Assessment of the Possibility of Identification of Black Toners on Printouts in Lexmark Printers Using SEM Method

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Abstract

Background: Document examiners are frequently asked to determine whether or not a printout has originated from a particular laser printer. The printer can rarely be identified unless some unique defects or irregularities of the printer are present on the printout.

Aim/Objective: The project aimed at obtaining the individual identification of printing and copying devices.

Material and Methods: This paper presents an overview of a systematic approach to characterizing and discriminating the toner of different laser printer cartridges using scanning electron microscopy. A total of 21 collected printouts were printed on 21 different Lexmark printing machine models. Sixteen printouts were obtained using printers with original toner cartridges, while five printouts were obtained using printers with replacements. Clear criteria were established for individual assessment.

Results: Based on the SEM images and established criteria, the details of topography of the toner structure were revealed. Based on this study, the differences or similarities of toners on tested printouts were determined.

Conclusion: The SEM technique can be successfully, simply, and rapidly applied to the analysis of toners on paper documents.

Keywords: Scanning electron microscopy, SEM/energy dispersive X-ray, toners, X-ray microanalysis

Introduction

A forensic examination of documents is a broad research area encompassing the so-called classical techniques of document examination including handwriting and technical examination. Counterfeiters benefiting from the latest technologies are capable of producing new documents in their entirety or changing the contents of existing documents by introducing alterations. The availability of appliances such as printers and multifunctional devices results in the majority of documents no longer being handwritten. There is a range of state of the commercial art technologies, such as photocopiers, printers, and other multifunctional devices at our disposal, produced by companies such as Konica Minolta, Canon, OKI, Hewlett–Packard, Lexmark, Epson, Dell, Ricoh, Brother, and Xerox.

From the forensic science, one of the essential problems faced by analysis, among others, appears to be the identification of the implementation device. From the data available, we know that synthetic resins, pigments, and additives make up the elements of toners. Each of these elements plays a specific role. Resins, making up the highest percentage content, ensure the right plasticity and serve as a medium, that is, the factor tying together all elements of the toner as well as the toner with the base. The pigment adds the appropriate color to the printing elements, whereas the various additives improve other properties of the components, such as electrostatic properties, sheen, wearability, and plasticity. At present, most electrophotographic devices use powdered toners (loose granular solid materials). The following techniques are used in the analysis of the composition of such toners: Fourier transform infrared (FTIR), conjugated gas chromatography around mass spectrometry with attachment to the pyrolysis, and scanning electron microscopy with X-ray microanalysis.

Studies using the infrared spectrometric method and scanning electron microscopy with X-ray analysis have demonstrated the ability to differentiate between toners available on the Polish Market. However, they failed to match the toner explicitly to the relevant device (whether printer or photocopier).
Almeida Assis et al.[6] proposed the use of diamond cell FTIR spectrometry methodology for the analysis of black toners. This methodology was considered to be nondestructive as it allows the forensic analysis of the questioned documents while preserving their integrity. Cartridge raw toner and processed toner of printed documents showed minimal differences on some spectra. Toners were classified into twenty different groups using their main chemical characteristics and relative peak.

The optical microscopy[7] is another method of analyzing paper-based documents. The microscopic observations revealed that the surface of the toner embossed on paper was highly elaborate and created a characteristic, easily observable structural pattern. Therefore, within the framework of present research, the project aimed at obtaining the individual identification of printing and copying devices. Our goal here is to assess the possibility to individually verify the toner on prints made with laser printers and laser multifunctional devices. In this project, 160 printouts obtained from the different laser printer devices such as Konica Minolta, Canon, OKI, Hewlett-Packard, Lexmark, Epson, Dell, Ricoh, Brother, and Xerox were tested. In consideration of the wide results obtained in this paper for the first time, only results obtained from the observation of the printouts from the Lexmark printing laser devices were presented. Printouts were selected on the grounds of quantity, diversity, and specific features. These features were given names to identify and distinguish them. Printouts from the Lexmark printing laser devices served as the research material.

Scanning electron microscopy

The method of the scanning electron microscopy (SEM), introduced as early as in the mid-1970s, enables detection and description of morphology, as well as the chemical composition in the entire volume of the tested samples or in individual grains and particles. The main advantage of the scanning electron microscopy (SEM) is the possibility to observe the surface of objects without their prior preparation. The samples are scanned with the electron beam deflected by the scanning coils. The signals from the surface of the sample, most often in the form of secondary electrons (SE) or reflected electrons (backscattered electron-BSE), reach the detector. The signal resulting from the detector controls the brightness of the image shown on the monitor. The magnification of the scanning microscope results is affected by the size of the sample’s scanned areas.[8]

Research capabilities of the SEM microscope arise from the parameters of an electron beam and the detection system properties. However, the classic high-vacuum version has its limitations. The high vacuum maintained in the electron optical column and in the chamber excludes the presence of preparations with a high vapor pressure. In addition, the surface of the sample should be conductive so that it does not accumulate electric charges on the surface, thus disturbing the image. Nonconductive samples can be coated with a thin layer of conductive material by means of sputtering. The coating materials used are the noble metals: chromium, gold and platinum, and carbon. Sputtered layers not only fulfill the role of an electric conductor but also protect the tested sample against the thermal influence of the electron beam.

The current technological solutions of the scanning electron microscopes (changeable vacuum and sufficiently big chamber) also enable the performance of nondestructive examination of documents. These solutions, notwithstanding their significant contribution to the extension of research capabilities in forensic science examination, if it comes to the examination of documents, they are not necessarily able to achieve high-resolution images, particularly with high magnifications of the order above 10,000 times, which is achievable in a high vacuum after sputtering. Nevertheless, they prove sufficient in accordance with the expertise requirements in forensic laboratories.

Materials and Methods

In this study, a total of 21 printouts from the Lexmark laser printers and laser multifunctional devices were analyzed. Sixteen printouts were obtained using original toner cartridges, while five printouts were obtained using their replacements. All printouts were printed on A4 paper format. Printouts were prepared using ordinary and commonly available printing paper – ePrimo of 80 g² basis weight. Only black toner was tested. The surface of the same letters was examined (o, n, and u). In case of every printout, three letters at the beginning, in the middle, and at the end of the text were analyzed, making it nine letters in total. Every letter was observed in a few places: at the curves, at the edges, in the corner, and at the letter joints. The samples could be divided into two groups, namely, the printouts obtained using printers with original toner cartridges and printouts obtained using printers with substitutes manufactured (replacements) by companies other than the original manufacturers. Substitute cartridges that are relatively cheaper are now commercially available in the market. The printouts were coded from 1 to 21 (SIMPLE ID). All printouts were obtained from four types of devices such as color laser printers, monochrome laser printers, color laser multifunctional devices, and monochrome laser multifunctional devices. The model of printer and the type of device used for the samples are shown in Tables 1 and 2.

The black toners were imaged using a scanning electron microscope (SEM) (Tescan Mira 3XMU) equipped with an energy-dispersive X-ray spectrometer (Oxford Instruments). The following detectors were used: SE and BSE. Samples were imaged in a magnification range from ×80 to ×130,000. The analytical conditions of observation were as follows: working distance – 15 mm and accelerating voltage in HV – 10 kV. The imaging process was carried out in the following sequence, for all samples. First, a single letter was imaged in a low magnification. In the next step, the irregularities on the surface of toner were tested. In higher magnification, two types of specific areas are visible: solid surface and granular surface. Finally, the surface of a single grain in a very high magnification (more than ×100,000) was examined.
Table 1: List of printers analyzed in this study - original toner cartridges

| Sample ID | Model | Type of devices      |
|-----------|-------|----------------------|
| 1         | E260/E360 | Monochrome laser printers |
| 2         | MS 610dn   | Monochrome laser printers |
| 3         | T650n/T652/T654n | Color laser printers |
| 4         | C734/C736   | Monochrome laser printers |
| 5         | X264/X363/X364 | Multifunctional devices |
| 6         | MX410       | Monochrome laser printers |
| 7         | X654de/X656de | Color laser printers |
| 8         | X463/X464/X466 | Color laser printers |
| 9         | X658de      | Multifunctional devices |
| 10        | MX710/MX810 | Color laser printers |
| 11        | X860de/X862de/X864de | Multifunctional devices |
| 12        | C540/C543/C544 | Color laser printers |
| 13        | X543/X544   | Multifunctional devices |
| 14        | X920de      | Multifunctional devices |
| 15        | X940e/X945e | Color laser printers |
| 16        | X950        | Color laser printers |

Table 2: List of printers analyzed in this study - replacements

| Sample ID | Model | Type of devices      |
|-----------|-------|----------------------|
| 1         | E240n | Monochrome laser printers |
| 2         | E250d | Monochrome laser printers |
| 3         | T640/T642/T644 | Color laser printers |
| 4         | T634   | Color laser printer |
| 5         | C780   | Color laser printer |

Sample preparation

Single letters have been previously extracted from the printouts and collected using commercially available 1/2” sample stubs provided with a conductive carbon adhesive tape. Before the analysis, the samples were chromium- or carbon-coated, using a QUORUM Q150T sputter, to facilitate a conductive surface.

Evaluation criteria of toners

- Distribution of toner surface on a single letter
- Characteristic features of solid surface
- Characteristic features of granular surface
- Size, shape, and structure of the grains.

Repeatability

The repeatability was evaluated using toner on printouts obtained from four printers: E260/E360 (1), C734/C736 (4), MX410 (6), and X920 (14). For each printout, six different samples were taken and analyzed separately by SEM. A few characteristic features (rods, irregular and regular structures of the grains and solid surface, and shape of the grains) were used for evaluation. It turned out that in terms of selected features, toner microstructure is the same in all printouts.

Results and Discussion

Based on the optical microscopes’ observations with the maximum magnification up to 2000 times, we can solely conclude that the surface of the black toner on printouts differs in all cases. Such surface creates a characteristic and unique pattern for every single printout. Two types of specific areas are visible: solid surface and granular surface. The granular surface tends to form small or large clusters of a varying density. The surface of the toner was observed to be differentiated in terms of distribution in these two areas. In some printouts, the granular layer prevails, and in others, the solid one. In other cases, the ratio of both surfaces is approximately equal, depending on the device used. In the case of printouts made from the same type of device, the structure of the toner surface is the same. It was an inspiration for a much detailed investigation, providing the possibility of more elaborated. For this purpose, the research material was analyzed using the scanning electron microscopy, ensuring even more detailed analysis of the topography of the toner surface embossed on the paper. The examination by SEM discloses in detail the topography of the toner and enables the application of the criteria mentioned above. Based on this study, we can determine the difference between specific types of toners generating the printout, that is, taking into consideration the structure of the toners’ surface distributed on the paper and the contribution of larger and smaller grains and their structural surface. The characteristic features of the original toners and their replacements are shown in Tables 3 and 4 and Figure 1.

We can, accordingly, observe the differences in the distribution of the toner layer. This layer can be solid, smooth, and covering the entire surface of the toner. Only a few single grains are visible on the coastline of the toner layer. The layer of the toner may contain parts of the solid surface, interrupted by smaller or larger, numerous, or single areas with the grains of varying density or it may be entirely granular. The analysis of the toner surface showed that in all cases, the toner layer on the printouts consists of both solid and granular surface. The difference relies on the ratio of one layer to another and their distribution patterns on the area of one letter, depending on the type of device used.

Toners are showing the great diversity in terms of structures observed on the surface of grains or just under the surface. These are spherical structures of different sizes from 0.05 to 1 µm in diameters, “bump” structures of the order up to circa 0.05 µm in diameter, straight, or bent “rod” structures with various lengths of up to about 4 µm, and irregular structures of about 1 µm in diameter. The grain surface is also characterized by a degree of density of the structure and the structure present on the grain surface, or structures visible just beneath grain surface, or in both combinations. For example, spherical grains have been observed in several printouts obtained from the printing devices such as X264/X363/X364, MX410, X543/X544, X658de, and X920de. These structures most often appear in large numbers from several tens to several hundred items, covering even entire surface of the grain. Structures in the form of “bump” appear in large most numbers from a few hundred to a dozen or so thousand items on the surface of the grain largely of models apart from X264/X363/X364, MX410, X654de/X656de, X658de, X940e/X945e, E240n,
T640/T642/T644, and T634 printers. Structures in the form of straight “rods” on the grain surface or under grain surface were observed in the amount from a few to several tens of items. These structures were observed on printouts obtained from printers such as E260/E360, MS610dn, T650n/T652/T654n, X460/X656de, MX710/MX810, X543/X544, E240n, and E250d. Several bent “rod” structures on printouts obtained from only two printers such as X264/X363/X364 and X463/X464/X466 were found.

Estimation of the actual quantity of structures in the grain is impossible; on account of technical restrictions SEM, we can see only a surface of grains. For example, structures in the form of straight “rods” can start with one ending on the surface and second to end deep inside grains. Other structures, as irregular structures presumably not only on the toner surface but also the inside grains, are appearing. Many structures are not being observed. The appearance of some structures is shown in Figure 1.

In Tables 3 and 4, they also entered into information about the appearance and the size of grains. The grains adopt spherical or cylindrical shapes and some of them look like perfect spheres, others are irregular with sharp or rounded edges or look like “potato,” and others are more or less elongated. The grains form clusters and chains or occur in the single form. The

| Sample ID | Model | Type of device | Solid surface | Granular surface |
|-----------|-------|----------------|---------------|------------------|
| 1   | E260/E360 | Monochrome laser printer | Irregular structures/diameter (µm) | Bumps/0.05 |
| 2   | MS 610dn  | 0.5 | 0.5 | 0.5-3.8 | Regular |
| 3   | T650n/T652/T654n | Color laser printer | Bumps/0.05 | 0.5-1.0 |
| 4   | C734/C736 | Straight and bent with different length | Bumps/0.05 | Spherical + irregular |
| 5   | X264/X363/X364 | Monochrome laser multifunctional device | Bumps/0.05 | Bumps/0.05 |
| 6   | MX410 | 0.5 | 0.5 | 0.2-2.0 |
| 7   | X654de/X656de | 0.1 | Straight and bent with different length | Different/1.0 |
| 8   | X463/X464/X466 | Straight and bent with different length | Spherical | Spherical/0.05 |
| 9   | X658de | 0.3 | Straight with different length | Spherical | Spherical/0.05 |
| 10  | MX710/MX810 | 0.3+cones | - | - |
| 11  | X860de/X862de/X864de | 0.5 | Spherical | Spherical |
| 12  | C540/C543/C544 | Color laser multifunctional device | Bumps/0.05 | Spherical |
| 13  | X543/X544 | 0.5 | Single/0.6-0.8 | Spherical |
| 14  | X920de | 1.0 | Bumps/0.05 | Single |
| 15  | X940e/X945e | Single/0.5 | Spherical | Spherical |
| 16  | X950 | - | Spherical, bumps/0.05 | Spherical |

In Tables 3 and 4, they also entered into information about the appearance and the size of grains. The grains adopt spherical or cylindrical shapes and some of them look like perfect spheres, others are irregular with sharp or rounded edges or look like “potato,” and others are more or less elongated. The grains form clusters and chains or occur in the single form. The
different sizes and shapes were observed. The grain diameter was calculated from 3 to 25 µm. A thorough description of distinctive features of toners was included in the more distant part of the article.

Before starting a series of examinations, ground test without the toner and trial tests were carried out to eliminate possible features coming from a coated layer of chromium or carbon. Letter samples without coating were examined.

Figure 2d shows analysis results in magnification ×15,300 for printouts obtained from T650n/T652/T654n monochrome laser printer. The analytical conditions of observation were as follows: working distance – 5 mm and accelerating voltage in HV – 5 kV. The presence of the same characteristics was confirmed for this kind of toner both before and after coating. In consideration of toner sensitivity to electron beam, the analysis was conducted merely up to ×15,300 of magnification.

#### Table 4: Characteristic feature of the toner surface on tested printouts - replacements

| Sample ID | Model   | Type of device               | Solid surface | Granular surface |
|-----------|---------|------------------------------|---------------|------------------|
|           |         |                              | Irregular/ diameter (µm) | Regular/ diameter (µm) | Rod structures/ length (µm) | Shape of the grain | Structures on the gran surface/ diameter (µm) | Grains diameter (µm) |
| 17        | E240n   | Monochrome laser printer     | 0.05-2        | -                | Single/2               | Irregular + spherical | Beneath surface/ irregular/0.05 | 5-15               |
| 18        | E250d   |                              | 5.0           | -                | Single/2               | Irregular + spherical | Irregular/0.3 Bumps/0.05 Single rods/2 | 4-8                |
| 19        | T640/T642/T644 | 1.0 | - | - | Irregular | Irregular/0.05-0.5 Beneath surface/ irregular >0.3 | 5-20               |
| 20        | T634    |                              | Numerous >1.0 | - | - | Spherical irregular elongated | Beneath surface/ irregular >0.3 | 6-15               |
| 21        | C780    | Color laser printer          | 2             | -                | -                      | Irregular elongated + spherical + cylindrical | Bumps/0.05 Beneath surface/ irregular/0.2 | 7-15               |

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**Figure 1:** SEM images of the characteristic feature of toner

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**Figure 2d:** Analysis results in magnification ×15,300 for printouts obtained from T650n/T652/T654n monochrome laser printer. The analytical conditions of observation were as follows: working distance – 5 mm and accelerating voltage in HV – 5 kV. The presence of the same characteristics was confirmed for this kind of toner both before and after coating. In consideration of toner sensitivity to electron beam, the analysis was conducted merely up to ×15,300 of magnification.
On the printouts obtained from three monochrome laser printers, significant differences in the distribution of the toner layer in such low magnifications ranging from ×100 to ×200 were observed.

In the case of the printouts obtained from the E260/E360 printer [Figure 2a], the solid surface is a small part of total surface of the one letter and occurs in the form of small areas – “patches” of different sizes; whereas, in the printouts obtained from the MS 610dn printer [Figure 2b], the area of solid surface constitutes even smaller part of the surface of the printout and is shown in the form of elongated areas.

On the printouts obtained from the T650n/T652/T654n printer [Figure 2c], the solid surface prevails and effectively covers the whole area of one letter. This observation was made with the magnification ranging from ×100 to ×1000.

Further analysis of the images, at the level of magnification exceeding ×20,000, has shown numerous details of the structure of the toner of solid surface. On the printouts obtained from the E260/E360 printer, characteristic features such as “waves,” “cracks,” “layer accumulation,” and other irregularities were visible in the area of the solid surface [Figure 2a]. In this layer, numerous, empty “holes” could be seen. Only the structure could be seen through the paper in the “holes” inside. Furthermore, on the solid surface, numerous structures of various shapes and sizes were observed. These are spherical structures, “bumps” about 0.05 µm in diameter, and single “rod” structure with different lengths and irregular structures.

The toners on printouts obtained from the printers such as MS 610dn [Figure 2b] and T650n/T652/T654n [Figure 2c] have a similar topography. Only one difference was observed. It was a varied length of the “rod” structures located just beneath the surface of the toner. The length of the rod structures was calculated from 0.5 to 1 µm. These structures occurred both in pairs, one next to another and were placed in different directions, usually in a sequence: the shorter-longer or longer-shorter.

In the case of the printout generated by a color laser printer (C734/C736), the toner layer creates two zones: the solid and granular [Figure 3]. The area of the solid surface is interrupted with numerous areas of the granular surface. The solid surface is in the form of small “patches.”

The solid surface was folded, and numerous “cracks” and “holes” without the grains inside were also discovered. On the solid surface, numerous structures of various and irregular shapes and sizes up to about 2 µm in diameter were visible. In addition, on the area of the solid surface, many agglomerates of the other “spherical” structures were observed. They were “bumpy” structures sized about 0.05 µm in diameter.

In the next group of the monochrome laser multifunction devices, the printouts from seven models were tested [Table 1]. In general, in all printouts, two types of areas were observed: the solid and granular surface. In the examined printouts obtained from the four printer models such as X264/X363/X364, MX410, X463/X464/X466, and X658de, a significant dominance of a granular surface was observed [Figure 4a, b, d, and e]. The solid zone occurs in the form of a few and small “patches.” In the case of printouts obtained from another three printer models: X654de/X656de, MX710/MX810, and X860de/X862de/X864de, the solid surface is in the form of numerous and large “patches” or covers the whole area of single letter [Figure 4c, f, and g]. The examination shows that it could be difficult to make individual identification of printouts exclusively by the features described above. More details were obtained with higher magnifications of the order of ×20,000.

On the printouts obtained from the printers such as X264/X363/X364 and MX410 [Figure 4a and b], the solid surface area was folded, and numerous “cracks,” “holes,” and other structures were observed. On this surface, numerous irregular structures are also visible. These structures were of different shapes and sizes.

On the printouts obtained from the X654de/X656de printer [Figure 4c], the solid surface was folded and its numerous “cracks” and “holes” were also detected. On this surface, numerous irregular structures of varying shapes (rod and grains) and sizes were observed. The structures on printouts obtained from the X654de/X656de printer [Figure 4c] revealed more differentiation in comparison with the printouts obtained from two different models of printer [X264/X363/X364 – Figure 4a and MX410 – Figure 4b].

In the case of the printouts obtained from the printers such as X463/X464/X466 [Figure 4d] and X658de [Figure 4e], the

![Figure 2: SEM images of the solid surface of toner on the printout obtained from monochrome laser printers: (a) E260/E360, (b) MS 610dn, (c) T650n/T652/T654n, (d) T650n/T652/T654n without coating](image)

![Figure 3: SEM images of the solid surface of toner on the printout obtained from color laser printer C734/C736](image)
solid surface was folded, and it also contained the “cracks,” “holes,” and other structures. On the solid surface or inside this surface, numerous irregular structures of different shapes were observed. In the case of the printouts obtained from the X463/X464/X466 printer [Figure 4d], the bent or straight “rod” structures of various lengths and structures and grains of different shapes and sizes were visible. However, in the case of the printouts obtained from the X658de printer [Figure 4e], a straight “rod” structure of varying lengths and other structures of various sizes and shapes were observed.

On the printouts obtained from the MX710/MX810 printer [Figure 4f], the solid surface was cracked and a numerous “holes” were visible. On the solid surface, a distortion, around the “holes” and “bulging” surface, looking like cones was noticed. On the toner surface or inside the surface, numerous small and irregular structures were found.

The last group was color laser multifunction devices. The observation was provided on the printouts derived from five models of printer [Table 1]. On the printouts obtained from all printers, the toner surface was created by solid and granular zones. In the case of the printers such as C540/C543/C544 [Figure 5a] and X543/X544 [Figure 5b], a solid surface was interspersed with the granular surface. The solid surface resembles “patches” of various sizes in the area of one letter. In the C540/C543/C544 printer [Figure 5a], the solid surface area of one letter is larger than area of one letter on the printouts obtained from the X543/X544 printer [Figure 5b]. In the case of printouts obtained from the subsequent three models of the printers such as X920de [Figure 5c], X940e/X945e [Figure 5d], and X950 [Figure 5e], the solid surface prevails over the granular surface. The granular surface creates the area – “patches” of various sizes and shapes depending on the model device. The difference is the orientation of both regions (solid and granular). Within the printed letters, different patterns were visible. The patterns of different sizes and shapes were observed. However, the individual identification is difficult solely on the basis of the presented features. More details can be seen in the magnification of ×30,000 on the toners surface.

On the printouts obtained from the C540/C543/C544 printer [Figure 5a], the solid surface was folded. Numerous “depressions” and small, empty “holes” were observed. In some places, on the solid surface, the outline of a grain shape was visible. Numerous spherical structures too with diameter of 0.05 µm and an irregular structure with different shapes and diameters of the order up to 1 µm were observed.

On the printouts from the X543/X544 printer [Figure 5b], several “depressions” on the solid surface were visible. The “depression” surface was smooth and no other structure was visible. In another area, numerous irregular structures of different sizes were visible. In other places of this solid surface, the spherical structures and “bump” structures of different sizes from 0.02 to 0.05 µm in diameter, forming large clusters, were observed. The “rod” structure of different lengths from 0.06 to 0.08 µm and another irregular structure of the order up to 0.5 µm of diameter were observed.

On the surface of the solid area of the toner on the printouts obtained from the X920de printer [Figure 5c], surface irregularities were observed. On this surface, several “cracks”
and “depressions” occur. The numerous spherical structures and “bump” structures of the order up to 0.5 µm in diameter were observed. These small structures create smaller or larger clusters or they occur in a single form. The small clusters are visible at the border of “depressions.” Inside the “depressions,” the toners’ surface was smooth without other structures.

A completely different structure of the toners’ surface was observed on the printouts obtained from the X940e/X945e printer [Figure 5d]. Here, the toner surface was quite smooth. There were only few surface irregularities. On the toner surface, numerous spherical structures or delicately visible shapes of the order up to circa 0.05 µm in diameter and a single irregular structure of the order up to circa 0.5 µm in diameter were observed. These structures created a few small clusters.

In the case of the printouts obtained from the X950 printer [Figure 5e], the solid surface was smoothly accompanied only by a few depressions. On the toners’ surface, spherical and “bumpy” structures of the order up to circa 0.05 µm in diameter were observed. This structure occurs as the single form or creates chains or smaller and larger clusters of various densities.

We can accurately look at the structure of the toner using the opportunities afforded by the scanning electron microscopy.

As previously mentioned, the toner powder is characterized by the presence of the grain. In higher magnifications of about 30,000 times, we can observe single grains. Hence, we can determine the size, shape, and structural surface of a single grain. It appears that the toners contained not only the different shapes of grains but also different structures of the surface. The grains adopt different shapes and some of them look like perfect spheres, others are irregular with sharp or rounded edges or look like “potato” or “the grain of rice,” and others are more or less elongated. The grains form clusters and chains or occur in the single form. A characteristic feature of the toner is the distribution of grains and the appearance of clusters, which are frequently in the middle of the line of print or on the inside line of the print and the manner in which the grains are connected: clusters, aggregates, and chains.

In the case of printouts obtained from the C734/C736 printer [Figure 6d], a granular surface is characterized by single grains and their clusters. The grains create the “clusters,” agglomerated together in the shape of “grapes” and “chains.” The grains are connected by a large part of their surface. At the border of two areas such as granular and solid surface, a molten surface with clearly visible grain shape is visible.

In other printouts obtained from three printer models such as E260/E360, MS 610dn, and T650n/T652/T654n [Figure 6a-c], the granular area showed a variation in the structure: molten grains, forming agglomerates with different shapes depending on the model used. In this case, we do not observe numerous grains, only the scratched fragments of the molten surface with delicately visible shapes of grains or several single grains connected to the molten surface by larger part of their surface. The monochrome laser printers can be distinguished from color laser printers [Tables 1 and 2] based on the described features.

In a group of the printouts obtained from the monochrome laser multifunction devices, some differences between models were observed. However, a few models have the same features as it was observed on the printouts obtained from the monochrome laser printers. For example, in the printer models such as X264/X363/X364 [Figure 6e], MX410 [Figure 6f], X654de/X656de [Figure 6g], and X463/X464/X466 [Figure 6h] in the grain area of the toner, the molten surface with delicately visible shapes of grains was observed. In this type of area, only single grains were visible or they were missing. This area had different sizes and was characterized by various shapes and relocation of the molten fragments or agglomerates, depending on the model device. While on the printouts obtained from three printer models such as X658de [Figure 6i], MX710/MX810 [Figure 6j], and X860de/X862de/X864de [Figure 6k], many more single grains with clearly visible grain shape were observed. However, the number of grains was much fewer than in the case of toners used in other printers. The molten fragments of the toner with different sizes and shapes and distribution on the area of the one letter were found. These features distinguished the tested printouts from other groups of devices. The printouts obtained from the color laser multifunctional devices revealed the difference on the examined models. However, it was significantly different from the group of the monochrome laser multifunctional devices. But, some similarities in the group of color laser printers were revealed. Numerous single grains were observed. With these grains, clearly visible structures in the form of “grapes” – C540/C543/C544 [Figure 6l] and “grapes” and “chains” – X543/X544 and X950 [Figure 6m and p] were created. In the case of described models of printers, the toner grains occur in pairs or in three connected grains or larger clusters. The grains were connected together by the smaller part of their surface. This type of grain connection is different than in the color laser multifunctional devices group, where

![Figure 5: SEM images of the solid surface of toner on the printout obtained from color laser multifunction devices: (a) C540/C543/C544, (b) X543/X544, (c) X920de, (d) X940e/X945e, (e) X950](image-url)
the grains are connected on the larger part of their surface. There are examples in the color laser multifunctional devices group, where in the granular area of the toner, the molten fragments and single grains were observed \([X920de – Figure 6n
and X940e/X945e – Figure 6o]. This feature characterizes to more extent the toners in monochrome laser printer group. However, the difference lies in the distribution and shape of these structures. In the magnification, 100,000 times or more the surface of single grain can be observed. This surface can be smooth, scratched, and folded. On the grain surface depressions, structures looking like “needles,” spherical structures, “bumps,” rods, or other irregular structures were observed.

On the printouts obtained from the monochrome laser printers, the grain surface in all free models of printers [Table 1] was different. Numerous single grains or their groups were observed in the middle part or outside the printed line of single letter. The irregular grains or their groups were distributed in layers one by one. On the grain surface or inside this surface, numerous small structures of various shapes were observed [Figure 7].

In the case of printouts obtained from the \textit{E260/E360} printer [Figure 7a] on the grain surface, the “bumps” structures of the order up to circa 0.05 µm in diameter were visible. A “bumpy” structure occurs in small clusters. The rod structures with lengths of about 1 µm and a few irregular structures were found. The grain diameter was calculated from 3 to 8 µm.

On the printouts obtained from the \textit{MS 610dn} printer [Figure 7b] on the grain surface of toner, the “bumps” structures of the order up to about 0.05 µm in diameter are visible. The grain surface was folded. A “bumpy” structure occurs in the clusters, especially on the border of the “depressions.” On the surface of single grain, numerous depressions were observed. The surface of “depressions” was usually smooth without another visible structure. On the grain surface, a “rod” structure was also visible of the length of about 0.5 µm. These structures occurred both in pairs, alongside. On the grain surface, a few irregular

\textbf{Figure 6:} SEM images of the granular surface of toner on the printout obtained from printers: (a) \textit{E260/E360}, (b) \textit{MS 610dn}, (c) \textit{T650n/T652/T654n}, (d) \textit{C734/C736}, (e) \textit{X264/X363/X364}, (f) \textit{MX410}, (g) \textit{X654de/X656de}, (h) \textit{X463/X464/X466} (i) \textit{X658de}, (j) \textit{MX710/MX810}, (k) \textit{X860de/X862de/X864de}, (l) \textit{C540/C543/C544}, (m) \textit{X543/X544}, (n) \textit{X920de}, (o) \textit{X940e/X945e}, (p) \textit{X950}.
structures of the order up to about 0.05 µm in diameter were found. The grain diameter was calculated from 5 to 10 µm.

In case of printouts obtained from the T650n/T652/T654n printer [Figure 7c], the grain surface of toner was similar to the surface of the grain on printouts obtained from the MS 610dn printer [Figure 7b]. Only the “rod” structures were of various lengths, calculated from 0.5 µm to 1 µm. These structures were chaotically located on the grain surface. The grain diameter was calculated from 4 to 8 µm.

On the printouts obtained from the color printer device [C734/C736 – Figure 7d] in the toner, grains of different shapes and surfaces were observed. Numerous single grains or groups...
of grains were observed in the middle part or outside the printed line of the single letter. A few grains have much more spherical shape, and the other grains were irregular. The grain surface was less or more folded, and on the surface, “depressions” and other irregularities were observed. On the surface of the grain, numerous small “bumpy” structures of about 0.05 µm in diameter were found. The grain diameter was calculated from 5 to 10 µm [Figure 7d].

In the granular area of the toner on the printouts obtained from the monochrome laser multifunctional devices in all tested models [Table 1], the grains melted in the surface of the toner with clearly visible grain shape were observed. In the case of printouts obtained from the printers such as MX410 [Figure 7f], X463/X464/X466 [Figure 7h], X658de [Figure 7i], MX710/MX810 [Figure 7j], and X860de/X862de/X864de [Figure 7k], numerous single grains or groups of grains were observed in the middle part or outside the printed line of single letter. In the printers such as X264/X363/X364 [Figure 7c] and X654de/X656de [Figure 7g], a few single grains or groups of grains were observed only outside the printed line of a single letter. In addition, the toner on printouts obtained from the X463/X464/X466 printer [Figure 7h] and MX710/MX810 printer [Figure 7j] was characterized by a clearly visible multilayer granular area — grains and their groups were on the top of each other. The grain shape and the grain surface were different in various printers.

On the printouts obtained from the X264/X363/X364 printer [Figure 7c] on the surface of the toner, irregular grains were observed. The grain surface was less or more folded. On the surface of a single grain, “depressions” and “bulge” were observed. Numerous structures of various shapes and sizes were visible on the grain surface, inside of grains, and under the grain surface. The agglomerates of spherical structures with about 0.05 µm in diameter on the grain surface were observed. Under the grain surface, “rod” structures with various lengths up to about 1 µm were visible. These structures were delicately bent and chaotically located. On the grain surface and beneath the grain surface, a single irregular structure of different sizes and shapes and its clusters were found. The grain diameter was calculated from 5 to 8 µm.

In the case of printouts obtained from the MX410 printer [Figure 7f] below the grain surface, straight “rod” structures with various lengths up to about 0.3 µm were visible. The characteristic feature of the toner in this model type was a diameter of grains, calculated at about 8 µm. The diameter was similar for all grains.

On the printouts obtained from the X654de/X656de printer [Figure 7g] on the surface of toner, the irregular grains with about 0.1 µm of diameter were observed. The grain surface was folded. On the surface of single grain, “depressions” and “rod” structures of various lengths from 0.4 µm to about 1 µm were visible. The grain diameter was calculated from 12 to 18 µm.

On the printouts obtained from the X463/X464/X466 printer [Figure 7h] on the surface of the toner, irregular grains were observed. Some grains resembled the shape of a “potato.” The grain surface was folded, and numerous “depressions” and other irregularities were observed. On the grain surface, the “bumpy” structures of various sizes from 0.05 to 1 µm of diameter were found. Below the grain surface, a few “rod” structures with various lengths of up to about 2 µm were visible. The grain diameter was calculated from 6 to 14 µm.

In the case of the toner on the printouts obtained from the X658de printer [Figure 7i], grains of different sizes were observed. Some grains had more spherical and others were of a slightly elongated shape. The grain surface was folded, and numerous “depressions” and other irregularities were observed. On the grain surface, numerous spherical structures of 0.05 µm in diameter were found under the grain surface, and on the grain surface, numerous “rod” structures of different lengths from 0.2 µm to about 1 µm and irregular structures with 0.3 µm in diameter were visible. The grain diameter was calculated from 8 to 25 µm.

The grains of the toner on the printouts obtained from the MX710/MX810 printer [Figure 7j] are characterized by irregular form. Various surfaces of the grains were observed. The grain surface was folded and numerous “depressions” and other irregularities were observed. On the grain surface, the “bumpy” structures of various shapes and sizes from 0.05 to 1 µm in diameter were found. Under the grain surface, the irregular structures of about 0.3 µm in diameter and a few “rod” structures with different lengths from 0.2 to about 1 µm were visible. The grain diameter was calculated from 5 to 10 µm.

In the case of printouts obtained from the X860de/X862de/X864de printer [Figure 7k], the grains of different shapes (spherical or elongated) were observed. The grain surface was folded and

Figure 8: SEM images of the solid surface of toner on the printout obtained from monochrome and color laser printers: (a) E240n, (b) E250d, (c) T640/T642/T644, (d) T634, (e) C780
numerous “depressions” and other irregularities were observed. On the grain surface, the “bumpy” structures of the order up to about 0.05 µm in diameter were found. In the few places, the surface of the grain was smooth without other structures present. This place looks like the patches on the grain surface. The grain diameter was calculated from 5 to 8 µm.

In the granular area of the toner on the printouts obtained from the color laser multifunctional devices in all tested models, the grains melted in the surface of the toner were observed. In the case of three printers such as C540/C543/C544 [Figure 7j], X543/X544 [Figure 7m], and X920de [Figure 7n], numerous single grains or their groups occur in the middle part or outside the printed line of the single letter. On the printouts obtained from the X940e/X945e printer [Figure 7o], a few single grains or their groups outside the printed line of a single letter were observed. While on the printouts obtained from the X950 printer [Figure 7p], numerous single grains or their groups only outside of the printed line of the single letter were visible.

Depending on the printer tested, the differences in the shape and surface of the grains were visible. A few spherical grains and delicately elongated in the four printers such as C540/C543/C544 [Figure 7j], X543/X544 [Figure 7m], X940e/X945e [Figure 7o], and X950 [Figure 7p] were observed. Other forms of irregular grains were found, like in the toner obtained from the C540/C543/C544 [Figure 7j] and X920de [Figure 7n] printers.

On the printouts obtained from the C540/C543/C544 printer [Figure 7j], the grain surface of toner was folded and “depressions” and other irregularities were observed. On the surface of grain, a numerous small “bumpy” structures up to about 0.05 µm of diameter were found. The grain diameter was calculated from 5 to 8 µm.

On printouts obtained from the X543/X544 printer [Figure 7m] on the grainy surface, a numerous small “bumpy” structures with various sizes from 0.02 to 0.1 µm in diameter were found. These structures create numerous clusters on the surface of the single grain. “Rod” structures of different lengths from 0.5 µm to about 1 µm located on the grain surface were visible. The grain diameter was calculated from 5 to 8 µm.

On the printouts obtained from the X920de printer [Figure 7n], the grains with smooth surface and some large “depressions” were observed. On the surface of the grain, numerous small “bumpy” structures up to about 0.05 µm in diameter were found. This small structure occurred in the form of single grains or created numerous clusters on the grain surface. The grain diameter was calculated from 5 to 8 µm.

In the case of printouts obtained from the printers such as X940e/X945e [Figure 7o] and X950 [Figure 7p], the grains with the folded surface were observed. On the grainy surface, numerous “depressions” and numerous small spherical structures up to about 0.05 µm in diameter were found. These small structures occurred in the form of single grains or created smaller or larger clusters on the grain surface. The grain diameter was calculated from 5 to 10 µm for the X940e/X945e printer [Figure 7o] and from 5 to 8 µm for the X950 printer [Figure 7p].

In addition, the replacements were used for analysis. On the printouts obtained from the four monochrome laser printers, significant differences in low magnifications from ×100 to ×200 were observed. In the case of printouts obtained from the Lexmark E240n [Figure 8a], Lexmark E250d [Figure 8b], Lexmark T640/T642/T644 [Figure 8c], and Lexmark T634 [Figure 8d] and Lexmark C780 [Figure 8e], a significant advantage of the solid surface was observed and/or completely covered the area of a single letter. The solid surface of toner was folded. On the solid surface, characteristic features such as “waves,” “cracks,” and “foundations” and characteristic structures with various shapes and sizes were visible. Numerous “cracks” were found on the printouts obtained from the E250d printer [Figure 8b]. On the printouts obtained from the Lexmark E240n [Figure 8a] and Lexmark T634 [Figure 8d] printers, on the toner surface and under this surface, numerous small structures of different shapes and sizes about 0.05 µm in diameter were visible. In other printers, the structure placed under the surface occurs at a much lower density. A smooth and clear area without another structure was observed. In the case of printouts obtained from the Lexmark E250d printer [Figure 8b], a few single “rod” structures with length about 2 µm were found.

In the case of printouts obtained from the monochrome laser printers such as Lexmark E240n [Figure 9a], Lexmark E250d [Figure 9b], Lexmark T640/T642/T644 [Figure 9c], and Lexmark T634 [Figure 9d] on the granular surface, single grains and their clusters were observed. These grains created the agglomerates and “chains.” The grains are connected by the large part of their surface. On the outside of the printed line of the single letter, less or more single grains were visible.

Figure 9: SEM images of the granular surface of toner on the printout obtained from monochrome and color laser printers: (a) E240n, (b) E250d, (c) T640/T642/T644, (d) T634, (e) C780
The printouts obtained from the color laser printer C780 [Figure 9e] were different to the tested printouts obtained from the monochrome laser printers. Numerous or a few single grains in the middle or outside the printed line of the single letter were spotted. The grains occurred in pairs or created large clusters. The grains are connected by lesser or larger part of surface together.

Further analysis was provided in higher magnification in about 100,000 times and more.

On the printouts obtained from the four monochrome laser printers, the grainy surface was quite different. On the printouts obtained from the E240n printer [Figure 10a], the grain surface was folded with numerous “depressions.” Numerous small structures of different shapes and sizes up to about 0.05 µm of diameter occurred beneath the surface. Single “rod” structures of the length about 2 µm, chaotically located on the surface of the grain, were visible. In the granular layer of toner, the irregular grains and single spherical grains were observed. The grain diameter was calculated from 5 to 15 µm.

On the printouts obtained from the E250d printer [Figure 10b], the grainy surface was delicately folded with some “depressions.” On the surface of the grains, some “bumpy” structures of about 0.05 µm in diameter were observed. These small structures created small agglomerates. Under the grainy surface, single “rod” structures with the length about 2 µm occurred. Different shapes of grains were observed: irregular, less or more spherical, and delicately elongated. The grain diameter was calculated from 4 to 8 µm.

In the case of printouts obtained from the T640/T642/T644 [Figure 10c] and T634 [Figure 10d] printers, the grain structure was similar. In both cases, under the surface of the grains, single irregular structures with various sizes up to about 0.05 µm of diameter were visible. The grains adopted spherical shapes, but more numerous were irregular. In the case of printouts obtained from the T640/T642/T644 printer, the grain surface was smooth and delicately folded. However, on the printouts obtained from the T634 printer on the grain surface, numerous small and round “depressions” were observed. The grain diameter was calculated from 5 to 20 µm for the T640/T642/T644 printer and from 6 to 15 µm for the T634 printer.

On the printouts obtained from the color laser printer C780 [Figure 10e], spherical grains, delicately elongated grains, and irregular grains were observed. The grain surface was folded and single “depressions” were visible. On the surface of grain, “bumpy” structures of about 0.05 µm in diameter and numerous irregular structures with different sizes up to about 0.2 µm in diameter were found. The “bumpy” structures occur in small clusters on the grain surface. Under the surface, numerous and small structures of different shapes and sizes were visible. The grain diameter was calculated from 7 to 15 µm.

In conclusion, attention should be paid to printouts from those devices whose grains show the presence of structures...
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visible just under grain surface or on it. Grains of this kind appear mainly in monochrome device printouts. The grain surface seems transparent up to a certain depth. Judging from microscope image contrast, we can conclude that different element composition is characteristic of these structures than of the grain itself. It can be clearly noted in BSE-mode picture [Figure 1], where brighter color structure can be observed. To determine the elemental composition microanalyzer EDS examinations were carried out. EDS analysis confirmed that the toner sample is rich in carbon (C), which proves the presence of organic components. Additional analysis demonstrated the presence of elements such as oxygen, iron, silicon, calcium, sulfur, aluminum, magnesium, manganese, and phosphorus. However, a serious problem is the size of the structures and limited capabilities of the EDS analyzer. The analyzer collects data also from the area around the analyzed structure. As a result, it cannot be certain that the outcome is from the analyzed structure only since it may also be from the surface. The problem could be solved by conducting research with the use of WDS analyzer.

**Conclusion**

Implementation of the scanning electron microscopy for forensic document research has created new opportunities for the identification of laser printing devices through the selection of group and individual prints derived from different models. Investigations have been carried out with a series of toner prints from 21 laser Lexmark printing devices. For this purpose, toners and their replacements were used. Microscopic images obtained allowed us to allocate characteristic features of toners for specific groups of devices: monochrome laser printers, color laser printers, monochrome multifunction devices, and color multifunction devices.

Clear criteria were established for individual assessment, which allowed to assign and select distinctive features of toners on the printouts obtained from the respective printer devices. On the printouts, step by step, the assessment of the characteristic features of respective toner was examined. The evaluation of the characteristic features of the toner on the printouts was carried out gradually, determining the distribution of the surface of solid and granular layer in low magnifications from ×100 to ×1000 (single visible letter). In higher magnification from ×2000 to ×15,000, specific features of the existed solid and granular surface were analyzed obviously after determining that the toner contained solid layer and/or granular layer. The last step of the analysis was to visualize individual grains and their surface in magnification over ×100,000. Based on the SEM images and established criteria, the details of topography of the toner structure were revealed. Based on this study, the differences or similarities of toners on tested printouts were determined.

Therefore, the differences in the distribution of the layer of toner were observed. Such a layer can be solid, smooth, and covering the entire surface. Only a few single grains on the coastline of toner of single letter are visible. The toner layer contains fragments of the solid surface, interrupted more or less by single or multiple areas with varying density of grains, or contain completely granular layer.

In some cases, certain features such as bent rod structure (X264/ X363/X364), a single spherical structure resembling balls scattered about on the surface of a toner (X940e/X945e), and clearly the same size (MX410) are individual for a specific print job. Toners, creating text on the printouts, are characterized by some features that allow you to distinguish among them, but only within certain limits. Unfortunately, many of these features are repeated in different models, which significantly impede their individual identification. However, these features allow you to specify the exact group of printing devices (monochrome, color).

The grain surface is also characterized by a degree of density of the structure and the structure present on the grain surface, or structures visible just beneath grain surface, or in both combinations.

Unfortunately, we were unable to get a printout of the same device using both original toner and its replacement. The question appears whether the same features are retained if we change the original for another original or its replacement. In the first case, within the same cartridge batch, we are going to observe the same features. Original toner shows repeatable features. In case of replacements, they will definitely differ. Finding the differences between replacements is much more difficult since one replacement is used for several different models. In this case, the toner typically has similar features, which makes it difficult to identify and even becomes impossible. Apparently a number of printouts were too low, so it does not give such a broad picture on the structure of the toners.

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**Conflicts of interest**

There are no conflicts of interest.

**References**

1. Merrill RA, Bartick EG, Taylor JH 3rd. Forensic discrimination of photocopy and printer toners I. The development of an infrared spectral library. Anal Bioanal Chem 2003;376:1272-8.
2. Munson T. The classification of photocopies by Py–GC–MS. J Forensic Sci 1989;34:352-65.
3. Chang WT, Huang CW, Giang YS. An improvement on Py–GC–MS. J Forensic Sci 1993;38:843-63.
4. Trzcińska BM, Brozek-Mucha Z. The possibilities of identifying photocopy toners by means of infrared spectroscopy (FT-IR) and scanning microscopy (SEM-EDX). Mikrochim Acta Suppl 1997; 14:235-7.
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5. Trzcińska BM. Classification of black powder toners on the basis of integrated analytical information provided by FTIR and XRF spectrometry. J Forensic Sci 2006;51:919-24.

6. Almeida Assis AC, Babosa MF, Valente Nabais JM, Custódio AF, Tropeceo P. Diamond cell Fourier transform infrared spectroscopy transmittance analysis of black toners on questioned documents. Forensic Sci Int 2012;214:59-66.

7. Szczepańczyk S, Konarowska U. Application of the optical microscopy for verification of documents obtained from the laser printer devices. Forensics problems 2012;276:65-8.

8. Łasińska A. Scanning electron microscopy in forensic science. Prosecutors Dep Law 2013;10:1-25.