# ECO-FRIENDLY PRINTING WITH AGRO-INDUSTRIAL WASTE: REVIEW OF ADVANCES IN FUNCTIONAL TEXTILES

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***Abstract:***
*The growing demand for sustainable and eco-friendly solutions in the textile and graphic industries highlights the urgent need to replace conventional synthetic dyes with natural alternatives. This review aims to provide an overview of recent advances in the use of agro-industrial waste as a valuable source of bio-based pigments and binders for printing applications. The main objective is to demonstrate how natural colorants such as anthocyanins or carbon-based pigments like biochar and activated carbon can serve as options for achieving both aesthetic and functional properties in textiles. Furthermore, the review discusses the role of eco-friendly binders in formulating stable printing pastes. Special attention is given to functional outcomes such as antimicrobial activity, antioxidant capacity, and dielectric properties. By summarizing current progress and challenges, the paper emphasizes the potential of natural pigments and agro-industrial by-products to drive innovation in textile printing. The review also identifies existing limitations, including colour stability and scalability of production, and highlights future perspectives for integrating natural pigments into advanced functional materials. In this way, the study contributes to building a foundation for sustainable, innovative, and environmentally responsible printing technologies.*

**Keywords:** *agro-industrial waste, natural pigments, eco-friendly printing, functional textiles.*

## 1. INTRODUCTION

Modern graphic and textile industries face increasing sustainability and environmental pressures, largely due to the extensive use of synthetic dyes and pigments. Although highly durable, their complex chemistry limits degradation, and their by-products may be toxic, allergenic, or carcinogenic, posing risks to human health and ecosystems [1]. Consequently, research is increasingly directed toward natural dyes and pigments, particularly plant- and mineral-based colourants incorporated into printing pastes. Extracts from agro-industrial waste (e.g., grape pomace and blackberry) are especially attractive because they combine colouring ability with antioxidant and antimicrobial potential, enabling value-added functional textiles while supporting waste valorisation [2].

## 2. NATURAL INKS

Natural inks and dyes are organic or mineral colourants applied to various substrates, including textiles. While synthetic dyes dominate industry because of their colour range and permanence, natural dyes are gaining renewed interest due to biodegradability, low toxicity, and bioactive potential, reflecting their long-standing use before the advent of synthetic colourants [3]. **Plant dyes** constitute the most numerous groups and include pigments such as anthocyanins, flavonoids, tannins, curcumin, and chlorophyll, which are extracted from flowers, leaves, roots, fruits, or bark [4]. This group of dyes often possesses additional functionalities (antimicrobial and antioxidative properties), making them suitable for obtaining functional textiles [5]. **Animal dyes**, although less frequently used in contemporary research, had historical significance. The most well-known example is carmine, a red colour obtained from the cochineal insect, which was used to dye luxury fabrics [6]. Also, in ancient times, Tyrian purple from sea snails of the genus *Murex* was used, which was a symbol of status and power [7]. **Mineral dyes** include pigments such as ochre, hematite, and ultramarine, which are distinguished by exceptional stability and resistance to light and washing. Their application in the textile industry is limited, but in combination with natural binders, they can contribute to specific functionalities, as UV resistance [4].

Contemporary research is focused on plant dyes due to their availability, low cost, and possibility of extraction from agro-industrial waste [5]. In this way, the ecological footprint of waste is simultaneously reduced and a new source of dyes for textile applications is obtained. The most common printing with natural dyes involves obtaining colour by grinding part of the plant into a powder sample and adding various thickening agents (alginates) which are subsequently treated as conventional screen dyes. Various researchers have investigated the intensity of colour (K/S) from alkanet [8][9][10], rhubarb [8][9], turmeric [9][11], marigold [11], madder [12], walnut bark [12][13], tamarisk, acacia [14], pomegranate bark [13][14], walnut shells, orange tree leaves, chamomile [13], henna [10][14][15], Japanese knotweed rhizome [16][17], sour dock [18][19], grape [5], or white pine [20][21]. Özgüney et al. investigated the applicability of Indian madder in printing, examining the effects of dye concentration and urea, type of fixing, temperature and duration of fixing, type of mordant and method of adding mordants [22]. El-Hennawi and colleagues (2012) used laccase enzymes instead of harmful mordants for fixing natural dyes [23]. Klančnik demonstrated the applicability of Japanese knotweed rhizome extract for screen printing on paper and textiles using synthetic thickeners [17]. In another investigation, Klančnik reported that methanolic extracts of *Impatiens glandulifera* flowers exhibit pronounced pH-dependent colour changes but also cause reduced paste viscosity at higher concentrations, limiting printability and leading to brownish colour shifts after drying due to non-anthocyanin phenolics [19].

Visual and spectrophotometric measurements of prints on various materials showed variations in colour depending on dye concentration and the basic colour of the substrate. Prints on virgin fibre paper (white) had a purple-blue tint, whilst on recycled paper (brown) they had a greenish-yellow tint. Alkaline printing paste gave greenish prints, whilst in the case of acidic printing paste, the prints were lighter and less chromatic [19]. Thakker, Sun and Bucknall used aqueous extracts of plant indigo *Indigofera tinctoria*, red quebracho *Schinopsis lorentzii* and plant flower forest flame *Butea monosperma*, which give blue, red and yellow colours, respectively. Black plant colour was obtained by mixing these primary colours. These colours were used in a cartridge ink-jet printer and properties of absorption and colour durability were investigated [24]. Savvidis et al. also used ink-jet printing dyes on a water basis prepared with natural pigments from henna Bixa orellana, acacia Acacia Catechu, pomegranate bark Punica granatum and yellow dock Rumex crispus. The physical and rheological properties of the prepared dyes—pH, conductivity, surface tension and viscosity—were measured over a period of 90 days to assess the stability of the dyes [15].

**Red Shades**

Anthocyanins are flavonoid pigments whose colour and stability depend on molecular structure and environmental factors such as pH and light [25]. In addition to colouration, anthocyanin-rich extracts exhibit antioxidative and antimicrobial properties [5] and are therefore widely explored as pH-responsive indicators in intelligent systems for food freshness monitoring [26]. Some examples relate to the production of agar/potato starch filled with anthocyanin [27]; cellulose nanofibril film for packaging based on anthocyanin from purple sweet potato and oregano essential oil [28]; purple sweet potato starch and pH indicator films based on peel connected with anthocyanin [29]; *Echium amoenum* anthocyanins incorporated into bacterial cellulose film [30]; gelatine films with anthocyanin [31]; and nanocomplexes of ovalbumin-propylene glycol alginate filled with anthocyanin [32]. However, stabilisation and delivery of these molecules is challenging because they are unstable under certain environmental conditions, leading to low bioavailability [33]. Ameri et al. developed a food-safe film for pH-indicator sensors on fish packaging using anthocyanin. Thanks to simple printing procedure, the solution is easily applicable to food packaging [34]. Betalain and betanin extracted from the peel of red pitaya and combined with anthocyanin extracted from the peel of Kyoho grape have been developed as dyes for piezoelectric inkjet printing and then printed as colorimetric indicators [35]. Sun and colleagues developed a printing dye based on natural pigment from purple cabbage and successfully applied it in fish packaging for monitoring freshness [36]. Liu and colleagues combined anthocyanins from black goji berries with xanthan gum, they used screen printing for applying these applications [37]. Alshammari et al. screen printed a thermochromic dye from red cabbage anthocyanins on paper, the experiment showed reddish-purple tones in the acidic domain and pH-dependent colour changes [38]. A common combination of anthocyanins is with chitosan [39] or sodium alginate [5].

**Blue Shades**

Blue hues from nature are rare because only a few classes of natural pigments stably give blue colour and this is usually under specific environmental conditions. The main sources of blue hues are indigo compounds – indigotin from *Indigofera/Isatis tinctoria* – which in oxidised form give stable blue and are a classic choice for textiles [40]. These plants have been used for centuries in art for obtaining blue pigment. Phycobilins such as phycocyanin from *Spirulina*, a blue-green alga that grows in both salt and fresh water, give an intensely blue pigment but is extremely sensitive to light and heat [41]. Genipin-derived pigments such as *gardenia blue* arise from the reaction of genipin with amines/proteins [42]. There are anthocyanin systems also in which blue is achieved by changing the pH value to a neutral or alkaline medium. For strong blue hues, stable formulas combined from gardenia blue pigment and gelatine gels have been shown, which are suitable for food colours [43], and there are also blue edible ink-jet dyes from this pigment [44]. For textiles, the latest review and experimental work examine natural indigo *Indigofera tinctoria* and proposes direct pigment printing with bio-based binders, thereby preserving blue hues [45].

**Yellow Shades**

Yellow hues in nature most often come from curcuminoids from turmeric, carotenoids – β-carotene, lutein – flavonols such as quercetin or crocin. These pigments differ in solubility and stability: carotenoids and curcumin are hydrophobic and sensitive to light/oxygen, whilst crocins are water-soluble, but also sensitive to light [46]. Eco-friendly pastes with acrylic or polysaccharide binders such as PVA, alginate, chitosan, combined with dye from turmeric, are successfully combined in screen-printing dyes and give pure yellow. The quality of the print and viscosity parameters have been described in detail by Khankaew and Panichayupakaranant [47]. Similarly, Sharma and colleagues developed an acrylic, functional water-based dye with curcumin for packaging, with antimicrobial effect and rapid drying time/adhesion to substrate [48]. For ink-jet dyes, Wang and colleagues used crocin (*Gardenia jasminoides)* and successfully printed on wool using ink-jet printing. The dye showed high stability, they added tannin after printing, which significantly improved colour durability, and the prints had antibacterial effect against *S. aureus* and *E. coli*. They obtained pure yellow hues [49]. Additionally, natural extracts rich in flavonoids/carotenoids such as Japanese knotweed rhizome gave sufficiently durable yellow hues for screen printing [16] as well as for inkjet printing [17].

**Black Shades**

Black pigments are crucial for contrast in textile and graphic printing; however, the extensive use of carbon black raises health and environmental concerns. Biochar produced by biomass pyrolysis and activated carbon represent promising alternatives, providing dark coloration while supporting waste valorisation. Activated carbon is widely used due to its high surface area, porosity, and surface functional groups that enable adsorption and purification functions [50][51]. Pigment obtained from pyrolysis of certified wood waste is used in aqueous dispersion for printing on various materials. The particles are smaller than 400 nm and irregular shaped, with black shade in print [52], while bamboo-derived activated carbon showed shades from light grey to dark grey and black, with excellent durability against washing, light and friction [53]. Coal and activated carbon from biomass have been extensively investigated for use in the textile industry. Particles of bamboo charcoal were added to spinning solutions during the production of polymer fibres [54][55], and the biomass wood coal was applied into fabrics by coating or printing techniques [39]. In one study, polyester non-woven fabric was treated with bamboo charcoal and acrylic resin to provide protection from infrared radiation and negative ions, as well as deodorisation [56]. In another study, activated carbon powder from coconut and palm kernel shells was applied by coating and pigment printing to cotton and polyester fabrics to provide deodorising properties against onion odour [57]. Çay and colleagues investigated the potential of using biochar from textile waste for improving the properties of textile materials. The addition of biochar improved moisture transport lowered drying time and increased water vapour permeability [58].

## 3. BINDERS AND MODIFIERS

Binders control the stability and rheology of printing pastes and enable uniform colour distribution, adhesion, and film formation on textile substrates. Sodium alginate is the most widely investigated bio-based binder due to its compatibility with aqueous extracts, adjustable viscosity, and limited interaction with colourants, while starch- and cellulose-based thickeners remain attractive biodegradable options but often require optimisation to improve mechanical durability. Polyvinyl alcohol (PVA) is a water-soluble, film-forming polymer with favourable barrier and abrasion resistance, frequently used as a mechanical reinforcement and printability enhancer in coating and printing systems [59]. Carrageenans, extracted from red marine algae, provide strong gel-forming and thickening behaviour and are increasingly explored in active and intelligent materials [60][61].

Current research increasingly favours multifunctional formulations based on binder–modifier combinations rather than single-component systems. Alginate- and carrageenan-based blends – often combined with CMC and anthocyanins – have been applied as pH- and TVB-N-sensitive indicators for fish and meat freshness monitoring [62][63][64]. In printed films and coatings, alginate/PVA and carrageenan/CMC mixtures offer tunable rheology and improved uniformity, while humectants such as glycerol support levelling during application [65][66]. Beyond packaging, alginate-based hydrogels filled with carbon nanomaterials (graphene/graphite) have enabled conductive inks for 3D printing and screen printing, supporting sensor and functional printing applications [67][68].

## 4. FUNCTIONAL PROPERTIES OF NATURAL INKS

One of the key advantages of natural dyes and pigments is not only improved environmental acceptability but also the potential to impart functional properties to printed textiles, enabling multifunctional and smart materials. The final performance of printed textiles depends on the substrate, colourant chemistry, printing method, and post-treatment, which jointly determine visual quality, durability (washing/light), and surface feel [69]. Compared with conventional finishing, printing allows a more economical and localised deposition of bioactive ingredients with controlled layer thickness and dosage, often reducing water and chemical consumption while enabling decoration and function within the same layer.

Bio-based binders combined with natural extracts can produce decorative prints with antimicrobial activity [5][70]. Chitosan is among the most frequently investigated antimicrobial additives and can be printed alone or combined with plant polyphenols to create functional patterns with attractive appearance [71][72]. In packaging-related systems, encapsulated essential oils applied by printing provide controlled-release effects (e.g., anti-odour/repellent layers) [73][74] , while pH-sensitive natural dyes (notably anthocyanins) are widely used in indicators responding to pH/TVB-N/VOC changes, from small "indicator spots" to full labels for fish and meat freshness monitoring [34][75][76]. Natural UV-absorbing compounds (e.g., flavonoids) can increase fabric UPF through printed layers, and mild crosslinking strategies can improve fixation and reduce leaching [69][77]. Green flame-retardant approaches based on phytic-acid-type systems have also been reported as simple coatings that can reduce flammability and, in some cases, moisture uptake [78][79]. Finally, biochar/graphite-based particles derived from agro-waste can be formulated into functional pastes for antistatic coatings, EMI shielding,

or adsorption layers; for wearable and sensor applications, additional optimisation of elasticity and wearability remains necessary [80][81].
Research conducted to date in the field of natural dyes and pigments shows that it is possible to formulate sustainable printing systems that imitate CMYK process colours. In this context, anthocyanins from agro-industrial waste represent the main source for magenta hues, whilst biochar, activated carbon and graphite give stable black and grey tones. The yellow component is in the literature often associated with readily available natural sources such as curcumin from turmeric or flavonoids from certain plant species, whilst cyan/blue is significantly harder to obtain from natural spectrum due to the limited distribution of plants which give stable blue pigments. For this reason, the largest number of investigations focus on the development of magenta and black components as the most promising for industrial application. This approach confirms that natural sources of colour, in combination with environmentally acceptable binders, can represent the foundation for the development of a process printing system with the potential to replace synthetic dyes in specialised applications of the textile and graphic industry.

## 5. CONCLUSIONS

This review indicates that eco-friendly printing based on natural pigments from agro-industrial waste is a promising route toward more sustainable textile production, offering both reduced environmental impact and opportunities for value-added functional textiles. Key conclusions are:

- Natural pigments from agro-industrial waste (e.g., anthocyanins, biochar, activated carbon) show strong potential to substitute conventional synthetic colourants in textile printing.
- Bio-based and eco-acceptable binders (sodium alginate, carrageenan) and supportive film-formers such as PVA enable stable, printable formulations across different printing techniques.
- Beyond coloration, natural-pigment prints can provide functional effects (antimicrobial, antioxidant, UV-protective), increasing product value.
- Remaining challenges include colour stability and scalability, requiring optimisation of extraction, formulation, fixation, and application parameters.
- Future work should prioritise standardised protocols for extraction and characterisation, together with long-term performance evaluation under real-use conditions.

Overall, the transition to sustainable textile printing is increasingly driven by environmental requirements and market demand. Addressing stability and scale-up limitations will be decisive for broader industrial adoption.

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