Pigment Color Printing on Cotton and Polyester Fabrics with Electron Beam Irradiation Curable Formulations

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Received 21 November 2007; accepted 29 April 2008
DOI 10.1002/app.28728
Published online 6 November 2008 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Accelerated electrons delivered by electron beam accelerator were used to fix pigment colors, incorporated in curable formulation containing diluting monomer and an oligomer, to cotton and polyester fabrics. Tetrahydrofurfurfryl acrylate monomer and trifunctional urethane-methacrylate (TFUMA) oligomer were used as curable base besides ethylene glycol. The fabrics were printed with these formulations and exposed to various doses of electron beam irradiation generated from the 1.5 MeV (25 kW) electron beam accelerator machine. The irradiation dose, formulation composition, and pigment color concentration were studied. The roughness and color fastness to rubbing, washing, and perspiration of the printed fabrics by electron beam irradiation, were compared to the same fabrics printed by conventional pigment printing pastes. The results showed that cotton and polyester fabrics printed with the pigment colors by electron beam irradiation displayed higher color strength than those fabrics printed by the conventional thermal curing at equal pigment color ratios depending on the kind of pigment color. The durability properties, in term of roughness, rubbing, washing, and perspiration of fabrics printed by electron beam irradiation are better than those printed by conventional thermal curing. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 111: 1892–1899, 2009

Key words: electron beam accelerator; pigment color; curable formulations; color strength; durability properties

INTRODUCTION

Pigment printing may be considered as the most commonly and extensively used technique for printing textiles because of its easy application to a variety of fabrics and relatively clean and environmentally friendly aspects.[1,2] On a global basis, about 50% of printed textiles are processed by pigment printing. However, pigment printing has a few problems such as the relatively high temperature cure, stiff hand, and poor crockfastness of printed goods.[3–5] Attempts to lower the curing temperature have received considerable attention not only because the high temperature process wastes energy, but also runs the risk of destroying substrates.[6]

Radiation curing by electron beam has become a well-accepted technology, which has found a large number of industrial applications mainly in the coating and printing fields.[7] It allows the rapid conversion of especially formulated 100% reactive liquids to solids. Initiation by electrons leads to free radical or cationic polymerization and crosslinking.[8] The formulation to be cured or crosslinked by electron beam irradiation which usually contains unsaturated monomers (e.g., vinyl pyrillidone, tetrahydrofurfuryl acrylate, and acrylic acid derivatives), oligomers (e.g., polyurethane acrylates, polyester acrylates, and polyether acrylates as well as epoxy acrylates), and other additives according to the desired properties.[9] Pigment printing with a formulation containing only pigment and reactive chemicals that can be cured rapidly at room temperature by electron beam or ultraviolet radiation is an attractive possibility. Primarily due to the elimination of water and solvents and the roughly 90% reduction in energy needed to evaporate them and cure the conventional binders in addition to the high production rates and smaller space requirements.[10] This work aims at using electron beam irradiation as an alternative for thermal curing in the pigment printing of cotton and polyester fabrics using formulation free from binders and thickeners. These formulations contain reactive chemicals (oligomers and monomers) and pigment colors. A comparative study between radiation and thermal curing of pigment prints in term of color measurements, mechanical properties will be conducted.

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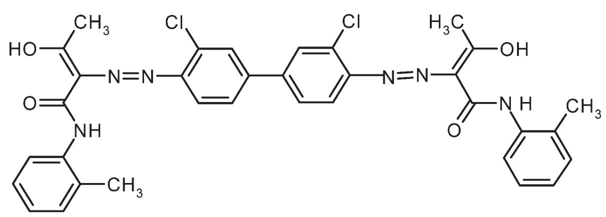
EXPERIMENTAL

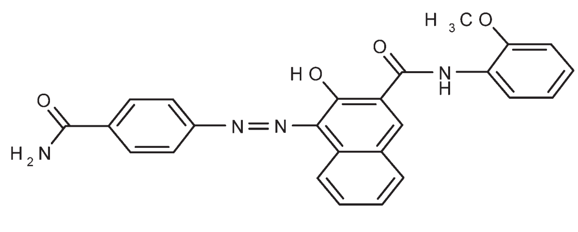
Materials

Cotton and polyester fabrics used in this work were received from Misr spinning and weaving (El-

Mehalla El-Kobrra). These fabrics were scoured, bleached, and mercerized, in which cotton fabric was 153 g/m^2 and polyester fabric was 215 g/m^2. The size of sample used for electron beam curing was 5 cm × 10 cm. The pigments used were kindly supplied by Clarinet. These pigments are Printofix Red H3BD-ET and Printofix Yellow HRNC-ET. The chemical structure and color index are shown below. The pigment colors were fixed on the textile surfaces by incorporating in electron beam curable formulations containing an oligomer and monomer. The oligomer used in this study was Trifunctional urethane-methacrylate (TFUMA), produced by Bomar Specialist, USA, while the diluting monomer tetrahydroxyfurfuryl acrylate monomer (THFA) was furnished by Sigma, Germany. Ethylene glycol was purchased from Merck, Germany. The synthetic thickener Alcoprint (PTP) and the synthetic binder Printofix MTB supplied by Clarient, were used in the conventional pigment printing.



C.I. Pigment yellow 110



C.I. Pigment red 215

Methods

Pigment printing by electron beam irradiation

The formulations for electron beam curing were prepared by dispersing the pigment color in the appropriate monomer (THFA monomer) with high shear polytron homogenizer. This dispersion was then stirred into molten oligomer (Trifunctional urethane acrylate oligomer) and mixed, after which ethylene glycol was added with continuous stirring until complete homogeneity. Printing was carried out on the surface of the fabrics with a floating coater with a thickness of 25 μm. The printed fabrics were exposed to accelerated electrons using the electron beam accelerator facility of the National Center for Radiation Research and Technology (1.5 MeV and 25 kW), Nasr city, Cairo, Egypt. The required doses were obtained by adjusting the electron parameters and conveyor speed.

Pigment printing by thermal curing

The conventional paste for printing was prepared by adding the synthetic thickener (25 g/kg) stepwise to water with stirring then the required amount of binder (100 g/kg) was then added to the stock thickening followed by addition of acid catalyst, diammonium phosphate (5 g/kg). The printing pastes were applied to fabrics through a flat silk screen. After drying, curing was conducted at 150°C for 5 min in an automatic thermostatic oven, produced by Warner Mathis, Switzerland. The printed samples by electron beam or thermal curing were soaped with aqueous solution containing (2 g/L) of nonionic detergent at 60°C for 15 min.

Measurements

Color measurements (ΔE)[11]

A microcolor unit manufactured by Dr. Bruno Lang GmbH, Konigsweg 10, D-1000, Berlin, Germany, was also used for color measurements. The L^*, a^*, b^* system used throughout this work is based on the CIF-color triangle (Commission International de E'claire units X, Y, Z). In this system, the L^* value represents the dark-white axis, a^* represents the green-red axis while the b^* represents the blue-yellow axis. The L^*, a^*, and b^* values of control and printed fabrics with the different methods were measured and the color difference ΔE^* of the fabrics after printing was determined as follows and expressed as color difference:

$$\Delta E^* = \{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2\}^{1/2}$$

Surface roughness

The surface roughness of the printed samples was measured by surface roughness measuring instrument SE 1770X, Kostaka lab, Company.

Fastness properties

The color fastness to crocking and perspiration was determined according to AATCC 8-1993.[12] Washing fastness tests were carried out according to BS1006 : C02 Test 2 with a soap solution (5 g/L, liquor ratio = 50 : 1) for 45 min at 48–50°C.[13]

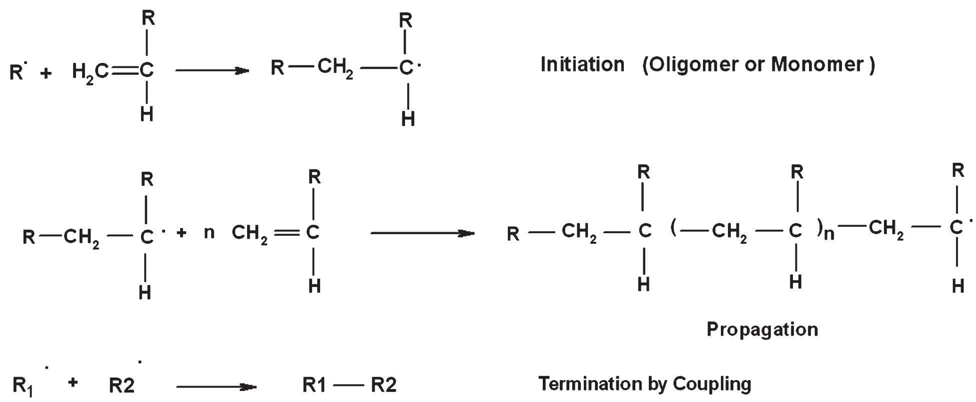


Figure 1 Mechanism of radical polymerization.

RESULTS AND DISCUSSION

Pigment printing on cotton and polyester fabrics by electron beam irradiation

Acrylates, methacrylates and their prepolymers (oligomers) show typical free radical addition polymerization. In free radical polymerization, a monomer or oligomer joins with a free radical and in effect, it forms a larger free radical that acts upon another monomer or oligomer and forms an even larger molecule, which is followed by coupling termination as it shown in the mechanism of a classical chain reaction in Figure 1. In the printing of substrates using the radiation curable formulation, in addition to the reactivity of monomers and oligomers towards EB curing, also, the compatibility between them in the formulation and the surface to be printed is very important factor, in this regards a preliminary experiments were conducted using various types of oligomers (e.g., epoxy acrylates, urethane acrylates, and acrylic acrylates) and monomers (e.g., tetrahydrofurfuryl acrylate and hexandioldiacrylae).

The initial experiments showed that TFUMA oligomer and THFA monomer are compatible with each other and to the surface of the cotton and polyester fabrics, they gave the desired properties of printing formulation in term of viscosity and stiffness, thus they became the application of choice.

Effect of irradiation dose

The color strength of cotton and polyester fabrics printed with different pigment colors incorporated in formulations containing constant composition of the monomer THFA and the oligomer TFUMA (25/75 wt %) is shown in Figure 2. It should be noted that all formulations contain a constant ratio of 2.5% ethylene glycol. The effect of electron beam irradiation as a fixation tool is very clear, in which the color strength of the exposed fabrics to electron beam irradiation is much higher than the unirradiated fabrics, regardless of the kind of fabrics used and the pigment color. The color strength of unirradiated fabrics is mainly resulted from the adhered paste on the surface even after washing and soaping. However, the cotton or polyester fabrics printed with the pigment yellow displayed the higher color strength than those printed with the pigment red by electron beam irradiation. In addition, irrespective of the color used, cotton fabrics printed with pigment colors displayed higher color strength than polyester fabrics at definite irradiation dose in the range of study. Generally, the dose of 75 kGy is the most effective one at which the highest color strength was achieved for cotton or polyester fabrics.

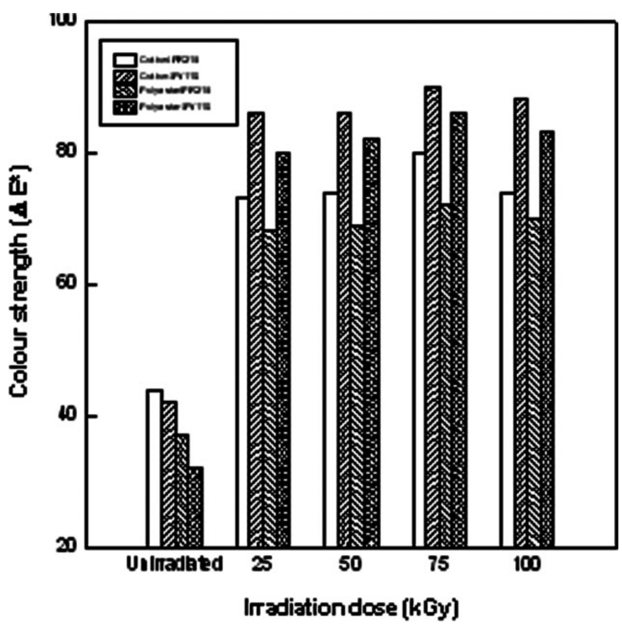


Figure 2 Effect of irradiation dose on the color strength of cotton and polyester fabrics printed by different pigment colors. Printing conditions: pigment ratio, 3%; ethylene glycol ratio, 2.5%; THFA/TFUMA composition, 25/75%.

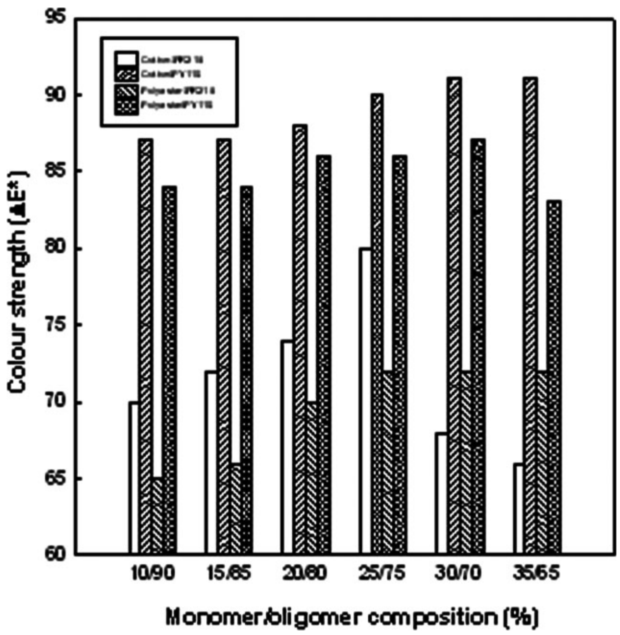


Figure 3 Effect of monomer/oligomer composition on the color strength of cotton and polyester fabrics printed by different pigment colors. Printing conditions: pigment ratio, 3%; ethylene glycol ratio, 2.5%; monomer (THFA); oligomer, (TFUMA); dose, 75 kGy.

Effect of monomer/oligomer composition

Figure 3 shows the effect of monomer/oligomer (THFA/TFUMA) composition on the color strength of cotton and polyester fabrics printed with different pigment colors. It should be noted that all formulations contain a constant ratio of ethylene glycol of 2.5 wt % and the fabrics were exposed to constant dose of 75 kGy of electron beam irradiation. It can be seen that the color strength increases constantly by increasing the monomer ratio from 10 to 25%, regardless of the kind of fabric or the pigment color. However, the color strength in the case of the printing with the yellow pigment color is still higher than that in the case of the printing with the red pigment color, regardless of the kind of fabric. It also seems that a composition of 25% of THFA and 75% of TFUMA is the best condition of monomer/oligomer composition displayed highest color.

Effect of pigment color ratio

The effect of pigment colors ratio on the color strength of the printed cotton and polyester fabrics is shown in Figure 4. It should be noted that the printing was conducted with formulations containing constant composition of 25% of THFA monomer and 75% of TFUMA oligomer and exposed to various doses of electron beam irradiation. Even though, the increase in color strength is not of practical importance, the printed cotton fabrics with the red pigment color displayed the relatively highest change by increasing the color ratio from 1 to 3% at any irradiation dose.

Further increase in the pigment red color ratio beyond 3% does not bring significant change in color strength. The increase in color strength of cotton fabrics printed with the red pigment color resulted from the increase in the pigment ratio from 1 to 3% and from 3 to 5% at the irradiation dose of 75 kGy was calculated to be $\sim$ 23% and 6%, respectively. As shown in Figure 3, the effect of electron beam irradiation is not significant when using low ratios of the red or yellow pigment colors; however, the major change was achieved by using relatively higher ratios.

Similar trends can be seen in the case of the printing of polyester fabrics with the different pigment colors as shown in Figure 4. The color strength increases greatly by increasing the red or yellow pigment colors ratios. In this regard, the color strength of polyester fabrics printed with the red pigment color and at the dose 75 kGy was increased by 11 and 0.2% when the ratio was increased from 1 to 3% and from 3 to 5%, respectively. The change in color strength of polyester fabrics printed with increased ratios from 1 to 3% and 3 to 5% of the yellow pigment color at the same dose of electron beam irradiation was calculated to be 10 and 1.2%, respectively. In addition, it seems that the irradiation dose is not an effective factor in the case of low ratios of the red or yellow pigment colors.

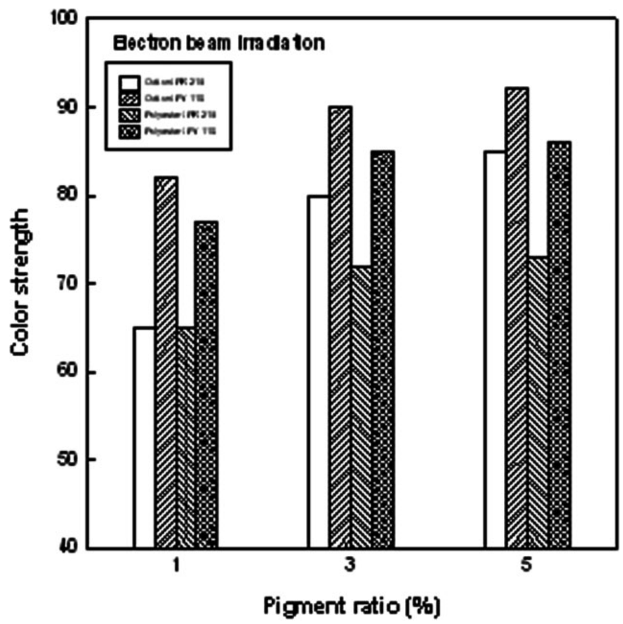


Figure 4 Effect of pigment ratio on the color strength of cotton and polyester fabrics printed by electron beam irradiation. Printing conditions: ethylene glycol ratio, 2.5%; THFA/TFUMA composition, 25/75%; dose, 75 kGy.

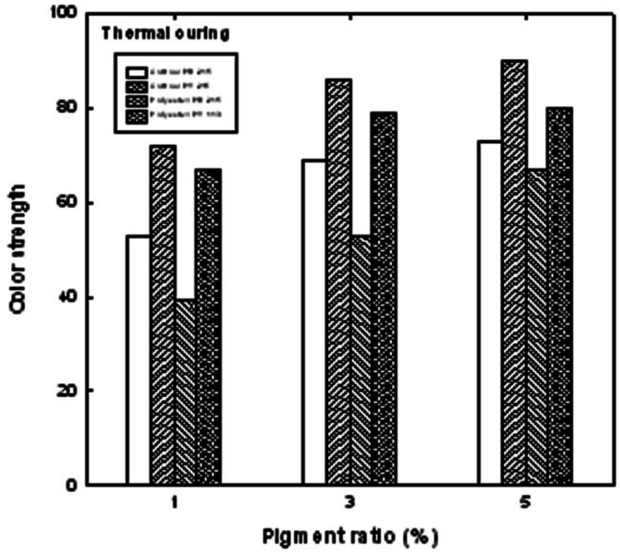


Figure 5 Effect of pigment ratio on the color strength of cotton and polyester fabrics printed by thermal curing. Printing conditions: Pritofix MTB binder; 10%; Alcoprint PTP thickener, 2.5%; thermal curing at 160°C for 5 min.

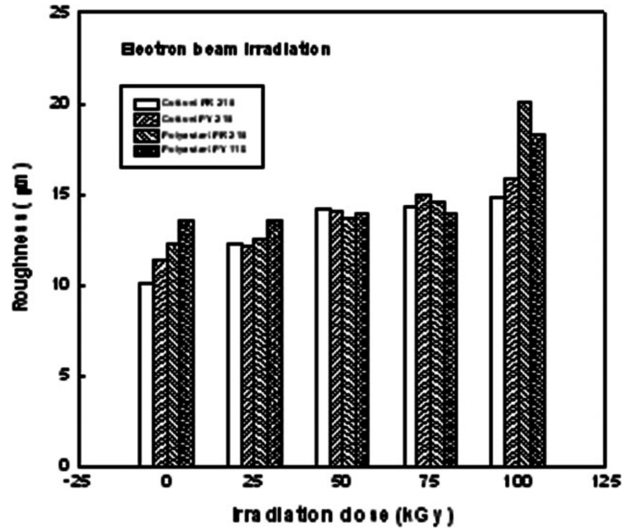


Figure 6 Roughness properties of cotton and polyester fabrics printed with different pigment colors by electron beam irradiation. Printing conditions: ethylene glycol ratio, 2.5%; THFA/TFUMA composition, 25/75%; pigment ratio, 3%; dose, 75 kGy.

On the basis of the results in Figures 2–4, few points may be indicated; (1) electron beam irradiation is responsible for the fixing of pigment color in the formulation and fixing the whole formulation to the fabric surface through crosslinking reactions. In this context, the exposure of the printed cotton fabric at a relatively lower dose of 25 kGy resulted in an increase in color strength of 66% in the case of printing with the red pigment color and 105% in the case of printing with the yellow pigment color. In the case of the printing of polyester fabrics with the red and yellow pigment colors, the increase was 84 and 150%, respectively. (2) The highest color strength was observed in the case of the printing of cotton and polyester fabrics with the yellow pigment color. However, the printing with the yellow pigment color displayed higher color strength on cotton fabrics than the printing of polyester fabrics. (3) The irradiation dose seems more effective factor rather than monomer/oligomer composition factor regarding the printing of either cotton or polyester fabrics. The increase of the ratio of the THFA monomer beyond 25% in the formulations causes a great drop in the color strength of cotton fabrics printed with the red pigment color and nearly has no effect on the color strength in the case of cotton fabrics printed with the yellow pigment color and polyester fabrics printed with both pigment colors. (4) The exposure of the coated fabrics with the different formulations to higher doses up to 100 kGy differs from fabric to another. Although the color strength of cotton fabrics printed with red and yellow pigment colors was decreased by 7.5 and 2.2% by increasing the irradiation dose from 75 to 100 kGy, the decrease in color strength of polyester fabrics printed with the same pigment colors was found to be 2.8 and 1.2% within the same range of irradiation doses, respectively. These findings indicate that higher irradiation doses cause oxidative degradation to cotton fabrics, whereas polyester fabrics seem to be more resistant against electron beam irradiation. (5) The chemical structure of the pigment colors seems to play an important role in the printing of cotton or polyester

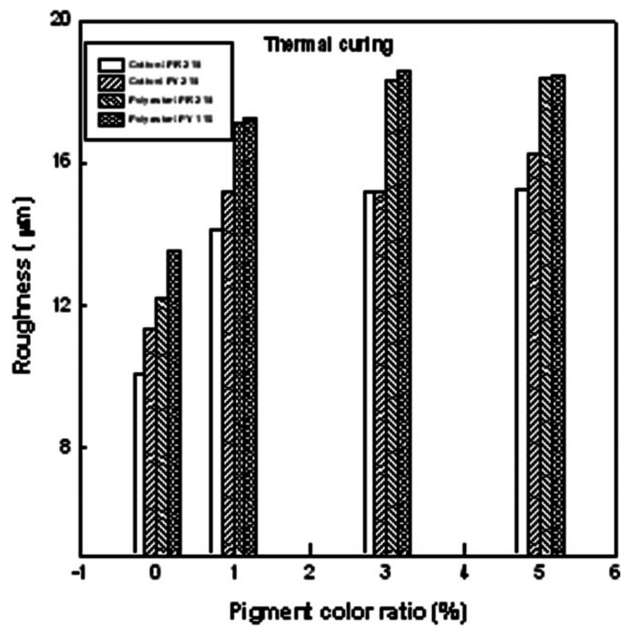


Figure 7 Roughness properties of cotton and polyester fabrics printed with different pigment colors by thermal curing. Printing conditions: Pritofix MTB binder; 10%; Alcoprint PTP thickener, 2.5%; thermal curing at 160°C for 5 min.

TABLE I
Rubbing Fastness of Cotton and Polyester Fabrics Printed with the Red and Yellow Pigment Colors by Electron Beam and Thermal Curing

	Electron beam irradiation (75 kGy)				Thermal curing (160°C)			
	Pigment red 215		Pigment yellow 110		Pigment red 215		Pigment yellow 110	
Kind of fabric	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
Cotton	4	3	3–4	2–3	3–4	2–3	4	2–3
Polyester	4	2–3	4	3	4	2–3	4	3

fabrics by electron beam irradiation. In this regard, the higher color strength offended by the yellow pigment color with respect to pigment red can be attributed to the presence of two active chlorine atoms in the molecular structure of the yellow pigment color. (6) The slight decrease in the color strength of the polyester fabrics occurred by increasing irradiation dose from 75 to 100 kGy is due to the presence of benzene rings along the structure of polyethylene terphthalate, which dissipates radiation energy.

Pigment printing on cotton and polyester by thermal curing

Pigment printing by thermal fixation is one of the most popular printing methods for many uses and for the easiest and simple method of application. Insoluble colors, which have no affinity for any fiber, are used in a finely dispersed form and the film forming binders are used to fix these pigments to the surface through crosslinking reactions. The binders are high molecular weight polymers that based on acrylates, vinyl acetate, etc., are essential components for printing by thermal curing.[13,14] Thickeners are necessary systems for pigment printing for the viscosity of printing pastes flow to be transferred onto the fabric easily.

In this work, the pigment printing on cotton and polyester fabrics by electron beam curing was compared with the pigment printing on cotton and polyester fabrics with the same colors by thermal curing. The pastes for thermal curing were prepared by incorporating the different pigment colors, with the commercial binder (Printofix MTB) and the synthetic thickener (Alcoprint PTP). Figure 5, shows the color strength of cotton and polyester fabrics printed with various ratios of the red and yellow pigment colors by thermal curing at 160°C for 5 min. It is clear that there is no optimum ratio of the pigment colors, in which the color strength was progressively, increase with increasing the ratio up to 5%, regardless of the pigment color or the fabric kind. However, it is interesting to compare the color strength of the printed fabrics by electron beam irradiation and thermal curing at similar conditions of printing. In this context, the monomer/oligomer composition is equivalent to the binder/thickener past and the electron beam irradiation of 75 kGy is equivalent to the thermal curing at 160°C for 5 min. Therefore, the color strength of cotton fabrics printed with 3% of the red and yellow pigment colors by electron beam irradiation was higher than those fabrics printed with the same ratio of the pigment colors by 16 and 5%, respectively (based on the color strength by thermal curing). Meanwhile, the printing of polyester fabrics with the same ratio of the pigment colors displayed an increase in color strength of 36 and 9% over those printed by thermal curing, respectively. In conclusion, cotton and polyester fabrics printed with different pigment colors by electron beam irradiation displayed higher color strength than the printing with the conventional thermal curing even at similar conditions.

TABLE II
Washing Fastness of Cotton and Polyester Fabrics Printed with the Red and Yellow Pigment Colors by Electron Beam and Thermal Curing

	Electron beam irradiation (75 kGy)						Thermal curing (160°C)					
	Pigment red 215			Pigment yellow 110			Pigment red 215			Pigment yellow 110		
Kind of fabric	SC.	SW.	Alt.	SC.	SW.	Alt.	SC.	SW.	Alt.	SC.	SW.	Alt.
Cotton	3–4	4	4	4–5	4–5	4–5	4–5	4–5	4–5	4	4	4
Polyester	4	4–5	4–5	4–5	4–5	4–5	4–5	4–5	4–5	4–5	4–5	4–5

TABLE III
Perspiration Fastness of Cotton and Polyester Fabrics Printed with the Red Pigment Color by Electron Beam and Thermal Curing

	Electron beam irradiation (75 kGy)						Thermal curing (160°C)					
	Acidic			Alkaline			Acidic			Alkaline		
Kind of fabric	SC.	SW.	Alt.	SC.	SW.	Alt.	SC.	SW.	Alt.	SC.	SW.	Alt.
Cotton	4	4–5	4–5	4	3	4	4–5	4–5	4–5	4–5	4–5	4–5
Polyester	4	4–5	4–5	4	3	4	4	4	4	4–5	4–5	4–5

Roughness properties of cotton and polyester prints by electron beam irradiation and thermal curing

Pigment printing by electron beam irradiation or thermal curing is s surface application; the major concern is the roughness properties, i.e., the uncomfortable felling of the wearing of the prints. This is understandable because the successful pigment printing by both methods implies the formation of crosslinking, which is essential for the adhesion of printing pastes in case of thermal method and the formulation in case of electron beam irradiation. Thus, the durability of printed fabrics in term of roughness is of major importance. Figure 6, shows the effect of electron beam irradiation on the roughness properties of cotton and polyester fabrics printed with different pigment colors incorporated in formulations containing constant ratios of the pigment colors and monomer/oligomer composition of (25/75%). It can be seen that the roughness of unirradiated polyester (coated only by the formulation) is higher than the cotton fabric at the same conditions. This can be explained based on the higher hydrophilicity of cotton fabrics than the synthetic polyester fabrics. In this regard, the formulation is highly adhered to the hydrophilic surface rather than the hydrophobic surface, which indicates that the surface of polyester is rougher than cotton. In addition, it can be noticed that the formulation containing the yellow pigment color is rougher than that the one containing the red pigment color. In general, the roughness was found to increase with increasing irradiation dose, in which there is no optimum dose to achieve the minimum roughness. This is understandable because the higher is the irradiation dose, the higher the crosslinking density and thus the solidification of the formulation will eventually increase.

Figure 7 shows the effect of pigment ratio on the roughness properties of cotton and polyester fabrics printed with different pigment colors incorporated in conventional pastes containing constant ratios of the Printofix MTB binder of 10% and the Alcoprint PTP synthetic thickener of 2.5% and cured at 160°C for 5 min. The increase in pigment ratio was accompanied by a slight increase in the roughness, regardless of the kind of fabrics or pigment color. However, still the printed fabrics with the yellow pigment color possess higher roughness values than those fabrics printed with the red pigment color.

On the basis of the roughness properties of the printed fabrics with either electron beam irradiation or thermal curing, few points may be indicated; (1) generally, the printing with the yellow pigment color makes the fabric more rough than the printed with the red pigment color in all the methods of printing.. Here too, this behavior can be attributed to the difference in molecular structure of pigment colors. (2) At roughly similar condition of printing by electron beam irradiation (3% pigment ratio and dose of 75 kGy) and thermal curing (3% pigment ratio and 160°C for 5 min), it can be seen that the printing of polyester fabrics by electron beam irradiation is more acceptable than the printing by thermal curing. However, there is nearly no difference between the roughness values of the cotton fabrics printed with electron beam irradiation or thermal curing.

TABLE IV
Perspiration Fastness of Cotton and Polyester Fabrics Printed with the Yellow Pigment Color by Electron Beam and Thermal Curing

	Electron beam irradiation (75 kGy)						Thermal curing (160°C)					
	Acidic			Alkaline			Acidic			Alkaline		
Kind of fabric	SC.	SW.	Alt.	SC.	SW.	Alt.	SC.	SW.	Alt.	SC.	SW.	Alt.
Cotton	4–5	4–5	4–5	4–5	4	4–5	4	4	4	4–5	4–5	4–5
Polyester	4–5	4–5	4–5	4	4–5	4–5	4	4	4–5	4–5	4–5	4–5

Color fastness properties of the printed fabrics by electron beam irradiation and thermal curing

The durability of pigment printing on cotton and polyester fabrics by electron beam irradiation and thermal curing was evaluated in term of fastness towards rubbing, washing, and perspiration using the gray scale according to I.S.O recommendations as shown in Table I–IV. The I.S.O recommendations was issued in such way that color difference in the National Bureau Standards (N.B.S) units 0, 4, 8, 16, and 32 have the fastness rating 5, 4, 3, 2, and 1 on the gray scale, respectively.

On the basis of the assessments shown in Tables I–III, several points may be addressed: (1) as shown in Table I for the rubbing properties, cotton fabrics printed with the red pigment color by electron beam irradiation are more resistant against rubbing than those cotton fabrics printed with the same pigment color by thermal curing, in the dry and wet state of assessment. However, polyester fabrics printed by electron beam irradiation or thermal curing displayed the same color fastness towards rubbing. (2) As shown in Table II, for the color fastness towards washing, cotton fabrics printed with the yellow pigment color by electron beam irradiation displayed slightly higher washing fastness towards washing than those printed with the same pigment color by thermal curing. On the other hand, polyester fabrics printed by electron beam irradiation or thermal curing displayed similar washing fastness properties. Meanwhile, the printing of cotton fabrics with the red pigment color by thermal curing is relatively better than the printing by electron beam irradiation with the same pigment color.

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