Received: 31 January 2023 | Revised: 24 March 2023 | Accepted: 30 March 2023

DOI: 10.1111/cote.12690

ORIGINAL ARTICLE

Coloration Technology

Supercritical carbon dioxide (SC-CO_2) dyeing of cellulose acetate: An opportunity for a "greener" circular textile economy

Peter J. Broadbent[1] | **Chris M. Carr**[1] | **David M. Lewis**[1] | **Muriel L. Rigout**[1] | **Ernst J. Siewers**[2] | **Narjes Shojai Kaveh**[2]

[1]School of Design, University of Leeds, Leeds, UK

[2]DyeCoo Textile Systems, Weesp, The Netherlands

Correspondence
Chris M. Carr, School of Design, University of Leeds, Leeds, UK.
Email: c.carr@leeds.ac.uk

Funding information
Innovate UK

Abstract

This article compares the dyeing of cellulose diacetate (cellulose-based) and polyester fabrics using supercritical carbon dioxide (SC-CO_2) and aqueous media. The benefits of dyeing in SC-CO_2 were clearly demonstrated in laboratory-based and pilot-scale studies in terms of increased colour strength, uniformity, fastness and the absence of auxiliaries such as dispersing agents or surfactants. In addition, the "super-levelling" nature of the SC-CO_2 medium was demonstrated in the reprocessing of polyester "waste textile" and the re-use of the "locked-in waste" colourant. The SC-CO_2 processing medium can be utilised to accurately colour "multiple life" polyester and cellulose acetate uniformly and to creatively tie-dye polyester and cellulose acetate fabrics. Through SC-CO_2 fluid technology, we can envisage a viable waterless circular manufacturing and recycling/remanufacturing framework for the predominantly polyester global fibre market coupled to the sustainably sourced, biodegradable cellulose diacetate as a replacement for cotton. The key technical and commercial advantages being the use of a single solvent dye class for both polyester and the cellulose diacetate, saving on energy costs, integrated simpler processing, reduced water usage and associated efficient recycling. Further, repositioning the cellulosic fibre industry towards using sustainable forests is attractive in terms of improved land, water and environmental management.

1 | INTRODUCTION

The production of the clothing globally doubled each year from 2000 to 2014 up to 100 billion garments per/year, the equivalent of 14 garments per person on the planet.[1] Further, apparel consumption is predicted to rise to 102 million tonnes in volume by 2030 (a 63% increase) with the associated value increasing from \$2.2T to \$3.3T.[2] However, despite this impressive manufacturing growth in the developed and emerging economies, the textiles industry still has significant problems with obvious challenges being the disposal of waste clothing derived from fast fashion and the relatively low recycling/remanufacturing of this waste into multiple lifetime products.[3–5] In addition, the water-intensive nature of textile production, the use of fertilisers in cotton agriculture and increasing competition for land all add to its perceived negative environmental impact.

Typically, dyeing and finishing of textiles requires high temperatures and account for up to 25% of the total energy used from fibre to fabric.[6,7] In addition, for cotton, the water consumed in processing may vary between 10,000 and 300,000 litres per 1000 kg of finished product,[8] coupled to estimates that up to ~1000–2000 tons of unfixed dyes were discharged in dyehouse effluent.[9] Further, the European Parliament[10,11] estimated that 20% of industrial water pollution comes from textile dyeing and that the total greenhouse emissions from textile production accounts for 10% of global emissions. Water has been identified by the World Bank as a key resource; recognising the extent of water pollution, assessing the magnitude of the impacts and formulating strategies to address pollution will be critical to improving public health, preserving ecosystems, and sustaining economic growth throughout the twenty-first century.[12]

With the textiles industry being identified as one of the most polluting sectors in the world, its manufacturing industry, their supply chains and global and regional retail brands are being challenged to reduce ecological impacts. This "stressed" economic and environmental framework is being further pressured by the current energy-cost crisis which is focusing the industry to establish new lower temperature, less polluting and more economic dyeing technologies. Within this context, the development of "waterless" textile dyeing, scouring and bleaching is regarded as significant since it offers a technology platform that completely revolutionises traditional textile "wet" processing.[13–18]

By increasing the temperature above 31°C and the pressure above 74 bar for carbon dioxide, a supercritical highly-compressed fluid phase is created.[19] The supercritical carbon dioxide (SC-CO_2) fluid possesses both the high diffusion properties of a gas, but also the solvation characteristics (ability to dissolve substances) of a liquid and easy movement of CO_2 as dye carrier through the fabric/yarn. Supercritical fluids are used in various industrial processes, such as, extraction, separation, concentration, purification, sterilisation, impregnation, enrichment, chemical reactions and drying.[20,21] For most solutes, the solvent power of SC-CO_2 is similar to that of light hydrocarbons, such as hexanes and pentanes, and it is this solubility that is an essential part of its application in dyeing and textile scouring/cleaning.

DyeCoo Textile Systems[22] have developed a patented and industrially-proven dyeing technology based on CO_2, instead of water, to dye polyester. The technology uses (reclaimed) heated and pressurised SC-CO_2 as the dyeing medium in a closed-loop process, with the dye dissolving in the SC-CO_2, exhausting into the polyester fibres and creating vibrant colours.

The technical advantages and impacts of SC-CO_2 dyeing for polyester are: zero water use in dyeing and therefore zero wastewater, zero processing chemicals (dispersing agents), less energy, less dyestuff, a high level of colour consistency across batches of textiles and reuse of 95% of the CO_2 within the process. In addition, on releasing the CO_2 pressure the fabric is left dry and no drying costs are incurred. Current DyeCoo commercial machines can process 800,000 kg of polyester per year, effectively saving 32 million litres of water and avoiding the use of 160,000 kg of chemicals. The total amount of CO_2 used would be five million kilogrammes (5000 metric tonnes), 95% of which (4750 metric tonnes) would be recycled back into the process.[23] Another benefit of the waterless dyeing technology is geographical "freedom" in that dyeing without water enables independence from clean water availability and creates new opportunities for the textile industry, allowing production to position closer to market, shorten lead times and disconnect from Earth's most valuable resource, water.

In the mid-1990s synthetic fibres overtook cotton as the predominant market fibres and this growth trajectory has continued, with polyester (polyethylene terephthalate) in 2020 dominating the global fibre market with 52% (57.1 million tonnes) of the total fibre usage.[24] Over the past 10 years, there has also been an increasing share of mechanically and chemically recycled polyester, enhancing the sustainability of the fibre. It is likely this trend will continue with increased sourcing of biobased and sustainable polyester. In contrast, cotton now only has 24% market share; however, the mindset of the retailers, consumers and statisticians is still focused on cotton as the "go to" fibre. To support cotton production, there have been initiatives focused on developing organic cotton and importantly "preferred" cotton sources which offer improved fibre yield, water usage, reduced fertiliser application and overall sustainability. In addition, while the level of cotton recycling is currently relatively low at ~1% of total cotton share, it is increasing, with an alternative "waste" management approach also established regenerating man-made cellulosic fibres such as Refibra, Ioncell, Infinna, SaxCell, and so forth, from the waste cellulosic feedstocks.

However, while fibre producers and academia look to create "new" man-made fibres from "waste" cellulosic feedstocks, there appears to be little consideration regarding the wider ongoing environmental challenge of water-based textile wet processing and coloration of cellulosic-based fibres and polyester/cellulosic blends. In this study we offer an alternative eco-friendly integrated pathway for coloration of man-made cellulose-based fibres that recognises the game-changing potential of SC-CO_2-based processing and the obvious, predominant market position of polyester fibre.

2 | EXPERIMENTAL

2.1 | Materials

The cellulose diacetate woven satin fabric, 100 g/m^2, was supplied by Whaley's, Bradford, UK and the polyester plain woven fabric, 191 g/m^2 was supplied by Denby Dale Clothing Ltd, Denby Dale, UK. The 100% Naia™ (cellulose diacetate) knitted jersey fabric, 98 g/m^2, and the 60/40 RPET (recycled polyester)/Naia blend knitted fabric, 105 g/m^2, were supplied by Eastman Chemical Company (USA) for the pilot scale studies.

The soaping agent Eriopon LAN, and the dispersing agent Matexil DA-AC (sodium polynaphthalene sulphonate) were supplied by Town End (Leeds) and Zeneca Colours, UK, respectively. Alizarin (97%) was purchased from Aldrich Chemicals, UK.

The Corangar Red PE-3469, Corangar Blue PE-3648, Corangar Blue PE-3618 and Corangar Yellow PE-3205 dyes were generously supplied by Colourtex, India, and are manufactured specifically for the SC-CO_2 dyeing range with no dispersing agents. The Corangar Red PE-3469, Blue PE-3648 and Yellow PE-3205 SC-CO_2 dyes were converted into the analogous aqueous disperse dyes (Leeds Disperse Dyes) at the University of Leeds, using the method described later, or were converted into the industrial disperse dyes at Colourtex India using traditional industrial dispersing agent/ball milling processing (Colourtex Disperse Dyes).

2.2 | Laboratory-scale SC-CO_2 dyeing of cellulose diacetate fabric with Corangar dyes

Cellulose diacetate woven fabric (10 g) was dyed with 0.5, 1 and 2% owf (on weight of fabric) Corangar Red PE-3469, Corangar Blue PE-3648 and Corangar Yellow PE-3205, respectively, in a DyeCoo Textiles Systems SC-CO_2 laboratory dyeing machine where each dyeing tube was filled with the woven cellulose diacetate fabric, the Corangar dye and CO_2 (212 g). The dyeing temperature was 85°C and the dyeing time was 1.5 h. After dyeing, the dyed cellulose acetate fabrics were immersed in the aqueous Eriopon LAN soaping agent solution (1% w/w) for 15 min at 100°C, with a liquor ratio of 20:1, in order to remove any unfixed dye. After the soaping process, the fabrics were then thoroughly rinsed in cold running water for 10 min and air dried.

2.3 | Conversion of SC-CO_2 Corangar dyes to analogous Leeds Disperse Dyes and laboratory-scale aqueous dyeing of cellulose diacetate fabric

The water-insoluble SC-CO_2 Corangar Red PE-3469, Corangar Blue PE-3648 and Corangar Yellow PE-3205 dyes were dissolved in acetone and drip-fed into an aqueous solution containing 10 g/L Matexil DA-AC dispersing agent/surfactant at 25°C. The dye/dispersing agent bath was vigorously stirred for 10 min and the 0.5, 1 and 2% owf dye dispersions (Leeds Disperse Dyes) were then used to dye the cellulose diacetate woven fabric at 85°C for 90 min. The fabric to liquor ratio was 1:10 and the dyebath pH was adjusted to 4 using a 0.1 M citric acid/0.2 M disodium hydrogen phosphate McIlvaine buffer (20 mL). After dyeing, the fabrics were soaped with Eriopon LAN solution (1% w/w) for 15 min at 100°C, with a liquor ratio of 20:1, then thoroughly rinsed in cold running water for 10 min and air dried.

The analogous commercially ball-milled manufactured disperse dyes (labelled Colourtex Disperse Dyes), kindly supplied by Colourtex India, were used as received and the woven cellulose diacetate was dyed in water as described earlier.

2.4 | Pilot scale SC-CO_2 dyeing of Naia™ (cellulose diacetate) fabrics and recycled polyester/Naia blend fabric with Corangar dyes

2 kg of Naia™ knitted fabrics were beam-dyed, 50 cm width, in the SC-CO_2 pilot machine at the DyeCoo Textile Systems facilities in the Netherlands. The SC-CO_2 dyeing was performed at 100°C with a pressure of 250 bar for 90 min using either 0.5% owf Corangar Red PE-3469, 0.7% owf Corangar Blue PE-3618 or with a mixture of Corangar dyes (1.54% owf in total) to produce a Black colour.

At the end of the dyeing process, the CO_2 was sent to a separator, where excess dye and any residues were removed by lowering the pressure. In this process, a minimum of 95% of CO_2 was recycled and recovered to the storage tank for reuse. The uniformly dyed fabric emerged from the vessel dry and ready to use, with no need of post-washing.

2.5 | Alizarin dyeing

Polyester fabric (10 g) was dyed with 2% owf alizarin, in the DyeCoo Textile Systems SC-CO_2 laboratory dyeing machine where each dyeing tube was filled with the polyester fabric, alizarin and CO_2 (147 g). The dyeing temperature was set as 120°C and the dyeing time was 1.5 h. After dyeing, the dyed polyester fabrics were rinsed with acetone in order to remove any surface-deposited dye and then finally air dried. Since alizarin is soluble in acetone, it is therefore a suitable solvent for removing surface-deposited dye in laboratory-based experiments. In any commercial processing, an aqueous-based or SC-CO_2-based scouring treatment would be employed, if necessary, and solvents would not be used.

2.6 | "Superlevelling" of SC-CO_2 Corangar and disperse dyes in SC-CO_2 medium

Undyed polyester fabric (5 g) and polyester fabric (5 g) dyed with 1% owf Corangar Red PE-3469 were treated together in the DyeCoo Textile Systems SC-CO_2 laboratory dyeing machine where each dyeing tube was filled with a piece of dyed and undyed polyester fabric, and CO_2 (147 g). The dyeing temperature was set as 120°C and the dyeing time was 1.5 h. After dyeing, the dyed polyester fabrics were rinsed with acetone in order to remove any surface-deposited dye and then finally air dried.

The dyeing procedure earlier was repeated again but in this case two polyester fabrics (5 g each) dyed with 1% owf Corangar Yellow PE-3205 and 1% owf Corangar Blue PE-3648, respectively, were treated in a single dyeing tube.

Similarly, a 2% owf alizarin dyed polyester fabric (5 g) and an undyed, randomly knotted polyester fabric (5 g) were similarly treated as earlier in the single-dyeing tube and afterwards.

2.7 | Transfer printing

Polyester fabrics were transfer printed with a standard colour calibration block print design at 200°C for 1 min using an Adkins Beta Major pneumatic swing press, with the sublimation dye print papers prepared on a Mimaki JV 150-160 inkjet printer.

2.8 | Colour wash fastness tests

The colour wash fastness analysis was performed using the ISO 105 C02 (Standard soap without optical brightening agent, SDC Enterprises Ltd) wash-fastness test at 50°C for 45 min. After wash testing the colour contrast, grey scale rating between the dyed and washed fabrics was determined visually in a light box and the staining of adjacent multi-fibre strip fabric was similarly rated in a D65 standard lighting colour matching cabinet.

The pilot-dyed samples were tested according to the ISO 105 C06 test method with standard ECE detergent with phosphates and the following programme A1M at 40°C for 45 min with 10 steel balls.

2.9 | Crock fastness

The wet and dry crock fastness analysis was performed using the ISO 105 X12:2016 crock fastness test. A sample of dyed fabric (14 cm × 5 cm) was placed on the testing bed of a James Heal CrockMaster rubbing fastness tester and rubbed with a desized, bleached wet and dry cotton rubbing cloth (16 mm in diameter) 10 times to and fro along a track (10.4 cm) on the specimen using a downward force of 9 N and a rate of one cycle per second. The colour contrast, grey scale rating between the used rub cloth and an unused rub cloth was determined visually in a light box.

2.10 | Light fastness

The light fastness analysis was performed according to the ISO EN 105 B02:2014 light fastness test. Samples of the dyed fabrics (5 cm × 1 cm) and blue wool reference standards were mounted on non-optically brightened white card. The left-hand quarter of the mounted specimens were masked and the samples exposed to xenon arc light using a James Heal TruFade xenon arc light fastness tester. Samples exposure continued until blue wool standard 6 had faded to a grey scale rating of 4 when the grey scale colour change of the samples and blue wool reference standards were determined.

3 | RESULTS AND DISCUSSION

3.1 | Overview

The use of SC-CO_2 technology for the coloration of polyester is well established.[25–27] In addition to the excellent fibre coloration and colour fastness, the process does not require the addition of auxiliaries, such as dispersing agents, that are normally associated with water-based dyeing of polyester. It also eliminates the need for reductive chemical clearing or post-washing. Accordingly, global retailers such as Nike, Adidas and Bon Prix already bulk dye textile fabrics for their apparel ranges and promote the commercial and environmental benefits of this manufacturing approach. For the world's dominant textile fibre, polyethylene terephthalate, this is undoubtedly an attractive twenty-first century processing technology.

However, while SC-CO_2 dyeing is successful, the inability to replicate the coloration of cotton, wool and silk dyeing using this waterless technology has slowed subsequent market penetration. This inability to dye these natural fibres is due to the hydrophobic nature of the SC-CO_2 solvent and its inability to break hydrogen bonds between the adjacent hydrophilic cellulose chains to allow fibre swelling and create pathways for dye diffusion. In addition, the commercially available dyes for cotton/wool are typically salts, which are insoluble in SC-CO_2. In contrast, "disperse" dyes are hydrophobic/non-polar and are soluble in the SC-CO_2 solvent but have little affinity for cotton. To address this incompatibility between the cotton/wool fibre and the non-polar "disperse" dye, a number of approaches have been explored:[28–36]

1. Modification of the disperse dye molecules by attaching a reactive group to allow covalent fixation to the cotton/wool substrates.
2. The addition of auxiliary agents, such as co-solvents and swelling agents, into the SC-CO_2 solvent to aid in the dissolution, transportation and fixation of the dye in the fibre.
3. The modification of the cotton/wool fibre substrate to increase the substantivity of the hydrophobic disperse dyes by covalently attaching bulk aryl residues to the polymeric substrate. This will increase the affinity of the modified cotton/wool substrate towards the non-polar disperse dyes.

In reviewing these approaches, it is apparent to the general reader and industrial dyer that none yet have achieved a commercial outcome due to significant market and processing disadvantages/limitations and that further alternatives need to be identified and developed.

Examination of the previous work by Lewis and Broadbent[34–41] indicated that the approach of modifying cotton and wool with reactive aryl hydrophobes was attractive in not only imparting disperse dyeability but also by improving dimensional stability to laundering, easy care/crease resistance performance and heat setting. Hence, this long-term and strategic research programme aimed to deliver single dye class coloration of polyester, cotton and wool as well as impart multi-functional properties to cotton and wool. In addition to this current journal publication, ongoing research work at Leeds is still focused on developing a simple "one-pot" fibre modification/SC-CO_2 dyeing technology for cotton, nylon, silk and wool to impart dye compatibility with polyester fibres and will be subsequently reported in this journal. However, in recognising and researching the need for introducing aryl-based species into the cellulosic polymer to allow disperse dyeing, it was apparent that in the current portfolio of industrial commodity fibres there is cellulose diacetate. This cellulosic-based polymer material is one of the earliest man-made fibres[42–44] and is manufactured by the reaction of cellulose pulp with acetic anhydride leading to the production of cellulose triacetate and the more common cellulose diacetate, Figure 1.

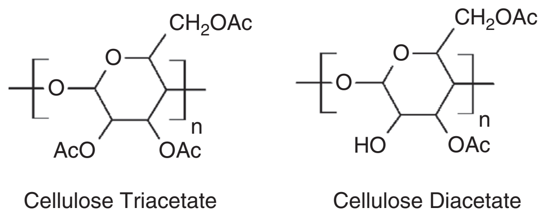

FIGURE 1 Structure of cellulose acetate polymers.

The aliphatic substituent in the cellulose backbone, while more polar than as the phenyl functionality nevertheless does allow commercial disperse dyeing.[43–45] Cellulose diacetate is mainly used in cigarette filters and contributes only 0.78% (0.9 million tonnes) to the global fibre market. However, the recent launch of the Eastman Naia™ (2017) and Naia™ Renew (2020) range of cellulose diacetate yarns opens the way for the development of apparel-based products[46] based on sustainable cellulose sources, favourable biodegradability and recycling opportunities.

While many would suggest it is an unlikely journey for cellulose diacetate to transition from being a "minor" fibre to becoming a viable commercial textile commodity that could displace cotton, it is perhaps worthwhile reflecting that at the time of the First International Wool Research Conference in Melbourne in 1955,[47] cotton and wool were the predominant global textile fibres and there were four Nobel prize winners present at the conference reflecting both the academic and commercial importance of this protein fibre. In addition, the previous year two young designers, Yves Saint Laurent and Karl Lagerfeld, were prize winners at the International Wool Secretariat Fashion Design competition in the best dress and coat categories, respectively, again reflecting the importance of wool fashion. In the subsequent years after this inaugural conference, there was a rapid strategic repositioning of the industry, with synthetic fibres growing rapidly and dramatically displacing wool in the apparel, homeware and carpet sectors. Currently, wool contributes only marginally more to the global fibre market (0.94%) than cellulose diacetate (0.78%) but is regarded as a higher value premium product.

3.2 | SC-CO_2 dyeing of cellulose diacetate

Examination of cellulose diacetate fabric dyed with comparable "disperse-type" red colourant using aqueous and SC-CO_2 media, Table 1, indicated the colour strength achieved in SC-CO_2 was much higher. Indeed the 0.5% owf application level of the Corangar Red PE-3469 was almost the equivalent of the 2% owf application level of the comparable aqueous disperse red dye. This colour difference was due to two related factors:

TABLE 1 Colour strength and fastness of Corangar Solvent Red PE-3469 supercritical carbon dioxide (SC-CO_2) dyed and equivalent water-based Leeds and Colourtex Disperse Red PE-3469 dyed cellulose diacetate woven fabrics (high energy dye).

				Cross-staining						Colour fastness		
Application (% owf)	*K/S* before wash	*K/S* after wash	Wash fastness[a]	Wool	Acrylic	PET	Nylon	Cotton	CA	Dry rub	Wet rub	Light
Red PE-3469, SC-CO_2 Application												
0.5	14.4	13.1	5	5	5	5	4–5	5	4–5	4–5	4–5	6+
1.0	26.6	24.8	4–5	4–5	4–5	4–5	3–4	4–5	3–4	5	5	ND
2.0	27.0	25.5	5	4–5	3–4	4	2–3	3–4	2–3	4–5	5	ND
Red PE-3469, Water Application												
0.5 (Leeds)	7.5	7.4	5	4–5	4–5	4–5	4	4–5	3–4	3–4	4–5	ND
1.0 (Leeds)	11.5	11.5	5	5	4–5	4–5	3–4	4–5	3–4	3–4	4–5	ND
2.0 (Leeds)	16.2	15.9	4–5	4–5	4	4	3	4–5	3	3–4	4	6+
1% (Colourtex)	8.5		3–4	4	4–5	4–5	4–5	5	4			
2% (Colourtex)	10.1		3–4	4–5	4–5	4–5	4–5	4–5	3–4			

[a]ISO 105 C02.
Abbreviation: ND, not determined.

1. The coloration process in the SC-CO_2 medium is not a "disperse" dyeing but rather a true "solvent dyeing" where the fibre structure is more accessible, the dye is fully soluble in the solvent and dispersing agents are not necessary. Indeed, the Corangar dyes are manufactured specifically for SC-CO_2 dyeing without dispersing agents. In Banchero's reviews in 2012 and 2020 he refers to the swelling/plasticising of polyester fibre/film in the SC-CO_2 medium aiding the penetration of the dye into the fibre.[27,48] In contrast, the inability of SC-CO_2 to swell and promote the diffusion of dyes into the interior of polar natural fibres is identified one of the main problems for their similar coloration.
2. In being a true solvent dyeing process, the influence of molecular weight and size on the dye penetration and colour yield that is apparent in aqueous dyeing,[43] is less pronounced in SC-CO_2 dyeing.

A comparison of the wash fastness of the red dyed cellulose diacetate indicated the wash fastness and cross-staining of the fabrics dyed in the SC-CO_2 medium was superior to that observed in the comparable aqueous dyed fabrics. In this laboratory-based and pilot scale study two wash fastness tests were evaluated in order to gain a better insight into the effect of bath temperature (50 or 40°C) and detergent composition (simple soap-based or multi-component ECE Reference detergent with phosphate) on dye substantivity. Under both sets of wash formulation conditions the SC-CO_2 dyed fabrics performed well.

Similarly, examination of the crock fastness of the Corangar Red dyed also indicated the fabrics dyed in the SC-CO_2 medium had superior performance, Table 1, again reflecting the improved penetration and uniformity. As expected, the light fastness of the red dyed substrates were excellent regardless of the dyeing media.

For a broader analysis of the colour range of SC-CO_2 dyes, the Corangar Blue PE-3468 and Corangar Yellow PE-3205 were similarly studied and the colour strength, build-up and fastness properties of the dyed fabrics established. Again, for the blue and yellow dyed fabrics, the SC-CO_2 dyed fabrics offered higher colour strength and better fastness over the comparable aqueous disperse dyed fabrics, Tables 2 and 3. While the light fastness of the yellow dyed substrates were excellent regardless of the dyeing media, the blue dyed cellulose acetates exhibited relatively poorer light fastness performance, which is most likely a reflection of the chromophore stability.

In the fashion and wider textile market a popular design colour is black. Therefore, an evaluation of the aqueous and SC-CO_2 black dyed fabrics, based on a trichromatic mixture of red, blue and yellow dyes, indicated again the SC-CO_2 dyed fabrics offered higher colour strength and better fastness over the comparable aqueous disperse dyed fabrics, Table 4.

A potential criticism of cellulose acetate as a fibre could be its "feel", but this property was historically addressed through the application of the industrial "S-Finish" to cellulose triacetate, which involved a controlled deacetylation of the fibre surface[43] imparting a more "cotton-like" cellulosic handle. The effect of this physical and chemical modification can now be fully characterised through modern textile analytical techniques. With modern finishing technology, the fibre interface and properties can also be effectively engineered and optimised similar to finishing of polyester and cotton. Nevertheless, the effect of surface modification on the dyeing properties may need to be characterised further, but it should not present a significant problem.

TABLE 2 Colour strength and fastness of Corangar Solvent Blue PE-3648 supercritical carbon dioxide (SC-CO_2) dyed and equivalent water-based Leeds and Colourtex Disperse PE-3648 dyed cellulose diacetate woven fabrics (medium energy dye).

Application (% owf)	*K/S* before wash	*K/S* after wash	Wash fastness[a]	Cross-staining						Colour fastness		
				Wool	Acrylic	PET	Nylon	Cotton	CA	Dry rub	Wet rub	Light
Blue PE-3648, SC-CO_2 Application												
0.5	6.1	5.6	4	4	4	4	3	4–5	4	4	5	3
1.0	7.9	7.1	4	4	2–3	3–4	2–3	4	3	3–4	4	ND
2.0	8.3	7.8	4–5	4	2–3	3–4	2–3	4	3	3	3–4	ND
Blue PE-3648, Water Application												
0.5 (Leeds)	1.7	1.3	4	4–5	3	4	3	4–5	4	3–4	5	ND
1.0 (Leeds)	2.2	1.9	4	4–5	2–3	3–4	2–3	4–5	3–4	3–4	4	ND
2.0 (Leeds)	4.0	3.5	4	4	2–3	3	2–3	4	3	3	3–4	3
1% (Colourtex)	2.3		3	4–5	4–5	4–5	4–5	4–5	4			
2% (Colourtex)	2.7		3	3–4	4–5	4	4	4–5	4			

[a]ISO 105 C02.
Abbreviation: ND, not determined.

TABLE 3 Colour strength and fastness of Corangar Solvent Yellow PE-3205 supercritical carbon dioxide (SC-CO_2) dyed and equivalent water-based Leeds and Colourtex Disperse PE-3205 dyed cellulose diacetate woven fabrics (medium energy dye).

Application (% owf)	*K/S* before wash	*K/S* after wash	Wash fastness[a]	Cross-staining						Colour fastness		
				Wool	Acrylic	PET	Nylon	Cotton	CA	Dry rub	Wet rub	Light
Yellow PE-3205, SC-CO_2 Application												
0.5	18.0	14.9	4	4	4	4	3–4	4–5	3	5	5	6^+
1.0	21.4	18.2	4–5	4	4	4	2–3	4–5	3	5	5	ND
2.0	23.7	20.2	4–5	4–5	4–5	4	3	4–5	3	5	4–5	ND
Disperse Yellow PE-3205, Water Application												
0.5 (Leeds)	7.8	5.5	4	4–5	5	4–5	2–3	4–5	3	4	5	ND
1.0 (Leeds)	9.6	7.4	4	4.0	4–5	3–4	2	4–5	2	3–4	4	ND
2.0 (Leeds)	12.9	8.5	3	3–4	4–5	3–4	2	4	2	3	4	6^+
1% (Colourtex)	10.0		3–4	4–5	4–5	3–4	2–3	4–5	2			
2% (Colourtex)	10.1		3	3–4	4–5	3	2	4	1–2			

[a]ISO 105 C02.
Abbreviation: ND, not determined.

3.3 | Pilot scale dyeing of cellulose acetate (Naia) and recycled polyethylene terephthalate (RPET)/cellulose acetate (Naia) blend fabrics

In any commercialisation process, there is the necessity to translate the academic study to a pilot-scale trial and demonstrate its applicability on a larger production level. Our laboratory- scale study uses sealed, pressurised stainless steel vessels where the fabric, dye and SC-CO_2 fluid are physically rotated and raised to the requisite dyeing temperature. On completion of the dyeing cycle the vessel is depressurised and the dry fabric removed. Typically, it is then aftersoaped in water or rinsed in acetone since the closed system does not have the capability for an *in situ* surface clearing. In contrast, while the commercial scale equipment is still essentially a sealed vessel with beam or yarn package holders, it allows the SC-CO_2 fluid with the dye to be circulated through the fabric via a pump. In addition, following the draining of spent SC-CO_2 dye liquor, the final dyed textile can be rinsed with fresh SC-CO_2 fluid or aftersoaped in water to remove any surface deposits if necessary. However, industrial experience with the large-scale dyeing has shown that due to the superlevelling properties of the SC-CO_2 fluid often no after-rinsing is necessary and the fastness of the dyeing is excellent, as illustrated in our pilot scale results. However, where there is the need the fabric can be SC-CO_2 rinsed or aftersoaped with minimal water usage.

TABLE 4 Colour strength and fastness of Corangar Solvent Black Trichromat supercritical carbon dioxide (SC-CO_2) dyed and equivalent water-based Leeds disperse dyed cellulose diacetate woven fabrics.

Application (% owf)	K/S before wash	K/S after wash	Wash fastness[a]	Cross-staining						Colour fastness	
				Wool	Acrylic	PET	Nylon	Cotton	CA	Dry rub	Wet rub
Black Trichromat, SC-CO_2 Application											
0.5	5.1	4.4	4	5	5	5	4	5	4	4–5	4–5
1.0	11.8	11.5	4	4–5	5	4–5	3–4	5	3–4	4–5	4–5
2.0	23.5	17.3	3–4	5	5	4–5	4	5	3–4	4	4
Disperse Black Trichromat, Water Application											
0.5 (Leeds)	6.5	5.5	3–4	4–5	3–4	4	3–4	4–5	3–4	4	5
1.0 (Leeds)	9.9	6.8	2–3	4–5	3	3–4	2–3	4–5	3	3	4
2.0 (Leeds)	10.1	8.9	3	4–5	2–3	3–4	2–3	4–5	2–3	3	4

[a]ISO 105 C02.

Figure 2 illustrates that the Naia™ cellulose acetate materials can be successfully dyed in the SC-CO_2 process with excellent dye-uptake, uniformity and colour strength. The black dyed fabric wound around the beam is shown in the pilot scale 5 kg beam dyeing machine, Figure 3, which replicates the construction of the larger commercial machines, and can be arranged for yarn dyeing as well as fabric coloration. Confirmation of the higher colour strength of the SC-CO_2 dyed fabrics, Tables 1, 3 and 5, relative to the aqueous control fabric achieved at laboratory scale, was also observed with the larger scale dyeings with Corangar Blue PE-3618 dye.

The colour depth of SC-CO_2 dyed Naia materials was at least twice higher than samples dyed in aqueous process. This benefit of the higher colour strength for equivalent dye application will clearly translate into lower dyestuff cost and further increase the attractiveness of SC-CO_2 dyeing. The pilot-scale dyed samples showed reasonable to excellent ISO 105 C06 wash fastness performance, even without a post-dyeing rinsing treatment, Table 5. Even the deep black-dyed fabrics fulfilled the commercial wash and rub fastness requirements without any further post-treatment and washing process.

4 | RECYCLING AND REMANUFACTURING IN POLYESTER AND CELLULOSE ACETATE

4.1 | Superlevelling properties of SC-CO_2 and potential for recycling/remanufacturing “waste” textiles

The textile industry has recognised that the fast fashion has beneficially contributed to the evolution of a global economy, but the downside has been the generation of large quantities of textile waste destined for landfill. There have been initiatives to collect the cellulosic material for regenerating new fibres and these developments will satisfy many of the environmental and sustainability challenges. However, this vision is limited and does not acknowledge the wider need for a coordinated “water” approach, which links waterless dyeing with waterless textile recycling and remanufacturing. In this section of the article, we highlight the potential for better usage of fibre, water and dye resources and illustrate a further aspect of novel creative design using SC-CO_2 technology.

One of the technical benefits of SC-CO_2 dyeing is the absence of any dyeing auxiliaries. This offers cost savings in terms of chemicals but also recognises the excellent levelling properties of the technology. This advantageous feature is illustrated in Figure 4, where a deep red dyed polyester fabric was treated with clean SC-CO_2 fluid in the presence of an identically sized undyed polyester fabric, resulting in the production of two identical, lighter red dyed fabrics due to highly efficient SC-CO_2 levelling action.

In recognising the efficiency of the “superlevelling” process, it offers opportunities for the disperse dye inside the coloured polyester fibre/fabric, which may have been discarded as waste, and could be reused as a source of colourant for the undyed polyester fibre/fabric. It is not pre-extracted, isolated or purified prior to any subsequent use in colouring the secondary polyester fabric but rather the dye is transferred by the SC-CO_2 fluid from the “primary” coloured fibre into the secondary white fibre/yarn/fabric or garment in an integrated process. This allows for the recovery of the valuable colourant for reuse *in situ* in the SC-CO_2 dyeing vessel and aids in preparing the waste fabrics/fibres for subsequent reuse.

CMYK is a subtractive colour model used in colour printing.[49] CMYK refers to the four inks used in colour printing: cyan, magenta, yellow and key (black), which

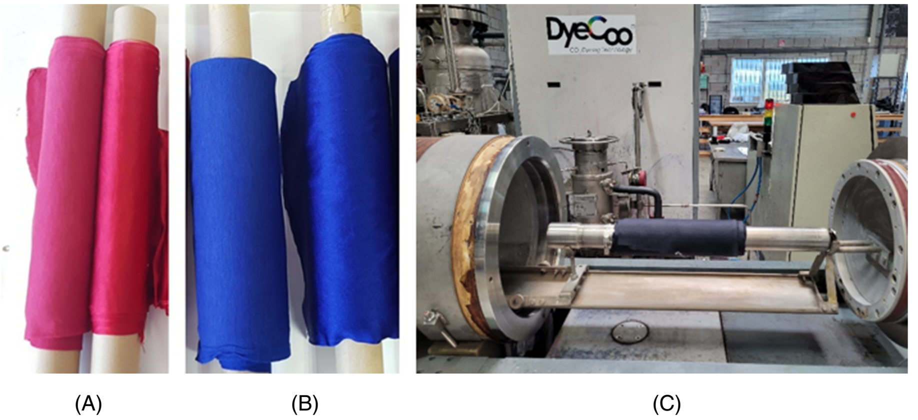

FIGURE 2 Pilot scale SC-CO_2 dyeing of cellulose diacetate (Naia™) and 60% RPET/40% Naia™ blend fabrics with (A) 0.5% Corangar Red PE-3469, (B) 0.7% Corangar Blue PE-3618 and (C) trichromatic mix of Corangar Black (1.54% owf dye content) located in the pilot-scale dyeing system.

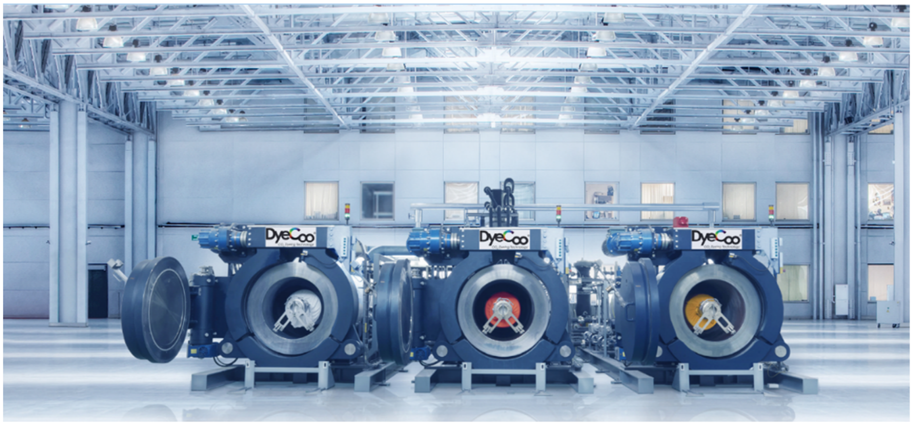

FIGURE 3 Commercial scale SC-CO_2 dyeing machines.

TABLE 5 Colour strength, ISO 105 C06: A1M wash fastness and rub fastness of pilot-scale supercritical carbon dioxide (SC-CO_2) Corangar solvent dyed cellulose diacetate (Naia) and recycled polyethylene terephthalate (RPET)/cellulose diacetate (Naia) blend fabrics.

			Cross-staining						Rub fastness	
Corangar dyed fabric	**K/S**	**Wash fastness**	**Wool**	**Acrylic**	**PET**	**Nylon**	**Cotton**	**CA**	**Dry**	**Wet**
0.5% owf Red PE-3469										
60/40 RPET/Naia	13.2	4	4/5	4/5	4	4	4/5	3/4	4/5	4
100% Naia fabric	28.3	4	4/5	4/5	4	4	4/5	3/4	3/4	4/5
0.7% owf Blue PE-3618										
60/40 RPET/Naia	25.9	4	4/5	4/5	4	4	4/5	3	4/5	4
100% Naia fabric	43.0	4	4/5	4/5	4	4	4/5	3	3/4	4
1.54% owf Black										
60/40 RPET/Naia	22.7	5	5	5	5	5	5	5	4/5	3/4
100% Naia fabric	31.0	5	5	5	4/5	4/5	5	4/5	3/4	4

when combined generate new colours, for example, mixing yellow and cyan produces green, Figure 5.

This colour mixing has been clearly demonstrated in our studies, Figure 6, where yellow and blue Corangar solvent dyed fabrics (dyed in SC-CO_2 but may be aqueous dyed) are placed in the clean SC-CO_2 fluid together, the blank dyebath raised to 120°C and maintained for 30 min. On releasing the SC-CO_2 bath, it was apparent that two spectrally-equivalent, uniformly coloured green fabrics have been produced, Figure 6, following the redistribution of the dye from both fabrics. What is remarkable about this process is the speed and efficiency of dye exchange and uniform levelling between the separate polyester materials

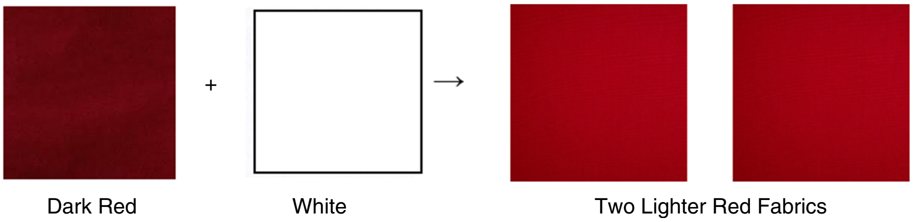

FIGURE 4 Transfer of dye from deep red dyed primary polyester fabric to white secondary fabric by SC-CO_2 dyeing process to produce two uniformly coloured red fabrics.

producing two fabrics of equivalent colour. This dye relocation could not be achieved in aqueous media and highlighted the "superlevelling" of SC-CO_2.

This predictable coloration process using the dye "locked" in the two "waste" fabrics was further demonstrated by combining three yellow, red and blue fabrics in the clean SC-CO_2 fluid, dyeing at 120°C and producing three identical black coloured fabrics.

The concept of superlevelling within a fabric can be further illustrated using transfer printed polyester fabric with typical colour calibration blocks used to assess the colour profile of a digital print, Figure 7. The transfer printed fabric was placed in a SC-CO_2 blank dyebath, the temperature raised to 120°C and maintained for 30 min. At the end of the dyeing cycle, the individual fabric colour blocks have homogenised producing a uniform silver grey colour, Figure 7A, again demonstrating the remarkable levelling properties of the SC-CO_2. In contrast, the identical transfer printed polyester fabric with distinct colour blocks similarly treated at 120°C in aqueous media did not level to any great extent, Figure 7B. This removal of a transfer print design could be particularly useful for erasing "dated unfashionable" designs on fabrics, such as on replica football kits, and can create a "second life" value added coloured garment/fabric.

Banchero in 2020 commented that "the tuneable solvent power of supercritical carbon dioxide could allow the dye and the solvent to be selectively recollected and recycled at the end of the coloration process".[27] However, from our studies exploiting the *in situ* superlevelling properties of SC-CO_2, it is apparent that there is no need for a distinct extraction/isolation of dye from first life textiles; rather the dyed textile can be regarded a "holding vehicle" for subsequent recoloration of other textiles. This would provide for an integrated, simpler and less expensive recycling and remanufacturing framework and in "stripping" the dyed primary waste material it facilitates easier recycling of the waste/byproduct. While the technical challenge of predictive subtractive coloration using primary coloured fabric and secondary undyed textiles is significant, it is not insurmountable. Indeed, this commercial opportunity has already been recognised by Nike in their SC-CO_2 processing publication in 2016 where the coloration principle of this circular economy has been patented.[50]

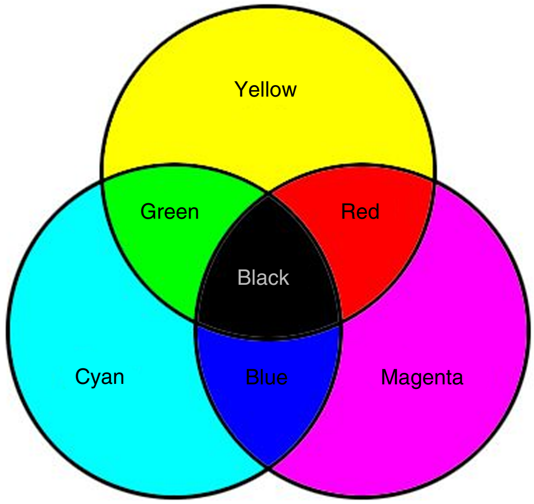

FIGURE 5 Diagrammatic illustration of subtractive colour mixing.

4.2 | Novel structural dyeing (tie-dyeing) of fabrics in the SC-CO_2 medium

Alizarin is a "natural" colourant found in madder, with purpurin, and imparts an intense yellow coloration under SC-CO_2 dyeing conditions. (In water-based dyeing it is typically red at neutral pH, while under the "acid" conditions of SC-CO_2 it is yellow.) In recognising the reuse application of the dye on non-polar fabrics such as polyester, SC-CO_2 alizarin dyed polyester fabric was placed in a SC-CO_2 fluid blank dyebath with white undyed polyester fabric which had been tied with knots. The temperature was then raised to 120°C and maintained for 30 min. At the end of the dyeing cycle, the polyester fabric that had been tied in knots exhibited a typical tie-dyed appearance where the knots restricted the penetration of the SC-CO_2 alizarin dye into the fabric and provided a means of introducing design features into polyester fabric using dye located in an adjacent fabric, Figure 8. In the unrestricted areas of the fabric the dye exhausted uniformly.

FIGURE 6 Subtractive colour mixing of yellow and blue SC-CO_2 dyed polyester fabrics to produce two uniformly coloured green fabrics.

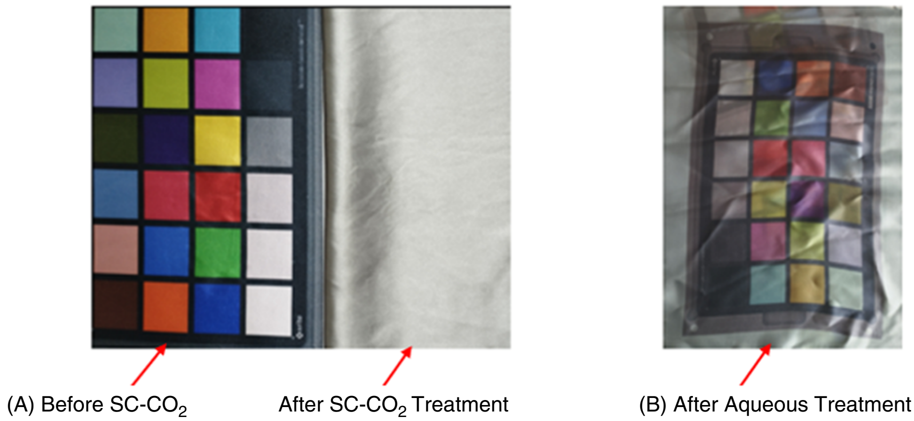

FIGURE 7 Effect of post-levelling of colour calibration blocks transfer printed on white polyester fabric. (A) Fabrics before and after superlevelling treatment in SC-CO_2 fluid. (B) Incomplete levelling following comparable aqueous treatment.

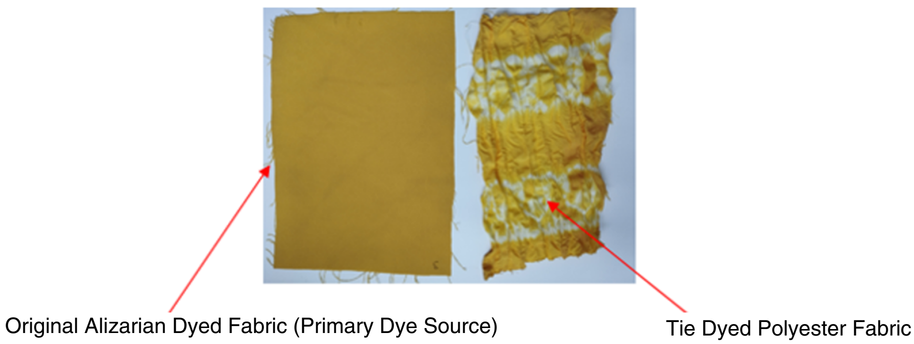

FIGURE 8 Redistribution of alizarin colourant from 2% owf dyed polyester and resultant tie-dyed white polyester fabric in the SC-CO_2 fluid.

5 | CONCLUSIONS

In this study we have demonstrated the SC-CO_2 dyeing of cellulose diacetate fabric with a range of Corangar SC-CO_2 solvent dyes and compared the coloration performance relative to traditional aqueous based disperse dyeing application. It was found that the SC-CO_2 based dyeing offered greater colour strength and improved fastness over the comparable traditional water-based coloration. Studies at DyeCoo Textile Systems, the Netherlands, validated these results and highlighted the technical and commercial applicability of the coloration technology, not only to polyester but also to cellulose diacetate.

Much research has focused on introducing aromatic functionalities into cotton to allow single dye class coloration of cotton and polyester to establish an integrated, more efficient commercial dyeing framework. We demonstrated that cellulose diacetate (incorporating aliphatic substituents) offers a sustainable, biodegradable cellulose-based fibre alternative to cotton that enables a truly "waterless" dyeing technology with no polluting effluent and lower operational costs. The recent launch by Eastman of their Naia™ range of cellulose diacetate coupled to SC-CO_2 dyeing technology offers transformational technology for the polyester dominated textile industry.

Coupled to these strategic innovations is the potential to exploit the "superlevelling" nature of SC-CO_2 fluids to enable more efficient recycling and remanufacturing of dyed/printed polyester and cellulose diacetate. As polyester is the predominant global textile fibre (52% of the market) in the apparel sector, the persuasive and logical next step in fibre evolution is to recognise that cellulose diacetate may well have a significant role going forward to play as a sustainable cellulose-based fibre in an eco-friendly twenty-first century textile industry. Twenty-five years ago, Lewis and Broadbent[34] asked the question "A universal dye for all fibres – are disperse dyes capable of fulfilling this vision?" Perhaps this question should now be revised to "A universal dye for all fibres – are SC-CO_2 solvent dyes capable of fulfilling this vision, coupled to an integrated SC-CO_2 based polyester/cellulose acetate circular manufacturing and recycling framework?" Perhaps in another 25 years we will have a definitive confirmation!

ACKNOWLEDGEMENTS

The authors acknowledge the primary funding through Innovate UK for the project, Resurgence of UK Manufacturing through Waterless Innovation, but also partial funding through the AHRC Future Fashion Factory initiative. The authors gratefully recognise the materials and technical contribution of Colourtex in providing the dyes and also advising and promoting SC-CO_2 coloration technology. The authors also thank Dr Peter Rhodes in reviewing the article and providing technical and editorial advice. Finally, the authors thank the Dyers' Company, London, for their ongoing commitment and encouragement in developing "green" technology for the dyeing industry.

ORCID

Peter J. Broadbent https://orcid.org/0000-0003-3671-2379
Chris M. Carr https://orcid.org/0000-0001-8541-4955
David M. Lewis https://orcid.org/0000-0001-7396-9793
Muriel L. Rigout https://orcid.org/0000-0002-4894-7768
Narjes Shojai Kaveh https://orcid.org/0000-0002-8810-0617

REFERENCES

1. Remy N, Speelman E, Swartz S. Style that's sustainable: a new fast-fashion formula. McKinsey & Co 2016 Accessed 29 January 2023.https://www.mckinsey.com/business-functions/sustainability/our-insights/style-thats-sustainable-a-new-fast-fashion-formula
2. Lehmann M, Arici G, Boger S, Martinez-Pardo C, Krueger F, Schneider M, Carriere-Pradal B, Schou D. Pulse of the Fashion Industry. 2019. Global Fashion Agenda, Boston Consulting Group, and Sustainable Apparel Coalition. https://media-publications.bcg.com/france/Pulse-of-the-Fashion-Industry2019.pdf
3. Stanescu MD. State of the art of post-consumer textile waste upcycling to reach the zero waste milestone. *Environ Sci Pollut Res*. 2021;28:14253-14270.
4. Marques AD, Marques A, Ferreira F. Homo sustentabilis: circular economy and new business models in fashion industry. *SN Appl Sci*. 2020;2:306-311. doi:10.1007/s42452-020-2094-8
5. Bailey K, Basu A, Sharma S. The environmental impacts of fast fashion on water quality: a systematic review. *Water*. 2022;14:1073-1084. doi:10.3390/w14071073
6. Uddin F. Energy management and energy crisis in textile finishing. *Am J Energy Res*. 2014;2(3):53-59.
7. UNIDO. Textile industry: Output of a seminar on energy conservation in textile industry, UNIDO and Ministry of International Trade and Industry (Japan). pp. 1–9 1992.
8. Shaik MA. Water conservation in textile industry. *Pak Text J*, November Issue. 2009;58:48-51.
9. Forgacs E, Cserhati T, Oros G. Removal of synthetic dyes from wastewaters: a review. *Environ Int*. 2004;30(7):953-971.
10. European Parliament. *The Impact of Textile Production and Waste on the Environment (Infographic)*. European Parliament; 2021 https://www.europarl.europa.eu/news/en/headlines/society/20201208STO93327/the-impact-of-textile-production-and-waste-on-the-environment-infographic
11. Leal Filho W, Perry P, Heim H, et al. An overview of the contribution of the textiles sector to climate change. *Front Environ Sci*. 2022;10:973102. doi:10.3389/fenvs.2022.973102
12. Damania R, Desbureaux S, Rodella A-S, Russ J, Zaveri E. Quality unknown–the invisible water crisis, World Bank Report 2019. Accessed 28 December 2022 https://openknowledge.worldbank.org/handle/10986/32245
13. Saus W, Knittel D, Schollmeyer E. Dyeing with supercritical carbon dioxide-an alternative to high temperature dyeing of polyester. *Textile-Praxis*. 1992;47:1052-1054.
14. Saus W, Knittel D, Schollmeyer E. Dyeing of textiles in supercritical carbon dioxide. *Text Res J*. 1993;63:135-142.
15. Cygnarowicz-Provo M, King JW, Marmer WN, Magidman P. Extraction of Woolgrease with supercritical carbon dioxide. *JAOCS*. 1994;71(2):223-225.
16. Zhang J, Zheng H, Zheng L. A novel eco-friendly scouring and bleaching technique of flax rove using supercritical carbon dioxide fluid. *J Eng Fibers Fabr*. 2017;12:44-51.
17. Eren HA, Avinc O, Eren S. Supercritical carbon dioxide for textile applications and recent developments. *IOP Conf Series: Mater Sci Eng*. 2017;254:82011. doi:10.1088/1757-899X/254/8/082011
18. Eren S, Avinc O, Saka Z, Eren HA. Waterless bleaching of knitted cotton fabric using supercritical carbon dioxide fluid technology. *Cellul*. 2018;25:6247-6267.
19. What is a supercritical fluid? https://www.scimed.co.uk/education/what-is-a-supercritical-fluid/. Accessed 29 January 2023.
20. Polikhronidi N, Batyrova R, Aliev A, Abdulagatov I. Supercritical CO_2: properties and technological applications-a review. *J Therm Sci*. 2019;28:394-430. doi:10.1007/s11630-019-1118-4
21. Tutek K, Masek A, Kosmalska A, Cichosz S. Application of fluids in supercritical conditions in the polymer industry. *Polymers*. 2021;13:729-746.
22. DyeCoo. https://dyecoo.com/. Accessed 28th December 2022.
23. DyeCoo. Internal Production Analysis. 2022.
24. Textile Exchange. Preferred Fiber & Materials Market Report 2021. Accessed 28th December 2022 https://textileexchange.org/app/uploads/2021/08/Textile-Exchange_Preferred-Fiber-and-Materials-Market-Report_2021.pdf
25. Agrawal BJ. Supercritical carbon-dioxide assisted dyeing of textiles: an environmental benign waterless dyeing process. *Int J Innov Res Creat Technol*. 2015;1:201-206.
26. Elmaaty TA, Mousa A, Gaafar H, Hebeish A, Sorour H. Development of textile dyeing using the green supercritical fluid technology: a review. *Mat Int*. 2020;2:373-390.
27. Banchero M. Recent advances in supercritical fluid dyeing. *Color Technol*. 2020;136:317-335.
28. Gebert B, Saus W, Knittel D, Buschmann SE. Dyeing natural fibres with disperse dyes in SCF CO_2. *Textile Res J*. 1994;64(7):371-374.

29. Schmidt A, Bach E, Schollmeyer E. The dyeing of natural fibres with reactive disperse dyes in supercritical carbon dioxide. *Dyes Pigm*. 2003;56:27-35.
30. Long J-J, Xiao G-D, Xu H-M, et al. Dyeing of cotton fabric with a reactive disperse dye in supercritical carbon dioxide. *J Supercrit Fluids*. 2012;69:13-20.
31. Long J-J, Ma Y-Q, Zhao J-P. Investigations on the level dyeing of fabrics in supercritical carbon dioxide. *J Supercrit Fluids*. 2011;57:80-86.
32. Luo X, White J, Thompson R, et al. Novel sustainable synthesis of dyes for clean dyeing of wool and cotton fibres in supercritical carbon dioxide. *J Clean Prod*. 2018;199:1-10.
33. Elmaaty TA, Kazumasa H, Elsisi H, et al. Pilot scale water free dyeing of pure cotton under supercritical carbon dioxide. *Carbohydr Polym Technol Appl*. 2020;1:100010. doi:10.1016/j.carpta.2020.100010
34. Lewis DM, Broadbent PJ. A universal dye for all fibres-are disperse dyes capable of fulfilling this vision? *J Soc Dye Colour*. 1997;113:159-164.
35. Özcan AS, Clifford AA, Bartle KD, Lewis DM. Dyeing of cotton fibres with disperse dyes in supercritical carbon dioxide. *Dyes Pigm*. 1998;36:103-110.
36. Özcan AS, Clifford AA, Broadbent PJ, Lewis DM. Dyeing of modified cotton fibres with disperse dyes from supercritical carbon dioxide. *J Soc Dye Colour*. 1998;114:169-173.
37. Lewis DM, Pailthorpe MT. The benzoylation of wool and its effect on dye affinity and shrink resistance. *J Soc Dye Colour*. 1983;99:354-357.
38. Lewis DM, Pailthorpe MT. The modification of wool with reactive hydrophobes. *J Soc Dye Colour*. 1984;100:56-63.
39. Broadbent PJ, Clark M, Croft SN, Lewis DM. Use of a novel water-soluble arylating agent to dye wool and wool blends with disperse dyes. *Color Technol*. 2007;123:29-33.
40. Liu H, Lewis DM. *Application of Sodium Benzoyl Thiosulphate from Aqueous Solution to Wool and its Effect on Substantivity of Disperse Dyes. Part 1: Synthesis and Characterization of Sodium Benzoyl Thiosulphate*. Vol 126. Coloration Technology; 2010:147-151.
41. Liu H, Lewis DM. *Application of Sodium Benzoyl Thiosulphate from Aqueous Solution to Wool and its Effect on Substantivity of Disperse Dyes. Part 2: Modification of Wool with Sodium Benzoyl Thiosulphate*. Vol 126. Coloration Technology; 2010:152-156.
42. Steinman HW. Cellulose acetate and fibers. In: Lewin M, Pearce EM, eds. *Handbook of Fiber Science and Technology: Volume 4-Fiber Chemistry*. Marcel Dekker; 1985:1001-1078.
43. Mears P. Dyeing of acetate and triacetate fibres, Chapter 5. In: Hawkyard C, ed. *Synthetic Fibre Dyeing*. Society of Dyers and Colourists; 2004:164-217.
44. Campbell B. The dyeing of cellulose diacetate. *J Soc Dye Colour*. 1966;82:303-313.
45. Choi J-H, Towns AD. Acetate dyes revisited: high fastness dyeing of cellulose diacetate and polyester–polyurethane. *Color Technol*. 2001;117:127-133.
46. Naia™. https://naia.eastman.com/. Accessed 28 December 2022.
47. Commonwealth Industrial Scientific Research Organisation. Proceedings of the First International Wool Textile Research Conference, Melbourne. 1955.
48. Banchero M. Supercritical fluid dyeing of synthetic and natural textiles–a review. *Color Technol*. 2012;129:2-17.
49. Rigg B, Colorimetry and the CIE System, in Colour Physics for Industry. Society of Dyers and Colourists, Bradford, UK. 1997 Editor McDonald R, 84–88.
50. Kelly MW, Nike Inc. Supercritical Fluid Rolled or Spooled Material Finishing. US Patent Publication Number US 2016/0244912 A1.

AUTHOR BIOGRAPHIES

Peter J. Broadbent studied in the Department of Colour Chemistry and Dyeing at the University of Leeds between 1983-1993 and awarded with his BSc, MSc and PhD degrees. Peter worked as a Research Fellow/Senior Research Fellow in the Department of Colour Chemistry and Dyeing (1993-2010), the School of Materials (Textiles and Paper), University of Manchester (2011-2012), and the School of Design, University of Leeds (2013-current). Peter's research interests include cosmetic science, the reactive dyeing of cotton, the dyeing of all types of textile fibres in supercritical carbon dioxide, security printing, inkjet printing, and the easy-care finishing of cotton.

Chris M. Carr Professor of Textile Technology at the School of Design, University of Leeds and Liveryman in the Dyers Company, London. Research interests include textile and hair chemistry, coloration, textile finishing, sustainable technologies, healthcare and cultural heritage science.

David M. Lewis Professor of Colour Chemistry & Head of Department until retirement in 2004 and currently active as Professor Emeritus at the University of Leeds. His research interests include cosmetic science, improving reactive dyeing of cotton, dyeing all types of textile fibres in super-critical carbon dioxide, security printing, ink-jet printing, durable flame proofing of cellulosic textiles. These interests have resulted in him being research director of two spin-out companies, have led to publication of 252 refereed papers, two books and 62 patents and was President of the SDC (1993/1994).

Muriel L. Rigout Associate Professor in Textile Printing and Design at the School of Design, University of Leeds and PhD graduate of the Department of Colour Chemistry, University of Leeds. Research expertise is focused on textile coloration and finishing, in particular novel colour effects, digital print technology, multi-sensory effect chemistries and applications, cosmetic science, eco-friendly processing and cultural heritage science.

Ernst J. Siewers Technical director of DyeCoo Textile systems B.V. Chemical engineer (TU-Delft), with >20 years in SC-CO_2 technology. Involved from the start in the design, development and implementation of DyeCoo's innovative CO_2-based textile dyeing technology.

Narjes Shojai Kaveh Principal research scientist of DyeCoo Textile Systems B.V. PhD in Chemical Engineering from TU-Delft and experienced in supercritical fluid technology with application to sustainable production technology and SC-CO_2 textile dyeing. Extensive practical experience in designing and performing high-pressure, high-temperature lab and pilot equipment, and product and process development.

How to cite this article: Broadbent PJ, Carr CM, Lewis DM, Rigout ML, Siewers EJ, Shojai Kaveh N. Supercritical carbon dioxide (SC-CO_2) dyeing of cellulose acetate: An opportunity for a "greener" circular textile economy. *Coloration Technology*. 2023;1-14. doi:10.1111/cote.12690