

Optical and Printing Properties of Deinked Office Waste Printed Paper

B. Ghanbarzadeh¹, M. Ataefard^{*1}, S. M. Etezaad², S. Mahdavi³

¹ Department of Printing Science and Technology, Institute for Color Science and Technology, P.O. Box: 16765-654, Tehran, Iran.

² Department of Environmental Research, Institute for Color Science and Technology, P.O. Box: 16765-654, Tehran, Iran.

³ Wood and Forest Product Division, Research Institute of Forest and Rangeland, Agricultural Research Education and Extension Organization (AREEO), P.O. Box: 315851343, Tehran, Iran.

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ABSTRACT

Deinking waste paper is done in various ways, and research on newer methods is also being done. Traditional deinking methods to remove ink from waste printing papers have a relatively high efficiency compared to contact methods, but in non-contact printing such as electrophotographic printing (LaserJet printers and photocopiers) due to Mixing of the ink with the paper texture during the stabilization of the ink with the help of high heat, the traditional deinking methods are less efficient and it is necessary to use newer methods. In this research, enzymatic deinking of office waste paper was done by a commercial cellulase enzyme in a neutral environment. 15 tests were conducted, and optical and printing properties including the paper were investigated. By measuring the reflection before and after printing and measuring the print density after printing, it was determined that the printability of deinked paper is within the desired range. Eric's number was 128 ppm for sample number 8 and 138 ppm for sample number 12, which has decreased compared to the control sample with an Eric number of 571 ppm. Measuring the reflection before and after printing and measuring the print density after printing, show that the printability of decomposed ink paper is in the desired range. Prog. Color Colorants Coat. 17 (2024), 75-84© Institute for Color Science and Technology.

1. Introduction

Management, the exceeding production, and the environmentally safe disposal of solid waste is one of the biggest challenges of modern society [1]. Before the 1970s, municipal solid waste disposal generally consisted of depositing wastes in open or excavated

landfills, accompanied by open burning to reduce waste volumes [2]. But today's developed countries try to recycle solid wastes especially paper waste, because a large amount of solid wastes in these countries is paper and paperboard products and it has a great effect on the environment and people's routine life [3]. Recycled paper has become an important source of raw

*Corresponding author: * ateefard-m@icrc.ac.ir

material for the pulp and paper industry [4]. Every ton of recycled fiber saves an average of 17 trees plus related pulping energy. Also, for every ton of paper which is recycled about 30,000 liters of water, 1200 to 1400 kWh of electricity, and 95 % of air pollution could be minimized [5]. The recycling of waste paper would be a greener way to reduce environmental pollution since recycling could reduce forest destruction and also give less water and air pollution than making virgin paper [6].

Deinking is one of the important processes in recycling paper which means the removal of the printing inks from the used paper to obtain brighter pulp [7]. Deinking is usually performed through two main steps: (1) the separation of ink particles from the fiber, and (2) the washing or floatation of the pulp slurry to remove the separated ink. One of the most important factors in choosing the deinking method is the printing process and ink type [8].

The ink used in electrophotography (toners), consists of thermoplastic polymers and charge control agents along with carbon black and/or other pigments [9]. During the printing process, this ink is physically bonded to the substrate because of high heat and pressure, making it difficult and expensive to remove by conventional methods. Also, in conventional thinking, greater amounts of chemicals like NaOH, Na₂-SiO₃, Na₂CO₃, H₂O₂, chelating agents, and surfactants are used which increases the amount of releasing pollutants and makes the method expensive and environmentally damaging [10-12]. These chemicals increase the chemical oxygen demand (COD) values of wastewater hence increasing the cost of wastewater treatment [12].

Therefore, now a day's enzyme deinking becomes a new method to solve this problem [3]. The enzymatic treatment favors ink detachment from paper fibers without discharge of pollutants, thus contributing to environmental compatibility so Enzyme deinking is a clean method in the paper recycling industry [8]. Recently, enzyme deinking has made rapid progress, especially in the case of old newspapers [14]. About non-impact printing paper, some studies have been done. Anderes et al. [15], did enzyme treatment with commercial cellulase, and evaluated the brightness and number of specks on the handsheet. Kok Chang Lee et al. [8], evaluate the use of crude enzyme derived from *Penicillium rolfii* c3-2(1) IBRL to remove the non-impact ink particles from laser-printed papers. Tsatsis et al. [14], investigated enzyme treatment with

five cellulase enzymes with a different activity at acidic pH, and check the effect of these enzymes on speck's surface and brightness on the final hand sheet. Ghanbarzadeh et al. [16], investigated Enzymatic deinking of office waste printed paper: Optimization via response surface methodology. Ataefard et al. [17], investigated enzyme treatment with intelligent modeling and optimization of environmentally friendly green enzymatic deinking of printed paper.

In those researches mechanical properties of hand sheets, were investigated, but the printing and optical properties were not considered. So it seems that more research must be conducted on enzyme treatment performance for office waste papers. Therefore, the present work for the first time, seeks to disclose the effects of processing parameters in enzyme deinking of office waste papers i.e. enzyme dosage and the treatment time and temperature to optimize treatment conditions with considering the printing process.

2. Experimental

2.1. Materials

A4 size paper with 80 g/m² base weight and 100 μm thickness was printed by using an HP1320 printer. The striped pattern covered about 50 % of the sheet surface. HP LaserJet CF287A-X-XC toner supplied by HP Co. was composed of styrene acrylate copolymers, iron oxide, wax, and amorphous silica. Commercial cellulase enzyme, Cellusoft CR was supplied from Novozymes Co., Denmark.

Activity of cellulase toward carboxymethyl cellulose (CMC), was measured by using the Bernfeld method. The reaction mixture contained different concentrations of the substrate in 20 mM phosphate buffer (pH 7) and 100 μl of the enzyme solution in total volume of 500 μl. For optimum pH determination, the reaction mixture contained 50 μL of 1 % (V/W) of substrate in mixed buffer (phosphate, citrate) (pH 5-12) and 100 μL of the enzyme solution in total volume of 500 μL. The reaction was incubated at 45 °C for 15 min and then stopped by adding 500 μL DNS (3, 5-dinitrosalicylic acid) to the reaction mixture followed by heating in boiling water for 10 min. The amount of reducing sugars released was determined by DNS at 540 nm. One unit of enzyme activity was defined as the amount of enzyme required to generate 1 μmol of reducing ends per minute under the catalysis conditions described above. Maltose was used as a standard for

estimating reducing sugar. The Maltose standard curve was constructed with different concentrations of maltose. Protein concentration was determined by the Bernfeld method.

2.2. Methods

All sheets were immersed in tap water for 24 h to swell the fibers. Pulping process was carried out in a laboratory disintegrator for about 10 min at 3500 rpm and a consistency of 12 %. Enzyme treatment was carried out at pH=7 and 12 % consistency for three variables, i.e. time, temperature and enzyme dosage, in three levels, as shown in Table 1. After treatment, pulp slurry was filtered, washed with water, and dried for 24 h at room temperature.

After enzymatic hydrolysis, the pulp slurry was subjected to the flotation cell to separate the released ink from fibers immediately to avoid redeposition. The pulp was diluted to approximately 0.8 % consistency by using distilled water before flotation. And Tween 80 (0.5 %) was added as a surfactant. Then, the pulp was floated for 12 min in a flotation cell at room temperature. The foam containing floated ink particles was removed in parallel with the air bubble from the bottom of the flotation unit. At the end of the flotation process, the deinked fibers were recovered on a laboratory mesh sieve 400 [18].

2.3. Characterization

For evaluation of physical properties of deinked pulp, TAPPI test method T 205 sp-95 was used to make hand sheets from the deinked pulp with 60g/m² base weight and 95 μ m thickness [19, 20]. The bursting strength of the handsheet was determined based on the TAPPI T403 method using a bursting strength tester (burst tester 109B, DRK Corporation) [21]. The whiteness of the handsheet was determined based on the TAPPI T 562 test method [22] and the brightness was measured using a brightness and whiteness tester (ZB-A colorimeter, China) [23].

The printability of the prepared handsheet after deinking was checked via printing in a controlled environment using a monochrome laser-jet printer (HP 1100). An Ihara S900 spectrophotometer (USA) was used to measure the optical ink density of the printed

handsheet.

Eric's number or effective residual ink concentration is the ratio of the light absorption coefficient of paper pulp or paper containing ink of the light absorption coefficient of ink, both of which are measured at 950 nm wavelength. Eric's number was measured by an Analytic Jena Specord 250 spectrophotometer made in America [24].

3. Result and Discussion

3.1. Properties of deinked paper

Paper opacity is a measure of the back cover of the paper, in the sense that if the paper opacity is suitable, the text or image printed on the paper will not be seen from the opposite side. Enzyme treatment was carried out at pH=7 and 12 % consistency for three variables, i.e. time, temperature and enzyme dosage, in three levels, as shown in Table 1. The results of measuring the opacity of deinked papers are shown in Table 1. Since these results are very close to each other and are in a suitable range, the pattern obtained for this result was not significant. This means that the defined variables had no effect on the paper opacity.

In the case of white paper, the amount of reflection of light from the surface of the paper is actually a measure of the color change of the paper surface. The closer the reflection spectrum of the paper is the reference sample; it can be said that the color change of the paper after deinking is less. In the case of deinked paper, the reflection of the samples was measured and compared to the reflection of white paper. Figure 1 shows the image of deinked papers, and Figure 2 shows the measurement results.

As seen in Table 2 and Figure 3, the light value (L^*) for unprinted white paper (neat) is 91, which is higher than all the treated samples. In terms of color, neat paper is in the range of green and blue. All samples are in the blue range in terms of b^* index, and in terms of a^* index, samples 1, 2, 3, 4, 6, 8, 9, 10, 11, 12, 13, 14 and 15 are in the range is red and samples 5 and 7 are in the green range, which can generally be said that the color of the paper has changed after deinking. The L^* index is between 84 and 89, which in most cases has not changed much compared to the neat paper.

Table 1: determined levels of variability and answers of Opacity test.

Run	Time (min)	Temperature (°C)	Enzyme dosage (mg/15gr oven dried fiber)	Opacity (%)
1	15	45	1.5	89.58
2	120	45	0.015	91
3	60	60	0.15	96.53
4	60	60	0.015	9.84
5	60	30	0.015	99.95
6	60	30	1.5	96.98
7	120	45	1.5	94.09
8	60	60	1.5	94.28
9	60	45	0.015	95
10	15	45	0.015	94.88
11	120	60	0.15	95.46
12	120	30	0.15	96
13	15	30	0.15	96.53
14	15	60	0.15	97.31
15	60	45	0.15	90.69

Table 2: L* a* b* values for unprinted white paper and de-inked papers.

Run	L*	a*	b*
1	88	5.07	-12.55
2	87.70	9	-6.8
3	88	12.77	-13.26
4	84.20	33.88	-20
5	89.90	-6	-9
6	88.40	3.4	-11.8
7	88	-3.5	-4.3
8	88.50	4.39	-11.8
9	87	15.9	-12.4
10	86	9.3	-9.4
11	88	5.6	-6.7
12	89	14	-10.8
13	88.85	17.7	-15.4
14	89	13.3	-13.6
15	88	14.7	-18
Neat	91	5.36	-15

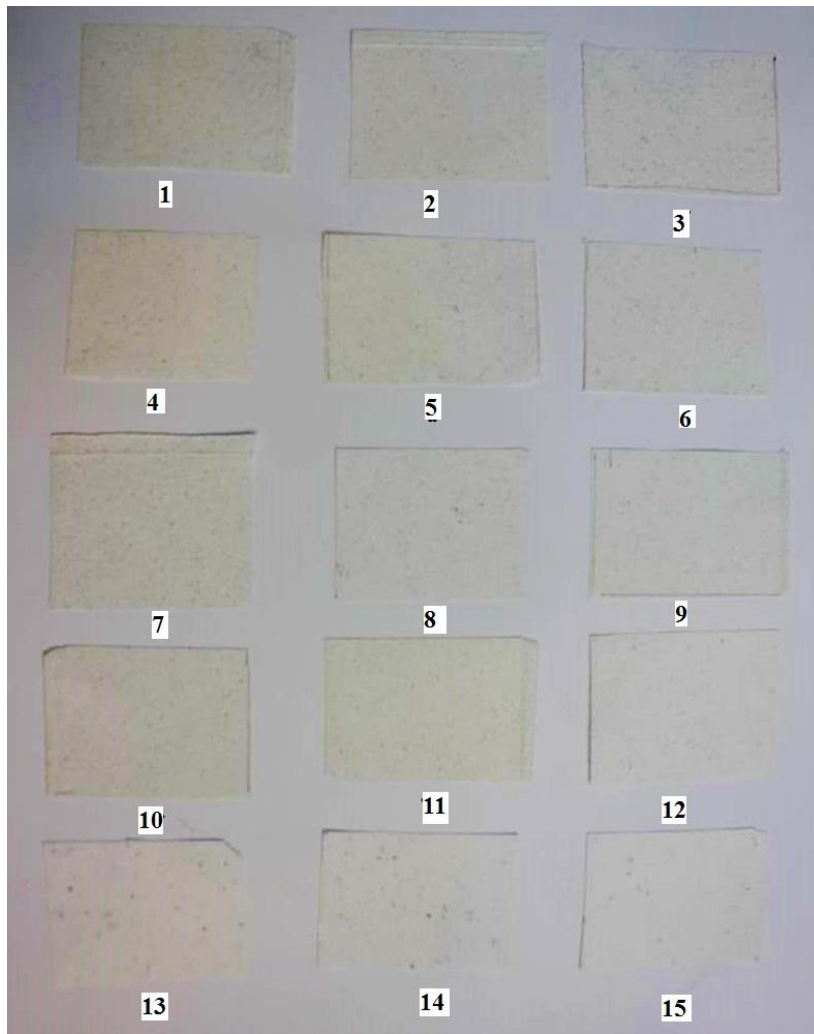


Figure 1: Image of deinked papers with 15 treatments.

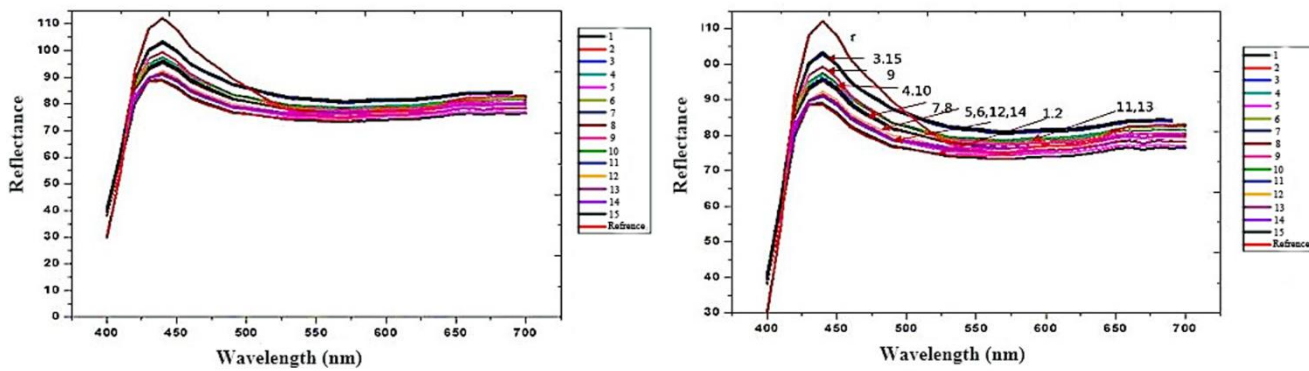


Figure 2: Reflection diagram of deinked paper and white paper

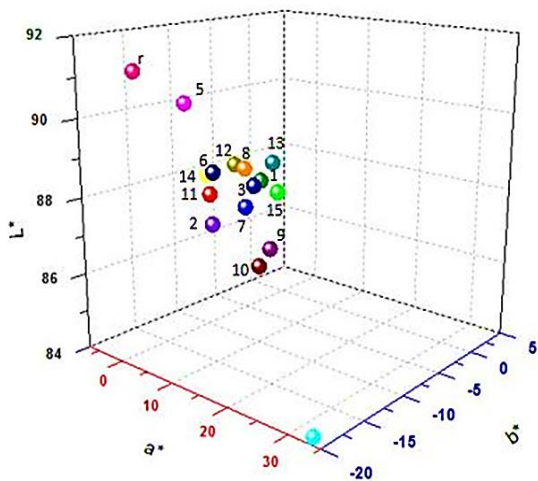


Figure 3: 3D diagram L* a* b* of deinked paper

3.2. Properties of deinked and printed paper

The paper used in this research is suitable for electrophotographic printing. Since the recycled paper will be used again in printing, the printability of the samples was investigated. It was printed by the HP 1320 printer with black toner and the reflection of the samples was measured. The reflection curve for an ideal black body is a flat curve with zero reflections. Figure 4 shows the reflection curve of the printed paper for each sample as well as the neat paper, which is his original paper.

Figure 5 shows the image of deinked and then printed papers. According to the diagram, all the samples in the blue and red range have been absorbed and the black color has become a little red. The reflection of the samples is higher than that of neat paper, which shows that the ink absorption on the paper surface has decreased. Sample 12 has the most similar reflective spectrum compared to the reference sample and has the highest absorption of ink among the de-inked paper. Sample 13 has the highest reflection in all wavelengths and has the lowest composite absorption.

The optical components of deinked and printed papers were measured and compared with the sample printed on neat paper (neat). Table 3 shows the values of these components for printed papers.

According to Table 3, and Figure 6, the part printed on the deinked papers is brighter than the one printed on neat paper, which indicates the reduction of absorption of black ink on the deinked papers. Also, all the samples and neat paper are in the range of red and

blue in terms of the color spectrum, which confirms the results obtained from the reflection spectrum.

Printing density is the ratio of the amount of light reflected from the white surface of the paper to the light reflected from the printed (black) surface of the paper. The printing density of reference number one paper and de-inked paper was measured and the results are shown in Table 4.

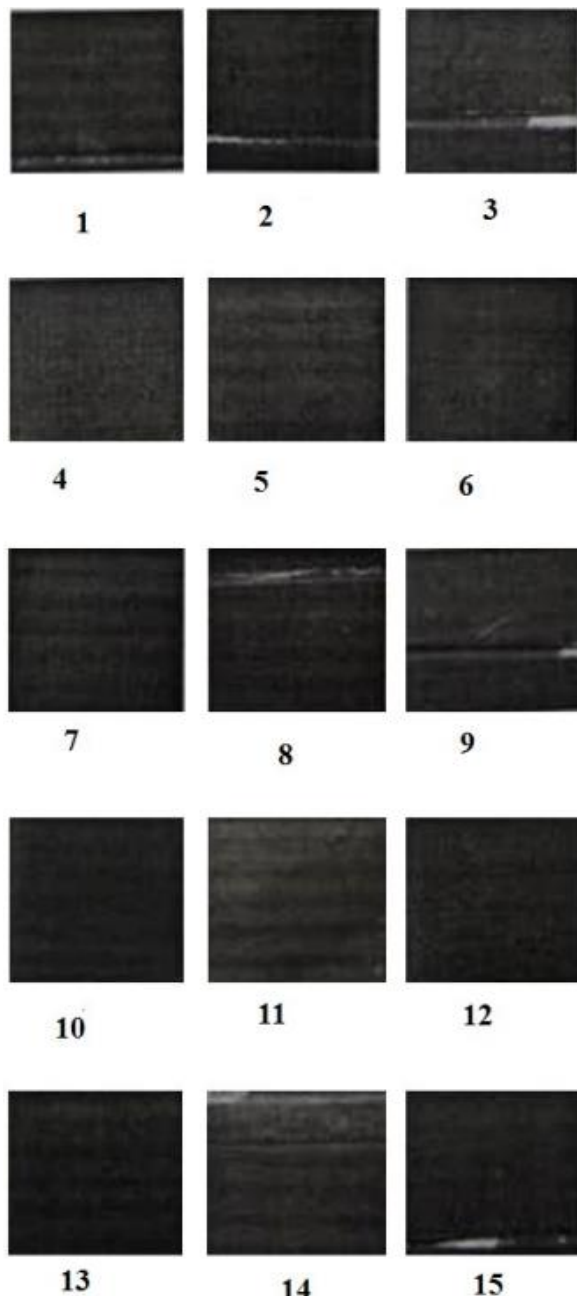


Figure 4: Image of deinked and printed papers.

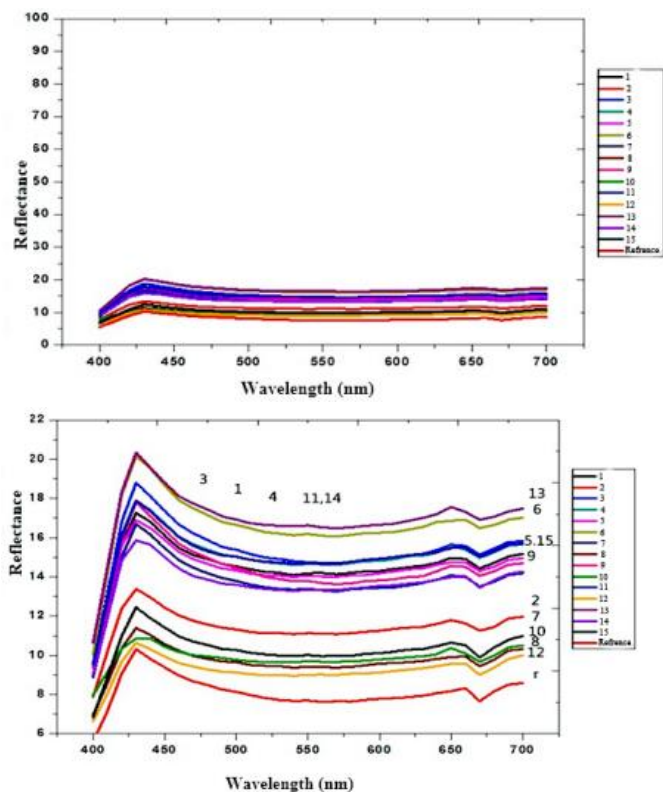


Figure 5: Reflectance diagram of the control sample and de-inked samples after printing.

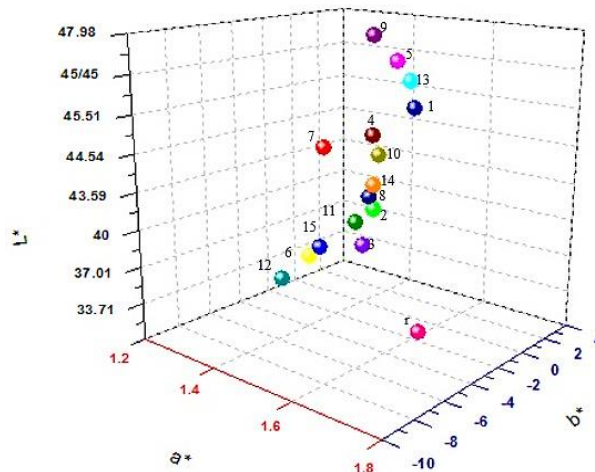


Figure 6: 3D diagram of L* a* b* deinked and printed paper.

Table 3: values of optical components for de-inked papers.

Run	L*	a*	b*
1	45.51	1.64	-9.03
2	43.51	1.53	-2.86
3	37.01	1.38	-2.64
4	45.45	1.38	-3.58
5	47.48	1.62	-3.94
6	36.19	1.33	-2.10
7	44.54	1.55	-3.90
8	43.59	1.54	-3.76
9	47.98	1.56	-3.76
10	44.76	1.54	-3.23
11	10	1.48	-2.74
12	33.71	1.24	-2.94
13	45.7	1.67	-4.27
14	44.28	1.54	-4.60
15	38.1	1.51	-3.50
Neat	33.18	1.96	-4.34

Table 4: Printing density values for reference paper and deinked papers.

Printing density	Run	Printing density	Run	Printing density	Run
0.87	11	0.97	6	0.76	1
1.06	12	0.78	7	0.82	2
0.76	13	0.83	8	0.95	3
0.381	14	0.99	9	0.77	4
0.96	15	0.81	10	0.76	5
1.4			Neat		

The print density of the witness sample is 1.4, and sample 12 has the highest print density among the test design samples, which shows that its print quality was the highest among all samples. This sample is in the range of blue and green colors before printing and after printing, but after printing, the degree of color saturation has decreased. In terms of the reflected spectrum before printing, sample 12 reflective is lower than neat paper, and regarding the reflective spectrum after printing, it has the closest reflective spectrum compared to control sample 1, which shows that the composite absorption is higher in this sample. And it has a better print quality that confirms the print density results.

Eric's number indicates the concentration of the effective residual ink. Eric's method is used to measure its reflectance in the infrared range of the spectrum. In this range, the size of the light absorption coefficient of the ink is several times larger than the size of the absorption coefficient of paper fibers and other compounds, and it provides a sensitive method for estimating the concentration of the remaining ink. During decomposing, after each step, the Eric number can be measured and the changes in the process can be checked by means of it. Eric's number was measured for handmade paper prepared for printed pulp and sample number 12 and sample number 8 which is the closest sample to the control sample. Eric's number for printed paper without deinking is 571 ppm, for sample number 12, it is 138 ppm, and for sample number 8, it is 129 ppm. After decomposition, Eric's number decreased by 76 % for treatment 12 and 77.5% for treatment 8. Sample number 8 has a lower Eric

number and more clarity (85 %) than sample 12 (82.59 %) [24].

3.3. Deinking mechanism

The Decomposition mechanism in the enzyme method is in two ways. In the first method, enzymes such as cellulase, xylanase, and pectinase are used, which hydrolyze the outer surfaces of the paper fibers in a limited way and the compound is separated from the paper. In this method, a part of the fine fibers is separated from the surface of the paper together with the ink. In the second method, enzymes such as lipase hydrolyze the oil-based carrier of the ink and the ink is separated from the surface of the fibers. In this method, only the ink is separated from the paper [25]. In this research, to investigate the deinking mechanism of SEM images (Figure 7), related to the sample of printed paper, de-inked paper, handmade paper made from pulp prepared from printed papers without de-inking, handmade paper made from pulp. The unprinted white paper was reviewed. Also, the residue obtained from enzymatic deinking was dried on filter paper and its SEM image was prepared. Figure 7 a, shows that the toner particles are bonded to the paper fibers. A comparison of Figure 7-b and Figure 7 c shows that the decompression has been done in a relatively favorable way. In Figure 7 d. We can see that the toner particles along with the fine fibers are separated and placed on the filter paper, which confirms the decompression mechanism in the first method.

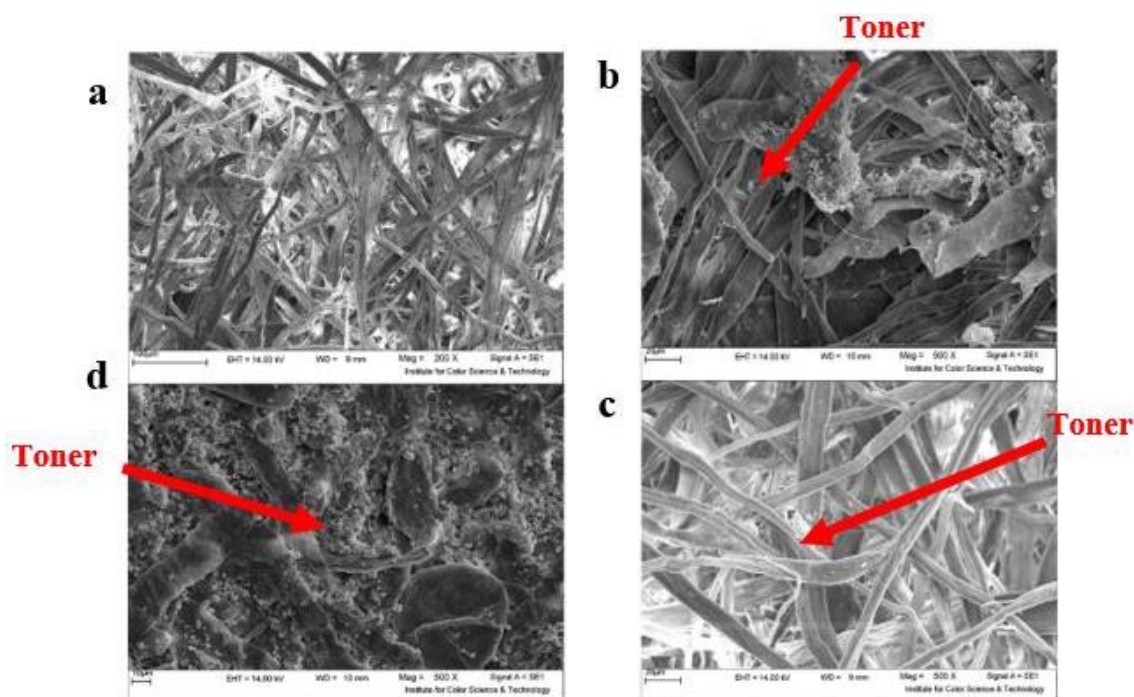


Figure 7: a) Handmade paper prepared from untreated pulp, b) Handmade paper from the white pulp, c) Deinked handmade paper, d) Effluent collected on filter paper.

4. Conclusion

In this research, deinking of electrophotographic printed papers was done with the help of commercial cellulase enzymes. To investigate the effects of deinking condition, i.e., treatment time, treatment temperature and enzyme dosage on the properties of hand sheet 15 runs was considered. Tensile strength, burst resistance, whiteness, and lightness of the paper, were analyzed. According to the reflection of the deinked papers, sample 12 with tensile strength of 8.9 N.m/g, the bursting strength of mN. 0.6 m²/g, whiteness (%) 72.88 and brightness (%) 82.59, has the closest reflective spectrum compared to the neat sample. In terms of the color spectrum, the neat paper

is in the range of green and blue, and sample 12 is in the range of red and blue. In sample 12, we have changed the color of the paper after deinking, but due to the high density of the print, it is suitable for the print. According to the results of the reflection of the samples after printing and the results of the print density, sample 12 with a print density of 1.06 has the highest print quality. According to the measured results of the optical components, it can be said that after Deinking of paper color has changed in component a* (red and green) and has not changed in the component b* (yellow and blue) and the change in a* is within the acceptable range.

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