

Permeation Characterization of the Sequence of Intersecting Ink and Seal Lines Using Field Emission Scanning Electron Microscope

Jiangchun Li^{1†}, Ziyi Wang^{2†}, Xudong Yang^{2†}, Mengyang Lu^{2†}, Xiaobo Xiong^{1*}, Yongqian Wang^{2*}

¹The Key Laboratory of Forensic Science, Hubei University of Police, Wuhan, Hubei, China

²Engineering Research Center of Nano-Geomaterials of Ministry of Education, Faculty of Materials Science and Chemistry, China University of Geosciences, Wuhan, Hubei, China

†Co-first Author

*Correspondence Author

Abstract

Objectives: Paper documents are playing an increasingly important role in people's daily work with the development of economy, society and culture. In the practice of judicial appraisal, the sequence of the intersections of ink and seal on suspicious documents can often provide critical information for the detection of criminal cases. The examination of sequence of intersecting seal and ink lines is to judge the sequence of seal and ink mark formation by certain technical means. *Methods:* A representative black signature pen, ink, and specific paper are selected to prepare experimental samples. Under the given experimental conditions, the field emission scanning electron microscope is used to perform micro-morphology on the cross-sectional characteristics of the samples and all the characterization results obtained are systematically analyzed to summarize the specificity of the sample. *Results:* The results showed that the proposed method can efficiently discriminate the Permeation Characterization of the sequence of intersecting seal and ink lines. *Conclusions:* This research is expected to be applied to forensic investigation for counterfeiting documents and bring new developments in the field of document inspection.

Keywords: Forged documents, sequence, intersecting lines, preparation, FESEM

I. Background

With the continuous improvement of legal society, the application scope of contracts, official documents, bills and other documents in daily life is gradually expanding, as well as increasing demand for authenticity judgment of such documents^{1, 2}. However, due to the development of modern forging technology, the field of document identification has faced many challenges over the years. The overlapping sequence of all kinds of handwriting and ink has become one of the important contents in document inspection and forensic science.

The sequence of intersecting lines is defined as the sequence of stamping and writing, printing or facsimile writing³⁻⁵. The examination of the intersecting lines is to judge the sequence of seal and ink by certain technical methods. Ink, seal and paper interacting with each other forms the basis of examining the sequence of intersecting lines when making documents⁶. The contact between the printer, pen and paper is an important factor affecting the property of the text as well. Meanwhile, the time of document printing, stamp and cross interface formation also affect sequence of intersecting lines⁷.

The main task of sequence of intersecting lines detection is to determine the order of the overlapping of handwriting and seals. Due to the different composition and preparation methods of various inks and seals, their overlapping parts will also present different characteristics. If there is a significant difference between the composition and production process of the two dyes at the intersection, it is easier to judge its overlap order⁸⁻¹⁰. Therefore, the determination of the sequence of crossing stroke can provide important information when investigating a fraud. At present, the research methods are as follows: microscopic observation^{11, 12}, fluorescence microscopy¹³, stripping and layer reduction¹⁴, spectral analysis¹⁵⁻¹⁸, etc. In addition to the limitations caused by the

intermiscibility and permeability of printing ink, the current research direction also limits the development of sequence of intersecting lines detection^{19, 20}. At present, the focus of sequence of intersecting lines detection method is mostly the analysis of physical and chemical properties of the cross interface, but there are few studies on sequence of intersecting lines in the non-cross state and different preparation time²¹.

To this aim, this paper focuses on the overlap order of ink and seal, analyzes the characterization and mechanism of the cross-interface over time, and look for the change rules after the cross interface is formed. This research provides a new test method for judging sequence of intersecting lines, so as to prove the authenticity of the document and judge the relative formation time of the document. At the same time, our results will shed light on document examination via a simple and efficient way and provide technical support for the court in economic and civil dispute cases.

II. Experimental

2.1 Materials

The black marker pen with the writing thickness of 0.5 mm, red round oil-based inkpad and 70 g/m² multifunction typing paper was produced by Deli group (China). Other experimental supplies have no obvious influence on the sample preparation process, so they are not screened in detail.

2.2 Experimental procedure

2.2.1 The preparation of samples written after sealing

Sample preparation method is as follows. Use a black marker pen on blank A4 paper to make three samples of the same content. Then the seal was used to stamp one of the samples when ink surfaces without moisture and stamp on another one three days later. A total of three samples named sealed sample, sample written immediately after seal and sample written three days after sealing were obtained, each one was stored in the natural environment of the laboratory. All specimens were stamped or written by the same person with similar and normal pressure (in accordance with the usual habits, rather than particular, to increase or decrease the pressure) under no other specific requirements.

2.2.2 The preparation of samples sealed after writing

Sample preparation method is as follows. Use a stamp pad on blank A4 paper to make three samples of the same content. Then the black marker pen was used to write on one of the samples when ink surfaces without moisture and write on another one three days later. A total of three samples named written sample, sample sealed immediately after writing and sample sealed three days after writing were obtained, each one was stored in the natural environment of the laboratory. All specimens were stamped or written by the same person with similar and normal pressure (in accordance with the usual habits, rather than particular, to increase or decrease the pressure) under no other specific requirements.

2.3 Characterization

The experiment used field emission scanning electron microscopy (FESEM, SU8010, Hitachi, Japan) directly observes the cross-section of the sample. The acceleration voltage is 20 kV, and the samples were stored in vacuum and room temperature.

2.4 Attention of experiment

By completing the experiment on the sequence of the intersections of ink and seal with time, we summarized the operational considerations of this experiment as follows:

- i)When preparing the sample, the upper surface of the sample section must be at the same level as the top surface of the sample table, otherwise it is easy to damage the section of the sample paper, thus affecting the detection.
- ii)Within the range of the shooting horizon, it should be possible to find typical features for the next search.
- iii)When taking a picture, first to find the morphology of the sample with common characteristics of the area in a lower multiple where all morphology features of each place are uniform, and then magnify the multiple to find a reference point as a special position.

III. Results and Discussion

3.1 The change rule of written sample

As shown in Figure 1, the cross-section morphology of the written sample was analyzed by FESEM. It can be clearly seen that the paper structure at the section has a regularity change. From Figure 1f, cross-section appeared some burr sharp fiber. And in Figure 1g, the burr shape fiber became denser and more uniform. In addition, morphology of Figure 1h had obvious changes that fiber swelling obviously increased and began to appear section collapse. At the same time, the paper was still stable and had no obvious change within the visible depth of field allowed by FESEM. Preliminary analysis, the situation is due to the ink volatilization.

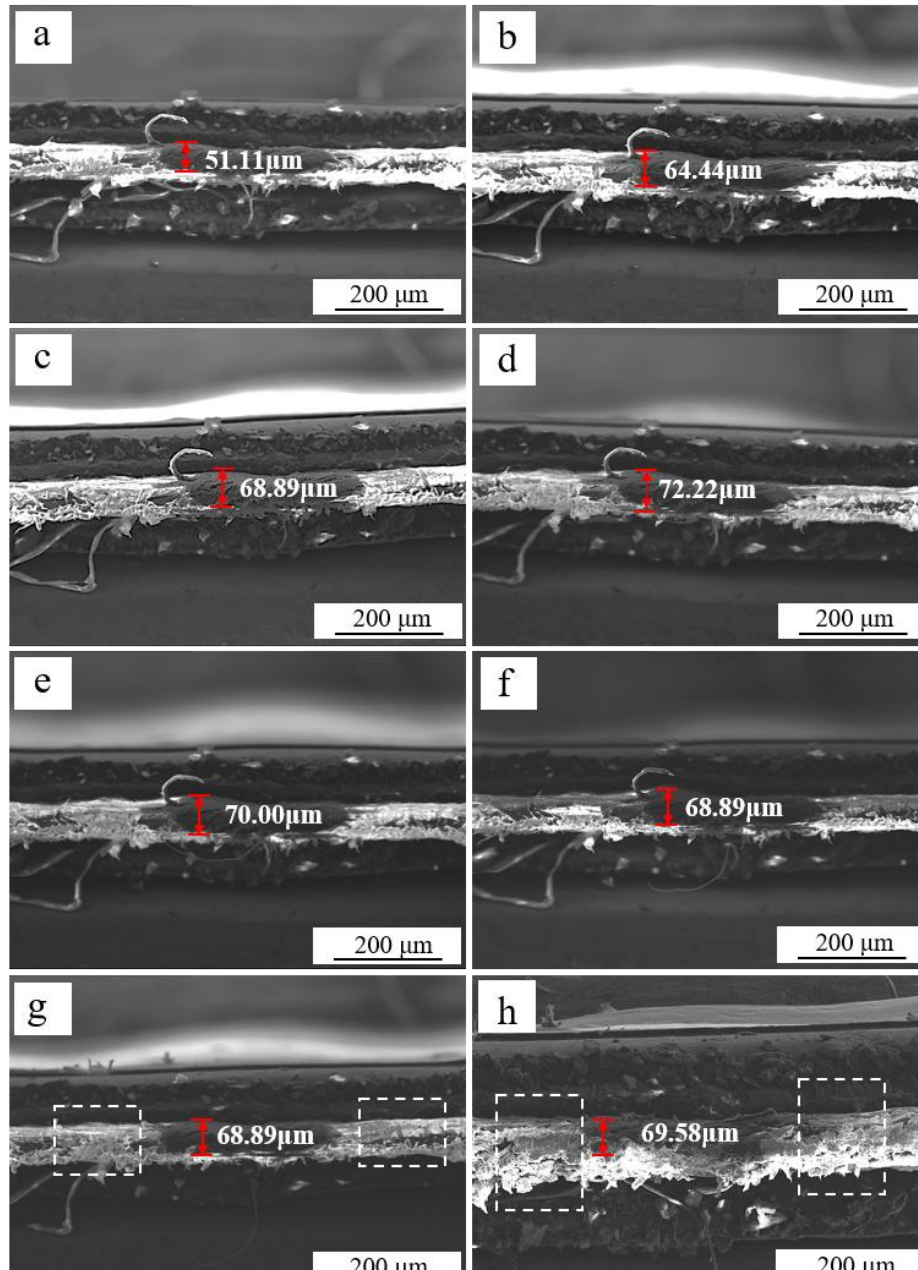


Fig 1: The cross-section morphology of written sample with time
(a) 1st day, (b) 3rd day, (c) 5th day, (d) 7th day, (e) 9th day, (f) 14th day, (g) 21st day, (h) 35th day

By analyzing the relative permeability (ΔH) change rule of written sample with time (Figure 2), the relative permeability curve of written sample can be obtained by calibrating and measuring the boundary of the permeation surface. It can be seen that the ink is evenly distributed in part of the section of the sample and the obvious boundary of the permeation surface can be observed in the section morphology. Meanwhile, ΔH of the sample grew with the preparation time over a period of time. On the 7th day, it began to decline to a certain extent and then reached a relatively stable state.

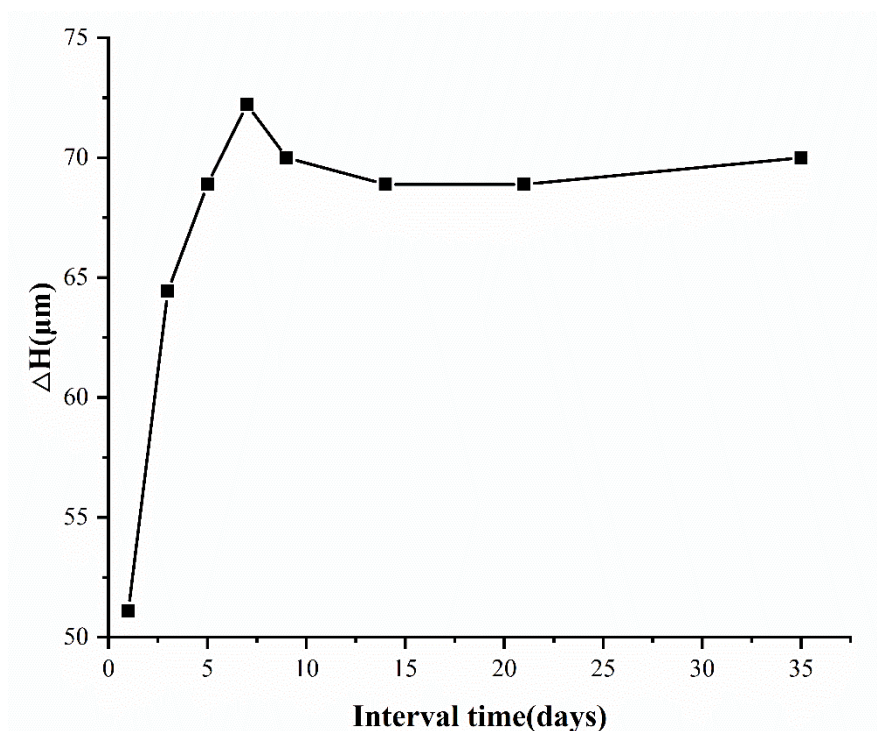


Fig 2:Relative permeability (ΔH) change rule of written sample with time

3.2 The change rule of sealed sample

Figure 3 showed the change rule of cross-section morphology of sealed sample. It can be seen that the penetration of seal in the depth of the paper is more uniform and the penetration ability is larger than the ink. On the 7th day (Figure 3d), the degree of paper fiber exposure at the seal section increased, which was significantly different from the burr paper as shown in Figure 1d. At the same time, the sealed cross section of 21st day (Figure 3g) was mottled and the fibers were stripped in clusters. In Figure 3f, it was obvious that only a small part of the section morphology of sample had section collapse, and more than 50% of the section morphology was stable.

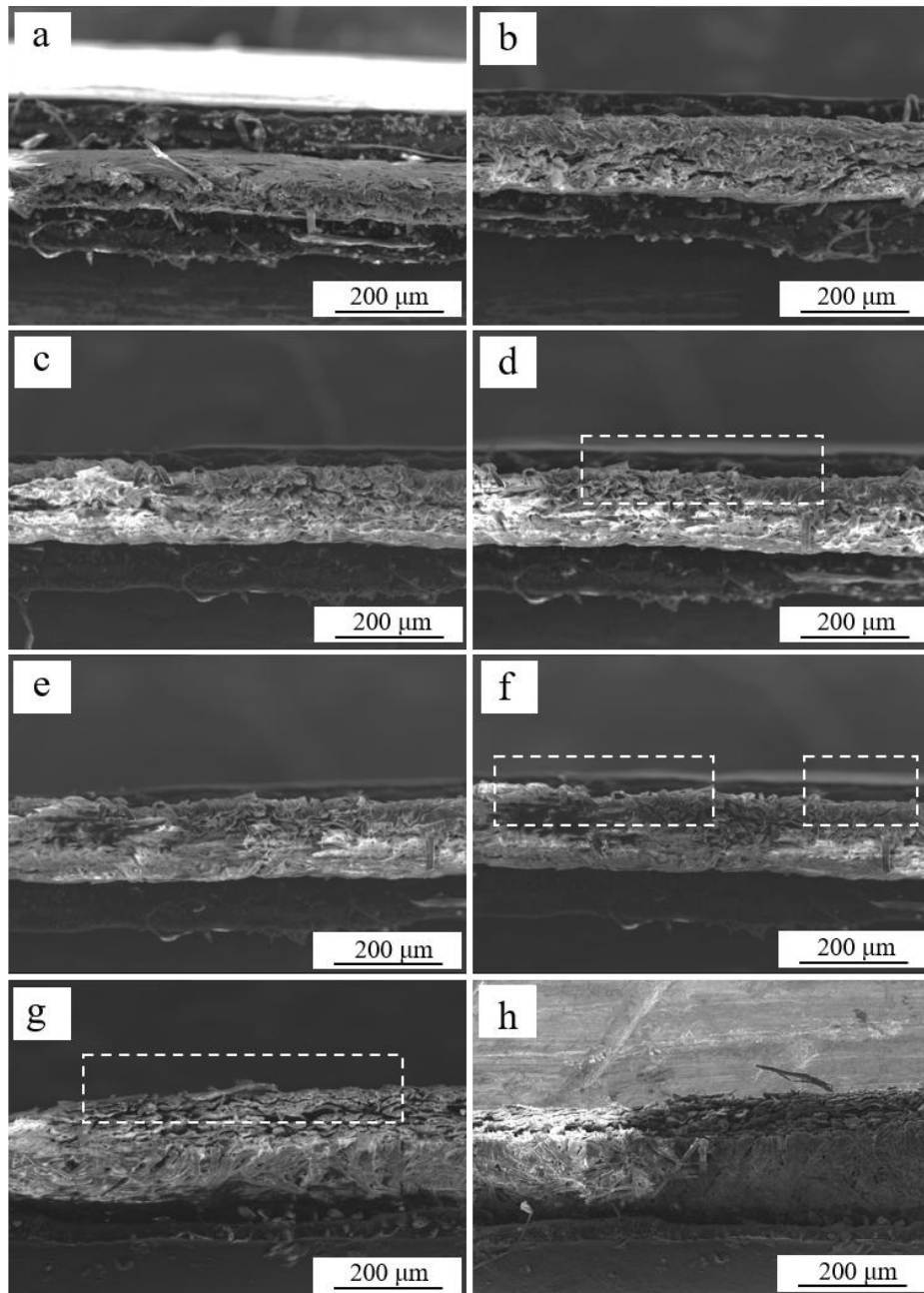


Fig 3: The cross-section morphology of sealed sample with time
(a) 1st day, (b) 3rd day, (c) 5th day, (d) 7th day, (e) 9th day, (f) 14th day, (g) 21st day, (h) 35th day

3.3 The change rule of samples sealed after writing

3.3.1 The change rule of sample sealed immediately after writing with time

Firstly, by comparing the blank area of the section with the cross area of ink, it can be seen that the microscopic morphology of the blank area without seal and ink penetration is stable, and there is still no obvious change when the sample is prepared on the 21st day (Figure 4g). However, in Figure 4d, the cross part showed significantly different from the 7th day of sample preparation. On the 35th day (Figure 4h), the microscopic morphology changed obviously because of the diffusion of ink and seal to blank area. The paper fiber section of the original cross area presented block-like peeling.

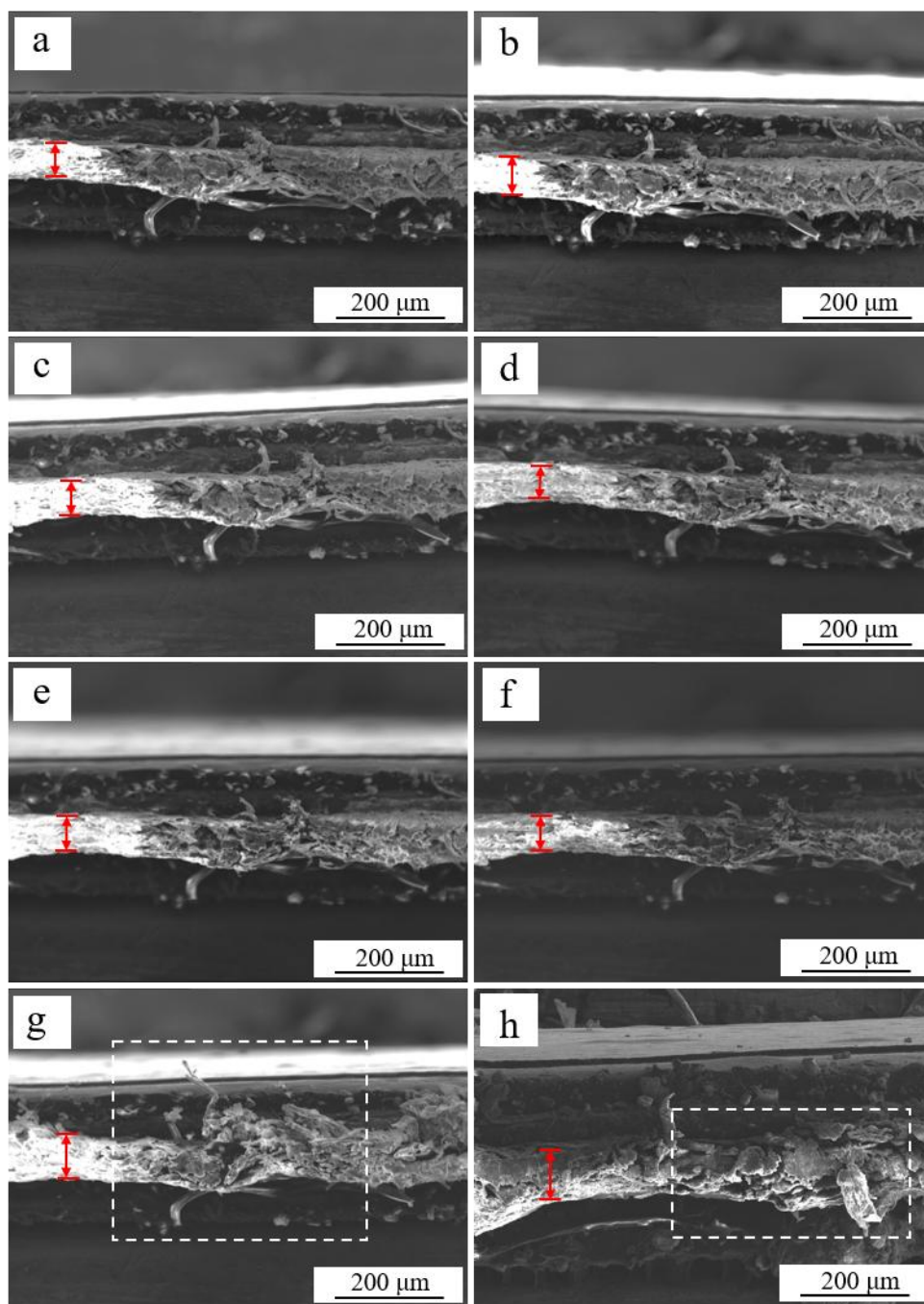
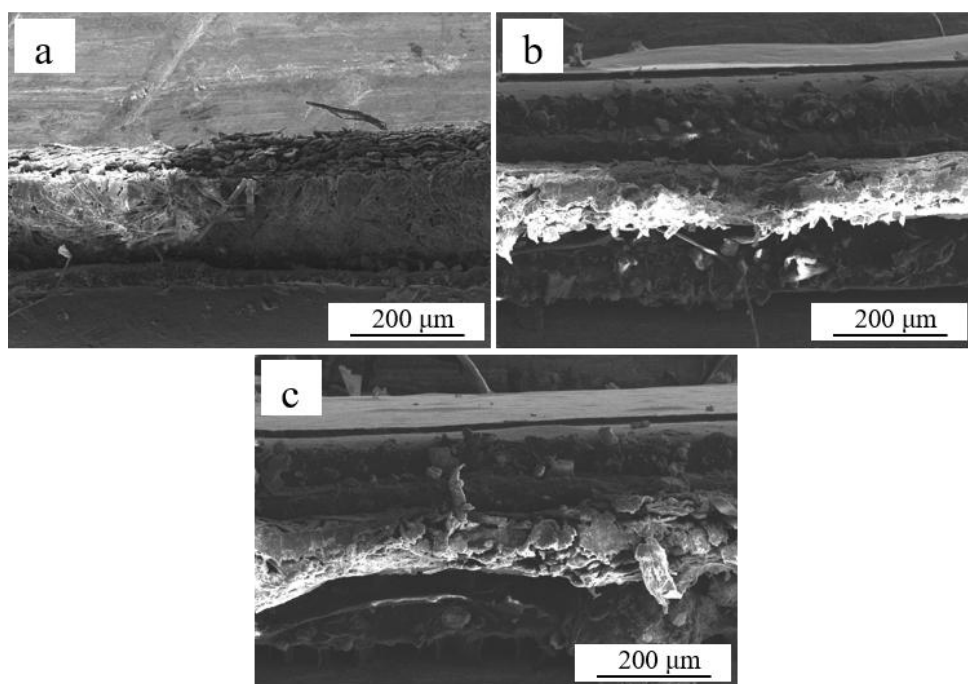


Fig 4: *The cross-section morphology of sample sealed immediately after writing with time (a) 1st day, (b) 3rd day, (c) 5th day, (d) 7th day, (e) 9th day, (f) 14th day, (g) 21st day, (h) 35th day*

As showed in Figure 5, by comparing and analyzing the cross-section morphology of the sample, it can be seen that the influence of seal and ink on the section of the paper is mainly caused by the evaporation of ink and the deposition of solute. The variation degree of the cross-section morphology of the paper is as follows: the cross section of ink and seal > ink > seal > blank.



***Fig 5: Comparison cross-section morphology of samples on 35th day
(a) sealed sample (b) written sample (c) sample sealed immediately after writing***

3.3.2 The change rule of sample sealed three days after writing

FESEM was used to observe the change rule of cross section morphology of sample sealed three days after writing with time. By observing Figure 6, it can be known that after writing on the paper, the ink penetrated at a certain depth vertically. In Figure 6g, the cross-section morphology of the sample did not change significantly. Preliminary analysis shows that after writing on paper, the water-based medium in the ink could not evaporate within a short time due to the adsorption effect of the paper on the ink.

During the sample preparation, the water-based medium in the ink has not completely volatilized, therefore, there is no obvious influence on the cross-section morphology. Figure 6h showed that the cross-section morphology of the cross area remained unchanged. It can be reasonably inferred that when the ink is completely penetrating the paper, the sealing on the ink can form a dense oil film, effectively blocking further evaporation of ink moisture. Hence in a long period of time, the cross-section morphology is in a stable state.

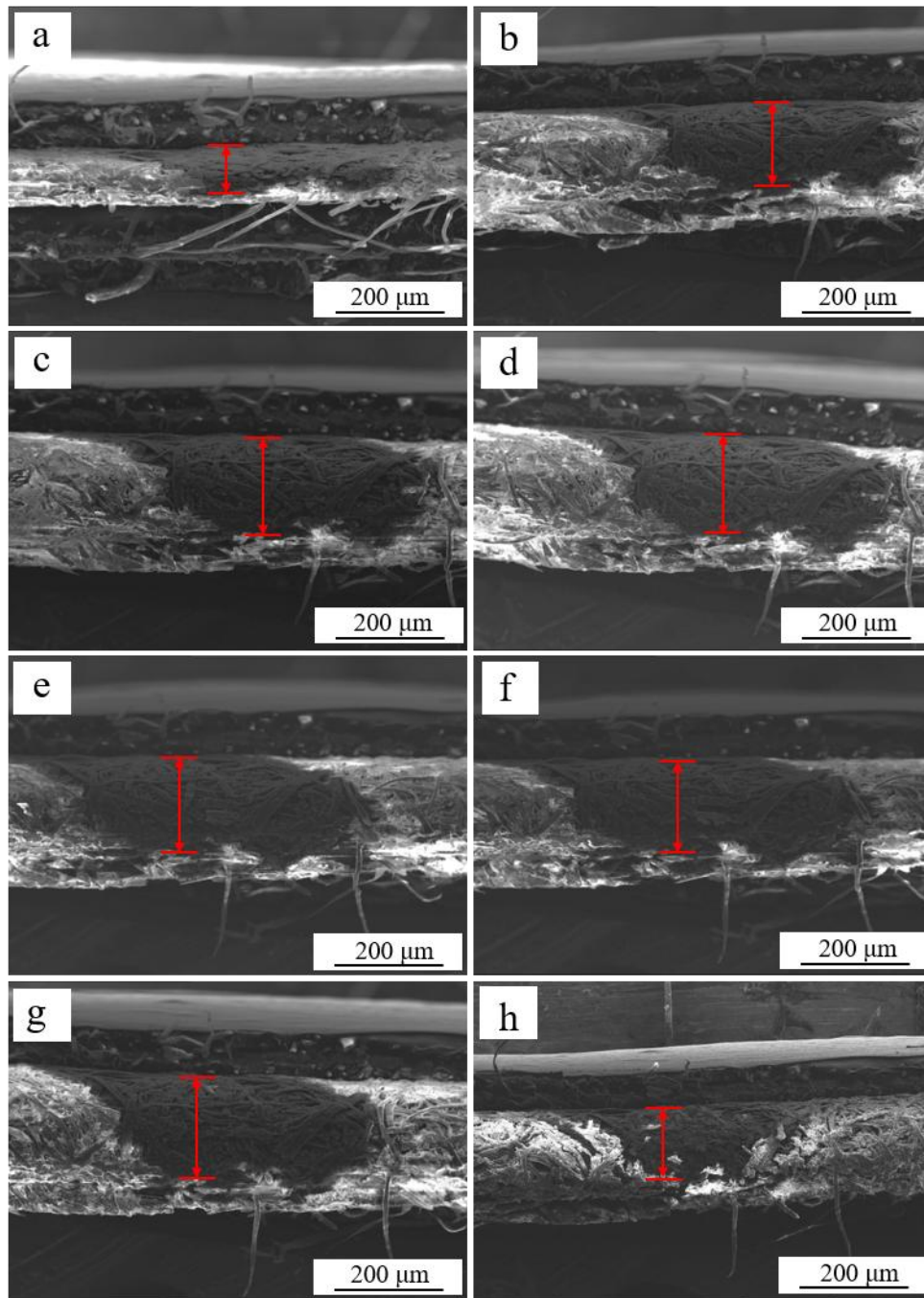


Fig 6:*The cross-section morphology of sample sealed three days after writing with time (a) 1st day, (b) 3rd day, (c) 5th day, (d) 7th day, (e) 9th day, (f) 14th day, (g) 21st day, (h) 35th day*

3.4 The change rule of samples written after sealing

3.4.1 The change rule of sample written immediately after sealing

The experimental results in Figure 7 were represented as microscope images of the section morphologies of the sample. It can be seen from the picture that on the 3rd day after it was made (Figure 7b), the cross-section microstructure changed greatly, but there was no obvious boundary between the degree of interpenetration of seal and ink. However, after 21st day, the surface morphology of the sample changed greatly and the paper surface appeared bulges and fibrous peeling (Figure 7g).

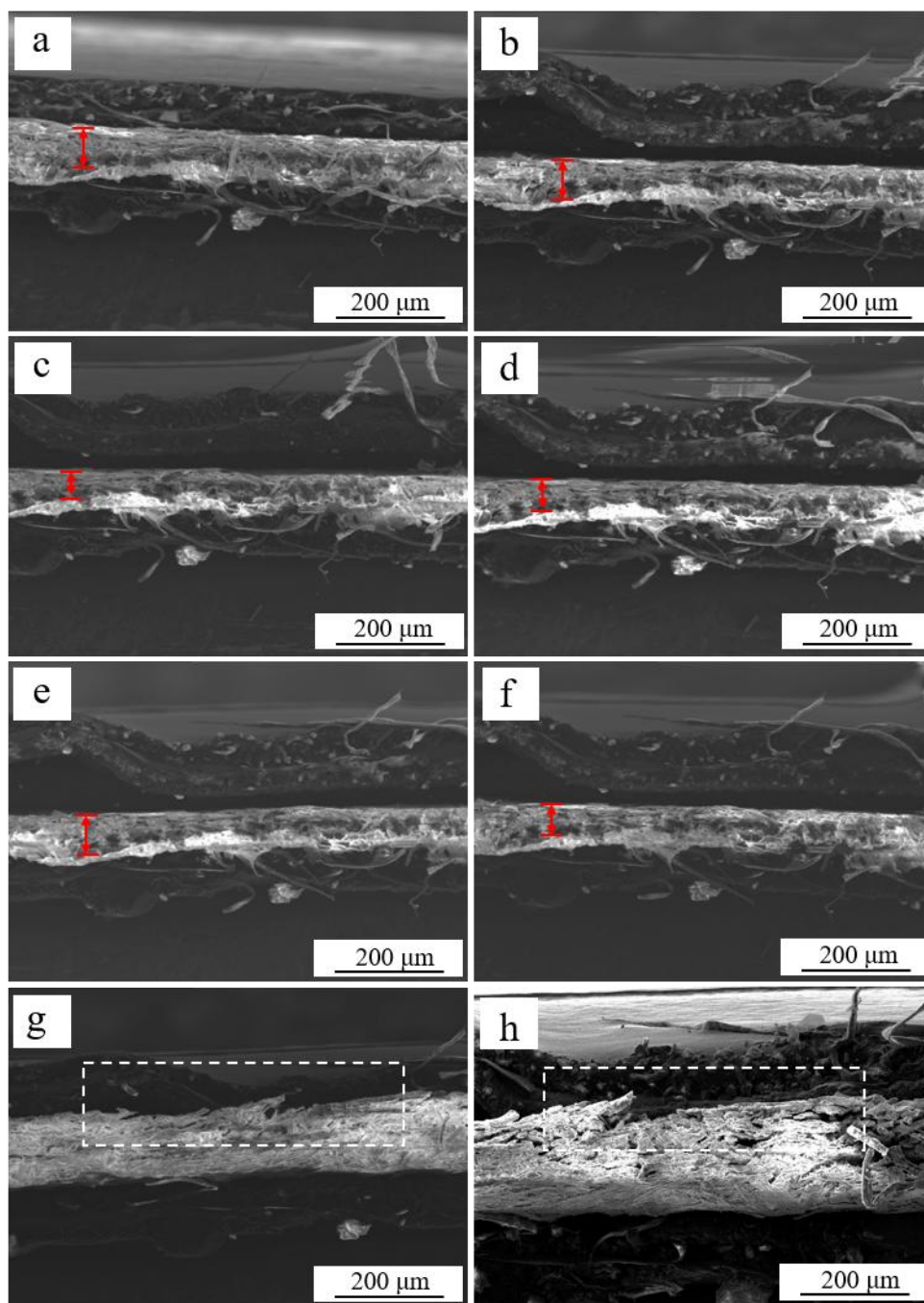


Fig 7: *The cross-section morphology of samples written immediately after sealing with time (a) 1st day, (b) 3rd day, (c) 5th day, (d) 7th day, (e) 9th day, (f) 14th day, (g) 21st day, (h) 35th day*

As shown in Figure 8, we compared cross-section morphology of different samples on 3rd day. It can be preliminarily determined that a dense oil film with a certain covering area formed on the sample surface after the paper was sealed, which prevented further penetration of ink after writing. In the storage environment of this experiment, the water-based medium in the ink naturally volatilized on the paper surface, so that the surface morphology of the paper with ink changed significantly, while the lower surface did not have marked change. This further confirms water-soluble inks in the evaporation rate faster than oil-soluble seal in paper fiber.

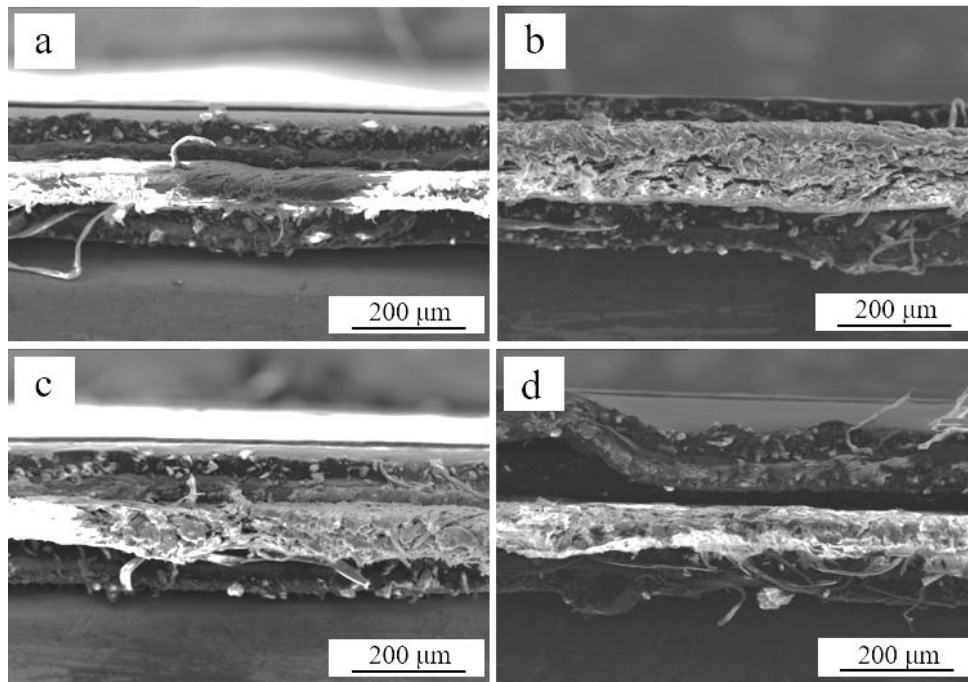


Fig 8: Comparison cross-section morphology of samples on 3rd day
(a) written sample, (b) sealed sample, (c) sample sealed immediately after writing, (d) sample written immediately after seal

3.4.2 The change rule of samples written three days after sealing

FESEM studies are carried out to study the section morphologies and Figure 9 showed the change rule of samples written three days after sealing with time. As can be seen from the Figure 9h, on the 35th day of sample preparation, there was no obvious change on the cross section. Figure 7 and Figure 9h showed the results which reveal that surface morphology on the cross section is greatly different and the change in the middle area is roughly the same, which may be because in Figure 7h when the seal film is initially formed, the ink makes the surface film damaged and its distribution is uneven. However, in the secondary formation process of the oil film, the uneven printing oil distribution causes a greater degree of damage to the paper surface structure. But three days later, the oil film has been completely formed on the surface of the paper and the water is basically all volatilized while ink in the process of permeation and volatilization has only a certain effect on the middle section of the paper.

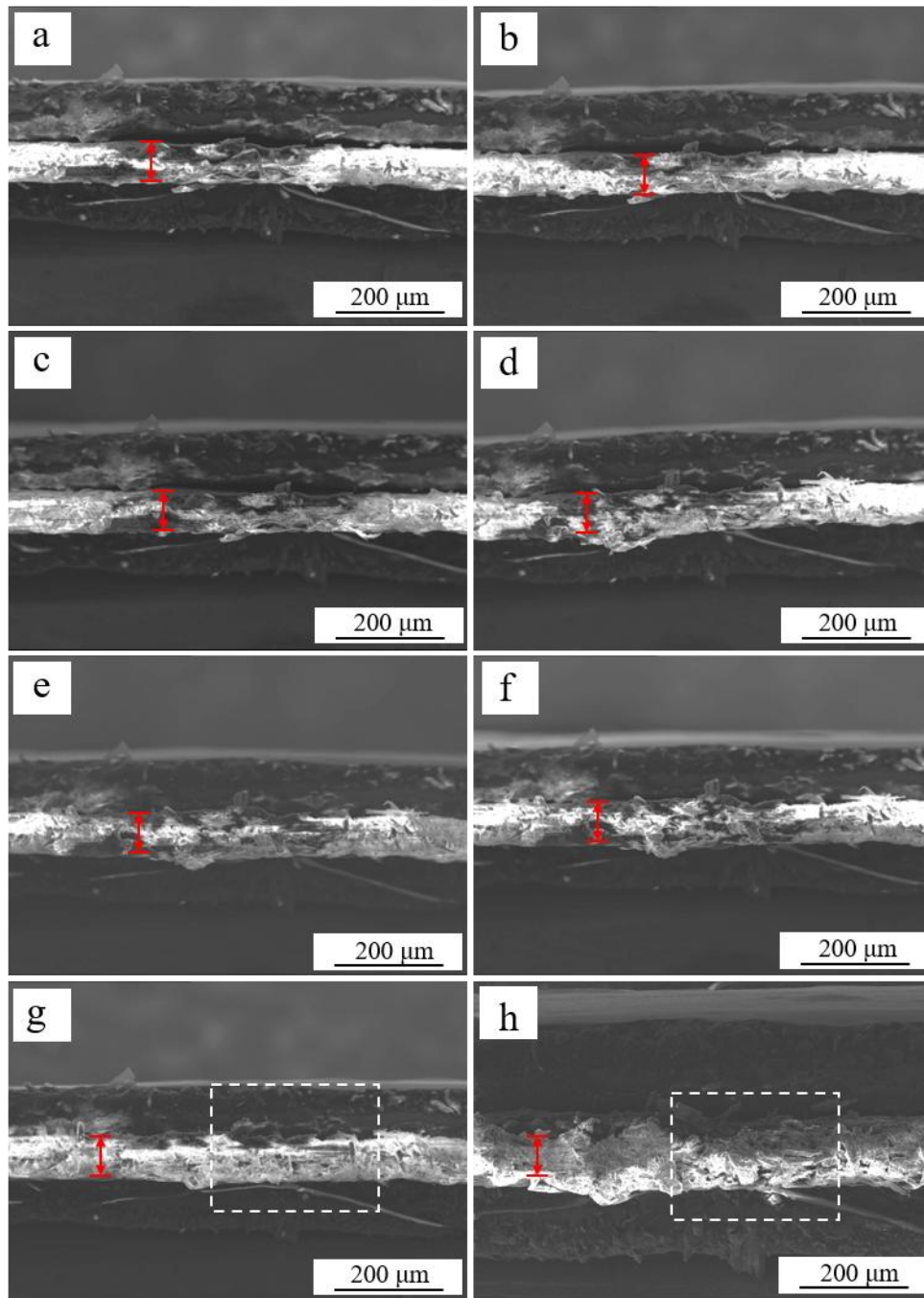


Fig 9: *The cross-section morphology of sample written three days after sealing with time (a) 1st day, (b) 3rd day, (c) 5th day, (d) 7th day, (e) 9th day, (f) 14th day, (g) 21st day, (h) 35th day*

IV. Conclusions

In summary, this study presents a new method of distinctively analyzing the sequence of intersecting ink and seal lines using SEM. On the basis of characterization results, we systematically observed the change rule of different samples. For the control group, seal can permeate the whole section quickly and evenly while the section penetration depth of ink with time has a certain change and it reached the maximum in the 7th day.

By analyzing the sequence, we found that when the samples sealed after writing preparation time was less than 3 days, the surface of the cross section had no obvious change, and the lump-like paper fiber peeling began to appear in the middle section after being stored for a period of time. When the sample preparation time is greater than or equal to 3 days, the cross-section morphology is in a relatively stable state and there is a relatively obvious boundary between ink and seal. Similarly, when the samples written after sealing preparation time is less than 3 days, the paper fibers on the cross-section are exposed and the middle fibers are stripped. When the seal sample preparation time is more than 3 days, in the longer preservation time, the section has no obvious change, and the boundary between ink and seal is not distinct.

It further shows that the research can determine the formation time of documents and provide technical support for the trial of court in economic and civil dispute cases.

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