 60 V for the current supplier. The lower currents requires more time for the experiments. This is because the driven force for the electrodialysis is a direct current field, which is proportional to the current supplied in the stack. Fig. 4 shows the conductivity changes in the dilute chamber under different initial current when the tobacco extract content is 20%. It is indicated that the conductivities in the dilute chamber are declined with the elapse of time. Because conductivity is the most direct evidence about the ionic strength in a solution, the decrease of conductivity means the desalination of salts. The conductivities in the tobacco extract decrease from the initial 22.4 mS/cm to the final



(a) (b)

Fig. 3 Effect of initial current on the voltage and current drops in ED stack

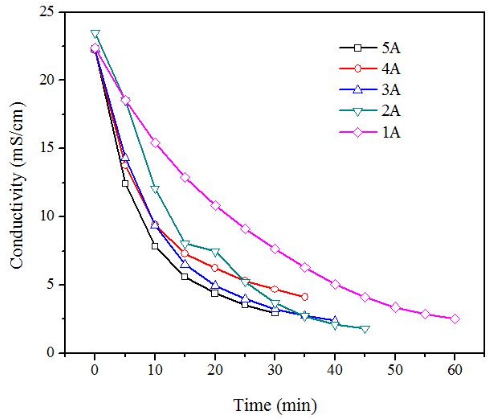


Fig. 4 The conductivities changes in the dilute chamber under different initial current

2-3 mS/cm, indicating the majorities of ions are removed by an electrodialysis process. Meantime, the high the current, the more swift the conductivity decreases. The conductivity gradient at 3-5 A are much higher than that at 1-2 A. This is because of a higher driven force for a higher applied current.

Fig. 5 shows the effect of the initial current on the ion removal rate when the tobacco extract content is 20%. It is clearly indicated that the majority of investigated ions are successfully removed by the electrodialysis process. More than 90% of chloride and nitrate ions and 85% of divalent ions are removed. There is no significant difference for the ions removal ratio under the investigated current. Generally, the removal ratio of divalent ions is slightly lower than that of the monovalent ions. The possible reason is that the monovalent ions have less hydration radium and little charge compared with the divalent ions. Fig. 6 shows the effect of initial current on the energy consumption of the electrodialysis process. It can be seen that the energy consumption at low currents is lower than the higher currents. This suggested that low current densities have higher current utilization rate, this is consistent with the results in the literature. Unfortunately, it is

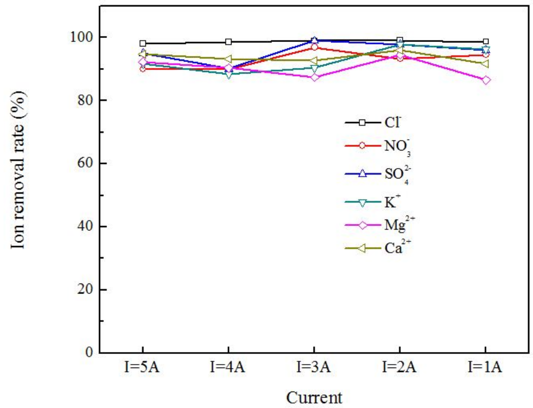


Fig. 5 The removal rate of ions for different initial current

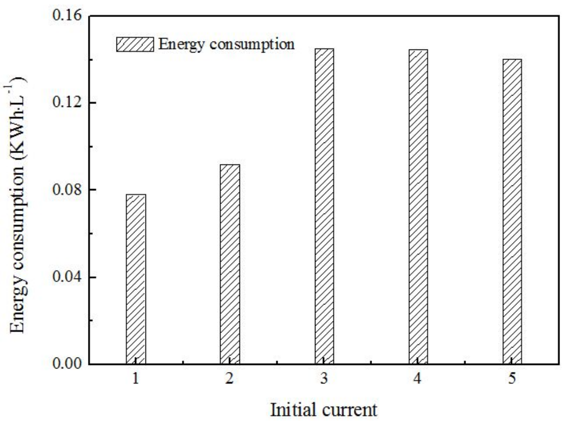


Fig. 6 The energy consumption of the electrodialysis process under different initial current

difficult to calculate the current efficiency due to the complex of ions in the tobacco extract. In fact, there is often a trade-off to be made between high and low current density during the practical applications of the ED process. Higher current densities are not attractive due to high energy costs related to increased ohmic drop and lower current densities are not attractive due to the large required membrane area. The current of 2 A is an optional chosen by considering both the energy consumption and desalting efficiency.

3.3 Effect of tobacco extract content

Due to the complex of the tobacco extract, the contents of extract have a significant effect on the desalination performance of the electrodialysis process. The casting extract usually has a solid content of 40% solution in the practical papermaking reconstituted tobacco process. But high solid contents lead to poor flowability and can easily cause fouling of the membranes considering the narrow flow passage in the ED stack. Therefore, an appropriate content of the tobacco extract must be chosen to improve the performance of the electrodialysis process. Fig. 7 presents the voltage and current changes in the ED stack under different tobacco extract. It indicates that the experiment time is incremented by an increase in the contents of the extract solution. This is a logically truth since the higher the content of tobacco extract, the higher the electrolytes ions will be. Fig. 8 shows the decrease of solution conductivity with the elapse of time. This is consistent with the voltage drop curves. It can be seen that a high desalination rate is achieved for tobacco sheet content lower than 20%. A tobacco extract content of 40% is not suitable for the electrodialysis due to poor flowability of solution. Fig. 9 shows the removal rate of different ions for different contents of tobacco extract. It can be seen that a higher removal rate is achieve for the investigated ions when the tobacco sheet content is lower than 20%. For tobacco extract content of 40%, the removal ratios of monovalent ions are as high as 92%, but the removal ratios of divalent ions are not acceptable. A possible reason for this is that the divalent ions are coagulated with some organic components at an elevated temperature during the concentrate process. Because the conventional concentrate process is usually achieved by multiple-effect evaporator, the tobacco extract components can easily degraded at a high temperature. Another possible reason is that

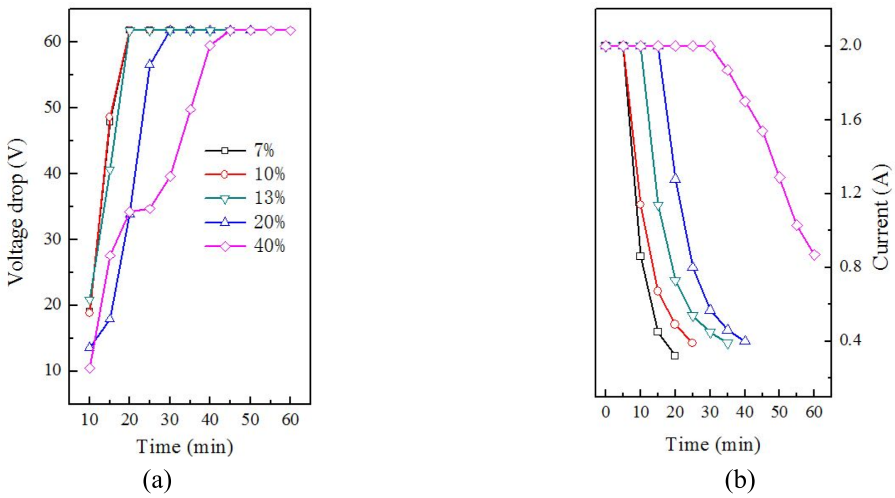


Fig. 7 Effect of tobacco extract content on the voltage and current drops in ED stack

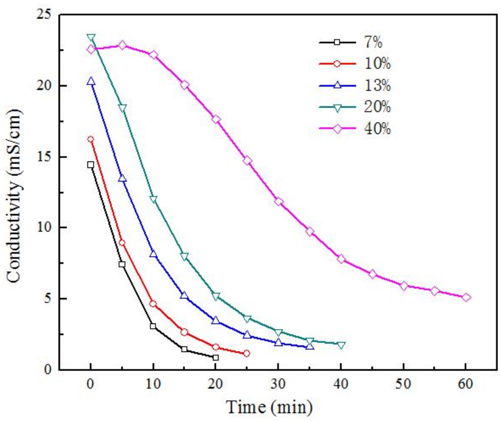


Fig. 8 The conductivities changes in the dilute chamber under different contents of tobacco extract

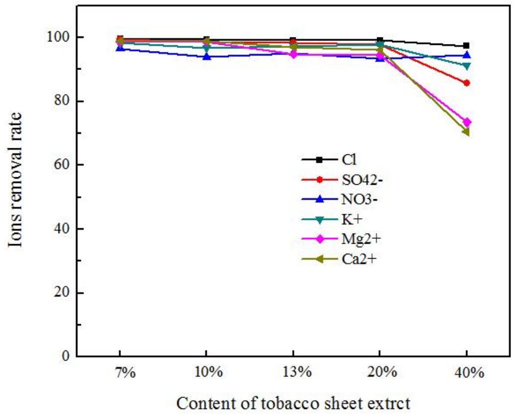


Fig. 9 The removal rate of ions for different contents of tobacco extract

a high concentration of multivalent metals may accelerate the fouling of membranes. The fouling of the electro-membranes for the desalting of tobacco extract will be discussed in our further study. Moreover, it is not economical for ED operating under too much low levels of extract even with a satisfactory desalination rate. Because additional energy consumption will be consumed for enriching the low levels of tobacco extract. A 20% tobacco extract is an appropriate content by considering both the desalination efficiency and treatment capacity.

3.4 The influence of desalting tobacco extract on the delivery of harmful mainstream tobacco smoke

According to health concerns, the low molecular weight aidehydes and ketones, such as formaldehyde, acetaldehyde, acetone, propanal, acrolein etc., have been classified as toxic and carcinogenic constituents (Zhou *et al*. 2011).Considerable efforts were devoted to selectively reduce these harmful components, commonly indexed by the ratio of special component content to total particulate matter (TPM). These volatile carbonyl compounds have been reported to be found in low levels in tobacco (Stedman 1968) and mainly generated from the pyrolysis of carbohydrate, including sugar, starch and cellulose etc., during cigarette combustion (Seeman *et al*. 2002).The inorganic components affect the cigarette mainstream smoke chemistry by impacting the combustion process. Fig. 10 shows the influence of desalting of tobacco extract on the selective reduction of five carbonyl compounds in the main stream smoke of cigarette. It indicates that the removal of inorganic ions lead to a considerable decrease of relative delivery of ammonia and phenol, whereas ED treatment of tobacco extract has no significant influence on crotonaldehydes delivery. The different effect of demineration of tobacco extract on aldehydes and ketones may be attributed to the difference of their formation pathways (Paine III *et al*. 2008), which suggests inorganic ions influence carbohydrate pyrolysis mechanisms. The preliminary results indicate that the removal of inorganic components is beneficial to selective decrease the harmful carbonyl components in cigarette smoke. Moreover, the smoking quality is evaluated by sensory test, which usually includes several indexes such as fragrance, strength, concentration, harmony, miscellaneous gas, irritation and remaining taste. Sensory evaluation results showed that the cigarette made from desalting tobacco sheet extract has satisfy tobacco fragrance, improved strength and concentration, and less miscellaneous gas emission. But more detailed experiments are needed to indentify the

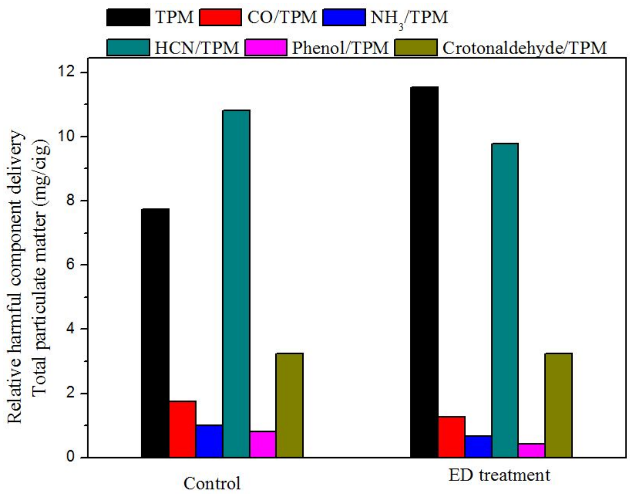


Fig. 10 The desalting of tobacco extract using ED on the delivery of harmful components in the tobacco smoke

influence of the individual ion on the harmful components delivery in the tobacco smoke. This can be achieved by a simulated back-addition of individual ions to their original levels in the tobacco extract. Nevertheless, the electrodialysis is a feasible platform technology to illustrate the influence of inorganic ions on the tobacco smoke.

4. Conclusions

Laboratory-scale experimental set-up was used to desalting of tobacco extract by using the electrodialysis process. Results indicated that the applied current was a dominant impact on the desalination performance. High currents lower than the limiting current density could accelerate the desalting efficiency but cause higher energy consumption. Low currents have high current utilization but require higher capital cost. A current of 2 A, or current density of ~11mA·cm^{-2}, is an optional choice by considering both the energy consumption and desalting efficiency. For tobacco extract at a 40% content, the removal ratios of monovalent ions are as high as 92% but are not acceptable for divalent ions. A 20% tobacco extract is an appropriate content by considering both the desalination efficiency and treatment capacity. The tobacco smoke result indicates that the removal of inorganic ions lead to a slight decrease of aldehydes in the total particulate matter (TPM). But the relative contents of two ketones delivery are considerablely decreased compared with the control cigarette. The ED technology is a potential technology not only to improve the smoking quality of cigarette but also to selectively decrease the harmful component delivery in cigarette smoke. But it requires the interdisciplinary collaboration between academia and industry from both the membranes science and tobacco industry to identify the detailed relationship between the inorganic components and cigarette smoke delivery.

Acknowledgments

This research is supported in part by the National Natural Science Foundation of China (Nos.

21206154, 21476220), and Anhui Key Laboratory of Tobacco Chemistry (No. 2014104).

References

Aghajanyan, A.E., Tsaturyan, A.O., Hambardzumyan, A.A. and Saghyan, A.S. (2013), "Obtaining the zwitterionic form of L-lysine from L-lysine monohydrochloride by electrodialysis", *Membr. Water Treat., Int. J.*, **4**(1), 1-9.

Ali, M.B.S. and Hamrouni, B. (2016), "Development of a predictive model of the limiting current density of an electrodialysis process using response surface methodology", *Membr. Water Treat., Int. J.*, **7**(2), 127-141.

Baker, R.R., da Silva, J.R.P. and Smith, G. (2004), "The effect of tobacco ingredients on smoke chemistry. Part I:Flavourings and additives", *Food Chem. Toxicol.*, **42**(S), 3-37.

BAT Technical Memorandum, 79-09-063, 18.9.79.

Burley stem washing (2004), November 04. Philip Morris. URL: http://industrydocuments.library.ucsf.edu/tobacco/docs/jtfx0151

Burley Stem Washing, URL: http://legacy.library.ucsf.edu/tid/vaz94g00/pdf

Chen, M.S., Xu, L., Tian, H.L., Yin, C.Y., Zhou, Z.L., Li, Y., Ma, J.G. and Zhong, F. (2014), "Effects of common ammonium salt on the thermal behavior of reconstituted tobacco sheet", *J. Therm. Anal. Calorim.*, **118**(3), 1747-1753.

Denitration of Venezuelan Burley Stem Part II: The Effect on Smoke Deliveries. URL: http://www.legacy.library.ucsf.edu/tid/zna11a99/pdf

Flischer, S., Spiegehalder, B. and Pressumann, R. (1989), "Performed tobacco-specific nitrosamines in tobacco-role of nitrate and influence of tobacco type", *Carcinogenesis*, **10**(8), 1511-1517.

Gaisch, H., Krasna, B. and Schulthess, D. (1986), "Continuous method of denitrating tobacco extracts", US Patent 4622982.

Huang, C.H. and Xu, T.W. (2008), "Electrodialysis-based separation technologies: acritical review", *AICHE J.*, **54**(12), 3147-3159.

ISO 4387 (1991), Cigarettes—determination of total and nicotine free dry particulate matter using a routine analytical smoking machine; Reference number ISO 4378:1991 (E), International Organization for Standardization: Geneva, Switzerland.

Jeanneret, C. and Morris, P. (1981), "Legislation 800124800224". URL: http://industrydocuments.library.ucsf.edu/tobacco/docs/klxk0000

Johnson, M.D., Schilz, J., Djordjevic, M.V., Rice, J.R. and Shields, P.G. (2009), "Evaluation of in vitro assays for assessing the toxicity of cigarette smoke and smokeless tobacco", *Cancer. Epidemiol. Biomarkers Prev.*, **18**(12), 3263-3304.

Krol, J.J., Wessling, M. and Strathmann, H. (1999), "Concentration polarization with monopolar ion exchange membranes: current-voltage curves and water dissociation", *J. Membr. Sci.*, **162**(1-2), 145-154.

Lee, H.J., Strathmann, H. and Moon, S.H. (2006), "Determination of the limiting current density in electrodialysis desalination as an empirical function of linear velocity", *Desalination*, **190**(1-3), 43-50.

Majewska-Nowak, K.M. (2013), "Treatment of organic dye solutions by electrodialysis", *Membr. Water Treat., Int. J.*, **4**(3), 203-214.

Mattina, C.F. and Selke, A.S. (1973), "Method of making reconstituted tobacco having reduced nitrates", US Patent 3847164.

Müller, A.L.H., Bizzi, C.A., Pereira, J.S.F., Mesko, M.F., Moraes, D.P., Flores, E.M.M. and Muller, E.I. (2011), "Bromine and chlorine determination in cigarette tobacco using microwave-induced combustionand inductively coupled plasma optical emission spectrometry", *J. Braz. Chem. Soc.*, **22**(9), 1649-1655.

Narkowicz, S., Polkowska, Z., Kielbratowska, B. and Namiesnik, J. (2013), "Environmental tobaccosmoke: Exposure, health effects, and analysis", *Crit. Rev. Environ. Sci. Technol.*, **43**(2), 121-161.

Nunes-Alves, A., Nery, A.A. and Ulrich, H. (2013), "Tobacco nitrosamine N-nitrosonornicotine as inhibitor of neuronal nicotinic acetylcholine receptors", *J. Mol. Neurosci.*, **49**, 52-61.

Paine III, J.B., Pithawalla, Y.B. and Naworal, J.D. (2008), "Carbohydrate pyrolysis mechanisms from isotopic labeling: Part 3. The Pyrolysis of d-glucose: Formation of C3 and C4 carbonyl compounds and a cyclopentenedione isomer by electrocyclic fragmentation mechanisms", *J. Anal. Appl. Pyrol.*, **82**(1), 42-69.

Seeman, J.L., Dixon, M. and Haussmann, H.J. (2002), "Acetaldehyde in mainstream tobacco smoke: formation and occurrence in smoke and bioavailability in the smoker", *Chem. Res. Toxicol.*, **15**(11), 1331–1350.

Seyler, T.H., Kim, J.G., Hodgson, J.A., Cowan, E.A., Blount, B.C. and Wang, L. (2013), "Quantitation ofurinary volatile nitrosamines from exposure to tobacco smoke", *J. Anal. Toxicol.*, **37**(4), 195-202.

Strathmann, H. (2011), "Membrane separation process: Current relevance and future opportunities", *AICHE J.*, **47**(5), 1077-1087.

Stedman, R.L. (1968), "The chemical composition of tobacco and tobacco smoke", *Chem. Rev.*, **68**(2), 53-207.

Tanaka, Y., Uchino, H. and Murakami, M. (2012), "Continuous ion-exchange membrane electrodialysis of mother liquid discharged from a salt-manufacturing plant and transport of Cl^- ions and SO_4^{2-} ions", *Membr.Water Treat., Int. J.*, **3**(1), 63-76.

Uhl (1977), RG RL Denitration separete burley processing.
URL: http://legacy.library.ucsf.edutid/kzm02a00/pdf

Verma, M.S., Lakshmanan, C.C. and Chaudhury, A.S. (2009), "A process for the manufacture of reconstituted tobacco sheet", Indian Patent 194046.

Wang, W.S., Wang, Y. and Yang, L.J. (2005), "Studies on thermal behavior of reconstituted tobacco sheet", *Thermochim. Acta*, **437**(1-2), 7-11.

Xu, T.W., Fu, R.Q., Yang, W.H. and Xue, Y.H. (2006), "Fundamental studies on a novel series of bipolar membranes prepared from poly (2,6-dimethyl-1,4-phenylene oxide) (PPO) - II. Effect of functional group type of anion-exchange layers on I-V curves of bipolar membranes", *J. Membr. Sci.*, **279**(1-2), 282-290.

Zarrelli, A., DellaGreca, M., Parolisi, A., Lesce, M.R., Cermola, F., Temussi, F., Isidori, M., Lavorgna, M., Passananti, M. and Previtera, L. (2012), "Chemical fate and genotoxic risk associated with hypochlorite-treatment of nicotine", *Sci. Total Environ.*, **426**(1), 132-138.

Zhang, Z.H., Ge, S.L., Jiang, C.X., Zhao, Y. and Wang, Y.M. (2004), "Improving the smoking quality of papermaking tobacco sheetextract by using electrodialysis", *Membr. Water. Treat., Int. J.*, **5**(1), 31-40.

Zhou, S., Wang, C., Xu, Y. and Hu, Y. (2011), "The pyrolysis of cigarette paperunder the conditions that simulate cigarette smouldering andpuffing", *J. Therm. Anal. Calorim.*, **104**(3), 1097-1106.

Zhou, S., Ning, M., Zhang, Y.P., He, Q., Wang, X.F., Zhu, D.L., Guo, S., Hong, N.N. and Hu, Y. (2014), "Significant removal of harmful compounds in mainstream cigarette smoke using carbon nanotubes mixture prepared by catalytic pyrolysis", *Adsorpt. Sci. Technol.*, **32**(6), 453-464.

CC