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Migration of luminescent ink components, a new approach for ink dating

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ABSTRACT

Many inks of wet stamps and of writing instruments such as ballpoint pens, felt-tip pens or roller pens contain luminescent components. Optical comparison of two or more unknown inks with the aid of IR-camera systems is a standard method in questioned document examination. It has been observed, that these components may migrate within the paper and especially along the stroke of an intersected line with another ink. In this study we describe and discuss the migration of these components.

We could show that migration data can be used for ink dating.

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1. Introduction

The determination whether or not a document was written or signed at the date on the document or if it has been produced later (backdating) is a frequent question in forensic document labs. This had been an unsolved problem for many decades until the solvent loss method was introduced [1–6]. Even though that the amount of phenoxyethanol and/or phenoxyethoxyethanol within ballpoint-pen ink strokes decreases with the age of a document, it is still a challenge to apply the solvent loss method. The main problem is, that solvent evaporation processes strongly depend on the composition of the ink and the paper [7]. Regression on an ageing curve of a questioned entry will not be successful, since the amount of the initial solvent is unknown and may strongly vary from ink to ink [8,9]. As in other comparative analyses and evaluating reports, it is the state of the art to work with hypotheses; e.g.:

Hypotheses 1 (H1): The date of the entry corresponds to the date on the document;
Hypotheses 2 (H2): The entry was applied after the date on the document, thus, the document has been backdated.

The document expert has to evaluate the probability to get the measured data if H1 is true and if H2 is true. Finally, he has to estimate the likelihood ratio to give his expert opinion. If it is undisputed that the questioned entry is older than a few months [10] (e.g., if the document was confiscated some months before it could be analyzed), evaporation processes are usually terminated and the solvent loss method cannot be applied anymore [11].

In the early 20th century, chloride and sulfate ion diffusion from iron gallus ink into the paper was studied for questioned document dating [12,13]. This was the first material migration method. The disappearance of iron gallus ink from the marked was the end of the chloride/sulfate diffusion method application.

In the early 21st century, José Balbuena started to analyze different optical behaviors on intersecting lines, depending on their sequence. He introduced a light system to determine the sequencing of any inks [14]. During this work, he also observed a luminescent zone beside the line on several inks.

Inks showing this phenomenon were used to study the migration process and to check, if the migration data can be used for ink dating.

As the luminescent zone is only detectable with the aid of equipment such as a fluorescent microscope or a digital camera/filter system, we call it – in this paper – "non-visible". The ink color is called "visible" when it is with the naked eye. Furthermore, we usually talk in this paper about "luminescence" for light emissions in the visible wavelength range.

2. Methods and materials

Intersecting lines of different inks and one ink without any intersection were applied on different paper qualities.

Inks:

A: Ballpoint pen, Stabilo, Ref n° 0800 M 97 3, blue ink
Stroke width: 0.3 mm
B: Ink pad, Troadat, Ref n° 6/4911C, red ink
Stroke width: 0.6 mm
J: Ballpoint pen, CARAN d' ACHE, Giant Cartridge "Goliath", 8428.000 medium black, black ink
Stroke width: 0.4 mm

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Table 1
Used inks and apparatus settings.

Ink/ink combination	Excitation wavelength	Detection wavelength	Magnification	Picture integration time
A over B and vice versa	620 nm	735 nm, long pass	46.20	4 s
J over Q and vice versa	485–610 nm	645 nm, long pass	31.83	37 ms
R	485–610 nm	645 nm, long pass	31.83	37 ms

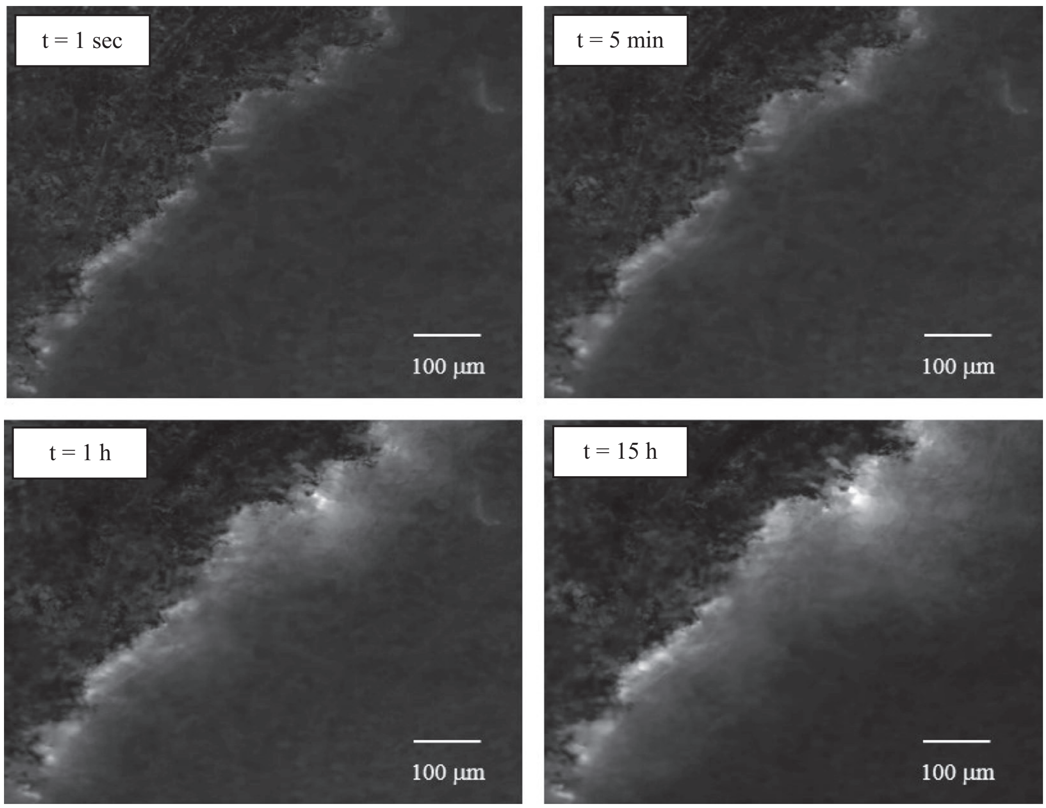

Fig. 1. Sequence of the diffusion process of luminescent components into the paper substrate: Fluorescence microscope pictures show the migration process with 1 s, 5 min, 1 h, and 15 h after the deposition of blue ink on the paper. The luminescent component(s) (white zone) migrates from the inked area (upper left area in the pictures) into the paper material (right lower area of the pictures).

Q: Ink pad, Trodat Printy 4822, red ink
Stroke width: 0.6 mm
R: Ballpoint pen, CARAN d' ACHE 826, blue ink
Stroke width: 0.3 mm

Paper brands:

- REFUTURA, Art. Nr. 228 080, 80 g/m^2, white recycling paper
- NAVIGATOR Universal, 80 g/m^2; silky touch; ultra bright, bleached office paper

The following instruments were used:

- Fluorescence microscopes of Olympus BX series with celSens software and Zeiss Axioplan with Axiomanager.Z2 software for illustration of the migrating zone
- Video comparator system VSC 6000 from Foster&Freeman and Docucenter Nirvis with PIA 7000 Software from Projectina for determination of the migration distances.

The setting on the detection unit was optimized for each sample type (paper and ink or ink combination) to get adequate luminescence and contrast. These parameters were kept constant during the whole experiment.

Sample preparation:

The crossing line sets of the ink combinations (see Table 1) were produced on the eve of their analysis. Each set consists of a large number of intersections of one ink combination. The second, intersecting line was applied one minute after the primary one. Pictures were taken with the aid of a video comparator apparatus according to the following protocol:

1st picture: immediately after the production of the line intersections
2nd picture: 2 h after the production of the line intersections
3rd picture: 4 h after the production of the line intersections
4th picture: 1 day after the production of the line intersections
5th picture: 1 week after the production of the line intersections
6th picture: 2 weeks after the production of the line intersections
7th picture: 3 weeks after the production of the line intersections
8th picture: 1 month after the production of the line intersections
9th picture: 2 months after the production of the line intersections
10th picture: 3 months after the production of the line intersections
Continuation: one picture per month
Last picture: 12 months after the production of the line intersections

3. Results

A piece of Navigator office paper was fixed on an Olympus fluorescence microscope. Magnification and focus were set. Ink of a blue Ballpoint pen, CARAN d' ACHE 826 (ink R) was applied on the paper, and pictures of the diffusion process were recorded from the first seconds to the first 15 h. Four pictures of the sequence are shown in Fig. 1.

To get an impression of luminescence effects, a picture of one intersection was taken with a fluorescence microscope (Fig. 2). The luminescence intensity is measured on one line within the intersection. The border of the migrating, luminescent ("non-visible") zone is not sharp. The highest intensity is close to

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