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Examination of black inkjet printing inks by capillary electrophoresis

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ABSTRACT

Counterfeiting of documents is a common phenomenon in the modern world. A large proportion of forgeries relates to inkjet printed documents. Hence there is an evident need to develop an effective and reliable method for the differentiation and identification of inkjet inks on questioned documents. The aim of the presented study was to investigate the possibility of applying micellar electrokinetic capillary chromatography (MECC) to forensic analysis of inkjet inks extracted from black and white printouts.

In order to achieve the above aim, a capillary electrophoresis system equipped with a diode array detector was used. The separation was performed using a fused silica capillary (60/50 cm total/effective length, 75 μm i.d.) with a background electrolyte composed of 40 mM sodium borate, 20 mM SDS and 10% (*v*/*v*) acetonitrile (pH 9.5) at 25 °C and 30 kV. Ink samples were extracted from black inkjet printouts with the use of dimethyl sulphoxide (DMSO).

Differentiation of inks was based on the number of significant peaks at different wavelengths, the relative migration times and the characteristic UV–Vis spectra. The electropherograms of the inks extracted from paper showed patterns which in most cases were distinctly different from each other. The greatest diversity of electrophoretic profiles was revealed for documents printed by Hewlett-Packard inkjet technology. A database of electrophoretic separation results of inks has been constructed for further forensic use.

1. Introduction

The study of various kinds of inks is one of the most important problems among the forensic document examiners. There are several papers presenting the utility of microscopic [1], spectroscopic [2–6] and chromatographic: thin layer chromatography (TLC) [7–9], high performance liquid chromatography (HPLC) [10–12] and gas chromatography (GC) [13,14], methods for the analysis of inkjet printing inks.

The increasing number of articles reporting the examination of various kinds of inks with the use of capillary electrophoresis (CE) confirms its constant development in recent decades. According to them, electroseparation techniques seem to be very promising in differentiation and determination of compounds from ballpoint, fountain, water-soluble fiber-tip, and many other pen inks. Since black printed documents are most often used in practice, black inks are more interesting to authors.

Fanali and Schudel [15] performed successful electrophoretic separation of black water-soluble fiber-tip pen inks for the first time. However, the authors did not deal with the extraction of ink from paper as the ink samples were taken directly from pens. Thus, they did not face the problems of dye solubility and of adhesion forces between dyes and the cellulose fibers. Electrophoretic separation of some black rollerball pen inks sampled from pen refills was also reported [16]. Although some obtained peaks were very broad (probably because of an incomplete resolution of the sample components), the research demonstrated the potential of CE to discriminate between liquid black inks. The first results from analysis of inks extracted from paper were published by Tsutsumi and Ohga [17]. They compared 26 extracts from filter and envelope paper and they found the two electropherograms of every ink to be the same except for an additional peak from envelope brighteners. Fountain pen inks, both liquid and extracted from paper, were also analyzed by Rohde et al. [18]. Good results were obtained in alkaline buffer, giving the possibility for simultaneous detecting of cationic and anionic analytes. With the addition of an organic solvent that caused less dye adsorption to capillary walls, better repeatability of results was achieved.

A very interesting idea was to apply fluorescence detection (apart from UV–Vis) at different excitation and emission

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wavelengths in comparisons between similar ink extracts and between ink extracts and the original ink composition – fountain [19] and ballpoint pen black inks [20]. Owing to such detection, the authors significantly improved the discrimination power of their method. Furthermore, they recommended particle induced X-ray emission (PIXE) as a complementary method giving sufficient information for comparison of the most problematical inks. The suitability of CE in the form of the micellar electrokinetic capillary chromatography (MECC) technique for forensic analysis of black ballpoint pen inks was assessed by Mania et al. [21]. The authors found the most effective extractant of inks from paper (pyridine: water, 1:1, *v*/*v*) and the optimum amount of a sample (6 dots of i.d. 0.8 mm). A quite different but comparably discriminating approach was proposed by Egan et al. [22]. In that study two different buffers systems were designed to analyze either cationic or anionic components of various black ballpoint pen inks. Another article [23] returned to the theme of comparison of roller pen inks. On the basis of electropherograms of 100 analyzed inks with patterns distinctly different from each other, it was stated that the capillary zone electrophoresis (CZE) technique is useful for comparing types of roller pen inks irrespective of the extraction process. A non-aqueous capillary electrophoresis (NACE) technique was developed for analyzing seven basic dyes contained in black ballpoint pen inks [24]. Supported by chemometrics (e.g. cluster analysis), the authors were able to differentiate 120 such kinds of inks.

Up till now, there has been no report presenting results of CE analysis of black inkjet printing inks. Thus, taking into account the above-mentioned successful applications of CE (in CZE, MECC, and NACE modes) to the analysis of various kinds of black writing inks and bearing in mind the positive results obtained for color inkjet printing inks [25–27], the authors decided to make an effort to fill in the gap in current analytical methodology of black and white questioned document examination. Therefore, the aim of this paper was to develop a rapid and cost-effective method of analyzing and discriminating between black inkjet printing inks.

For this purpose, a systematic study was undertaken with the use of the MECC technique due to its numerous advantages, such as small sample and reagent requirements, simplicity of sample preparation, high resolution, minimum amount of nascent waste, possibility of connection to various kinds of detectors and low costs of exploitation. Furthermore, the capacity of MECC to separate sample analytes from a range of compound classes within a single run makes it an ideal technique for the criminalistic analysis of multicomponent mixtures such as black inkjet printing inks.

2. Experimental

2.1. Materials and samples

The chemicals used throughout the experiments were sodium tetraborate decahydrate and dimethylformamide (DMF) produced by POCH (Poland), sodium dodecyl sulphate(VI) (SDS), methoxyethoxyethanol (MEE) made by Sigma–Aldrich (Germany), methanol (MeOH), acetonitrile (ACN), ethanol (EtOH), dimethyl sulphoxide (DMSO), hydrochloric acid and sodium hydroxide produced by Merck (Germany). All reagents and solvents were of HPLC grade or p.a. purity.

A printout of pure black ink HP 11 (C4810A) was taken from the Hewlett-Packard Business Inkjet 1200 printer. An extract of this ink was used during optimization and validation steps. Test pages of black ink samples were printed by printers listed in Table 1. All printouts were prepared using ordinary and commonly available printing paper – PolSpeed (International Paper, Poland).

Table 1
Black inkjet printing inks analyzed in the study.

No.	Producer	Model	Cartridge number
1	Hewlett-Packard	Deskjet 2500C	HP 10 (C4844AE)
2		Business Inkjet 1200	HP 11 (C4810A)
3		Deskjet F380	HP 21 (C9351A)
4		Deskjet 3740	HP 27 (C8727A)
5		Deskjet 3550	HP 27 (C8727A)
6		Deskjet 710C	HP 45 (51645AE)
7		Deskjet F4280	HP 300 xL (CC641)
8		Photosmart C4280	HP 350 xL (CB335)
9		PSC 1510	HP 338 (C8765EE)
10	Canon	Pixma iP1800	PG-37
11		Pixma iP1900	PG-37
12		Pixma MP210	PG-37
13		Pixma iP4500	PGI-5 BK and CLI-8BK
14	Brother	DCP-135C	LC-970 BK
15		DCP-350C	LG-1000 BK
16	Epson	Stylus D92	T0711
17	Lexmark	Lexmark X2530	34 Black
18		Lexmark Z615	16 Black

2.2. Preparation procedures

The background electrolyte (BGE) was prepared using an adequate amount of stock solutions of 100 mM borate buffer (obtained by dissolving sodium tetraborate) and 100 mM SDS and pure ACN. Ready solutions were sonicated, filtered through 0.45 μm Minisart RC15 cellulose syringe filters (Sartorius, Germany) and stored in darkness until used. They were replaced after every five runs.

Before daily operation, the capillary was conditioned with MeOH for about 5 min, then with: 1 M HCl for 5 min, 0.1 M NaOH for 10 min, water for 2 min, and finally, with the BGE for 10 min. Between single runs, the capillary was rinsed with the same solvents for a shorter time (for 5, 2, 2, 1, and 1.5 min, respectively).

To prepare stock sample solutions used throughout optimization and validation stages, 12 squares (each of 0.9 cm × 1.2 cm) from a test page were cut out and extracted in 0.5 mL of DMSO supported by 15 min of sonication. After that, the extract were transferred to new bottles in order to separate them from paper particles and centrifuged (13000 rpm for 5 min) in Eppendorf vial (to dispose of possible small pieces of paper and paper fibers). 5 μL of extract prepared in this way was evaporated at 190 °C. The oxidation of dyes was avoided by blowing with dry nitrogen gas. The residue was diluted in 10 μL of injection solution (BGE:water, 1:1 *v*/*v*) and after that, sample was ready to be directly injected into the CE instrument.

Real ink samples were collected in the form of 25 dots (ø 0.8 mm) of printed paper. Ink was extracted to 5 μL of DMSO with the use of a sonication bath. Every successive step of sample preparation (centrifugation, evaporation and dilution) was the same as presented above.

2.3. Instrumentation

Electrophoretic measurements were performed with the P/ACE MDQ Capillary Electrophoresis System (Beckman-Coulter, USA) equipped with a diode array detector (DAD). The injection was made by applying a pressure of 0.7 psi for 6 s. Separation was carried out in a 60 cm (50 cm effective length) polyimide-coated fused silica capillary of 75 μm ID (Beckman-Coulter, USA) for 15 min with +30 kV applied. The electropherograms were recorded in spectral range from 190 to 600 nm (number of diods 256, scan data rate collection 4 Hz). All peaks were integrated at the same wavelength – 220 nm – in order to make comparison more reliable. The capillary cassette and the sample storage module were maintained at constant temperature – 25 and 10 °C, respectively.

Table 2
Efficiency of extraction of black Hewlett-Packard ink in the four most effective extractants.

Printer number	Extraction efficiency [%] after 15 min of sonication			
	DMF	DMSO	BGE[a]	MEE:DMF:DMSO:EtOH:H_2O (1:1:1:1:1, *v*/*v*/*v*/*v*)
1	75	100	75	75
2	100	75	100	75
3	100	100	100	100
4	50	100	100	25
5	50	75	75	75
6	50	100	75	75
7	25	75	75	75
8	25	75	25	50
9	25	75	75	50
10	0	25	0	0
11	0	25	0	0
12	25	25	0	0
13	25	25	75	25
14	25	75	50	50
15	100	100	100	100
16	25	50	0	0
17	50	25	25	0
18	25	0	25	0

[a] Solution of 40 mM borate buffer with 20 mM SDS and 10% acetonitrile.

3. Results and discussion

3.1. Optimization of the method

The examination aimed at selecting the optimal extractant for black inkjet printing inks proceeded analogically to that performed for color inkjet printing inks [28]. Two circular pieces (i.d. = 0.4 cm) of printed paper were punched out and transferred to an extracting vial. Ink was extracted by adding 2 mL of 1 of 24 tested extracting agents. The vials were capped to prevent evaporation and left for 30 min to extract. The extraction efficiency was evaluated visually using a 5-degree scale from 0 (no extraction) to 100% (all the ink was extracted, the remaining paper was white). The first observation was made after 30 min of the extraction process and then the extraction of inks was supported by putting the samples into an ultrasonic bath. The effects of that step were monitored after 5 and 15 min.

24 different solutions were examined in terms of extraction efficiency and the best results obtained for some of them are presented in Table 2. Pure DMSO was chosen as the most effective extracting agent. Since the extracts based on DMSO could not be injected directly into the CE system, they were evaporated by heating up to 190 °C under nitrogen gas.

The influence of the number of dots cut out from the printout and taken for analysis was investigated. For this purpose, 10, 15, 20, 25 and 30 dots were punched, extracted and analyzed. On the basis of the obtained results, it was ascertained that 25 is the optimum number of dots in a sample, giving 7 well-shaped peaks of at least double the intensity compared to a 10-dot sample.

In order to determine the optimum composition of BGE in terms of the number, intensity and resolution of peaks in electrophoretic profiles, 10 solutions containing borate buffer, SDS and ACN were investigated. Concentrations of the components were set within ranges 40–120 mM, 10–30 mM and 10–30% *v*/*v*, respectively, as presented in Table 3.

As the primary criterion of optimization, the following function was used: $F = n^2/(t_m \cdot RSD_{med})$, where n is the number of obtained peaks, t_m is the migration time of the latest peak, and RSD_{med} is the median of relative standard deviations of migration times of peaks (of those whose area ≥1000 area units) [26]. The F values obtained for the examined solutions are presented in Table 3.

Table 3
Optimization of the composition of BGE solutions with regard to function *F* (see text).

No.	Borate buffer [mM]	SDS [mM]	ACN [% *v*/*v*]	n	t_m [min]	RSD_{med}	F
1	40	10	10	6	8.9	0.09	43.1
2	120	10	10	5	13.2	2.46	0.8
3	120	10	30	3	20.0	0.53	0.9
4	40	10	30	3	8.2	0.49	2.3
5	40	30	10	5	9.2	0.17	16.1
6	120	30	10	5	20.0	4.03	0.3
7	120	30	30	3	20.0	0.60	0.7
8	40	30	30	3	20.0	0.20	2.2
9	80	20	20	6	12.2	0.35	8.4
10	40	20	10	7	11.7	0.19	22.1

In accordance with *F*-values obtained, the solutions containing 40 mM of borate buffer and 10% of ACN revealed the most effective influence on components of black printing inks separation. Additional visual comparison of electropherograms, allowing the intensity of individual peaks to be estimated, inclined us to choose the solution containing 40 mM borate buffer, 20 mM SDS and 10% ACN as the optimal one. It offered a good compromise between resolution and analysis time and provided higher intensity of dyes peaks. Moreover, such concentrations of the components were effective in reducing adsorption effects onto the capillary wall and minimizing rinsing time.

It is worth noting that the chosen BGE was of the same composition as the one selected for color inks [26]. The possibility of using the same BGE for separation of ink components is beneficial with regard to examination of unknown black printouts with some color elements (e.g. stamps).

The effect of normal stacking mode was also investigated. Four injection solutions of different concentration of components were examined: pure BGE and BGE with water in ratios 1:1, 1:9 and 1:99 (*v*/*v*). A stacking effect was observed that was especially great in the case of less intensive peaks (peak 2 increased 1.5 times, peak 4 almost doubled). The best results (the greatest signal intensity) were obtained for BGE diluted in ratio 1:1.

3.2. Repeatability and reproducibility of the method

The validation study focused on the precision of the method in terms of repeatability and reproducibility of the migration time of peaks. Due to the fact that the formula of inks is a trade secret and no reference samples were available, it was not possible to determine the accuracy.

Verification of the repeatability and reproducibility was conducted with the use of stock extract. To estimate the influence of injection and instrumental parameters one sample was analyzed 5 times (an example of an electropherogram is shown in Fig. 1). To check the influence of the preparation process, 5 portions of stock solution of the extract were taken, prepared and analyzed consecutively. Moreover, 5 samples of black ink were independently cut out for printouts, extracted, prepared (in accordance with the procedure for real sample preparation), and analyzed to estimate the precision of the whole method. Reproducibility was examined with the samples being analyzed in the following way: (a) every day for 5 days (5 samples per day), (b) on one day by two analysts (5 samples per analyst), and (c) on one day using two different capillaries (5 samples per capillary). The results obtained are presented in Table 4.

By looking at the RSD values of the 5 repetitions of a sample, the conclusion is that the sample injection and the instrumental parameters did not have a great impact on the precision of the method (in all cases RSD was less than 0.3%). An additional step of sample preparation – extractant evaporation – resulted in slightly

Table 4
The repeatability and the reproducibility of MECC separation of black ink depending on days, analysts and capillaries (Business Inkjet 1200 printer).

Analysis	Parameter	EOF	Peak 1	Peak 2	Peak 3	Peak 4
1 sample analyzed 5 times	$\bar{t}_m$ [min]	3.0	6.2	6.6	6.8	8.3
	RSD [%]	0.3	0.3	0.2	0.2	0.2
5 samples independently prepared	$\bar{t}_m$ [min]	3.0	6.4	6.7	7.0	8.3
	RSD [%]	0.9	1.3	1.3	1.4	1.6
5 samples independently extracted	$\bar{t}_m$ [min]	3.0	6.1	6.4	6.7	7.7
	RSD [%]	0.8	2.0	2.0	1.9	2.3
5 samples on 5 days	$\bar{t}_m$ [min]	3.0	6.3	6.7	6.9	8.3
	RSD [%]	1.1	1.8	1.9	2.1	2.2
5 samples by 2 operators	$\bar{t}_m$ [min]	3.0	6.3	6.7	6.9	8.3
	RSD [%]	1.0	1.6	1.6	2.0	0.6
5 samples using 2 capillaries	$\bar{t}_m$ [min]	3.0	6.2	6.5	6.8	7.8
	RSD [%]	1.0	3.0	3.4	3.8	6.3

worse repeatability (RSD = 1.6%) in comparison to that obtained for color inks (1.1%) [26]. On the other hand, the sampling conditions (printed areas selected to be punched) and extraction process, the most serious sources of errors, triggered an increase of RSD to 2.3%. It seems that in the case of black inks, the printouts were more homogeneous.

As shown above, different days of analysis and different analysts affected reproducibility three times less than changing the capillary (about 6%). This indicates that the precision of the method depends mainly on variations of the capillary. That is why, the analyst should devise a method for repetitive preparation of a new capillary, taking much notice of its length.

3.3. Application of the method

To verify the suitability of the proposed method, electrophoretic profiles were created for all of the collected printouts made by printers of different producers and models (see Table 1) and then they were compared with each other. The results revealed that the inks could be divided into two groups in terms of brands. The first group contained the results for samples taken from Hewlett-Packard printers (Fig. 2), which were evidently different from electropherograms of other producers. The extracts contained so many colorful components (originating from the color undercoat of printouts) that the peaks corresponding to them were of sufficient intensity to be clearly distinguished. In some cases, dyes coming from all three colors (yellow, magenta and cyan) of inks (Fig. 2a) could be detected, but in other cases two (Fig. 2b) or one (Fig. 2c) dye was visible.

The second group included the results obtained for inks of producers other than Hewlett-Packard. Their electrophoretic profiles were characterized only (or predominantly) by peaks of colorless components – additives absorbed in the range of 220–330 nm (Figs. 3 and 4) and, as a consequence, they were less informative than those obtained for Hewlett-Packard inks. However, there was some possibility of distinguishing between some of the inks in this group.

The electrophoretic profiles of Canon inks (Fig. 3) were very typical and similar for each of the examined Canon printers – only one, but very intensive, peak at about 11.4 min was observed. Electropherograms obtained for inks produced by Brother revealed many additives but at a very low concentration (see Fig. 4a). Inks originating from Epson and Lexmark printers contained mainly carbon black (insoluble pigment), hence their electrophoretic profiles were without any characteristic peaks (Fig. 4b and c).

In order to confirm the possibilities of the optimized method in the field of document examination, an investigation of two (simulated) questioned documents was performed. Each of them was specially designed and prepared with the use of different printers. On the first document (simulating an invoice), an additional two digits in the date were introduced, while on the second one (simulating a letter of attorney) an extra sentence was added. The inks from both fragments of printouts – original and suspicious – were taken for analysis. To avoid autosuggestion, the producer and model of the particular printers were not known to the person performing analysis. So the method was tested by printouts that could be considered as ‘real samples’.

In the first case, only 12 dots of printed paper were taken for extraction as the examined area (two digits) was very small. As seen in Fig. 5, the electropherograms obtained were quite different: two additional peaks at about 6.6 and 8.0 min were visible in profile b.

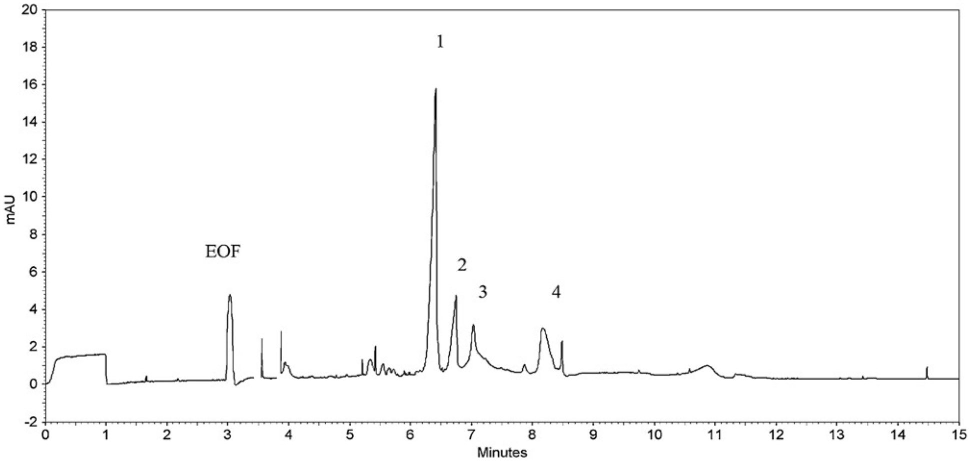

Fig. 1. Electropherogram of black ink taken from Hewlett-Packard Business Inkjet 1200 printer, obtained in BGE consisting of: borate buffer (40 mM), SDS (20 mM) and ACN (10%, *v*/*v*).

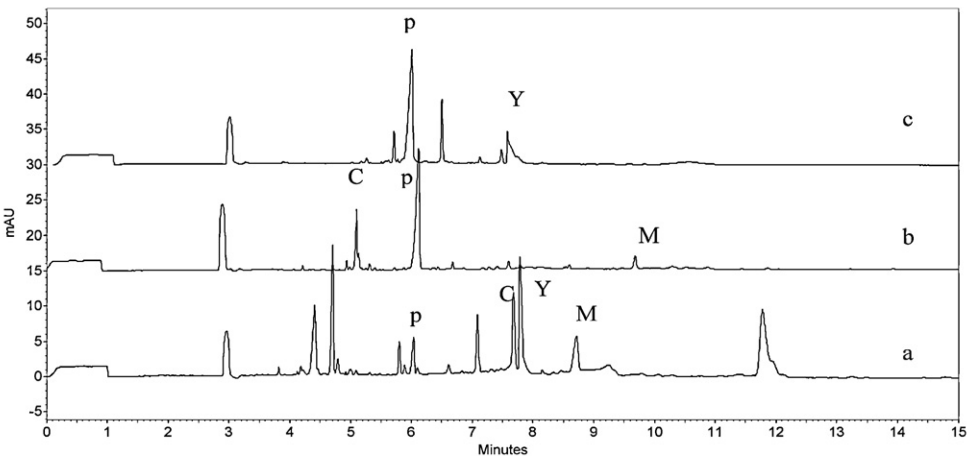

Fig. 2. Electropherograms of black inks taken from different models of Hewlett-Packard printers: (a) Deskjet F4280, (b) Deskjet 3740 and (c) Photosmart C4280, Y – peak coming from yellow, M – magenta, and C – cyan ink, p – peak from paper.

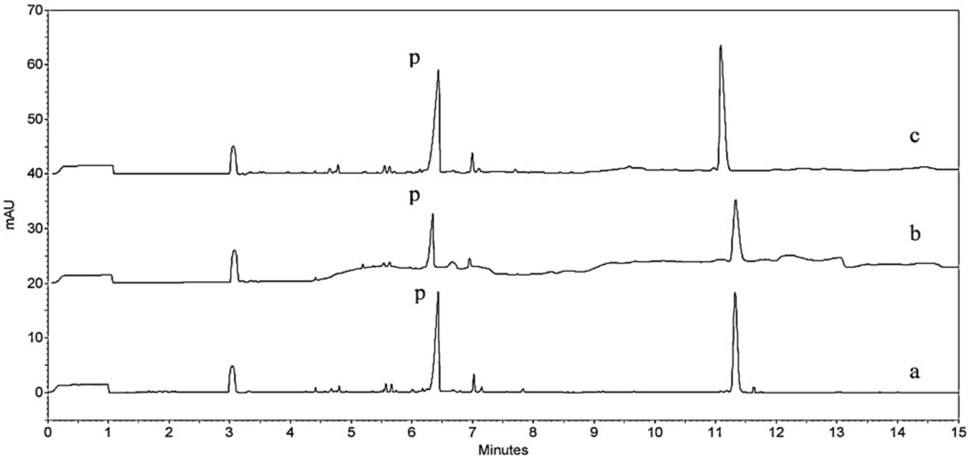

Fig. 3. Electropherograms of black inks taken from different models of Canon printers: (a) Pixma iP4500, (b) Pixma MP210 and (c) Pixma iP1800, p – peak from paper.

This was proof that a different printer was used to change the date. What is more, according to the obtained results (the peak at 8.0 coming from magenta ink), one can ascertain that it was a printer which uses cartridges with color inks during the process of black letter printing. In all likelihood, it was a Hewlett-Packard printer.

The electrophoretic profiles obtained in the latter case are presented in Fig. 6. The peak at about 6.6 min (coming from components absorbing in the UV range) made a difference. Another dissimilarity was revealed at about 8.2 min: there was a single peak in electrophoretic profile *a* and a double peak in profile *b*. That

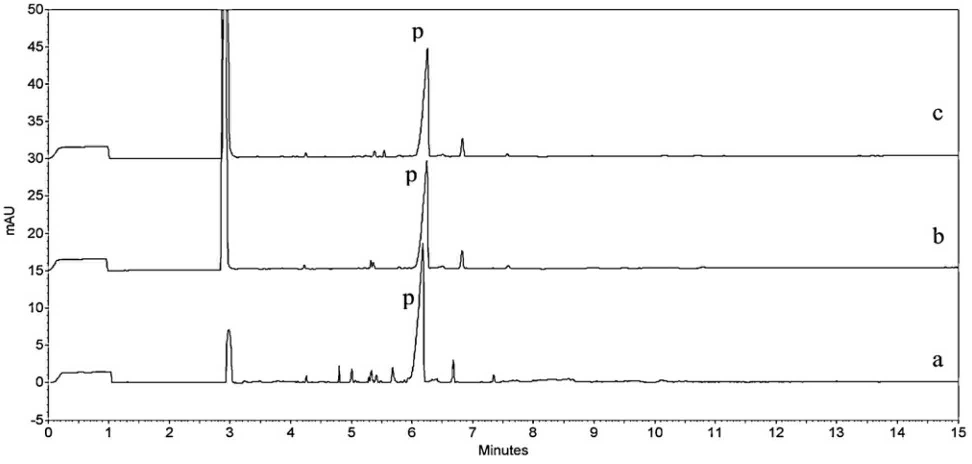

Fig. 4. Electropherograms of black inks taken from different printers: (a) Brother DCP-135C, (b) Epson Stylus D92 and (c) Lexmark Z615, p – peak from paper.

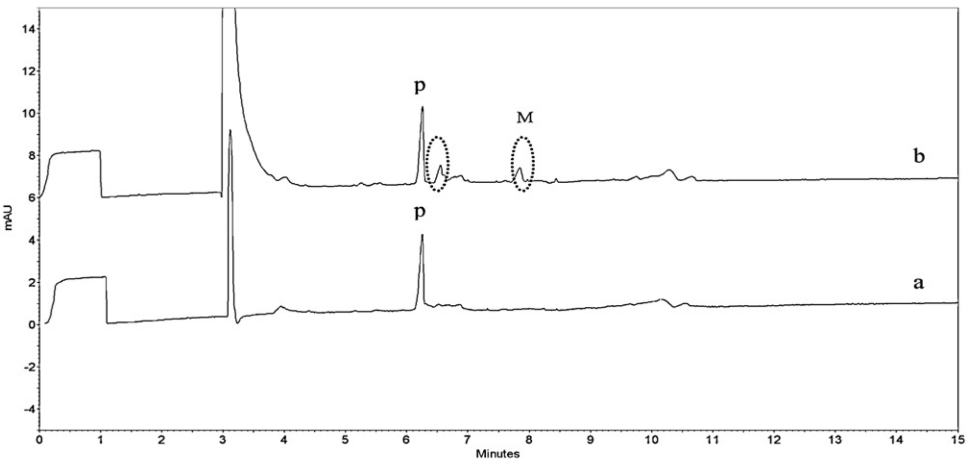

Fig. 5. Electropherograms of inks extracted from an "invoice": (a) original document, (b) questioned fragment, M – peak coming from magenta ink, p – peak from paper.

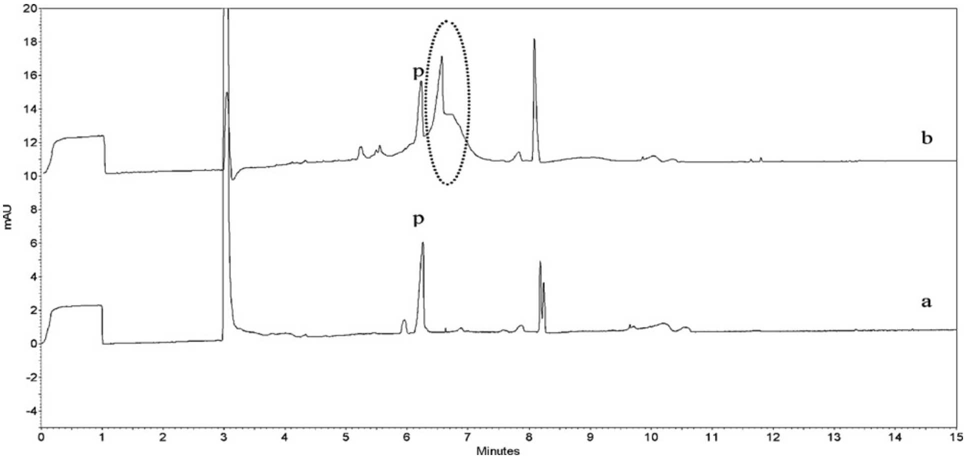

Fig. 6. Electropherograms of inks extracted from a "letter of attorney": (a) original document, (b) questioned fragment, p – peak from paper.

is why, the conclusion was drawn with high probability that the analyzed document had been counterfeited.

4. Conclusions

Examinations carried out previously [26] and at present have revealed that in spite of different composition of color and black inks, the methods proposed for their analysis by capillary electrophoresis for forensic purposes are quite similar. For instance, under well-defined separation conditions (capillary temperature – 25 °C, sample garage temperature – 10 °C, and internal diameter of capillary – 75 μm), the composition of BGE components was found to be the same (40 mM sodium borate, 20 mM SDS, and 10% *v*/*v* ACN) and the mixture of BGE and water in 1:1 volume ratio is the most effective injection solution. The optimum number of taken dots were the same – 25 pieces (ø 0.8 mm) of printed paper. In such conditions the step that is most critical in terms of quality of electrophoretic profiles is extraction of inks with the use of as effective a reagent as possible. Although two specific extractants were carefully chosen for color and black inks, efforts should be continued to find mixtures that are even more effective in both cases.

It is worth noting that the thickness of the ink surface on the analyzed printout depends on many factors (e.g. type of ink, its amount in cartridge and printer model and settings) and this can influence the results of analysis. Therefore, in every case, the document examiner should take an individual and carefully considered decision about the minimum number of dots to be taken. What is more, some black inks consist mainly of carbon black particles, which are not soluble and consequently unable to be extracted. Nevertheless, this discovery gave a basis for making an assumption about the possible producer of a given ink.

It was ascertained that the method is capable of offering satisfactory results in terms of precision, provided that special attention is paid to the selection and cutting out of printed paper pieces and to the extraction step. Furthermore, a case should be analyzed using the same capillary, preferably by the same operator. Different days of analysis, in such a case, have less importance.

At this stage of the research, it can be stated that the possibility of differentiation of inks originating from different models of printers depends on the producer of the printer. In the case of Hewlett-Packard inks, no difficulties were encountered in differentiating between their electrophoretic profiles. It is also possible to differentiate Hewlett-Packard inks from inks of other producers. However, a comparison of black prints printed using various makes of ink seems to be much more difficult than in the case of color prints. Cases such as the comparison of the same brand inks (e.g. Canon inks) or inks that are difficult to extract (e.g. Brother, Epson and Lexmark inks) were more complicated, thus the differentiation was very problematic or even unfeasible.

Summarising, the proposed method fulfilled the basic criteria of questioned document examination: it enabled us to obtain reliable analytical results with rather small document destruction and in a relatively short time. It proved to be suitable and effective; however, it still needs to be improved, especially in the case of inks that are difficult to extract. Therefore, additional examinations are planned, encompassing a wider sample set of black inks purchased from different manufacturers. The searchable database of electropherograms of the numerous black inks available in Poland (created in this study) will enable us to evaluate the usefulness of this technique in routine forensic examinations.

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