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REMOVAL OF LASER PRINTER TONER FROM THE PAPER SURFACE USING SOME CHEMICAL METHODS

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Abstract

In this article, organic solvents are applied to a range of toner- paper combinations to determine their ability to remove toner. Paper sample printed from HP Laserjet 2300dn is soaked in 300 mlof 17 alone solvent and 133 solvent mixtures for 30 minat room temperature.Visual effect of solvent on toner and paper can be classified into 6 groups A, B, C, D, E and F.The effect of varying the solvents proportions in the mixture is explored. Solvents give good dissolving results for paper sample printed from HP Laserjet 2300dn applied on paper samples printed from different brands and same brands of different models.The evaluation of removal efficiency and re-usability of paper and its application in document forgeryare performed by optical tests on paper surfaces. Also the effect of solvents on mechanical, physical and optical properties of paper is studied. Removal of toner from security paper and its using in creating forged documents is applied in this study. Optical tests and SEM examination indicate that it is possible to remove toner from paper without damaging and discolouring the substrate.

Keywords: Toner removal, Paper surface, Laser printer, Optical tests

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1. Introduction

Numerous types of documents can be produced and/or personalized using several printing methods such as offset, dye sublimation, thermal printing, inkjet, electro photography, among others. In line with the advancement of information, the demand for printers has shown enormous increase[1].The technological development of the printing devices is significant, and high quality printers can easily be found on the market at a reasonable price [2]. Laser printers are being extensively used in offices, educational institutions and commercial establishments [3,4]. Use of laser printers has increased dramatically over the last 20 years because of the speed, simplicity, and accessibility of photo-printing and inexpensive[5].

Toner is dry ink that creates the image on paper during the printing process used in most copiers and some large printers [6,7,8]. Formulations of toners are different from that of conventional ink [9]. Common black dry toners are constituted by opaque solid material imbedded in a matrix of organic binder which bonds to the

surface of the paper by fusion. Typical binders include variations of styrene, methacrylate polymers and epoxy resins, sometimes cured with other organic components. As colored pigments, in black toners carbon black or iron oxide are employed, while organic pigments are employed in other color toners. Other than these principal components, toners comprehend also many adjutants as waxes (e.g. polypropylene), used as release agents, and silicic acid (amorphous silicon dioxide), used as anti-binder.The options for how to remove toner from paper surface were: doing work (as abrasive), introducing heat (as laser) or changing the local electron structure (by, for instance, adding a solvent).A major disadvantage of laser printers is that they are now more accessible for illegal activities by removing toner from paper surface (removing original text) and using removed paper in fraud, false documents, anonymous letters, confidential materials, and acts of terrorism [10].

This study representsthe removing of toner from ordinary and security papers surfaces by some chemical methods as using solvents without any side effects on paper to re-printing new text on removed paper.

2. Experimental

2.1. Specimen preparation [11]

Paper samples printed from different brands and same brands of different models (listed in Table 1) were collected after 1 hr the toner was dry to the touch. With respect to sample 1,significant volume (300 ml) of 17 standard laboratory alone organic solvents(name of solvents listed in Table 2) and mixture of each alone solvent with the others by proportion 1:1 put in a big plate (its size increase about size of A4 paper to kept paper un-rolled) and printed paper sample soaked in it for 30 min at room temperature and plate covered to prevent solvent evaporation. Solvent mixtures prepared by adding 150 ml of one solvent and complete to total volume (300 ml) by another solvent. After soaking the paper sample go away from solvent and sweep by cotton to remove any toner particle suspended on paper and then left to air dry at room temperature. Documented security paper printed by laser printer involving hand signatures and stamp also removed. The effect of removal on security features of paper, signatures and stamp, re-usability and its using in creating forged documents is studied.

Table 1. Specifications of the used samples

Brand	Model	Nomenclature
Hp LaserJet	2300dn (compatible toner)	1
Hp LaserJet	P1005 (compatible toner)	2
Hp LaserJet	P1005 (original toner)	3
Hpcolour LaserJet	4700 (Original toner)	4
LEXMARK	E350d (original toner)	5
EPSON aculaser	M 2000 (original toner)	6

Table 2. Standard laboratory organic solvents

No	Solvent
1	Tiner
2	Ethanol
3	Isopropanol
4	Methanol
5	Chloroform
6	Carbon tetrachloride
7	Toluene
8	n-Hexan
9	Benzene
10	m-Xylene
11	2-Butanol
12	Diethyl-ether
13	Ethyl-acetate
14	THF (Tetrahydrofurane)
15	Dichloromethane
16	Acetone
17	1-Butanol

2.2. Optical Tests

Optical tests are appliedon removed paper to investigate effect of solvent on toner and paper.

2.2.1. Image scanned test[9]

All removed paper samples is taken by scanning using a 1200 dpi (dots per inch) scanner (CanoScanLiDE 100,Canon Co.) to evaluate the removal efficiency expressed by the residual ink speck population on a given sample paper. Also, the re-printed paper sample with a new text scanned to make qualitatively compared to freshly printed paper of the original and new texts.

2.2.2. Ultraviolet light test[12]

Ultraviolet (U.V) light at wavelength 365 nm in a VSC 6000 (Foster and Freeman, UK) using to examine if solvent mixtures have more dissolving toner effect exhibit any fluorescent effect on paper samples under U.V light or not.

2.2.3. Transmitted light test

Transmitted light in a DOCUCENTER 4500 (Projectina, Switzerland) using to determine if any solvent possess any damage effect on coating layer or cellulose fiber of paper.

2.2.4. Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM) of paper samples is performed by a JSM-5140 (JEOL, USA) using an accelerating voltage of 30 KV, magnification of 150 x.

Small squares of print, 2 mm×2 mm, are cut from one letter with a "thick" application of toner. The toner sample is removed from the paper by making vertical cuts on each side of the sample, then inserting the blade under the toner in a plane parallel to the paper, removing a minimum of paper fibers. The instrument design employed a 50 µm working distance to provide optimal sample/detector geometry. The scan is restored over the surface of an area of toner removed.

2.3. Paper Properties

2.3.1. Mechanical properties (tensile, elongation, bursting andtearing tests) [13]

In all mechanical properties (burstingstrength test , tearing resistance test (longitudinal (L) direction, depth (D) direction) , tensile strength test (L-direction, D-direction), elongation test (L-direction, D-direction) 5500 R Universal Testing Machine (Instron, USA) is used. This type of machine has a self-calibration, zero adjusting and automatic balance, which are done daily before testing or during testing. This testing instrument is accompanied by a highly reliable system for evaluating the mechanical properties. Measuring drum of sensitivity ± 0.01 mm is used for dimensions evaluation. The tests specimens are conditioned at 23°C with a humidity of 60 %. For bursting, tensile and elongation each sample is measured five times and three times for tearing.

2.3.2. Physical properties (thickness measurements)[14]

Thickness of paper samples are measured using a Gauge model No. 11/2704. Five thickness measurements were taken on each sample.

2.3.3. Optical properties (brightness,whiteness andgloss measurement)[9]

Brightness and whiteness of paper samples are measured using a hunterlab color /difference meter ASTMD-2244. Gloss of paper samples is carried out according to multi angle gloss meter model gm-2000. Five gloss measurements are taken on each sample at 20°C, 60°C and 85°C.

3. Results and Discussion

3.1. Image Scanned Test[15]

After removal process, all paper samples scanned by cannon scanner and according to visual effect of solvent on toner and paper the results can be classified into 6 groups A, B, C, D, E and F. Group Aincludes 5 alone solvents and 17solvent mixtures.Solvents of this group don't exhibit anyvisual effecton toner or paper.

Group Bincludes10 alone solvents and 43 solvent mixtures. A solvent of this group involvessome blurring of printed areas but no significant removal withno effect on paper.

Group Cincludes 1 alone solvent and 60 solvents mixtures. Solvent of this group lead to the degree of blackcolour of toner is decreasebut the printed text is still clear and legible.

Solvent of groups A, B and C don’t give good resultsdue to solubility parameters for both alone and solventmixturesare far away about the solubility parameters of polymer resin present in toner.

Group Dincludes 1 alone solvent and 5 solvent mixtures. Solvents of this group possess damage effect on coating layer and cellulose fiber of paper surface under and around the removed area. The damage effect due to solubility parameters for both alone and solventmixtures close form solubility parameters of cellulose fiber of paper causing removal coating layer and dissolve cellulose fiber.

Group E includes 5 solvent mixtures. Solvents of this group remove significant quantities of toner and the printed text almost disappeared. This may be due to solubility parameter of solvent mixtures is near from solubility parameters of polymer resin present in toner. Also, change in proportion for some solvent mixtures present in groups C, D and E doesn't improve above results of toner removal.

Group Fincludes3 solvent mixtures. Solvents of this group more effective dissolving and completely toner removal therefore, the printed text disappeared. Complete miscibility is to be expected between the polymer and solvent components if their solubility parameters are very close to or even identical to each other. Some alone and solvent mixtures present in different groups listed in Table 3for example.

Table 3.Represents solvents; alone and as a mixtures of each group for example

Groups	alone	mixture
A	methanol and isopropanol	n-butanol + isopropanol and n-butanol + ethanol
B	ethyl acetate and diethyl ether	toluene + benzene and m-xylene + acetone
C	THF	chloroform + ethyl acetate and 2-butanol + acetone
D	tiner	n-butanol+ CCl_4 and n-butanol + acetone
E		ethyl acetate + ethanol and 2-butanol + THF
F		methanol + benzene (2:1), methanol + dichloromethane (2:1) and methanol + ethyl acetate

Visual effect of groupsA, B, C, D, E and Fon toner and paper shows in Figure1.

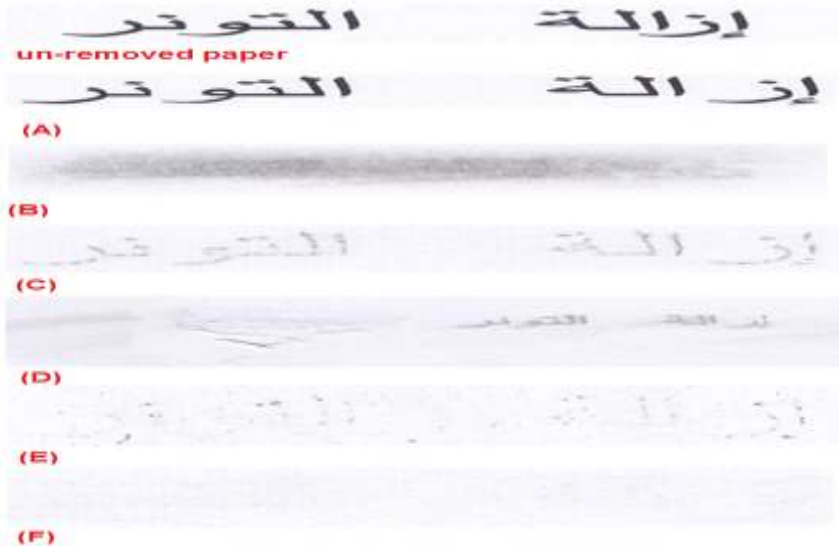

Figure 1.Visual effect of solvents in groups A,B,C,D,E and F on toner and paper

More effective dissolving mixtures on sample 1don't possess the same dissolving effect when applied on paper samples from 2 to 6 (listed in Table1) due to polymer resin present in toner of sample 1 differs about polymer resin present in other tonersamples or may be polymer resin in all samples the same but other additives present in toner decrease its dissolving. Figure 2 shows the effect of one mixture (methanol + ethyl acetate)from three more effective dissolving mixtures on samples from 2 to 6.

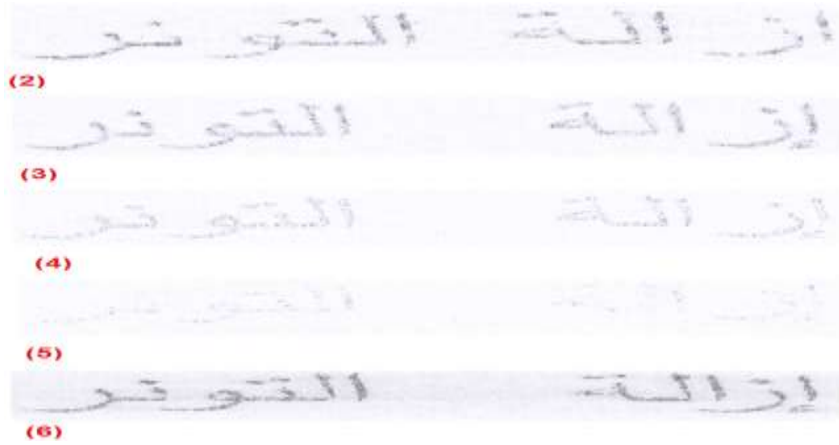

Figure 2. Effect of methanol + ethyl acetate (1:1) on paper samples no.2, 3, 4, 5 and 6

3.2. Ultraviolet light Test[12]

After removal process, paper samples removed by acetone + dichloromethane (1:1) (group B) and methanol + dichloromethane(2:1) (group F)beside the un-removed paper when placed under effect wavelength of U.V light at 365nm no fluorescence is observed in case of solvent mixture of group F while in case solvent mixture of group B removed area show darker than un-removed area. This may be due to in case of solvent mixture of group F when paper sample remain to dry at room temperature the solvent is evaporated and its effect don't appear under U.V light while in case solvent mixture of group B remain. Effect of U.V light on un-removed paper and paper samples results from solvent mixture of group Fshows in Figure3.

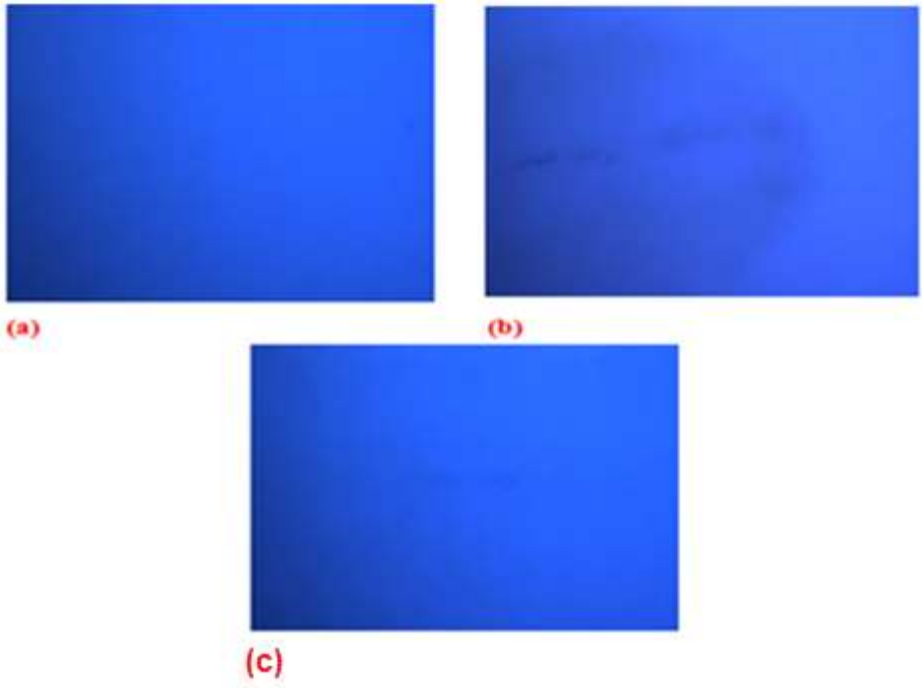

Figure 3. Effect of ultraviolet light sources on (a) un-removed, (b) removed paper by acetone + dichloromethane (1:1) and (c) removed paper by methanol + dichloromethane (2:1)

3.3. Transmitted light Test [16]

After removal process, paper samples results from solvent mixtures of group F (more effective dissolving mixture) when placed under effect of transmitted light sources no damage effect is observed on coating layer or cellulose fiber of paper sample when compared with un-removed paper. This due to solubility parameters

for solvent mixture is far away about the solubility parameters of cellulose fiber of paper sample. Effect of transmitted light sources on un-removed and removed papers results from one solvent mixture in group D and two solvent mixtures of group F shows in Figure 4.

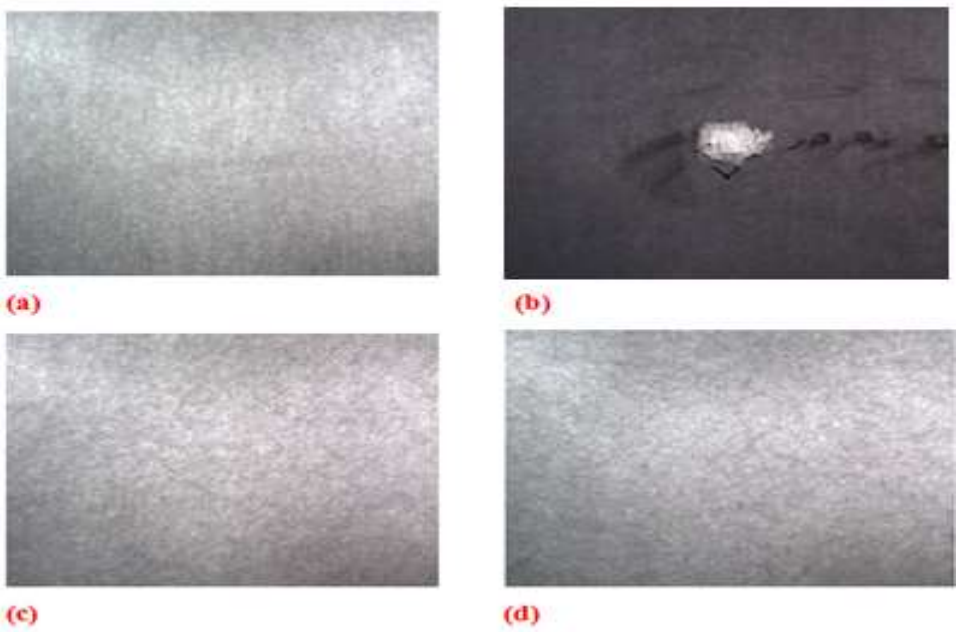

Figure 4. Effect of transmitted light sources on (a) un-removed, (b) removed paper by ethyl acetate+diethyl ether (1:1), (c) removed paper by methanol + ethyl acetate (1:1) and (d) removed paper by methanol + dichloromethane (2:1)

3.4. Scanning Electron Microscopy (SEM)[17]

SEM images taken from the un-removed paper sample for comparison with taken from paper samples removed by solvent mixture of group F can be seen that, the removal process doesn't seem to produce any visible effect on white paper of removed sample. Also, there is no clear difference between the appearances of white paper resulting from removal of toner when compared with un-removed paper. Cellulose fibers in both removed and un removed areas seem to possess bonding and be kept densely packed, indicating that solvent mixture don't created damage in the paper zones that were covered by toner-print.

This may be due to in laser printer toner particle fixed on surface of paper and don't penetrate to cellulose fiber, and solvent mixture evaporated when paper sample drying after removal process, therefore removal process take place without any effect on paper. SEM images taken from un-removed and removed papers samplesare shown inFigure5.

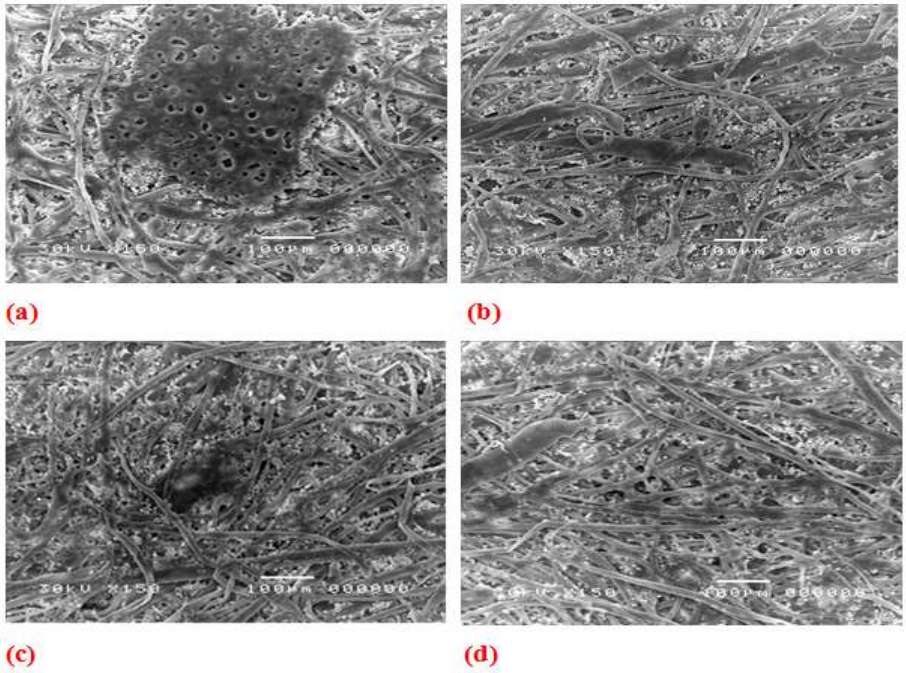

Figure 5. Scanning electron microscopy images for (a) un-removed paper, (b) removed paper by methanol + benzene (2:1), (c) removed paper by methanol+ ethyl acetate (1:1) and (d) removed paper by methanol+ dichloromethane (2:1)

3.5. Mechanical Properties

3.5.1. Tensile test

In order to display the experimental data obtained in a comprehensible form, we evaluated the mechanical characteristics caused by the solvent mixture procedure on two samples. Values of tensile strength reflect the detailed structure of the paper and the properties of its individual fibers, i.e., the dimension and strength of fibers, their arrangements, and inter fiber bonding. The results obtained for tensile strength of the paper after soaking in solvent mixture are presented in Table4.

Table 4. The mean values of tensile test

Sample	Tensile test			
	L-Direction		D-Direction	
	kg/cm	N/cm	kg/cm	N/cm
Un-removed	5.203	51.021	1.869	18.329
Methanol + benzene	4.653	45.634	1.668	16.353
methanol + ethyl acetate	4.599	45.098	1.776	17.414

The results show no significant difference among the obtained values of un-removed and removed paper after using two solvent mixtures under the same experimental conditions. These results confirmed the weak impact of the solvent mixture on tensile strength. The application of solvent mixture caused a little decrease of tensile strength of paper. It should be noted here that paper removed samples demonstrated good stability, since after using solvent mixture decrease in tensile strength is very close and lower than almost 1 with respect to un-removed paper. The values of tensile strength monitored upon solvent mixture procedures are related to the extent of oxidative and hydrolytic damage to cellulosic fibers. The decrease in tensile strength of paper samples is mainly due to swelling of cellulose fiber by solvent penetration during removing and may be partially due to the fact that coating materials are impregnated into the cellulose structure of paper and interfere with fiber to fiber interaction. Also such interference by the coating materials causes a reduction of interaction force between the fibers of coated papers, consequently resulting in decreased tensile strength of paper[13, 18].

3.5.2. Elongation test

Elongation at break (%E) shows the ability of a film to stretch before it breaks. Elongation can be related to the paper's ability to conform and maintain conformance to a particular contour, and is also regarded as one of the most important criteria for the satisfactory behavior of paper in applications. Table5 shows the percent elongation in L–Direction which decreased slightly as a result of toner removal by solvent mixture, also decreased slightly in D–Direction in case of (methanol + ethyl acetate), while increased in case of methanol + benzene. Solvent mixture has a positive effect on elongation responses of the papers. This solvent mixture decreases the intermolecular forces along polymer chains and increases the free volume and chain mobility, imparting increased flexibility and stretch ability. Elongation of paper samples increase with using solvent mixture. This may be caused by a stress relaxation in the base paper during the removing process when the base paper was exposed to the solvent mixture in the removing solution. Removed papers are higher in elongation than un-removed paper, which means that solvent mixture improve paper strength and ductility [18, 19,20].

Table 5. The mean values of elongation test

Sample	Elongation test			
	L-Direction		D-Direction	
	cm	%	cm	%
Un-removed	0.534	5.34	0.324	3.42
Methanol + benzene	0.468	4.68	0.416	4.16
methanol + ethyl acetate	0.446	4.46	0.32	3.2

3.5.3. Bursting test[13]

The mechanical properties of paper determine its durability and resistance to environmental stress. To investigate the effect of solvent mixture on paper samples, the bursting strength of removed paper is measured as shown in Table6. The bursting strength of removed paper is decreased compared to the un-removed paper, but this difference is not statistically significant. For this reason, in principle, there is no problem of toner removal by solvent mixture. Most importantly, solvent mixture which is sufficient to remove toner on paper samplesdoesn't change its bursting strength.

Table 6. The mean values of bursting test

Sample	Bursting strength, (kg/cm^2)
Un-removed	2.37
Methanol + benzene	2.50
methanol + ethyl acetate	2.60

3.5.4. Tearing test

The strength properties (burst, tensile and tear) of paper are attributed to the fibre strength and the number of inter fibre bonds. From results listed in Table 7.it is clear that the tearing load paper samples of removed toner are higher than (in L–Direction) that reference paper. This may be due to the solvent mixture allow to a higher degree of polymerization (D.P) leads to increasing the inter fiber bonds and crosslinking between the fibers in the paper therefor tearing load in L-Direction. On the other hand, in D–Direction solvent mixture interferes the fibre bonding, which reduces the tearing load and number of inter fibre bonds [20, 21].

Table 7. The mean values of tearing test

Sample	Tearing load			
	L-Direction		D-Direction	
	Kg	N	Kg	N
Un-removed	34	333.426	49.33	483.795
Methanol + benzene	38	372.653	47.33	464.181
methanol + ethyl acetate	36	353.039	48.00	470.719

3.6. Physical Properties (thickness measurements)

The paper thickness obtained and the solvent mixtures influence on thisthickness can be seen in Table8. No difference is observed between the thickness of the un-removed and removed paper. The average thickness values of the un-removed and removed paper are statistically equal. From analysis of the results, it is possible to observe that the constant in thickness for both reference and removed paper. The solvent mixtures are volatile therefore don't formed other coating layer on the

surface of the paper and thickness increase, or deposition on to the cellulosic substrate lead to thickness decrease, therefor become constant of measurable paper thickness [22,23,24].

Table 8. The mean values of thickness measurements

Sample	Whiteness	Brightness	gloss		
			20°C	60°C	80°C
Un-removed	155.0	93.6%	0.1	4.18	4.32
Methanol + benzene	154.9	93.2%	0.1	3.94	3.28
methanol+ ethyl acetate	153.1	93.0%	0.1	3.96	4.22

3.7. Optical Properties

Optical properties whiteness, gloss and brightness of the un-removed and removed paper listed in Table 9. The results show that no significant change in this properties, due to using of solvent mixture in removal process. The brightness of paper depend on the number of individual particles in the paper structure, the grammage, the number of surfaces in the structure and on the differences in refractive index between the particles and the surrounding medium. Image of SEM shown after removal process slightly toner particles not removed and still remain on fillers of paper which possess higher light scatterings than cellulose fibres causing slightly reduce in optical properties including the whiteness, gloss and brightness [21,25,26,27].

Table 9. The mean values of whiteness, brightness and gloss tests.

sample	Thickness, mm
Un-removed	0.11
Methanol + benzene	0.11
methanol+ ethyl acetate	0.11

Case study 1

Case study included forgery by removal of original text and change in the value of the money amountin new re-printed text as shown in Figure6.

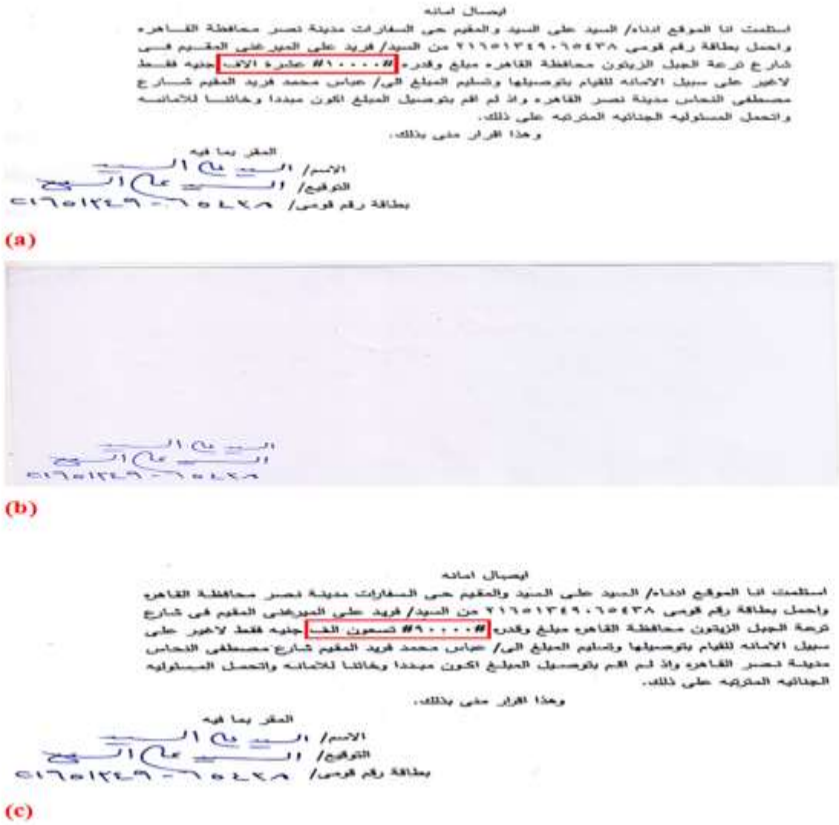

Figure 6. Images of: (a) original text before removal, (b) after removal and (c) re-printed new text

Case study 2

This case study involved removal of toner from security paper has stamp and signature without any effect on security features present in paper and also without any effect on stamp and signature as shown in Figure7.

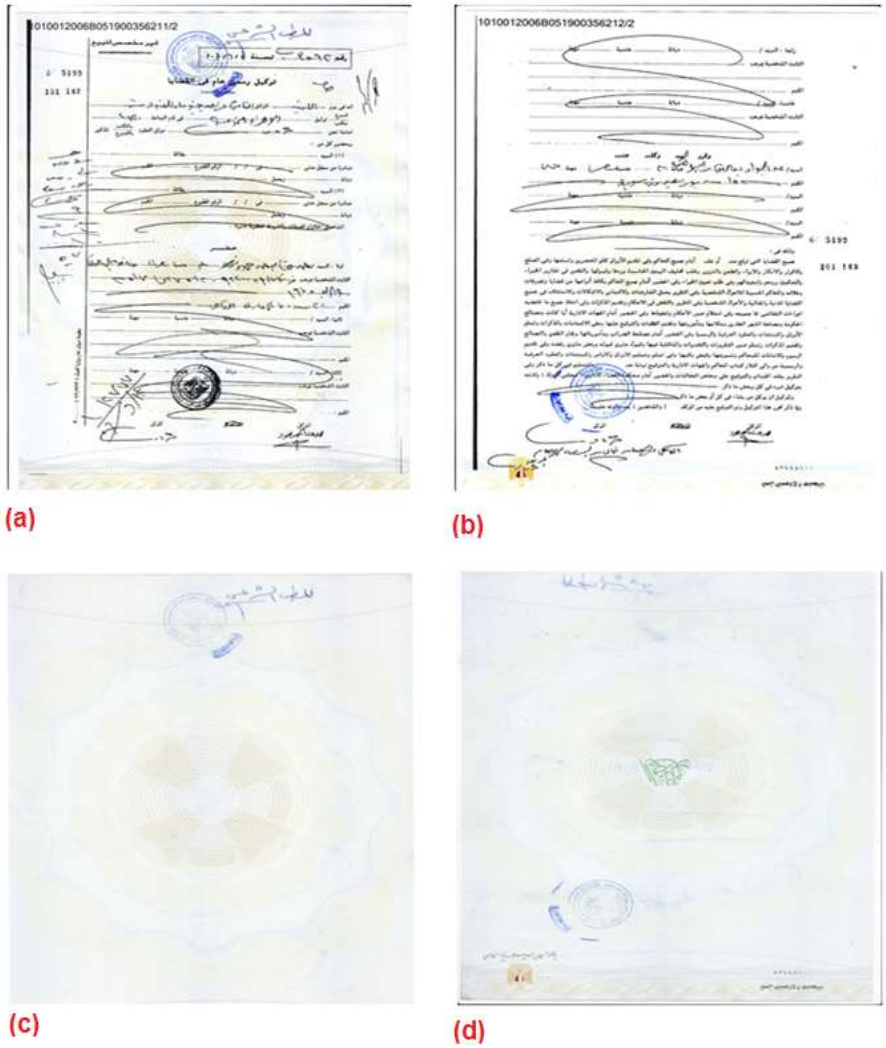

Figure 7. Images of security paper:(a) face involved signature and stamp before removal, (b) back involved stamp onlybefore removal,(c) face involved signature and stamp after removal and (d) back involved stamp only afterremoval

4. Conclusion

Paper samples printed by Hp LaserJet 2300dn (sample 1) soaked in 17 standard laboratory alone organic solvents and in 133 solvent mixtures to remove toner from paper. After removal, scanned paper image is classified into 6 groups. From this removal study, we have identified an efficient mixed solvent consisting of methanol

+ benzene (2:1), methanol+ dichloromethane (2:1) and methanol+ ethyl acetate (1:1) for use in toner removal from both ordinary and security paper. These mixed solvents are found to have similar structural features and solubility parameters close in proximity to that of the polymeric binders in toner. It is found that the mixed solvents play a key role in the removal process by rapid dissolution of toner for detachment from paper fibers. The optical tests, SEM andpaper properties (mechanical,physical, optical) are explored for paper removed by efficient mixed solvent. All tests show no significant difference between un-removed and removed paper therefor removed paper can be re-used to create forgery document.More effective dissolving mixtures are applied on paper sample from 2 to 6, but not give good results as sample 1.

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