Just coloured paper: Toning paper using natural dyes

Susan Catcher

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Introduction
The use of natural colours to tint paper to line, mount or in-fill when conserving Western or Eastern objects is already common practice, with traditional watercolour, Japanese paint sticks and Chinese pigment chips being available to colour both paper and silk. Dyes from natural plant material such as yasha, derived from the Japanese alder cone, Alnus japonica and indigo prepared from leaves of the Polygonum tinclonium plant are commonly used in conservation and these have been adequately explored in a previous paper. However, this use of natural colourants could be extended to embrace some of the more traditional dyes made from indigenous plants such as lichens, berries and tree cones or bark still used in the traditional craft of the dyeing of textiles and paper.

Dyes
A dye is a coloured substance that has an affinity for the substrate to which it is being applied, in this case paper, as opposed to a pigment that is usually an insoluble powder mixed with a binder to produce paint. Dyes are generally applied in an aqueous solution and often require a mordant to improve their fastness. The choice of indigenous dye material was initially governed by the season (autumn/winter) and via a literature search as to what might constitute a good uptake on cellulose fibres, that is, cotton. Driven historically by the textile trade, there are plenty of recipe books with a choice of plants and mordants being recommended for use on wool, silk and cotton fibres.²

Dyes that work well with wool and silk, being proteins, do not necessarily give a good colour on cellulose and especially not paper, which cannot be boiled. So it was of interest to discover that the internationally acclaimed South Korean artist, Kwang-Young Chun, was exhibiting vibrant, naturally coloured paper structures developed from his earlier monochrome, 3D pieces. He uses units called bojagi,³ or triangular pieces wrapped in old printed mulberry paper, which are dyed using plant material: gardenia seed heads, sappanwood, tea, indigo and mugwort leaves (Fig. 1).


2 V. Thurstan, The Use of Vegetable Dyes (The Reeves Dryad Press, 1977); R. Adrosko, Natural Dyes and Home Dying (Dover, 1971).

3 The traditional wrapping of objects for safekeeping or protection during transport, as explained in J.C. Welchman, 'Terraform: The Art of Kwang Young Chun’, in Kwang Young Chun Mulberry Mindscapes (New York: Skira Rizzola, 2014), 27.

Fig. 1 Coloured bojagi
Having used yasha, a dye from the Japanese alder cone, and indigo for many years to colour sheets of paper for lining and the in-filling of large missing areas of museum objects such as Chinese wallpapers and Western posters, it made me wonder if there was a potential to use other plant material to create a variety of natural colours to choose from. Therefore, I decided, first, to recreate the colours that Chun used, but, secondly, I also wanted to see what range of colours I could obtain from British indigenous plants and whether this was a viable alternative to toning with pigments. Occasionally, over a period of time toned in-fills stand out as the pigment used to colour the missing area has faded at a slower rate than the organic pigments of the object. If natural dyed papers were seen to fade at similar rate to natural pigments, then why not use these as an alternative?

Methodology

One litre of cold tap water (pH 8) was added to 100 gm of cut-up or powdered plant material and placed in a saucepan where it was brought up to just below boiling point. The length of cooking time varied depending on the part of the plant and if a specific recipe was being followed, as for weld and woad. In general, barks and harder material were heated for two hours, whereas leaves and fruits were heated for about 30 minutes. The resulting dye was strained and a first direct dyeing was applied warm (50° C) to the paper either by brush or by dipping. The left-over dye was then cooled and a pH reading taken to decide on the appropriate mordant. The mordant was chosen in relation to both a common usage on cellulosic fibres, and the pH reading of the initial dye. Some dye classes are pH sensitive, so alum (potassium aluminium sulphate) and chalk (calcium carbonate) were used either separately or in combination. Two grams of mordant was put into 500 ml of the remaining dye solution and heated up to approximately 40° C until the salt dissolved. The resultant dye was strained and then brushed warm onto the paper or dipped as before and allowed to dry.

Mordants are usually an inert salt used to ‘fix’ the dye onto the fibre and generally have an affinity for both the fibre and colouring matter. They form an insoluble precipitate within the fibre structure holding onto the dye and thus prevent fading. Pliny the Elder understood this and in AD 70 wrote:

\[ \ldots \] the white cloth being first stained in various places, not with dyestuffs, but with drugs, which have the property of absorbing colours. These applications do not have to appear on the cloth, but when the cloths are afterwards plunged into a cauldron containing the dye liquor they are withdrawn fully dyed of several colours, according to the different properties of the drugs which have been applied \[ \ldots \]

Application

Sample papers were chosen to represent commonly used paper in both Eastern and Western conservation: Chinese xuan zhi containing a large percentage of grass fibres, normally rice and a smaller percentage of than pi; Japanese sekishu shi made of mainly kozo fibres from the inner bark of the mulberry tree with some wood pulp; and a Western rag paper made up of cotton fibres. All the fibres consisted of cellulose polymers, so natural dyes that have an affinity for cotton were chosen and the mordant likewise. The dye was applied by brush on a table to the two Oriental papers using a slightly different technique for each: the Chinese xuan zhi was initially lightly humidified with the tips of the brush dipped in the dye solution, flapped to allow the air to reduce creases that take up the dye and hold it in the fold, creating a darker line. The dye is then applied using a Chinese paibi to the smooth side in three sweeps and a fill-in along the length of the sheet.
A second sheet is placed directly over the first and repeated until a block of paper has been constructed. The last sheet is just smoothed into place to soak up the dye from the lower sheets.

The Japanese *sekishu shi* is also brush dyed, but the table is initially coated with the dye to enable an even take-up, before a sheet of paper is brushed smooth side up with a charged Japanese dye brush. A new sheet is again placed directly on top and the dye brush smooths the sheet into position (Fig. 2). Both the Chinese and Japanese papers are removed in a block and placed flat to dry on felts. The *xuan zhi* block is divided when dry as its short fibre has little wet strength; the *sekishu shi* paper is separated out when slightly damp, having a longer fibre and good wet strength.

Western rag paper does not dye evenly using the brush method, and was therefore dipped dyed for a period of 10 minutes before being removed and pegged on a line to dry. The even uptake of dye using this method was more difficult to control.

**A little dye chemistry**

Plants usually contain a mixture of natural dyes and are grouped according to their chemical structure, with 50% being classed as flavonoid. There are four main types of flavonoids: flavones, flavonols, anthocyanidins and anthcyanins, each made up of water-soluble compounds derived from a 2-phenyl-1, 4-benzopyrone. They are all pH sensitive and water soluble, so the dye is easily extracted. Flavones and flavonols have yellowish colours, examples being weld and onion skins. The yellow becomes much deeper in solutions of high pH, with the colours of flavones tending not to fade in strong light, unlike the flavonols, although they are generally much paler (Fig. 3). Anthocyanidins and anthcyanins are the most highly coloured of the flavonoids and are responsible for the scarlets, reds, violets and blues found in many flowers, fruits and vegetables, such as the dye extracted from blackberries and raspberries (Fig. 4). Anthocyanidins bond to glucose molecules to become anthcyanins. They are soluble in water and are easily extracted into weak acidic solution. As with all flavonoids, the colour is pH dependent.

Other important groups are the anthracenes, which subdivides into anthraquinones where madder dye is identified, and betanin, giving colour to beetroot. Betanin is a betacynin and, like anthcyanins, is glucosidic. However, its molecules contain nitrogen, making it less reactive to pH changes. Tannins are present in most plant tissues, being produced from the flavonoids, especially the anthcyananins, when tissues break down, but are also found in quantity in the bark of oak and oak galls (Fig. 5).
There are two main types of carotenoid: carotene is the orange colourant found in orange fruit, vegetables and orange flowers, whilst xanthophyll is the yellow of saffron and that of plant leaves. Most are hydrocarbons, although some also contain oxygen. They are fat soluble and do not dissolve in highly polar solvents such as water but in organic solvents that are not too polar. For the purposes of this study, this group is not represented. However, chlorophyll is: there are four types of chlorophyll, one of them, chlorophyll b, can be extracted from nettles, spinach and mugwort (Fig. 6). The resultant colour is rarely used to dye textiles but is to be found in the food industry. Lastly, indigoid provides the blue colourant of indigo and woad (Fig. 7). It is a natural vat dye and are generally more light-fast as opposed to acid dyes, which are anionic and applied in an acid dye bath. Most natural dyes are acid dyes.
Table 1  Summary of the common dye characteristics for each of the chemical class of colours examined.

<table>
<thead>
<tr>
<th>Dye</th>
<th>Chemical class</th>
<th>Characteristics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ghardinia</td>
<td>Flavonoid</td>
<td>Gives yellow to brown colours, depending on mordant; are all pH sensitive.</td>
</tr>
<tr>
<td>Weld</td>
<td>Flavonoid</td>
<td></td>
</tr>
<tr>
<td>Onion skins</td>
<td>Flavonoid</td>
<td></td>
</tr>
<tr>
<td>Tea</td>
<td>Flavonoid</td>
<td></td>
</tr>
<tr>
<td>Sappanwood</td>
<td>Homoisoflavonoid</td>
<td>Various red shades but mordant and pH dependent.</td>
</tr>
<tr>
<td>Blackberry</td>
<td>Anthocyanin</td>
<td>Anthocyanin dyes give the reds, violets and scarlet colours.</td>
</tr>
<tr>
<td>Raspberry</td>
<td>Anthocyanin</td>
<td></td>
</tr>
<tr>
<td>Indigo</td>
<td>Indigoid</td>
<td>Blue or purple colour and require no mordant as are natural vat dyes.</td>
</tr>
<tr>
<td>Woad</td>
<td>Indigoid</td>
<td></td>
</tr>
<tr>
<td>Oak bark</td>
<td>Tannin</td>
<td>Brown, grey or black. Can be applied directly or as a mordant dye.</td>
</tr>
<tr>
<td>Oak Galls</td>
<td>Tannin</td>
<td></td>
</tr>
<tr>
<td>Beetroot</td>
<td>Betacyanin</td>
<td>Similar to anthocyanins but not pH sensitive.</td>
</tr>
<tr>
<td>Mugwort</td>
<td>Chlorophyll</td>
<td>Green of the chlorophyll masks the yellow dye of the plant.</td>
</tr>
<tr>
<td>Nettle</td>
<td>Chlorophyll</td>
<td></td>
</tr>
</tbody>
</table>

The chemical interaction between the dye and the fibre can have a number of variables: the class of dye and the type of fibre, which in this study is cellulose from plant material; including several possible chemical interactions from simple hydrogen bonding to the chemical reaction of the mordant. Each of these contributes to the uptake of the dye, the colour produced and how permanent the colour is when exposed to light. But the loss of colour can be due to many factors affecting both the chemical and physical state of the dye: whether the chemical structure of the dye is symmetrical or not, what chemical class it is, the size of the dye molecules and the substitute group on the dye molecule. Most natural dyes fade, and this has been well documented, but it is the rate of fade and if there is a change of hue that is of interest here.

Many of the museum objects we work on contain light-sensitive media requiring low lux levels for a limited time exposure. At the Victoria and Albert Museum this is usually expressed as 50 lux for two years in every 10 for an average of 10 hours a day under UV-protected glass (36,000 lux hours). The use of some modern watercolour pigments or dyes made specifically for paper, such as Cartesolâyes, do not necessarily mimic the rate or type of fade exhibited by the organic pigments used in some Eastern paintings or prints. Fading rate curves first described by Giles in the 1960s and confirmed by Patricia Cox Crews 20 years later, define five fading curve types: Type I, a fading rate which decreases steadily over time; Type II, a linear or constant fade; Type III, a linear or constant fade; Type IV, an initial darkening followed by a slow fade; and, lastly, Type V, which fades increasingly with time (Fig. 8). Most natural dyes fall in the Type II fading curve with indigo a Type III and consequently considered the most light-fast of all natural dyes.

Fading experiments

Each sample was cut to size (70 x 50 mm) and attached to an acid-free board with a dab of wheat-starch paste to the upper edge to hold it in place. An aluminium foil covering was put over the top half of the samples and held in place by elastic bands to prevent light seepage. Each strip was exposed to ‘natural’ light (which fell at between 0–6000 lux for approximately

8 This is a very simplified chemical description and a more detailed study can be found in J. Kirby, M. van Brommel and A. Verhecken, Natural Colorants for Dyeing and Lake Pigments (Archetype Publications Ltd, 2014).


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Fig. 8 Giles diagram of fading rate curves re-plotted as percentage changes in concentration over time.

Fig. 9 Light readings over the 4-week period.

10 hours a day) averaging 3250 lux, through a north-facing Pilkington UV-filtered glass for four weeks. Blue dyed wool indicator strips of 1–4 were also attached\(^\text{13}\) and photographs taken every week to document any changes. A light metre was placed in the same orientation to the samples, and the sample strips were moved around in a predetermined order each week to try to even out any light irregularities (Fig. 9).

BOIC LFS4—January 1987
BOIC LFS3—August 1985
BOIC LFS2—November 1985
BOIC LFS1—November 1982
It must be mentioned that this was not an empirical measurement but a visual response in order to initially reject dyes that either faded rapidly or changed hue so as to make them unsuitable for long-term conservation. After a week, sappanwood direct dye on all the papers showed a change of hue; after two weeks, sappanwood (mordanted), beetroot and direct-dyed samples on all three papers showed one noticeable fade, with blackberry fading on the sekishu shi; after four weeks, onion, weld, gardenia, beechwood, nettle and blue wool indicator strip BOIC LFS1—November 1982 showed one noticeable fade.

Table 2 Coloured dyes showing one noticeable fade on the three paper samples over a 4-week period in natural light.

<table>
<thead>
<tr>
<th>Week 1</th>
<th>Week 2</th>
<th>Week 3</th>
<th>Week 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rag Paper</td>
<td>Xuan zhi</td>
<td>Sekishu shi</td>
<td></td>
</tr>
<tr>
<td>Sappanwood (direct)</td>
<td>Sappanwood (direct)</td>
<td>Sappanwood (direct)</td>
<td></td>
</tr>
<tr>
<td>Sappanwood (alum)</td>
<td>Sappanwood (alum)</td>
<td>Sappanwood (alum)</td>
<td></td>
</tr>
<tr>
<td>Beetroot (direct)</td>
<td>Beetroot (direct)</td>
<td>Beetroot (direct)</td>
<td>Blackberry (CaCO₃)</td>
</tr>
<tr>
<td>Blackberry (CaCO₃)</td>
<td>Blackberry (CaCO₃)</td>
<td>Ghardenia (direct)</td>
<td></td>
</tr>
<tr>
<td>Indigo (direct)</td>
<td>Onion (direct)</td>
<td>Onion (direct)</td>
<td>Beecwood (direct)</td>
</tr>
<tr>
<td>Onion (direct)</td>
<td>Beechwood (direct)</td>
<td>Nettle (direct)</td>
<td></td>
</tr>
<tr>
<td>Beechwood (direct)</td>
<td>Nettle (direct)</td>
<td>Weld (Na₂CO₃)</td>
<td></td>
</tr>
</tbody>
</table>

To verify the above results, another three sets of samples were prepared and were illuminated in a light box under UV protection Perspex using 20 F20W/AD light bulbs for 36 hours at 9–10,000 lux. This is the equivalent of 50 lux during a 10-hour day for a two-year period (36,000 lux hours). Light readings were taken at specific points to ascertain the areas where the lux levels were highest/lowest. Each sample strip was then placed in the area of the highest light levels and moved around every 12 hours, to again even out the irregularities (Fig. 10).
Conclusion
Initially I was interested in the range of colours that could be obtained from a variety of indigenous plant material as inspired by Kwan Young Chun, but it soon became obvious that the use of these natural dyes to tone paper could be utilized in conservation to mimic the natural fading qualities of organic painted or printed pigments such as dayflower blue or safflower red. It is a common fact that natural dyes fade, and it was this characteristic that I wanted to pursue. The interaction of the dyes on three different conservation papers meant that the parameters were widened, but the different fibre combinations did have a bearing on the uptake of colour and the rate of fade. Generally, the dyed colour was strongest on the rag paper, with the sekishu shi weakest. This could be attributed to the different weight of the papers and the density of the fibres as opposed to the method of dye penetration or fibre uptake. But in the initial natural light fading experiment, the mulberry fibre seemed to fade more readily and over a larger number of colours, in comparison to the rag paper and xuan zhi. This was a fairly brutal test, with the natural light levels averaging 3250 lux over 224 hours, equalling approximately 200 years at 50 lux. However, the results of the controlled test using a light box were more encouraging, with only the sappanwood direct dye demonstrating a visual change in hue and extensive fading on all paper samples. The experimentation is still ongoing and will be repeated several times to give more conclusive results. The samples will be CIE L*A*B* Colour Space scored to identify any minute colour changes.

Two samples of toned lining papers have been used on two museum objects, one an oriental book cover using a direct beetroot dyed sekishu shi and the other a xuan zhi dyed with mordanted beechwood to line a Chinese panel depicting a Qing dynasty courtesan, currently being displayed in the ‘Shoes, Pleasure or Pain’ exhibition (Fig. 11). Both these will be monitored for changes to the dyed paper and the object colour as I want to produce a dyed paper that fades at a similar rate as a light-sensitive museum object. The two fading rates will be compared by photographic documentation (Chinese courtesan) and CIEL*A*B* colour model of both the book cover and vegetable dyed lining paper.

Fig. 11 Xuan zhi dyed with beechwood (direct dye) used as a lining

Acknowledgements
Thanks to Kwang-Young Chun who invited me to his home and shared his ideas, Rosalie Kim, Samsung Curator of Korean Art at the V&A, who arranged the trip and Seungmi Baik, Assistant Curator at the National Museum of Korea.

Abstract
The use of natural colours to tint paper to line, mount or in-fill when conserving Western or Eastern objects is already common practice, with traditional watercolour, Japanese paint sticks and Chinese pigment chips being available to colour both paper and silk. Dyes from natural plant material such as yasha, derived from the Japanese alder cone, Alnus japonica and indigo prepared from leaves of the Polygonum tinclonium plant are also commonly used in conservation. It was of interest to discover that the internationally acclaimed South Korean artist, Kwang-Young Chun, was exhibiting vibrant, naturally coloured paper structures, which are dyed using plant material: gardenia seed heads, sappanwood, tea, indigo and mugwort leaves. This led the author to wonder if this use of natural colourants could be extended to embrace some of the more traditional Western dyes made from indigenous plants such as lichens, berries and tree cones or bark still used in the traditional craft of the dyeing of textiles and paper. Results from paper dyed with a variety of plant material will be discussed as to whether this is a viable option especially with regard to light fading.

Biography
Susan Catcher initially qualified in textiles and used this specialism to work as a Volunteer teacher in Kenya for three years. Whilst living in a mud hut she studied for a BA honours degree in art history with the Open University and on returning to the UK she attended Camberwell College of Arts, London and graduated with an MA in Conservation in 1998. After a period of freelance work, she completed an internship in paper conservation at the Victoria and Albert Museum and never really left. She is currently a Senior Paper Conservator in charge of loans with an interest in mixed-media and ephemeral objects whilst also specializing in East Asian art. She was the lead conservator for the recently acclaimed exhibition ‘Masterpieces of Chinese Painting’ and has given several papers related to this topic including one on a Chinese erotic painting and a 17th Century export wallpaper. She has recently spent a period of time at Nanjing Museum, China to refine her ink rubbing and scroll mounting skills. She is an accredited member of Icon.