CHAPTER-3

MATERIALS AND METHODS
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3.1 Determination of nature and source of photocopies

First two objectives were collectively achieved by the analyses of photocopies. The examination of photocopied documents for determination of their nature and source included physical and chemical analyses of the photocopies. The physical analysis included examination of toner type, toner fusion, splattering effect of toner and defect marks. The chemical examination referred to analyses of toners (both raw and processed) with analytical techniques.

3.1.1 Collection of samples

3.1.1.1. Collection of black photocopier samples

The survey for the usage of toners in photocopiers concluded that the refilled industry had taken over the market. Refilled toners of different brands were used in the photocopiers irrespective of their make and model. Keeping this thing in mind, collection of samples was designed. Photocopied documents were collected three times from sixty two photocopiers of twenty five different makes and models. Raw toners were also collected from the respective photocopiers. Raw toners of twenty eight brand names were found used in sixty two photocopiers (Table-1). Photocopies as well as their raw toners were collected from same photocopiers after a gap of one month to check the consistency in the results.

3.1.1.2. Collection of coloured photocopiers samples

Samples of the coloured processed toners were collected; two pages were designed for collecting the samples, one containing bars of different colours (Fig. 1) and other containing text matter in different colours (Fig. 2). Then both pages were printed from nine colour laserjet printers and photocopied from nineteen coloured photocopiers. After the gap of one month, both the pages were again collected from the same coloured laserjet
printers and photocopiers to detect the variation due to time. In this way, one hundred and twelve sheets of processed toners were collected from twenty eight colour photocopiers and laser printers (Table-2). Ten raw toners containing all the four colours; yellow, cyan, magenta and black were collected. These samples include the toners from original manufacturers as well as refilled toner (Table-3).

3.1.2. Analysis of samples

Black and colour (raw as well as processed) toner samples were analysed for its physical and chemical characteristics to determine their nature and source.

3.1.2.1. Physical examination of black photocopies

The physical characteristics such as toner type, toner fusion, splattering effect of the toner and trash marks were produced on the photocopy during its transit through the photocopier machine. The defect marks develop over a period of time through use and abuse of photocopier which appeared on the photocopy in form of trash marks. These physical characteristics, if utilized properly could limit and established possible source of photocopier (Totty and Baxendale 1981, Holland 1984, Gilmour 1994, Buglio 1999). Black photocopied samples were classified on the basis of these characteristics. The samples were examined under Olympus SZX7 Stereomicroscope at magnification 4x40X for the presence of various physical characteristics and trash marks. The photographs were taken with 8 megapixel Camedia-8080 Olympus digital camera and images were then transferred to the computer.

3.1.2.1.1. Toner type

The photocopiers mainly used two types of toners, dry and wet toners. Dry toners were characterized by the glossy appearance of the toner particles on the surface of paper. Liquid toners dyed individual paper fibres and appeared as thin coating through which
paper fibres could be seen (James, 1987; Totty 1990a). The black photocopies were examined and classified on the basis of the presence of these characteristics (Table-4).

3.1.2.1.2. Fusion methods

Final stage of the copying process included fusion of the toner to paper surface to form a permanent copy. Different photocopiers used different toner fusion methods such as radiant heat, heat and pressure, cold pressure, blotter or air dry. These methods had marked effect on the final appearance of the toner on the paper surface and utilized to classify the photocopied samples. Surface morphology of black processed samples was examined for the different fusion methods (Table-4).

3.1.2.1.3. Splattering effect of toner

During the application of toners on the latent image present on the drum, toner particles showed splattering effect on the edges of the strokes. Splattering effect of toner could be used to differentiate photocopied documents from laser printed documents (Gilmour, 1994). Direction of splattering effect of toner had been observed to have good evidential value and therefore used for the classification of the photocopiers in the present study. Thus, black processed samples were examined and classified on the basis of direction of splattering effect of the toner (Table-4).

3.1.2.1.4. Trash marks

The scratches on the glass platen, photosensitive drum, fusing roller and impressions of residual toner appeared on the photocopies in the form of trash marks. The trash marks due to glass platen, photosensitive drum are individualistic and could be used for identification of photocopied documents (Totty and Baxendale, 1980; Totty, 1990b). However, the marks from fusing roller impression of residual toner could be used to classify the
photocopies (Gerhart, 1992; Buglio, 1999; Laporte 2004). These trash marks have been generated consistently on the copies, until the defective component of the machine is removed or replaced. Surface of black photocopies were examined and classified for all types of trash marks.

3.1.2.1.4.1. **Glass platen defect marks**

Scratch marks on the glass platen appeared in the form of trash marks on the surface of photocopy. These types of marks would always show up in same place if the document is photocopied in 1:1 ratio until the glass platen had replaced (Totty and Baxendale, 1980; Levinson, 2001; Totty 1990b). All the photocopies were examined for the same and the results were tabulated (Table-5). These marks when present have been differentiated from each other on the basis of their number, position, shape and size (Table-6).

3.1.2.1.4.2. **Drum defect marks**

Scratch or dent present on the drum appeared on the photocopies in the form of drum defect marks. Due to small diameter of drum, these trash marks repeated on a single copy with the position of marks migrating from one copy to another. The distance between the two drum defect marks could be quite helpful in measuring the circumference of the drum (Totty and Baxendale, 1980; Levinson, 2001; Totty 1990b). The drum defect marks have been differentiated on the basis of different shape, size, location at the page and circumference of drum (Tables-5 and 7). The sequencing of the pages had also been determined from the distance between two drum defect marks present on two consecutive pages.

3.1.2.1.4.3. **Roller defect marks**
The damage of the Teflon coating of the roller due to wear and tear caused by the knob of metal prong, which rested on the roller and appeared on the surface of photocopy in form of vertical line. These marks appeared in the same relative position in same make and model of machine. Though, this type of wear lack individuality but would greatly reduce the number of suspect copiers (Gerhart 1992, Laporte 2004). The distance between the two vertical defect marks helped in establishing the link between roller and the photocopies. The photocopied samples were examined for the same and the results were tabulated (Tables-5 and 8)

3.1.2.1.4.4. Trash marks due to residual toners

The impressions appeared on the copy due to inability of the cleaning pad to remove residual toner from the surface of photosensitive drum. These impressions were significant, identifiable and unique as to number, size, shape and distribution and they would be spaced on the paper at a distance apart which is vertically equal to that of circumference of the fusing roller. It could be used to improve the classification of the photocopiers further (Totty and Baxendale, 1980; Buglio, 1999). In the present study, trash marks due residual toners were examined (Table-9).

3.1.2.2. Physical examination of colour photocopies

Coloured laser printed and photocopied documents were examined for their printing characteristics such as Counterfeit Protection System (CPS) code and ‘Toner deposit pattern’.

3.1.2.2.1. Counterfeit Protection System (CPS) code

Anti-counterfeiting technology has been incorporated in the coloured photocopiers or laser printer to avoid the fraudulent use. The counterfeit protection system code comprised of a pattern of yellow dots and nearly invisible to naked eye. This pattern covered the entire
surface of the document but could be more easily seen in area of the document, where there is no printing (Tweedy, 2001 and Li et al. 2004). This counterfeit protection system code would be quite useful to identify the source of coloured photocopy and printout and could be visualized by VSC and Microscopic methods. In present study, VSC method and microscopic methods were applied to examine the CPS code of the samples.

3.1.2.2.1.1. Video Spectral Comparator (VSC) method

The CPS code in form of yellow dots on the colour copies and printouts is visualized by using Video Spectral Comparator 2000/HR of Foster and Freeman Ltd. The colour copy/printout was placed in the VSC chamber and the image was displayed on the monitor in VSC software. The copies/printouts were illuminated with Ultraviolet Light by pressing ‘Ultraviolet’ option with band pass filter <400mm and long pass filter off. The image was focused by pressing the +ve or –ve sign of focus option of VSC software and by rotating the knob for fine focusing. This dotted pattern was seen on the entire surface of the paper. Then the images were saved into the computer hard drive through VSC software. Similarly, images of CPS code from the surface of printout/copies of all the coloured samples were captured. These images were then interpreted on the basis of the patterns of yellow dots.

3.1.2.2.1.2. Microscopic Method

The technique given by Tweedy (2001) was modified in the following way. The printouts and photocopies were examined under Olympus SZX7 Stereomicroscope (magnification 4x40X). The photographs were taken with Camedia-8080 Olympus (8 megapixel) camera and images were then transferred to the computer. The captured image of the sample was opened in the Adobe Photosshop CS2 software. The cursor was moved on the ‘select’ and the command ‘Colour Range’ was selected and yellow filter was applied. Then the option ‘Level’ was opened from the ‘Adjustment’ of ‘Image’ icon of the window and range was
increased to maximum. Same process was repeated till the yellow dots were visualized. Then, this processed image was saved on the hard drive by selecting option ‘save as’. The images of all samples were processed similarly and the results were interpreted. The results obtained with both methods were recorded in the Table-10

3.1.2.2.2. **Printing characteristics (Toner deposit pattern)**

In colour photocopier and laser printer, four colours that is, yellow, magenta, cyan and black toners were used for printing the picture in layers (Mizrachi et al. 1998). The image is created by using a four colour pass process. Each of the four colours is applied to the drum one at a time in the following order: Y, M, C, and then K (https://www.office.xerox.com/partners/.../05_systemoutline.pdf). The image formed on the drum is then transferred to the transfer belt. The full four colour image formed on the transfer roller, from the transfer belt, is then transferred to the media. In the present study, the coloured laser printed and photocopied documents were examined for this toner deposit pattern with Digital microscope (Olympus MIC-D with magnification of 132K) having an LED (Light emitting diode) light source attached. Images were captured by 5-megapixel inbuilt camera and were directly transferred to the computer. The images of all the samples were then interpreted on the basis of their toner deposit patterns. The results obtained were recorded in the Table-11.

3.1.2.3. **Scanning Electron Microscopy (SEM)**

The surface morphology of the toner deposits on the paper surface was examined under Scanning Electron Microscopy. This technique helped to determine different fusion methods used in the photocopiers (Totty 1990, Brandi et al. 1997).

3.1.2.3.1. **Sample preparation for SEM**

Various colour and black photocopies and laser printouts were examined under Scanning Electron Microscope model JSM-6100 at 800X, 15KV, 10µm, WD-39 to determine the
fusion methods used. A small piece (1mm diameter) of the printed letter was taken from the samples and mounted on stub with double adhesive tape, then it was placed in the gold coating chamber (unit) of-Model Fine Coat Ion Spatter JFC-1100 to make the sampling material conductive. The gold coated samples were removed and subjected to Scanning Electron Microscope to identify fusion method used in the production of photocopies and printouts. The electrographs were taken and results were presented in Tables 12 and 13

3.1.2.4. Chemical examination

Numbers of possible source machines are quite large, therefore any additional classification method, that is, examination of chemical properties of the toners by instrumental methods has further improved the class characteristics.

3.1.2.4.1. Sample preparation of black photocopier samples

The processed toners were removed thermally from the documents by modifying the technique given by Munson, 1989 (Saini and Saroa, 2008). To accomplish this, thin copper foil was placed over the sampling area. Heat was supplied by a soldering iron for 2 to 5 minutes. The heat transferred some of the toner from the document to the copper foil. Three lifts were sufficient for dark photocopies, while six lifts were required for light photocopies. The toner was then washed from the foil with chloroform into a sampling vial. A very small amount of raw toner was dissolved in chloroform into a small sampling vial, and the dyes from the raw toners were extracted.

3.1.2.4.2. Sample preparation of colour photocopier samples

Four colour bars (bars no. 2, 3, 5 and 8 of yellow, cyan, magenta and black colour, respectively) of all the samples were selected because they had maximum amount of a particular colour (along with other colours) of toner particles present in them. The processed colour toners were removed thermally from the documents and colour toners of all the four colour bars were separately extracted with chloroform. In another sampling
vial, the extracts from all the four bars were mixed together also. This was done because in colour photocopier/laser printer, the four colours like yellow, magenta, cyan and black were used for printing the picture in layers, hence 4-colours of toner analyzed and considered as a mixture (Mizrachi et al., 1998). So, monochrome toner particles were prepared from full colour copiers (Seipp, 1997). Similarly, small amount of toners from all of four colours of raw toners (from the cartridges) were taken and extracted with chloroform separately as well as they were mixed together also (as stated above) to extract dye components from it. These extracts were used to analyze all toner samples by TLC, UV and FTIR techniques. These techniques have been used to classify the photocopies and their copiers, based on the mixture of all the colours.

3.1.2.4.3. Thin Layer Chromatography of black photocopier samples

3.1.2.4.3.1. Solvent-stationary phase

Sixty two black toners (both processed and raw) were analyzed with Thin Layer Chromatography. Four solvent systems (Table-14) were used for separation of dyes in the toners. The solvent systems were kept in chamber for saturation for 1 hour at room temperature (35°C). A precoated silica gel 60F (Merck) plate without fluorescence was used as the stationary phase.

3.1.2.4.3.2. Spotting of extracted samples and visualization of spots

The extracts of the samples of raw and processed toners were spotted on precoated silica gel 60F (Merck) TLC plates with the help of fine capillary tubes. Spotted TLC plates were placed in preheated oven at 95°C for 3 minutes to remove the extraction solvent (Pagano et al. 2000), then cooled to room temperature. The plates were placed in a saturated solvent chamber until the toner migrated 5 cm from its origin.

Each developed TLC plate was dried at room temperature and observed in the day light and was then exposed to ultraviolet light (long and short wave). The colour and
fluorescence of the developed spots were recorded. The boundaries of the spots seen under ultraviolet light and day light were marked and the hRf value of each spot was calculated and the results were recorded (Table-15). The experiments were repeated three times to verify the consistency in the results.

3.1.2.4.4. Thin Layer Chromatography of coloured photocopier samples

3.1.2.4.4.1. Solvent-stationary phase

The extracted samples of processed and raw colour toners were analysed with Thin Layer Chromatography. Sixteen solvent systems (Table-16) were tried to separate the dye components of raw as well as processed colour toners. The solvent systems were kept in chamber for saturation for 1 hour at room temperature (35°C). A precoated silica gel 60F (Merck) plate without fluorescence was used as stationary phase.

3.1.2.4.4.2. Spotting of extracted samples and visualization of spots

The extracts of four colours toners (raw as well as processed) along with mixture of these four colour toners of all the samples were spotted manually on precoated TLC plates and the procedure was followed as in case of black toners. The hRf value of each spot had been measured (Tables-17 and 18). Whole experiments were repeated for three times to verify the consistency in results.

3.1.2.4.5. UV Spectroscopy of black and coloured photocopier samples

Sixty two black toners (both processed and raw) were analysed with UV spectrophotometer model Perkin Elmer Lamda 45. The spectrum of solvent (chloroform) was taken, in which toners were dissolved, to set value to zero. The extracts of toners (both raw as well as processed) of all the samples were firstly filtered by using Whatman filter paper, then filtered solution was subjected to UV spectrophotometer and spectrum of the solution was recorded in UV-range (200nm to 300nm) at absorption mode-range -3 to +8
A. In this way, spectra of both processed and raw black toners of all the samples were recorded (Table-19). Similarly, analysis of twenty eight processed and ten raw colour toners was done with UV spectrophotometer and spectra of both processed and raw colour toners of all the samples were recorded (Tables-20 and 21).

3.1.2.4.6. Fourier Transform Infrared Spectroscopy of black and coloured photocopiers samples

Analysis of sixty two samples of black toners (both raw and processed) was done with FTIR spectrophotometer model Perkin Elmer- Spectrum RXI FTIR System. Measurements were performed in transmission mode. Spectra obtained at 2.0 cm\(^{-1}\) resolution with 4 scans in range of 450-4000 cm\(^{-1}\). The extracts of black processed toners of all the samples were poured on to the KBr transparent pallets and kept for drying. In case of raw toners, toners of all the samples were mixed with KBr powder and the pallets were prepared. Then these pallets were subjected to FTIR analysis. FTIR spectra were obtained and the results were recorded (Table-22). Similarly, analysis of twenty eight processed and ten raw colour toners was done with FTIR spectrophotometer model and FTIR spectra were recorded and results were tabulated (Tables-23 and 24).

3.2. Determination of authenticity of the photocopied documents

Authenticity of a photocopied document can be questioned regarding any alteration made to it such as transplantation, deletion or additions present. For this, collection of samples and their examination was carried out for the identification of alteration in typed and printed materials, handwritten material and signatures of photocopied documents.

3.2.1. Collection of samples

3.2.1.1. Determination of alteration
Determination of the authenticity of the photocopied documents was carried out to achieve the third objective. Black and coloured photocopies of the cheques, receipts, detail-marks certificate and letter pads were prepared. Alterations such as addition, deletion and transplantation were designed in the typed and printed portion, handwriting portion and signature portion of 50 black and 20 colour photocopies (Tables-25 and 26). Deletions were done by using erasure and correction fluid. Then additions were made at those places. These altered 50 black documents were then photocopied from two black photocopiers (one of poor quality and other of good quality) up to 3rd generation. Similarly, colour photocopies of twenty coloured altered documents were also prepared from one colour photocopier up to 3rd generation.

3.2.1.2. Determination of generation

Recopying of photostat may hide or eliminate most of the evidences of manipulation or alteration of an original document. So, an attempt has been done for the determination of the generation of photocopy. For this, a text page was designed in Microsoft word and got printed from printer, and then the printout was photocopied from a photocopier, which constituted the 1st generation copy. Then a copy of this (1st generation photocopy) was obtained from the same photocopier (2nd generation photocopy) and this photocopy was recopied again from the same photocopier, thereby forming the 3rd generation photocopy of the document. In the same way, the photocopies upto 3rd generations were obtained from 102 photocopiers (Table-27).

3.2.2. Analysis of samples

Analysis of black and coloured photocopies was carried to determine the authenticity by detecting for the presence of the alteration in photocopies. They were also examined for the determination of their generation.

3.2.2.1. Determination of the altered photocopied documents
Authenticity of the collected 300 black and 60 coloured photocopied samples was examined in the typed, printed, handwritten portion and signature portion of documents. Following characteristics were examined under Olympus SZX7 Stereomicroscope (4x40X) to indicate the manipulation in the document and results were presented in Tables-28 and 29.

i. Difference in quality of strokes.

ii. Difference in brightness of the strokes.

iii. Extra deposition of toner on the background.

iv. Discontinuity of dots of printed matter on the background

v. Difference in slant, size and design of the letters

vi. Difference in spacing between the letters as well as lines

vii. Difference in alignment of the matter

3.2.2.2. Determination of generation of the photocopied documents

Expansion of strokes and baseline printing due to disappearance of sharp angles, appearance of discontinuity in strokes, loss of smoothness of edges of the strokes and duplication of defect marks could be seen in photocopies in comparison to the original documents (Hilton, 1979). So, black photocopies were examined, upto 3rd generation for duplication of defect marks and for expansion of strokes and baseline printing for the determination of generation.

3.2.2.2.1. Duplication of defect marks

The pattern of flaws (defect marks) are duplicated with each copying positioning in close proximity to each other, which indicated that copy is the result of several recopying from the same machine (Hilton, 1979). Therefore, all samples were examined from 1st generation to 3rd generation for presence and nature of the duplication of flaw marks on the
photocopies. Examination was also done to notice nature of different generations of flaw marks present upto 3rd generation copy.

3.2.2.2.2. **Extent of expansion of strokes and baseline printing**

The degree and direction of expansion of strokes of the letter could be used as a tool to ascertain the generation of photocopies from the original document. Image processing is one of the useful techniques in the examination of questioned documents. Present study has been made to utilize image processing technique that is ‘Adobe Photoshop CS2’ in the detection of recopying or generation of photocopied documents. An attempt has also been made to classify different photocopier based on the features present in the work of a particular photocopier. The images were processed in following way:

All the samples (1st generation to 3rd generation) of 102 photocopiers along with the original document were scanned by using a digital coloured HP laser-jet 3030 scanner. The scanned images were then processed by software ‘Adobe Photoshop CS2’ by performing the following steps:

Step-1: The scanned images of 1st and 3rd generation of a sample were opened into Photoshop CS2 page (Fig. 3).

Step-2: A new blank file of size 21cmX16cm (A4) was opened in Photoshop CS2 page (Fig.4).

Step-3: The scanned Images of 1st and 3rd generation were dragged into it by clicking ‘Moving File Option’ of the Photoshop so that the image of 1st generation was placed over the 3rd generation image (Fig. 5).

Step-4: Then, ‘Layer Option’ was opened by clicking ‘Option of Window’ or by pressing F9. ‘Opacity option’ was varied to see which part of 3rd generation character was expanding beyond the 1st generation character (Fig. 6).
Step-5:-Then, the overlapped image was saved by ‘Merge Visible’ or ‘Flatten Image’ or pressing keys (Shift+ Cr +E) simultaneously (Fig. 7).

Similar procedure was applied to the scanned images of original document and 1st generation, 1st and 2nd generation copies and 2nd and 3rd generation copies of samples. Similarly the scanned images of all photocopies (from 1st generation to 3rd generation) from 102 photocopiers were processed using ‘Adobe Photoshop CS2’ to determine the generation of photocopies from the extent and direction of expansion of strokes of the letter and baseline of printing. The extent (percentage) of expansion or enlargement of strokes of the letters from original document upto its third generation were studied (Table-30). The photocopiers were classified from direction of expansion of stroke of the letter and baseline of printing and the results were tabulated in Table-31.