



Rapid communication

Development of latent fingermarks on thermal paper: Preliminary investigation into use of iodine fuming

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ABSTRACT

Thermal paper finds its extensive use in the modern day life and could act as a vital piece of physical evidence carrying latent fingermarks. A large number of citations are available in literature suggesting various techniques to develop these marks but all are suffering with one or the other drawbacks such as complex and cumbersome procedure, pre- or post-treatment, background coloration and efficiency to develop aged fingermarks. In present study, a very simple and novel method involving iodine fuming has been suggested to develop fingermarks which were not only permanent but also without any background coloration. The suggested method does not involve any pre- or post-treatment of the substrate and was able to develop very old fingermarks (upto >1 year). In this study an attempt has been made to explain the reaction mechanism of the process. In case of different types of thermal papers, presence of different substituents on leuco dye (lactone ring) structure resulted in development of different colored fingermarks upon reaction with iodine. Sebaceous material laden marks have been found to be more intensely developed as compared to eccrine marks, and the difference was more pronounced in case of aged fingermarks.

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1. Introduction

In the present era of information and technology, fast printing procedures like thermal printing are being increasingly employed. Thermal printing requires special type of thermal paper being used exclusively in the routine activities like fax machines, ATM receipts, store bills, bus tickets and in laboratory and medical instruments such as spectrophotometer, electrocardiograph, etc. Due to this high usage and handling, probability of encountering this type of paper bearing latent fingermarks as possible physical evidence has also increased.

Thermal (thermosensitive) papers were introduced by National Cash Register Company in 1968 using the color forming reaction between leuco dyes and co-reactants [1]. Leuco dyes are essentially colorless solids, which forms color in presence of acidic components, electron-accepting compounds such as oxygen, iodine and heat. This reaction involves opening of the lactone ring structure that forms colored fluorane cation by extension of the conjugated double bond system. The ring opening of the lactone structure is reversible under basic conditions, which is the primary reason for the print fading on the thermal paper with the passage of time [2].

When subjected to heat, thermal papers produce black color which is indicative of the presence of unsubstituted leuco dyes while presence of substituted fluoran compounds form different color(s) such as red, green, yellow, black [3].

Development of latent fingermarks on thermal paper presents the crime scene investigators with a unique problem. Treatment with conventional techniques like ninhydrin petroleum ether or 1,8-diazafluoren-9-one (DFO), produces black color on thermosensitive side that conceals any fingermarks developed resulting in partial or no identification [4]. Alkyl analogues of ninhydrin soluble in the non-polar solvents have been reported to partially develop the fingermarks on the thermal paper [5,6]. Fuming of fax and thermal papers with dimethylaminocinnamaldehyde (DMAC) has also been reported to produce fluorescent fingermarks with no background coloration [7]. Treatment of thermal paper with various visualization reagents was carried out by the fingerprint and footwear forensics user group at Home Office Scientific Development Branch [8]. They concluded that pre-dipping of thermal paper in ethanol prior to processing with ninhydrin or DFO is the most effective method of preventing the blackening of thermal receipts. It is mentioned further that DFO and ninhydrin can both be used after DMAC in a sequential process. Latent fingermark development on thermal paper by metallo-porphyrin compound [(TPP) Sn (OH)₂] has been reported to produce better results in comparison to ninhydrin/DFO in different weather conditions [9]. Use of heptane as solvent with ninhydrin rendered

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comparatively less damage to the paper but resulted in fading of printed matter on the thermal paper. Ninhydrin along with HFE-7100 solvent has been reported to develop latent fingermarks on the thermal paper with minimum damage and discoloration of the paper surface [10]. Polar properties of ethyl acetate and HFE-7100 in conjunction with 1,2-indanedione have been reported to produce consistent fingermark development on different types of thermal paper [11,12].

Treatment of the thermosensitive side of thermal paper surface with DFO or ninhydrin petroleum ether (NPB) leads to blackening of the surface. Removal of blackening with acetone has been reported to produce pale and diffused fingermarks. Blackening of the surface could be minimized while developing latent marks with INON (2-Hydroxy-2-(3,5,5-trimethyl-hexyloxy)-indan-1,3-dione) or using ninhydrin sublimated in vacuum [13,14]. Reversal of the lactone structure ring opening to decolorize the blackened paper (as a result of treatment with ninhydrin petroleum ether formulation) for longer periods has also been reported by using "G3" solution containing pyrrolidone-based compounds [15]. Majority of the techniques discussed above are amino acid sensitive and could be used to process porous surfaces due to prolonged binding of amino acids with paper fibers [16].

In cases, where sebaceous component is predominantly more than the amino acid content, these techniques may not be able to produce the desirable results on porous substrates. For such cases, physical developer has been a method of choice due to its operational efficiency in terms of development [17]. Oil red O has also been recommended as possible alternative to physical developer due to the complex and destructive properties of the later [18]. Mean fingermark quality of the marks developed with oil red O has been found to be superior as compared to physical developer on different types of thermal papers [19].

Application of heat with relatively low temperature (80 °C) using hair dryer to develop latent fingermarks on thermal papers has also been identified as a potential alternative to conventional solvent-based methods [20].

Fuming treatment for development of latent fingermarks with chemical vapors has been recommended in lieu of solvent-based developing techniques due to possible non-destructive nature of the former. Materials included in these techniques consist of iodine, bromine, mercuric bromide, osmium tetroxide, etc. [21]. Use of fluorescing vapor phase chemicals like anthracene, rhodamines, etc., to develop latent fingermarks on paper surface has also been investigated [22]. Exposure of the thermal paper surface to fumes of hydrochloric acid, acetic acid, ethanol, methanol, isopropyl alcohol, HFE-7100 and n-hexane has been reported to develop identifiable quality fingermarks on thermal paper [23]. It was suggested to use acetic acid to develop fingermarks on thermal paper but their reaction mechanism or the permanency of the ridges has not been discussed. Rather caution has been mentioned regarding use of these chemicals. Highly corrosive nature of the hydrochloric acid has also been acknowledged [24].

It is strange to note that no work has been reported till date on the use of iodine fuming of thermal paper, although, the method has been known for years and being used as one of the first steps in sequential development process. Though, chemically, iodine crystals and resulting vapors have been recognized as corrosive and toxic but not classified as a carcinogenic [25].

Iodine fuming technique is sensitive towards the sebaceous component of the latent mark residue. Fingermarks developed on ordinary paper with iodine fuming have been found to be of non-permanent nature as iodine sublimates at room temperature resulting in fading of the developed prints. Champod et al. referring about limited sensitivity of this technique mentioned that prints older than 3–5 days are unlikely to be detected [16].

In the present study, thermal paper with latent marks has been treated with iodine fuming to visualize those latent fingermarks. Electron-accepting properties of the iodine and its reaction with leuco dyes (main constituent of the thermal paper composition) were explored to develop fingermarks on thermal paper surface. Response of eccrine and sebaceous fingermarks, fresh as well as aged, to iodine fuming has been reported.

2. Material and methods

Different type of fax and thermal papers were collected from the market for the deposition of latent fingermark samples listed as follows:

1. Mitsubishi fax paper.
2. Oddy fax paper.
3. D.P. Print paper (used in issuing receipts).
4. ATM paper receipts (collected from bank machines).

Latent fingermarks were impinged on the collected thermal papers by different individuals ($n = 10$) of unknown donor capabilities. Individuals were requested to wash their hands to make them free from any greasy matter and after sometime asked to impinge their latent fingermarks on thermal paper samples. These were labeled as eccrine fingermarks. Individuals were further requested to impinge another set of fingermarks by "charging" the finger tips by gentle rubbing of the same against cheeks or forehead. These fingermarks were predominantly sebaceous. In this way, two sets of latent fingermarks from each individual were obtained.

Fresh as well as aged fingermarks upto 1 year were tested for development during the course of study. Experiments for different thermal paper types were repeated several times during the course of study.

For observation regarding development of aged fingermarks on thermal paper, few samples were kept in the closed plastic box for upto 1-year period.

Iodine (Ranbaxy Inc., India) was used for fuming without any further purification. Standard fuming cabinet method [26] was used for developing latent fingermarks. Thermal paper samples were hanged inside the cabinet and few iodine crystals were placed in a petridish over the heating apparatus with thermostat inside the fuming cabinet. Iodine crystals were heated accordingly to produce mild amount of fumes sufficient enough to develop fingermarks, subjected to visual observation. Developed fingermarks were subjected to short UV treatment (5–10 min) as well as kept in dark to observe any difference between intensity. Developed marks were photographed using digital camera (Canon IXUS., Japan).

3. Results and discussions

Most important observation in the present study was the permanency of the developed fingermarks with iodine fuming technique on thermosensitive side of the thermal paper, while in case of treatment of the non-glossy (non-thermal) side of the paper surface, as usual, non-permanent fingermarks were observed, primary reason being sublimation of iodine. Developed fingermarks were identifiable showing third level details without any background coloration. Also, iodine treatment did not result in running of ink or fading of the printed material.

It was observed, however, that the degree of permanency of developed fingermarks (degree of durability/fade resistance) is dependent upon the type of thermal paper surfaces used in the study. "Mitsubishi" and "Oddy" brand fax papers showed permanency of developed marks for longer period as compared to bank ATM receipts and "D.P." brand store receipt paper. But, in all the cases the extent of fading of the developed marks never hindered the visibility and identification (Fig. 1) and developed marks remained permanent for months irrespective of type of papers.

It was also observed that treatment of different types of thermal paper resulted in development of differently colored fingermarks. Fresh fingermarks developed on Mitsubishi, Oddy and ATM receipts were green/greenish black in color with sharp ridge details while on D.P. print paper the same were black/brown/yellowish. Following para illustrating mechanism of the reaction will explain the reason for having different color of developed fingermarks.



Fig. 1. Fresh fingerprint showing clear ridge details developed on Mitsubishi brand fax paper and photographed after 475 days.

3.1. Mechanism of the fingerprint development

As mentioned earlier, fluoran leuco dyes used in the thermal papers undergo lactone ring opening in presence of acidic or electro accepting compounds like oxygen, iodine, etc. Iodine being a strongly electron-accepting compound oxidizes the leuco dye structure and form a stable structure with ring opening (extended conjugated double bond system), that ultimately leads to color formation (Fig. 2).

In case of latent fingerprint development, sebaceous component of the latent print residue provides a template for iodine to react with the leuco dye molecules present in the thermal paper. Due to the high surface area of the fatty acids, the iodine gets physically adsorbed on to the fingerprints and further, leads to more intensified oxidation reaction between iodine and leuco dye. On rest of the thermal paper surface, while iodine would have the same type of reaction with leuco dyes but the reaction was found to be too weak leading to almost no background coloration which affects neither contrast nor the legibility of the printed matter.

Electron acceptor initiated color development could be credited to the electron transfer process. Fluorane lactone (spiro[isobenzofuran-1,9'-xanthene]-3-one) (Fig. 2) is regarded as an electron

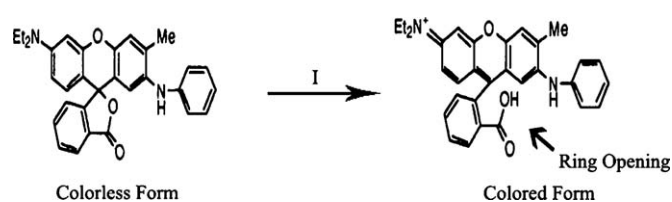


Fig. 2. Proposed mechanism of oxidation of the leuco dyes to produce colored complex.



Fig. 3. Fresh latent fingerprint developed on thermal paper by iodine fuming and kept in short UV (left) and in dark (right) for 15 min.

donor due to presence of dialkylamino group. Compatible electron acceptors like iodonium and sulphonium salts give efficient reactions with lactone structure in polymer film or melted stages [27].

Solution having reaction product of fluorane dye and iodonium salt has been reported to exhibit black color on exposure to the UV radiation [28] but in the present study, exposure of the developed fingerprints to UV treatment light and keeping them in dark revealed no difference in the fingerprint intensity (Fig. 3).

Variation in degree of fading in the developed fingerprints could be attributed to difference in the compositions of different types of thermal papers. One of the constituents of the top coat of the thermal paper are the antioxidant stabilizers, e.g. Cyanox 425 antioxidant, Cyasorb UV 531 light stabilizer, which reduce the reversibility of the oxidation reaction (reason for fading of developed marks) and aid in preserving the mark [29]. Also, grade assigned to a particular thermal paper type defines the contrast, legibility and whiteness and thus, paper type with higher grade has higher resistance and stability of printing [30]. Other factors that contribute towards degree of fading is paper reprocessing techniques, binder, dye precursors, color formers, paper fillers and protective coatings including sensitivity of particular type of paper towards environmental conditions [31].

Presence of substituents on the xanthene ring structure in the fluorane can lead to various colors in the reaction such as vermilion, red, yellow and black [32]. This statement explains for the formation of different colored fingerprints upon reaction with iodine on thermal papers. As most of the thermal paper technologies available in the market are patented, therefore, exact composition, i.e. type of leuco dye present is not known. But, according to the color produced few generalizations have been made as per literature [33]:

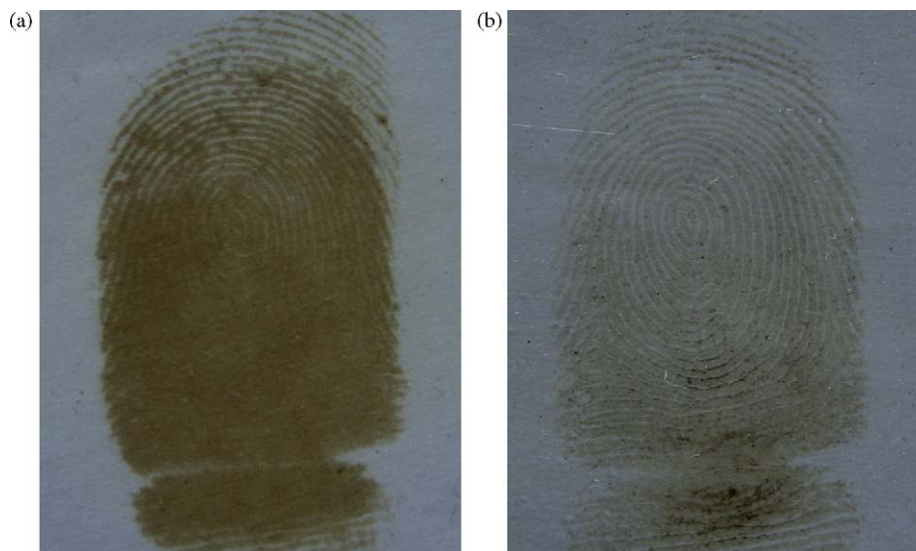


Fig. 4. One-week-old (a) sebaceous and (b) eccrine latent fingerprint developed on thermal paper surface showing difference in intensity.

1. *Green color* is produced by presence of two tertiary amino groups at 2' or 6' positions. Presence of secondary amino groups at 2'-position develops dark or greenish black color, e.g. 2'-n-butylamino-6'-diethylaminofluoran ($R = n-C_4H_9$).
2. *Black color* is produced by methyl group at 3'-position (e.g. 2'-anilino-6'-diethylamino-3'-methylfluoran) which causes steric hindrance to the anilino group that result in twisting of anilino group from xanthene plane. This causes electron transfer hindrance from anilino group to xanthene group resulting in absorbance shift to 570 nm (violet color). An absorption maxima of 470 nm (yellow) is also not affected by presence of methyl group. Complimentary colors, yellow and violet show additive effect to produce black color.
3. *Brown color* is produced by replacement of amino group at 2'-position with azomethine group, e.g. 6'-diethylamino-2'-ethylidenaminofluoran.

Difference between sebaceous and eccrine laden fingerprints was found to be less pronounced in case of fresh fingerprints. Intensity of the freshly developed sebaceous and eccrine fingerprints

was comparable while after few hours decrease in intensity of eccrine marks was observed. For further support of this observation, sebaceous and eccrine marks were deposited and kept for a period of 7 days. On development under the same conditions, the sebaceous marks were found to be distinctively intense than that of eccrine ones (Fig. 4). Generally, latent fingerprint residue when deposited on the porous surface like paper tends to get absorbed quickly. Rate of absorption of water soluble deposits is more than non-water soluble deposits. Sebaceous components remain on the surface for a short period of time in case of porous surface but after some time, fat dries in case of poor donors and in case of good donors, it diffuses and saturates the print area (if the quantity of fatty material is considerable) [34].

In case of aged fingerprints, development of clear and permanent ridge details could be achieved upto 1 year. Aged fingerprints were observed to be less intense than that of the fresh fingerprints but the ridges were identifiable and permanent in nature (Fig. 5). In cases where the sample was overexposed to iodine vapors, white colored ridges against dark background was observed. Water insoluble components in the latent print residue

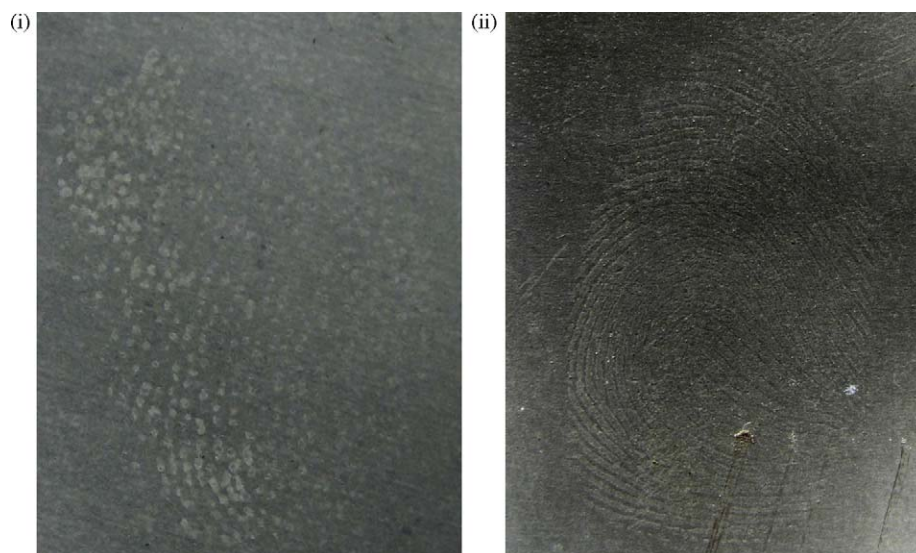


Fig. 5. Ridge development seen in case of upto year old fingerprints on (i) Oddy brand and (ii) Mitsubishi brand thermal paper by iodine fuming.

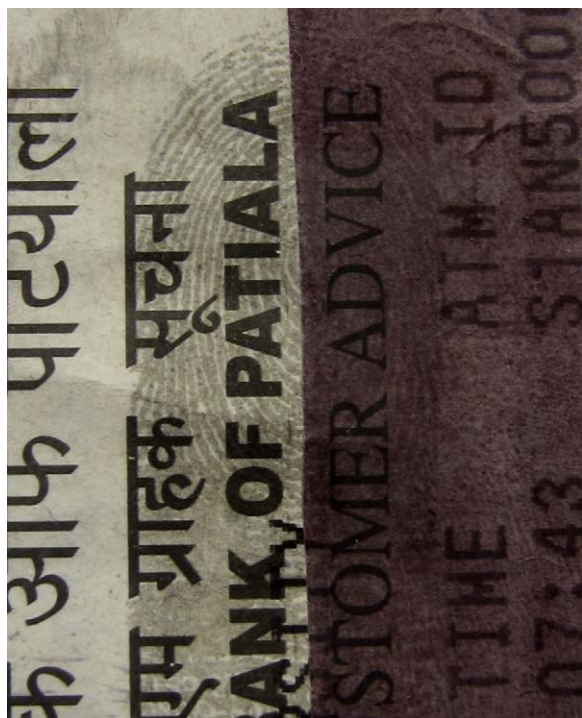


Fig. 6. Latent fingerprint developed on thermal (ATM) paper by iodine fuming (left) followed by DFO (right) in sequence.

have been reported to have two types of fractions, i.e. labile and robust. Iodine is likely to be more sensitive to the “robust fraction” (very less change in the characteristics of the fraction over a period of time) of the lipid moiety of the fingerprint residue [35]. This could be the main factor in development of aged fingerprints while the reduced intensity is due to the drying or diffusion of the sebaceous material. In comparison with the referenced works, DMAC has been reported to produce 2 weeks old fingerprint on the thermal paper [7] while 24 h-old fingerprints were developed by NPB/DFO/IND [15]. All the above-mentioned techniques plus iodine fuming have different sensitivities towards different components of latent fingerprint residue, which could lead to variation in results.

Iodine fuming technique is placed at the top of the sequential processing scheme in the development of latent fingerprints due to the non-permanency of results. Therefore, it became pertinent to observe its effect on other techniques which could be applied afterwards in the sequential processing. In the present work, as we could achieve the permanency of the developed marks, practically it was not required to proceed further with other methods but just for academic interest, we applied DFO and indanedione methods to see whether they affect the substrate or developed marks in any way. The fingerprints developed with iodine were of permanent nature and further treatment with DFO and indanedione could not affect the developed marks but resulted in blackening of the paper surface (Fig. 6) [15]. Further, if permanent and clear fingerprints with no background coloration was possible with iodine, there was no need to treat it further with any other technique. Detailed studies with respect to different sequential processing schemes in case of thermal paper are a subject matter of further studies in future.

Fresh fingerprints were also developed on 1-year-old thermal paper samples. Paper with and without printing was kept in ambient conditions for 1 year. This was observed to see if there was any change in the dye characteristics of the paper within this time period that might ultimately affect the developed print quality.



Fig. 7. Fresh latent fingerprint developed on 1-year-old thermal paper sample by iodine fuming showing clear ridge details.

Thermal paper samples did not show any apparent change in the surface or color during the 1-year storage period and the developed fingerprints were as permanent as developed on the fresh thermal paper samples (Fig. 7).

Further, treatment and developing time in case of iodine fuming of thermal paper samples was considerably far less and the simplest one as compared to other conventional methods like NPB (ninhydrin petroleum ether)/INON/Indanedione. In case of all the later techniques, procedures consist of more than one step, may need pre- or post-treatment, may take more time as well as spoiling the paper due to undesired conditions for example, removal of blackening of the surface by another solution adds one more step to the whole technique [15]; petroleum ether-based formulations are likely to cause damage to the thermal paper samples and heptane tends to fade the developed image [12]. No such problem was observed in case of iodine fuming for thermal paper, in the present study.

4. Conclusions

Iodine fuming method has been recommended as viable methods to develop fresh as well as aged latent fingerprints on thermal paper with no background coloration. The fingerprints developed by iodine fuming were not permanent in case of normal porous surface as paper but they were found to be permanent in case of thermal paper. Oxidation of leuco dyes present in the thermal paper surface by the iodine has been found to be possible reason for permanency of iodine developed fingerprints. Treatment with iodine fumes developed clear and legible fingerprints with third level details visible. Various colors of developed fingerprints were observed on different types of thermal papers which could be attributed to different substituents present on the fluorane leuco dye structure. No difference could be found in fresh eccrine and sebaceous marks, but in case of aged marks, marked difference was observed in both with respect to intensity and permanency of fingerprints.

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