Eco-friendly Flame-retardant Xuan Paper *via* Layer-by-layer Assembly Organic/Inorganic Composite Coating on Pulp Fiber

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Positively charged chitosan (CS) and negatively charged montmorillonite (MMT) were layer-by-layer (LBL) assembled on Xuan paper pulp fiber to fabricate an organic/inorganic flame-retardant coating. The Xuan paper achieved a limiting oxygen index (LOI) value of 24.6% when the pulp fiber was treated by 20 bilayers (BLs) of CS/MMT coating, and its vertical flame properties also improved with less after-flame and after-glow times compared to the untreated one. These results demonstrated that the flame retardancy of Xuan paper could be enhanced by this organic/inorganic composite coating. Thermogravimetric (TG) analysis results revealed that LBL assembly of CS/MMT coating on pulp fiber improved the thermal stability at higher temperature and promoted char formation, showing the obvious condensed phase flame retardant action. Scanning electron microscopy results further confirmed that the treated Xuan paper by CS/MMT coating on pulp fiber promoted the char formation and formed the stable covering layer. The ink wetting property of Xuan paper was not influenced through the LBL assembly flame-retardant treatment of the pulp fibers by CS and MMT. This research provides a new approach for flame-retardant Xuan paper without influencing its ink wetting property using the eco-friendly organic/inorganic composite flame-retardant system.

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Keywords: Xuan paper; Flame retardant; Eco-friendly; Layer-by-layer assembly; Organic/inorganic hybrid

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INTRODUCTION

Chinese Xuan paper as an important art carrier is widely used in traditional calligraphy and painting due to its high durability and aging resistant characteristics (Köklükaya *et al.* 2015; Wu *et al.* 2016). The artworks that are painted or drawn on Xuan paper could be preserved for thousands of years (Feng *et al.* 2020). Thus, Xuan paper is given the reputation of “the king of paper” (Tang and Smith 2013). As one outstanding representative of traditional cultures, the traditional handicraft of making Xuan paper was included the 2009 UNESCO Intangible Cultural Heritage of Humanity list (Shao *et al.* 2019).

However, Xuan paper made from the cellulosic raw materials of blue sandalwood bark and rice straws has high flammability, which makes it susceptible to fire disasters. Numerous precious art treasures carried by paper have been destroyed during the long history of human civilization (Dong and Zhu 2020). The recent fire disasters of Brazil's

National Museum Fire in 2018 and the huge fire at Paris' Notre Dame Cathedral in 2019 was a big loss to human civilization history. Improving the flame retardancy of these papers is one of the most important ways to reduce the fire hazards of precious artworks. Therefore, endowing flame retardancy to Xuan paper is extremely important (He *et al.* 2019; Xu *et al.* 2019; Chen *et al.* 2020).

There are many methods to improve the flame retardancy of papers, such as adding, dipping, coating, and spraying methods (Nassar *et al.* 1999; Sha and Chen 2016; Wang *et al.* 2017a; Pan *et al.* 2018). The most common way to improve the flame retardancy of paper is by adding the flame retardants into pulp fibers during the papermaking process (Wang *et al.* 2017b). Inorganic flame retardants are the most widely used in the addition method due to their lower cost, for instance, aluminum hydroxide and magnesium hydroxide (Yang *et al.* 2017; Zhou *et al.* 2020). However, the needed usage levels of these inorganic flame retardants are usually large, which may greatly influence the physical properties of paper (Wang *et al.* 2019). Organic halogen containing and/or phosphorus containing compounds are the effective flame retardants (Chen *et al.* 2018; Fang *et al.* 2019). However, these flame retardants cause severe environmental pollution problems and damage to the human body. In particular, halogen-containing flame retardants produce dioxin and toxic gases during combustion, and consequently their use has been banned (Fang *et al.* 2017a,b). Eco-friendly organic/inorganic hybrid flame retardants or flame-retardant systems with high efficiency have attracted increased attention in the last few decades, such as the nacre-mimetic organic/inorganic composite flame retardant system (Launey *et al.* 2010; Ding *et al.* 2017). The authors reported a layer-by-layer (LBL) assembly fabricating the nacre-mimetic organic/inorganic composite flame retardant system by chitosan (CS) and montmorillonite (MMT) on Xuan paper (Chen *et al.* 2019). The flame retardancy of Xuan paper was greatly improved by this nacre-mimetic flame retardant system, while the color and ink wetting property of treated Xuan paper were influenced. When the LBL assembly coating of CS/MMT on Xuan paper was over 10 bilayers (BLs), its ink wetting property will be greatly reduced. The LBL assembly method has been used for paper modification by adding functional polyelectrolytes during the papermaking process (Wågberg *et al.* 2002).

In this paper, to maintain the original properties of Xuan paper, organic/inorganic flame retardant coating of CS/MMT was fabricated on Xuan paper pulp fiber by the LBL assembly method before forming the paper sheet. The flame retardancy of Xuan papers treated by different BLs of CS/MMT on pulp fiber was determined. Thermogravimetric analysis (TGA) was used to determine the thermal stability of original and treated Xuan papers. The ink wetting property of Xuan paper before and after treatment was also compared in this research.

EXPERIMENTAL

Materials

Xuan paper (38 g/m^2) was purchased from Anhui Yuchen Paper Co., Ltd. (Anhui, China). Chitosan (CS, viscosity: 50 to 800 mPa·s) and concentrated hydrochloric acid (HCl) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Sodium montmorillonite (Na-MMT) was purchased from Zhejiang Fenghong New Material Co., Ltd. (Zhejiang, China).

LBL Assembly Treatment of Xuan Paper

The powder was dispersed in deionized water under the ultrasonication for 1 h and then magnetically stirred for 24 h to prepare uniform 1 wt% Na-MMT dispersion solution. The 1.0 wt% CS solution was prepared by dissolving in deionized (DI) water, which was adjusted the pH value to 4 using 1 M HCl solution and then stirred for 24 h.

The original Xuan paper was smashed into the pulp fiber by a mechanical pulverizer. The Xuan paper pulp fiber was dispersed into the positively charged 1 wt% CS solution for 5 min first, and then rinsed with deionized water for 30 s with the filtration cloth and oven-dried at 60 °C for 2 h. Then, the pulp fiber was dispersed into the negatively charged Na-MMT dispersion for 5 min. After that the material was repeatedly rinsed and dried, thus obtaining one bilayer (BL) of CS/MMT. The pulp fiber LBL assembly was repeated with CS and MMT until 1, 5, 10, and 20 BL were obtained, which were named by XP-1BL, XP-5BL, XP-10BL, and XP-20BL, respectively. The original Xuan paper was named XP-0. The LBL assembly CS and MMT process of pulp fiber is shown in Fig. 1. Afterwards, the same amount (5 g) of pulp fiber was added into the Ai-CP-200 type water circulation handsheet former (Shandong Animite Instrument Co., Ltd., Jinan, China) to prepare the paper sheet with the diameter of 20 cm that was dried on the AT-G2Q type paper sheet dryer (Shandong Animite Instrument Co., Ltd., Jinan, China) at 80 °C for 1 h.

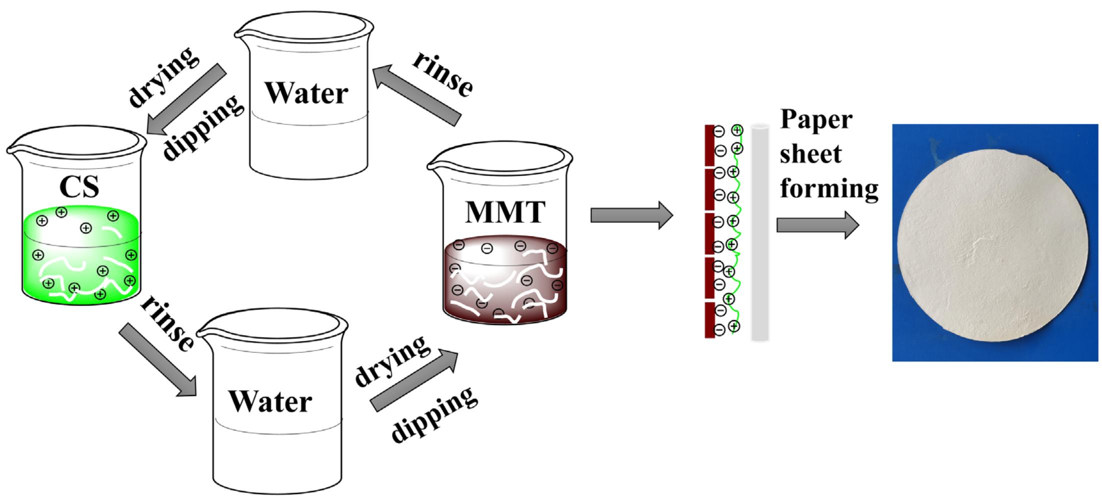

Fig. 1. Schematic representation of the LBL assembly CS/MMT on Xuan paper pulp fiber

Characterization

Fourier transform infrared (FTIR) spectra of Xuan paper before and after LBL assembly coating with CS/MMT were determined on a Thermo Nicolet Avatar 6700 FTIR (Thermo Fisher Scientific, Waltham, MA, USA). The flame retardancy of Xuan Paper before and after treatment was determined by limiting oxygen index (LOI) and vertical burning test. The LOI value was preformed according to the ASTM D2863 (2006) testing procedure on a JF-3 oxygen index instrument (Jiangning Analysis Instrument, Nanjing, China). The vertical burning test was conducted according to the ASTM D6413 (2008) testing procedure on a YG(B)815D-I flame retardant performance tester (Darong Textile instrument Co., Ltd., Wenzhou, China). Thermal stability of Xuan paper before and after treatment was investigated on a DTG-60H thermal analyzer (Shimadzu, Kyoto, Japan) from room temperature to 800 °C with a heating rate of 10 °C/min both under nitrogen and air atmosphere. The morphology of Xuan paper and char residues were viewed using a Hitachi S-4800 scanning electron microscope (SEM) (Hitach Ltd., Tokyo, Japan). Ink dispersion performance of Xuan paper was evaluated by the dispersal behavior of the same volume ink droplets that were dropped on the Xuan paper's surface from the same height (1 cm) and recorded with a digital camera.

RESULTS AND DISCUSSION

FTIR and SEM Images of Xuan Papers

The FTIR spectra of original and treated Xuan papers are shown in Fig. 2. In the spectra of untreated Xuan paper, there were peaks at 3440 and 1650 cm^{-1}, which correspond to the stretching vibration of O-H and the absorption of adsorbed water (Fang *et al.* 2020b). The spectra of treated Xuan paper also had the same peak at 3440 cm^{-1}. The peak at 1640 cm^{-1} was assigned to the O-H bending peak of the adsorbed water on the MMT surface, and the peaks at 1040 cm^{-1}, 524 cm^{-1}, and 461 cm^{-1} were attributed to the characteristic absorption peaks for the oxide band of metals in the MMT (Günister *et al.* 2007). The peak at 1558 cm^{-1} was assigned to the stretching vibration of $-NH_3^+$ due to the protonated $-NH_2$ in CS (Liang *et al.* 2019). The peak at 1379 cm^{-1} was assigned to the bending vibration of C-H in CS. The result of FTIR spectra showed that CS and MMT had been successfully applied on Xuan paper.

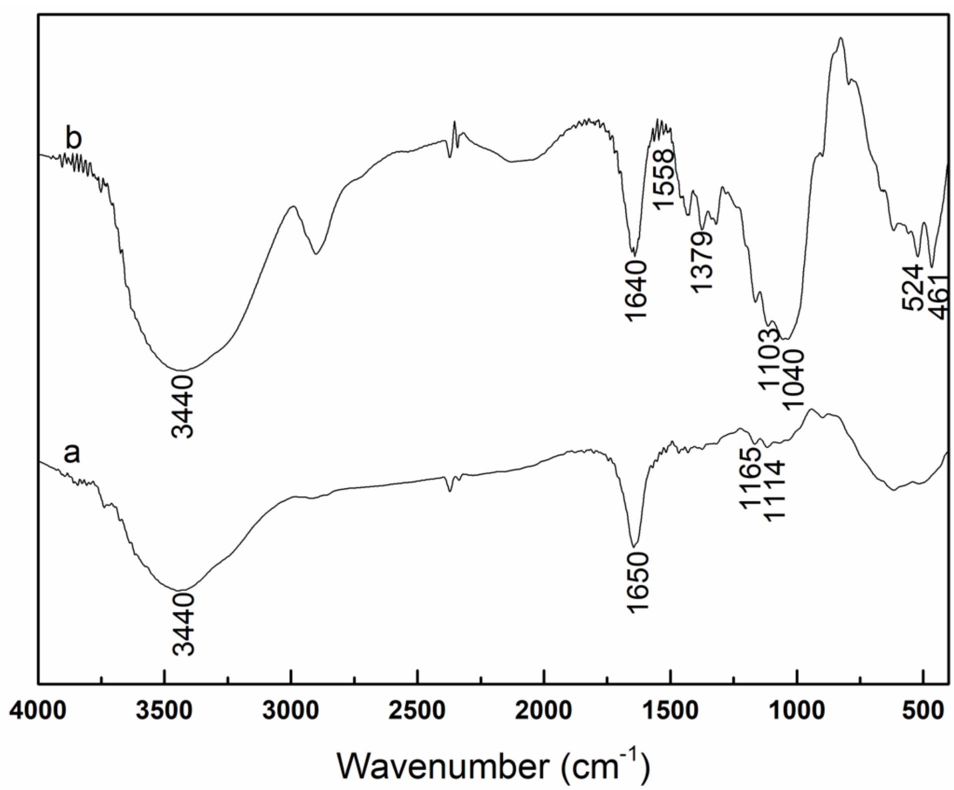

Fig. 2. FTIR spectra of (a) untreated and (b) XP-20BL

The SEM images of original and treated Xuan paper are shown in Fig. 3. As shown in the figure, the surface of treated Xuan papers changed obviously compared with the untreated one. The surface of Xuan paper was clean and smooth, while the surface was covered by CS/MMT coating after the LBL assembly treatment. The covering area of the coating layer on paper fiber increased with the BL. It was almost completely covered when the LBL assembly of CS/MMT was over 10 BL. From the results of FTIR spectra and SEM images of untreated and treated Xuan papers, it was concluded that the CS and MMT formed the organic and inorganic hybrid coating on Xuan paper fiber.

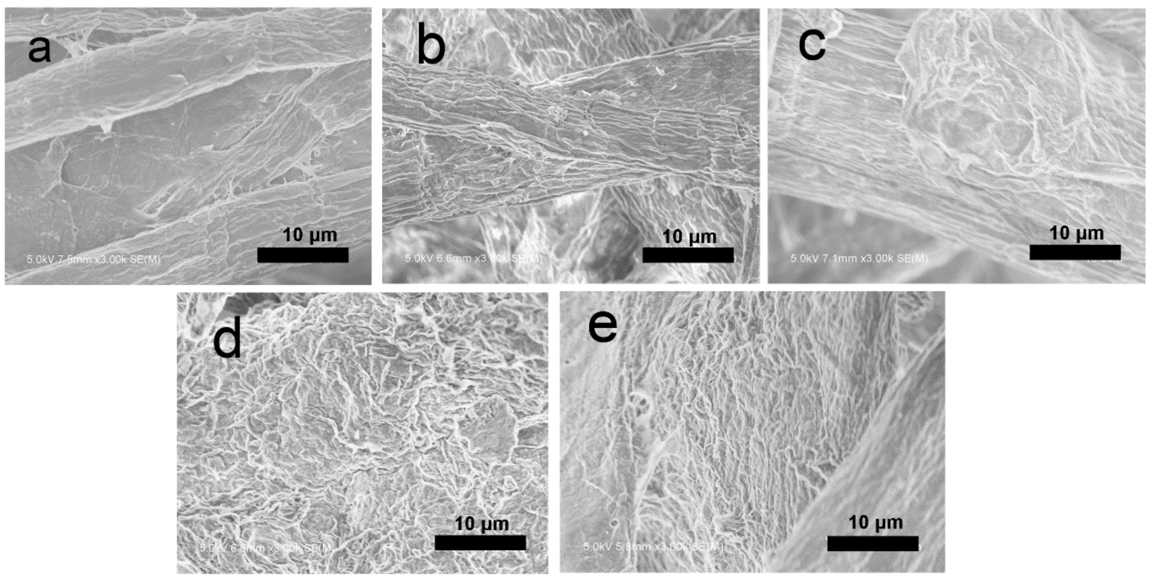

Fig. 3. The morphology of the untreated and treated Xuan papers: (a) XP-0, (b) XP-1BL, (c) XP-5BL, (d) XP-10BL, and (e) XP-20BL

Flame Retardancy of Xuan Paper

The LOI values and vertical flame test of Xuan papers are shown in Table 1. The digital photographs of Xuan papers after vertical flame are shown in Fig. 4.

Table 1. Flame Retardancy of Uncoated and Coated Xuan Paper

Sample	LOI (%)	Vertical Burning Test		
		Char Length (cm)	After-flame Time (s)	After-glow Time (s)
XP-0	18.0	Burn out	7.8	25.2
XP-1BL	19.5	Burn out	7.6	24.0
XP-5BL	20.0	Burn out	7.2	20.1
XP-10BL	21.0	Burn out	2.0	4.7
XP-20BL	24.6	Burn out	3.1	2.4

As shown in Table 1, the flame retardancy and vertical flame properties of Xuan paper after treatment by CS/MMT coating were all improved. The LOI value of untreated Xuan paper was only 18.0%, and it increased with the increasing of CS/MMT BLs. The LOI value of XP-20BL just reached 24.6%, which was less than 26%, indicating the flame retardancy of treated Xuan paper could be enhanced but not to a great extent. The result of vertical flame test also supported the same conclusion.

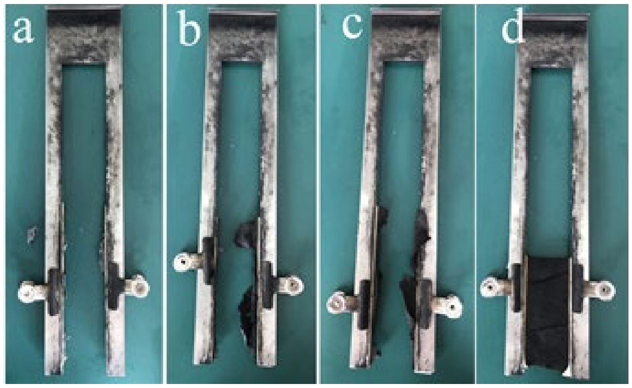

Fig. 4. The digital images of the uncoated and coated Xuan paper after vertical testing: (a) XP-0, (b) XP-5BL, (c) XP-10BL, and (d) XP-20BL

All the Xuan papers were burnt out, while both the after-flame time and after-glow time were reduced with the CS/MMT BLs. They were greatly reduced from 7.8 s and 25.2 s for untreated paper to 3.1 s and 2.4 s, respectively, for the treated paper. The morphology of Xuan papers after burning exhibited a clear difference between the untreated Xuan paper and the treated ones, as shown in Fig. 4. The residual char of Xuan paper after burning increased with the treated CS/MMT BLs. The shape of Xuan paper was completely maintained after the vertical burning when it was treated by 20 BL CS/MMT. This result showed that CS/MMT coating on Xuan paper fiber would promote char formation during the combustion (Rahman *et al.* 2022). Therefore, the flame retardancy of Xuan paper could be enhanced after LBL assembly with CS/MMT coating on pulp fiber, but it is not satisfactory. This might have been because the main flame-retardant component of the CS/MMT coating is the inorganic flame retardant MMT, which formed a barrier layer to inhibit the diffusion of flammable gas and heat transfer, the flame-retardant efficiency of inorganic flame retardants greatly relies on its usage amount. The CS is usually used as the carbon source and blowing agent in the intumescent flame-retardant system, which promotes the char formation of substrate. Hence, the CS/MMT flame-retardant coating should reach a certain amount for the satisfactory flame retardancy of Xuan paper.

Thermal Stability of Xuan Paper

The TG and DTG curves of Xuan papers before and after treatment are shown in Fig. 5, and the corresponding data are given in Table 2.

Table 2. TGA Data of Uncoated and LBL Assembly-coated Xuan Paper Under Nitrogen and Air A

Atmosphere	Samples	$T_{-10\%}$ (°C)	Stage 1		Stage 2		Residue at 800 °C (wt%)
			T_{max} (°C)	Mass Loss Rate at T_{max} (%/°C)	T_{max} (°C)	Mass Loss Rate at T_{max} (%/°C)	
N_2	XP-0	288	348	2.36	488	0.20	0
	XP-10BL	292	349	1.95	505	0.24	13.95
	XP-20BL	270	350	1.47	512	0.22	24.10
Air	XP-0	287	340	2.45	470	0.33	0
	XP-10BL	290	342	1.92	495	0.24	13.90
	XP-20BL	256	338	1.40	498	0.21	23.46

$T_{-10\%}$ is the initial degradation temperature, defined as the temperature of 10 wt% mass loss, and T_{max} is the temperature of maximum mass loss rate.

From Fig. 5a and Table 2, both the untreated Xuan paper and treated papers displayed two mass loss stages under nitrogen atmosphere. The first mass loss stage was due to the Xuan paper to thermal degradation to form the volatile products (levoglucosan) and unstable products. And the second stage of mass loss was due to the unstable products to further decompose forming stable char. The initial degradation temperature ($T_{-10\%}$) of the treated Xuan paper was not influenced when it was treated by CS/MMT lower than 10 BL. The $T_{-10\%}$.was reduced to 270 °C in the case of X-20BL, compared with 288 °C for the untreated one. The lower initial degradation temperature was possibly due to the lower degradation temperature of CS. The mass loss rate at the T_{max} of treated Xuan papers decreased with CS/MMT BL number. The T_{max} of the treated Xuan paper increased, especially the T_{max} of the second decomposition stage. This result showed that CS/MMT coating improved the thermal stability of Xuan paper at higher temperature. The CS/MMT coating on Xuan paper fiber would promote the char formation, as shown by the char residue at 800 °C. The XP-10BL and XP-20BL retained 14.0% and 24.1% char residue at 800 °C, compared with no char residue of untreated paper (Choi *et al.* 2018).

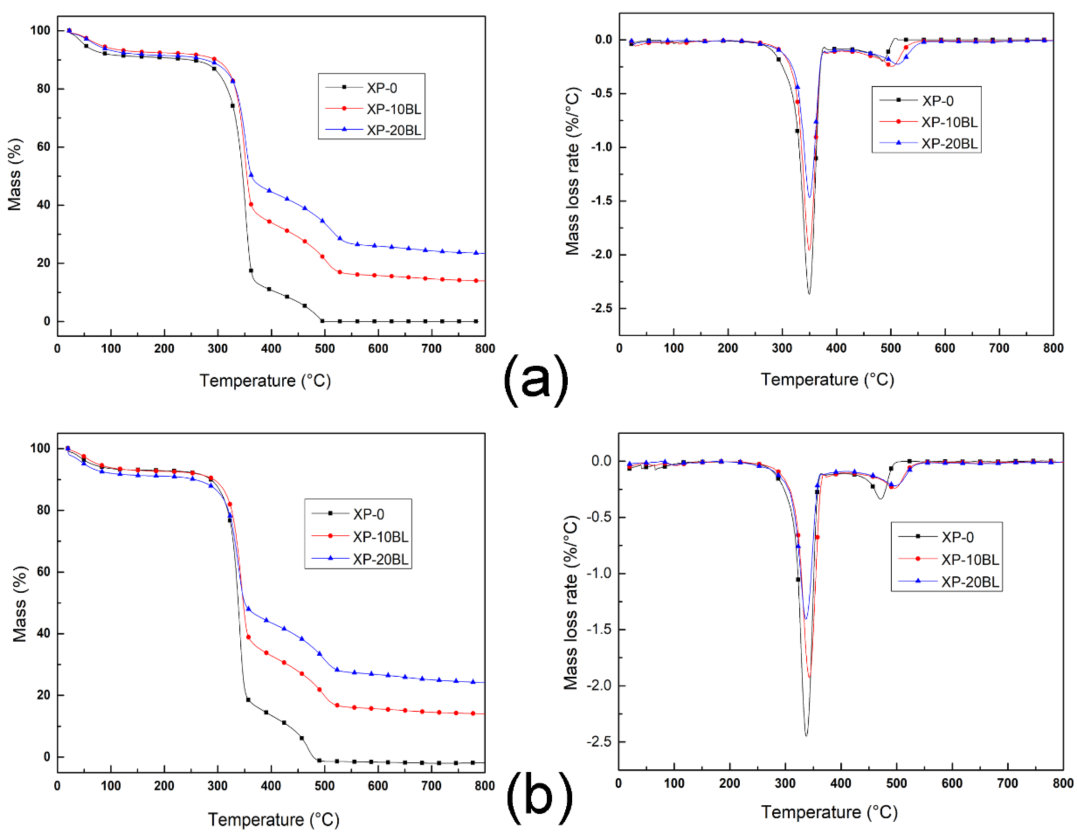

Fig. 5. TG and DTG curves of untreated and treated Xuan papers: (a) under nitrogen and (b) air atmosphere

From Fig. 5b and Table 2, there were also two mass loss stages for both the untreated and treated Xuan papers under air atmosphere. The initial degradation temperature of treated Xuan paper with 20 BL CS/MMT coating also was reduced compared to untreated one, which may be for the same reason under nitrogen atmosphere. The T_{max} and the mass loss rate at the T_{max} of the two stages showed the same tendency with that under nitrogen atmosphere. The char residue at 800 °C for XP-10BL and XP-20BL were 13.90% and 23.46%, respectively. This result also demonstrated LBL assembly treated pulp fiber with CS and MMT could promote the char formation of Xuan paper under air atmosphere

Therefore, LBL assembly treated Xuan paper improved the thermal stability at higher temperature and promoted the char formation both under nitrogen and air atmosphere. This showed LBL assembly treated Xuan paper by CS and MMT mainly act through the condensed phase to form the char layer on their surface to inhibit flammable gas diffusion and heat transfer.

Morphology of the Char Residues

The morphology of the char residues of Xuan papers after vertical flame test is shown in Fig. 6.

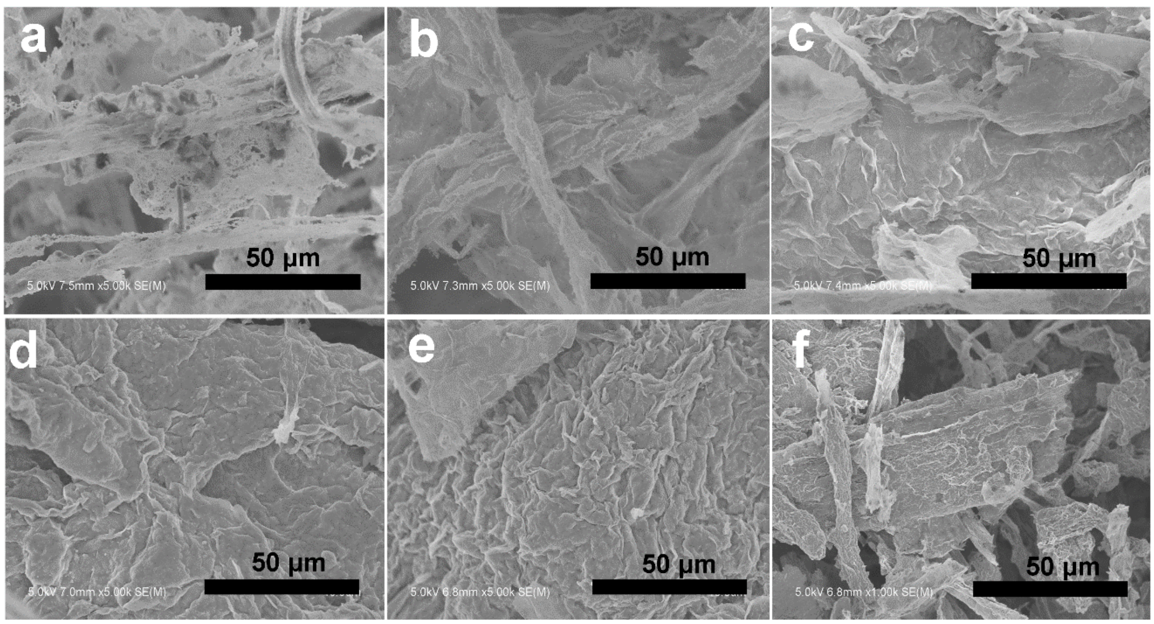

Fig. 6. The morphology of char residuals of untreated and treated Xuan papers: (a) XP-0, (b) XP-1BL, (c) XP-5BL, (d) XP-10BL, (e) XP-20BL (×5000), and (f) XP-20BL (×1000)

As shown in Fig. 6, the untreated Xuan paper could not form char, while the treated Xuan papers could form the stable char after the vertical flame test. There was also the covering layer present on the char residue surface of treated Xuan paper. The covering layer became thicker as the treated CS/MMT BLs was increased. This result showed that the treated Xuan paper promoted the char formation and formed a stable covering layer. The char layer and stable covering layer are beneficial for inhibiting the combustible gas diffusion and blocking heat transmission (Fang *et al.* 2020a). Thus, the LBL treatment was able to improve the flame retardancy of Xuan paper.

Ink Wetting Performance of Xuan Paper

The ink wetting digital images of the untreated and treated Xuan papers are shown in Fig. 7. As shown, ink wetting spreading images on the untreated and treated Xuan papers showed nearly the same spreading area. Even for the Xuan paper fiber treated with 20 BL CS/MMT coating, the spreading area of ink was almost not influenced (Dong and Zhu

2018). Therefore, the ink wetting property of Xuan paper will not be influenced through LBL assembly of flame-retardant treatment of the pulp fibers by CS and MMT. Although the flame retardancy of treated Xuan paper was worse than the LBL assembly coating on Xuan paper by CS and MMT in the authors' previous research (Fang *et al.* 2020b), the ink wetting property was much better.

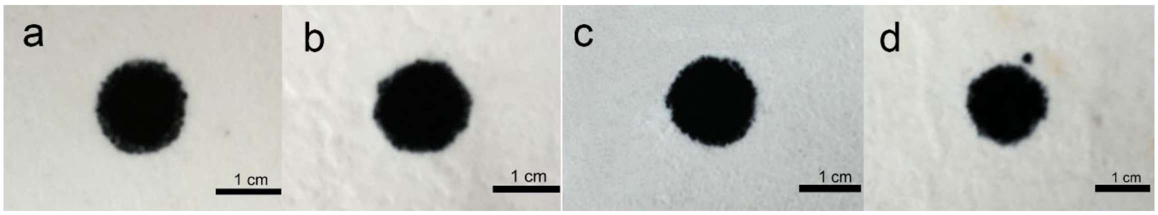

Fig. 7. The digital images of the ink wetting performance of uncoated and coated Xuan paper: (a) XP-0, (b) XP-1BL, (c) XP-10BL, and (d) XP-20BL

CONCLUSIONS

1. Xuan paper was treated to improve its flame retardancy and to reduce the fire risk. In this study, positive charged organic chitosan and negative charged inorganic MMT were layer-by-layer assembled on pulp fiber to fabricate the CS/MMT coating for improving flame retardancy. The FTIR and SEM results of treated Xuan paper showed that the CS/MMT coating was constructed on Xuan paper fiber.
2. Xuan paper fiber treated with 20 BL CS/MMT achieved a LOI value of 24.6%, and its vertical flame properties also improved with less after-flame and after-glowing times compared with the untreated one. The XP-20BL was all burned out, while its shape was completely maintained. These results demonstrated that the flame retardancy of Xuan paper could be enhanced after LBL assembly of the CS/MMT coating on pulp fiber, but it was not satisfactory. This may have been due to poor the flame retardant efficiency of inorganic flame retardant MMT, which greatly depended on its usage amount.
3. The TGA results revealed that LBL assembly of the CS/MT coating on Xuan paper fiber improved the thermal stability at higher temperature and promoted the char formation, showing the obvious condensed phase flame-retardant action. The SEM results further confirmed that the treated Xuan paper by CS/MMT coating on paper fiber promoted the char formation and formed the stable covering layer.
4. Additionally, the ink wetting property of Xuan paper was not influenced through the LBL assembled flame retardant treatment of the pulp fibers by CS and MMT. This research provides a new approach for the flame retardant Xuan paper without influencing its ink wetting property using the eco-friendly organic/inorganic hybrid flame retardant system.

ACKNOWLEDGMENTS

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REFERENCES CITED

ASTM D2863 (2006). "Standard test method for measuring the minimum oxygen concentration to support candle-like combustion of plastics (oxygen index)," ASTM International, West Conshohocken, PA, USA.

ASTM D6413 (2008). "Standard test method for flame resistance of textiles (vertical test)," ASTM International, West Conshohocken, PA, USA.

Chen, C. H., Lin, C. H., Hon, J. M., Wang, M. W., and Juang, T. Y. (2018). "First halogen and phosphorus-free, flame-retardant benzoxazine thermosets derived from main-chain type bishydroxydeoxybenzoin-based benzoxazine polymers," *Polymer* 154, 35-41. DOI: 10.1016/j.polymer.2018.08.003

Chen, X. Q., Pang, G. X., Shen, W. H., Tong, X., and Jia, M. Y. (2019). "Preparation and characterization of the ribbon-like cellulose nanocrystals by the cellulase enzymolysis of cotton pulp fibers," *Carbohydrate Polymers* 207, 713-719. DOI: 10.1016/j.carbpol.2018.12.042

Chen, Z., Xiao, P., Zhang, J., Tian, W., Jia, R., Nawaz, H., Jin, K., and Zhang, J. (2020). "A facile strategy to fabricate cellulose-based, flame-retardant, transparent and anti-dripping protective coatings," *Chemical Engineering Journal* 379, article ID 122270. DOI: 10.1016/j.cej.2019.122270

Choi, K., Seo, S., Kwon, H., Kim, D., and Park, Y. T. (2018). "Fire protection behavior of layer-by-layer assembled starch–clay multilayers on cotton fabric," *Journal of Materials Science* 53(16), 11433-11443. DOI: 10.1007/s10853-018-2434-x

Ding, F., Liu, J., Zeng, S., Xia, Y., Wells, K., Nieh, M. P., and Sun, L. (2017). "Biomimetic nanocoatings with exceptional mechanical, barrier, and flame-retardant properties from large-scale one-step coassembly," *Science advances* 3(7), article ID e1701212. DOI: 10.1126/sciadv.1701212

Dong, L. Y., and Zhu, Y. J. (2018). "Fire-resistant inorganic analogous Xuan paper with thousands of years' super-durability," *ACS Sustainable Chemistry & Engineering* 6(12), 17239-17251. DOI: 10.1021/acssuschemeng.8b04630

Dong, L. Y., and Zhu, Y. J. (2020). "Fire-retardant paper with ultrahigh smoothness and glossiness," *ACS Sustainable Chemistry & Engineering* 8(47), 17500-17507. DOI: 10.1021/acssuschemeng.0c06665

Fang, Y., Zhou, X., Xing, Z., and Wu, Y. (2017a). "An effective flame retardant for poly (ethylene terephthalate) synthesized by phosphaphenanthrene and cyclotriphosphazene," *Journal of Applied Polymer Science* 134(35), article ID 45246. DOI: 10.1002/app.45246

Fang, Y., Zhou, X., Xing, Z., and Wu, Y. (2017b). "Flame retardant performance of a carbon source containing DOPO derivative in PET and epoxy," *Journal of Applied Polymer Science* 134(12), article ID 44639. DOI: 10.1002/app.44639

Fang, Y., Liu, X., and Tao, X. (2019). "Intumescent flame retardant and anti-dripping of PET fabrics through layer-by-layer assembly of chitosan and ammonium polyphosphate," *Progress in Organic Coatings* 134, 162-168. DOI: 10.1016/j.porgcoat.2019.05.010

Fang, Y., Liu, X., and Fei, W. (2020a). "Nacre-mimetic CH/MMT fabrication coating on PET fabric for improving anti-dripping performance," *International Journal of Clothing Science and Technology* 32(6), 803-812. DOI: 10.1108/IJCST-10-2019-0154

Fang, Y., Liu, X., Zheng, H., and Shang, W. (2020b). "Bio-inspired fabrication of nacre-mimetic hybrid nanocoating for eco-friendly fire-resistant precious cellulosic Chinese Xuan paper," *Carbohydrate Polymers* 235, article ID 115782. DOI: 10.1016/j.carbpol.2019.115782

Feng, J., Ma, J., Pang, X., and Tang, B. (2020). "Non-destructive quantitative analysis of nano-mechanics of aged Xuan paper," *Journal of Cultural Heritage* 46, 155-158. DOI: 10.1016/j.culher.2020.06.002

Günister, E., Pestreli, D., Ünlü, C. H., Atıcı, H., and Güngör, N. (2007). "Synthesis and characterization of chitosan-MMT biocomposite systems," *Carbohydrate Polymers* 67(3), 358-365. DOI: 10.1016/j.carbpol.2006.06.004

He, S., Liu, C., Chi, X., Zhang, Y., Yu, G., Wang, H., Li, B., and Peng, H. (2019). "Bio-inspired lightweight pulp foams with improved mechanical property and flame retardancy *via* borate cross-linking," *Chemical Engineering Journal* 371, 34-42. DOI: 10.1016/j.cej.2019.04.018

Köklükaya, O., Carosio, F., Grunlan, J. C., and Wågberg, L. (2015). "Flame-retardant paper from wood fibers functionalized *via* layer-by-layer assembly," *ACS Applied Materials & Interfaces* 7(42), 23750-23759. DOI: 10.1021/acsami.5b08105

Launey, M. E., Munch, E., Alsem, D. H., Saiz, E., Tomsia, A. P., and Ritchie, R. O. (2010). "A novel biomimetic approach to the design of high-performance ceramic-metal composites," *Journal of the Royal Society Interface* 7(46), 741-753. DOI: 10.1098/rsif.2009.0331

Liang, B., Shu, Y., Wan, P., Zhao, H., Dong, S., Hao, W., and Yin, P. (2019). "Genipin-enhanced nacre-inspired montmorillonite-chitosan film with superior mechanical and UV-blocking properties," *Composites Science and Technology* 182, article ID 107747. DOI: 10.1016/j.compscitech.2019.107747

Nassar, M. M., Fadali, O. A., Khattab, M. A., and Ashour, E. M. (1999). "Thermal studies on paper treated with flame-retardant," *Fire and Materials* 23(3), 125-129. DOI: 10.1002/(SICI)1099-1018(199905/06)23:3<125::AID-FAM677>3.0.CO;2-X

Pan, Y., Liu, L., and Zhao, H. (2018). "Recyclable flame retardant paper made from layer-by-layer assembly of zinc coordinated multi-layered coatings," *Cellulose* 25(9), 5309-5321. DOI: 10.1007/s10570-018-1922-0

Rahman, M. Z., Kundu, C. K., Nabipour, H., Wang, X., Song, L., and Hu, Y. (2022). "Flame retardant and hydrophilic coatings on undyed and dyed polyamide 6.6 textiles: Clarifying the processing and performance," *Progress in Organic Coatings* 163, article ID 106600. DOI: 10.1016/j.porgcoat.2021.106600

Sha, L. Z., and Chen, K. F. (2016). "Surface modification of ammonium polyphosphate-diatomaceous earth composite filler and its application in flame-retardant paper," *Journal of Thermal Analysis and Calorimetry* 123(1), 339-347. DOI: 10.1007/s10973-015-4941-1

Shao, Y. T., Zhu, Y. J., Dong, L. Y., and Zhang, Q. Q. (2019). "A new kind of nanocomposite Xuan paper comprising ultralong hydroxyapatite nanowires and cellulose fibers with a unique ink wetting performance," *RSC Advances* 9(69), 40750-40757. DOI: 10.1039/C9RA08349A

Tang, Y., and Smith, G. J. (2013). "Fluorescence and photodegradation of Xuan paper: The photostability of traditional Chinese handmade paper," *Journal of Cultural Heritage* 14(6), 464-470. DOI: 10.1016/j.culher.2012.11.002

Wågberg, L., Forsberg, S., Johansson, A., and Juntti, P. (2002). "Engineering of fibre surface properties by application of the polyelectrolyte multilayer concept. Part I: Modification of paper strength," *J. Pulp Paper Sci.* 28(7), 222-228.

Wang, N., Liu, Y., Xu, C., Liu, Y., and Wang, Q. (2017a). "Acid-base synergistic flame retardant wood pulp paper with high thermal stability," *Carbohydrate Polymers* 178 123-130. DOI: 10.1016/j.carbpol.2017.08.099

Wang, N., Liu, Y., Liu, Y., and Wang, Q. (2017b). "Properties and mechanisms of different guanidine flame retardant wood pulp paper," *Journal of Analytical and Applied Pyrolysis* 128, 224-231. DOI: 10.1016/j.jaap.2017.10.007

Wang, S., Yang, X., Wang, F., Song, Z., Dong, H., and Cui, L. (2019). "Effect of modified hydrotalcites on flame retardancy and physical properties of paper," *BioResources* 14(2), 3991-4005. DOI: 10.15376/biores.14.2.3991-4005

Wu, S., Wu, X. L., and Chu, P. K. (2016). "Ink dispersion on qianlong Xuan paper with improved ink expression," *Journal of Materials Science & Technology* 32(2), 182-188. DOI: 10.1016/j.jmst.2015.12.014

Xu, F., Zhong, L., Xu, Y., Feng, S., Zhang, C., Zhang, F., and Zhang, G. (2019). "Highly efficient flame-retardant kraft paper," *Journal of Materials Science* 54(2), 1884-1897. DOI: 10.1007/s10853-018-2911-2

Yang, F., Zhang, Y., and Feng, Y. (2017). "Adding aluminum hydroxide to plant fibers using *in situ* precipitation to improve heat resistance," *BioResources* 12(1), 1826-1834. DOI: 10.15376/biores.12.1.1826-1834

Zhou, R., Ming, Z., He, J., Ding, Y., and Jiang, J. (2020). "Effect of magnesium hydroxide and aluminum hydroxide on the thermal stability, latent heat and flammability properties of paraffin/HDPE phase change blends," *Polymers* 12(1), article no. 180. DOI: 10.3390/polym12010180

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