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**Abstract:**  $\mu$ -XRF is one of the powerful non-destructive analytical techniques in forensic science application. Thanks to the elemental mapping and the spectrum search function, XGT-9000 enables to reveal intentional alternation and identify the ink used on a document.

**Keywords:**  $\mu$ -XRF, Forensic science, Ink identification, Black ink, Spectrum search

## Introduction

Identifying types of ink on handwriting texts with the analytical instrument is significant in forensic science because it makes it possible to clarify its authentication and origin. The difference in the chemical composition of inks reveals an intentional alternation on the document.

Recent research has reported the investigation of inks using analytical techniques.<sup>[1,2,3]</sup> Our previous study reported that a Raman microscope allowed us to identify two different black printer ink types.<sup>[4]</sup> However, substances in ink are subject to damage by Raman laser, which makes the laser set in low power. In this case, the acquisition time gets lengthy. XRF is one of the elemental analysis techniques to differentiate inks by the inorganic constituents. The advantages of  $\mu$ -XRF are as follows: non-destructive technique, higher signal intensity and faster imaging of ink sample than Raman.

In this application note, the X-ray Analytical Microscope XGT-9000 (Figure 1) allows one to differentiate and identify black inks using elemental mapping and spectrum search function.



Figure 1. XGT-9000

## Sample Preparation and Experimental Condition

Five commercial black pens (Table 1) were prepared to obtain the spectra and form a database for the spectrum search function. Point analysis was conducted on each ink line written on paper with an ultra-high intensity probe for 100 sec under full vacuum condition.

Table 1. Description of black pens used to form a database.

ID	Brand	Model
a	Company-P	Model-L
b	Company-M	Model-6
c	Company-Z	Model-M
d	Company-T	Model-M
e	Company-P	Model-B

The “HORIBA” characters were written with two different black pens (ink-c and ink-e in Table 1) on paper. Elemental mapping was performed on the area covering the character with an ultra-high intensity probe under full vacuum condition. The pixel time was 30 msec. The primary X-ray generator was operated at 30 kV and 1000  $\mu$ A in all the measurements.

## Elemental Maps to Visualize Intentional Alternation

Figure 2 shows the optical image and elemental maps. Al, S, and Fe were detected in a part of the paper. The elements are known as the composition of the fillers in the paper. Ca and Ti were detected in all the areas. These elements are used in this paper as  $\text{CaCO}_3$  and  $\text{TiO}_2$ . Cr and S displayed specific distributions on the different area of the “HORIBA” characters. It suggested that two different inks were used to write the characters.

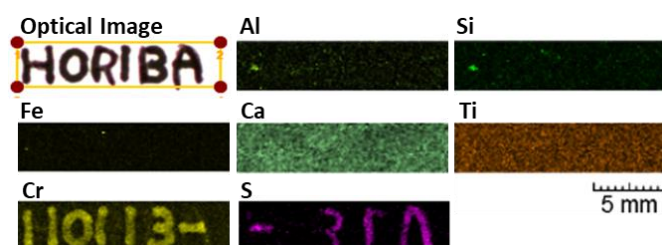


Figure 2. The optical image and elemental maps of the HORIBA characters on paper acquired with XGT-9000.

## Spectrum Search to Identify Ink used on the Handwriting

Spectrum search is a function of XGT-9000 software to load a spectrum of an unknown material and search the best hits from the spectra of known samples. This function helps to identify unknown materials.

To identify ink used to write the HORIBA characters, the spectrum search function was applied to the two extracted spectra on the layered image in Figure 3 (a). The spectrum in yellow and the spectrum in pink shown in Figure 3 (b) were extracted from the Cr-rich area and the S-rich area, respectively. The five commercial black ink spectra in Table 1 are also shown in Figure 3 (b). It is difficult to find the most similar spectrum to each extracted spectrum visually.

(a) Layered image (Cr-map and S-map)

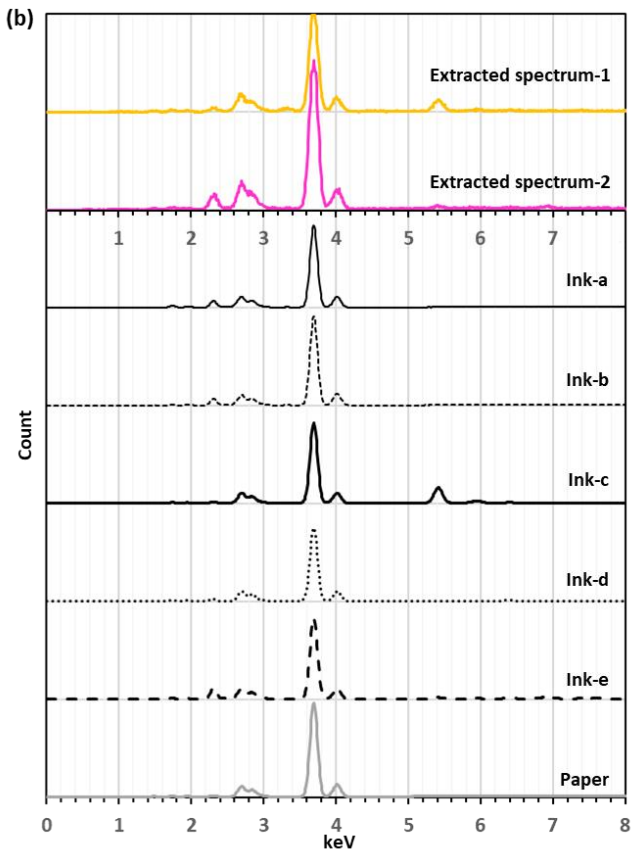
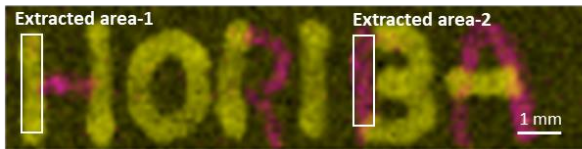


Figure 3. (a) The layered map of Cr map and S map (b) The extracted spectra from the map and the spectra of the five commercial black ink and paper.

Figure 4 shows the similarity results of the five spectra to the two extracted spectra using the spectrum search function. The smaller value in Figure 4 means higher similarity to the extracted spectrum. It suggests that the ink-c and ink-e are the most possible ink used in the HORIBA characters. The result was consistent with our sample preparation.

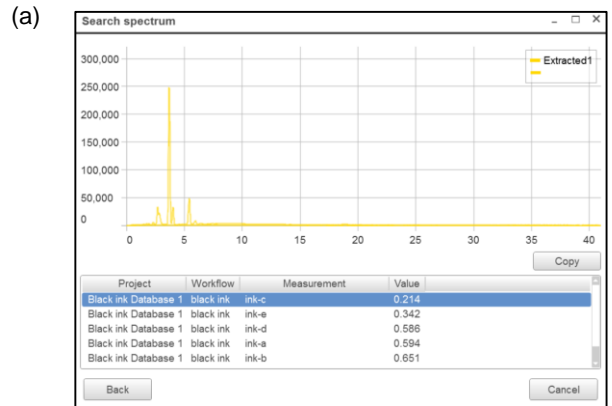


Figure 4. The similarity results of spectrum search for (a) the extracted spectrum-1 and (b) the extracted spectrum-2.

## Conclusion

XGT-9000 is a powerful non-destructive tool to visualize intentional alternation and to identify ink used in a document.

## Reference

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