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Simultaneous removal of SO_2 and NO using M/$NaClO_2$ complex absorbent[☆]

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ABSTRACT

A complex absorbent containing NaClO (M) and $NaClO_2$ was used to investigate simultaneous removal of SO_2 and NO from flue gas. The various influencing factors, such as molar ratio of M to $NaClO_2$, solution pH, reaction temperature, SO_2 concentration and NO concentration on removal efficiencies of SO_2 and NO were studied experimentally, and the optimal conditions were established, in which molar ratio of M to $NaClO_2$ was 4.0, solution pH was 5.5, reaction temperature was 50 °C. Under the optimal conditions, the removal efficiencies of SO_2 and NO reached 100% and 85%, respectively. In addition, the reaction mechanism of simultaneous desulfurization and denitration using complex absorbent was proposed.

1. Introduction

China has become one of the countries which have most serious pollution problems of sulfur dioxide (SO_2) and nitrogen oxides (NOx) in the recent years [1,2]. SO_2 as the precursor of acid rain, and NOx as the precursors of acid rain and photochemical smog, are important air pollutants because of serious harm for human health and ecological environment. In general, the removal of SO_2 and NOx from coal-fired flue gas is carried out by installing the flue gas desulfurization and denitration equipments separately at the rear of coal-fired boiler. But the individual treatment strategy will result in expensive investment and operating cost. At present, wet limestone/gypsum process has become the mainstream technology of flue gas desulfurization (FGD) at home and abroad, which has attained high SO_2 removal efficiency, yet it is not much effective for NOx removal because NO is insoluble in water. Therefore, this problem has been extensively researched, including mediated electrochemical oxidation (MEO) [3,4], semidry processes using highly reactive absorbent [5], TiO_2 photocatalytic oxidation [6] and wet scrubbing processes using chemical oxidants in which and many reagents [7–13] have been added into aqueous solutions as absorbents to achieve simultaneous removal of SO_2 and NOx, including $NaClO_2$/NaOH, $FeSO_4$/H_2SO_4, $KMnO_4$/NaOH, Fe(II)EDTA, Na_2SO_3, $FeSO_4$/Na_2SO_3, H_2O_2, urea, and so on. $NaClO_2$ is considered as an extremely effective absorbent [14–24], but its industrial application is difficult because of the high price and the low removal efficiency of NO at low $NaClO_2$ concentration.

In order to reduce the cost of simultaneous removal of SO_2 and NOx and to improve removal efficiency of NO, a complex absorbent that was made up of NaClO (M) and $NaClO_2$ was used to simultaneous removal of SO_2 and NOx from simulated flue gas in this paper. The influencing factors, such as molar ratio of M to $NaClO_2$, solution pH, reaction temperature, SO_2 concentration and NO concentration were investigated, and the optimal experimental conditions were established. Meanwhile, the presumed reaction mechanism of simultaneous desulfurization and denitration was discussed according to the experimental results and previous work. This work has great academic significance and application value.

2. Experimental

2.1. Experimental apparatus

Fig. 1 shows the schematic diagram of experimental apparatus including a flue gas simulation system, an absorption reactor and a flue gas analysis system. A small bubbling device at the height of 15 cm with volume of 1 L was used as the absorption reactor.

2.2. Reagents

Sodium chlorite used was technical-pure grade (Tianjin Tanggu Chemical Reagent Factory, Tianjin, China). Sodium hypochlorite (M) was analytical pure reagent (Tianjin Chemical Factory, Tianjin, China). Hydrochloric acid was analytical pure reagent (Zhenxing Chemical Factory, Tianjin, China). Sodium hydroxide was analyt-

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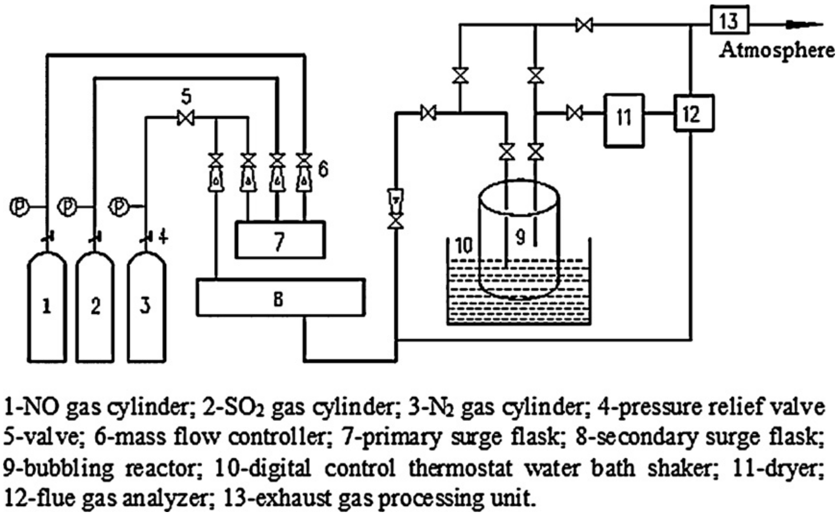


Fig. 1. Schematic diagram of experimental apparatus.

ical pure reagent (Huadong Chemical Reagent Factory, Tianjin, China).

2.3. Experimental method

During the experiments, SO_2, NO and N_2 were metered through mass flow controllers (MFC) and mixed in two surge flasks, in which SO_2 and NO were diluted by N_2 to the desired concentrations, the simulated flue gas formed. The oxidization and absorption reactions occurred when simulated flue gas entered into bubbling reactor. The concentrations of SO_2 and NOx were measured by flue gas analyzer (MRU95/3 CD, MRU Company, Heilbronn, Germany) before and after absorption to calculate the removal efficiencies of SO_2 and NO. Then, the simulated flue gas was discharged into the atmosphere after being treated by the exhaust gas processing unit. In order to avoid the destroy of flue gas analyzer, the desiccator was used to remove the moisture content in simulated flue gas after absorption.

In addition, the reaction temperature was regulated by the digital control thermostat water bath shaker (THZ-82 type, Jiangsu Jintan Shunhua Instrument Co., Ltd, Changzhou, China). The solution pH was adjusted by mixed acid and alkali (phosphoric acid, acetic acid, boric acid and sodium hydroxide) buffer, during the preparation of complex absorbent, reagents, M and $NaClO_2$ were added to the buffer solution with required pH. Hence, the pH and volume of reaction solution kept almost constant.

3. Results and discussion

3.1. Effect of molar ratio of M to $NaClO_2$ on the removal efficiencies of SO_2 and NO

When the concentration of $NaClO_2$ retained constant, the effect of molar ratio of M to $NaClO_2$ on removal efficiencies of SO_2 and NO was investigated experimentally and the results are shown in Fig. 2. From Fig. 2, one finds that the molar ratio of M to $NaClO_2$ has a slight effect on desulfurization and remarkable effect on denitration. The removal efficiency of NO increases significantly with an increase of M concentration between 0 and 0.4 mmol L^{-1}, however, the change occurs at 0.4 mmol L^{-1}, thereafter, remains stable with increasing M concentration at above 0.4 mmol L^{-1}, indicating that M has obvious promotion on denitration. According to previous work [25], using $NaClO_2$ solution for simultaneous desulfurization and denitration, when the removal efficiency of NO reached 85%, the concentration of $NaClO_2$ was required to be 4 mmol L^{-1}. But in this work, it was 0.1 mmol L^{-1} when the same removal efficiency of NO was obtained, the usage is reduced to about 1/40 comparing with using $NaClO_2$ alone.

From the aspect of economy, the price of $NaClO_2$ is obviously higher than that of M. Hence, it means that the absorbent cost can be greatly reduced by increasing the consumption of M and reducing that of $NaClO_2$ when the M/$NaClO_2$ solution is applied for simultaneous desulfurization and denitration. Fig. 3 shows the effect of

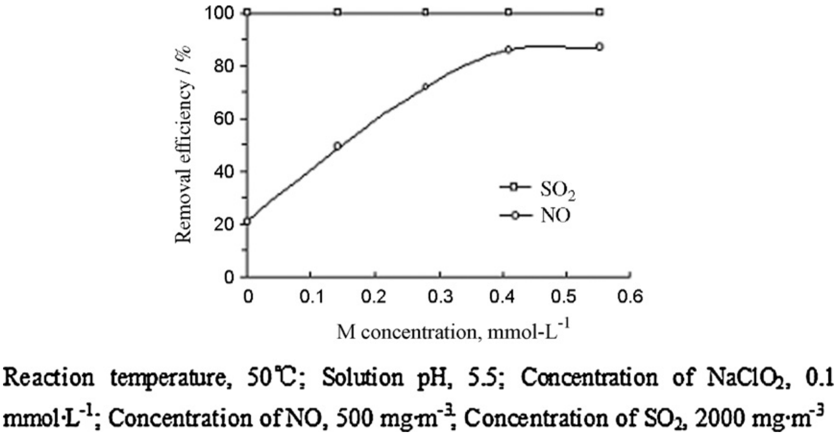


Fig. 2. Effect of the concentration of M on the removal efficiency of NO.

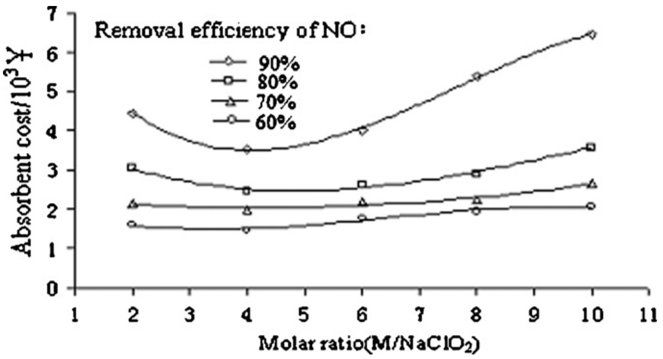


Fig. 3. Effect of molar ratio of M to $NaClO_2$ on the absorbent cost.

molar ratio of M to $NaClO_2$ on absorbent cost in which the price of $NaClO_2$ is 16 000 ¥/t, and the price of M is 700 ¥/t. As shown in Fig. 3, overall, the absorbent cost increases with an increase of desired removal efficiency. No matter which of 60%, 70%, 80% and 90% the removal efficiency of NO is, the cost decreases with increasing the molar ratio of M to $NaClO_2$ between 2.0 and 4.0, however, the change occurs at 4.0, thereafter, it increases gradually with the increase of molar ratio. Therefore, the optimal molar ratio of M to $NaClO_2$ was selected as 4.0.

3.2. *Effect of solution pH on the removal efficiencies of SO_2 and NO*

The previous studies indicated that the oxidation–reduction potentials relevant to $NaClO_2$ and M are greatly affected by solution pH, hence, during the removal reactions, the oxidative absorption characteristics of SO_2 and NO will vary with solution pH. So the effect of solution pH on the removal efficiencies of SO_2 and NO was investigated experimentally and the results are shown in Fig. 4.

As shown in Fig. 4, the removal efficiency of SO_2 is slightly affected by solution pH and retains 100% with the change of solution pH. The removal efficiency of NO is significantly affected by solution pH, however, it is higher and almost retains stable when solution pH varies from 2.4 to 4.0, thereafter, decreases with an increase of solution pH between 4.0 and 9.2.

The phenomenon may be attributed to the generation of chlorine dioxide in acid solution because of $NaClO_2$ decomposition [26] and the reaction of $NaClO_2$ with M [27].

$$5ClO_2^- + 4H^+ \rightarrow 4ClO_2 + Cl^- + 2H_2O \quad (1)$$

$$ClO^- + H^+ \rightarrow HClO \quad (2)$$

$$HClO + ClO_2^- + H^+ \rightarrow Cl_2O_2 + H_2O \quad (3)$$

$$Cl_2O_2 + ClO_2^- + H^+ \rightarrow 2ClO_2 + Cl^- \quad (4)$$

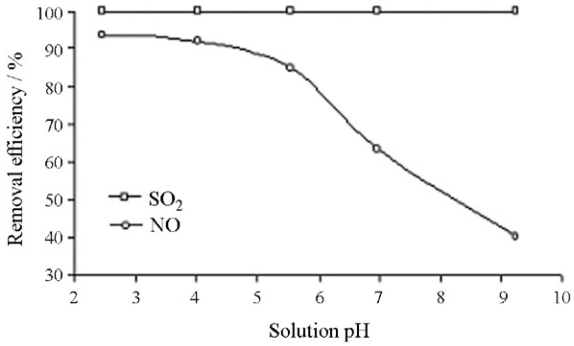
$$HClO + Cl^- + H^+ \leftrightarrow Cl_2 + H_2O \quad (5)$$

The conversion of $NaClO_2$ to ClO_2 can be promoted through Eqs. (2)–(4) in the presence of M. The molecule of ClO_2 contains 19 valence electrons, including an unpaired electron which can transit between the oxygen atom and chlorine atom. So the molecule of ClO_2 is like a free radical [28], and can help in achieving higher removal efficiency of NO because of its special structure and strong oxidative ability. Based on the consideration of equipment corrosion problems and industrial application to WFGD process, the optimal pH value of M/$NaClO_2$ solution was selected as 5.5.

3.3. *Effect of reaction temperature on the removal efficiencies of SO_2 and NO*

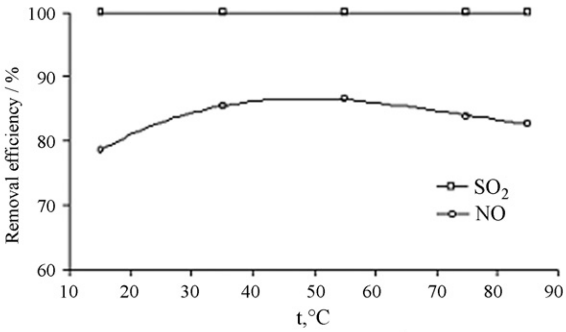
The reactions between SO_2 and absorbent, and NO and absorbent belong to gas–liquid reactions. Reaction temperature plays an important role on diffusion behavior, dissolution and reaction characteristics of molecules or ions in solutions. So its effect on removal efficiencies of SO_2 and NO was investigated experimentally and the results are shown in Fig. 5.

It can be seen from Fig. 5 that the removal efficiency of SO_2 is slightly affected by reaction temperature and almost retains 100%, but the removal efficiency of NO has a change of increasing at first and then dropping with the rise of reaction temperature. The highest removal efficiency of NO is achieved at 40–50 °C. For this, there are two reasons. On one hand, with the rise of the reaction temperature, the thermal decomposition of $NaClO_2$ [29] and the reaction producing ClO_2 between $NaClO_2$ and M [27] become more rapid and more remarkably affects the absorption of SO_2 and NO. Meanwhile, there is a promoting effect on the absorption of NO because the diffusion rates of NO molecules and the species in complex absorbent are accelerated, which lead to an increase of the oxidation and absorption rates. On the other hand, the absorption of NO is inhibited because of the reduction of the solubility with the rise of the reaction temperature. When the promoting effect is greater than the inhibiting effect, the removal efficiency of NO increases, otherwise, it decreases. The optimal reaction temperature was selected as 50 °C.



Reaction temperature, 50℃; Concentration of $NaClO_2$, 0.1 mmol·L^{-1}; Concentration of additive M, 0.4 mmol·L^{-1}; Concentration of NO, 500 mg·m^{-3}; Concentration of SO_2, 2000 mg·m^{-3}

Fig. 4. Effect of solution pH on the removal efficiencies of SO_2 and NO.



Solution pH, 5.5; Concentration of NaClO$_2$, 0.1 mmol·L^{-1}; Concentration of additive M, 0.4 mmol·L^{-1}; Concentration of NO, 500 mg·m^{-3}; Concentration of SO$_2$, 2000 mg·m^{-3}

Fig. 5. Effect of reaction temperature on the removal efficiencies of SO$_2$ and NO.

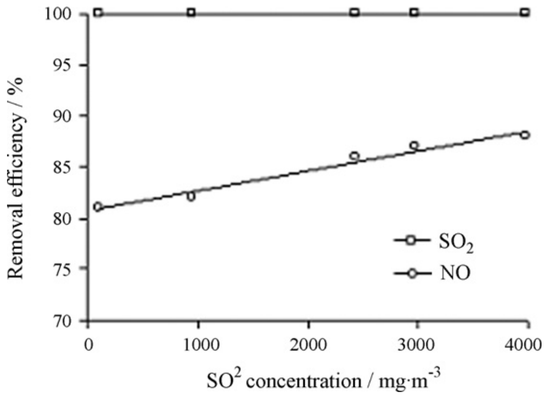


Fig. 6. Effect of SO$_2$ concentration on the removal efficiencies of SO$_2$ and NO.

3.4. Effect of SO$_2$ concentration and NO concentration on the removal efficiencies of SO$_2$ and NO

The effect of SO$_2$ concentration on removal efficiencies of SO$_2$ and NO is shown in Fig. 6 when the concentration of NO is 500 mg m^{-3}, and the effect of NO concentration on removal efficiencies of SO$_2$ and NO is shown in Fig. 7 when the concentration of SO$_2$ is 2000 mg m^{-3}. And the experiments were carried out under the conditions in which the reaction temperature was 50 °C, solution pH was 5.5, the concentration of NaClO$_2$ was 0.1 mmol L^{-1}, the concentration of M was 0.4 mmol L^{-1}.

As shown in Fig. 6, the removal efficiency of SO$_2$ retains 100% and the removal efficiency of NO increases with an increase of SO$_2$ concentration, indicating that the promotion of SO$_2$ on the oxidation and absorption of NO exists in this system, the reason may be resulting from hydrolysis of SO$_2$ [30,31], in which the products, such as $HSO_3{}^-$ and $SO_3{}^{2-}$, catalyze the NOx absorption.

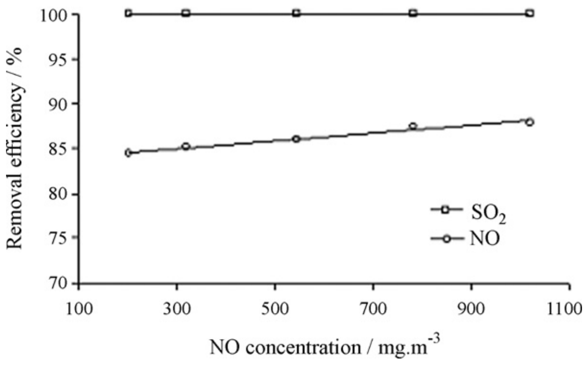


Fig. 7. Effect of NO concentration on the removal efficiencies of SO$_2$ and NO.

As seen from Fig. 7, the removal efficiency of SO$_2$ retains 100% and the removal efficiency of NO increases slightly with an increase of NO concentration. The reason might be that the gas–liquid mass transfer rate of NO is proportional to the concentration driving force according to film theory and the increase of gas partial pressure makes the concentration driving force between gas phase and liquid phase increase so that the gas–liquid mass transfer rate of NO is enhanced when the concentration of NO is increased.

3.5. Parallel tests

The parallel tests were carried out under the optimal conditions in which the reaction temperature was 50 °C, solution pH was 5.5, molar ratio of M to NaClO$_2$ (NaClO$_2$ concentration of 0.1 mmol L^{-1}, M concentration of 0.4 mmol L^{-1}) was 4.0, the concentration of NO was 500 mg m^{-3}, the concentration of SO$_2$ was 2000 mg m^{-3}. The results are shown in Table 1. As shown in Table 1, the better reproducibility of removal efficiencies of SO$_2$ and NO in tests indicates that this method has the stable performance and could provide a reference for industrial application.

4. Reaction mechanism

For complex absorbent solution, the possible existence forms of chlorine species are $ClO_2{}^-$, $HClO_2$, ClO_2, HClO, ClO^-, Cl^- and Cl_2, respectively, in which, $ClO_2{}^-$ and HClO are the main forms of Cl(III) species and Cl(I) species, respectively, when solution pH is 5.5. The experimental results of simultaneous desulfurization and denitration with complex absorbent indicate that the oxidative absorption of NO is enhanced effectively in the presence of M, namely M takes part in the solution reactions. When the oxidative absorption of SO$_2$ and NO with complex absorbent solution occurs, ClO$_2$ and Cl$_2$ are produced by the reaction Eqs. (1)–(5), and there might be mass transfers of ClO$_2$ and Cl$_2$ from liquid phase to gas phase because their partial pressures in gas phase are smaller than the gas–liquid equilibrium partial pressures. Meanwhile, there are mass transfers of SO$_2$ and NO from gas phase to liquid phase because their partial pressures in gas phase are greater than the gas–liquid equilibrium partial pressures. So the oxidation reactions might happen in gas phase and liquid phase at the same time.

Lee et al. [32] considered that ClNO$_2$, NO$_2$ and ClNO were produced in the oxidation of NO using NaClO$_2$. Zhong [33] reported that

Table 1
Results of parallel tests.

	No.					Average	Standard deviation
	1	2	3	4	5		
Removal efficiencies of SO_2, %	100	100	100	100	100	100	0
Removal efficiencies of NO, %	85.5	85.2	84.5	85.4	84.9	85.1	0.406

SO_3 and Cl_2 were produced in the oxidation of SO_2 using $NaClO_2$. So the possible reactions in gas phase are listed as follows.

$$ClO_2 + SO_2 \rightarrow SO_3 + ClO \quad (6)$$

$$ClO + SO_2 \rightarrow SO_3 + Cl \quad (7)$$

$$ClO_2 + NO \rightarrow NO_2 + ClO \quad (8)$$

$$ClO + NO \rightarrow NO_2 + Cl \quad (9)$$

$$Cl + NO \rightarrow ClNO \quad (10)$$

$$Cl + NO_2 \rightarrow ClNO_2 \quad (11)$$

$$Cl + Cl \rightarrow Cl_2 \quad (12)$$

When the reactions take place in liquid phase, the possible existence forms of sulfur species are SO_3, $SO_2{\cdot}H_2O$, $HSO_3{}^-$, $SO_3{}^{2-}$, $HSO_4{}^-$ and $SO_4{}^{2-}$, respectively, in which, $HSO_3{}^-$ and $SO_4{}^{2-}$ are the main existence forms of S(IV) species and S(VI) species, respectively, when solution pH is 5.5. For nitrogen species, the possible existence forms are NO, NOCl, NO_2, NO_2Cl, HNO_2, $NO_2{}^-$, HNO_3 and $NO_3{}^-$, respectively, in which, $NO_2{}^-$ and $NO_3{}^-$ are the main existence forms of N(III) species and N(V) species, respectively, when solution pH is 5.5. So the possible reactions in liquid phase are listed in the supplementary supporting information.

In this work, the gas phase oxidation of SO_2 and NO could be neglected because little ClO_2 and Cl_2 are transferred into gas phase from the solution containing low concentrations of $NaClO_2$ and M.

The oxidation reactions of the reductant with $ClO_2{}^-$ are considered to be carried out in two steps [34]. The products are HClO in first step and Cl^- in the second step. So the reactions (Eqs. (18), (27), (31)) are inhibited due to the increase of HClO concentration in the presence of M. But there is more ClO_2 generated through reaction Eqs. (2)–(5) in the presence M, which enhances the oxidative absorption of SO_2 and NO. At the same time, the performance of the solution in desulfurization and denitration of the solution is strengthened through the reaction Eqs. (19), (28), and (32). As seen from the whole process, the reactions of $NaClO_2$ self-decomposition and the interaction between M and $ClO_2{}^-$ consume H^+ which is produced by the absorption reactions of SO_2 and NO. So the simultaneous removal of SO_2 and NO using complex absorbent might be considered as H^+ catalytic process, and the performance of desulfurization and denitration is enhanced with the decrease of solution pH.

5. Conclusions

(1) M in complex absorbent could promote the conversion of $NaClO_2$ to ClO_2, and take part in oxidation reactions of SO_2 and NO. The existence of $ClO_2{}^-$, ClO_2 and M could improve the removal efficiencies of SO_2 and NO.

(2) Compared with $NaClO_2$ solution used for simultaneous desulfurization and denitration, the absorbent cost for given removal efficiency is greatly reduced due to the decrease of $NaClO_2$ dosage in the presence of M.

(3) Considering the application to FGD process, the optimal conditions for simultaneous desulfurization and denitration using complex absorbent containing M and $NaClO_2$ were determined in which the reaction temperature was 50 °C, solution pH was 5.5, and molar ratio of M to $NaClO_2$ was 4.0. Under the optimal conditions, 100% of SO_2 and 85% of NO were removed experimentally.

(4) The reaction mechanism for simultaneous desulfurization and denitration using complex absorbent containing M and $NaClO_2$ could be considered as the H^+ catalytic process. The removal efficiencies of SO_2 and NO are increased with the decrease of solution pH value.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.cej.2010.02.060.

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