The Printability of Wool Fabric with Synthetic Food Dyes

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Abstract:
Dyes used for textile dyeing and printing have many different, complex chemical structures and are produced using risky chemical methods from petrochemical sources, endangering both human health and the environment. Today, most synthetic food dyes are also derived from petroleum, but they are edible and less toxic than those used to dye textiles. The current study is an attempt to reduce the risks on human health and the environment that resulting from the usage of synthetic textile dyes via evaluating printability of wool fabric with three synthetic food dyes. The chosen colors are Brilliant Blue FCF, Ponceau 4R, and Tartrazine. The effect of different factors on the color strength of the printed samples was studied, i.e. concentrations of acid donor, urea, solvent, and dyes. The impact of steaming time and temperature on dye fixation was investigated. Light, rubbing, washing fastness and ultraviolet protection factor "UPF" values of the printed woolen samples were evaluated. The printed samples gave high values of UPF and color fastness properties were ranging between moderate to excellent.

Keywords:
Textile printing, wool, food dyes.

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1. Introduction:

Dyes are non-nutritive food additives used mainly to give color to increase the appeal and recognition of food products, which in turn increases consumer acceptability (5, 16). Human culture has traditionally included color additives; drug colorants usage was noted in ancient Egyptian writings, and historians believe that food coloring probably improved the quality of food products (29).

In ancient times, colorants were obtained from natural sources, but nowadays they are obtained also from synthetic ones. Natural dyes are chosen over many other types when it related to food, but industrially, natural food colors don't meet quality standards since they fade quickly (24). Artificial food dyes offer numerous benefits in comparison to natural dyes. For instance, brighter and more consistent color, reduced price, and less sensitive to technological processing and storage circumstances. The product color is thus crucial for its appeal to consumers (23, 25). In fact, the use of artificial food dyes "AFDs" has dramatically expanded in the last few decades (19). Children represent the majority of consumers of processed foods that are presented in bright colors (28). Plant-based or naturally derived extracted dyes have no harmful health consequences. e.g. paprika "E 160 c", carotenes "E 160 a", cochineal "E 120", curcumin "E 100" and Betanin "E 162" (20). Synthetic dyes are prepared chemically and can potentially have detrimental impacts on one's health. Most of them commonly composed of aromatic rings and azo functional groups, certain dyes also comprise intricate, substantial, and intense structures derived from petroleum or tar coal (24). There is a great deal of debate about the safety of synthetic dyes, many of which have been tested for toxicity in animal experiments before being used in food. It's noteworthy to know that certain food dyes are considered safe in one nation but illegal for ingestion by humans in another, which makes determining their safety very difficult (15).

To help in identification of synthetic food colors, they were given an official number and a common name, which may vary from one country to another, The International Numbering System (INS) is a globally recognized system used to classify any components related to food. It assigns numbers ranging from 100: 199 for color additives whether they are accepted to use or not, as defined by the Codex Alimentarius ("Book of Food") established in 1963 by the Food and Agriculture Organization (FAO) and World Health Organization (WHO).

The European Union (EU) utilizes the INS system and applies an "E" prefix (representing Europe) to natural and synthetic food colors that have been authorized by the European Food Safety Authority (EFSA) (18). In contrast to the European Union; in the United States, synthetic colors that are permitted to use in food, pharmaceutical, and cosmetic but do not occur naturally are assigned "FD & C" numbers (10). Table (1) contains different names of some artificial food dyes and their hazardous effects.

Table (1): Different names of some artificial food dyes and their hazardous effects

Common name	Color index	E number	INCI name	FD&C name
Brilliant Blue FCF	C. I. 42090 (Food Blue 2)	E 133	Acid Blue 9	Blue No.1
Ponceau 4R	C. I. 16255 (Food red 7)	E 124	Acid Red 18	-------
Tartrazine	C. I. 19140 (Food Yellow 4)	E 102	Acid Yellow 23	Yellow No. 5

*Note: - INCI: International Nomenclature Cosmetic Ingredient - FD&C: Federal Food, Drug, and Cosmetic.

Brilliant Blue FCF is an artificial dye belongs to the triphenylmethane compounds (30), utilized in a variety of baked goods, drinks, desserts, medications, cosmetics, and other items. In 1969, the United States Food and Drug Administration (USFDA) approved it for use in meals and swallowed medications. This dye was banned in eleven European nations prior to 1993; however, following the establishment of the EU, it was approved as a safe food additive (22). It can provide several tones of green when combined with tartrazine (9). Many investigations have documented harmful effects on humans and animals, including attention deficit disorder and children hyperactivity, gastrointestinal origin cancers, lymphoma, and convulsions in rodents (22). Brilliant Blue FCF was approved by "USFDA" as an additive for food, cosmetics, and medications with an allowed daily intake (ADI) of 12.0 mg/kg bw/day (milligrams per kilogram of body weight per day) (2).

Ponceau 4R is an artificial strawberry red azo dye. It can be utilized in many different foods, medicine, and cosmetics. EFSA has allowed it (11), although it was banned by USFDA and is regarded as carcinogenic in certain nations, such as the USA, Norway, and Finland (14). The "Joint FAO/WHO Expert Committee on Food Additives" (JECFA) suggests a highest ADI of 4 mg/kg bw/day (7), but European ADI is 0.7 mg/kg bw/day (8, 11). At permitted dietary exposures, there is no evidence of cancer-causing, genetic-damaging, nerve-damaging, or reproductive and developmental-damaging effects; however, exceeding this limitation may cause allergies, particularly if paired with acetylsalicylic acid intolerance. It might also have traces of chemicals that could cause cancer, the occurrence of asthmatic symptoms, animal malignancies, and DNA damage have been documented

Tartrazine is an artificial lemon yellow azo dye that is widely employed as a food coloring agent. It is one of the most contentious color additives in terms of safety and is used in candy, ice cream, breakfast cereal, soups, jellies, cakes, beverages, and other foods (3). Tartrazine is employed in pharmaceuticals to impart a yellow, orange, or green color to liquids, capsules, pills, lotions, or gels. Additionally, it finds application in various other items including household cleaning products, paper disposable plates, pet meals, crayons, inks, face paints, and cosmetics (12). Ttartrazine elicits the greatest amount of allergy and intolerance reactions compared to other food azo dyes, especially in asthmatics and aspirin-intolerant individuals (20). Tartrazine may contain carcinogens and induce hyperactivity in children (28). The maximum acceptable dose (ADI) is 7.5 mg/kg bw/day according to EFSA approval, and 5.0 mg/kg bw/day according to USFDA (13).

Color is the main attraction of any fabric too. Synthetic dyes used for textile dyeing and printing have many different, complex chemical structures and are produced from petrochemical sources using dangerous chemical procedures, which provide risks to both the environment and human health. The utilization of these colors has a detrimental impact on all living organisms. Environmentalists are becoming increasingly concerned about them due to their harmful nature. In an attempt to reduce the risks on human health and the environment that come with the use of synthetic textile dyes, the current study examines the printability of wool fabric using three artificial food colors that have comparatively lower toxicity. The printed wool samples were evaluated for ultraviolet protection factor (UPF) values and color fastness to light, rubbing, and washing.

2. Materials and Methodology:

2.1. Materials:

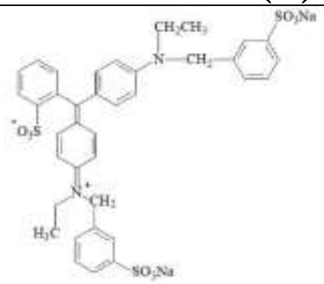
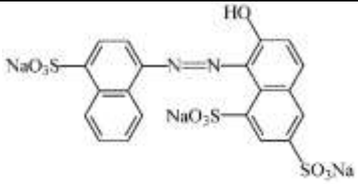
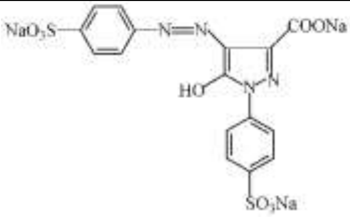
2.1.1. Fabric:

Mill-scoured 100% wool fabric (210 g/m^2), twill weave 2/2, is supplied by Misr Company for Spinning and Weaving, El Mehalla El Kobra, Egypt.

2.1.2. Dyestuffs:

Blue food dye (Brilliant Blue FCF), Red food dye (Ponceau4R), Yellow food dye (Tartrazine). All the dyes are in a powder form, supplied by Kamena Products Corporation, Egypt. Their chemical structures are demonstrated in table (2).

Table (2): Chemical structures of the dyestuffs

Dye name	Chemical structure (20)	Chemical Classification
Brilliant Blue FCF		Triphenylmethane
Ponceau 4R		Azo
Tartrazine		Azo

All the dyes are water-soluble sodium salt of sulphonic acids. They can be classified as acid dyes according to their application.

2.1.3. Thickening agent:

Meypro gum (Adprint EG) supplied by Adachi, India.

2.1.4. Chemicals:

Ammonium sulphate, urea, glycerin (99.5 %), and non-ionic detergent, all chemicals were of a laboratory grade.

2.2. Methodology:

2.2.1. Printing process & printing paste recipe:

Wool samples were printed with the three dyes via flat silk screen technique. The suggested formulation for the printing paste used is provided in grams per kilogram (g/kg) as given in table (3).

Table (3): Formulation of the printing paste

Weigh (g/kg)	Ingredients
20	Dye
30	Ammonium sulphate
30	Urea
30	Glycerol
500	Mypro gum (10%)
390	Water

Any decrease or increase in the weight of any ingredient accompanied by an increase or decrease in water to balance the printing paste to one kilogram. Ammonium sulphate concentrations (0, 10, 20, 30, 40 and 50 g/kg) were added to the printing paste to obtain the suitable pH degree to fix the dyes on wool fabrics, as well as different concentrations of urea (0, 10, 30, 50, 70, 90 and 110 g/kg) and glycerin (0, 10, 30, 50, 70and 90 g/kg) which act as solvents and hygroscopic agents to enable dyes to penetrate wool fiber to bond with. After printing, the samples were dried at ambient conditions prior to steam fixation.

The fixation process was carried out at various steaming temperatures (105, 110, 115, 120, 125, and 130 ºC) for different times (10, 20, 30, 40, and 50 minutes) in order to determine the ideal fixation conditions. Under optimal printing conditions the impact of different dye concentrations on the color strength of printed wool samples was assessed.

Following fixation, the printed samples had a cold water rinse, followed by washing with 2 g/l solution of non-ionic detergent, L. R. 1:50, at 65 ºC for 20 minutes. The samples were then rinsed with hot water, subsequently rinsed again with cold water and finally dried under ambient conditions.

2.2.2. Color strength (K/S) measurement:

The K/S of the printed wool was assessed using a Hunter lab spectrophotometer. The Miniscan Diffuse SAV, Stdz Mode, utilizes the Kubelka and Munk equation: K/S = (1-R)2/ 2R. In this equation, K, S, and R represent absorption coefficient, scattering coefficient and reflectance in the order given.

2.2.3. Ultraviolet protection factor (UPF) evaluation:

UPF of the printed samples were determined based on the Australian/New Zealand Standard AS/NZS 4399:1996 using Jasco V-750 Spectrophotometer. The UPF is a scientific measurement that quantifies the level of ultraviolet (UV) protection offered by cloth to the skin (26). UPF of fabrics is determined by many variables including fiber content, weave construction, dyes, and finishing process. For clothing materials to be categorized to provide high

UV protection, the UPF should range between 40 and 50+.

2.2.4. Color fastness properties:

Color fastness to washing was assessed using IS 687: 1979. Rubbing and light color fastness were evaluated using ISO 105-X12 (1987) and ISO 105-B02 in the order given.

3. Results and discussion:

3.1. Effect of ammonium sulphate concentration:

To examine the effect of ammonium sulphate concentration on K/S of wool fabric, printing pastes were prepared for each color with 20 g/kg dye, 30 g/kg urea, 30 g/kg glycerin, 500 g/kg mypro gum (10%) and different concentrations of ammonium sulphate. After printing, the samples were dried and steamed for 120°C for 40 minutes. The results are presented in figure (1) shows that K/S increases by increasing ammonium sulphate concentration. It may be due to the structural features of the dye and the fibers. During steaming, ammonium sulphate decomposes, ammonia liberates and the remaining sulphuric acid decreases the pH of the printing paste. Under acidic conditions, the amino groups present in wool fibers undergo protonation, resulting in the formation of positively charged amine and amide groups. Since all the three dyes contain sulphonyl groups (–SO3Na) and some also have hydroxyl (–OH) and/or carboxyl groups (–COOH), they ionized in acidic medium and acquire negative charge, so they would interact through ionic reaction with the protonated terminal amino groups (–NH3+) of wool. This ionic attraction forms salt linkages and would increase the printability of the fibers (6, 17). Also hydrogen bonds would form between the hydroxyl and carboxyl groups of the dye and (–NH) groups present in wool fibers (17, 21). The optimal concentration for prints using blue and red dyes was 30 g/kg, whereas the most effective concentration for prints using yellow dye was 40 g/kg.

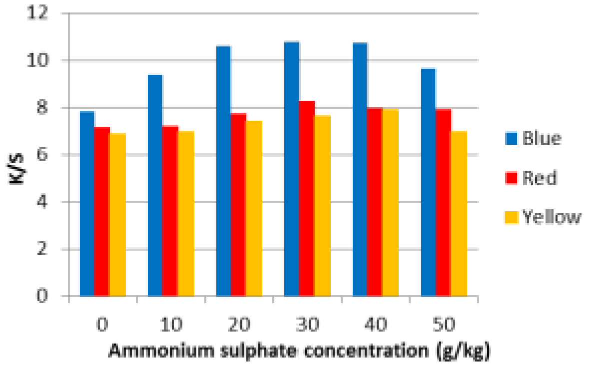


Fig. (1): Effect of ammonium sulphate concentration on K/S of the printed samples

3.2. Effect of urea concentration:

The relationship between K/S values and urea concentration (zero, 10, 30, 50, 70, 90 and 110 g/kg paste) is illustrated in figure (2). It was known that urea plays a favorable role on the color strength (K/S) in fabric printing because during steaming it acts as a water retaining agent that absorbs a sufficient amount of moisture to swell the fibers, increases solubility, disaggregation, and migration of the dye from the thickener film into the fiber matrix and accordingly improves the yield of dye reaction with the fabric. A concentration of 70 g/kg yielded the best results for prints with red and yellow dyes and 110 g/kg gave the best for blue.

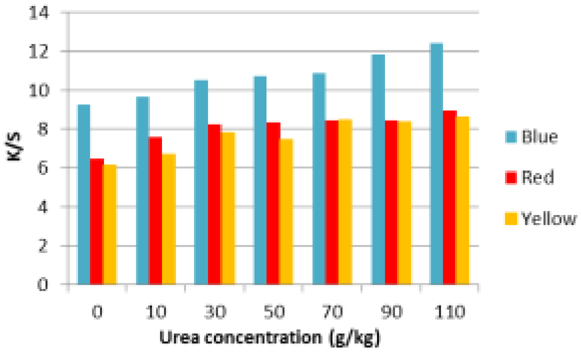


Fig. (2): Effect of urea concentration on K/S of the printed samples

3.3. Effect of glycerin concentration:

K/S of wool printed samples with the three food dyes is studied through adding different concentrations of glycerin to their recipes and the results are plotted in figure (3). It is observed that the optimum concentration of glycerin is 30 g/kg for yellow and red dyes and 70 g/kg for the blue. These results may be attributed to the hygroscopic property of glycerin, which enhances condensation of water on the fabric surface during steam fixation. This leads to the ease of penetration of the dye molecules through the fine microporous structure of the fabric followed by binding it (17).

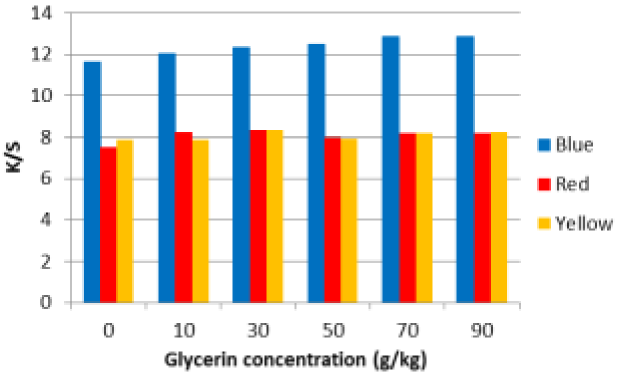


Fig. (3): Effect of glycerin concentration on K/S of the printed samples

The optimum concentration of printing pastes ingredients is presented in table (4). it is obvious that the blue dye needs higher amounts of urea and glycerin in the printing paste than the red and yellow dyes, it may be attributed to the structure of the blue dye which makes it more exposed to self-association and aggregation owing to strong intermolecular van der Waals-like attraction forces between the dye molecules, hence needs more amounts of solubilizing agents. Also the presence of auxochromes such as hydroxyl and/or carboxyl groups in the structure of the red and yellow dyes may influence their solubility because they are ionizable groups which are capable of imparting water solubility to the dye.

Table (4): Optimum concentration of printing pastes ingredients (g/kg)

Dye name	Dye concentration	ammonium sulphate	urea	glycerin	Mypro gum (10%)	water
Brilliant Blue FCF	20	30	110	70	500	270
Ponceau4R	20	30	70	30	500	350
Tartrazine	20	40	70	30	500	340

3.4. Effect of steaming temperature and time:

Wool samples were printed with 20 g/kg dye, 500 g/kg mypro gum and the chosen concentration of ammonium sulphate, urea and glycerin for each dye as shown in table (4), then the samples were dried at ambient conditions and subjected to saturated steam fixation at different temperatures for different intervals of time. The results were plotted in figures (4 & 5). Concerning fixation temperature, it is obvious in figure (4) that the K/S of the printed samples increases with the increase of steaming temperature. This enhancement in the color strength values is a direct result of the dissolution of dye molecules in the condensed water which eases its migration from the surface of the fabric into its fine microporous structure to bind with. The temperature of 120ºC is chosen to fix the fabrics to avoid fiber yellowing.

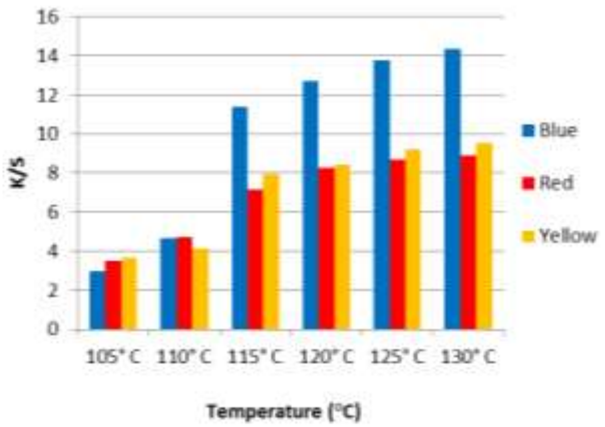


Fig. (4): Effect of steaming temperature on K/S of the printed samples (time = 40 min.)

Figure (5) displays the effect of steam fixation time on K/S of wool samples. It's obvious that increasing the time duration of steam fixation to 30 minutes is clearly associated with an enhancement in K/S values of the resulting prints, most probably because increasing steaming time enables sufficient dye dissolution, adsorption onto the fabric surface, diffusion and retention of dye molecules into the fiber matrix, thus allows for increased dye fixation. So the production of level and well penetrated printing is usually favored by increasing the time of steaming. Further increase in fixation time over 30 minutes has an adverse influence on the level of color strength of the printed samples, it can be because of negatively impacting the characteristics of the thickener film and the surface of the fabric; consequently, the coloring degree reduces (1). So the optimum chosen steaming condition is at 120 ºC for 30 minutes.

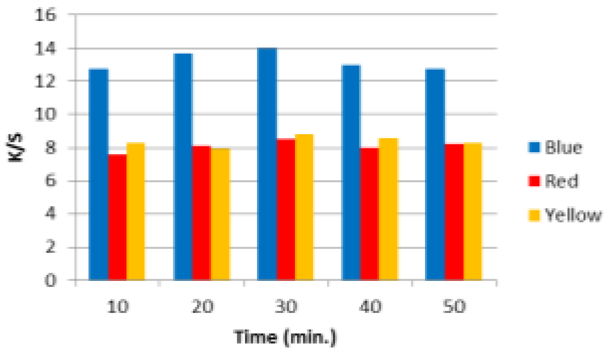


Fig. (5): Effect of steaming time on K/S of the printed samples

3.5. Effect of dye concentration:

To examine the correlation between dye concentration and K/S of the printed wool fabric, different printing pastes containing the optimum concentration of each ingredients and different concentration of each dye (10, 20, 30, 40 and 50 g/kg) were prepared. After printing, the samples were dried and steam fixed at 120 ºC for 30 minutes. It is clear in figure (6) that K/S of wool samples increases upon increasing the dye concentration in the range studied as a consequence of an increase in the quantity of dye molecules that can attach to the fiber and hence higher K/S values were obtained.

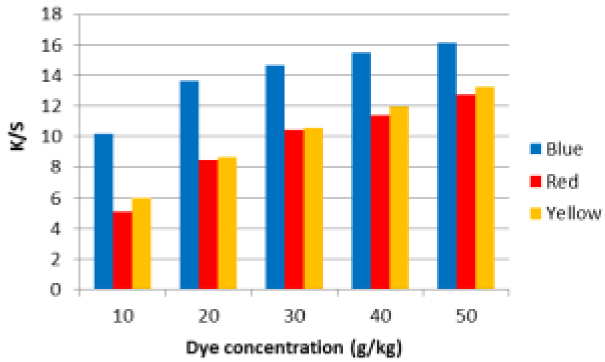


Fig. (6): Effect of dye concentration on K/S of the printed samples

3.6. Ultra violet protection factor (UPF) of printed wool samples:

The Ultra violet protection factor of printed wool samples were measured at the different concentrations of each dye and their results are presented in table (5). The results disclose that increasing dye concentration for each dye separately resulted in an improvement of UPF values, therefore enhancing the level of UV protection for the darker hues achieved, this might be explained as a greater degree of UV radiation blocking and/or absorption (27). All the prints achieved excellent protection category with UPF rating of 50+.

Table (5): Ultra violet protection factor of printed wool samples

Dye concentration (g/kg)	UPF of Brilliant Blue FCF	UPF of Ponceau 4R	UPF of Tartrazine
10	163	351	192
20	228	413	255
30	333	491	321
40	390	874	326
50	390	891	327

3.7. Color fastness properties:

Color fastness properties were evaluated at the middle concentration of the dye (30 g/kg) and the results were revealed in table (6). The results indicate that the three dyes gained acceptable overall fastness properties when printed on wool fabric. Wash fastness ranged between average and excellent. Rubbing fastness ranged from good to excellent. It may be due to the strength of linkages between the dyes and wool fibers. The light fastness tends to be moderate to very good. Light fastness depends on the chemical structure of dyes. Generally azo dyes show good to very good light fastness properties as obtained for red and yellow but dyes which contain triphenylmethane groups usually have very bright shades but relatively low levels of light fastness (4) as in case of the blue dye.

Table (6): Color fastness properties of printed wool sample

Printed Sample	Washing fastness			Rubbing fastness		Light fastness
	Alteration	Staining on cotton	Staining on Wool	Dry	Wet	
Brilliant Blue FCF	4-5	4	4	5	4-5	3-4
Ponceau 4R	4-5	3	3	5	4	5
Tartrazine	4-5	3-4	3-4	5	4	6

4. Conclusion:

The present study confirms the possibility of printing wool fiber with three different food dyes. The printed samples acquire good overall color fastness properties and excellent protection category with UPF rating of 50+.

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