

## ACCEPTED MANUSCRIPT

This is an early electronic version of an as-received manuscript that has been accepted for publication in the Journal of the Serbian Chemical Society but has not yet been subjected to the editing process and publishing procedure applied by the JSCS Editorial Office.

Please cite this article as M. M. Nikodijević, D. Z. Troter, S. T. Stojanović, M. T. Stojanović Krasić, and S. S. Konstantinović, *J. Serb. Chem. Soc.* (2026) <https://doi.org/10.2298/JSC260413033N>

This “raw” version of the manuscript is being provided to the authors and readers for their technical service. It must be stressed that the manuscript still has to be subjected to copyediting, typesetting, English grammar and syntax corrections, professional editing and authors’ review of the galley proof before it is published in its final form. Please note that during these publishing processes, many errors may emerge which could affect the final content of the manuscript and all legal disclaimers applied according to the policies of the Journal.





*J. Serb. Chem. Soc.* **00(0)** 1-10 (2026)  
JSCS-13906

## Dyeing of polyester fabric with disperse dye in a natural deep eutectic solvent as and eco-friendly medium

MILENA M. NIKODIJEVIĆ<sup>1\*</sup>, DRAGAN Z. TROTTER<sup>1</sup>, SANJA T. STOJANOVIĆ<sup>2</sup>, MARIJA T. STOJANOVIĆ KRASIĆ<sup>2</sup>, AND SANDRA S. KONSTANTINOVIĆ<sup>1</sup>

<sup>1</sup>University of Niš, Faculty of Technology, Bulevar Oslobođenja 124, 16000 Leskovac, Serbia,

<sup>2</sup>University of Niš, Faculty of Medicine, Blvd. Dr Zorana Đinđića 81, 18000 Niš, Serbia.

(Received 13 April; revised 12 May; accepted 19 June 2026)

**Abstract:** The present study proposed a sustainable, cleaner, alternative, safer and environmentally friendly method for dyeing polyester fabric using a natural deep eutectic solvent as a dyeing medium. Besides enabling water conservation and reducing wastewater generation, the process also successfully eliminated the use of toxic and environmentally harmful auxiliaries. Dyeing performance, measured by CIELab and K/S values, was compared to conventional dyeing. The NADES dyeing method achieved higher K/S values and eliminated the need for additional auxiliaries, such as levelling and dispersant agents. The polyester fabric structure remains intact, as confirmed by physical property results. SEM analysis shows that the surface morphology of all polyester samples is smooth and uniform with no visible changes, indicating the dyeing process did not affect the fiber surface. Fastness ratings are consistent across all dyed polyester samples.

**Keywords:** betaine; deep eutectic solvent; dyeing; polyester; sustainability.

### INTRODUCTION

Polyester is the most hydrophobic synthetic fiber, with a compact crystalline structure and desirable qualities such as affordability, availability, softness, drapability, comfort, and a lightweight feel against the skin. As a result, it is widely used in both wearable and decorative textiles and garments. However, its high hydrophobicity presents challenges in the common industrial dyeing process with disperse dyes in aqueous solutions, which requires high temperatures, pressure, and a high fabric-to-solvent ratio to achieve optimal dyeing. As a result, residual disperse dyes and auxiliary chemicals remain in the water, producing large volumes of wastewater with high chemical and biochemical oxygen demand.<sup>1-3</sup>

\* Corresponding author. E-mail: [nikmilena94@gmail.com](mailto:nikmilena94@gmail.com)  
<https://doi.org/10.2298/JSC260413033N>

An alternative dyeing technique uses a specific class of solvents called deep eutectic solvents (DESs) as non-aqueous dyeing media. DESs are binary or ternary mixtures made from Lewis or Brønsted acids and bases. One component acts as a hydrogen bond donor (HBD), and the other as a hydrogen bond acceptor (HBA). When combined in a specific molar ratio, these components form a network of hydrogen bonds, leading to a significant reduction in the system's lattice energy. This leads to a substantial or, as commonly termed, "deep" - decrease in melting point, much lower than that of the pure components. If these components are naturally derived, the solvents are classified as natural deep eutectic solvents (NADESs).<sup>4-8</sup>

NADESs represent the latest generation of "green" solvents prepared from natural, cheap, biodegradable, non-toxic, environmentally friendly, and safe components, making them suitable for designing, tailoring, and modifying different technological processes. NADESs application in food, cosmetics, and drug-related products is even declared acceptable or even more desirable in comparison to some conventionally used organic solvents (especially volatile, toxic, expensive, and strictly regulated).<sup>4-8</sup>

Choline chloride, a quaternary ammonium salt acting as a hydrogen bond acceptor (HBA), is characterized by its low cost, biodegradability, non-toxicity, and exceptional ability to form intermolecular interactions with various organic and inorganic compounds.

However, it is produced synthetically and is only available commercially as a synthetic product. Additionally, its strong hygroscopic nature poses challenges in handling and storage, often requiring the use of desiccants under certain conditions.

In recent years, betaine has emerged as an excellent, sustainable substitute for choline chloride. It is an inexpensive, non-toxic, biodegradable, and renewable organic salt that exhibits high resistance to humidity. Commercially available betaine is bio-based, typically sourced as a by-product from sugar refining.<sup>9-12</sup> Glycerol and urea were selected as hydrogen bond donors, since glycerol is a non-toxic, recyclable, and renewable alcohol widely used in cosmetics and pharmaceuticals, while urea is a safe, cheap, and readily available compound.<sup>7,8</sup>

The main reason for selecting this specific ternary (three-component) system over more common binary mixtures lies in its synergistic effects. According to the literature, the formulation of ternary eutectic solvents by incorporating both an alcohol (glycerol) and an amide (urea) disrupts the crystalline lattice more effectively, which significantly lowers the viscosity and melting point of the solvent compared to binary alternatives like betaine-urea.<sup>13,14</sup> Also, our previous work showed that this exact ternary system is highly effective for polyester fabric dyeing, achieving enhanced dye solubility and excellent kinetic penetration into the hydrophobic matrix of polyester fibers.<sup>15</sup> Therefore, building directly upon our

established findings, this study aimed to further explore and optimize the process parameters for this specific green solvent system.

Thus, the aims and novelties of the current study are:

- Developing a novel, greener, and more sustainable technique for dyeing polyester fabric with Disperse Red 60 using NADES - an eco-friendly liquid medium prepared from betaine hydrochloride, glycerol, and urea;
- Characterizing the dyed fabric through color strength determination (K/S values), thermogravimetric analysis, and surface morphology testing.

By incorporating NADES into the dyeing process, two main goals were achieved simultaneously:

- eliminating the need for toxic and environmentally harmful dyeing auxiliaries such as dispersants and carriers,
- reducing water consumption and the volume of wastewater generated.

#### EXPERIMENTAL

##### *Materials and chemicals*

Raw, unprocessed 100% polyester fabric (surface mass  $75 \text{ g}\cdot\text{m}^{-2}$ ) was kindly donated by Yumco (Vranje, Republic of Serbia). C.I. Disperse Red 60 dye (chemical structure shown in Fig. 1) was purchased from Hoechst (Frankfurt, Germany). NADES components included betaine hydrochloride (99%, Tokyo Chemical Industry, Tokyo, Japan; chemical structure in Fig. 1), glycerol (Ph Eur grade, Meilab, Belgrade, Serbia), and urea (99%, Centrohem, Stara Pazova, Serbia). Other dyeing chemicals were glacial acetic acid (Zorka Pharma-Hemija DOO, Šabac, Serbia), distilled water, leveling agent Kollasol Lok, and dispersing agent CHT-Dispergator XHT-S (both from CHT GmbH, Tübingen, Germany). Sodium hydroxide and sodium hydrosulfite were obtained from Centrohem (Stara Pazova, Serbia), and non-ionic detergent Sarabid DLO from CHT GmbH (Tübingen, Germany).

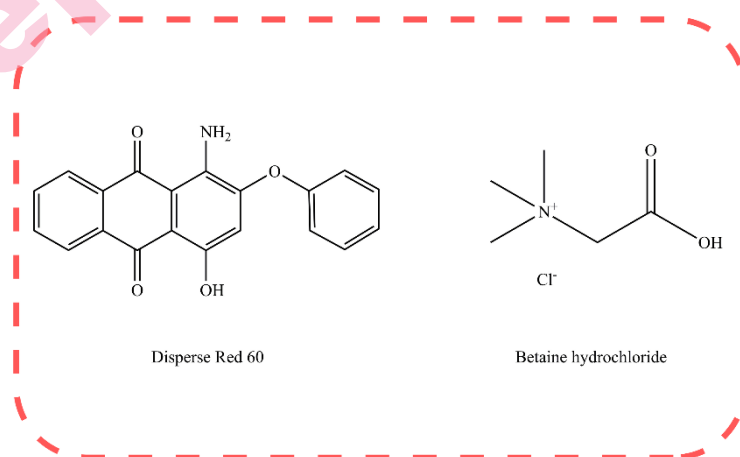


Fig.1. Chemical structures of Disperse Red 60 and betaine hydrochloride.

*Preparation of NADES*

An Erlenmeyer flask containing equimolar amounts of betaine hydrochloride, glycerol, and urea was sealed with a glass stopper and heated to 40 °C while stirring continuously at 1100 rpm. After 20 minutes, a homogeneous, stable, clear, and transparent liquid formed. The prepared NADES solution was kept sealed and stored in a desiccator with CaCl<sub>2</sub> until further use.

*Pre-dyeing processing*

Polyester fabric was washed in a 2 g·L<sup>-1</sup> solution of saturated detergent Sarabid LDR (CHT GmbH, Tübingen, Germany) for 30 minutes at 60 °C with a 1:30 fabric-to-liquor ratio. After washing, the fabric was air-dried at room temperature for 24 h and ironed. It was then cut into 5×5 cm squares using a surgical scalpel.

*Dyeing*

Conventional dyeing method in Ahiba Nuance Top Speed dyeing machine (Allstates Textile Machinery Inc., Williamston, South Carolina, United States): 0.6% acetic acid, 1% leveling agent, 1% dispersing agent, temperature of 120 °C, duration of 110 min, bath ratio of 1:50, pH value of 4, and different dye concentrations (1, 2, 3, 4, and 5% w/w).

NADES dyeing method: NADES without any dyeing auxiliaries, temperature of 100 °C, duration of 60 min, pH value of 4, and different dye concentrations (1, 2, 3, 4, and 5% w/w).

*Post-dyeing processing*

After dyeing, the polyester fabrics were subjected to reductive cleaning using 2 g·L<sup>-1</sup> sodium hydroxide and 2 g·L<sup>-1</sup> sodium hydrosulfite at 60 °C for 15 minutes, followed by rinsing with cold water and drying in a laboratory oven at 100 °C for 10 minutes.

*Evaluation of color strength of dyed polyester fabric*

The determination of the color strength of dyed polyester fabric was carried out by K/S (Kubelka-Munk function) values using Daticolor Spectraflash SF600Ks (New York, USA). K/S values were calculated using the equation.<sup>16</sup>

$$K/S = (1-R)^2/2R \quad (1)$$

where *K*, *S*, and *R* denote the adsorption coefficient, the scattering coefficient, and the reflectance at complete opacity, respectively. Generally, the higher the K/S value, the higher the dye uptake on fabric, *i.e.*, the deeper the color on the fabric.

*Surface morphology test*

Scanning electron microscopy (SEM) was used to analyze the morphology of undyed, conventionally dyed, and NADES-assisted dyed samples. The samples were sprayed with an alloy of gold and palladium (85%/15%) under vacuum in a Fine Coat JEOL JFC-1100 Ion Sputter (JEOL Ltd., Tokyo, Japan). The metalized samples were scanned using a JEOL Scanning Electron Microscope JSM-5300 (JEOL Ltd., Tokyo, Japan) under a magnification of 750 times, a voltage of 20 kV, and a vacuum of 1.33·10<sup>-5</sup> Pa.

*Color fastness of dyed polyester fabric*

Adequate procedures provided by the specific ISO methods were employed for the determination of dyed fabrics' color performance properties. The wash, light, and rub fastness of dyed polyester fabric were measured by ISO 105-C06: 2010, ISO 105-B02: 2014 (en), and ISO 105-X12: 2016 protocols.<sup>17</sup> The fastness of dyed polyester fabric was evaluated at standard

depths of 1:1 and 2:1. The dyed sample specimen was subjected to a sublimation test according to the ISO 105-POI 1993 standard.<sup>18</sup>

#### *Physical properties of polyester*

Physical property (tensile strength and elongation at break) measurements of undyed, conventionally dyed, and NADES-assisted dyed samples were performed on a MesdanLab Strength Tester dynamometer, in accordance with the standard ISO 13934-1:2013.<sup>19</sup>

### RESULTS AND DISCUSSION

#### *Comparison of color strength and CIELab parameters of polyester fabric dyed by conventional and NADES-assisted processes*

The K/S color strength values of conventionally and NADES-assisted dyed polyester samples in the range of  $\lambda_{\max} = 400\text{-}700$  nm are presented in Fig.2. The maximum wavelength for the used disperse dye was  $\lambda_{\max} = 524$  nm. Higher K/S values are noticeable for the samples dyed by the NADES-including dyeing technique. Moreover, the highest K/S values were observed at the highest dye concentration of 5% for both dyeing procedures.

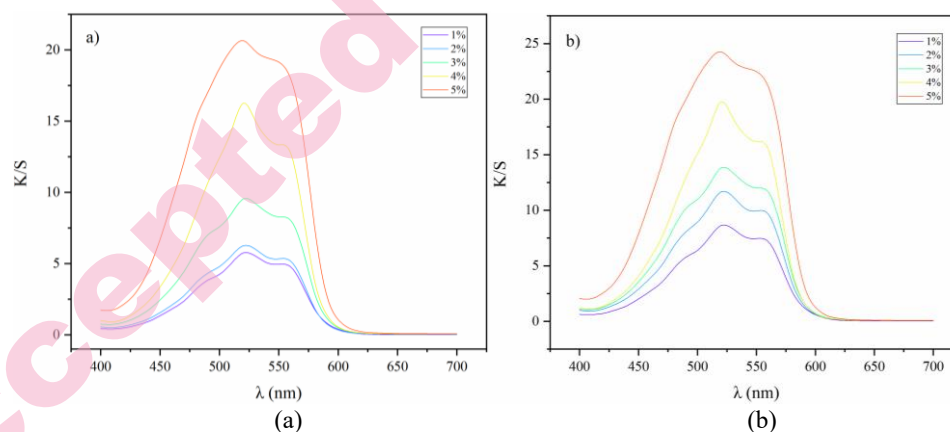


Fig.2. Comparison of the K/S values obtained for conventionally (a) and NADES (b) dyed polyester fabrics at different dye concentrations.

CIELab parameters for conventionally and NADES-assisted dyed polyester samples are presented in Table 1.

TABLE I. CIELab coordinates of undyed, conventionally dyed, and NADES dyed polyester fabrics at different dye concentrations.

Dye conc. (%)	Type of sample	L*	a*	b*	C*	H*
-	Undyed	91.23	0.73	2.02	2.14	70.11
1	Conventionally dyed	61.85	45.71	3.58	45.85	355.52
	NADES-assisted dyed	47.94	58.58	6.56	58.66	356.98
2	Conventionally dyed	52.60	57.71	10.65	57.71	359.92
	NADES-assisted dyed	45.63	63.80	12.65	60.61	360.01
3	Conventionally dyed	50.26	57.78	11.52	57.80	360.23
	NADES-assisted dyed	44.45	60.33	13.52	64.03	362.25
4	Conventionally dyed	46.49	62.42	13.52	62.64	361.63
	NADES-assisted dyed	42.98	63.27	15.58	65.53	362.03
5	Conventionally dyed	45.65	63.42	15.23	64.52	362.52
	NADES-assisted dyed	40.19	65.72	17.08	65.02	362.75

The highest  $L^*$  parameter values are noticeable for the undyed sample, while the lowest are observed for the NADES-assisted dyed fabric with the maximum dye concentration (5%), strongly indicating both a higher dye adsorption and darker achieved shades. Positive  $a^*$  parameter values for all dyed samples indicate redness. Positive  $b^*$  parameter values in the raw sample and all dyed samples indicate redness.  $C^*$  values are higher in all dyed samples compared to the undyed samples, describing the distance from the black-white axis and good brilliance. Parameter  $H^*$  has the lowest values for the undyed sample, while the values for the dyed samples are similar.

These results can be explained by various factors. NADESs possess the potential for softening or partially dissolving various polymer-type matrices, thus facilitating dye penetration. Due to improved dye penetration, the fabric becomes darker. Therefore, careful choice of solvent and its adjustment according to the desired properties greatly affect the efficiency of the dyeing process.<sup>20</sup>

As shown in Fig. 3, a visual comparison of samples at a 5% dye concentration demonstrates that the introduction of NADES significantly enhances dyeing efficiency. This is supported by the highest K/S value and the visibly darker shade obtained for the NADES-assisted sample compared to the conventional one.

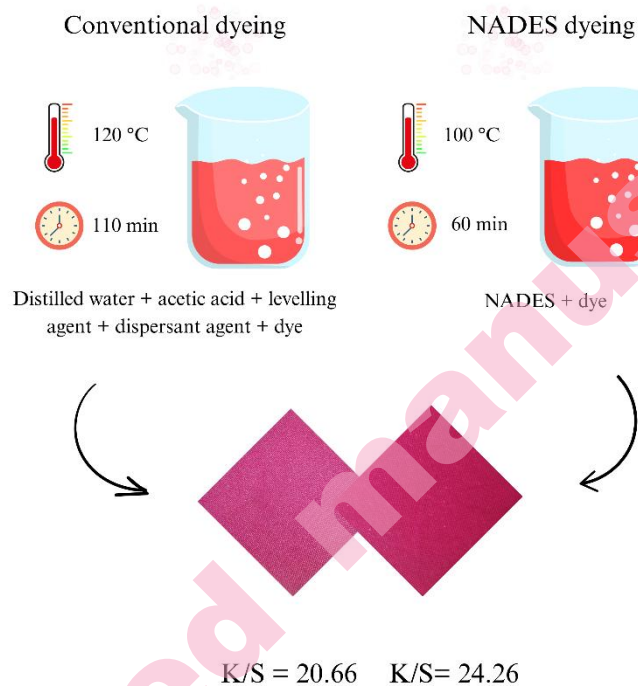


Fig.3. Comparison between conventional and NADES-assisted dyeing at the maximum dye concentration.

*SEM analysis/morphological properties of the polyester fabrics*

SEM images for the undyed, conventionally dyed, and NADES-assisted dyed samples are shown in Fig.4. Smooth surface morphology is noticeable, without any evident visual changes among all polyester fabrics.

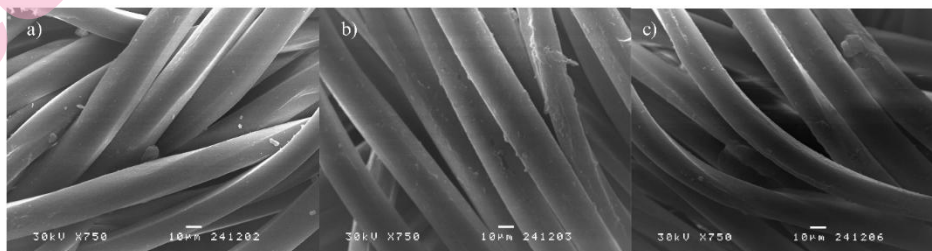


Fig.4. SEM images of a) undyed, b) conventionally dyed and c) NADES dyed polyester fabrics.

*Color fastness properties (washing, light, rubbing and sublimation fastness) of dyed polyester*

Color fastness property ratings of conventional and NADES-assisted dyed samples are displayed in Table II. It is noticeable that these values are similar for both dyeing processes, indicating there is no deterioration occurring when NADES-assisted dyeing is applied. Therefore, no dye residues were left on the polyester surface post-dyeing.

Table II. Wash, light, sublimation and rubbing fastness for the conventionally dyed, and NADES dyed polyester fabrics.

Type of sample	Wash fastness		Wet rubbing fastness		Dry rubbing fastness		Light fastness	Sublimation fastness
	Change	Staining	Change	Staining	Change	Staining		
Conventionally dyed	4-5	4-5	4	4	4-5	4-5	5-6	4-5
NADES dyed	4-5	4-5	4	4	4-5	4-5	5-6	4-5

*Mechanical-physical properties of polyester*

In order to provide a deeper understanding regarding the influence of NADES-containing dyeing solution on the strength of the polyester fabric, tensile strength and elongation at break measurements were conducted; results are presented in Table III. It is noticeable that the tensile strength values are the highest in the undyed sample compared to the conventionally dyed and NADES dyed samples. Conventional and NADES dyed samples have similar tensile strength values, the warp and weft directions. The breaking elongation values are the highest for the undyed sample, although similar values were obtained for the conventionally dyed and NADES dyed polyester fabric in the warp and weft direction.

TABLE III. Tensile strength and elongation at break values for undyed, conventionally dyed, and NADES-dyed polyester fabrics.

Type of sample	Tensile strength (Kgf)		Elongation at break (%)	
	warp	weft	warp	weft
Undyed	85.04	50.17	42.5	28.1
Conventionally dyed	84.84	49.86	41.4	27.7
NADES-assisted dyed	84.64	49.66	40.2	27.1

## CONCLUSIONS

A sustainable, eco-friendly technique for dyeing polyester fabrics with Disperse Red 60 dye in the presence of betaine-based NADES is proposed. In the described process, NADES serves several roles: as a “greener” dyeing medium, dispersing agent, and homogenization agent for the dyeing bath. The incorporation of NADES in the process resulted in obtaining stable dyed fabric with higher *K/S* values and improved fastness properties, simultaneously excluding the need for

adding a carrier or dispersant into the dyeing solution. Moreover, the structure of the polyester fabric was not degraded, which is confirmed by the results of the analysis of the physical properties of the fabric. Based on the SEM, all polyester fabric samples demonstrate a uniform and smooth surface morphology, without any noticeable visual alterations between the treated and untreated samples. The wash and light fastness of conventional and NADES-dyed polyester are almost similar, although a slight advantage is given to NADES-dyed polyester.

*Acknowledgements:* This work was supported by the Republic of Serbia - Ministry of Science, Technological Development and Innovation (Programs for financing scientific research: 451-03-33/2026-03/200133 and 451-03-34/2026-03/200133 assigned to the University of Niš, Faculty of Technology, Leskovac).

### ИЗВОД

#### БОЈЕЊЕ ПОЛИЕСТАРСКЕ ТКАНИНЕ ДИСПЕРЗНОМ БОЈОМ У ПРИРОДНОМ ДУБОКОМ ЕУТЕКТИЧКОМ РАСТВАРАЧУ КАО ЕКОЛОШКИ ПРИХВАТЉИВОМ МЕДИЈУМУ

МИЛЕНА М. НИКОДИЈЕВИЋ<sup>1</sup>, ДРАГАН З. ТРОТЕР<sup>1</sup>, САЊА Т. СТОЈАНОВИЋ<sup>2</sup>, МАРИЈА Т. СТОЈАНОВИЋ КРАСИЋ<sup>2</sup>, САНДРА С. КОНСТАНТИНОВИЋ<sup>1</sup>

<sup>1</sup>Универзитет у Нишу, Технолошки факултет, Булевар ослобођења 124, 16000 Лесковац и  
<sup>2</sup>Универзитет у Нишу, Медицински факултет, Булевар др Зорана Ђинђића 81, 18000 Ниш, Србија.

У раду је предложена одржива, чистија, алтернативна, безбеднија и еколошки прихватљива метода за бојење полиестарске тканине коришћењем природног дубоког еутектичког растварача као средства за бојење. Поред тога што омогућава уштеду воде и смањује стварање отпадних вода, процес је такође успешно елиминисао употребу токсичних и еколошки штетних помоћних средстава. Перформансе бојења, мерене CIELab и K/S вредностима, упоређене су са конвенционалним бојењем. NADES методом бојења постигнуте су веће K/S вредности и елиминисана је потреба за додатним помоћним средствима, као што су средство за разливање и средство за дисперговање. Структура полиестарске тканине остала је нетакнута, што потврђују резултати физичких својстава. SEM анализа показује да је површинска морфологија свих узорака полиестра глатка и уједначена без видљивих промена, што указује на то да процес бојења није утицао на површину влакана. Оцене постојаности су сличне код свих обојених узорака полиестра.

(Примљено 13. априла; ревидирано 12. маја; прихваћено 19. јуна 2026.)

### REFERENCES

1. A. M. Al-Etaibi, M. A. El-Asasery, *Polymers* **14** (2022) 1703 (<https://doi.org/10.3390/polym14091703>)
2. S. Eren, İ. Yiğit, K. Kutlay, Z. Kaya, C. Basrik, H. A. Eren, *Polymers* **16** (2024) 735 (<https://doi.org/10.3390/polym16060735>)
3. S. H. Ramugade, S. Nagaiyan, *J. Indian Chem. Soc.* **100** (2023) 100960 (<https://doi.org/10.1016/j.jics.2023.100960>)
4. K. A. Pishro, M. H. Gonzalez, *RSC Adv.* **14** (2024) 14480–14504 (<https://doi.org/10.1039/D4RA01610F>)

5. N. Hussain, S. Hussain, M. Mehdi, M. Khatri, S. Ullah, Z. Khatri L. V. Langenhove, I. S. Kim, *Polymers* **13** (2021) 2594 (<https://doi.org/10.3390/polym13162594>)
6. P. Kaur, N. Rajani, P. Kumawat, N. Singh, J. P. Kushwaha, *Colloids. Surf. A. Physicochem. Eng. Asp.* **539** (2018) 85–91 (<https://doi.org/10.1016/j.colsurfa.2017.12.013>)
7. D. Z. Troter, Z. B. Todorović, D. R. Đokić-Stojanović, B. S. Đorđević, V. M. Todorović, S. S. Konstantinović, V. B. Veljković, *J. Serb. Chem. Soc.* **82** (2017) 1039–1052 (<https://doi.org/10.2298/JSC170225065T>)
8. M. Zdanowicz, S. Paszkiewicz, M. El Fray, *Prog. Polym. Sci.* **161** (2025) 101930 (<https://doi.org/10.1016/j.progpolymsci.2025.101930>)
9. Q. Chen, N. He, J. Fan, F. Song, *Polymers* **14** (2022) 1783 (<https://doi.org/10.3390/polym14091783>)
10. A. Mero, S. Koutsoumpou, P. Giannios, I. Stavarakas, K. Moutzouris, A. Mezzetta, L. Guazzelli, *J. Mol. Liq.* **377** (2023) 121563 (<https://doi.org/10.1016/j.molliq.2023.121563>)
11. D. O. Abranches, L. P. Silva, M. A. Martins, S. P. Pinho, J. A. Coutinho, *Chem.Sus.Chem.* **13** (2020) 4916–4921 (<https://doi.org/10.1002/cssc.202001331>)
12. F. M. Fuad, M. M. Nadzir, *J. Mol. Liq.* **360** (2022) 119392 (<https://doi.org/10.1016/j.molliq.2022.119392>)
13. T. Sathasivam, T. Wu, U. A. loka Weerasinghe, P. Y. M. Yew, X. L. Quek, Y. J. Eng, D. Kai, *Bioresour Technol.* **442** (2026) 133753 (<https://doi.org/10.1016/j.biortech.2025.133753>)
14. M. C. Gutiérrez, M. L. Ferrer, C. R. M. F. del Monte, *Langmuir* **25** (2009) 5509 (<https://pubs.acs.org/doi/10.1021/la900552b>)
15. M. M. Nikodijević, D. Z. Troter, S. S. Konstantinović, *Adv. Technol.* **13** (2024) 45 (<https://doi.org/10.5937/savteh2402045N>)
16. S. S. Pawar, S. Maiti, S. Biranje, K. Kulkarni, R. V. Adivarekar, *Heliyon* **5** (2019) e01606 (<https://doi.org/10.1016/j.heliyon.2019.e01606>)
17. M. Hosseinezhad, K. Gharanjig, H. Imani, N. Razani, *Environ. Sci. Pollut. Res. Int.* **18** (2020) 7249 (<https://doi.org/10.1007/s11356-020-11041-2>)
18. ISO 105-PO1: 1993. Textiles - Tests for color fastness. Part PO1: Color fastness to sublimation, ISO; 1993. <https://www.iso.org/standard/3871.html> (accessed 31 March 2025).
19. ISO 13934-1:2013, 2013. Textiles - Tensile properties of fabrics, Part 1: Determination of maximum force and elongation at maximum force using the strip method. <https://www.iso.org/standard/60676.html> (accessed 31 March 2025).
20. M. Zheng, Y. Sun, C. Li, Y. Lu, Y. Dai, Z. Wang, *Color. Technol.* **139** (2023) 552–564 (<https://doi.org/10.1111/cote.12673>).