

Article

Eco-Friendly Disperse Dyeing and Functional Finishing of Nylon 6 Using Supercritical Carbon Dioxide

Tarek Abou Elmaaty ^{1,*}, Eman Abd El-Aziz ¹, Jaehuyk Ma ², Fathy El-Taweel ³ and Satoko Okubayashi ²

¹ Department of Textile Dyeing & Finishing, Faculty of Applied Arts, Damietta University, Damietta 34512, Egypt; E-Mail: emanabdelaziz@hotmail.com

² Department of Advanced Fibro Science, Kyoto Institute of Technology, Kyoto 606-8585, Japan; E-Mails: boyx@navar.com (J.M.); okubay@kit.ac.jp (S.O.)

³ Department of Chemistry, Faculty of science, Damietta University, Damietta 34512, Egypt; E-Mail: fathyeltaweel@yahoo.com

* Author to whom correspondence should be addressed; E-Mail: tasaid@du.edu.eg; Tel./Fax: +20-572-353-098.

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Abstract: In this work, a supercritical carbon dioxide assembly was successfully constructed for dyeing Nylon6 fabric. Primary experiments were carried out to confirm the possibility of bringing the dyeing up to factory scale. A series of disperse azo dyes with potential antibacterial activity were applied to dye the fabric under our study in supercritical carbon dioxide (*scCO*₂). The factors affecting the dyeing conditions (*i.e.*, dye concentration, time, temperature and pressure) and functional properties were discussed and compared with those in aqueous dyeing. The comparison revealed that elimination of auxiliary chemicals such as salt, carrier or dispersing agent has no diverse effect on dyeing. The color strength of the dyed fabric evaluated by using K/S measurements increased by increasing dye concentration from 2% to 6% owf. (on weight of fabric). The nylon6 fabrics dyed in supercritical carbon dioxide have good fastness properties, and especially light fastness compared with conventional exhaustion dyeing. Antibacterial activity of the dyed samples under supercritical conditions was evaluated and the results showed excellent antibacterial efficiency.

Keywords: eco-friendly disperse dyeing; polyamide 6 fabric; supercritical carbon dioxide; antimicrobial disperse dyeing; combined process

1. Introduction

Using supercritical carbon dioxide ($scCO_2$) instead of water in textile dyeing can preserve energy, lower water use and prevent pollution. This dyeing method offers many advantages compared with conventional aqueous dyeing; no carrier or dispersing agent is required, residual dyestuff can be collected and carbon dioxide can be recycled [1–3]. It is an environmentally friendly technique, as it may replace the traditional wet-dyeing method [4].

Polyamide fibers have particular properties such as high tensile strength, elasticity and good mechanical and chemical resistance, *etc.* Correspondingly, polyamide fabrics are utilized in considerable applications of the textile industry [5,6].

The dyeability of synthetic hydrophobic fibers such as polyamide, polyester, polyacrylonitrile and polypropylene in $scCO_2$ poses a great challenge to dyestuff chemists. As a consequence in the last few decades, various researchers focused their efforts on the synthesis of new dyes for these fibers [7–12]. The success of dyeing synthetic textiles, particularly polyester [13–23], with disperse dyes in $scCO_2$ prompted research application of this technique to other synthetic fibers such as polypropylene [24], and aramid fibers [25] or alternative natural fabrics like cotton [26–28].

However, only a limited number of studies have been published on dyeing of polyamide textiles using $scCO_2$. The dyeing of nylon 6-6 with hydrophobic-reactive and disperse-reactive dyes using supercritical carbon dioxide as a solvent was reported [29,30]; a covalent force was formed successfully between the terminal amine group of nylon 6-6 and the vinylsulphone group of the dye molecule. The work indicated that both solubility and affinity have an effect on the dye uptake of nylon 6-6 with hydrophobic reactive and disperse-reactive dyes using supercritical carbon dioxide as a solvent system. Light fastness was acceptable for common applications and washing fastness was superior.

In an earlier study [10], we explored the dyeing of polyester fabrics with antibacterial disperse-azo dyestuffs which were synthesized in our program, employing a supercritical carbon dioxide dyeing technique. Working with antibacterial dyes in textiles integrated the dyeing and finishing process and resulted in a more effective technique in terms of water and energy management. The obtained result showed that this process was absolutely as adequately efficient as the typical procedure and led us to study the behavior of the synthesized dyes on other synthetic fabrics. In this context, the purpose of this work was to provide a one-step dyeing and finishing process for nylon 6 fabrics with antimicrobial disperse dyes through supercritical processing.

2. Experimental Section

2.1. Fabric and Dyes

A 100% polyamide 6 plain plane weave fabric (70 g/m²) supplied by Shikisen-sha company (Osaka, Japan) was used as dyeing substrate. Figure 1 shows the chemical structure of dyes employed in our research and was prepared according to the literature [31].

dye	Name (IUPAC)
1	2-(1,5-dimethyl-3-oxo-2-phenyl-2,3-dihydro-1H-pyrazol-4-yl)-2-oxo-N'-(p-tolyl) acetohydrazonoyl cyanide
2	2-(1,5-dimethyl-3-oxo-2-phenyl-2,3-dihydro-1H-pyrazol-4-yl)-N'-(2-methoxyphenyl)-2-oxoacetohydrazonoyl cyanide
3	2-([1,1'-biphenyl]-4-yl)-N'-(1,5-dimethyl-3-oxo-2-phenyl-2,3-dihydro-1H-pyrazol-4-yl)-2-oxoacetohydrazonoyl cyanide
4	N'-(2-chloro-4-methylphenyl)-2-oxo-2-(p-tolyl) acetohydrazonoyl cyanide
5	N'-(2-chlorophenyl)-2-oxo-2-(p-tolyl) acetohydrazonoyl cyanide

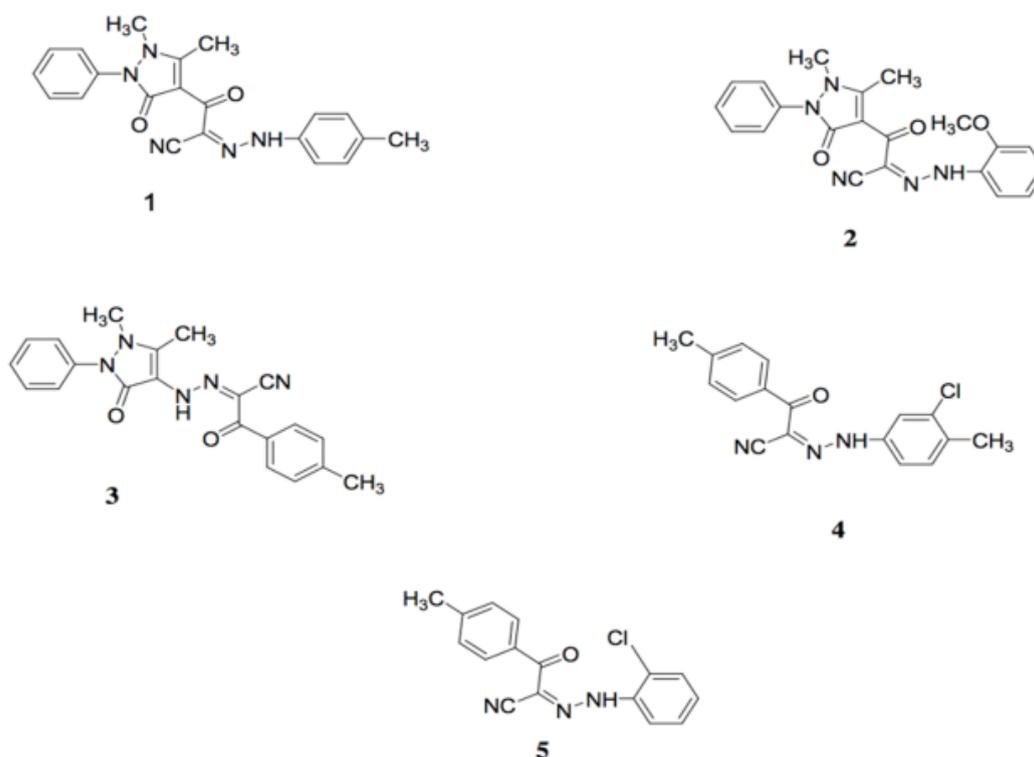


Figure 1. The chemical structure of dyes.

2.2. Dyeing Apparatus

Figure 2 is a diagram of the whole apparatus. The liquefied CO₂ departing from the cylinder moved inward to a cooling unit and was infused into a high-pressure syringe pump (model Jasco P11-2880 plus, Jasco, Easton, PA, USA). High-pressure CO₂ ultimately ran out into a dyeing autoclave. The dyeing

autoclave (Jasco EV-3, Jasco, Easton, PA, USA) as shown in Figure 2 is a 50 cm³ stainless steel autoclave outfitted with a steel screw-tube, a pressure sealed magnetic stirrer, and a quick-release cap.

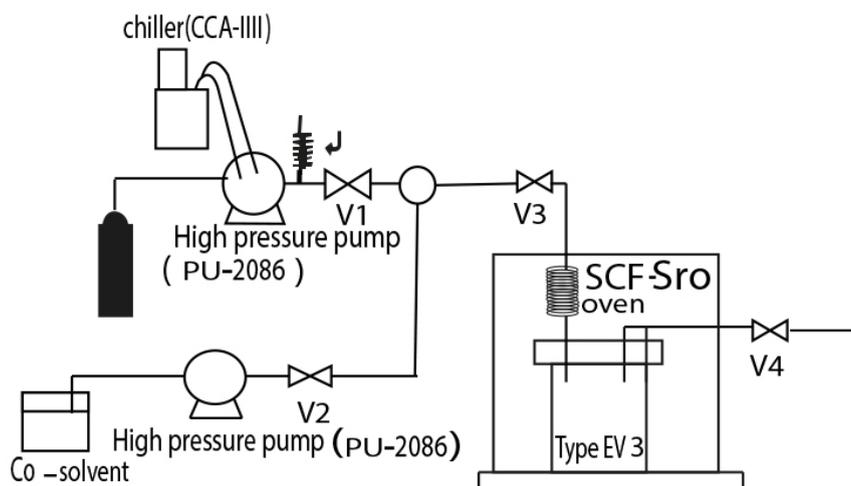


Figure 2. Supercritical CO₂ apparatus.

2.3. Procedures

2.3.1. ScCO₂ Dyeing

Polyamide 6 fabric (usually 3 × 10 cm) was wrapped around a stainless steel cylinder coil bearing perforated holes (0.5 cm diameter) and seated inside the autoclave. The purified dye was loaded on the base of the surface of the cylinder, and the amount of dye used varied from 2% to 6% owf. The autoclave was then sealed and heated to the desired temperature. At the same time, CO₂ was pumped through into the vessel and kept at a working pressure by stirring. The head temperature of the pump was maintained at −5 °C using a chiller. The circulation system was activated as the pressure reaches 10 MPa. The stream of the fluid was introduced using the magnetic drive under the column at 750 rpm. The fluid flowed from the inside to the outside of the cylinder. After a definite reaction time (1 and 3 h), the CO₂ released by shutting off the valve slowly until the pressure of the dyeing vessel reached atmospheric pressure. After dyeing, the fiber was removed, soaped at a temperature of 60 °C for 15 min, and then rinsed with water.

2.3.2. Aqueous Dyeing

As shown in Figure 3, the dye bath (1:20 liquor ratio) containing 5 g/dm³ carrier, 4% ammonium sulphate was adjusted to pH 5.5 and brought to 60 °C. The polyamide 6 fabric was added at this temperature and run for 15 min. 2.0% to 6.0% (owf.) of the dyes under our study were dissolved in a solution of (2 g/dm³), an anionic dispersing agent, followed by the dye being precipitated in a fine dispersion. The fine dispersion was then added, the temperature was raised to the boiling point over a period of 45 min, and dyeing was continued at the boiling point for about one hour. After dyeing, the samples were soaped with a detergent and some NaOH in a bath containing 2% nonionic detergent at a temperature of 60 °C for 15 min, then rinsed in water and dried at room temperature.

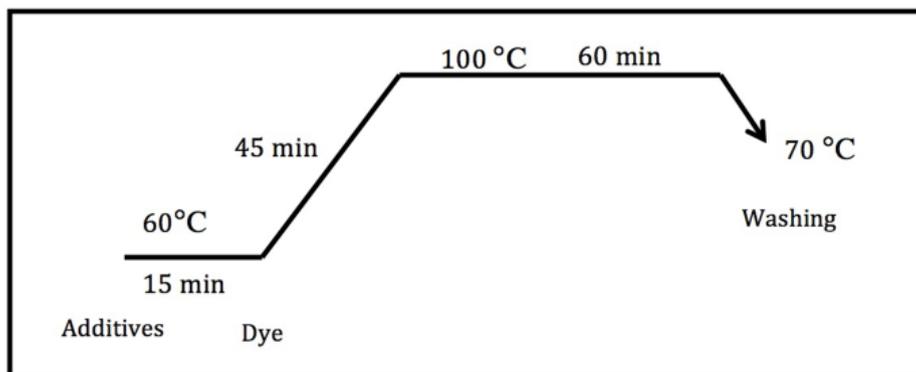


Figure 3. Exhaustion dyeing curve.

2.4. Measurements

Color strength (K/S) values of the dyed polyamide 6 fabrics were evaluated using the (Konica Minolta spectrophotometer CM-3600 d) spectrophotometer (Minolta, Tokyo, Japan).

Fastness properties, mainly washing, rubbing, and light fastness, of the dyed polyamide 6 fabrics were evaluated according to JIS L 0844, JIS L 0849, and JIS L 0842:2004 test methods, respectively [32–34].

The color parameters of the dyed polyamide fabric were measured using the (Konica Minolta spectrophotometer CM-3600 d) spectrophotometer. The following CIELAB coordinates were measured: lightness (L^*), chroma (C^*), hue (h), the degree of redness (+ve) and greenness (–ve) (a^*), and the degree of yellowness (+ve) and blueness (–ve) (b^*).

The antibacterial activity assessment on G+ve bacteria (*Staphylococcus aureus* and *Bacillus subtilis*) and G–ve bacteria (*Escherichia coli* and *Pseudomonas aeruginosa*) was conducted qualitatively according to the AATCC Test Method (147-1988) and expressed as zone of growth inhibition ZI (mm).

2.5. Statistical Analysis

All tests have been performed by taking the average of three sample readings. The standard error of the mean was calculated according to the equation given below and found to be ± 0.1 .

$$SE_{\bar{x}} = \frac{S}{\sqrt{n}} \quad (1)$$

where S = sample standard deviation, n = number of observations of the sample.

3. Results and Discussion

The main task of the current work is to introduce a one-step procedure for producing polyamide 6 fabric with antimicrobial functionality under supercritical carbon dioxide medium. The effect of dyeing parameters such as dye type, concentration, temperature, time and pressure as well as a comparison of the supercritical dyeing method with traditional aqueous dyeing have been investigated. The results obtained, along with appropriate discussion, are presented below.

3.1. Dyeing Properties of Hydazonopropanenitrile Dyes

3.1.1. Effect of Dye Concentration

Figure 4 shows that at the same dyeing conditions (120 °C, 15 MPa, 60 min), the color strength of nylon 6 fabric increased by increasing the dye concentration from 2% to 6% owf., but the increment in color depth becomes smaller when the concentration surpasses 4% for dyes number 2, 4 and 5. This may be attributed to the fact that these dyes have strong saturation at low concentration.

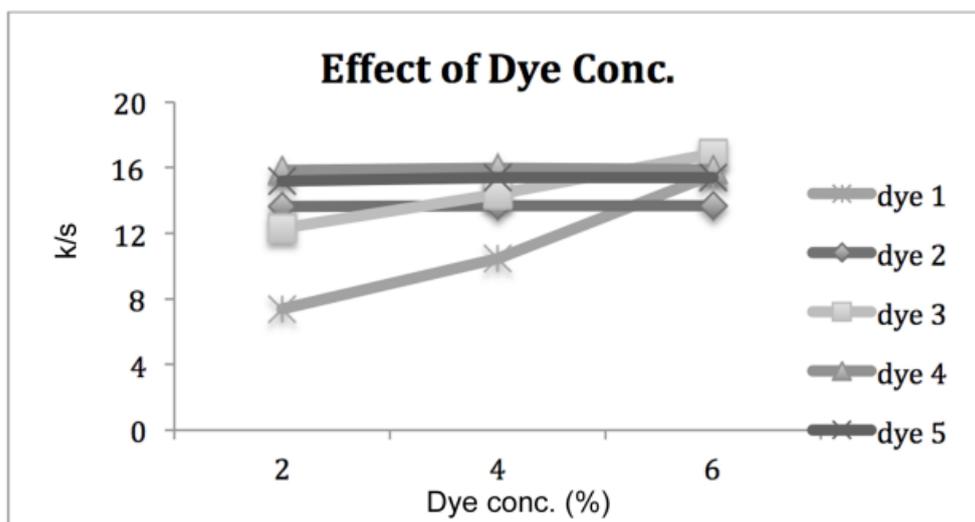


Figure 4. Effect of dye concentration.

A comparison of color strength of the *scCO*₂ and the aqueous dyed fabrics is shown in Figure 5. It was indicated that, without adding salt, carrier or dispersing agent, the appreciable color strength (K/S) of the samples dyed in *scCO*₂ was superior to those dyed in water. It can be observed that in Figure 5a, the sample dyed with dye 1, 2 and 3 in *scCO*₂ with 2% conc. has a higher K/S value than samples dyed in water with 6% dye conc. (Figure 5c). This may be attributed to the fact that dye uptake was improved by a large margin when using *scCO*₂ as a dyeing medium. For dyes 4 and 5, both have higher K/S values in supercritical conditions than in water conditions, but with a smaller border than those detected in dyes 1, 2 and 3. This means dyeing in a *scCO*₂ system exhibited significant advantages compared with traditional water dyeing. The exclusive dyeing procedure of the *scCO*₂ system is the main reason [35].

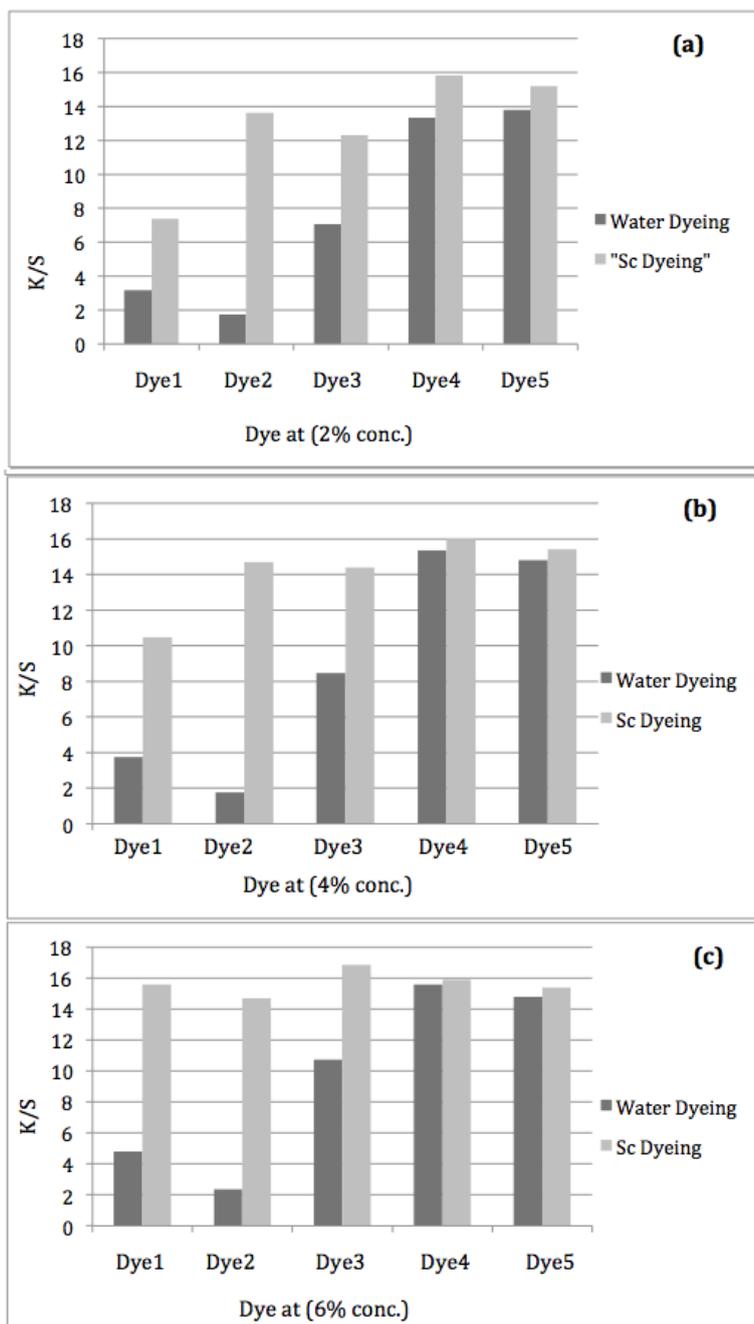


Figure 5. Comparison of color strength of *scCO*₂ and the aqueous dyeing (a) at 2% dye conc; (b) at 4% dye conc; (c) at 6% dye conc.

3.1.2. Effect of Dyeing Temperature

As shown in Figure 6a, the dye adsorption or uptake, characterized as color strength of K/S, remarkably increased with increasing system temperature, especially for temperatures higher than 100 °C. The significant effect of temperature could be explained by the fact that, higher system temperatures lead to higher activities of the molecules of dyestuff and supercritical carbon dioxide fluid, as well as an increase in flexibility of the nylon polymer chains. The rubbery and amorphous regions of the polymer were increased compared to the harder and more brittle areas, resulting in greater permeability of the dye molecules [4,36].

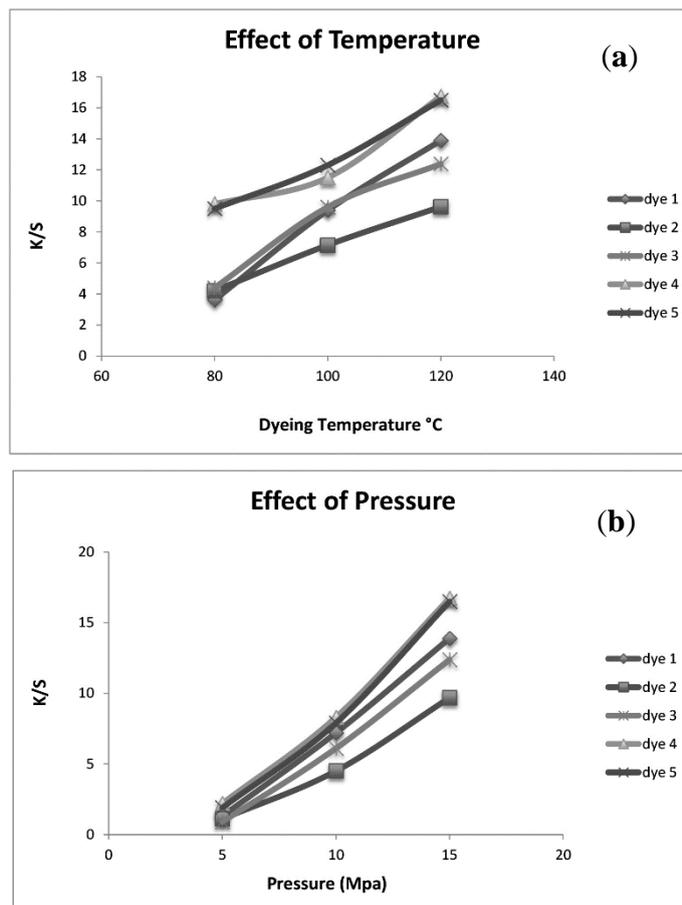


Figure 6. (a) Effect of dyeing temperature; (b) Effect of dyeing pressure.

3.1.3. Effect of Dyeing Pressure

As shown in Figure 6b, the dye uptake expressed as the color strength K/S was remarkably improved with an increasing system pressure. This behavior could be made clear by the fact that increasing system pressure led to an increase in the density of supercritical carbon dioxide fluid, which consequently increased its solvent power. Hence the dyes could be readily dissolved, as well as enhancing swelling of the nylon fibers in the supercritical dyeing medium, resulting in a higher dye adsorption and enhancement in color strength value [36].

3.1.4. Effect of Dyeing Time

The relationship between dye adsorption and dyeing time (1h and 3h) in $scCO_2$ is demonstrated in Figure 7a–c. It is seen that dye uptake expressed as color strength (K/S) increased with increasing dyeing time, at all dye concentrations (2%, 4% and 6%). The improvement in K/S values reflected the positive impact of increasing dyeing time which led to an adequate and uniform adsorption of the dye by the fibers, as well as uniform penetration and diffusion of the dye into the fabric, which resulted in the enhancement of the uptake of the dye into the fabric [36,37]. However, this result was not consistent with all dyes, since the K/S value of the fabric dyed with dye 2 for 1 h is higher than that of the fabric dyed with dye 2 for 3 h as shown in Figure 7a. This may be attributed to the decomposition of the dye with prolonged heating leading to a lower K/S value.

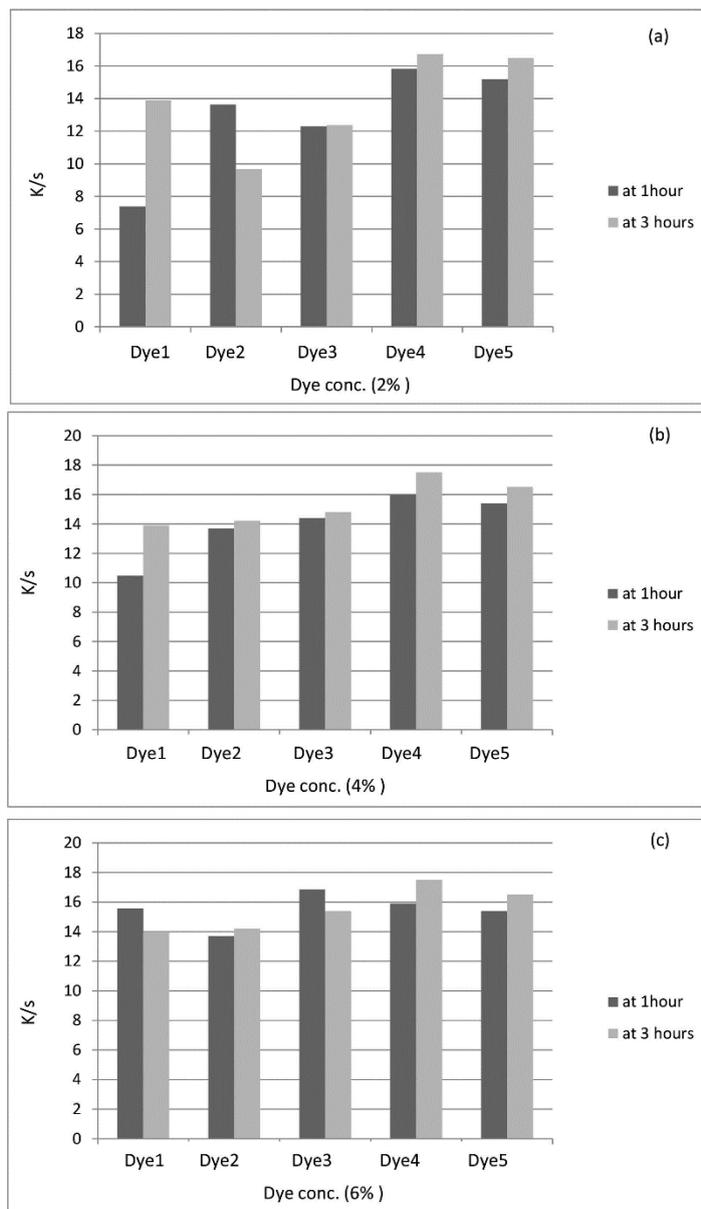


Figure 7. (a) Effect of dyeing time at 2% dye conc; (b) at 4% dye conc; (c) at 6% dye conc.

3.2. Color Fastness

The color fastness of the dyed nylon 6 fabrics with the proposed dyes was evaluated and recorded in Table 1. The washing fastness rating of the dyed nylon 6 fabric under supercritical medium was excellent for both fading and staining with ratings ranging from 4 to 5, while those of exhaustion dyeing were excellent for staining and ranged from moderate to excellent for fading (2–5). This result is probably due the high affinity of the colored hydrazonopropanenitrile dyes for nylon 6 fibers. Furthermore, all the dyed fabrics presented very good rubbing fastness, indicating good diffusion and penetration of the dyes under our study into fiber substrates. Nevertheless, the poor light fastness of all the water dyed fabrics was noticed. On the contrary, the dyed nylon 6 fabrics under supercritical conditions had relatively excellent light fastness (rating 4–5), which should be attributed to its higher level of dye molecule aggregation and superior depth of shade.

Table 1. Fastness properties of dyed nylon 6 samples.

Dye	ScCO ₂ Dyeing				Aqueous Dyeing			
	Rubbing Fastness	Washing Fastness		Light Fastness	Rubbing Fastness	Washing Fastness		Light Fastness
		Color Change	Staining			Color Change	Staining	
1	5	5	5	4	5	5	5	1-2
2	4-5	5	5	4	5	5	5	1
3	4-5	5	5	4-5	2-3	5	5	1
4	4-5	4-5	5	4-5	3-4	2-3	5	1
5	5	4-5	5	5	4	2-3	4	1-2

Table 2. Antibacterial activity of dyed samples.

Dye	ZI of the Dyed Nylon in scCO ₂				ZI of the Dyed Nylon in Water			
	G-ve		G+ve		G-ve		G+ve	
	<i>Pseudomonas Aeruginosa</i>	<i>Escherichia coli</i>	<i>Bacillus subtilis</i>	<i>Staphylococcus Aureus</i>	<i>Pseudomonas aeruginosa</i>	<i>Escherichia coli</i>	<i>Bacillus subtilis</i>	<i>Staphylococcus S. aureus</i>
1	12	13	13	13	12	13	12	13
2	13	12	14	13	12	13	14	13
3	14	13	14	13	13	13	13	13
4	13	12	14	13	12	12	12	13
5	12	11	12	11	11	11	12	11

ZI: zone of inhibition.

3.3. Antimicrobial Activity

The antimicrobial activities of the nylon fabric samples dyed in both supercritical and water media were screened using an agar-well diffusion technique against four different microbial cultures. The antimicrobial results (Table 2) attained were found to be fairly good owing to the presence of potentially active function groups, e.g., chloro, cyano and antipyrine moiety in the structure of dyes [38]. The results can be interpreted in terms of nonspecific action, *i.e.*, antibacterial activity can be achieved either by causing damage to bacterial cells or by means of restriction of a specific bacterial target [39]. The dyeing technique has practically no effect on the imparted antimicrobial properties.

3.4. Color Assessment

The color of the supercritical dyed nylon 6 fabrics was evaluated using the CIELAB system in terms of L^* , a^* , and b^* (Table 3). The color coordinates recorded in Table 3 indicated that the dye has good affinity for nylon 6 fabric and favored the following characteristics:

The dyes in our study displayed good affinity for nylon 6 fabrics at the given temperature and present generally bright and deep hues ranging from yellow to orange.

The color hues of the dyes on nylon 6 fabrics were shifted towards the yellowish direction on the yellow-blue axis according to the positive values of b^* .

The color hues of the dyes on nylon 6 fabric were shifted towards the greenish direction on the red-green axis as indicated from the negative value of a^* .

Table 3. Color coordinates of the dyed nylon samples.

Dye	L^*	C^*	H	a^*	b^*
1	88.11	79.1	97.84	-10.79	78.36
2	90.75	90.34	100.11	-12.4	88.94
3	92.12	76.33	102.71	-16.79	74.46
4	90.04	95.11	99.76	-14.31	83.16
5	92.73	81.86	102.51	-17.69	79.75

4. Conclusions

The conventional dyeing process using water as a solvent has drawbacks. Different agents have to be added for treatment of hydrophobic material; after dyeing, a consequent drying process with high energy consumption is imperative; and a large amount of wastewater is used. In contrast, dyeing with $scCO_2$ is water-free. The results confirm that hydrazonopropanenitrile azo dyes are convenient for dyeing nylon6 fabrics in $scCO_2$ and help expand the application of supercritical technology. The innovative supercritical model was designed for cleaner production of antimicrobial nylon6 fabrics.

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Author Contributions

Satoko Okubayashi and Tarek Abou Elmaaty designed experiments; Fathy El-Taweel synthesized the dyes; Eman Abd El-Aziz and Jaehuyk Ma performed the experiments; Tarek Abou Elmaaty and Eman Abd El-Aziz wrote the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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