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Use of atomic force microscopy in the forensic application of chronological order of toners and stamping inks in questioned documents

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ABSTRACT

This paper describes the application of the atomic force microscopy (AFM) as a nano-indentation method and introduces a new method of identifying the chronological order of the application of the toner and stamping ink on the surface of documents by removing either of them. Various toners were used as samples for the AFM nano-indentation method. The chronological order of the application of the toner and stamping ink with either the toner placed over the stamping ink or the stamping ink placed over the toner, could be identified, regardless of the kinds of toners made by various companies. This paper provides the new approach for physically removing the toner and checking the material below it to identify questioned documents, which allows the method to be used to appraise documents forensically. Blind testing has shown that the method to analyze the chronological order of toner-printed documents and the seal stamping on them could accurately identify the order in all samples, while minimizing damage to the samples.

1. Introduction

Seals date back to around a thousand years ago, and their history, systems, and types have changed in diverse ways according to the times. The seal was first introduced to Korea in approximately 2nd century BCE. In Korea, Japan, China, and other East Asian nations, seals are widely used as a mean of identifying a person [1]. In South Korea, about 200 administrative documents per year require the submission of registered seal impressions. This requires the verification of seal impressions printed on documents related to the transfer of ownership of major properties, the establishment of liens, and other important legal acts [2]. Seals are crafted by carving a person/institute's name on the surface of relevant documents. Seal impressions refer to the traces that are created by stamping ink seals on paper, etc.

There are, currently, several methods of seal impression forgery that are used in crimes. The first is the method of direct carving by hand or machine. Mechanical-carving forgery uses the original film that contains the photographed true seal impression, and carves a seal using a photoelectric carving machine. The second method is photoengraving, such as lithography which photographs the original seal impression with the same dimensions, exposes the metal blockpress plate or the photosensitive letterpress plate, and lets it corrode or be developed. Lastly, a method involves transcription, which covers the original seal impression with an absorbent medium. In this case, the seal impression forgery is confirmed by attaching the medium and fixing the seal impression part on it. By applying a certain force onto it, a strong friction occurs, and a copy of the impression is on the surface of the medium. The copy of the impression in then transferred onto another sheet of paper.

In addition to these impression forgery methods, there is a method of copying the original seal impression on a piece of blank paper, behind the original document, when arranging an agreement, then printing different content on that blank paper. In this case, courts can not solve disputes between the parties involved, because the seal impression itself is the original one; thus, the chronological order of the application of the toner and the stamping ink would need to be identified.

A study was published, reporting that polarized microscope-assisted visual observation enabled the chronological application of the toner and the stamping ink. Yoshimitsu et al. [3] and Yoshida argued that the use of a red filter enables the observation of letter cuttings and the appraisal of forgery [4]. However, these two

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methods make it difficult to observe seal impressions since the amount of the pigment-based ink is small. It was likely that they depend on subjective experience rather than objective data. Unlike these methods, Lee et al. used the TOF-SIMS, which analyzed the components of the red stamping ink present on the paper surface, and confirmed the manufacturer of the ink, as well as detected the components of the stationery ink or the stamping ink present on the paper surface, to analyze the chronological printing order [5]. They analyzed the particular components of the stamping ink applied on the paper surface; but only when the weight percentage of Ca and Pb is over 10 (wt.%) they can be detected, thus limiting the method. This limitation came from the relatively large background spectrum of the paper. Also, there is a possible damage of the samples in the process of mounting the samples before analyzing them. The method would be complicated because the spectrum of paper's background could be detected in the presence of ink on the paper. Marquis et al. used the digital microscopy, which analyzed chronological order of toners and rollerball pen ink on the paper surface [6], While the optical(digital) microscope observations are relatively simple, the method can be useful when the crossing area is relatively big. On the other hand, this study is much more useful when the crossing area is very small (even almost invisible area). And the method of identifying the chronological order of the application of ink-printed text and stamping ink [7], toner and seal ink [8] were investigated on the surface of documents

Recently, red seal inks were investigated by not only the micro-attenuated total reflectance (ATR) FTIR spectroscopy [9,10] but also laser desorption ionization mass spectrometry [11].

1.1. Toners and stamping inks

With the spread of office automation devices, many toner-printed documents are being produced, and so it is significant to appraise them forensically. Various toners have been developed along with plain paper copiers (PPCs). They are classified into wet and dry toners. Dry toners currently prevail, and they can be classified into one-component toners and two-component toners. One-component toners use only toners, whereas two-component toners use a carrier (metal) and a toner (coloring powder). The use of one-component toners has been increasing due to the low prices associated with the smaller-sized and simpler machines, but two-component toners would have prevailed in the market [12]. Toners are also widely used in electrostatic presses, electronic platemaking machines, and laser printers, which all use the electrophotography system. The electrophotographic process has six subprocesses: electrification, exposure, development, transcription, fixing, and cleaning. Of these, toners are directly involved in the development, transcription, fixing, and cleaning processes, and they thereby require diverse properties (electrical, mechanical, thermal, and rheological) for each of these processes. Also, toner components include thermoplastic resins, salt or pigments, charge control agents, magnetic pigments, and fluidization agents. The electrophotographic process attaches the toner onto paper, etc., and makes the height a few micro meters.

The toner is attached to paper because not only the heat and pressure involve in the fusing process but also paper has many pores [13–16]. This toner penetration pattern is influenced by the fixing temperature, pressure, and property changes in paper and toners. For instance, it is influenced by the surface energy, surface illuminance, distribution of sizes, viscosity, surface tension, and thickness of the toner layer.

As the fixed temperature is raised, the toner viscosity decreases, which allows the toner to flow into the pores in the paper. Thus, higher fixed temperatures would allow the toner to more deeply penetrate the paper pores. Also, a higher fixing pressure prompts the toner to flow into the paper pores from the paper surface The larger the contact surface of the toner with the paper is, the greater the increase in the amount of its penetration will be, which would improve its attachment to the paper [17].

Red stamping ink has long been used with the development of seals, and is now being highly used in Asian countries (e.g. Korea, China, Japan, etc.). Stamping ink is normally red, as it uses red pigments such as vermilion, cadmium red, and lake dyes. Premium varieties of stamping ink use silver red and cadmium red, whereas ordinary stamping ink uses a mixture of lake dyes. In Asian countries, red stamping ink pigments have traditionally used vermilion since a procedure incorporating sulfur and mercury into a synthesized vermilion had recorded in the 8th century BCE [1]. Colors can be formulated to range from orange to dark red depending on the manufacturing method. Pigments are not dissolved in acids and alkalis, but in aquaregia and sodium sulfide. These pigments are not easily eliminated, but some of them turn black due to sunlight or heat.

The dry manufacturing method includes a chemical reaction of mercury with sulfur with each to produce mercury sulfide representing a black color; and further heats and sublimates it to be red color. The wet manufacturing method mixes mercury with potassium pentasulfide, for 2-3 days at over 45 °C. This lets the supernatant flow out, and treats the dregs with potassium hydroxide. These substances were widely used to make premium pigments, water colors, lacquers, and stamping ink. However, they are now being replaced by other pigments such as cadmium lead since they are more expensive. Stamping ink also contains vehicles and viscous agents. Vehicles such as castor oil are used to adjust the pigment concentration and weight while viscous agents such as resins are used to adjust the stamping ink viscosity. Vehicles and viscous agents have hydrophobic properties.

1.2. AFM principle and the proposed method

The AFM is a device that is designed to use the force of elements between the cantilever-end probe and the surface of the sample and measure the shape of the surface [18]. The interaction force between the cantilever-end probe and the sample surface can change the cantilever's state (bending/amplitude), and this change can be sensed by a change in the laser beams projected on the cantilever's upper side. Using this process, the structure of the sample surface can be imaged.

AFM analysis has the following strengths. It has a high resolving power with sub-nano scale, and it can also measure not only the morphology of the surface of samples with organic and inorganic components, but also the surface roughness, lateral force, phase imaging, hardness, and adhesion [19]. Also, AFM provides an advantage over conventional optical microscopy. It can implement images in three dimensions, x, y, and z (normal to the sample surface) while 2D image could usually obtained through conventional optical microscopy. Even it has been applied in various biosensor applications [20].

Ott et al. measured the modified surface of toner particles in diverse ways as well as the adhesion of the toner particles using AFM [21]. The same method for the adhesion of paper and the toner was not applied, however, to the research on the toner and stamping ink. Also, Kasas et al. [22], using AFM, identified the surface information on the crossings of the printer ribbon and the ballpoint pen ink in documents to determine the chronological order of the printing. In the method, the samples can be analyzed without damaging them, which is its strength; but this method makes it difficult to measure when the ink-applied portions of the surface are damaged. Reports have yet to be made regarding the analysis of the chronological sealing order of the stamping ink and toner-printed outputs using AFM.