Extending the Durability of Old Books by Atomized Deacidification and Reinforcement Treatments

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Old books suffer from aging and deterioration spurred by acidification, oxidation, and other factors. To preserve these important historical documents, it is important to implement deacidification and reinforcement methods to extend their durability. In this study, microdroplets of 75 g/L sodium hydroxide solution were atomized before being utilized to neutralize acidity in the paper. As well, styrene acrylic latex was diluted to 10 times for atomization to function as a reinforcing agent. In addition to studying these methods individually, the effects of simultaneous deacidification and reinforcement were also studied.

Keywords: Old books; Papermaking; Deacidification; Reinforcement

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INTRODUCTION

Old books are an important carrier for recording human history and cultural traditions. In addition to being widely used for academic purposes, books are an integral part of many historical relic collections. Due to the degradation of paper over time, some precious original manuscripts have become yellowed, brittle, and riddled with holes, rendering them unreadable. The damage that occurs in old books is mainly due to natural aging, acidification, and oxidation (Area and Cheradame 2011). Such damage generally occurs slowly over time, and any delay in preserving aging books will lead to further damage. Old books are non-renewable, so it is important to maintain their authenticity and original contents, especially for those that exist in limited copies.

Cellulose fibers are the main component of paper. The aging of paper is mainly due to the degradation of cellulose, which hydrolyzes easily in acidic conditions. Acidic substances from hydrolysis of the acetyl groups in the hemicellulose component of the fibers, the adsorption of atmospheric pollutants (SO_2, NO_x) (Menart *et al.* 2014), as well as the additives from the papermaking process (Basta *et al.* 2006) may cause hydrolysis of paper cellulose.

Unbound paper sheets have been conventionally deacidified by their immersion in aqueous solutions containing a pH buffer compound such as magnesium carbonate followed by drying under constraint to preserve flatness (Baty *et al.* 2010). Though such procedures are well justified for high-valued items and rare books that need rebinding, there has been a need for rapid methods that do not require the complete wetting of paper. A class of methods known as mass deacidification has been developed and practiced mainly to extend the useful lifetimes of library books (Baty *et al.* 2010).

The basic principle of the deacidification of old paper is to neutralize the acidic materials present in the surface and interior of the paper. The known mass deacidification methods can be roughly classified into gas-phase and liquid-phase methods (Williams and Kelly 1976; Fan and Guo 2018). Known gas-phase deacidification processes include diethyl zinc (DEZ) (Library of Congress 1988; Cunha 1987; Yasue 1997; Zeronian and Needles 1989) and the Book Preservation Associates (BPA) (Cunha 1987) method. The DEZ process, involves the treatment of paper with gaseous diethyl zinc (Zeronian and Needles 1989). The BPA progress involves the treatment of paper with ammonia gas and gaseous ethylene oxide, which react to form ethanolamine, diethanolamine, and triethanolamine (Cunha 1987). The alkali retention after the gas method treatment involving ammonia gas is low (Yan *et al.* 2016; Fan and Guo 2018). The inefficient alkali retention and safety hazards of gas methods make them not commonly used anymore.

Liquid-phase mass deacidification methods can be regarded as more versatile than the gas-phase methods. Liquid-phase methods mainly include the Wei T'o system (Library of Congress 1988; Cunha 1987; Yasue 1997; Zeronian and Needles1989; Kolar 2008; Brandis 1994) and the Battelle method (Andres *et al.* 2008). The Wei T'o process involves the treatment of paper with methoxy magnesium methyl carbonate (MMMC) dissolved in a mixture of methanol and freon solvent. The Battelle process can be regarded as a modified Wei T'o process that replaces MMMC by magnesium and titanium ethoxides and the freon solvent by hexamethyldisiloxane (HMDO) (Andres *et al.* 2008). Besides, in early work at the British Library (Hon 1989; Hubbe *et al.* 2018) methanol was used as a nonaqueous solvent, and $Ba(OH)_2$ as the alkaline compound to extend the lifetime of paper. But methanol has been found to solubilize some ink components that might do harm to paper protection. The CSC Booksaver system, employing carbonated methyl propoxylate (CMP) in a propanol-fluorocarbon solvent mixture, became available in 2002 (Dupont *et al.* 2002; Gibert Vives *et al.* 2004; Wagner et al. 2008; Henniges *et al.* 2012; Hubbe *et al.* 2018). The addition of magnesium ions catalyzes the acceleration of cellulose by iron and copper ions natural oxidation (Zeronian and Needles 1989; Kolar *et al.* 2005). These methods are currently applied in the restoration of acidic books and ancient relics, but they have several unavoidable drawbacks such as powder sediment (Zervos and Moropoulou 2006; Area and Cheradame 2011; Wang *et al.* 2013).

Another mass deacidification method in current practice uses a suspension of MgO particles in a nonaqueous carrier liquid (Baty *et al.* 2010; Jablonsky *et al.* 2013). This method has demonstrated effectiveness for distribution of the alkaline particles throughout the paper structure, though doubts have been raised regarding the completeness of the deacidification under typical conditions of treatment (Hubbe *et al.* 2017).

In addition to deacidification, reinforcement is another important factor that must be considered. If an old book is only deacidified, then the damage can only be decelerated and not reversed. A reinforcement process is needed to modify the mechanical performance of an old book to make it strong enough for holding/reading. The reinforcement generally includes the Viennese process (Cunha 1987), organosilane reagent (Ipert *et al.* 2006; Souguir *et al.* 2011; Souguir *et al.* 2012), lamination (Letnar and Vodopivec 1997), sizing/impregnation (Leijonmarck *et al.* 2013), paper splitting (Galinsky and Haberditzl 2004), and leafcasting (Kruth 1988). By implementing such reinforcement processes, the strength properties of old books can be improved obviously, and this may be a viable alternative or an addition to deacidification process in the preservation of paper.

Up to this point, most of the research done on the preservation of old books has focused on the chemicals used in the process. NaOH is the well-known strong base, which is beneficial to efficient and rapid deacidification. In the early 2000s, alkaline nanoparticles started to be utilized for deacidification; such work was based on an idea of easy penetration into the surface and interior of paper (Giorgi *et al.* 2005; He *et al.* 2019). In order to improve the deacidification efficiency, an ultrasonic atomizer, which can make micro-particles (around 4 to 5 μm), could help penetrate deacidifying agent and reinforcing agent into paper.

Based on the results of previous study (Chen *et al.* 2018), it has been shown that sodium hydroxide can be efficient in deacidification on old books. But the defect of this method is that it is difficult to control the proper dosage and homogenous penetration of NaOH during treatment process. If the amount of alkaline substances retained in the paper after treatment is too high, then the paper might be damaged. Considering the reasons above, the ultrasonic atomization method was selected in an attempt to improve the uniform penetration of tiny droplets of relatively concentrated NaOH. The operating principle of ultrasonic atomization method requires the use of a relatively concentrated aqueous solution to obtain sufficiently high treatment levels in the paper without excessive moistening (Hanus *et al.* 2008). Thus the same amount of agent can add more alkaline substances onto paper. The atomization process will inevitably bring in moisture to the paper sheet. This will cause more problems such as forming wrinkles on the paper and even accelerating the hydrolysis of fibers in the natural drying process afterwards. To avoid these problems, it is good to select the higher concentration to bring less moisture in a relatively short period of time. During ultrasonic atomization method deacidification, strict control of NaOH usage is obtained by controlling the atomization time, because the pH of treated paper needs to be maintained at 7.0 to 8.0 in order to prevent accelerating air-oxidation of paper. Meanwhile, the mechanical strength is not significantly decreased, and the color difference is controlled within 1.5, which can avoid being naked to the naked eye (Chen *et al.* 2018).

This is the first report of the application of sodium hydroxide with ultrasonic atomization method for the acidification of old books. This work used an ultrasonic atomizer with microdroplets and nanodroplets of deacidifying and reinforcing solutions are applied to old books separately. The deacidification conditions and mechanism was investigated with the aim of achieving good conservation without any damage at room temperature and atmospheric pressure. The degradation of paper fibers from the point of paper structure and the aging principle were also considered in this study.

EXPERIMENTAL

Materials

The paper samples were taken from the 1988 Xiangtan University textbook, *Environmental Engineering Principles*. Sodium hydroxide (NaOH) was purchased from Nanjing Chemical Reagent Co. (Nanjing, China). Styrene acrylic latex was used as the reinforcing agent (Guangzhou Zhencheng Chemical Co., Guangzhou, China). The styrene acrylic latex (Guangzhou Zhencheng Chemical Co., Guangzhou, China) was diluted to one-tenth of the as-received strength before use, with the viscosity of 4.69 cP after mixing at 180 rpm.

Paper preparation

The paper was prepared according to the standard GB/T 450-2008 (2008). To absorb more microdroplets, the paper samples were placed in a vacuum-drying (DZF 6050, China) oven at 40 °C under a vacuum degree of 0.08 MPa for 12 h before being immediately subjected to ultrasonic atomization (WH-2000, China). The idea was to remove some of the bound and free water, to enable the paper to absorb more microdroplets later in the treatment.

Through the deacidification method, 75 g/L of NaOH solution was used and the diluted styrene acrylic latex was prepared for the strengthening of the paper. The solutions were prepared as microdroplets (around 4 to 5 μm) to penetrate the paper.

Paper can withstand the moisture of atomization for 90 min as pre-tested (while longer than 90 min, paper started to shrink during the drying process). Too much exposure to moisture will cause the paper to shrink during drying, so it is important to set a time limitation per round, which was chosen as 30 min per round in this paper.

During atomization, two separate fans were used to circulate air if needed. This treatment option will be referred to as the 'fan ventilation'.

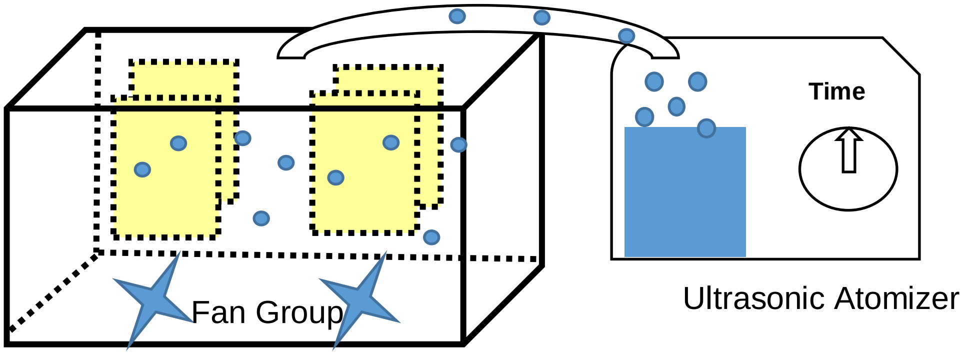

Fig. 1. The designed atomization device

After atomization, the treated samples were naturally air-dried for around 4 h at room temperature (25 °C). This process prevented the paper from absorbing too much NaOH and raising the pH value excessively. The control sample had a pH value of approximately 4.5 and a machine direction (MD) tensile index of 26.0 Nm/g.

Methods

Characterization of paper – Determination of pH on paper surface

To avoid the destructive effects of measurements such as cold extraction, the surface pH method was utilized in this study (Strlič *et al.* 2007; Roberson *et al.* 1976). The pH value of the paper surface is an important indicator to evaluate the degree of deacidification (Strlič *et al.* 2007; Xingling *et al.* 2008). During the test, a drop of distilled water (0.5 mL) was added on the surface of paper and then put on the flat electrode (planar pH composite electrode, China), each sample was tested 3 times to calculate the results. The Chinese standard GB/T 13528-92 (1992) was used to determine the pH of the sample.

Determination of machine direction (MD) tensile index of paper

The samples were placed in an constant temperature and humidity chamber for 24 h (23 ± 1 °C, 50 ± 2% relative humidity (RH)) according to the ISO 187-90 (1990) standard. The tensile strength (L&W CE062, Sweden) was measured according to the standard GB/T 12914-2008 (2008). Because the fibers were arranged mainly in the MD, the paper strength performance was different in all directions. The tensile strength was tested in the MD and the tensile index was calculated accordingly (Liang *et al.* 2017). A total of 10 specimens of each sample (100-mm-long × 15-mm-wide) were tested to ensure repeatability of the results.

Calculation of paper color difference

The whiteness, L^*, a^*, and b^* values were tested as standard ISO2470-1-2009, (L&W Elrepho 070, Sweden, R457 D65) before and after atomization were measured using the whiteness meter. The color difference was calculated according to Eq. 1,

$$\Delta E^* = (2\Delta L^* + 2\Delta a^* + 2\Delta b^*)^{\frac{1}{2}} \tag{1}$$

where ΔL^* is the difference in lightness and darkness, Δa^* is the difference in red and green, Δb^* is the difference in yellow and blue, and ΔE^* is the total color difference of the sample.

Calculation of relative change (RC) in pH

The relative change in pH was calculated to evaluate how efficiently the method neutralized the acidity in the paper. The calculation for relative change in pH can be seen in Eq. 2,

$$RC\ (\%) = \frac{pH\ of\ treated\ paper - pH\ of\ untreated\ paper}{pH\ of\ untreated\ paper} \times 100 \tag{2}$$

where RC is the relative change in pH (%).

RESULTS AND DISCUSSION

Extending the Durability of Old books Using Only the Deacidification Method

The deacidification mechanism is assumed to involve a neutralization reaction between the acidity in the paper and OH^- from the NaOH in standard conditions. The atomization time of the deacidifying agent was controlled and the fan ventilation was used in combination with the atomization device. The changes in the surface pH values and deacidification efficiency, MD tensile index, and color difference were determined. Results are shown in Fig. 1, Fig. 2, and Table 3, respectively.

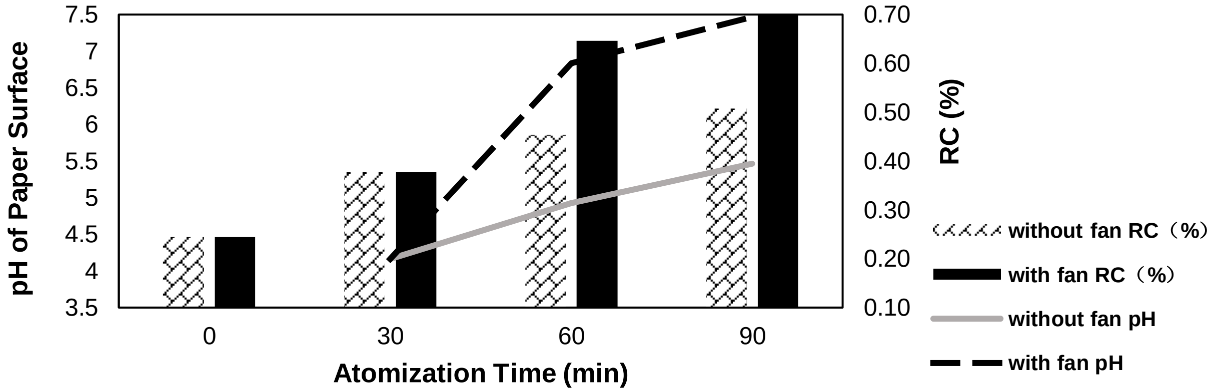

Fig. 2. Effect of NaOH atomization time on the relative change (RC) in the pH and the paper surface pH

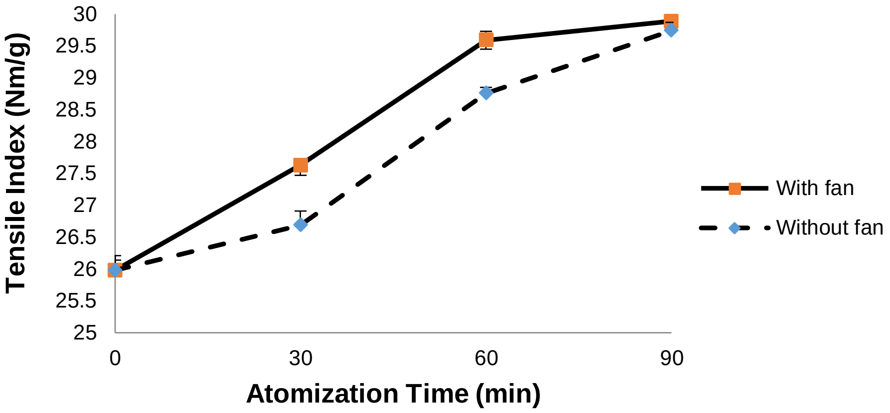

Fig. 3. Effect of NaOH atomization time on the MD tensile index

Table 1. Effect of Deacidification Time on Paper Color Difference

Conditions	△*L**	△*a**	△*b**	△*E**
Deacidified 30 min	-1.66	-0.32	-1.34	1.47
Deacidified 60 min	0.03	-0.08	0.49	0.50
Deacidified 90 min	0.10	0.01	1.10	1.10

The original pH of the sheet was 4.5, and the original MD tensile index was 26.0 Nm/g. The post-treatment surface pH of all samples increased considerably and showed a positive trend with increasing atomization time of NaOH solution. The surface pH increased from 4.5 to 7.6 as the atomization time was increased from 0 to 90 min, respectively. As the pH of paper is being determined by wetted with a drop of water, it is not possible to test local alkaline spots and local acidic spots (Hubbe *et al.* 2017). It was shown that the neutralization reaction effectively inhibited the acid hydrolysis of cellulose fibers. However, the growth rate leveled off after 60 min of ultrasonic atomization. The relative change in pH showed a similar trend of quickly rising from 19% to 60% and then slowly rising to 69%. In the initial deacidification stage, neutralization was the dominant reaction that occurred between the alkaline agent and the acidic substances in the paper sheet. While neutralization continued over the course of the reaction, most of the acidic substances were neutralized before the reaction was complete. As the atomization time increased in the later stage, the change of pH was moderate.

Modification the atomization device with addition of the fan ventilation was found to remarkably improve the uniformity of the atomized droplets, which allowed better dispersity of the microdroplets in the paper sheet. The increase in pH was also more apparent in the samples with the fan ventilation. There was a positive correlation between the change in pH and the relative change in pH. After 90 min of atomization, the efficiency of the samples with the fan group was nearly double that of the ones without use of fans. However, this change was not clear when the atomization time was only 30 min, as longer time led to better dispersity. It has been recommended that the pH values achieved after conservation should be kept in the optimal range of 7.0 to 8.5 (Yueer *et al.* 2016), where there is an appropriate alkali dosage. Certain alkali reserves in papers can prevent or slow down further hydrolysis of old books by neutralizing the accumulation of acidic substances.

The use of the fan ventilation allowed the deacidification process to achieve the best preservation range to enhance their durability.

The MD tensile index of the paper also slightly increased accompanying the neutralization, and it was increased by up to 115% when the atomization time was 90 minutes. Compared to the groups without the fan ventilation, the simultaneous trend in the MD tensile index between the samples with fans also confirmed that the fan ventilation had a beneficial effect on the uniformity. The whole deacidification stage had little effect on the chromatic aberration of the paper, which kept the color change within a range that was hard to be perceived by the naked eye (Chen *et al.* 2018). With the increased atomization time, more alkaline droplets had penetrated into the paper and neutralized the acidity of paper.

The observed increase in strength can be understood possibly in terms of a localized wetting of spaces between cellulosic fibers. When a film of water becomes established between solid surfaces, it can exert a capillary force that draws the surfaces together (Page 1993). As the water evaporates, the capillary forces draw the surface into molecular contact, allowing new hydrogen bonds to form between the cellulosic surfaces (Campbell 1959). Though the structure of the paper might change in unpredictable ways during partial moistening and redrying, there is a possibility that the final strength can be increased.

Besides, as the fibers absorbed water, and the distance between cellulose got shorter, some hydrogen bonding formed. As a result, the MD tensile index increased in Fig. 3. Moreover, the use of alkaline particles might influence the lignin of paper, which caused the paper color difference.

Extending the Durability of Old books Using Only the Reinforcement Method

The strength property is important factor for extending the durability of old books Moreover, the effects of the reinforcing agent on the durability of old books were also investigated. The effect of atomization time on the tensile index is shown in Fig. 4. The effects of atomization time on the pH, relative change in pH, and color can be seen in Table 2.

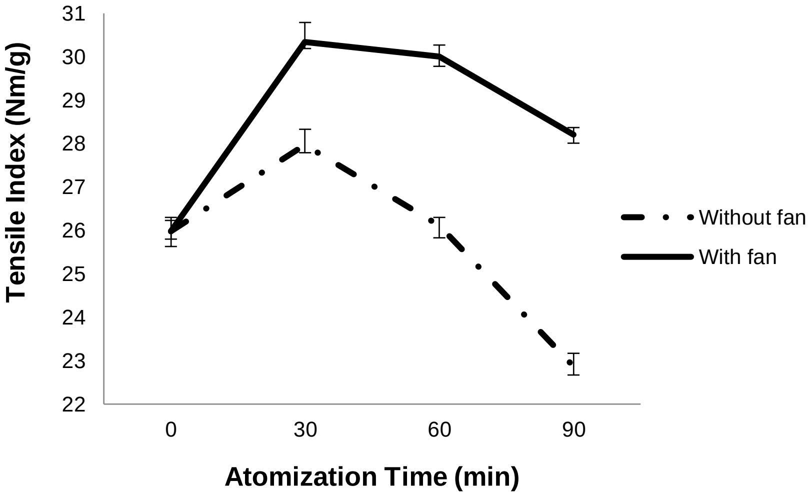

Fig. 4. Effect of the reinforcing agent atomization time on MD tensile index

Table 2. Effect of the Reinforcing Agent Atomization Time on pH, RC, and Color Difference

Conditions	pH	RC (%)	$\triangle L^*$	$\triangle a^*$	$\triangle b^*$	$\triangle E^*$
Reinforced 30 min	5.33	19.51	-2.26	-0.45	-1.07	2.54
Reinforced 60 min	5.23	17.26	0.08	-0.27	-0.82	0.87
Reinforced 90 min	5.09	14.13	-0.06	-0.36	-1.52	1.56

RC = Relative change in pH

A reinforcing agent is commonly used for strengthening both in papermaking and for cellulosic materials. It can be impregnated into the fibers to form a film that can cover the paper and modify the fiber surfaces. Moreover, the reinforcing agent is micron-size, has good permeability, and can easily penetrate the inter-fiber layers and gaps to form an adhesive bond. It can easily form a film on the surface of the paper. The addition of a reinforcing agent clearly increased the MD tensile index of the paper, by up to 117%. However, an atomization time past 30 min caused the MD tensile index to decrease due to the accumulated moisture. The concentration of the reinforcing agent was extremely low, approximately 10%, which resulted in a viscosity of 4.69 cP after mixing at 180 rpm. As time increased, the paper was exposed to more moisture. In this process, the water molecules destroyed the hydrogen bonds between the cellulose and within the fibers.

The reinforcing agent had a limited effect on increasing the MD tensile index and inhibiting moisture. In contrast, the deacidifying agent alone showed a positive, linear improvement in the MD tensile index (Fig. 4). These results verified the uniform effect of the atomizing treatment when using the fan ventilation.

The reinforcing agent is a weak, alkaline substance, with a pH value of approximately 9. The pH of the paper slightly increased as the reinforcing agent was utilized. However, as the atomization time increased, the moisture content also increased, so the weakly alkaline reinforcing agent provided insufficient alkaline content to neutralize the acidic substances in the paper. As a result, the change in pH tended to be minor with the change in moisture content. There may be some degree of chromatic aberration change on the paper, but a small chromatic aberration change can be maintained if the humidity is controlled properly.

Extending the Durability of Old Books Based on Both the Deacidification and Reinforcement Methods

With the use of the fan ventilation, the paper was first deacidified and then reinforced. By controlling the atomization time, the synergistic effect is shown in Table 3 and Table 4. The fan group was used in all of the samples.

Table 3. Effect of Atomization Time on MD Tensile Index

Conditions	Not reinforced	Reinforced (30 min)	Reinforced (60 min)	Reinforced (90 min)
Deacidified 30 min	26.69	28.12	29.01	29.90
Deacidified 60 min	28.76	29.89	30.57	30.02
Deacidified 90 min	29.74	30.17	30.94	30.15

Table 4. Effect of Atomization Time on Paper pH, RC, and Color Difference

Conditions	pH	RC (%)	△L*	△a*	△b*	△E*
Deacidified 30 min Reinforced 30 min	6.10	36.77	0.27	-0.10	-0.85	0.90
Deacidified 60 min Reinforced 30 min	7.42	66.37	-2.37	-0.01	-0.64	2.45
Deacidified 90 min Reinforced 30 min	7.83	75.45	-1.38	-0.01	-0.40	1.44
Deacidified 30 min Reinforced 60 min	6.26	40.36	0.38	-0.16	-0.57	0.71
Deacidified 60 min Reinforced 60 min	7.14	59.98	-2.39	0.10	0.84	2.53
Deacidified 90 min Reinforced 60 min	7.45	67.04	-0.60	-0.04	0.07	0.61
Deacidified 30 min Reinforced 90 min	6.12	37.22	1.52	0.10	-0.34	1.56
Deacidified 60 min Reinforced 90 min	6.62	48.32	3.63	0.15	0.23	3.64
Deacidified 90 min Reinforced 90 min	7.08	58.74	-1.66	-0.32	-1.34	1.47

As shown in Table 3, comparison with the original MD tensile index was 26.0 Nm/g, the samples treated with the deacidification solution in addition to the reinforcing agent yielded stronger paper samples, as the acidity in the paper sheet was thoroughly neutralized (Table 4). Compared to the increased time of deacidification at the same reinforced time, pH values showed an increasing trend due to the sufficient neutralization. But, with the increased reinforced time, the pH values were seen to first slightly rise and then to decrease. That might be due to the moisture change of paper and possibly experimental error. The maximum increase in the tensile index (19.1%) was found when the paper was deacidified for 90 min and reinforced for 60 min (Table 3). The bonding effect of the reinforcing agent was also increased with prolonged time at the same degree of deacidification. However, the tensile index did not always increase with increased reinforcing agent. There was a slight decrease in strength after the moisture content of the paper sheet became saturated. Higher humidity promotes hydrolysis (Graminski *et al.* 1979; Du Plooy 1981; Zou *et al.* 1996; Welf *et al.* 2005). When the atomization time of the reinforcing agent reached 60 min, the strengthening effect of the reinforcing agent reached its maximum benefit.

In Table 4, the difference in paper colors was sporadic and likely due to the degree of aging and the uneven distribution of acidic substances. The deacidification and reinforcement methods influenced the paper color. Generally, if the color difference ΔE^* is less than 1.5, the visual change in the paper color is not recognized by the naked eye (Lin *et al.* 2018). The use of alkaline particles might influence the lignin of paper to form more chromophoric groups, which caused the paper color difference. Besides, the use of SAE particles might make the paper more blue under the tests, due to its specific film property (Fei *et al.* 2012). As a result of balance, the color difference showed a slightly increase and after a bit decrease trend.

In addition to removing the acidic substance, the use of the reinforcing agent after the deacidification method can make the paper have better strength behavior to extend its life. This improved the strength, which played a positive role in the durability of the paper. However, with extended reinforcing agent atomization time, the pH decreased. The pH and relative change in pH of the groups that were reinforced longer than 60 min were outperformed by the control group. When the deacidification agent was atomized for 90 min and the reinforcing agent was atomized for 60 min, optimal conditions were achieved. Additionally, the color difference change was only 0.61, which was hard to detect with the naked eye.

The results of the present work in terms of surface pH, paper strength, and minimal change in appearance can be regarded as encouraging regarding the use of nebulized NaOH solution droplets and strengthening aid solution droplets for treatment of acidic papers. Future steps in research can include an evaluation of the storage stability. For instance, by use of accelerated aging assays it is possible to estimate the rates of further long-term degradation of paper strength (Zervos and Moropoulou 2006). Such testing is needed to evaluated the hypothesis that nebulized treatment with small droplets of NaOH, with insufficient application amounts to fully wet the paper, is able to achieve comprehensive and uniform neutralization, as well as good resistance to further aging.

CONCLUSIONS

1. The treatment that combined both deacidification and reinforcement methods was the most effective measure in raising the surface pH and increasing the paper strength of old books. Such neutralization of pH can be expected to extend the useful lifetime. The maximum MD tensile index improvement of 19.1% was seen when using the fan ventilation, atomizing the deacidification agent for 90 min, and atomizing the reinforcing agent for 60 min. If the acid was removed thoroughly *via* deacidification followed by reinforcing, then the enhanced effect obtained was higher than that of only deacidification or reinforcement.
2. Ventilation with two fans was added to the process to improve the dispersion of the agent. The fan ventilation allowed the NaOH solution and the reinforcing agent to penetrate the paper sheet more uniformly, neutralizing the acidic substance and forming inter-fiber bonds. The samples with the fan ventilation exhibited a more obvious pH increase and better relative change in pH compared to those without the fan ventilation.
3. Variations in paper moisture have an important role in the preservation of old books. As the atomization time progressed, the moisture in the sheet accumulated, resulting in a gentle change in pH. The MD tensile properties of the sheet did not always increase linearly with prolonged reinforcement atomization. At a certain point, the moisture content in the sheet became saturated, which caused a decrease in the MD tensile index. The alkaline hydrolysis of the cellulose was also promoted in this high-humidity and strong alkaline environment. Meanwhile, high moisture makes fibers swell and also accelerate the hydrolysis of fibers, which ultimately led to the decrease in the tensile strength of the paper.

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