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Contribution of Plant Transfer Printing to Sustainable Fashion

Irena Šabarić, Ana Sutlović *, Jana Filipčić and Franka Karin

Faculty of Textile Technology, University of Zagreb, Prilaz baruna Filipovića 28a, 10000 Zagreb, Croatia; irena.sabaric@ttf.unizg.hr (I.Š.)
* Correspondence: ana.sutlovic@ttf.unizg.hr; Tel.: +385-1-4877-354

Abstract: Nowadays, there is a growing awareness of environmental protection, new findings in the field of sustainable chemistry, the use of biodegradable materials, and the increased use of eco-friendly textile products. For this reason, natural dyes are being used more and more frequently, giving rise to a new way of decorating textiles, namely, plant transfer printing, popularly known as "eco-printing", in which the shape and/or pigment of a plant is transferred to the textile. In addition, the great interest of the young generation in the application and research into the use of natural dyes can create incentives for cultural and social sustainability through the preservation of national heritage. Plant transfer printing is a method that combines scientific technology and artistic design with corresponding benefits for the eco system. The very fact that the patterns are unique and unpredictable brings out the notion of artistic freedom. In the work, plant transfer printing was carried out on undyed cotton material and on material dyed with pomegranate peels, walnut leaves, coffee, and aleppo pine bark. The influence of the pH value and the capillarity of the fabric, as well as the treatment of the leaves with iron(II) sulphate heptahydrate solution, on the aesthetics of the print and the colour fastness during washing was investigated. Based on the optimised parameters and a sustainable fabric design, the clothing collection "Hamadryad", inspired by Greek mythology, was realised.

Keywords: plant transfer printing; eco-printing; natural dyes; sustainable fashion

1. Introduction

Since the dawn of the human species, man has achieved colourful effects on textiles with natural dyes derived from mineral, vegetable, and animal sources. Natural dyes have been the only type of dye for a long series of years and humans have perfected their use and passed on traditional knowledge over generations. The production and use of synthetic dyes gradually replaced natural dyes, and, today, they have almost no commercial significance in the textile industry [1–7]. The modern textile industry faces the problem where it is both one of the world's most strategically important industries and one with the greatest negative impact on the environment. On a global scale, the textile and clothing industry is one of the largest, but, unfortunately, also one of the most environmentally damaging industries. Environmental problems are usually related to the consumption of energy, water, and chemicals, direct CO_2 emissions, and solid waste [8–10]. However, with the growing awareness of global warming, climate change, the limitation of water resources, and the problem of chemical waste disposal, dyeing with natural dyes [1–7] and plant transfer printing can be seen as a good solution and a step in the right direction in terms of environmental protection and the preservation of our planet [8–13].

One of the health-acceptable methods of textile dyeing, and certainly with a great contribution to human well-being, is the application of the plant transfer printing method on textile material with the aim of producing eco-friendly textile products. Plant transfer printing, i.e., eco-printing, has recently been developed mainly by boosting the creativity of fashion designers who are concerned with sustainable fashion. By promoting the idea of "eco-design", they want to help raise awareness of the harmful effects of the fast fashion and

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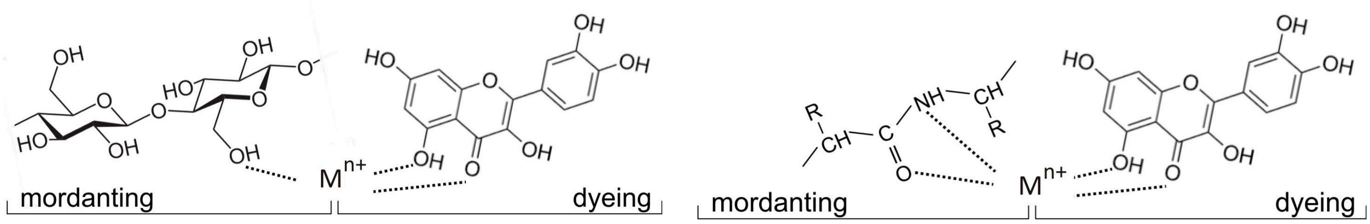
textile industries on environmental pollution and global warming. Eco-printing represents an improved, more environmentally friendly way of dyeing textiles by using natural plant dyes in the production process and, on the other hand, achieving very interesting, non-uniform prints that appeal to the artistic side of textile design. Natural materials from all parts of plants are used, i.e., roots, leaves, flowers, fruits, and bark. They serve as templates to achieve interesting colour shades and patterns on textiles [1–4]. In addition to the colour, the textiles processed in this way have multifunctional properties. This applies, in particular, to the benefits for human health. They offer good protection against UV radiation and have anti-fungal properties, and less unpleasant odours have been observed precisely because of their antibacterial and bacteriostatic properties [14–20].

Precisely because the natural plant dyes mainly belong to the group of flavonoid acid mordant dyes, they have been used throughout history mainly for protein materials. The author Ismal points out that plant transfer printing is suitable for applying these dyes to common cellulosic materials such as cotton, viscose, and Tencel, as well as their blends with polyester [9].

Sahu emphasises that the application of this method has an impact on the proportion of the positive consumer attitude according to their impact on the environment [10]. Manuja and colleagues have demonstrated good colour fastness using this technique and emphasise the importance of using natural dyes given their multifunctional properties [11]. Authors Tri and Nooryan [12] contribute to the combination of proven screen printing and vegetable transfer printing and to the subjective assessment of fastness. Considering the impossibility of spectrophotometric measurements, Nurmasitah [12] and team have taken a commendable step by evaluating the clarity of the subject, colour sharpness, uniformity of colour, and colour absorption based on the scores of 40 observers. However, this evaluation method also needs to be extended due to the statistical acceptability.

Despite its name, plant transfer printing belongs to the group of contact dyeing and can be carried out according to different principles. There is a physical (mechanical) method, popularly known as the Hapa-Zome technique, in which mechanical forces are used to imprint the plant material into the textiles, and another chemical method, in which natural mordant dyes and metal salts are used together. By using water vapour with natural materials such as flower petals or different leaves, very fascinating and unique patterns can be achieved. Plant transfer printing combines the chemical aspect of dyeing with the artistic freedom of designing plant prints, creating unique and very interesting designs. The term plant transfer printing refers to the direct use of plants in this process without extracting their natural substances. As the material is not uniform, the end result will certainly vary and it is not possible to achieve absolute repeatability of patterns, but each one will be unique [8–13]. However, to achieve an "eco" premise, both technical and chemical knowledge is required. By using the optimal chemicals and adjusting the process parameters, the print will look as clear and sharp as possible. In addition, the term "eco-printing" must emphasise the use of renewable and sustainable plant materials and the exploitation of invasive species. No protected plant species should be used or plant habitats jeopardised when using this technique. The modern application of natural plant dyes makes it imperative to use waste from other industries, easily renewable plants, and invasive species while caring for the preservation of biological diversity [21–26].

The principle for achieving colour effects with the plant transfer printing process is based on the fact that most natural plant dyes belong to the group of flavonoid acid mordant [1–5]. The use of acid mordant dyes is based on the treatment of textiles with metal salts (mordants), which influence the colour hue and colour fastness achieved. If the goal is to achieve lighter colours, aluminium salt is used as a dye. Plant transfer printing, however, emphasises the contrasting structure of the leaves and the textile substrate and usually uses iron salt, giving darker shades [1–7]. The chemical bonding of the dye molecules in the fibre structure takes place during fixation, and this is where colour fastness is achieved. During fixation, depending on the composition of the raw material, a reaction occurs between the imide, carbonyl, or hydroxyl groups of the textile material, metal ions from the mordant, and hydroxyl or carbonyl groups of the natural dye. Metal complexes with different colours are formed; i.e., the colours obtained and their properties are the result of the formation of ligands: fibre–metal ion–natural dye (Figure 1) [2–4,21].



(**a**) (**b**)

Figure 1. Treatment of textiles with natural mordant dyes: (**a**) cellulosic material; and (**b**) protein material [4,21].

Plant transfer printing is also associated with the notion of a sustainable method that seeks to minimise the negative impact of the textile industry on the environment. The use of natural dyes can be considered a sustainable method of processing, mainly because it results in the production of a small number of garments that are both unique and not mass-produced, so that there is no large demand for raw materials. It also promotes healthy and safe working conditions without health risks, as no harmful chemicals are used. Among other things, the amount of waste is reduced, and, as leaves, flowers, bark, and twigs are the main raw materials, it is possible to recycle/reuse them. As no strong chemicals are used, there is also no risk of polluting the soil, air, and water. At the same time, printing on plants can be a very effective way of reusing old clothes, giving old clothes a completely new outfit instead of buying new clothes. According to statistical estimates, the consumption of clothing will increase by 63% by 2030 and the amount of annual textile waste will reach 102 million tonnes. If the goal is to preserve the environment and not neglect future generations, sustainable fashion is a big step in the right direction [19,20].

By analysing the process parameters that affect the aesthetics and fastness of the colouration and the realisation of the finished garment, this research represents a contribution to sustainable fashion and is a bold step towards the possibility of moving from the laboratory scientific testing of samples to the creation of a clothing collection. In view of the exceptional colouring heterogeneity and colourfulness of the samples obtained by plant transfer printing, the success of the process presented in the paper is based on the author's many years of experience in the use of natural dyes for dyeing and textile printing.

2. Materials and Methods

2.1. Textile Material

We used 100% chemically bleached cotton fabric in plain weave from producer, Čateks d.o.o., Čakovec, Croatia. Textile has the following structural characteristics: weight 191.45 g/m^2, density in warp direction 26 cm^{-1}, and density in weft direction 25 cm^{-1}.

2.2. Plant Selection for Transfer Printnig

For the plant transfer printing, the plants available in the surrounding area, i.e., in the centre of the city of Zagreb, Croatia, were selected, which, according to the literature, could be used for this purpose [1–4]. These were the leaves of various maple species (e.g., silver-leaf maple, honeydew maple, and chub maple), walnut, birch, and red-leaved plum.

2.3. Plant Selection for Dyeing Process

The textile material was dyed with plant extracts from pomegranate bark, walnut leaves, aleppo pine collected in Croatia, and coffee grounds bought in a local shop.

2.4. Plant Transfer Printing on Uncoloured Fabric

The plant transfer printing on uncoloured fabric was tested with the following parameters:

- Influence of fabric pH—printing was performed in an acidic medium (pH 4 adjusted with tartaric acid, $C_4H_6O_6$, Kemika, Zagreb, Croatia), a neutral medium, and an alkaline medium (pH 8 adjusted with Na_2CO_3, Kemika, Zagreb, Croatia);
- Influence of capillarity—printing was performed under neutral conditions in all combinations of dry and wet fabric and leaves;
- Influence of leaf mordanting—leaves were treated for 10, 30, 60, and 120 min in a solution of 10 g/L iron(II) sulphate heptahydrate, $FeSO_4 \cdot 7H_2O$ (Kemika, Zagreb, Croatia);
- The influence of leaf positioning—the front and back of the leaves were used (neutral conditions, and dry substances)

2.5. Plant Transfer Printing on Pre-Dyed Fabric

Plant transfer printing was carried out on fabrics pre-dyed with natural plant dyes according to the following steps:

- Extraction of the dye was performed in soft water containing 100 g/L of plant raw material. The extraction was performed in a bath ratio of 1:40 (considering the mass of the plant matter) at 100 °C for 60 min. The bath was then cooled for 12 h and the extract was decanted.
- The dyeing process was performed in the prepared solution at a bath ratio of 1:30 bath ratio (considering the mass of the textile) at 100 °C for 60 min.
- The plant transfer printing was carried out on dry pre-dyed fabric with dry leaves immersed for 30 min in a solution of 10 g/L $FeSO_4 \cdot 7H_2O$.

2.6. Fixation

Fixation was carried out with steam at the same time as the printing process. Leaves treated with iron salts were placed on the fabric. Aluminium foil is placed on top; everything is wrapped around a wooden roll, wrapped again with aluminium foil, and tied with string. The fabric prepared in this way is placed in a pot of hot water for 90 min, and then wrapped and cooled for 60 min.

2.7. Wash Fastness

The wash fastness of cotton fabrics was tested in a laboratory apparatus for wet and dyeing processes—Polycolor, Mathis, Basel, Switzerland. The test was performed according to the standard ISO 105-C06:2010 (A2S) Textiles-Colour fastness tests-Part C06: Colour fastness in domestic and commercial laundry [26] using 2 g/L standard detergent (James Heal ECE A, without optical brighteners and without phosphates), with a bath ratio of 1:20 (considering the mass of the textile), a temperature of 40 °C, and a soaking time of 30 min. The alkalinity of the solution was pH 8.

Considering the appearance of the colourful pattern on the textile and the purpose of the test, the evaluation was based on the designer's subjective assessment.

2.8. Applying the Recipe—Creating a Clothing Collection

In the final phase, the clothing collection "Hamadryad", inspired by Greek mythology, i.e., Greek nymphs who were born and died together with the tree they were connected to, was realised from the created patterned textiles. In realising the collection, the inspiration of the designer and the freedom to use plant transfer printing to achieve the desired unique design were respected.

3. Results and Discussion

The results shown vary considerably depending on the process parameters: the pH value, use of the mordant, capillarity of the fabric, and properties of the plant material. The adjustment of these parameters is the result of the knowledge and experience of a textile chemist and has a considerable influence on the intensity and sharpness of the print, as well as on the washing fastness. However, the choice of the "best" result certainly depends on the designer's attitude and the market's requirements for the aesthetics and purpose of the textile material.

3.1. Printing on Uncoloured Fabric

3.1.1. Effect of the pH Value of the Fabric

Figure 2 shows the effect of the pH value of the fabric on the quality of the plant transfer printing. The research was carried out by pre-treating the fabrics in acidic, neutral, and alkaline media. The leaves were pre-treated for 45 min in a solution of 10 g/L $FeSO_4{\cdot}7H_2O$. It was found that the palest and dullest samples are obtained in an acidic medium, and sharper contours are more visible in a neutral medium, while the samples are most intense in an alkaline medium. The transition of plant pigments into textiles is also the best in an alkaline medium, and brown and green shades are obtained, while grey tones are obtained in an acidic medium, which is due to the use of iron salt as the mordant.

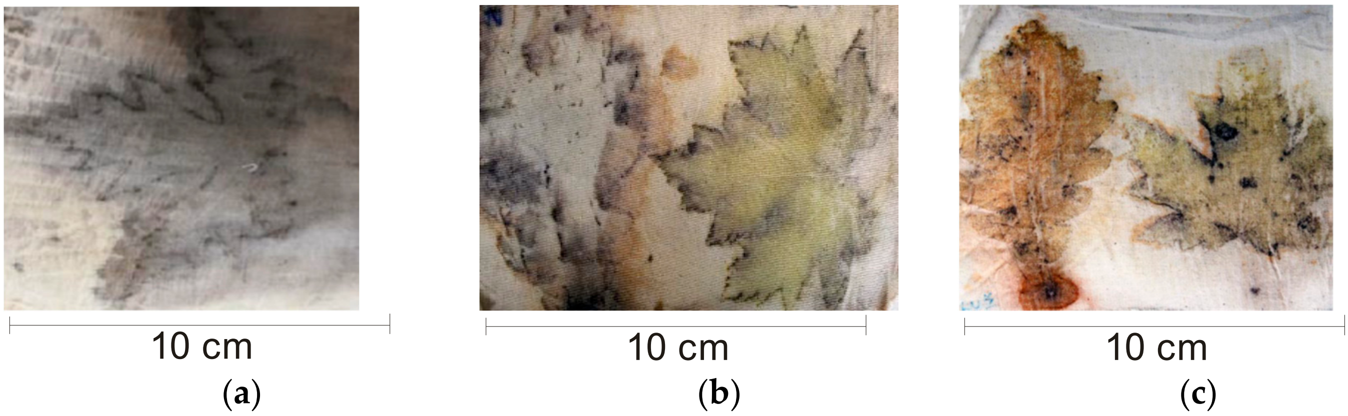


Figure 2. Effect of the pH value of the fabric on plant transfer printing: (**a**) pH 4; (**b**) pH 7; and (**c**) pH 8.

It is important to note that it was not possible to objectify the colouristic analysis, since the specific colouration is characterised by a multitude of shaded transitions and the simultaneous presence of different shades of a single hue. Therefore, the colouristic analysis was carried out on the basis of the theoretical guidelines for harmonic and contrasting relationships between colours.

When the pH value shifts to alkaline, an increase in colour chroma is observed. The grey to grey-green hues seen in Figure 2a change to colours of the yellow-orange and yellow-green spectrum, in an alkaline medium (Figure 2c). The intensity of the colouration, which results from the specific ratio of saturation (chroma) and lightness, is also more pronounced in the same samples, and the specific colour characteristic of the natural plant dyes is more pronounced. In terms of the harmonious and contrasting relationship between the colours, in Figure 2c, the relationship of analogous colours can be observed, i.e., of different colours but in a spectral sequence, which is exactly characteristic of the harmony of natural colours.

In addition, the non-uniformity of colouration and the specific shading as well as the presence of different shades of the same hue are visible, due to which, as already mentioned, the colouration obtained could not be objectively analysed.

3.1.2. Influence of the Capillarity of the Fabric

The investigations into the influence of the capillarity of the cotton fabric and leaves are shown in Figure 3. Combinations of dry or wet fabric and dry or wet leaves are shown. The research was carried out in a neutral medium using iron salt as a mordant. It can be observed that the best contact and, therefore, the sharpest print is achieved with dry textiles and dry leaves. Any source of moisture disturbs this relationship and leads to a blurring of the sample. Depending on the design idea, it is possible to create a pattern of varying sharpness by combining wet and dry leaves.

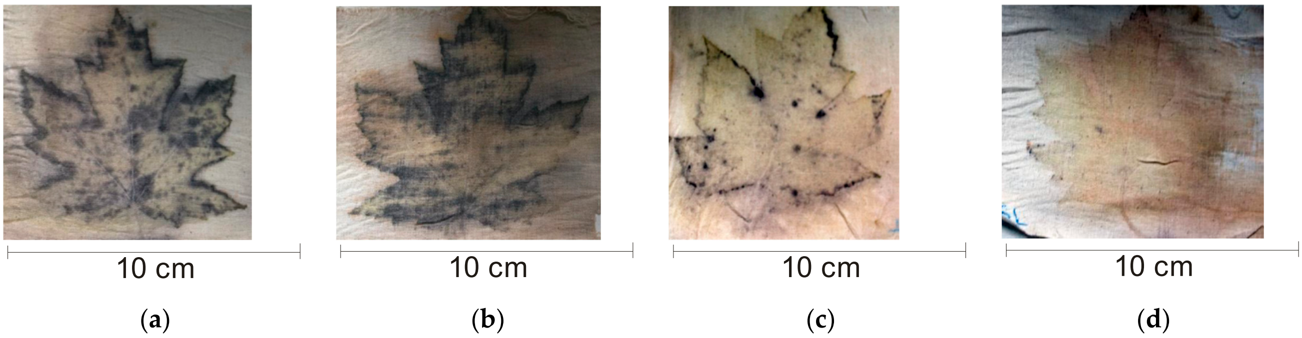


Figure 3. Influence of the capillarity of the fabric on plant transfer printing: (**a**) dry textile–dry leaves; (**b**) dry textile–wet leaves; (**c**) wet textile–dry leaves; and (**d**) wet textile–wet leaves.

Figure 3a shows the print created by the use of dry fabric and dry leaves. A sharp contour of the leaf and the leaf veins can be recognised. When using wet leaves on the same dry fabric (Figure 3b), the pattern is still clearly visible, but the inner structure is destroyed. This phenomenon is logical and to be expected due to the capillary movement of the dye on a wet surface. If the goal is greater sharpness, a contribution to this can be expected if a larger-diameter roller is used for fixing, which helps to reduce the number of layers of fabric and the passage of moisture through the layers. When wet (3c) or dry (3d) leaves are used on a wet fabric, a reduction in colour intensity is visible. In these combinations, the contour is pale and barely visible; i.e., the effect of the print pattern is lost. The evaluation of these patterns as satisfactory or unsatisfactory depends on the designer's ideas for the realisation of the textile design and clothing design. From the aspect of colour analysis, the colour of the already mentioned yellow-green to yellow-orange spectrum can be observed in addition to grey. No sample can be said to be completely intense, as the characteristic colour obtained with plant pigments, although harmonious, is generally of lower intensity. However, a comparison of Figure 3a–d shows that the colour is more pronounced when the textile or textile web I is wet, although the sharpness of the contour is lost.

3.1.3. Influence of Time Mordanting of Leaves

Leaves treated in a solution of 10 g/L $FeSO_4 \cdot 7H_2O$ were placed on a pH-neutral dry fabric for the following periods: 10, 30, 60, and 120 min. The results shown in Figure 4 indicate that extending the mordanting time does not have a good effect on the print. After a longer treatment of the leaves in the metal solution, dark spots can be observed on the sample, which may indicate iron deposits. An excess of iron on the textile material can have a negative effect on the quality of the textile; i.e., damage can occur over time. If the designer wants to create a garment, iron deposits can have negative effects on the skin and cause allergic reactions. This negative phenomenon can be avoided by optimising the metal salt concentration. For the reasons mentioned above, metal salts should be used in the lowest possible concentration. It is also necessary to use containers adapted to the selection of plants.

3.1.4. The Influence of Leaf Positioning—Front/Back

Figure 5 shows that the choice of leaf side, i.e., front or back, can influence the appearance of the sample obtained. By using the front side of the leaves, patterns with a more accentuated colouration will be obtained, and, with the reverse side, the structure of the leaf itself (veins and petiole) will be expressed. In addition to a more intensive pigment deposition, the positioning of the front of the leaves on the textile led to a higher colour

Chroma, which is reflected in the more intensely perceptible yellow-orange colouration of the sample in Figure 5a compared to Figure 5b.

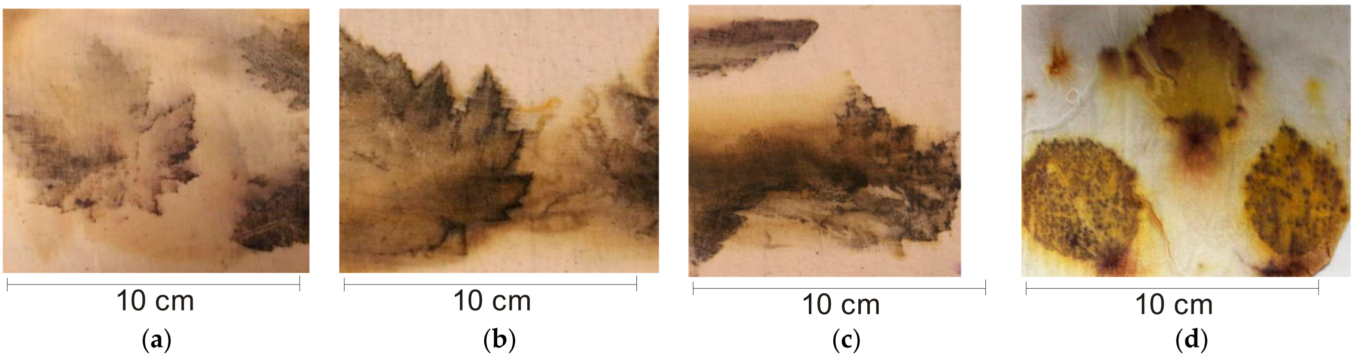


Figure 4. Influence of time mordanting of leaves on plant transfer printing: (**a**) 10 min; (**b**) 30 min; (**c**) 60 min; and (**d**) 120 min.

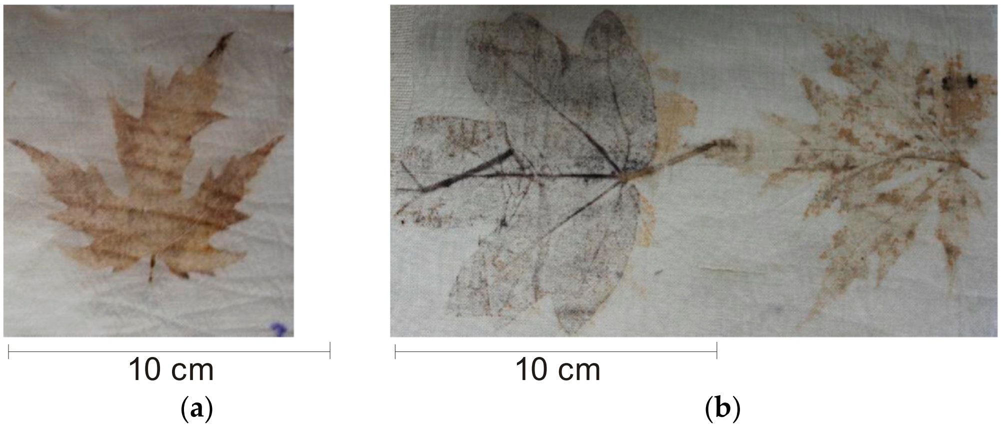


Figure 5. The influence of leaf positioning on plan transfer printing: (**a**) front of the leaves; and (**b**) back of the leaves.

3.2. Plant Transfer Printing on Pre-Dyed Fabric

Sustainable textile design was also realised through a combination of plant transfer printing and pre-dyeing of the textile material (Figure 6). Based on previous results, plant transfer printing was performed with dry dyed fabric and dry leaves pre-treated in a solution of 10 g/L $FeSO_4{\cdot}7H_2O$ for 30 min. Depending on the choice of plants, a different colour is obtained, i.e., a chromatic yellow fabric from pomegranate bark, a reddish one from Aleppo pine bark, and soft pastel achromatic colours without an accentuated tone from walnut and coffee leaves. With such combinations, a sustainable designer can choose an accentuated contrast or a subtle pattern.

The colours obtained are the result of the chemical structure of the dyes obtained by extraction (Figure 7) and their reaction with iron ions, as shown in Figure 1 [2,3]. The peel of the pomegranate (*Punica granatum* L.) contains 28% of the tannins punicalagin and punicalin, which are responsible for the yellow colour (Figure 7a). Walnut leaves (*Juglans regia* L.) contain the brown pigment juglone (Figure 7b) as a glycoside. The most common chemical compound in coffee is caffeine (Figure 7c), an alkaloid from the xanitol group, which is responsible for the brown colour. Procyanidin (Figure 7d) from the flavonoid class is responsible for the red colour extracted from the bark of the aleppo pine [2,3]. The final colour of the fabric depends on three parameters: the raw material composition of the textile material, the chemical structure of the natural dye, and the choice of metal. Their interaction creates a metal complex with a specific colour. If one of these three parameters changes, the colour of the textile changes. It is also known that dark colours are achieved by combining natural dyes and iron salts [1–7].

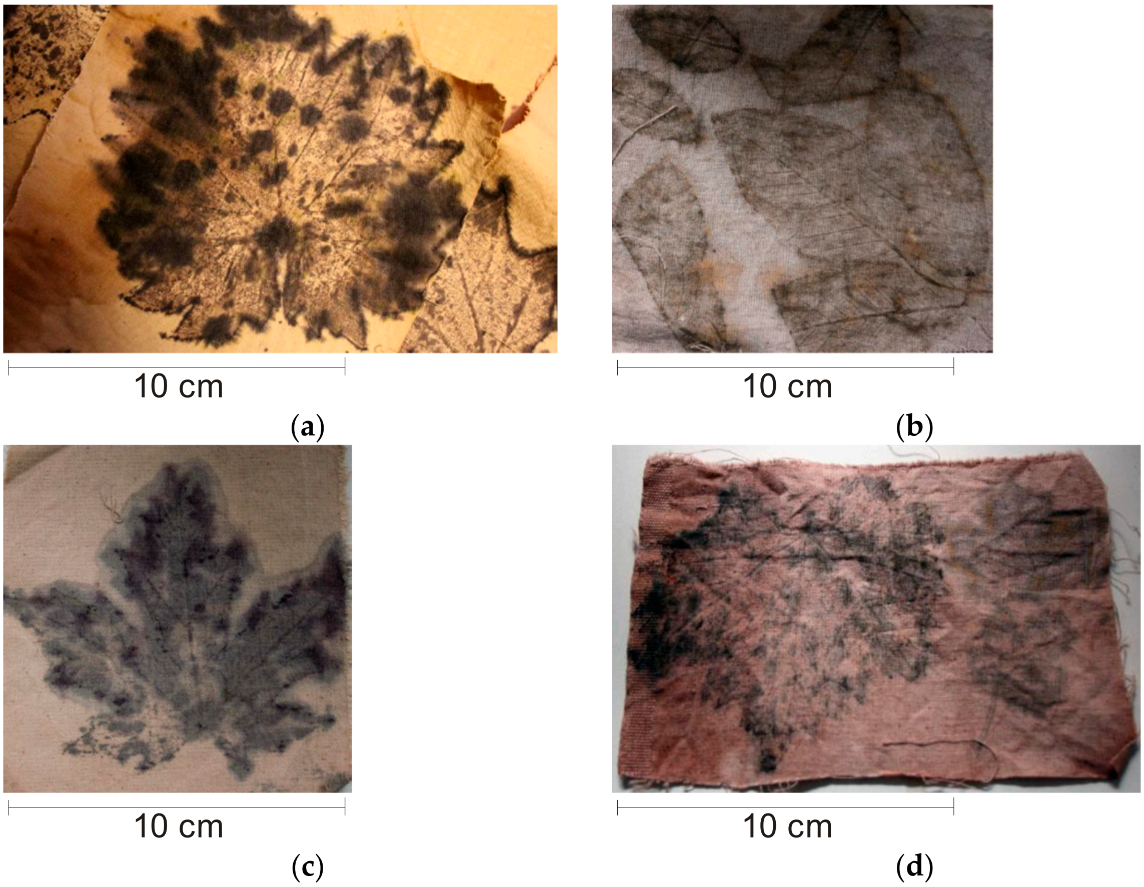


Figure 6. Samples of plant transfer printing on pre-dyed textiles: (**a**) pomegranate peels; (**b**) walnut leaves; (**c**) coffee; and (**d**) aleppo pine bark.

The strongest colour contrast was achieved when printing on a substrate dyed with a natural dye from pomegranate peels. The colours of the yellow spectrum with their brilliance and high level of lightness, as well as their medium to high Chroma, have the property of highlighting the printed pattern and placing it visually in the foreground (Figure 6a). It is, therefore, not only a matter of contrast due to the different intensities of black-brown and yellow colours, but also this specific property of the colour yellow.

Even the achromatic colour of the background with a high lightness in Figure 6c has a pronounced difference in intensity with the printed pattern of very low lightness and Chroma, but does not lead to such a strong contrast visually. In the case of example 6d, if the effect achieved is considered from a colouristic point of view, a complementary relationship between the reddish background and the greenish print with a low lightness can be defined, which brings the entire composition into balance without, however, creating an accentuated contrast.

3.3. Wash Fastness Test

The fastness to washing test (Figures 8 and 9) is an important parameter for the colouring of textiles intended for the manufacture of clothing. The samples are found to have a satisfactory stability. After washing, the samples become even more intense, which may be due to the chemical reactions on metal salts and plant substances (Figure 1). In an alkaline detergent solution, the ionisation of the molecule can occur (Figure 7), which can lead to a change in the colour obtained [4,5,17,18].

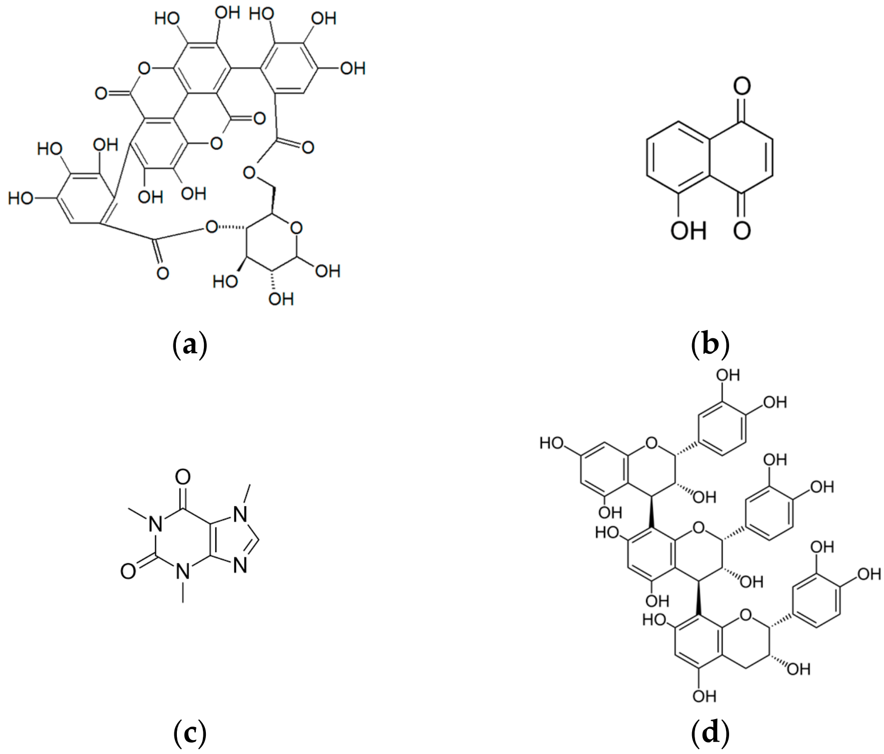


Figure 7. Chemical structures: (**a**) punicalin (4,6-gallagyl-D-glucose); (**b**) juglone (5-hydroxy-1,4- naphthoquinone); (**c**) caffeine (1,3,7-trimethylxanthine); and (**d**) procyanidin (2-(3,4-Dihydroxyphenyl)-2-((2- (3,4-dihydroxyphenyl)-5,7-dihydroxychroman-3-yl)oxy)chroman-3,4,5,7-tetraol).

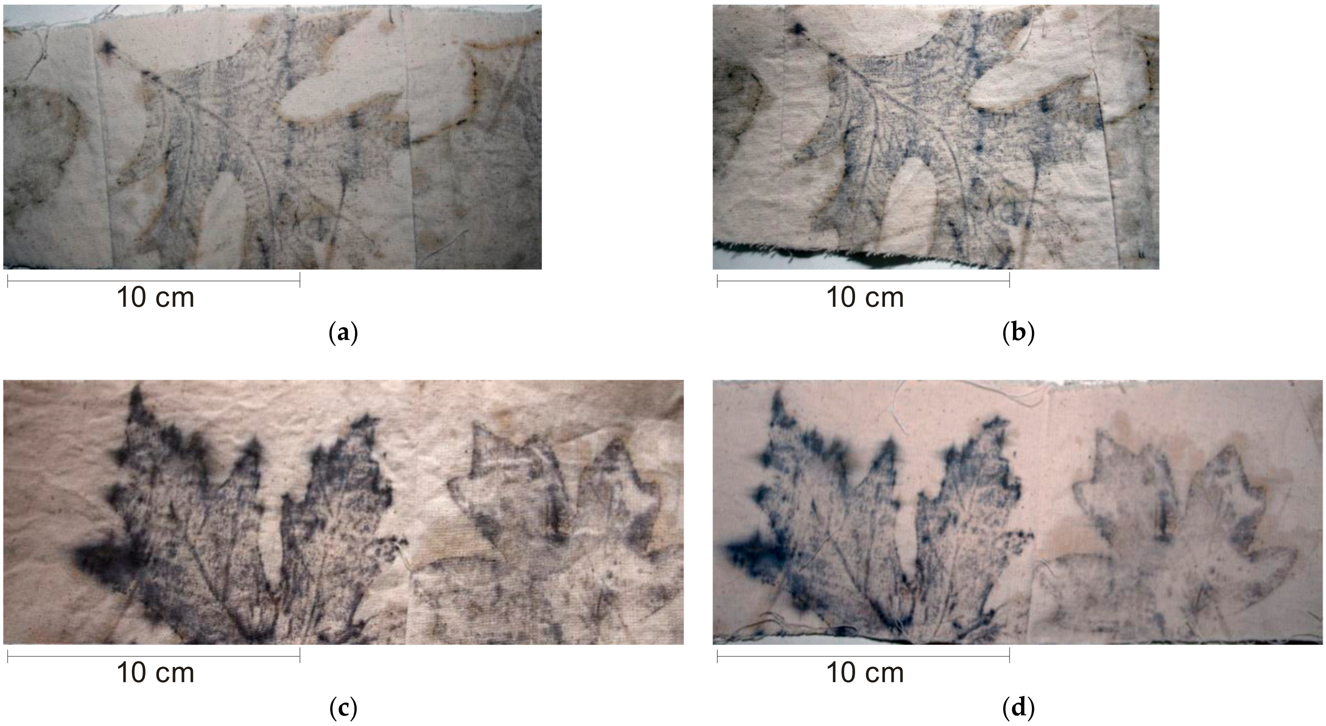


Figure 8. Colour fastness to washing of undyed samples: (**a**,**c**) before washing process; and (**b**,**d**) after washing process.

Figure 9. Colour fastness to washing of pre-dyed samples before washing process/after washing process.

It can also be seen from the chemical structures shown in Figure 7 that, in addition to the colour, these compounds also have hydroxyl or carbonyl groups in favourable positions for the formation of metal complexes with trivalent iron ions. It is precisely for this reason that dark-coloured imprints of plant structures were obtained. In addition, the iron ions form co-ordination bonds with the textile material, which ensure good colour fastness.

3.4. "Hamadryad" Clothing Collection

Based on the results of the textile design obtained by combining technological parameters, fabrics with transfer printing on undyed fabrics and fabrics dyed in pomegranate bark were selected for the realisation of the collection. Plant transfer printing was performed in an alkaline medium with dry dyed fabric and wet and dry leaves pre-treated in a solution of 10 g/L $FeSO_4 \cdot 7H_2O$ for 30 min. This choice emphasises the pattern of leaves, which is in line with the creations of the clothing collection (Figure 10a) inspired by Greek nymphs, i.e., the trees they were associated with.

The realisation of the "Hamadryad" collection (Figure 10b) is a true example of sustainable textile design. The collection consists of classic garments such as skirts, shirts, jackets, and trousers, with interventions in cuts that attempt to imitate trees. The garments from the collection are easy to mix and match and can be an interesting detail in combination with everyday clothing.

Figure 10. Selection from the "Hamadryad" clothing collection: (**a**,**c**) fashion illustrations; and (**b**,**d**) realisations.

When applying the plant transfer printing technique in the realisation of textile design and clothing collections, it is important to emphasise the co-operation between sustainable textile chemistry and design. From a technological point of view, it would be logical to choose methods that achieve sharp contours and visible motifs. However, designers often opt for the technique of plant transfer printing, which gives them blurred contours and indistinct motifs. It is important that the process is carried out in such a way as to obtain a final product that satisfies the market with all its characteristics in terms of aesthetics, functionality, and colour fastness. It is precisely for this reason that the impossibility of carrying out measurements commonly used in colour analysis, such as spectrophotometric measurements, becomes a less important problem.

4. Conclusions

The result of plant transfer printing is based on chemical and technological knowledge about the application of natural plant dyes. By varying the process parameters, different colours and effects can be achieved, the choice of which depends on the attitude of the sustainable designer.

By using iron(II) sulphate heptahydrate as a mordant, it has been confirmed that the reaction of natural mordant dyes and the use of iron(II) ions produce dark colours, i.e., high-contrast prints. This contrast can be further enhanced by the choice of plants for pre-dyeing the textile material. For sharper contours in textile printing, it is also necessary to prepare dry material and dry herbs that have previously been treated with metal salts.

The "Hamadryad" clothing collection was realised through the synergy of textile chemistry and sustainable design. Considering the fact that it is inspired by Greek mythology, i.e., Greek nymphs who were born and died together with the tree they were connected to, the contribution to sustainable design was confirmed by the choice of cutting and plant printing technique. Clothing produced in this way can help to raise the market's awareness of its impact on environmental protection.

This research is a direct contribution to the imperative of understanding this connection, to understand and target natural colourants and to develop branding techniques to achieve effects with natural colourants in the context of sustainable fashion, as well as textile and clothing production.

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